Control of Toxic Chemicals in Puget Sound

Phase 3: Primary Sources of Selected Toxic Chemicals and Quantities Released in the Puget Sound Basin

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Control of Toxic Chemicals in Puget Sound

Phase 3: Primary Sources of Selected Toxic Chemicals and Quantities Released in the Puget Sound Basin

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Abstract

During 2010, the Washington State Department of Ecology conducted a study to (1) identify the primary sources of toxic chemicals in the Puget Sound basin and (2) estimate annual releases of these chemicals. This source identification and inventory was undertaken as part of the Puget Sound Toxics Loading Analysis project and complements the other loading studies done as part of the project.

Fourteen chemicals and chemical groups (chemicals of concern [COCs]) were addressed, and the quantities of COCs released annually from numerous sources were estimated. Estimates of COC releases were based on data obtained from various information resources; no sampling or other field data collection was conducted for this project.

The study estimated that petroleum is released in the largest quantity annually among the COCs evaluated, followed by zinc which is the only other COC released at a rate greater than 1,000 metric tons (t) per year. Lead, polycyclic aromatic hydrocarbons, copper, and triclopyr are released at rates greater than 100 t/yr, and nearly 35 t of phthalates are released annually. Releases of arsenic, cadmium, mercury, polychlorinated biphenyls, polybrominated diphenyl ethers, polychlorinated dioxins and furans, and nonylphenol are generally near one t/yr or less.

Recommendations are made to increase the scope, accuracy, and usefulness of this large-scale assessment of chemical releases. These recommendations include refining COC release estimates, increasing the number of COC release estimates, and re-evaluating the list of chemicals addressed.

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Executive Summary

The overall goal of the present project (*Primary Sources)* is to balance the chemical loading data generated from the Puget Sound Toxics Loading Analysis (PSTLA) with information on chemical releases in order for the Washington State Department of Ecology, the Puget Sound Partnership, and others to develop and implement a toxics reduction and control strategy.

Specific objectives of the *Primary Sources* project are to:

- Identify the primary sources of selected chemicals released to the Puget Sound basin.
- Make preliminary estimates of the quantity of chemicals released annually from each source.

This report describes methods used to calculate quantities of chemicals released from a variety of sources and presents results of these calculations. The annual release estimates calculated in this report are preliminary and are not intended to be highly precise. Instead, they provide information about the relative magnitude of the chemical amounts released from different sources, and the reader is encouraged to use the results in that context.

The chemicals of concern (COCs) selected for the *Primary Sources* project are nearly identical to the core group of chemicals analyzed for the various toxicant loading studies carried out under PSTLA:

- Arsenic
- Cadmium
- Copper
- Lead
- Mercury
- Zinc
- Polychlorinated biphenyls (PCBs)
- Polybrominated diphenyl ethers (PBDEs)
- Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs)
- Polycyclic aromatic hydrocarbons (PAHs)
- Phthalate esters
- Triclopyr
- Nonylphenol
- Petroleum hydrocarbons

The annual quantities of these COCs that are released from numerous sources are estimated. The geographical study area (Study Area) includes Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. portion of the watershed for Puget Sound and the Straits.

For the purpose of the present project, the term *source* is strictly defined as *the object or activity from which a COC is initially released to environmental media or released in a form which can be mobilized and transported in an environmental pathway*. The term *primary source* has been adopted for the present report to distinguish the initial release of a COC from a secondary release, such as mobilization of a chemical from a toxic cleanup site. In addition to distinguishing a *primary source* of a COC from a secondary release or a natural source, the definition of *primary source* also distinguishes an initial release of a COC from its presence in transport/delivery pathways such as stormwater, atmospheric transport, and wastewater treatment plants. Transport and delivery pathways were not addressed in this report.

Summary of Results

A summary of the total annual releases estimated for each COC is shown in Table ES-1. Petroleum is estimated to be released in the largest quantity annually, followed by zinc which is the only other COC released at a rate greater than 1,000 metric tons (t) per year. Lead, PAHs, copper, and triclopyr are released at rates greater than 100 t/yr, and nearly 35 t of phthalates are released annually. Arsenic, cadmium, mercury, PCBs, PBDEs, PCDD/Fs, and nonylphenol are generally released at rates near one t/yr or less.

The diversity of major sources among COCs is illustrated in Table ES-1. In general, industrial, commercial, and institutional point sources do not account for the largest releases of COCs. Instead, a variety of diffuse sources account for a majority of the COC releases.

Runoff and leaching of chemicals from roofing materials generally appears to be a more important source of metals than organic chemicals. For two metals – cadmium and zinc – release from roofing materials appears to account for the majority of the total release, and nearly 20% of arsenic is released through roof runoff. Annual zinc releases from roof runoff were estimated to be particularly large (1,300 t/yr). There are also substantial releases of copper and lead from roof materials (27 t/yr and 18 t/yr, respectively). Leaching of metals from plumbing components also accounts for substantial releases of copper and zinc (39 t/yr and 30 t/yr, respectively).

Vehicle and road-related COC releases occur primarily through wear of vehicle components, combustion of fuel, and leaks of motor oil and fuel. Abrasion of brake pads accounts for a large annual release of copper (37 t/yr), and tires are a substantial source of zinc (82 t/yr). Vehicle-related fuel combustion releases large quantities of COCs, accounting for 14% (42 t/yr) of the total PAH release (due to gasoline and diesel fuel combustion) and 10% (0.9 g TEQ[1]/yr) of the total PCDD/F release (due primarily to diesel fuel combustion).

Petroleum represents the largest COC quantity released annually to roadways and other impervious surfaces. Most of the motor oil lost via drips and leaks (6,100 t/yr) likely occurs along roadways or impervious surfaces – parking lots and driveways – connected to roadways. In addition, gasoline is released at a rate of 570 t/yr during on-road vehicle fueling at the pump. PAHs contained in uncombusted petroleum are also released along with the petroleum at a rate of approximately 11 t/yr.

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[1] Toxic Equivalent

Table ES-1. Total Releases and Largest Sources for Each Chemical of Concern (COC).

COC	Total Release in the Puget Sound Basin[a] (t/yr)	Major Sources
Petroleum	9,200	Motor oil drips and leaks, and improper disposal of used oil. Gasoline spillage during fueling.
Zinc	1,500	Roofing material leaching. Vehicle tire abrasion.
Lead	520	Ammunition and hunting shot use. Loss of fishing sinkers and wheel weights. Roofing material leaching. Aviation fuel combustion.
Total PAHs	310	Woodstoves and fireplace combustion emissions. Vehicle combustion emissions. Creosote-treated pilings, railroad ties, and utility poles.
Copper	180 - 250	Pesticides use on urban lawns and gardens.[b] Residential plumbing component leaching. Brake pad abrasion. Roofing material leaching. Vessel anti-fouling paint leaching.
Triclopyr	150	Herbicide use on crops and golf courses.
Phthalates	34	Personal care products. Polymer (primarily PVC) off-gassing. Industrial, commercial, and institutional air emissions. Roofing material leaching.
Total PCBs	2.2	Electrical equipment spills and leakage.[b] Residential trash burning. Building sealant (caulk) volatilization and abrasion.
Cadmium	0.96	Roofing material leaching.
Mercury	0.54	Consumer product improper disposal. Crematoria and industrial plants air emissions.
Total PBDEs	0.68	Furniture, computer monitors, and other components of residential and commercial indoor environments.
Arsenic	0.79	Industrial air emissions. CCA-treated wood leaching. Roofing material leaching.
Nonylphenol[c]	0.18	Industrial, commercial, and institutional air emissions.
PCDD/Fs	0.000009[d]	Backyard burn barrels.

t: metric ton (appr. 2.2. tons).

[a] The Study Area which includes the Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. portion of the watershed for Puget Sound and the Straits.

[b] Estimate is highly uncertain.

[c] Sources were not fully assessed.

[d] Expressed as Toxic Equivalents (TEQs).

Combustion sources other than petroleum account for major releases of PCDD/Fs, PAHs, and mercury. In the Study Area, combustion emissions from backyard burn barrels account for three-quarters (7.3 g TEQ/yr) of the total PCDD/Fs released. Woodstoves and fireplaces are the largest source of PAHs (110 t/yr; 38% of total PAH release). As much as one-quarter of the mercury release to air in the Study Area may be through combustion emissions. Much of this may be due to fossil fuel combustion, although mercury emissions from crematoria and cement plants may originate from the source material rather than the fuel. A large source of PCBs (280 kg/yr; 13% of the total PCB release) is also released from a combustion source: residential trash burning.

Since few of the COCs assessed are typically associated with agriculture, annual releases from agricultural uses are generally small. Triclopyr, the only COC used exclusively as a pesticide (herbicide), is an exception with large quantities (130 t/yr) applied to crops in the Study Area. There are also some releases of metals in fertilizers applied to agricultural crops, with zinc releases being the largest (41 t/yr).

Copper is used in agriculture as both a pesticide and a micronutrient. Total annual crop usage is estimated to be 15 t/yr, with 10 t/yr of copper used as an agricultural pesticide alone. A potentially large release of copper (up to 73 t/yr) is due to its use as an urban pesticide, although the accuracy of this estimate is highly uncertain. The release of copper from vessel anti-fouling paint is also substantial (26 t/yr). All pesticidal uses of copper combined account for anywhere between 10% and 60% of the anthropogenic copper release in the Study Area.

PAH releases from creosote-treated railroad ties, marine pilings, and utility poles account for over 33% (110 t/yr) of the PAHs released annually in the Study Area.

PCB releases from electrical equipment leakage appear to be substantial, possibly as much as 2,000 kg/yr. However, this amount may also include indoor leakage or PCB which is immediately cleaned up and contained; therefore, this likely over-estimates actual PCB release in the Study Area by a large degree.

PBDE and phthalate releases are difficult to assess due to their passive emissions from materials containing them. For PBDEs, their loss via air emissions and dust particles into air and dust from commercial offices and homes, followed by subsequent release to the outdoor environment, appears to be the major release pathway. Phthalates may be released in the same manner, although releases from domestic products – including personal care products (e.g. fragrance, nail polish) – may be more important sources. For both of these COCs, attachment to indoor dust and subsequent release to sanitary sewers may also be an important release pathway.

Limitations and Uncertainty

The information used to calculate the quantities of COCs released annually from primary sources was obtained through peer-reviewed literature, government and non-governmental organization publications, government databases, direct communications with experts, readily available marketing data, and other miscellaneous information resources. *No sampling or other field data collection was conducted for this Primary Sources project.*

All of the COC release estimates calculated for this project have an associated uncertainty due largely to the assumptions required to compute the estimates; in some cases the estimates of the COC releases may be highly imprecise. For each release estimate calculated, the assumptions used to derive the estimate were described and a discussion of the uncertainty associated with each assumption was addressed. In some cases, there was uncertainty surrounding the rates used for COCs released from specific objects or activities (i.e. release rates). In other cases, there was uncertainty about the appropriateness of using particular information to scale releases to the Study Area.

Due to the uncertainty associated with all of the release estimates, the reader should view the results as a gauge against which to compare the relative magnitude of chemical releases, but the absolute value for each release estimate should be viewed with caution.

The reader is also reminded that estimated quantities of COCs released in the Study Area do not necessarily translate to equivalent loads being delivered to Puget Sound. A variety of factors affect a chemical's behavior and fate once released in the environment, which in turn governs its presence in delivery and transport pathways. In many cases, the specific COC source and the mechanism of release may have more environmental relevance then the absolute quantity released. Mechanisms of COC release and the receiving environmental media (air, water, or soil) following release are implicitly or explicitly mentioned along with each source addressed. However, an analysis and discussion of COC behavior, fate, and degree of attenuation following release are beyond the scope of this project.

The reader is also cautioned that quantities of COCs released do not simply translate to a potential for harm; differences in mobility, location of release, persistence, bioavailability, and comparative toxicities are more important factors. However, an assessment of the relative hazard posed by COCs is beyond the scope of the present report.

Information on COC sources will be brought together with other PSTLA information on loading, pathways, and relative hazards of COCs in a subsequent report (*Assessment Report*) which synthesizes information to better focus priorities for toxic chemical reduction and control strategies. For instance, the *Assessment Report* will compare the COC release estimates presented in this report to COC loads delivered to Puget Sound through various pathways. These release/delivery comparisons may help to explain the relationships between COC sources and COC loading/pathways, thereby helping to guide decisions about source control. The *Assessment Repo*rt will also provide an assessment of the relative hazards posed by COCs in various environmental media across the Puget Sound region.

Recommendations

Recommendations for strategies to control toxic chemicals in the Puget Sound basin are beyond the scope of the present report. However, recommendations to increase the scope, accuracy, and usefulness of this large-scale assessment of toxic chemical releases are as follows:

- Refine COC release estimates.

  The need to refine estimates is largely dependent on the users of these data and their ability to make decisions based on either the rough estimates presented here or on a requirement for more precise estimates.

- Increase the number of sources for which COC releases are estimated.

  The following primary sources should receive top priority:

  - Evaluation and assessment of major zinc sources not addressed in the report including releases to marine waters from sacrificial anodes, leaching from guard rails, streetlight and road sign standards, chain link fences, and other outdoor galvanized material, as well as use of zinc salts for moss control on rooftops.
  - Assessment of all major sources of nonylphenol.
  - Assessment of the direct release of petroleum to water from 2-stroke outboard engines.
  - Assessment of the proportion of indoor and outdoor leakage of PCBs from transformers and capacitors.
  - Assessment of PBDEs from auto shredder and electronics recycling facilities.

- Re-examine the list of chemicals comprising the most important chemicals in the Puget Sound basin based on new information or new ways to evaluate existing information.

  Conducting an evaluation to determine the most important contemporary toxic chemicals in Puget Sound was beyond the scope of the present study. A thorough evaluation of these chemicals would help target the primary sources to be investigated in future projects.

Introduction

Background and Purpose

The Puget Sound Partnership has identified the need to study toxic chemical loadings to Puget Sound in order to understand the relative contributions from various sources of contaminants (toxicants) to the ecosystem. To accomplish this goal, a multi-agency effort is underway to assess the loading and effects of toxicants in the Sound, commonly referred to as the Puget Sound Toxics Loading Analysis (PSTLA).

To date, the bulk of effort in the PSTLA has been devoted to studying the loading of toxicants through various pathways, such as direct atmospheric deposition to Puget Sound, surface water runoff, wastewater treatment plants, and water exchange at the ocean boundary. However, relatively little attention has been given to obtaining information about how these toxicants are initially released to the environment.

For the Washington State Department of Ecology (Ecology), the Puget Sound Partnership, and other agencies, preventing the initial release of contaminants rather than treatment or cleanup is typically the preferred solution to contend with pollution. For instance, a major priority identified in the 2020 Puget Sound Action Agenda (PSP, 2008) is to "*Prevent water pollution at its source [since] many of our efforts have focused on cleaning up degraded waters and sediments, but insufficient resources have been devoted to stopping pollutants before they reach our rivers, beaches, and species*."

The present study (*Primary Sources*) is a preliminary attempt to gather and compile information on the sources of the most common and important toxicants studied among the various loading studies. The goal is to balance the loading data with information on the releases of chemicals in order for Ecology, the Puget Sound Partnership, and others to develop and implement a toxics reduction and control strategy. The strategy will address the most important sources of toxic chemicals.

Objectives

Specific objectives of the *Primary Sources* project are to:

- Identify the primary sources of selected chemicals released to the environment.
- Make preliminary estimates of the quantity of chemicals released annually from each source.

It is widely recognized that the specific source, mechanism of release, and medium in which a chemical is released is often more important than the absolute quantity released in determining the delivery of a chemical to surface waters or other delivery pathways. While the project team recognizes that none of the PSTLA projects alone provide enough information to guide decisions for reducing toxicants in Puget Sound, each project provides a piece of the puzzle to shape the larger picture.

Information on sources of chemicals of concern (COCs) will be brought together with other PSTLA information on loading, pathways, and relative hazards of COCs in a subsequent report which synthesizes information to better focus priorities for control strategies (*Assessment Report*; Ecology, 2011a). For instance, the *Assessment Report* will:

- Provide comparisons between COC release estimates presented in this report to COC loads delivered to Puget Sound through various pathways.
- Compare the relative hazards posed by COCs through a toxicity screening process.

These release/delivery/hazard comparisons may help to explain the relationships between COC sources and COC delivery, and will consider the relative importance of COCs, thereby helping to guide decisions about source control.

This report describes methods used to calculate quantities of chemicals released from a variety of sources in the Puget Sound region and presents results of these calculations. As mentioned previously, these are preliminary estimates and are not intended to be highly precise. The reader is therefore encouraged to focus on the relative magnitude of the chemical amounts released among sources rather than to isolate information about a release from a single source.

Scope, Approach, and Organization of the Project

Scope

Numerous elements of the project scope required definitions and scope boundaries prior to identifying and quantifying chemical releases. These elements included the selection of the chemicals to be addressed, definition of primary sources, and the geographical study area and resolution of the project. Each element is discussed in the following sub-sections.

Selection of Chemicals

The list of chemicals considered for this project was based on the 17 toxic chemicals or chemical groups (i.e. COCs) addressed in the initial (Phase 1) project to estimate loading of chemicals to Puget Sound (Hart Crowser et al., 2007). The list was selected by a Chemicals of Concern Workgroup that had been convened to recommend a list of chemicals based on previous work and best professional judgment. The workgroup sought to choose chemicals *"… that harm or threaten to harm the Puget Sound ecosystem and those that represent, or serve as an indicator for, a particular class of chemicals."*

The workgroup settled on a list of chemicals that largely mirrored those identified by the then lead agency for Puget Sound (Puget Sound Action Team; Redman et al., 2006). This list of COCs became the core group of chemicals analyzed for many of the subsequent loading studies, although numerous chemicals were added and some excluded for particular studies.

COCs were selected not only for their potential to be harmful, but also to represent a group of chemicals that may be present in a variety of transport/delivery pathways such as atmospheric deposition, surface water runoff, and wastewater treatment plant effluent.

To assess whether the list of 17 chemicals analyzed in loading studies was appropriate for this *Primary Sources* study, a cross-program Ecology workgroup was convened to provide suggestions. Chemicals frequently detected in Puget Sound-wide studies, those that frequently exceeded water or sediment quality standards, and chemicals leading to consumption advisories were among the criteria used by the group to evaluate chemicals. In the end, the group agreed that the 17 chemicals appeared to be suitable for the *Primary Sources* project, but future work should include additional evaluation of each chemical's importance as well as evaluation of other chemicals.

COCs selected for the *Primary Sources* project are as follows:

- Arsenic
- Cadmium
- Copper
- Lead
- Mercury
- Zinc
- Polychlorinated biphenyls (PCBs)
- Polybrominated diphenyl ethers (PBDEs)
- Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs)
- Polycyclic aromatic hydrocarbons (PAHs)
- Phthalate esters
- Triclopyr
- Nonylphenol
- Petroleum hydrocarbons

This list deviates from the original list of 17 due to exclusion of two chemical groups – DDT compounds and oil & grease[2] – which were not deemed appropriate for the *Primary Sources* study.

DDT sources were not inventoried in the analysis of primary sources since it was banned nearly four decades ago, and DDT had no uses other than as an agricultural insecticide and limited use as an urban pesticide.

Unlike other COCs, oil & grease does not refer to one or more specific chemicals, but instead refers to a group of chemicals with loosely related chemical and physical properties (including non-volatile hydrocarbons, vegetable oils, animal fats, waxes, soaps, greases, and related materials). The working definition of oil & grease is based on the analytical method to determine the concentration in water or soil. More specifically, oil & grease is the fraction of a sample that is extractable by n-hexane, and is sometimes referred to as hexane extractable material (HEM).

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[2] The original list of 17 chemicals included "oil or petroleum product" which included petroleum hydrocarbons as well as oil & grease.

The present study also addresses PAHs as a single chemical group whereas the loading studies address these chemicals as three distinct groups: low molecular weight PAHs, high molecular weight PAHs, and carcinogenic PAHs.

PAHs were addressed in total since: (1) they are typically released as mixtures of low- and high-molecular weight constituents, (2) expressing them as total PAHs simplifies the discussion, and (3) splitting total PAHs into sub-groups is generally only needed where they are compared to environmental criteria or used in health assessments.

Definition of Primary Sources

The term *source* is used in a variety of ways with regard to chemicals in the environment. For instance, stormwater, wastewater treatment plants, and atmospheric deposition are commonly referred to as sources of chemicals. However, these are more accurately termed pathways, since they only serve to transport and mobilize chemicals from one location to another, or, in the case of wastewater treatment plants, act as a focal point for chemical transport (and treatment).

For the purpose of the present project, the term *source* is strictly defined as: *the object or activity from which a COC is initially released to environmental media or released in a form which can be mobilized and transported in an environmental pathway*.

The term *primary source* has been adopted for the present report to distinguish the initial release of a COC from a secondary release, such as a release from a toxic cleanup site. The following examples illustrate the distinction between primary sources and pathways and secondary sources:

- Examples of *primary sources*
  - PAHs released to air from wood burning
  - Copper released to air, stormwater, and roadside ditches from brake pad wear
  - PCBs released to soil from transformer leakage
  - Triclopyr released to roadside ditches during herbicide application

- Examples of *pathways and secondary sources*
  - Atmospheric deposition
  - Stormwater
  - Treated wastewater discharges from wastewater treatment plants
  - Leaching or mobilization of contaminated soil from a cleanup site

The present report addresses only ongoing COC releases from primary sources.

Geographical Study Area and Resolution

The geographical study area (hereafter referred to as Study Area) includes Puget Sound, the U.S. portion of the Straits of Georgia and Juan de Fuca, and the entire U.S. portion of the watershed for Puget Sound and the Straits (Figure 1). This is consistent with study areas used in previous and ongoing projects in the PSTLA (e.g. Hart Crowser et al., 2007; Envirovision et al., 2008).

The Study Area was not divided into the 14 study areas used to develop loading estimates in other studies. Calculating release estimates for COCs by the 14 study areas would have required additional effort and time, yet added little to the overall goal of the project. However, the data generated in the report generally lends itself to re-calculation at a higher resolution (e.g. the 14 study areas) if this appears warranted.

In some cases, data were available to calculate releases based on the precise geographical area and boundaries of the Study Area as described above. In other cases, the 12-county Puget Sound region – Clallam, Island, Jefferson, King, Kitsap, Mason, Pierce, San Juan, Skagit, Snohomish, Thurston, and Whatcom Counties – was used as a surrogate for the Study Area (Figure 2). While some county boundaries do not fall fully within the Study Area, the majority of population and development within those counties does, rendering any discrepancies minor.

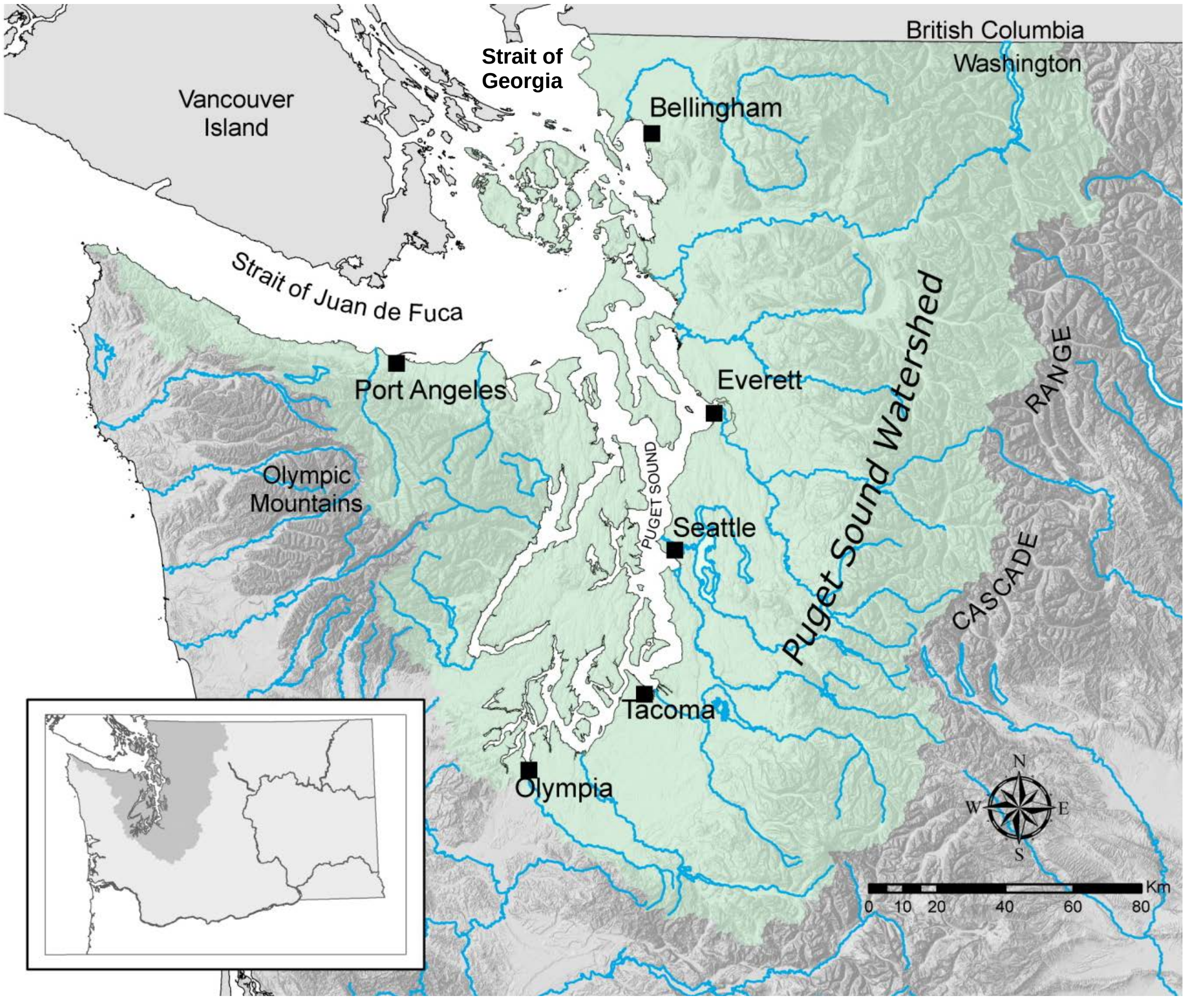


Figure 1. Map of Study Area: Watershed Area for Puget Sound and the U.S. Portion of the Strait of Georgia and the Strait of Juan de Fuca.

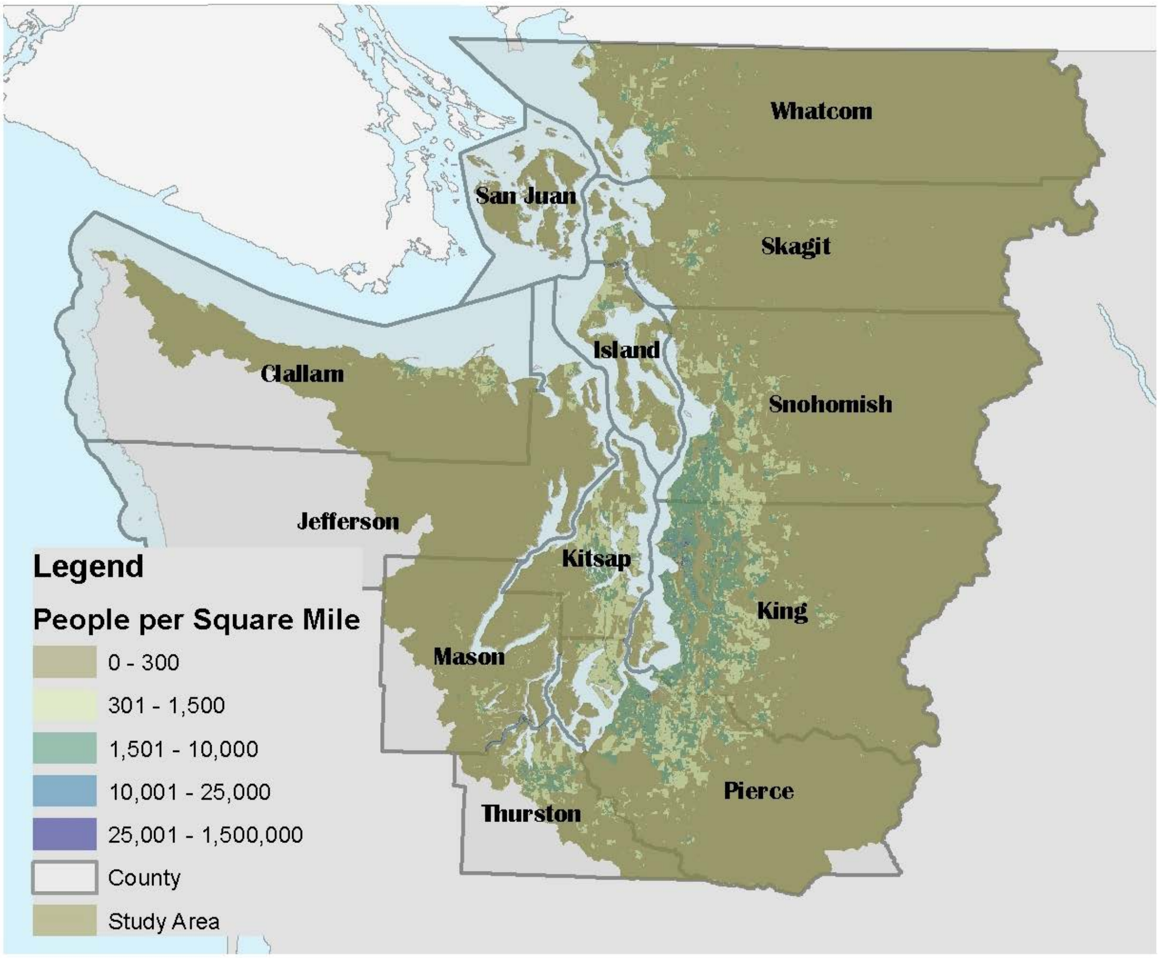


Figure 2. Map of Puget Sound Counties and Population Density within the Study Area.

Approach

All of the information used to calculate the quantity of COCs released from primary sources was obtained through peer-reviewed literature, government and non-governmental publications, government databases, direct communications with experts, marketing data, and other miscellaneous information resources. No sampling or other field data collection was conducted for this project.

Identification of the primary sources for most COCs was initially done through common knowledge or information from other efforts to characterize primary sources of COCs, such as the Oregon Department of Environmental Quality P3 Project (Oregon Insider, 2009). As the searches for primary source information proceeded, so too did the list of primary sources that were potentially worthy of investigation. In general, the project team focused on calculating release estimates from diffuse sources, beginning with the sources potentially releasing the largest amount of COCs.

Early on in the process of preparing this report, it was recognized that detailed and thorough analyses of each COC release from each source was not possible due to a lack of available information and the staff time allotted for this project. Therefore, most of the release estimates are rough estimates, and it was not possible to calculate releases for every primary source identified.

In general, release estimates were calculated by determining a release rate per unit or activity (e.g. brake pad or woodstove use) and multiplying by the number of units or amount of activity (e.g. vehicle kilometers travelled, number of woodstoves and amount of firewood consumed) in the Study Area. If no data could be found to derive a release estimate using this methodology, information on releases for a particular region (e.g. watershed, nation) was sought. These estimates were scaled to the Study Area based on an appropriate scaling factor (e.g. land area, population) to yield release estimates for the Study Area. In other cases, ready-made release estimates were available for the 12-county Puget Sound region, such as the data from the Toxics Release Inventory (TRI) and from the Air Emissions Inventory (Ecology, 2007a).

An important aspect of deriving COC release estimates was documentation of the assumptions used, limitations of the data, and uncertainty associated with each of the major variables. Each of the release estimates includes an evaluation of these components in order to provide (1) readers with a gauge of the confidence in the estimates, and (2) a guide for further refinement of the estimates where desired.

Most of the release estimates derived in this report represent either a median value, mean value, or mid-point of a data range. The associated term (e.g. mean, median, mid-point) was included with the value to describe what was represented by the estimate. In cases where the final estimated value was based on a combination of these terms (e.g. the sum of a mean value and a median value), or in cases where the most reasonable estimate was based on the judgment of the project team, the final estimate was labeled as the ‘best estimate’ and an explanation was provided.

In some cases, there was sufficient information to calculate a range of estimates for a particular COC source. These ranges were shown where the data availability and type of data permitted their calculation. In most cases, however, there were not enough data available to calculate an error term about the estimate. Instead, narrative descriptions were used to describe the uncertainty associated with the estimates.

Report Organization

The main body of this report is divided into sections for each COC. Within each COC section are sub-sections for each primary source. For each primary source, a release estimate for the Study Area is calculated; the methodology, variables, and assumptions are described; and a short discussion of the uncertainty is included. A summary of releases from each of the primary sources is included at the end of each section.

In certain instances, the methodology for deriving an estimate is relatively complex or otherwise requires a lengthy explanation. In these cases, the methodology is included in the appendices and is only summarized in the main text. Other supporting information is also included in the appendices and referenced in the main text.

A Summary and Conclusions chapter at the end of this report includes a compilation of the COC release estimates from each source category.

Release Estimates for Sources of Toxic Chemicals

Arsenic

Sources Assessed

Assessment of ongoing anthropogenic arsenic releases included the following sources:

- Roofing material runoff
- Fertilizer use
- Non-structural CCA-treated wood
- Industrial, commercial, and institutional facilities reporting under the TRI requirements
- Residential (non-wood) fuel use

Other possible sources of arsenic release include its use as animal feed additives (e.g. roxarsone) and herbicides (e.g. monosodium methyl arsenate [MSMA]), runoff from polyvinyl chloride (PVC) roofing material, and base metal smelting and refining operations. Releases from these sources, beyond those reported in the TRI, were not assessed.

Estimate of Arsenic Released from Roof Runoff

Data on arsenic in roof runoff are limited to leaching from asphalt shingles reported by Clark (2010). Clark (2010) is one of the few investigators of metals in roof materials who attempted to account for atmospherically deposited metals.

Following the study design described in Clark et al. (2008), runoff from test panels of roofing materials was compared to a Plexiglass 'blank.' The contaminant values from the Plexiglass results were then subtracted from the roof panel results. This resulted in negative runoff concentrations when the runoff from the Plexiglass panel was higher than that of the roofing material, suggesting that for that particular storm event, the roofing material may have stored some of the metals load. Contaminant release varied between and during storm events, with the initial rainfall, or first flush, often having the highest contaminant load. Arsenic concentrations over three runoff events ranged from -1.0 to 1.43 ug/l. The mean arsenic concentration was 0.26 ug/l.

The annual asphalt shingle rooftop runoff volume was used to translate arsenic concentrations in roof runoff into release loads for the Study Area (Table 1). The method used to calculate roof runoff volumes is described in Appendix B. The range of annual arsenic release for asphalt shingles was -0.59 to 0.84 metric tons (t)/yr, with a mean of 0.15 t/yr.

Table 1. Estimates of Arsenic Release from Asphalt Roofing Material.

Roof Type	Total Area of Roof Type (m^2)	Total Runoff Volume of Roof Type (l/yr)	Arsenic Concentrations in Runoff (ug/l)	Arsenic Release in Study Area (t/yr)
Asphalt Composite	3.96×10^8	5.84×10^{11}	-1.02 – 1.43 0.26 (mean)	-0.59 – 0.84 0.15 (mean)
			Total Estimate:	-0.59 – 0.84 0.15 (mean)

t: metric ton

Uncertainty

The greatest uncertainty in estimates of arsenic released from rooftops is due to lack of data on roof materials other than asphalt shingles, leaving approximately 27% of the total roof area in the Study Area unaccounted for. Furthermore, the assumption that all asphalt shingles contain arsenic, and that arsenic is present at similar concentrations among asphalt shingles, may not be correct; different commercial products may produce different results. Roofing age, unaccounted for here, may also play a factor, as release rates may vary over the life of a roof. Clark (2010) provides the only data on arsenic in runoff from roof materials.

Scale-up information for estimates of arsenic releases from roof materials appears to be fairly reliable. Accuracy may be increased by collecting information on roof type fractions from jurisdictions in addition to those discussed in Appendix B.

Estimate of Arsenic Released from Fertilizer Application

Estimates of arsenic released from fertilizers were based on the reported concentrations in fertilizers and amounts distributed in the state annually. Producers of lime and fertilizer products registered for distribution in Washington State report metals concentrations for each of their products to the Washington State Department of Agriculture (WSDA). WSDA also collects information on the amount of fertilizer and lime distributed within the state annually based on fertilizer grade, not specific product.

WSDA calculated a statewide estimate of metals loading from fertilizers and lime products for the 2006-2007 reporting year, applying representative metals concentrations to the tonnage amounts for each grade of material (Maxwell, 2008). To estimate the arsenic load in the Study Area, a factor of 0.0164 was applied to the statewide arsenic load for each material to reflect the portion of the total statewide crop area (4,387,169 acres; NASS, 2007) that falls within the Study Area (71,680 acres; Envirovision et al., 2008).

Total arsenic releases from fertilizers are shown in Table 2. In general, arsenic releases corresponded with the amount of each material distributed. The total arsenic release was estimated to be 0.063 t/yr.

Table 2. Estimates of Arsenic Release from Fertilizer.

Material	Material Distributed in Washington During 2006-2007 Reporting Year (t)	Arsenic Release in Washington (t/yr)	Arsenic Release in Study Area (t/yr)
Nitrogen Material	618,550	0.77	0.013
Phosphate Material	115,474	1.53	0.025
Potash Material	94,161	0.15	0.002
Miscellaneous Fertilizer	163,652	0.55	0.009
Sulfur	25,954	0.26	0.004
Gypsum	38,409	0.14	0.002
Natural Organics	32,866	0.13	0.002
Liming Material	55,579	0.32	0.005
Total as Fertilizer	**1,144,645**	**3.85**	**0.063**
		Total Estimate:	0.063

Uncertainty

The largest source of uncertainty may be the assumption that fertilizer application practices within the Study Area are identical to other regions of Washington State. An annual estimate of arsenic release also assumes that fertilizer use during the 2006-2007 reporting year is representative of current use. The validity of these assumptions has not been assessed.

An additional source of uncertainty may be due to the reporting conventions used by WSDA. Reported arsenic levels may over-estimate actual levels since WSDA used detection limits to substitute for non-detected results, possibly resulting in an upward bias of the reported concentrations.

Estimate of Arsenic Released from Non-Structural CCA-Treated Wood

Wood pressure-treated with the pesticide chromated copper arsenate (CCA) has traditionally been used widely in outdoor applications due to its fungicidal and insecticidal properties. Although after 2003 it was banned from most residential applications, its use remains wide-spread in industrial, commercial, and limited residential application where its use is still permitted. Due to its durability, CCA-treated wood remains in service years and decades after its initial installation.

Although arsenic leaching rates are available in the published literature, little information is available on the volume of CCA-treated wood in the Study Area. In the absence of information necessary to conduct a Puget Sound basin-wide scale-up based on rates of arsenic leaching from CCA-treated wood, information about copper leaching from the San Francisco Bay Clean Estuary Partnership (TDC, 2004) (corrected for a mathematical error) was combined with published measured relative leaching rates of arsenic and copper from CCA-treated wood (Townsend et al., 2005). A per capita release rate was calculated for the San Francisco Bay area then scaled up to the Study Area using census data for the 12-county Puget Sound region (4,475,300; U.S. Census Bureau, 2010). Based on these factors, the estimated arsenic releases from CCA-treated wood yields a Study Area-wide release estimate of 0.044-0.50 t/yr. The mid-point of this range is 0.27 t/yr (Table 3).

Table 3. Estimates of Arsenic Release from Non-Structural CCA-Treated Wood.

Source	Estimate of Total Copper Release in SF Bay Watershed (t/yr)	Per Capita Copper Release (g/person/yr)	Paired Ratios Of Arsenic-to-Copper Release Rates	Per Capita Arsenic Release (g/person/yr)	Arsenic Release in Study Area (t/yr)
Non-Structural CCA-Treated Wood	0.064 – 0.13 0.095 (mid-point)	0.0091 – 0.018 0.014 (mid-point)	1.08 – 6.2	0.0099 – 0.11 0.061 (mid-point)	0.044 – 0.50 0.27 (mid-point)
				Total Estimate:	0.044 – 0.50 0.27 (mid-point)

Uncertainty

Uncertainties associated with arsenic release estimates are primarily related to (1) the reliability and accuracy of assumptions about copper releases calculated in the San Francisco Bay area study, and (2) the assumptions that the Study Area and San Francisco Bay area are identical with regard to CCA-treated wood use and leaching and that the measured ratio of arsenic to copper that leaches from CCA-treated wood is the same as the actual ratio for CCA-treated wood in the Study Area. Since the literature shows a range of arsenic and copper leaching rates from CCA-treated wood, a preferable method for calculating releases in the Study Area is to obtain regional information on CCA wood use and local conditions related to leaching rates.

Estimate of Arsenic Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Industrial facilities reported total mean annual arsenic (and arsenic compounds) releases of 0.013 t/yr for the five TRI categories assessed (Table 4). There were no reported releases from military facilities. A single wood-treating facility, Superior Wood Treating in Pierce County, accounted for all of the arsenic releases, with most of the release to surface water. Reported annual arsenic release from this facility was generally on the order of 10 pounds or less, but the annual mean was elevated due to a high reported water release during 2001 (250 pounds).

Table 4. Estimates of Mean Annual Arsenic Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 1999-2008.

Release by Facility Type	Percent of Total	Arsenic Release in Study Area (t/yr)
Wood Treatment	100%	0.013
Total	100%	0.013
Release by Medium	**Percent of Total**	**Arsenic Release in Study Area (t/yr)**
Fugitive Air Emissions	5%	0.00066
Stack Air Emissions	4%	0.00053
Surface Water Discharge	91%	0.012
Other On-Site Land Disposal	0%	0.000
Discharge or Transfer to POTWs	0%	0.000
Total	100%	0.013
Total Estimate:		0.013

POTWs: Publicly-owned treatment works

Uncertainty

Criteria and limitations of TRI reporting requirements are in Appendix C.

Estimate of Arsenic Released from Modeled Air Emission Sources

Air emissions of arsenic inventoried during 2005 for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Point source air emissions account for the bulk of releases (0.28 t/yr), but the exact source was not identified in the Inventory Report (Table 5). Smaller releases of arsenic (0.01 t/yr) were due to combustion of distillate oil (heating oil) as a residential heating source. Total arsenic emissions were 0.29 t/yr.

Table 5. Estimates of Arsenic Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	Arsenic Release in Study Area (t/yr)
Point Sources - Industrial, Commercial, Institutional (primarily Title V AOP)	0.28
Residential Fuel Use, except Wood	0.01
Total Estimate:	0.29

AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

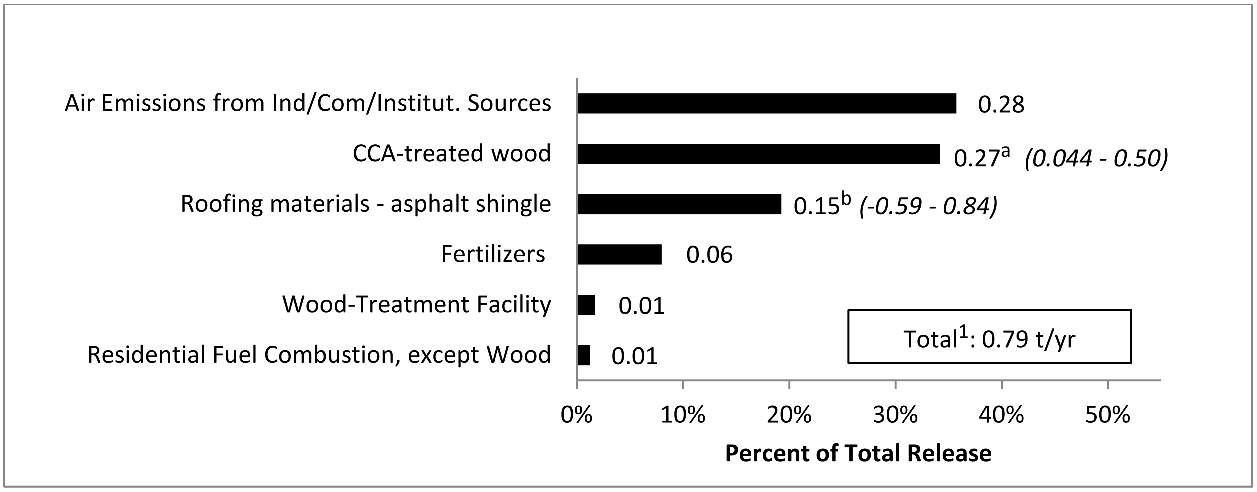
Uncertainty

Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of Arsenic Release Estimates

Based on the best estimates available, total arsenic releases from the sources assessed are approximately 0.8 t/yr (Figure 3). However, there is a large amount of uncertainty around estimates of arsenic released from roof runoff and CCA-treated wood, sources which potentially release the bulk of anthropogenic arsenic in the Study Area. For other sources – fertilizer application, a wood treatment facility that releases arsenic primarily to surface water, and other industrial/commercial/institutional facilities emitting arsenic to air – releases are comparably small or moderate, but there are no data to gauge the uncertainty around these estimates.

Large historical sources of arsenic in the region include the Asarco Smelter in Tacoma which emitted arsenic for decades and may have resulted in large swaths of the Puget Sound area with elevated arsenic (PTI, 1991; San Juan, 1994). Review of the literature suggests that use of arsenic-containing pesticides would likely represent the largest ongoing releases of anthropogenic arsenic in the Study Area if patterns of use are similar to other regions of the U.S. (e.g. Solo-Gabriele et al., 2003). However, golf course representatives meeting with the U.S. Department of Agriculture’s National Agricultural Statistics Service (NASS) in 2008 did not report any use of monosodium methyl arsenate (MSMA), and the expected usage of MSMA in the Study Area is therefore low (Kelly McLain, WSDA, written communication).



[1]Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a]Mid-point of range [b]Mean

Figure 3. Total Arsenic Release in the Study Area (values shown are t/yr).

Cadmium

Sources Assessed

Assessment of ongoing anthropogenic cadmium releases included the following sources:

- Roofing material runoff
- Fertilizer use
- Vehicle tire wear
- Vehicle brake pad wear
- Industrial, commercial, and institutional facilities reporting under the TRI requirements
- Residential fuel use, including wood
- Locomotive emissions

Other possible sources of cadmium release include use as anti-corrosives on vessels, base metal smelting and refining, use in paints, and plastics manufacturing. Releases from these sources beyond those reported in the TRI were not assessed.

Estimate of Cadmium Released from Roof Runoff

Cadmium concentrations have been reported in runoff from asphalt shingles, metal roofing, and built-up roofing. For the following estimates, the *built-up roof* category includes runoff concentrations from both rubber and general commercial roof areas. Built-up roofing is a general term that can be applied to many flat, membrane, or torch-down roof types.

Steuer et al. (1997) provide data on roof runoff in a small residential catchment where runoff is primarily from asphalt shingle roofs, although some runoff from the upper painted sides of buildings and galvanized or metal roofing components may have been included in their samples. The mean cadmium concentration in runoff from this study was 0.7 ug/l, higher than a cadmium concentration of 0.12 ug/l in roof runoff isolated to residential areas as reported by Davis et al. (2001).

Steuer et al. (1997) and Clark (2010) also reported a combined mean cadmium concentration of 1.41 ug/l in built-up roofing (individual means ranged from 0.9 – 1.91 ug/l). Concentrations from built-up roofing are similar to cadmium in runoff from generalized commercial areas as reported by Davis et al. (2001) and Steuer et al. (1997) (mean of 0.93 ug/l, range of 0.6 - 1.3 ug/l).

The relatively narrow range of cadmium concentrations in asphalt shingle and built-up roofing materials also appears to hold true for metal roof materials. Clark (2010) reported that runoff from aluminum and Galvalume roofing materials had a mean cadmium concentration of 0.76 ug/l, with individual means of 0.24 and 1.3 ug/l, respectively.

Annual rooftop runoff volumes for each roof type were used to translate cadmium concentrations in runoff into release loads. The method used to calculate runoff volumes is described in Appendix B. Total annual cadmium release for metal, asphalt shingle, and built-up roofs were estimated to be 0.03, 0.4, and 0.1 t/yr, respectively (Table 6). The total 'best estimate' load is 0.6 t/yr based on the sum of estimates for the cadmium release from asphalt shingles and mean releases from metal and built-up roofs.

Table 6. Estimates of Cadmium Release from Various Roofing Materials.

Roof Type	**Total Area of Roof Type (m^2)**	**Total Runoff Volume of Roof Type (l/yr)**	**Cadmium Concentration in Runoff (ug/l)**	**Cadmium Release in Study Area (t/yr)**
Metal	2.96×10^7	4.29×10^{10}	0.24 – 1.3 0.76 (mean)(a)	0.01 – 0.06 0.03 (mean.)
Asphalt Composite	3.96×10^8	5.84×10^{11}	0.7	0.4
Built-up	7.40×10^7	1.03×10^{11}	0.9 – 1.9 1.4 (mean)	0.09 – 0.20 0.1 (mean)
			Total Estimate:	0.5 – 0.7 0.6 (best estimate)

(a) Based on runoff concentrations from aluminum and Galvalume roof materials (Clark, 2010).
t: metric ton

Uncertainty

The greatest uncertainty in estimates of cadmium released from rooftops is most likely due to the scarcity of cadmium release data in the literature. As mentioned previously, atmospheric deposition was not accounted or controlled for in the runoff studies used to derive release estimates with the exception of Clark (2010), and could potentially result in an upward bias of results. Roofing age, unaccounted for here, may also play a factor, as release rates may vary over the life of a roof. In addition, runoff data were available for only three of eight roof types, leaving approximately 10% of the roof area unaccounted for.

Scale-up information for estimates of cadmium releases from roof materials appears to be fairly reliable. Accuracy may be increased by collecting information on roof type fractions from jurisdictions in addition to those jurisdictions discussed in Appendix B.

Estimate of Cadmium Released from Fertilizer Application

Estimates of cadmium released from fertilizers were based on the reported concentrations in fertilizers and amounts distributed in the state annually, then scaled to the Study Area based on estimated proportion of crop area in the Study Area (0.0164). Details are included in the section describing arsenic releases from fertilizers.

Total cadmium releases are shown in Table 7. Cadmium releases from phosphate fertilizer accounted for approximately four-fifths of total fertilizer release. The total cadmium release was estimated to be 0.26 t/yr.

Table 7. Estimates of Cadmium Release from Fertilizer.

Material	Material Distributed in Washington During 2006-2007 Reporting Year (t)	Cadmium Release in Washington (t/yr)	Cadmium Release in Study Area (t/yr)
Nitrogen Material	618,550	0.238	0.004
Phosphate Material	115,474	12.856	0.210
Potash Material	94,161	0.077	0.001
Miscellaneous Fertilizer	163,652	2.445	0.040
Sulfur	25,954	0.013	0.000
Gypsum	38,409	0.009	0.000
Natural Organics	32,866	0.031	0.001
Liming Material	55,579	0.047	0.001
Total as Fertilizer	**1,144,645**	**15.7**	**0.26**
		Total Estimate:	0.26

Uncertainty

The largest source of uncertainty may be the assumption that fertilizer application practices within the Study Area are identical to other regions of Washington State. An annual estimate of cadmium release also assumes that fertilizer use during the 2006-2007 reporting year is representative of current use. The validity of these assumptions has not been assessed.

An additional source of uncertainty may be due to the reporting conventions used by WSDA. Reported cadmium levels may over-estimate actual levels since WSDA used detection limits to substitute for non-detected results, possibly resulting in an upward bias of the reported concentrations.

Estimate of Cadmium Released from Vehicle Tire Wear

Releases of cadmium due to vehicle tire wear were calculated using estimates of tire wear rates and cadmium concentrations reported in tire material, then scaled to the Study Area using annual vehicle kilometers travelled (VKTs) for the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Table 8 shows a summary of the variables used to calculate cadmium release from tire wear. For each tire, a wear rate of 38 mg tire material per km travelled was assumed for all vehicle types considered, with differences being only the number of tires per vehicle. Although total annual tire wear is substantial (approximately 11,000 t/yr), cadmium concentrations in tire material are in the low parts per million range, yielding relatively low overall cadmium releases. Estimates of total cadmium release from tire wear is <0.001 to 0.06 t/yr, with an overall mean of 0.03 t/yr.

Table 8. Estimates of Cadmium Release from Vehicle Tire Wear.

Vehicle Type	Number of Tires per Vehicle	Tire Wear Rate (mg/km/tire)	Cadmium Concentration in Tire Material (mg/kg)	Vehicle Kilometers Travelled in Study Area (km/yr)	Cadmium Release in Study Area (t/yr)
Motorcycle	2	38	0 – 5 3 (mean)	2.3E+08	<0.001 (mean)
Passenger Car	4			3.6E+10	0 – 0.03 0.02 (mean)
Light Truck	4			1.5E+10	0 – 0.01 0.007 (mean)
Bus	8			1.2E+08	<0.001
Single Unit Truck	8			2.8E+09	0 – 0.004 0.002 (mean)
Combination Truck	18			3.5E+09	0 – 0.01 0.007 (mean)
				Total Estimate:	<0.001 – 0.06 0.03 (mean)

Uncertainty

Overall estimates of tire wear rates, cadmium concentrations in tire material, and VKTs appear to be reliable since the estimates are based on published information and WSDOT data (see Appendix E). Tire wear rates reported in the literature appear to be realistic when checked against what may be considered a reasonable tread loss over the life of a tire (see Appendix E for this analysis). However, both the tire wear rates and cadmium concentrations may vary considerably based on tire brand, with wear rates also strongly influenced by driving conditions. Although the VKTs are based on the 12-county Puget Sound region, they are a reasonable proxy for the Study Area.

Estimate of Cadmium Released from Vehicle Brake Pad Wear

Releases of cadmium due to vehicle brake pad wear were calculated using estimates of brake pad wear rates and cadmium concentrations reported in brake pad material, then scaled to the Study Area using annual VKTs in the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Wear rates for brake friction materials were estimated from various literature values and were found to range from 3 mg/km travelled for motorcycles to 245 mg/km travelled for combination trucks (Table 9). Different vehicle types employ different types of braking systems, with motorcycles, passenger cars, and light trucks primarily using disc brakes and heavier vehicles using drum brakes almost exclusively. There are few data on cadmium in drum brake friction materials, and they have not been investigated extensively since most of the abraded material is retained in the brake drums (Sinclair Rosselot, 2006).

Based on the differences in braking systems among vehicle types, mean cadmium concentrations were calculated only for motorcycles, passenger cars, and light trucks although maximum releases from other vehicle types are also calculated by assuming maximum cadmium concentrations also apply to drum material (Table 9). Mean cadmium releases due to brake pad wear is estimated to be 0.002 – 0.06 t/yr, with a best estimate of 0.01 t/yr.

Table 9. Estimates of Cadmium Release from Vehicle Brake Pad Wear.

Vehicle Type	**Brake Pad Wear Rate per Vehicle (mg/km)**	**Cadmium Concentration in Brake Pad Material (mg/kg)**	**Vehicle Kilometers Travelled in Study Area (km/yr)**	**Cadmium Release in Study Area (t/yr)**
Motorcycle	3	2.7 – 29.9 13 (mean)	2.3E+08	<0.001 - <0.001 <0.001 (mean)
Passenger Car	16		3.6E+10	0.002 – 0.017 0.008 (mean)
Light Truck	16		1.5E+10	<0.001 - 0.007 .003(max)
Bus	110	29.9 (max)	1.2E+08	<0.001 (max)
Single Unit Truck	129		2.8E+09	0.011 (max)
Combination Truck	245		3.5E+09	0.025 (max)
			Total Estimate:	0.002 – 0.06 0.01 (best estimate)

Uncertainty

Estimates of mean cadmium releases due to brake pad wear are highly uncertain due primarily to the difficulty obtaining reliable estimates of wear rates. Much of the wear rate uncertainty is due to a wide range of assumptions possible for assigning the proportion of disc brake use among various vehicle types. For instance, in the present analysis an inconsequential proportion of passenger cars and light trucks are assumed to use drum brakes. However, Sinclair Rosselot (2006) assumes that approximately one-third of these vehicles have rear drum brakes. If reliable estimates were available for the concentration of cadmium in drum lining materials, the proportion of cars with drum brakes could have been considered in the current analysis.

Cadmium concentrations in brake pads range by an order of magnitude, with little assessment of confidence about the mean concentration of 13 mg/kg. Preferable methods for calculating chemical releases from brake pad wear would consider automobile make/models independently and calculate releases based on the brake pads and VKTs associated specifically with each make/model. However, for each vehicle make/model there are typically a variety of both factory-installed or after-market pads available, which may contain different amounts of metals concentrations, suggesting that data on VKTs by each car make/model may not by itself improve the level of certainty in release estimates.

As noted in the analysis provided in Appendix E, other variables such as driving habits and types of roads travelled are also considerations in determining releases from brake pad wear, but a detailed evaluation of these variables are beyond the scope of the present report.

Estimate of Cadmium Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Industrial facilities reported total mean annual cadmium (and cadmium compound) releases of 0.0005 t/yr for the five TRI categories assessed (Table 10). The mean release was based on a single report of stack air release of 10 pounds from Spectrum Glass in Snohomish County during 2000.

Table 10. Estimates of Mean Annual Cadmium Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 1999-2008.

Release by Facility Type	**Percent of Total**	**Cadmium Release in Study Area (t/yr)**
Specialty Glass Manufacturer	100%	0.0005
Total	100%	0.0005
Release by Medium	**Percent of Total**	**Cadmium Release in Study Area (t/yr)**
Fugitive Air Emissions	100%	0.0005
Stack Air Emissions	0%	0
Surface Water Discharge	0%	0
Other On-Site Land Disposal	0%	0
Discharge or Transfer to POTWs	0%	0
Total	100%	0.0005
Total Estimate:		0.0005

POTWs: Publicly-owned treatment works

Uncertainty

Criteria and limitations of TRI reporting requirements are in Appendix C.

Estimate of Cadmium Released from Modeled Air Emission Sources

Air emissions of cadmium inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Point source air emissions account for the bulk of releases (0.042 t/yr), but the exact source(s) were not identified in the 2005 Air Emissions Inventory report (Table 11). Smaller releases of cadmium were due to combustion of distillate oil (heating oil) as a residential heating source, use of certified and non-certified woodstoves, and from locomotives. Total cadmium emissions were estimated to be 0.06 t/yr.

Table 11. Estimates of Cadmium Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	Cadmium Release in Study Area (t/yr)
Locomotive Emissions	0.004
Point Sources - Industrial, Commercial, Institutional (primarily Title V AOP)	0.042
Residential Fuel Use, except Wood	0.007
Woodstoves and Fireplaces	0.006
Total Estimate:	0.06

AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

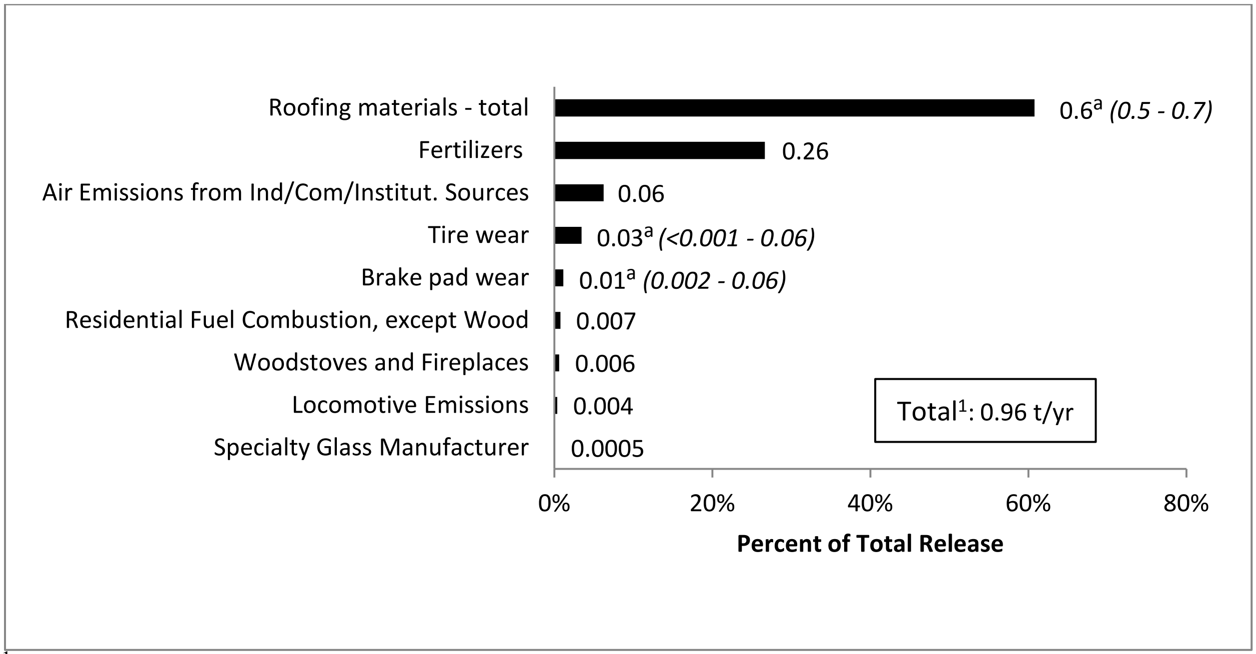
Uncertainty

Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of Cadmium Release Estimates

Based on the best estimates available, total cadmium releases from the sources assessed are approximately 0.96 t/yr (Figure 4). Leaching from roofing materials accounts for two-thirds of the cadmium release. However, there are few data to gauge the uncertainty of cadmium release from asphalt composite shingles, the roof type releasing the largest cadmium loads in the Study Area. There are also no ranges available for cadmium in fertilizers, the second largest release assessed for the Study Area. The resulting ranges around total cadmium release therefore reflect a lack of data rather than a high degree of certainty around the estimates.

Aside from roof runoff and fertilizer applications, there appear to be only small releases of anthropogenic cadmium in the Study Area. The magnitude of releases from base metal smelting and refining and use in paints and plastics manufacturing in the Study Area are not known, but no releases were reported under TRI for these types of facilities. No information was sought on the use of cadmium as anti-corrosives on vessels.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mean

Figure 4. Total Cadmium Release in the Study Area (values shown are t/yr).

Copper

Sources Assessed

Assessment of ongoing copper releases was limited to the following sources:

- Urban lawn and garden use of pesticides
- Plumbing fixtures, pipe, and solder leaching
- Vehicle brake pad wear
- Roofing material runoff
- Vessel anti-fouling paint
- Agricultural use of pesticides
- Fertilizer and micronutrient use
- Vehicle tire wear
- Aquatic-use algaecides in pools, fountains, spas, etc
- CCA-treated wood
- Military, industrial, commercial, and institutional facilities

Other possible sources of copper release include paints and base metal smelting and refining. Releases from these sources, beyond those reported in the TRI, were not assessed due to time limitations.

Estimate of Copper Released from Indoor Residential Plumbing

Copper may be released from residential plumbing through the use of copper piping and brass fittings which generally contain > 50% copper. Water supply lines may contain brass fittings as well (copper piping is not typically used in distribution systems) and may also be responsible for copper releases.

The release of copper and other metals from plumbing components is largely dependent on water quality variables such as pH and hardness. In 1991, the U.S. Environmental Protection Agency (EPA) published the Lead and Copper Rule (LCR; 40 CFR Part 141) requiring water suppliers to take actions to reduce corrosion, which is largely responsible for the release of copper, lead, and other metals (EPA, 2004). The LCR also requires monitoring for copper and lead at customers' taps, and sets action levels for copper and lead at the tap. The action level for copper is 1.3 mg/l in 10% of taps sampled; water suppliers must take action to control corrosion when this concentration is exceeded.

Results of water sampling from taps within the 12-county Puget Sound region during 2010 were obtained from the Department of Health Drinking Water Database (WDOH, 2011). The median copper concentration was 90 ug/l (n=4,220), within the range of annual median values obtained from a similar dataset in California (84 – 268 ug/l; Kimbrough, 2009). Copper concentrations at the 25th and 75th percentiles for the Puget Sound counties were 20 and 310 ug/l, respectively.

Domestic water consumption rates for households summarized in Appendix F were used to estimate per capita copper loads in the Study Area. Water consumption rates were based on a national survey of domestic water consumption (Mayer et al., 1999). Mean per capita water consumption rates (gal/day) were reported for the following domestic uses: toilet - 18.5; clothes washer - 15.0; shower - 11.6; faucet - 10.9; baths - 1.2; dishwasher - 1.0; and leakage and other uses - 11.1. Total per capita indoor consumption was 69.3 gal/day.

The per capita indoor domestic water consumption was scaled up to the Study Area by population (4,475,300). The consumption per household was then calculated (2.53 residents per household; U.S. Census Bureau, 2010) for a total of 1,768,893 households consuming 310 million gal/day. An estimated 500,000 households in the Study Area use on-site (septic) systems (Ecology, 2007b). Thus, the total indoor water consumption for the Study Area discharged to POTWs is estimated to be 81 billion gal/yr (307 billion liter/yr), and the indoor consumption discharged to septics is estimated to be 32 billion gal/yr (121 billion l/yr).

Applying the copper concentrations obtained from WDOH (2011) and per capita water consumption rates reported by Mayer et al. (1999), indoor plumbing releases an estimated 39 t/yr of copper, with a range of 8.6 to 130 t/yr (Table 12).

Table 12. Estimates of Copper Release from Indoor Residential Plumbing.

Per Capita Indoor Water Consumption Rate (gal/day)	Receiving System Type	Indoor Water Consumption in Study Area (gal/yr)	Indoor Water Consumption in Study Area (l/yr)	Copper Concentration in Tap Water (ug/l)	Copper Release in Study Area (t/yr)
69.3	POTWs	8.12 x 10^{10}	3.07 x 10^{11}	20 – 310[a] 90 (median)	6.1 – 95[a] 28 (median)
	Septic	3.20 x 10^{10}	1.21 x 10^{11}		2.4 - 38[a] 11 (median)
				Total Estimate:	8.6 – 130[a] 39 (median)

[a] 25th - 75th percentile
t: metric ton
POTWs: Publicly-owned treatment works

Uncertainty

Releases of copper from residential plumbing may vary depending on the chemistry of the domestic source water and treatment by the water utility prior to distribution. To assess the copper contribution from source water, WDOH tap water results for 2010 were compared to results of paired source water data. For the 135 pairs of results available, median copper concentrations in the source water (10 ug/l) were much lower than the median concentration of the paired tap water (50 ug/l), with a median of the differences of 41 ug/l. This indicates that plumbing within the distribution system and the buildings contributes a significant amount of the copper found at the tap.

It is notable that LCR compliance data potentially represents the highest concentrations of copper within a given water service provider, since the rule requires monitoring of homes and buildings that are at the highest risk of copper and lead contamination (EPA, 2004). Therefore, the copper concentrations used for release estimates may be biased high compared to the overall population of tap water.

Water used outdoors may also carry contaminants leached from plumbing into the environment, and outdoor water usage represents 58% of the total residential water budget (Mayer et al., 1999). However, concentrations of copper and other metals may be different than concentrations at indoor taps. Many outdoor faucets are plumbed directly from the distribution system, with no residence time in the indoor system and less exposure to components which may contribute metals. As no information specific to outdoor plumbing was found, release estimates presented here do not include water used outdoors.

Scale-up water consumption data are based on a national survey conducted in a variety of geographical areas. There is no obvious reason they would not be representative of the water consumption habits for the Study Area. However, since the survey data are over a decade old, they may over-estimate water consumption rates which may have fallen in recent years due to water conservation education, installment of low-flow fixtures, and use of water-saving appliances such as dishwashers and clothes washers.

Estimate of Copper Released from Vehicle Brake Pad Wear

Releases of copper due to vehicle brake pad wear were calculated using estimates of brake pad wear rates and copper concentrations reported in brake friction material, then scaled to the Study Area using annual VKTs in the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Wear rates for brake friction materials were estimated from various literature values and were found to range from 3 mg/km travelled for motorcycles to 245 mg/km travelled for combination trucks (Table 13). Different vehicle types employ different types of braking systems, with motorcycles, passenger cars, and light trucks primarily using disc brakes and heavier vehicles using drum brakes almost exclusively. Mean copper concentrations in drum friction material were found to be much lower than in disc brake pads (2,179 mg/kg and 49,552 mg/kg, respectively).

Copper releases associated with brake pad wear were calculated by applying concentrations for each friction material to the wear rates associated with that material. The mean copper concentrations for disc brake pads were calculated for motorcycles, passenger cars, and light trucks only; the mean copper concentration for drum material was applied to all other vehicle types (Table 13). The mean copper release due to brake wear is estimated to be 37 t/yr.

Table 13. Estimates of Copper Release from Vehicle Brake Pad Wear.

Vehicle Type	Brake Pad Wear Rate Per Vehicle (mg/km)	Copper Concentration in Brake Pad Material (mg/kg)	Vehicle Kilometers Travelled in Study Area (km/yr)	Copper Release in Study Area (t/yr)
Motorcycle	3	49,552	2.3E+08	0.03
Passenger Car	16		3.6E+10	24
Light Truck	16		1.5E+10	10
Bus	110	2,179	1.2E+08	0.028
Single Unit Truck	129		2.8E+09	0.78
Combination Truck	245		3.5E+09	1.8
			Total Estimate:	37

Uncertainty

Estimates of the mean annual copper release due to brake pad wear is highly uncertain due primarily to difficulty obtaining reliable estimates of wear rates. Much of the wear rate uncertainty is due to the wide range of assumptions that are possible for assigning the proportion of disc brake use among various vehicle types. For instance, in the present analysis an inconsequential proportion of passenger cars and light trucks are assumed to use drum brakes. If reliable estimates were available for the concentration of copper in drum lining materials, the proportion of cars with drum brakes could have been considered in the current analysis.

Preferable methods for calculating chemical releases from brake pad wear would consider automobile make/models independently and calculate releases based on the brake pads and

VKTs associated specifically with each make/model. However, for each vehicle make/model there are typically a variety of both factory-installed and after-market pads available which likely contain different amounts of metals concentrations. This suggests that data on VKTs by each car make/model may not by itself improve the level of certainty in release estimates. Sinclair Rosselot (2006) reported that copper concentrations in brake friction material may range from near 0% to 20% (200,000 mg/kg). As noted in the analysis provided in Appendix E, other variables such as driving habits and types of roads travelled are also considerations in determining releases from brake pad wear, but a detailed evaluation of these variables is beyond the scope of the present report.

Estimate of Copper Released from Roof Runoff

Copper may be used as a constituent in roofing materials as an integral or non-integral (i.e. by-product) chemical, incorporated for its algaecidal properties or as the primary structural element as in solid copper roofs. As with other metals, leaching rates of copper may depend on a number of factors such as age of the roof, pH, storm intensity, and incline angle (Wallinder et al., 2004).

Literature data are available for copper runoff from metal roofs, including copper roofs, and for generalized roofing materials in residential and commercial areas. Pennington and Webster-Brown (2008) reported copper concentrations in runoff from a variety of copper roofs, with values ranging from 684 – 2,870 ug/l copper, with a mean of 1,530 ug/l. Boulanger and Nikolaodis (2003) reported a similar range of concentrations in steady-state runoff from both a roof installed in 1931 (1,040 ug/l) and one installed in 1993 (2,660 ug/l). As expected, copper concentrations in metal roofing material, specifically aluminum and galvanized roofing materials, were typically lower (mean = 355 ug/l) (Good, 1993; Tobiason, 2004).

Data on copper runoff associated with asphalt shingle roofs are scarce, even though asphalt shingles represent the most common type of material in residential areas in the western U.S. (Dodson, 2007). Steuer et al. (1997) provide data on roof runoff in a small residential catchment where roof runoff is primarily asphalt shingle, although some runoff from the upper painted sides of buildings and galvanized or metal roofing components may have been included in their samples. Mean copper concentrations of runoff reported in Steuer et al. (1997) were 10 ug/l copper, similar to runoff in other studies isolated to residential areas (Bannerman et al., 1993; Davis et al., 2001), suggesting that copper concentrations around 10 ug/l are typical for residential rooftops. Steuer et al. (1997) also reported copper concentrations averaging 23 ug/l in commercial roofing typically composed of flat rubberized or tar-sealed roofs.

The studies used for copper runoff concentrations were not conducted in controlled settings, and samples collected from rooftop runoff likely contained atmospherically deposited chemicals as well as chemicals leached from the roofing materials. Very few studies have accounted for the effects of contribution of wet or dry deposition, and none of those studies appear to have relevant copper data. In studies where attempts are made to correct for metals in atmospheric deposition (e.g. Clark, 2010), values at the low end are often negative; rooftops would seem to have a metals-capturing effect. While this is unlikely to be the case, especially where sampling and analytical error may account for the apparent metals loss, users of the data should be aware that deposition likely contributes at least some of the metals measured in roof runoff.

Annual rooftop runoff volumes for each roof type were used to translate copper concentrations in runoff into release loads. The method for the calculation of runoff volumes is described in Appendix B. Total annual releases for copper, metal, asphalt shingle, and built-up roofs were 4.1, 15, 5.8, and 2.4 t/yr, respectively (Table 14). The overall 'best estimate' release (27 t/yr) was based on the sum of estimates for asphalt composite and built-up roof types and the mean estimates from copper and other metal roof types.

Table 14. Estimates of Copper Release from Roofing Material.

Roof Type	**Total Area of Roof Type in Study Area (m^2)**	**Total Runoff Volume of Roof Type in Study Area (l/yr)**	**Copper Concentrations in Runoff (ug/l)**	**Copper Release in Study Area (t/yr)**
Copper	1.87×10^6	2.41×10^9	1,530 – 1,850 1690 (mean)	3.7 - 4.4 4.1 (mean)
Metal	2.96×10^7	4.29×10^{10}	4 - 705 355 (mean) (a)	0.17 - 30 15 (mean)
Asphalt Composite	3.96×10^8	5.84×10^{11}	10	5.8
Built-up	7.40×10^7	1.03×10^{11}	23	2.4
			Total Estimate:	12 – 43 27 (best estimate)

(a) Based on runoff concentrations from aluminum painted metal and galvanized roof materials (Good, 1993 and Tobiason, 2004).

Uncertainty

The greatest uncertainty in estimates of copper released from rooftops is most likely due to representativeness of the release data obtained from the literature. As mentioned previously, atmospheric deposition was not accounted or controlled for in the runoff studies used to derive release estimates, and could potentially result in an upward bias of results. In addition, runoff data were available for only four of eight roof types, leaving approximately 10% of the total roof area unaccounted for.

No release estimate was calculated specifically for algae-resistant asphalt shingle roofs containing copper-coated ceramic granules. Barron (2000) reported that algae-resistant asphalt shingle roofs with copper-coated ceramic granules had a mean copper content twice that of ordinary composition shingles (24.5 and 12.7 mg/kg, respectively) and 14 times the copper concentration in runoff (13 versus 0.9 $ug/cm^2/yr$). These copper-granulated roof materials are increasing in popularity, particularly in wet climates (Barron, 2000), yet estimates of copper releases for the Study Area could not be calculated due to the lack of information about the proportion of roofs containing these shingles. Roofing age, unaccounted for here, may also play a factor in copper release rates, as these rates may vary over the life of a roof.

Scale-up information for estimates of copper releases from roof materials appears to be fairly reliable. Accuracy may be increased by collecting information on roof type fractions from jurisdictions in addition to those discussed in Appendix B.

Estimate of Copper Released from Vessel Anti-Fouling Paint – Recreational Vessels

Copper is a common biocide for vessels moored in marine waters for extended periods and is also the primary ingredient in most anti-fouling paints applied to vessel bottoms. The total amount of copper released annually from recreational vessel anti-fouling paint was calculated by: (1) estimating the number of vessels moored in the Study Area; (2) calculating the wetted surface areas (WSAs) of the moored fleet; and (3) applying unit-area copper leaching rates found in the literature. An extended discussion of the scenarios and methods used to calculate vessel numbers and WSAs is in Appendix G.

The number of moored vessels was estimated under three scenarios based on Washington State Department of Licensing (DOL) and marina inventory data (BST, 2001 and 2010a). Estimates ranged from 27,727 to 46,232 vessels for the 12-county Puget Sound area.

Three methods were used to calculate the total recreational vessel WSA for the Study Area. Nine possible results were produced from the combination of each method applied to each vessel number scenario. Estimates of WSA ranged from 6.44×10^9 to 1.32×10^{10} cm^2.

Rates on passive copper leaching from anti-fouling paints were found to range from ≤ 3 to 8.2 $ug/cm^2/d$ (Schiff et al., 2003; Valkirs et al., 2003; Dobalian and Arias, 2005). Leaching rates were available for epoxy, ablative, and vinyl paint; there were no data for sloughing copper-based paints. Rates were based largely on *in-situ* leaching of copper from fiberglass panels and appeared to be appropriate for estimating releases in the Puget Sound aquatic environment. Copper leaching rates used to calculate releases are shown in Appendix G, Table G-1.

Total annual copper releases from recreational vessels were calculated from all possible combinations of vessel estimates (three), WSA computation methods (three), and leaching rates (six) for a total of 54 values. Copper release estimates ranged from 7.1 to 39 t/yr, with a mean of 18 t/yr and a median value of 16 t/yr. Differences in results were driven mostly by the leaching rates which varied by nearly a factor of 3. A summary of input parameters used to calculate annual copper loads leached from anti-fouling paint of recreational vessels are shown in Table 15.

Table 15. Estimates of Copper Release from Anti-Fouling Paint of Recreational Vessels.

Number of Recreational Vessels Moored in Study Area	**Total WSA of Recreational Vessels Moored in Study Area (cm^2)**	**Leaching Rates ($ug\ Cu/cm^2/day$)**	**Copper Release in Study Area (t/yr)**
27,737 – 46,232	$6.44 \times 10^9 - 1.32 \times 10^{10}$	≤3 – 8.2	7.1 – 39 18 (mean) 16 (median)
		Total Estimate:	7.1 – 39 18 (mean) 16 (median)

WSA: wetted surface area

Uncertainty

Perhaps the greatest source of uncertainty for these estimates is due to uncertainty associated with the number of recreational vessels moored year-round in the Study Area, although the number of permanent slips and estimates of their usage compare well with the approximate number of licensed pleasure craft over 25 feet in length (a length for which trailering may be less likely). A thorough survey of recent slip occupancy and empirical data on vessel length distribution would be required to develop accurate estimates of recreational vessel numbers and calculations of Study Area WSA.

A number of factors affect the actual release rate of copper from anti-fouling paint. These include the type of paint, the age of the paint coating, the degree of biofilm covering the paint, the chemistry of the water, the amount of mechanical cleaning, and the amount of movement that a boat undergoes. While in-water hull cleaning can cause a spike in copper concentrations, it may be short-lived (Valkirs et al., 1994). Other studies have found that the ratio of overall release from passive leaching to hull cleaning is 20:1 (Dobalian and Arias, 2005).

Due to the number of factors potentially affecting leaching rates, it is also uncertain if the values provided in the literature reflect conditions in the Study Area. However, a thorough study of *in-situ* leaching rates for a variety of conditions with a variety of paint products would be necessary to determine values with a high level of confidence.

Estimate of Copper Released from Vessel Anti-Fouling Paint – Commercial Vessels

Annual release of copper from commercial vessels was calculated using the same method as for recreational vessels. Based on Washington State Department of Revenue data, there were an average of 2,003 commercial vessels registered in the 12-county Puget Sound area during 2004 and 2005 (BST, 2010b), an estimate consistent with a recent search of Department of Revenue (DOR) records (DOR, 2010).

Total WSA values were calculated for the commercial fleet using a single formula for deriving the WSA for each vessel based on length (Appendix G). Estimates were from 3.27×10^9 to 3.29×10^9 cm^2, depending on estimates of the number of boats considered trailerable. Overall, the number of trailerable boats registered as commercial vessels has little impact on the total WSA for the entire commercial fleet.

Leaching rates used to calculate copper releases from commercial vessels were the same as used for recreational vessels (Appendix G, Table G-1). Total annual copper releases from commercial vessels were calculated from the two WSA estimates and the six leaching rate values mentioned previously, for a total of 12 release estimates. Copper release estimates ranged from 3.6 to 9.8 t/yr, with a mean of 5.9 t/yr and a median value of 4.8 t/yr. Differences in results were driven mostly by the leaching rates which varied by nearly a factor of 3. A summary of input parameters used to calculate annual copper loads leached from anti-fouling paint of recreational vessels is shown in Table 16.

Table 16. Estimates of Copper Release from Anti-Fouling Paint of Commercial Vessels.

Number of Commercial Vessels Moored in Study Area	**Total WSA of Commercial Vessels Moored in Study Area (cm^2)**	**Leaching Rates (ug Cu/cm^2/day)**	**Copper Release in Study Area (t/yr)**
1,878 – 2,003	$3.27 \times 10^9 – 3.29 \times 10^9$	$\leq 3 – 8.2$	3.6 – 9.8 5.9 (mean) 4.8 (median)
		Total Estimate:	3.6 – 9.8 5.9 (mean) 4.8 (median)

WSA: wetted surface area

Uncertainty

For commercial vessels, the greatest source of uncertainty may be in the accuracy of leaching rate values. A number of factors affect the actual release rate of copper from anti-fouling paint. These include the type of paint, the age of the paint coating, the degree of biofilm covering the paint, the chemistry of the water, the amount of mechanical cleaning, and the amount of movement that a boat undergoes. While in-water hull cleaning can cause a spike in concentrations, it may be short-lived (Valkirs et al., 1994). Other studies have found that the ratio of overall release from passive leaching to hull cleaning is 20:1 (Dobalian and Arias, 2005).

Estimates of total annual copper releases from commercial vessels may also be complicated by uncertainty about days per year spent in the Study Area. Fishing vessels, cargo vessels, sea going tugs and barges, as well as other classes of commercial vessels likely spend at least part of the year in waters outside of the Study Area, but the extent is unknown. The release estimates calculated for commercial vessels are based on the assumption that all non-trailerable commercial vessels are lying in slips and remain in Puget Sound a mean of 365.25 days per year. This may result in an upward bias of estimates of annual copper release, the accuracy of leaching rates notwithstanding.

Due to the number of factors potentially affecting leaching rates, it is also uncertain if the values provided in the literature reflect conditions in the Study Area. However, a thorough study of *in-situ* leaching rates for a variety of conditions with a variety of paint products would be necessary to determine values with a high level of confidence.

It appears that there is a high degree of certainty about the number and size of commercial vessels in the Study Area. The resulting WSA estimate for commercial vessels should also be reasonably accurate, although the degree of confidence would greatly benefit from empirical verification of the formula used to estimate WSA.

Estimate of Copper Released from Vessel Anti-Fouling Paint – Naval Vessels

Loads of copper leached from naval vessel hulls were estimated for three Navy facilities in Puget Sound: The Puget Sound Naval Shipyard (PSNS) at Bremerton, the Naval Station at Everett (NAVSTA Everett), and the Trident Submarine Base at Bangor (Bangor). Estimates of copper loads from PSNS were available in the published literature, while loads from NAVSTA Everett and Bangor were derived from a variety of information sources. Assumptions about vessel numbers and methods for WSA calculations are in Appendix G. Input parameters used to calculate annual copper loads leached from anti-fouling paint of naval vessels are shown in Table 17.

Estimates of copper leached from naval vessel hulls at PSNS were obtained from Johnson and Grovhoug (1999) and Brandenberger et al. (2008). Johnson and Grovhoug (1999) estimated a copper load of 3.4 t/yr using a comparatively high leaching rate of 17 ug/cm^2/day for active duty vessels and 1 ug/cm^2/day for inactive vessels. Brandenberger et al. (2008) estimated a much lower copper load (0.88 t/yr) due primarily to assumptions of an overall lower leach rate for calculating the load (3.9 ug/cm^2/day for active and inactive vessels) and fewer vessels in port. Both of these studies incorporated in their load estimates assumptions about the typical amount of time per year the ships spent in port or Puget Sound.

No published values of copper released from vessels at NAVSTA Everett or at Bangor were available. Estimates were therefore based on a number of assumptions and information available about the vessels that comprise the respective fleets at each port. The identities of the ships homeported in the NAVSTA Everett and Bangor fleets were found on Wikipedia (2010a; http://en.wikipedia.org/wiki/Naval_Station_Everett and 2010b; http://en.wikipedia.org/wiki/Naval_Base_Kitsap#Bangor, respectively), and verified with the Naval Vessel Registry (2010, www.nvr.navy.mil/). Specifications for each vessel (NAVSTA Everett-one aircraft carrier, two destroyers, and three frigates; Bangor-eleven submarines) were found in the Naval Vessel Registry.

The estimated copper load from the NAVSTA Everett vessels ranged from 0.13 to 0.56 t/yr, depending on the leaching rates used (3.9 or 17 ug/cm^2/day). The copper load from Bangor was estimated to range from 0.24 to 1.0 t/yr based on the same leach rates. Ships were assumed to be in port or Puget Sound 120 days per year.

Based on the sum of total copper loads from all three ports, the total copper load leached from hulls of naval vessels stationed in Puget Sound was estimated to be 1.2 to 5.0 t/yr, with a mid-point value of 3.1 t/yr (Table 17).

Table 17. Estimates of Copper Release from Anti-Fouling Paint of Naval Vessels.

Location	**Number of Naval Vessels Moored in Study Area**	**Assumed Time in Port (day/yr)**	**Total WSA of Naval Vessels Moored in Study Area (cm^2)**	**Leaching Rates (ug Cu/cm^2/day)**	**Copper Release in Study Area (t/yr)**
Bremerton PSNS (a)	48 active 18 inactive	NA	NA	17 (active) 1 (inactive)	3.4
Bremerton PSNS (b)	39	NA	NA	3.9	0.88
Everett Naval Station	6	120	2.73×10^8	3.9 – 17	0.13 – 0.56
Bangor Sub Base	11		5.06×10^8		0.24 – 1.0
				Total Estimate:	1.2 – 5.0 3.1 (mid-point)

WSA: wetted surface area.
(a) Johnson and Grovhoug (1999). (b) Brandenberger et al. (2008)

Uncertainty

Several variables introduce uncertainty into these estimates. While the number and types of vessels associated with each port is fairly well documented, information about the average number of days each year Navy vessels were docked at port or within Puget Sound was not readily available from the Navy or elsewhere. Based on the discussion in Brandenberger et al. (2008), the time in port or Puget Sound for Navy ships stationed at Everett and Bangor was assumed to be 120 days per year.

Other factors mentioned previously in regards to copper leaching from pleasure craft (type and age of paint, hull biofilm, water chemistry, mechanical cleaning, vessel movement) were also not considered due to the amount and complexity of information required. However, given the more than four-fold difference in leaching rates used to calculate the load estimates, it seems likely that the actual load from Navy vessels falls within the range of estimated values.

Estimate of Copper Released from Urban Lawn and Garden Pesticide Use

Home owners, as well as professional landscapers and pesticide applicators, use copper-containing lawn and garden products. Copper hydroxide is used widely as a fungicide in urban applications; copper formulations such as copper sulfate, copper ethanolamine, and metallic copper may also be used although in lesser quantities (TDC, 2004; ODA, 2009). Like agricultural applications of copper (mentioned in subsequent sections), residential use of copper is potentially significant but difficult to quantify due to the lack of local use data.

Two sources of information were used to gauge the rate of copper pesticide use for urban applications in the Study Area: estimates based on reported data from California Department of Pesticide Regulation (CDPR) and from Oregon Department of Agriculture (ODA). CDPR requires reporting for most agricultural and commercial pesticide applications, except for home-and-garden use and most industrial and institutional uses (CDPR, 2011). Record-keeping is required at the ‘point of first sale’ for all pesticides, but it is unclear the extent to which this captures retail sales to homeowners.

ODA collected non-household pesticide use data for several years and produced reports for reporting years 2006 through 2008 (ODA, 2011). Attempts were made to assess household pesticide use through homeowner surveys, but these attempts were met with limited success (ODA, 2009).

As part of a large effort to assess copper sources to San Francisco Bay, TDC (2004) used CDPR data to estimate the amount of copper used for urban landscaping use in the Bay watershed. TDC (2004) estimated 113 t/yr copper was used for urban landscaping, including estimates of unreported use based on sales data, with copper hydroxide accounting for 70% of the use. The San Francisco Bay watershed estimate is equivalent to a per capita rate of 16.2 g/person/yr based on the Bay area population estimate (6,994,500) at the time of the estimate (TDC, 2004).

Estimates of copper pesticide use reported in Oregon were obtained from a breakdown of the information contained in the 2008 Pesticide Use Reporting System Annual Report (ODA, 2009; Sunny Jones, ODA, written communication 2011). According to the ODA, 0.9 t copper as copper hydroxide was used in Oregon outdoor urban areas statewide during 2008; copper use from other formulations was minimal. Based on the 2010 Oregon population of 3,831,074 (U.S. Census Bureau, 2011), the per capita copper pesticide use in urban areas is 0.24 g/person/yr. However, this estimate does not include unreported uses by homeowners.

A range of estimates of urban pesticide copper use for the Study Area is based on the per capita rates derived from the Oregon and California estimates described previously. Applying these per capita rates to the Puget Sound population of 4,475,300 translates to an estimated release of 1.1 to 73 t/yr in the Study Area (Table 18).

Table 18. Estimates of Copper Release from Urban Lawn and Garden Pesticide Use.

Basis for Estimate	Total Release (t/yr)	Per Capita Release (g/person/yr)	Copper Release in Study Area (t/yr)
Oregon (statewide)	0.9	0.24	1.1
San Francisco Bay watershed	113	16.2	73
		Total Estimate:	1.1 – 73

Uncertainty

There are many uncertainties associated with the estimates for copper released as an urban pesticide. Since it is not feasible to derive a ‘best estimate’ based on the available data, a range of estimates was derived. However, without information specific to the Study Area, it is not clear if the actual rate of copper release from urban pesticide use falls within this range.

The available data on pesticide usages from California and Oregon are not sufficient to assess which rate, if either, may be applicable to the Study Area. The per capita rate derived from the Oregon data may represent an under-estimate due the lack of estimated copper release by homeowners, and because the data are from a largely rural state. The estimates for the San Francisco Bay watershed would appear to be a reasonable basis for a Study Area estimate since both areas are basins containing large metropolitan areas of similar geographical size and population. However, the Bay area estimate was derived from data collected statewide in California, and may not be representative of actual use patterns within the San Francisco Bay watershed.

Manufacturer sales and distribution data for copper-containing products for urban use would be the best source of information to refine release estimates, but these data are proprietary and not freely available. A well-designed survey with a few willing retailers or purchased marketing data would likely provide enough information to greatly increase the confidence in release estimates from this source of copper.

Estimate of Copper Released from Agricultural Pesticide Use

Copper products are used widely in agricultural applications due to copper's herbicidal and fungicidal properties. As with all agricultural pesticides in Washington, the lack of reporting requirements limits the feasibility of obtaining accurate use data. However, several sources of information were used to piece together estimates of copper use as agricultural pesticide in the Study Area.

Actual reported acreages of crops on which copper was applied statewide were provided by the WSDA, the National Agricultural Statistics Service, and the Washington Agricultural Statistics Service (WSDA, 2010a and b; NASS, 2010; WASS, 2004). These agencies also reported the seasonal copper application rates, percentages of acres treated, and the acreages in the Study Area for the following crops; apples, pears, potatoes, onions, berries (raspberries, blackberries, boysenberries, and marionberries), and "caneberries" (includes some orchard/vineyard and lavender crops). The resulting estimate of copper usage on these crops in the Study Area is shown in Table 19 as "Crops with statewide copper use data." The estimated annual copper released during treatment of these crops is 2 t/yr.

In addition to the 22,740 acres of crops in the Study Area for which copper application rates are known (at least on a statewide basis), there are an additional 61,135 acres of crops for which copper is approved (WSDA, 2010a), but there is no information on actual application rates. Based on labels from several manufacturers, copper application rates for the variety of crops grown in Washington are 0.1 – 22.7 kg active ingredient (a.i.)/acre (Bionide, 2009; Loveland, 2006; W. Neudorff, 2009; Syngenta, 2007).

When the range of application rates was multiplied by 100% of acreage for all crops in the Study Area for which copper may be used, the total amount of copper applied for agricultural purposes ranged from 10 to 1,900 t/yr. However, it appears that the low end of this range is a more reasonable estimate based on the amount of copper applied to acreages with some information on known application rates. If, for instance, the low end of the label application rates (0.1 kg a.i./acre) is applied to the acreage of Study Area crops with some copper use data (22,740 acres), the resulting annual usage would be 2.3 t/yr, similar to the 2 t/yr estimate based on known usage rates. Therefore, the estimate for copper usage on the remaining crops was based on the lowest label application rate.

Table 19 shows the results of annual copper usage estimated for the 61,135 acres with no reported usage rates (8 t/yr). Combined with copper loads from crops with "known" application rate data, total copper release is 10 t/yr.

Table 19. Estimates of Copper Release Due to Agricultural Pesticide Applications.

Crop Information Type	Acres in Study Area	Percent of Acres Treated with copper (a)	Statewide Copper Application Rates (kg a.i./acre)	Label Application Rates (kg a.i./acre)	Basis for Copper Release Estimate	Copper Release in Study Area (t/yr)
Crops with statewide copper use data (b)	22,740	1.3 – 44.5%	0.23 – 2.7	0.1 – 22.7	Acres in Study Area x % Acres Treated x Statewide Appl. Rates	2
Crops without statewide copper use data (c)	61,135	No data available	No data available	0.1 – 22.7	Acres in Study Area x Lowest Label Appl. Rate	8
					Total Estimate:	10

(a) May include multiple applications on same acreage

(b) Apples, pears, potatoes, berries (raspberries, blackberries, boysenberries, and marionberries), and "caneberries" (includes some orchard/vineyard and lavender crops)

(c) Thirty-three different crops

a.i.: active ingredient

Uncertainty

There are large uncertainties associated with agricultural copper use. Highly accurate estimates are not feasible without accurate reporting on the acreages treated and actual rates of copper application.

Estimate of Copper Released from Agricultural Micronutrient Application

An estimate of copper released from micronutrients was calculated from the reported amounts distributed in the state annually. Producers of lime and fertilizer products registered for distribution in Washington State report metals concentrations for each of their products to the WSDA. However, copper is not tested as a component of fertilizer products and is only reported when sold as a micronutrient.

WSDA calculated a statewide estimate of copper compounds sold as micronutrients as reported in the annual tonnage reports (WSDA, 2005-2008). To estimate the copper load in the Study Area, a factor of 0.0164 was applied to the statewide copper load for micronutrients to reflect the portion of the total statewide crop area (4,387,169 acres; NASS, 2007) that falls within the Study Area (71,680 acres; Envirovision et al., 2008).

Mean annual copper releases for the years 2005 through 2008 are shown in Table 20. The total copper release was estimated to be 5.4 t/yr.

Table 20. Estimates of Annual Mean Copper Release from Agricultural Micronutrient Applications.

Material	Copper Release in Washington (t/yr)	Copper Release in Study Area (t/yr)
Micronutrients	333	5.4
	Total Estimate:	5.4

Uncertainty

The largest source of uncertainty may be the assumption that micronutrient application practices within the Study Area are identical to other regions of the State. The validity of this assumption has not been assessed. Also, reported concentrations may over-estimate actual concentrations since WSDA used detection limits to substitute for non-detected results.

Estimate of Copper Released from Vehicle Tire Wear

Releases of copper due to vehicle tire wear were calculated using estimates of tire wear rates and copper concentrations reported in tire material, then scaled to the Study Area using annual VKTs in the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Table 21 shows the variables used to calculate copper release from tire wear. A tire wear rate of 38 mg/km travelled was assumed for all vehicle types considered, with differences being only the number of tires per vehicle. Copper concentrations in tire material vary considerably, with the lowest reported concentration in the low parts per million. Estimates of total copper release from tire wear are 0.02 to 5.4 t/yr, with a mean of 1.9 t/yr.

Table 21. Estimates of Copper Release from Vehicle Tire Wear.

Vehicle Type	Number of Tires per Vehicle	Tire Wear Rate (mg/km/tire)	Copper Concentration in Tire Material (mg/kg)	Vehicle Kilometers Travelled in Study Area (km/yr)	Copper Release in Study Area (t/yr)
Motorcycle	2	38	2 – 490 174 (mean)	2.3E+08	<0.001 – 0.009 0.003 (mean)
Passenger Car	4			3.6E+10	0.011 – 2.7 0.96 (mean)
Light Truck	4			1.5E+10	0.004 – 1.1 0.39 (mean)
Bus	8			1.2E+08	<0.001 – 0.017 0.006 (mean)
Single Unit Truck	8			2.8E+09	0.002 – 0.41 0.15 (mean)
Combination Truck	18			3.5E+09	0.005 – 1.2 0.41 (mean)
				Total Estimate:	0.02 – 5.4 1.9 (mean)

Uncertainty

Overall estimates of tire wear rates, copper concentrations in tire material, and VKTs appear to be reliable since the estimates are based on published information and WSDOT data (see Appendix E). Tire wear rates reported in the literature appear to be realistic when checked against what may be considered a reasonable tread loss over the life of a tire (see Appendix E for this analysis). However, both the tire wear rates and copper concentrations may vary considerably based on tire brand, with wear rates also strongly influenced by driving conditions. Although the VKTs are based on the 12-county Puget Sound region, they are a reasonable proxy for the Study Area.

Estimate of Copper Released from Aquatic-Use Algaecide Application

Ecology permits the use of copper-based algaecides in irrigation canals under the NPDES and State Waste Discharge General Permit programs (Ecology, 2008). The Pesticide Incident Reporting and Tracking (PIRT) Panel reported that 77 t (active ingredients) of copper compounds and chelated copper were used in 16 of the 97 irrigation districts in Washington during 2007 (PIRT, 2008). However, none of the permits issued to irrigation districts for copper application were in the Study Area, a pattern which appears to be typical from year to year.

Prior to 2001, Ecology issued permits for the application of copper-based algaecides in lakes but discontinued the practice due to concerns about effects to salmonids as well as accumulation of copper in sediments after repeated usage. However, Ecology is currently considering allowable uses of copper to control invasive aquatic species, particularly zebra and quagga mussels (Ecology, 2010a-Draft).

Although there is currently no use of copper-containing aquatic pesticides under permitted conditions in the Study Area, unpermitted uses may exist for pools, spas, fountains, and small ponds. Since data are unavailable on use rates, loads calculated for the San Francisco Bay area (TDC, 2004) were used to estimate copper releases from this source category. TDC (2004) estimated a maximum of 95,000 pounds of copper used for these applications annually, with approximately 5,000 pounds per year discharged to storm drains. A per capita release rate was calculated for the San Francisco Bay area then scaled up to the Study Area using census data for the 12-county Puget Sound region (4,475,300; U.S. Census Bureau, 2010). Based on these factors, the estimated copper releases from aquatic use algaecides in the Study Area is 1.5 t/yr (Table 22).

Table 22. Estimates of Copper Release from Aquatic Use Algaecides.

Source	Estimate of Total Copper Release in San Francisco Bay Watershed (t/yr)	Per Capita Release (g/person/yr)	Copper Release in Study Area (t/yr)
Aquatic Use Algaecides	2.3	0.32	1.5
		Total Estimate:	1.5

Uncertainty

There are many uncertainties associated with the estimate for copper released as a result of unpermitted use as an algaecide, and the degree of confidence in the final number is low. TDC (2004) estimates of the amount of copper used, as well as the amount released, are uncertain. Therefore the estimate for the Study Area is scaled down from an uncertain estimate, and further evaluation is warranted due to the relatively high quantity of copper potentially released from this source.

Estimate of Copper Released from Non-Structural CCA-Treated Wood

Wood pressure-treated with the pesticide chromated copper arsenate (CCA) has traditionally been used widely in outdoor applications due to its fungicidal and insecticidal properties. Although after 2003 CCA was banned from most residential applications, its use remains wide-spread in industrial, commercial, and limited residential applications where its use is still permitted. Due to its durability, CCA-treated wood remains in service years and decades after its initial installation.

The literature provides estimates for copper leaching rates of new and weathered CCA-treated wood. For instance, Rasem et al. (2010) estimate copper leaching at rates of 0.52 – 2.24 mg/m^2/d for newly treated wood, 0.25 – 0.58 mg/m^2/d for weathered wood, and 5.45 mg/m^2/d in wood used for marine applications. Lebow et al. (1999) found that after 10 years 2-3% of copper content leached from CCA lumber products and 21-24% leached from submersed pilings.

Although copper leaching rates are obtainable in the published literature, little information is available on the volume of CCA-treated wood in the Study Area. In the absence of information necessary to conduct basin-wide, scale-up rates of copper leaching from CCA-treated wood, information from the San Francisco Bay Clean Estuary Partnership (TDC, 2004) was used as a proxy. A per capita release rate was calculated for the San Francisco Bay area then scaled up to the Study Area using census data for the 12-county Puget Sound region (4,475,300; U.S. Census Bureau, 2010). Based on these factors, the estimated copper releases from CCA-treated wood in the Study Area are 0.04 - 0.08 t/yr. The mid-point of this range is 0.06 t/yr (Table 23).

Table 23. Estimates of Copper Release from Non-Structural CCA-Treated Wood.

Source	Estimate of Total Copper Release in San Francisco Bay Watershed (t/yr)	Per Capita Copper Release (g/person/yr)	Copper Release in Study Area (t/yr)
Non-Structural CCA-Treated Wood	0.064 – 0.13 0.095 (mid-point)	0.0091 – 0.018 0.014 (mid-point)	0.04 – 0.08 0.06 (mid-point)
		Total Estimate:	0.04 – 0.08 0.06 (mid-point)

Uncertainty

Uncertainties associated with copper release estimates are primarily related to (1) the reliability and accuracy of assumptions about copper releases calculated in the San Francisco Bay area study and (2) the assumption that the Study Area and San Francisco Bay area are identical with regard to CCA-treated wood use and leaching. Since the literature shows a range of copper leaching rates from CCA-treated wood, a preferable method for calculating releases in the Study Area is to obtain regional information on CCA wood use and local conditions related to leaching rates.

Estimate of Copper Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Industrial and military facilities reported total mean annual copper (and copper compound) releases of 31 t/yr for the five TRI categories assessed (Table 24). The highest overall reported copper releases were from the U.S. Army (Fort Lewis) and Navy (Bremerton) bases, and from aluminum smelters and other metal works.

Fort Lewis was the dominant discharger of copper and copper compounds, with an annual mean release of 25 t/yr reported in the "other disposal" on-site release category, presumably due to munitions (shell casing) disposal on firing ranges. The Bremerton PSNS had the single largest fugitive air emissions of copper (2.5 t/yr) as well as discharges to water (0.50 t/yr). Copper emissions from stacks were much lower than fugitive air emissions, with the largest releases from the Intalco Aluminum Corporation in Whatcom County (0.98 t/yr). A large number of facilities reported copper discharges to POTWs, and no single industrial category stood out as a large source. TTM Technologies, Inc., a King County manufacturer and distributor of circuit boards, had the highest mean annual copper release to POTWs (0.09 t/yr).

RSA Microtech, a Skagit County producer and distributor of micronutrient fertilizers, had modest (≈ 0.01 kg/yr) releases of copper to air and POTWs during 2000, its only reporting year. It appears that RSA no longer operates in the Puget Sound area, and copper release data from 2000 were not included in the calculations presented here.

Table 24. Estimates of Mean Annual Copper Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 1999-2008.

Release by Facility Type	Percent of Total	Copper Release in Study Area (t/yr)
Army Base	80%	25
Naval Shipyard	17%	5.1
Metal Foundries	<1%	0.22
All Others	3%	0.83
Total	100%	31
Release by Medium	**Percent of Total**	**Copper Release in Study Area (t/yr)**
Fugitive Air Emissions	13%	4.0
Stack Air Emissions	<1%	<0.031
Surface Water Discharge	2%	0.62
Other On-Site Land Disposal	84%	26
Discharge or Transfer to POTWs	<1%	<0.031
Total	100%	31
Total Estimate:		31

POTWs: Publicly-owned treatment works

Uncertainty

Criteria and limitations of TRI reporting requirements are in Appendix C.

Estimate of Copper Released from Modeled Air Emission Sources

Air emissions of copper inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Point source air emissions account for all of the inventoried releases (0.44 t/yr), but the exact source was not identified in the Air Emissions Inventory report (Table 25). Based on a county-by-county breakdown of air emissions, it appears that up to one-fifth of the point source emissions of copper may have also been reported under TRI. But this appears to be a minimal amount overall (<0.1 t/yr), and data are not available to positively conclude that this amount was double-counted for the present report.

Table 25. Estimates of Copper Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	Copper Release in Study Area (t/yr)
Point Source - Industrial, Commercial, Institutional (primarily Title V AOP)	0.44
Total Estimate:	0.44

AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

Uncertainty

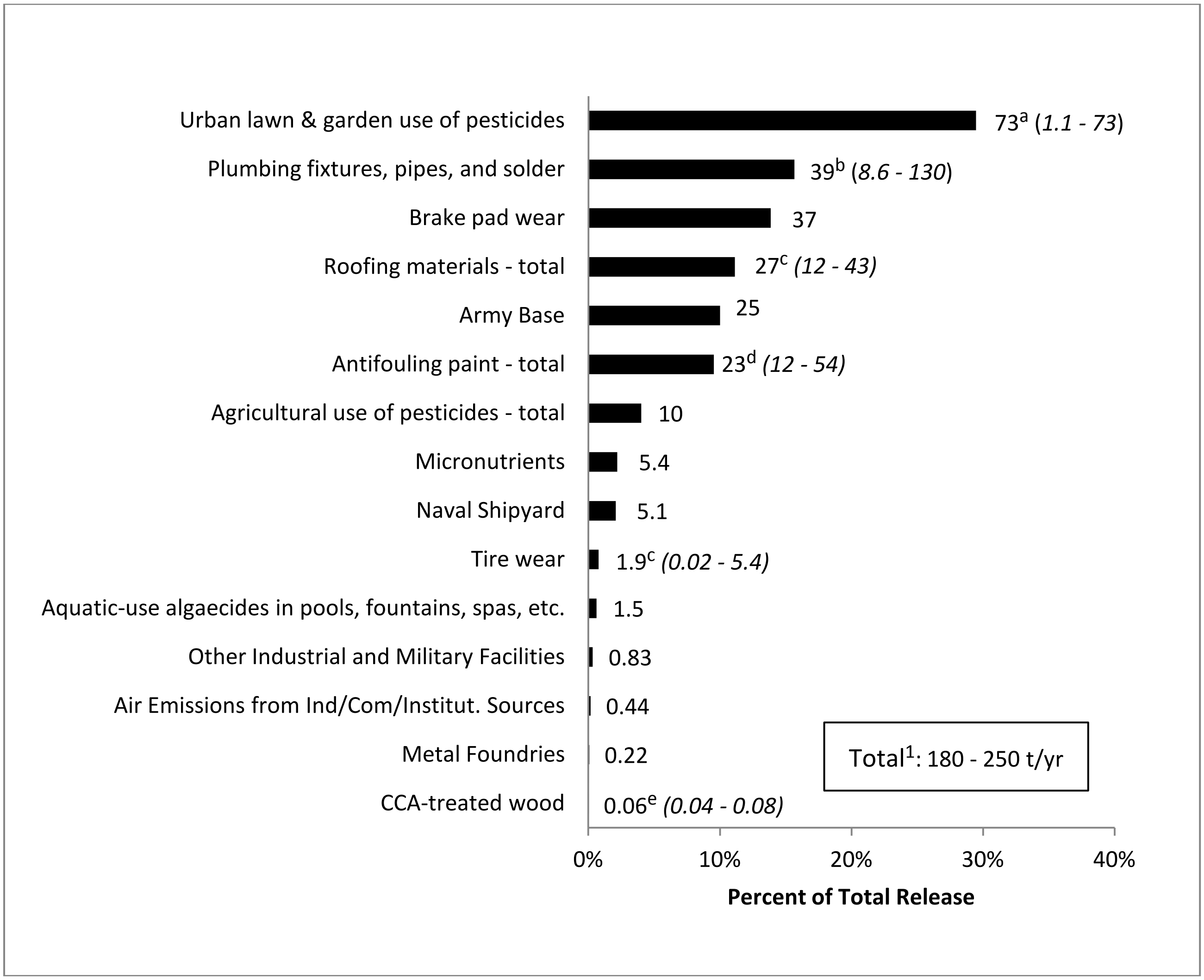
Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of Copper Release Estimates

Total annual copper releases from the 15 sources assessed totaled approximately 180 – 250 t (Figure 5). The largest single source of release is potentially urban lawn & garden pesticide use, contributing as much as 40% of the total copper release based on the high estimate for this source and the lowest estimates for other sources. Conversely, urban pesticide use may account for less than 1% of the copper release if the low urban pesticide estimate is used.

All pesticidal uses of copper combined (lawn & garden, anti-fouling paint, agriculture, pools & spas, treated wood) account for anywhere between 10% and 60% of the anthropogenic copper release, depending on the estimates used. Leaching from plumbing components and brake pad wear each appear to account for approximately 15% - 20% of the total copper release based on ‘best estimates.’ The Fort Lewis Army Base had the largest release of copper (approximately 10% of the total) among all of the industrial and military facilities reporting under TRI.

The uncertainty in the estimated overall copper release is driven mainly by uncertainty in the estimates for urban use of copper-containing pesticides. The two estimates derived for this source are vastly different, with the higher estimate representing the greatest potential source of anthropogenic copper in the Study Area. However, there is no applicable information to assess whether either of the estimates of copper released from this source is realistic.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] High end of range

[b] Median

[c] Average

[d] Sum of means for recreational and commercial and mid-point for naval vessels

[e] Mid-point of range

Figure 5. Total Copper Release in the Study Area (values shown are t/yr).

Lead

Sources Assessed

Assessment of ongoing anthropogenic lead releases included the following sources:

- Ammunition and hunting shot use
- Fishing sinker losses
- Wheel weight losses
- Roofing material runoff
- Aviation fuel use
- Vehicle brake pad wear
- Vehicle tire wear
- Plumbing fixtures, pipe, and solder leaching
- Fertilizer use
- Industrial, commercial, and institutional facilities reporting under the TRI requirements
- Residential fuel use, except wood
- Locomotive emissions

Other possible sources of lead release include use in paints, vehicle maintenance and repair activities, fireworks, and base metal smelting and refining. Releases from these sources beyond those reported in the TRI were not assessed.

Estimate of Lead Released from Lead Ammunition

Lead remains the preferred metal for most types of ammunition used in hunting and target practice, although efforts to find and promote alternatives appear to be gaining ground (Ecology and WDOH, 2009). In Washington, lead shot has been prohibited for all waterfowl, coot, and snipe hunting since a nation-wide phase-in of non-toxic shot was implemented during 1986-1991 (Ecology and WDOH, 2009). In preparing the Chemical Action Plan (CAP) for lead, Ecology and Health (2009) attempted to estimate the quantity of lead ammunition currently used by hunters in Washington. Estimates of statewide lead ammunition use were based on estimates of nation-wide and statewide ammunition use and sales (Table 26). Statewide ranges of 40 to 1,226 t/yr were scaled to the Study Area based on the fraction of the state population (67.2%), resulting in the Study Area estimates of 27 to 823 t/yr.

Possible shortcomings of each estimate are described in the CAP. Estimates based on retail sales and tax revenue data often do not distinguish between lead and other metal ammunition. Data based on hunter surveys are generally limited to specific game and assumptions about the number of shots fired. Authors of the CAP evaluated the various estimates and concluded that the state revenue analysis of lead shot provided the best estimate (556 t/yr). Scaled to the Study Area by population, the best estimate for ammunition use is 373 t/yr.

Table 26. Estimates of Lead Release from Ammunition Use.

Lead Ammunition Use in Washington* (t/yr)	Basis for Estimate	Lead Release in Study Area (t/yr)
40	Washington share of tax revenues paid by retailers of hunting goods	27
85	State estimate of shot used in upland bird hunting	57
520	TTB estimate for all ammunition sold nation-wide	349
556	State revenue analysis for proposed tax on lead shot	373
1,226	USGS national estimate of total ammunition use	823
	Total Estimate:	27 – 823 373 (best estimate)

*Source: Ecology and WDOH, 2009
t: metric ton
TTB: Tobacco Tax and Trade Bureau
USGS: U.S. Geological Survey

Uncertainty

Estimates of ammunition use in the Study Area were based on statewide estimates calculated in the Lead CAP (Ecology and WDOH, 2009) where the shortcomings are discussed (e.g. lack of distinction between lead and other metal ammunition, data limited to specific game). These shortcomings are therefore carried through to Study Area estimates, although the best statewide estimate (556 t/yr statewide) appears to avoid these limitations and should be considered reliable. No data were available to assess the geographical distribution of ammunition use with the state.

Another variable which has not been evaluated is the relationship between lead ammunition use and the environmental release of lead. For instance, even the best estimate for statewide lead ammunition use does not distinguish between ammunition sold and actually used, nor does it distinguish between lead used in firing ranges where it may be collected and recycled after being fired.

Estimate of Lead Released from Fishing Sinkers

Losses of lead fishing sinkers statewide were estimated during preparation of the Lead CAP (Ecology and WDOH, 2009). Most estimates were derived from national estimates of sales or production of lead sinkers (Table 27). The only state-specific estimate calculated in the CAP is based on annual state fishing license sales (approximately 787,000) and assumptions about the fractions of fly-fishers (30%) and annual sinker loss for the remainder of anglers (4 ounces per year). Statewide ranges of 48 to 80 t/yr were scaled to the Study Area based on the fraction of the state population (67.2%), resulting in the Study Area estimates of 32 to 54 t/yr, with a median value of 36 t/yr.

Table 27. Estimates of Lead Release from Fishing Sinker Loss.

Lead Fishing Sinker Loss in Washington* (t/yr)	Basis for Estimate	Lead Release in Study Area (t/yr)
48	EPA estimate based on sinkers (<2 cm) produced nation-wide	32
52	CWS estimate based on sinkers produced in U.S.	35
54	Industry estimate on fishing sinker sales nation-wide	36
63	Statewide estimate based on fishing license sales and sinker loss rates	42
80	CWS estimate of U.S. sales of fishing sinkers	54
	Total Estimate:	32 – 54 36 (median)

*Source: Ecology and WDOH, 2009
CWS: Canadian Wildlife Service

Uncertainty

It appears that statewide estimates of lead fishing sinker losses based on national sales and production data are consistent with an independent estimate based on state-specific data. Therefore the statewide estimates, from which Study Area estimates are based, should be considered reliable.

Recreational fishing patterns across the state may vary substantially. For instance, there may be more fishable freshwaters outside of the Study Area, but a high proportion of fished marine waters may occur within the Study Area (no data were evaluated to distinguish lead fishing weight losses between fresh and marine waters). No directional bias is therefore evident for Study Area release estimates based on scaling statewide data.

Estimate of Lead Released from Wheel Weights

Lead has historically been the preferred metal for balancing vehicle wheels due to its density, malleability, and relatively low cost. Since balancing weights are not permanently affixed to wheels, losses to the environment are common, and weights may accumulate on urban streets due to increased stop-and-go traffic and wheel jarring due to potholes, curbs, or roadway bumps. Once released from wheels, the weights are vulnerable to pulverization from traffic and exposure to stormwater.

The use of lead wheel weights is currently being phased out in Washington (Ecology and WDOH, 2009). Most new car manufacturers have already switched to non-lead wheel weights. Les Schwab, one of the state's largest tire retailers, voluntarily switched to steel weights in all of its stores beginning in 2009. That same year, the state enacted a law requiring alternatives to lead wheel weights at time of tire replacement or repair (vehicles with wheel diameters >19.5 inches or gross vehicle weight >14,000 pounds are exempt). This law went into effect on January 1, 2011 (RCW 70.270).

Ecology and WDOH (2009) estimated annual loss of lead wheel weights statewide for the Lead CAP (Table 28). Estimates were based largely on national averages scaled to Washington. While estimates of the fraction of wheel weights lost ranged from 3% to 15%, the estimates of the quantities released were fairly uniform among the information sources. Ecology and WDOH (2009) considered a national study by USGS to contain the most reliable and applicable estimate for statewide wheel weight loss (40 t). Scaled to the Study Area by using the fraction of the state population (67.2%), the best estimates of lead released from wheel weights was 28 t/yr.

Table 28. Estimates of Lead Release from Wheel Weight Loss.

Wheel Weight Loss in Washington* (t/yr)	Basis for Estimate	Lead Release in Study Area (t/yr)
30	National estimate originally based on data from urban roads in Albuquerque, NM	20
35	National estimate based on data from Albuquerque, NM	24
40	USGS national estimate of wheel weight loss	28
41	Statewide estimate based on vehicle registrations	29
	Total Estimate:	20 – 29 28 (best estimate)

*Source: Ecology and WDOH, 2009
USGS: U.S. Geological Survey

Uncertainty

It appears the national estimates for lead wheel weight losses are consistent across information sources. A relatively high level of precision for estimates of wheel weight loss would be expected due to the relative ease at which the variables can be measured (e.g. number of registered vehicles from local governments, number of replacement weights from tire installers).

Although there appears to be a high level of certainty in the estimates of lead released from wheel weight loss, these estimates may quickly become out-dated as phase-out actions are underway.

Estimate of Lead Released from Roof Runoff

Lead runoff concentrations were assessed from four types of roof materials: asphalt shingle, metal, wood shake/shingle, and built-up roofing. No applicable data were found for lead runoff for other roofing materials.

Data on lead associated with runoff from asphalt shingle roofs are scarce, even though it is the most common type of material in residential areas of the western U.S. (Dodson, 2007). Chang and Crowley (1993) reported a mean lead concentration of 56 ug/l from asphalt shingles. Steuer et al. (1997) provide data on roof runoff in a small residential catchment where roof runoff is primarily asphalt shingle, although some runoff from the upper painted sides of buildings and galvanized or metal roofing components may have been included in their samples. Mean lead concentrations of runoff from this study were 25 ug/l, similar to another such study done in 1993

(Bannerman et al., 1993 [21 ug/l]), but much higher than a study from 2001 (Davis et al., 2001 [1.5 ug/l]), possibly reflecting a higher contribution from lead in atmospheric deposition during the early-to-mid-1990s.

Steuer et al. (1997) also reported comparatively high concentrations of lead (52 ug/l) in commercial roofing typically composed of flat rubberized or tar-sealed roofs. In this case, concentrations were similar to runoff isolated to commercial and industrial roofs reported by Davis (2001) (62 – 64 ug/l), but still substantially higher than both Bannerman (1993) (8 – 9 ug/l) and the rainfall-corrected lead values from rubberized roofing materials as recorded by Clark (2010) (-1.7 – 8.7 ug/l). The mean lead concentration in runoff from built-up roofs used for release estimate calculations was 27 ug/l (Table 29). Clark also recorded lead in runoff from wood shingles, after correcting for atmospheric deposition, ranging from -2.5 to 3.2 ug/l, with a mean concentration of 0.81 ug/l.

The available literature suggests that metal roofs, including weathered metal with aluminum paint, corrugated aluminum, and Galvalume roofing, appear to result in relatively low lead runoff concentrations, averaging 5.4 ug/l. This may simply be a reflection of the rainfall-corrected Clark (2010) data which heavily influenced the computation of the mean. In one of the few datasets from the Pacific Northwest, Good (1993) found relatively low lead concentrations (mean of 8 ug/l) in runoff from a weathered metal sawmill roof located near the outer Washington coast.

All of these data should be used with caution due to the lack of correction for atmospheric deposition, with the exception of these reported by Clark (2010). Phase-out of leaded gasoline occurred from 1973 – 1996, with the bulk of the reductions occurring in the mid-1970s; by 1995, lead emissions from gasoline were estimated to be less than 1% of the level prior to initiation of the phase-out (EPA, 1996). Therefore, runoff sampling conducted as late as the mid-1990s may contain high levels of atmospherically deposited lead, and lead concentrations in rainfall probably vary by geographical location as well. For instance, Chang and Crowley (1993) reported lead concentrations of 23 ug/l in East Texas rainwater while analyzing roof material runoff for water quality parameters. Clark (2010) data suggests that lead levels in central Pennsylvania rainwater had diminished substantially by the late-2000s; lead levels in runoff from control panels designed to represent atmospheric deposition ranged from 0.66 – 3.3 ug/l.

Annual rooftop runoff volumes for each roof type were used to translate lead concentrations in runoff into release loads. The method for the calculation of runoff volumes is described in Appendix B. Total annual release loads for wood shake/shingle, metal, asphalt shingle, and built-up roofs are 0.04, 0.23, 15, and 2.7 t/yr, respectively (Table 29). The combined 'best estimate' load (18 t/yr) is the sum of lead release from asphalt composite roofs and mean releases from wood, metal, and built-up roof types.

Table 29. Estimates of Lead Release from Roofing Material.

Roof Type	Total Area of Roof Type in Study Area (m^2)	Total Runoff Volume of Roof Type in Study Area (l/yr)	Lead Concentration in Runoff (ug/l)	Lead Release in Study Area (t/yr)
Wood Shake/Shingle	3.62×10^7	5.45×10^{10}	-2.5 – 3.3 0.81 (mean)	-0.14 – 0.18 0.04 (mean)
Metal	2.96×10^7	4.29×10^{10}	2.1 – 6.1 5. 4 (mean)(a)	0.09 - 0.26 0.23 (mean)
Asphalt Composite	3.96×10^8	5.84×10^{11}	25	15
Built-Up	7.40×10^7	1.03×10^{11}	1.3 – 52 27 (mean)	0.13 – 5.3 2.7 (mean)
			Total Estimate:	15 - 20 18 (best estimate)

(a) Based on runoff concentrations from metal roof with aluminum paint, corrugated aluminum, and Galvalume roof materials

Uncertainty

The greatest uncertainty in estimates of lead released from rooftops is most likely due to representativeness of the release data obtained from the literature. Some of the runoff data used to calculate release loads was corrected for atmospherically deposited lead; other data were not. The disparity in release loads for various roof types may reflect differences in accounting for lead in rainfall. Furthermore, roofing age, unaccounted for here, may also play a factor, as release rates may vary over the life of a roof.

Ideally, estimates of lead released from rooftops should include only:

1. Data generated well after the phase-out of leaded gasoline (for on-road vehicles) was complete in 1996.
2. Studies geographically isolated to the Pacific Northwest and possibly an even higher resolution of geographical representativeness, depending on the specific use of the loading data.
3. Studies corrected for lead (and other metals) in rainfall or with accompanying atmospheric deposition data.

An example of a seemingly valuable study that in fact contains none of these qualities was published by Yaziz et al. (1989). The study found high (uncorrected) lead concentrations (102 – 271 ug/l, median of 204 ug/l) in Malaysian roof runoff well before the beginning of the Malaysian phase-out of leaded gasoline in the late 1990s - early 2000s (Earth Summit Watch, 2010).

The example above is provided as a cautionary note. Unfortunately, it appears there is a bounty of unusable literature such as the Yaziz et al. study apparently due to an active evaluation of roof runoff for drinking water use in developing countries during the 1980s and 1990s.

Scale-up information used to estimate lead releases from roof materials in the Study Area appears to be fairly reliable. Accuracy may be increased by collecting information on roof type fractions from jurisdictions in addition to those discussed in Appendix B. Runoff data available for the four roof types used to calculate lead release estimates accounted for approximately 96% of the roof area in the Study Area.

Estimate of Lead Released from Aviation Fuel

The most commonly used non-jet aviation fuel (Avgas 100LL) contains approximately the same concentration of lead (2.1 g/gal) as typical vehicle gasoline prior to the initial phase-out of leaded gasoline in the 1970s (EPA, 2002). As cited in the Lead CAP (Ecology and WDOH, 2009), 11,437,910 gallons of Avgas 100LL were sold in Washington during 2006. Scaled to the Study Area by using the fraction of the state population (67.2%), an estimated 7.7 million gallons of Avgas 100LL containing 16 t/yr of lead are sold and released in the Study Area (Table 30).

Table 30. Estimates of Lead Release from Aviation Fuel.

Amount of Avgas 100LL Sold in Washington (gallons)	Amount of Avgas 100LL Sold in Study Area (gallons)	Lead Concentration in Avgas (g/gal)	Lead Release in Study Area (t/yr)
11,437,910	7,681,060	2.1	16
		Total Estimate:	16

Uncertainty

Uncertainty about the release of lead from aviation fuel is centered around the difference between the location of where the fuel is sold and where it is actually used. The release estimate assumes that all of the fuel sold in the Study Area is used in the Study Area. Although it seems unlikely that all purchases of Avgas 100LL are used in the Study Area – it is likely common for planes departing from the Study Area to fly outside of the region boundaries – the difference may be accounted for by planes flying into the Study Area from other locations. There are no available data to determine if these patterns of aviation fuel use balance out to equal the amount sold.

Another uncertainty involves the scaling of statewide Avgas 100LL use to determine Study Area use. There does not appear to be a reason why the estimates are not proportional based on population, and there is no obvious direction in bias if one exists.

Estimate of Lead Released from Vehicle Brake Pad Wear

Releases of lead due to vehicle brake pad wear were calculated using estimates of brake pad wear rates and lead concentrations reported in brake pad material, then scaled to the Study Area using annual vehicle kilometers travelled (VKTs) in the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Wear rates for brake friction materials were estimated from various literature values and were found to range from 3 mg/km travelled for motorcycles to 245 mg/km travelled for combination trucks (Table 31). Different vehicle types employ different types of braking systems, with motorcycles, passenger cars, and light trucks primarily using disc brakes and heavier vehicles using drum brakes almost exclusively. There are few data on lead in drum brake friction materials, and they have not been investigated extensively since most of the abraded material is retained in the brake drums (Sinclair Rosselot, 2006). Based on the differences in braking systems among vehicle types, mean lead concentrations were calculated only for motorcycles, passenger cars, and light trucks although maximum releases from other vehicle types are also calculated by assuming maximum lead concentrations also apply to drum material (Table 31).

Lead releases due to brake pad wear are estimated to be 0.04 – 13 t/yr. The combined 'best estimate' release (2.6 t/yr) is the sum of the mean release estimates for motorcycles, passenger cars, and light trucks.

Table 31. Estimates of Lead Release from Vehicle Brake Pad Wear.

Vehicle Type	Brake Pad Wear Rate Per Vehicle (mg/km)	Lead Concentration in Brake Pad Material (mg/kg)	Vehicle Kilometers Travelled in Study Area (km/yr)	Lead Release in Study Area (t/yr)
Motorcycle	3	50 – 6,594 3,126 (mean)	2.3E+08	<0.001 – 0.005 0.002 (mean)
Passenger Car	16		3.6E+10	0.03 – 3.8 1.8 (mean)
Light Truck	16		1.5E+10	0.011 – 1.6 0.75 (mean)
Bus	110	6,594 (max)	1.2E+08	0.083 (max)
Single Unit Truck	129		2.8E+09	2.4 (max)
Combination Truck	245		3.5E+09	5.6 (max)
			Total Estimate:	0.04 – 13 2.6 (best estimate)

Uncertainty

Estimates of mean lead releases due to brake pad wear are highly uncertain due primarily to the difficulty obtaining reliable estimates of wear rates. Much of the wear rate uncertainty is due to a wide range of assumptions possible for assigning the proportion of disc brake use among various vehicle types. For instance, in the present analysis an inconsequential proportion of passenger cars and light trucks are assumed to use drum brakes. If reliable estimates were available for the concentration of lead in drum lining materials, the proportion of cars with drum brakes could have been considered in the current analysis.

Lead concentrations in brake pads range by two orders of magnitude, with little assessment of confidence about the mean concentration of 3,126 mg/kg. Preferable methods for calculating chemical releases from brake pad wear would consider automobile make/models independently and calculate releases based on the brake pads and VKTs associated specifically with each make/model. However, for each vehicle make/model there are typically a variety of both factory-installed or after-market pads available which may contain different amounts of metals concentrations. This suggests that data on VKTs by each car make/model may not by itself improve the level of certainty in release estimates. As noted in the analysis provided in

Appendix E, other variables such as driving habits and types of roads travelled are also considerations in determining releases from brake pad wear, but a detailed evaluation of these variables are beyond the scope of the present report.

Estimate of Lead Released from Vehicle Tire Wear

Releases of lead due to vehicle tire wear were calculated using estimates of tire wear rates and lead concentrations reported in tire material, then scaled to the Study Area using annual VKTs in the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Table 32 shows the variables used to calculate lead release from tire wear. A tire wear rate of 38 mg/km travelled was assumed for all vehicle types considered, with differences being only the number of tires per vehicle. Lead concentrations in tire material vary considerably, with the lowest reported concentration in the low parts per million. Estimates of total lead release from tire wear are 0.01 to 1.8 t/yr, with a mean of 1.2 t/yr.

Table 32. Estimates of Lead Release from Vehicle Tire Wear.

Vehicle Type	Number of Tires per Vehicle	Tire Wear Rate (mg/km/tire)	Lead Concentration in Tire Material (mg/kg)	Vehicle Kilometers Travelled in Study Area (km/yr)	Lead Release in Study Area (t/yr)
Motorcycle	2	38	1 – 160 107 (mean)	2.3E+08	<0.001 – 0.003 0.002 (mean)
Passenger Car	4			3.6E+10	0.006 – 0.9 0.59 (mean)
Light Truck	4			1.5E+10	0.002 – 0.4 0.24 (mean)
Bus	8			1.2E+08	<0.001 – 0.006 0.004 (mean)
Single Unit Truck	8			2.8E+09	0.001 – 0.1 0.09 (mean)
Combination Truck	18			3.5E+09	0.002 – 0.4 0.25 (mean)
				Total Estimate:	0.01 – 1.8 1.2 (mean)

Uncertainty

Overall estimates of tire wear rates, lead concentrations in tire material, and VKTs appear to be reliable since the estimates are based on published information and WSDOT data (see Appendix E). Tire wear rates reported in the literature appear to be realistic when checked against what may be considered a reasonable tread loss over the life of a tire (see Appendix E for this analysis). However, both the tire wear rates and lead concentrations may vary considerably based on tire brand, with wear rates also strongly influenced by driving conditions. Although the VKTs are based on the 12-county Puget Sound region, they are a reasonable proxy for the Study Area.

Estimate of Lead Released from Indoor Residential Plumbing

Lead may be released from residential plumbing through the use of brass fittings and, in some cases, lead piping and lead solder. Water supply lines may also contain brass fittings and lead piping. Although the 1986 amendments to the Safe Drinking Water Act banned new uses of piping with lead content >8% lead and solder with lead content >0.2%, older homes and water service utilities may contain older pipes and solders with high lead content. EPA limits the amount of lead which can leach from any end-point device as an acceptable method to meet the Safe Drinking Water Act requirements. The current leaching limit is 11 ug/l, slated to be reduced to 5 ug/l in 2012 (Ecology and WDOH, 2009).

The release of lead and other metals from plumbing components is largely dependent on water quality variables such as pH and hardness. In 1991, EPA published the Lead and Copper Rule (LCR; 40 CFR Part 141) which required water suppliers to take actions to reduce corrosion that was largely responsible for release of lead, copper, and other metals (EPA, 2004). The LCR also requires monitoring of lead and copper at customers' taps, and sets actions levels for copper and lead at the tap. The action level for lead is 15 ug/l in 10% of taps sampled; water suppliers must take action to control corrosion when this concentration is exceeded.

Results of water sampling from taps within the 12-county Puget Sound region during 2010 were obtained from the Department of Health Drinking Water Database (WDOH, 2011). The median lead concentration was 0.5 ug/l with 25th and 75th percentile values of 0.5 and 2 ug/l, respectively (n=4,220). Lead concentrations in the Puget Sound region were well below values obtained during similar sampling efforts in California during 1998 – 2006 (annual medians ranging from 2.7 to 7.7 ug/l; Kimbrough, 2009); the reason for the differences is not clear.

Domestic water consumption rates for households described previously for copper and summarized in Appendix F were used to estimate annual loads in the Study Area. Applying the lead concentrations obtained from WDOH (2011) and per capita water consumption rates reported by Mayer et al. (1999), indoor plumbing releases an estimated 0.2 t/yr lead, with a range of 0.2 to 0.9 t/yr (Table 33).

Table 33. Estimates of Lead Release from Indoor Residential Plumbing.

Per Capita Indoor Water Consumption Rate (gal/day)	Receiving System Type	Indoor Water Consumption in Study Area (gal/yr)	Indoor Water Consumption in Study Area (l/yr)	Lead Concentration in Tap Water (ug/l)	Lead Release in Study Area (t/yr)
69.3	POTWs	8.12×10^{10}	3.07×10^{11}	0.5 - 2[a] 0.5 (median)	0.1 – 0.6[a] 0.2 (median)
	Septic	3.20×10^{10}	1.21×10^{11}		0.06 – 0.2[a] 0.06 (median)
				Total Estimate:	0.2 – 0.9[a] 0.2 (median)

[a] 25th - 75th percentile

POTWs: Publicly-owned treatment works

Uncertainty

Releases of lead from residential plumbing may vary depending on chemistry of the domestic source water and treatment by the water utility prior to distribution. To assess the lead contribution from source water, WDOH tap-water results were compared to results of paired source water data. For the 135 pairs of results available, median lead concentrations in the source water were the same as from tap water (0.5 ug/l), with a median of the differences of zero, indicating that plumbing within the distribution system and the buildings contributes no measurable lead.

LCR compliance data potentially represents the highest concentrations of lead within a given water service provider, since the rule requires monitoring of homes and building that are at the highest risk of copper and lead contamination (EPA, 2004). Therefore, the lead concentrations used for release estimates may be biased high compared to the overall population of tap water.

It is not clear why lead concentrations in the California dataset are much higher than those from the Puget Sound region. Age of the data does not appear to be a factor since no temporal trend was seen, and there was no indication that source water for the California households was particularly aggressive or mild in terms of corrosion, leaching, or other factors promoting lead release from plumbing components (Kimbrough, 2009).

Water used outdoors may also carry contaminants leached from plumbing into the environment and outdoor water usage represents 58% of the total residential water budget (Mayer et al., 1999). However, concentrations of lead and other metals may be different than those at indoor taps. Many outdoor faucets are plumbed directly from the distribution system, with no residence time in the indoor system and less exposure to components which may contribute metals. As no information specific to outdoor plumbing was found, release estimates presented here do not include water used outdoors.

Study Area water consumption data were based on a national survey conducted in a variety of geographical areas. There is no obvious reason these data would not be representative of the water consumption habits for the Study Area. However, since the survey data are over a decade old, they may over-estimate water consumption rates which may have fallen in recent years due to water conservation education, installment of low-flow fixtures, and use of water-saving appliances such as dishwashers and clothes washers.

Estimate of Lead Released from Fertilizer Application

Estimates of lead released from fertilizers were based on the reported concentrations in fertilizers and amounts distributed in the state annually, then scaled to the Study Area based on estimated portions of crop area. Details are included in the section describing arsenic releases from fertilizers.

Total lead releases are shown in Table 34. In general, lead releases corresponded with the amount of each material distributed. The total lead release was estimated to be 0.04 t/yr.

Table 34. Estimates of Lead Release from Fertilizer.

Material	Material Distributed in Washington During 2006-2007 Reporting Year (t)	Lead Release in Washington (t/yr)	Lead Release in Study Area (t/yr)
Nitrogen Material	618,550	0.50	0.008
Phosphate Material	115,474	0.55	0.009
Potash Material	94,161	0.21	0.003
Miscellaneous Fertilizer	163,652	0.33	0.005
Sulfur	25,954	0.22	0.004
Gypsum	38,409	0.10	0.002
Natural Organics	32,866	0.24	0.004
Liming Material	55,579	0.32	0.005
Total as Fertilizer	**1,144,645**	**2.5**	**0.04**
		Total Estimate:	0.04

Uncertainty

The largest source of uncertainty may be the assumption that fertilizer application practices within the Study Area are identical to other regions of the State. An annual estimate of lead release also assumes that fertilizer use during the 2006-2007 reporting year is representative of current use. The validity of these assumptions has not been assessed. Also, reported concentrations may over-estimate actual concentrations since WSDA used detection limits to substitute for non-detected results.

Lead in House Paint

Prior to 1955, lead content of house paint was as high as 50%; this level began to decrease only after initial voluntary reduction by paint manufacturers to 1% followed by a 1978 federal regulation limiting lead content of house paint to 0.06% (600 mg/kg) (Ecology and WDOH, 2009). High lead-content paint is still permitted for some specialty applications, including traffic paint, non-consumer marine paint, and paint for bridges (Ecology and WDOH, 2009). Smaller-scale applications are still allowed for artist use, touch-up paint, and some appliances and fixtures.

Although there is a substantial amount of lead remaining in painted buildings, the exact amount of lead-based paint (defined as ≥0.5%) is nearly impossible to ascertain due to the difficulties associated with tracking materials used three or more decades previous. Some buildings painted with lead-based paint may have been renovated or demolished. Accurate estimates of lead release from high lead-content paint is confounded by the fact that some paint may be exposed and weathered, while other paint may be maintained or otherwise encapsulated by over-coatings and therefore not releasing lead; the proportion of weathered to maintained/encapsulated lead-based paint is not known.

Based on a national survey scaled to Washington State and the Study Area by age category and population, there are about 210,000 houses in the Study Area with lead levels in soil above 200 mg/kg, 135,000 above 400 mg/kg, and 80,000 above 1,200 mg/kg; the majority of the lead remains in the upper ten centimeters of soil (Ecology and WDOH, 2009).

With few area-specific data to estimate lead in house paint, national estimates provided by Davis and Burns (1999) appear to offer the best data from which to derive Study Area estimates. They cite several studies which estimate the painted surface area in the U.S. (32 billion square feet) and the percent with lead-based paint (up to 27%). Scaled from the national amount by population, there are approximately 126 million square feet (12 million square meters) of exterior building surfaces with lead-based paint in the Study Area.

Davis and Burns (1999) found the mean quantity of lead released from painted surfaces was 164 ug/m^2 when sprayed with synthetic rainwater; increasing spray intensity and paint age resulted in increases in the amount of lead released. However, unlike roofs which have a planar exposure to rain, it is virtually impossible to estimate vertical wall exposure to rain, at least within the scope of the present project. Therefore, while releases of lead from lead-based paint may be substantial, they are not estimated here.

Lead in Traffic Paint

Yellow traffic paint typically contains 1% - 3% lead. Lower lead, or “lead free”, alternatives typically contain closer to 600 mg/kg, or less than 0.5% lead (5,000 mg/kg) maximum by definition (Paul Fabiniak, Ecology Hazardous Waste and Toxics Reduction Program, personal communication). It is unknown how much traffic paint is used within the Study Area, and of that, how much lead it contains. The rate of paint abrasion or leaching is also unknown, although it seems likely that a high percentage of the paint material may enter the environment if allowed to remain on the road surface for an extended period.

Ecology encourages traffic paint applicators to use the lower lead formulations (Tom Boucher, Ecology Hazardous Waste and Toxics Reduction Program, personal communication; Fabiniak, personal communication). A survey of applicators indicated that many already use the lower lead alternatives; however, some applicators continue to use higher lead-content paint. King County Department of Transportation (responsible for the maintenance of roughly 1,700 miles of unincorporated county roads) recently switched to lower lead alternatives, reducing the use of lead on King County roads by an estimated 7,000 pounds (Fabiniak, personal communication).

Estimate of Lead Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Industrial and military facilities reported total mean annual lead (and lead compounds) releases of 42 t/yr for the five TRI categories assessed (Table 35). The highest overall reported lead releases were U.S. Army (Fort Lewis) and Navy (Bremerton) bases, paper mills, and steel manufacturers.

Fort Lewis was the dominant discharger of lead and lead compounds, with an annual mean release of 39 t/yr reported in the "other disposal" on-site release category, presumably due to munitions disposal on firing ranges. Fort Lewis had the single largest fugitive air emissions of lead (0.09 t/yr), and lead stack emissions were highest from Nucor Steel in King County (0.4 t/yr). Paper mills accounted for the bulk of lead discharges to water, with the Kimberly-Clark mill in Snohomish County reporting nearly half of the lead release to water (0.5 t/yr.) Overall lead releases to POTWs were comparatively small, with Burlington Environmental (King County) releasing approximately one-quarter of the total (0.03) t/yr.

Table 35. Estimates of Mean Annual Lead Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 1999-2008.

Release by Facility Type	**Percent of Total**	**Lead Release in Study Area (t/yr)**
Army Base	89%	39
Naval Shipyard	4%	1.8
Pulp and Paper Mills	2%	0.7
All Others	5%	2.3
Total	100%	43
Release by Medium	**Percent of Total**	**Lead Release in Study Area (t/yr)**
Fugitive Air Emissions	<1%	<0.4
Stack Air Emissions	3%	1
Surface Water Discharge	3%	1
Other On-Site Land Disposal	93%	40
Discharge or Transfer to POTWs	<1%	<0.4
Total	100%	43
Total Estimate:		43

POTWs: Publicly-owned treatment works

Uncertainty

Criteria and limitations of TRI reporting requirements are in Appendix C.

Estimate of Lead Released from Modeled Air Emission Sources

Air emissions of lead inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Point source air emissions account for the bulk of releases (0.5 t/yr), but the exact source was not identified in the Air Emissions Inventory report (Table 36). Smaller releases of lead were due to combustion of distillate oil (heating oil) as a residential heating source and from locomotives.

Table 36. Estimates of Lead Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	Lead Release in Study Area (t/yr)
Locomotive Emissions	0.01
Point Sources - Industrial, Commercial, Institutional (primarily Title V AOP)	0.5
Residential Fuel Use, except Wood	0.02
Total Estimate:	0.5

AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

Uncertainty

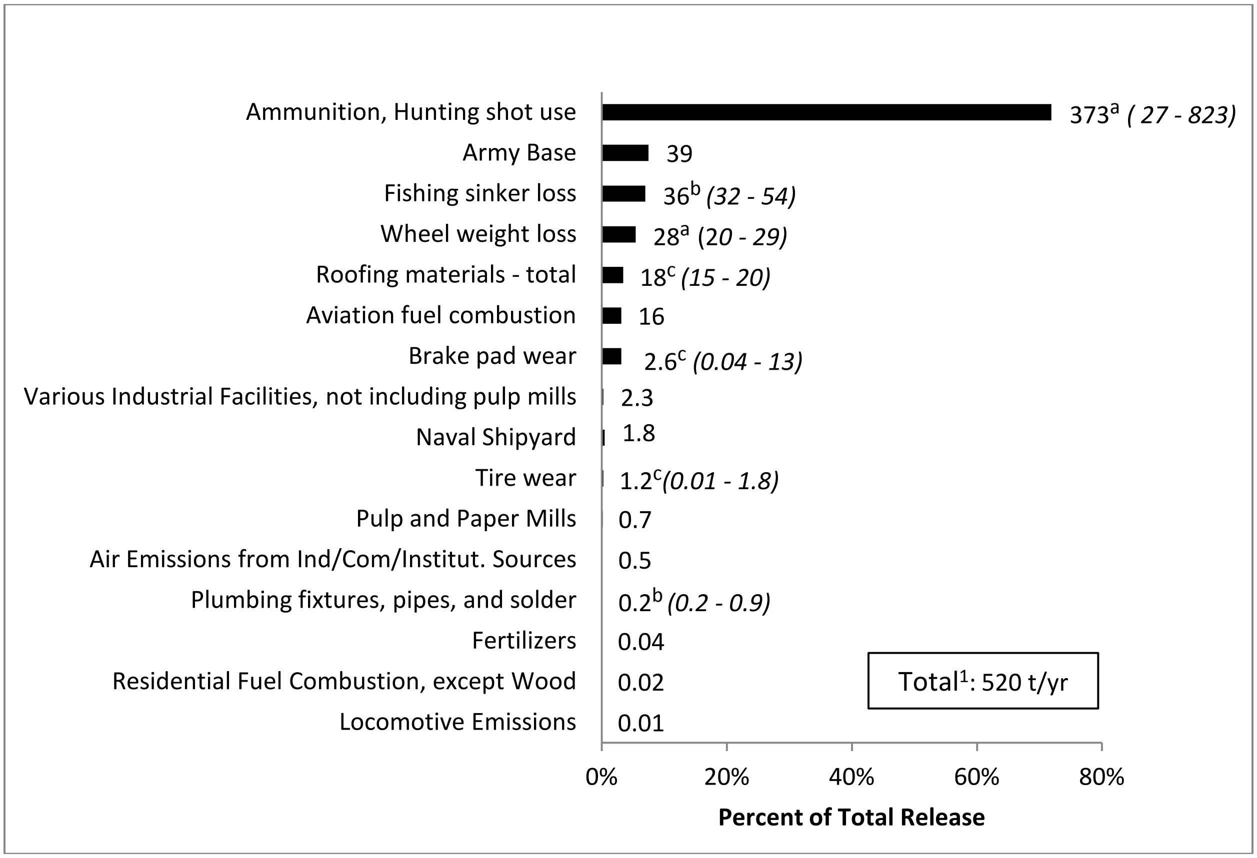
Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of Lead Release Estimates

Based on the best estimates available, total lead releases from the sources assessed are approximately 520 t/yr (Figure 6). The largest single source is ammunition and hunting shot (>72%). Lead released from Fort Lewis (7%) may also be in the form of spent ammunition, suggesting that ammunition and shot account for three-quarters of total lead releases. The estimated annual loss of fishing weights and wheel weights together account for 12% of lead releases. Therefore, at least 90% of the annual lead released to the Study Area is in the form of solid metal. Another 3% is released to air, presumably through combustion emissions. It appears that aside from wheel weight loss, a relatively small amount is released from vehicles during tire and brake pad wear (1%).

The overall uncertainty in estimates of lead release is driven mainly by a wide range of possible estimates for ammunition use. Based on the full range of ammunition use rates, total anthropogenic lead releases in the Study Area range from 150 to 1,000 t/yr. Lead releases from vehicle component wear (brake pads and tires) are small compared to bulk metal losses (ammunition, fishing sinkers, and wheel weights), but these estimates contain a high degree of uncertainty due to variable lead concentrations in component parts. At the high end of release estimates for these sources, lead emissions may be substantial (>20 t/yr).

It appears that most of the major anthropogenic sources of lead in the Study Area were assessed with the possible exception of lead in paint, both road and house paint. This source of lead may be substantial. However, due to the confounding factors discussed in the section on paint (e.g. varying age of buildings, paint encapsulation and removal), estimates of lead released from this source on a basin-wide scale would likely prove very difficult.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Most reasonable estimate

[b] Median

[c] Mean

Figure 6. Total Lead Release in the Study Area (values shown are t/yr).

Mercury

Sources Assessed

In 2003, Ecology and the Washington State Department of Health (WDOH) published the Mercury Chemical Action Plan (Mercury CAP) which inventoried uses and releases of mercury within Washington State (Ecology and WDOH, 2003). Actions undertaken since 2003, both voluntarily and under the legislative mandate provided under The Mercury Education and Reduction Act of 2003 (MERA), have both reduced the total amount of mercury used in products and increased the collection and recycling of existing mercury-containing items. MERA mandated labeling light bulbs, banned most mercury in schools, and banned or limited the sale of many mercury-containing products such as manometers, thermometers, thermostats, and switches. Ecology reports the amount of mercury collected annually since the 2003 Mercury CAP (Ecology, 2010b).

Ongoing mercury releases described in the present report are based largely on those detailed in the Mercury CAP, with adjustments made for reported collections since 2003. Unlike other COCs, mercury disposed to landfills is considered a primary source (i.e. initial release to the environment) due to its presence as a gas in some products or its potential to volatilize and escape in gaseous form.

Assessment of ongoing anthropogenic mercury releases included the following sources:

- Fuel oil, gasoline, and diesel fuel combustion
- Thermostat disposal
- Fluorescent lamp bulb disposal
- Household fever thermometers
- Crematoria emissions
- Disposal of auto convenience switches
- Button cell battery disposal
- Dental amalgam excretion
- Fertilizer use
- Military, industrial, commercial, and institutional facilities
- Residential fuel use, except wood

Estimate of Mercury Released from Fuel Oil, Gasoline, and Diesel Fuel Combustion

Residual fuel oil is the heavy oil remaining following removal of all other distillate fractions of petroleum and is used largely in ships and in industrial boilers. The 2003 Mercury CAP estimated that 0.013 tons of mercury was released statewide through residual oil (bunker fuel) combustion based on a mercury emission of 32 ug/gal for residual No. 6 (Ecology and WDOH, 2003). Based on consumption of residual fuel oil in the Study Area during 2009 (363 million gal/yr of residual oil Nos. 5 and 6; EIA, 2010), the annual mercury release is estimated to be 0.01 t/yr (Table 37).

Mercury releases through gasoline and diesel fuel combustion were obtained from the National Emissions Inventory (EPA, 2011) for the 12-county Puget Sound region. Estimated emissions of mercury from gasoline and diesel combustion are very small (Table 37). The Mercury CAP presented rates of mercury releases from these sources up to four orders of magnitude higher than reported in the National Emissions Inventory. However, the National Emissions Inventory is much more specific about the type of combustion source (e.g. vehicle use, boiler use) which translates to more accurate estimates of mercury emissions from specific fuels (Gary Palcisko, Ecology Air Quality Program, written communication).

Table 37. Estimates of Mercury Release from Fuel Oil, Gasoline, and Diesel Combustion.

Fuel Type	**Consumption in Study Area (gal/yr)**	**Mercury Emission Rate (ug/gal)**	**Mercury Release in Study Area (t/yr)**
Residual Fuel Oil Nos. 5 and 6 (bunker fuel)	3.63×10^8	32*	0.01
Gasoline (On-Road)	--	--	0.0001
Gasoline (Non-Road)	--	--	0.0001
Diesel (On-Road)	--	--	0.00004
Diesel (Non-Road)	--	--	0.00003
		Total Estimate:	0.01

t: metric ton

*Note that the emission rate is for Residual No. 6.

Uncertainty

The robustness of the dataset from which the residual fuel oil emission factor was used is unknown. The largest source of uncertainty for gasoline and diesel combustion may be that emissions are consistent from region-to-region, and that these have not changed since 2005. No data were reviewed to evaluate the validity of these assumptions.

Estimate of Mercury Released from Thermostat Disposal

Estimates of mercury released through disposal of thermostats were calculated based on information about the amount of mercury in thermostats and the number disposed of annually. Data on both the mercury content and disposal rates vary considerably and are explained below.

The Mercury CAP states that each thermostat is expected to contain roughly 3 grams of mercury per switch, and may have up to six switches (Ecology and WDOH, 2003). According to the Thermostat Recycling Corporation (TRC), household thermostats contain a mean of 1.4 mercury switches, with at least 2.8 g of mercury per switch, or about 4 g total. Industrial-sized thermostats may have multiple switches and thus have higher amounts of mercury (TRC, 2010). The Interstate Mercury Education and Reduction Clearinghouse (IMERC) reported that from 2001 to 2007 the amount of mercury used in thermostats nationally decreased by 73%, from 14.6 to 3.66 tons (Wienert, 2009). Washington and other states have restricted the sale of mercury thermostats, and active collection programs are in place.

Mercury-containing thermostats have been common for over 50 years (Leopold, 2002). The EPA estimated that in 1994 there were 70 million thermostats in residences across the U.S. and that 2-3 million are disposed of annually (Leopold, 2002). EPA also estimated that 4 – 6 million new mercury thermostats entered the market each year in the 1990s (both domestic and imports). Applying the IMERC value of 4 g per thermostat, IMERC reports provide a basis for estimating the number sold nationally from 2001 (3.3 million) to 2007 (875,000). Adding the EPA and IMERC annual sales to the 1994 EPA estimate of existing thermostats, while subtracting annual disposals, yields an estimated 82 million mercury thermostats in use nationally during 2008. Scaled by population, there are roughly 1.2 million thermostats containing an estimated 4.8 t of mercury in the Study Area.

Since thermostats can last 40 years or more, they will continue to enter the waste stream for decades. Using EPA's disposal rate of 2 - 3 million units per year suggests that all thermostats would not be disposed of until 2036 - 2050. Based on this disposal rate, 0.12 – 0.18 t of mercury from thermostats enters the Study Area waste stream annually (Table 38). This estimate agrees well with the Mercury CAP estimate of 0.19 t/yr statewide, 0.12 t of which can be assigned to the Study Area based on population apportionment.

Ecology currently expects that about 18 kg of mercury is collected in thermostats per year statewide (Diana Olegre, Ecology Hazardous Waste and Toxics Reduction Program, personal communication). TRC reports collecting a mean of 15.4 kg of mercury per year statewide from 2006-2009 (Mark Tibbetts, National Electrical Manufacturers Assn,, personal communication). Thermostats are also collected alongside thermometers as moderate risk waste, and it is unknown what portion of that waste stream belongs to each product (Al Salvi, Ecology Waste 2 Resources Program, personal communication). However, King County estimated that 4 – 16 thermostats are collected at their household hazardous waste facilities per week (Armstrong et al., 2002).

Scaling this collection rate up by population (2.34 x King County population), the Study Area would be expected to collect 486 – 1,943 thermostats and 2 – 8 kg of mercury (at 4 g mercury/ thermostat) each year. Adding the TRC thermostat collection estimate (10.32 kg mercury for the Study Area) to the collection estimates based on King County rates suggests that total mercury collections from thermostats in the Study Area yields approximately 0.012 – 0.018 t/year (Table 38).

The thermostat mercury disposal rate was based on the difference between the estimated national disposal rate (Leopold, 2002), scaled to the Study Area, and the estimated collection rate (Table 38). The mid-point of the estimated range (0.11 – 0.16 t/yr) is 0.13 t/yr.

Table 38. Estimates of Mercury Release from Thermostat Disposal.

Quantity of Mercury Retired Annually in Thermostats in Study Area (t/yr)	Quantity of Mercury in Thermostats Recycled Annually in Study Area (kg/yr)	Quantity of Mercury in Thermostats Disposed Annually in Study Area (t/yr)
0.12 – 0.18	0.012 – 0.018	0.11 – 0.16 0.13 (mid-point)
	Total Estimate:	0.11 – 0.16 0.13 (mid-point)

Uncertainty

The total number of in-use thermostats in the Study Area may not be accurate since the baseline amount (i.e. 1994 EPA estimate) and the disposal rates are scaled from either national or statewide estimates. The disposal rate may also have changed significantly over time.

Furthermore, failure to remove thermostats prior to building demolition results in the release of mercury as well. While both Ecology and King County have undertaken efforts to estimate the amount released from the demolition of buildings containing mercury thermostats, the complex variables involved prevented them from achieving a reasonable estimate (Maria Peeler, Ecology Hazardous Waste and Toxics Reduction Program, personal communication).

Estimate of Mercury Released from Fluorescent Lamp Tube Disposal

Using data from IMERC, Wienert (2009) reported the total amount of mercury in lighting sold nationally. Scaling by population to the Study Area and applying a mean mercury content of 11.5 mg of mercury per tube lamp (Leopold, 2002; PSI, 2008) provides an estimated sales of 11 – 12 million tube lamps in the Study Area annually from 2001 – 2007. Comparing these numbers to annual recycling totals (Newman, 2010), assuming a four-year span between production and disposal, results in a 32% recycling rate by 2008 (see Appendix H for supporting information and calculations). Because it is likely that 100% of disposed tubes are broken, applying a 32% recycling rate to the 2007 total (0.14 t of mercury when scaled to the Study Area), results in 0.1 t/yr of mercury released through fluorescent lamp disposal (Table 39).

Table 39. Estimates of Mercury Release from Fluorescent Lamp Tube Disposal.

Quantity of Mercury in Fluorescent Lamp Tubes Sold/Retired Annually in Study Area (t/yr)	Quantity of Mercury in Fluorescent Lamp Tubes Recycled Annually in Study Area (t/yr)	Quantity of Mercury in Fluorescent Lamp Tubes Disposed Annually in Study Area (t/yr)
0.14	0.04	0.1
	Total Estimate:	0.1

Uncertainty

The total amount of mercury used in lamps for Washington State reported by IMERC (Wienert, 2009) comes directly from lamp manufacturers and is considered reasonably certain since manufacturers have the best information on mercury content and sales of lamps, and are required by law to report to IMERC. The recycling rate has the potential to greatly affect the amount available for release. Points of possible error in the recycling rate include the assumption that each bulb has a four-year lifespan and that the total number manufactured in one year will equal the total disposed of four years later. Furthermore, the mean amount of mercury estimated in each lamp (11.5 mg) may differ from the actual mean. Scaling by population may also present some error as consumption rates in the Study Area may be different from the U.S. overall, and the recycling rate of the Study Area may differ from the state overall.

Furthermore, this estimate treats all mercury lamp materials as tube lamps, applying a mercury per lamp factor for tube lamps to the total mercury used in all lamps, and applying a weight-per-bulb ratio for tube lamps to the total weight of all mercury lamps collected as reported by lamp recyclers, though tube lamps represent only 75% of the mercury lamp market (IMERC, 2009). As recyclers combine various types of bulbs and report a combined weight of all materials collected, it is not possible to estimate the number of compact fluorescent lamps (CFLs) collected versus tube lamps or other types of bulbs. Thus, this estimate treats all sales and all collections as tube lamps, though the actual lamp type may differ.

Increased promotion of energy-efficient fixtures has led to greater use of CFLs. The increased use of CFLs may result in even higher consumption of mercury bulbs, as evidenced by the lack of decrease in total mercury used in lamps over the last several years (Wienert, 2009) despite the reduction in mercury per bulb (PSI, 2008).

The amount of mercury released from broken fluorescent bulbs into the atmosphere has been estimated to be 17-40% within two weeks, with one-third of the release occurring during the first eight hours (Aucott et al., 2004). The remainder becomes homogenized in the waste material, from which it is emitted over time (Lindberg et al., 2005). It is likely that 100% of the disposed bulbs are broken.

Estimate of Mercury Released from Household Fever Thermometers

Using two different estimates of the number of broken thermometers and assuming that each thermometer contained 0.5 g of mercury, the 2003 Mercury CAP estimated that 0.005 – 0.14 tons of mercury were released via broken thermometers statewide each year (Ecology and WDOH, 2003).

At the time of the Mercury CAP publication, many retailers had voluntarily stopped selling mercury thermometers, and all sales were banned in 2006 under MERA. The Mercury CAP provides a statewide baseline total of roughly 1.5 million thermometers in 2003, estimated by adding the existing number of thermometers in households in 2001 (976,701) to 272,568 additions (545,136 sold, minus 50% broken) in both 2002 and 2003. Applying the Mercury CAP estimate of 0.5 g of mercury per thermometer to this number results in 0.76 t of mercury remaining in household fever thermometers as of 2003.

Ecology reported that from 2003 – 2009, 1.38 t (3,050 lb) of mercury was recovered in thermometers (Ecology, 2010b). While this value includes thermostats as well as thermometers, the number of thermostats is estimated to be negligible, as the total thermostat mercury estimated to be recovered via this collection stream is only 2 - 8 kg, a minimal portion of the total 1.38 t reported (see discussion of King County thermostat collection in previous sub-section on thermostats). Thus, from 2003 – 2009, 1.38 t, nearly twice the 2003 in-use estimate of 0.76 t, was collected.

It is unknown exactly how many thermometers may remain in use and how many have been collected; however, it seems reasonable to assume that some households continue to use mercury thermometers. Of the remaining mercury, it is likely that some is released via broken thermometers. However, because the number is uncertain, the lower Mercury CAP estimate of less than 5 kg a year is used as a continued release estimate (Table 40).

Table 40. Estimate of Mercury Release from Household Fever Thermometers.

Quantity of Mercury Collected In Thermometers in Washington, 2003 – 2009 (t)	Mercury Release in Study Area (t/yr)
1.38	0.005
Total Estimate:	0.005

Uncertainty

It is uncertain how many thermometers remain in use and how many are disposed of/broken each year. Furthermore, the estimate of the total mercury collected relies on rough conversions from the total weight of thermometers and thermostats collected, including shipping containers (Ecology, 2010b). The Mercury CAP estimates rely on a per thermometer mercury content of 0.5 g, but the amount of mercury per thermometer can range from 0.5 to 54 g (IMERC, 2010).

Estimate of Mercury Released from Crematoria

The Mercury CAP estimated a release rate of 1 gram of mercury to air per cremation, resulting in an estimated statewide mercury release of 0.026 t during the early 2000s based on a cremation rate of 59% during 2000 (Ecology and WDOH, 2003). Cremation rates have since risen to nearly 70%, with the Cremation Association of North America (CANA, 2008) reporting 32,272 cremations in Washington State during 2008 (rate of 67%), and WDOH reporting nearly the same rate (33,155 cremations, or 68%). Applying the 1 g/cremation emission rate used in the Mercury CAP to the number of cremations in Washington, scaled down by population to the Study Area, results in 0.022 t of mercury emissions during 2008.

A second estimate is provided by EPA in their estimation of annual mercury release nationwide from cremations (3 t during 2005; EPA, 2010). The national estimate scaled to Washington State by percent of nation-wide cremations (3.65%), then further down to the Study Area by population, yields an estimated release of 0.07 t.

Table 41 summarizes the two release estimates of annual mercury release from crematoria. The mid-point of the range (0.02 – 0.07 t/yr) is 0.05 t/yr.

Table 41. Estimates of Mercury Release from Crematoria.

Information Source	Basis for Estimate	Mercury Release in Study Area (t/yr)
Mercury CAP, CANA	21,672 cremations/yr (Study Area) x 1 g mercury/cremation	0.02
EPA	3 t/yr mercury released in U.S. scaled to Study Area by population	0.07
	Total Estimate:	0.02 – 0.07 0.05 (mid-point)

Mercury CAP: Mercury Chemical Action Plan (Ecology and WDOH, 2003)
CANA: Cremation Association of North America (CANA, 2008)
EPA: U.S. Environmental Protection Agency, 2010b

Uncertainty

Better emission factors would vastly improve the estimate for mercury released from crematoria. The estimates provided here may under-estimate actual amounts released since (1) the Mercury CAP notes that ranges of less than 1 to more than 5 grams of mercury per cremation were found, and (2) the United Kingdom and Sweden estimated that 11 to 32% of their mercury emissions came from crematoria, respectively (Ecology and WDOH, 2003). The state cremation rate may also differ from that of the Study Area.

Estimate of Mercury Released from Disposal of Auto Convenience Switches

The 2003 Mercury CAP estimated that 0.115 t of mercury in auto switches entered auto scrap each year statewide (Ecology and WDOH, 2003). From 2006 to 2010, 0.134 t of mercury was recovered from auto scrappers in Washington State, yielding an annual mean of 0.027 t, a 23% recovery rate when compared to the CAP disposal estimate (ELVS, 2010). The remainder (0.09 t statewide, 0.06 t for the Study Area) presumably ends up in the waste stream delivered to electric arc furnaces (EAFs).

A second estimate was derived using an industry recovery rate of 40-50% (Jan Brydsen, Ecology Auto Recycling Specialist, personal communication), although no mass of recovered mercury was provided. Assuming the recovered mass reported by ELVS (2010) represented a 40-50% recovery rate, an estimated 0.02 – 0.03 t/yr is unrecovered. Table 42 summarizes the estimated mercury entering the waste stream.

Table 42. Estimates of Mercury Release from Disposal of Auto Convenience Switches.

Basis for Estimate	Annual Quantity of Mercury Retired in Auto Convenience Switches in Study Area (t/yr)	Annual Quantity of Mercury Recovered in Auto Convenience Switches in Study Area (t/yr)	Annual Quantity of Mercury from Auto Convenience Switches Entering the Waste Stream in Study Area (t/yr)
Mercury CAP	0.077	0.018	0.06
Jan Brydsen, personal communication/ ELVS, 2010	0.036 – 0.045	0.018	0.02 – 0.03
		Total Estimate:	0.02 – 0.06 0.04 (mid-point)

Mercury CAP: Mercury Chemical Action Plan (Ecology and WDOH, 2003)

Uncertainty

Some of the mercury from auto convenience switches is likely released at shredding facilities, of which there are two in the Study Area, but the amount is unknown. It is also unknown what proportion of cars recycled statewide is disposed of at Study Area shredders, and thus the scaling factor (based on population) may not be appropriate. There is one EAF in the Study Area, but the amount of auto scrap processed there is also unknown. The shredding industry estimates that in Washington one-half to one-third of their auto scrap is sent to the EAF depending on market prices (Pinky Feria, Ecology Hazardous Waste and Toxics Reduction Program, written communication). Shredders from outside of the Study Area also likely send scrap to the EAF facility, but again, it is unknown how much. It has been estimated that one-third of the mercury in EAF feedstock enters the atmosphere (Cerreno et al., 2002); mercury emissions from such facilities are presumably reported to the TRI.

The sale of new cars containing mercury switches is prohibited in Washington. The state also encourages removal by paying $3.00 for each recovered switch.

Estimate of Mercury Released from Button Cell Battery Disposal

The Mercury CAP estimated that 0.040 t of mercury in button cell batteries was disposed of statewide during 2001 (Ecology and WDOH, 2003). From 2003 to 2009, 0.061 t of mercury in such batteries was recovered statewide, yielding an annual mean recovery of 0.009 t (Ecology, 2010b), a 22% recovery rate when applied to the Mercury CAP annual disposal mean.

IMERC data indicate a downward trend in the amount of mercury used in batteries over the last decade, from 0.058 t statewide during 2001, to 0.041 t during 2007 (Wienert, 2010). It is unclear if this reduction stems from a reduction in the number of batteries manufactured overall, or a reduction in the amount of mercury used per battery.

An estimated rate of disposal was calculated by using the most recent IMERC value for mercury in battery use (0.041 t/yr) and subtracting the recovery rate reported by Ecology (0.009 t/yr), yielding a statewide disposal estimate of 0.032 t/yr. Scaled to the Study Area by population, this yields an estimated mercury release of 0.02 t/yr (Table 43).

Table 43. Estimates of Mercury Release from Button Cell Battery Disposal.

Annual Quantity of Mercury Used in Button Cell Batteries in Study Area (t/yr)	Annual Quantity of Mercury Recovered from Button Cell Batteries in Study Area (t/yr)	Annual Quantity of Mercury from Button Cell Battery Disposal in Study Area (t/yr)
0.027	0.006	0.02
	Total Estimate:	0.02

Uncertainty

The amount of mercury used in batteries may be considered fairly accurate, though the disposal rate may not equal production as assumed.

Estimate of Mercury Released from Excretion and Disposal of Dental Amalgam

Mercury released from dental amalgam excretion was estimated using a rate of 17.2 ug mercury/day from adults (defined as age 20 or over) reported by the Association of Metropolitan Sewage Agencies (AMSA, 2000). Applying this excretion rate to the current Study Area population, reduced to 74% to exclude children and teens (Ecology and WDOH, 2003; Conway and Associates, 2006) results in a mercury release of 0.020 t/yr to the Study Area (Table 44).

The 2003 Mercury CAP estimated that over 0.183 t of mercury was generated by dental facilities each year (Ecology and WDOH, 2003). From 2003 to 2009, 1.159 t of mercury was recovered from dental facilities, for an annual mean of 0.166 t (Ecology, 2010b). Based on the Mercury CAP estimate of mercury generation, this represents a 90% recovery rate.

Comparing the 2009 collection rates to the total mercury sold in amalgams during 2007 (0.218 t) (Wienert, 2009) suggests a recovery rate of 103%. However, other studies have shown that even with best management practices, 4-5% of the mercury in dental offices continues to escape down the drain (Maria Peeler, Ecology Hazardous Waste and Toxics Reduction Program, personal communication; Fan et al., 2002). Estimates of mercury disposed from dental offices were derived from the 2007 sales volumes for the Study Area and a 4-5% loss rate, resulting in an estimated 0.01 t of mercury release (Table 44).

Table 44. Estimates of Mercury Release through Dental Amalgam Excretion and Disposal.

Release Type	**Mercury Release Rate (ug/day/person)**	**Study Area Population**	**Fraction of population ≥ 20 years of age**	**Mercury Sold in Amalgams During 2007 in Study Area (t)**	**Rate of Amalgam Loss in Dental Office Drains**	**Mercury Release in Study Area (t/yr)**
Excretion	17.2	4,475,000	0.74	--	--	0.02
Disposal	--	--	--	0.218	4-5%	0.01
					Total Estimate:	0.03

Uncertainty

Much lower excretion rates were suggested by exposure values reported by Richardson (1995), but a correlating release rate was not found. Mercury amalgam loss rates in dental offices may under-estimate actual releases since loss rate estimates (4-5%) may be based on facilities in high compliance areas, such as King and Snohomish Counties, which strictly adhere to best management practices. Dental offices outside of high compliance areas may have higher amalgam loss rates (Maria Peeler, Ecology Hazardous Waste and Toxics Reduction Program, personal communication).

Estimate of Mercury Released from Fertilizer Application

Estimates of mercury released from fertilizers were based on the reported concentrations in fertilizers and amounts distributed in the state annually, then scaled to the Study Area based on estimated portions of crop area. Details are included in the section describing arsenic releases from fertilizers.

Total mercury releases are shown in Table 45. In general, mercury releases corresponded with the amount of each material distributed. The total mercury release was estimated to be 0.002 t/yr.

Table 45. Estimates of Mercury Release from Fertilizer.

Material	Material Distributed in Washington During 2006-2007 Reporting Year (t)	Mercury Release in Washington (t/yr)	Mercury Release in Study Area (t/yr)
Nitrogen Material	618,550	0.035063	0.000573
Phosphate Material	115,474	0.013472	0.000220
Potash Material	94,161	0.028123	0.000459
Miscellaneous Fertilizer	163,652	0.018008	0.000294
Sulfur	25,954	0.001134	0.000019
Gypsum	38,409	0.003175	0.000052
Natural Organics	32,866	0.001361	0.000022
Liming Material	55,579	0.005715	0.000093
Total as Fertilizer	**1,144,645**	**0.106**	**0.002**
		Total Estimate:	0.002

Uncertainty

The largest source of uncertainty may be the assumption that fertilizer application practices within the Study Area are identical to other regions of the State. An annual estimate of mercury release also assumes that fertilizer use during the 2006-2007 reporting year is representative of current use. The validity of these assumptions has not been assessed. Also, reported concentrations may over-estimate actual concentrations since WSDA used detection limits to substitute for non-detected results.

Other Potential Sources of Mercury Release

Manometers

Prior to the publication of the 2003 Mercury CAP, an effort to remove manometers from dairies resulted in 0.025 t of mercury collected statewide, representing most of the manometers used in dairies. Other users of manometers include water systems, and 1.7 t of mercury was recovered from manometers and barometers throughout the state during 2003 – 2009. It is unknown how many manometers may remain in use.

Artisan Mining

Statewide, 0.212 t of mercury has been collected from artisan miners (using placer mining techniques) since 2006. Gold deposits have been documented within the Study Area (Ecology and WDOH, 2003), and small-scale placer mining may be releasing mercury into the environment. However, it is unknown how much remains uncollected and how much is released into the environment or where it may be released (Diana Olegre, Ecology Hazardous Waste and Toxics Reduction Program, personal communication).

Medical Waste Autoclaves and Retorts

The Mercury CAP provided a highly uncertain estimate that 0.048 t mercury amalgam improperly disposed of as “red bag” infectious medical waste was treated at medical autoclaves or retorts (Ecology and WDOH, 2003). Since the Mercury CAP, both Ecology and DOH have undertaken outreach efforts to educate hospitals and clinics, and Ecology has collected 0.122 t of such mercury, with 0.045 t being collected in 2009 alone.

Estimate of Mercury Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Industrial facilities reported total mean annual mercury (and mercury compound) releases of 0.096 t/yr for the five TRI categories assessed (Table 46). The highest overall reported mercury releases were from cement plants, petroleum refineries, steel producers, and paper mills.

The Lafarge North America cement plant in King County was the largest single discharger of mercury and mercury compounds, with an annual mean release of 0.024 t/yr reported in stack air emissions. The BP Cherry Point Refinery in Whatcom County had the single largest fugitive air emissions of mercury (0.0002 t/yr), accounting for approximately two-thirds of the fugitive air releases.

The Georgia Pacific pulp mill in Whatcom County had some of the highest historical air emissions (both fugitive and stack), including a fugitive air release of 0.23 tons reported during 1999, but the mill discontinued reporting after 2000 and was permanently shuttered soon afterwards (data from this facility were excluded from the release calculations here). The City of Tacoma Steam Plant, a municipal waste incinerator, historically had some of the highest reported stack emissions of mercury, but the plant shut down soon after its last reporting period in 2001 (data from this facility also excluded here).

The Kimberly-Clark paper mill in Snohomish County reported nearly half of the mercury release to water (0.002 t/yr), and the waste processor, Burlington Environmental (King County) was responsible for all of the reported releases of mercury to POTWs during 1999-2008 (0.0001 t/yr). The Shell Oil Skagit County refinery accounted for nearly all of the mercury released in the “other disposal” category (0.014 t/yr), with the remainder made up by the Kimberly-Clark facility in Snohomish County (0.001 t/yr).

One of the largest mercury emitters is Washington’s lone coal-fired power plant, the TransAlta Centralia Generating plant. TRI data show that in 2008 and 2009 this plant emitted 0.142 t and 0.0161 t mercury, respectively. Although it is not included as a source because its location falls outside of the Study Area boundary, it is upwind of the Study Area and at least two nearby lakes in the Study Area have sediment mercury levels that appear to reflect deposition from the plant (Furl and Meredith, 2010).

Table 46. Estimates of Mean Annual Mercury Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 1999-2008.

Release by Facility Type	**Percent of Total**	**Mercury Release in Study Area (t/yr)**
Cement Plants	42%	0.040
Petroleum Refineries	35%	0.033
Steel Mills	16%	0.016
Pulp and Paper Mills	6%	0.0060
All Others	<1%	0.0006
Total	100%	0.096
Release by Medium	**Percent of Total**	**Mercury Release in Study Area (t/yr)**
Fugitive Air Emissions	<1%	<0.001
Stack Air Emissions	79%	0.076
Surface Water Discharge	4%	0.0040
Other On-Site Land Disposal	16%	0.015
Discharge or Transfer to POTWs	<1%	<0.001
Total	100%	0.096
Total Estimate:		0.096

Uncertainty

Criteria and limitations of TRI reporting requirements are in Appendix C.

Estimate of Mercury Released from Modeled Air Emission Sources

Air emissions of mercury inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Point source air emissions account for the bulk of releases (0.050 t/yr), but the exact source was not identified in the Air Emissions Inventory report (Table 47). Smaller releases of mercury (0.007 t/yr) were due to combustion of distillate oil (heating oil) as a residential heating source. Total mercury emissions were 0.057 t/yr.

Examination of the TRI data and a county-by-county breakdown of the Air Inventory data suggest that some of the air emissions data have been double-counted, but the exact amount is unknown. Air emissions of mercury were highest in Skagit, King, and Whatcom Counties which correspond with individual facilities or facility types reporting high mercury stack emissions under TRI.

Table 47. Estimates of Mercury Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	Mercury Release in Study Area (t/yr)
Point Sources - Industrial, Commercial, Institutional (primarily Title V AOP)	0.050
Residential Fuel Use, except Wood	0.007
Total Estimate:	0.057

AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

Uncertainty

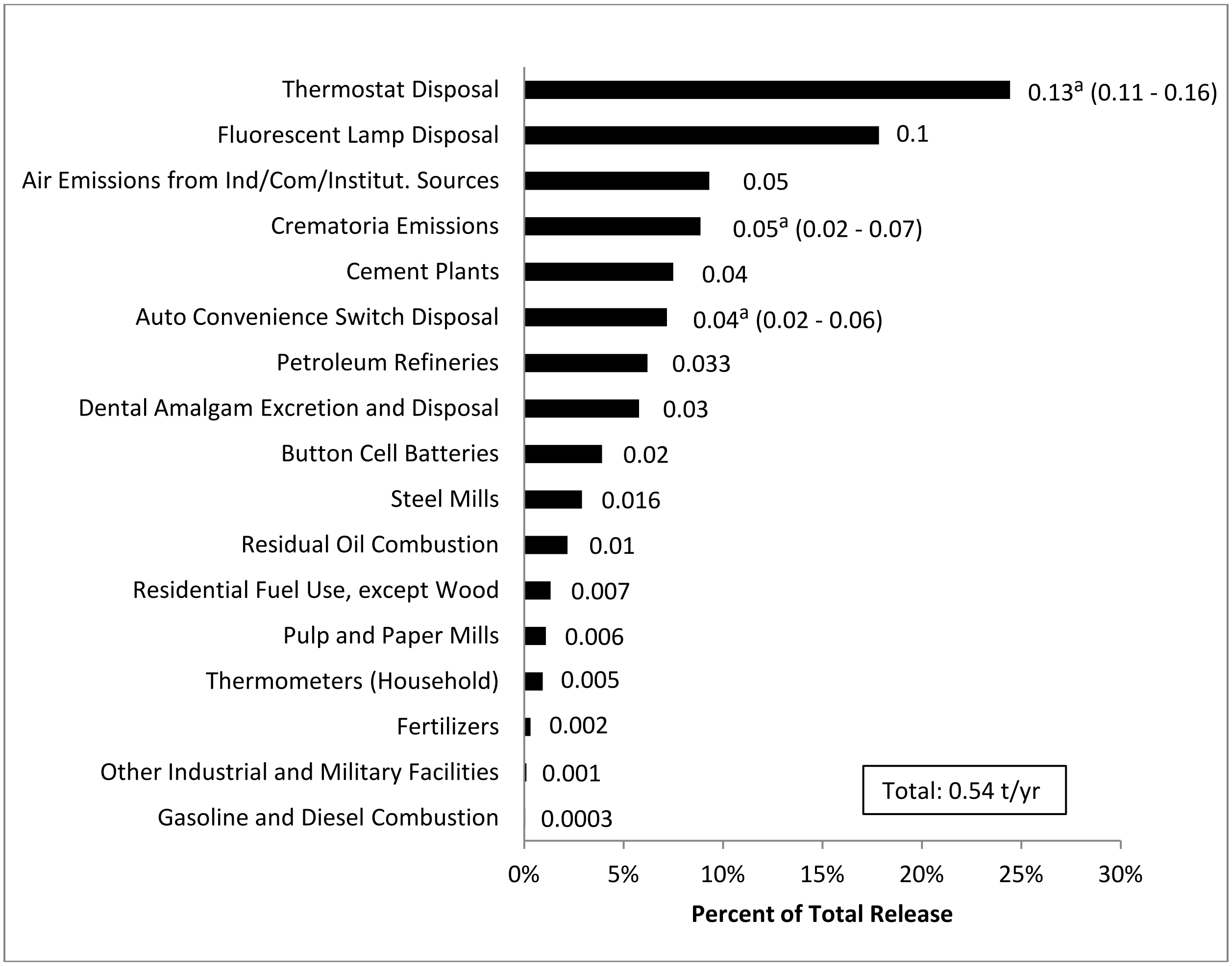
Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of Mercury Release Estimates

Based on the best estimates available, total mercury releases from the sources assessed are approximately 0.54 t/yr (Figure 7). The largest source of mercury may be due to thermostat and fluorescent lamp disposals (24% and 18%, respectively).

The combined air emissions from point sources (stationary industrial, commercial, and institutional sources, generally operating under a Title V Air Operating Permit) as categorized in the Air Emissions Inventory (Ecology, 2007a) account for 9% of the total. Air emissions from major industrial facilities reporting under TRI represent approximately 18% of the total mercury release, and four-fifths of these are stack air emissions. In addition, some of the residual fuel oil emissions may be due to industrial boilers as well as ships. Therefore, the industrial mercury emissions were likely double-counted, suggesting the total releases counted (among sources considered) may over-estimate actual values.

No estimates of error or ranges of possible values were available for most of the release estimates. Therefore, the narrow range of possible values for the total anthropogenic mercury release in the Study Area (0.5 – 0.6 t/yr) should not be construed as a high degree of precision in the estimates. This is particularly true for estimates of mercury releases based on disposal rates of consumer goods, where numerous assumptions were used.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 7. Total Mercury Release in the Study Area (values shown are t/yr).

Zinc

Sources Assessed

Assessment of ongoing anthropogenic zinc releases included the following sources:

- Roofing material runoff
- Vehicle tire wear
- Fertilizer and micronutrient use
- Plumbing fixtures, pipe, and solder leaching
- Petroleum leaks, spills, and improper disposal of oil
- Vehicle brake pad wear
- Military, industrial, commercial, and institutional facilities

Other possible sources of zinc release include releases to marine waters from sacrificial anodes, leaching from guard rails, leaching from streetlight and road sign standards, leaching from chain link fences and other galvanized materials, and use of zinc salts for moss control on rooftops. Releases from these sources were not assessed.

Estimate of Zinc Released from Roof Runoff

Zinc is a common constituent of nearly all roofing materials and is readily leached, particularly in acidic rainwater. Zinc runoff data are available for virtually all roof types since zinc is one of the most commonly analyzed constituents of stormwater runoff. However, the various studies reviewed had different objectives, thus the data are not directly comparable. In some cases, researchers sampled experimental panels with controlled variables. In other cases, runoff from actual roofs was sampled. Sample size and the duration of the sampling also varied among studies.

Many of the sample data reflect not only zinc contributions from the roofing material discussed below, but also gutters, vents, flashing, and other components typically found as part of a roof. In order to present a full range of potential release values, reported averages for each roof type were used, with means or medians for specific roof types providing both the high and the low values for that particular type, with all reported means or medians averaged to derive the overall mean value. The intention was to provide as complete a picture of roofing releases as possible, while not obscuring any of the specific roofing sources.

Mean zinc concentrations in asphalt shingle roof runoff were 1,330 ug/l, with means or medians ranging from 318 – 2,330 ug/l (Chang and Crowley, 1993; Steuer et al., 1997; Chang et al., 2004). Although they did not correct for zinc in rainfall, Chang and Crowley (1993) reported a mean zinc concentration of 980 ug/l in East Texas rainwater, higher than values recorded in other areas of the country. While elevated regional zinc levels may be contributing factors in the higher concentrations of runoff reported by Chang and Crowley (1993) and Chang et al. (2004) for asphalt and wood shingles, the values reported by Chang et al. (2004) for aluminum roofing (514 – 16,600 ug/l) were similar to those reported by both Clark (2010) (30 – 16,929 ug/l) and

Tobiason (2004) (420 – 14,700 ug/l) in other regions of the country. This indicates that high atmospheric zinc deposits alone do not explain the comparatively high levels reported for wood and asphalt in the Texas-based studies.

In Chang et al. (2004) and Chang and Crowley (1993), as well as in studies of zinc runoff from residential areas (e.g. Bannerman et al., 1993; Steuer et al., 1997; Davis et al., 2001), runoff samples collected for analysis may have contacted galvanized flashing or been channeled through gutters made of galvanized material, potentially enriching sample zinc concentrations. Runoff results from Chang et al. (2004) may have been particularly influenced by contributions from a galvanized gutter system installed as part of their experimental panel set-up. The area of gutter to roof would be much larger on the test panels than on a real roof, perhaps explaining some of the increase seen over reports from residential areas (Bannerman et al., 1993; Steuer et al., 1997; Davis et al., 2001). The higher proportion of gutter area to roof area does not fully explain the elevated concentrations reported by Chang et al. (2004); however, as the Chang and Crowley (1993) samples had similar concentrations and were taken from actual roofs.

Runoff from built-up roofing appears to have lower zinc concentrations, with a mean of 221 ug/l (means or median ranged from 93.7 – 348 ug/l) reported by Clark (2010) and Steuer et al. (1997). These concentrations are lower than zinc in runoff isolated to commercial areas reported by Bannerman et al. (1993) and Davis et al. (2001) (mean = 706 ug/l), but as mentioned previously, disparities in zinc concentrations may be driven by exposure of runoff to galvanized metal. In a study of zinc from various industrial roof types in western Washington, Golding (2006) found that the runoff from roofs with galvanized ducting or downspouts contained zinc at twice the concentration of runoff from roofs with little or no galvanized materials (mean conc. = 241 ug/l for galv.; 105 ug/l for non-galv.).

A recent study of contaminants in roadway runoff from the Highway 520 Bridge over Lake Washington illustrates the importance of galvanized metals as a source of zinc in runoff. In this study by King County (2005), zinc was found at very high levels in drainage from the floating bridge (430 – 3,000 ug/l), and was nearly all in the dissolved phase which is unusual for a road runoff study. Further investigation revealed that the high levels of zinc were due to leaching caused by a failure of the galvanizing material in the drain downspouts (which partially explained the high proportion of zinc in the dissolved form), making the study of little use for representing zinc concentrations in road runoff.

Numerous studies have reported high concentrations of zinc in metal roofing materials, and there appears to be as much variability within specific metal roof types as among different types. For instance, zinc in galvanized metal runoff was found to range by three orders of magnitude (124 – 212,000 ug/l) in a study by Chang et al. (2004), and Clark (2010) who, after correcting for atmospherically deposited zinc, found concentrations ranging from 29.8 to 17,000 ug/l in runoff from corrugated aluminum roof panels. Overall, the mean zinc concentrations in runoff from aluminum, Galvalume, and galvanized steel roofs were 4,530 ug/l (mean of Chang et al., 2004 and Clark, 2010), 25 ug/l (Clark, 2010), and 5,200 ug/l (mean of Yaziz et al., 1989; Good, 1993; Tobiason, 2004; Chang et al., 2004), respectively. The overall mean used to calculate release loads from metal roofs was 2,860 ug/l, a mean of all metal roof types.

Other roofing materials with associated zinc runoff data include clay tiles (1,080 ug/l,; Chang and Crowley, 1993), concrete tiles (mean of 94 ug/l; Yaziz et al., 1989), and wood shake/shingle (mean of 7,390 ug/l; Chang and Crowley, 1993; Chang et al., 2004; Clark, 2010). The latter may have elevated concentrations due to impregnation with zinc as biocide.

Annual rooftop runoff volumes for each roof type were used to translate zinc concentrations in runoff into release loads. The method for the calculation of runoff volumes is described in Appendix B. Total annual releases loads for wood shake/shingle, metal, asphalt shingle, built-up, clay tile, and concrete tile roofs were 400, 120, 780, 23, 0.5, and 2.1 t/yr, respectively (Table 48). The combined load was 1,300 t/yr.

Table 48. Estimates of Zinc Release from Roofing Material.

Roof Type	**Total Area of Roof Type in Study Area (m^2)**	**Total Runoff Volume of Roof Type in Study Area (l/yr)**	**Zinc Concentrations in Runoff (ug/l)**	**Zinc Release in Study Area (t/yr)**
Wood Shake/Shingle	3.62×10^7	5.45×10^{10}	201 – 16,317 7,390 (mean)	11 – 890 400 (mean)
Metal	2.96×10^7	4.29×10^{10}	24.6 – 11,788 2,860 (mean)(a)	1.1 – 510 120 (mean)
Asphalt Composite	3.96×10^8	5.84×10^{11}	318 – 2,330 1,340 (mean)	190 - 1,400 780 (mean)
Built-Up	7.40×10^7	1.03×10^{11}	93.7 – 348 221 (mean)	9.6 – 36 23 (mean)
Clay Tile	3.03×10^5	4.50×10^8	1,080	0.49
Concrete Tile	1.59×10^7	2.23×10^{10}	48 – 193 94 (mean)	1.1 – 4.3 2.1 (mean)
			Total Estimate:	210 – 2,800 1,300 (mean)

t: metric ton

(a) Based on runoff concentrations from aluminum, Galvalume, and galvanized steel roof materials

Uncertainty

As with roofing releases reported for other metals, the greatest degree of uncertainty appears to be related to leaching data. Leaching data from controlled experiments and with regionally relevant environmental conditions would provide more appropriate data than data currently available, and would greatly improve accuracy of the calculated release loads. Roofing age, unaccounted for here, may also play a factor, as release rates may vary over the life of a roof.

Zinc concentrations in roof runoff are largely influenced by various components of the roofing system. Data presented here reflect representative zinc concentrations from types of roofs, not necessarily the zinc leached from isolated roofing material. Therefore, the usefulness of these data depends on their application. The data appear to be relevant for evaluating zinc roof runoff across a large area. However, if data on zinc concentrations leached from specific materials are needed, these data have limited applicability.

Furthermore, the values reported by Clark (2010) for specific roofing materials (absent of any additional components), and corrected for atmospheric deposition, possibly suggest that some materials actually retain contaminants in some storm events, and release them in others. Other researchers also note that the intensity and length of a storm, as well as the time between storms will have a large impact on the overall runoff value; sample concentrations for the same roofs can vary greatly, as evidenced by the large ranges reported in Chang and Crowley (1993).

While combining data from various studies offers an indication of the range of potential releases, the actual mean runoff values for any given roof type may be different than what has been calculated here. Also, adjustments based on regional conditions could be made – for instance, by selecting only data with runoff matching Puget Sound regional pH, or by correcting for regional atmospheric deposition flux – but without a thorough evaluation of the appropriate correction factors, adjustments may simply result in additional layers of uncertainty without improving accuracy.

Notwithstanding the shortcomings previously described, the amount of zinc runoff and leaching data in the literature, along with the variety of roof types for which these data are available are useful to gauge the degree to which zinc is released from rooftops. The scale-up information used to calculate zinc releases from roof materials appears to be fairly reliable. Like other calculated estimates of roofing release loads, accuracy would be increased by further collection of information on roof type fractions from counties in addition to Pierce and Snohomish Counties. Estimates of zinc releases would also greatly benefit from more detail about specific roof materials documented by counties and other local jurisdictions (e.g., describing a roof as corrugated aluminum or galvanized steel, rather than simply as “metal”).

Estimate of Zinc Released from Vehicle Tire Wear

Releases of zinc due to vehicle tire wear were calculated using estimates of tire wear rates and zinc concentrations reported in tire material, then scaled to the Study Area using annual vehicle kilometers travelled (VKTs) for the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Table 49 shows the variables used to calculate zinc release from tire wear. A wear rate of 38 mg/km travelled was assumed for all vehicle types considered, with differences being only the number of tires per vehicle. Zinc concentrations in tire material are higher than other metals, with a mean concentration of 7,434 mg/kg. Estimates of total zinc release from tire wear are 4.8 to 150 t/yr, with a mean of 82 t/yr. The estimate of mean zinc release is slightly different than that shown in Appendix E (77 t/yr) since the latter estimate is based on a median annual zinc release rather than an arithmetic mean. However, the arithmetic mean is shown here to remain consistent with methods employed to obtain tire wear release estimates for other metals.

Table 49. Estimates of Zinc Release from Vehicle Tire Wear.

Vehicle Type	Number of Tires per Vehicle	Tire Wear Rate (mg/km/tire)	Zinc Concentration in Tire Material (mg/kg)	Vehicle Kilometers Travelled in Study Area (km/yr)	Zinc Release in Study Area (t/yr)
Motorcycle	2	38	430 – 13,484 7,434 (mean)	2.3E+08	0.008 – 0.24 0.13 (mean)
Passenger Car	4			3.6E+10	2.4 – 74 41 (mean)
Light Truck	4			1.5E+10	0.97 – 31 17 (mean)
Bus	8			1.2E+08	0.015 – 0.47 0.26 (mean)
Single Unit Truck	8			2.8E+09	0.36 – 11 6.3 (mean)
Combination Truck	18			3.5E+09	1.0 – 32 18 (mean)
				Total Estimate:	4.8 – 150 82 (mean)

Uncertainty

Overall estimates of tire wear rates, zinc concentrations in tire material, and VKTs appear to be reliable since the estimates are based on published information and WSDOT data (see Appendix E). Tire wear rates reported in the literature appear to be realistic when checked against what may be considered a reasonable tread loss over the life of a tire (see Appendix E for this analysis). However, both the tire wear rates and zinc concentrations may vary considerably based on tire brand, with wear rates also strongly influenced by driving conditions. Although the VKTs are based on the 12-county Puget Sound region, they are a reasonable proxy for the Study Area.

Estimate of Zinc Released from Fertilizer and Micronutrient Application

Estimates of zinc released from fertilizers and micronutrients were based on the reported concentrations in fertilizers and amounts distributed in the state annually, as well as the reported amounts of micronutrients, and then scaled to the Study Area based on estimated portions of crop area. Details are included in the section describing arsenic releases from fertilizers and copper releases from micronutrients.

Total zinc releases from these sources are shown in Table 50. Zinc release from micronutrient use accounted for approximately 91% of the total release from this source. The total zinc release was estimated to be 41 t/yr.

Table 50. Estimates of Zinc Release from Fertilizer and Micronutrients.

Material	Material Distributed in Washington During 2006-2007 Reporting Year (t)	Zinc Release in Washington (t/yr)	Zinc Release in Study Area (t/yr)
Nitrogen Material	618,550	3.049	0.050
Phosphate Material	115,474	172.0	2.810
Potash Material	94,161	1.337	0.022
Miscellaneous Fertilizer	163,652	32.85	0.537
Sulfur	25,954	3.556	0.058
Gypsum	38,409	0.575	0.009
Natural Organics	32,866	8.059	0.132
Liming Material	55,579	1.962	0.032
Total as Fertilizer	**1,144,645**	**223.4**	**3.650**
Total as Micronutrients	**2,257**	**2,257**	**36.887**
		Total Estimate:	41

Uncertainty

The largest source of uncertainty may be the assumption that fertilizer and micronutrient application practices within the Study Area are identical to other regions of the State. An annual estimate of zinc release also assumes that fertilizer use during the 2006-2007 reporting year is representative of current use. The validity of these assumptions has not been assessed. Also, reported concentrations may over-estimate actual concentrations since WSDA used detection limits to substitute for non-detected results.

Estimate of Zinc Released from Residential Plumbing

Zinc may be released from residential plumbing through the use of brass fittings and galvanized piping. Water supply lines may also contain brass fittings and galvanized piping.

Recent tap water sampling conducted by WDOH for LCR compliance was used to calculate copper and lead releases from residential plumbing. However, this sampling effort did not include results for zinc at the tap. In order to estimate zinc releases from residential plumbing, data from LCR compliance sampling in California were used to represent zinc concentrations at the tap.

In a study characterizing lead, copper, zinc, and other metals in water reclamation plant influent, Kimbrough (2009) evaluated data collected for LCR compliance in six southern California water utility service areas during 1998 through 2006. Source water for the service area had a pH above 7.0 and "… often above 8.0 …", and therefore did not appear to have conditions which would promote accelerated or extreme corrosion of plumbing components. Residences sampled for LCR compliance appeared to have a typical mixture of plumbing components: plastic (PVC) piping and fittings, copper piping, brass fittings, and fixtures, galvanized piping, solder with high and low lead content, and possibly lead piping. Therefore, these residences may be representative of both newer and older houses.

Thirty-six separate sample data populations were reported by Kimbrough (2009) for the LCR compliance samples collected at bathroom and kitchen taps. Median zinc values for the data were reported for the four years during which they were sampled (1998, 2003, 2005, and 2006), and an overall median was reported for the entire data compilation; individual data were not published. The overall median for zinc in tap water was 69 ug/l. The lowest median level of zinc was from the 2003 dataset (47 ug/l), while the highest median value was from 2005 (216 ug/l).

Domestic water consumption rates for households described previously for copper and summarized in Appendix F were used to estimate annual loads in the Study Area. Applying the copper concentrations obtained from Kimbrough (2009) and per capita water consumption rates reported by Mayer et al. (1999), indoor plumbing releases an estimated 30 t/yr zinc, with a range of 20 to 93 t/yr (Table 51).

Table 51. Estimates of Zinc Release from Residential Plumbing.

Per Capita Indoor Water Consumption Rate (gal/day)	**Receiving System Type**	**Indoor Water Consumption in Study Area (gal/yr)**	**Indoor Water Consumption in Study Area (l/yr)**	**Zinc Concentration in Tap Water (ug/l)**	**Zinc Release in Study Area (t/yr)**
69.3	POTWs	8.12×10^{10}	3.07×10^{11}	47 – 216[a] 69 (median)[b]	14 – 66 21 (median)
	Septic	3.20×10^{10}	1.21×10^{11}		5.7 – 26 8.4 (median)
Total Estimate:					20 – 93 30 (median)

[a] 25th - 75th percentile

[b] Overall median value among all data assessed

Uncertainty

Releases of zinc from residential plumbing may vary depending on chemistry of the domestic source water and treatment by the water utility prior to distribution. Zinc concentrations used for release estimates were from households in southern California. While there was no indication that source water for these households was particularly aggressive or mild in terms of corrosion, leaching, or other factors promoting/preventing zinc release from plumbing components, site-specific water chemistry data for the Study Area would be preferable for calculating releases. Median copper and lead tap water concentrations reported for the Puget Sound region (WDOH, 2011) were lower than those from California reported by Kimbrough (2009). This could suggest that zinc concentrations from the California dataset are comparably high as well, if water chemistry variables promote metals leaching.

LCR compliance data potentially represent higher concentrations of zinc than the overall population within a given water service provider, since the rule requires monitoring of homes and buildings that are at the highest risk of lead and copper contamination (EPA, 2004), and high zinc levels may be associated with elevated copper and lead since they are all components of brass. Therefore, the zinc concentrations used for release estimates may be biased high compared to the overall population of tap water samples. However, Kimbrough (2009) used a concentration of 200 ug/l as a distinction between high and low zinc concentrations, and the median value used to calculate release estimates (69 ug/l) was below this level by a factor of 3, indicating this value was not atypically high for the population sampled.

Study Area water consumption data were based on a national survey conducted in a variety of geographical areas. There is no obvious reason the data would not be representative of the water consumption habits for the Study Area. However, since the survey data are over a decade old, they may over-estimate water consumption rates which may have fallen in recent years due to water conservation education, installment of low-flow fixtures, and use of water-saving appliances such as dishwashers and clothes washers.

Estimate of Zinc Released from Motor Oil Leaks and Improper Disposal of Used Motor Oil

Zinc released through leaks and drips of crankcase oil from vehicles and from improper disposal of used oil following oil changes was estimated by applying information on zinc concentrations in used oil to estimates of oil loss from drips and leakage, and improper disposal of used motor oil.

Zinc is added to engine crankcase oil to reduce engine wear, and nominal concentrations are typically 900 – 1,200 mg/kg, although oil containing up to 2,000 mg/kg may be used in high-performance racing engines (mobiloil.com; valvoline.com). Denton (2004) conducted a review of information on zinc and other chemicals in used motor oil, and found that data published since 1993 reported similar zinc levels in used motor oil (mean concentrations in four studies ranged from 800 to 1,240 mg/kg, with a mean of 1,100 mg/kg).

Motor oil releases to the environment were calculated using the methodology and assumptions discussed in the Petroleum section. The quantities of petroleum released from various sources and associated zinc releases are shown in Table 52. The range of mean zinc concentrations in used oil and the mean of the means reported by Denton (2004) were used to represent zinc content of used oil. The estimated range of zinc loss was 5.7 – 8.9 t/yr, with a mean of 7.9 t/yr (Table 52).

Table 52. Estimates of Zinc Release Due to Motor Oil Leaks and Improper Disposal of Used Motor Oil.

Material	**Zinc Concentration (mg/kg)**	**Release From Leaks and Drips in Study Area (t/yr)**		**Release From Improper Disposal of Used Motor Oil in Study Area (t/yr)**		**Zinc Release in Study Area (t/yr)**
		Petroleum	**Zinc**	**Petroleum**	**Zinc**	
Used Motor Oil	800 – 1,240 1,100 (mean)	6,100	4.9 – 7.7 6.8 (mean)	970	0.78 – 1.2 1.1 (mean)	5.7 – 8.9 7.9 (mean)
					Total Estimate:	5.7 – 8.9 7.9 (mean)

Uncertainty

The range of reported zinc levels in both new and used motor oil appears to be consistent among the limited information reviewed. However, more information to verify these values would be preferable.

Volumes of motor oil released have associated uncertainties. These uncertainties are discussed in detail in the Petroleum section.

Estimate of Zinc Released from Vehicle Brake Pad Wear

Releases of zinc due to vehicle brake pad wear were calculated using estimates of brake pad wear rates and zinc concentrations reported in brake pad material, then scaled to the Study Area using annual VKTs in the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Wear rates for brake friction materials were estimated from various literature values and were found to range from 3 mg/km travelled for motorcycles to 245 mg/km travelled for combination trucks (Table 53). It should be noted that different vehicle types employ different types of braking systems, with motorcycles, passenger cars, and light trucks primarily using disc brakes and heavier vehicles using drum brakes almost exclusively. There are few data on zinc in drum brake friction materials, and these materials have not been investigated extensively since most of the abraded material is retained in the brake drums (Sinclair Rosselot, 2006).

Based on the differences in braking systems among vehicle types, mean zinc releases were calculated only for motorcycles, passenger cars, and light trucks, although maximum releases from other vehicle types are also calculated by assuming maximum zinc concentrations also apply to drum material (Table 53). Mean zinc releases due to brake pad wear are estimated to be 0.22 to 44 t/yr, with a mean of 7.1 t/yr.

Table 53. Estimates of Zinc Release from Vehicle Brake Pad Wear.

Vehicle Type	Brake Pad Wear Rate per Vehicle (mg/km)	Zinc Concentration in Brake Pad Material (mg/kg)	Vehicle Kilometers Travelled in Study Area (km/yr)	Zinc Release in Study Area (t/yr)
Motorcycle	3	270 – 21,800 8,676 (mean)	2.3E+08	<0.001 – 0.015 0.006 (mean)
Passenger Car	16		3.6E+10	0.16 – 13 5.0 (mean)
Light Truck	16		1.5E+10	0.064 - 5.2 2.1(mean)
Bus	110	21,800 (max)	1.2E+08	0.28 (max)
Single Unit Truck	129		2.8E+09	7.8 (max)
Combination Truck	245		3.5E+09	19 (max)
			Total Estimate:	0.22 – 44 7.1 (mean)

Uncertainty

Estimates of average zinc releases due to brake pad wear are highly uncertain due primarily to the difficulty obtaining reliable concentration estimates for drum lining materials. Much of the wear rate uncertainty is due to the wide range of assumptions that are possible for assigning the proportion of disc brake use among various vehicle types. For instance, in the present analysis an inconsequential proportion of light trucks and passenger cars are assumed to use drum brakes. However, Sinclair Rosselot (2006) assumes that approximately one-third of these vehicles have rear drum brakes. If reliable estimates were available for the concentration of zinc in drum lining materials, the proportion of cars with drum brakes could have been considered in the current analysis.

Preferable methods for calculating chemical releases from brake pad wear would consider automobile make/models independently and calculate releases based on the brake pads and VKTs associated specifically with each make/model. However, for each vehicle make/model there are typically a variety of both factory-installed and after-market pads available which likely contain different amounts of metals concentrations, suggesting that data on VKTs by each car make/model may not by itself improve the level of certainty in release estimates. As noted in the analysis provided in Appendix E, other variables such as driving habits and types of roads travelled are also considerations in determining releases from brake pad wear, but a detailed evaluation of these variables is beyond the scope of the present report.

Estimate of Zinc Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Industrial and military facilities reported total mean annual zinc (and zinc compound) releases of 14.8 t/yr for the five TRI categories assessed (Table 54). The highest overall reported zinc releases were steel manufacturers and galvanizers, U.S. Navy (Bremerton PSNS), paper mills, and petroleum refiners.

The Bremerton PSNS had the highest single fugitive air emissions of zinc (1.05 t/yr), while the highest stack emissions were from Nucor Steel in King County (4.34 t/yr). Paper mills accounted for the bulk of zinc discharges to water, with the Kimberly-Clark mill in Snohomish County reporting more than half of zinc release to water (2.55 t/yr) and accounting for all of the zinc released via the “other disposal” category. The Bremerton PSNS had the highest zinc release to POTWs (0.02 t/yr).

RSA Microtech, a Skagit County producer and distributor of micronutrient fertilizers, had substantial releases of zinc to air and POTWs during 2000, its only reporting year. It appears that RSA no longer operates in the Puget Sound area, and zinc release data from 2000 were not included in the calculations presented here. Zinc releases from the Georgia Pacific pulp mill in Whatcom County and the City of Tacoma Steam Plant were also excluded from the calculations.

Table 54. Estimates of Mean Annual Zinc Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 1999-2008.

Release by Facility Type	**Percent of Total**	**Zinc Release in Study Area (t/yr)**
Steel Mills	35%	4.3
Pulp and Paper Mills	30%	3.7
Naval Shipyard	14%	1.8
Petroleum Refineries	9%	1.1
Steel Galvanizers	6%	0.73
All Others	6%	0.77
Total	100%	12
Release by Medium	**Percent of Total**	**Zinc Release in Study Area (t/yr)**
Fugitive Air Emissions	18%	2.2
Stack Air Emissions	45%	5.6
Surface Water Discharge	29%	3.6
Other On-Site Land Disposal	7%	0.87
Discharge or Transfer to POTWs	<1%	<0.12
Total	100%	12
	Total Estimate:	12

Uncertainty

Criteria and limitations of TRI reporting requirements are in Appendix C.

Estimate of Zinc Released from Modeled Air Emission Sources

Air emissions of zinc inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Point source air emissions accounted for all of the releases (12 t/yr), but the exact source was not identified in the Air Emissions Inventory report (Table 55).

Examination of the data on a county-by-county basis and comparisons to TRI-reported stack emissions suggests there is little, if any, double-counting of zinc emissions.

Table 55. Estimates of Zinc Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	Zinc Release in Study Area (t/yr)
Point Sources - Industrial, Commercial, Institutional (primarily Title V AOP)	12
Total Estimate:	12

AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

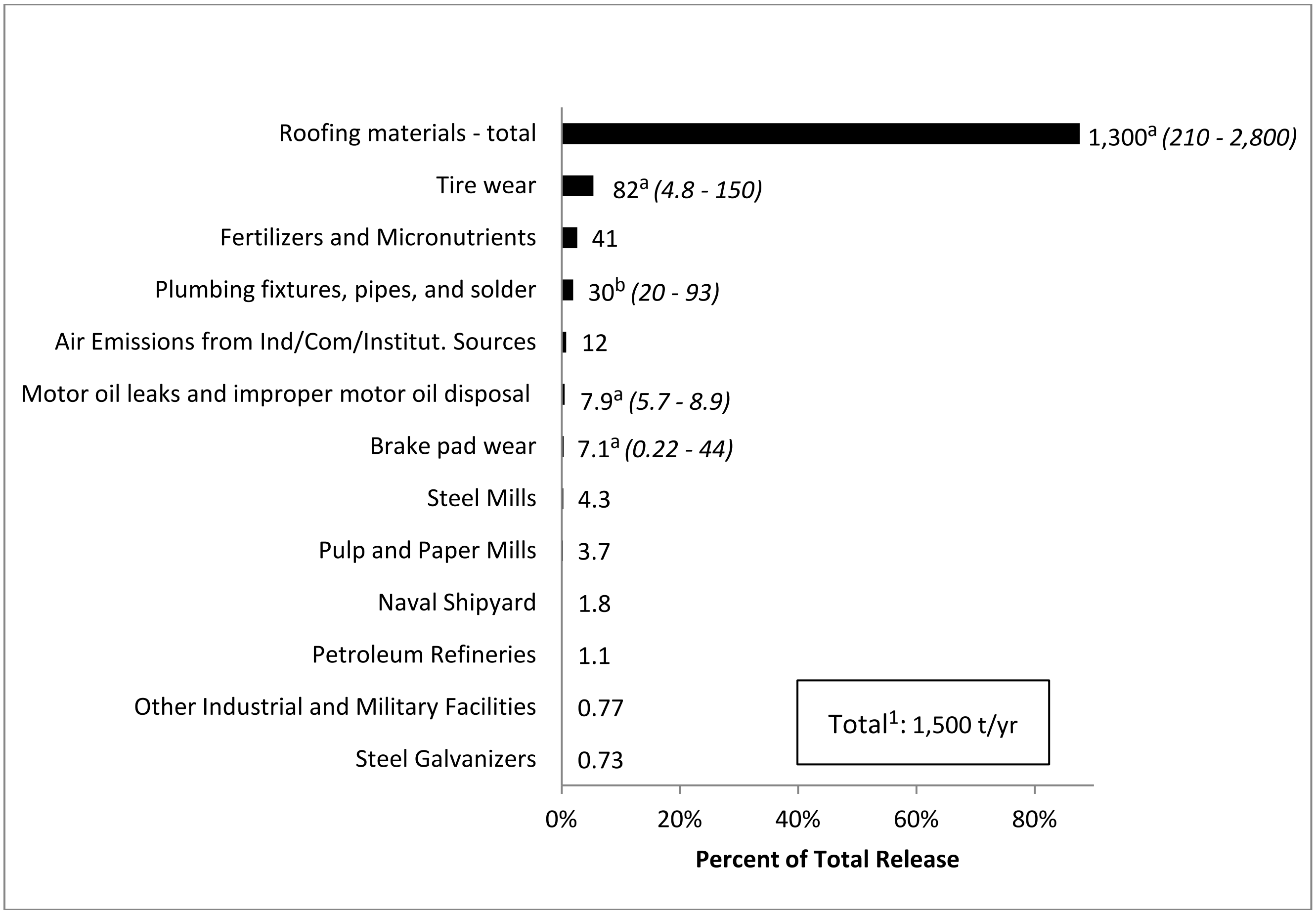
Uncertainty

Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of Zinc Release Estimates

Zinc releases from the sources assessed total approximately 1,500 t/yr (Figure 8). The largest source of zinc release is from roofing material, accounting for approximately 90% of the total. Another component of buildings (plumbing) accounts for an additional 1% of the total. Zinc releases from vehicles account for 6% of the total, with most of that contributed by tire wear. Releases from industrial, commercial and institutional air emissions were minor (<2% of the total).

Estimates of zinc releases from the sources assessed may substantially under-estimate total anthropogenic releases. Possible sources not assessed here include releases to marine waters from sacrificial anodes, leaching from guard rails, leaching from streetlight and road sign standards, leaching from chain link fences and other galvanized materials, and use of zinc salts for moss control on rooftops. These un-assessed sources could potentially add up to thousands of metric tons per year. However, no estimates were made to gauge the magnitude of release from these un-assessed zinc sources.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mean

[b] Median

Figure 8. Total Zinc Release in the Study Area (values shown are t/yr).

PCBs

Sources Assessed

Assessment of ongoing PCB releases included the following sources:

- PCBs in Open Systems
  - Building sealant and caulk loss
- PCBs in Closed Systems
  - Electrical equipment leakage (capacitors, transformers)
  - Residential trash burning

Other possible sources of PCB release include manufacturing and use of pesticides and paints, historic use of pavement seam caulking material, and re-manufacturing and recycling of carbonless copy paper. Releases from these sources were not assessed.

Polychlorinated biphenyls (PCBs) were manufactured commercially in the U.S. from 1929 until their ban in 1979 after the negative health and environmental impacts associated with PCBs became apparent (EPA, 1999). The EPA (1997) estimated that as of 1977, U.S. manufacturers had produced a total of 635,000 t of PCBs. Prior to federally imposed use restrictions, the PCB market spanned a wide range of end products. While electrical equipment represents the majority of PCB use –77% of total use from 1929-1975 according to the EPA (1997) – their chemical stability and plasticizing properties made them useful in a variety of applications. PCB applications as plasticizers, hydraulics fluids and lubricants, and carbonless copy paper accounted for 9.2%, 6.4%, and 3.6% of their uses, respectively.

PCBs in Open Systems

In order to assess environmental release of PCBs from their primary sources, their usage in products may be divided into open and closed systems. Closed systems represent more than three-quarters of the historical PCB use and are typically limited to sealed electrical units (capacitors and transformers). Open systems represent a broader array of historical PCB uses:

- Adhesives and tapes
- Antifouling compounds
- Asphalt
- Brake linings
- Carbon copy paper
- Caulking/Sealants
- Ceiling tiles
- Coal-tar enamel coatings
- Electrical cable insulation
- Fabric
- Flame retardant coatings
- Floor tiles
- Floor waxes and sealers
- Foam
- Glues
- Grout
- Hydraulic and heat transfer systems
- HVAC components
- Inks
- Insulation
- Lubricants
- Paint
- Pesticide extenders
- Plastics
- Roofing
- Roughcast plaster
- Rubber gaskets/parts
- Siding
- Sound-proofing materials
- Varnish
- Waterproofing compounds
- Window glazing

While many materials are known to contain open-PCB stocks, little information with which to compile an inventory on these materials was found, except for sealants. Paints, ceiling and floor tiles, gaskets, fabrics, and other materials containing PCBs remain in use, continuously releasing PCBs into the environment. However, neither the quantity of PCBs in these materials nor the amount of these materials in the Study Area is known. A summation of known maximum concentrations for various materials is shown in Appendix I, Table I-1.

Background on Building Sealants

Of the open-system PCB uses, sealants and other building materials are among the most durable and likely to continue to be a source of PCBs to the environment. Sealants plasticized with high levels of PCBs, as well as other contaminated building materials, have been found in a number of places including municipal water reservoirs, swimming pools, office and institutional buildings, apartments, homes, and schools (Anne Casey, Northeast Analytical, Inc., written communication; Herrick et al., 2004; Kohler et al., 2005; Lefkowitz, 2005; Robson et al., 2010; Rudel et al., 2008). These materials produce elevated PCB concentrations in both the indoor and outdoor surroundings (Astebro et al., 2000; Herrick et al., 2007a; Kohler et al., 2005; Sundahl et al., 1999; Weis et al., 2003).

German researchers reported indoor air contamination up to 40,000 ng/m^3 in an area with PCB-laden plaster (Weis et al., 2003). Only limited, local efforts to address the continued use and mismanagement of these materials have been made in the U.S. (Jackie McQueen, EPA, personal communication; Watnick, 2010), while European governments began addressing this issue in the 1990s (Astebro et al., 2000; Sundahl et al., 1999). Environmental Health and Engineering (2010) estimates that 60% of the U.S. building stock may be affected by PCB contaminated materials.

Added as an after-market plasticizer, the concentration of PCBs found in sealants varies widely; amounts within the same building or room can vary by orders of magnitude (Weis et al., 2003; Ann Casey, Northeast Analytical, Inc., written communication). Astebro et al. (2000) reported that Thiokol® plasticized with up to 35% PCB was common. The highest concentration reported in the literature comes from a Swiss school constructed between 1966 and 1967 where researchers found concentrations up to 58% (Kohler et al., 2005 citing Sundahl et al., 1999).

Kohler et al. (2005) suggest a minimum functional concentration of 1% PCB in order to achieve plasticizing properties, and numerous reported concentrations fall well above this level. Researchers in Finland reported finding a mean PCB content of 10-20% (Priha et al., 2003; Pyy and Lyly, 1998). Sundahl et al. (1999) reported a range of 4-8% for one 8-story building with badly deteriorated joints sealants, applying a mean PCB concentration of 6% to estimate a total PCB store of 90 kg for the building sealants overall. Because PCBs migrate into surrounding materials, recontamination of replacement caulking from surrounding materials also occurs, and may explain the observances of PCB concentrations below 1% in some caulk material (Robson et al., 2010; Tom McSimmons, EPA, personal communication).

Most published inventories of PCBs in sealants have focused on masonry and institutional buildings; however, Robson et al. (2010) reported finding PCB sealants in one single-family dwelling (of 14 sampled). Sundahl et al. (1999) reported that 100 to 500 t of PCBs were added to sealants in Sweden. Kohler et al. (2005) estimated a total store of 50 to 150 tons of PCBs in masonry building sealants across Switzerland. Robson et al. (2010) recently published a similar inventory for the city of Toronto, Canada, in which they estimated 1.6 to 231 t of PCBs remain in sealants throughout the city. Looking at individual building sites, both Astebro et al. (2000) and Sundahl et al. (1999) observed contamination to the surrounding areas. Applying Kohler's suggested minimal functional concentration of 1%, Robson et al. (2010) calculated a mean regional sealant PCB loss rate of 9% over 50 years of exposure.

Congener analysis supports the theory of loss through volatilization, with the ratio of lower-to-higher chlorinated congeners originally present in commercial Aroclor mixtures reduced in present-day sealant samples. Robson et al. (2010) reported a near lack of trichlorobiphenyls in sealant samples, though the compounds were present in original Aroclor mixtures at percent levels (ATSDR, 2000). Herrick et al. (2007b) also found higher blood serum levels of lower chlorinated congeners in workers removing PCB-contaminated caulking than in the mean population. Investigations in Sweden have shown that people living in buildings with PCB caulking have higher PCB levels in their blood than those living in non-PCB buildings within the same community (Johansson et al., 2003).

Researchers throughout the U.S. have also reported finding PCBs in sealants and building materials. Herrick et al. (2004) found PCBs in 13 of 24 (54%) buildings sampled in the Boston area, with eight of those samples (33%) exceeding the regulatory bulk waste level of 50 mg/kg. Sampling in the northeastern U.S. revealed that a number of schools and public buildings contain PCB-laden sealants at tens of thousands of parts per million (Lefkowitz, 2005). The New York school district enacted a remediation protocol and pilot study in response to a lawsuit initiated by parents after the discovery of PCB contamination in the districts' schools. In the western U.S., PCB-containing expansion joints have been reported in water basins in both Colorado and California (EPA, date unknown; Sykes and Coate, 1995), and in pavement joints in Washington State (Golder, 2007). Recently, PCBs were shown to have migrated to surrounding carpet and soils from construction materials in a public building in King County, Washington (Ervin, 2010).

While it is known that the era for PCB use in building materials and other products is limited, an exact date range is unknown and varies by material. For example, PCBs were intentionally added to paints as early as the 1930s. For other materials the initial use may have been as late as the 1950s. In 1973 the Organization for Economic Cooperation and Development urged member nations to cease using PCBs in open systems (Kohler et al., 2005), and all but limited PCB production was banned in the U.S. in 1979.

According to the EPA (1997), Monsanto, the U.S. manufacturer of PCBs, did not market PCBs for open systems until 1957, and then voluntarily withdrew its PCB production for such uses in 1972. The EPA therefore concludes that in the U.S., the usage of PCBs in open systems was limited to 1957 – 1972. However, studies investigating the prevalence of PCB sealants have found them in buildings constructed from 1945-1980. U.S. workers that applied caulking in the 1970s reported that PCB sealants were common, and recalled that the formulation changed in the late 1970s, indicating that PCB use in sealants in the U.S. may have continued after 1972 (Herrick et al., 2004).

The era for PCB use in sealants in other countries appears to parallel the U.S. timeframe. In Switzerland, PCBs were purportedly used only during the 1955-1972 period, after which Switzerland also banned their use in open systems. However, Kohler et al. (2005) reported finding PCB-containing sealants in buildings built both before 1954 and after 1977. In a Toronto, Canada study, three of 11 buildings constructed between 1945 and 1960 had detectable concentrations of PCBs in their sealants (the exact date of the specific samples remains unclear), but the results suggest that either the use of PCBs in sealants occurred earlier than reported, or PCB-containing caulking material introduced during renovations obscure the exact PCB sealant era based on building age (Robson et al., 2010).

Estimate of PCBs Released from Building Sealants

Release of PCBs from building sealants (caulking material) in the Study Area was conducted by:

1. Reviewing the available literature for information on the types and ages of buildings most likely to contain caulking with PCBs.
2. Sampling available county assessor's information to estimate the volume of candidate buildings and develop an inventory of caulking material likely to contain PCBs within the Study Area.
3. Reviewing the available literature for data on PCB concentrations and release rates in caulking material.
4. Applying literature values to estimate the mass of PCBs contained in caulk and PCBs released from caulk in the Study Area.

A review of the literature on the types and ages of buildings likely to contain PCBs in caulk was discussed previously. Based on this information, it appears that commercial and institutional masonry buildings (or buildings with significant masonry components) constructed from 1945-1980 are the best candidates for containing caulking materials with PCBs. Since the literature reported only one instance of PCB-contaminated caulking in single family homes (Robson et al., 2010), such residences were excluded, though some homes may contain PCB-contaminated sealant materials. Additional materials may also be present in non-masonry buildings, around windows, doors, and other places where sealants are used, as well as pavements and sidewalks (University of Massachusetts, 2010), although fewer data are available on PCB-containing caulk in these items and the quantity of sealants used in association with these items are difficult to estimate. Therefore, the focus of PCB releases was limited to sealants in the narrow group of buildings described above.

The total volume of commercial masonry buildings built between 1945 and 1980 was calculated by obtaining a per capita estimate of volume from two counties within the Study Area – Pierce and Snohomish Counties – containing a combined one-third of the Study Area population. County assessors' databases were queried for relevant information on target buildings. Building volume estimates were then scaled up to the entire Study Area in proportion to population.

The number of buildings meeting the target criteria in Pierce and Snohomish Counties combined was 14,469, with a combined area of 61 million square feet. The total volume of these buildings was 782 million cubic feet base on a mean number of stories per building (1.29 for Pierce, 1.14 for Snohomish) and a mean story height of 10.3 feet (both counties). Scaled up for the Study Area, the total volume of buildings meeting the criteria was 2.3 billion cubic feet (66 million cubic meters). Details of the methodology used to derive the building volume estimates are in Appendix J.

Estimates of sealant amounts were calculated by applying a mean sealant mass per volume of 55 g/m^3 based on information provided in Robson et al. (2010). Based on the sealant mass per volume of 55 g/m^3 and a masonry building volume in the Study Area of 66 million cubic meters, total sealant mass was estimated to be 3,655 t.

A number of studies contain relevant information on PCB concentrations in caulking sealant material. Published data on detectable concentrations ranged from 0.56 mg/kg (Herrick et al., 2004) to 583,000 mg/kg (Kohler et al., 2005, citing Sundahl et al., 1999). Appendix I, Table I-2, shows a summary of average detected concentrations from a variety of studies. The mean concentration of PCBs in sealants from these reported values was 38,600 mg/kg.

Few of these studies reported on the frequency at which PCBs were detected in caulking sealants analyzed. Robson et al. (2010) and Herrick et al. (2004) reported detection frequencies of 15% and 54%, respectively, but these were based on relatively few detections (12 and 13, respectively). A compilation of data on PCBs in caulking sealants by Kohler et al. (2005) provides a summary of 1,348 results with a 48% frequency of detection (646 detects). In addition to providing a comparatively large dataset, Kohler et al. (2005) have also included information on the distribution of PCB concentrations among detected results. This is particularly useful information given the wide range of reported PCB concentration. Kohler et al. (2005) do not report median or mean PCB concentrations, but the distribution of PCB concentrations indicate that the median concentration is in the 1,000 – 10,000 mg/kg range.

Estimates of PCB quantity in Study Area sealants were calculated based on the distribution of PCB concentrations reported by Kohler et al. (2005). Table I-3 (Appendix I) displays the variables used in the calculations and resulting inventory. Based on these calculations, there are approximately 39,000 – 79,000 kg of PCBs in Study Area sealants, with a mid-point estimate of 59,000 kg.

Little information is available on release rates of PCBs from caulking material. As mentioned previously, volatilization appears to be the primary route of loss. However, data suggest that PCBs may not remain in vapor phase much past their initial release from caulk. Secondary receiving media may include adjoining materials (e.g. other building material, carpet), or adsorption to dust and films. Weathering of caulk through fragmentation and abrasion may also release PCBs directly in particle form. Since these routes of PCB release are impossible to accurately quantify without empirical data and modeling, a long-term gross loss estimate calculated by Robson et al. (2010) – a mean regional sealant PCB loss rate of 9% over 50 years of exposure – was used to estimate the PCB loss rate from sealants in the Study Area. Averaged over 50 years, a release rate coefficient of 0.0018/yr (0.09 divided by 50 years) was used to estimate annual PCB release to the Study Area.

Table 56 shows estimates of PCBs released to the Study Area from sealants. Estimates were based on the inventory of PCBs in caulk and a release rate coefficient of 0.0018/yr. Total releases were calculated to be 71 – 140 kg/yr, with a mid-point value of 110 kg/yr.

Table 56. Estimates of PCBs Release from Building Sealants.

Volume of Buildings in Study Area Potentially Containing PCB Sealants (m^3)	**Quantity of Sealants in Study Area Potentially Containing PCBs (kg)**	**Quantity of PCBs in Sealants in Study Area (a) (kg)**	**Release Rate Coefficient**	**Total PCBs Release in Study Area (kg/yr)**
65,450,693	3,654,788	39,000 – 79,000 59,000 (mid-point)	0.0018/yr	71 – 140 107 (mid-point)
Total Estimate:				71 – 140 110 (mid-point)

(a) See Appendix I, Table I-3 for derivation of PCBs in Sealants

Uncertainty

There are no regional data to validate the assumptions used in the PCB release estimates. While it can reasonably be assumed that caulking material containing PCBs is still in use in the Study Area and that PCBs are released from the caulking material, there is no information to assess whether regional rates of use and releases are the same as those reported in the literature.
In terms of relative confidence of the variables used to calculate release estimates, the PCB concentrations in caulking materials are likely to be the most reliable. The frequency of detection and the distribution of concentrations provided by Kohler et al. (2005) are useful in estimating a reasonably narrow range of PCB concentrations in order to calculate a PCB inventory.

Estimates of building volumes for masonry buildings appears to be fairly reliable – although the construction era of 1945 to 1980 may over-estimate the number of candidate buildings with PCBs in caulk – but there are no data to validate the caulk mass per building volume (55 g/m^3) assumption. Furthermore, the caulking may not be limited to commercial masonry buildings. Compounding this uncertainty is a lack of information on the amount of original caulk replaced. Given the age of the buildings, it seems likely that renovations and remodeling have occurred on a substantial percentage of these building.

The final variable for which there is a low degree of confidence is the PCB release rate. The release rate coefficient assumes a consistent rate of release over 50 years, an unlikely scenario since evidence points to an initial relatively rapid release of the lighter, more volatile PCB congeners. Congeners remaining after this initial loss through volatilization would be heavier and less volatile on average, and overall PCB concentrations – particularly PCBs near the caulk surface – would be lower as well. However, it is possible that even as loss through volatilization becomes slower, loss rates from abrasion and brittleness of weathered and aged material may increase. In addition, secondary releases (i.e. releases from the initial receiving mediums) may also increase over time through disturbance, weathering, or other processes.

PCBs in Closed Systems

Estimate of PCBs Released from Transformers and Large Capacitors

Although many PCB-containing transformers and large capacitors have been retired, a substantial number may remain in service. The estimated lifetime of Askarel (PCB-containing) transformers may be as much as 85 years (EPRI, 2000). Capacitors last approximately 20 years, and the cessation of PCB production in 1977 means that even the oldest of the capacitors should have already reached the end of their useful life, but some PCB transformers may remain operational for decades to come.

Prompted by concern over the risks of fires, the EPA maintains a transformer registry. For cities located within the Study Area, historic registrations totaled 104 individual units with 60,150 kg of PCB oil. However, as of 2006, only 29 units with 2,077 kg PCB oil were registered, a 72% reduction in individual units and a 97% reduction in PCB oil weight. The registry may not accurately reflect true conditions, as some units are not registered (a discussion on the estimated number of units remaining in service follows). Also, some previously registered units that have been retired or retro-filled with non-PCB oil are not accounted for in the most recently published registration data. For example, four units registered to the Bonneville Power Administration under the most recent registration data are no longer in service (Joe Sharpe, Bonneville Power Administration, written communication).

The 2007 Great Lakes Binational Toxics Strategy Progress Report (EPA/EC, 2007) estimates that there were 73,000 PCB transformers in use in 2005; however, the registry showed only 14,700. In the EPA's own accounting, they state that the data maintained by EPA "is not particularly useful for determining the amount of PCB equipment that is remaining in service" (EPA/EC, 2007). Discrepancies uncovered between the current registry and the stated use of several equipment owners, combined with the limitations described in the 2007 Progress Report (EPA/EC, 2007), lead to the conclusion that the registry does not provide an accurate inventory of existing equipment.

Therefore, no current inventory of PCB equipment exists in the U.S; however, various attempts have been made to estimate the number of units remaining in use. As of 2005, the EPA estimated that 73,000 PCB transformers and 1,290,000 PCB capacitors remained in use throughout the U.S. (EPA/EC, 2007). Scaled down from the national population to the Study Area (U.S. Census Bureau, 2010), an estimated 1,064 transformers and 18,805 large PCB capacitors remained in use in the Study Area in 2005.

While both the size of transformer and capacitor units and the amount of PCBs per unit vary, Askarel transformer fluid is typically 60-70% PCB by weight. The quantity of oil per transformer unit ranges from 150-1,890 liters and weighs 235-2,932 kg, resulting in a PCB content of 141-2,052 kg per transformer (EPA, 1987). Capacitors are typically filled with nearly pure PCB oil (EPRI, 2000). Discussed in terms of large capacitors (over three pounds) and small capacitors, the largest contain as much as 35 kg PCB (EPRI, 2000), with the typical large, high-voltage capacitor weighing about 54 kg (120 lbs) and containing 11 kg (25 lb) of PCBs. Small capacitors, used in motors and other applications, generally contain 0.1-0.6 kg of PCBs (EPA, 1982). Lamp ballast capacitors contain 46-70 grams of PCB per unit (EPA, 1982).

During a 1982 inventory of utility industry equipment, 2.3% of large capacitors were found to have developed small leaks annually and 0.77% developed moderate leaks annually (EPA, 1982). Small leaks were defined as those resulting in a visible sheen on the outside of the equipment; moderate leaks were those in which fluid ran off of the equipment. An annual leakage rate was not provided for PCB transformers. However, the study reported finding small leaks in 12% and moderate leaks in 4% of inspected PCB transformers (EPA, 1982); the risk of leakage increases as equipment ages. The 1982 EPA study also reported leakage rates for mineral oil transformers, which EPA later used, along with the leakage rate for PCB capacitors, to calculate a total annual PCB leakage amount from all electrical equipment, both utility and non-utility owned (EPA, 1987).

Leakage rates from mineral oil transformers and PCB capacitors were applied to the total number of units estimated to be in use to derive a total annual leakage amount. Using the 1982 data, annual leakage rates of 0.23 kg PCBs/unit for transformers and 0.06 kg PCBs/unit for capacitors were calculated. Applying these to the estimated number of Study Area electrical equipment units results in a total release of 7 to 250 kg/yr for transformers and 1,100 kg/yr for large capacitors (Table 57).

Table 57. Estimates of PCBs Release from Transformers and Large Capacitors.

Equipment Type	Number of Units in Service in Study Area	Basis for Estimate	PCB Spill/Leak Rate (kg/yr/unit)	Total PCBs Release in Study Area (kg/yr)
Transformers	29	Registered	0.23	7
	1,064	Scaled from national estimate		250
	29 – 1,064	Summary of estimates above		7 – 250 130 (mid-point)
Large Capacitors	18,805	Scaled from national estimate	0.06	1,100
			Total Estimate:	1,100 – 1,400 1,200 (mid-point)

Uncertainty

Differences in the possible number of in-use transformers are a large source of uncertainty. There is no regional inventory of Askarel transformers still in use, and the registration program appears to vastly under-estimate the number of transformers possibly due to lack of aggressiveness in tracking or lack of participation by transformer owners.

The equipment leakage rates should be considered to be fairly reliable, though based on older data that may not reflect current operating conditions. The number of electrical units used indoors versus outdoors is unknown, and the estimates do not account for any spill response efforts, thus actual amounts released into the environment may only be a portion of the estimated total. Further emissions are likely through direct volatilization from equipment, although the exact amounts are not known.

Estimate of PCBs Released from Small Capacitors

Capacitors containing less than three pounds of PCB oil are considered small capacitors. Small capacitors containing PCBs have been used in a number of items including motors, appliances, and lamp ballasts. A typical small capacitor unit contains 0.1-0.6 pounds (45 - 270 grams) of PCB oil, with lamp ballasts typically containing about 45 - 70 grams per ballast (EPA, 1982).

Globally, one-third of all PCB production may have gone into lamp ballasts (Panero et al., 2005). In 1992 the University of Illinois estimated that 10-25% of U.S. household white goods (major appliances) contained capacitors with PCBs (Panero et al., 2005). Though it is known that many small PCB capacitors were manufactured prior to 1978, estimates of the number still in use vary.

EPA (1982) estimated that historically there were 870 million small capacitors in use throughout the U.S. in 1977 in industrial machines and small appliances. EPA (1987) also estimated a 10% annual disposal rate in 1982. Applying annual disposal rates of 10 and 20% to the national estimate for 1977 and scaling to the Study Area by proportion of national population in the Study Area (1.46%) yields a range of 8,000 to 380,000 total small capacitors remaining in use in the Study Area during 2010.

Estimates for PCB lamp ballasts currently in use are an order of magnitude higher than the 1982 EPA estimate for small capacitors. These estimates place the number of ballast units remaining in use nationally between roughly 300 million (U.S. Army, 2001) and 500 million (Missoula County, 2010). In 1998, the EPA cited an unnamed industry source that estimated one billion ballasts were currently in use (EPA, 1998a). The EPA (1998a) reference suggests that the current number of PCB-containing ballasts in use nationally would be somewhere between 280 million, assuming a mean annual disposal rate of 10% from 1998 to 2010, and 69 million, assuming a mean annual disposal rate of 20% from 1998 to 2010. Scaling the 1998 estimate by population, and applying the 10 and 20% disposal rates, yields a range of 970,000 to 4 million such ballasts remaining in-use in the Study Area in 2010.

An effort was made to determine a disposal rate from available data; however, only an incomplete picture of disposal volumes could be constructed. Washington State tracks PCB waste disposed of under the Washington State PCB Waste rule (WPCB) as well as PCB-ballasts disposed of under the Moderate Risk Waste rule (MRW). Combining disposal data from both WPCB and MRW for 2007-2009 provides an estimated mean annual disposal rate of 5,400 small capacitor units. However, few disposals are captured by these waste streams, and under WPCB, the specific type or origin of the waste is not always provided (Tom Cusack, Ecology Hazardous Waste and Toxics Reduction Program, personal communication; Kathleen Kaynor, Ecology Hazardous Waste and Toxics Reduction Program, written communication; Al Salvi, Ecology Waste 2 Resources Program, personal communication).

Due to uncertainties surrounding the number of units remaining in use, a range of 8,000 to 4 million was used to estimate releases. While small capacitors may contain 45-270 grams per capacitor unit (EPA, 1982), most of the remaining units are likely lamp ballasts, which typically contain only 45-70 grams of PCB oil. Applying 45-70 grams of PCB per unit to the number of estimated units in use (8,000 to 4 million) results in an estimated 400 to 300,000 kg of PCBs remaining in small capacitors within the Study Area. Applying a leakage rate of 4.2 kg per metric ton (0.42%) of PCB, calculated from the 1982 inventory of large capacitors (i.e. total mass of PCBs leaked/total mass of PCBs in large capacitors; EPA, 1982), there is a total leakage of approximately 1 to 1,000 kg from small capacitors in the Study Area in 2010 (Table 58). The mid-point of this range is 500 kg.

Table 58. Estimates of PCBs Release from Small Capacitors.

Number of Small Capacitors in Use in Study Area	Quantity of PCBs Contained in Each Small Capacitor (g)	Quantity of PCBs Contained in Small Capacitors in Study Area (kg)	PCB Leakage Rate (yr^{-1})	Total PCBs Release in Study Area (kg/yr)
8,000 – 4,000,000	45 - 70	400 – 300,000	0.0042	1 – 1,000 500 (mid-point)
			Total Estimate:	1 – 1,000 500 (mid-point)

Uncertainty

Differences in the possible number of in-use small capacitors are a large source of uncertainty, as are disposal rates which are based on estimates nearly three decades old. The equipment leakage rates should be considered to be fairly reliable if they are identical to rates for large capacitors and transformers. The number of electrical units used indoors versus outdoors is unknown, thus actual amounts released into the environment may only be a portion of the estimated total. Further emissions are likely through direct volatilization from equipment, although the exact amounts are not known.

Estimate of PCBs Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

There were no reported PCB releases to air, water, "other disposal", or POTWs during 1999-2008. Pulp mills and waste processors reported releases to other receiving media (e.g. regulated landfills), but these were not defined as releases for the present study.

Estimate of PCBs Released from Modeled Air Emission Sources: Trash Burning

Air emissions of PCBs inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Residential trash burning accounted for all of the releases (280 kg/yr, Table 59).

Table 59. Estimates of PCBs Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

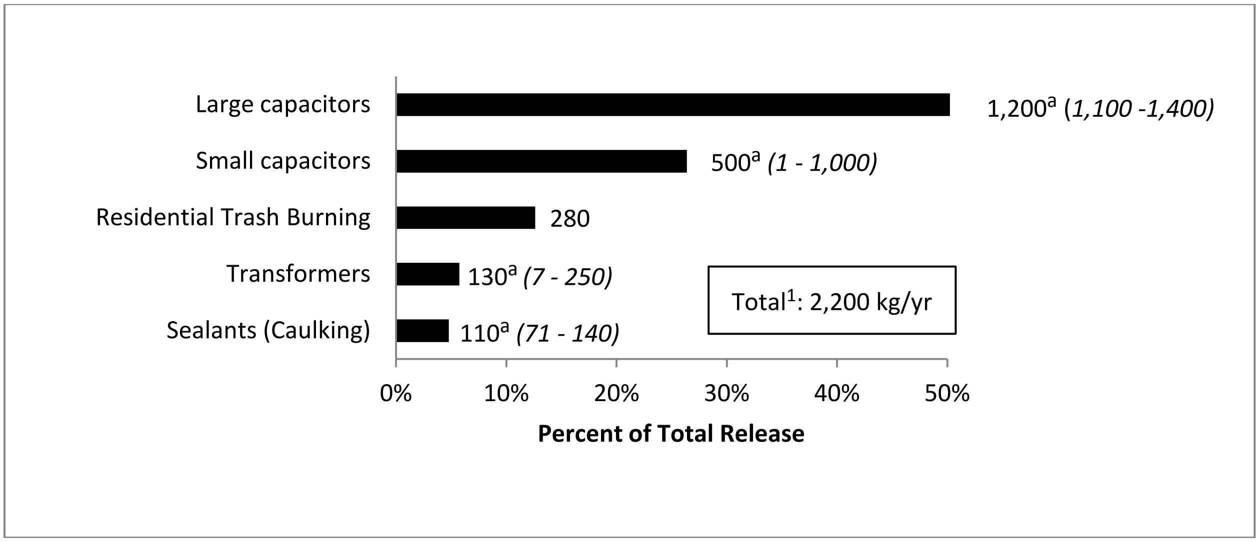
Air Emission Source	Total PCBs Release in Study Area (kg/yr)
Residential Trash Burning	280
Total Estimate:	280

Uncertainty

Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of PCB Release Estimates

Based on the best estimates available, total PCB releases from the sources assessed are approximately 2,200 kg/yr, with a range of 1,500 to 3,000 kg/yr (Figure 9). One-half of PCB release is contributed by large capacitors, with leakage from all electrical equipment (large and small capacitors, transformers) accounting for more than three-quarters of the total PCB release in the Study Area.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 9. Total PCB Release in the Study Area (values shown are kg/yr).

The PCB release estimates presented here are highly uncertain due to a number of factors. First, the products and materials into which PCBs were incorporated (electrical equipment and caulk) are a minimum of three decades old, and therefore it is impossible to obtain accurate estimates of their continued use and PCB release rates based on the available information. Second, it is unclear the degree to which PCBs are released *to environmental media or released in a form which can be mobilized and transported in an environmental pathway* (see definition of sources in the Introduction section). This latter point is applicable to caulking sealants, but is especially true for leakage from electrical equipment since much of the leakage may occur indoors, on containment structures, or may otherwise be contained or cleaned up following leakage. Since releases from electrical equipment account for at least three-quarters of the total PCB release, the values presented here likely over-estimate actual releases by a substantial degree.

There are likely ongoing PCB releases from materials other than those assessed here. They may include manufacturing and use of pesticides and paints, historic use of road and pavement seam caulking material, and re-manufacturing and recycling of carbonless copy paper. PCB releases from manufacturing and re-manufacturing activities are likely to be small, but releases from road and pavement caulking may be substantial based on studies done on private pavement surfaces and anecdotal street sweeping information (Tony Paulson, U.S. Geological Survey, written communication). However, due to the age of the material and the continual replacement of road pavement, estimates of PCB releases from pavement sealants would prove extremely difficult on a region-wide scale.

PBDEs

Background

Polybrominated diphenyl ethers (PBDEs) are bromine-containing organic compounds widely used as flame retardants. The PBDE molecule consists of two phenyl ring molecules linked together by an oxygen atom. Each phenyl can have from zero to five bromine atoms attached.

As there are 10 locations on the PBDE molecule to which a bromine atom can attach, 209 unique compounds (also called congeners) can exist. Congeners vary from each other only by the number and location of bromine atoms on the molecule. The use, behavior in manufactured materials, and behavior once released of specific congeners vary substantially depending on the number of bromine atoms in the congeners.

PBDE manufacturers produced three major PBDE blends referred to as Penta, Octa, and Deca. Each blend contained a range of PBDE congeners, and the following shows the PBDE congeners found in each group according to Ecology and WDOH (2006):

- Penta : <1% tri-BDEs, 24-38% tetra-BDEs, 50-60% hexa-BDEs
- Octa: 10-12% hexa-BDEs, 43-44% octa-BDEs, 31-35% nona-BDEs, <1% deca-BDE
- Deca: <3% non-BDEs, 97-98% deca-BDE

Penta was used widely in polyurethane foam and textiles, while the heavier blends (Octa and Deca) were used primarily in polymers and electronics. The heavier blends tend to be less volatile than the lighter PBDEs, although once in the environment the heavier PBDEs may degrade to the lighter congeners. Heavier congeners such as Deca may also bind to dust more strongly than the lighter congeners.

Manufacturers of many different materials and products have used PBDEs as flame retardant additives since the 1960s. These products include fabrics, television sets, computers, polymer resins such as acrylonitrile butadiene styrene (ABS) and high impact polystyrene (HIPS), textile coatings, carpet, polyurethane foams, cushions, mattresses, and insulation for wire and cables.

PBDEs are not chemically bonded to the matrices of those materials and products; therefore, they potentially escape from their matrix through volatilization, sloughing, and product aging. Products and materials partially composed of or treated with PBDEs release PBDEs to the environment during the useful lifetime of the product or material (i.e., while the product or material is still in use). Volatilization has been suggested as one of the primary mechanisms of the release of PBDEs to the environment (Lorber and Cleverly, 2010). Although PBDEs generally are not volatile compounds, the increased temperatures found in electronic products can cause PBDEs to be released into the home and office through this mechanism.

Those PBDEs released through volatilization quickly settle out of the air and can be found coating surfaces and adsorbing onto particulate matter. PBDEs adsorb preferentially onto organic particles which, given the prevalence of organic matter in homes and offices, increases their presence in house and office dust. In addition, air transport of PBDE-containing particulate matter is believed to be one of the primary mechanisms for transport of PBDEs into remote areas and throughout the planet.

Beginning in the late-1990s, concerns began to emerge over the accumulation of PBDEs in breast milk and animal tissues and their potential toxicity. Pressure to limit or ban PBDEs continued to mount until three major manufacturers of Penta and Octa voluntarily ceased production beginning in 2004. At least 11 states have currently banned the use of Penta and Octa in their state. Three states have banned Deca in specific applications and two have banned Deca in all applications. Washington State banned Deca for use in mattresses beginning in 2008. After conducting an alternative assessment and identifying a viable alternative in 2009, Deca was banned from televisions, computers, and residential upholstered furniture in Washington State as of January 1, 2011. In December 2009, three major Deca manufacturers agreed to phase out the use of Deca in the U.S. by 2012.

Sources Assessed

Assessment of ongoing PBDE releases included the following sources:

- Releases to indoor commercial office space
- Releases to indoor residential space

These sources of PBDE represent combined releases from individual products used in the home and office such as, for example, televisions, computer monitors and other electronic equipment, foam padding for furniture, and wire insulation. Assessment of PBDE sources through combined releases in the home and office integrates the individual components from which PBDEs are initially released. Not all of the PBDEs found in these sources are released directly to the environment. However, using the amounts of PBDEs found in these spaces compensates for not including other potential sources that are not well studied and quantified.

PBDEs are released to the environment through numerous sources such as:

- Manufacture and transport of the PBDE mixtures
- Formulation and manufacture of specific products
- Products during use
- Products during disposal and release at end-of-life

This list is not intended to be comprehensive but to provide an idea of the broad scope of potential PBDEs releases. Some of these sources can be locally important. Some research has been done on the release of PBDEs during manufacture (DEFRA, 2004) but little has been on releases during transport of the PBDEs themselves. Some limited research has been conducted on PBDE releases at the end-of-life, primarily from landfills, recycling facilities, and auto and metal shredders. Assessing direct environmental releases from all of these individual sources are nearly impossible based on the current level of knowledge.

Estimate of PBDEs Released Due to Emissions to Indoor Commercial Office Air and Dust

Many of the furnishings and equipment in a typical office emit vaporous and dust-bound PBDEs. Hard plastic equipment such as computers and monitors, furniture, carpets, wire insulation, piping, and many other materials contain PBDEs which are released, adhere to dust particles, and either blow away or adsorb onto the floor and other surfaces.

Detailed analyses of PBDE loss from individual components (e.g. computers, wire cables insulation, polyurethane foam) are difficult to quantify due to the variables associated with each component (e.g. PBDE composition, age of component) as well as environmental variables such as temperature which partially drive the rate of loss. When PBDE losses from individual components are compounded with factors affecting their escape to the outdoor environment (e.g. remaining in vapor phase, attaching to dust particles, re-sorption to organic materials), factors such as the amounts adsorbed onto surfaces, the amount of PBDEs that settle with particulate matter and are found in indoor dust samples, accurate release estimates, etc. require more sophisticated modeling than is available for the present study.

The following are primary release mechanisms of PBDEs from the office to the outdoor environment:

1. Vacuuming and sweeping and removal to landfills, POTWs, incinerators, etc.
2. Release to POTWs via cleaning of surfaces including floor washing, dusting, etc. and through hand washing, showering, etc.
3. Direct transport of material via foot traffic
4. Capture in filters from heating, air conditioning, etc. and subsequent disposal to landfills or to incinerators
5. Direct release of particles through air exchange

It is difficult to quantify which of the above is the primary release mechanism of PBDEs from the office directly to the environment. Work has been done to quantify the release through air exchange (Zhang, 2008; Harrad et al., 2008), and the amount of total PBDEs found in office dust has been studied (Leonards et al., 2001; Santillo et al., 2001). Leonards et al. (2001) in particular indicated that for deca-BDE, air particles and dust are the most important human exposure pathways. For the purposes of this study, the amount of total PBDEs found in office indoor air and dust will be used to estimate the overall releases of PBDEs to the environment.

In order to avoid the requirements of modeling numerous individual component emission rates, the indoor environment as a whole was considered to be a source for the purpose of calculating PBDE releases and data on PBDE releases, from the indoor environment were sought. Zhang (2008) conducted a detailed study employing environmental measurements and modeling of PBDE releases due to the combination of both volatilization and dust transport to assess the rate that indoor environments emitted PBDEs to the outdoors. Sampling of ambient air from 20 indoor environments in Toronto revealed total PBDE emissions ranging from 150 to 970 ng/hr for each square meter of office floor space. These rates were applied to estimates of the area of office space in the Study Area based on data from the U.S. Energy Information Administration (EIA, 2003) and the U.S. Census Bureau (2010). PBDE releases ranged from 120 to 750 kg of total PBDEs per year, with a mid-point of 430 kg/yr (Table 60).

Table 60. Estimate of Total PBDE Emission from Indoor Office Air.

Commercial Office Area in Study Area (a) (m^2)	PBDEs Measured	Unit Emission ($ng/m^2/hr$)	Total PBDEs Release in Study Area (kg/yr)
8.77 x 10^7	BDE-28, 47, 66, 99, 100, 153, 154	150 – 970	120 – 750 430 (mid-point)
		Total Estimate:	120 – 750 430 (mid-point)

(a) Includes offices, warehouse and storage, education, lodging, service, public assembly, religious, non-mall retail, health care, and food service & sales

Watkins et al. (2011) collected dust samples from 31 offices in the Boston area. Samples were analyzed for 21 major PBDE congeners. Results from two groups of co-eluting congeners (BDE-28/33 and BDE-85/155) were combined and reported as a single number. The primary PBDE congeners found in these dust samples were from the penta BDE mixture (congeners 47, 99, 100, 153, &154) and the deca BDE mixture (congener 209). Using these data, the concentrations of PBDEs in office dust were calculated to range from <0.1 to 106,000 ng/g, with a geometric mean of 6,854 ng/g (no median or arithmetic mean values were reported).

No studies attempting to identify the amount of dust generated in offices were found. However, a value of 40 pounds of dust per year from a 1,500 square foot house has been extensively reported (Brooks, 2011), equal to a unit area dust production of 130 g/m^2. Although offices are typically occupied for only one-third the amount of time as a house, the amount of PBDEs in house dust has been correlated with the number of electronics in the house (Allen et al., 2008), and offices generally have a much higher concentration of electronics than the home environment. Using the commercial office area identified in Table 60 (8.77 x 10^7 m^2) and assuming the same rate of dust generation as a home, the mean (geometric) amount of PBDEs in dust generated in an office is estimated to be 78 kg/yr (Table 61).

Table 61. Estimate of Total PBDE Release from Indoor Office Dust.

Total Commercial Office Area in Study Area (m^2)	Rate of Dust Production ($g/yr/m^2$)	Total Dust Generated in Offices in Study Area (g/yr)	PBDEs Measured	PBDE Concentration in Dust (ng/g)	Total PBDEs Release in Study Area (kg/yr)
8.77 x 10^7	130	1.14 x 10^{10}	BDE-28/33, 47, 49, 66, 75, 85/155, 99, 100, 138, 153, 154, 183, 196, 197, 204, 206, 207, 208, 209	<0.1 – 106,000 6,854 (geo. mean)	<0.001 – 1,200 78 (geo. mean)
				Total Estimate:	78 (geo. mean)

Uncertainty

PBDE emissions from commercial office space depend largely on the type of materials contained in the offices (e.g. computers, carpets) and the rate of air emissions. Estimates based on data from Zhang (2008) were from samples of 20 indoor environments, suggesting a fairly robust representation of office space. However, these releases are based solely on one study, leading to concerns that the amounts observed are not representative of office emissions in general. The impact of this assumption has not been evaluated.

In addition, Leonards et al. (2001) and Santillo et al. (2001) calculated office dust concentrations from European Parliament buildings which may not accurately reflect the concentration found in U.S. offices. Numerous studies of household dust samples (e.g. Hites 2004; Stapleton et al., 2005) have shown concentrations as much as an order of magnitude higher in the U.S. than in equivalent European samples. This variability has not been evaluated and is not included in the calculations.

Two additional sources of uncertainty associated with the commercial office estimates are common among all PBDE release estimates:

1. Since most studies fail to analyze the full suite of PBDE congeners, the data used to comprise "total PBDEs" typically vary among studies and adds a confounding factor when comparing study results.
2. The decline of PBDE uses in new products during the past several years may already be resulting in decreases in environmental releases, introducing a confounding variable when comparing studies of different ages and possibly making sampling data obsolete from as recently as the mid-2000s.

Since Zhang (2008) analyzed only seven PBDEs, and Leonards et al. (2001) and Santillo et al. (2001) only 21 congeners, the resulting release as "total PBDEs" may under-estimate the total of all congeners. With regard to age of the data, the Zhang (2008) data are comparatively recent by most standards, and are well within the typical lifetimes of major PBDE-containing products (typically seven to ten years). The Leonards et al. (2001) and Santillo et al. (2001) data are considerably older and may no longer accurately reflect dust concentrations in offices as manufacturers move away from Deca for newer electronic equipment.

Estimate of PBDEs Released Due to Emissions from Indoor Residential Air

Many of the furnishings of typical residences contain PBDE fire retardants that slowly volatilize or mobilize through release of dust particles. PBDEs are components of polyurethane foam-filled furniture such as sofas and chairs, curtains and carpets with foam or rubberized backings, bedding, plastic items such as televisions and toys, electrical wire coatings, adhesives, and paints and lacquers. The fate of PBDEs released from products and materials within the home is complex, similar to that for indoor offices and commercial areas.

The annual release of PBDEs from indoor residential areas to the outdoor environment was estimated using measured concentrations of PBDEs in U.S. and Canadian homes (Harrad et al., 2010). They reported mean indoor air PBDE concentrations of 100 pg/m^3 in 74 Ottawa, ON locations and 3,200 pg/m^3 in 24 Boston, MA locations. These concentrations were applied to national estimates of median residential housing floor space (1,769 sq ft; HUD and Commerce, 2008), converted into volume by assuming each story was 3 m in height, then scaled to the Study Area (12-county Puget Sound region) according to estimates of the number of houses 1,768,893; U.S. Census Bureau, 2010). The indoor-to-outdoor air exchange rate was assumed to be the same as for indoor offices (0.75/hour; Zhang, 2008).

Based on the variables described above, PBDEs were released at an estimated rate of 0.6 to 18 kg/yr to the outdoor environment. The mid-point of this range is 9.5 kg/yr and a summary of these estimates are in Table 62.

Table 62. Estimate of Total PBDE Emission from Indoor Residential Air.

Residential Volume in Study Area (a) (m^3)	**PBDEs Measured**	**Unit Emission (pg/m^3)**	**Total PBDEs Release in Study Area (kg/yr)**
8.73 x 10^8	Sum(tri-hexa-BDEs)	100 – 3,200	0.6 – 18 9.5 (mid-point)
		Total Estimate:	0.6 – 18 9.5 (mid-point)

(a) Assumes a mean story height of 3 m

Uncertainty

PBDE emissions from residential space to the outdoor environment depend largely on the type of materials contained in the homes (e.g. computers, carpets) and the rate of air exchange. Estimates based on data from Harrad et al. (2010) were from numerous locations (98) in North America, and therefore likely are representative of typical residential indoor environments.

Two additional sources of uncertainty associated with PBDE release estimates are described in the section on releases from commercial office space: inconsistency among the complement of PBDE congeners analyzed, and the age of the reported data. Since some of the locations evaluated by Harrad et al. (2010) were assessed only for tri- through hexa-substituted PBDEs, the resulting release as "total PBDEs" may under-estimate the total of all congeners. With regard to age of the data, the Harrad et al. (2010) data are comparatively recent by most standards, and are well within the typical lifetimes of major PBDE-containing products (typically seven to ten years).

Estimate of PBDEs Released to Indoor Residential Dust

The second major release of PBDEs to the environment is through household dust. As indicated earlier for office space emissions, there are numerous pathways for dust to be released to the environment. In order to estimate PBDE releases, numerous studies on the levels of dust found in homes were evaluated. Stapleton et al. (2004) measured PBDE concentrations in 16 house dust samples. The study only reported the results for 7 congeners (BDE-28, BDE-47, BDE-99, BDE-100, BDE-154, BDE-183, and BDE-209). The total PBDE concentrations ranged from 310 to 31,000 ng/g, with a mean of 5,946 ng/g and a median of 4,130 ng/g.

In a subsequent report, Stapleton et al. (2005) measured 22 PBDE congeners from 16 homes in Washington, DC and one home in Charleston, SC. The median total concentration for all PBDEs was 4,250 ng/g. Zota et al. (2008) evaluated levels of three PBDEs (BDE-47, BDE-99, and BDE-100) in 49 California homes and found the median sum of the concentrations to be 8,336 ng/g for 39 Richmond homes and 2,643 ng/g for 10 homes in Bolinas.

The amount of PBDEs released from house dust was calculated from these values. Although the amount of dust generated in a home is dependent on a wide range of factors (e.g. amount of electronic equipment, age of the house and furnishings, the amount of cleaning), the value of 40 pounds of dust as reported for a standard six-room house of 1,500 square feet was used (130 g/m^2; see discussion in previous sub-section). The U.S. Census Bureau (2010) estimates that there are 1,768,893 homes in the Puget Sound area with a median area of 1,769 sq ft; HUD and Commerce, 2008). Based on this information, it is estimated that 160 kg of PBDEs are released to house dust annually (Table 63) in the Study Area.

Table 63. Estimates of Total PBDE Release to Indoor Residential Dust.

Total Residential Area in Study Area (m^3)	**Rate of Dust Production (g/yr/m^2)**	**Total Dust Generated in Residences in Study Area (g/yr)**	**Median PBDE Concentrations in Dust (ng/g)**	**Total PBDEs Release in Study Area (kg/yr)**
2.91 x 10^8	130	3.78 x 10^{10}	4,130 (Stapleton et al., 2004)	160
			4,250 (Stapleton et al., 2005)	160
			8,336 (Zota et al., 2008)	320
			2,643 (Zota et al., 2008)	100
			Total Estimate:	100 – 320 160 (median)

Uncertainty

There is a large degree of uncertainty associated with the estimate of PBDE releases from house dust. Due to the numerous pathways to which house dust is subjected, it is difficult to approximate exactly how much of the house dust is directly released to the environment and how much might be sequestered in landfills. In addition, numerous studies have tested house dust and reported a wide range of concentration values; therefore, these estimates are only provided as a worst-case estimate of the amount of PBDEs released to the environment from house dust. Lastly, there are numerous other pathways which release PBDEs directly to the environment, and some of these pathways are described below. Therefore, the estimates from house dust are used to compensate for some of these omissions.

Automobiles may be a source of the dust that attaches to fabric and that may subsequently be released to the environment. PBDEs may also be released through particulate transfer as individuals enter and exit a vehicle. Harrad et al. (2010) reported that the interior dust of U.S. automobiles had PBDE concentrations ranging from 2,600 ng/g (sum of tri- through hexa-substituted congeners) to 49,000 ng/g (Deca only). Incorporating automobile-derived PBDE dust would increase the high-end estimate of releases, but these calculations were not included here due to the lack of information about the relative contributions between automobile and house dust.

There are two additional sources of uncertainty associated with PBDE release estimates: Inconsistency in the suite of congeners analyzed, and the age of the data considered.

The concentrations of PBDEs in house dust used to calculate releases can differ by orders of magnitude. The 16 residences analyzed by Stapleton et al. (2004) varied for the 7 PBDE congeners from 310 to 30,140 ng/g dry mass, a variation of two orders of magnitude. In addition, these samples were collected in 2004 and may not reflect the trend away from brominated flame retardants adopted by many computer and television manufacturers. Similar variability was also seen in Stapleton's subsequent report of 17 residences for 22 PBDE congeners (Stapleton et al., 2005). Total PBDE concentrations varied between 780 and 30,100 ng/g dry mass.

Even greater variability was seen in Zota et al. (2008). For the 39 house dust samples from Richmond residences, the values just for BDE-99 varied from 102 to 170,000 ng/g, a difference of more than three orders of magnitude. None of these studies attempted to quantify the age of the electronics and furniture in the residences tested to see if these variables may have played a factor in the observed PBDE concentrations.

Estimate of PBDEs Released from Automobile and Electronics Recyclers

Recyclers of automobiles and electronics are a potentially large source of PBDE release to the environment due to the sheer mass of PBDE-containing materials they handle (e.g. PBDE-impregnated upholstery padding and plastics). Within the Study Area, three large recycling facilities have been identified which have the potential to be large emitters and dischargers of PBDEs. Two of them are auto-recyclers with the bulk of the activity occurring outdoors, while the third facility recycles primarily electronics with disassembly occurring indoors (Pinky Feria, Ecology Hazardous Waste and Toxics Reduction Program, personal communication).

The actual releases of PBDEs from these sources have not been quantified, and published data on PBDEs from auto and electronics recyclers elsewhere are typically comprised of indoor (i.e. warehouse) or outdoor downwind air concentrations. For instance, Cahill et al. (2007) showed that outdoor air samples collected immediately downwind of an automobile-shredding operation in California had total PBDE concentrations up to 1,900 pg/m^3 while the shredder was operating, higher than when the shredder was off. They attributed the PBDE emissions primarily to particulate-associated PBDEs released during operation, although they noted that some of the lighter congeners prevailed during times of non-operation, apparently due to volatilization. There are other instances of air PBDE concentrations reported in the literature, but the variables needed to compute quantities released to the environment are not available.

In addition to air emissions, PBDE releases from these facilities through leaching and entrainment of dust particles in runoff may also contribute to overall releases. Treated stormwater runoff from the largest of these facilities (25 acres) was measured at 2.7 to 4 ng/l total PBDE, and inputs to the system included even higher levels of total PBDEs ranging from 134 to 237 ng/l (Alex Stone, Ecology Hazardous Waste and Toxics Reduction Program, personal communication). Storm events over-ran the treatment system twice per year on average, potentially releasing relatively high quantities of PBDE-enriched solids (53 mg/kg) normally retained during treatment (Podger, 2009). However, the combined (treated and untreated) stormwater load of PBDEs from this facility was estimated to be only 12 grams per year, orders of magnitude lower than estimates from other sources addressed in the present report.

Estimate of PBDEs Released from Landfills

PBDEs may also be released to the environment from landfills which receive consumer products containing high levels of PBDEs. Although limited work has been done in this area, Danon-Schaffer (2010) studied the impact of PBDE-containing products upon releases from landfills. Danon-Schaffer (2010) studied 27 landfills across southern Canada and 11 dump sites in the Canadian North. She reported that there was wide variability in the results both in terms of the total concentrations of PBDEs and in the distribution of congeners. Northern sites tended to have lower levels of PBDEs than southern ones. Even with this limitation, some of the northern sites contained significant amounts of PBDEs in the landfill leachates. Danon-Schaffer (2010) reported that landfill leachate samples contained total PBDE levels ranging from less than 100 to greater than 1,000,000 pg/l.

Landfill leachate potentially is another significant pathway of PBDEs released to the Study Area. Unlike most landfills in Eastern Washington that recycle landfill leachate, landfills in Western Washington typically send leachate to local POTWs for treatment. Although PBDEs are only minimally soluble in water, PBDEs adsorbed to particles have proven to be an important transport mechanism and may play a significant role in PBDE release. This pathway, however, has not been evaluated in Washington and will not be included in release estimates in this report.

Estimate of PBDEs Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

There were no reported PBDE releases in the TRI, as PBDEs are not one of the chemicals included in the report requirements.

Estimate of PBDEs Released from Modeled Air Emission Sources

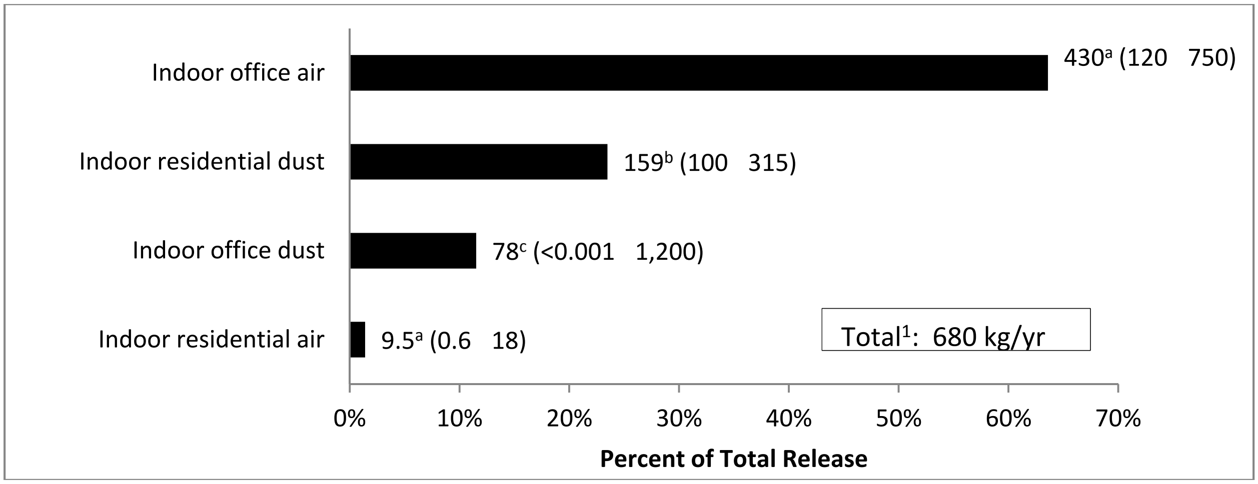
There were no PBDE emissions modeled for the Air Emissions Inventory.

Summary of PBDE Release Estimates

Based on the ‘best estimates,’ total PBDE release from the four sources assessed totaled approximately 680 kg/yr (Figure 10). Indoor office space air accounted for approximately 67% of the total release, while indoor residential air accounted for just 1%. Indoor residential dust was the other major contributor at 23%, while the indoor office dust contributed 12% to the overall releases.

The summary information in Figure 10 suggests a high concentration of PBDEs is found in the office air environment compared with the home environment where house dust contains the highest concentration. This contradicts the values for dust where the office dust levels are approximately one-half of what is found in the home. The reasons for this discrepancy remain unclear. One factor may be the greater predominance of air circulation in the office, greater numbers of computers and other office equipment containing PBDEs, increased movement, and other factors that would favor the suspension of PBDE-containing dust particles in office air. In addition, the volume of office space is only a fraction of the amount of floor space in homes in the target area, which could lead to concentrating PBDEs in the office environment. Lastly, the air calculations are based on air space amounts which, as a concentration, cannot be easily compared with the surface area (square footage) used for the home dust calculations.

One other major uncertainty is related to the amount of data on indoor air. As dust in the home has been identified as one of the major PBDE routes of exposure for sensitive populations like young children and babies, house dust has been extensively sampled. There has been limited work on PBDE concentrations in office air and dust (Harrad, 2010). As additional data on PBDE concentrations in office air are generated, the estimated amounts of PBDEs from office air may change.



[1] Sum of best estimates. Best estimates are either the mean, mid-point, median, or most reasonable estimate for each source.

[a] Mid-point of range

[b] Median

[c] Geometric mean

Figure 10. Total PBDE Release in the Study Area (values shown are kg/yr).

PCDD/Fs

Sources Assessed

Assessment of ongoing PCDD/F releases included the following sources:

- Nonpoint source combustion
- Industrial, military, commercial, and institutional facilities

Other possible sources of PCDD/F release include manufacturing and use of pesticides. Releases from these sources were not assessed.

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs; a.k.a. dioxins) are a class of persistent bioaccumulative compounds that are ubiquitous in the environment at low concentrations. Unlike many other environmental contaminants, PCDD/Fs were not produced intentionally, but instead are formed and released as by-products of industrial production and combustion of certain chlorinated materials. As a result, industrial and combustion sources are responsible for most of the PCDD/F releases.

PCDD/Fs are generally present as mixtures of the 17 compounds that make up the group of toxic dioxins and furans[3]. The concentrations of these mixtures are commonly expressed as the mixture's toxicity translated to that of the most toxic members, TCDD[3] and PeCDD[4]. Each of the 17 PCDD/Fs are assigned a toxicity factor relative to that of TCDD and PeCDD, and the sum of these relative toxicities are termed the toxic equivalent (TEQ). The TEQ of an environmental sample is generally useful shorthand for assessing comparisons to regulatory thresholds and for assessing risks. Sampling and source data are commonly expressed as TEQs, and TEQ is therefore adopted as the convention used for assessing releases in the present document.

One other characteristic also distinguishes PCDD/Fs from most other environmental toxicants: their analysis requires expensive analytical techniques and is only performed by a limited number of commercial laboratories. As a result, they are commonly excluded from screening level investigations and are generally analyzed only if they are the focus of an environmental investigation. Since environmental PCDD/F data are not as prevalent as for other chemicals, there are fewer data on sources of their release to the environment.

Estimates of PCDD/F Release from Backyard Burn Barrels

EPA (2006a) conducted national inventories of PCDD/F releases for the three "reference" years (1987, 1995, and 2000) and concluded that PCDD/F formation in backyard burn barrels has overtaken municipal waste incineration as the major source of dioxin to the environment. This reflects a 100-fold decrease in PCDD/Fs from municipal waste incinerators from 1987 to 2000 due to tightened controls on air emissions and the closing of many plants.

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[3] Compounds with chlorine atoms in the 2,3,7,8 positions

[3] 2,3,7,8-tetrachlorodibenzo-*p*-dioxin

[4] 1,2,3,7,8-pentachlorodibenzo-*p*-dioxin

Other major sources of dioxins nationally have also reduced their dioxin emissions and discharges substantially from 1987 to 2000, including order-of-magnitude decreases by medical waste incinerators, bleached chemical pulp and paper mills, and cement kilns. There are no medical or municipal waste incinerators currently operating in the Study Area (Yake et al., 1998; Gary Palcisko, Ecology Air Quality Program, written communication). Overall PCDD/F releases nationally have decreased from 13,965 g TEQ during 1987 to 1,422 g TEQ during 2000 (EPA, 2006a).

While PCDD/F releases from most major source categories declined substantially from 1987 through 2000, releases from backyard burn barrels nationally have remained relatively consistent from 1987 (604 g TEQ) through 2000 (498 g TEQ). Estimates of PCDD/F releases from backyard burn barrels in the Study Area were scaled from the 2000 national release estimate based on the proportion of the U.S. population residing in the Study Area (1.46%). Estimated PCDD/F releases in the Study Area were 7.3 g TEQ/yr (Table 64).

Table 64. Estimates of Annual PCDD/F Release from Backyard Burn Barrels.

PCDD/Fs Release from Burn Barrels in the U.S. (g TEQ*/yr)	**PCDD/Fs Release from Burn Barrels in the Study Area (g TEQ*/yr)**
498	7.3
Total Estimate:	7.3

*Based on 1998 WHO TEFs

Uncertainty

The largest sources of uncertainty are the assumptions that burn barrel activity and material combusted is consistent from region-to-region, and that these have not changed since 2000. No data were reviewed to evaluate the validity of these assumptions.

It should also be noted that TEQs were calculated using 1998 World Health Organization toxicity equivalency factors (1998 WHO TEFs); in 2005, WHO revised toxic equivalency factors (TEFs) for several compounds (van den Berg et al., 2006). Differences between TEQs calculated using the 1998 WHO TEFs and 2005 WHO TEFs depend on the make-up of the PCDD/F mixture. If, for instance, PCDD/Fs released from backyard burn barrels are dominated by octachlorinated dioxins and furans, a TEQ calculated using the 2005 WHO TEFs would be greater than those reported above because octachlorinated dioxins and furans are assigned triple the toxicity in the 2005 WHO TEFs than in the 1998 WHO TEFs. Conversely, mixtures with a high proportion of pentachlorinated furans would have a slightly lower TEQ using the 2005 WHO TEFs since TEFs assigned to pentachlorinated furans have decreased by 67%.

Estimate of PCDD/Fs Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Releases from industrial, commercial, and institutional facilities were assessed only for the 2008 reporting year. Releases were reported in total grams of PCDD/Fs during previous years, making comparisons to the release estimates expressed as TEQs untenable. During 2008, PCDD/F releases of 0.565 g TEQ/yr were reported for the five TRI categories assessed (Table 65). The highest overall reported PCDD/F releases were from pulp and paper mills, cement plants, petroleum refiners, and wood-treaters.

The Simpson Tacoma Kraft mill was responsible for approximately 63% of total PCDD/F TEQ releases among industrial, commercial, and institutional facilities, with 0.142 g TEQ/yr in stack air emissions and 0.216 g TEQ/yr in releases to water. A comparatively small amount of PCDD/F TEQ was released through fugitive air emissions, with the BP Cherry Point refinery in Whatcom County as the largest emitter (0.001 g TEQ/yr). There were no releases in the "other disposal" category or to POTWs.

Table 65. Estimates of Annual PCDD/F Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 2008.

Release by Facility Type	**Percent of Total**	**PCDD/Fs Release in Study Area (g TEQ*/yr)**
Pulp and Paper Mills	86%	0.49
Cement Plants	8%	0.048
Petroleum Refineries	3%	0.015
Wood-Treaters	<1%	0.00060
All Others	3%	0.015
Total	100%	0.57
Release by Medium	**Percent of Total**	**PCDD/Fs Release in Study Area (g TEQ*/yr)**
Fugitive Air Emissions	<1%	<0.001
Stack Air Emissions	51%	0.29
Surface Water Discharge	48%	0.27
Other On-Site Land Disposal	0%	0.00
Discharge or Transfer to POTWs	0%	0.00
Total	100%	0.57
Total Estimate:		0.57

*Based on 2005 WHO TEFs

In their statewide assessment of PCDD/F sources, Yake et al. (1998) estimated a release to water of 0.15 g TEQ/yr from pulp and paper mills, not including the Georgia Pacific pulp mill in Whatcom County (ceased production around 2000) or the Rayonier mill in Clallam County (closed in 1997). This estimate was fairly close to the 2008 PCDD/F releases to water for pulp and paper mills reported in the 2008 TRI (0.26 g TEQ). Yake et al. (1998) did not estimate air emissions from pulp and paper mills, but their estimate for cement kilns (0.46 g TEQ/yr) was an order of magnitude higher than the 2008 TRI air releases from this source category (0.05 g TEQ), and four times higher for hog fuel boilers (0.06 g TEQ/yr reported by Yake et al.; 0.015 g TEQ in 2008 TRI).

Uncertainty

Criteria and limitations of TRI reporting requirements are in Appendix C.

Estimates of PCDD/Fs Released from Modeled Air Emission Sources

Air emissions of PCDD/Fs inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Emissions from heavy-duty diesel vehicles accounted for more than one-half of the total PCDD/F emissions (0.81 g TEQ/yr), followed by woodstoves and fireplaces (0.38 g TEQ/yr). Emissions of less than 1.0 mg TEQ/yr were also reported, but not included in Table 66.

It appears that none of the large PCDD/F sources discussed previously (burn barrels or industrial facilities) were included among the air emissions included in the 2005 Air Emissions Inventory.

Table 66. Estimates of PCDD/F Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	**PCDD/Fs Release in Study Area (g TEQ*/yr)**
Agricultural Equipment Emissions	0.005
Airport Service Equipment Emissions	0.003
Commercial Equipment Emissions	0.019
Construction Equipment Emissions	0.18
Heavy-Duty Diesel Vehicle Emissions	0.81
Heavy-Duty Gasoline Vehicle Emissions	0.004
Industrial Equipment Emissions	0.030
Lawn and Garden Equipment Emissions	0.010
Light-Duty Diesel Vehicle Emissions	0.037
Light-Duty Gasoline Vehicle Emissions	0.079
Logging Equipment Emissions	0.005
Oil Field Equipment Emissions	0.000
Recreational Boat Emissions	0.011
Recreational Equipment Emissions	0.001
Woodstoves and Fireplaces	0.38
Total Estimate:	1.6

*Based on 2005 WHO TEFs

Uncertainty

Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

PCDD/Fs Released from Other Sources

The ten highest PCDD/F sources reported in the national inventory represent approximately 93% of all releases. All have either (1) been addressed in previous sections (e.g., burn barrels, heavy-duty diesel vehicles, industrial wood [hog fuel] combustion, diesel off-road equipment [agricultural equipment, logging equipment]), (2) are not applicable to the Study Area (municipal waste incineration, medical waste incineration, coal-fired utility boilers, ethylene dichloride/vinyl chloride production, sintering plants), or (3) are not categorized as primary releases for the present report (municipal wastewater treatment plant sludge incineration and land application). Other PCDD/F sources which have had historically high PCDD/F releases, such as pulp and paper mills and cement kilns, have become less prominent nationally, yet release estimates for the Study Area were nevertheless calculated. Therefore, it seems likely that the PCDD/F releases discussed in previous sections account for most of the releases to the Study Area.

While most releases appear to be accounted for, there are a number of minor PCDD/F releases which are poorly defined. However, the sum of these minor sources could potentially become substantial relative to sources currently accounted for.

Two examples are combustion of waste oil mixed with solvents in automotive shop space heaters, and incidental PCDD/F formation during production of certain pesticides. While the latter has been known for some time – PCDD/F impurities in herbicides used in Agent Orange is probably the best known example – the issue was recently re-visited by investigators in Australia (Holt et al., 2010) to determine the current level of PCDD/F impurities and the implications for PCDD/F loading at a national scale. They found substantial PCDD/F concentrations in the pesticide pentachloronitrobenzene (not commonly used in Washington) and lesser concentrations in 2,4-D (commonly used in Washington) among other pesticides. At the national loading scale in Australia, PCDD/Fs from pesticide impurities are potentially as large as major known sources (e.g. waste incineration, pulp and paper mills).

In Washington, a study to assess PCDD/Fs in ash from the combustion of mixed waste oil in automotive shop space heaters was conducted to characterize the toxicity to fish as well as to determine appropriate disposal methods for the material (Delistraty and Stone, 2007). Although the study was designed neither to estimate the occurrence of waste oil burning nor to estimate fugitive PCDD/F emissions, the results of PCDD/Fs in ash residue suggest that this is potentially a large source of PCDD/Fs.

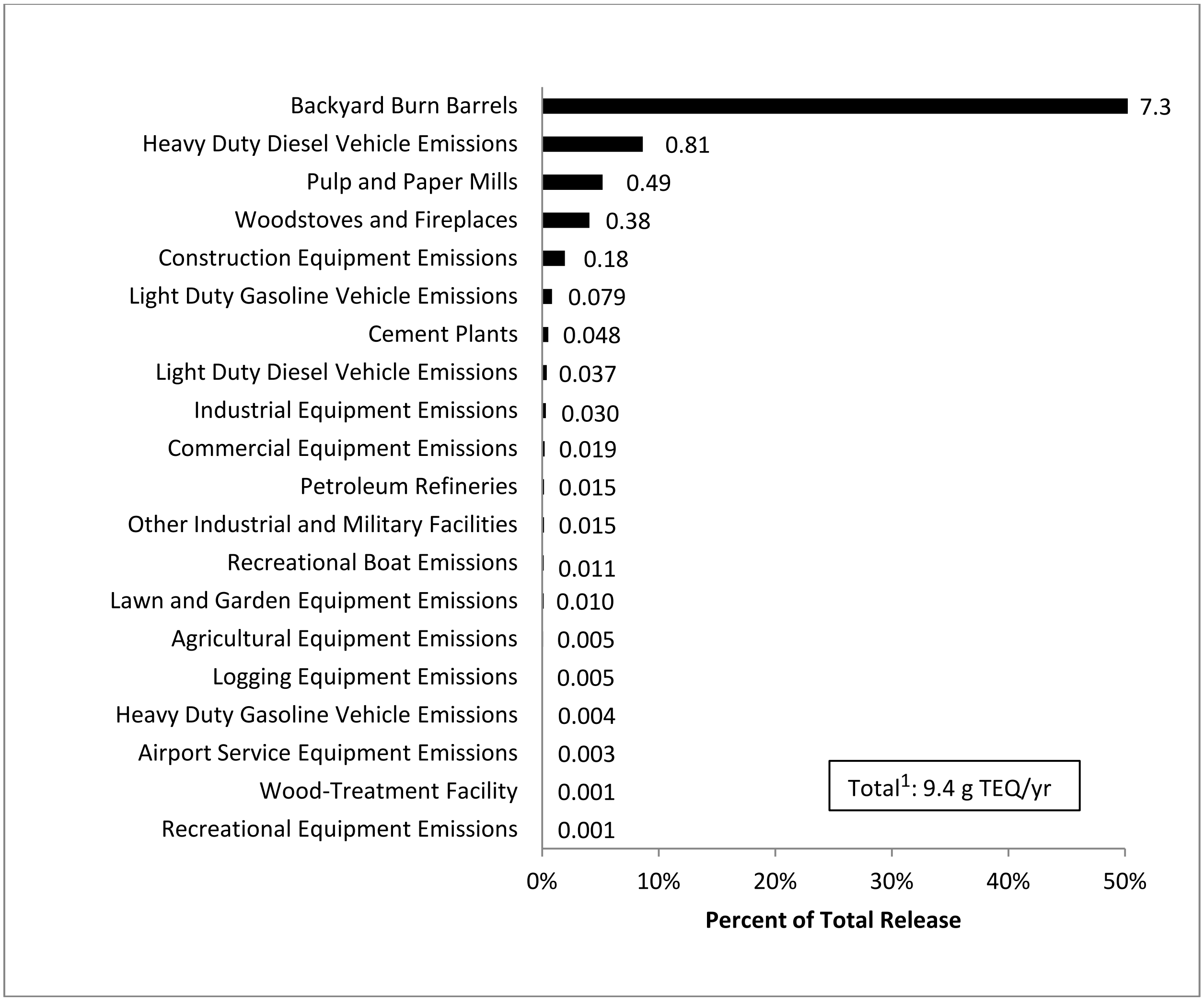
These studies both illustrate that diffuse, unregulated, or minimally regulated PCDD/F sources are potentially significant and are not likely to be captured in conventional estimates. Compounding the uncertainty about these sources is the high expense of dioxin analysis which tends to discourage research or screening-level analysis as an investigative option.

Summary of PCDD/F Release Estimates

Based on the best estimates available, total PCDD/F releases from the sources assessed are approximately 9.4 g TEQ/yr (Figure 11). The largest single source is backyard burn barrels, accounting for three-quarters of the PCDD/F release to the Study Area.

Combustion emissions to air account for 97% of the dioxin release. The remaining release is to water, primarily from pulp and paper mills. This represents a substantial change from the previous two-to-three decades when PCDD/F discharges to water from pulp mills represented the bulk of all releases to Washington State (EPA, 1991).

The largest PCDD/F release estimates were derived from national estimates scaled to the Study Area by population. No data were found to assess whether population is an appropriate scaling factor for these circumstances, and the accuracy of these release estimates are therefore highly uncertain.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

Figure 11. Total PCDD/F Release in the Study Area (values shown are g TEQ/yr).

PAHs

Sources Assessed

Assessment of ongoing PAH releases included the following sources:

- Creosote-treated railroad ties, utility poles, and marine pilings
- Petroleum leaks, spills, and improper disposal of oil
- Vehicle tire wear
- Roofing material runoff
- Asphaltic and coal-tar based pavement erosion and leaching
- Nonpoint source combustion
- Industrial, military, commercial, and institutional facilities

Other possible sources of PAH release include improper disposal of paints and wood preservatives, gasoline spills, jet and aviation fuel spills and combustion, petroleum products such as specialty oils and greases, combustion of logging debris and shooting targets. Releases from these sources were not assessed due to time limitations.

Polycyclic aromatic hydrocarbons (PAHs) are a class of compounds characterized by two or more fused aromatic rings composed of carbon and hydrogen. There are hundreds of such compounds, but most studies have focused on 16 compounds that were designated as “priority pollutants” in the federal Clean Water Act. In size order, these 16 compounds are:

- Naphthalene
- Acenaphthylene
- Acenaphthene
- Fluorene
- Phenanthrene
- Anthracene
- Fluoranthene
- Pyrene
- Benzo(a)anthracene*
- Chrysene*
- Benzo(b)fluoranthene*
- Benzo(k)fluoranthene*
- Benzo(a)pyrene*
- Dibenzo(a,h)anthracene*
- Benzo(g,h,i)perylene
- Indeno(1,2,3-c,d)pyrene*

The seven asterisked PAHs have been designated by EPA as probable human carcinogens. Most estimates of PAHs in this report refer to these 16 PAHs or these 16 plus perylene. Perylene was analyzed in addition to the 16 priority pollutant PAHs in a comprehensive study of PAH sources to the New York/New Jersey (NY/NJ) Harbor (Valle et al., 2007), which provided a much cited resource for estimates presented here.

Estimate of PAHs Released from Creosote-Treated Railroad Ties

Estimates of PAH releases from creosote-treated railroad ties were derived from release rates published in a 2007 study of PAH sources to New York/New Jersey Harbor and estimates of the number of railroad ties in the Study Area.

The authors of the NY/NJ Harbor study estimated a PAH loss rate from creosote-treated railroad ties of 16.67 g/tie/yr over the lifetime of a tie (Valle et al., 2007). Assumed lifetimes were 30 years for railroad ties. Losses modeled by Valle et al. (2007) were equally split between air emissions and rainwater.

According to WSDOT (Blake, 2007) there are 3,196 miles of freight rail lines in Washington and 794 miles in the Study Area. Using an average of 3,249 ties per mile of track (RTA, 2010), there are a total of 2,579,186 ties in the Study Area. The PAH release rate applied to the total number of creosote-treated railroad ties results in an estimated 43 t/yr of PAH released to the Study Area (Table 67).

Table 67. Estimates of Total PAH Release Due to Emissions and Leaching from Creosote-Treated Railroad Ties.

Product	Number in Study Area	PAH Release Rate (kg/tie/yr)	Total PAH Release in Study Area (t/yr)
Railroad Ties	2,579,186	0.01667	43
		Total Estimate:	43

t: metric ton

Uncertainty

Creosote-treating of railroad ties is EPA-approved. This remains the primary treatment option for railroad ties, although concrete ties now comprise 6.5% of the market share in North America, and the American Wood Protection Association has recently approved ammoniacal copper zinc arsenate as an accepted preservative for ties (RTA, 2010).

Railroad ties from inactive rail lines are not included in the estimate, which may under-estimate actual PAH releases. Blake (2007) indicates that in addition to the 794 miles of active track, there are approximately 485 mile of railway in the Study Area that have been abandoned. Although abandoned railways may have had ties removed or may contain old ties no longer leaching PAHs, there may be some abandoned railroad where PAH losses from ties continue to occur.

Estimate of PAHs Released from Creosote-Treated Marine Pilings

Estimates of PAH releases from creosote-treated marine pilings were derived from release rates published in a 2007 study of PAH sources to New York/New Jersey Harbor and estimates of the number of marine pilings in the Study Area.

Rate of PAH loss from marine pilings is complex due to differences in exposure media (air, water, sediment) for each piling, and lack of uniformity in size and age for each piling. The authors of the NY/NJ Harbor study estimated a PAH loss rate from creosote-treated marine pilings based on the amount of PAH in creosote-treated marine pilings and lifetime losses of PAH using different approaches from two studies. Leaching data from both studies were used in the NY/NJ Harbor report to estimate long-term releases based on a first-order decay model and a diffusion-based model leading to a final estimate of 23% loss. For this report we used the same value of 23% loss of PAH over the 30-year lifetime of a marine piling. (Valle et al., 2007).

Dimensions of marine pilings vary due to their application; ranging from small diameter pilings used for private docks and finger piers in shallow water, to large pilings used for dolphins, wing walls, and navigation aids in relatively deep waters. Based on WSDOT's Creosote Removal Initiative for state ferry terminals, the average piling removed was 63.5 ft^3 (WSDOT, 2010). According to Valle et al. (2007), 8% of a typical piling is exposed to air (5.1 ft^3), 62% is exposed to water (39.4 ft^3), and 30% is buried below sediment (19.0 ft^3). Applying the PAH loss rates from Valle et al. (2007) to the mean size piling in Puget Sound yields a loss rate of 0.54 kg PAH/piling/yr, with 0.062 kg/piling/yr to air and 0.48 kg/piling/yr to water.

No data were found on the numbers of pilings in the Study Area. According to the U.S. Census Bureau, there are 79 marinas in the Puget Sound area. Many of the marinas still contain creosote pilings, but the sum of these is not known. Creosote pilings used for commercial, industrial, and recreational piers likely numbers in the tens of thousands.

Permits are required to install marine pilings, and creosote-treated wood is currently not allowed for new installations, so new use of creosote-treated pilings in Puget Sound is minimal. Creosote piling removal is being conducted by a number of organizations. Washington State Department of Natural Resources had removed 6,756 pilings as of April 27, 2010 and plans to remove thousands more (Lisa Kaufman, Washington Department of Natural Resources, personal communication). WSDOT's Creosote Removal Initiative removed 1.5 million board feet of pilings and timber (approximately 2,000 pilings) from its ferry terminals, representing 10% of the amount planned for removal in the coming decade (WSDOT, 2010).

Given the rate of ongoing piling removal, the number of marinas in Puget Sound, and the amount of industrial, commercial, and recreational waterfront, a rough estimate of 100,000 creosote-treated pilings remaining in the Study Area appears reasonable, although this estimate may differ by several factors above or below the actual number.

Based on an estimate of 100,000 creosote-treated pilings in the Study Area and a PAH release rate of 0.062 kg/piling/yr to air and 0.48 kg/piling/yr to water, the total release of PAHs from creosote-treated pilings is estimated to be 54 t/yr (Table 68).

Table 68. Estimates of Total PAH Release Due to Emissions and Leaching from Creosote-Treated Marine Pilings.

Product	**Number in Study Area**	**PAH Release Rate (kg/piling/yr)**	**Total PAH Release in Study Area (t/yr)**
Marine Pilings	100,000	0.062 (air) 0.48 (water)	6.2 (air) 48 (water)
		Total Estimate:	54

Uncertainty

There is a low level of certainty in the PAH release estimate for marine pilings primarily due to uncertainty surrounding the number of marine pilings in the Study Area. There are no known surveys to estimate the number of pilings on a large geographic scale, although there may be methods available to obtain piling numbers. Shoreline Master Plans may contain an inventory of pilings by type (e.g. wood, steel, concrete) at a local level. Habitat restoration efforts may also contain documentation of piling numbers, also at a local level. GIS coverages of over-water structures may be useful to fill in data about open water pilings. These local data and GIS sources could be used to patch together a fairly accurate number of creosote-treated pilings given a large dedicated effort.

PAH release rates from creosote pilings should also be investigated to a greater extent. As mentioned previously, the rate of PAH release is complex, and additional loss rate estimates should be obtained. Since the rate of loss is dependent on factors such as age and size of piling, temperature of air and water, and the surface area exposed to water and air, rate loss calculations may require more sophisticated modeling than applying simple numerical rate values.

Estimate of PAHs Released from Creosote-Treated Utility Poles

Estimates of PAH releases from creosote-treated utility poles were derived from release rates published in a 2007 study of PAH sources to New York/New Jersey Harbor and estimates of the number of utility poles in the Study Area.

The authors of the NY/NJ Harbor study estimated a PAH loss rate to air from creosote-treated utility poles of 212 g/pole/yr over the lifetime of a pole (Valle et al., 2007). Assumed lifetimes were 35 years for utility poles.

Feldman and Shistar (1997) reported 333,886 poles owned by Puget Sound Energy. Based on the number of customers served by Puget Sound Energy (1.1 million; Puget Sound Energy, 2010), the number of utility poles owned by the remaining electrical service providers in the Study Area were extrapolated by scaling to the number of customers (0.93 million) served by the remaining providers (Clallam County PUD, 2010; Mason County PUD 1, 2010; Mason County PUD 3, 2010; Seattle City Light, 2009; Snohomish County PUD, 2010; Tacoma Power, 2010). The total number of utility poles in the Study Area is estimated at 617,765.

Based on the assumption that the percentage of utility poles treated with creosote in the Study Area is identical to that of the NY/NJ Harbor (13%; Valle et al., 2007), the total number of creosote-treated poles in the Study Area is 80,309. The PAH emission rate applied to the total number of creosote-treated poles results in an estimated 17 t/yr of PAH released to the Study Area (Table 69).

Table 69. Estimates of Total PAH Release Due to Emissions from Creosote-Treated Utility Poles.

Product	Number in Study Area	PAH Release Rate (kg/pole/yr)	Total PAH Release in Study Area (t/yr)
Utility Poles	80,309	0.212	17
		Total Estimate:	17

Uncertainty

The number of utility poles in the Study Area may be slightly under-estimated; San Juan County and eastern Jefferson County were not included in the inventory of poles. In addition, the assumption that the same rate of creosote treatment of poles in the Study Area is the same as for NY/NJ Harbor (13%) has not been validated.

Valle et al. (2007) applied railway tie PAH release rates to utility poles, due to the lack of specific peer-reviewed studies on long-term release information for utility poles. While this seems reasonable, it is possible that the two uses of creosote-treated wood have different emissions.

Estimate of PAHs Released from Petroleum Spills, Leaks, and Improper Disposal of Used Motor Oil

PAHs released through large petroleum spills (≥ one gallon) and minor oil spills, as well as leaks and drips of crankcase oil from vehicles and from improper disposal of used motor oil following oil changes, was estimated. Concentrations of individual PAHs from one or more petroleum materials were available in the literature and cited in the NY/NJ Harbor report (Valle et al., 2007). These concentrations are shown in Appendix K. Motor oil and other forms of petroleum released to the environment were calculated using the methodology and assumptions discussed in the subsequent *Petroleum* section. The quantities of petroleum released from various sources and associated PAH releases are shown in Table 70.

The composition of individual PAHs among crude oil and refined petroleum products is inconsistent, although naphthalene is generally found at the highest concentrations. Total PAH concentrations are highest in diesel and heating oil (8,111 mg/kg), lowest in crude oil (497 mg/kg), and at intermediate concentrations in asphalt and lubrication/motor oils (1,012 – 1,397 mg/kg). Approximately 93% of the petroleum-related PAH release was due to release of motor oil through leaks and drips and through improper disposal. The total release of PAH from petroleum sources was estimated to be 11 t/yr based on the sum of PAHs released from all sources and materials combined.

Table 70. Estimates of Total PAH Release Due to Petroleum Spills, Leaks, and Improper Disposal of Used Motor Oil.

Material	Total PAH Conc. (mg/kg)	Quantity Released Annually in Study Area from Spills ≥ one gallon (t/yr)		Quantity Released Annually in Study Area from Leaks and Drips (t/yr)		Quantity Released Annually in Study Area from Improper Disposal of Used Motor Oil (t/yr)		Total PAH Release in Study Area (t/yr)
		Petroleum	**PAH**	**Petroleum**	**PAH**	**Petroleum**	**PAH**	
Diesel, Kerosene, and Heating Oil	8,111	47.8 (b)	0.39	nr	nr	nr	nr	0.39
Crude Oil	497	8.30 (b)	0.004	nr	nr	nr	nr	0.004
Lube and Motor Oil	1,012	2.93 (b)	0.003	nr	nr	nr	nr	0.003
Other Petroleum	4,069 (a)	102 (b) - 176 (c) 139 (mean)	0.42 - 0.72 0.57 (mean)	nr	nr	nr	nr	0.42 - 0.72 0.57 (mean)
Used Motor Oil	1,375	nr	nr	6,100	8.5	970	1.4	9.9
Total Estimate:								11 (best estimate)

nr : not reported

(a) Mean of PAH concentrations in diesel and heating oil, crude oil, lubricating and motor oil, and No. 6 heavy oil (asphalt).

(b) Sum of RDA and Environmental Report Tracking System (ERTS) estimates (see Table 91)

(c) Sum of Environmental Research Consulting (ERC) and ERTS estimates (see Table 91)

Uncertainty

There is a substantial degree of uncertainty about PAH releases due to the lack of additional data on PAH concentrations in petroleum, as well as uncertainties surrounding petroleum release rates. Estimates here rely heavily on the NY/NJ Harbor report (Valle et al., 2007) for data on PAH concentrations in petroleum materials. While this report appears to have comprehensive data on PAH sources, confidence in the reported values for PAH concentrations would be improved through a more extensive literature search. One notable omission in the PAH concentrations is the lack of data for gasoline and jet and aviation fuels.

In addition to uncertainty about PAH concentrations, the volume of petroleum released also has associated uncertainty. These uncertainties are discussed in detail in the Petroleum section.

Estimates of PAHs Released from Vehicle Tire Wear

Releases of PAH due to vehicle tire wear were calculated using estimates of tire wear rates and PAH concentrations reported in tire material, then scaled to the Study Area using annual vehicle kilometers travelled (VKTs) for the 12-county Puget Sound region. A complete description of the methodology, assumptions used, and the variables applied are in Appendix E.

Table 71 shows the variables used to calculate PAH release from tire wear. A wear rate of 38 mg/km travelled was assumed for all vehicle types considered, with differences being only the number of tires per vehicle. PAH concentrations in tire material were estimated to be 89 mg/kg based on data reported by Valle et al. (2007). Most of the PAH in tire material is pyrene (54 mg/kg), followed by phenanthrene (12 mg/kg), fluoranthene (11 mg/kg), chrysene (8 mg/kg), and benzo(a)pyrene (4 mg/kg). The estimates of total PAH release from tire wear is 0.98 t/yr.

Table 71. Estimates of Total PAH Release from Vehicle Tire Wear.

Vehicle Type	**Number of Tires per Vehicle**	**Tire Wear Rate (mg/km/tire)**	**Total PAH Concentration in Tire Material (mg/kg)**	**Vehicle Kilometers Travelled in Study Area (km/yr)**	**PAH Release in Study Area (t/yr)**
Motorcycle	2	38	89	2.3E+08	0.002
Passenger Car	4			3.6E+10	0.49
Light Truck	4			1.5E+10	0.20
Bus	8			1.2E+08	0.003
Single Unit Truck	8			2.8E+09	0.075
Combination Truck	18			3.5E+09	0.21
				Total Estimate:	0.98

Uncertainty

Overall estimates of tire wear rates, PAH concentrations in tire material, and VKTs appear to be reliable since the estimates are based on published information and WSDOT data (see Appendix E). Tire wear rates reported in the literature appear to be realistic when checked against what may be considered a reasonable tread loss over the life of a tire (see Appendix E for this analysis). However, both the tire wear rates and PAH concentrations may vary considerably based on tire brand, with wear rates also strongly influenced by driving conditions. Although the VKTs are based on the 12-county Puget Sound region, they are a reasonable proxy for the Study Area.

Estimate of PAHs Released from Roof Runoff

PAH concentrations have been reported in roof runoff isolated from residential areas and presumed to stem primarily from asphalt shingles (Steuer et al., 1997). The mean PAH concentration in runoff from asphalt shingles was 0.61 ug/l. Steuer et al. (1997) also reported a mean PAH concentration of 2.06 ug/l in runoff isolated from commercial areas where roofing material was typically composed of flat-rubberized or tar-sealed roofs, which has been categorized as built-up roofing for estimated releases calculated here.

Steuer et al. (1997) reported roof runoff concentrations for 11 PAHs (nine high molecular weight PAHs and two low molecular weight PAHs), all of which were detected in both residential and commercial areas except for anthracene which was not detected in runoff from either area. None of the individual PAHs had concentrations substantially higher than any other. The phenanthrene concentration (0.24 ug/l) was highest in residential (asphalt shingle) runoff while fluoranthene was found at the highest concentration in commercial (built-up roofing) areas (0.48 ug/l). Interestingly, concentrations of individual PAHs in runoff from residential driveways reported by Steuer et al. (1997) were nearly identical to concentrations from the built-up roofs, possibly suggesting similar materials with similar leaching rates used in each application.

Annual roof runoff volumes for each asphalt shingle and built-up roof type were used to translate PAH concentrations in runoff into release loads. The method for the calculation of runoff volumes is described in Appendix B. Total annual PAH releases for asphalt shingle and built-up roofs were 0.36 and 0.21 t/yr, respectively (Table 72). The combined load was 0.57 t/yr.

Table 72. Estimates of Total PAH Release from Roofing Material.

Roof Type	Total Area of Roof Type in Study Area (m^2)	Total Runoff Volume of Roof Type in Study Area (l/yr)	Total PAH Concentrations in Runoff (ug/l)	Total PAH Release in Study Area (t/yr)
Asphalt Composite	3.96×10^{8}	5.84×10^{11}	0.61	0.37
Built-up	7.40×10^{7}	1.03×10^{11}	2.06	0.21
			Total Estimate:	0.57

Uncertainty

The greatest uncertainty in estimates of PAH released from rooftops is most likely due to scarcity of PAH release data in the literature. Uncertainty is compounded due to the lack of accounting for PAHs in atmospheric deposition in the available literature. Roofing age, unaccounted for here, may also play a factor, as release rates may vary over the life of a roof. Runoff concentrations used to derive release estimates could therefore potentially result in an upward bias of results.

Scale-up information for estimates of PAH releases from roof materials appears to be fairly reliable. Accuracy may be increased by further collection of information on roof type fractions from counties in addition to Pierce and Snohomish Counties.

Estimate of PAHs Released from Cigarette Smoke

PAH emissions were estimated using *per-cigarette* PAH emission factors provided by Valle et al. (2007) and various sources of information on the number of cigarettes consumed in the Study Area.

Seven individual PAHs are reported in cigarette smoke, with phenanthrene present at the highest level (2.6 ug/cigarette; Valle et al., 2007). The total PAH quantity was reported to be 6.79 ug/cigarette. Appendix L shows emissions rates for individual PAHs.

Two estimates of cigarette numbers consumed annually in the Study Area were calculated using data from Washington State Departments of Health (WDOH) and Revenue (DOR). WDOH estimated there were 840,000 smokers in Washington State (WDOH, 2010). Scaling to the Study Area by population (U.S. Census Bureau, 2010) yielded a Study Area smoker number of 564,000. The Centers for Disease Control estimates that each smoker consumes 16.8 cigarettes per day (CDC, 2005), yielding a total Study Area consumption rate of 3.46 billion cigarettes/yr.

A higher total cigarette consumption rate is provided by DOR which reported a state per capita consumption of 1,106 cigarettes/yr during 2006 (DOR, 2007). Scaled up to the Study Area by population (U.S. Census Bureau, 2010) yielded a Study Area consumption rate of 4.95 billion cigarettes/yr.

Estimated release rates based on PAH emissions and cigarette consumption rates are shown in Table 73. PAH release estimates ranged from 0.024 to 0.034 t/yr. The mid-point of this range was 0.029 t/yr.

Table 73. Estimates of Total PAH Release from Cigarette Smoke.

Cigarette Consumption in Study Area (cigarettes/yr)	Total PAH Emission Rate (ug/cigarette)	Total PAH Release in Study Area (t/yr)
$3.46 \times 10^9 – 4.95 \times 10^9$ 4.21×10^9 (mid-point)	6.79	0.024 – 0.034 0.029 (mid-point)
	Total Estimate:	0.024 – 0.034 0.029 (mid-point)

Uncertainty

Little is known about the accuracy of the PAH emission rates from cigarette smoke. Furthermore, only seven of the individual PAHs have reported emission rates. If the emission rates are accurate for the PAHs with reported data, emission rates for total PAHs may be an under-estimate if additional PAHs are measured.

Scale-up information for cigarettes appears to be reasonably accurate. Consumption data collected with either DOR or WDOH survey goals in mind yielded results with a relative percent difference of 35%. Although DOR includes estimates of illegal cigarette sales (24% of total sales) in its estimate of per capita consumption, this figure is a source of imprecision in the overall estimate by DOR. However, error in the illegal sales estimates would not substantially change the estimate of overall PAH releases from cigarette smoke.

Estimates of PAHs Released from Asphalt Pavement

An estimate of PAHs released from the wearing and leaching of asphalt pavement was calculated using data on PAH concentrations in asphalt, as well as leaching and erosion rates found in the published literature. Release rates were then scaled up to the asphalt pavement surface in the Study Area calculated using several sources of information.

Asphaltic concrete pavement consists of a mixture of rock or sand aggregate and the asphalt itself, which is the heavy undistillable portion of petroleum which contains PAHs. The asphalt content of pavement generally ranges from 5-10% by weight, with a mean content of 7.2% (Kupiainen, 2007; Simon and Sobieraj, 2006; Wess et al., 2004). Birgisdottir et al. (2007) found that mean PAH concentrations in four asphaltic pavement samples averaged 1.79 mg/kg. Kriech et al. (2002) analyzed PAH concentrations in six different samples of asphalt and found a mean concentration of 19.6 mg/kg; when coupled with the typical asphalt composition of pavement (7.2%), the resulting concentration in pavement was 1.4 mg/kg.

Pavement erosion rates were obtained from Kupiainen (2007) and classified into two groups: 6.87 mg pavement per vehicle kilometer travelled (VKT) for light vehicles (motorcycles, passenger cars, and light trucks), and 33.5 mg/VKT for heavy vehicle (single-unit trucks, busses, and combination trucks). These erosion rates were applied to VKTs for each class as previously described in discussions of brake and tire wear. Total erosion of pavement in the Study Area was estimated to be 567 t/yr. PAH release through pavement erosion was calculated by applying PAH concentrations to pavement erosion (approx. 0.0009 t/yr, Table 74).

PAH leaching rates were obtained from modeled leaching rates for five PAHs based on diffusion coefficients measured from tank leaching tests (Birgisdottir et al., 2007). Since the leaching rates were modeled for a 25-year period, the reported leaching rate for total PAHs (0.554 mg/m^2) was divided by 25 to yield an annual leach rate of (0.022 mg/m^2).

Total surface area of pavement in the Study Area was estimated by summing the areas covered by roads (Envirovision et al., 2008) and other pavemented surfaces such as parking lots. The surface area of non-road pavement such as parking lots (87.60 square miles; 230 million square meters) was estimated by subtracting roads (255.94 mi^2; 660 million m^2) and rooftops (214.51 mi^2; 560 million m^2; see calculation of rooftop surfaces in Appendix B) from the entire impervious surface of the Study Area (558.06 mi^2; 1.45 x 10^9 m^2) (NOAA, 2006). The leaching rate for PAHs was applied to the total pavemented area to yield an estimated PAH release of 0.020 t/yr due to leaching from asphalt pavement (Table 74).

Table 74. Estimates of Total PAH Release from Asphalt Pavement Wear and Leaching.

Release Process	**Pavement Erosion in Study Area (t/yr)**	**Total PAH Concentration of Pavement (mg/kg)**	**Pavement Surface in Study Area (m^2)**	**Total PAH Leaching Rate for Pavement (mg/m^2)**	**Total PAH Release in Study Area (t/yr)**
Erosion	567	1.4 – 1.79	--	--	0.0008 – 0.001 0.0009 (mid-point)
Leaching	--	--	8.9 x 10^8	0.022	0.020
				Total Estimate:	0.021

Uncertainty

It is not known if any of the variables used to calculate Study Area releases (erosion rates, leaching rates) are accurate for the Study Area, although the data were obtained from reliable sources. Scaling information is as accurate as reasonably achievable since it is based on well-documented WSDOT traffic data and standardized GIS coverage data.

One shortcoming of these release estimates is the limited set of PAHs analyzed for leaching rates. Although the five PAHs used for modeled leaching rate estimates – fluoranthene, fluorene, naphthalene, phenanthrene, and pyrene – appeared to have the highest leach rates, leaching rate estimates derived for other PAHs may have substantially increased the PAH release estimate for asphalt. In addition, the assumption of a consistent and equal leaching rate over a 25-year period is likely an unreasonable assumption, but no data are available to provide an improved estimate of changes in leaching rates over time.

Estimates of PAHs Released from Coal-Tar and Asphalt Sealants

Coal-tar and asphalt-based sealants are black liquids sprayed or painted onto asphalt parking lots and driveways to protect the asphalt from damage caused by UV degradation, gas and oil, and water. Coal tar is a by-product formed during the carbonization and gasification of coal, processes used to generate coke and coal gas. Asphalt is derived from the refinement of crude petroleum (Mahler et al., 2005).

Coal tar-based sealants represent a far more significant PAH source than do asphalt-based versions. Studies published by the USGS first identified coal-tar sealants as a potentially important PAH source in 2005 (Mahler et al., 2005). The City of Austin, Texas found a median PAH content in asphalt-based sealants of 50 ppm, while that of coal-tar sealants was over 50,000 ppm (City of Austin, 2005-Draft).

Subsequent research indicated that PAHs from sealants posed a more significant problem east of the Rockies. PAH concentrations in dust and runoff from seal-coated pavement east of the Rockies were a thousand times higher than in western cities, including Seattle (Van Metre et al., 2008). This was expected, since coal-tar sealants are more commonly used east of the Rockies and asphalt-based sealants are more commonly used west of the Rockies. Furthermore, PAH levels in runoff from both asphalt-sealed and unsealed parking lots were similar, indicating that the asphalt sealant itself was not a major source (Van Metre et al., 2008).

A 2010 paper (Van Metre and Mahler) expanded their analysis of PAH sources to 40 urban lakes across the U.S. that are among those sampled by the USGS National Water Quality Assessment (NAWQA) Program. Surprisingly, this recent study suggests that about half of PAHs in sediments in urban lakes come from coal-tar sealants, including two urban lakes in western Washington. However, the total levels of PAHs were consistently much lower west of the Rockies.

Looking at the New York Harbor watershed, Valle et al. (2007) estimated annual sales of 1.4 million gallons of coal-tar sealant. Using release rates from Mahler et al. (2005), and assuming a constant release rate for all annual storm events, they estimated an annual release of 900-5800 kg for the NY Harbor, or 0.64 to 4.14 g of PAHs per gallon of sealant.

Exact coal-tar sealant usage estimates for Washington were not found, but industry estimated a usage of 400,000-600,000 gallons in 2004 (WSDOT, 2007). Annual use may have diminished, as preference has shifted toward a blended product of 20% coal-tar pitch and 80% asphalt emulsion, which greatly reduces the release of PAHs (WSDOT, 2007). WSDOT itself has historically used less than 3,000 gallons of coal-tar sealants per year. In 2011, Governor Christine Gregoire signed into law a ban on coal-tar (defined as having PAH content > 1%) sealants that prohibits the sale of these products after January 1, 2012 and prohibits their use after July 1, 2013.

In order to estimate PAH releases to the Study Area, the estimated use of coal-tar sealants statewide (400,000 – 600,000 gallons/year) was scaled to the Study Area by using the Study Area fraction of the state population (0.672; U.S. Census Bureau, 2010). Applying the release rates calculated in Valle et al. (2007) results in approximately 0.17 – 1.7 t of PAHs entering Study Area runoff from coal-tar sealants annually (Table 75).

Table 75. Estimates of Total PAH Release in Runoff from Coal-Tar Sealants.

Coal-Tar Sealants Used in Study Area (gallons)	**PAH Contribution to Runoff (g/gallon of sealant)**	**Total PAH Release in Study Area (t/yr)**
270,000 – 400,000	0.64 – 4.14	0.17 – 1. 7 0.92 (mid-point)
	Total Estimate:	0.17 – 1. 7 0.92 (mid-point)

Uncertainty

This estimate may be high since it does not account for a shift toward asphalt-based sealants and coal-tar asphalt mixtures. Also, this estimate does not account for volatilization of PAHs from the sealant.

Estimate of PAHs Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Industrial, commercial, and institutional facilities reported mean annual total PAH releases of 8.8 t/yr for the five TRI categories assessed (Table 76). The highest overall reported releases were from paper mills, aluminum plants, and petroleum refineries.

Air emissions of PAHs accounted for 95% of PAH release from industrial facilities among the categories assessed. Tesoro Refining in Skagit County and Intalco Aluminum Corp. in Whatcom County had the largest fugitive and stack air PAH emissions, at 0.8 t/yr and 2.2 t/yr, respectively. The Shell Oil Refinery in Skagit County had the largest mean release to water (0.01 t/yr). Petroleum refineries and terminals accounted for nearly all of the release through the “other disposal” category, with the BP Cherry Point Refinery in Whatcom County as the largest releaser (0.2 t/yr). Only one facility reported PAH releases to POTWs: the ConocoPhillips Renton Terminal (0.0001 t/yr).

Table 76. Estimates of Mean Annual PAH Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 1999-2008.

Release by Facility Type	**Percent of Total**	**PAH Release in Study Area (t/yr)**
Pulp and Paper Mills	36%	3.2
Aluminum Mills	31%	2.7
Petroleum Refineries	26%	2.3
All Others	7%	0.58
Total	100%	8.8
Release by Medium	**Percent of Total**	**PAH Release in Study Area (t/yr)**
Fugitive Air Emissions	27%	2.4
Stack Air Emissions	69%	6.1
Surface Water Discharge	<1%	<0.088
Other On-Site Land Disposal	4%	0.35
Discharge or Transfer to POTWs	<1%	0.0001
Total	100%	8.8
Total Estimate:		8.8

Uncertainty

In addition to the criteria and limitations of TRI reporting requirements described in Appendix C, the list of PAHs used for TRI reporting varies from the list of “priority pollutant” PAHs, making direct comparisons imprecise. The TRI reports anthracene, benzo(g,h,i)perylene, naphthalene, and phenanthrene, in addition to 21 additional polycyclic aromatic compounds (PACs)[4]. Of the 21 PACs, only six are also Clean Water Act “priority pollutant” PAHs. Since there is no method available for assessing the individual constituents of the TRI PAC list, direct comparisons between the TRI PACs and the “priority pollutant” PAHs cannot be made.

Estimates of PAHs Released from Modeled Air Emission Sources

Air emissions of PAHs inventoried for the 12-county Puget Sound region were reported by Ecology’s Air Quality Program (Ecology, 2007a). Woodstoves accounted for the greatest releases (110 t/yr), followed by emissions from light-duty and heavy-duty vehicles using gasoline (40 t/yr) (Table 77). Total PAH emissions in the Air Emissions Inventory totaled 170 t/yr.

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[4] Benz(a)anthracene; Benzo(b)fluoranthene; Benzo(j)fluoranthene; Benzo(k)fluoranthene; Benzo(j,k)fluorene; Benzo(r,s,t)pentaphene; Benzo(a)phenanthrene; Benzo(a)pyrene; Dibenz(a,h)acridine; Dibenz(a,j)acridine; Dibenzo(a,h)anthracene; 7H-Dibenzo(c,g)carbazole; Dibenzo(a,e)fluoranthene; Dibenzo(a,e)pyrene; Dibenzo(a,h)pyrene; Dibenzo(a,l)pyrene; 7,12-Dimethylbenz(a)-anthracene; Indeno(1,2,3-cd)pyrene; Methylcholanthrene; 5-Methylchrysene; 1-Nitropyrene

Table 77. Estimates of Total PAH Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	**Total PAH Release in Study Area (t/yr)**
Agricultural Equipment Emissions	0.044
Airport Service Equipment Emissions	0.017
Commercial Equipment Emissions	2.0
Construction Equipment Emissions	1.3
Consumer/Commercial Solvent Emissions	Excluded
Gas Station Emissions	1.2
Heavy-Duty Diesel Vehicle Emissions	1.8
Heavy-Duty Gasoline Vehicle Emissions	11
Industrial Equipment Emissions	0.30
Lawn and Garden Equipment Emissions	5.0
Light-Duty Diesel Vehicle Emissions	0.21
Light-Duty Gasoline Vehicle Emissions	29
Locomotive Emissions	0.49
Logging Equipment Emissions	0.05
Point Sources - Industrial, Commercial, Institutional (primarily Title V AOP)	5.2
Railroad Maintenance Equipment Emissions	0.002
Recreational Boat Emissions	0.86
Recreational Equipment Emissions	0.94
Residential Fuel Use, except Wood	0.038
Residential Trash Burning	6.5
Residential Yard Waste Burning	0.15
Woodstoves and Fireplaces	110
Total Estimate:	173

Excluded: excluded from estimates due to questionable data
AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

Uncertainty

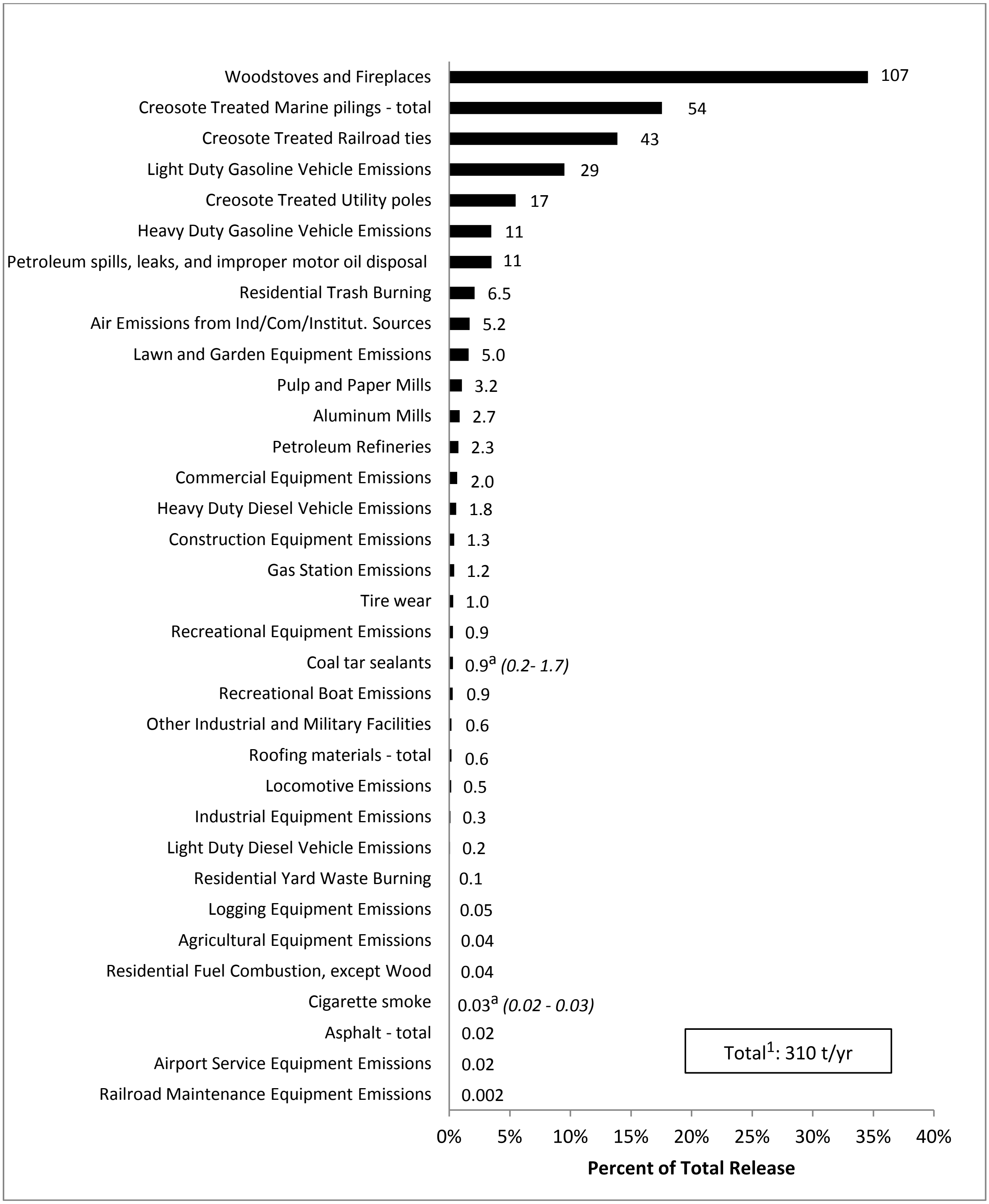
Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of PAH Release Estimates

Total annual PAH releases from sources inventoried total approximately 310 t/yr in the Study Area (Figure 12). Generally speaking, the releases can be placed into two categories: combustion emissions and releases from creosote-treated wood. Combustion releases account for slightly more than one-half of the PAH release in the Study Area, mostly due to woodstove and fireplace use, and smaller amounts due to residential trash burning and industrial emissions. The remainder of the combustion emissions is from petroleum fuel combustion, primarily due to gasoline use in vehicles.

Creosote-treated wood accounts for over one-third of the PAH release, with marine pilings (54 t/yr), railroad ties (43 t/yr), and utility poles (17 t/yr) representing the major sources. Releases from large petroleum spills (≥ one gallon) and minor petroleum drips leaks, spillage, and improper disposal of motor oil account for PAH release of approximately 11 t/yr due to the PAHs contained in uncombusted petroleum.

Uncertainty associated with PAH releases is largely tied to the accuracy of modeled air emission rates, yet data were not examined to assess accuracy of these estimates. PAH releases from creosote-containing products are based largely on published release rates from regions of the U.S. other than the Puget Sound area and should be validated to increase the confidence in these estimates.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 12. Total PAH Release in the Study Area (values shown are t/yr).

Phthalates

Sources Assessed

Assessment of ongoing phthalate releases included the following sources:

- Losses from plasticized polyvinylchloride (PVC) products
- Losses from non-polymer uses (e.g. sealants, paints, inks)
- Fragrance and other personal care product use
- Vehicle brake pad wear
- Industrial, commercial, and institutional facilities

Other possible sources of phthalate release include use of automobile components and insect repellants. Releases from these sources were not assessed.

Phthalates are 87% of the 10.4 billion pounds per year world market for plasticizers, with bis(2-ethylhexyl) phthalate (a.k.a. di(2-ethylhexyl) phthalate [DEHP]) as the most common plasticizer for polyvinylchloride (PVC), constituting more than 50% of the phthalates produced (SPGW, 2007; ECB, 2008). Approximately 90-95%, and possibly as much as 97%, of DEHP is used to plasticize PVC products, which may contain up to 60% DEHP, although 30% may be more typical (ECB, 2008). Roughly 200-300 million pounds of DEHP are produced in the U.S. each year, with unknown amounts contained in imported products. According to ATSDR (2002), relatively little is imported (4 million pounds in 1998) or exported (14-27 million pounds per year in 1994-1998).

Plasticized PVC products are widely used and include exterior siding and roofing materials, automobiles, wires/cabling, advertizing banners, flooring, weather stripping, upholstery, garden hoses, swimming pool liners, footwear, clothing, food containers, tablecloths, shower curtains, rainwear, and toys. Rigid PVC products, such as pipes and windows, do not contain phthalates or other plasticizers. In Western Europe, DEHP emissions were estimated at 300 tons per year from indoor uses and 2,600 tons per year from exterior uses after measuring emissions to the air from PVC products (ATSDR, 2002). The population of Western Europe is about 400 million people, compared to about seven million people in Washington State.

The primary source of phthalates to the Study Area is emissions from plasticized PVC products (SPWG, 2007). Phthalates are not covalently bound to PVC polymer chains and migrate out over time (Rudel and Petrovich, 2009). The amount of phthalates that leach out of the PVC into air is quite variable. Factors that affect the rates of volatilization include the temperature and surface area of the material; emission rates among phthalates may also be quite variable. At a certain point, the PVC undergoes glassification when it becomes stiff and brittle, and very little phthalate continues to off-gas. Once emitted, phthalates tend to adhere to dust particles rather than remain in vapor phase (ATSDR, 2002; SPWG, 2007).

Non-plasticizer (non-polymer) uses of DEHP are a small percentage (<10%) of overall use. These uses include PCB replacement (dielectric fluids for electric capacitors), de-foaming agent in paper manufacturing and detergents, chemical intermediate for insect repellent, cosmetics, lacquers, munitions, ceramics, printing inks, adhesives, sealants, and industrial lubricants.

Estimates of Phthalates Released from PVC and Non-Polymer Uses

The use estimates for DEHP in Study Area PVC and non-polymer uses were extrapolated by work done in Sweden for the European Union (EU); no estimates for DEHP use were found for the U.S. Sweden's National Chemicals Inspectorate prepared the 2003 draft Risk Reduction Strategy and on the 2008 EU Risk Assessment Report (RAR) for DEHP (ECB, 2008). Sweden estimated PVC and non-polymer use in each category, primarily based on information from industry.

The estimates from Sweden were used to estimate releases in the Study Area, based on the assumption that releases are proportional to the amount used. The amount of DEHP used in the Study Area was scaled by population, compared to the amount used in the U.S. An estimated 1,340 metric tons (t) of DEHP are used annually in the Study Area, with 1,270 t used for polymers and 70 t for non-polymer uses. This is based on approximately 2,000 t for Washington's share of the 200-300 million pounds of DEHP used in the U.S. each year.

The emission estimates to air for indoor use were based on a general emission factor of 0.05% per year and the expected product lifetime. Emission estimates for outdoor uses were based on the amount of PVC and non-polymers used, expected lifetime, the normal thickness of the material, and measured emissions. For car undercoating, the emissions were based on a Danish study of DEHP in car wash water; the number of cars registered in the Study Area was used to calculate a release estimate.

Total indoor release of DEHP was 0.5 t/yr, with emissions from wall coverings and flooring accounting for almost one-half of the total (Table 78). Outdoor DEHP releases were estimated to be 10.2 t/yr. An estimated 16 t/yr from buried cable sheathing was not included in the release total because the majority of releases from these cables are expected to occur deep underground and should not contribute to surface releases. Better models of groundwater exposure are needed to understand the environmental contribution of such releases. Other releases to soil such as car undercoating and sealants may be secondarily carried to soil through water transport or other pathways. Overall, of the 16 diffuse sources of phthalate release from PVC and non-polymer use as described by ECB (2008), approximately 3.9 t/yr were released to soil, 5.4 t/yr to water, and 0.96 t/yr to air for a total of 10.2 t/yr.

Table 78. Estimates of DEHP Release from PVC and Non-Polymer Uses.

Product Use	DEHP in Products in Study Area (t/yr)	DEHP Release in Study Area - Air (t/yr)	DEHP Release in Study Area - Water (t/yr)	DEHP Release in Study Area - Soil (t/yr)	Total DEHP Release in Study Area (t/yr)
PVC (Polymer)					
Indoor PVC					
Wall coverings	272	0.14	--	--	0.14
Flooring	203	0.10	--	--	0.10
Films, sheets, coated products	197	0.10	--	--	0.10
Cables	168	0.08	--	--	0.08
Hoses and profiles	157	0.08	--	--	0.08
Total Indoor PVC	990	0.5	--	--	0.5
Outdoor PVC					
Cables- above ground	55	0.35	--	--	0.35
Car undercoating	19	--	1.6	1.6	3.3
Coil coated roofing	14	0.01	0.72	0.72	1.4
Coated fabric	58	0.01	0.60	0.60	1.2
Shoe soles	110	0.0004	0.10	0.10	0.20
Roofing material	2.8	0.0003	0.07	0.07	0.14
Hoses and profiles	16	0.0004	0.04	0.04	0.09
Total Outdoor PVC	280	0.4	3.2	3.2	6.7
Total PVC	**1,270**	**0.9**	**3.2**	**3.2**	**7.2**
Non-Polymer					
Lacquers and paint	6.8	0.009	1.4	0.46	1.9
Sealants, adhesives, etc.	52	0.005	0.8	0.27	1.1
Printing inks	7.8	0.08	--	--	0.08
Ceramics	0.14	--	--	--	--
Total Non-Polymer	**67**	**0.09**	**2.2**	**0.7**	**3.0**
Total PVC + Non-Polymer	**1,340**	**1.0**	**5.4**	**3.9**	**10**
				Total Estimate:	10

t: metric ton

Uncertainty

Emissions of phthalates from products may vary widely due to the DEHP composition with and among various materials and also environmental factors that partially drive rates of loss. While the broad range of product categories addressed is useful because of its inclusiveness, the rate of DEHP loss from each category is likely a gross estimate, and actual loss rates based on empirical evidence would provide more useful data.

Scaling DEHP releases from PVC is based on the assumption that consumption patterns of phthalate-containing materials in the Study Area are identical to that of Sweden. While there are no data to validate or dispute this assumption, it appears to be reasonable.

Finally, the estimates for DEHP likely under-estimate releases of all phthalates. The use of DEHP corresponds to 51% of the total phthalate consumption used as plasticizer in the EU (ECB, 2008). Thus, the releases of all phthalates may be considerably higher than estimates for DEHP alone.

Estimates of Phthalates Released from Cosmetics and Personal Care Products

According to testing data from U.S. and Swedish non-governmental organizations, diethyl phthalate (DEP) is the primary phthalate used in cosmetics and personal care products, with some products also containing di-n-butyl phthalate (DBP). There appears to be little use of DEHP in these products (DiGangi and Norin, 2002; Houlihan et al., 2002; EPA, 2009).

Table 79 shows the average use of major cosmetic and personal care products (EPA, 2009) and measured phthalate concentrations as reported by the Environmental Working group (Houlihan et al., 2002). Average hair spray usage (aerosol and pump combined) was 3.11 g/day for the fraction of the population that used it (32%) based on market research data (EPA, 2009). Solid deodorant antiperspirant usage was 0.5 g per use with 1.1 uses per day, also based on market research data (EPA, 2009). Phthalate concentrations in hair spray (246 mg/kg) and deodorant (318 mg/kg for solid) were obtained from a report by the Environmental Working Group (Houlihan et al., 2002). Spray deodorant contains comparatively high concentrations of phthalates (887 mg/kg; Houlihan et al., 2002), but no data were available for usage rates.

The reported concentration of phthalates in fragrances (12,314 mg/kg) was three orders of magnitude higher than for body lotion (18 mg/kg) (Houlihan et al., 2002), although the usage rate for body lotion was much higher than the average rate for fragrances (4.54 and 0.53 g/person/day, respectively) (EPA, 2009).

Accurate estimates of phthalate concentrations in nail polish were difficult to obtain. The Environmental Working Group reported phthalates listed on the labels of two-thirds of the labels examined at an average concentration of 5% (Houlihan et al., 2002), and the Food & Drug Administration (FDA) reported phthalates at an estimated concentration of 10% (FDA, 2010). Although the concentration of phthalate in nail polish appeared to be high (40,000 mg/kg was used as an estimated mean), the usage of nail polish based on market data (EPA, 2009) appeared to be low (0.021 g/person/day).

Products usage rates were applied to the Study Area population. It was assumed that all phthalates associated with cosmetics and personal care products are released to either POTWs through sanitary sewers or to septic systems. Release estimates to each are presented based on the Study Area population residing in homes discharging to POTWs (3,210,300; approximately 72% of the population) and homes discharging to septic systems (1,265,000; approximately 28% of the population). The proportion of discharges to drains at offices and other workplaces, restaurants, hotels, schools, gyms, etc. was not estimated.

Product usage rates and phthalate concentrations were used to estimate annual phthalate releases for the Study Area (Table 79). The highest releases appear to be a result of fragrance use (11 t/yr), followed by nail polish (1.4 t/yr). Total annual release of phthalates from cosmetics and personal care products is approximately 13 t/yr.

Table 79. Estimates of Phthalate Release from Cosmetics and Personal Care Products.

Product	Amount of Product Used in Study Area (g/person/day)	Phthalate Conc. (mg/kg)	Total Phthalate Release to POTWs in Study Area (t/yr)	Total Phthalate Release to Septics in Study Area (t/yr)	Total Phthalate Release in Study Area (t/yr)
Fragrance	0.528	12,314	7.6	3.0	11
Nail polish	0.021	40,000	1.0	0.4	1.4
Hair spray (aerosol and pump spray)	0.9952	246	0.3	0.1	0.4
Deodorant (solid)	0.55	318	0.2	0.1	0.3
Body lotion	4.543	18	0.1	0.04	0.1
Deodorant (spray)	unknown	887	--	--	--
Total	--	--	9.2	3.6	13
				Total Estimate:	13

Uncertainty

There are numerous cosmetics and personal care products available to consumers, and it seems likely that phthalate compositions may vary widely among products. Therefore, additional sources of information, if available, should be used to increase the accuracy of the phthalate concentrations (and type of phthalate) assessed for phthalate releases.

Usage rates of products are likely to be as accurate as any available information since the rates are based on market research data. However, if products within each category (i.e. brand names) contain variable concentrations of phthalates, then data on usage rates at the brand name level would be required for accurate release estimates.

The largest uncertainty about phthalate releases from cosmetics and personal care products may be the degree to which product (phthalate) usage equates to phthalate release either to POTWs or to septic systems. The assumption underlying these releases is that all products are eventually discharged to POTWs or to septic systems, but there are no data to assess the validity of this assumption.

Estimate of Phthalates Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

Industrial, commercial, and institutional facilities reported total mean annual phthalate releases of 0.98 t/yr for the five TRI categories assessed (Table 80). The highest overall reported phthalate releases were from a plastic manufacturer, paint and coatings manufacturers, and a boat manufacturer.

Air emissions accounted for 99% of phthalate release from industrial facilities among the categories assessed. The plastics manufacturer, Achilles USA, of Snohomish County had the bulk of air emissions, with a mean annual stack release of 0.86 t/yr. Fugitive air emissions were roughly an order of magnitude lower than from stack air, with the King County chemical distribution company, Univar USA, responsible for approximately two-thirds of the phthalates through fugitive emissions (0.033 t/yr). Chemcentral, a King County chemical distributor, had the only reported phthalate releases to water (0.011 t/yr) and to the "other disposal" category (0.0002 t/yr). Achilles USA and Chemcentral reported the only phthalate releases to POTWs, with most of this release from Achilles USA (0.001 t/yr).

All of the reported phthalate releases were for DEHP except for U.S. Marine/Brunswick Family Boat Co, Inc. which reported dimethyl phthalate stack air releases.

Table 80. Estimates of Mean Annual Phthalate Release from Industrial, Commercial, and Institutional Facilities as Reported in the Toxics Release Inventory, 1999-2008.

Release by Facility Type	**Percent of Total**	**Phthalate Release in Study Area (t/yr)**
Plastics Manufacturer	88%	0.86
Boat Manufacturer	5%	0.05
Chemical Distribution	4%	0.04
Paint and Coatings Manufacturers	3%	0.03
Total	100%	0.98
Release by Medium	**Percent of Total**	**Phthalate Release in Study Area (t/yr)**
Fugitive Air Emissions	5%	0.05
Stack Air Emissions	94%	0.92
Surface Water Discharge	1%	0.01
Other On-Site Land Disposal	<1%	<0.01
Discharge or Transfer to POTWs	<1%	<0.01
Total	100%	0.98
	Total Estimate:	0.98

Uncertainty

Criteria and limitations of TRI reporting requirements are in Appendix C.

Estimates of Phthalates Released from Modeled Air Emission Sources

Air emissions of phthalates inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Point source air emissions accounted for all of the releases (9.6 t/yr), but the exact source was not identified in the Air Emissions Inventory report (Table 81).

DEHP accounts for approximately one-half of the total phthalate release in the Air Emissions Inventory; butylbenzyl phthalate accounts for an additional one-quarter, and dimethyl phthalate makes up most of the remainder. Examination of the data on a county-by-county basis and comparisons to TRI-reported stack emissions suggests as much as 1 t/yr may be double-counted.

Table 81. Estimates of Phthalate Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	Phthalates Release in Study Area (t/yr)
Point Sources - Industrial, Commercial, Institutional (primarily Title V AOP)	9.6
Total Estimate:	9.6

AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

Uncertainty

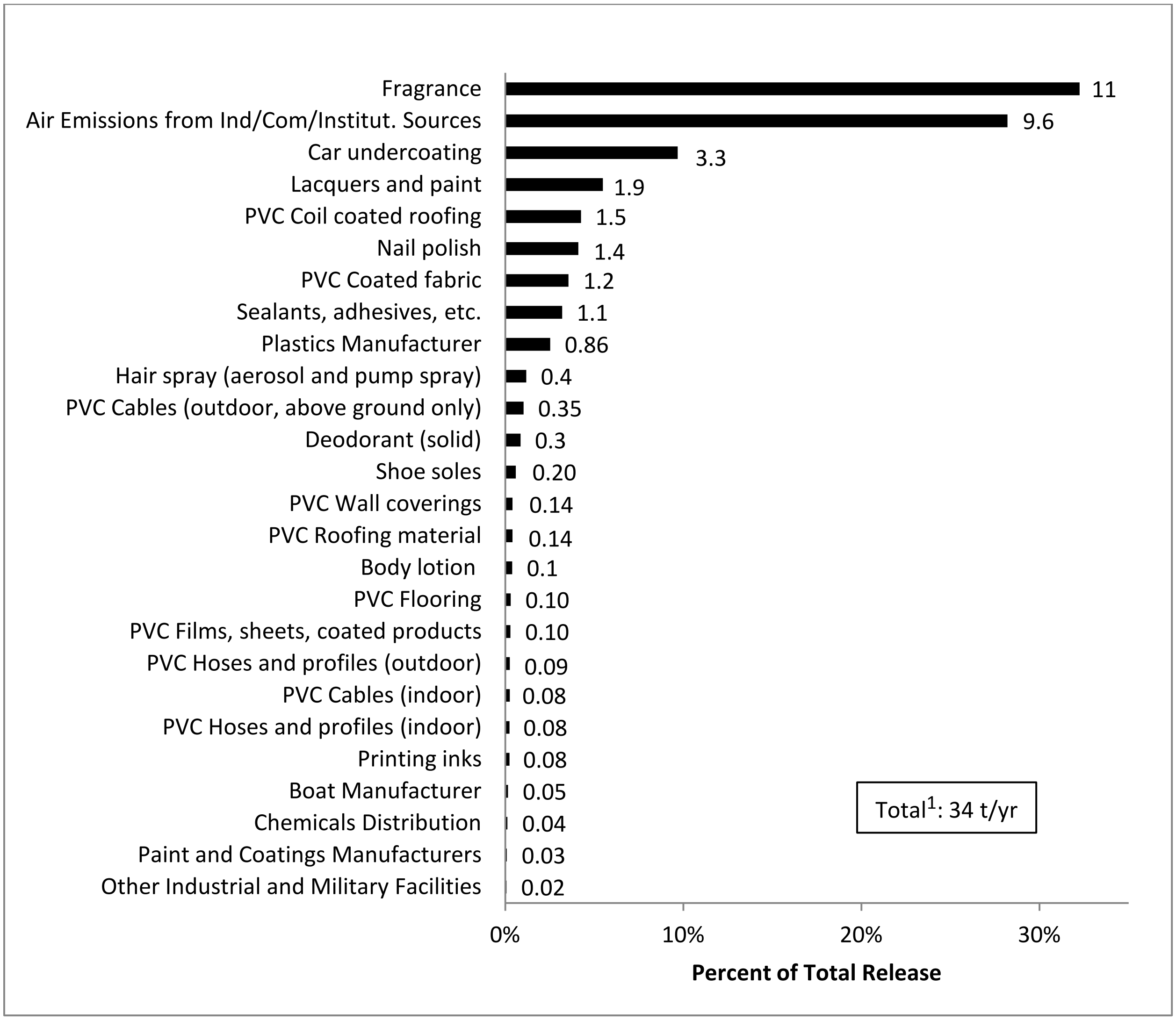
Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of Phthalate Release Estimates

Total phthalate releases from the sources assessed are approximately 34 t/yr (Figure 13). Phthalate release from fragrances is the largest single source, accounting for approximately 33% of the total. Other personal care products account for an additional 6% of the total release.

Releases of DEHP from PVC account for approximately 20% (7 t/yr) of the phthalate release in the Study Area, but this does not take into account the additional 16 t/yr released from buried cable. Overall, releases from non-polymer sources appear to exceed those from polymer (such as PVC) sources. Point source (industrial, commercial, institutional) air emissions account for 28% of the total phthalate release in the Study Area.

The data evaluated to obtain release estimates did not include ranges for the variables used. Therefore, it is difficult to assess the factors that may result in uncertainty or to gauge the magnitude of uncertainty. Based on the data that were available, it appears that the emission rate values may be the largest source of uncertainty.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

Figure 13. Total Phthalate Release in the Study Area (values shown are t/yr).

Triclopyr

Sources Assessed

Assessment of ongoing triclopyr releases included the following sources:

- Crop and golf course use
- Right-of-way maintenance
- Aquatic weed control
- Forest herbicide use (state forests only)
- Urban residential and commercial use

Other possible sources of triclopyr release include use as forest herbicides on private forests, and additional residential and urban use. Releases from these sources were not assessed due to lack of available information on use.

Estimates of Triclopyr Released from Crop and Golf Course Use

Triclopyr release on cropland was estimated from acreage and type of crops registered for triclopyr usage and product label application rates. According to WSDA, 59,600 acres of cropland and 9,200 acres of golf courses in the Study Area are registered for triclopyr use (WSDA, 2010c). Grass hay makes up the bulk of the cropland acreage with triclopyr usage (52,400 acres), followed by pasture, grass seed, and sod farms (approximately 1,800 acres each), and Christmas trees (1,400 acres). Orchard crops (apples, apricots, filberts) combined only comprise 500 acres registered for triclopyr use in the Study Area. Since the actual acreage treated was not reported, it was assumed that all of the registered acreage was treated.

WSDA (2010c) also provided the active ingredient types (i.e. BEE or TEA)[5] for which each crop is registered. Since there were no reported application rates, product labels for the BEE and TEA salts were reviewed to determine recommended application rates for each crop of interest (Alligare, 2010a and 2010b). Product labels provide ranges of maximum application rates but no information on optimum rates or minimum rates. Therefore, maximum application rates were used to estimate triclopyr releases on crops in the Study Area.

Estimates of total triclopyr released (as acid equivalent[6]) on crops and golf courses in the Study Area ranged from 60 to 240 t/yr, with 150 t/yr as the mid-point of this range (Table 82). Grass hay potentially accounts for the bulk of triclopyr usage due primarily to the high percentage of total acreage registered for triclopyr.

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[5] triclopyr BEE = 3,5,6-trichloro-2-pyridinyloxyacetic acid, butoxyethyl ester;
triclopyr TEA = 3,5,6-trichloro-2-pyridinyloxyacetic acid, triethylamine salt

[6] The acid equivalent is the active chemical without salt component. This is the chemical typically measured in environmental samples. For the TEA salt, the mass of acid equivalent is obtained by multiplying the mass of the triclopyr TEA by 0.715. For the BEE salt, the mass of acid equivalent is obtained by multiplying the mass of the triclopyr BEE by 0.721.

Table 82. Estimates of Triclopyr Release Due to Use on Crops and Golf Courses.

Crop	Acreage Treated (with Maximum Application Rates)				Total Triclopyr Release in Study Area (t acid equivalent/yr)
	Triclopyr BEE (2 -8 lbs/ac)	Triclopyr BEE (2 lbs/ac)	Triclopyr BEE (0.5 - 1 lbs/ac)	Triclopyr TEA (0.75 - 1.875 lbs/ac)	
Apples	335	--	--	--	0.3 - 1
Apricots	1	--	--	--	0.001 - 0.004
Filberts	177	--	--	--	0.2 - 0.6
Grass Hay	52,397	--	--	--	50 - 190
Grass Seed	1,768	--	--	--	2 - 6
Pasture	--	1,827	--	--	2
Sod Farm	--	--	1,751	--	0.4 - 0.8
Golf Courses	9,161	--	--	--	8 - 30
Christmas Trees	--	--	--	1,390	0.5 - 1
Other Conifers	--	--	--	11	0.004 - 0.009
				Total Estimate:	60 – 240 150 (mid-point)

t: metric ton

Uncertainty

Estimates on the number of cropland and golf course acreages registered for triclopyr are based on a reliable source (WSDA), as are the presumable application rates for triclopyr (product labels). However, based solely on these sources of information, there is no reasonable method to evaluate the actual acreages treated since there are no reporting requirements for treatment. The estimated releases calculated therefore represent maximum releases on registered cropland. Triclopyr use may also occur on non-registered cropland, but the occurrence and extent cannot be assessed with additional information.

The accuracy and confidence in the release estimates provided here could be greatly improved through required reporting of treated acreage. Since this is not a requirement in Washington, an indirect method to obtain release estimates is to use known application rates (due to mandatory reporting requirements) on identical crops under similar geographical/climatologically conditions, then scale the application rates to registered cropland in the Study Area.

Another method to estimate actual use data is to obtain regional triclopyr sales or other marketing data. This type of information would help avoid under-reporting and indirectly address treatment to non-registered acreage.

Estimates of Triclopyr Released from Terrestrial Use for Right-of-Ways

Triclopyr is used for right-of-way maintenance of roads by state agencies and possibly local governments, and also for right-of-way maintenance of rail tracks by railroads. Triclopyr is applied primarily as the formulated product Garlon® 3A, (triclopyr TEA; Dow AgroSciences, 2003), with smaller amounts of Garlon® 4 (triclopyr BEE; Dow AgroSciences, 2008a) and Crossbow® (triclopyr TEA; Dow AgroSciences, 2002) also used.

Quantities of triclopyr TEA and triclopyr BEE used along state highways within each of the WSDOT maintenance regions in the Study Area during 2007, 2008, and 2009 were provided by WSDOT (Ray Willard, WSDOT, written communication). The mean annual application rate of triclopyr (as the acid equivalent) was 0.34 t/yr (Table 83). The total of WSDOT's estimated rate of application for triclopyr was approximately 22 kilograms per square mile (kg/sq mi), about triple a calculated national estimate of 7.7 kg/sq mi (EnviroVision et al., 2008; EPA, 1998b).

The Washington State Department of Natural Resources (DNR) provided estimates of triclopyr applied along DNR roads in the Puget Sound and surrounding regions from 2005 through 2009 (Keith Yonaka, DNR, written communication). Annual triclopyr (acid equivalent) means for the South Puget Sound, Pacific Cascades, Olympic, and Northwest Regions were 521 kg, 36 kg, 54 kg, and 0 kg, respectively. However, all except the South Puget Sound Region estimates are uncertain due to lack of documentation or due to doubts about the drainage where applications were conducted. Therefore, only the South Puget Sound Region estimate was retained to represent the Study Area. All applications for DNR forest road maintenance were the Garlon® 4 formulation (Table 83).

The usage of triclopyr on railroad rights-of-way in the Study Area was scaled down from an estimate of annual national usage (approximately 45,000 pounds; EPA, 1998b). The formulation of triclopyr used was not specified and therefore was assumed to be an acid equivalent mass. Reported lengths of the treated railroad rights-of-way in the U.S. are 132,006 miles (Maps of the World, 2006), 140,249 miles (Weatherford et al., 2008), and 160,741 miles (North American Transportation Statistics, 2010). Resulting use rates based on these national data are 0.13 – 0.15 kg/track mile. When applied to track mileage in the Study Area – 749 miles (Blake, 2007) – the estimated triclopyr usage for the Study Area is approximately 0.1 t/yr (Table 83).

Based on the estimation methods described above, annual triclopyr usage on right-of-ways for state highways, state forest roads, and railroads was similar. Total triclopyr release through these applications was estimated to be 0.9 t/yr.

Table 83. Estimates of Triclopyr Release Due to Use for Right-of-Way Maintenance.

Right-of-Way	**Total Triclopyr Release in Study Area (t acid equivalent/yr)**
State Highway	0.3
State Forest Road	0.5
Railroad	0.1
Estimate of Triclopyr Release from Right-of-Way Maintenance (t acid equivalent/yr)	0.9

t=metric ton

Uncertainty

There is large uncertainty in the estimates of triclopyr usage to maintain right-of-ways. Overall, it appears that the usage amounts calculated here under-estimate the actual triclopyr usage due to lack of data for power line right-of-ways and for road right-of-ways managed by counties, private forest owners, and in national forests.

Estimates obtained where data were available are also uncertain. For instance, documentation of triclopyr usage on forest roads is spotty among DNR regions. National usage data from EPA may be out-of-date and the scaling method for estimating triclopyr release in the Study Area may not reflect accurate proportionality.

Estimates of Triclopyr Released from Permitted Aquatic Pesticide Use

Aquatic use of triclopyr to control nuisance vegetation in freshwater lakes is permitted under Ecology's Aquatic Pesticide General Permit. During 2009, Ecology issued permits for 14 applications in the Study Area, primarily to control Eurasian water milfoil (*Myriophyllum spicatum*). Approximately 135 surface acres in the Study Area were treated using the commercial product Renovate® containing 4 – 14% of triclopyr TEA as the active ingredient (Dow AgroSciences, 2008b). A summary of the aquatic use of triclopyr in the Study Area is shown in Appendix M. The amount of triclopyr (as acid equivalent) used during applications in 2009 totaled approximately 0.7 t/yr (Table 84).

Table 84. Estimates of Triclopyr Release Due to Use as an Aquatic Pesticide.

Number of Acres Treated	Mean Rate of Triclopyr Treatment (kg acid equivalent/acre)	Total Triclopyr Release in Study Area (t acid equivalent/yr)
135.33	5.0	0.7
	Total Estimate:	0.7

Uncertainty

Ecology relies on self-reporting by aquatic pesticide applicators to track amounts of chemicals used and compliance with the issued permits. There is no information available to assess the reported versus actual application amounts. In addition, there is no available information to assess un-permitted aquatic use of triclopyr.

Data used for release estimates of aquatic triclopyr usage included only 2009 data. Therefore, it is not known if the data reflect annual means as the data provide no gauge of inter-annual variation.

Estimates of Triclopyr Released from Forest Use

Use of triclopyr as a forestry herbicide was estimated for state (DNR) forest lands in the Study Area. Estimates of the usage on state forest lands were provided by two sources of information provided by DNR.

The first estimate was based on a "ball-park" figure of 800 gallons per year of triclopyr BEE applied to DNR-managed forest lands in Washington (Scott McLeod, DNR, written communication). Scaled by the relative amount of DNR forest land in the Study Area (30.6%) yielded an annual mean of 244 gallons, a triclopyr acid equivalent of 0.44 t/yr (Table 85).

A second estimate on triclopyr usage provided by DNR was based on a ten-year record (1999-2008) of triclopyr BEE applications in DNR's Habitat Conservation Plan (HCP) planning units for Forest Management Activities, excluding right-of-way and all non-forest applications (Bob Aulds, DNR, written communication). The North Puget Sound planning unit had the highest amount of triclopyr application, followed by the Straits and South Puget Sound units, with a total Garlon® 4 application rate among all units of 636 gallons (1.2 t triclopyr acid equivalent) per year (Table 85).

Table 85. Estimates of Triclopyr Release Due to Use as a Forest Herbicide.

Basis For Estimate	**Estimated Amount Used in Study Area (gallons)**	**Total Triclopyr Release in Study Area (t acid equivalent/yr)**
State – DNR-Managed	244 (triclopyr BEE)	0.4
State – DNR-Managed (HCP planning units)	636 (triclopyr BEE)	1.2
Total Estimate:		0.4 – 1.2 0.8 (mid-point)

HCP: Habitat Conservation Plan

Uncertainty

There is large uncertainty in the estimates of triclopyr usage as a forest herbicide. Overall, it appears that the usage amounts calculated here under-estimate the actual triclopyr usage due to lack of data for private forests and national forest use.

Estimates obtained from available data also have a large degree of uncertainty. For instance, reported triclopyr applications on HCPs are limited to the state managed portions of HCPs and may therefore exclude any triclopyr used for private or national forest portions of HCPs.

Estimates of Triclopyr Released from Urban Residential and Commercial Use

Triclopyr usage on residential and commercial land covers was estimated from national estimates of triclopyr usage on these land use types. EPA's Registration Eligibility Decision (RED) for Triclopyr (EPA, 1998b) estimated that triclopyr was applied to 0.23% of the 33 million residential and commercial acres nation-wide. A total of 40,000 pounds of triclopyr (active ingredient, salt not specified) was applied to these 75,000 acres, resulting in an application rate of 0.17 kg acid equivalent/acre.

Areas of residential and commercial/industrial land covers in the Study Area are published in Envirovision et al. (2008) and were originally based on land covers from the 2001 National Land Cover Dataset (MRLC, 2001). Total acreages for residential and commercial/industrial land covers in the Study Area are 1.02 million and 66,300 acres, respectively. Based on the national estimates of treatment rates, the residential and commercial/industrial land covers in the Study Area treated with triclopyr are 2,379 and 152 acres, respectively. Based on these areas and application rates on the national averages, the annual amounts of triclopyr applied to residential and commercial/industrial lands in the Study Area are estimated to be 0.4 and 0.03 t acid equivalent/yr, respectively (Table 86). The total amount of triclopyr used domestically was estimated to be 0.4 t acid equivalent/yr.

Table 86. Estimates of Triclopyr Release Due to Use in Urban Residential and Commercial Areas.

Land Type	**Area Treated in Study Area (acres)**	**Mean Rate of Triclopyr Treatment (kg acid equivalent/acre)**	**Total Triclopyr Release in Study Area (t acid equivalent/yr)**
Residential	2,379	0.17	0.4
Commercial/Industrial	152	0.17	0.03
		Total Estimate:	0.4

Uncertainty

The primary source of uncertainty in the estimate for domestic use of triclopyr is due to the use of national application rates for the Study Area. There were no data found to assess whether national triclopyr application rates are representative of Study Area rates, or if application rates differ among regions of the nation.

In addition to the lack of information on geographical differences, the triclopyr rate of use for residential and commercial areas published in EPA's RED may be out of date since this document was published during 1998. Rates of triclopyr use by homeowners, landscapers, and others performing weed and brush control may have changed substantially in the ensuing 12 years. Information to improve the accuracy of regional and contemporary triclopyr use would be vastly improved through obtaining market data, although this may require purchasing these data from one or more sources.

Estimate of Triclopyr Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

There were no reported triclopyr releases in the TRI.

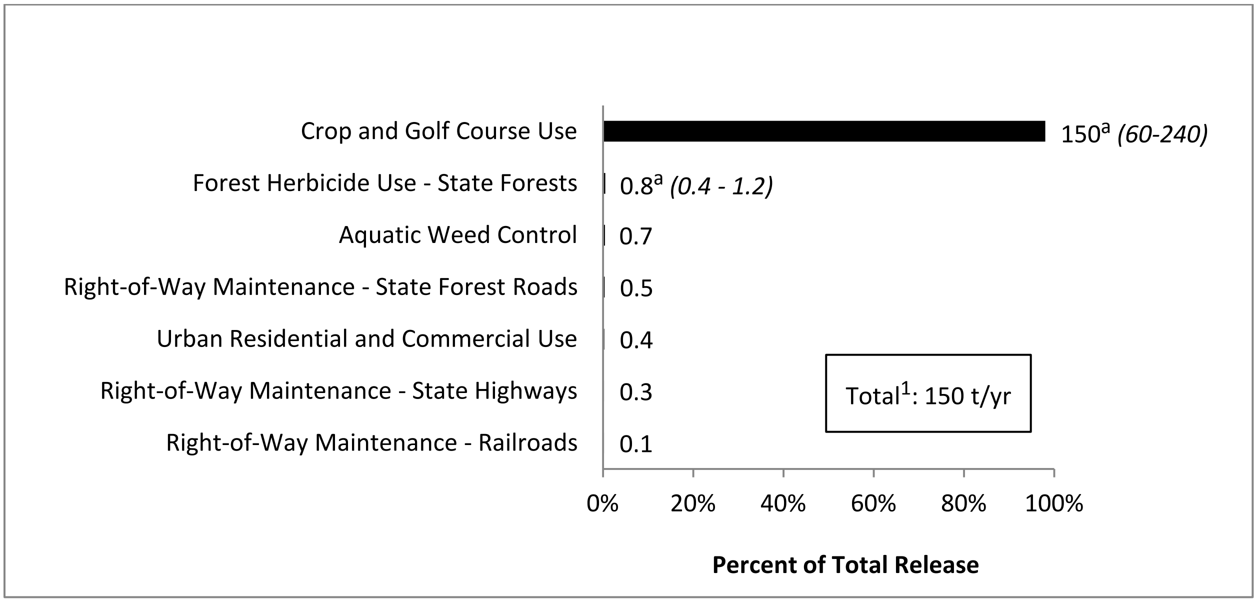
Estimate of Triclopyr Released from Modeled Air Emission Sources

There were no triclopyr emissions modeled for the Air Emissions Inventory.

Summary of Triclopyr Release Estimates

Total triclopyr release from the five categories assessed totaled approximately 60 – 240 t/yr, with a mid-point of this range of 150 t/yr (Figure 14). Agricultural uses of triclopyr accounted for 76 – 94% of the total release, with golf course use accounting for 5 – 22%. The remaining triclopyr use was primarily for right-of-way maintenance (0.7%), aquatic weed control (0.5%), and domestic use (0.3%). The small amounts calculated for domestic use and forestry use likely under-estimate actual usage rates for these applications.

All of the estimates presented here are highly uncertain due to the difficulty encountered in obtaining release data. Unlike other COCs addressed in this report, the only intended use of triclopyr (as an herbicide) is defined as a release, and there are no known unintentional releases. Therefore, if accurate information on its use is available, release data should also contain a high degree of accuracy.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 14. Total Triclopyr Release in the Study Area (values shown are t/yr).

Nonylphenol

Sources Assessed

Assessment of ongoing nonylphenol releases was limited to the following sources:

- Industrial, commercial, and institutional facilities

Other possible sources of nonylphenol release include use of parent compounds (alkylphenol ethoxylates) as additives in gasoline, as industrial detergents, and as carrier agents for pesticides. Releases from these sources were not assessed due to time limitations.

Estimate of Nonylphenol Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

The TRI does not require reporting for nonylphenol.

Estimates of Nonylphenol Released from Modeled Air Emission Sources

Air emissions of nonylphenol inventoried for the 12-county Puget Sound region were reported by Ecology's Air Quality Program (Ecology, 2007a). Point source air emissions accounted for all of the releases (0.18 t/yr), but the exact source was not identified in the Inventory report (Table 87).

Table 87. Estimates of Nonylphenol Release from Air Emission Sources Reported in the 2005 Air Emissions Inventory.

Air Emission Source	Nonylphenol Release in Study Area (t/yr)
Point Sources - Industrial, Commercial, Institutional (primarily Title V)	0.18
Total Estimate:	0.18

AOP: Air Operating Permit (see Appendix D for definition of a Title V AOP)

Uncertainty

Limitations and uncertainty of releases estimated in the Air Emissions Inventory are discussed in Appendix D.

Summary of Nonylphenol Release Estimates

Total nonylphenol release totaled approximately 0.18 t/yr, all from point-source air emissions. No effort was made to calculate nonylphenol releases from diffuse (nonpoint) sources, and nonylphenol was not reported under TRI.

Petroleum

Sources Assessed

Assessment of ongoing petroleum releases included the following sources:

- Motor oil drips and leaks
- Minor gasoline spillage from fueling vehicles and non-road equipment
- Improper disposal of used motor oil
- Major (>20 gallons) and minor (≥ one gallon and ≤ 20 gallons) petroleum spills

Other possible sources of petroleum release include aviation fuel leaks and uncombusted oil and fuel discharge from marine and freshwater vessel engines. Releases from these sources were not assessed.

Estimates of Petroleum Released from Leaks, Incidental Drippage, and Other Releases from Motor Vehicles

Oil released from vehicles through leaks, incidental drippage, and through burn-off or other means (e.g. combustion, blow-by) were estimated using data on vehicle miles travelled (VMT) and estimates of motor (crankcase) oil loss between oil changes.

Data on the number of VMT in the Study Area were obtained from the Federal Highway Administration (see Appendix E). Among the six vehicle categories analyzed, passenger cars had the highest annual VMT (23 billion), followed by light trucks (9.3 billion) and heavy trucks (single and combination trucks; 3.9 billion VMT).

Assumptions about oil loss rates were made on the basis of anecdotal information concerning oil loss rates (e.g. AA1Car.com; Performanceoiltechnology.com; About.com). Estimates of oil loss rates presented in these forums were typically one quart lost per 1,000 – 3,000 miles for passenger cars and up to one gallon per 10,000 miles for heavy trucks and buses. In contrast, Ecology's Waste 2 Resources Program estimates one-half quart loss per 5,000 miles (Jay Shepard, Ecology Waste 2 Resources Program, personal communication). Based on this information, loss rates of one-half quart per 5,000 miles for motorcycles and one quart per 5,000 miles for passenger cars, light trucks, heavy trucks, and buses were used to calculate motor oil loss rates for the Study Area.

Table 88 shows annual rates of oil released from motor vehicles via leaks, incidental drippage, combustion, blow-by, and other means based on assumptions about loss rates. Total releases were highest for passenger cars (3,800 t/yr) followed by light trucks (1,600 t/yr) and combination trucks (360 t/yr). Total petroleum released to the Study Area from motor oil loss was estimated to be approximately 6,100 t/yr.

Table 88. Estimates of Motor Oil Release Through Motor Oil Leaks, Incidental Drippage, and Other Releases from Vehicles.

Vehicle Type	Release Rate (Qts/5,000 mi)	Release Rate (mg/mi)	Vehicle Miles Travelled (mi/yr)	Motor Oil Release (t/yr)
Motorcycle	0.5	84 (a,b)	1.4×10^{8}	12
Passenger car	1	168 (b,c)	2.3×10^{10}	3,800
Light truck			9.3×10^{9}	1,600
Bus			7.2×10^{7}	12
Single unit truck			1.7×10^{9}	290
Combo. truck			2.2×10^{9}	360
			Total Estimate:	6,100

(a) Equivalent to one-half quart loss per 5,000 miles.

(b) Specific gravity of 0.89 used to convert volume to mass. Specific gravity is median value (range = 0.885 - 0.9) obtained from Material Safety Data Sheets from Quaker State® motor oil (all grades), Citgo non-detergent SAE 30 motor oil, Chevron Supreme motor oil (all weights), and Ashland Valvoline® non-detergent SAE 30 motor oil.

(c) Equivalent to one quart loss per 5,000 miles.

Uncertainty

While there were no published data found for oil loss rates from vehicles through leaks, incidental drippage, combustion, blow-by, and other means, it appears that the release rates used (one-half quart per 5,000 miles for motorcycles; one quart per 5,000 miles for cars, trucks, and buses) are probably no more than a factor of 2 from consensus values, and is likely within a factor of 3 from actual values. Scale-up values obtained from the Federal Highway Administration are likely to have a high degree of accuracy.

Motor oil is unique among the chemicals evaluated for the present study because its use on a unit (i.e. vehicle) scale is generally well-documented. As a result, it follows that its rate of loss should be easily quantifiable with a reasonable level of effort. Surveys conducted of large retail oil change services would likely provide the most accurate information on crankcase oil volumes and miles driven between oil changes. Another method to estimate oil loss is to conduct large-scale surveys of oil sold and oil recycled, although this would likely prove to be a less accurate alternative due to variables such as oil purchased outside of the survey area and uncertainties about the accuracy of recycled volumes.

The lack of information on uncombusted gasoline and diesel fuel released from vehicles is a notable data gap in the overall estimate of petroleum releases. Although the fraction of volume lost may be much smaller than motor oil, the higher rate of consumption – two to three orders of magnitude greater – may yield substantial releases at a large scale. Fuel and oil released from vessel engines is also a potentially important source of petroleum release, but losses from this source were not estimated.

Estimates of Petroleum Released from Spillage from On-Road Vehicle and Non-Road Equipment Fueling, and Filling Portable Fuel Containers

Spillage from gasoline filling and fueling activities can lead to substantial releases of petroleum when scaled up to regional geographic scales. As part of their effort to estimate and model air emissions, EPA (2006b) calculated gasoline spillage rates from filling vehicles and portable fuel containers (PFCs) at the pump, spillage during transfer of fuel in PFCs, and spillage during the fueling of non-road equipment (e.g. chainsaws, wood-chippers). EPA also estimated emissions of gasoline vapor from sources related to these filling and fueling activities, such as vapor displacement during fueling and evaporative emissions due to container wall permeation and diurnal volume change (i.e. breathing) during storage.

Nation-wide gasoline releases from residential and commercial PFC usage alone was estimated to be 315,000 tons during 2005 (EPA, 2006b), with approximately 60% of the loss from evaporation of volatile organic compounds (VOCs) through breathing and permeation of container walls. Total spillage from filling, transport, and fuel transfer accounted for approximately 31%, with the remaining 9% due to vapor displacement during filling and transfer.

Estimates of gasoline releases due to spillage from filling and fueling activities in the Study Area are shown in Table 89. Releases due to vapor displacement and evaporation were discounted since these may be volatile components of gasoline and therefore may not fully represent the entire chemical composition of gasoline.

Spillage rates used to calculate releases were 0.3128 grams per gallon (83 mg/l) during on-road vehicle fueling and during initial filling of PFCs at the pump: 6.71 – 13.89 grams per gallon (1,771 – 3,669 mg/l) during transport of PFCs, depending on the type of container and whether the usage was residential or commercial; and 1.5 – 201.4 grams per gallon (390 – 53,000 mg/l) during non-road equipment fueling, depending on the type of equipment (EPA, 2006b). Chain saws had the highest rates of spillage per gallon, possibly due to their comparatively small fuel tanks and resulting requirement for frequent re-fueling. For all spillage categories, individual spillage events were assumed to be small and are therefore not captured in the Environmental Report Tracking System (ERTS; discussed in subsequent sub-sections on petroleum releases) or other data compilations of spills.

Total gasoline release from on-road vehicle filling was estimated using the release (spillage) rate of 83 mg/l and the statewide gasoline consumption during 2008 (63.9 million barrels; EIA, 2010), then scaled to the Study Area by using the fraction of the state population (67.2%) based on the 12-county Puget Sound area census for 2009 (U.S. Census Bureau, 2010). Total gasoline release to the Study Area from this source was 570 t/yr.

Total gasoline released in Washington due to spillage from PFCs and non-road equipment was estimated by EPA (2006b) to be 2,039 t during 2005. Estimates of gasoline released to the Study Area were scaled to the Study Area by using the fraction of the state population (67.2%). Based on the population scale-down, the total amount of gasoline spilled from PFCs and non-road equipment in the Study Area was 1,400 t/yr, with the largest amounts occurring during equipment fueling (930 t/yr) and spillage during transport (430 t/yr). The amount of gasoline spilled during PFC filling at the pump was a relatively small percentage of the total (<1%).

Total gasoline release to the Study Area was estimated to be 1,900 t/yr (Table 89). Approximately 29% of this was due to spillage during filling of on-road vehicles.

Table 89. Estimates of Gasoline Release from Filling On-Road Vehicles and Portable Fuel Containers (PFCs) at the Pump, and from Fueling Non-Road Equipment.

Activity	**Amount Released in Study Area Based on EPA (2006b) (t/yr)**	**Amount of Gasoline Consumed in Study Area (l/yr)**	**Release Rate (mg released/ liter fuel used)**	**Gasoline Release in Study Area (t/yr)**
Spillage During Filling at Pump – On-Road Vehicles	--	6.82×10^9	83	570
Spillage During Filling at Pump – PFCs	15	--	--	15
Spillage During PFC Transport	430	--	--	430
Spillage During Non-Road Equipment Fueling	930	--	--	930
			Total Estimate:	1,900

Uncertainty

Uncertainty in the estimates for gasoline spillage from PFCs is ultimately due to any uncertainty in the EPA analysis since the present estimates rely on the reported spillage rates and the modeled values for Washington. The only other variable applied to the release estimates is the scale-down population fraction, likely to be a robust number based on census data. The population scale-down number also contains the underlying assumption that spillage rates and gasoline consumption rates are identical across Washington, an assumption which appears to be reasonable.

Although there was no formal analysis of uncertainty in the EPA (2006b) report on PFCs, peer reviewers of the report pointed out that spillage at the pump should have been based on a rate of container filling, rather than a *per gallon* basis. The logic is that spillage at the pump occurs with fuel nozzle removal and is identical whether a vehicle or PFC is filled. Since PFCs are generally smaller than vehicle fuel tanks, the *per gallon* spillage rate for PFCs should be much higher than for vehicles. Therefore, the estimate for spillage during filling at the pump may be an under-estimate, but as noted it has a relatively small contribution to the overall release from PFC spillage.

One uncertainty of these estimates is due to the location of gasoline spillage at the pump. Presumably, gas station pump areas are controlled environments; they are typically covered, may have oil/water separators or separate basins to collect runoff from dispensing areas, and may be required to have spill response materials available for larger spill events. However, casual observers are aware of constant track out of spills by vehicles, particularly during wet weather, drive-off of vehicles with gasoline continuing to drip from gas cap/gas door area, substantial spills from “topping-off” or inattentiveness of motorists, and a general lack of spill clean-up material readily at hand. Therefore, it is unlikely that these controlled areas actually control all or most of the spillage. The degree to which spillage at fuel-dispensing areas remains confined is not known; the estimates for gasoline releases assumes that all of the spillage is released to the environment (i.e. does not remain confined).

One notable omission from the EPA report on PFCs is the absence of estimated diesel fuel losses from filling and fueling activities. Estimates of gasoline spillage from these sources were initially conducted by air quality agencies to calculate volatile organic compounds (VOCs) emitted to the air, and diesel may have therefore been omitted due to the comparatively low degree of VOCs. The quantity of diesel spilled from filling and fueling activities could theoretically be made by simply substituting diesel fuel for gasoline in the methodology to calculate releases, but *per gallon* spillage estimates for gasoline would not likely apply to diesel fuel. Diesel vehicles and non-road equipment likely have much larger fuel tanks than those using gasoline, requiring fewer transfers *per gallon* resulting in a much lower *per gallon* spillage rate. Therefore, no estimates have been made for diesel fuel releases from these sources. Jet and aviation fuel released through these activities were also not estimated.

Estimates of Motor Oil Released Due to Improper Disposal of Used Oil

Oil released from vehicles through improper disposal by vehicle owners/operators following oil changes was estimated by applying survey results of oil disposal habits to the estimated number of oil changes performed annually in the Study Area. These estimates used the same VMTs as used for calculated releases from leaks and drippage (see previous sub-section), except the heavy-duty vehicle categories were excluded, since oil changes for heavy-duty vehicles are likely to be performed by service outfits and not by owners at their homes.

Rates of improper disposal of motor oil from motorists performing oil changes were based on a national survey published in 2002 by the U.S. Department of Transportation (U.S. DOT, 2002). The survey found 21% of adults changed their own oil, and 16% of people who change their own oil disposed of the used oil improperly, yielding an improper disposal rate of 3.2%.

The number of oil changes in the Study Area was based on an assumption of 5,000-mile intervals between changes. When applied to VMTs estimated for each vehicle category, there are 4.5 million oil changes per year for passenger cars, 1.9 million changes per year for light trucks, and 29,000 changes per year for motorcycles.

Oil volume per disposal event was assumed to be the full crankcase oil capacity (including filter) of the vehicle. The 74 passenger vehicles and 17 light-duty trucks for which Motorcraft® supplies filters have median oil capacities of 5.0 quarts and 6.0 quarts, respectively (Motorcraft Oil, 2004). An internet search of motorcycle engine crankcase oil capacities suggests that 2.5 quarts is a reasonable estimate of the average (e.g. ehow.com; fixya.com; v-twinforum.com; yahoo.com).

Table 90 shows annual rates of motor oil released in the Study Area due to improper disposal of used oil. Improper disposal of used oil from passenger cars accounted for two-thirds of the improper disposal (640 t/yr), with light-duty trucks accounting for nearly all of the remainder (320 t/yr). Improper disposal of used oil from motorcycles accounted for approximately 2 t/yr. Total release from all vehicle categories totaled 960 t/yr.

Table 90. Estimates of Motor Oil Release Through Improper Disposal of Used Oil.

Vehicle Type	Vehicle Miles Travelled in Study Area (mi/yr)	Oil Changes per Year in Study Area	Annual Oil Changes in Study Area Resulting in Improper Disposal	Average Oil Change Volume (Qt)	Motor Oil Release in Study Area (t/yr)
Motorcycle	1.4×10^{8}	29,000	965	2.5	2 (a)
Passenger car	2.3×10^{10}	4,500,000	151,969	5.0	640 (a)
Light truck	9.3×10^{9}	1,900,000	62,717	6.0	320 (a)
				Total Estimate:	960

(a) Specific gravity of 0.89 used to convert volume to mass. Specific gravity is median value (range = 0.885 - 0.9) obtained from Material Safety Data Sheets from Quaker State® motor oil (all grades), Citgo non-detergent SAE 30 motor oil, Chevron Supreme motor oil (all weights), and Ashland Valvoline® non-detergent SAE 30 motor oil.

Uncertainty

Rates of improper used oil disposal used for the release estimates may be inaccurate due to both regional and chronological factors. The improper disposal rate (3.2%) assumes that residents in the Study Area are represented by respondents of the national survey. It also assumes that used oil disposal practices have not changed during the past decade. Education efforts and disposal facilities implemented regionally during the past two decades may have substantially reduced historical rates of improper oil disposal.

Other variables applied to derive the release estimates appear to be reliable and accurate. Data on the crankcase oil capacities may be refined by analyzing a fleet breakdown by vehicle type, but the current values appear to be reasonable representations of vehicle fleets.

Estimates of Petroleum Released from Large Spills

Data on Major Spills (>20 gallons)

Estimates of petroleum released in the Study Area through major spills (>20 gallons) were obtained from two sources: The Washington Oil Spill Resource Damage Assessment (RDA) account (Ecology, 2011b) and an oil spill risk analysis conducted for Ecology's Spills Program (ERC, 2009). Data from the RDA and from ERC may overlap to some degree; it appears that ERC relied heavily on RDA data for their risk analysis.

The RDA data are an accounting of compensation paid or direct restoration completed by liable parties that have spilled petroleum or other materials to waters of the state from February 1991 through January 2011. Damages are assessed on a case-by-case basis, but in general appear to be proportional to the volume of material spilled.

Most of the spills were from commercial vessels and appear to have been directly to marine waters or the Lake Washington-Lake Union-Ship Canal system. There were 361 spills in the Study Area with recorded or estimated spill volumes ranging from 20 to 277,200 gallons. A complete list of the spills is in Appendix N, Table N-1.

Spilled materials considered for damage claims include a variety of asphalt, crude oils, diesel, gasoline, kerosene, jet fuels, lubricating oils, and non-petroleum oils such as hydraulic fluid and cooking oil. The total amount of material spilled in the Study Area from 1991- early 2011 for which damages were assessed was 484,188 gallons, over half of which (277,200 gallons) was gasoline spilled during the 1999 Olympic Pipeline Spill (OPS) in Bellingham. Excluding the OPS, there was 204,969 gallons of petroleum spilled during the period of record, only 3% of which was gasoline (6,150 gallons). Diesel, on the other hand, was 42% of the total excluding OPS (85,619 gallons), followed by crude oil at 26% (53,614 gallons) and other petroleum at 18% (37,864 gallons).

The "other petroleum" category includes 13,371 gallons of spills in which multiple materials were reported under one volume, including mixtures with diesel (12,049 gallons), gasoline (3,849 gallons), and jet fuel (1,302 gallons. Volumes of the various petroleum materials spilled averaged over the period of record are summarized in Table 91.

For their oil spill risk analysis, ERC (2009) reported that 328,705 gallons of oil and petroleum products were spilled into waters of the Study Area during 1995-2008 (Appendix N, Table N-2). The majority of this, however, was due to the 1999 OPS. Excluding the OPS volume from the ERC data yields a total of 51,505 gallons of material spilled (Table 91), approximately one-half the total for the same period as listed in the RDA database (103,742 gallons). The reason for the difference is not clear from the documentation provided in the report. However, ERC reported only 119 spill events in all Washington waters as opposed to 281 events in the Study Area listed in the RDA for the same period, indicating that a number of the RDA events were excluded from ERC's risk analysis.

The ERC (2009) risk analysis does not include the volume of specific materials actually spilled. Instead, the spill volumes are categorized by regulated sector. The OPS aside (the only pipeline spill listed), cargo vessels were responsible for approximately 40% of the spill volume, followed by oil tankers (25.5%), tank barges (13.5%), fishing vessels (11%), and terminals (9.5%). Marinas and passenger vessels together accounted for about 0.5% of the total spill volume. These categories appear to be consistent with the RDA data, although it is not clear if U.S. Navy support and warship vessels are included in the ERC data; Navy vessels accounted for approximately 16,000 gallons of spillage during the ERC reporting period.

Data on Minor Spills (≥ one gallon and ≤ 20 gallons) and Additional Data on Major Spills

Estimates of petroleum released in the Study Area through minor spills (≥ one gallon and ≤ 20 gallons) and additional data on major spills were obtained from Ecology's Environmental Report Tracking System (ERTS) (Ecology, 2010c). ERTS contains data collected largely through a telephone hotline and online reporting approach to response of material spills and other complaints about chemical releases (e.g. odor complaints). Information from the hotline calls and online submittals are placed in a database and prioritized for subsequent investigation. While this database contains a fairly comprehensive inventory of spill events, data on release amounts are generally un-quantified. For instance, oil sheens may be reported on water surfaces which pose difficulties for investigators attempting to estimate the quantity, and sometimes the type of material, spilled.

Since a complete tally of the material quantities listed in ERTS could potentially result in an over-estimate of spillage considered relevant for the present analysis, some of the ERTS data were excluded. Direct spills from vessels were excluded to avoid over-counting spills captured in the RDA and ERC datasets. Propane leaks to air were excluded, as were spills of anti-freeze and other non-petroleum materials.

According to ERTS, there were 48,249 gallons of material spilled into freshwater, marine water, soil, and unknown media during 2009, the period of record used to analyze ERTS data (Table 91). Spill volumes were tallied for diesel, gasoline, crude oil, fuel (heating) oil, jet and aviation fuel, lube and motor oil, and other petroleum products. Of the 1,222 incidents considered, only 663 (54%) reports included an associated volume. No volume estimates were made for the remaining incidents.

Spills to freshwater accounted for 49% of the spill volume, followed by spills to soil (33%) and marine waters (3%). Spills to unknown or un-documented media represented 15% of the total spill volume during 2009. A summary of the ERTS data used for release estimates is in Appendix N, Table N-3.

Assuming no overlap between the ERTS data (with exclusions noted) and the RDA or ERC data, the total annual mass of petroleum spilled ranged from 220 t/yr (sum of RDA values and ERTS values) to 230 t/yr (sum of ERC values and ERTS values). This is approximately 120-130% higher than the mean annual amount reported for the period 2000 – 2006 in the Phase I Loading Study (104 t; Maroncelli, 2009). Each of these estimates point to a relatively small percentage of the total spillage reaching marine waters.

Table 91. Estimates of Petroleum Release Due to Large Spills (≥ one gallon).

Material	**Period of Record**	**Data Source**	**Mean Volume Spilled in Study Area (gallons/yr)**	**Petroleum Release in Study Area (t/yr)**
Diesel	Feb 1991 – Jan 2011	RDA (Ecology, 2011b)	4,331	14
Gasoline			14,332	38
Crude Oil			2,712	8.3
Lube Oil			83	0.3
Kerosene and Heating Oil			27	0.08
Jet Fuel			986	2.8
Other Petroleum (a,b)			1,915	6.2
Total Petroleum			24,389	70
Non-Petroleum			102	0.4
Oil and Petroleum (b)	1995-2008	ERC, 2009	25,002	80
Diesel	2009	ERTS (Ecology, 2010c)	9,405	31
Gasoline			5,712	15
Crude Oil			0	0
Lube and Motor Oil			768	2.6
Fuel (Heating) Oil			662	2
Jet and Aviation Fuel			1,847	5.2
Other Petroleum (b,c)			29,855	96
Total Petroleum			48,249	150
			Total Estimate:	220(d) – 230(e)

(a) Includes asphalt, transformer oil, waste oil, and other products.
(b) Specific gravity of 0.85 used to convert from volume to mass.
(c) Includes asphalt/creosote, hydraulic, mineral, other, waste/used, and unknown oils.
(d) Sum of RDA and ERTS
(e) Sum of ERC and ERTS
Shaded areas indicated total petroleum releases from each data source.

Uncertainty

Unlike many other estimates of chemical releases, spill data are region-specific and not extrapolated from releases in other regions or extrapolated from literature values. However, actual spill data are limited to available documentation; there are no data available to estimate the magnitude of unreported spills, and therefore the quantities reported in the present assessment may be an under-estimate of actual spillage in the Study Area.

Estimated spill volumes from the RDA (Ecology, 2011b) and ERC (2009) may be relatively accurate due to rigors of the damage assessment documentation process. However, quantities estimated from the RDA and ERC represent only about 10% of the total spill quantity. ERTS data comprise the remaining 90%, and due to the nature and mission of ERTS (a tracking system to prioritize responses), the accuracy of the reported spill volumes is dubious.

One notable caveat regarding the use of a mean annual spill volume estimate from the RDA data is that the annual means used for the estimate (February 1991-January 2011) may be somewhat out of date since it appears that the size of spills is decreasing. The total spill volumes for 2004-2008 averaged 2,603 gallons per year, compared to 10,476 gallons per year for 1999-2004 (excluding the 1999 Olympic Pipeline Spill in Bellingham), and 12,633 gallons per year for 1992-1998. Only two spills of 500 gallons or more were reported since 2006, whereas 55 spills of that size occurred during the previous years for which data are available.

Leaking Underground Storage Tanks

Estimates of petroleum released from underground storage tanks (USTs) were initially pursued by reviewing Ecology's Toxics Cleanup Program UST database and application of estimated release rates from each tank. According to Ecology (2010d), there are approximately 7,800 underground tanks storing petroleum products in the Study Area. Most of the registered tanks (57%) have a storage capacity of 10,000 – 20,000 gallons and approximately 55% are registered for storage of unleaded gasoline. Appendix O shows the distribution of tank sizes and the number of tanks registered for each petroleum substance.

There are no known empirical data that can be used with confidence to estimate petroleum releases from USTs. Ecology's Toxics Cleanup Program documents the number of site cleanups involving petroleum USTs, but the program does not routinely attempt to estimate the volume leaked at each site (Mike Blum, Ecology Toxics Cleanup Program, personal communication). Assuming volumes of leaked material could be back-calculated at each site, there is no reliable method to re-create the chronology of the spill, precluding the ability to make estimates of annual release rates.

Underground Storage Tank Regulations (Ch. 173-360 WAC) provide thresholds for the performance of leak detection monitoring systems and tank tightness testing. In general, the maximum sensitivity to assess tank tightness is a 0.1 gallon per hour leak rate, although smaller tanks and specialty tanks may use different criteria to determine the threshold definition of a leaking tank. Assuming that a 0.099 gal/hr leak rate for each tank could "fly under the radar" for leak detection and tank tightness regulation, annual leakage for each tank would be 868 gallons per year, with a Study Area total of 6.8 million gal/yr.

Since the threshold value for UST leak detection and tank tightness appears to be unrealistically high for estimating releases, and no other values are known which may provide reasonable estimates for release rates, release estimates of petroleum from USTs have been deferred pending additional information.

Estimate of Petroleum Released from Industrial, Commercial, and Institutional Facilities – Toxics Release Inventory (TRI) Reporting

There were no reported petroleum releases in the TRI.

Estimate of Petroleum Released from Modeled Air Emission Sources

Petroleum was not included in the 2005 Air Emission Inventory (Ecology, 2007a) apparently due to the infeasibility of measuring petroleum as a single component (petroleum is composed of many constituent chemicals).

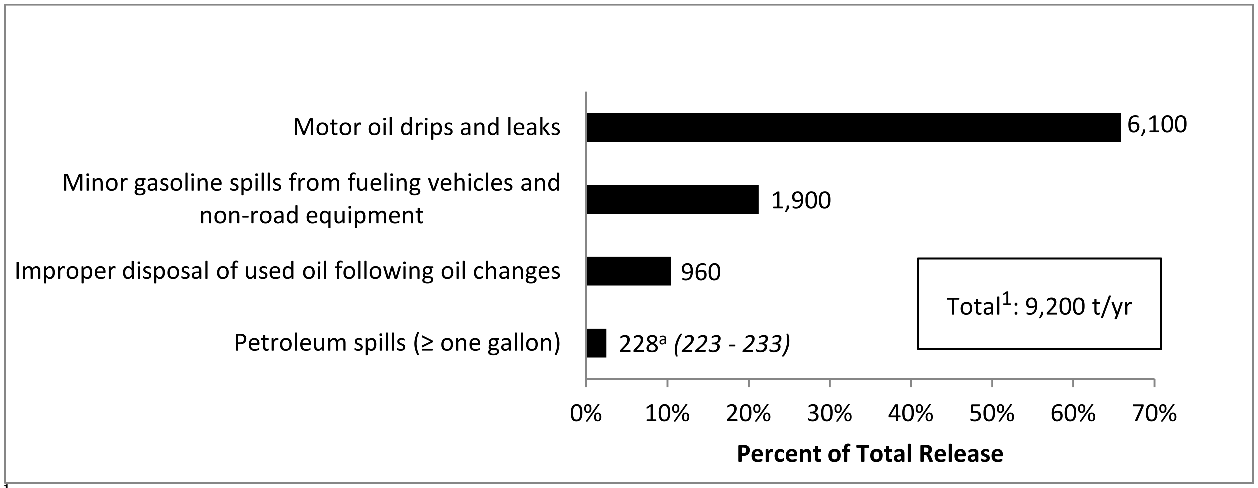
Summary of Petroleum Release Estimates

Total petroleum release from the four sources assessed totaled approximately 9,200 t/yr (Figure 15). Motor oil drips and leaks accounted for two-thirds of the total petroleum release, with other minor spills from fueling activities accounting for an additional one-fifth of the total. Large spills (≥ one gallon) accounted for a small portion (2%) of total petroleum released in the Study Area.

Based on the release estimates obtained and the types of sources assessed, it is difficult to ascertain the degree of uncertainty for petroleum releases in the Study Area. Estimates of large spills, particularly major spills (>20 gallons) are reasonably well-documented since they carry implications for liability. However, these spills make up less than one-fiftieth of the annual petroleum release to the Study Area. In contrast, drips, leaks, and spillage from filling pumps and portable containers make up the bulk of annual petroleum release, but lack of information on the accuracy of typical drip quantity, as well as accuracy of the number of drips, results in a high level of uncertainty for region-wide release estimates.

In some cases, release estimates may be simply out of date due to conventional practices for handling petroleum products. Improper disposal of motor oil following replacement may be the best example of petroleum handling practices which have changed during the past several decades; release estimates derived from current disposal practices may be lower than those presented here.

The lack of information on uncombusted gasoline and diesel fuel released directly from vehicles is a notable data gap in the overall estimate of petroleum releases. Fuel and oil released from vessel engines is also a potentially important source of petroleum release, but losses from these sources were not estimated here.



[1] Sum of best estimates. Best estimates are either mid-point, median, mean, or most reasonable estimate for each source.

[a] Mid-point of range

Figure 15. Total Petroleum Release in the Study Area (values shown are t/yr).

Summary and Conclusions

General Considerations

The present report identifies the major primary sources of selected toxic chemicals and provides preliminary estimates of the annual quantity released from each source in the Puget Sound region. Fourteen chemicals and chemical groups (chemicals of concern [COCs]) are addressed, and the quantities of COCs released annually from numerous sources were estimated.

Sources addressed were limited to primary sources, defined as *the object or activity from which a COC is initially released to environmental media (air, water, soil) or released in a form which can be mobilized and transported in an environmental pathway*. Many of the sources assessed were diffuse sources which required an understating of release rates from specific objects or activities in order to calculate release estimates. Other estimates were based on regional or national studies, or on data from regulatory programs. Releases were calculated for most of the COC sources identified; recommendations are made to estimate releases from known sources not addressed in this report.

COC release estimates were calculated for the entire Study Area (Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. portion of the watershed for Puget Sound and the Straits). Except for several industrial and military facilities, there were no geographical distinctions made for releases. All of the releases presented in the report are expressed as a total annual mass released, generally in terms of metric tons (t) or kilograms (kg) per year.

All of the COC release estimates calculated for this project have an associated uncertainty due largely to the assumptions required to compute the estimates; in some cases the estimates of the COC releases may be highly imprecise. For each release estimate calculated, the assumptions used to derive the estimate, along with a discussion of the uncertainty associated with each estimate, were discussed. In some cases, there was uncertainty surrounding the rates used for COCs released from specific objects or activities (i.e. release rates), while in other cases there was uncertainty about the appropriateness of using particular information to scale releases to the Study Area. A summary of uncertainties for each release estimate and a categorical rating of each uncertainty level are presented in Appendix P.

Due to the uncertainty associated with all of the release estimates, the reader should view the results as a gauge against which to compare the relative magnitude of chemical releases, but the absolute value for each release estimate should be viewed with caution.

The reader is also reminded that estimated quantities of COCs released in the Study Area do not necessarily translate to equivalent loads being delivered to Puget Sound. A variety of factors affect a chemical's behavior and fate once released in the environment, which in turn governs its presence in delivery and transport pathways. In many cases, the specific COC source and the mechanism of release may have more environmental relevance then the absolute quantity released. Mechanisms of COC release and the receiving media following release are implicitly or explicitly mentioned along with each source addressed. However, an analysis and discussion of COC behavior, fate, and degree of attenuation following release are beyond the scope of this project.

The reader is also cautioned that quantities of COCs released do not simply translate to a potential for harm; differences in mobility, location of release, persistence, bioavailability, and comparative toxicities are more important factors. However, an assessment of the relative hazard posed by COCs is beyond the scope of the present report.

Information on COC sources will be brought together with other Puget Sound Toxics Loading Analysis (PSTLA) information on loading, pathways, and relative hazards of COCs in a subsequent report (*Assessment Report*) which synthesizes information to better focus priorities for toxic chemical reduction and control strategies. For instance, the *Assessment Report* will compare the COC release estimates presented in this report to COC loads delivered to Puget Sound through various pathways. These release/delivery comparisons may help to explain the relationships between COC sources and COC loading/pathways, thereby helping to guide decisions about source control. The *Assessment Repo*rt will also provide an assessment of the relative hazards posed by COCs in various environmental media across the Puget Sound region.

Summary of Total Releases

A summary of the total annual releases estimated for each COC is shown in Table 92. Petroleum is estimated to be released in the largest quantity annually, followed by zinc which is the only other COC released at a rate greater than 1,000 metric tons (t) per year. Lead, PAHs, copper, and triclopyr are released at rates greater than 100 t/yr and nearly 35 t of phthalates are released annually. Arsenic, cadmium, mercury, PCBs, PBDEs, PCDD/Fs, and nonylphenol are generally released at rates near one t/yr or less.

The diversity of major sources among COCs is illustrated in Table 92. In general, industrial, commercial, and institutional point sources do not account for the largest releases of COCs. Instead, a variety of diffuse (nonpoint) sources account for a majority of the COC releases.

Table 92. Total Releases and Largest Sources for Each Chemical of Concern (COC).

COC	Total Release in the Puget Sound Basin[a] (t/yr)	Major Sources
Petroleum	9,200	Motor oil drips and leaks, and improper disposal of used oil. Gasoline spillage during fueling.
Zinc	1,500	Roofing material leaching. Vehicle tire abrasion.
Lead	520	Ammunition and hunting shot use. Loss of fishing sinkers and wheel weights. Roofing material leaching. Aviation fuel combustion.
Total PAHs	310	Woodstoves and fireplace combustion emissions. Vehicle combustion emissions. Creosote-treated piling, railroad ties, and utility poles.
Copper	180 - 250	Pesticides use on urban lawns and gardens.[b] Residential plumbing component leaching. Brake pad abrasion. Roofing material leaching. Vessel anti-fouling paint leaching.
Triclopyr	150	Herbicide use on crops and golf courses.
Phthalates	34	Personal care products. Polymer (primarily PVC) off-gassing. Industrial, commercial, and institutional air emissions. Roofing material leaching.
Total PCBs	2.2	Electrical equipment spills and leakage.[b] Residential trash burning. Building sealant (caulk) volatilization and abrasion.
Cadmium	0.96	Roofing material leaching.
Mercury	0.54	Consumer product improper disposal. Crematoria and industrial plants air emissions.
Total PBDEs	0.68	Furniture, computer monitors, and other components of residential and commercial indoor environments.
Arsenic	0.79	Industrial air emissions. CCA-treated wood leaching. Roofing material leaching.
Nonylphenol[c]	0.18	Industrial, commercial, and institutional air emissions.
PCDD/Fs	0.000009[d]	Backyard burn barrels.

t: metric ton (appr. 2.2 tons).

[a] Study Area: Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. watershed for Puget Sound and the Straits; also referred to as the Study Area.

[b] Estimate is highly uncertain.

[c] Sources were not fully assessed.

[d] Expressed as Toxic Equivalents (TEQs).

Summary of Releases by Category

Sources addressed in the report are summarized here by categories according to the physical objects, activities, mechanisms of release, or the environmental medium to which a COC is released. A complete listing of COCs with accompanying information useful for categorizing COC releases is included as Appendix Q.

COCs Released from Roofing Materials

Runoff and leaching of chemicals from roofing materials is a more important source of metals than organic chemicals. For two metals – cadmium and zinc – release from roofing materials accounts for a majority of the total release, and nearly 20% of arsenic is released through roof runoff. Annual zinc releases from roof runoff were estimated to be particularly large (1,300 t/yr). There is also a substantial release of lead from roof materials (18 t/yr), although it is a small percentage of the total annual lead release in the Study Area

Approximately 10% of the copper released annually in the Study Area is potentially due to roof runoff. The amount of copper released from rooftops (27 t/yr) may under-estimate the actual release since leaching from copper-impregnated asphalt composite roof materials appears to be under-represented in the information used to calculate the release estimate. Unlike other metals, most of the copper release is from metal (non-copper) roof material. Asphalt composite roofing accounts for the bulk of roofing release of other metals due to the large area covered by this roofing material.

COCs Released from Residential Sources Other Than Roof Runoff

Sources other than roof runoff associated with residential areas include COC releases from components of residential buildings as well as activities that typically occur in residential areas. Leaching of metals from plumbing components accounts for comparatively small rates of lead discharges to sanitary sewers (0.2 t/yr), but the quantities of zinc and copper released annually are substantial (30 t and 39 t, respectively).

The only copper source that potentially surpasses residential plumbing is the use of copper as an urban pesticide (up to 73 t/yr), although the accuracy of this release estimate is highly uncertain. Residential plumbing and urban pesticide use combined account for nearly 50% of the total copper release in the Study Area, although it is likely that some of the urban copper pesticide release actually occurs in commercial areas, and therefore copper use in residential areas may be over-stated. Another pesticide, triclopyr, also occurs in residential areas, but the estimated annual quantity used (0.4 t/yr) likely under-estimates actual releases due to the difficulty in obtaining reliable triclopyr use data.

For PCDD/Fs and PAHs, the largest single sources are associated with residential activities. Based on national survey data, backyard burn barrels now surpass industrial and institutional sources as the largest single source of PCDD/Fs. In the Study Area, combustion emissions from backyard barrels account for approximately 75% of the total PCDD/Fs release based on the national survey data. Combustion releases from woodstoves and lawn & garden equipment contribute an additional 4% of the PCDD/F total.

Woodstoves (and fireplaces) are also a very large source of PAHs (110 t/yr; 33% of the total PAH release). Other PAHs releases associated with combustion activities in residential areas include residential trash burning (2% of total), lawn & garden equipment (2% of total), and non-wood fuel use and yard waste burning (< 1% of total combined). Residential (non-wood) fuel use also accounts for 1%-2% of the arsenic, cadmium, and mercury released annually in the Study Area.

PBDEs and phthalates are also released from residential areas in substantial quantities, primarily through passive means (volatilization, attachment to dust) that originate from indoor consumer products or from personal care products (e.g. fragrance, nail polish) which are discharged to sanitary sewers. The overall releases of PBDEs from residential indoor air and dust is estimated to be approximately 170 kg/yr, or about 25% of the total PBDE release in the Study Area. Like PBDEs, phthalate releases are difficult to quantify because of their complex behavior, but releases from personal care products alone are nearly 40% of the total release to the Study Area.

COCs Released from Vehicle Component Wear and Other Vehicle and Roadway Sources

Vehicle and road-related sources of COCs potentially encompass a wide variety of sources addressed for the present report. Abrasion of vehicle components is a large source of metals except for arsenic and mercury, and a minor source of PAHs. Brake pad wear accounts for approximately 15% of the total annual copper release and much smaller percentages for other metals. Tire wear accounts for substantial zinc and cadmium releases (82 and 0.03 t/yr, respectively), and possibly other chemicals as well due to the sheer quantity of tire material worn annually (11,000 t). The amount of PAHs released from tire abrasion (1 t/yr) is small compared to other PAH sources.

Lead releases from tire and brake pad wear are approximately 4 t/yr combined, but these sources are small compared to the loss of lead wheel weights (28 t/yr). Although this represents the best estimate of current lead wheel weight loss in the Study Area, a decline in coming years appears imminent due to a recent ban on the use of lead for wheel weights.

For PAHs, vehicle-related fuel combustion accounts for a large portion of releases. Gasoline combustion in light-duty vehicles alone accounts for nearly 10% of the total annual release, with an additional 4% from heavy-duty gasoline vehicles, and another 0.7% contributed by diesel vehicles (light- and heavy-duty combined). Other road and vehicle-related sources of PAHs also account for substantial releases, including PAHs released from petroleum drippage and leaks (4%), gas station emissions (0.4%), tire wear (0.3%), and abrasion and leaching of asphalt and coal-tar sealants (0.3%). In all, nearly 20% of the total annual PAH release is associated with vehicle and road use.

Other COC releases associated with vehicles and roads include approximately 10% of the total PCDD/F release contributed by vehicle combustion emissions (primarily heavy-duty diesel vehicles). Comparatively small quantities of triclopyr (< 1% of total triclopyr usage) are used along state highway right-of-ways. However, the total amount of triclopyr used for roadway maintenance may under-estimate actual usage since no estimates were obtained for usage along city and county roads or interstate highways.

Petroleum represents the largest COC quantity released annually to roadways (and other impervious surfaces). Most of the motor oil lost via drips and leaks (6,100 t/yr) likely occurs along roadways or impervious surfaces – parking lots and driveways – connected to roadways. In addition, gasoline is released at a rate of 570 t/yr during on-road vehicle fueling at the pump.

COCs Released from Industrial, Commercial, and Institutional Facilities

Releases from industrial, commercial, and institutional facilities are generally based on point source emissions from Ecology's 2005 Air Emissions Inventory or from self-reported Toxics Release Inventory (TRI) data. Point source air emissions accounted for the largest industrial-related releases of arsenic, cadmium, zinc, PAHs, phthalates, and nonylphenol based on data in the Air Emissions Inventory.

Large lead and copper releases, 8% - 10% of the totals for the Study Area, were self-reported (i.e. TRI) in the "other disposal" category for the Fort Lewis Army Base. No details are available to interpret the type of material or the exact mechanism of release ("other disposal" is largely interpreted as placing material in waste piles), but considering the large release of lead in ammunition throughout the Study Area, the lead release may simply be an estimate of lead ammunition used, with the copper release also possibly associated with ammunition use.

There are no TRI reporting requirements for PBDEs, and their emissions are not modeled in the Air Emissions Inventory. However, based on data on PBDE concentrations in indoor commercial air, 75% of total PBDE releases are due to release of air and generation of dust from indoor commercial office space.

COCs Released from Combustion

Unlike the categories described previously, the combustion category describes COC releases from a single mechanism. In general, combustion is a minor release mechanism for metals other than mercury, although lead emissions from aviation fuel combustion are substantial (16 t/yr). As much as of 25% of the mercury release to air in the Study Area may be through combustions emissions. Much of this may be due to fossil fuel combustion, although mercury emissions from crematoria and cement plants may originate from the source material rather than the fuel.

As mentioned previously, backyard burn barrels account for 75% of the PCDD/F release, and combustion sources account for nearly all of the remaining PCDD/F releases in the Study Area. PCBs are also formed and released through backyard trash burning, with an estimated 281 kg/yr released from this source.

Most of the PAHs released to the Study Area are emitted through combustion. As described previously in this summary, woodstove use is the largest source (110 t/yr), with emissions from vehicles releasing another 42 t/yr. In total, combustion emissions of PAHs account for approximately 60% of all annual PAHs releases in the Study Area.

COCs Released from Agricultural Activities

Since few of the COCs assessed are typically associated with agriculture, releases from agricultural uses are generally small. Triclopyr, the only COC used exclusively as a pesticide (herbicide), was an exception with large quantities (130 t/yr) potentially applied to crops in the Study Area. There are also some releases of metals in fertilizers applied to agricultural crops, with zinc releases the largest by far (41 t/yr). Arsenic, cadmium, lead, and mercury are applied at rates less than 0.3 t/yr.

Copper is used in agriculture as both a pesticide and a micronutrient. Total usage of copper on agricultural crops in the Study Area was estimated to be 15 t/yr, with approximately 10 t/yr of copper used as an agricultural pesticide alone.

COCs Released as Pesticides and from Wood Preservatives

Of the COCs assessed, only copper, PAHs, and triclopyr were identified as being currently used in the Study Area as pesticides or for wood preservation. Copper use as an urban pesticide is potentially the largest single copper source in the Study Area. When all pesticidal uses of copper are combined (lawn & garden use, anti-fouling paint, agricultural use, pools & spas, treated wood), they account for anywhere between 10% and 60% of the anthropogenic copper release.

PAHs released from creosote-treated wood also falls into the pesticide and wood-preservative category. Together, PAHs released from railroad ties, marine pilings, and utility poles account for more than 33% of the PAHs released annually in the Study Area.

As noted previously, triclopyr application to crops comprises the largest release of triclopyr, followed by use on golf courses or road right-of-way maintenance. A comparatively small amount of triclopyr (0.7 t/yr, 0.4% of the total) is applied directly to surface waters to control aquatic weeds.

Recommendations

Recommendations for strategies to control toxic chemicals in Puget Sound are beyond the scope of the present *Sources* report. However, recommendations to increase the scope, accuracy, and usefulness of this large-scale assessment of toxic chemical releases are as follows:

Refine COC Release Estimates

As stated in the Introduction, most of the COC release calculations presented in this report represent rough estimates. The *Uncertainty* discussions following the presentation of each release estimate serve as a guide for refining the estimates by identifying variables and their associated level of uncertainty.

The need to refine estimates is largely dependent on the users of these data and their ability to make decisions based on (1) the rough estimates presented here or (2) a requirement for more precise estimates.

Increase the Number of Sources for which COC Releases are Estimated

While the authors (project team) attempted to address the largest releases for each COC, there are a number of primary sources of these COCs that were not addressed. In future efforts, the project team recommends that addressing the following primary sources should receive top priority:

- Evaluation and assessment of major zinc sources not addressed in the report including: releases to marine waters from sacrificial anodes; leaching from guard rails, streetlight and road sign standards, chain link fences, and other outdoor galvanized material; and use of zinc salts for moss control on rooftops. Zinc releases from these sources have the potential to be large when compared to sources assessed in the present report.
- Assessment of all major sources of nonylphenol.
- Assessment of the direct release of petroleum to water from 2-stroke outboard engines.
- Assessment of the proportion of indoor and outdoor leakage of PCBs from transformers and capacitors.
- Assessment of PBDEs from auto shredder and electronics recycling facilities.

To obtain reliable release estimates for these sources, information on the number of units or the amount of activity causing releases must be established. The collection of this information is likely to be particularly time-consuming for the sources of zinc, nonylphenol, petroleum, and PCBs recommended for further assessment. For the PBDEs, release estimates from auto shredder and electronics recycling facilities may require site-by-site evaluation or other extensive investigation.

Re-examine the List of the Most Important Chemicals in the Puget Sound Basin based on New Information or New Ways to Evaluate Existing Information

Evaluating the most important toxic chemicals in Puget Sound was beyond the scope of the present study. The COCs were originally selected not only for their potential to be harmful, but also to represent a group of chemicals that may be present in a variety of transport/delivery pathways such as atmospheric deposition, surface water runoff, and wastewater treatment plant effluent. While it appears that the current list of COCs generally reflects conventional thinking on the most important chemicals in Puget Sound, no studies have been conducted recently to comprehensively evaluate the relative importance of all toxicants known to be present in the Puget Sound basin, including those that represent emergent issues.

This recommendation is not specific to this project alone. A thorough evaluation of the most important contemporary chemicals in Puget Sound would provide invaluable information to guide future work, including prioritizing the chemicals and associated primary releases to be assessed in future projects.

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Appendices

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Appendix A. Glossary, Acronyms, and Abbreviations

Glossary

Anthropogenic: Human-caused.

Basin: A drainage area or watershed in which all land and water areas drain or flow toward a central collector such as a stream, river, or lake at a lower elevation.

Built-up roof: A general term that can be applied to many flat, membrane, or torch-down roof types.

Congener: In chemistry, congeners are related chemicals. For example, polychlorinated biphenyls (PCBs) are a group of 209 related chemicals that are called congeners.

Nonpoint source: Pollution that enters any waters of the state from any dispersed land-based or water-based activities, including but not limited to atmospheric deposition, surface-water runoff from agricultural lands, urban areas, or forest lands, subsurface or underground sources, or discharges from boats or marine vessels not otherwise regulated under the NPDES program. Generally, any unconfined and diffuse source of contamination. Legally, any source of water pollution that does not meet the legal definition of "point source" in section 502(14) of the Clean Water Act.

Point source: Sources of pollution that discharge at a specific location from pipes, outfalls, and conveyance channels to a surface water. Examples of point source discharges include municipal wastewater treatment plants, municipal stormwater systems, industrial waste treatment facilities, and construction sites that clear more than 5 acres of land.

Source: For the purpose of the present project, the term *source* is strictly defined as: the object or activity from which a COC is initially released to environmental media (air, water, or soil) or released in a form which can be mobilized and transported in an environmental pathway.

Study Area: The geographical Study Area includes Puget Sound, the U.S. portions of the Straits of Georgia and Juan de Fuca, and the entire U.S. portion of the watershed for Puget Sound and the Straits.

Toxicant: Toxic contaminant.

12-county Puget Sound region: Clallam, Island, Jefferson, King, Kitsap, Mason, Pierce, San Juan, Skagit, Snohomish, Thurston, and Whatcom Counties.

Acronyms and Abbreviations

a.i.	active ingredient
a.k.a.	also known as
ABS	acrylonitrile butadiene styrene
ADVMT	average daily vehicle miles traveled
AOP	Air Operating Permit
ATSDR	Agency for Toxic Substances and Disease Registry
BEE	butoxyethyl ester

BNSF	Burlington Northern Santa Fe
CAP	Chemical Action Plan
CCA	chromated copper arsenate
COC	chemical of concern
DBP	di-n-butyl phthalate
DDT	dichlorodiphenyltrichloroethane
DEHP	di(2-ethylhexyl) phthalate [a.k.a. bis(2-ethylhexyl) phthalate]
DEP	diethyl phthalate
DNR	Washington State Department of Natural Resources
DOL	Washington State Department of Licensing
DOR	Washington State Department of Revenue
EAF	electric arc furnace
ECB	European Chemicals Bureau
Ecology	Washington State Department of Ecology
e.g.	For example
EIA	U.S. Energy Information Administration
EPA	U.S. Environmental Protection Agency
EPCRA	Emergency Planning and Community Right to Know Act
ERC	Environmental Research Consulting
ERTS	Environmental Report Tracking System
GIS	geographic information system
HCP	Habitat Conservation Plan
HPAH	high-molecular weight polycyclic aromatic hydrocarbon
HVAC	heating, ventilation, and air conditioning
IMERC	Interstate Mercury Education and Reduction Clearinghouse
LPAH	low-molecular weight polycyclic aromatic hydrocarbon
MERA	Mercury Education and Reduction Act
MRW	moderate risk waste
NAICS	North American Standard Industry Classification System
NASS	National Agricultural Statistics Service
NAVSTA	naval station
NPDES	National Pollutant Discharge Elimination System
NY/NJ	New York/New Jersey
OPS	Olympic Pipeline Spill
PAH	polycyclic aromatic hydrocarbon
PBDE	polybrominated diphenyl ether
PBT	persistent, bioaccumulative, and toxic substance
PCB	polychlorinated biphenyl
PCDD/F	polychlorinated dibenzo-*p*-dioxin and dibenzofuran
PFC	portable fuel containers
PIRT	Pesticide Incident Report Tracking
POTW	Publicly-owned treatment works
PPA	Pollution Prevention Act
PSNS	Puget Sound Naval Shipyard
PSP	Puget Sound Partnership
PSTLA	Puget Sound Toxics Loading Analysis
PUD	Public Utility District

PVC	polyvinyl chloride
RAR	Risk Assessment Report
RDA	Resource Damage Assessment
RED	Registration Eligibility Decision
TEA	triethylamine salt
TEF	toxic equivalency factor
TEQ	toxic equivalent
TRI	Toxics Release Inventory
UP	Union Pacific
U.S.DOT	United States Department of Transportation
UST	underground storage tank
VKT	vehicle kilometers travelled
VMT	vehicle miles travelled
VOC	volatile organic compound
WAC	Washington Administrative Code
WASS	Washington Agriculture Statistics Service
WDOH	Washington State Department of Health
WEDS	Washington Emissions Data System
WHO	World Health Organization
WPCB	Washington State PCB Rule
WSA	wetted surface area
WSDA	Washington State Department of Agriculture
WSDOT	Washington State Department of Transportation
WWTP	wastewater treatment plant

Units of Measurement

ft	foot
g	gram
kg	kilogram, a unit of mass equal to 1,000 grams.
km	kilometer, a unit of length equal to 1,000 meters.
l	liter
m	meter
mg	milligram
mg/kg	milligrams per kilogram (parts per million)
mg/l	milligrams per liter (parts per million)
mi	mile
ng/g	nanograms per gram (parts per billion)
t	tonne (metric ton; equal to 1,000 kg; appr. 2.2 tons)
ug	micrograms
ug/l	micrograms per liter (parts per billion)

Appendix B. Methodology for Roof Runoff Volume Calculations

1. Calculation of total roof area fractions

a. Rooftop (building) footprints GIS covers were requested from the 12 Puget Sound counties; only Kitsap and Thurston Counties possessed building footprints (Kitsap County, 2006; Thurston County, 2006). Building footprint data were also obtained from the Cities of Bellevue and Bellingham (City of Bellevue, 2007; Gill, 2007).

b. Assigning equal area to rooftops and the building footprints, footprint covers were laid over land cover data (MRLC, 2001) to determine the fraction of rooftop area within each of the four major land cover categories addressed in the other PSTLA studies: residential, commercial/industrial, agriculture, and forest/field.

c. Rooftop fractions by land cover category were averaged across the four jurisdictions. The mean rooftop fractions were: Residential – 0.081; commercial/industrial – 0.245; agriculture – 0.009; and forest/field – 0.005. Forest/field roof area fractions from Bellevue and Bellingham (0.037 and 0.008, respectively) were not included in the means since they do not appear to be representative of the majority of forest areas within the Study Area.

d. Rooftop fractions of each land cover were applied to the total area of the respective land covers within each of the 14 Puget Sound study areas, minus highways, used in the phase 2 surface runoff loading project (Envirovision et al., 2008). The result is an estimated rooftop area for each land cover type in each of the 14 study areas. Note the footprints represent a flat roof and do not account for roof angles, thus the total area of roofing materials may be under represented.

2. Assignment of roof types to roof footprints

a. Roof type frequencies were obtained from Assessor's Office parcel databases from the Pierce and Snohomish Counties (Pierce County, 2010; Snohomish County, 2010).

b. Roof type fractions (presented in Table B-2) from the two counties were averaged separately by residential and commercial parcel zones. Market summary data from trade publications supplemented the county data (ARMA, 2010; Dodson, 2007; Dodson Publications, 2009; Johns Manville, 2009). Roof type fractions for agricultural and forest/field land covers were computed as means of residential and commercial roof type fractions weighted to the relative land areas of these land covers over the entire basin. As a result, the roof type fractions for agriculture and forest/field showed more resemblance to residential roof types than to commercial roof types. Major roof type fractions for residential zoned areas were: Asphalt shingles – 0.82; wood shingle/shake – 0.086; and metal – 0.041. For commercial zoned areas, major roof type fractions were: Built-up roofing – 0.62; asphalt shingles – 0.21; and metal – 0.15. For agriculture and forest/field land covers, major roof type fractions were: Asphalt shingles – 0.72; built-up roofing – 0.12; wood shingle/shake – 0.073; and metal – 0.057.

c. Roof type fractions for each land cover were multiplied by total roof areas in each of the four land cover categories within each of the 14 study areas. The result is an estimated roof type area for each roof type for each land cover type in each study area.

3. **Calculation of runoff volumes for each roof type**

   a. Based the PRISM precipitation model (Oregon State University, 2002), mean annual precipitation for each of the four land covers within each of the 14 study areas were multiplied by each roof type area for each of the four land covers within each of the 14 study areas, excluding highways. The result is an estimate of the annual runoff volume for each of the roof types in each study area. It was assumed that evaporative loss and rooftop infiltration (i.e. runoff coefficient <1) was negligible, and therefore all precipitation was accounted for in the runoff volume.

Steps 1 through 3 above may be expressed as the formula:

$[(\text{Rooftop frx})_{LC} * (\text{Area})_{SA,LC}] * [(\text{Roof type}_{(1,2..n)}\ \text{frx})_{LC}] * [(\text{Precip (m/yr)})_{SA,LC} * (1{,}000\ \text{l/m}^3)]$

$=$ Annual runoff volume for roof type$_{(1,2..n)}$ for each study area (l/yr)

Where:

$(\text{Rooftop frx})_{LC}$ = rooftop fraction for the specific land cover
$(\text{Area})_{SA,LC}$ = area for specific study area and land cover (m^2)
$(\text{Roof type}_{(1,2..n)}\ \text{frx})_{LC}$ = fraction of roof type 1, or frac. of roof type 2, … or fraction of roof type n for
each of the four land cover categories
$(\text{Precip})_{SA,LC}$ = annual precipitation for specific study area and land cover (m/yr)

Roofing Data Summary Tables

Table B-1. Total Roof Area and Runoff by Roof Type

Roof Type	Sum of Roof Area (m^2)	Sum of Runoff (l/yr)*
Asphalt Shingles	3.96E+08	5.84E+11
Built-up	7.40E+07	1.03E+11
Clay Tile	3.03E+05	4.50E+08
Concrete Tile	1.59E+07	2.23E+10
Copper	1.87E+06	2.41E+09
Masonite	1.57E+06	2.32E+09
Metal	2.96E+07	4.29E+10
Other	1.60E+06	2.18E+09
Wood Shingle/Shake	3.62E+07	5.45E+10

* Sum of all 14 study areas and land covers

Table B-2. Roof Type Fractions

Roof Type[a]	**Commercial**	**Residential**	**Agriculture & Forest/ Field/Other**[b]
Built-up	0.62	0.033	0.15
Metal	0.15	0.041	0.061
Copper	0.0027	0.000025	0.00055
Asphalt Shingles	0.21	0.82	0.70
Concrete Tile	0.014	0.021	0.019
Clay Tile	--	0.00066	0.00053
Wood Shingle	--	0.086	0.070
Masonite	--	0.0034	0.0027
Other	0.017	--	0.0033

[a] Information showing a contribution of contaminants of concern to runoff was not found for all roof types.

[b] Mean of Commercial and Residential Roof Types weighted by relative roof area for the entire Puget Sound

Table B-3. Roof Area Fractions

Land Cover	**Rooftop Fraction**
Commercial	0.24
Residential	0.081
Agricultural	0.009
Forest/Field/Other	0.005

Table B-4. Total Roof Area by Land Cover

Land Cover	**Total Roof Area (m^2)**
Commercial/Industrial	6.39E+07
Residential	3.30E+08
Agriculture	1.34E+07
Forest/Field/Other	1.49E+08
All	5.56E+08

Table B-5. Total Area, minus Highways by Study Area Land Cover (m^2)

Land Cover	Admiralty Inlet	Commencement Bay	Hood Canal (North)	Hood Canal (South)	Elliott	Main Basin	Port Gardner
Commercial/Industrial	1.08E+06	3.18E+07	4.51E+05	5.87E+05	5.60E+07	7.04E+07	1.91E+07
Residential	4.86E+07	3.99E+08	4.80E+07	9.19E+07	3.15E+08	8.95E+08	4.48E+08
Agriculture	2.04E+07	7.55E+07	8.57E+05	5.49E+06	5.56E+07	2.16E+07	1.43E+08
Forest/Field/Other	2.24E+08	2.18E+09	2.96E+08	2.32E+09	8.79E+08	1.07E+09	4.14E+09

Land Cover	San Juan Islands	Sinclair/ Dyes Inlet	South Sound (East)	South Sound (West)	Strait of Georgia	Strait of Juan de Fuca	Whidbey Basin
Commercial/Industrial	5.90E+06	7.21E+06	2.68E+07	9.68E+06	1.57E+07	6.49E+06	9.59E+06
Residential	9.72E+07	1.42E+08	5.14E+08	2.55E+08	2.85E+08	1.31E+08	4.05E+08
Agriculture	7.07E+07	2.56E+06	1.32E+08	3.55E+07	5.47E+08	8.70E+07	3.28E+08
Forest/Field/Other	4.93E+08	2.23E+08	2.05E+09	1.27E+09	2.78E+09	2.91E+09	8.80E+09

Source: Envirovision et al., 2008.

Table B-6. Mean Precipitation by Study Area Land Cover (mm)

	Admiralty	Commencement Bay	Hood Canal North	Hood Canal South	Elliot	Main Basin	Port Gardner
Agriculture	623	1158	1034	1742	1210	1219	1234
Commercial/Industrial	567	1044	1096	1534	989	1006	1090
Forest/Field/Other	671	1785	1015	2263	1933	1602	2559
Residential	624	1205	1034	1951	1213	1103	1528

	San Juan Islands	Sinclair	South Sound (East)	South Sound (West)	Strait of Georgia	Strait of Juan de Fuca	Whidbey
Agriculture	671	1136	1124	1296	1159	639	1083
Commercial/Industrial	664	1239	1069	1336	1097	646	968
Forest/Field/Other	728	1239	1586	1554	2258	2026	2466
Residential	678	1201	1192	1385	1336	891	1565

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Appendix C. Summary of Toxics Release Inventory (TRI) Reporting Requirements and Release Categories Assessed for the Present Report (source: EPA, 2010)

Summary of Toxics Release Inventory

Section 313 of the Emergency Planning and Community Right to Know Act (EPCRA) of 1986 was enacted to facilitate emergency planning, to minimize the effects of potential toxic chemical accidents, and to provide the public with information on releases of toxic chemicals in their communities. The Pollution Prevention Act (PPA) of 1990 mandates collection of data on toxic chemicals that are treated, recycled, and combusted for energy recovery. Together, these laws require facilities in certain industries, which manufacture, process, or use toxic chemicals above specified amounts, to report annually on disposal or other releases and other waste management activities related to these chemicals.

The U.S. Environmental Protection Agency (EPA) maintains this information in a national database called the Toxics Release Inventory (TRI), which is available to the public via the Internet at www.epa.gov/tri. The TRI contains detailed information on nearly 650 chemicals and chemical categories that over 23,000 industrial and other facilities manage through disposal or other releases, recycling, energy recovery, or treatment. The data are collected from industries including manufacturing, metal and coal mining, electric utilities, commercial hazardous waste treatment, and other industrial sectors. Information on the North American Standard Industry Classification System (NAICS) can be found at www.epa.gov/tri/lawsandregs/naic/ncodes.htm.

Facilities must report release and other waste management information pursuant to EPCRA Section 313 if they: (1) have 10 or more full-time employees or the equivalent; (2) are in a covered NAICS code; and (3) use TRI-listed chemicals above a designated threshold – 25,000 pounds manufactured or processed, or 10,000 pounds otherwise used. Thresholds for the following EPA PBT chemicals assessed for the present report are much lower:

- Benzo(g,h,i)perylene – 10 pounds
- Dioxin and dioxin-like compounds – 0.1 grams
- Polycyclic aromatic compounds category –100 pounds
- Polychlorinated biphenyl (PCBs) – 10 pounds
- Mercury – 10 pounds
- Mercury compounds – 10 pounds

Once a facility meets these criteria, they are required to submit a TRI report for that calendar year, regardless of whether they released any of these chemicals to the environment; i.e. a facility may be required to report under TRI, even if there are no environmental releases (Diane Fowler, Ecology Hazardous Waste and Toxics Reduction Program, written communication).

Many of the TRI release categories involve material recycling or confined and regulated disposal. Therefore, only a limited number of release categories – those that meet the present project definition of environmental release – were considered for release estimates in the present report. They are described in the following section.

Toxics Release Inventory Release Categories Assessed

Fugitive Air Emissions

Fugitive air emissions are all releases to air that are not released through a confined air stream. Fugitive emissions include equipment leaks, evaporative losses from surface impoundments and spills, and releases from building ventilation systems. Data from Section 5.1 on the TRI Form R.

Stack or Point Source Air Emissions

Stack or point source air emissions occur through confined air streams such as stack, vents, ducts, or pipes. Data from Section 5.2 on the TRI Form R.

Surface Water Discharges

Releases to water include discharges to streams, rivers, lakes, oceans, and other bodies of water. This includes releases from confined sources, such as industrial process outflow pipes or open trenches. Releases due to runoff, including stormwater runoff, are also reportable to TRI under this category. Data from Section 5.3 on the TRI Form R.

Other On-site Land Disposal

Other land disposal is the disposal of the toxic chemical to land at the facility that does not fall into one of the other on-site land release categories found in Sections 5.5.1 through 5.5.3 on the TRI Form R. Other disposal includes such activities as placement in waste piles and spills or leaks. Data from Section 5.5.4 on the TRI Form R.

POTWs (Metal and Metal Compounds Only)

Transfers to publicly-owned treatment works (POTWs) of metals and metal compounds only. Because metals are not destroyed by sewage treatment processes, amounts of metals and metal category compounds reported in Section 6.1 are considered transfers to disposal or other releases. Data from Section 6.1, metals and metal compounds only, on the TRI Form R.

Transfers to POTWs (Non Metals)

The total amount of the toxic chemical in the waste stream transferred from the facility to all Publicly-owned treatment works (POTWs) during the calendar year (January 1 - December 31). POTW refers to a municipal sewage treatment plant. The most common transfers will be conveyances of the toxic chemical in facility wastewater through underground sewage pipes; however, trucked or other direct shipments to a POTW are also included in this estimate. Data from Section 6.1 on the TRI Form R. Transfers to POTWs of metals and metal category compounds are presented separately from those of the other non-metal TRI chemicals.

References for Appendix C

EPA, 2010. TRI Explorer. www.epa.gov/triexplorer/. Accessed September 8, 2010.

Fowler, Diane, 2010. E-mail dated October 7, 2010. Washington State Department of Ecology, Hazardous Waste and Toxics Reduction Program, Olympia, WA.

Appendix D. Air Emissions Inventory

Air emissions releases for a subset of COCs were reported in the Washington State Base Year 2005 County Inventories prepared by Ecology's Air Quality Program (Ecology, 2007). Air emissions inventories are prepared every four-to-five years as a tool for planning, forecasting, rule-making, and other management activities, as well as to meet federal reporting requirements. The air emission inventory was conducted at the county level and therefore estimates of releases to the Study Area were based on summed inventories from the 12-county Puget Sound area.

In general, the methodologies for conducting the air emissions inventory is similar to the methodology used for other categories of release estimates in the present report; unit release (emission) rates are determined and scaled-up to a large geographic area using variables such as housing, population, vehicle travel, etc. at the appropriate scale. Meteorological data also play a central role in calculating emissions. The air emission estimates in the Inventory are based heavily on modeling outcomes, particularly EPA-provided models such as MOBILE6.2, rather than on empirical release data from the literature. Like the present study, there are cases where release estimates are scaled down from national data or are equated to Washington from similar scaling variables from other states.

Due to the reliance on modeling, the accuracy and precision of the air release estimates are not evaluated in the Inventory report. The "scale-up" variables appear to be based on the most up-to-date information, but there is no information to assess to accuracy of release rates for chemical constituents. The Inventory report does not include empirical release rate information or model calibration data to assess validity of the models. Finally, the purpose of the air emissions inventory – as a management tool – does not necessarily indicate that generating the most accurate release estimates possible were the primary objective of the report. Although the air emissions inventory provides the most complete air release data available for toxicants, the reader should take the preceding factors into account when evaluating uncertainty in the release estimates.

Air emissions for the following sources were inventories for COCs addressed in the present report:

- Agricultural Equipment Emissions
- Airport Service Equipment Emissions
- Commercial Equipment Emissions
- Construction Equipment Emissions
- Consumer/Commercial Solvent Emissions
- Gas Station Emissions
- Heavy-Duty Diesel Vehicle Emissions
- Heavy-Duty Gasoline Vehicle Emissions
- Industrial Equipment Emissions
- Lawn and Garden Equipment Emissions
- Light-Duty Diesel Vehicle Emissions
- Light-Duty Gasoline Vehicle Emissions
- Locomotive Emissions

- Logging Equipment Emissions
- Oil Field Equipment Emissions
- Point Sources - Industrial, Commercial, Institutional Sources (primarily Title V)
- Railroad Maintenance Equipment Emissions
- Recreational Marine Vessel Emissions
- Recreational Equipment Emissions
- Residential Fuel Use, except Wood
- Residential Trash Burning
- Residential Yard Waste Burning
- Woodstoves and Fireplaces

For most COCs, emissions were calculated for only a small subset of these sources. The following provides additional detail of these air emission sources:

Agricultural Equipment Emissions

Agricultural equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Airport Service Equipment Emissions

Airport service equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Commercial Equipment Emissions

Commercial equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Construction Equipment Emissions

Construction equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Consumer/Commercial Solvent Emissions

Consumer and commercial solvent emissions addresses non-industrial solvents that are used in consumer or commercial applications. It includes solvent used as propellants, aids in product drying through evaporation, or as co-solvents or cleaning agents.

Gas Station Emissions

Emissions from gasoline service stations result from the evaporation of gasoline vapors during underground tank filling, underground tank breathing and emptying, vehicle refueling, and losses from fuel trucks in transit.

Heavy-Duty Diesel Vehicle Emissions

Emissions from fuel combustion and evaporation were estimated. Brake and tire wear were also considered, primarily to estimate emissions of particulate matter as road dust. The AQ inventory used the WSDOT information from the national Department of Transportation's Highway Performance Monitoring System to estimate the mean daily vehicle miles traveled (ADVMT). The AQ inventory also took into account different fuels and variations within a calendar year. It is included in the Inventory report under the Onroad Mobile Sources category.

Heavy-Duty Gasoline Vehicle Emissions

Emissions from fuel combustion and evaporation were estimated. Brake and tire wear were also considered, primarily to estimate emissions of particulate matter as road dust. The AQ inventory used the WSDOT information from the national Department of Transportation's Highway Performance Monitoring System to estimate the mean daily vehicle miles traveled (ADVMT). The AQ inventory also took into account different fuels and variations within a calendar year. It is included in the Inventory report under the Onroad Mobile Sources category.

Industrial Equipment Emissions

Industrial equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Lawn and Garden Equipment Emissions

Lawn and garden equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Light-Duty Diesel Vehicle Emissions

Emissions from fuel combustion and evaporation were estimated. Brake and tire wear were also considered, primarily to estimate emissions of particulate matter as road dust. The AQ inventory used the WSDOT information from the national Department of Transportation's Highway Performance Monitoring System to estimate the mean daily vehicle miles traveled (ADVMT). The AQ inventory also took into account different fuels and variations within a calendar year. It is included in the Inventory report under the Onroad Mobile Sources category.

Light-Duty Gasoline Vehicle Emissions

Emissions from fuel combustion and evaporation were estimated. Brake and tire wear were also considered, primarily to estimate emissions of particulate matter as road dust. The AQ inventory used the WSDOT information from the national Department of Transportation's Highway Performance Monitoring System to estimate the mean daily vehicle miles traveled (ADVMT). The AQ inventory also took into account different fuels and variations within a calendar year. It is included in the Inventory report under the Onroad Mobile Sources category.

Locomotives

Air emission estimates from locomotives are based on diesel combustion emissions from line haul, passenger, and switchyard activities. Estimates were only based on Class I railroads – Burlington Northern Santa Fe (BNSF) and Union Pacific (UP) are the only Class I railroads operating in Washington – although Amtrak is also included in the estimates.

Logging Equipment Emissions

Logging equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Oil Field Equipment Emissions

Oil field equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Point Sources - Industrial, Commercial, Institutional Sources (primarily Title V)

Point sources are defined as industrial, commercial, or institutional stationary sources. Major point sources that have a potential to emit large quantities of pollutants (≥10 tons per year or more; see Ecology 2007a for more detail on major emitters) generally operate under a Title V Air Operating Permit. All Title V sources (major) are included in the point source inventory. Local air agencies and Ecology's regional offices and Industrial Section collect emissions data from major emitters and report them to Ecology for inclusion in the Washington Emissions Data System (WEDS) database. Point source facilities that voluntarily adopt enforceable permit limits that reduce their potential for emissions – also called synthetic minor sources – may be also be included in the inventory at the discretion of the local regulating authority.

Railroad Maintenance Equipment Emissions

Railroad maintenance equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Recreational Marine Vessel Emissions

Recreational marine vessel emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Recreational Equipment Emissions

Recreational equipment emissions include emissions from gasoline, diesel, compressed natural gas, and liquefied petroleum gas fueled equipment. It is included in the Inventory report under the Nonroad Mobile Sources category.

Residential Fuel Use, Excluding Wood

Fuel use (distillate oil, natural gas, and liquefied petroleum gas) for 2005 was obtained from the Energy Information Administration. Coal was not considered, since less than 500 housing units in the state use coal as their heating source. The regional use was estimated using temperature, population and other parameters.

Residential Trash Burning

Air emissions from residential trash burning are based on outdoor burning of household waste. The inventory was based on the number of households burning trash, and the amount of trashed generated and burned per household.

Residential Yard Waste Burning

Emissions from residential yard waste burning were estimated from the number of households burning yard waste and the size of the piles based on outdoor burning habits in Idaho, Oregon, and Washington.

Woodstoves and Fireplaces

Residential woodstove and fireplace use includes a variety of certified and non-certified wood-burning devices used to provide residential heat. In addition to the use of woodstoves and fireplaces, the air emission estimates also accounted for the type of wood or other material (e.g. pellets, presto logs) used in wood-burning devices.

Reference for Appendix D

Ecology, 2007. Washington State Base Year 2005 County Inventories. Prepared by Sally Otterson, Washington State Department of Ecology, Air Quality Program, Olympia, WA.

Appendix E. Methodology Used to Estimate Metals Releases from Vehicle Brake Pads and Tire Wear

July 14, 2011

Technical Memorandum

Anthony J. Whiley, P.E.
Washington State Department of Ecology, Water Quality Program

RE: Automotive copper and zinc loading to road surfaces

Analysis Overview

This analysis provides an estimate of the annual loading of zinc and copper to road surfaces associated with automotive tire and brake pad wear within twelve counties situated within the greater Puget Sound basin. These estimates are for deposition to road surfaces as opposed to receiving waters and are based primarily on the number of vehicle kilometers travelled (VKT) within each of the study counties in 2008. A finer resolution to these loading estimates could be undertaken and would be warranted, particularly if water quality impacts associated with automotive-related metals deposition is the ultimate interest.

The distribution of this loading throughout the road network is not even. For instance, for a given kilometer of rural highway it is expected that the loading of brake pad-related copper is lower than that of an urban intersection. Highway tire wear rates of passenger vehicles are different from non-highway wear rates. In addition, differences in driving behavior, type, and age of vehicle, road surface, weather conditions, the characteristics of the brake and tire material, and their level of maintenance, are among the many variables that could be considered in making this type of estimate. While recognizing this complexity, the approach ultimately undertaken by this analysis steered in the direction of simplicity due in large part to the lack of data available to support a more detailed type of analysis that more fully accounted for these many variables.

The ultimate objective of this work is to:

- Determine the relative magnitude of copper and zinc deposition associated with automotive brake pad and tire wear occurring within the greater Puget Sound basin.
- Quantify that load by proximity and the types of vehicles primarily associated with it.

The study methods can also be applied to examine the loading of other constituents present within brakes and tires.

Methods

The estimation of automotive metals loading associated with brake pad and tire wear was based on three primary factors: (1) the annual vehicle kilometers travelled (VKT); (2) brake pad and tire wear rates; and (3) the representation of copper and zinc within brakes pads and tires, respectively.

To provide a greater level of specificity to the loading estimates, the vehicle kilometers travelled data was further defined by period (month), proximity (county), and representation among vehicle types. In addition, brake pad and tire wear rates were defined by vehicle type. These factors were incorporated into two equations used to estimate loading (below): equation 1 was used to estimate zinc loading associated with tire wear, while equation 2 was used to estimate copper loading associated with brake pad wear.

Equation 1. Zinc loading associated with tire wear.

$$\frac{Zn}{Vehicle\ Type - County}\left(\frac{kg}{yr}\right) =$$

$$\sum\left(\frac{\mathbf{Kilometers\ Travelled}}{\mathbf{Month}}\right)\left(\frac{km}{mo}.\right) * \mathbf{Tire\ wear\ rate}\left(\frac{mg}{km-Tire}\right) * \mathbf{Tire\ Number} * \mathbf{Zn\ Tire\ Concentration}\ \left(\frac{mg}{kg}\right) *(10^{-12})$$

Equation 2. Copper loading associated with brake pad wear.

$$\frac{Cu}{Vehicle\ Type - County}\left(\frac{kg}{yr}\right) =$$

$$\sum\left(\frac{\mathbf{Kilometers\ Travelled}}{\mathbf{Month}}\right)\left(\frac{km}{mo}.\right) * \mathbf{Brake\ Pad\ Wear\ Rate}\left(\frac{mg}{km-vehicle}\right) * \mathbf{Cu\ Brake\ Pad\ Concentration}\ \left(\frac{mg}{kg}\right)*(10^{-12})$$

There is considerable variability for many of the parameters used to estimate the loading of copper and zinc associated with automobile use. To account for these various uncertainties, a Monte Carlo-type analysis was applied. The analysis took the form of generating results for 1000-iterations of loading estimates through the application of the Excel formula: NORMINV(rand(), mean, standard deviation). The formula generates a random parameter value based on sample mean and associated standard deviation. (The underlying assumption in the use of this formula is that the parameter distribution is normal.) The NORMINV() function was applied to the parameters: vehicle kilometers travelled, wear rates, and metals composition and, in the case of passenger cars and light trucks, the representation of brake pads as opposed to drums. Mean parameter values and assumed variability are included in Table 1 with further discussion in the report section titled: “Brake pad and tire wear rates and metals composition”.

Loading estimates were based at the county level by vehicle type and roadway system (highway, non-highway). Percentiles were determined from the 1000 loading estimates and a series of box plot figures generated from those estimates. The box plots provide a graphical display of the estimated loading variability.

Table 1. Assumed values and associated variability*****.

Parameters and assumed variability		
Brake Pads / Drums		
Parameter	**Mean value**	**Assumed standard deviation**
Vehicle kilometers travelled	=====	5% of parameter value
Copper representation	Pads =49552 mg Cu/kg brake material Drums = 2179 mg Cu/kg brake material	Pads = 12699 mg Cu/kg brake material Drums =2439 mg Cu/kg brake material
Representation of pads vs. drums on passenger cars and light trucks	1.66 axles full pads 0.34 axles front pads, rear drums	10% of parameter value
Brake pad / drum wear rates	Refer to Table 3	10% of parameter value
Tires		
Parameter	**Mean value**	**Assumed standard deviation**
Vehicle kilometers travelled	=====	5% of parameter value
Zinc representation	7434 mg Zn/kg tire material	3771 mg Zn/kg tire material
Tire wear rate	38 mg tire material/km travel	26 mg tire material/km travel

*Data sources and discussion included in section “Brake pad and tire wear rates and metal composition”.

Vehicle Miles Travelled (VMT)

Within Washington State, an annual estimation of vehicle miles travelled (VMT) for the state highway system is conducted by the Washington State Department of Transportation (WSDOT). (WSDOT reports vehicle travel in units of miles though this report uses kilometers.) The estimate is conducted in part to fulfill an annual reporting requirement of the Federal Highway Administration’s Performance Monitoring System (www.wsdot.wa.gov/mapsdata/tdo/annualtrafficreport.htm). The data is reported at the county level. In addition, as part of that requirement, VMT are also reported for non-highway or local road automotive use. This estimate is conducted at the local level (city and county) and reported annually to WSDOT. Within each county, both the highway and non-highway VMT estimates are further defined by rural and urban settings, based on Census Bureau designations.

From this information, and particular to this analysis, these annual VMT data were further divided into a monthly distribution by county and vehicle type. Twelve counties draining to greater Puget Sound were included in this assessment including Clallam, Island, Jefferson, King, Kitsap, Mason, Pierce, San Juan, Skagit, Snohomish, Thurston, and Whatcom. Vehicle types were divided into the following groups: motorcycle, passenger car, light truck, bus, single unit truck, and combination truck. The representation of these vehicle types on the urban and rural highway network is based on traffic survey data collected by WSDOT. The vehicle representations are expressed as a percent of the total annual VMT, providing an annual average use level. For this analysis, it was assumed that the representation of vehicle types present for the highway system is the same as that for the non-highway system. In addition, it was assumed that vehicle representation shows no seasonal (monthly) variation.

The standard deviation applied in the analysis for vehicle kilometers travelled was assumed ±5% around the reported levels (www.wsdot.wa.gov/mapsdata/tdo/annualtrafficreport.htm). This VMT categorization scheme was conducted to examine metals loading in terms of proximity, seasonal variation, and to determine the relative source levels by vehicle type. An outline of this classification scheme is presented in Table 2.

Table 2. The classification scheme applied to WSDOT vehicle miles travelled (VMT) data.

<table>
<tr><td rowspan="5">County</td><td colspan="5">Clallam, Island, Jefferson, King, Kitsap, Mason, Pierce, San Juan, Skagit, Snohomish, Thurston, Whatcom</td></tr>
<tr><td rowspan="4">Functional Use</td><td colspan="4">Highway, Non-Highway</td></tr>
<tr><td rowspan="3">Setting</td><td colspan="3">Urban, Rural</td></tr>
<tr><td rowspan="2">Reporting Period</td><td colspan="2">Monthly</td></tr>
<tr><td>Vehicle Type</td><td>Motorcycle, Passenger Car, Light Truck, Bus, Single Unit Truck, Combination Truck</td></tr>
</table>

Brake pad and tire wear rates and metals composition

Brake pad copper composition and wear rates

While a number of studies have been conducted to examine metal levels in brake pads, in particular copper levels, there is a wide range in their findings. Part of this variability has to do with the application of differing analytical procedures for the extraction, processing, and analysis of the brake pad samples examined. Further complicating matters is the fact that the representation of copper present in brake pads is highly variable. Brake pad composition is a complex of materials and considered proprietary; therefore, there is not a uniform process to their manufacture (BPP, 2011). Levels vary among the automobile manufacturers as well as among their various vehicle models. In addition, the copper representation in brake pads tends to be significantly higher in those originally equipped with the vehicle (OEM) in comparison to replacement pads. Not discussed among the various brake pad studies is the relation between the price of copper and its representation in brake pads, which also could be a factor affecting variability. Despite recent economic concerns, which caused a short though rapid drop in the price of copper, prices have risen by a factor of 5 since 2003. This may be a factor in the variability and, in particular, in the lower copper representation in the aftermarket pads in which profit margins are lower.

Study results define the average copper represented in brake pads, based on a broad spectrum of manufacturers and vehicle models, at between 4 and 12%, with similar levels in materials reported for Europe (Table A-1). A weighted average based on original manufacturers equipment (OEM) brake pads from various manufacturers and their respective vehicle models, determined the representation of copper at 4.2% (Armstrong, 1994). Similarly, an overall weighted average, based on the percentage of vehicles equipped with OEM, as opposed to replacement pads, determined a copper representation of 5.2% (Hjortenkrans, 2007). The OEM copper representation was found to be 13% while the replacement pads were significantly lower at 0.015%. An assessment of brake pads, also considering OEM as opposed to replacement, though including differences in pad location, designated for front wheel brakes as opposed to rear wheel brakes, found average copper levels of 11.8% and 9.2% for OEM front and rear, respectively, and 7.2% and 5.1% for front and rear replacements (Westerlund, 2001). An analysis of copper levels in OEM pads, based on a compilation of manufacturers and models, which together represented about 40% of the passenger car traffic in the San Francisco Bay area, over an 8-year period (1998-2005), found an average of 5% (Rosselot, 2006).

Appropriately, due to its high representation among traffic, the primary focus of brake pad study has concerned passenger as opposed to commercial vehicles. It is generally assumed that commercial vehicles utilize brake drums that, due to lower braking temperatures, contain less copper. What material that is worn is further contained within the brake drum, resulting in a lower overall emission level (Rosselot, 2006). When analyzed, brake drums associated with commercial vehicles were found to have a fraction of the copper typically found in those associated with passenger cars; 0.2% as opposed to 5% (Uexkull, 2003).

This analysis assumes an average representation of copper present in the brake-pads of motorcycles, passenger vehicles, and light trucks at 5%, with a standard deviation of 1.3% (Brake Pad Partnership, 2005) (Table 3). The representation of brake-pads in light trucks and passenger vehicles is variable depending on manufacturer, model, year of production, among other factors. Almost all recent light trucks and passenger cars have brake-pads present on the front axle with representation on the rear axle the reason for the variability. This analysis assumes that 66% of passenger cars and light trucks have full brake-pad coverage with 34% of the vehicles having front brake-pads and rear drums. This assumption is consistent with prior reported estimates (Rosselot, 2006) and those provided by several local automobile repair shops. Around these figures, a standard deviation of 10% was assumed.

It is assumed that commercial vehicles utilized brake drums that have an average copper representation of 0.22% with a standard deviation of 0.24% (Uexkull, 2003). The vehicles assumed to represent commercial vehicles include buses, single unit and combination trucks.

An analysis of deposition rates and composition of traffic-related roadway deposits, conducted prior to the wide use of the brake pads, determined a copper deposition rate of 0.0801 mg Cu/km-axle (Shaleen, 1975). The assumption of a 0.2% copper composition applied to an average estimated drum brake wear rates per axle of 43 mg/km-axle (average of bus, single unit and combination trucks) (Table 2), results in a copper deposition rate of 0.086 mg Cu / km-axle, similar to the pre-brake pad loading level. This estimate provides further foundation to the assumption of lower copper representation of brake drums as opposed to brake pads.

The average wear rate, by vehicle type, of brake pads (drums) is based on the distance traveled to achieve a 70% loss in mass of the original braking material. Table 3 includes estimates of brake material wear rates applied in this analysis. The estimates for automobile, bus, and light goods truck wear rates were those reported in Westerlund (2001) while the rates used for single unit and combination trucks were those from Rosselot (2006). The assumed brake pad wear rate for motorcycles was 3 mg/kg, based on the ratio of the average vehicle weight to pad number (341 kg/pad for automobile (www.nhtsa.gov/cars/rules/CAFE/DomesticCarFleet.htm) as opposed to 68 kg/pad for motorcycle (http://motorcycle-specs.com/Index.asp). Therefore, motorcycles have an 80% lower vehicle weight to pad metric in comparison to automobiles. For this analysis, it was assumed that standard deviation associated with brake pad wear is 10% of the mean value.

Table 3. Assumptions applied to calculate copper loading associated with brake pad wear.

Vehicle Type	**Per Vehicle Brake pad/drum wear rate (mg/Km)**	**Per Vehicle No. axles**	**Per Vehicle Wear rate based on axles (mg/axle-km)**	**Representation of copper in brake material (mg Cu/kg)**		**Per Vehicle Average copper emission rate (mg Cu/km)**
				Average	**Standard Deviation**	
Motorcycle	3	1	3	49552 [4]	12699	0.15
Automobile [1]	16 [2]	2	8	49552	12699	0.66
Bus	110 [2]	3	37	2179 [5]	2439	0.24
Light Truck [1]	16 [2]	2	8	2179	2439	0.66
Single Unit Truck	129 [3]	3	43	2179	2439	0.28
Combination Truck	245 [3]	5	49	2179	2439	0.53

1. Assumes a weighting factor of 1.66 axles with brake-pads and 0.34 axles with drums, ± 10%.
2. Brake Pad Partnership, 2005
3. Uexkull, 2003
4. Westerlund, 2001
5. Rosselot, 2006

Vehicle tire zinc composition and wear rates

The representation of zinc in automobile tires is about 1% of composition by weight (Councell, 2004). Reported zinc composition levels for retread and non-retread tires were found to average 1.2% and 0.94%, respectively (Hjortenkrans, 2007). Compilations of literature values found reported zinc compositions ranging between 0.04 to 1.6% with a mean level of 0.7% (7434 mg Zn/kg tire tread) (EMEP, 2006; Councell, 2004).

Reported tire wear rates range between 10 and 97 mg thread/km with median levels approximately 40 mg/km (Councell, 2004). (Appendix A contains a calculation of tire wear rate for a common passenger car, finding a wear rate of 38 mg/km.) Few studies focus on the tire wear rates of commercial vehicles though reported rates are comparable to those found for passenger vehicles (EMEP, 2006). This is likely due to the larger number of tires supporting the increased weight of commercial vehicles, balancing stresses.

An analysis of deposition rates and composition of traffic-related roadway deposits determined a zinc deposition rate of 0.987 mg Zn / km-axle (Shaheen, 1975). Considering a tire-wear rate of 38 mg/km, a 1% Zn composition, and a weighted-average tire per axle of 2.14 (based on the representation of the various vehicle-types considered and their associated tire number – refer to Table 4), would result in a loading rate of 0.813 mg Zn / km-axle, similar to the prior estimate. This is not unexpected in that while the advent of the brake pad resulted in increased copper composition for passenger vehicles, the level of zinc in automotive tires has remained at a similar level for an extended period (Councell, 2004). Also, the estimated lower loading rate is appropriate given that there are additional automotive sources of zinc than tires, associated with, for instance, the wearing of metal parts (i.e. brake pads) along with being an additive to lubricants and petrol.

This analysis assumes an average tire wear rate of 38 mg/km, with a standard deviation of 26 mg/km. These metrics are derived from literature-compiled rates reported in Councell (2004) and EMEP (2006). The average zinc composition associated with this wear is assumed to be 7,434 mg Zn/kg tire tread (0.74%) with a standard deviation of 3771 mg Zn/kg tire tread (EMEP, 2006). (The standard deviation was estimated based on half the reported range divided by the square root of three.) The assumed number of axles and associated tires by vehicle type is presented in Table 4.

Table 4. Assumptions applied to calculate zinc loading associated with tire wear.

Vehicle Type	**Tire Wear Rate (mg/Km)**	**Per Vehicle No. Axles**	**Per Vehicle No. Tires**	**Per Vehicle No. Tires per axle**	**Representation of Zinc (mg Zn/kg)**		**Per Vehicle Zn Emission Rate (mg Zn/km)**
					Average	**Standard Deviation**	
Motorcycle	38 [1]	1	2	2.00	7434 [2]	3771	0.56
Automobile	38	2	4	2.00	7434	3771	1.13
Bus	38	3	8	2.67	7434	3771	2.26
Light Truck	38	2	4	2.00	7434	3771	1.13
Single Unit Truck	38	3	8	2.67	7434	3771	2.26
Combination Truck	38	5	18	3.60	7434	3771	5.08

1. Councell, 2004
2. EMEP, 2006

Considering proximity of loading

The metals loading rates (mg/km) presented in Tables 3 and 4 are based on vehicle type, their associated brake pad and tire wear rates, and the average composition of copper and zinc, respectively. It is recognized that there are various pathways that brake pad and tire material can take including introduction to air, retention on the vehicle under carriage, and direct deposition to the street surface. However for this analysis, the loading estimates are based on the assumption that the brake-pad and tire material, once emitted, is all eventually deposited to the road surface in proximity (at county level) to the point of travel.

Applying the wear rates implies that there is an equitable distribution to the metals loading. However, in reality the proximity of the loading will be quite varied. For instance, some tire wear will always occur, to varying levels, with movement of the vehicle. In contrast, brake pad wear only occurs with application of the brake mechanism. So, for highway travel under conditions of low traffic volume, very little brake pad wear can be expected per kilometer travelled. However, braking at higher speeds in more urbanized highway sections (in high to moderate traffic volumes) results in considerably higher wear rates. The highest metals loading associated with brake pad wear is expected to occur in urbanized highway and non-highway (local) roadways due to the increased frequency and intensity of braking. Recognizing this variability, the vehicle kilometers travelled and, ultimately, loading, was categorized by type of road system (highway and non-highway) and setting (urban and rural), providing a surrogate of traffic intensity.

Results

While an assessment of variability was included when calculating annual loading rates, for this discussion only the overall median values will be considered. This is mainly to keep the discussion focused on relative comparisons. Variability has not been completely ignored, but instead is considered in the series of figures found throughout the results section in the form of box plots. Regarding the interpretation of the box plot graphic: the upper and lower sides of the central box indicate the 75th and 25th percentiles of the dataset; the dot within the box is the median (50th percentile); while the upper and lower circles (at end of upper and lower whisker extensions) are the 90th and 10th percentile of the load estimates.

Vehicle kilometers travelled

An estimated 58 billion (10^9) kilometers were travelled throughout the Puget Sound study area in 2008. 55% of the travel occurred on the highway system with the remainder (45%) occurring on the non-highway (local) road system (Figure 1). Among the Puget Sound counties considered by this analysis, travel within King County represented approximately 45% of the annual total, more than twice that of Pierce County (17%), the county with the next greatest level. Travel within King, Pierce, and Snohomish counties together accounts for 77% of the greater Puget Sound area total (Table 5).

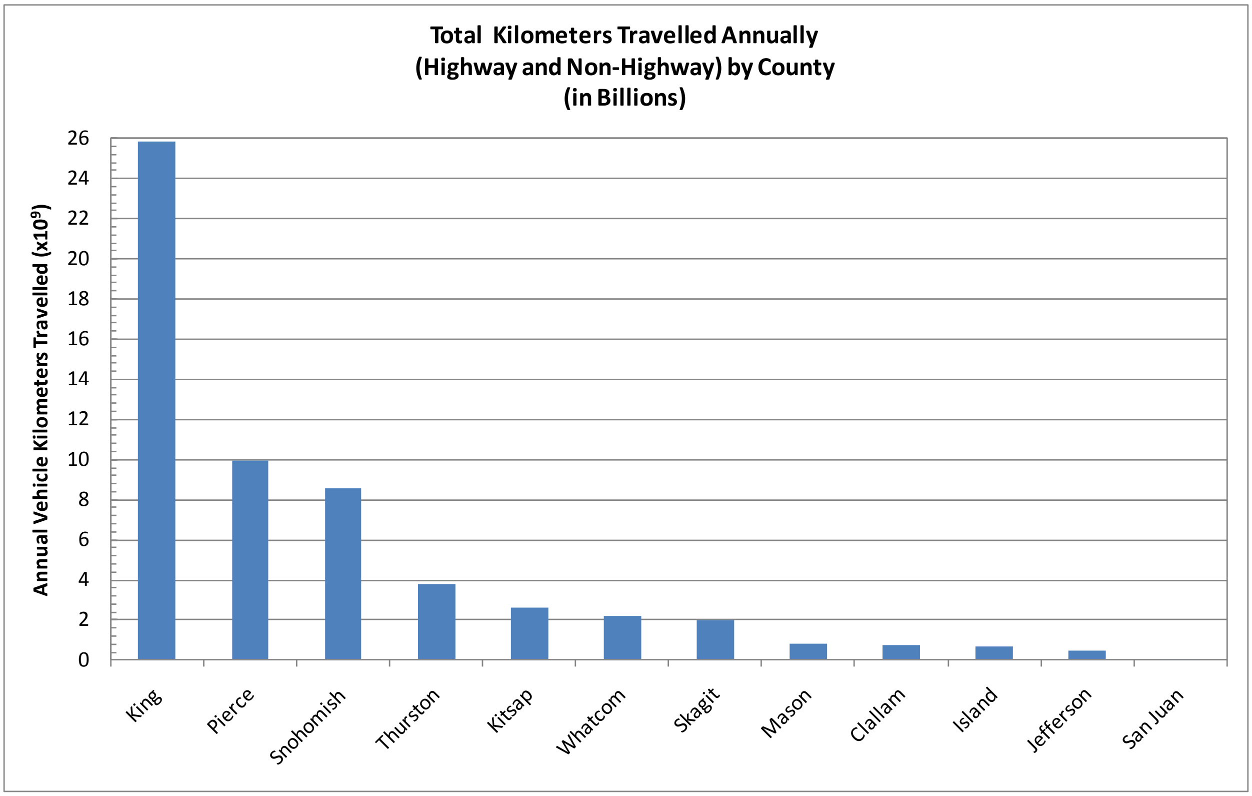

Figure 1. Total vehicle kilometers travelled on both highway and non-highway roads, by county.

Travel within these counties is polarized. For instance, 91% of the annual VKT in King, Pierce, and Snohomish Counties occurs in their urbanized western portions, along the Interstate-5 corridor (Figure 2).

In contrast, the majority of the VKT occurring in counties situated outside of the Interstate-5 corridor (including Clallam, Island, Jefferson, and Mason), is rural-based. For these counties, approximately 77% of the annual VKT occurs on rural roads, with the other 23% occurring in an urbanized setting. (San Juan County has no highway road system or urbanized setting.) Thurston, Skagit, and Whatcom counties are also bisected by Interstate-5 but the road systems in these counties remain primarily rural with a smaller urbanization base. For this reason, the travel in these counties is more evenly split between the urbanized and rural road systems. Kitsap County, though also outside of the I-5 corridor, has a level of urbanization and highway road system more reflective of the Puget Sound urban core (King, Pierce, and Snohomish Counties) and, for this reason, is an anomaly among the study counties.

Table 5. The annual VKT (considering all vehicle types) by county and setting.

Annual (2008) Vehicle Kilometers Travelled (VKT), By County - All Vehicles Combined

County	**Highway (VKT)**		**Non-highway (VKT)**		**Total**	**% of Total**
	Urban	**Rural**	**Urban**	**Rural**		
Clallam	97,299,288	378,013,946	133,126,937	144,611,190	753,051,361	1.30
Island	43,158,368	287,366,106	144,593,651	187,968,248	663,086,374	1.15
Jefferson	35,009,427	305,600,742	59,605,294	87,337,644	487,553,106	0.84
King	12,300,087,970	1,223,423,233	12,047,277,095	288,829,021	25,859,617,319	44.76
Kitsap	1,110,058,763	341,035,909	901,614,425	247,285,322	2,599,994,419	4.50
Mason	24,547,226	442,867,264	71,457,049	265,670,768	804,542,306	1.39
Pierce	4,512,323,884	460,243,337	4,783,037,335	226,948,614	9,982,553,170	17.28
Skagit	596,466,763	779,742,298	379,555,153	214,187,995	1,969,952,209	3.41
San Juan	0	0	0	59,569,789	59,569,789	0.10
Snohomish	4,566,913,715	834,985,220	2,887,046,507	297,244,484	8,586,189,926	14.86
Thurston	1,156,157,579	786,993,578	1,199,591,739	662,203,992	3,804,946,889	6.59
Whatcom	629,810,232	606,256,865	607,778,181	353,887,944	2,197,733,222	3.80
Total =	25,071,833,215	6,446,528,497	23,214,683,366	3,035,745,010	57,768,790,088	100.00
% of Total=	43.40	11.16	40.19	5.25	=======	=======

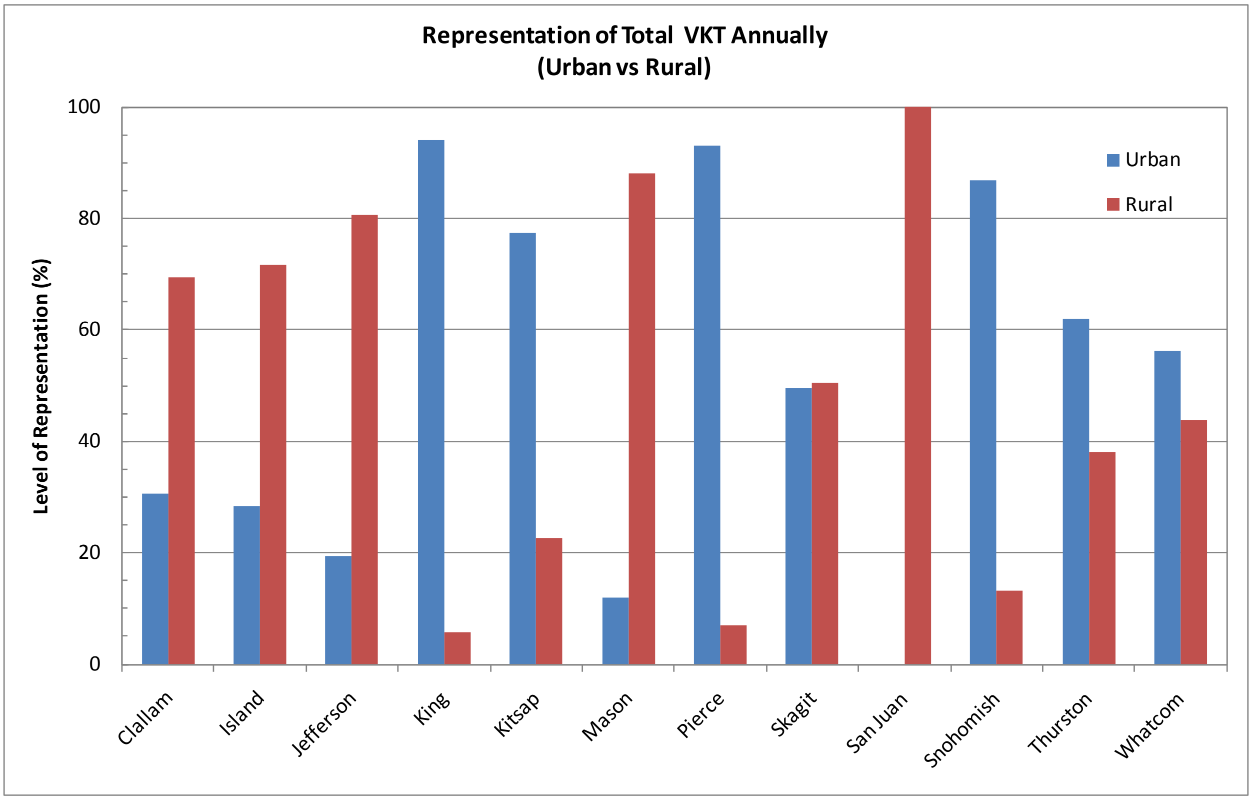

Figure 2. The representation of total annual vehicle kilometers travelled (VKT) for highway and non-highway roads by setting, urban or rural.

In this analysis, VKT is a major factor in estimating automotive-related metals loading and, as will be shown, the majority of the automotive metals loading within the Puget Sound study area occurs within King, Pierce, and Snohomish Counties.

Seasonal use

The distribution of travel by month is presented in Figure 3. Within the Study Area, the greatest travel period occurs in July and August with 9.3% and 9.5%, respectively, of the annual total. As expected, lower travel levels occur during the winter months with an annual low occurring in December, accounting for 6.9% of the total. While December sets an annual low to the VKT metric, it is only 2.6% lower than the August peak, indicating an underlying steady and relatively high travel base to the seasonal variation.

The seasonal use pattern is important to recognize in terms of potential water quality impacts. From the seasonal VKT pattern, the greatest automotive-related metal loading occurs during the summer months. Travel during July and August account for about 19% of the annual total. This is a period when precipitation, and therefore storm water runoff, is at an annual low. For this reason, it is expected that these loadings accumulate on road surfaces and are only transported to receiving waters at greater levels following the onset of more sustained and intense precipitation events in the fall and winter.

Representation by vehicle-type

Among the vehicle types considered by this study, passenger cars are the dominant form of transportation within the Puget Sound counties, accounting for approximately 63% of all VKT in 2008 (Figure 4). Light trucks are a distant second at 26%. The total for the other vehicles considered including combination trucks (6.0%), single unit trucks (4.8%), motorcycles (0.4%), and buses (0.2%) together account for the remaining 11%.

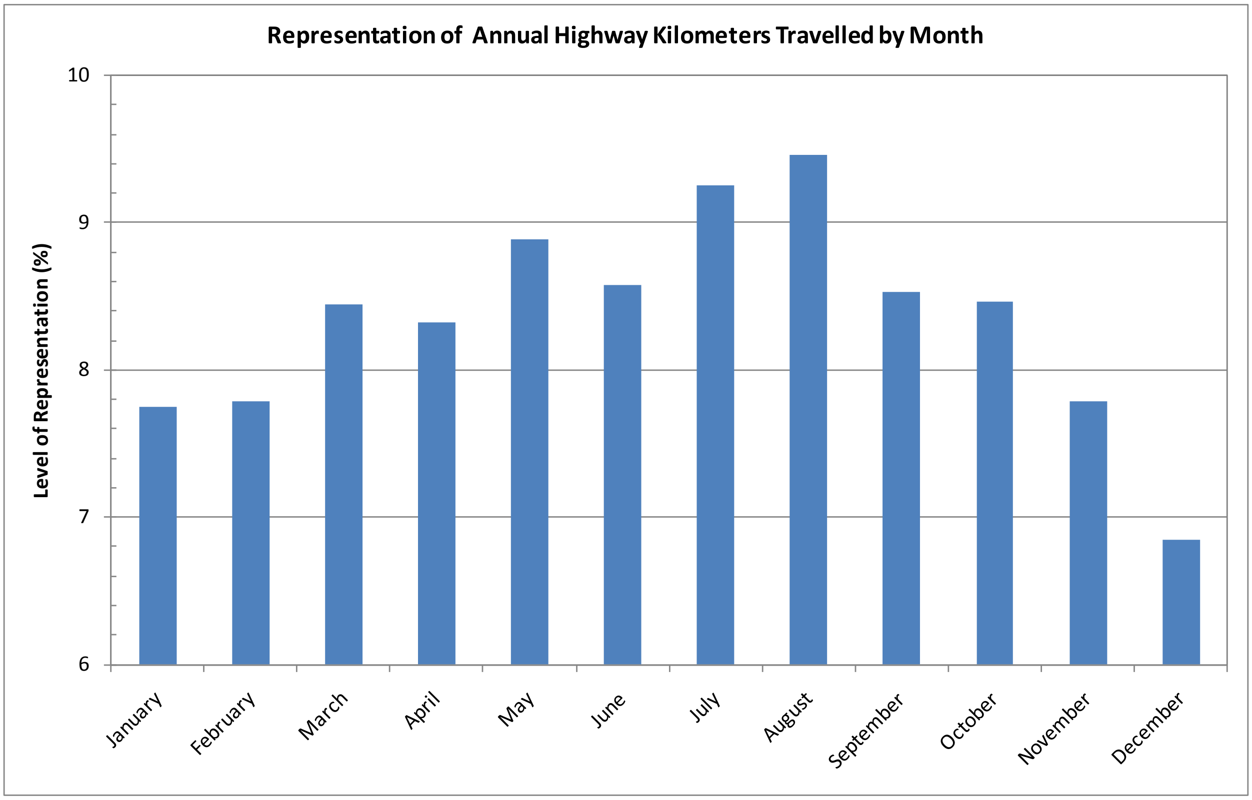

Figure 3. Monthly variation to annual highway-based vehicle kilometers travelled.

Based on these statistics, passenger cars within the urban road system of King County alone account for approximately 26% (15 billion VKT) of the 58 billion VKT total for all vehicle travel within the Study Area in 2008. As it will be shown, the dominance of passenger car travel within the Puget Sound basin, particularly King County, also results in it being the major source for metals loading associated with tire and brake pad wear.

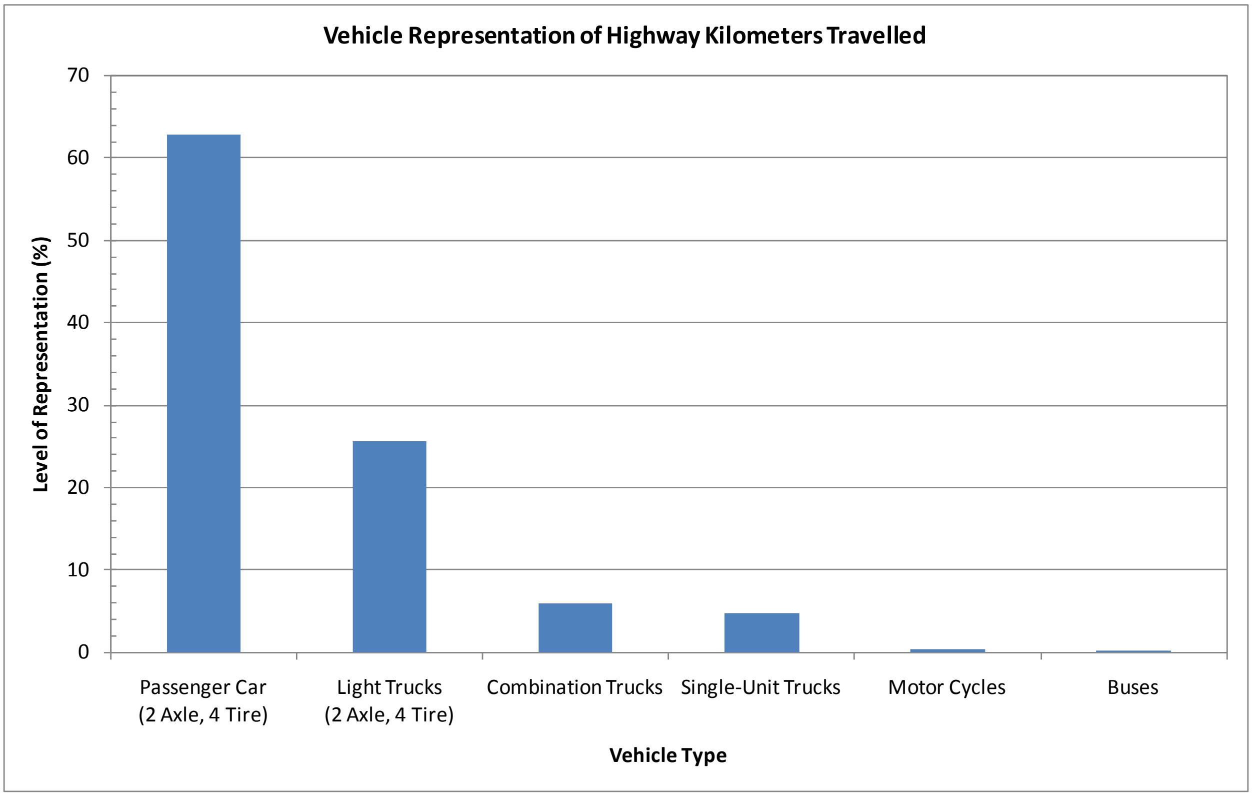

Figure 4. The representation of the six vehicle types of the total 2008 VKT occurring within the Puget Sound counties considered by this analysis.

Automotive metals loading

Zinc loading associated with tire wear

The average annual zinc load to road surfaces, associated with automotive tire wear, is estimated at 80 tonnes (t) for the Puget Sound counties considered (Table 6). (1 tonne (t) = 1000 kilograms). Approximately 44% of the zinc load (35 t) is associated with vehicle travel occurring solely within King County (Table 6, Figure 5). Together, King, Pierce, and Snohomish counties accounted for 76% (61 t) of the estimated 2008 total.

A tire-based zinc yield can be calculated for each county by dividing its annual load by the total lane kilometers (highway and local roads) within its jurisdiction. (The lane kilometer metric is a measure of the linear length of road accounting for the number of lanes present.) Considering the entire study area, the overall yield is 914 grams per kilometer per year (g/km-yr). Study area yields varied from 1222 g/km-yr for King County to 77 g/km-yr for San Juan County (Figure 6). Counties with the highest loads tended to also have the highest yields indicating a relationship between VKT and road length (refer to Appendix A). The zinc yields for Pierce, Snohomish, and Thurston Counties were similar with an overall median of 982 g/km-yr. (The increased yield for Thurston County indicates that there is a higher level of vehicular travel relative to the lane kilometers compared to the other counties.) As reference, an assessment in the United Kingdom found a tire-based zinc yield of 1435 g/km-yr, while in Germany highway-based yields were estimated at 810 g/km-yr (Councell et al., 2004).

Table 6. The annual (2008) zinc loading associated with tire wear (kg) by county and road system.

Annual zinc loading associated with tire wear (kg) by county and road system.						
County	**Highway**		**Non-Highway**		**Total (by County)**	**% Representation**
	Urban	**Rural**	**Urban**	**Rural**		
Clallam	135.8	633.3	172.5	188.5	1130.0	1.4
Island	60.2	481.4	187.3	245.0	974.0	1.2
Jefferson	48.9	512.0	77.2	113.8	751.9	0.9
King	17164.6	2049.6	15606.5	376.4	35197.2	44.1
Kitsap	1549.1	571.3	1168.0	322.3	3610.7	4.5
Mason	34.3	741.9	92.6	346.3	1215.0	1.5
Pierce	6296.9	771.1	6196.1	295.8	13559.9	17.0
Skagit	832.4	1306.3	491.7	279.2	2909.5	3.6
San Juan	0.0	0.0	0.0	77.6	77.6	0.1
Snohomish	6373.0	1398.9	3740.0	387.4	11899.3	14.9
Thurston	1613.4	1318.5	1554.0	863.1	5349.0	6.7
Whatcom	878.9	1015.7	787.3	461.2	3143.1	3.9
Total	34987.3	10800.0	30073.2	3956.7	79817.2	100.0
% Representation	43.8	13.5	37.7	5.0	=====	=====

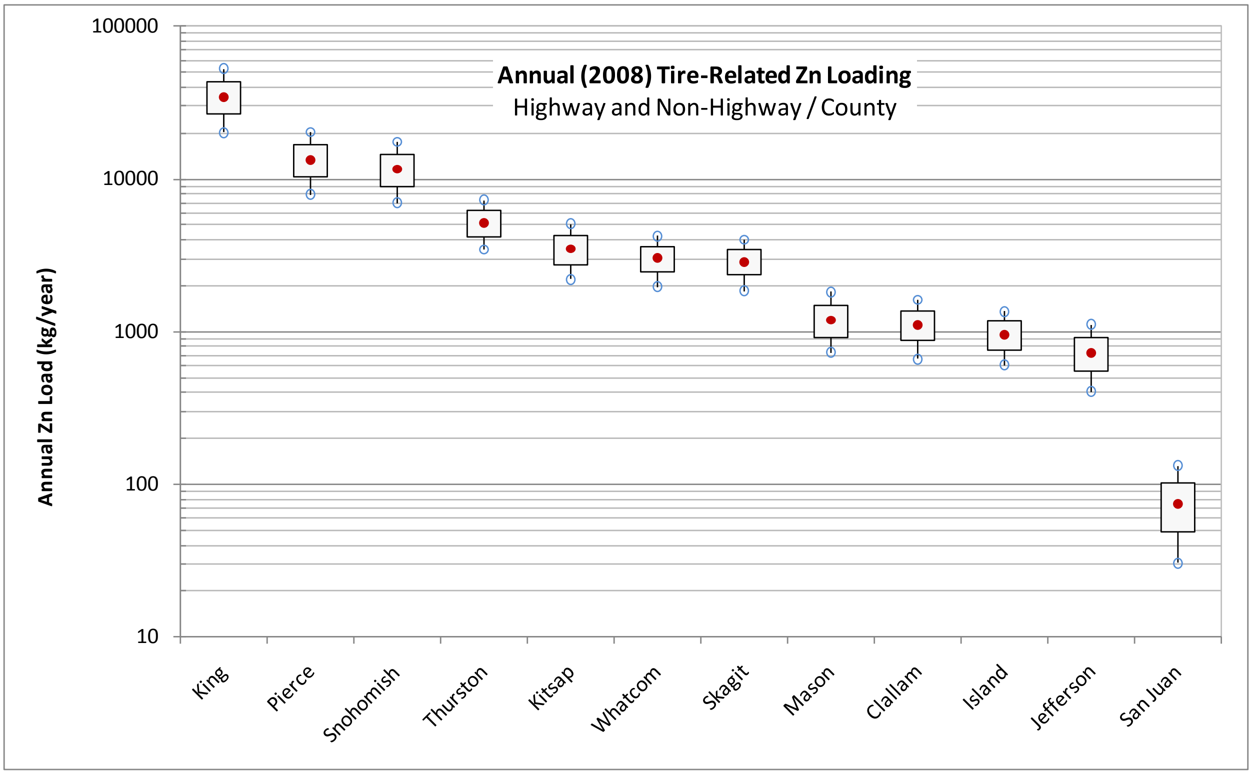

Figure 5. The annual zinc load (kilograms) estimated for the combined highway and non-highway road system, by county.

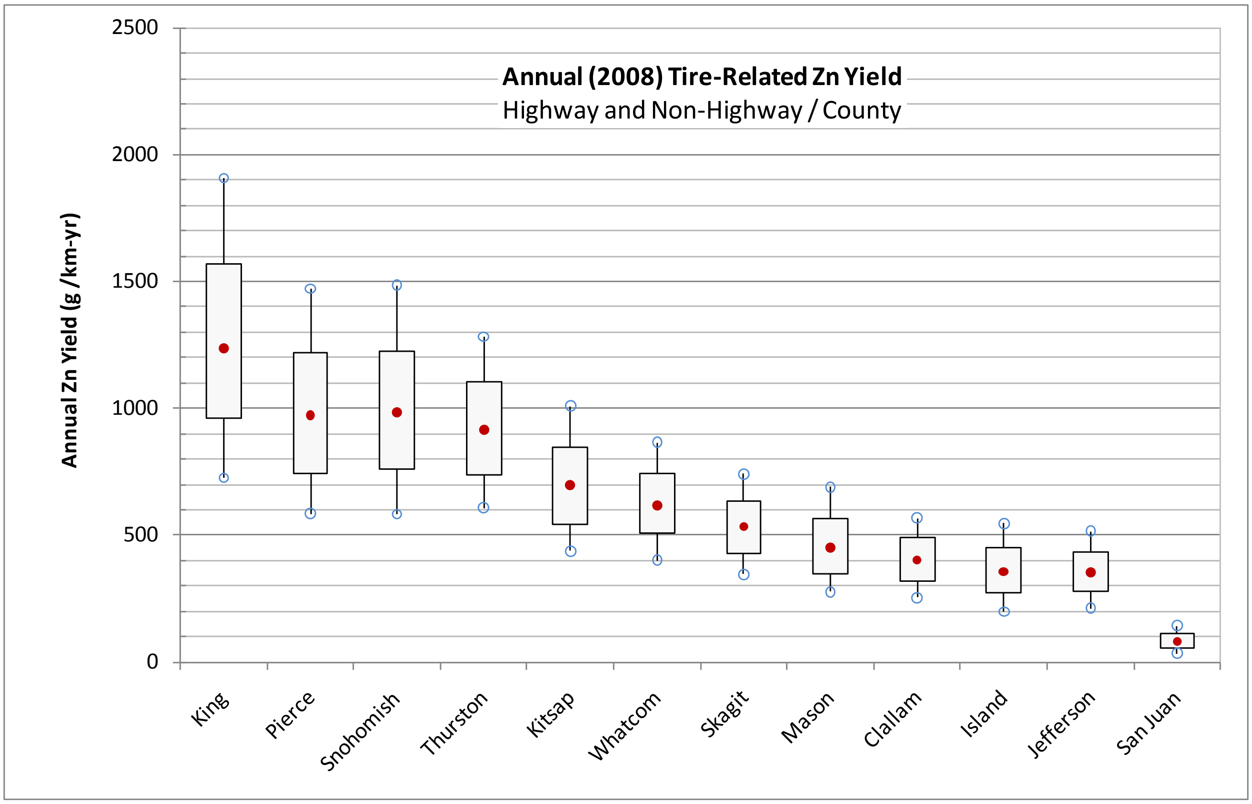

Figure 6. The zinc yield (g/km-yr) based on the annual load and total lane miles (highway and local), by county.

Highway-based zinc loading

Focusing solely on highway-based loading and considering both urban and rural settings, the zinc load in King County accounts for 41% (19 t) of that estimated for all the Puget Sound counties considered (45.8 t) (Figure 7). In comparison, both Snohomish and Pierce Counties had significantly lower load levels at approximately 7.8 t and 7.1 t, respectively. Together, the estimated loading for King, Snohomish, and Pierce counties represents about 74% (34 t) of the Puget Sound total for highway-based zinc loading. As observed, within the Puget Sound counties the vast majority of the load occurs within the urbanized setting representing 76% (35 t) of the total highway-based zinc load with the majority occurring with the King, Snohomish, and Pierce urbanized corridor which represent 65% (30 t) of the total.

Non-highway-based zinc loading

Zinc loading occurring on the non-highway-based (local) road system exhibited a similar pattern as observed for the highway system though at a lower level, 34 t as opposed to 46 t (Figure 8). Again, the greatest load occurs in the urbanized roads of King, Snohomish, and Pierce counties, which together represent 78% of the total load attributed to this grouping. The estimated zinc load associated with the urbanized road system of King County, in particular, was about 16 t or 47% of the annual total. The urbanized local road systems of Pierce and Snohomish Counties accounted for 19% (6.5 t) and 12% (4.1 t) respectively, of the annual total.

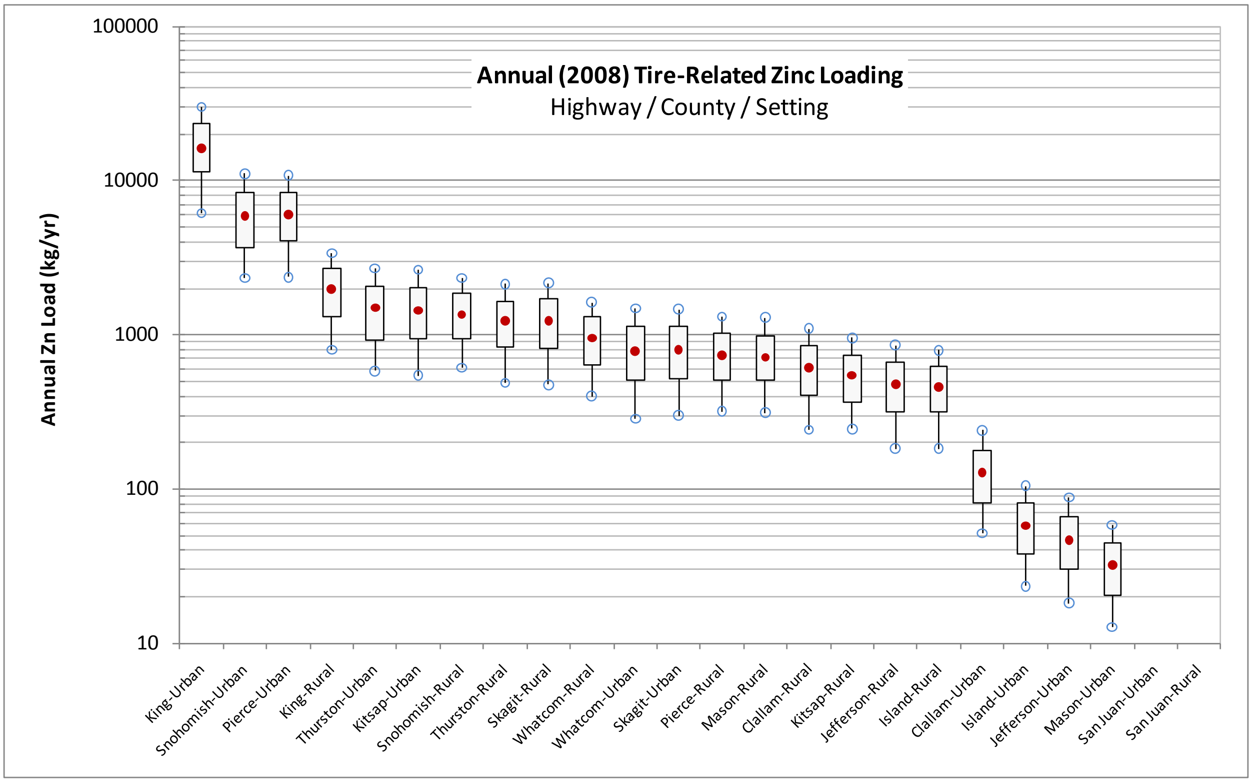

Figure 7. The annual zinc load for the highway system based on county and setting.

Zinc loading by vehicle-type

Based on vehicle type, the major source of tire-related zinc loading are passenger vehicles, representing about 50% and 53% for the total annual loading occurring in the highway and non-highway road systems, respectively (Table 7, Figures 9 and 10). As previously indicated, the majority of this loading occurs on urbanized roads. Zinc loading associated with combination trucks and light trucks comprise the next greatest sources at 24% and 19% of the highway-based load. Light trucks contribute a greater percentage of the non-highway based load at 27% as opposed to combination trucks at 10%. Combination trucks are primarily highway-based and this is reflected in these loading rates.

Together, the three most dominant tire-based zinc sources – passenger cars, light trucks, and combination trucks, for the highway and non-highway road system, comprise 92% and 90%, respectively, of the total loading. The zinc loading associated with passenger cars just in the urbanized road system of King County represents 21% (17 t) of the entire annual load associated with highway and non-highway travel throughout the Study Area.

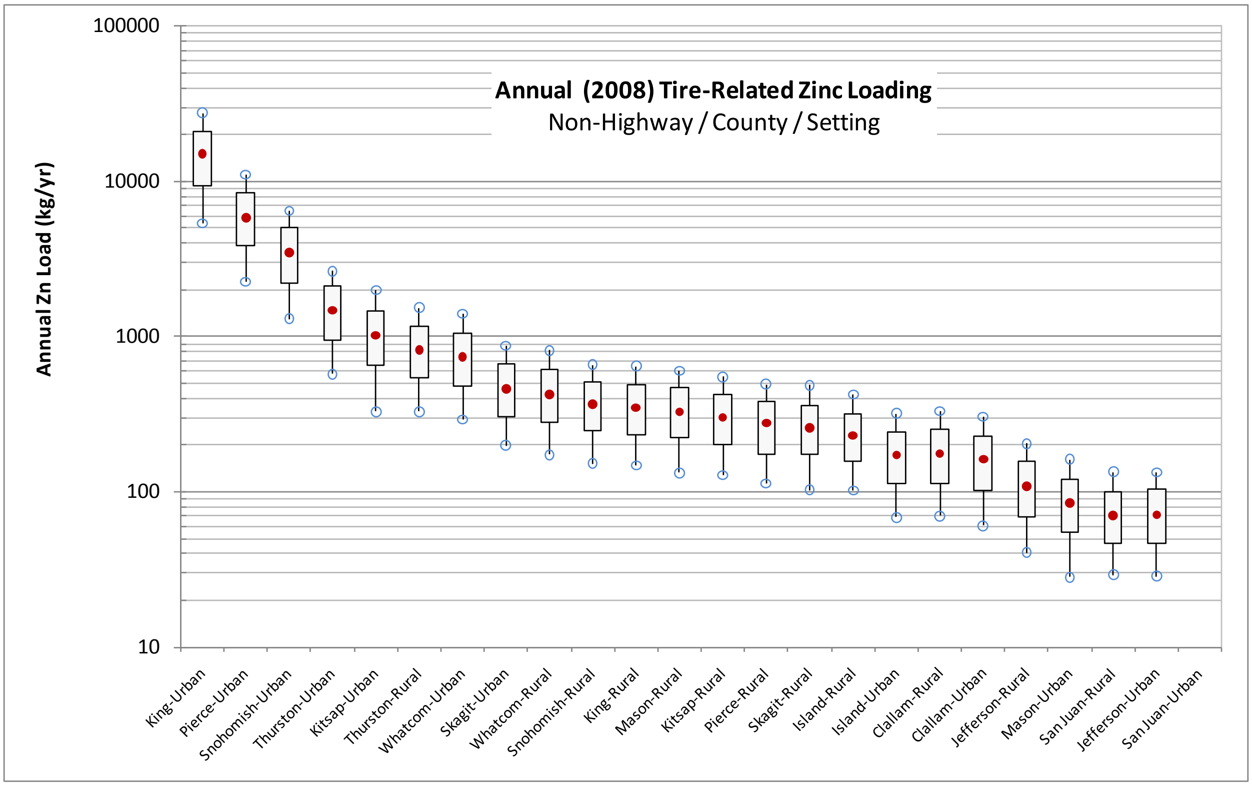

Figure 8. The annual zinc load for the non-highway road system by county and setting.

Table 7. The annual (2008) zinc loading associated with tire wear (kg) by county and road system.

Annual zinc loading associated with tire wear (kg) by vehicle type and road system.						
Vehicle-Type	**Highway**		**Non-Highway**		**Total (by Vehicle Type)**	**% Representation**
	Urban	**Rural**	**Urban**	**Rural**		
Motor Cycles	40.8	9.3	54.3	16.6	121.0	0.2
Passenger Car (2 Axle, 4 Tire)	18383.3	4189.8	15792.6	1950.8	40316.5	50.5
Light Trucks (2 Axle, 4 Tire)	7014.3	1763.5	8209.5	1098.3	18085.6	22.7
Buses	130.3	43.4	110.2	13.7	297.6	0.4
Single-Unit Trucks	2494.8	812.1	2712.4	526.0	6545.2	8.2
Combination Trucks	6923.8	3982.1	3194.3	351.2	14451.3	18.1
Total (by road system and setting)	34987.3	10800.0	30073.2	3956.7	79817.2	100.0
% Representation	43.8	13.5	37.7	5.0	=====	=====

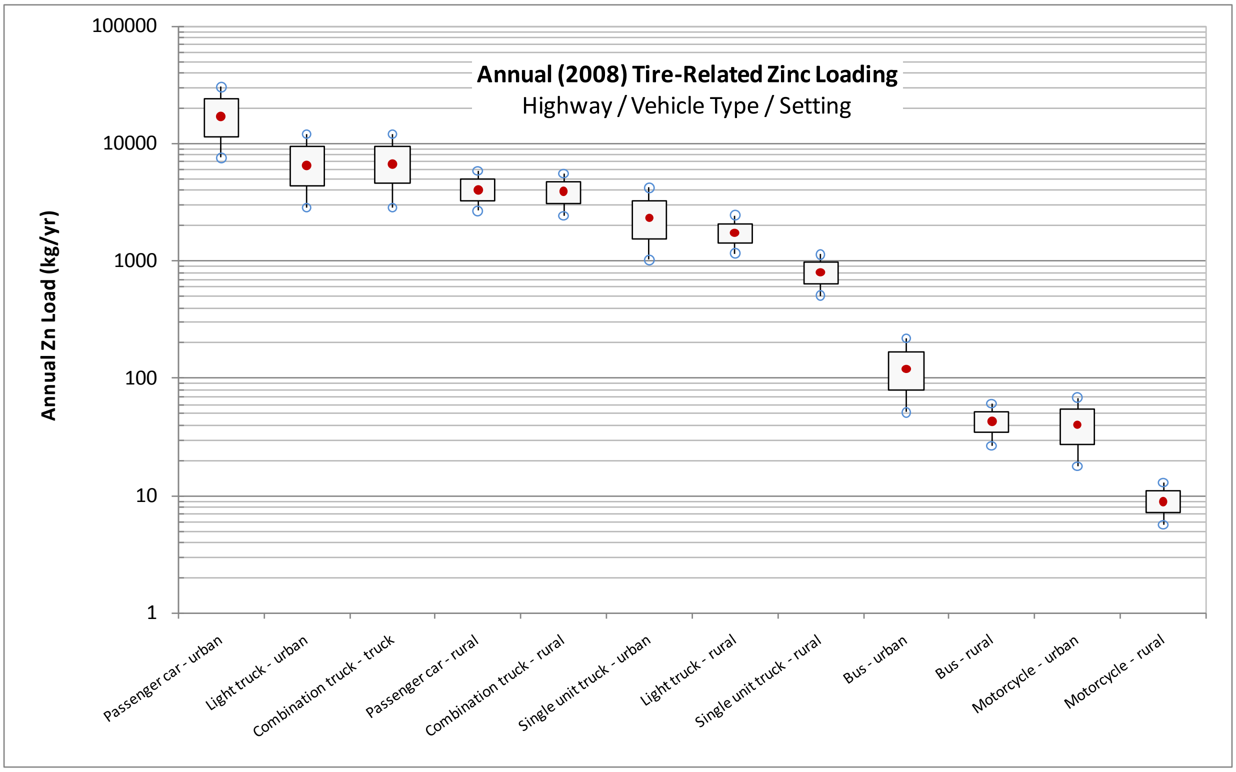

Figure 9. The annual zinc load estimated for the highway road system by vehicle type and setting.

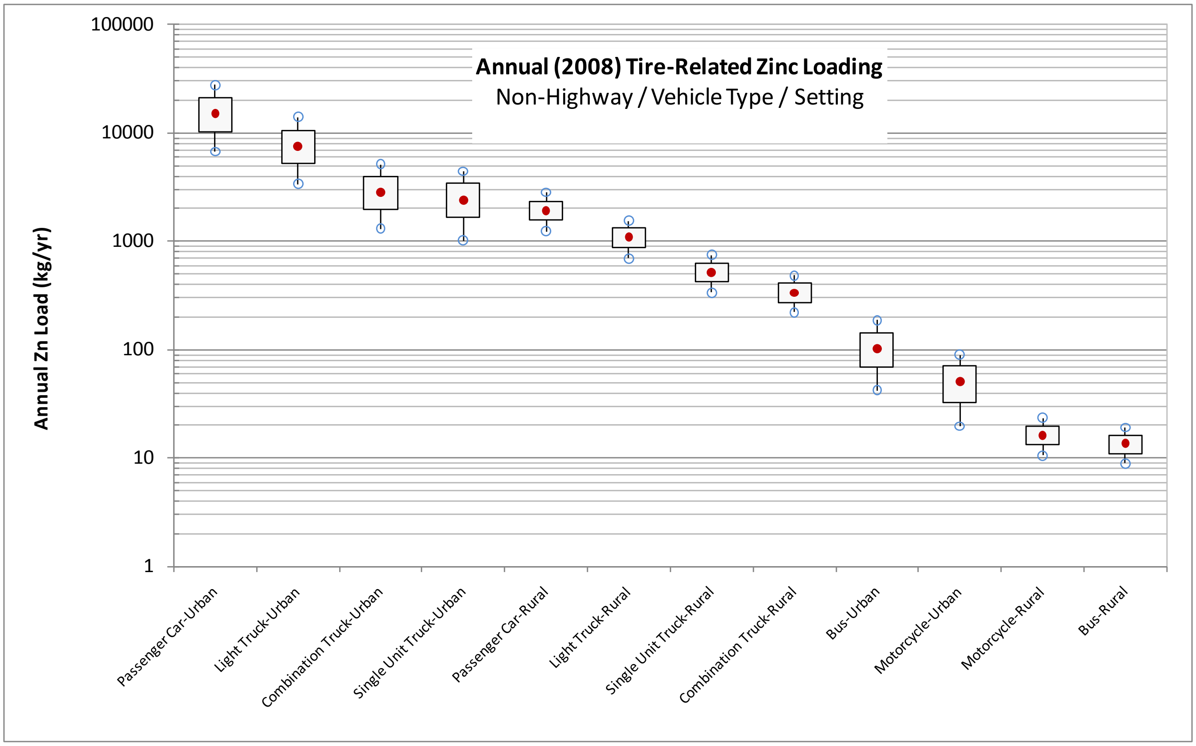

Figure 10. The annual zinc load estimated for the non-highway (local) road system by vehicle type and setting.

Copper loading associated with brake pad wear

The estimated annual copper load associated with automotive brake pad wear for the Puget Sound study area is 37 t. Approximately 45% of the load (16 t) is associated with travel solely within King County, followed by Pierce (17%, 6 t) and Snohomish (15%, 5 t). Together, the copper load in these counties accounts for 77% of the annual total (Table 8, Figure 11).

Table 8. The annual copper loading associated with brake wear (kg) by county and road system.

Annual copper loading associated with brake wear (kg) by county and road system.						
County	**Highway**		**Non-Highway**		**Total (by County)**	**% Representation**
	Urban	**Rural**	**Urban**	**Rural**		
Clallam	62.0	236.0	84.9	90.5	473.4	1.3
Island	27.5	179.4	92.2	117.6	416.7	1.1
Jefferson	22.3	190.8	38.0	54.7	305.8	0.8
King	7842.4	763.7	7681.8	180.8	16468.7	44.9
Kitsap	707.8	212.9	574.9	154.8	1650.3	4.5
Mason	15.7	276.5	45.6	166.3	503.9	1.4
Pierce	2877.0	287.3	3049.8	142.0	6356.2	17.3
Skagit	380.3	486.8	242.0	134.0	1243.1	3.4
San Juan	0.0	0.0	0.0	37.3	37.3	0.1
Snohomish	2911.8	521.2	1840.9	186.0	5460.0	14.9
Thurston	737.2	491.3	764.9	414.4	2407.8	6.6
Whatcom	401.6	378.5	387.5	221.5	1389.0	3.8
Total	15985.6	4024.4	14802.5	1899.8	36712.2	100.0
% Representation	43.5	11.0	40.3	5.2	=====	=====

The overall automotive copper yield for the Study Area is 424 g/km-yr though varied from 588 g/km-yr for King County to 39 g/km-yr for San Juan County (Figure 12).

Of the total estimated load, approximately 55% (20 t) is associated with highway-based travel, 80% of which occurs within the urban road network (Figure 13).

As discussed earlier, some level of tire wear occurs once the vehicle is in motion. However, brake-pad wear only occurs on actuation of the brake mechanism. For this reason, it is expected that the loading of copper associated with brake-pad wear occurs more prominently in urban settings where, due to higher traffic volumes (highway) and management measures (traffic lights, stop signs) higher brake use and, therefore, wear occurs. Therefore, a conservative estimate of the brake pad copper load can be determined by assuming that it occurs solely in the urbanized setting of the highway and non-highway roadways.

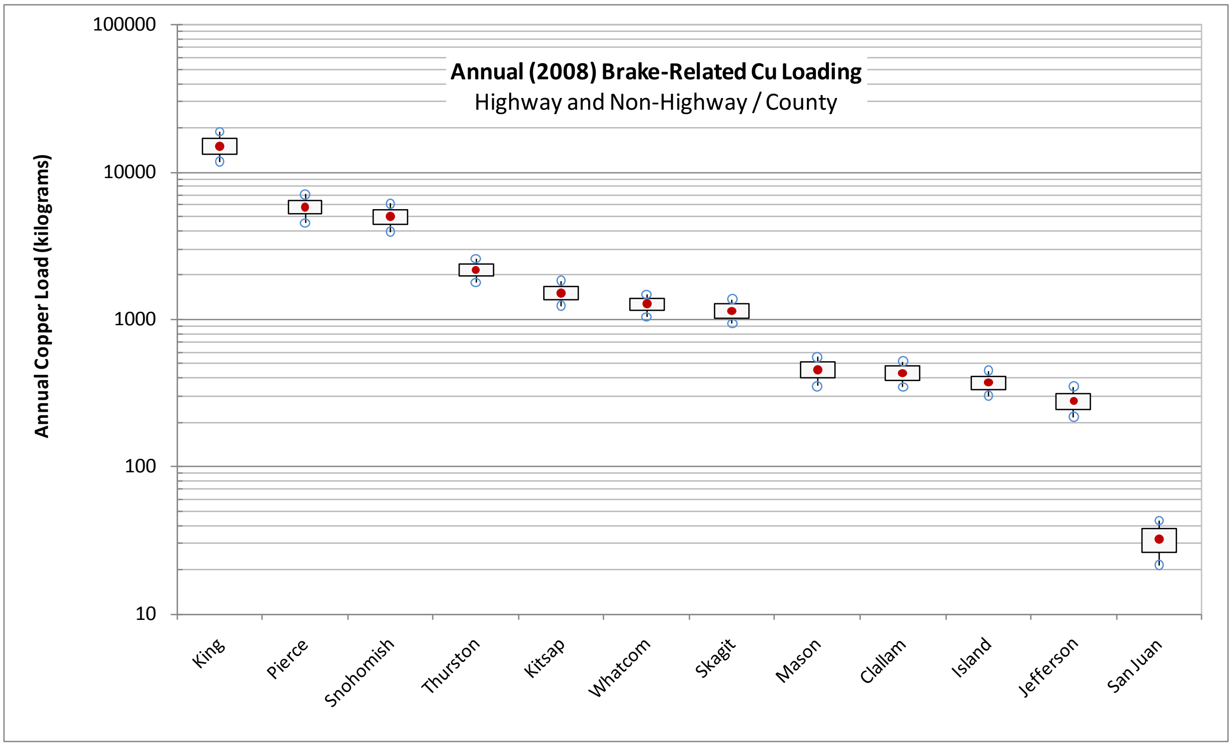

Figure 11. Annual Copper loading associated with brake wear for the highway and non-highway road system by county.

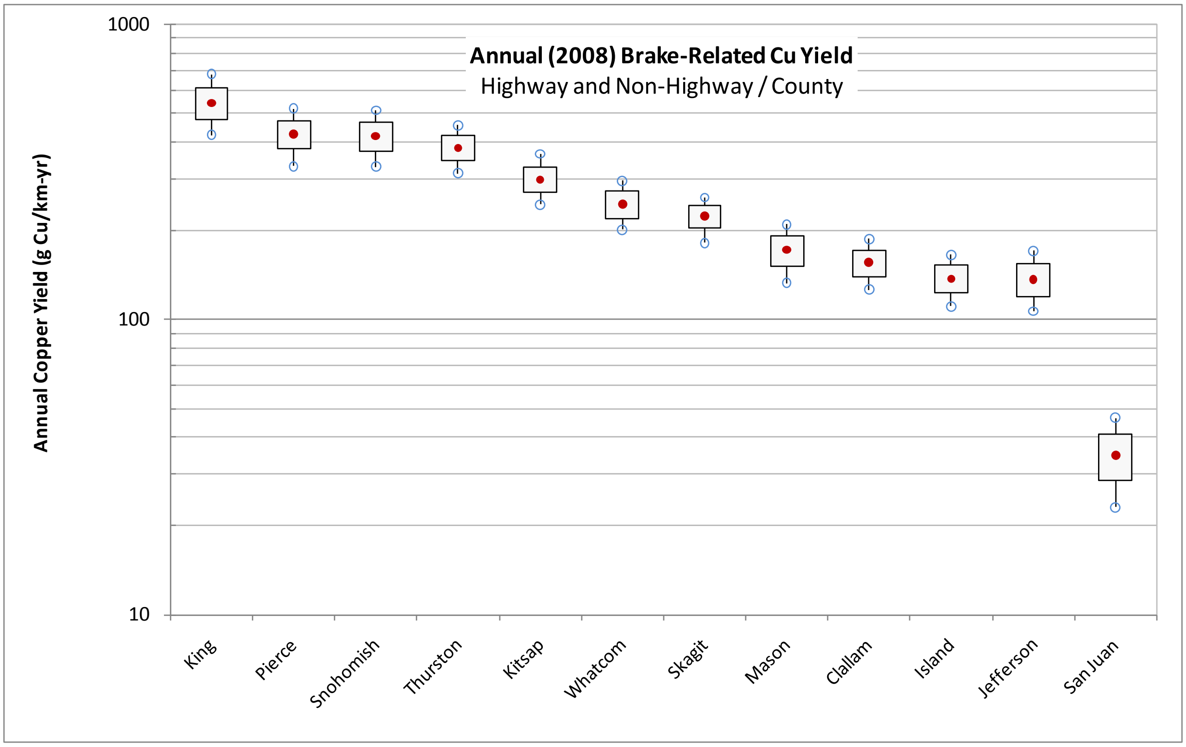

Figure 12. The copper yield (g/km-yr) based on the annual load and total lane kilometers, by county.

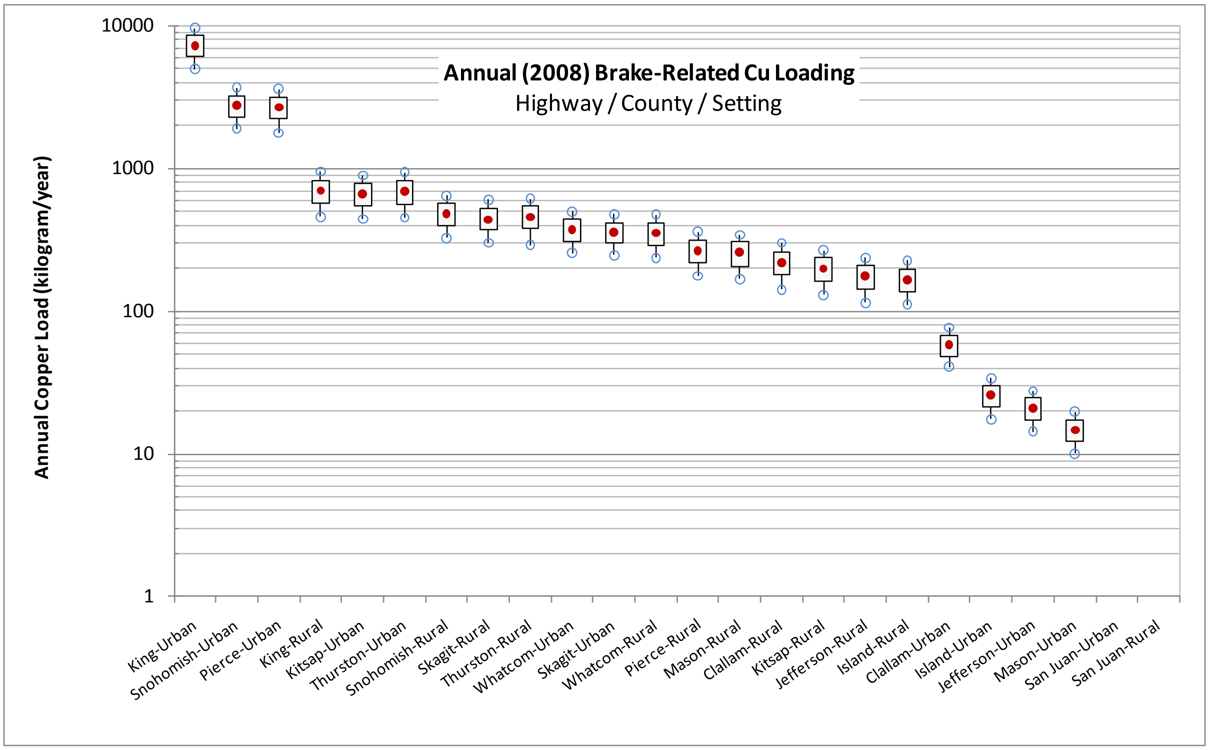

Figure 13. Annual copper loading associated with brake wear for the highway system, by county and setting.

Highway-based copper loading

Focusing just on the urbanized highway setting, the highest level of loading occurs in King County at 7.8 t, representing 49% of the total (Figure 13). In comparison, the estimated copper load for Snohomish and Pierce counties is approximately 3 t each. Together, the copper loading associated with brake pads (drums) within these counties comprises 85% of the annual load estimated for the urban highway designation.

Non-highway-based copper loading

A similar loading pattern is present for the non-highway road system. The majority of the copper loading occurs in King County, 47% of the 17 t, followed by Pierce (19%) and Snohomish (12%), with the majority of the load in each of these counties occurring in the urban setting.

Copper loading by vehicle-type

Passenger cars provide the majority of the copper load representing 65% (24 t) of the total highway-based and non-highway-based annual loads (Table 9, Figure 14) followed by light trucks at 29% (11 t). In both cases, loading occurs primarily in the urban setting representing about 85% of the annual total. The representation of the copper load attributed to the other vehicle types is significantly lower either due to lower copper levels in drum-brakes (combination trucks, buses, single-unit trucks) or a low VKT presence (motorcycle). Copper loading associated with passenger cars within urban King County (highway and non-highway roads) is estimated at 10 t or 27% of the Study Area total.

Table 9. The annual copper loading associated with brake wear (kg) by vehicle type and road system.

Annual copper loading associated with brake wear (kg) by vehicle type and road system.						
Vehicle-Type	**Highway**		**Non-Highway**		**Total (by Vehicle Type)**	**% Representation**
	Urban	**Rural**	**Urban**	**Rural**		
Motor Cycles	10.7	2.4	14.3	4.4	31.8	0.1
Passenger Car (2 Axle, 4 Tire)	10802.2	2461.9	9279.9	1146.3	23690.3	64.5
Light Trucks (2 Axle, 4 Tire)	4121.7	1036.2	4824.0	645.4	10627.2	28.9
Buses	13.8	4.6	11.7	1.5	31.6	0.1
Single-Unit Trucks	310.3	101.0	337.4	65.4	814.1	2.2
Combination Trucks	726.9	418.1	335.4	36.9	1517.2	4.1
Total (by road system and setting)	15985.6	4024.3	14802.5	1899.8	36712.2	=====
% Representation	43.5	11.0	40.3	5.2	=====	=====

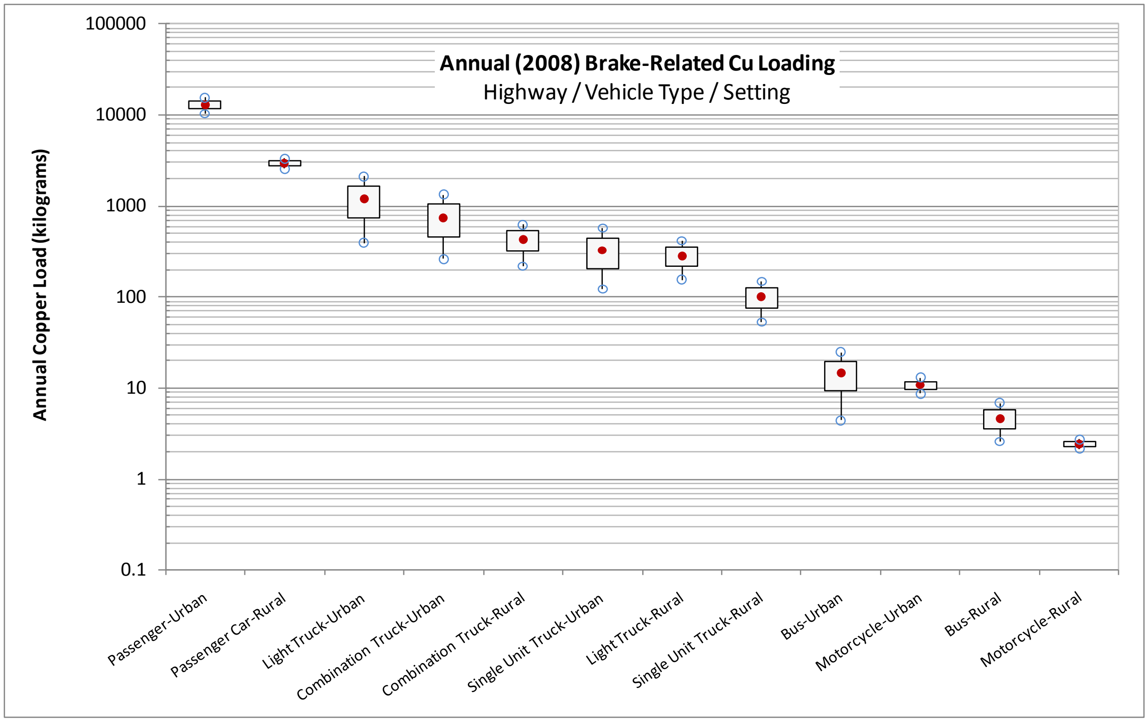

Figure 14. Annual copper loading associated with brake wear for the highway system, by vehicle type and setting.

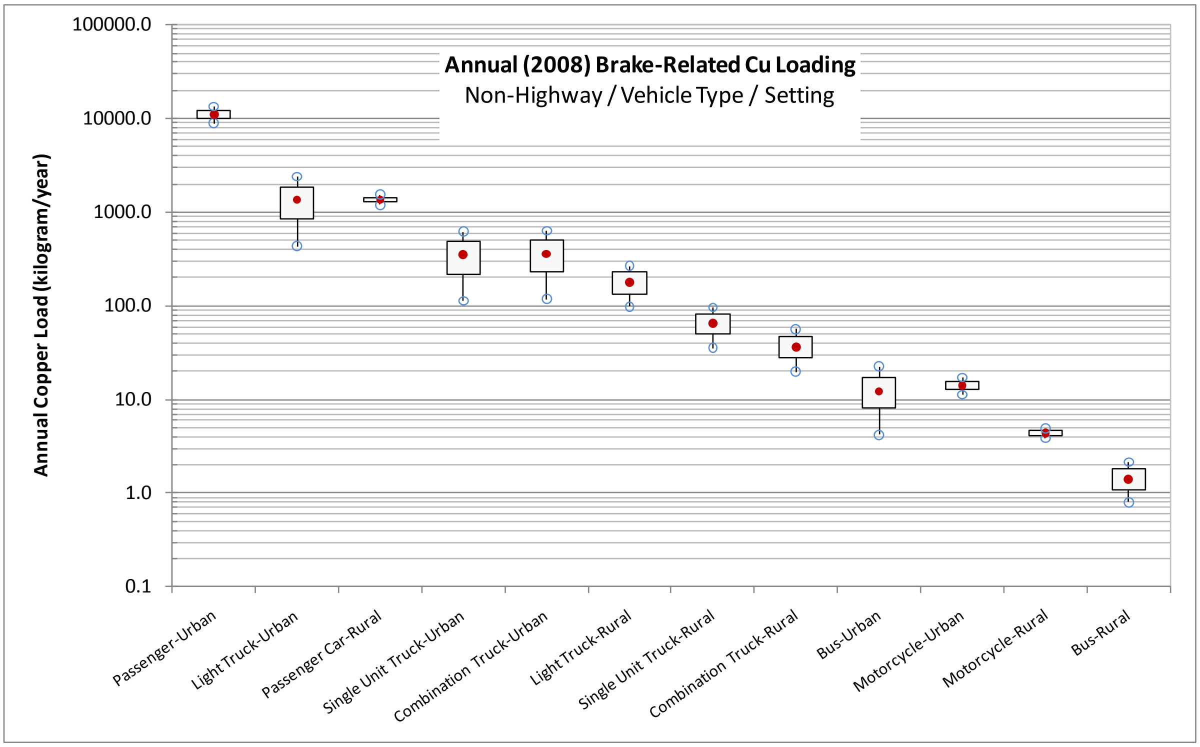

Figure 15. Annual copper loading associated with brake wear for the non-highway road system, by county and setting.

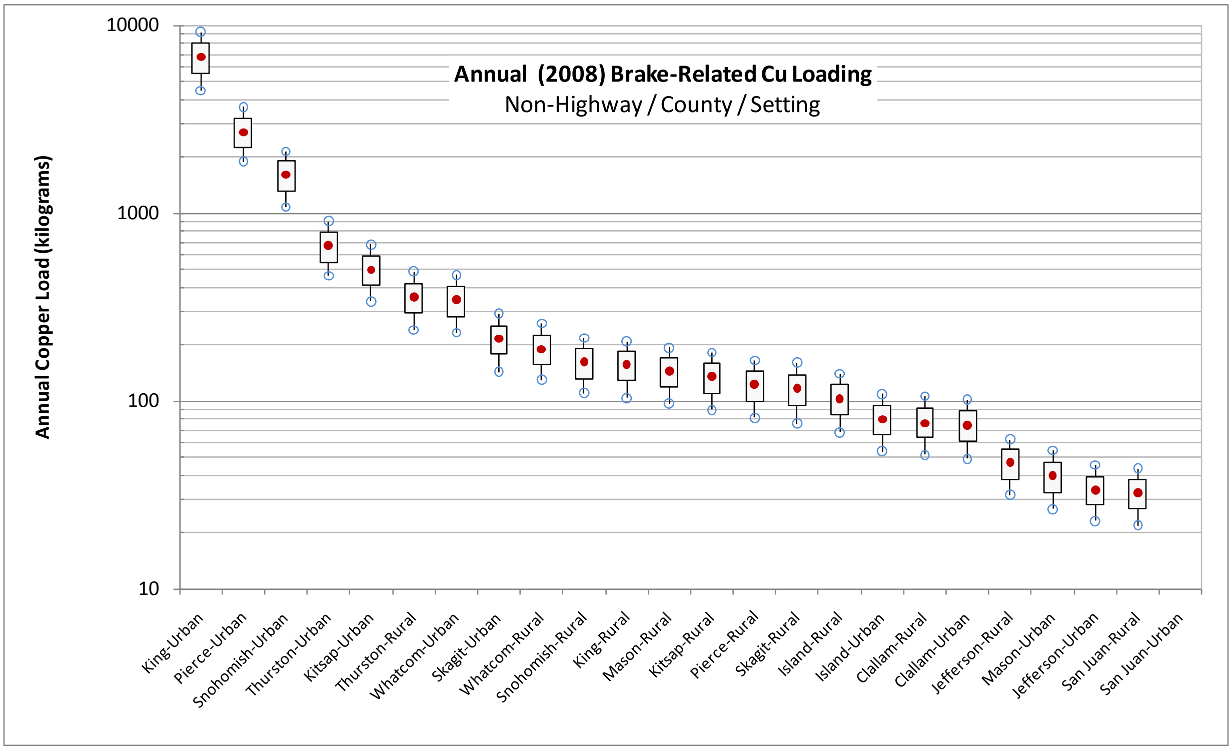

Figure 16. Annual copper loading associated with brake wear for the non-highway road system, by vehicle type and setting.

Conclusions

The conclusions of this analysis are summarized by the vehicle kilometers travelled metric because of its importance in defining loading characteristics, followed by tire-related zinc and brake pad-related copper loading.

Vehicle kilometers travelled

- 2008 vehicle kilometers travelled (VKT) were estimated at 58 billion kilometers for the Puget Sound study area: 55% occurring on highway and 45% on local roads.
- 77% of the annual VKT occur in three counties: King (45%), Pierce (17%), and Snohomish (15%).
- Within these counties, 93% of the VKT occurs in urbanized roadways, representing 71% of all VKT throughout the Study Area.
- Passenger cars are the dominant form of travel, accounting for 63% of all VKT.
- Passenger car travel within King County alone represents 26% of the Study Area total.

Tire-related zinc loading

- The annual (2008) zinc load to road surfaces within the Study Area is estimated at 80 t.
- The majority of the annual load occurs in King County (44% or 35 t), followed by Pierce (17%) and Snohomish (15%).
- The zinc yield (based on lane miles) for the study is 914 g/km-yr though ranged from 1222 g/km-yr for King County to 77 g/km-yr for San Juan County.
- As with the VKT, the majority of the load occurs on urban-based roadways. Urban (highway and local road) travel in King, Pierce, and Snohomish Counties together accounted for 70% (55 t) of the annual study area total. The majority of this load, approximately 41% or 33 t, occurs in urbanized roadways within King County.
- The major source of tire-related zinc loading is passenger vehicles, representing about 49% and 52% of the total annual loading occurring in the highway and non-highway (local) road systems, respectively.
- Passenger vehicles in the urbanized road system (highway and local) of King County contribute 22% to the entire zinc load estimated for the Study Area.

Brake pad-related copper loading

- The estimated annual (2008) copper load to road surfaces within the Study Area is 37 t.
- The majority of the annual load occurs in King County (45% or 16 t), followed by Pierce (17%, 6 t) and Snohomish (15%, 5 t) together accounting for 77% of the total.
- The copper yield (based on lane miles) for the study is 425 g/km-yr, though it ranged from 589 g/km-yr for King County to 40 g/km-yr for San Juan County.

- As with the VKT, the majority of the load occurs on urban-based roadways. Loading occurring on urban-situated highway and local roads in King, Pierce, and Snohomish Counties together accounted for 71% (26 t) of the annual study area total. The majority of this load, approximately 42% or 15 t, occurs in urbanized roadways situated within King County.
- The major sources of brake pad-related copper loading are passenger cars followed by light trucks representing about 65% and 29%, respectively of the estimated total annual copper load.
- Passenger cars within urbanized King County contribute approximately 10 t or 27% of the total estimated for the Study Area.

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Appendix A [of Appendix E]: Additional Discussion, Figures, and Tables

Table A-1 [of Appendix E]. Automotive brake and tire constituent levels (EMEP, 2006).

Material	Tire Composition (mg/kg)			Brake Composition (mg/kg)		
	Average	Min	Max	Average	Min	Max
Aluminum (Al)	324	81	470	2050	330	3770
Arsenic (As)	1	====	====	10	====	====
Barium (Ba)	125	1	370	38520	2640	74400
Bromine (Br)	20	====	====	40	====	====
Calcium (Ca)	892	113	2000	7700	1100	14300
Cadmium (Cd)	3	0	5	13	2.7	29.9
Chlorine (Cl)	250	====	====	1500	====	====
Chloride (Cl-)	600	====	====	1500	====	====
Cobalt (Co)	13	1	25	6	====	====
Chromium (Cr)	12	0	30	669	115	1200
Copper (Cu)	174	2	490	51112	370	142000
Ele. Carbon (EC)	153000	====	====	26100	====	====
Iron (Fe)	1712	2	4600	209667	115000	399000
Potassium (K)	280	180	380	524	190	857
Lithium (Li)	1	0	2	56	====	====
Magnesium (Mg+2)	166	32	360	44570	6140	83000
Manganese (Mn)	51	2	100	2460	1700	3220
Molybdenum (Mo)	3	====	====	10000	====	====
Sodium (Na+)	645	610	680	7740	80	15400
Ammonium (NH4+)	190	====	====	30	====	====
Nickel (Ni)	34	1	50	463	133	850
Nitrate (NO3-)	1500	====	====	1600	====	====
Organic Carbon (OC)	360000	====	====	107000	====	====
Phosphorus (P)	====	====	====	====	====	====
Lead (Pb)	107	1	160	3126	50	6594
Rubidium (Rb)	====	====	====	50	====	====
Sulfur (S)	1100	====	====	12800	====	====
Antimony (Sb)	2	====	====	10000	====	====
Selenium (Se)	20	====	====	20	====	====
Silicon (Si)	1800	====	====	67900	====	====
Sulfate (SO4-)	2500	====	====	33400	====	====
Tin (Sn)	====	====	====	7000	====	====
Strontium (Sr)	14	0	40	520	81.4	740
Titanium (Ti)	378	====	====	3600	====	====
Vanadium (V)	1	====	====	660	====	====
Zinc (Zn)	7434	430	13494	8676	270	21800

(The mg/kg ratio is an expression of parts per million. To express the concentration as a percent, or parts per 100, divide by 10,000. For instance, the assumed average concentration of zinc in tires is 10,000 mg/kg representing about 1% of the composition.)

Check on tire wear rate

As a check on the tire wear rate, assume that the average passenger car tire is described by that equipped on the most popular car in 2008, the Toyota Camry. That tire is described by the following characteristics inscribed on its sidewall: P215/60/R16. The "P" indicates that it is a passenger car tire. The "215" denotes the nominal section width in millimeters. The nominal section width is a measure that is close to the tread width so it can be assumed that the tread width for this tire is approximately 215 millimeters (mm) or 21.5 centimeters (cm). Assuming that the tread occupied 90% of the width then the effective width, the portion of the tire in contact with the roadway, is 19.4 cm. The wheel diameter ("R") is 16 inches or approximately 40.6 centimeters. The "40" in the tire numerical description refers to the aspect ratio, which is the sidewall height, expressed as a percentage of the nominal section width (approximately the tread width). Therefore, the sidewall height is approximately 60% of the nominal section width (21.5 *0.6) or 12.9 cm. The entire wheel diameter is then 66.4 cm with a radius of 33.2 cm, resulting in a circumference of 208.6 cm.

The typical tire has a thread depth of $10/32^{nd}$ of an inch or 0.79 cm, and is considered completely worn when it has a tread depth of $2/32^{nd}$ of an inch or 0.16 cm. Therefore, the tread loss from new to obsolete is 0.63 cm. The entire volume of tire loss is then (19.4 cm * 208.6 cm* 0.63 cm) or 2550 cm^3. The density of tread is approximately 1180 mg/cm^3. When the density is multiplied by the volume of rubber loss, the result is 3,009,000 mg (3.0 kg) of material loss over the life of a passenger car tire. Assuming that the average tire lasts 50,000 miles (approximately 80,000 kilometers) then the wear rate is determined by dividing the weight of the material lost (3,009,000 mg) by the kilometers travelled (80,000 km), resulting in 38 milligrams of tire loss per kilometer travelled (kg/km).

Appendix F. Domestic Water Consumption Rates

Table F-1. Domestic Water Consumption Rates
(sources: Mayer et al., 1999; Ecology, 2007; U.S. Census Bureau, 2010)

Daily Mean Consumption Rates for U.S.	**gal/person/day**	**liter/person/day**
Toilet water use	18.5	70
Clothes washer water use	15.0	57
Shower water use	11.6	44
Faucet water use	10.9	41
Baths water use	1.2	5
Dishwasher water use	1.0	4
Other water use	1.6	6
Leakage water use	9.5	36
Total indoor domestic water consumption	69.3	262
Population Statistics for Study Area		
Total population	4,475,300	
Persons per household	2.53	
Total households	1,768,893	
Households using on-site septic systems	500,000	
Households discharging to WWTPs	1,268,893	
Population discharging to WWTPs	3,210,300	
Annual Consumption Totals for Study Area	**gal/yr**	**liter/yr**
Toilet water use	2.17E+10	8.21E+10
Clothes washer water use	1.76E+10	6.65E+10
Shower water use	1.36E+10	5.15E+10
Faucet water use	1.28E+10	4.83E+10
Baths water use	1.41E+09	5.32E+09
Dishwasher water use	1.17E+09	4.44E+09
Other water use	1.87E+09	7.10E+09
Leakage water use	1.11E+10	4.21E+10
Total indoor domestic water consumption	8.12E+10	3.07E+11

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Appendix G. Methodology Used to Estimate Copper Releases from Vessel Anti-Fouling Paint

Copper Leaching Rates

Table G-1. Rates of Copper Leaching from Anti-Fouling of Recreational Vessels.

Test Conditions	Rate (ug Cu/cm^2/day)	Reference
static *in-situ* panels in harbor, 5 paints -- after 120 days	≤ 3	Valkirs et al., 2003
Navy vessels, ablative coatings, mean, *in-situ*	3.8	“
pleasure craft, modified epoxy, mean *in-situ*	8.2	“
hard vinyl paint, *in-situ*, fiberglass panels	3.7	Schiff et al., 2003
modified epoxy paint, *in-situ*, fiberglass panels	4.3	“
combination from literature	6.5	Dobalian and Arias, 2005

Vessel Wetted Surface Area Calculations

Calculation of copper releases from anti-fouling bottom paint requires estimation of total vessel wetted surface areas (WSAs) for the Study Area. The following variables are required to estimate Study Area WSAs:

- Number of vessels in the Study Area
- Types of vessels (sail, power, commercial, naval)
- Lengths and preferably other dimensions (beam, displacement) for each vessel type.
- Formulas to convert vessel dimensions to WSA for each vessel type

Calculations of WSAs were done independently for three vessel classifications – recreational, commercial, and naval vessels – due to differences in the sources of the vessel registration and size data, as well as the different size distributions among the three categories. For instance one of the formulas used to linearly extrapolate WSA estimates for recreational vessels was deemed inappropriate for vessels over 80’ or so, a size cutoff which encompasses nearly all recreational vessels but excludes a large portion of the commercial fleet.

Methods and assumptions used to calculate WSAs for each vessel category are described below:

Recreational Vessel WSA Calculations

Estimate Of Recreational Vessel Numbers

The number of vessels moored in the Study Area was estimated under three scenarios based on Department of Licensing (DOL) 2008 registration data (BST, 2010a) and 2001 marina inventory data (BST, 2001). Estimates ranged from 27,727 to 46,232 vessels. There were no data to distinguish saltwater and freshwater moorages, although most marinas slips are located in the Puget Sound and Strait of Juan de Fuca with few freshwater moorage facilities outside the Lake

Washington/Lake Union system and the Snohomish River. Estimates also assume that all boats registered in Clallam and Jefferson Counties are used are within the Study Area and that all Clallam County marina slip use is within the Study Area.

The 2008 DOL registration data are summarized in the table below.

Table G-2. DOL Vessel Registrations for the 12-County Puget Sound Area.

Size Class	16' to 20'	21' to 30'	31' to 40'	41' to 50'	51' to 60'	Over 60'	Total
No. in Class	67,019	32,534	9,417	3,278	639	364	175,466
Pct. in Class	38.2%	18.5%	5.4%	1.9%	0.4%	0.2%	100.0%

SCENARIO A – DOL Registered Vessels ≥ 21'

In this scenario, all boats ≥ 21' were considered to be moored in marine waters of the Study Area, resulting in 45,232 vessels. Boats < 21' were excluded because they were considered to be trailerable and not likely to contain anti-fouling paint since they were less likely to spend extended durations in the water

SCENARIO B – DOL Registered Vessels ≥ 30' plus one-half of DOL Registered Vessels 21'-30'

In this scenario, all boats ≥ 30' plus one half of the DOL registered vessels 21'-30' were considered to be moored in marine waters of the Study Area, resulting in 29,965 vessels. Boats < 21' and one half of boats 21'-30' were excluded because they were considered to be trailerable and not likely to contain anti-fouling paint.

SCENARIO C- Marina Slip Use

This scenario uses a 2001 recreational marina use survey conducted by BST (BST, 2001). The number of boats in permanently occupied marina slips plus boat houses was 27,737. All boats were assumed to contain anti-fouling paint since they were permanently moored.

Formulas Used To Calculate WSA For Recreational Vessels

METHOD 1 – Interlux Method

For this method, calculation of the WSA was based on information provided by Interlux (2006), a major paint manufacturer:

$$WSA = 0.85(L)(B)$$

Where: WSA = Wetted surface area (ft^2)
L= Overall length (ft)
B = Beam (ft)

METHOD 2 – Fitted Line Method

This method used interpolated and extrapolated values from a line fitted from known length and WSA values for a variety of sail and power boats (Epoxyproducts.com, 2009). For sailboats, the formula is:

$$WSA = 9.8692(L)$$

For power boats, the formula is:

$$WSA = 10.623(L)$$

Where: WSA = Wetted surface area (ft^2)
L= Overall length (ft)

METHOD 3 – Grovhoug Method

A third method developed by Grovhoug et al. (as cited in Johnson et al., 1998) to estimate loading from pleasure craft in the vicinity of naval harbors used the formula:

$$WSA = 0.2021(L^2) - 0.2197(L) + 3.5571$$

Where: WSA = Wetted surface area (ft^2)
L= Overall length (ft)

While this method supposedly provides more accurate results because it is empirically based, it appears to produce surface area results approximately 30% lower than other methods, possibly because it was derived exclusively from sailboat data (Johnson et al., 1998).

WSA Estimates For Recreational Vessels Based On Scenarios A,B,C And Methods 1,2,3

Table G-3 shows a matrix of resulting WSA values for all possible combinations of scenarios and methods as described previously. All results fell within a factor of two, with the highest WSA from SCENARIO A x the “fitted line Method” (1.32×10^{10} cm^2) and the lowest value produced from SCENARIO C x the “Grovhoug Method” (6.44×10^9 cm^2).

Table G-3. Matrix of Scenarios and Input Parameters Used to Calculate Wetted Surface Areas of Recreational Vessels.

	SCENARIO A –DOL Registered Vessels ≥ 21’	SCENARIO B – DOL Registered Vessels ≥ 30’ plus one-half of DOL Registered Vessels 21’-30’	SCENARIO C- Marina Slip Use
	Number of Vessels	Number of Vessels	Number of Vessels
	46,232	29,965	27,737
	Wetted Surface Area (cm^2)*		
Interlux Method 1– WSA = 0.85(L)(B)	1.20×10^{10} (a,b,c)	9.17×10^{9} (a,b,c)	1.17×10^{10} (b,d)
Fitted Line Method 2– WSA = 9.869(L) (sailboats) WSA = 10.623(L) (power boats)	1.32×10^{10} (a,c,e)	9.23×10^{9} (a,c,e)	1.06×10^{10} (d,e)
Grovhoug Method 3– WSA = $0.20(L^2) - 0.22(L) + 3.56$	8.40×10^{9} (a,c)	6.44×10^{9} (a,c)	8.20×10^{9} (d)

DOL=Department of Licensing
WSA=wetted surface area
L= length overall
B=Beam
(a) Assumes L=100’ for all boats >60’
(b) Assumes B=L/3 when beam is not provided
(c) Assumes boat length is mid-point for each size class (e.g. 31’-40’ boats are 35.5’)
(d) Assumes L=40’ for all permanently moored boats
(e) Assumes 50% sailboats and 50% power boats
* $1\ \text{cm}^2 = 0.001077\ \text{ft}^2$

Commercial Vessel WSA Calculations

Estimate Of Commercial Vessel Numbers

The number of commercial vessels registered by DOR in the Study Areas during 2004 and 2005 was provided by BST (2010b). The mean number of commercial vessels registered in the 12-county Puget Sound area for 2004 and 2005 was 2,003 vessels. Unlike DOL information which grouped data into size classes for recreational vessels, the DOR data provide length data for each commercial vessel registration, except in the few cases where data were missing. There were no data to distinguish saltwater and freshwater usage, although it seems likely that most usage is in marine waters.

It is notable that while most commercial vessels are likely to be used in marine waters, a substantial portion may be used outside of the Study Area for certain periods of the year, although there are no data to estimate the use within or outside of the Study Area. Boats not registered within the Study Area also enter the Study Area, but again no data to form estimates of this was found. Since leaching rates of copper in bottom paint are normally expressed in units of loss per day, the final release load calculations are the appropriate step to apply these in/out of

Study Area fractions. However, the present calculations are simply to calculate WSAs, and therefore the fraction of time spent in/out of the Study Area does not apply here.

SCENARIO A – DOR Registered Vessels ≥ 21'

In this scenario, all boats ≥ 21' were considered to be moored in marine waters of the Study Area, resulting in 2,003 vessels. Boats < 21' were excluded because they were considered to be trailerable and not likely to contain anti-fouling paint. It was recognized that this probably excludes some commercial vessels, such as log broncs, that remain moored year-round, but this number is thought to be comparatively small.

SCENARIO B – DOR Registered Vessels ≥ 30' plus one-half of DOR Registered Vessels 21'-30'

In this scenario, as in the DOL Scenario B, all boats ≥ 30' plus one half of the DOR registered vessels 21'-30' were considered to be moored in marine waters of the Study Area, resulting in 1,878 vessels. Boats < 21' and one half of boats 21'-30' were excluded because they were considered to be trailerable and not likely to contain anti-fouling paint. It was recognized that this probably excludes some commercial vessels, such as log broncs, that remain moored year-round, but this number is likely to be comparatively small.

Formula Used To Calculate WSA For Commercial Vessels

GROUVHOG METHOD 1

The method developed by Grovhoug et al. (as cited in Johnson et al., 1998) to estimate loading from pleasure craft in the vicinity of naval harbors was used to estimate WSAs for the Study Area commercial fleet:

$$WSA = 0.2021(L^2) - 0.2197(L) + 3.5571$$

Where: WSA = Wetted surface area (ft^2)
L= Overall length (ft)

While this method supposedly provides more accurate results because it is empirically based, it appears to produce surface area results approximately 30% lower than other methods, possibly because it was derived exclusively from sailboat data (Johnson et al., 1998). However, the "Grovhoug method" has several advantages over the "Interlux method" and "fitted line method" used for recreational vessels. These latter methods are based on linear relationships which appear to hold up well for craft <80', but much of the commercial fleet is much longer, with some vessels exceeding 600'. In addition, the "Grovhoug method" does not require beam dimensions, which appear to be missing from much of the DOR registration records.

WSA Estimates For Commercial Vessels Based On Scenarios A,B And Method 1

Table G-4 shows a matrix of resulting WSA values for the two boat number scenarios described previously. Results were similar (3.29 x 10^9 cm^2 and 3.27 x 10^9 cm^2) since elimination of trailerable-sized boats has little impact on the total WSA for the commercial fleet.

Table G-4. Matrix of Scenarios and Input Parameters Used to Calculate Wetted Surface Areas of Commercial Vessels.

	SCENARIO A – DOR Registered Vessels ≥ 21’	SCENARIO B – DOR Registered Vessels ≥ 30’ plus one-half of DOR Registered Vessels 21’-30’
	Number of Vessels	Number of Vessels
	2,003	1,878
	Wetted Surface Area (cm^2)*	
Grovhoug Method– WSA = 0.20(L^2) - 0.22(L) + 3.56	3.29 x 10^9 (a)	3.27 x 10^9 (a)

DOR=Department of Revenue

WSA=wetted surface area

L= length overall

(a) Assumes L=68.4’ where length data are missing (68.4’ is mean length of commercial vessels registered in the 12-county Puget Sound region during 2004 and 2005)

* 1 cm^2 = 0.001077 ft^2

Naval Vessel WSA Calculations

Estimate Of Naval Vessel Numbers

WSAs of naval vessels were estimated for three Navy facilities in Puget Sound: The Puget Sound Naval Shipyard (PSNS) at Bremerton, the Naval Station Everett (NAVSTA Everett), and the Trident Submarine Base at Bangor (Bangor). The number of vessels at each location was obtained from various sources. The number of vessels at PSNS was reported by Johnson and Grovhoug (1999) to be 48 active and 18 inactive. However, Brandenberger et al. (2008) reported a total of only 39 vessels at PSNS. Specific ships homeported in the NAVSTA Everett and Bangor fleets were found on Wikipedia 2010a; http://en.wikipedia.org/wiki/Naval_Station_Everett and 2010b; http://en.wikipedia.org/wiki/Naval_Base_Kitsap#Bangor, respectively), and verified with the Naval Vessel Registry (2010, www.nvr.navy.mil/). Specifications on each vessel (NAVSTA Everett-one aircraft carrier, two destroyers, and three frigates; Bangor-eleven submarines) were found in the Naval Vessel Registry. A summary of the naval vessel numbers is shown in the Table G-5 below.

Table G-5. Number of Naval Vessels in the Study Area.

Location	Number of Vessels	Type
Bremerton PSNS	48 active 18 inactive	Various
Bremerton PSNS	39 total	Various
Everett Naval Station	6 active	1 Aircraft Carrier 2 Destroyers 3 Frigates
Bangor Sub Base	11 (activity unknown)	11 Submarines

It is notable that while naval vessels are likely to be used exclusively in marine waters, a substantial portion may be used outside of the Study Area for certain periods of the year, although there are no data to estimate the use within or outside the Study Area. Since leaching rates of copper in bottom paint is normally expressed in units of loss per day, the final release load calculations are the appropriate step to apply these in/out of Study Area fractions. However, the present calculations are simply to calculate WSAs, and therefore the fraction of time spent in/out of the Study Area does not apply here.

Formula Used To Calculate WSA For Naval Vessels

METHOD 1 – WSA Reported for PSNS

For vessels at the PSNS, Johnson and Grovhoug (1999) and Brandenberger et al. (2008) reported estimates of total annual copper releases (t/yr) from ship hulls and copper release rates expressed as ug/cm^2/day. Dividing the annual load by release rates with unit conversions yields a total WSA for the fleet at PSNS, although WSAs for individual vessels are not known.

METHOD 2 - NAVSEA

The WSA was calculated using the formula developed by the Naval Sea Systems Command (as cited in Johnson et al., 1998):

$$WSA = 1.7(L)(d) + (V/d)$$

Where: WSA = Wetted surface area (ft^2)
L= Overall length (ft)
d= molded mean draft at full displacement
V= molded volume of displacement

WSA Estimates For Naval Vessels Based On Vessel Numbers And Methods 1,2

Table G-6 shows a matrix of resulting WSA values reported for PSNS and for vessels at Everett and Bangor using the NAVSEA method. Total WSA for PSNS + Everett + Bangor is 1.26×10^9 cm^2 when the Johnson and Grovhoug (1999) WSA is used for PSNS, and 1.40×10^9 cm^2 when the Brandenberger et al. (2008) value is applied.

Table G-6. Matrix of Scenarios and Input Parameters Used to Calculate Wetted Surface Areas of Naval Vessels.

		Number of Vessels		Number of Vessels		Number of Vessels		Number of Vessels
	Bremerton PSNS	48 active 18 inactive (a)	Bremerton PSNS	39 (b)	Everett Naval Station	6	Bangor Sub Base	11
	Wetted Surface Area (cm^2)*							
WSA Reported for PSNS – METHOD 1		4.76×10^8 (a)		6.16×10^8 (b)		na		na
NAVSEA Method 2- WSA = 1.7(L)(d) + (V/d)		na		na		2.73×10^8		5.06×10^8

WSA=wetted surface area

(a) Johnson and Grovhoug (1999)

(b) Brandenberger et al. (2008)

L= length overall

WSA=wetted surface area

V= molded volume of displacement

d= molded mean draft at full displacement

* 1 cm^2 = 0.001077 ft^2

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Appendix H. Products Containing Mercury

Mercury Products

IMERC reports the amount of mercury used in products in the U.S. (Wienert, 2009) (summarized in Table H-1). Scaling by population to the Study Area results in 1.7 tons of mercury products in 2001. The total amount was reduced to 0.91 tons by 2007.

Table H-1. Mercury Sold in Study Area Products (t/yr)

Product	2001	2004	2007
Switches & Relays*	0.76	0.68	0.41
Dental Amalgam	0.41	0.40	0.22
Thermostats*	0.19	0.19	0.051
Lamps	0.13	0.13	0.14
Miscellaneous	0.068	0.032	0.037
Batteries	0.039	0.034	0.027
Chemicals and Solutions	0.014	0.012	0.019
Sphygmomanometers*	0.028	0.015	0.011
Thermometers*	0.022	0.018	0.0040
Manometers*	0.013	0.017	0.00
Barometer	0.0024	0.0016	0.00
Totals	1.7	1.5	0.91

*Sales in Washington State banned/restricted under MERA

Reductions

Since the publication of the Mercury CAP in 2003 Ecology has tracked the amount of mercury reduced/collected from various products and facilities, presented in Table H-2. Scaled to the Study Area, Ecology estimates that 5.4 t of mercury have been collected, with 1.2 kg collected in 2009 alone.

Table H-2. Mercury Collected in Study Area Products (kg), 2003 - 2009

Product/Source	2009	TOTAL
Thermometers (numbers include some thermostats)	0.0060	0.93
Manometers, Barometers	0.27	1.1
Schools K - 12	0.0000	0.79
Hospitals	0.12	0.82
Medical Waste ("Red Bags")	0.030	0.082
Mining	0.091	0.14
Bulk Mercury	0.43	--
Thermostats	0.012	0.050
Auto Convenience Switches	0.022	0.053
Utility Switches & Relays	0.0000	0.034
Assorted Switches	0.0004	0.0004
Button Cell Batteries	0.024	0.041
Fluorescent Lamps	0.016	0.12
Dental Amalgam Waste	0.23	0.78
Total	**1.2**	**5.4**

Mercury Lamps

Scaling from a national estimate and assuming a 20% recycling rate, the Mercury CAP (Ecology and WDOH, 2003) identified fluorescent tube lamps as the largest source of mercury to landfills in Washington, contributing about 198 - 229 kg annually. Scaled to the Study Area population, this totals 133 - 154 kg. This Mercury CAP assumes mercury content of 20 mg/lamp (Ecology and WDOH, 2003).

The amount of mercury per lamp has decreased over the last decade. Electro-Canada reported that a 1990s baseline mean of 43 mg/lamp had been reduced to an 11.4 mg/lamp mean by 2003 (PSI, 2008). The EPA estimated the mean content to be 48.2 mg for a four-foot tube in 1989 and 11.6 mg in 1999 (Leopold, 2002). Further reductions since these reports are likely. In 2001 Canada required the mercury content of lamps to be 70% of the 1990 levels by 2005, and 80% by 2010. In the U.S. such efforts are limited to individual states and voluntary efforts, similar regulatory requirements do not exist on a federal level.

Lamp manufacturers report the amount of mercury used in lamps sold in the U.S. to IMERC triennially. While the amount of mercury per lamp has been decreasing, total mercury use has not, indicating a growth in sales. This seems consistent with the increased promotion of energy reduction measures that rely on mercury lighting as an alternative to incandescent.

Using the total mercury amounts (Wienert, 2009), and applying an estimate of the amount of mercury per lamp, 11.5 mg (the mid-point of Electro-Canada and EPA's estimates), the number of lamps sold in the Study Area for 2001, 2004, and 2007 can be estimated from the IMERC mercury totals, Table H-3.

Table H-3. Calculation of the Number of Bulb Units in the Study Area

		2001	2004	2007
Mercury in Bulbs (t)	National	10	9.6	11
	State	0.22	0.21	0.23
	Study Area	0.15	0.14	0.15
		Number of Bulb Units in Study Area		
mg mercury/bulb	11.5	12,000,000	11,000,000	12,000,000

1. Wienert, 2009.
2. PSI, 2008 and Leopold, 2002.

Based on the 2003 Mercury CAP estimate there would be about 9 million tube lamps in the Study Area. Cascadia (2007) reported sales of 11- 13 million tube lamps statewide in 2004, which would place the total number in the Study Area at 7.4 to 8.7 million. Based on the ratio of mercury used in various bulb types reported by IMERC for 2001 and 2004 (Table H-4), fluorescent tubes contain 75% of the mercury used in lighting, meaning that the tube lamp estimates given in the Mercury CAP (Ecology and WDOH, 2003) and the Cascadia (2007) report represent only 75% of the mercury lamps. Thus, an estimate of 11,000,000 – 12,000,000 mercury tube lamps in the Study Area, produced by applying the value of 11.5 mg per lamp to the total mercury used in lamps reported by Wienert (2009) seems in line with the other lamp estimates. (Note that this estimate combines all mercury lamps into the category of tube lamp, though they are only 75% of the total market.)

Table H-4. Mercury in Lamps Sold in the U.S. by Type (kg/yr)

Lamp Type	**2001 Total Mercury (All Companies)**	**2004 Total Mercury (All Companies)**
Fluorescent	7,600	6,500
Compact Fluorescent Lamps (CFLs)	400	670
HID*		0
- Metal Halide	970	1,100
- Ceramic Metal Halide	N/A	14
- High Pressure Sodium	180	200
- Mercury Vapor	92	97
Total HID Lamps	1,200	1,400
Mercury Short-Arc	5	8
Neon	500	480
Miscellaneous**	19	11
TOTAL	9,700	9,100

* The 2001 data does not break out HID lamps by specific types; a few manufacturers provided this information.

** This category includes some HID lamps. It was not possible to separate them from the other lamps in the category.

N/A = not applicable

Source: IMERC Fact Sheet

Cascadia (2007) provides an estimate of the number of bulbs recycled in 2004. Estimates of the number of bulbs collected for 2005 – 2008 can be made by taking the total tons of material recycled (Newman, 2010) and applying a weight of 0.6125 pounds per tube lamp (Salvi, 2010), then scaling to the Study Area by population, Table H-5. (Again, all lamps recycled are being combined into the category of tube lamp. As recyclers report a gross weight of collections, the numbers of tube lamps recycled versus CFLs in unknown.)

Table H-5. Number of Mercury Bulbs Recycled

Year Recycled		2004	2005	2006	2007	2008
Recycled Materials (tons):			730	1,100	980	1,600
Number of Tubes (0.6125 lb/tube)	State	2,500,000[1]	2,400.000	3,400,000	3,200,000	5,200,000
	Study Area	1,700,000	1,600,000	2,300,000	2,100,000	3,500,000

1. Cascadia, 2007

Comparing the number of bulbs recycled to the number estimated to be made using the 11.5 mg mercury/lamp factor (PSI, 2008 and Leopold, 2002), and applying a 4 year life expectancy (Cascadia 2007), indicates that recycling rates have increased, going from 14% in 2005 to 32% in 2008, Table H-6. Other estimates of the state recycling rate include 17 – 23 % (Cascadia, 2007) and 37% for King County specifically (Cascadia, 2007, citing King County).

Table H-6. Mercury Bulb Recycling Rate

	Bulbs	% Recycled
Sold, 2001	12,000,000	14%
Recycled, 2005	1,600,000	
Sold, 2004	11,000,000	32%
Recycled, 2008	3,500,000	

References for Appendix H

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Appendix I. Products Containing PCBs.

Table I-1. Known Maximum Concentrations of Non-Liquid Open-System Products Containing PCBs.

Material	PCB content (mg/kg)
Adhesive tape	1,400
Anti-fouling compounds	No data available
Caulking[1]	310,000
Ceiling tiles [2]	53
Cloth/paper insulating material	12,000
Coal-tar enamel coatings	1,264
Dried paint [3]	63,300
Dried paint [4]	97,000
Fiberglass insulation	39,158
Fire retardant coatings	No data available
Flooring and floor wax/sealant [5]	No data available
Fluorescent light ballast potting	No data available
Foam rubber insulation	13,100
Foam rubber parts	1,092
Grout	9,100
Insulating materials in electric cable	280,000
Plastics/plasticizers	13,000
Processed cork ventilation system gasket material	6,400
Roofing/siding material	22,000
Rubber parts	84,000
Sound-dampening material	No data available
Thermal insulation	73,000
Waterproofing compounds	No data available
Wool felt gaskets	688,498

Source: EPA, 1999 (Unless otherwise noted).

1 Kohler et al., 2005 (citing Sundahl et al., 1999) reported concentrations up to 583,000 mg/kg.

2 Weis et al. (2003) reports ceiling panels with 110,000 mg/kg PCB flame retardant coating.

3 Non-degraded gray chlorinated rubber-based paint, Federal specification TT-P-912; PCBs added presumably to prevent brittleness.

4 Semi-gloss paint; white and light blue, Amercoat 33HB with red Amercoat 86 primer.

5 Rudel et al., 2008 found elevated concentrations in indoor air and residents' blood in residence containing PCB wood floor varnish.

Table I-2. Reported Mean PCB Concentrations in Sealants

Study	Sample Population	PCB Conc. (mg/kg)
Robson et al., 2010[a]	13/95 buildings in Toronto, Canada	4,600
Sundahl et al., 1999	1 concrete building in Sweden (8-story)	60,000
Astebro et al., 2000	1 apartment in Sweden (7 story)	130,000
Melymuk et al., 2008	6/20 in Toronto, Canada	22,100
Herrick et al., 2004[b]	13/24 buildings in Boston	9,637
Lefkowitz, 2005	57 detects from Northeastern U.S.	51,543
Chang et al., 2002	9 samples from NE University	6,073
Pyy and Lyly, 1998[c]	Pre-Fab Apts, Finland	24,500
Mean		38,557

[a] Geometric mean

[b] Targeted sample population

[c] Mean of the two means given (24,000 and 25,000)

Table I-3. Derivation of Inventory for PCBs in Sealants in the Study Area

Sealant Quantity for Study Area (kg)[a]	Percent of Sealants with Detectable PCB Conc.[b]	PCB Concentration Bin (mg/kg)[b]	Bin Percent of Total[b]	PCB Quantity for Bin – Mid Point (kg)	PCB Quantity for Bin – Low Estimate (kg)	PCB Quantity for Bin – High Estimate (kg)
3,654,788	47.9%	20 - <50	12.10%	7	4	11
		50- < 100	7.72%	10	7	14
		100- <1,000	18.99%	180	33	330
		1,000 - <10,000	18.15%	1,750	320	3,200
		10,000 - <100,000	23.16%	22,000	4,100	41,000
		>100,000	20.03%	35,000	35,000	35,000
		Sum Total	**100%**	**59,000**	**39,000**	**79,000**

[a] Study Area target building volume of 6.64 x 10^7 m^3(See Appendix J for supporting information) * 55 g caulk/m^3 of building (as presented in Robson and Melymuk 2010).

[b] Estimates of PCB concentrations in Study Area sealants were calculated based on the distribution of PCB concentrations reported by Kohler et al. (2005). The distribution of these concentrations appears to be consistent with other published data and they have the added benefit of being grouped by concentration bin. The total PCB inventory was calculated by assuming that 48% of the Study Area caulk material had measurable PCBs (per Kohler et al., 2005), multiplying by the percent and mid-point concentration represented by each "bin", then summing the total quantities. High and low estimates were calculated by using the respective high and low concentration values from each range in the calculations. Based on these calculations, there are approximately 39,500 – 79,200 kg of PCBs in Study Area sealants, with a mid-point estimate of 59,300 kg.

References for Appendix I

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Appendix J. Methodology Used to Calculate the Volume of PCB-Containing Sealants in the Study Area

Estimates of PCB-containing sealants in the Study Area were limited to masonry buildings constructed between 1945 and 1980. While other structures also contain PCB-caulking, not enough information was found to calculate an estimate of the amount of PCBs in such materials.

The total volume of commercial masonry buildings built between 1945 and 1980 was calculated using assessor's data from Pierce and Snohomish Counties; together, these counties account for approximately one-third of the Study Area population. County assessors' parcel databases were queried for relevant information on target buildings. Using building data, a per capita estimate of sealant volume for each county was calculated. Building volume estimates were then scaled up to the entire Study Area in proportion to population.

Pierce County Dataset

For Pierce County, there were 13,067 buildings classified as commercial, industrial, or multiple unit built between 1945-1980. Removing blank and zero values from the number of stories and story height fields from this subset of buildings resulted in 12,729 buildings with a mean of 1.3 stories per building and a mean story height of 10.3 feet. Because Pierce County does not provide construction types for commercial buildings, a masonry fraction calculated from Snohomish County of 0.49 was applied to the 78,526,707 total square feet for the buildings of interest in Pierce County to obtain an estimate of 38,331,384 square feet of masonry buildings of interest in Pierce County.

Snohomish County Dataset

For Snohomish County, improvement types classified as commercial built from 1945-1980 were included in the dataset. This classification includes commercial and industrial buildings as well as apartment complexes. Removing any buildings with a blank construction type classification resulted in 4,962 buildings with a total of 46,947,491 square feet. Further reducing this subset to represent masonry buildings, including only those with a predominant construction type of either fire resistant or reinforced concrete, resulted in 1740 buildings with a total of 22,916,564 square feet, and a mean of 1.1 stories per building. Comparing the total square feet to the number of masonry square feet resulted in a masonry fraction of 0.49. Because Snohomish County does not report story heights, the Pierce County mean story height of 10.3 feet was applied to Snohomish County.

Masonry Building Volume

Building volume was calculated using Total Volume = sq ft masonry x mean stories/building x mean story height. Comparing the values obtained from the 2 counties to their populations resulted in a total masonry building volume of 15 cubic meters per capita. Applying this value to the Puget Sound population of 4,475,300 resulted in a total volume of commercial masonry buildings built between 1945 and 1980 in the Puget Sound study area of 66,450,000 cubic meters (Table J-1).

Table J-1. Estimated Volume of Commercial Masonry Buildings in the Study Area Constructed 1945-1980.

	Total Comm/Inst. Bldg Area (ft^2)	**Masonry Fraction**	**Total Masonry Area (ft^2)**	**Mean Stories per Bldg**	**Mean Ht. per Story (ft)**	**Masonry Bldg Volume (ft^3)**	**Masonry Bldg Volume (m^3)**	**Population**	**Per Capita Masonry Bldg Volume (m^3)**	**Study Area Masonry Bldg Volume (m^3)**
Pierce Co.[a]	7.85×10^{7}	0.4	3.83×10^{7}	1.3	10.3	5.12×10^{8}	1.45×10^{7}	796,836	18	--
Snohomish Co.[b]	4.69×10^{7}	0.4	2.29×10^{7}	1.1	10.3	2.70×10^{8}	7.64×10^{6}	694,571	11	--
Pierce & Snoho. Combined	1.25×10^{8}	--	6.12×10^{7}	--	--	7.82×10^{8}	2.21×10^{7}	1,491,407	15[c]	--
Study Area	--	--	--	--	--	--	--	4,475,300	15	6.64×10^{7}

a Pierce County, 2010.

b Snohomish County, 2010.

[c] Pierce & Snohomish Counties combined masonry building volume divided by Pierce & Snohomish Counties combined population

References for Appendix J

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Appendix K. Concentrations of Individual PAHs in Various Petroleum Materials

Table K-1. Concentrations of Individual PAHs in Various Petroleum Materials (source: Valle et al., 2007)

	Lubricants and Motor oils	**Crude Oil**	**No. 6 Heavy (Asphalt)**	**No. 2 (Diesel and Heating Oil)**	**Used Motor Oil**
	mg/kg	**mg/kg**	**mg/kg**	**mg/kg**	**mg/kg**
LPAH					
Acenaphthene	NR	NR	NR	NR	0.67
Acenaphthylene	NR	NR	NR	NR	0
Anthracene	22	3	240	37	13
Fluorene	NR	50	NR	3,600	16.33
Naphthalene	590	400	NR	4,000	268.67
Phenanthrene	79	26	482	429	65.33
HPAH					
Benz(a)anthracene*	63	7	196	2	48.67
Benzo(a)pyrene*	17	1	NR	NR	17
Benzo(b)fluoranthene*	NR	0	22	NR	10.33
Benzo(k)fluoranthene*	NR	NR	NR	NR	6
Benzo(g,h,i)perylene	28	NR	NR	NR	33.33
Chrysene*	35	3	44	1	NR
Dibenz(a,h)anthracene*	NR	NR	NR	NR	8.33
Fluoranthene	38	5	23	41	467.67
Indeno(1,2,3-cd)pyrene*	40	NR	NR	NR	3.67
Pyrene	100	2	90	1	416
TOTAL PAH	1,012	497	1,097	8,111	1,375

*Carcinogenic PAHs
NR=Not Reported

Reference for Appendix K

Valle, S., M.A. Panero, and L. Shor, 2007. Pollution Prevention and Management Strategies for Polycyclic Aromatic Hydrocarbons in the New York/New Jersey Harbor. New York Academy of Sciences, New York, NY. 170 pages.

Appendix L. Individual PAH Emission and Release Rates for Cigarettes and Tires

Table L-1. Individual PAH Emission and Release Rates for Various Materials (source: Valle et al., 2007)

	Cigarette Smoke	**Tire Material**
	ug/cigarette	**mg/kg**
LPAH		
Acenaphthene	NR	NR
Acenaphthylene	NR	NR
Anthracene	0.76	ND
Fluorene	NR	NR
Naphthalene	NR	NR
Phenanthrene	2.6	11.8
HPAH		
Benz(a)anthracene*	0.27	ND
Benzo(a)pyrene*	NR	3.9
Benzo(b)fluoranthene*	0.54	ND
Benzo(k)fluoranthene*	NR	ND
Benzo(g,h,i)perylene	NR	ND
Chrysene*	0.67	8.2
Dibenz(a,h)anthracene*	NR	NR
Fluoranthene	0.95	11.1
Indeno(1,2,3-cd)pyrene*	NR	NR
Pyrene	1.0	54.1
TOTAL PAH	6.79	89.1

*Carcinogenic PAHs
NR=Not Reported
ND=Not Detected

Reference for Appendix L

Valle, S., M.A. Panero, and L. Shor, 2007. Pollution Prevention and Management Strategies for Polycyclic Aromatic Hydrocarbons in the New York/New Jersey Harbor. New York Academy of Sciences, New York, NY. 170 pages.

Appendix M. Permitted Aquatic Applications of Triclopyr in the Study Area

Table M-1. Permitted Aquatic Applications of Triclopyr in the Study Area
(source: Kathy Hamel, Ecology’s Water Quality Program, written communication)

Permit Number	Lake Name	County	Amount (Lbs)	Acres Treated	Dates Treated	EPA Registration Number	Plants Targeted	Product Name	Percent a.i. (labels)	Amount Applied (lbs)
994154	Spring Lake	King	240	1.5	08/05/09	67690-42	Eurasian milfoil	Renovate OTF	14.0%	**33.6**
994149	Beaver Lake	Skagit	120	1	7/30/09	62690-42	Myriophyllum spicatum	Renovate OTF	14.0%	**16.8**
994174	Cottage Lake	King	320	2	08/06/09	67690-42	Eurasian milfoil	Renovate OTF	14.0%	**44.8**
994060	Blue Lake	Thurston	2240	11	06/23/09	67690-42	Variable Milfoil	Renovate OTF	14.0%	**313.6**
994069	Lk. WA - Newport Shores	King	2880	24	5/27/09	62690-42	Myriophyllum spicatum	Renovate OTF	14.0%	**403.2**
994083	Mason Lake	Mason	4000	21.35	8/11/09	67690-50	Myriophyllum spicatum	Renovate Max G	4.0%	**160.0**
994083	Mason Lake	Mason	4000	21.35	8/11/09	67690-50	Myriophyllum spicatum	Renovate Max G	4.0%	**160.0**
994089	Clear Lake 1	Thurston	360	1.38	06/24/09	67690-42	Variable Milfoil	Renovate OTF	14.0%	**50.4**
994089	Clear Lake 1	Thurston	1000	5	08/18/09	67690-42	Variable Milfoil	Renovate OTF	14.0%	**140.0**
994108	Lk. WA - Hunts Point	King	4200	35	5/28/09	62690-42	Myriophyllum spicatum	Renovate OTF	14.0%	**588.0**
994112	Erie Lake	Skagit	300	3	7/29/09	62690-42	Myriophyllum spicatum	Renovate OTF	14.0%	**42.0**
994113	Big Lake	Skagit	80	0.5	7/28/09	62690-42	Myriophyllum spicatum	Renovate OTF	14.0%	**11.2**
994114	Campbell Lake	Skagit	600	6	7/29/09	62690-42	Myriophyllum spicatum	Renovate OTF	14.0%	**84.0**
994125	Lk. WA - Madison Estates	King	270	2.25	5/26/09	62690-42	Myriophyllum spicatum	Renovate OTF	14.0%	**37.8**

Reference for Appendix M

Hamel, Kathy, 2010. Written communication, March 8, 2010. Washington State Department of Ecology, Water Quality Program, Olympia, WA.

Appendix N. Washington Oil Spill Inventories

Table N-1. Washington Oil Spill Resource Damage Assessments, Feb 1991 – Jan 2011 (source: Ecology, 2011)

Date	Waterway		Material	Vessel/Facility		Volume (gal)
2/22/1991	Fidalgo	Bay	Crude	Texaco		40,000
6/30/1992	Blair	Waterway	IFO-180	Sun	Rose	850
7/17/1992	Elliott	Bay	Gasoline	Samson	Tug	70
10/11/1992	Elliott	Bay	Diesel	Arctic	Alaska	30
12/15/1992	Elliott	Bay	Diesel	Arctic	Alaska	500
1/7/1993	Elliott	Bay	Diesel	Arctic	Alaska	800
3/2/1993	SanJuan	Islands	Diesel/lube	F/V	Rover	495
4/15/1993	Sinclair	Inlet	JP-5	USS	Camden	5,400
4/21/1993	Blair	Waterway	IFO-180	Nosac	Forest	6,260
4/25/1993	Port	Townsend	Diesel	Port of	Port Townsend	900
5/4/1993	Bellingham	Bay	Diesel	Dutchie	C	60
6/1/1993	Tumwater	Creek	Diesel	Peninsula	Fuel	35
7/21/1993	Lake	Washington	Diesel	A.H.	Powers	20
8/3/1993	Point	Wells	Diesel	Great	Pacific	100
8/5/1993	Pier	91	Diesel	Arctic	Alaska	50
8/5/1993	Harbor	Island	Diesel	F/V	Excellence	3,387
8/8/1993	Elliott	Bay	Bunker-C	Pacific	NorthernOil	80
8/13/1993	SanJuan	Islands	Diesel&Lube	F/V	Radio	360
9/6/1993	Lake	Washington	Diesel	Stormy	Sea	30
9/25/1993	Springbrook	Creek	Diesel	Pilchuck	Construction	35
10/15/1993	Lake	Washington	Diesel	F/V	Anela	50
11/23/1993	McNeil	Island	Diesel	WA	Dept.ofCorrections	25
11/25/1993	Keyport		Diesel	Navy	Shore Facility	560
12/17/1993	Miller	Creek	HeatingOil	R.	Roullard	50
12/22/1993	Sinclair	Inlet	JP-5	USS	Nimitz	308
12/31/1993	Mitchell	Creek	Diesel	ALRT	Corp.	54
1/7/1994	Elliott	Bay	Diesel	Island	Tug&Barge,WandaS	40
1/25/1994	Lake	Union	Diesel	Foss	Maritime	300
2/1/1994	Manchester		Diesel	USS	Camden	30
2/15/1994	Duwamish	River	Diesel&Gas	Northwest	EnviroServices,Inc.	3,146
5/4/1994	Elliott	Bay	Diesel	JJW	Trucking	200
5/10/1994	Lake	Union	Diesel	Golden	Dawn	85
6/6/1994	Sinclair	Inlet	Diesel	USS	Sacramento	200
6/14/1994	Lake	Union	Gasoline	Matthew		50
6/29/1994	Lake	Union	Diesel	Arctic	IV	1,000
7/18/1994	Duwamish	Estry	Diesel&Lube	Joe	C	700
8/9/1994	Sinclair	Inlet	Diesel	U.S.Navy	TugYTB-768	325
9/11/1994	Lake	Union	Diesel&Lube	Omar		200
10/3/1994	Budd	Bay	HydraulicOil	Holbrook	Inc.	25
10/15/1994	Lake	Union	Diesel	Tyson	Seafood,ArcticI	25
10/25/1994	Skagit	River	Diesel	Crowley	Marine	200
10/27/1994	Sinclair	Inlet	JP-5	USS	Sacramento	3,700
11/5/1994	Lake	Union	Diesel	F/V	Sitkof	100
11/13/1994	Hood	Canal	Diesel	NOAA		80
12/17/1994	Shilshole	Bay	Diesel	Jupiter		50
12/31/1994	San Juan	Islands	Diesel	Crowley Marine,	Crowley 101	26,900
1/20/1995	Duwamish	Estry	Diesel	Polar	Cub	200
1/25/1995	Sinclair	Inlet	JP-5	U.S.Navy	BargeYON-102	2,520
1/25/1995	Seely	Lake	HydraulicOil	Johnson	Control	50

Table N-1 (Cont'd). Washington Oil Spill Resource Damage Assessments, Feb 1991 – Jan 2011 (source: Ecology, 2011)

Date	Waterway		Material	Vessel/Facility		Volume (gal)
1/26/1995	Lake	Washington	Diesel	Tripoli		30
1/30/1995	Lake	Union	Diesel	Coastline	PacificLtd.,Daphne	200
2/17/1995	Days	Island	Diesel	NX	Pression	250
2/20/1995	March	Point	HeavyGasOil	Texaco		20
2/20/1995	Thea Foss	Water Way	Heavy Oil	Tacoma	Public Schools	50
2/23/1995	Middle	Waterwy	Diesel	Catherine		200
4/22/1995	Everett	Marina	Diesel	Martinique		55
5/24/1995	Port	ofEverett	Diesel	Aleksandra	Kollontoy	100
6/2/1995	Elliott	Bay	Diesel	Northern	Victor	30
7/16/1995	Port	Gamble	Diesel	Betty	Jean	25
7/18/1995	Elliott	Bay	Diesel	Rybakcautoky		100
8/9/1995	Blair	Waterway	Bunker-C	Gastello		50
8/16/1995	Mt.	Pilchuck	Diesel	Snohomish	County	557
8/19/1995	Lake	Union	Gasoline	Pelican		40
8/26/1995	Sail	River	Diesel	Olympic	FishCompany	78
9/14/1995	Liberty	Bay	Diesel	Sea	Nest	75
9/14/1995	Everett	Harbor	Diesel	USS	DavidR,Ray	50
9/29/1995	Lake	Washington	Diesel	Diane		50
10/21/1995	T.Foss	Waterway	Bunker-C	WSDOT		50
10/31/1995	Strait	ofGeorgia	Crude	TOSCO		85
11/12/1995	Shilshole	Bay	Diesel	Omar		120
1/4/1996	Lake	Union	HydraulicOil	Muskrat		30
1/5/1996	Guemes	Channel	Diesel	Commodore		241
1/6/1996	Blair	Waterway	CrudeOil	U.S.		25
2/6/1996	March	Point	Diesel	Neptune		378
2/8/1996	Puget	Sound	Diesel	Burlington	Northern	3,200
2/26/1996	Sinclair	Inlet	Diesel	U.S.Navy	TugYTB-812	100
3/25/1996	Lake	Union	Diesel	Northern	Lady	450
4/16/1996	Lake	Union	Diesel	Polar	Queen	37
4/22/1996	T.Foss	Waterway	Diesel	Isswat		35
5/15/1996	Swinomish	Channel	Diesel	Expeditions	3	100
6/11/1996	Sinclair	Inlet	Diesel	USS	Nimitz	50
6/17/1996	Ebey	Slough	Diesel&Gas	Olympic	PipeLineCompany	1,000
8/5/1996	Hall	Creek	Diesel	Daniels,	Inc.	50
8/16/1996	SanJuan	Islands	Diesel	F/V	Kansas	650
8/26/1996	Sinclair	Inlet	JP-5	USS	Nimitz	1,300
9/22/1996	Three	TreePoint	Diesel	William	J	30
9/30/1996	Fidalgo	Bay	CrudeOil	Texaco		123
10/5/1996	Lake	Union	Diesel	Nor	PacIII	50
10/18/1996	Kingston		Diesel	Island	Tug&BargeGailS	300
11/10/1996	Sinclair	Inlet	LubeOil	USS	AbeLincoln	100
11/20/1996	Sinclair	Inlet	JetFuel	Barge	SWOB-8	300
12/14/1996	Lake	Union	Diesel	F/V	Zenith	100
12/22/1996	Tongue	Point	Diesel	E/Z		50
2/1/1997	Eld	Inlet	Diesel	Barge	Joan	25
2/24/1997	Sinclair	Inlet	JetFuel	USS	CarlVinson	200
2/28/1997	Everett	USNavy	LubeOil	USS	Callahan	120
3/2/1997	Port	Angeles	Bunker-C	ITT	Rayonier	75
3/18/1997	Hylebos	Creek	Diesel	Mabro	Corporation	80
5/27/1997	Sinclair	Inlet	Jet Fuel	U.S. Navy,	USS Rainier	25
6/9/1997	Lake	Union	Diesel	Hessa	Fjord	100
7/28/1997	I-90	Wetland	Diesel	Prime	TimeExpress	100
7/28/1997	Budd	Bay	Diesel	Duffer		50

Table N-1 (Cont'd). Washington Oil Spill Resource Damage Assessments, Feb 1991 – Jan 2011 (source: Ecology, 2011)

Date	Waterway		Material	Vessel/Facility		Volume (gal)
8/5/1997	Strait	ofGeorgia	IFO&JP-5	TOSCO		1,302
8/26/1997	Hylebos	Waterway	Diesel	Fascination		40
10/6/1997	Sinclair	Inlet	Jet Fuel	U.S. Navy,	USS Carl Vinson	50
12/12/1997	Cherry	Point	JetFuel	FortyNiner	Transp.,FNT-340	30
1/1/1998	Sitcum	Waterway	IFO	M/V	Anadyr	7,457
1/9/1998	Harbor	Island	Diesel	Olympic	T&B,CatherineQuigg	35
1/29/1998	Everett	Harbor	Diesel	U.S. Navy,	USS Paul Foster	50
2/10/1998	Bellingham	Bay	Diesel	Tug	TotemIII	36
3/7/1998	Sinclair	Inlet	Diesel	U.S. Navy,	USS Sacramento	30
3/29/1998	Sinclair	Inlet	Diesel	U.S.Navy	Barge309	25
4/3/1998	Sinclair	Inlet	JP-5	USS	Kittyhawk	4,200
4/13/1998	Sinclair	Inlet	Diesel	USS	Kittyhawk	800
4/15/1998	Sinclair	Inlet	Jet Fuel	U.S. Navy,	USS Carl Vinson	870
6/9/1998	Capitol	Lake	Road Sealant	WSDOT		200
7/14/1998	Lake	Union	Diesel	Vessel	Continuum	300
7/15/1998	TheaFoss	Waterway	Asphalt	Statewide	Parking	31
7/28/1998	Zittel's	Marina	Diesel	F/V	Debuster	25
8/1/1998	Hood	Canal	Waste Oil	U.S. Navy,	Bangor Sub Base	30
8/3/1998	SanJuan	Islands	Diesel	F/V	Starlight	75
8/28/1998	Bellingham	Bay	Gasoline	Hilton	HarborMarina	70
9/21/1998	White	River	Diesel	McKinlay	&Co.Trucking	86
10/13/1998	Everett	Harbor	Diesel	U.S Navy,	USS David R. Ray	62
10/21/1998	Hylebos	Waterway	HydraulicOil	F/V	KiskaEnterprise	30
11/22/1998	Strait	JuandeFuca	Diesel	F/V	LadyValerie	150
12/8/1998	Central	PugetSound	Hydraulic	M/V	Sealth,	120
1/5/1999	Everett	Harbor	Diesel	USS	Abraham Lincoln	800
4/29/1999	Elliott	Bay	LubeOil	Todd	Shipyard	350
5/20/1999	Commencement	Bay	Diesel	Vessel	SeawayII	50
6/5/1999	Strait	JuandeFuca	LubeOil	SS	OceanPhoenix	60
6/8/1999	Manchester		Diesel	USS	Juneau	522
6/10/1999	Whatcom	Creek	Gasoline	Olympic Pipe	Line Company	277,200
6/27/1999	Strait	Georgia	CrudeOil	M/V	ArcoTexas	300
7/7/1999	Elliott	Bay	Diesel	F/V	AlaskanCommand	25
7/28/1999	Manchester		Diesel	USCGC	PolarStar	84
8/18/1999	LakeWA	Ship Canal	Diesel	F/V	DeepPacific	350
9/1/1999	Sinclair	Inlet	Diesel	US. Navy,	USS Camden	900
9/4/1999	Lake	Union	Diesel	Horizon	Fisheries	25
9/6/1999	Budd	Inlet	Diesel	Vessel	ArchibaldEley	100
9/10/1999	Port	Townsend	IFO	Port	TownsendPaper	200
10/19/1999	Elliot	Bay	Bunker-C	Pacific	NorthernOil	30
10/22/1999	Port	Townsend	Diesel	US. Navy,	USS Bridge	400
11/11/1999	Samish	Bay	Diesel	Tug	Noble	125
11/18/1999	Elliot	Bay	Bunker-C	SS	GreatLandTOTE	1,273
12/12/1999	Possession	Sound	Diesel	Dunlap	Towing	288
12/13/1999	Possession	Sound	HydraulicOil	Manson	Construction	50
12/30/1999	Elliott	Bay	Gasoline	Seacoast	Towing	35
1/19/2000	East	Waterway	Diesel	Island	Tug&Barge,EstherS	290
2/3/2000	Hwy	12wetland	Diesel	USFWS,	NisquallyRefuge	25
2/14/2000	Neah	Bay	Diesel	F/V	DoryLynn	250
2/17/2000	DesMoines	Creek	Heating	Wesley	Homes	150
2/22/2000	Issaquah	Creek	Diesel	Trism,	Inc.Trucking	100
2/25/2000	LakeWA	Ship Canal	LubeOil	F/V	SouthernWind	70
3/27/2000	Sinclair	Inlet	Diesel	US. Navy,	USS Camden	35

Table N-1 (Cont'd). Washington Oil Spill Resource Damage Assessments, Feb 1991 – Jan 2011 (source: Ecology, 2011)

Date	Waterway		Material	Vessel/Facility		Volume (gal)
4/4/2000	Strait	ofJuandeFuca	Diesel	Vessel	ArchibaldJ.Eley	40
5/2/2000	Liberty	Bay	Diesel	Vessel	Laverne	60
5/12/2000	LakeWA	Ship Canal	HydraulicOil	F/V	Northwind	318
5/14/2000	Elliott	Bay	Diesel	Sea	CoastTowing	35
6/10/2000	Puget	Sound	Diesel	Barge	LuckyBuck	496
6/19/2000	Commencement	Bay	Diesel	Alaskan	Command	400
6/21/2000	LakeWA	Ship Canal	Diesel	F/V	SeaFisher	80
8/7/2000	Port	Townsend	Diesel	Vessel	Cavok	95
8/22/2000	Neah	Bay	Diesel	Forks	Sand&Gravel	30
8/24/2000	Port	Orchard	Gasoline	Harris	Transportation	731
10/15/2000	Mitchell	Bay	Diesel	VesselHydah		50
10/20/2000	North	Creek	Diesel	Snohomish	FireDistrict-1	355
11/5/2000	LakeWA	Ship Canal	Diesel	F/V	Columbia	200
11/14/2000	Port	Hadlock	Diesel	Vessel	Norden	35
11/15/2000	Elliott	Bay	Diesel	Crowley	TugStalwart	1,667
11/21/2000	Lake	Union	Diesel	F/V	WesternQueen	70
12/5/2000	Lake	Union	Diesel	Sea	&ShoreConstruction	1,063
12/7/2000	Ditch	toBuddBay	WasteOil	Dunlap	Towing	150
12/20/2000	Commencement	Bay	Diesel	M/V	RubinPearl	103
1/3/2001	Lake	Union	Diesel	F/V	CoastalMerchant	111
1/26/2001	Lynch	Creek	Diesel	Associated	PetroleumProducts	215
1/29/2001	Port	AngelesHarbor	CrudeOil	T/V Prince	William Sound	504
2/20/2001	Everett	Harbor	JP-5	USS	Abraham Lincoln	45
3/15/2001	Guemes	Channel	Gasoline	Newport	Petroleum	200
4/15/2001	Oakland	Bay	LubeOil	Simpson	Timber	71
4/18/2001	Lincoln	Ditch	Diesel	Fields	Corporation	300
5/1/2001	Bellevue	Lake	Diesel	Safeway	Corporation	1,075
5/3/2001	LakeWA	Ship Canal	Diesel	Sheryl	Belcher,F/VSeaLassie	150
5/10/2001	Halls	Creek	Diesel	Vitamilk	DairyProducts	70
6/3/2001	Sitcum	Waterway	Bunker-C	CSX	Reliance	61
6/4/2001	Elliott	Bay	Diesel	F/V	Cleveland	70
6/13/2001	Strait	ofGeorgia	crude	M/V	OverseasBoston	2,436
6/18/2001	Blair	Waterway	ANScrude	M/V	Overseas	84
6/21/2001	Sinclair	Inlet	Diesel	US. Navy,	USS Sacramento	80
7/6/2001	Drayton	Harbor	Diesel	F/V	Glory	200
8/6/2001	Port	AngelesHarbor	LubeOil	F/V	Tunek	35
8/28/2001	Strait	ofGeorgia	Gasoline	ITB	Baltimore	150
9/19/2001	Lake	Union	Diesel	F/V	FierceContender	750
10/5/2001	Puget	Sound	Diesel	BNSF	Railway	982
10/23/2001	Lake	Washington	Diesel	Tug	SeaPrince	203
10/27/2001	Pier	91	WasteOil	F/V	Arctic	40
11/11/2001	Elliott	Bay	MDO	Kinder-	Morgan	2,136
11/18/2001	Port	MadisonBay	Diesel	Vessel	High	100
12/12/2001	Paradise	River	Diesel	National Park	Service	2,000
12/13/2001	Elliott	Bay	Diesel	BNSF	Railway	2,336
1/4/2002	Gig	Harbor	Diesel	F/V	St.Mary	250
1/22/2002	Maytown	Wetland	Diesel	Hammell	Transport	125
1/28/2002	Blind	Bay,ShawIsland	Diesel	Vessel	EightBells	50
2/8/2002	Eagle	Harbor	Diesel	Caicos	Corp.,TugWinslow	50
2/19/2002	Everett	Harbor	Oil/water mix	US. Navy,	USS Abraham Lincoln	2,000
2/21/2002	Commencement	Bay	Diesel	University of	Puget Sound	122
4/5/2002	UW	Wetland	Diesel	North	AmericanVanLines	200
4/24/2002	Ohanapecosh	River	Diesel	National Park	Service	985

Table N-1 (Cont'd). Washington Oil Spill Resource Damage Assessments, Feb 1991 – Jan 2011 (source: Ecology, 2011)

Date	Waterway		Material	Vessel/Facility		Volume (gal)
5/7/2002	Port	Gamble	Diesel	Vessel	Gratitude	28
5/9/2002	Everett	Harbor	Diesel	USS	Fife	60
5/16/2002	Deer	Creek	Gas&Diesel	Mid-Mac	Enterprises	1,050
5/19/2002	Port	AngelesHarbor	Bunker-C	M/V	GazDiamond	1,188
5/22/2002	Salmon	Bay	Diesel	F/V	Pathfinder	270
5/24/2002	Commencement	Bay	Diesel	Simpson	Timber	50
6/12/2002	Sinclair	Inlet	Diesel	US. Navy,	USS Camden	42
7/2/2002	Sinclair	Inlet	Diesel	US. Navy,	USS Camden	30
7/5/2002	Compressor	Creek	Diesel	Pierce	Co.PublicWorks	1,919
7/16/2002	Tibbetts	Creek	Gasoline	Lee	&EatesTankLines,Inc.	2,076
8/19/2002	SanJuan	Channel	Diesel	VesselCrossfire	-	30
8/25/2002	Rosario	Straits	Diesel	VesselNamaste	Lyons,	100
10/12/2002	McNeil	Island Wetland	Diesel	DOC, McNeil	Island	2,576
10/22/2002	Hylebos	Waterway	Bunker-C	M/V	Kaede	50
11/11/2002	Friday	Harbor	Diesel	Vessel	IlimaNalani	45
11/25/2002	Elliott	Bay	HydraulicOil	American	DivingService	50
1/4/2003	Salmon	Bay	HydraulicOil	F/V	Alyeska	74
1/11/2003	Blair	Waterway	HydraulicOil	General	Construction	50
3/12/2003	Oakland	Bay	Diesel	Evergreen	Fuel	50
3/22/2003	Eld	Inlet	Diesel	Sea-Nik I,	Edenstrom	150
4/7/2003	Fife	wetland	Diesel	Western	Cartage	50
6/16/2003	Swinomish	Channel	Diesel	F/V	LadyIndependence	91
7/12/2003	Scriber	Creek	Gasoline	Harris	Transportation	2,244
7/17/2003	Commencement	Bay	Diesel	Vessel	Jimmyboy	60
8/21/2003	Elliott	Bay	Diesel	F/V	Excellence	144
9/18/2003	Fidalgo	Bay	Diesel	F/V	MissMattie	225
10/12/2003	Acme	Wetland	Diesel&Gasoline	McEvoy	OilCo.	2,919
10/28/2003	Admiralty	Inlet	Diesel	F/V	Martle	700
11/15/2003	Sinclair	Inlet	Diesel	PSNS	-	35
11/28/2003	Port	Townsend	Diesel	Caicos	Corporation	70
12/22/2003	Southgate	Creek	HeatingOil	Vince	Christianson	300
12/30/2003	Port	Madison	Bunker-C	Foss Barge	248 P-2	4,637
1/1/2004	Strait	ofGeorgia	Diesel	Fred	Williamson,VesselAllura	500
1/1/2004	Strait	ofGeorgia	Diesel	Vessel	PacificMako	450
1/13/2004	Lacey	Wetland	Diesel	St Martins	College	600
3/26/2004	Salmon	Bay	LubeOil	F/V	FrontierMariners	218
4/18/2004	Port	Susan	Gasoline	Everett	Brown	50
5/2/2004	Salmon	Bay	Diesel	F/V	CapeWindy	270
5/19/2004	Eld	Inlet	Diesel	James Day,	Vessel Kuan Yin	30
5/26/2004	Friday	Harbor	HydraulicOil	VesselPanther	-	95
6/4/2004	Duwamish	River	Diesel	M.	Diamond,VesselGoliathon	52
6/25/2004	Friday	Harbor	Diesel&HydraulicOil	Vessel	AndrewZ	85
7/8/2004	Hylebos	Waterway	Diesel	R/V	PointDefiance	30
8/13/2004	Salmon	Bay	LubeOil	F/V	Diomedes	124
8/16/2004	Neah	Bay	HydraulicOil	F/V	Alyeska	400
9/9/2004	Highwood	Lake	Diesel	The	Firs	100
10/14/2004	Dalco	Passage	Crude	Conoco-	Phillips	7,200
11/4/2004	Kent	Wetland	WasteOil	Kent	SchoolDistrict	200
12/13/2004	Seattle	Pond	Diesel	Schnieder	Trucking	35
1/18/2005	Strait	ofGeorgia	Diesel	Sirius	Maritime	416
2/13/2005	Steilacoom	Marina	Diesel	Vessel	Tranquility	65
2/14/2005	Strait	ofGeorgia	IFO-380	Marine	Petrobulk	109
3/24/2005	Steilacoom	Marina	Diesel	F/V	Islander	50

Table N-1 (Cont'd). Washington Oil Spill Resource Damage Assessments, Feb 1991 – Jan 2011 (source: Ecology, 2011)

Date	Waterway		Material	Vessel/Facility		Volume (gal)
5/5/2005	Grays	Harbor	Diesel	F/V Loree	-	50
6/23/2005	Lake WA	ShipCanal	LubeOil	M/V	Florence Filberg	330
7/6/2005	Blair	Waterway	Diesel	Totem	OceanTrailerExpress	75
7/12/2005	Port	Madison	Diesel	F/V	Kristina	50
7/22/2005	Hylebos	Waterway	Diesel	F/V	JyhShen	100
8/8/2005	Hammersley	Inlet	Diesel	F/V	JoyIrene	50
8/31/2005	Gig	Harbor	Diesel	Harborview	MarinaFire	450
8/31/2005	Swinomish	Channel	Diesel	F/V	Renamarie	206
10/17/2005	Port	Gamble	Diesel	Tug	Active	200
10/28/2005	Port	Townsend	Diesel	U.S. Navy,	USS Rodney Davis	228
12/18/2005	Mats	Bay	Diesel	Tug	Yarder	50
1/22/2006	Duwamish	Waterway	Diesel&Lube	Tug Island Brave		2,094
1/28/2006	Skagit	Bay	Diesel	F/V	Rover	65
1/31/2006	Muck	Creek	Diesel	Milgard	Windows	26
3/29/2006	Lake	Union	Diesel	F/V	Defender	48
5/29/2006	Puget	Sound	Diesel	Pleasure	VesselDreamCatcher	43
6/15/2006	Everett	Harbor	Diesel	Port of	Everett	472
7/26/2006	Thornton	Creek	Gasoline	Sentinel	Petroleum	84
8/3/2006	TheaFoss	Waterway	Gasoline	Jeremiah	Firth	35
9/4/2006	Duwamish	River	Diesel	Olympic	Tug&Barge	43
9/15/2006	Port	Townsend	Diesel	F/V	SilverLady	110
1/7/2007	Puyallup	River	Diesel	Organically	GrownCompany	115
1/9/2007	LakeWA	Ship Canal	Diesel	M/V	CoastalPilot	50
1/9/2007	TheaFoss	Waterway	Diesel	Key	Bank	272
1/23/2007	Drayton	Harbor	Diesel	F/V	CapeUltika	40
1/25/2007	Dyes	Inlet	UsedLube	Sears,	Roebuck&Co	50
2/5/2007	Gig	Harbor	Diesel	Vessel	X-TA-SEA	90
2/6/2007	Tacoma	Wetland	Diesel	Gerald	Meyer	100
2/28/2007	Elliot	Bay	IFO-380	M/V	SongaHua	93
2/28/2007	Franklin	Creek	Diesel	Hogland	Transfer	50
3/12/2007	Enumclaw	Ditch	Diesel	Small	&SonsOilDistributing	63
3/22/2007	Rosario	Strait	Diesel	Vessel	Destiny	150
4/15/2007	Steamboat	Slough	Diesel	Tug	IslandChampion	374
5/5/2007	Strait	ofJuandeFuca	Diesel	Vessel	SpiritQuest	350
5/9/2007	Swinomish	Channel	Diesel	Vessel	Eagle	206
6/11/2007	Squalicum	Harbor	Diesel	F/V	Heidi	60
6/26/2007	Horse	Creek	Kerosene	QFC	Foods	36
6/28/2007	I-90	Wetland	HydraulicOil	Cranes	4Rent	90
7/10/2007	Hylebos	Waterway	Diesel	F/V	Bowfin	90
8/17/2007	Drayton	Harbor	Diesel	F/V	MuirMilach	260
9/22/2007	Tulalip	Bay	Diesel	F/V	St.Nicholas	70
9/28/2007	Blair	Waterway	Diesel	Graymount	Western	25
10/22/2007	Swinomish	Channel	Diesel	F/V	NewQueen	115
10/29/2007	MtVernon	Ditch	Diesel	Skip	Hamilton	45
11/20/2007	Eagle	Harbor	Diesel	M/V	CommanderToad	30
11/23/2007	Crescent	CreekTributary	Diesel	CFM	Excavating	25
12/1/2007	Guemes	Channel	Diesel	M/V	Trident	40
12/3/2007	McAllister	Creek	Diesel	Swift	Transportation	75
12/3/2007	LakeWA	Ship Canal	WasteOil	Ballard	Oil	50
12/10/2007	Hylebos	Waterway	Diesel	F/V	Bowfin	496
1/17/2008	LakeWA	Ship Canal	Diesel	F/V	CapeHorn	75
1/18/2008	Green	RiverTributary	TransformerOil	Seattle	CityLight	40
3/1/2008	Hylebos	Creek	Diesel	Gresham	Transfer,Inc.	30

Table N-1 (Cont'd). Washington Oil Spill Resource Damage Assessments, Feb 1991 – Jan 2011 (source: Ecology, 2011)

Date	Waterway		Material	Vessel/Facility		Volume (gal)
3/12/2008	Skagit	Bay	Diesel	Truck	CityTruckStop	84
4/4/2008	Salmon	Bay	Diesel	Vessel	ChristineK	42
5/5/2008	Commencement	Bay	Diesel	F/V	Caldo	30
5/8/2008	Maddox	Creek	Diesel	Birch	EquipmentRental	25
6/6/2008	Salmon	Bay	Diesel	Trident	Seafoods,F/Vbillikin	230
6/9/2008	Strait	ofGeorgia	Diesel	Tug Tiger-K,	SeaTransportation	120
6/19/2008	Skagit	River	TransformerOil	Seattle	CityLight	180
6/20/2008	Friday	Harbor	Diesel	P/V	AfricanGrace	25
7/15/2008	Duwamish	Waterway	Diesel	Tug	JamesT.Quigg	161
8/18/2008	TheaFoss	Waterway	Diesel	M/V	OverBudget	100
11/7/2008	Lake	Union	Diesel	Roadway	Express	100
11/26/2008	Bellingham	Bay	Diesel	Tug BarentsSea,K-	SeaTransp.	25
12/24/2008	Neah	Bay	Diesel	F/V	MyAmmi	100
1/11/2009	LakeWA	ShipCanal	Diesel	P/V	SapphireSea	200
1/20/2009	Hylebos	Waterway	CookingOil	Meeker	MiddleSchool	35
2/3/2009	Central	PugetSound	Diesel	Tug	CatherineQuigg	211
2/5/2009	Steamboat	Slough	Diesel	F/V	ElusiveDream	200
2/20/2009	Orcas	Is.WestSound	Diesel	P/V	Coho	50
2/28/2009	Lake	Union	Gasoline	Aaron	Hubner	35
3/17/2009	Puget	Sound	HydraulicOil	M/V	HyundaiRepublic	159
4/18/2009	Lake	Washington	Diesel	Tug	SeaPrince	252
7/30/2009	Central	PugetSound	Diesel	P/V	TwilaDawn	30
8/28/2009	Cattle	Pass	Diesel	F/V	Anna J	450
9/5/2009	Oak	Harbor	Diesel	P/V	Never Satisfied	220
11/4/2009	Roche	Harbor	Gasoline	P/V	Liquid Assests	100
11/5/2009	Sinclair	Inlet	JP-5	U.S. Navy,	USS Abraham Lincoln	489
11/10/2009	Duwamish	River	Diesel	P/V	Angel Rae	665
1/7/2010	Horse	Creek	Diesel	Waste	Management	35
1/12/2010	Port of	Everett	Gasoline	Elizabeth	Weeks	50
2/28/2010	Duwamish	Waterway	Lube Oil	Tug	Island Wild	121
3/2/2010	Friday	Harbor	Diesel	P/V	Eldorado	28
3/8/2010	Sinclair	Inlet	Diesel	M/V	Mist Cove	456
3/14/2010	Bellingham	Wetland	Jet fuel	Bellingham	Fuel Service	60
4/3/2010	Hylebos	Waterway	Fish Oil	Oil	F/V Island Enterprise	83
4/15/2010	Commencement	Bay	Diesel	P/V	Heron	70
4/22/2010	Bellingham	Bay	Diesel	F/V	Muir Milach	49
4/29/2010	Puget	Sound	Diesel	Long	Painting Company	114
5/28/2010	John's	Creek	Cooking	Oil	Waste Management	140
6/22/2010	Lake WA	Ship Canal	Diesel	Tug	Rustler	216
6/29/2010	Port of	Everett	Diesel	P/V	Sea Dragon	30
7/9/2010	Possession	Sound	Gasoline	Valcrig	Pruglo	50
8/6/2010	Puget	Sound	Gasoline	Michael	Dunsmore	80
9/10/2010	Elliott	Bay	Diesel	Tug	Howard Olsen	36
10/29/2010	Blaire	Waterway	Diesel	Manson	Construction	177
6/29/2010	Port of	Everett	Diesel	P/V	Sea Dragon	30
7/9/2010	Possession	Sound	Gasoline	Valcrig	Pruglo	50
8/6/2010	Puget	Sound	Gasoline	Michael	Dunsmore	80
11/26/2010	Clover	Creek	Diesel	Certified	Cleaning Services	100
12/20/2010	Strait of	Juan de Fuca	Diesel	P/V	Scout	300
1/14/2011	Lake WA	Ship Canal	Diesel	P/C	Kokanee	50

Table N-2. Gallons of Petroleum Spilled into Waters of the Study Area by Regulated Sector, 1995-2008 (source: ERC, 2009)

Geographic Zone	Oil Tankers	Tank Barges	Cargo Vessels	Fishing Vessels	Passenger Vessels	Terminals	Pipelines	Tank Trucks	Marinas	Total
Juan de Fuca	0	0	0	0	0	0	0	0	0	**0**
Inner Straits	1,650	40	0	95	0	235	0	0	0	**2,020**
Rosario Strait	3,586	465	0	0	0	0	277,200*	0	70	**281,321**
Whidbey	0	0	0	0	0	0	0	0	0	**0**
N. Puget	570	1,625	3,135	1,318	0	0	0	0	0	**6,648**
C. Puget	0	4,737	1,609	1,362	120	2,136	0	0	30	**9,994**
S. Puget	7,334	60	15,656	1,476	0	2,576	0	0	50	**27,152**
Hood Canal	0	0	0	0	0	0	0	0	0	**0**
Lake Union	0	25	0	1,495	0	0	0	0	50	**1,570**
Total	**13,140**	**6,952**	**20,400**	**5,746**	**120**	**4,947**	**277,200**	**0**	**200**	**328,705**

* Bellingham (Olympic Pipeline Spill) gasoline pipeline leak, 1999

Table N-3. Summary of Spills Incidents Tracked in the Environmental Report Tracking System (ERTS) During 2009 (source: Ecology 2010).

	Receiving Medium				
	Fresh	**Marine**	**Soil**	**Unknown**	**Total**
Diesel					
Volume of Spills with Reported Quantity (gal)	2,173	706	5,168	1,358	9,405
No. Incidents with Reported Quantity	43	16	32	39	130
Median Volume of Spills with Reported Quantity (gal)	20	5	28	10	--
No. Incidents w/o Reported Quantity	12	7	40	22	81
Gasoline					
Volume of Spills with Reported Quantity (gal)	333	35	5,093	251	5,712
No. Incidents with Reported Quantity	26	5	15	32	78
Median Volume of Spills with Reported Quantity (gal)	6	1	3	5	--
No. Incidents w/o Reported Quantity	13	4	47	26	90
Crude Oil					
Volume of Spills with Reported Quantity (gal)	0	0	0	0	0
No. Incidents with Reported Quantity	0	0	0	0	0
Median Volume of Spills with Reported Quantity (gal)	0	0	0	0	--
No. Incidents w/o Reported Quantity	0	0	0	1	1
Lube and Motor Oil					
Volume of Spills with Reported Quantity (gal)	164	16	298	290	768
No. Incidents with Reported Quantity	18	8	18	29	73
Median Volume of Spills with Reported Quantity (gal)	2	1	10	2	--
No. Incidents w/o Reported Quantity	16	8	22	20	66
Fuel (Heating) Oil					
Volume of Spills with Reported Quantity (gal)	10	0	452	200	662
No. Incidents with Reported Quantity	1	0	5	1	7
Median Volume of Spills with Reported Quantity (gal)	10	0	5	200	--
No. Incidents w/o Reported Quantity	2	1	5	1	9
Jet and Aviation Fuel					
Volume of Spills with Reported Quantity (gal)	1,073	501	101	172	1,847
No. Incidents with Reported Quantity	6	2	2	12	22
Median Volume of Spills with Reported Quantity (gal)	10	251	51	5	--
No. Incidents w/o Reported Quantity	0	1	0	0	1
Other Petroleum (a)					
Volume of Spills with Reported Quantity (gal)	19,955	144	4,910	4,846	29,855
No. Incidents with Reported Quantity	60	19	182	92	353
Median Volume of Spills with Reported Quantity (gal)	9	1	5	5	--
No. Incidents w/o Reported Quantity	66	16	169	60	311

(a) Includes asphalt/creosote, hydraulic, mineral, other, waste/used, and unknown oils

References for Appendix N

Ecology, 2010. Environmental Report Tracking System (ERTS) for 2009.

Ecology, 2011. Washington Oil Spill Resource Damage Assessments: 1991-2011. www.ecy.wa.gov/programs/spills/restoration/RDA%20web%20page%20report-by%20date%203-11.pdf. Accessed 4/6/2011.

ERC, 2009. Oil Spill Risk in Industry Sectors Regulated by Washington State Department of Ecology Spills Program for Oil Spill Prevention and Preparedness. Prepared for Washington State Department of Ecology by Environmental Research Consulting, Cortlandt Manor, NY.

Appendix O. Underground Storage Tanks in the Study Area

Table O-1. Number and Material Stored in Registered Underground Storage Tanks in the Study Area (source: Ecology, 2010).

Material	Tank Capacity (gallons)							Total
	111 - 1,100	1,101 - 2,000	2,001 - 4,999	5,000 - 9,999	10,000 - 19,999	20,000 - 29,000	50,000	
Diesel	107	66	126	258	871	186	12	**1,626**
Biodiesel Blend	0	1	4	2	8	2	0	**17**
Unleaded Gasoline	71	46	153	668	2,971	355	0	**4,264**
Leaded Gasoline	53	11	28	121	519	18	0	**750**
Alcohol Blended Gasoline	0	1	0	2	17	2	0	**22**
Bunker C	0	0	0	2	0	0	0	**2**
Motor Oil	6	2	14	9	4	0	0	**35**
Heating Fuel	467	7	12	10	19	24	0	**539**
Kerosene	8	3	2	16	8	6	0	**43**
Aviation Fuel	6	0	6	12	58	22	21	**125**
Used Oil/Waste Oil	316	12	14	9	3	1	0	**355**
Other Petroleum	7	2	16	6	1	2	0	**34**
Total	**1,041**	**151**	**375**	**1,115**	**4,479**	**618**	**33**	**7,812**

Reference for Appendix O

Ecology, 2010. Database listings of registered underground storage tanks in Washington. Accessed at fortress.wa.gov/ecy/tcpwebreporting/reports.aspx.

Appendix P. Summary of Uncertainties Associated with Release Estimates

Table P-1. Summary of Uncertainty Associated with Release Estimates.

Activity/ Source Category	Specific Source	Arsenic	Cadmium	Copper	Lead	Mercury	Zinc	PCBs	PBDEs	PCDD/Fs	PAH	Phthalates	Nonylphenol	Triclopyr	Petroleum
Buildings and Grounds	Backyard Burn Barrels									M					
	Indoor office space dust								MH						
	Indoor office space air								MH						
	Indoor residential air								MH						
	Indoor residential dust								MH						
	Lawn and Garden Equipment Emissions									MH	MH				
	Plumbing fixtures, pipes, and solder			MH	MH		MH								
	PVC Cables (indoor)											MH			
	PVC Coil coated roofing											MH			
	PVC Flooring											MH			
	PVC Hoses and profiles (indoor)											MH			
	PVC Hoses and profiles (outdoor)											MH			
	PVC Roofing material											MH			
	PVC Wall coverings											MH			
	Residential Fuel Use, except Wood	MH	MH		MH	MH					MH				
	Residential Trash Burning							MH			MH				
	Residential Yard Waste Burning										MH				
	Roofing materials - asphalt shingle	M													
	Roofing materials - total		M	M	M		M				M				
	Sealants (Caulking)							MH							
	Urban lawn & garden use of pesticides			MH										MH	
	Woodstoves and Fireplaces		MH							MH	MH				
Industrial and Institutional Point Sources	Aluminum Mills										M				
	Army Base			M	M										
	Boat Manufacturer											M			
	Cement Plants					M				M					
	Crematoria Emissions					M									
	Ind, Comm, Inst Sources (primarily Title V)	MH	MH	MH	MH	MH	MH				MH	MH	MH		
	Metal Foundries			M											
	Naval Shipyard			M	M		M								
	Other Industrial and Military Facilities			M		M	M			M	M	M			
	Paint and Coatings Manufacturers											MH			

Table P-1 (Cont'd). Summary of Uncertainty Associated with Release Estimates.

Activity/ Source Category	Specific Source	Arsenic	Cadmium	Copper	Lead	Mercury	Zinc	PCBs	PBDEs	PCDD/Fs	PAH	Phthalates	Nonylphenol	Triclopyr	Petroleum
Industrial and Institutional Point Sources	Petroleum Refineries					M	M			M	M				
	Plastics Manufacturer											M			
	Pulp and Paper Mills				M	M	M			M	M				
	Residual Fuel Oil Combustion					M									
	Specialty Glass Manufacturer		M												
	Steel Galvanizers						M								
	Steel Mills					M	M								
	Various Industrial Facilities, not including pulp mills				M										
	Wood-Treatment Facility	M								M					
Miscellaneous Material Use	Auto Convenience Switch Disposal					MH									
	Button Cell Batteries					M									
	Fertilizers	MH	MH		MH	MH									
	Fertilizers and Micronutrients						MH								
	Fluorescent Lamp Disposal					M									
	Lacquers and paint											MH			
	Large capacitors							H							
	Micronutrients			MH											
	Petroleum spills (large)														M
	Printing inks											MH			
	PVC Cables (outdoor, above ground)											MH			
	PVC Coated fabric											MH			
	PVC Films, sheets, coated products											MH			
	Sealants, adhesives, etc.											MH			
	Shoe soles											MH			
	Small capacitors							H							
	Thermometers (Household)					H									
	Thermostat Disposal					M									
	Transformers							H							
Non-Point Combustion Sources	Agricultural Equipment Emissions									MH	MH				
	Airport Service Equipment Emissions									MH	MH				
	Aviation fuel combustion				MH										
	Cigarette smoke										M				
	Commercial Equipment Emissions									MH	MH				
	Construction Equipment Emissions									MH	MH				
	Industrial Equipment Emissions									MH	MH				
	Locomotive Emissions		MH		MH						MH				
	Logging Equipment Emissions									MH	MH				
	Railroad Maintenance Equipment Emissions										MH				
	Recreational Boat Emissions									MH	MH				
	Recreational Equipment Emissions									MH	MH				

Table P-1 (Cont'd). Summary of Uncertainty Associated with Release Estimates.

Activity/ Source Category	Specific Source	Arsenic	Cadmium	Copper	Lead	Mercury	Zinc	PCBs	PBDEs	PCDD/Fs	PAH	Phthalates	Nonylphenol	Triclopyr	Petroleum
Outdoor Product Use	Ammunition, Hunting shot use				**LM**										
	Fishing sinker loss				**LM**										
Personal Care Products	Body lotion											**MH**			
	Dental Amalgam Excretion and Disposal					**MH**									
	Deodorant (solid)											**MH**			
	Fragrance											**MH**			
	Hair spray (aerosol and pump spray)											**MH**			
	Nail polish											**MH**			
Pesticides and Wood Preservation	Agricultural use of pesticides - total			**MH**											
	Antifouling paint - total			**M**											
	Aquatic Weed Control													**H**	
	Aquatic-use algaecides in pools, fountains, spas, etc.			**M**											
	CCA-treated wood	**M**		**M**											
	Creosote Treated Marine pilings - total										**MH**				
	Creosote Treated Railroad ties										**MH**				
	Creosote Treated Utility poles										**MH**				
	Crop and Golf Course Use													**H**	
	Forest Herbicide Use - State Forests													**LM**	
	Right-of-Way Maintenance—State Forests													**M**	
Vehicles and Roads	Asphalt - total										**M**				
	Brake pad wear		**M**	**M**	**M**		**M**								
	Car undercoating											**M**			
	Coal tar sealants										**H**				
	Gas Station Emissions										**MH**				
	Gasoline and Diesel Combustion					**M**									
	Heavy Duty Diesel Vehicle Emissions									**MH**	**MH**				
	Heavy Duty Gasoline Vehicle Emissions									**MH**	**MH**				
	Improper disposal of used oil following oil changes														**M**
	Light Duty Diesel Vehicle Emissions									**MH**	**MH**				
	Light Duty Gasoline Vehicle Emissions									**MH**	**MH**				
	Minor gasoline spills from fueling vehicles and non-road equipment														**LM**
	Motor oil drips and leaks														**M**
	Motor oil leaks and improper disposal						**MH**								
	Petroleum spills, leaks, and improper motor oil disposal										**MH**				

Table P-1 (Cont'd). Summary of Uncertainty Associated with Release Estimates.

Activity/ Source Category	Specific Source	Arsenic	Cadmium	Copper	Lead	Mercury	Zinc	PCBs	PBDEs	PCDD/Fs	PAH	Phthalates	Nonylphenol	Triclopyr	Petroleum
Vehicles and Roads	Right-of-Way Maintenance - Railroads													**MH**	
	Right-of-Way Maintenance – State Highways													**LM**	
	Tire wear		**LM**	**LM**	**LM**		**LM**				**LM**				
	Wheel weight loss				**LM**										

LM=low-medium
M=medium
MH=medium-high
H=high

Appendix Q. Summary of Release Estimates for All COCs

Table Q-1. Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Arsenic	**TOTAL**					**0.79 (0 - 1.7)**	**t/yr**	**100%**
Arsenic	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.28	t/yr	36% (0% - 100%)
Arsenic	Pesticides and Wood Preservation	CCA-treated wood	Leaching	Solubilized in water	Soil, Surface water	0.27 (0.04 - 0.5)	t/yr	34% (0% - 100%)
Arsenic	Buildings and Grounds	Roofing materials - asphalt shingle	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	0.15 (0 - 0.84)	t/yr	19% (0% - 100%)
Arsenic	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.06	t/yr	8% (0% - 100%)
Arsenic	Industrial and Institutional Point Sources	Wood-Treatment Facility	Unknown release	Unknown form	Water	0.01	t/yr	2% (0% - 100%)
Arsenic	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	1% (0% - 100%)
Cadmium	**TOTAL**					**0.96 (0.84 - 1.2)**	**t/yr**	**100%**
Cadmium	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	0.59 (0.5 - 0.7)	t/yr	61% (53% - 68%)
Cadmium	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.26	t/yr	27% (22% - 31%)
Cadmium	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release	Dust, Vapor	Air	0.06	t/yr	6% (5% - 7%)
Cadmium	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.03 (<0.01 - 0.06)	t/yr	3% (<1% - 6%)
Cadmium	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.03 (<0.01 - 0.06)	t/yr	1% (<1% - 7%)
Cadmium	Buildings and Grounds	Residential Fuel Use, except Wood	Dispersal of dust following wear	Particulate matter, Fugitive dust	air	0.01	t/yr	<1%
Cadmium	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	<1%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Cadmium	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Cadmium	Industrial and Institutional Point Sources	Specialty Glass Manufacturer	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	<0.01	t/yr	<1%
Copper	**TOTAL**					**180 - 250 (120 - 390)**	**t/yr**	**100.0%**
Copper	Buildings and Grounds	Urban lawn & garden use of pesticides	Direct application to soil or vegetation	Solid, Liquid	Soil, Vegetation	1.1 - 73	t/yr	0.6% - 29% (0.3% - 38%)
Copper	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	39 (8.6 - 130)	t/yr	16% - 22% (4% - 45%)
Copper	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	37	t/yr	15% - 21% (10% - 31%)
Copper	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	27 (12 - 43)	t/yr	11% - 16% (3% - 29%)
Copper	Industrial and Institutional Point Sources	Army Base	Unknown release	Unknown form	Unknown	25	t/yr	10% - 14% (6% - 21%)
Copper	Pesticides and Wood Preservation	Antifouling paint - total	Leaching, Ablation	Solubilized in water, Particulate matter in water	Marine surface water, Marine sediment	23 (12 - 54)	t/yr	9% - 13% (3% - 34%)
Copper	Pesticides and Wood Preservation	Agricultural use of pesticides - total	Direct application to soil or vegetation	Solid, Liquid	Soil, Vegetation	10	t/yr	4% - 6% (2% - 8%)
Copper	Miscellaneous Material Use	Micronutrients	Direct application to soil	Solid, Liquid	Soil	5.4	t/yr	2% - 3% (1% - 5%)
Copper	Industrial and Institutional Point Sources	Naval Shipyard	Fugitive air release, Undefined release to surface water	Dust, Vapor, Undefined form released to surface water	Air, Surface water, Other	5.1	t/yr	2% - 3% (1% - 4%)
Copper	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	1.9 (0.02 - 5.4)	t/yr	<1% - 1% (<1% - 4%)
Copper	Pesticides and Wood Preservation	Aquatic-use algaecides in pools, fountains, spas, etc.	Direct application to water in contained pools (swimming pools, fountains, etc.)	Solid, Liquid	POTWs, Soils	1.5	t/yr	<1% - 1% (<1% - 1%)

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Copper	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	0.83	t/yr	<1%
Copper	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	0.44	t/yr	<1%
Copper	Industrial and Institutional Point Sources	Metal Foundries	Fugitive air release	Dust, Vapor	Air	0.22	t/yr	<1%
Copper	Pesticides and Wood Preservation	CCA-treated wood	Leaching	Solubilized in water	Soil, Surface water	0.06 (0.04 - 0.08)	t/yr	<1%
Lead	**TOTAL**					**520 (150 - 1,000)**	**t/yr**	**100%**
Lead	Outdoor Product Use	Ammunition, Hunting shot use	Intentional loss	Soilid metal	Soil, Surface water	370 (27 - 820)	t/yr	72% (13% - 87%)
Lead	Industrial and Institutional Point Sources	Army Base	Unknown release	Unknown form	Unknown	39	t/yr	8% (4% - 25%)
Lead	Outdoor Product Use	Fishing sinker loss	Unintentional loss	Soilid metal	Surface water, Aquatic sediment	36 (32 - 54)	t/yr	7% (3% - 31%)
Lead	Vehicles and Roads	Wheel weight loss	Unintentional loss	Solid metal	Impervious surfaces, Roadside areas	28 (20 - 29)	t/yr	5% (2% - 18%)
Lead	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	18 (15 - 20)	t/yr	3% (2% - 12%)
Lead	Non-Point Combustion Sources	Aviation fuel combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	16	t/yr	3% (2% - 10%)
Lead	Industrial and Institutional Point Sources	Various Industrial Facilities, not including pulp mills	Unknown release	Unknown form	Unknown	2.3	t/yr	<1% (<1% - 2%)
Lead	Industrial and Institutional Point Sources	Naval Shipyard	Unknown release	Unknown form	Unknown	1.8	t/yr	<1% (<1% - 1%)
Lead	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	2.6 (0.04 - 13)	t/yr	<1% (<1% - 8%)
Lead	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	1.2 (0.01 - 1.8)	t/yr	<1% (<1% - 1%)
Lead	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Water	0.66	t/yr	<1%
Lead	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Fugitive air release, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	0.53	t/yr	<1%
Lead	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	0.21 (0.2 - 0.9)	t/yr	<1%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Lead	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	0.04	t/yr	<1%
Lead	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	t/yr	<1%
Lead	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	<1%
Mercury	**TOTAL**					**0.54 (0.47 - 0.61)**	**t/yr**	**100%**
Mercury	Miscellaneous Material Use	Thermostat Disposal	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.13 (0.11 - 0.16)	t/yr	24% (20% - 31%)
Mercury	Miscellaneous Material Use	Fluorescent Lamp Disposal	Volatilization	Vapor	Air	0.10	t/yr	18% (16% - 20%)
Mercury	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.05	t/yr	9% (8% - 11%)
Mercury	Industrial and Institutional Point Sources	Crematoria Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05 (0.02 - 0.07)	t/yr	9% (4% - 12%)
Mercury	Industrial and Institutional Point Sources	Cement Plants	Volatilization, Combustion	Vapor, Aerosols, Particulate matter	Air	0.04	t/yr	8% (7% - 9%)
Mercury	Miscellaneous Material Use	Auto Convenience Switch Disposal	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.04 (0.02 - 0.06)	t/yr	7% (4% - 12%)
Mercury	Industrial and Institutional Point Sources	Petroleum Refineries	Volatilization, Combustion	Vapor, Aerosols, Particulate matter	Air	0.03	t/yr	6% (5% - 7%)
Mercury	Personal Care Products	Dental Amalgam Excretion	Human Excretion	Excrement	POTWs, Groundwater	0.02	t/yr	4% (3% - 4%)
Mercury	Miscellaneous Material Use	Button Cell Batteries	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	0.02	t/yr	4% (3% - 4%)
Mercury	Industrial and Institutional Point Sources	Steel Mills	Unknown release	Unknown form	Unknown	0.02	t/yr	3%
Mercury	Industrial and Institutional Point Sources	Residual Fuel Oil Combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	2%
Mercury	Personal Care Products	Dental Office Amalgam Waste	Wastewater	Liquid	POTWs, Groundwater	0.01	t/yr	2%
Mercury	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	t/yr	1%
Mercury	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Surface water	0.01	t/yr	1%
Mercury	Miscellaneous Material Use	Thermometers (Household)	Volatilization, Leaching, Washout	Vapor, Liquid, Particle-bound	Air, Groundwater, Soil	<0.01	t/yr	<1%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Mercury	Miscellaneous Material Use	Fertilizers	Direct application to soil	Solid, Liquid	Soil	<0.01	t/yr	<1%
Mercury	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	<0.01	t/yr	<1%
Mercury	Vehicles and Roads	Gasoline and Diesel Combustion	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Zinc	**TOTAL**					**1,500 (300 - 3,200)**	**t/yr**	**100%**
Zinc	Buildings and Grounds	Roofing materials - total	Leaching, Corrosion	Solubilized in water, Particulate matter in water	Surface water, POTWs, Groundwater	1,330 (210 - 2,800)	t/yr	87% (37% - 97%)
Zinc	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	82 (4.8 - 150)	t/yr	5% (<1% - 33%)
Zinc	Miscellaneous Material Use	Fertilizers and Micronutrients	Direct application to soil	Solid, Liquid	Soil	41	t/yr	3% (1% - 13%)
Zinc	Buildings and Grounds	Plumbing fixtures, pipes, and solder	Leaching	Solubilized in water	POTWs	30 (20 - 93)	t/yr	2% (<1% - 25%)
Zinc	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	12	t/yr	<1% (<1% - 4%)
Zinc	Vehicles and Roads	Motor oil leaks and improper disposal				7.9 (5.7 - 8.9)	t/yr	<1% (<1% - 3%)
Zinc	Vehicles and Roads	Brake pad wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	7.1 (0.22 - 44)	t/yr	<1% (<1% - 13%)
Zinc	Industrial and Institutional Point Sources	Steel Mills	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	4.3	t/yr	<1% (<1% - 1%)
Zinc	Industrial and Institutional Point Sources	Pulp and Paper Mills	Undefined release to surface water	Undefined form released to surface water	Surface water	3.7	t/yr	<1% (<1% - 1%)
Zinc	Industrial and Institutional Point Sources	Naval Shipyard	Fugitive air release	Dust, Vapor	Air	1.8	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Petroleum Refineries	Unknown release	Unknown form	Unknown	1.1	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Unknown	0.77	t/yr	<1%
Zinc	Industrial and Institutional Point Sources	Steel Galvanizers	Unknown release	Unknown form	Unknown	0.73	t/yr	<1%
PCBs	**TOTAL**					**2,100 (1,500 - 2,800)**	**kg/yr**	**100%**
PCBs	Miscellaneous Material Use	Large capacitors	Leakage	Liquid	Soil, Impervious surfaces	1,100	kg/yr	52% (40% - 75%)
PCBs	Miscellaneous Material Use	Small capacitors	Leakage	Liquid	Soil, Impervious surfaces	500 (1 - 1,000)	kg/yr	24% (<1% - 41%)

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PCBs	Buildings and Grounds	Residential Trash Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	280	kg/yr	13% (10% - 19%)
PCBs	Miscellaneous Material Use	Transformers	Leakage	Liquid	Soil, Impervious surfaces	130 (7 - 250)	kg/yr	6% (<1% - 15%)
PCBs	Buildings and Grounds	Sealants (Caulking)	Volatilization, Abrasion and fragmentation from weathering	Vapor, Sorption to dust particles	Air, Fugitive dust	110 (71 - 140)	kg/yr	5% (3% - 9%)
PBDEs	**TOTAL**					**680** **(220 - 2,300)**	**kg/yr**	**100%**
PBDEs	Buildings and Grounds	Indoor office space air	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	430 (120 - 750)	kg/yr	64% (7% - 88%)
PBDEs	Buildings and Grounds	Indoor residential dust	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	160 (100 - 320)	kg/yr	23% (5% - 72%)
PBDEs	Buildings and Grounds	Indoor office space dust	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	78 (<0.01 - 1,200)	kg/yr	12% (<1% - 84%)
PBDEs	Buildings and Grounds	Indoor residential air	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	9.5 (0.6 - 18)	kg/yr	1% (<1% - 8%)
PCDD/Fs	**TOTAL**					**9.4**	**g TEQ/yr**	**100%**
PCDD/Fs	Buildings and Grounds	Backyard Burn Barrels	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	7.3	g TEQ/yr	77%
PCDD/Fs	Vehicles and Roads	Heavy Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.81	g TEQ/yr	9%
PCDD/Fs	Industrial and Institutional Point Sources	Pulp and Paper Mills	Combustion, Undefined release to surface water	Aerosols, Vapor, Undefined form released to surface water	Air, Surface water	0.49	g TEQ/yr	5%
PCDD/Fs	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.38	g TEQ/yr	4%
PCDD/Fs	Non-Point Combustion Sources	Construction Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.18	g TEQ/yr	2%
PCDD/Fs	Vehicles and Roads	Light Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.08	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Cement Plants	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05	g TEQ/yr	<1%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PCDD/Fs	Vehicles and Roads	Light Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Industrial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.03	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Commercial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Petroleum Refineries	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Air, Surface water	0.02	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Recreational Boat Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Buildings and Grounds	Lawn and Garden Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Agricultural Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Logging Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Vehicles and Roads	Heavy Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Airport Service Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Non-Point Combustion Sources	Recreational Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	g TEQ/yr	<1%
PCDD/Fs	Industrial and Institutional Point Sources	Wood-Treatment Facility	Unknown release	Unknown form	Unknown	<0.01	g TEQ/yr	<1%
PAH	**TOTAL**					**310**	**t/yr**	**100%**
PAH	Buildings and Grounds	Woodstoves and Fireplaces	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	110	t/yr	34%
PAH	Pesticides and Wood Preservation	Creosote Treated Marine pilings - total	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Surface water, Air	54	t/yr	18%
PAH	Pesticides and Wood Preservation	Creosote Treated Railroad ties	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Soil, Air, Surface water	43	t/yr	14%
PAH	Vehicles and Roads	Light Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	29	t/yr	10%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Pesticides and Wood Preservation	Creosote Treated Utility poles	Leaching, Washout, Volatilization	Solubilized in water, Vapor	Soil, Air, Surface water	17	t/yr	6%
PAH	Vehicles and Roads	Heavy Duty Gasoline Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	11	t/yr	3%
PAH	Vehicles and Roads	Petroleum spills, leaks, and improper motor oil disposal	Leakage, Spillage, Direct release, Improper disposal	Liquid	Impervious surfaces, Soils, Stormwater, POTWs, Landfills	11	t/yr	3%
PAH	Buildings and Grounds	Residential Trash Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	6.5	t/yr	2%
PAH	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Combustion, Volatilization	Aerosols, Vapor, Particulate matter	Air	5.2	t/yr	2%
PAH	Buildings and Grounds	Lawn and Garden Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	5.0	t/yr	2%
PAH	Industrial and Institutional Point Sources	Pulp and Paper Mills	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	3.2	t/yr	1%
PAH	Industrial and Institutional Point Sources	Aluminum Mills	Volatilization, Fugitive air release, Combustion	Vapor, Dust, Aerosols, Particulate matter	Air	2.7	t/yr	<1%
PAH	Industrial and Institutional Point Sources	Petroleum Refineries	Fugitive air release, Volatilization, Combustion	Dust, Vapor, Aerosols, Particulate matter	Air	2.3	t/yr	<1%
PAH	Non-Point Combustion Sources	Commercial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	2.0	t/yr	<1%
PAH	Vehicles and Roads	Heavy Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	1.8	t/yr	<1%
PAH	Non-Point Combustion Sources	Construction Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	1.3	t/yr	<1%
PAH	Vehicles and Roads	Gas Station Emissions	Volatilization	Vapor	Air	1.2	t/yr	<1%
PAH	Vehicles and Roads	Tire wear	Abrasion	Particulate matter	Impervious surfaces, Roadside areas, Air, POTWs	0.98	t/yr	<1%
PAH	Non-Point Combustion Sources	Recreational Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.94	t/yr	<1%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Vehicles and Roads	Coal tar sealants	Leaching, Abrasion	Solubilized in water, Particulate matter in water	stormwater, fugitive air, dust	0.92 (0.17 - 1.7)	t/yr	<1%
PAH	Non-Point Combustion Sources	Recreational Boat Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.86	t/yr	<1%
PAH	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Unknown release	Unknown form	Air, Surface water, Other	0.58	t/yr	<1%
PAH	Buildings and Grounds	Roofing materials - total	Leaching	Particulate matter, Solubilized in water	Surface water, POTWs	0.57	t/yr	<1%
PAH	Non-Point Combustion Sources	Locomotive Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.49	t/yr	<1%
PAH	Non-Point Combustion Sources	Industrial Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.30	t/yr	<1%
PAH	Vehicles and Roads	Light Duty Diesel Vehicle Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.21	t/yr	<1%
PAH	Buildings and Grounds	Residential Yard Waste Burning	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.15	t/yr	<1%
PAH	Non-Point Combustion Sources	Logging Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.05	t/yr	<1%
PAH	Non-Point Combustion Sources	Agricultural Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	t/yr	<1%
PAH	Buildings and Grounds	Residential Fuel Use, except Wood	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.04	t/yr	<1%
PAH	Non-Point Combustion Sources	Cigarette smoke	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.03 (0.02 - 0.03)	t/yr	<1%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
PAH	Vehicles and Roads	Asphalt - total	Leaching, Abrasion	Solubilized in water, Particulate matter in water	stormwater, fugitive air, dust	0.02	t/yr	<1%
PAH	Non-Point Combustion Sources	Airport Service Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	0.02	t/yr	<1%
PAH	Non-Point Combustion Sources	Railroad Maintenance Equipment Emissions	Combustion emissions	Aerosols, Particulate matter, Vapor	Air	<0.01	t/yr	<1%
Phthalates	**TOTAL**					**34**	**t/yr**	**100%**
Phthalates	Personal Care Products	Fragrance	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	11	t/yr	32%
Phthalates	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	9.6	t/yr	28%
Phthalates	Vehicles and Roads	Car undercoating	Washout, Vapor	Liquid, Vapor	Surface water, Soil, Air	3.3	t/yr	10%
Phthalates	Miscellaneous Material Use	Lacquers and paint	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.9	t/yr	5%
Phthalates	Buildings and Grounds	PVC Coil coated roofing	Leaching, Volatilization	Liquid, Vapor	Surface water, POTWs, Air	1.5	t/yr	4%
Phthalates	Personal Care Products	Nail polish	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	1.4	t/yr	4%
Phthalates	Miscellaneous Material Use	PVC Coated fabric	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.2	t/yr	4%
Phthalates	Miscellaneous Material Use	Sealants, adhesives, etc.	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	1.1	t/yr	3%
Phthalates	Industrial and Institutional Point Sources	Plastics Manufacturer	Volatilization	Vapor	Air	0.86	t/yr	3%
Phthalates	Personal Care Products	Hair spray (aerosol and pump spray)	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.4	t/yr	1%
Phthalates	Miscellaneous Material Use	PVC Cables (outdoor, above ground)	Volatilization	Vapor, Sorption to soil	Air	0.35	t/yr	1%
Phthalates	Personal Care Products	Deodorant (solid)	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.29	t/yr	<1%
Phthalates	Miscellaneous Material Use	Shoe soles	Abrasion	Dust particles	Surface runoff, POTWs, Fugitive dust	0.2	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Wall coverings	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.14	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Roofing material	Leaching, Volatilization	Solubilized in water, Vapor	Surface water, POTWs, Air	0.14	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Flooring	Volatilization, Abrasion	Vapor, Dust particles	Air, Fugitive dust	0.1	t/yr	<1%
Phthalates	Miscellaneous Material Use	PVC Films, sheets, coated products	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.1	t/yr	<1%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Phthalates	Buildings and Grounds	PVC Hoses and profiles (outdoor)	Volatilization	Vapor	Air	0.09	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Cables (indoor)	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.08	t/yr	<1%
Phthalates	Buildings and Grounds	PVC Hoses and profiles (indoor)	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.08	t/yr	<1%
Phthalates	Miscellaneous Material Use	Printing inks	Washout, Volatilization	Liquid, Vapor	POTWs, Groundwater, Air	0.08	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Boat Manufacturer	Volatilization	Vapor	Air	0.05	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Chemicals Distribution	Volatilization	Vapor	Air	0.04	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Paint and Coatings Manufacturers	Volatilization	Vapor, Sorption to dust particles	Air, Fugitive dust	0.03	t/yr	<1%
Phthalates	Industrial and Institutional Point Sources	Other Industrial and Military Facilities	Volatilization	Vapor	Air	0.02	t/yr	<1%
Triclopyr	**TOTAL**					**150 (63 - 240)**	**t/yr**	**100%**
Triclopyr	Pesticides and Wood Preservation	Crop and Golf Course Use	Direct application to vegetation	Liquid	Vegetation and soils	150 (60 - 240)	t/yr	98% (95% - 99%)
Triclopyr	Pesticides and Wood Preservation	Forest Herbicide Use - State Forests	Direct application to vegetation	Liquid	Vegetation and soils	0.8 (0.4 - 1.2)	t/yr	<1% (<1% - 1%)
Triclopyr	Pesticides and Wood Preservation	Aquatic Weed Control	Direct application to surface water	Liquid or granular	Surface water	0.68	t/yr	<1% (<1% - 2%)
Triclopyr	Pesticides and Wood Preservation	Right-of-Way Maintenance--State Forests	Direct application to vegetation	Liquid	Vegetation and soils	0.5	t/yr	<1%
Triclopyr	Buildings and Grounds	Urban lawn & garden use of pesticides	Direct application to soil or vegetation	Liquid	Soil, Vegetation	0.43	t/yr	<1%
Triclopyr	Vehicles and Roads	Right-of-Way Maintenance - State Highways	Direct application to vegetation	Liquid	Vegetation and soils	0.3	t/yr	<1%
Triclopyr	Vehicles and Roads	Right-of-Way Maintenance - Railroads	Direct application to vegetation	Liquid	Vegetation and soils	0.1	t/yr	<1%
Nonylphenol	**TOTAL**					**0.18**	**t/yr**	**100%**
Nonylphenol	Industrial and Institutional Point Sources	Ind, Comm, Inst Sources (primarily Title V)	Unknown release	Unknown form	Air	0.18	t/yr	100%

Table Q-1 (Cont'd). Summary of Release Estimates for All COCs.

COC	Activity/Source Category	Specific Source	COC Release Mechanism	Form of Release	Initial Receiving Medium	Best Estimate of Release (Range)	Unit	Percent of Total (Range)
Petroleum	**TOTAL**					**9,300**	**t/yr**	**100%**
Petroleum	Vehicles and Roads	Motor oil drips and leaks	Leakage	Liquid	Impervious surfaces	6,100	t/yr	66%
Petroleum	Vehicles and Roads	Minor gasoline spills from fueling vehicles and non-road equipment	Spillage	Liquid	Impervious surfaces, Soils	1,900	t/yr	21%
Petroleum	Vehicles and Roads	Improper disposal of used oil following oil changes	Direct release, Improper disposal	Liquid	Stormwater, Soils, POTWs, Landfills	960	t/yr	10%
Petroleum	Miscellaneous Material Use	Petroleum spills (large)	Spillage	Liquid	Surface water, Soil, Impervious surfaces	228 (223 - 233)	t/yr	3% (2% - 3%)