TACOMA SMELTER PLUME SITE

CREDIBLE EVIDENCE REPORT:

The ASARCO Tacoma Smelter
and
Regional Soil Contamination in Puget Sound

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prepared by

Gregory L. Glass
Environmental Consultant
Seattle, Washington

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Tacoma-Pierce County Health Department
and
Washington State Department of Ecology

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1.0 INTRODUCTION

ASARCO's primary copper smelter in Ruston, Washington operated for almost 100 years. Final copper smelting occurred in 1985, with some processing of high-arsenic materials continuing into 1986.

While the Tacoma Smelter was still operating, it was included as part of the Commencement Bay Superfund Site. After smelter closure in 1986, studies of contamination both at the former smelter property and in surrounding, largely residential areas were performed under the Superfund program. The U.S. Environmental Protection Agency, Region 10 has issued Records of Decision (RODs) identifying required cleanup actions for the smelter site and areas within approximately one mile of the smelter. The remedial actions identified in these RODs have not yet been completed for either of the two designated uplands Operable Units.

Since 1999, the Washington State Department of Ecology, in cooperation with local health departments, has been investigating the much larger spatial extent of soil contamination associated with Tacoma Smelter emissions. Ecology has defined a Model Toxics Control Act (MTCA) site, based on the results to date, that includes several hundred square miles of King County and Pierce County within the central Puget Sound region. That site is called the Tacoma Smelter Plume Site. The boundaries of the site as currently defined are not considered to be final; additional investigations to extend sampling in King and Pierce Counties, as well as initial investigations in portions of Kitsap and Thurston Counties, are being considered by Ecology based on evaluations of the spatial patterns revealed so far. The Tacoma Smelter Plume Site includes areas more than 20 miles from the former smelter location in Ruston.

The spatial extent of the Tacoma Smelter Plume Site is defined by soil arsenic concentrations. The Tacoma Smelter has long been recognized as a major arsenic emissions source; for many years, it was the sole domestic supplier of arsenic for the United States. Smelter emissions were also a source for other metals (e.g., lead, cadmium, antimony, and mercury, among others), as noted in ASARCO's own reports and in various source tests conducted at the smelter. Information on soil concentrations of these additional emitted metals is useful for source determinations, even though those concentrations do not exceed unrestricted land use cleanup levels, as defined under MTCA, over most of the area included in the Tacoma Smelter Plume Site.

The results of the extensive soil sampling and analysis programs sponsored by Ecology since 1999 are reported elsewhere. This report presents and discusses evidence that Tacoma Smelter emissions were a primary source for the regional-scale soil contamination that defines the Tacoma Smelter Plume Site.

The body of this report consists of four main sections. Section 1.0 briefly describes

Tacoma Smelter operations and the chronology of numerous smelter-related studies, as well as the scope of the credible evidence report. Section 2.0 reviews the information sources used to support the credible evidence evaluations. Section 3.0 provides a short summary and overview of the multiple lines of reasoning that support identification of the Tacoma Smelter as a principal source. The individual rationales are then presented and discussed in more detail in Section 4.0. Section 5.0 presents the conclusions of the credible evidence analyses. An extensive list of references is included as Section 6.0.

1.1 TACOMA SMELTER OPERATIONS

The facilities and operations of the Tacoma Smelter are described in some detail in a number of previous documents (see PSAPCA 1981; see also PEDCo 1976; USEPA 1983a; USEPA 1984). Those references should be consulted to expand upon the brief descriptions provided here. Note that smelter operations, and particularly pollution control systems, changed over time.

The Tacoma Smelter began operation in 1890 as a lead smelter. It was purchased by ASARCO in 1905 and a process of conversion to copper smelting began. After the 1912 closure of ASARCO's Everett Smelter, where the first U.S. arsenic recovery plant had been constructed, arsenic recovery facilities were added to the Tacoma Smelter. By 1917 a tall stack was constructed and electrostatic precipitators were installed to reduce particulate emissions. The first sulfur control system was added by 1950 when a sulfuric acid plant came on line; that plant was later enlarged and a liquid SO2 plant was added, ultimately bringing sulfur control up to 45 percent. Particulate controls were improved in the late 1970s through installation of baghouses for the roaster and arsenic plant gas streams. Numerous additional steps were taken, especially after 1970, to reduce air emissions (see PSAPCA 1981).

The Tacoma Copper Smelter operated as a custom (toll) smelter that specialized in the processing of complex ores - those high in arsenic and other non-copper metals. It was the only domestic smelter of its type. Ore concentrates (copper sulfides) and other feed materials were delivered to the smelter from many U.S. and foreign sources. Those sources varied over time. Materials from different sources were commonly blended to achieve a more nearly constant metallurgy for the smelting process; however, smelter feed characteristics probably varied over the smelter's operating history. Annual smelter throughput and copper production varied with economic conditions, as well as in response to labor strikes and the effects of meteorological curtailment practices after 1970. Production declined markedly after 1970 (see PSAPCA 1981), corresponding to the period when studies of smelter impacts were conducted.

A traditional roaster-reverb pyrometallurgical smelting process was used at the Tacoma Smelter from 1930 through closure. Copper production included five main steps. Concentrates were first roasted to remove some of the sulfur and preheat the charge to the reverberatory furnace. The calcines produced by roasting were then melted in a continuously fired reverberatory furnace, where a copper matte separated from a lighter slag layer. Slag was tapped and taken to a slag dumping area where it was spray-cooled. The copper matte was oxidized and enriched in converter furnaces, run as batch operations over 10 to 12 hours. The resulting blister copper (98 to 99 percent copper) was then sent to anode casting furnaces where it was formed into plates for refining. The final step was electrolytic refining, where high-purity copper was produced and gold, silver, and other valuable trace elements were recovered as additional byproducts. The Tacoma Smelter's refinery operations were closed in 1979, after which copper anodes were shipped by rail to ASARCO's Amarillo, Texas refinery for final processing.

The elements contained in concentrates and other feedstocks processed at the Tacoma Smelter thus ended up in one of the following materials streams: copper anodes or refined copper; slag; sulfuric acid or liquid SO2; recovered arsenic trioxide and metallic arsenic; valuable trace elements recovered in refining operations; captured dusts (some high-lead dusts from treatment of converter gas streams were sent to ASARCO's East Helena, Montana lead smelter); or as air emissions. Many of the collected dusts from particulate control systems at the smelter were recycled to one of the in-plant smelting operations (i.e., as revert materials) or to the arsenic plant. (For an example of a materials balance for arsenic, see USEPA 1983a).

Air emissions occurred at the Tacoma Smelter as both tall stack and low-level, or fugitive, emissions. The 565-foot main stack, constructed by 1917, was intended to release collected gas streams (and entrained particulates) from various smelter operations at a higher elevation where they could be better dispersed in ambient air, thus reducing the concentrations of ground-level air contaminants. Tall stacks do not reduce the mass of contaminant emissions; their use of dispersion to improve ground-level air quality is accompanied by a wider distribution of contaminants. During the 1970s ASARCO contemplated construction of a "superstack" over 1,000 feet tall, but it was never built. Fugitive emissions arose from a number of materials handling and smelting processes (see USEPA 1983a for a list of potential fugitive sources). The elevations for releases of fugitive emissions ranged from ground level (e.g., slag dumping) to the top of buildings such as the converter building, where emissions from the converter furnaces were uncontrolled during "roll-out" charging and skimming operations.

Over time pollution control systems were installed for all of the major gas streams from smelting operations; many "housekeeping" measures and modifications to smelter facilities were also undertaken to reduce fugitive emissions. Particulate control of the main gas streams was accomplished using electrostatic precipitators and baghouses, as well as a wet-scrubbing system for treating converter off-gases prior to sulfur recovery. The sulfuric acid plant and liquid SO2 plant provided partial sulfur control, as already noted. To further reduce ambient SOx concentrations in the communities surrounding the Tacoma Smelter, a Meteorological Curtailment Program was started in 1970 and upgraded over time. That intermittent control program would suspend or curtail certain smelting operations when forecast meteorological conditions and process emissions were expected to result in a violation of ambient SOx standards. The Meteorological Curtailment Program resulted in an increasing number of hours of operational curtailment over time as it was upgraded to be more effective. By 1979 there were 3,605 hours of total or partial production curtailment (PSAPCA 1981). Meteorological curtailment occurred primarily when winds were calm or from the smelter toward the Ruston and North Tacoma neighborhoods. One effect of the Meteorological Curtailment Program was thus to increase the proportion of smelter emissions that occurred during northerly and northeasterly winds and the relative impacts to downwind areas in those directions.

The primary impacts of fugitive emissions were limited to areas close to the smelter (within one, or at most a few, miles). Those close-in areas could also be affected by upset conditions (e.g., releases from fallout from stack fires) during smelter operations, as well as fumigation of the tall stack plume (see Lutrick 1971). The effects of smelter operations at greater distances and on a regional scale, to the extent they occurred, were attributable to tall stack emissions, dispersion of the stack plume, and wet and dry deposition (including ground-level plume impact) of contaminants.

1.2 CHRONOLOGY OF STUDIES

Concerns about smelter impacts on human health, livestock, agricultural and forest productivity, and environmental contamination arose soon after the establishment of large-scale smelting industries in the United States. The "smelter smoke" problem led to numerous lawsuits and several extensive scientific studies (see, for example, Quinn 1989; Holmes et al. 1915; MacMillan 1973). After resolution of the "smelter smoke" lawsuits in the first decades of the twentieth century, relatively little work on smelter impacts appears to have been done until the rise of environmental concerns in the late 1960s.

The modern era of smelter studies began with the adoption of air pollution legislation and regulations and documentation of high exposures and body burdens, particularly in young children, in communities surrounding smelters. The Puget Sound Air Pollution Control Agency (PSAPCA; now the Puget Sound Clean Air Agency) was created by the Washington State Legislature in 1967 to regulate air pollution sources in a four-county region of central Puget Sound, including Pierce County, where the Tacoma Smelter was located. The first PSAPCA order to ASARCO to comply with the agency's adopted regulations for SOx controls was issued in 1968. (For a chronology through 1981 of PSAPCA efforts to control Tacoma Smelter air emissions, see PSAPCA 1981, Draft EIS, Appendix I). PSAPCA retained the role of lead agency for Tacoma Smelter air pollution issues through smelter closure in 1986.

In the early 1970s, monitoring of blood lead levels in young children living close to ASARCO's El Paso, Texas lead smelter showed significant community exposures were occurring (Landrigan et al. 1975). This led to a national monitoring survey of communities near U.S. nonferrous smelters (Baker et al. 1977). Newspaper accounts of the El Paso findings in 1972 alerted the Washington State Department of Social and Health Services (DSHS), which conducted urinary and hair arsenic analyses on elementary school children in Ruston, where the Tacoma Smelter was located, before the nationwide survey was organized. The results showed significantly elevated levels compared to a control population (Milham and Strong 1974). These findings led PSAPCA to focus on arsenic emissions as well as criteria pollutants such as total particulates and SOx.

Based on these early findings, both ASARCO and DSHS embarked on an extended series of community studies of urinary arsenic levels to evaluate ongoing community exposures. The cumulative data set for urinary arsenic results from both ASARCO and DSHS studies was compiled and evaluated by DSHS (see Wicklund and Harter 1984; see also PSAPCA 1981). Both PSAPCA and ASARCO performed ambient air monitoring at extended networks of monitoring stations; these monitoring networks continued to collect data for some time even after smelter closure. PSAPCA, in cooperation with USEPA, continued to identify and quantify smelter emissions sources and attempted to bring the smelter into compliance with air pollution regulations. In addition to PSAPCA's regulations, USEPA rulemaking on other related aspects of smelter air pollution controls continued - for example, with respect to nonferrous smelter orders (NSOs), national emissions standards for hazardous air pollutants (NESHAPs), including arsenic, and rules for the use of supplemental control systems such as the Tacoma Smelter's Meteorological Curtailment Program. Studies and data collection related to smelter air impacts and community arsenic exposures were thus more or less continuous.

Broader studies of environmental impacts from Tacoma Smelter operations, including soils investigations, tended to cluster into specific time periods, both during the era of smelter operations and after smelter closure. A brief and generalized overview of the chronology of studies related to the Tacoma Smelter is provided in the following paragraphs.

The Tacoma Smelter required variances from PSAPCA to continue to operate out of compliance with air pollution regulations. Numerous studies were performed during the period when PSAPCA considered variance applications from ASARCO, which extended through completion of a 1981 Environmental Impact Statement on continued smelter operations (PSAPCA 1981). Those studies included various environmental media in both uplands and marine (Puget Sound) areas surrounding the smelter.

PSAPCA granted ASARCO five-year variances for continued operation in 1981, but with a strict schedule for compliance establishing interim performance and reporting milestones. The pace of environmental studies decreased as attention was focused on activities related to that compliance schedule. Studies continued on a few topics of interest - for example, related to slag impacts, precipitation chemistry, and acid precipitation impacts. The next period of increased study activity occurred as a result of EPA's designation of the Commencement Bay Superfund Site, which included the Tacoma Smelter. An interagency Superfund Air Work Group was organized in 1983 to consider the application of Superfund to the operating Tacoma Smelter. That Air Work Group identified a list of studies that should be performed to provide additional information (e.g., additional ambient air monitoring for metals; an evaluation of re-entrainment of contaminated dusts; lab analyses of archived soil samples to support time trend analyses of soil contamination; arsenic speciation studies; an evaluation of soil metals mobility; a survey of drinking water sources; a survey of metals in garden soils; source apportionment modeling). Numerous studies were completed with the support of the member agencies. This period of initial Air Work Group studies also coincided with EPA's NESHAPS rulemaking for arsenic - which produced ambient air, exposure, and health risk modeling studies - as well as with an independent bee biomonitoring project (Bromenshenk et al. 1985).

The Interagency Air Work Group recommended that a large-scale "integrated" study be performed to determine which exposure pathway(s) contributed to the observed elevated urinary arsenic levels in children living near the smelter. The purpose of the study was to determine what types of actions (e.g., reducing ambient air arsenic concentrations versus capping or removing contaminated soils) would reduce those urinary arsenic levels. Funding for the study was obtained from the Agency for Toxic Substances and Disease Registry, Centers for Disease Control, with additional contributions from Ecology. The study was performed by a team of researchers from the University of Washington, resulting in a final Exposure Pathways Report (Polissar et al. 1987; see also Kalman et al. 1990 and Polissar et al. 1990). Data for this study were collected during 1985 and 1986, just as the Tacoma Smelter was being closed.

Smelter closure was recognized by several researchers as creating an opportunity for "natural experiments" in which measurements with and without smelter emissions could be made and compared. Another University of Washington research team received funding from EPA, Region 10 to perform an extensive smelter closure study (Vong et al. 1986). Other independent studies related to long-range transport and acid precipitation were also performed at the time of smelter closure. Continued air quality monitoring by ASARCO and PSAPCA documented the decline in ambient air contamination after closure.

The focus of Superfund attention regarding the communities surrounding the Tacoma Smelter shifted to the residual contamination of soils after smelter closure. Ecology and EPA conducted sampling and analysis studies of soil contamination beginning in 1987, culminating in EPA's Remedial Investigation Report (Bechtel Environmental, Inc. 1992) and Record of Decision (ROD) for soil cleanup actions in Ruston and North Tacoma. As ASARCO undertook the cleanup actions required by the ROD, detailed property-by-property sampling results became available for a large number of properties in the area within approximately one mile of the smelter. A database of those property-by-property sampling results is maintained at the Tacoma-Pierce County Health Department. Additional sampling was also performed on the smelter property as part of the Superfund investigations of that operable unit (see Parametrix, Inc. 1989).

Interest in the extent of contamination beyond EPA's Ruston/North Tacoma cleanup area was rekindled by the results of two independent studies of contamination in relatively undisturbed soils at distances of 5 to 7 miles northeast and south-southwest of the smelter. Significantly elevated soil arsenic concentrations were found in both studies (City of Tacoma and Glass 1999 [see also Golding 2001]; King County DDES 1999). Ecology initiated a series of soil investigations in 1999 that led to defining the Tacoma Smelter Plume Site under MTCA. These ongoing soil investigations of regional-scale contamination are discussed further in

Section 2.2.

Individual studies related to the Tacoma Smelter are identified and discussed in this report only as necessary for the credible evidence evaluations. Several earlier reports provide listings and chronologies of environmental and exposure monitoring studies through 1992 (see PSAPCA 1974a; PSAPCA 1981; Lowry et al. [undated, 1983?]; Glass and Lowry 1983; Lowry and Hughes 1984; Bechtel Environmental, Inc. 1992).

1.3 SCOPE OF CREDIBLE EVIDENCE REPORT

The credible evidence evaluations consider the cumulative information available from all studies related to the Tacoma Smelter, including historic studies. The extensive soil sampling conducted in recent Tacoma Smelter Plume Site investigations (see Section 2.2) provides information on arsenic and lead concentrations over an area of approximately 450 square miles in central Puget Sound. The analyses in this report are based on the complete "footprint study" results for that 450 square mile region, as well as additional trace element results for the King County portion of the region (including Vashon-Maury Island and the King County Mainland). The complete results of the Vashon-Maury Island Child-Use Areas study are also available. Still in progress are three additional investigations: extensive sampling of child-use properties on the King County Mainland and in Pierce County, and the analysis of selected samples from the Pierce County footprint study for additional trace elements. Results for all three of those additional investigations should be available by the end of 2003.

Data from the footprint studies are believed to be more relevant than the results from child-use areas for the credible evidence analyses. The footprint studies provide better spatial coverage and focus primarily on relatively undisturbed forest soils where impacts from miscellaneous local sources are minimized, enhancing data interpretations. Sampling in relatively undisturbed forest soils has also been found to reflect general upper bounds for smelter-related soil contamination in comparison to more disturbed soils on developed properties. Preliminary (unvalidated) results for approximately 20 percent of the Pierce County trace element samples have also been reviewed and appear highly consistent with the King County trace element results. Nevertheless, it should be noted that the evaluations in this report are based on a point in time, and that additional results will become available in the near term that may be used to supplement the credible evidence analyses.

Soil sampling and analysis results have documented regional-scale contamination by arsenic and other elements over an area of hundreds of square miles. The credible evidence evaluations focus on that regional-scale impact. It is not within the scope of this report to evaluate and assign or apportion sources for the results of each individual sample result, or small local subregions. The definition of the Tacoma Smelter Plume Site has been based on the aggregate database and overall spatial pattern of soil concentrations, not on individual results.

The credible evidence evaluations also consider multiple types of information (see Section 4.1) in assessing the role of the Tacoma Smelter as a source for the documented regional-scale contamination. The scope for the analyses is therefore not limited to soil sampling and analysis results. Spatial, temporal, and chemical patterns in various environmental media, as well as information on Tacoma Smelter operations, are used in the analyses. This broader scope for source evaluations supports multiple rationales whose consistency and completeness are important factors in reaching conclusions.

Analyses of additional trace elements for selected soil samples were included in the Tacoma Smelter Plume investigations specifically to support source evaluations. The extensive set of study results available for this report does not, however, include all possible approaches to source evaluations. Other study designs - for example, statistical source apportionment studies, isotopic composition studies, tree ring sampling, or peat bog or lake sediment sampling and depth profiling - were considered and have been used in studies of impacts at other smelters. It should therefore be noted that this report, while considering all <u>available</u> types of information relevant to the Tacoma Smelter, does not include all <u>possible</u> types of relevant information.

Finally, the credible evidence analyses consider the possible impacts on a regional scale of other potential sources, including both air emissions sources and widespread land application practices. The Tacoma Smelter is thus not considered in isolation when evaluating the patterns and magnitude of documented soil contamination in the central Puget Sound region.

2.0 INFORMATION SOURCES

A large body of information related to Tacoma Smelter operations and impacts was compiled and used to support the credible evidence evaluations. In this report, the term "historic smelter studies" is used to refer to all investigations and reports that predate Ecology's Tacoma Smelter Plume studies, which started in 1999. Many of the historic smelter studies had been assembled and reviewed for previous milestone reports on the Tacoma Smelter (e.g., PSAPCA 1974a, PSAPCA 1981, Black & Veatch 1988a, and Bechtel Environmental, Inc. 1992; see Section 1.2). Besides environmental monitoring studies, historic smelter studies include all available reports from emissions source tests at the Tacoma Smelter, emissions estimates, and other data related to smelter operations through closure in 1986.

A large number of historic smelter studies was reviewed. In addition, various investigations sponsored by Ecology and the local health departments in King County and Pierce County as part of the Tacoma Smelter Plume studies were also evaluated:

- The magnitude and extent of regional soil contamination were studied in a phased series of six investigations. Those soil studies included extensive sampling in both relatively undisturbed forest soils and at child-use areas (e.g., schools, parks, and childcare centers). The primary contaminants of interest were arsenic and lead, but the soil investigations also included analyses of numerous samples for multiple additional trace elements.

- A literature search was performed to identify and obtain reports on environmental impacts at other smelters, worldwide. This literature database provides a highly relevant context for interpreting the results of Tacoma Smelter studies.

- Finally, Historical Research Associates, Inc. of Seattle, Washington was retained to perform a historical review study for possible sources of arsenic with regional-scale impacts. The resulting study was supplemented by an examination of various uses for arsenic and additional review of some specific potential sources.

Some of the compiled information used in the credible evidence evaluations represents cumulative impacts over a time period including all years of smelter operations. For example, soils and sediments are believed to be long-term repositories for deposition from air emissions sources. It should be noted, however, that many of the types of information represented in the compiled database for this report reflect conditions only at the time of the study in question - for example, source test information and ambient air and precipitation chemistry monitoring data. The available studies are almost exclusively from the period of 1970 and later. Smelter production levels may have been higher, and pollution control systems were almost certainly less extensive and less effective, in the years before 1970 that are not represented in the available studies. Therefore, the available database may in some respects be unrepresentative of the full history of smelter emissions and environmental impacts.

Each of the basic information sources used for this report is briefly described in the following subsections. Information from more than one of the identified information sources is often linked to support a rationale for source identification (see Section 4.0). That is, the presentation of the credible evidence analyses is focused on types of reasoning much more than on individual studies or data types.

2.1 HISTORIC TACOMA SMELTER STUDIES

2.1.1 Smelter Operations

A characterization of Tacoma Smelter operations, for the purpose of understanding the nature and magnitude of air emissions, could include (among other information) the following: analysis of feedstock elemental composition; source tests for specific identified emissions sources and smelting operations; evaluations of the effectiveness of pollution control systems, including upset conditions; elemental mass balances for the facility as a whole; and an overall air emissions inventory. The inventory could provide information on elemental emissions by particle size range, by year of operating history, distinguishing between tall stack and various low-level, fugitive emissions sources.

Specific information of this type for the years before the creation of the Puget Sound Air Pollution Control Agency (PSAPCA) is all but absent. (See Varner 1983 for one existing estimate from ASARCO of possible pre-PSAPCA emissions levels). During the period of smelter operations when PSAPCA was the primary regulatory agency responsible for air pollution monitoring and control (1967 through 1986), ASARCO had substantial reporting requirements to PSAPCA related to smelter operations and emissions. PSAPCA also initiated or coordinated with EPA on many source tests of smelter operations, compiling the resulting source test reports in its files.

Some years after the closure of the Tacoma Smelter, when it was no longer an operating source subject to ordinary PSAPCA regulation (other than for windblown dusts or releases during remedial activities), PSAPCA destroyed all of its accumulated records related to the smelter (Nehen 2002). However, many of the relevant records and source tests from PSAPCA files had previously been obtained as part of earlier smelter studies (e.g., PSAPCA 1981 and Bechtel Environmental, Inc. 1992). The last files review at PSAPCA was conducted some years after smelter closure. It is possible that additional information no longer available at PSAPCA exists within EPA files or ASARCO files. A keyword database search of EPA's library holdings was performed as part of the credible evidence data compilation, which largely confirmed the list of studies already in hand. The Administrative Record and site file at EPA Region 10 for the Tacoma Smelter site (including the Tacoma Smelter and Ruston/North Tacoma Operable Units) was not reviewed. Other reports may exist at other EPA offices (e.g., the Office of Air Quality Planning and Standards). No records request has been made to ASARCO to identify and obtain additional information on smelter operations.

While undoubtedly incomplete in many respects, a set of source test reports and emission estimates was compiled for the credible evidence evaluations. Those references are listed in Table 1 (for complete reference information, see Section 6.0). Table 1 includes a brief comments section identifying the type of information provided in each referenced report.

2.1.2 Environmental Studies

A general chronology of historic environmental studies of the Tacoma Smelter is provided in Section 1.2. Most of those studies had already been compiled for earlier reports on Tacoma Smelter impacts (e.g., PSAPCA 1981; Bechtel Environmental Inc. 1992) and were therefore available from various project files. A few additional reports were identified and obtained during the period of Tacoma Smelter Plume studies. Historic environmental studies include those performed by regulatory agencies, resource agencies, ASARCO, students and academic researchers, municipalities (e.g., in connection with land use or property cleanup actions), and others.

A substantial number of soil investigations is included among the historic environmental studies. Those soil investigations included sampling at diverse types of properties, including: forests; roadsides and road dusts; home gardens; urban and suburban lawns; schools, parks, and playgrounds; and vacant lots. Many of the earliest soil studies had only a small number of samples and limited spatial coverage. Later studies included somewhat larger data sets and/or spatial coverage (e.g., Heilman and Ekuan 1977; Lowry 1983; the USEPA Urban Soils Monitoring Program repeated studies of Pierce County soils; Polissar et al 1987; Black & Veatch 1988a; Bechtel Environmental Inc. 1992). All of the previous soils data were reviewed as part of the study design process for Tacoma Smelter Plume soils investigations (see Section 2.2). Lists of historic soils studies are included in those study design reports. The diverse sampling designs and limited scopes of these historic soils studies make them less important than the results of the Tacoma Smelter Plume studies for source identification. (See Chirenje et al. 2002 for a useful discussion of the relevance of land use for soil sampling and data interpretation, an issue considered at length in designing Tacoma Smelter Plume studies).

Historic environmental studies also include numerous investigations of other media, such as ambient air, precipitation chemistry, collected dustfall, garden vegetables, non-garden vegetation (e.g., grass and tree leaves and needles), bee biomonitoring, marine and aquatic water quality, marine and freshwater sediments, marine biota, drinking water and ground water, house dusts, and urinary, blood, and hair contaminant levels. Most historic studies focus on SOx, arsenic, lead, and cadmium as smelter-related contaminants of concern; occasional studies include analyses for additional metals. As already noted, lists of these historic studies are available in existing reports (see especially PSAPCA 1981 and Bechtel Environmental Inc.

1992) and individual historic studies are included in the Section 6.0 references list only if they are specifically discussed in the credible evidence evaluations.

2.2 TACOMA SMELTER PLUME SITE STUDIES

Ecology and the local health departments adopted a systematic, phased approach to soil investigations for the Tacoma Smelter Plume Site beginning in 1999. Two sequential study phases, each conducted in three geographic regions, resulted in six separate soils studies. In each geographic region, initial "footprint" sampling was followed by targeted sampling of child-use areas where exposures to young children were of greatest concern. The three "footprint" studies are described in Section 2.2.1. The three child-use area studies are described in section 2.2.2. Additional trace element analyses were performed for selected samples from all three geographic regions, to support evaluations of sources for the documented soil contamination. Those trace element studies are described in Section 2.2.3.

2.2.1 Footprint Studies

The first phase of Tacoma Smelter Plume Site soils investigations consisted of "footprint" sampling to determine the regional-scale magnitude and extent of soil contamination. Footprint studies focused on sampling in relatively undisturbed forest soils. Footprint studies were performed for three geographic regions in the following sequence: all of Vashon-Maury Island (with some exploratory investigations added along the King County Mainland shoreline); an area of several hundred square miles on the King County Mainland, roughly northeast of the Tacoma Smelter; and an area of several hundred square miles in northwestern and western Pierce County.

The study designs for all three footprint studies were similar, but they differed in some details. Elements of the study design that varied included metals analyzed (cadmium was included for many of the Vashon-Maury Island samples); depth intervals sampled; archiving of some samples without chemical analysis; the frequency and layout of soil boring clusters (within sampling locations) to provide data on local variability; and sampling density, determined in part by the availability of forested properties to sample. The Pierce County footprint study also included numerous samples from residential properties in the urbanized areas nearest the Tacoma Smelter, because forested properties were all but absent in that area. Residential soils were included to provide spatial coverage of the defined study area for Pierce County, recognizing that they are more disturbed than forest soils and that soil contamination patterns will reflect those differences. Arsenic and lead were the two primary contaminants of concern analyzed in all processed samples in all three studies.

The database produced from each study is briefly described by the number of sampling locations and the total number of soil results (not including QA/QC sample analyses such as field duplicates, rinsate analyses, or requested reanalyses), as follows:

- <u>Vashon-Maury Island</u> (see PHSKC and Glass 2000): 177 sampling locations and 417 total samples analyzed. Those totals include 16 King County Mainland shoreline locations with 48 sample results. The data set for Vashon-Maury Island alone therefore includes 161 sampling locations and 369 sample results (covering approximately 40 square miles).

- <u>King County Mainland</u> (see Washington State Department of Ecology 2002): 59 additional sampling locations and 576 additional samples analyzed. Including the exploratory mainland sampling performed in the initial Vashon-Maury Island study, the total data set for the mainland includes 75 sampling locations and 624 sample results (covering approximately 200 square miles).

- <u>Pierce County</u> (see TPCHD 2003; report in progress): 194 sampling locations and 1,409 total samples analyzed. The density of sampling was increased in residential areas where only comparatively disturbed properties were available for sampling, in the expectation that spatial variability was increased and greater sampling density was required to define spatial patterns and upper-bound soil contamination levels. The total of 194 sampling locations included 105 developed residential properties and 89 forest locations (covering approximately 200 square miles). The numbers of disturbed and undisturbed samples were 974 and 435, respectively.

The combined data set from all three footprint studies provides arsenic and lead results for 430 sampling locations and 2,402 samples.

2.2.2 Child-Use Area Studies

The second phase of Tacoma Smelter Plume Site soils investigations consisted of sampling at targeted child-use areas such as schools, preschools, parks, camps, and childcare centers. These properties reflect varying degrees of soil disturbance (e.g., grading, excavation or importation of soils, mixing/tilling, and amending), but as a group they represent greater disturbance than in the comparatively undisturbed forest soils sampled in the footprint studies. In each of the three geographic regions, the footprint study was completed before child-use areas were sampled. The Vashon-Maury Island Child-Use Areas study was performed first (and in the overall sequence of studies it preceded the King County and Pierce County footprint studies). The child-use areas sampling in King County and Pierce County was performed in two concurrent studies with shared study design (see Washington State Department of Ecology et al. 2002).

Sampling at child-use areas followed a consistent approach involving arsenic and lead analyses of soils collected in "decision units" defined by children's activities and property characteristics. The first study on Vashon-Maury Island differed in some details from the two later studies. It attempted to survey all identified child-use areas; the King County and Pierce County studies, in areas of much greater population and development density, determined a maximum number of child-use areas that could be sampled and used a prioritization system based on several characteristics of child-use areas to select properties to be studied. While all areas of Vashon-Maury Island were included in the child-use areas sampling, only those portions of the King County and Pierce County footprint study regions that were judged likely to have soil contaminant levels above Ecology's selected Interim Action trigger levels for arsenic and lead were included for child-use property sampling. The ability to obtain access agreements affected the final list of child-use areas sampled in all three studies.

The total depth and number of intervals sampled were also greater in the Vashon-Maury Island study, and were varied by type of child-use area and location. That initial study of child-use areas was designed to provide additional characterization information supporting comparative evaluations of soil contamination between undisturbed and disturbed properties. The two later studies focused on Interim Action decisions and did not have as an objective further comparisons of contamination patterns between disturbed and undisturbed soils. Beaches were also sampled only in the Vashon-Maury Island study. All beach samples had low arsenic and lead concentrations, and sampling of beaches was therefore dropped for later studies.

The database produced from each study is briefly described by the number of child-use areas sampled and the total number of soil results (not including QA/QC sample analyses), as follows:

- <u>Vashon-Maury Island</u> (see PHSKC and Glass 2001): 34 child-use areas and 1,503 total samples analyzed. This study included the collection and analysis of additional samples to support the first trace element study (see Section 2.2.3 below) and the comparative evaluation of soil contamination at disturbed versus undisturbed areas. Four forested areas adjacent to child-use areas were sampled, as "forest fringe" areas; that resulted in 80 additional samples analyzed. Four of the locations with higher contaminant concentrations from the Vashon-Maury Island footprint study were also resampled and analyzed only for arsenic, to ensure that a wide range of arsenic concentrations would be available for the trace element study. Resampling produced an additional 36 sample results. One of the Vashon-Maury Island child-use areas was also resampled using a different sampling design (including composited samples) to provide confirmation data as Ecology's Interim Action levels were first being applied within the Tacoma Smelter Plume Site.

- <u>King County Mainland</u> (report in progress): 97 child-use areas and 2,544 total samples analyzed. All samples have been collected and analyzed.

- <u>Pierce County</u> (report in progress): to date, 55 child-use areas and 1,091 total samples analyzed. The Pierce County study is nearing completion. Access agreements are still being discussed for a few additional properties. Final samples are expected to be collected by the end of Summer 2003.

The combined data set from all three child-use area studies will provide results for at least 186 properties and 5,138 samples (not including the Vashon-Maury Island forest fringe and footprint location resamples).

2.2.3 Trace Element Studies

Analyses of additional trace elements were performed for selected samples from all three geographic regions, to support source evaluations. Trace element analyses were not added to the overall Tacoma Smelter Plume Site program until the Vashon-Maury Island footprint study had been completed and all archived soil samples discarded. This additional study component was therefore incorporated into the next study in the overall sequence of studies - the Vashon-Maury Island Child-Use Areas study. Trace element analyses were subsequently incorporated into the King County Mainland and Pierce County footprint studies.

The basic design for trace element analyses included two study phases for each of the three geographic regions: 1) a small number of samples was first analyzed for a large number of elements (approximately 35) to identify those elements that appeared to be good tracers of smelter impacts; 2) a small number of targeted trace elements (3 or 4) was then analyzed for the large remaining set of selected trace element samples. Selection criteria were developed and applied to the selection of samples for both study phases (e.g., good spatial coverage, a broad range of arsenic concentrations, and varying lead-to-arsenic ratios).

The samples selected for trace element analyses from Vashon-Maury Island and from Pierce County included both disturbed and undisturbed sampling locations; all samples selected from the King County Mainland were from undisturbed locations.

The database for trace element results associated with each geographic region is briefly described by the number of sampling locations represented and the total number of sample results (not including QA/QC sample analyses), as follows:

- Vashon-Maury Island (see Glass 2003): 27 sampling locations and 166 samples selected from the child-use areas study, including the forest fringe and footprint location resamples. Undisturbed samples numbered 41, from 8 locations; disturbed samples (from various types of child-use areas) numbered 125, from 22 locations. (The sum of undisturbed and disturbed locations is 30 because 3 locations contributed both undisturbed and disturbed samples).

- King County Mainland (see Glass 2003): 33 sampling locations and 68 total samples selected from the footprint study. This set of Tacoma Smelter Plume Site soil samples was supplemented by 8 additional soil samples from two eastern

Washington locations that were formerly orchards. The analysis of this additional type of samples with arsenic and lead contamination (from agricultural use of lead arsenate pesticides) provided an opportunity to determine how well trace element analyses could discriminate between Tacoma Smelter and orchard sources.

o Pierce County (sample analyses in progress): 82 sampling locations and 173 samples selected from the footprint study samples. Undisturbed samples numbered 85, from 40 locations; disturbed samples (from residential properties) numbered 88, from 42 locations. Preliminary results (prior to final validation) have been reported for an expanded initial set of samples (n=34) analyzed for an extended list of ten elements. Analyses of the remaining samples (n=139) for three selected primary trace elements should be completed and validated before the end of 2003.

The total trace element analysis data set for all three geographic regions will include 407 selected samples from 142 locations, covering almost 450 square miles. The initial trace element report that evaluates all results from Vashon-Maury Island and the King County Mainland includes results for 234 samples from 60 locations, covering approximately 250 square miles (Glass 2003). The primary targeted trace elements for analyses of samples for all three geographic regions were antimony (Sb), bismuth (Bi), and indium (In).

2.3 LITERATURE REVIEW: OTHER SMELTERS

The Tacoma Smelter was one of many metals smelters in the world. Individual smelters differ in the chemical characteristics of their feedstocks, the technical design of smelting processes, pollution control systems design and effectiveness, height of emissions releases (stack heights), location with respect to local topography and wind fields, and annual and cumulative production levels - all factors that can affect the magnitude and extent of environmental impacts from smelter operations. Those differences notwithstanding, smelters as a group have been identified as among the most important air emissions sources (see, for example, Nriagu 1984; Nriagu 1991; Chow 1995; Dudka and Adriano 1997).

A literature review was performed to identify and obtain copies of studies of environmental contamination around other smelters worldwide. The objective of this effort was to document the basic patterns of environmental contamination around smelters whose operations were in many respects similar to those of the Tacoma Smelter. The consistency in certain features of that contamination - for example, the distances to which contamination was observed, the relationship to wind rose patterns, and the association among multiple elements related to smelter feedstocks and emissions - was of particular interest. Unlike the Tacoma Smelter, which was located in a highly developed urban area, many smelters are located in remote and undeveloped regions where other significant emissions sources are absent. Such remote smelters offer interesting comparative cases where interpretation of the source for documented regional-scale contamination is simple and unmistakable.

Several approaches were used to identify other relevant smelter studies. A few reports were available from earlier project files (e.g., PSAPCA 1981; Black & Veatch 1988a; Bechtel Environmental Inc. 1992). Multiple key word database searches were conducted at the University of Washington and NOAA libraries in Seattle (using, among others, the Compendex and Agricola databases). The annual indices in Pollution Abstracts (published by Data Courier, Inc., Louisville, Kentucky) for the period 1979 through 1995 were key-word searched at the University of Washington Engineering Library. The cumulative USEPA library holdings were also searched online. Citation index files were used to identify references citing a few key early reports (e.g., Germani et al. 1981, Small et al. 1981). Additional key word searches on smelter-related environmental contamination and specific smelter names were conducted on the Internet through various search engines. Direct written and telephone contacts were also made to government agencies and researchers with respect to a few smelters of special interest (e.g., the Ronnskar, Sweden smelter) requesting listings of relevant environmental impact studies. Finally, several journals of particular interest were systematically reviewed to identify additional articles of interest.

The literature review resulted in the compilation of several hundred documents. Table 2 provides a listing, organized by country and smelter, of selected references judged to be of greatest interest for the credible evidence evaluations. Full reference information for these studies is provided in Section 6.0. The extensive set of studies included in Table 2 is by no means exhaustive; the study of smelter impacts remains a very active research topic (e.g., see Metals In The Environment - Research Network 1998 to present). The selected studies are representative of the very large set of smelter investigations worldwide and are sufficient to support the comparative evaluations included as part of the credible evidence analyses.

2.4 HISTORIC RESEARCH ASSOCIATES STUDY

After completion of the Vashon-Maury Island Footprint Study (PHSKC and Glass 2001), as the potential spatial scale of soil arsenic contamination became apparent, Ecology and Public Health - Seattle & King County decided to review possible historic sources of arsenic releases that might have contributed to regional-scale soil contamination. The agencies interest in researching other possible sources coincided with the extension of footprint studies from rural Vashon-Maury Island toward the urbanized and industrialized metropolitan Seattle area. Historical Research Associates, Inc. of Seattle, Washington was retained by PHSKC to perform a review for historical sources. That review focused on identifying substantial air emissions sources, or widespread land application practices, for arsenic that may have had regional-scale impacts. Localized sources that may have affected individual properties or small areas were not the focus of this research effort. The historical study focused on arsenic as the primary indicator contaminant associated with smelter emissions.

At the start of the historical research project, numerous references were consulted to develop a list of arsenic uses and potential emissions sources. Discussions between PHSKC and Historical Research Associates identified four areas in which the research would focus: smelters and other metals industries; steel mills; coal use; and agricultural use of arsenical chemicals. Information was also sought on other possible sources (e.g., incinerators), but less intensively, both by Historical Research Associates and PHSKC.

Historical Research Associates conducted research at a number of libraries, including the University of Washington, Washington State University, Seattle Public Library, and Vashon Public Library. The state archives were also reviewed for information on specific companies. Research contacts were made at the Museum of History and Industry and with local organizations and individuals to compile additional information, as well as with the Puget Sound Clean Air Agency. Extensive reviews of historical Seattle area directories for the period 1879 through 1969, at five-year intervals, and information from other documents were used to identify and locate specific industrial enterprises.

A final report was prepared (Mighetto and Schnaiberg 2001) that included a discussion of the research approach and findings, as well as supplemental materials in appendices (e.g., maps of Vashon Island agricultural land use in 1939 and Seattle area industrial land use in 1953). Information obtained on individual companies from directories research was compiled in an extensive spreadsheet. That spreadsheet and copies of relevant documents (e.g., on agricultural use of arsenical chemicals in Western Washington) were also provided to PHSKC at the end of the study.

The Historical Research Associates study addressed Vashon-Maury Island and the greater Seattle area in King County, which were the focus of footprint studies of soil contamination by PHSKC. It did not include similar historical research for areas of Pierce County that are also now included in the Tacoma Smelter Plume Site.

3.0 SUMMARY OF CREDIBLE EVIDENCE RATIONALES

A diverse set of data evaluations and rationales supports the credible evidence analyses. Each of the six major lines of reasoning includes the review and assessment of a substantial body of information. This section provides a brief overview of the cumulative rationales for identifying the Tacoma Smelter as a primary source for the documented regional-scale soil contamination in the central Puget Sound region. This overview will serve as an introduction and framework for the more detailed discussions in Section 4.0. It also points to the breadth and consistency of the total range of evaluations supporting the credible evidence analysis.

The six primary types of analyses included in the credible evidence evaluations can be labeled and summarized as follows:

- <u>Pathways (see Section 4.2)</u>. A complete pathway from smelter feedstocks through smelter emissions, airborne transport, and deposition to soils can be documented. The Tacoma Smelter was well-known as the U.S. copper smelter with the highest percentage of arsenic in its feedstock; arsenic production levels approached and at times exceeded 10,000 tons per year. Annual arsenic emissions measured in the hundreds of tons per year, with cumulative air emissions of several tens of thousands of tons over the operating history of the Tacoma Smelter. Other trace elements were also released in lesser amounts, as shown by various source test results. The typical annual wind rose represents the wind field affecting transport and dispersion of smelter emissions. Modeling studies of the deposition of arsenic using such annual wind rose information show regional-scale impacts. Monitoring of precipitation chemistry during individual storms showed strong upwind/downwind differences and increased deposition of arsenic plus other smelter-related metals (e.g., antimony) over a regional-scale area. Closure of the smelter resulted in changes in environmental chemistry at multiple long-range monitoring locations.

- <u>Spatial Patterns (see Section 4.3)</u>. Monitoring studies of multiple environmental media have shown gradients in contaminant concentrations associated with distance and wind direction from the Tacoma Smelter. Bee biomonitoring and precipitation chemistry studies (both single storm and long-duration monitoring) during smelter operations both provided contaminant contour plots demonstrating such gradients. Puget Sound and Lake Washington sediment profiles also show gradients with distance. The extensive Tacoma Smelter Plume footprint studies demonstrate such gradients in regional soils, with the greatest impacts in the two major downwind directions according to the annual wind rose (northeast and southwest). The footprint study data set is consistent with, but greatly expands upon, the results from numerous earlier soils investigations. That data set also reflects the development and application of a systematic approach to soil sampling (focused on comparatively undisturbed forest soils) at a regional scale.

- Chronologies (see Section 4.4). Studies of the chronology of Tacoma Smelter impacts include both short-term and long-term time scales. Pollution rose studies dating back more than 25 years demonstrated that short-term variations in ambient particulate arsenic concentrations as far away as Seattle were highly correlated with wind directions; higher levels occurred when winds came from the Tacoma Smelter. The concentrations of arsenic and other trace elements in ambient air were shown to be much higher during periods of smelter operations than during strike periods. Several studies also demonstrated decreases in impacts after closure versus during smelter operations. The long-term chronology of smelter impacts has been studied through chemical analyses of age-dated sediment cores in several downwind locations in Puget Sound and Lake Washington. The historic record for arsenic contamination in sediment profiles closely follows the history of smelter operations.

- Trace Element Patterns (see Section 4.5). The multi-element pattern of contamination in soils affected by smelter emissions has been shown to differ substantially from the pattern in orchard soils where lead arsenate was used. Air emissions of a number of volatile elements, including arsenic, have been associated with high-temperature smelting processes; source tests and emissions estimates for the Tacoma Smelter confirm releases of additional trace elements. Trace element analyses for a number of elements in Tacoma Smelter Plume soils from King County demonstrate strong, highly statistically significant correlations with arsenic and with each other. Trace elements such as antimony, bismuth, and indium (among others) therefore have similar regional spatial patterns as arsenic and indicate a common source. (The preliminary trace element results for soil samples from Pierce County are consistent with the King County results).

- Worldwide Smelter Environmental Impacts (see Section 4.6). A very large number of environmental studies exists for other smelters worldwide. Those studies have documented impacts in a variety of environmental media, including soils, sediments, vegetation, snowpack, and precipitation. Long-range transport of smelter trace elements and SOx in ambient air for several hundred miles has been demonstrated. The spatial scale for documented impacts is up to thousands of square kilometers, at distances of as much as 100 km or more downwind. The spatial pattern has typically shown contaminant concentration gradients, with an approximately exponential decrease with distance and a strong relationship to wind patterns. Soil contamination has been found mostly in near-surface soils, decreasing rapidly with depth. Many studies at other major smelters have documented contamination by multiple smelter-related elements, with a pattern of strong pairwise correlations among those elements. The spatial extent of detected contamination for any one element depends on factors such as the magnitude of smelter emissions, particle sizes within the smelter plume, and background soil concentrations and variability. Modeling studies and sampling during strike or post-closure versus operating periods have further confirmed smelter impacts. These findings are highly consistent with the results of Tacoma Smelter investigations. Studies at other smelters include many for smelters sited in remote regions where other credible sources are lacking and the interpretation of results is particularly clear.

o <ins>Mass Emissions/Other Potential Sources (see Section 4.7)</ins>. Cumulative arsenic emissions from the Tacoma Smelter amounted to tens of thousands of tons. Semi-quantitative estimates (derived from elevated soil arsenic concentrations) of the deposition of arsenic to regional uplands indicates a total deposition of at least several thousand tons. A historic review of potential air emissions sources did not identify any other arsenic sources in the metropolitan Seattle area that were of a commensurate magnitude. Historic sources such as a secondary lead smelter, gasworks facilities, municipal incinerators, steel mills, cement plants, and coal-fired powerplants and regional domestic and industrial coal use were judged likely to have at most localized or relatively small magnitude impacts on soil arsenic concentrations. Surveys of urban soil chemistry in a number of other cities, as well as nationwide sampling of stream sediments and several urban-to-rural gradient studies, further indicate that typical urban activities and industries are associated with only relatively small increases from geochemical arsenic levels. A historic review of possible agricultural use of lead arsenate or other arsenic-containing chemicals in Western Washington compiled numerous documents (e.g., agricultural bulletins) indicating that such use was recommended. However, the footprint sampling targeted forested areas (with a preference for larger acreages of more mature trees) where past agricultural land use was improbable. Moreover, a map of Vashon Island agricultural land use in 1939 showed that the spatial pattern of soil arsenic and lead contamination differed substantially from the pattern of agricultural areas. The trace element analyses further confirmed that use of lead arsenate could not explain the soil analysis results on Vashon Island. The use of arsenic-containing cacodylic acid in forest thinning operations was also shown in several studies to have only limited effects on general arsenic levels in forest soils. The review of other potential sources of arsenic thus failed to identify other credible sources of sufficient magnitude or pattern of use to explain the regional-scale soil contamination pattern.

4.0 RATIONALES FOR THE TACOMA SMELTER AS SOURCE

4.1 APPROACH

The Tacoma Smelter Plume Site studies have confirmed the occurrence of widespread soil contamination by arsenic and heavy metals in the central Puget Sound region. In evaluating the potential source(s) for that contamination, based on the compilation of available information (see Section 2.0), several approaches have been taken. Those approaches are identified here and discussed individually in the following subsections of the report.

The first approach for evaluating the role of the Tacoma Smelter as a source considers whether a reasonable and complete pathway for contaminants exists from the smelter to the contaminated region. This includes consideration of smelter emissions, particularly arsenic emissions, and the air transport and deposition of those emissions to surrounding soils. The Tacoma Smelter was only one of many pyrometallurgical facilities worldwide. The operations of these smelters were in most respects quite similar. Therefore, information from the numerous studies of impacts around other smelters helps to establish that there is a reasonable pathway for regional-scale contamination from their operation.

Another group of approaches considers the characteristic features of Tacoma Smelter emissions. Any significant source of contaminant releases can be characterized by features such as its place, the time scales of emissions, and the composition of releases (a chemical pattern). The impacts of Tacoma Smelter emissions can therefore be recognized by their spatial patterns (in various sampled media), their chronological patterns (at a variety of time scales), and the relationships among multiple contaminants within the region of impacts. Studies at other smelters support and confirm the interpretations of data from studies of the region near the Tacoma Smelter with respect to these patterns.

A third general approach considers the impacts from the Tacoma Smelter in the context of other potential sources. A historic review of other potential sources of arsenic, including their mass emissions and other characteristics, can provide information on the comparative roles of the smelter and other sources. The scale of impacts from various sources (scale defined both in terms of spatial scale and magnitude of resulting soil contamination) is an important consideration given the focus of this credible evidence report on regional-scale contamination.

The extensive literature on impacts from other smelters includes many applications of similar approaches to source identification. A variety of additional approaches has also been used, based on study designs not represented in the compilation of information used for this credible evidence evaluation. Some of those study designs are only applicable to an operating smelter with ongoing emissions; such approaches are no longer possible for the Tacoma Smelter. Other approaches are still feasible but have not yet been applied to the Tacoma Smelter.

Examples of additional approaches to source identification include the following:

- chemical and physical characterization of single particles, for example in downwind ambient air, to identify unique tracers (Bradley et al. 1981; Bradley and Buseck 1983; Pina et al. 2000). Henderson et al. (1998) examined single metal-enriched particles in humus samples downwind of a smelter, using scanning electron microscopy, and showed them to be similar to smelter dusts.
- multielement statistical analysis of ambient particulates and back-trajectory modeling of air mass transport (Gullu et al. 1995; Swietlicki and Krejci 1996; Polissar et al. 2001; Poirot et al. 2001)
- stable isotope ratio studies (Gulson et al. 1981; Sturges and Barrie 1989; Angle et al. 1995; Rabinowitz 1995; Blais 1996; Novak et al. 2003; Hou et al. undated; Savard et al. undated)
- chronologic and spatial studies using peat bog samples (Shotyk 1996; Kempter and Frenzel 2000; Cotizas et al. 2002; Nieminen et al. 2002; Novak et al. 2003)
- tree ring sampling and dendrochronologies (Robitaille 1981; Baes and McLaughlin 1984; Savard et al. undated; see also Riehl 1979 for an exploratory study on Vashon-Maury Island, downwind of the Tacoma Smelter)
- extensive spatial biomonitoring studies, for example using mosses, lichens, or forest floor humus (Fabiszewski et al. 1987; Sloof and Wolterbeek 1991; Gawel et al. 2001; Scerbo et al. 2002; Suchara and Sucharova 2002; Fernandez et al. 2002)

In some studies two approaches have been combined - for example, through the analysis of stable isotope ratios for lead within peat bog profile or dendrogeochemistry studies, or using multivariate statistical techniques to evaluate the results of national-scale lichen sampling.

The six approaches used to evaluate the Tacoma Smelter's role as a regional-scale source of soil contamination (see summary in Section 3.0 and detailed discussions below) are deemed sufficient to support the credible evidence findings. As noted above, however, these six approaches do not exhaust the range of possible approaches to source identification.

4.2 PATHWAYS

A complete pathway can be demonstrated between the Tacoma Smelter and the surrounding region where soil contamination by arsenic and other trace elements has been documented. Arsenic and other trace elements were present in the smelter's feedstock materials.

The smelting processes resulted in emissions of elements contained in those feedstocks, with arsenic emissions especially prominent. The annual wind rose describes the physical field into which trace elements were emitted; transport and deposition of emitted elements have been modeled and are consistent with environmental monitoring results. Ground-level impacts from tall stack plume looping behavior have been measured, and regional synoptic sampling of precipitation chemistry during single storm events has documented the transport of smelter emissions to tens of km downwind. Long range transport of smelter emissions has also been shown through correlating ambient particulate chemistry and wind directions at downwind locations and through changes associated with smelter closure.

The Tacoma Smelter specialized in the smelting of complex ores and concentrates. The typical roaster charge (blended from various sources) has been characterized as containing 3 to 4 percent arsenic (USEPA 1983a), a uniquely high value among U.S. copper smelters. An overall arsenic mass balance for the plant was also developed as part of USEPA's NESHAPs rulemaking (USEPA 1983a). That mass balance showed that most of the input arsenic was recovered as arsenic trioxide or metallic arsenic product, with main stack releases that were a small fraction of total arsenic inputs yet still elevated compared to other U.S. smelters. An earlier process flow analysis for arsenic is very similar and gives an arsenic content in the roaster charge as 3.8 percent (see PEDCo Environmental, Inc. 1976).

Nelson and Roberts (1975) analyzed the roaster charge composition on several days when they tested the efficiency of an electrostatic precipitator versus a pilot baghouse at the Tacoma Smelter. Values for lead, copper, arsenic, antimony, nickel, cadmium, mercury, and selenium were determined. The arsenic content on three days was 3.3, 3.8, and 3.8 percent. NEA, Inc. (1984) analyzed a roaster charge sample from the Tacoma Smelter for numerous elements, identifying elements such as copper, arsenic, lead, zinc, antimony, bismuth, silver, nickel, cadmium, and selenium. In the NEA sample, the roaster charge contained 6.5 and 8.8 percent arsenic in the coarse and fine size fractions, respectively. A sample of concentrates from a single high-arsenic source, the Lepanto (Philippines) concentrates, had 7.0 percent arsenic. Paulson et al. (1976) report the copper, arsenic, lead, zinc, antimony, and nickel composition of four samples of concentrates supplied from ASARCO Tacoma Smelter feedstock materials. These four samples were provided to represent a range of compositions typical of Tacoma Smelter operations, including one "arseniferous concentrate" from a single high-arsenic source, for testing the applicability of electric-arc furnace smelting technology for Tacoma. The high-arsenic source assayed at 11.4 percent arsenic; the other three blended concentrate samples ranged from 1.92 percent (low-arsenic green charge) to 4.13 percent (high-arsenic green charge).

The total throughput of arsenic at the Tacoma Smelter can be estimated from the annual amount of materials smelted and the blended, average arsenic content of those feedstocks. The resulting estimates are supported by annual arsenic production figures as reported by several sources. (Note: figures are variously given as arsenic or arsenic trioxide [As2O3]; the arsenic content of arsenic trioxide is 75.7 percent). Annual smelter throughput figures for 1971 through 1980, as provided by ASARCO, are reported by PSAPCA (1981; see Final EIS, Table II-1).

Typical throughput levels are 300,000 to 400,000 tons of smelted materials per year, with a decline below those levels in the last years of smelter operation. At an (assumed) average arsenic content of 3.8 percent, the total arsenic content of 300,000 to 400,000 tons of smelted materials is estimated at 11,400 to 15,200 tons per year. Arsenic trioxide production at the Tacoma Smelter captured more than three quarters of the arsenic content of smelted materials; somewhat less than one-quarter ended up in slag (which had a nominal arsenic content of about one percent), and smaller proportions were in produced copper anodes, in high-lead dusts sent to another smelter for lead recovery, or were released in air emissions (see PEDCo 1976, USEPA 1983a). The USEPA (1983a) arsenic balance based on data from 1982, during the period of reduced smelter throughput in the final years of plant operation, cites a feed rate of 2,074 pounds of arsenic per hour for 8,544 annual hours of operation, or an annual total of 8,860 tons of arsenic in smelted materials.

ASARCO (undated, 1975?) provides an arsenic balance for the Tacoma Smelter for 1974 and the first half of 1975. Arsenic production figures given are 4,578 tons for the 1st half of 1974, 2,279 tons for the second half of 1974 (including a labor strike period when the plant was shut down), and 4,015 tons for the 1st half of 1975. Nelson (1977) states that "current production rates [at the Tacoma Smelter] are about 1,000 tons of arsenic as arsenic trioxide per month" (page 31). The U.S. Bureau of Mines for many years withheld data on U.S. arsenic production to avoid disclosing individual company confidential information. ASARCO, however, released its company-wide arsenic production data to the Washington State Department of Social and Health Services (see Varner 1984). Tacoma Smelter production is broken out separately for the years 1946 through 1960. From 1947 through 1960, except for 1959, total As2O3 production at the Tacoma Smelter ranged between 8,051 and 13,334 tons per year (equivalent to 6,095 to 10,094 tons of arsenic per year). Loebenstein (1994) gives a figure of 9,900 metric tons (equivalent to 10,890 tons) of arsenic in arsenic trioxide produced in 1970, when the Tacoma Smelter was the only domestic supplier of arsenic.

In summary, an average feedstock arsenic content of 3 percent or more resulted in total arsenic throughput at the Tacoma Smelter approaching or exceeding 10,000 tons per year during years of full production. Various reports also document the occurrence of other trace elements in smelter feedstocks.

Randt (1975) provides data on the monthly mercury content of ores smelted at Tacoma in 1974 and 1975. The annual throughput of mercury at the smelter based on these data was about 8 to 9 tons.

Multielement analyses of collected dusts from the Tacoma Smelter have been performed for several studies. NEA, Inc. (1984) provides results for an extended list of elements for dusts collected from several pollution control systems at the Tacoma Smelter. Roberts and Nelson (1975) collected and analyzed dusts on the upstream and downstream sides of the Number 1 electrostatic precipitator and a pilot baghouse at the Tacoma Smelter. Samples were analyzed for arsenic, lead, cadmium, nickel, copper, mercury, and antimony. A sample of dusts collected at the pilot baghouse was also analyzed for the same seven elements. Statnick (1974) also provides data on elemental composition of dusts from the converter electrostatic precipitator inlet side, including arsenic, cadmium, chromium, copper, mercury, lead, and zinc. ASARCO data on the contents of recovered dusts from the two electrostatic precipitators, for 1976 to 1977, are also available (Roberts 2003) and provide information on gold, silver, lead, copper, zinc, arsenic, antimony, and mercury.

Tacoma Smelter slag has also been analyzed in several studies. A summary of trace element results from various slag analyses is provided in Glass and SAIC (1992; see Appendix F, Table F-1), including results for arsenic, cadmium, copper, lead, mercury, nickel, zinc, and antimony. Keystone/NEA (1991) performed analyses of slag samples for an extended list of elements.

These analyses of collected smelter dusts and slag provide additional information on the occurrence of arsenic and other trace elements in the smelting processes at the Tacoma Smelter.

Selected references for source tests and emissions estimates related to Tacoma Smelter operations are listed in Table 1. A summary of estimated historic main stack emissions for arsenic, lead, and mercury is provided by Varner (1983). The annual emissions for arsenic (as arsenic, not arsenic trioxide) decreased from 555 tons in 1960 to 70 tons in 1982 (last complete year of data); Varner also cites a 1953 company report estimating 630 tons in 1953. He states that pre-1960 arsenic emissions were about the same magnitude as in the 1960s. Based on these data, cumulative arsenic emissions from the Tacoma Smelter were tens of thousands of tons, making it one of the largest sources for arsenic emissions in the world. Emissions in the early decades of Tacoma Smelter operations are particularly uncertain; in the years before installation of electrostatic precipitators emissions may have been much larger than even the highest figures cited by Varner (compare Galbraith et al. 1995, who estimated emissions at the Anaconda, Montana smelter for 1911 through 1918 at up to 75 tons per day).

Varner (1983) lists annual lead emissions for the main stack declining from 226 tons in 1960 to less than 14 tons in 1982. Lead emissions were likely much higher in the initial years of operation as a lead smelter. Mercury emissions are cited for a shorter period; they decreased from 904 pounds in 1974 (including shutdown for a one month strike) to 330 pounds in 1982.

A number of other reports provide additional emissions estimates. Note that most of these estimates are for years well after peak emissions occurred, when additional pollution control systems had been installed and plant production was reduced somewhat (e.g., because of meteorological curtailment practices; see PSAPCA 1981). Labbe (1973) provides a summary report for 1972 emissions to air and surface water. Air emissions are reported as follows (in tons): arsenic, 132.6; lead, 82; zinc, 13.9; copper, 5.8; cadmium, 1.2; selenium, 0.2; nickel, 0.2; and silver, 0.2. Similar ASARCO monthly reports for January 1976 through September 1977 (including a two month strike period in 1977) provide estimated losses at the two electrostatic precipitators. Elemental percentages for particulates downstream of the electrostatic precipitators (mass-weighted averages of the two gas streams) for the period were arsenic, 41.7 percent; lead, 8.9 percent; zinc, 2.6 percent; copper, 2.1 percent; and mercury, 0.2 percent (Roberts 2003). Roberts and Nelson (1975) provide data for samples downstream of the Number 1 electrostatic precipitator and a pilot baghouse, quantifying amounts of arsenic, lead, antimony, copper, nickel, cadmium, and selenium. Statnick (1974) measured mass emissions rates from the two electrostatic precipitators, reporting the following results (in pounds per hour): arsenic, 58; lead, 25; zinc, 16; copper, 4.8; cadmium, 1.3; and chromium, 0.1.

NEA, Inc. (1984) also performed multielement analyses of samples of emissions from three sources at the Tacoma Smelter: the Number 1 brick flue (downstream of the Number 1 electrostatic precipitator), the Number 4 converter secondary hood, and a reverberatory furnace slag skimming operation. Those samples were analyzed as both coarse and fine fractions. These data were used to chemically characterize emissions sources for NEA's ambient arsenic source apportionment study.

PSAPCA, as the primary regulatory agency responsible for evaluating and controlling impacts from Tacoma Smelter air emissions, developed an extensive emissions inventory including tall stack and fugitive emissions sources from plant operations. ASARCO reports, agency source tests, and various emission factors were used in developing the emissions inventory. Estimates were derived for 1977 and 1979 for arsenic (as arsenic trioxide), lead, antimony, cadmium, nickel, and mercury (Roberts 2003). An inventory for the period 1971 through 1980 for arsenic and total particulate emissions from various sources was included in the PSAPCA EIS (PSAPCA 1981; see Appendix F in the Draft EIS). Several studies of slag dumping emissions were performed over time (see PSAPCA 1974; Henderson and Brand 1977; Henderson 1981; PSAPCA 1982), resulting in some modifications to the emissions estimates for that source of fugitive emissions. Other portions of the emissions inventory considered data for converter building emissions (Labbe 1975; Randt 1975), anode furnace emissions (Randt 1975), and roaster baghouse mercury emissions (Labbe 1977).

Arsenic mass flows through the smelting process, including tall stack arsenic emissions, have been developed by PEDCo Environmental, Inc. (1976) and EPA (USEPA 1983a). The results are generally consistent with the estimates by Varner (1983) for the time of the study. A number of specific source tests used for the 1983 arsenic process flow summary are summarized in USEPA (1983a; see Appendix C; see also USEPA 1979a, 1979b; USEPA 1984). A study was also performed to evaluate the effectiveness of a proposed air curtain hooding system for use with Tacoma Smelter converters (PEDCo Environmental, Inc. 1984). That study provides data on emission rates of six elements - arsenic, lead, antimony, bismuth, cadmium, and selenium - for various converter operating modes (e.g., charging, blowing, and skimming).

The source tests and emissions estimates from various reports identify the Tacoma Smelter as a dominant source for arsenic emissions and an important source for lead, antimony, and other trace elements.

Ambient particulates collected very near the Tacoma Smelter have been analyzed for multiple elements in several studies. GCA (1974) reports results for a list of elements including arsenic, bismuth, cadmium, copper, mercury, indium, nickel, lead, antimony, and zinc, among others. NEA, Inc. (1984) analyzed 10 ambient samples for an extended list of elements, including arsenic, copper, lead, antimony, mercury, selenium, and silver, among others. McClannan (1974; McClannan and Rossano 1975; see also Section 4.4 below) demonstrated significant increases for eight elements - arsenic, lead, zinc, selenium, antimony, manganese, cadmium, and mercury - during smelter operations versus shutdown during a strike period. Extensive ambient air monitoring data are also reviewed in the PSAPCA EIS (PSAPCA 1981).

Dustfall samples collected near the Tacoma Smelter have been analyzed on several occasions. Materials collected over a four month period in 1975 at three locations were analyzed for arsenic, lead, cadmium, mercury, copper, nickel, selenium, and antimony (Roberts 1975). Fallout collected on a nearby building roof contained 14.5 percent arsenic, 0.43 percent lead, and 0.24 percent cadmium (Franks 1981). Multiple samples of fallout collected after a stack fire in July 1984 were analyzed for arsenic, copper, lead, zinc, and antimony (Austin 1984). Data on arsenic, cadmium, and lead contents of dustfall from the demolition of the tall stack in 1993 are also available (ASARCO 1993).

These ambient particulate and dustfall studies identify highly elevated levels of arsenic associated with smelter operations, and elevated levels of a number of additional trace elements.

Annual winds near the Tacoma Smelter are dominated by winds from the southwest and northeast. PSAPCA has published wind roses for several monitoring locations in its annual summary reports. A representative annual wind rose from 1980 is shown on Figures 1 and 2. (Note that on those figures the arrows, scaled to frequency, are drawn pointing in the direction toward which the winds blow rather than the direction from which they blow, as is commonly done for wind roses). The wind field characterized by the annual wind rose (combined with other variables including wind speeds and atmospheric stability classifications) controls the physical transport of smelter emissions. In general, the downwind impacts from smelter emissions are expected to be greater in magnitude and extend farther in those directions with more frequent winds.

Meteorological information, including a representative annual wind rose, and smelter emissions estimates have been used to model impacts in the region surrounding the smelter. An SO2 modeling study was performed by H.E. Cramer, Inc., Salt Lake City, Utah for USEPA; the results are described in the PSAPCA EIS (PSAPCA 1981; see Appendix E in the Draft EIS). The deposition of arsenic and cadmium from smelter emissions was modeled as part of the PSAPCA EIS, using the Industrial Source Complex - Long Term deposition model (see PSAPCA 1981, Final EIS, section V.2). EPA also modeled ambient arsenic concentrations as part of risk evaluations performed for its NESHAPs rulemaking for arsenic (see USEPA 1983c). The results of these modeling studies show the expected relationship to wind frequencies and produce spatial patterns that are very similar to those observed in environmental monitoring studies of the region surrounding the Tacoma Smelter (see Section 4.3 below).

Vong (1982) modeled sulfate wet deposition from Tacoma Smelter emissions. The results to the northeast showed a gradient extending more than 100 km from the smelter. The deposition of arsenic and sulfate in the region downwind of the smelter was modeled by Luecken et al. (1989; see also Payton 1988) and compared to single storm precipitation chemistry monitoring data collected in 1985 prior to smelter closure. A version of the Multi-Pollutant Atmospheric Deposition and Depletion model developed by Battelle Northwest Labs was used. Modeling results reproduced the general spatial patterns shown in the monitoring data.

The physical transport and deposition of arsenic and other trace elements in the region downwind of the Tacoma Smelter is well documented in a series of synoptic precipitation chemistry studies by University of Washington researchers. The wind directions were known during these studies, establishing the direction of plume transport. Upwind samples had very low concentrations of smelter-related elements, and the downwind samples showed strong gradients in concentrations and correlations among multiple elements. These precipitation chemistry studies are described in Section 4.3 below; changes in precipitation chemistry patterns associated with smelter closure are discussed in section 4.4.

Lutrick (1971) performed mobile sampling with an integrating nephelometer (an instrument measuring light scattering) in the area south of the Tacoma Smelter. Sampling occurred over several days when winds were from the north, resulting in plume travel toward the sampling area. Short-term looping behavior of the tall stack plume was observed by Lutrick, who found high ground-level impacts at distances between two and six miles from the smelter. Similar looping behavior of the tall stack plume at the Sudbury, Canada smelter at very similar distances (3 to 10 km), with high ground-level contaminant concentrations, has been reported by Chan and Lusis (1985).

The concentrations of arsenic and antimony in ambient particulates in Seattle have been shown to be much higher when winds were from the south (i.e., coming from the direction of the smelter) than when they were from the north. Such pollution rose patterns indicate that Tacoma Smelter emissions had impacts as far downwind as Seattle and Lake Washington (see Section 4.4 below). Dethier (1979) measured precipitation chemistry and metals deposition rates to an alpine lake basin 80 km northeast of the Tacoma Smelter. That lake basin is downwind of the Tacoma Smelter during storms with typical southwest winds. Dethier (1979) suggests that the measured deposition rates for copper and arsenic show impacts from smelter emissions. Smelter closure studies that compared pollutant concentrations during smelter operations and after shutdown at far-downwind locations (e.g., in southern British Columbia; see Faulkner 1987) offer further confirmation that smelter emissions had measurable impacts over long distances downwind (see Section 4.4 below), well beyond the region of Tacoma Smelter Plume site soil sampling to date.

4.3 SPATIAL PATTERNS

A major objective of the Tacoma Smelter Plume footprint studies was to characterize the magnitude, extent, and spatial pattern of soil arsenic and lead contamination in the region surrounding the former Tacoma Smelter site. The cumulative results from the footprint studies provide the best information on spatial patterns, for several reasons. The design of these soil sampling studies provides the best regional coverage among all available data sets. Soils are recognized as a long-term sink for arsenic and other trace metal contaminants, and reflect cumulative deposition. Moreover, soil sampling in the footprint studies focused on relatively undisturbed forest soils where the influence of other arsenic sources is largely controlled, anthropogenic physical disturbance of the soil profile is minimized, and the interpretation of results is clearest (see Chirenje et al. 2002 for the importance of land use types; see Section 4.7 below). A consistent approach to sampling and analysis was used in all three footprint studies, resulting in a homogeneous data set. All lab analytical results went through a data validation process, assuring a data set of documented high quality. Finally, the data represent current conditions (while many historic studies collected data as much as 30 years ago).

Spatial patterns in contamination around the Tacoma Smelter have been studied through sampling of various environmental media in many studies since the early 1970s (see Section 2.1.2). Those results are considered less definitive than the information from the Tacoma Smelter Plume footprint studies because the earlier studies were smaller in spatial scale and sample size, had less carefully controlled and less powerful sampling designs, sampled media that were not as representative of total spatial impacts or were affected by other contaminant sources or processes (e.g., diluting the "signal" from smelter impacts), or were otherwise more limited. On the other hand, earlier studies included sampling of media only affected when the smelter was still operating (e.g., bee biomonitoring and precipitation chemistry studies); such studies could not be replicated today. A listing of selected studies providing information on the spatial extent and spatial pattern of Tacoma Smelter impacts is provided in Table 3. Although the earlier study designs are limited in certain respects, their results are largely consistent and mutually supportive. The results from historic studies will be briefly described before the results from Tacoma Smelter Plume footprint sampling are presented. Environmental sampling results are consistent with modeling of Tacoma Smelter impacts, which are discussed in Section 4.2 along with evidence for long-range transport of smelter emissions.

There are numerous historic studies of arsenic and other metals in soils near the Tacoma Smelter (see PSAPCA 1981 and Bechtel Environmental Inc. 1992). The results of those historic studies were compiled and reviewed as part of the footprint study design process. Interpretation of the cumulative results was complicated by the inconsistent sampling designs used across studies (e.g., sampling soils from gardens, residential lawns, or forested properties and collecting different depth intervals for analysis). The total spatial area represented by the historic studies was smaller than was eventually included in the footprint studies. Many of the earlier studies also included a comparatively small number of samples. Despite these limitations, the cumulative data set from early soil studies showed a strong general pattern of exponential decrease in contaminant concentrations with increasing distance from the smelter (mostly for arsenic, lead, and cadmium, and occasionally including other trace elements), as well as an association with wind direction frequencies.

One of the largest early studies was performed by Heilman and Ekuan (1977) and collected soil samples from 72 private gardens, most within 10 miles of the smelter. The samples were analyzed for many trace elements. Scatterplots of soil concentrations versus distance from the smelter (especially in the two primary downwind directions) showed substantially elevated levels near the smelter and a curvilinear decrease with distance, described as approximating an exponential decay curve. Heilman and Ekuan (1977) reported highly statistically significant exponential decreases for multiple elements, including arsenic, cadmium, lead, copper, mercury, and antimony, among others.

Simpson (1983) evaluated the spatial structure of soil arsenic concentrations using 270 sampling results from several studies, with most coming from a second large survey of arsenic in garden soils (Lowry 1983). The statistical approach used was kriging. Simpson reported a kriging solution that described the structure of the data set (semi-variogram) in terms of the correlations among the data as a function of spacing between samples and orientation. She also discussed the implications of those findings for future study designs.

Garden soils reflect the opposite end of the spectrum from undisturbed soils, with substantial physical mixing and the use of various soil amendments being typical in gardens. The results of the Heilman and Ekuan (1977) and Lowry (1983) garden soil surveys, with significant concentration versus distance relationships shown, therefore suggest the magnitude of smelter impacts was large. It is not surprising, given the disturbance of garden soils, that estimates of spatial extent from garden soil surveys would be smaller than similar estimates from sampling less disturbed soils. Three studies of soils at distances of about 5 to 7 miles from the smelter, in two downwind directions (University Place, Pierce County and Maury Island, King County), that were completed as the footprint studies began in fact demonstrated that sampling in less disturbed soils would show a greater magnitude and extent of soil contamination (see King County Department of Development and Environmental Services 1999; City of Tacoma and Glass 1999; Golding 2001).

Crecelius (1974) mapped arsenic and antimony concentrations in surface sediments throughout Puget Sound. He noted that the distributions of these elements from the smelter are affected by both wind and water; the mapped distributions therefore reflect more than just air emissions and deposition. Crecelius states: "The arsenic and antimony distributions in the surface sediments of Puget Sound indicate that the Tacoma copper smelter is the major anthropogenic source of these two metals in the region. Within 8 to 15 km of the Tacoma smelter, arsenic and antimony concentrations rise to 2-3 times background values" (page 25). The detectability of smelter impacts in sediments is affected by the diluting effects of other large sources of comparatively clean sediments (e.g., the Puyallup River).

The U.S. Geological Survey has also collected data on concentrations of arsenic and heavy metals in freshwater sediments in the Puget Sound Basin. MacCoy and Black (1998) report data for surface sediments (top 2-3 cms) from 18 locations. Eight locations designated as urban land use sites (in Auburn, Tukwila, Juanita, Bellevue, Steilacoom, Des Moines, Bothell, and Seattle) have arsenic concentrations of 10 to 33 ppm (dry weight basis, screened to <63 micron size fraction), elevated in comparison to forest and reference locations. Those eight urban locations are also downwind of the Tacoma Smelter, and the results may reflect the extensive spatial effects of smelter arsenic releases; on a national level, arsenic was not found to be enriched in urban stream sediments (see Rice 1999 and Section 4.7).

University of Washington researchers have performed a number of studies of precipitation chemistry over a large region downwind of the Tacoma Smelter. The setting of the Tacoma Smelter was characterized by these researchers as providing close to an ideal situation for understanding the impacts of a large point source of pollution. The air masses passing over the Tacoma Smelter during most precipitation events come from the southwest, where clean air masses cross the Washington coast and do not encounter other significant sources upwind of the smelter. The smelter itself was recognized as being the dominant regional source for SO2 and arsenic, and possibly other trace elements as well. Upwind precipitation arsenic levels (at locations southwest of the smelter) were found to be quite low. Carpenter et al. (1978) report the results of a two-year program of sampling and analyzing rain water at 12 locations from Olympia in the southwest to near Anacortes in the north. More than 200 samples were collected in all. The average (upwind) arsenic concentration near Olympia over the two-year study period was 0.4 ppb. Carpenter et al. (1978) state: "These results confirm that arsenic concentrations in the precipitation samples decrease with distance north of the Tacoma smelter, consistent with the fact that southerly winds usually accompany rainstorms in this region. Arsenic concentrations still average 4 ppb at the Edmonds station north of Seattle, and 2 ppb even further north at Everett. Thus, elevated arsenic concentrations are still found at least 50 km downwind of the smelter" (page 469).

University of Washington researchers also conducted a series of single-storm monitoring studies in which a large number of rain collection stations was established over a region extending approximately 80 km downwind and including some upwind locations. Collected samples were analyzed for major anions and cations, pH, and various trace elements including arsenic. Results for the first synoptic study of this type were reported by Larson et al. (1975). Isopleth maps for concentrations of pH, sulfate, arsenic, and antimony constructed from the results across all collection locations clearly showed the effects of Tacoma Smelter emissions on downwind rain chemistry. Larson et al. (1975; see also Harrison et al. 1977) concluded that the smelter influence was detected to a distance of about 40 km downwind and that arsenic and antimony appear to be good elemental tracers of the plume to a distance of about 25 km. Additional storms were monitored, using a similar synoptic collection design, over the years. Knudson et al. (1977) used multivariate statistical methods (factor analysis) to evaluate rain chemistry results and identified a factor associated with smelter emissions that included arsenic, antimony, copper, and cadmium, all of which showed a spatial pattern related to the smelter on contour maps. As part of the Tacoma Smelter Closure Study, 14 storms were monitored (Vong et al. 1986; see also Moseholm 1986; Vong et al. 1987; Vong, Larson et al. 1988; Vong, Moseholm et al. 1988; Peterson II 1991). A classification of collection locations was made that identified those within about 25 km downwind as being smelter-impacted (Vong et al. 1988), although some data evaluations noted changes associated with smelter closure at greater downwind distances (see discussion in Section 4.4). Sulfate, arsenic, copper, lead, indium, and antimony were enhanced in downwind samples during the period of smelter operations (Vong et al. 1987).

The impacts of smelter emissions have also been studied using bee colonies (Bromenshenk et al. 1985, 1991). Based on their travel patterns, bee colonies are identified with ambient air trace element contaminants over local areas of about 7 square km (Bromenshenk et al. 1985). A regional network of beekeepers was recruited to provide data from 72 locations over an area of 7500 square km. Kriging techniques were applied to the collected data to produce contour maps of arsenic content in bees over the sampling region. The results showed a clear pattern associated with the Tacoma Smelter, which the authors characterized as extending "at least to the Lake Sammamish Plateau" (Bromenshenk et al. 1985, page 633). In a separate investigation, smaller bee colonies were established along a five-station transect across Vashon Island (south to north). The biomass of bees per colony was shown to increase as the distance from the smelter increased, and the biomass values were inversely correlated with the arsenic content of the bees (Bromenshenk et al. 1991).

A few studies of the trace element contents of vegetation have included enough sampling locations to evaluate spatial patterns. Crecelius and Piper (1973) measured the copper content of Douglas Fir tree needles over a region extending to about 25 km from the Tacoma Smelter. Contouring of the results (using an "enveloping isopleth" approach to show the maximum extent of effects, given relatively high variability) produced a set of contours centered on the Tacoma Smelter and extending in the dominant downwind direction (NNE) to the limits of sampling near West Seattle. The Heilman and Ekuan (1977) study of garden soils also included sampling and analysis of many types of garden produce. The concentrations of multiple elements in multiple leafy crop types showed a statistically significant exponential decay with increasing distance from the smelter, similar to the soil results. Arsenic, antimony, cadmium, lead, copper, and mercury were among the elements with significant findings.

The Tacoma Smelter Plume footprint studies provide soil arsenic and lead results for 430 sampling locations. The sampling design for these studies included collection and analysis of multiple depth intervals in each soil boring. It also incorporated multiple closely-spaced borings at many sampling locations so that variability at a variety of spatial scales could be evaluated. Even closely-spaced borings were often found to have significant variation in contaminant concentrations, and multiple depth intervals within a boring similarly displayed considerable variation (e.g., see PHSKC and Glass 2000; compare Arrouays et al. 1996). For summary data evaluations, the total footprint data set was reduced to retain only the maximum reported concentration for arsenic or lead at a sampling location (any depth interval at any boring). These maximum values for arsenic and lead do not necessarily occur in the same sample at a location.

Focusing on these maximum values is equivalent to determining "enveloping isopleths" when evaluating spatial patterns in soil contamination. A listing of the maximum arsenic and lead results, including sampling location distance and direction from the smelter and location type (forested or residential), is provided as Attachment A.

The maximum arsenic and lead concentrations for all footprint sampling locations in King County and Pierce County to date are shown on regional maps produced by Ecology, reproduced here as Figures 1 and 2, respectively. When maximum arsenic or maximum lead concentrations are plotted versus distance from the Tacoma Smelter, a strong pattern of exponential decay with increasing distance results. The data can also be plotted by wind rose sector (a 16-fold division of the compass, each division therefore representing 22.5 degrees). These wind direction-segregated scatterplots still show exponential decay relationships, with the magnitude and spatial extent of soil contamination showing good agreement with wind frequencies. These wind direction-segregated scatterplots were used to define child-use sampling areas for the King County Mainland and Pierce County Child-Use Area studies.

The spatial pattern for soil contamination is most clearly shown, and nothing essential is lost, by focusing on the two primary downwind directions according to the annual wind rose near the smelter: areas to the north-northeast (between compass bearings 12.25 and 57.25 degrees) and to the south-southwest (between compass bearings 192.25 and 237.25 degrees). To accommodate the requirements of the data plotting program used and separate the data for the two downwind directions, distances from the smelter for sampling locations to the south-southwest of the smelter have been arbitrarily assigned negative values. The two primary downwind directions chosen to illustrate spatial patterns in soil contamination surrounding the smelter include 174 of the 430 total sampling locations, 112 to the north-northeast and 62 to the south-southwest. All of the data to the north-northeast represent undisturbed forested locations. The 62 sampling locations to the south-southwest include 28 residential properties to provide spatial coverage in areas where forested properties suitable for sampling are lacking. Residential sampling was used in the more heavily urbanized areas of Pierce County closest to the smelter. The sampling design increased the density of sampling locations in residential areas in anticipation of greater variability there due to increased soil disturbance. The results from residential sampling locations are also considered likely to under-represent the maximum smelter impacts because of mixing and dilution from soil disturbance; in regions where both residential and forested properties were sampled, this low bias was in fact observed.

The scatterplots for maximum arsenic and maximum lead concentrations versus distance from the smelter for the two primary downwind directions are provided as Figures 3 and 4, respectively. Locations to the south-southwest are on the left in these figures; locations to the north-northeast are on the right. Strong concentration gradients with approximately exponential decay shapes are demonstrated in these figures for two separate directions from the smelter and for two smelter-related elements. (See the discussion in Section 4.5 for results for additional trace elements). Regression analyses of these spatial patterns, using log arsenic values and linear distances, show highly statistically significant results; for example, for the 112 results to the north-northeast, the R-squared value is 46 percent and the p-value is <0.00001. Note that almost all of the plotted maximum arsenic concentrations for these two downwind directions exceed Ecology's designated "urban background" value of 20 ppm. The maximum arsenic and maximum lead results shown in Figures 3 and 4 have similar patterns and are highly correlated statistically, even though a small number of anomalous lead results is noted.

The spatial gradients for footprint study soils data shown in Figures 3 and 4, and for other wind directions as well, strongly indicate that the Tacoma Smelter is the primary source for the observed regional soil contamination.

4.4 CHRONOLOGIES

The chronologic patterns of emissions and impacts from the Tacoma Smelter occurred at a variety of time scales, from short-term (day-to-day or even hour-to-hour, based on smelter operations and wind patterns) to long-term (emissions occurred in a defined time period from 1890 to 1986). Several types of studies provide information that can be associated with such chronologic patterns; Table 4 provides a listing of some of the relevant Tacoma Smelter studies. The results of various chronologic studies confirm that the Tacoma Smelter was an emissions source with measurable regional-scale downwind impacts.

Just as a wind rose can be used to summarize the pattern of winds by direction, a pollution rose can be used to summarize measurable air pollution data - for example, ambient SOx or particulate arsenic concentrations - by wind direction (and wind speed). A consistent association between air pollution and wind direction can provide a strong indication of the source location (i.e., it indicates a transport pathway). The Tacoma Smelter was the dominant regional source for both SOx and arsenic emissions, and pollution roses related to those air pollutants (and sometimes other trace elements as well) have been characterized for Seattle monitoring locations, approximately 25 miles downwind from the Tacoma Smelter. Buchan (1967) reported that the highest SO_2 values at a University of Washington monitoring location were associated with winds from the SSW, regardless of wind speed. He demonstrated by simple diffusion calculations that mass SOx emissions from the smelter were consistent with these measurements, and further noted that arsenic levels in Seattle ambient particulates had been reported among the highest among U.S. cities. In a later Seattle study, Hatfield (1976) collected and analyzed size-fractionated ambient particulates. Hatfield found that arsenic, cadmium, and copper, which were all found predominantly in the fine fraction, were significantly correlated and that there was a suggested association of elevated arsenic levels with southerly winds.

Crecelius (1974), in his foundational study of the geochemistries of arsenic and antimony in the Puget Sound region, measured their concentrations (dry weight basis) in collected ambient particulate samples at the University of Washington campus. The concentrations for both arsenic and antimony were an order of magnitude higher when winds were from the south (i.e., from the direction of the smelter) versus the north, and were highly correlated. The lower values with northerly winds were comparable to reported values in other U.S. cities. Crecelius states: "The dramatic relationship between wind direction and arsenic concentration in atmospheric dust indicates that the Tacoma smelter is the major contributor of atmospherically transported arsenic to Seattle and Lake Washington. No other sources of arsenic to the atmosphere appear to be significant" (page 86).

PSAPCA (1981) also provides SO2 wind roses for five monitoring locations, including Northeast Tacoma, Maury Island, and the King County Mainland (Kent). For all five stations, SO2 concentrations rarely exceeded de minimis values when winds blew toward the Tacoma Smelter and most frequently exceeded de minimis values when winds were from the smelter toward the monitoring location (see Appendix D in PSAPCA 1981).

Thus, the chronologic association of pollutant concentrations with wind directions as shown in various pollution rose studies identifies the Tacoma Smelter as a dominant source for some pollutants at locations as distant as Northeast Tacoma, Maury Island, Kent, and Seattle.

Labor contract negotiations at the Tacoma Smelter periodically resulted in smelter shutdown until a new contract was signed. Such labor strikes created opportunities for "natural experiments" in which measurements during strike versus smelter operating periods could be compared. Ambient particulates were collected and analyzed at the time of the 1974 strike from 10 stations all located within one mile of the smelter (McClannan 1974; McClannan and Rossano 1975). The average ambient concentrations (in ug/m3) for arsenic, lead, zinc, selenium, antimony, manganese, cadmium, and mercury during 4 days of smelter operation were all higher than during 14 days of the strike. Arsenic concentrations increased by a factor of 12.3; other trace element enrichment factors were lower. Smelter operations were thus clearly associated with increased emissions of arsenic and other trace elements. McClannan (1974) notes that even during strike periods the concentrations of arsenic and other elements were significantly higher than in Seattle, likely reflecting contaminated dust resuspension near the Tacoma Smelter. McClannan also notes that during the 4 days of sampling during smelter operation, the ambient concentrations of multiple elements (e.g., arsenic, lead, antimony, and cadmium) varied synchronously at nearly all of the monitoring locations; this also strongly implicates a single source.

PSAPCA also compared ambient air arsenic measurements at three monitoring stations during a prolonged strike period in 1977 to values from preceding and succeeding operating periods. As in the McClannan study, a large factor of increase (more than 7-fold) was observed during smelter operations (PSAPCA 1981). The somewhat smaller factor of increase in the PSAPCA study probably reflects the more variable wind conditions during the much longer data evaluation period.

The final closure of the Tacoma Smelter in 1986 similarly created an opportunity for "natural experiments" to document downwind changes associated with the chronology of smelter closure. One closure study provides interesting information on the spatial scale of smelter plume transport. Faulkner (1987) reports the findings for monitoring of precipitation chemistry over several years, both before and after smelter closure, along a transect of Canadian stations from western Vancouver Island to the lower Fraser River Valley (over 100 miles from the Tacoma Smelter). Substantial and statistically significant decreases in arsenic are reported comparing pre- and post-closure results, with all locations showing post-closure values at essentially background levels. Faulkner comments: "Considering the meteorology of the Pacific Northwest and southwestern B.C., it is almost certain that the ASARCO smelter was the source of the arsenic monitored in precipitation in B.C." (page 21). Thus, arsenic was shown to be an excellent tracer for long-range transport of the smelter plume (compare Bigg et al. 1978 and Carras and Williams 1981 for smelter plume transport distances; see also Nerin et al. 1994 for arsenic as a long-distance tracer for a source with much lower arsenic mass emissions). Arsenic and sulfate correlated strongly before but not after closure. Significant reductions in monitored sulfate concentrations were also reported, although the interpretation of the sulfate findings is more complicated.

Welch et al. (1992) monitored sulfate concentrations in a number of lakes in the Alpine Lakes Wilderness Area, northeast of the smelter (approximately 50 miles), and in precipitation at Snoqualmie Pass before and after smelter closure. They report a statistically significant decrease in sulfate in the lakes between 1983 and 1988. Considering the comparative annual SO2 emissions from the Tacoma Smelter and Mt. St. Helens for the years 1980 through 1988, as well as the timing of lake sulfate decreases by lake type (fast-flush versus slow-flush), Welch et al. conclude that both may have been responsible for the trend of decreasing lake sulfate.

A research team from the University of Washington performed a study of precipitation chemistry bracketing the time of smelter closure (see Vong et al. 1986). Synoptic rain water sampling was conducted for 14 separate storms over a regional network of approximately 40 sampling locations. The collected samples were analyzed for major anions and cations as well as a list of trace elements, including arsenic. Differences in the chemistry of rain water samples from events before versus after smelter closure were reported in a series of publications and University of Washington theses. These smelter closure rain chemistry studies were enhanced by the relatively clean background characteristics of rain water southwest of the smelter (upwind), as confirmed by the inclusion of upwind sampling stations for all monitored storms.

Multivariate statistical analyses were used to classify sampling locations into groups, including one downwind group (to a distance of about 25 km) identified as affected by smelter emissions (see Vong, Larson et al. 1988). Variables used for this classification included arsenic, antimony, copper, lead, sulfate, and hydrogen ion concentrations (pH). The mean upwind sampling location sulfate levels were observed to vary somewhat between 1985 (pre-closure) and 1986 (post-closure) monitoring events. Therefore, the evaluation of smelter closure impacts on rain water chemistry was performed by looking at upwind versus downwind values as a function of time of sampling, or year (see Vong, Moseholm et al. 1988). These upwind-downwind differences for sulfate and hydrogen ion concentrations were found to have significantly decreased after smelter closure at the downwind locations classified as smelter-impacted based on the initial multivariate statistical analyses (Vong, Moseholm et al. 1988; Moseholm 1986).

Peterson II (1991) evaluated the trace element results from the smelter closure rain chemistry study. He found that arsenic, antimony, copper, and selenium decreased in concentration with increasing distance from the smelter and were associated with measured sulfate. Differences in trace element chemistry for storms before versus after smelter closure were statistically evaluated (using multivariate discriminant analysis). Statistically significant decreases in element enrichment factors were found, with arsenic and antimony - two of the primary smelter-related elements - contributing most to the observed differences. These statistically significant findings extended beyond the group of downwind sampling locations within 25 km of the smelter and included locations in the metropolitan Seattle region. Thus, Peterson II (1991) reports rain chemistry effects associated with smelter closure, based on arsenic and antimony data, over a larger region than Vong, Moseholm et al. (1988) did, based on sulfate and hydrogen ion data.

Scattarella (1988) evaluated the lead data from the UW smelter closure study. Significantly higher lead concentrations in rain samples were identified downwind. Lead values decreased in all parts of the study region, including upwind, in the period after smelter closure. Frequent not-detected results after closure, however, precluded statistical analysis of the upwind-downwind differences by year as was done by Vong, Moseholm et al. (1988). (Note that smelter closure coincided with the rapid phase-down of lead in gasoline). Scattarella used several other statistical approaches to assess the role of smelter emissions on downwind lead values. Using arsenic as a trace element for lead and lead:arsenic ratios, Scattarella concluded that the smelter was the dominant source for lead within about 25 miles downwind (accounting for 55 to 90 percent or more of the observed lead) and a contributing source (about 40 percent) farther downwind in the metropolitan Seattle region.

In addition to soils, sediments are a long-term environmental sink for arsenic and other metals. (Crecelius [1974] has also shown that most of the arsenic entering Puget Sound is discharged to the Pacific Ocean through the Strait of Juan de Fuca). The ability to age-date sediment cores provides a long-term chronologic record for the deposition of these contaminants. Pathways leading to sediment contamination include direct deposition from air (and runoff from upland areas) and current-driven water pathways. Specific sediment sampling locations can be characterized as largely reflecting either the air or water pathway; for example, locations that are upwind of smelter emissions based on wind roses could still be affected if they were "downcurrent" based on an understanding of Puget Sound circulation patterns. Locations that are downcurrent also could be more affected by direct water discharges (e.g., NPDES outfalls) or slag disposal/runoff from slag cooling rather than air emissions.

Crecelius (1974) mapped concentrations of arsenic and antimony in surface sediments in Puget Sound. He states that within about 15 km of the Tacoma Smelter surface sediment concentrations for arsenic and antimony are at least two to three times background values. The spatial distributions in surface sediments are affected by factors such as tidal current patterns and dilution at some locations by large volumes of relatively clean sediments in inflowing rivers (e.g., Puyallup River), as well as potential contributions from multiple sources.

Sediment cores collected at several Puget Sound and Lake Washington locations have been age-dated and analyzed by depth for arsenic and other trace elements. Crecelius (1974; see also Crecelius et al. 1975, Carpenter et al. 1978) analyzed sediment cores from four locations in Puget Sound: near Fox Island, Quartermaster Harbor on Vashon-Maury Island, East Passage (the channel east of Vashon-Maury Island), and North Seattle. Lead-210 analyses were used to age-date three of the four sediment cores (Fox Island omitted). Arsenic and antimony depth profiles were developed and associated with the ages determined from lead-210 analyses.

The North Seattle core had essentially background levels for arsenic, unrelated to depth; the core included sediments from about 1955. Crecelius notes elevated arsenic and antimony concentrations in the Fox Island core and attributes them primarily to slag transport by tidal currents from the smelter (although soils on Fox Island have also been shown to be contaminated from air deposition). The East Passage core had a much higher sediment accumulation rate and was not deep enough to go back before about 1960. A trend of increasing, above-background concentrations from 1960 through 1971 was observed in the East Passage sediment core.

Crecelius (1974) describes the Quartermaster Harbor location as largely representative of air transport rather than tidal currents. It is located in the dominant downwind direction from the smelter stack; adjacent uplands soils on Vashon-Maury Island had been shown to have substantially elevated arsenic and antimony concentrations. Puget Sound currents do not significantly affect Quartermaster Harbor. The arsenic:lead ratio in sediments was also noted to be much closer to stack emissions than to smelter slag. Arsenic concentrations approximated the Puget Sound background level (around 10 ppm) until about 1890 - the year the Tacoma Smelter started operating - and then showed a steady rise through 1971, reaching a maximum concentration of 54 ppm. The depth profile for antimony was similar. Crecelius et al. (1975) comment: "It is clear from these cores that at least a portion of the arsenic introduced to Puget Sound by the Tacoma Smelter has accumulated in bottom sediments" (page 330). In his calculated arsenic budget for Puget Sound, Crecelius (1974) estimated that smelter arsenic emissions falling onto Puget Sound as precipitation or dustfall were much greater than the total input from all rivers, and were exceeded only by the incoming (deep) seawater arsenic.

As previously noted, Crecelius (1974) documented that when winds were from the Tacoma Smelter toward the University of Washington campus in Seattle, near Lake Washington, ambient particulate matter had strongly elevated arsenic and antimony concentrations. Crecelius (1974) also examined sediments from Lake Washington as well as three other Western Washington Lakes (Crecelius 1974, 1975). Surface sediments were collected and analyzed from 14 locations. Arsenic and antimony concentrations were elevated, less so in shallow areas where coarser river sediments settled out and diluted. Five sediment cores were collected and analyzed.

Arsenic concentrations were elevated near the surface and decreased with depth to typical background concentrations (about 10 ppm). Antimony showed a very similar pattern with concentrations about one-tenth those of arsenic. Lead-210 dating for three cores established sedimentation rates. Based on that age-dating, the arsenic and antimony concentrations in Lake Washington sediment cores began increasing about the time when the smelter began operating (1890). A calculated arsenic budget for Lake Washington showed that rainfall and dustfall accounted for almost half the total inputs. An independently calculated rate of arsenic accumulation in Lake Washington sediments was about equal to the amount needed to close the arsenic budget (considering all estimated inputs and discharges through the lake outlet).

The U.S. Geological Survey also collected a number of sediment cores and analyzed them for multiple trace elements in the Reconstructed Trends National Synthesis Study, part of the National Water-Quality Assessment (NAWQA) Program. Cores were collected from single locations in Lake Washington and Lake Ballinger in 1998; both locations are NNE from the smelter in the primary downwind direction. The core from Lake Washington was chemically analyzed to a depth of 33 cm; assuming a sediment accumulation rate of 3 mm/yr (0.3 cm/yr), this is equivalent to 110 years and a start date of around 1888, just prior to the start of smelter operations. Arsenic concentrations showed a marked increase near the bottom of the sediment core, corresponding to the approximate date of the start of smelter operations, increasing to a maximum value (36.2 ppm) by the late 1930s more than 5 times the lowest value (6.4 ppm) at the bottom of the analyzed core. The Lake Ballinger core was analyzed to a depth of only 22.5 cm; at the reported sediment accumulation rate of 0.45 cm/yr, this represents only a 50 year record and a start date of around 1948. The arsenic concentrations in the Lake Ballinger core are elevated compared to the earliest depth intervals in the Lake Washington core, ranging up to 21.5 ppm. The top depth intervals show a reduction in arsenic levels corresponding to the period after closure of the Tacoma Smelter.

Crecelius and Piper (1973) similarly evaluated lead concentrations in two age-dated Lake Washington sediment cores. They noted that the depth profiles for lead were at background levels prior to about 1880, after which they increased to the present with a secondary maximum at about 1916 to 1920. This is shortly after the Tacoma Smelter converted from producing lead to copper. Indirect evidence, including measurements of lead and copper in Douglas Fir tree needles and comparative estimates of lead mass emissions from the smelter, leaded gasoline, and historic coal use in the Seattle metropolitan area, is cited for considering Tacoma Smelter impacts on Lake Washington sediment lead levels. (Note that the ambient particulate monitoring near Lake Washington, described above [see Crecelius 1974], was completed after this sediment lead study).

Lefkovitz et al. (1997) analyzed six age-dated sediment cores from the main basin of Puget Sound to continue a program for documenting historical trends in contamination. The six cores were collected in 1991 and covered an area from about Edmonds in the north to near Point Robinson, Maury Island in the south. Age-dating was performed using lead-210 analyses (six cores) and cesium-137 analyses (five cores). The highest concentrations of arsenic were found in the two southernmost cores, east of Vashon-Maury Island. In the two cores with sufficient depth to provide information from well before 1890 (near Maury Island and West Seattle), sediment arsenic concentrations were at background levels until starting to increase about 1890. The maximum arsenic concentrations occurred at around 1960; note that reported arsenic emissions from the Tacoma Smelter after 1960 were substantially reduced from earlier years (see Varner 1983). Antimony showed a similar pattern. Lead also had a synchronous increase at the southernmost location, with a sharp increase after about 1890. The substantial impacts of Tacoma Smelter lead emissions have in fact been used to adjust age dates for sediment cores in Puget Sound, as noted by Lefkovitz et al. (1997).

Sediment studies in Puget Sound and Lake Washington, to distances of up to 50 km downwind from the Tacoma Smelter, thus confirm trace element contamination patterns consistent with the known operating period and history of the Tacoma Smelter.

Finally, Riehl (1979) performed an exploratory study downwind of the Tacoma Smelter (Vashon-Maury Island) for one additional type of chronologic study - chemical analyses of tree rings. The arsenic content of individual annual growth rings was analyzed and evaluated in relation to smelter production history. The approach was considered exploratory and not fully validated.

4.5 TRACE ELEMENT PATTERNS

A detailed discussion of Tacoma Smelter trace element emissions and King County soil analyses for trace elements is provided as a separate report (Glass 2003). A summary of the major components and findings of that report is provided here. Readers are referred to Glass (2003) for additional information and extended citations to references (not all of which are repeated in Section 6.0 in this report).

The presence of trace elements in copper smelting processes is readily demonstrated through analyses of feedstock materials, collected flue dusts, emitted ambient particulates, and environmental media in surrounding areas. Early smelter studies documented the releases of multiple trace elements (see Harkins and Swain 1907; Glass 2003). Improved sampling and analytical methods in more recent studies have expanded the list of trace elements associated with non-ferrous smelters and copper smelters in particular (Parker 1978; Schwitzgebel et al. 1978; Bennett and Knapp 1989). Nelson (1977) lists antimony, arsenic, bismuth, cadmium, copper, indium, lead, selenium, tellurium, thallium, and zinc as elements associated with non-ferrous smelter feedstocks. Airborne sampling within a smelter plume, with trace element analyses of collected particulates, was performed by Chan et al. (1980). Small et al. (1981) performed in-plume sampling of ambient particulates downwind of five Arizona copper smelters, reporting enriched concentrations of numerous trace elements. Each smelter was shown to have a different pattern of elemental enrichment factors, reflecting smelter feedstock and process differences. Germani et al. (1981), in a companion paper, report on detailed trace element analyses of concentrates and captured or emitted particulates from five copper smelters. A general theory for the release of volatile chalcophilic trace elements from copper smelters is presented by Germani et al. (1981). Analyses of collected flue dusts from pollution control systems at copper smelters also document the occurrence of many trace elements (see Steele et al. 1994; Glass 2003).

Black & Veatch (1988b) provides a listing of 20 elements in addition to arsenic identified as potential airborne contaminants from copper smelting. These elements vary in their mass emission rates and comparative magnitude versus other sources. Several authors have provided reviews of trace elements associated with various sources including copper smelters (Lee and von Lehmden 1973; Chow 1995; Dudka and Adriano 1997; Nriagu and Pacyna 1988; Nriagu 1992; for arsenic, see Matschullat 2000). Copper smelting is generally identified as an important emissions source for trace elements. The U.S. Environmental Protection Agency's rulemaking for National Emission Standards for Hazardous Air Pollutants for Primary Copper Smelters notes that emissions from this source category include a number of trace elements listed as hazardous air pollutants (USEPA 1998a, 2001).

A number of reports provide information on trace elements associated with the Tacoma Smelter. The most extensive chemical analyses were performed by NEA, Inc. (1984) as part of a source apportionment study for the Puget Sound Air Pollution Control Agency (PSAPCA). Various materials from feedstocks to collected flue dusts, ambient particulates, and dustfall were analyzed for up to 30 elements; the analytical method used (XRF) had elevated detection limits that reduced the study's ability to quantify trace elements occurring at low concentrations (generally below several hundred parts per million). Nevertheless, numerous trace elements were identified in the various media sampled. A list of other source tests at the Tacoma Smelter is provided in Table 1. As noted on the table, many of the studies analyzed for a list of trace elements in addition to arsenic. Different studies targeted different lists of elements; most of the lists appear to have been chosen based on mass emissions rankings or estimates, or on elements of particular concern for human health. None of the trace element lists from these source tests should be considered to offer a complete characterization of elements released from smelter operations. Cumulatively, the source tests listed in Table 1 confirm smelter releases of a number of trace elements besides arsenic.

At various times, the USEPA, PSAPCA, and ASARCO developed mass emissions estimates for various elements from Tacoma Smelter operations. Table 1 also identifies a number of references providing such emission estimates, which were developed based on mass balance analyses or calculations from process source tests or emissions factors. With the exception of Varner (1983), these estimates are point-in-time annual emissions estimates rather than historical summaries. The references listed in Table 1 provide some information on about ten trace elements, but should not be considered complete either with respect to time (emissions histories) or the identification of all trace elements emitted by the Tacoma Smelter.

Many of the environmental studies of areas near the Tacoma Smelter focused on arsenic, lead, and cadmium as contaminants of concern. However, selected studies included an expanded list of trace element analytes (see Glass 2003). Ambient air particulates were collected by GCA (1974) and analyzed for 13 elements; strong correlations among pairs of elements and concentrations well in excess of normal urban values were found for multiple trace elements. McClannan (1974) measured eight trace elements in ambient particulates during strike and operating periods and found substantial increases when the Tacoma Smelter was operating. Heilman and Ekuan (1977) analyzed garden soils from 70 residential gardens for up to 37 elements and reported almost 20 elements had a concentration versus distance from the smelter pattern (approximately exponential decay curves). Black & Veatch (1988a) reported eight of thirteen trace elements analyzed in yard soil samples from Ruston and North Tacoma had a universal pattern of strong pairwise correlations, interpreted as showing a smelter source. Bechtel Environmental, Inc. (1992) similarly found elevated concentrations for six elements in addition to arsenic in soil samples. Fallout samples representing a four-month collection period (Roberts 1975) and an episodic release from a stack fire (Austin 1984) were analyzed for various trace elements (see Table 1), with deposition quantified. A series of regional precipitation chemistry investigations by researchers from the University of Washington also included analyses for multiple trace elements (see Larson et al. 1975; Knudson et al. 1977; Vong and Waggoner 1983; Vong et al. 1985; Vong et al. 1986; Moseholm 1986; Vong et al. 1987; Vong, Larson et al. 1988; Peterson 1991). In these precipitation chemistry studies a number of trace elements were found to be associated with the Tacoma Smelter.

Studies at a number of other smelters support the finding that multiple trace elements are associated with smelter emissions and are found at elevated concentrations in environmental media in surrounding regions (see Section 4.6 below and Glass 2003). Notable study locations include, among others, the Ronnskar, Sweden copper smelter (Nordgren et al. 1986; Johnson et al. 1992); the Bunker Hill lead smelter in Idaho (Roberts 1973; Ragaini et al. 1977); the San Manuel, Arizona copper smelter (Gabriel and Patton 1994, 1995); the Flin Flon copper/zinc smelting complex in Canada (Franzin et al. 1979; Zoltai 1988; Pip 1991; Hendersen et al. 1998; McMartin et al. 1999, 2002); a smelting region in northern France (Sterckeman et al. 2002); and the Kola Peninsula region of northwestern Russia (Niskavaara et al. 1996; Reimann et al. 1997; Gregurek et al. 1998, 1999; Reimann et al. 2000; Reimann 2002; Haugland et al. 2002).

The Tacoma Smelter Plume Site soil investigations included analyses for multiple trace elements in samples selected after the initial arsenic and lead results were reported (as described in Section 2.2.3). Samples included in the trace element studies were selected to meet several criteria, where possible: 1) a broad range of arsenic and lead concentrations; 2) spatial coverage of the entire Tacoma Smelter Plume region; 3) multiple depth intervals within borings (to account for different contaminant mobilities in the evaluations); 4) the inclusion of "anomalous" sample results (based on contaminant magnitude and location, or lead-to-arsenic ratio); 5) the inclusion of both forested (undisturbed) and developed (disturbed) sampling locations; and 6) some samples from multiple borings within one property (to evaluate local variability). After initial lab analyses of small groups of samples for approximately 35 elements, **antimony, bismuth, and indium** were selected as the primary target analytes for the bulk of the selected samples. Silver and mercury were also selected as additional target analytes for exploratory analyses using smaller numbers of samples. These five elements were among a group of ten or more that appeared to be effective tracers based on the analyses for the extended list of trace elements. The reduced list of elements analyzed for most trace element samples was required by budget limitations.

It should be noted that the selection of target elements was not based on their rankings using mass emissions. The best trace elements for smelter impacts in soils are those with the highest "signal-to-noise" ratios, and therefore take into account the emissions rate in comparison to typical (geochemical) background concentrations in soils and the likely emissions and soil impacts from other sources. Both the strength (enrichment over background) and the specificity (correlation with arsenic and other elements) of candidate trace elements were considered in selecting the primary target analytes. (Note that Sturges and Barrie [1989] concluded that indium was also the best tracer element for Canadian smelter impacts in ambient particulates, and that Gullu et al. [1995] identified a copper smelter factor contributing to ambient aerosol chemistry in upstate New York that included arsenic, antimony, and indium, among other tracers. Gabriel and Patten [1995] also showed large reductions in arsenic, antimony, and indium concentrations, among other elements, in honey mesquite leaves during an Arizona smelter strike versus an operating period).

Results of trace element analyses for 234 soil samples from King County (including Vashon-Maury Island and the mainland) have been evaluated (see Glass 2003). Those results are summarized here. Preliminary values for an additional 34 samples from Pierce County showed similar results. Evaluations of final trace element results for a total of 173 soil samples from Pierce County are expected to be completed by the end of 2003.

The King County trace element data set included 234 results for arsenic, lead, antimony, bismuth and indium; 78 results for silver; and 47 results for mercury. The range of soil arsenic concentrations included in this data set was 1.6 to 230 ppm (dry weight basis); the maximum arsenic value was about half the maximum concentration reported in the King County footprint studies. The 234 selected samples provided coverage of the entire King County study area (approximately 250 square miles). Additional results were obtained for eight samples from former orchard lands in Eastern Washington; all eight of those samples were analyzed for the extended list of trace elements, and thus had results for all seven elements noted above as primary target analytes for the King County samples.

Statistical correlation analyses of the King County trace element results were performed for both individual sample results (up to 234, depending on the pair of elements) and on maximum values from individual borings (up to 92). Data distributions were positively skewed; all of the data were log-transformed for correlation analyses. The results were similar and showed strong associations for additional trace elements with arsenic and with each other. The results based on maximum values within borings, which partly address different environmental mobilities for different elements in soils, are summarized here (figures and tables are from Glass

2003). Note that additional elements from the initial extended list also showed high correlations with arsenic, albeit based on a much smaller number of sample analyses.

The pairwise correlations for arsenic, lead, antimony, bismuth, indium, and silver (omitting mercury, for which a smaller number of results is available) are summarized in Table 5. The correlation coefficients range from 0.60 (arsenic versus lead) to 0.94 (bismuth versus indium); all are statistically significant at $p<0.00005$. Arsenic, antimony, bismuth, and indium exhibit particularly strong correlations; the three primary target trace elements thus appear well chosen. These strong correlations are illustrated by scatterplots (log-scaled) for arsenic versus antimony (Figure 5) and bismuth versus indium (Figure 6). The pattern of very high correlations among multiple trace elements commonly associated with copper smelting argues for a common source - the Tacoma Smelter - for soil contamination over the sampling region.

The very high correlations mean that the trace elements also show similar patterns for soil concentration versus distance (and direction) from the Tacoma Smelter. Thus, multiple elements have concentration gradients that decrease with distance and direction. This pattern can also be illustrated using scatterplots. Figure 7 shows the gradient for arsenic versus distance for trace element samples (compare to Figure 3, which shows the results for all locations in the two primary downwind directions). The similarity in patterns in Figures 7 and 3 indicates that despite the inclusion of a small number of "anomalous" sample locations within the trace element study, it is nonetheless representative of the data set for the entire study area. Figure 8 shows the similar spatial gradient for bismuth. These gradients for multiple trace elements commonly associated with copper smelting further confirm that the Tacoma Smelter is the source.

Finally, the inclusion of analyses for orchard soils allows comparisons to be made of the trace element patterns for soils where arsenic and lead occur based on agricultural practices (use of lead arsenate as a pesticide) versus smelter emissions. The trace element-to-arsenic ratios for antimony, bismuth, indium, and silver are shown to be substantially different in soils affected by these two sources (see Glass 2003), with the degree of difference increasing as arsenic concentrations increase. For example, compared to orchard soils, smelter-impacted soils are on average enriched in antimony and bismuth more than 6-fold at arsenic concentrations of 50 ppm and more than 10-fold at arsenic concentrations of 100 ppm. Thus, both the sampling design (targeting undisturbed forest locations) and these trace element source-discrimination results support a conclusion that the regional-scale soil contamination observed in King County is not attributable to the use of lead arsenate (see also Section 4.7).

4.6 CONSISTENCY WITH OTHER SMELTER STUDIES

Over the past 30 years, numerous studies of the environmental impacts in regions surrounding many smelters worldwide have been conducted. A review of that extensive literature supports the following general summary statement: elevated concentrations of multiple elements have been documented in multiple environmental media around large smelters, with spatial gradients in contamination showing approximately exponential decay in concentrations and a strong association with regional wind patterns. The various elements with elevated concentrations have shown a pattern of high correlations. The contaminated regions have been up to thousands of square km in size, with elevated concentrations extending to distances of tens of km from the smelter. These findings are consistent with the interpretations of data from Tacoma Smelter studies and the identification of the Tacoma Smelter as a primary source for regional soil contamination in central Puget Sound.

Table 2 lists selected studies of impacts near other smelters, from among a larger set compiled during the literature review performed for this credible evidence analysis. Over 100 reports for smelters in the U.S. and 15 other countries are included. These reports document similar patterns of contamination associated with smelter operations and emissions at major smelters worldwide. It is notable that several well-studied smelters are located in remote regions where other credible sources do not exist; the observed regional-scale environmental contamination surrounding such remote smelters is clearly the result of smelter emissions. Smelters that are most favorably located with respect to interpreting the results of environmental studies include, among others, those in Ronnskar, Sweden; Flin Flon, Manitoba, Canada; San Manuel, Arizona; the former Anaconda smelter in Montana; and the smelting complex on the Kola Peninsula, Russia.

The discussion here will not attempt an exhaustive listing of the results of all of the compiled smelter studies. Rather, several of the important features of contamination from smelter emissions will be highlighted and illustrative results from the literature cited in support.

Copper smelters are an important source of arsenic emissions. Elemental assays of ores, concentrates, and recovered flue dusts, as well as source tests of air emissions, document the presence of arsenic in smelting processes. Commercial supplies of arsenic are primarily produced as a by-product of non-ferrous smelting operations, including copper smelting (see USEPA 1998b and U.S. Bureau of Mines annual Mineral Facts and Problems). Thus, arsenic is especially associated with copper smelters processing high-arsenic materials, but it is also recognized in emissions from smelters not recovering arsenic as a by-product.

- In an early biomonitoring survey of children living near U.S. smelters, Baker et al. (1977) reported that children living near 10 of 11 U.S. copper smelters had elevated urinary arsenic levels compared to children living in 3 control communities without smelters. (Note: the Tacoma Smelter, already studied by Milham and Strong [1974], was not included in this nationwide survey).

- Beckman (1978) identifies the Ronnskar, Sweden copper smelter as a producer and major emitter of arsenic. It was built in about 1930 to process complex ores with high-arsenic content from northern Sweden. Beckman notes that historic emissions are uncertain, but for the period 1967-1978 are estimated at an average of 115 tons per year.

- Cumulative arsenic emissions from the Flin Flon, Canada smelter for the period 1931 to 1995 are estimated at over 4,000 tons (Bonham-Carter and McMartin 1997; McMartin et al. 1999).

- Chan and Lusis (1985) provide estimates of annual emissions of arsenic and other elements at the Sudbury smelting complex, based on source tests. For the period 1973 to 1981, annual arsenic emissions are estimated at 125 tonnes (1 tonne equals 1,000 kg or 1.1 tons).
- Newhook et al. (2003) characterize the ambient concentrations of arsenic (and other trace elements) near Canadian smelters. Arsenic values are consistently elevated compared to background levels. Based on this compilation of monitoring data and derived risk estimates, regions near copper smelters are identified as having comparatively high risk indices.

- Diaz-Barriga et al. (1993) state that the San Luis Potosi, Mexico smelter, located in an urban area, recovers arsenic as a by-product with a production capacity of 8,500 tons of arsenic trioxide per year. Elevated levels of arsenic in soil, dust, air, and children's urine and hair are reported indicating significant arsenic emissions occurred.

- Measurements of the arsenic content of ores, concentrates, recovered flue dusts, and emitted stack particulates at five "low-arsenic" copper smelters located in Arizona documented the significant enrichment of arsenic content in copper smelting processes (Germani et al. 1981). Measurements of the arsenic concentrations in ambient in-plume samples collected by aircraft downwind of five Arizona copper smelters documented an enrichment by more than two orders of magnitude above background values (Small et al. 1981).

- Galbraith et al. (1995) note that historic emissions estimates for the Anaconda, Montana smelter, which operated from 1884 to 1980, are incomplete, but cite an estimate that from 1911 to 1918 they may have been up to 75 tons per day.

Smelter emissions can be transported long distances. Sampling and chemical analysis studies to trace plumes as well as multivariate statistical evaluations of ambient particulate data sets demonstrate that smelter plumes can transport emitted contaminants long distances (hundreds of km).

- The Mount Isa, Queensland, Australia copper and lead smelters are located in a remote, undeveloped region. The absence of other sources facilitated the tracing of these smelter plumes for long distances. The smelter plumes were traced to distances of over 500 km under these favorable conditions (Bigg et al. 1978;

Carras and Williams 1981).

- Saltzman et al. (1986) collected and analyzed aerosol samples off the west coast of South America. Measurements of sulfate, arsenic, antimony, and selenium in collected samples were interpreted as identifying the smelting of sulphide ores as a likely source; such smelters are located at distance upwind (e.g., northern Chile).

- Gullu et al. (1995) analyzed ambient aerosol samples in upstate New York and identified a factor representing copper smelters, based on high factor loadings for arsenic, antimony, indium, cadmium, and the arsenic:selenium ratio. Canadian smelters located at distance to the north were implicated as the source contributing to this factor.

- Polissar et al. (2001) identify the Canadian smelters as the primary source for ambient arsenic measured in Vermont, based on multivariate analysis of chemical results and back-trajectory modeling.

- Sturges and Barrie (1989) measured lead isotope ratios and trace element concentrations at a rural sampling location in Ontario, Canada. The authors concluded that northern Canadian smelters were a contributing source to measured lead values, and that indium and arsenic were useful tracers of smelter impacts at large distances downwind.

- Eldred et al. (1983) reported that sulfate measurements at a number of sampling locations in the southwest U.S. (Arizona, New Mexico, Utah) during the period 1979 to 1981 were two to three times higher during periods of smelter operations compared to strike periods when most of the smelters were shut down. The sampling locations were hundreds of km downwind of the region where the copper smelters were located.

- Precipitation sampling at two locations in southeastern Arizona, located from less than 100 to about 400 km from regional copper smelters, during 1984 to 1985 showed an association of low pH and high sulfate concentrations when winds were from the direction of the smelters (Blanchard and Stromberg 1987). The authors identified a smelter "fingerprint" based on arsenic, antimony, cadmium, copper, lead, and zinc concentrations in collected precipitation.

<u>The spatial extent of contamination from smelter emissions can be regional in scale (extending tens of km from the source)</u>. Monitoring studies of sufficient scope have documented the large spatial scale of environmental contamination from smelter emissions. Smelter impacts have a general spatial pattern of sharply decreasing concentrations with increasing distance from the smelter; some studies have focused only on the areas closer to the smelter where larger

effects occur, and have not attempted to define the full spatial extent of smelter impacts.

- Cartwright et al. (1976) surveyed contaminant concentrations in surface soils near the Port Pirie, Australia lead smelter. Soil contamination decreased exponentially with distance and was detectable to 40 to 65 km depending on direction. Topographic elevation was also found to influence soil contamination levels. The Port Pirie smelter is described as an isolated plant located in an otherwise non-industrial region (mostly agricultural land use). Total atmospheric deposition within the 5,000 square km study area was estimated at 30,000 tonnes of lead and 300 tonnes of cadmium. Koh and Judson (1986), in a study of sheep near Port Pirie, reported similar spatial patterns and cite an estimate of total lead deposition of 40,000 tonnes in a contaminated region of at least 3,400 square km. Langley (2002) provides an estimate of 80,000 tonnes of lead deposited to land and detectable soil impacts as far as 60 km downwind of the smelter.

- Reif et al. (1989) measured contaminant concentrations in soil and in sheep near the La Oroya, Peru zinc smelter. Gradients in contaminant concentrations were reported for distances up to 56 km from the smelter.

- Nordgren et al. (1986) sampled soils in coniferous forest stands in the region surrounding the Ronnskar, Sweden copper smelter. Samples were collected in two different directions from the smelter to maximum distances of 40 and 55 km. The soil samples were analyzed for multiple trace elements. Soil contaminant concentrations showed a strong spatial pattern with approximately exponential decay to at least the largest distance sampled, 55 km. Johnson et al. (1992) reported a strong gradient in lake sediment contaminants for 12 sampled lakes at distances up to 80 km from the Ronnskar smelter.

- Lobersli and Steinnes (1988) studied soil and vegetation trace element concentrations along two transects from the Sulitjelma, Norway copper smelter. The maximum distance for sampling locations was 27 km in the primary downwind direction. Environmental contamination was shown to follow the prevailing wind direction and was still considerable at the largest sampling distance of 27 km. Several contaminants showed an exponential decrease in concentrations with increasing distance from the smelter.

- Impacts from the smelter at Flin Flon, Manitoba, Canada have been monitored in a series of studies. Franzin et al. (1979) estimated the annual deposition of trace elements, including arsenic, within about 250 km of the smelter based on bulk precipitation samples collected for one year or on snow samples from frozen lake surfaces. Inverse correlations of contaminant concentrations versus distance were found. Assuming baseline deposition rates per metal, affected distances were estimated from a power curve model of the concentration versus distance relationship and ranged from about 30 km to more than 200 km. Total deposition amounts within the affected distances ranged from a few tons (arsenic) to over one thousand tons (zinc). Zoltai (1988) reported exponentially decreasing concentrations of arsenic and other metals in surface peat layers up to 100 km from the smelter. Henderson et al. (1998) performed geochemical mapping for humus and till samples in the region around the Flin Flon smelter. Arsenic and other trace elements associated with the smelter showed a "bulls eye" pattern centered on the smelter, with spatial gradients decreasing with distance following a power law form. Estimated distances to reach background concentrations varied by element from 70 to 104 km. The spatial gradients extended farther in the distances of the prevailing winds.

- Dumontet et al. (1992) studied soil trace element concentrations along two downwind transects from the Rouyn-Noranda, Canada copper-zinc smelter. Samples were collected to distances of 27.5 and 42.5 km along the two transects. A gradient of decreasing concentrations with increasing distance was found to the maximum distances sampled. Similar results were reported for sampling in peat along the same transects (Dumontet et al. 1990).

- McMartin et al. (2002) summarize humus sampling results and gradients near the Trail, British Columbia, Canada smelter in comparison to those for the Flin Flon and Rouyn-Noranda smelters. Contaminant concentration data from all three smelter regions fit an inverse power law function with affected distances measured in tens of km from the smelter.

- Impacts of the large smelting complex at Sudbury, Canada have been studied repeatedly. A large area of vegetation damage, over 2,000 square miles in extent, has been reported (Hutchinson and Whitby 1974). An early study confirmed strong spatial gradients for contaminant concentrations in soils and collected rain water, indicating metals deposition had occurred to distances in excess of 50 km (Hutchinson and Whitby 1974). A later study reported surface soils showed elevated levels up to 70 km from the smelters (Freedman and Hutchinson 1980), and estimated that less than half of the nickel and copper emitted were deposited within a distance of 60 km, based on dustfall-rainfall collection and snow sampling.

- Beyer et al. (1984) found spatial gradients for zinc and cadmium that were strongly decreasing with increasing distance from two zinc smelters at Palmerton, Pennsylvania. The authors note that the concentration versus distance gradients extended to sampling locations up to 100 km from the smelters.

- Studies at the Anaconda, Montana smelter site confirmed widespread regional soil contamination. Soil arsenic background concentrations were determined to be 6 to 16 ppm. A kriging evaluation of soils data collected over an area of about

300 square miles estimated surface soil arsenic levels in over 3,000 cells of 70 acres each (over 330 square miles) to be between 29 and 1,856 ppm (i.e., all above background levels). The pattern of kriging results matched primary wind directions, with valley channeling in this region of mountainous topography (USEPA, Region VIII 1996).

- Blais et al. (1999) analyzed sediments from ten lakes within a radius of 110 km from the large smelting complex at Nor'ilsk, northern Siberia, Russia. A spatial pattern of elevated metals concentrations was reported for distances at least up to 60 km from the smelters. Comparisons of top and bottom sediments from the collected cores also showed significant enrichment factors for multiple elements in the region up to at least 60 km from the smelters.
- The Kola ecogeochemistry project conducted extensive sampling to document the regional distribution of a number of elements in various environmental media in a large region (188,000 square km) surrounding the complex of smelters on the Kola Peninsula, Russia (Reimann 2002). Transboundary impacts from Russia to northern Finland and Norway occur from these smelters. The distances to background levels for various elements in sampled media are up to 100 km or more (Reimann 2002), depending on wind direction. The contaminant concentrations decrease following a power law function with increasing distance from the smelters; the estimated cumulative deposition to distances of 100 to 200 km approaches cumulative smelter emissions (Reimann et al. 2000).

<u>Multiple elements associated with smelting operations are found at elevated concentrations in surrounding regions, and are highly correlated</u>. Smelters are known to emit a number of volatile elements occurring in ores and concentrates (see Glass 2003; Germani et al. 1981). Studies of environmental impacts in areas near smelters have selected different groups of elements for analysis. Most studies have included multiple smelter-related elements, but relatively few have attempted to monitor extended lists of trace elements associated with smelting and emitted in lesser quantities. In nearly all studies, the spatial patterns of contamination shown by multiple smelter-related elements are highly similar, reflecting strong correlations among those elements. The strong correlations and similar spatial patterns, as well as identification of the elements in smelter emissions, are interpreted as indicating a common source.

- Beavington (1975) reported highly significant correlations among copper, zinc, lead, cadmium, and nickel in garden soil and vegetable samples collected near the smelter at Wollongong, New South Wales, Australia.

- Nordgren et al. (1986) found that arsenic, copper, lead, zinc, nickel, and selenium, among other elements, showed very similar spatial patterns of decreasing concentrations with distance. These elements were grouped together in a multivariate statistical analysis in a common factor reflecting soil contamination levels.

- Sterckeman et al. (2002) sampled tilled agricultural plots around the Noyelles-Godault lead-zinc smelter in northern France. They reported similar spatial patterns for silver, arsenic, bismuth, cadmium, copper, mercury, indium, lead, antimony, tin, thallium, and zinc. All of these elements were also found to be elevated in samples of collected dusts from two emissions control systems at the smelter.

- Ball et al. (1983) evaluated the spatial distributions of arsenic and antimony in soils surrounding the Garfield, Utah copper smelter. The authors conclude: "A strong linear relationship between arsenic and antimony concentrations suggests a similar source and mode of dispersion for the two contaminants" (page 347).
- Ragaini et al. (1977; see also Roberts 1973) analyzed surface soils and ambient particulates collected near the Kellogg, Idaho lead smelter for more than 30 trace elements. Elements with high enrichment factors included cadmium, antimony, silver, lead, gold, zinc, selenium, arsenic, indium, nickel, copper, and mercury. The ranking of elements by enrichment factors was very similar for soils and ambient particulates, and the elements varied synchronously over time with lead in ambient particulates. These findings indicated the smelter was the source.

- Gabriel and Patten (1994) measured 27 elements in soils and honey mesquite in the region surrounding the San Manuel, Arizona copper smelter. A covarying group of elements including copper, arsenic, and antimony showed high pairwise correlations in soil samples. In honey mesquite bark samples, indium joined copper, arsenic, and antimony in a covarying group of elements. Those four elements also showed a statistically significant relationship with distance from the smelter for bark samples and were identified as particularly important for identification of the smelter as a source.

- McMartin et al. (1999) describe the regional soil contamination near the Flin Flon, Manitoba, Canada smelter by arsenic, cadmium, copper, mercury, lead, and zinc. The general spatial gradients for these six elements are similar; the authors suggest that differences in the slopes of the decay curves mirror the rankings of the elements' volatilities.

- As already noted, Germani et al. (1981) and Small et al. (1981) documented the enrichment of multiple elements in feedstocks, collected flue dusts, and in-plume ambient samples at five Arizona copper smelters. Elements associated with sulfur (chalcophilic elements) are noted as being strongly enriched at smelters and include copper, zinc, arsenic, selenium, silver, cadmium, indium, antimony, tungsten, and gold (Small et al. 1981).

- Many of the Kola Peninsula, Russia studies included analyses for multiple elements. In an early pilot study, Niskavaara et al. (1996) analyzed up to 40 elements for samples from 10 different environmental media over a region of 12,000 square km. They report that the area of contamination was clearly outlined for a number of elements, including copper, arsenic, nickel, cobalt, antimony, and cadmium, among others.

Modeling studies of smelter impacts are consistent with environmental measurements. Mathematical models of the air dispersion and atmospheric deposition of smelter-emitted contaminants have been developed for several smelters. The results of such models provide another approach to understanding the scale of smelter impacts.

- Hellstrom (1980) modeled the impacts of Ronnskar, Sweden copper smelter emissions of lead, copper, and cadmium on the surrounding region. Atmospheric deposition by wind direction was modeled to a distance of 70 km. Within that region about 60 percent of the emissions are estimated to be deposited (more for larger particle size fractions, less for smaller particle size fractions). The gradients for concentration versus distance were approximately exponential decay functions. Modeled values were compared against measured snowpack values, showing reasonable agreement.

- Godin et al. (1985) estimated a model for soil contaminant concentrations near the Noyelles-Godault lead-zinc smelter in northern France, based on soil sampling and analysis results. Concentrations were found to be inversely proportional to distance from the smelter and proportional to the square root of the wind frequency. The model was then used to estimate cumulative impacts from multiple sources (two lead-zinc smelters, a coal-fired power plant, and a waste incinerator) and the mass deposition from individual sources. Contaminant contour maps based on model results showed good agreement with measured soil values (in tilled agricultural soils). Estimated total deposition was 10,000 tonnes for lead (1 tonne equals 1,000 kg) and 100 tonnes for cadmium.

- Rupp et al. (1978) applied atmospheric transport and deposition models to estimated cadmium emissions from the East Helena, Montana lead smelter. Impacts to ambient air concentrations and deposition to soils were modeled as a function of particle size to distances of 8 miles from the smelter. Both measures were shown to sharply decrease with increasing distance from the smelter. Comparisons with monitoring data showed the model produced reasonable results.

- Dillon (1995) applied air dispersion modeling to the roaster stack emissions at the Yellowknife, Northwest Territories, Canada gold mining operation. SO2 and arsenic ambient air concentrations were modeled and compared to monitoring results, with reasonable agreement. Areas exceeding regulatory standards were delineated using the model and were centered on the roaster stack.

Chronologic studies demonstrate the large contributions of smelters. Several studies report on environmental measurements during operating versus strike periods at copper smelters. Age-dated sediment cores have also been evaluated for contaminant profiles in relation to smelter operating histories. These results provide chronologic evidence for smelter impacts.

- Ambient air samples collected 13 km downwind of the Caletones, Chile copper smelter, at a rural site, showed a large increase in arsenic concentration during smelter operations versus during a strike period (Romo-Kroger et al. 1994).

- Gabriel and Patten (1995) measured trace elements in honey mesquite leaves along distance and elevation gradients around the San Manuel, Arizona copper smelter. Samples were collected and analyzed during periods of smelter operation and periods of shutdown due to a labor strike. The elements copper, arsenic, antimony, tungsten, and indium showed concentrations substantially reduced, by up to 95 percent, during the strike period compared to the operating period.

- Nriagu et al. (1982; see also Nriagu 1983) determined contaminant concentration profiles in age-dated freshwater lake sediment cores in the region near the Sudbury, Ontario, Canada smelting complex. The authors relate the magnitudes and depth profiles of nickel, copper, lead, cobalt, and zinc in the sediment cores (by distance from the smelter and by depth) to the history of smelting activities in the region and the characteristics of smelted ores, finding reasonable correspondence.

- As noted above, sulfate concentrations in the southwestern region increased significantly when smelters were operating versus when most were shut down by a labor strike (Eldred et al. 1983).

Multiple environmental media show similar patterns of contamination near smelters. Sampling has been performed in a variety of environmental media to evaluate smelter impacts, including ambient particulates, rainfall, snow, dustfall, humus, soil, peat, vegetation (moss, tree needles, garden vegetables, grass), stream and lake water, stream and lake sediments, and human and animal tissues (biomonitoring). Contaminant concentrations that decrease with distance from the smelter have been reported for numerous types of sampling. At a number of smelters studies of multiple environmental media have been performed.

- At the Flin Flon, Canada smelter impacts have been monitored through bulk precipitation and snow samples (Franzin et al. 1979; Phillips et al. 1986), forest humus and soils (Hogan and Wotton 1984; Bonham-Carter and McMartin 1977;

Henderson et al. 1998), peat (Zoltai 1988), deer mice and blueberries (cited in Pip 1991), garden soils and produce (Pip 1991), and lake sediments (cited in Henderson et al. 1998).

- Environmental sampling near the Port Pirie, Australia smelter has included, among other media, soils (Cartwright et al. 1976), wheat crops (Merry et al. 1981), and sheep tissues (Koh and Judson 1986).

- The region around the Sudbury smelting complex has been extensively studied. Sampled media include, among others, mineral soils (Hutchinson and Whitby 1974, 1977; Rutherford and Bray 1979; Freedman and Hutchinson 1980; Hazlett et al. 1983; Dudka et al. 1995; Ontario Ministry of the Environment 2001; Adamo et al. 2002), wetland soils (Taylor and Crowder 1983), native vegetation (Hutchinson and Whitby 1974, 1977; Freedman and Hutchinson 1980; Hazlett et al. 1983; Ontario Ministry of the Environment 2001), bog vegetation (Arafat and Glooschenko 1982), dustfall and rainfall (Hutchinson and Whitby 1977; Freedman and Hutchinson 1980; Jeffries and Snyder 1981; Chan et al. 1982), snow (Freedman and Hutchinson 1980), lake sediments and suspended particulates (Nriagu et al. 1982; Nriagu 1983), and lake water (Nriagu 1983).

- Studies of the Kola Peninsula, Russia smelting complex have included sampling, among other media, of rainfall (Karaban and Gytarsky 1995), native vegetation (Kozlov et al. 1995; Niskavaara et al. 1996; Rautio et al. 1998; Steinnes et al. 2000; Haugland et al. 2002), humus and moss (Ayras et al. 1997), snow (Niskavaara et al. 1996; Barcan and Sylina 1996; Gregurek et al. 1998; Gregurek et al. 1999), soil (Niskavaara et al. 1996; Reimann et al. 1997; Barcan and Kovnatsky 1998; Steinnes et al. 2000; Haugland et al. 2002), and stream water and sediment (Niskavaara et al. 1996).

4.7 ABSENCE OF OTHER HIGH-EMISSIONS SOURCES

The Tacoma Smelter, which for many years was the sole U.S. producer of arsenic, was generally recognized as the largest single arsenic emissions source in the country. Studies of the impacts from Tacoma Smelter operations often have used arsenic as the primary tracer element. Cumulative emissions were on the order of tens of thousands of tons (see Varner 1983 and Section 4.2). A semi-quantitative estimate of the excess arsenic in surficial soils in the region surrounding the Tacoma Smelter, based on the data collected in the Tacoma Smelter Plume footprint studies, readily demonstrates that a source (or sources) needs to account for thousands of tons of arsenic deposition.

A rationale for identifying the Tacoma Smelter as the primary source of regional soil contamination is the absence of other credible sources of sufficient magnitude to produce impacts on such a regional scale. Nriagu and Pacyna (1988) have summarized information on worldwide arsenic emissions from various industries. The largest emissions are from pyrometallurgical non-ferrous smelters, and within that class the most important type is copper/nickel smelting. The USEPA (1982) provided information on all U.S. copper smelters, among other identified sources for arsenic emissions. The Tacoma Smelter feedstock averaged 3.8 percent arsenic, more than an order of magnitude larger than at any other U.S. copper smelter. Considering both feedstock arsenic content and copper production capacity, the Tacoma Smelter was noted to have more arsenic throughput than all other U.S. smelters combined - in fact, more than five times as much. USEPA (1998b) has also summarized information on arsenic emission factors for a large number of potential sources. These studies provide supporting information to identify the Tacoma Smelter as a uniquely large arsenic emissions source in the U.S.

The Puget Sound Air Pollution Control Agency (now the Puget Sound Clean Air Agency) compiled information on air emissions of toxic chemicals from sources within its four-county jurisdictional region. Recent arsenic emissions from all other listed sources were trivial compared to the emissions from the Tacoma Smelter when it was operating (e.g., see PSAPCA 1993 for 1989 Toxic Emissions Inventory). Reporting under the Toxic Release Inventory provisions of Superfund (Section 313 of the Emergency Planning and Community Right-to-Know Act of 1986) also shows that there are no other significant current sources of air emissions in the region.

The Historic Research Associates (HRA), Inc. study (Mighetto and Schnaiberg 2001) was performed to review possible historic sources of arsenic with respect to their potential for regional-scale impacts. As part of the scoping process for that study, numerous references were reviewed to identify historic arsenic uses and emissions sources (including, for example, Tyler and Petar 1934; Walsh et al. 1979; Encyclopedia Britannica 1980; Lederer and Fensterheim 1983; Nriagu and Pacyna 1988; Loebenstein 1994; USEPA 1998b; the U.S. Bureau of Mines periodicals Minerals Yearbook and Mineral Facts and Problems; standard chemical references such as the Kirk-Othmer Encyclopedia of Chemical Technology and Ullmann's Encyclopedia of Industrial Chemistry; and compiled literature reports of arsenic impacts from specific industries or uses). HRA performed a general review of Seattle's industrial development but focused on four source categories: agricultural use of arsenical chemicals in Western Washington, and especially on Vashon-Maury Island; coal use; the metals industry, particularly primary and secondary non-ferrous smelters; and steel mills.

Many different uses of arsenic were identified - for example, various herbicide or pesticide uses based on its toxicity, and multiple uses as a pigment or alloying agent based on its chemical properties. The number of possible sources for arsenic emissions, including a wide range of emission factors, was also large (see USEPA 1998b). It is certain that some arsenic releases occurred from these types of uses and sources in the central Puget Sound region. The review of possible historic sources of arsenic contamination, however, did not identify any arsenic emissions sources or uses that could account for the observed spatial scale and magnitude of regional soil contamination. The review of other possible sources will be discussed under three headings: impacts from the large collection of typical activities in urban regions (the "urban plume" effect, including direct applications to soils as well as air emissions); specific categories of air emissions sources; and land applications of arsenic including uses in agriculture and silviculture.

The areas downwind of the Tacoma Smelter include some rural locations such as Vashon-Maury Island and Fox Island, but much of the mainland region is in the heavily urbanized Seattle-Tacoma corridor. The cumulative effects of many contaminant sources in urban areas, including those that are of small magnitude but widespread (e.g., area sources), have been shown to result in increased urban contamination levels for some contaminants. Detailed sampling studies have documented this effect for lead (see Mielke, Adams et al. 1989; Mielke 1991, 1999; Mielke, Gonzalez et al. 1999). A number of other elements have also been studied in investigations of the urban-to-rural gradients in contamination (e.g., Grigal and Ohmann 1989; Pouyat and McDonnell 1991; Fergusson and Stewart 1992; Callender and Rice 2000) and found to exhibit such gradients. One U.S. study was identified that included arsenic among the elements analyzed. Rice (1999) reports the results of a U.S. national streambed sediment sampling program (541 sediment samples from 20 regions) in which nine elements were analyzed. Seven elements - cadmium, chromium, copper, lead, mercury, nickel, and zinc - were enriched in samples from urban areas compared to agricultural or forested areas. Arsenic and selenium were not comparatively enriched in urban areas. The 50th and 75th percentile values for all arsenic data were 6.3 and 9.2 ppm, respectively; Rice (1999) suggests that this reflects some degree of enrichment compared to geochemical values (average crustal abundance). However, the lack of a gradient for sediment arsenic concentrations indicates that the concentration of residential and industrial land uses in urban areas, with all of the consequent uses and emissions of arsenic, does not represent a significant cumulative arsenic source on a regional scale (in contrast to other elements such as lead). Typical urban emissions sources would include, among many others, the use of coal and oil for residential heating, motor vehicle emissions, and the distributed use of wood treated with copper chromated arsenic (CCA).

Birke and Rauch (2000) performed an extensive investigation of urban geochemistry in the metropolitan Berlin region, with several thousand soil samples collected and analyzed. Arsenic was included among numerous trace elements analyzed. The median arsenic concentrations for a variety of land uses, from industrial to high-density and low-density residential, agricultural, and woodlot, were all in the narrow range of 2.8 to 4.3 ppm. This pattern was quite different from that of lead, or other elements where urban emissions showed large effects. The detailed urban geochemistry characterization of Berlin thus supports the findings of Rice (1999) that typical urban sources of arsenic do not result in notable urban-to-rural gradients.

Ecology's statewide survey of soil background concentrations (conducted cooperatively with USGS) did not include focused sampling in urban areas. No general survey of urban soils in Washington has been performed, but small data sets are available for some areas (e.g., Port

Angeles and Bellingham; see the discussion in Washington State Department of Ecology 1999, Volume II, pages B-111 et seq.). One Seattle study included arsenic analyses of soils in recycling yard setouts in multiple residential neighborhoods and reported that 24 samples ranged from 6.9 to 18.6 ppm (Herrera Environmental Consultants, Inc. and Glass 1994). The results from Washington studies in urban areas consistently have shown arsenic concentrations to be below 20 ppm. That general finding is also supported by studies in other cities. Carey et al. (1980) report results for five U.S. cities; data categorized as urban or suburban sampling locations, and from well-maintained lawns or unmaintained soils, from all five cities demonstrate some variability but all have geometric mean values for arsenic below 20 ppm. Studies using various sampling designs report mean soil arsenic of 10.2 ppm in Christchurch, New Zealand (Fergusson et al. 1986); 2.8 ppm (median) in Ottawa, Canada (Rasmussen et al. 2001); 3.42 ppm (in street sediment) in Sault Ste. Marie, Canada (Stone and Marsalek 1996); and 3.9 ppm (median) in Berlin, Germany (Birke and Rauch 2000). Thus, urban soils appear to be only marginally contaminated by arsenic in the context of the much higher regional soil contamination reported in the Tacoma Smelter Plume studies. This supports a finding that typical urban sources are not major contributors to that regional arsenic contamination.

Air emissions of arsenic typically result from high-temperature processing of materials with measurable arsenic content. The arsenic volatilizes and is only partially captured by pollution control devices such as electrostatic precipitators or fabric baghouses. Therefore, factors such as the arsenic content of processed materials, mass throughput, effectiveness of control systems, and stack gas release characteristics (height, temperature) largely determine the magnitude and impacts of an air emissions source. Among the high-temperature processes that can be considered are primary and secondary smelters, incinerators (burning municipal solid waste, biomass/wood, or medical wastes), steel mills and foundries, cement plants, coal and fuel oil combustion (external combustion in large utility boilers, industrial boilers, historic gasworks plants, or distributed residential areas), and motor vehicle gasoline combustion (internal combustion), as well as smaller and less significant sources such as rockwool insulation plants, chemical manufacturing plants, munitions plants (lead shot), glass manufacturing, brickworks, and crematoria.

A recent report summarizes air emissions factors for many of these potential arsenic sources (USEPA 1998b). A nominal historic arsenic emission factor for the Tacoma Smelter can be developed based on about 300,000 to 400,000 tons of feedstocks processed annually at an arsenic content of about 4 percent, with arsenic air emissions in the range of 100 tons per year to several hundred tons per year (Varner 1983). The result is an arsenic emissions factor in the range of 0.5 to 1 lb/ton feedstock, or more. The USEPA arsenic emission factors for other high-temperature processes are several orders of magnitude smaller, and this difference is not made up by larger mass throughputs. Thus, other identified sources have annual emissions that are only a very small fraction of those associated with Tacoma Smelter emissions.

Lead batteries used by motor vehicles typically include a small percentage of arsenic.

Secondary lead smelters that reprocess such batteries therefore are widely recognized as a source of arsenic emissions. Numerous investigations of secondary lead smelters are reported in the literature. Typical arsenic-to-lead ratios in feedstock materials, impacted soils near the plants, and ambient particulates near operating facilities are summarized by Kimbrough and Suffet (1995) and Eckel et al. (2002). The typical ratios reported by these authors, on the order of 10:1000 to 50:1000, are consistent with data from the Harbor Island (Seattle) secondary lead smelter site (see NEA, Inc. 1982). They are far smaller than the arsenic-to-lead ratios associated with the Tacoma Smelter, where arsenic emissions exceeded lead emissions (see Varner 1983) except during the initial years of operation as a lead smelter. With smaller annual throughput, much lower mass arsenic emissions, and shorter stacks, secondary lead smelter impacts on surrounding soils are limited spatially in comparison to those of the primary copper smelter in Tacoma (see Roberts, Hutchinson et al. 1974; Roberts, Gizyn et al. 1974; Linzon et al. 1976; Temple et al. 1977; Linzon 1982; Bisessar 1982; Brown et al. 1985; Sturges and Harrison 1986; Small et al. 1995; Rieuwerts and Farago 1996; Farago et al. 1999; Rieuwerts et al. 1999).

Coal was widely used for residential and commercial heating in the early 1900s. It also fueled various industrial processes, including gasification plants. The Seattle-Tacoma metropolitan corridor was like most other developed urban areas in its use of coal as a primary energy source at that time. The cumulative impacts from arsenic emissions from such coal use would already be represented in urban soil arsenic levels, which as noted are only marginally elevated above rural levels on average. While coal sources vary in their arsenic content, occasionally exceeding 100 ppm, most coals - especially western coals - have concentrations below 10 ppm (Mighetto and Schnaiberg 2001). Assuming coal used in Seattle was not high-arsenic, that per capita consumption for all uses was about 3 tons per year (see Crecelius and Piper 1973; this per capita estimate is consistent with coal production and use figures given by Mighetto and Schnaiberg 2001), that a representative early 1900s population was around 200,000, and that 10 percent of the arsenic content of the coal was emitted (with the remainder ending up in the ash), an estimate of annual arsenic emissions from coal use can be calculated. The result is less than 1 ton per year compared to hundreds of tons per year from the Tacoma Smelter (see Varner 1983). If all of the emitted arsenic from coal use was evenly deposited over a Seattle urbanized area of 50 square miles (and a substantial fraction would likely be transported over greater distances) and incorporated into the top six inches of the soil profile, the resulting increase in soil arsenic concentration from 25 years of coal use would be on the order of 10 ppm, consistent with the reported typical urban values of under 20 ppm (including a background geochemical contribution on the order of 5 ppm). Locations of more intensive coal use (e.g., gasification plants or cement kilns) could have had somewhat larger impacts on a localized area.

Coal-fired power plants have been noted as an important source category for cumulative emissions of trace elements, including arsenic. The total estimated emissions from this category reflect the large number of facilities; emissions from an individual plant may be comparatively modest. Numerous reports discuss arsenic associated with coal-fired power plant operations (e.g., see Zoller et al. 1974; Klein et al. 1975; Swaine 1977; Coles et al. 1979; Ondov et al.

1979a, 1979b; Smith et al. 1979; Wangen and Turner 1980; Wangen 1981; Germani and Zoller 1988; see Lind et al. for similar discussion of a plant fueled by biomass). There is one coal-fired power plant (two units) in western Washington, at Centralia. That plant has been in operation only since the early 1970s, and thus has an emissions chronology that does not match the findings of chronologic studies (see Section 4.4). The reported air emissions of arsenic from the Centralia plant are small, less than 100 pounds per year.

Motor vehicles are not generally identified as a substantial source for arsenic air emissions (ignoring resuspended dusts from traffic). Arsenic is not among the elements found to be significantly enriched near heavily-traveled roadways; it was not included among 28 trace elements analyzed in motor vehicle emissions by Ondov et al. (1982). The low emissions reflect low arsenic content in vehicle fuels, a situation quite different than lead (see Mielke et al. 1989; Mielke 1999). Since urban areas have substantially higher traffic densities than outlying areas, and since urban-to-rural gradients in soil and sediment arsenic are lacking, motor vehicles are confirmed not to be an important arsenic emissions source. PSAPCA's Toxic Emissions Inventory for 1989 lists only 221 pounds of arsenic emissions for all of King County from motor vehicles (PSAPCA 1993). Scattarella (1988) cites data from a study by Kalman and Larson of the University of Washington in which ambient air samples were collected in Seattle above the eastbound on-ramp of the Evergreen Point floating bridge across Lake Washington and analyzed for trace elements including arsenic. The lead-to-arsenic ratios in those samples were more than one to two orders of magnitude higher than similar ratios for the Tacoma Smelter. This also supports a finding that motor vehicle emissions are an insignificant source for arsenic.

Seattle has some history in the first half of the twentieth century of using open burning and incineration for handling municipal solid waste. Proposals to construct large municipal solid waste incinerators in Seattle in the last quarter of the century were not brought to completion. Refuse incineration has been shown to be a source for arsenic and other trace elements (see Greenberg et al. 1978; Law and Gordon 1979). In New York City, a review of the historic use of incinerators and chronologic contaminant profiles in lake sediments suggested that incinerators may have been under-recognized as an emissions source for lead and other trace elements (Chillrud et al. 1999; Walsh et al. 2001). Early applications of incineration with less developed pollution control systems may have had larger arsenic emission factors than those given by USEPA (1998b). A study by Bache et al. (1991) reported on trace metals contamination in the area surrounding a municipal refuse incinerator which operated without particulate control devices. Samples were collected upwind and downwind of the facility. Comparisons showed that impacts for multiple elements were largely localized within about 1 km of the incinerator after 7 years of operation. The cumulative effects of open burning and incineration of solid waste in Seattle and other cities would be included in urban soil sampling results. As with impacts from historic coal burning and motor vehicle emissions, historic incineration practices do not appear to have produced significant regional-scale increases in soil arsenic levels.

Two large cement plants are located in South Seattle. The PSAPCA 1989 Toxic Emissions Inventory does not list any arsenic emissions from those facilities (PSAPCA 1993). A review of PSAPCA files for the Seattle cement kilns located some test burn reports and other information that confirmed maximum arsenic emissions of only a few pounds per year, consistent with the USEPA (1998b) arsenic emission factors.

Several steel mills have operated in Seattle for long periods of time (see Mighetto and Schnaiberg 2001). Among national steel mills, they are characterized as small to moderate in capacity. PSAPCA's Toxics Emissions Inventory for 1989 lists only a de minimis quantity (8 pounds) of arsenic emissions for the largest of the three Seattle steel mills (PSAPCA 1993). This is consistent with the arsenic emission factor given by USEPA (1998b). The only other arsenic emissions source for King and Pierce Counties listed by PSAPCA (1993) is residential wood combustion, with a two-county total of under 500 pounds for 1989.

Additional high-temperature processes with potential arsenic emissions, including among others small-scale gold and silver smelting (see Mighetto and Schnaiberg 2001), iron foundries, manufacture of metal alloys, or a Tacoma rock wool insulation plant using smelter slag as a feedstock material, may have resulted in moderate localized impacts but were too small to have regional-scale effects.

Arsenic impacts in soils can result from various uses of arsenical chemicals as well as from high-temperature volatilization, transport, and deposition processes. The design of the footprint studies for the Tacoma Smelter Plume Site focused on relatively mature (second-growth) forested areas where most direct applications of arsenical chemicals would be precluded; this study design was adopted to minimize artifacts affecting data interpretations. The possible influence of agricultural and silvicultural uses of arsenical chemicals was nevertheless considered.

Most of the forested sampling locations in the footprint studies are believed to have regenerated naturally rather than through intensive silvicultural management. Arsenical chemicals (cacodylic acid and monosodium methanearsonate) are known to have been used in managed forests for thinning and insect control (Newton 1986). The widespread use of such chemicals at footprint soil sampling locations is considered very unlikely and in any event could not account for the strong gradients and spatial patterns observed (see Section 4.3). Two studies of the changes in soil arsenic concentrations after use of these arsenical chemicals showed them to be very small (Norris et al. 1983; Newton 1986). Thus, silvicultural uses of arsenical chemicals can be eliminated as an alternate source of the observed regional-scale contamination.

Agricultural uses of arsenical chemicals as insecticides and herbicides were widespread from the late 1800s through about 1950. In that period the largest quantity uses of arsenic were for agricultural chemicals. Their extensive use in eastern Washington orchards, resulting in continuing elevated soil arsenic concentrations today, is well-known. HRA's historic study of arsenic also documented many specific recommendations for the use of these arsenical chemicals in western Washington (see Mighetto and Schnaiberg 2001). Vashon-Maury Island has a history of extensive commercial berry farming; a variety of other types of farming occurred on a smaller scale. Specific recommendations for use of arsenicals on berry crops were documented by HRA (Mighetto and Schnaiberg 2001). There are anecdotal reports from some Island residents that arsenical chemicals were widely used on Vashon-Maury Island, and one interesting comment that in some areas the heavy contamination from the smelter made use of chemical insecticides unnecessary (Eernisse 1976). It is possible that some of the second-growth forested areas sampled in the Vashon-Maury Island footprint study had previously been used for agriculture, although many probably were not. A 1941 University of Washington thesis by Cole included a map of land uses on Vashon Island in 1939; that map is reproduced as Appendix 1 of the HRA report (Mighetto and Schnaiberg 2001). That map shows that the pattern of agricultural land use on Vashon Island is not consistent with the spatial pattern of soil arsenic contamination documented in the footprint study. Moreover, the trace element analyses of selected footprint samples from Vashon-Maury Island provide conclusive evidence that the arsenic contamination is associated with smelter emissions and not agricultural chemicals (see Section 4.5). It is certainly possible that individual land parcels within the Tacoma Smelter Plume Site that have a history of agricultural use have elevated soil arsenic (and lead) concentrations from applications of arsenical chemicals. However, the regional-scale impacts shown in the footprint studies, with sampling in wooded areas, are not the result of agricultural chemical use.

As already noted, the footprint sampling design was developed to characterize regional-scale patterns of soil contamination and to reduce the likelihood that miscellaneous small sources for arsenic would affect the results. Sampling locations were chosen within one homogeneous land use classification - relatively mature forested areas - to the maximum extent possible. Land use type can be an important factor in determining soil contamination, because different sources can be associated with various types of land uses and because different physical and chemical processes occur in soils with different land uses (see Chirenje et al. 2002). Thus, defining the spatial area impacted by smelter emissions does not mean that every property within that area is still impacted, nor does it mean that there are no other sources contributing to soil contamination. The DuPont site in southern Pierce County, within the mapped smelter plume area, had soil arsenic contamination associated with the use of arsenical herbicides to eliminate vegetation along rail lines at an explosives manufacturing plant, reducing fire danger. The South Tacoma Field site similarly had documented soil arsenic contamination unlikely to have been associated with smelter plume fallout. In Denver, use on residential lawns of a chemical herbicide containing arsenic has been interpreted as affecting soil contamination patterns in an area that is also near a smelter (Folkes, Helgen et al. 2001; Folkes, Kuehster et al. 2001). Soils very close to CCA-treated wood uses have been shown to have some elevation in arsenic concentrations. Soils at firing ranges have been shown to have elevated arsenic concentrations resulting from the arsenic content of lead shot (used to increase sphericity). Other arsenic sources affecting non-forested properties may have to be considered in some cases as additional sampling data are reviewed, but they do not affect the interpretation of regional-scale soil contamination from the footprint studies.

Some atypical arsenic emissions sources have been reported in the literature with regional-scale impacts. For example, wind-blown dusts from the dry lakebed at Owens Lake in California has been associated with transport of arsenic-enriched dusts several hundred kilometers downwind (Reheis et al. 2002). The burning of high-arsenic coal at a power plant in Slovakia with low levels of pollution control has resulted in the emissions of several thousand tons of arsenic to surrounding areas (Ranft et al. 2003). Volcanic emissions in Washington might be classified as such an atypical source, but can hardly account for the spatial patterns of soil arsenic in the Tacoma Smelter Plume area. No atypical sources of arsenic have been identified with likely regional impacts over the study area.

In summary, no source has been identified, other than Tacoma Smelter emissions, that could have affected the large regional-scale pattern of soil arsenic contamination shown by sampling relatively undisturbed forested properties. Other sources that may have contributed to soil contamination are viewed as the superposition on this dominant regional-scale pattern of more localized impacts affecting comparatively small areas..

5.0 CONCLUSIONS

An extensive body of information was compiled and reviewed for this credible evidence analysis. Major sources of information included the following:

- historic studies of the Tacoma Smelter, including source tests of smelter operations and environmental monitoring studies.
- soil sampling and analysis studies conducted as part of Ecology's Tacoma Smelter Plume Site investigations, including trace element analyses.
- a literature review of investigations around other smelters worldwide.
- a historical review of other possible sources of arsenic with regional-scale impacts in central Puget Sound, together with an associated literature review of arsenic sources, uses, and chemical source characterizations.

The evaluations of this information used six approaches: 1) establishing that a reasonable pathway from Tacoma Smelter operations to regional soil contamination exists; 2) analyzing spatial patterns in contamination and concentration gradients; 3) analyzing temporal patterns in contamination in relation to smelter operations; 4) assessing the patterns of contamination among multiple trace elements associated with smelting operations; 5) documenting a consistent pattern of contamination around smelters from literature reports on numerous smelters, including those located where other credible sources are lacking; and 6) establishing that the Tacoma Smelter was the dominant regional emissions source for arsenic in Puget Sound, and that no other credible sources can be identified with regional-scale impacts.

The results of these evaluations provide six consistent and mutually supporting rationales to identify the Tacoma Smelter as the source for regional-scale soil contamination of the central Puget Sound region by arsenic and other smelter-related metals, as documented in the Tacoma Smelter Plume Site investigations. A summary of these rationales in provided in Section 3 of the report.

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TABLES

FIGURES

ATTACHMENT A

MAXIMUM ARSENIC AND LEAD CONCENTRATIONS for FOOTPRINT STUDY SAMPLING LOCATIONS

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Table 1

Selected References
Tacoma Smelter Source Tests and Emissions Estimates

Source Tests

Reference	Comments
Statnick 1974	determined trace element emissions from roaster/reverb and converter gas streams for As, Cd. Cr, Cu, Pb, Zn
GCA 1974	three high-vol air sampling stations adjacent to Tacoma Smelter; daily filters analyzed for 13 elements
PSAPCA 1974	source test, fugitive emissions from slag dumping
Nelson and Roberts 1975	emissions from electrostatic precipitator and pilot baghouse determined; size fractionation and emissions for As, Sb, Cd, Cu, Pb, Hg, Ni
Labbe 1975	source test, roof top of converter building; air loadings for As, Cd, Pb, Hg, Zn
Roberts 1975	analysis of deposition to three fall-out jars (4-month collection period) near Tacoma Smelter; As, Sb, Cd, Cu, Pb, Hg, Ni, Se
Labbe 1977	test of Hg emissions from experimental baghouse on roaster gas stream
USEPA 1979a	arsenic emissions from roaster baghouse, arsenic kitchen baghouse, and reverberatory furnace electrostatic precipitator
USEPA 1979b	arsenic emissions from flue and fugitive gases from converter operation
Henderson 1981	mass balance estimates of As, Pb, Zn emissions from slag dumping
PSAPCA 1982	source test for As emissions from slag skimming at reverb furnace
USEPA 1983a	various source tests for proposed NESHAP rule for high-

	arsenic primary copper smelter at Tacoma (see Appendix C)
USEPA 1984	As emission tests for reverb furnace and arsenic plant gas streams
PEDCo 1984	converter cycle emissions tests; As, Sb, Bi, Cd, Pb, Se
NEA 1984	source apportionment (receptor modeling) study; chemical composition and particle size fraction characterization of multiple smelter sources, multiple elements
Austin 1984	fallout samples after tall stack fire analyzed for As, Sb, Cu, Pb, Zn
ASARCO 1993	deposition of As, Cd, Pb in fallout jars from tall stack demolition

Emissions Estimates

Reference	Comments
Labbe 1973	estimated 1973 air emissions for As, Cd, Cu, Pb, Ni, Se, Ag, and Zn
Randt 1975	estimated 1975 emissions of SO2, total particulate, and arsenic trioxide from tall stack, converter building, and anode furnace
PSAPCA 1981	air emissions inventory (Appendix F to Draft EIS) providing annual SO2, total particulate, and arsenic trioxide emissions from 1971 through 1980
USEPA 1983a	arsenic material balance for smelter processes; estimates tall stack and fugitive arsenic emissions
Varner 1983	estimated annual As, Pb, and Hg emissions for 1960 through 1983
Roberts 2003	(from PSAPCA files) emissions estimates, various years, for As, Sb, Cd, Cu, Pb, Hg, Ni, Zn

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Table 2

Selected Studies
World Smelters

Country	Smelter	References
Australia	Wollongong, New South Wales (Cu smelter)	Beavington 1975 Beavington 1977
	Port Pirie (Pb/Zn smelter)	Cartwright et al. 1976 Merry et al. 1981 Koh and Judson 1986 Langley 2002
Peru	La Oroya (Zn smelter)	Reif et al. 1989
Chile	Caletones (Cu smelter)	Romo-Kroger and Llona 1993 Romo-Kroger et al. 1994
	offshore	Saltzman et al. 1986
Argentine	Lastenia (Pb smelter)	Fernandez-Turiel et al. 2001
Mexico	Torreon (Pb/Zn smelter) and Chihuahua	Benin et al. 1999
	San Luis Potosi (Cu/Zn smelter)	Diaz-Barriga et al. 1993
Sweden	Gusum (brass mill)	Nordgren et al. 1983
	Ronnskar (Cu smelter)	Beckman 1978 Hellstrom 1980 Nordgren et al. 1986 Johnson et al. 1992 Pennanen et al. 1996
	Landskrona (Pb smelter)	Farago et al. 1999
Finland	Harjavalta (Cu smelter)	Fritze et al. 1989 Pennanen et al. 1996 Veijalainen 1998
Norway	Sulitjelma (Cu smelter)	Lobersli and Steinnes 1988

France	Noyelles-Godault (Pb/Zn smelter) and Auby (Zn smelter)	Godin et al. 1985 Sterckeman et al. 2002
Czech Republic	national survey (including Pribram smelting area)	Suchara and Sucharova 2002
	Krompachy and Rudnany	Mankovska et al. 1989
Poland	Legnica (Cu smelter)	Kowalinski et al. 1979 Kabata-Pendias et al. 1981
	western Poland (Cu smelter)	Zwozdziak and Zwozdziak 1982
United Kingdom	Bristol-Avonmouth (Pb/Zn smelter)	Little and Martin 1972 Cameron and Nickless 1977
Russia	Noril'sk (Cu/Ni smelters)	Blais et al. 1999
	Kola Peninsula, including Nikel (Cu/Ni smelter), Zapoljarnij (Cu/Ni smelter), and Monchegorsk (Cu smelter)	Karaban and Gytarsky 1995 Kozlov et al. 1995 Barcan and Sylina 1996 Niskavaara et al. 1996 Reimann et al. 1997 Ayras et al. 1997 Rautio et al. 1998 Barcan and Kovnatsky 1998 Gregurek et al. 1998 Gregurek et al. 1999 Reimann et al. 2000 Steinnes et al. 2000 Haugland et al. 2002 Reimann 2002
Ghana	Obuasi (Au smelter)	Amasa 1975
Canada	Flin Flon, Manitoba (Cu/Zn smelter)	Franzin et al. 1979 Hogan and Wotton 1984 Phillips et al. 1986 Zoltai 1988 Pip 1991 Bonham-Carter and McMartin 1997 Henderson et al. 1998 McMartin et al. 1999
	Thompson, Manitoba (Ni smelter)	Wotton et al. 1986

	Afton, British Columbia (Cu smelter)	Robertson and Price 1986
	Belledune, New Brunswick (Pb smelter)	Pilgrim and Hughes 1994
	Rouyn-Noranda, Quebec (Cu/Zn smelter)	Dumontet et al. 1990 Dumontet et al. 1992
	Trail, British Columbia (Pb/Zn smelter)	John et al. 1975 Lynch et al. 1980
	Yellowknife, NW Territories (Au smelter)	Hocking et al. 1978 Canadian Public Health Association 1977 Hazra et al. 1977 Stanley Associates Engineering Ltd. 1986 Ferguson Simek Clark 1994 Dillon Ltd. 1995
	multiple sites	McMartin et al. 2002 Newhook et al. 2003 Metals in the Environment-Research Network (MITE-RN) 1998 to present
	Sudbury, Ontario including Coniston, Falconbridge, and Copper Cliff smelters (Cu/Ni smelters)	Hutchinson and Whitby 1974 Hutchinson and Whitby 1977 Rutherford and Bray 1979 Chan et al. 1980 Freedman and Hutchinson 1980 Jeffries and Snyder 1981 Arafat and Glooschenko1982 Nriagu et al. 1982 Chan et al. 1982 Taylor and Crowder 1983 Hazlett et al. 1983 Nriagu 1983 Chan and Lusis 1985 Dudka et al. 1995 Ontario Ministry of the Environment 2001 Adamo et al. 2002
United States	Anaconda, Montana (Cu smelter)	Galbraith et al. 1995 St. Clair et al. 1995 USEPA 1996

	East Helena, Montana (Pb smelter)	Rupp et al. 1978 Pagenkopf and Maughan 1984
	Kellogg, Idaho (Pb smelter)	Roberts 1973 Ragaini et al. 1977
	Palmerton, Pennsylvania (Zn smelter)	Buchauer 1973 Beyer et al. 1984
	Garfield, Utah (Cu smelter)	Ball et al. 1983
	El Paso, Texas (Pb smelter)	Landrigan et al. 1975
	Missouri (Pb smelters)	Bolter et al. 1975
	San Manuel, Arizona (Cu smelter)	Gabriel and Patten 1994 Gabriel and Patten 1995
	Arizona (Cu smelters)	Germani et al. 1981 Small et al. 1981 Eldred et al. 1983 Blanchard and Stromberg 1987
	multiple smelters	Baker et al. 1977 Hartwell et al. 1983
	Upstate New York (Canadian smelters influence)	Gullu et al. 1995

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Table 3

Selected Spatial Extent Studies

Study Type	Reference	Comments
Bee Biomonitoring	Bromenshenk et al. 1985 Bromenshenk et al. 1991	kriging maps of As and Cd in bees over a 7500 square km region; bee biomass and As content over a Vashon Island transect of sampling sites
Sediments	Crecelius 1974	As and Sb in surface sediments in Puget Sound and regional lakes
Vegetation	Crecelius and Piper 1973	Pb and Cu in Douglas Fir tree needles
	Heilman and Ekuan 1977	numerous trace elements measured in garden vegetables from 70 regional gardens
Precipitation Chemistry	Larson et al. 1975	single-storm (synoptic) precipitation chemistry monitoring study over region downwind of Tacoma Smelter
	Harrison et al. 1977	evaluation of single-storm precipitation chemistry monitoring data for source of acid rain and trace elements
	Knudson et al. 1977	multivariate statistical analysis of single-storm precipitation chemistry monitoring data and identification of affected downwind region
	Carpenter et al. 1978	two-year monitoring of arsenic in rainfall at Puget Sound sampling locations from Olympia to near Anacortes

	Vong et al. 1986 Moseholm 1986 Vong et al. 1987 Vong, Larson et al. 1988 Vong, Moseholm, et al. 1988 Peterson II 1991	monitoring of regional precipitation chemistry in 14 storms over a period before and after closure of the Tacoma Smelter (Smelter Closure Study); classification of monitoring locations by source of impacts; evaluation of downwind region with changes after smelter closure
Soil Sampling	Heilman and Ekuan 1977	numerous trace elements measured in soils from 70 regional gardens
	Lowry 1983 Simpson 1983	As and Cd in regional garden soils; kriging analysis of soils data
	PHSKC and Glass 2000	Vashon-Maury Island footprint sampling study
	Washington State Department of Ecology 2002	King County Mainland footprint sampling study
	TPCHD 2003	Pierce County footprint sampling study
Air Modeling	PSAPCA 1981	H.E. Cramer SO2 modeling study; As and Cd deposition modeling study (using ISC-LT model)
	Vong 1982	sulfate wet deposition modeling study
	PEDCo 1983 USEPA 1983c	regional ambient arsenic modeling for USEPA NESHAPs rulemaking
	Luecken et al. 1989	As and sulfate deposition modeling study (using MPADD model)
Long-Range Transport	Lutrick 1971	integrating nephelometer study located ground-level plume impacts two to six miles from the Tacoma Smelter
	Crecelius 1974	ambient particulate arsenic concentration in Seattle highly

		correlated with wind direction
	Dethier 1979	precipitation chemistry monitoring at Copper Lake, 80 km NE of Tacoma Smelter
	Faulkner 1987	precipitation chemistry monitoring along the U.S./British Columbia border
	Welch et al. 1992	precipitation chemistry monitoring at Cascade Mountain lakes at time of smelter closure

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Number of Pages: 3
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Table 4

Tacoma Smelter
Chronology Studies

Type	Reference	Comments
Sediment Profiles	Crecelius 1974 Crecelius et al. 1975 Carpenter et al. 1978	As and Sb determined in age-dated sediment core in Quartermaster Harbor (Vashon-Maury Island)
	Crecelius 1974 Crecelius 1975	As determined in multiple sediment cores from Lake Washington
	Crecelius and Piper 1973	Pb determined in sediment cores from Lake Washington
	Lefkovitz et al. 1997	six age-dated sediment cores from main basin of Puget Sound, analyzed for multiple metals
Smelter Strike	McClannan 1974 McClannan and Rossano 1975	As, Sb, Cd, Pb, Hg, Mn,Se, Zn determined for daily airborne particulate samples, 10 locations; operating-to-strike period ratios all showed increases
Pollution Rose	Buchan 1967	SO2 wind rose shows highest values with SSW winds; supported by smelter plume diffusion calculations; high ambient As in Seattle also noted
	Crecelius 1974	As and Sb in ambient particulate samples at U. of Washington campus strongly associated with wind direction

	Hatfield 1976	As, Cd, Cu mostly in fine fraction of ambient particulates and correlated; suggested relationship of higher arsenic with winds from southern quadrant (p=0.11)
	PSAPCA 1981	SO2 wind roses for multiple monitoring stations (see Appendix D)
Smelter Closure	Vong et al. 1986	overview of the ASARCO Smelter Closure Rain Chemistry Study (U. of Washington); samples collected at multiple locations for multiple storms pre- and post-closure
	Faulkner 1987	precipitation chemistry at stations along US/Canada border showed significant As decrease after closure
	Moseholm 1986 Vong et al. 1988	excess sulfate in rain samples showed significant decrease to 25 km downwind after closure
	Scattarella 1988	smelter emissions shown to have downwind impacts on Pb in rainfall
	Peterson II 1991	rainwater chemistry showed influence of smelter source (As, Sb, Cu, Se, and sulfate); post-closure rainfall chemistry discriminated largely for decreased As, Sb
	Welch et al. 1992	decreasing trend in alpine lake sulfate associated with smelter emissions time trend up to and after closure

Filename: Crtable4.doc
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Number of Words: 327 (approx.)
Number of Characters: 1,874 (approx.)

Tacoma Smelter Plume Site
Credible Evidence Report

Table 5
Trace Element Correlations
Based on Maximum Concentrations by Boring

Inter-Element Correlations

	Arsenic	Lead	Antimony	Bismuth	Indium
Lead	0.6037				
	(92)				
	0.0000				
Antimony	0.8030	0.7210			
	(92)	(92)			
	0.0000	0.0000			
Bismuth	0.8682	0.7465	0.8849		
	(92)	(92)	(92)		
	0.0000	0.0000	0.0000		
Indium	0.8407	0.6969	0.8878	0.9447	
	(92)	(92)	(92)	(92)	
	0.0000	0.0000	0.0000	0.0000	
Silver	0.7664	0.6734	0.6890	0.8260	0.8295
	(38)	(38)	(38)	(38)	(38)
	0.0000	0.0000	0.0000	0.0000	0.0000

NOTES:

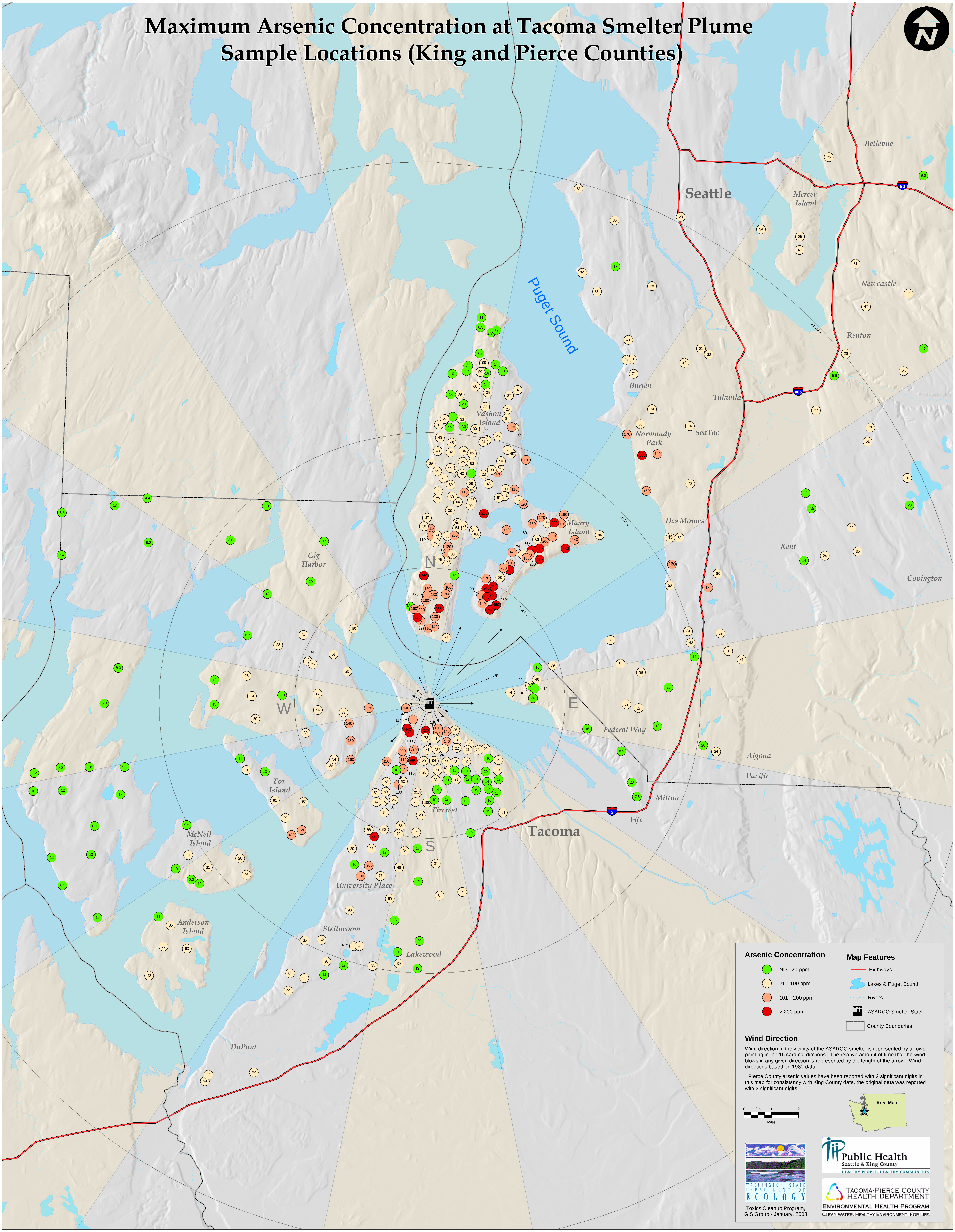
1. Entry formats are:

0.6037	correlation coefficient
(92)	number of samples
0.0000	significance level (0.0000 means < 0.00005)

2. All data distributions are right-skewed; therefore correlations (parametric) are calculated using Log concentrations.
3. Arsenic and lead results are from OnSite lab analyses; other results are from Battelle lab analyses.
4. For 9 samples without OnSite lead results, Battelle results are used instead as surrogate values.
5. Correlations are based on results from Vashon-Maury Island and King County Mainland

	locations including undisturbed and disturbed soils.	

Maximum Arsenic Concentration at Tacoma Smelter Plume Sample Locations (King and Pierce Counties)



Arsenic Concentration

●	ND - 20 ppm
●	21 - 100 ppm
●	101 - 200 ppm
●	> 200 ppm

Map Features

Highways

Lakes & Puget Sound

Rivers

ASARCO Smelter Stack

County Boundaries

Wind Direction

Wind direction in the vicinity of the ASARCO smelter is represented by arrows pointing in the 16 cardinal dirctions. The relative amount of time that the wind blows in any given direction is represented by the length of the arrow. Wind directions based on 1980 data.

* Pierce County arsenic values have been reported with 2 significant digits in this map for consistancy with King County data, the original data was reported with 3 significant digits.

Area Map

0 0.5 1 2
Miles

WASHINGTON STATE DEPARTMENT OF ECOLOGY

Toxics Cleanup Program,
GIS Group - January, 2003

Public Health
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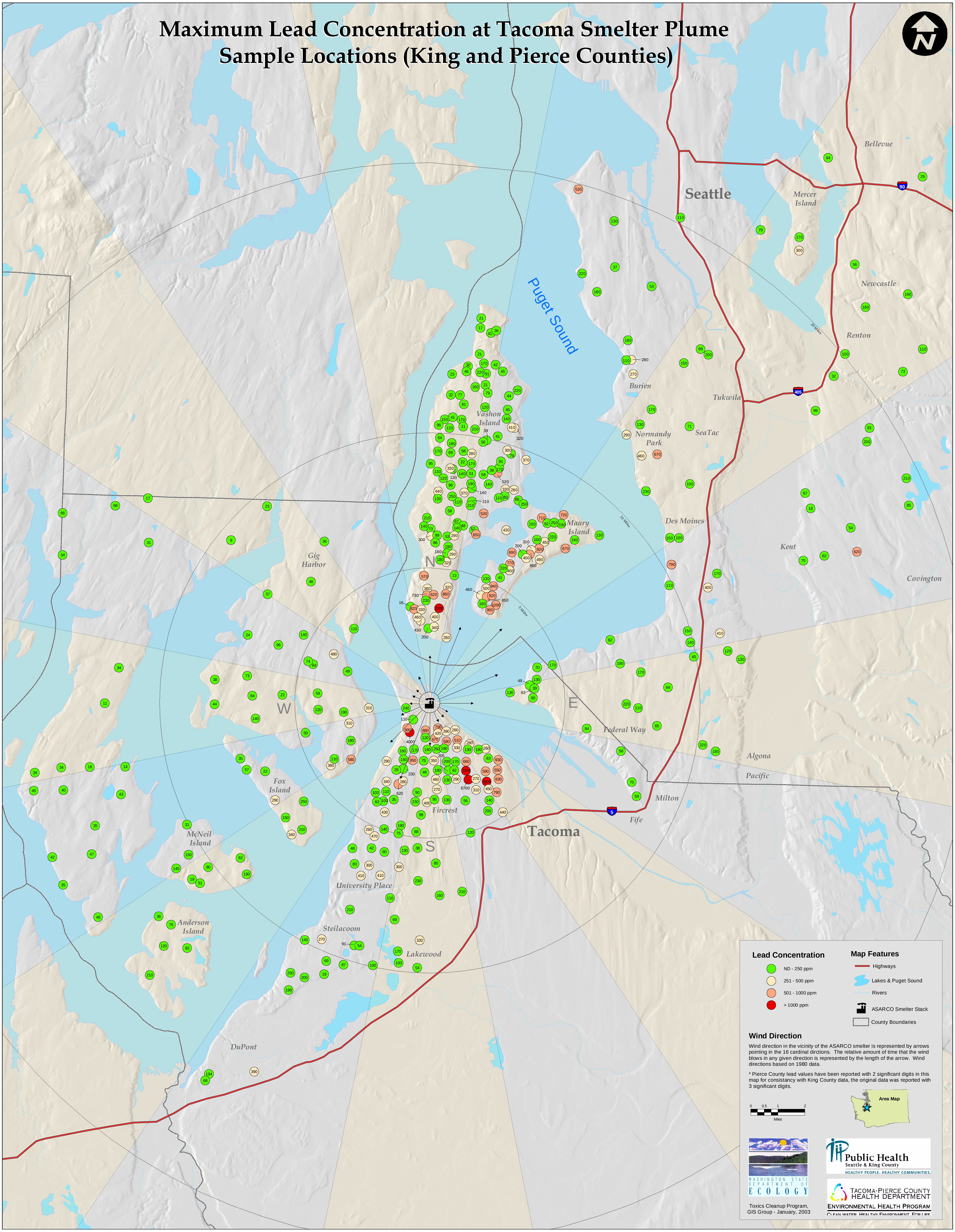
TACOMA-PIERCE COUNTY HEALTH DEPARTMENT
ENVIRONMENTAL HEALTH PROGRAM
CLEAN WATER. HEALTHY ENVIRONMENT. FOR LIFE.

Maximum Lead Concentration at Tacoma Smelter Plume Sample Locations (King and Pierce Counties)



Lead Concentration

- 🟩 ND - 250 ppm
- ● 251 - 500 ppm
- ● 501 - 1000 ppm
- 🟥 > 1000 ppm

Map Features

- Highways
- Lakes & Puget Sound
- Rivers
- ASARCO Smelter Stack
- County Boundaries

Wind Direction

Wind direction in the vicinity of the ASARCO smelter is represented by arrows pointing in the 16 cardinal dirctions. The relative amount of time that the wind blows in any given direction is represented by the length of the arrow. Wind directions based on 1980 data.

* Pierce County lead values have been reported with 2 significant digits in this map for consistancy with King County data, the original data was reported with 3 significant digits.

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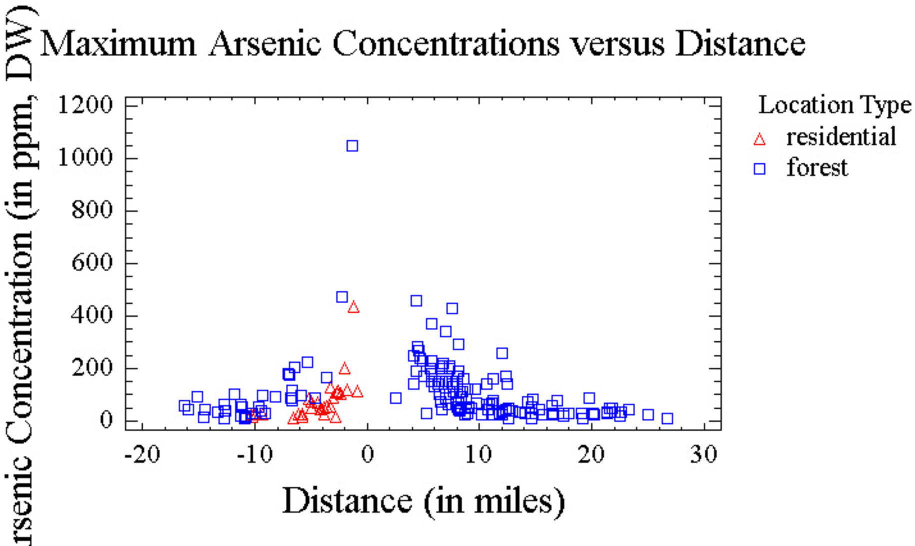
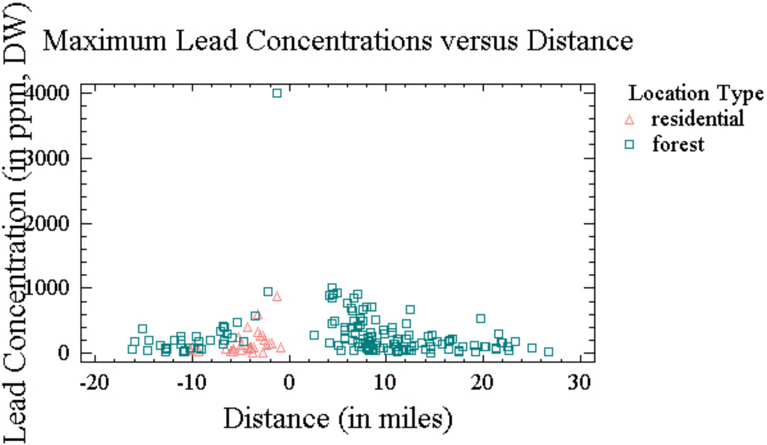
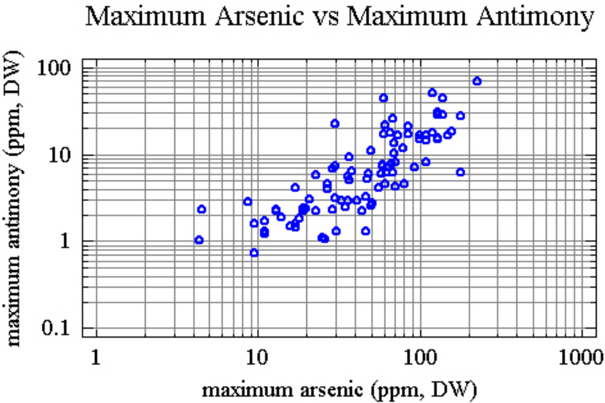
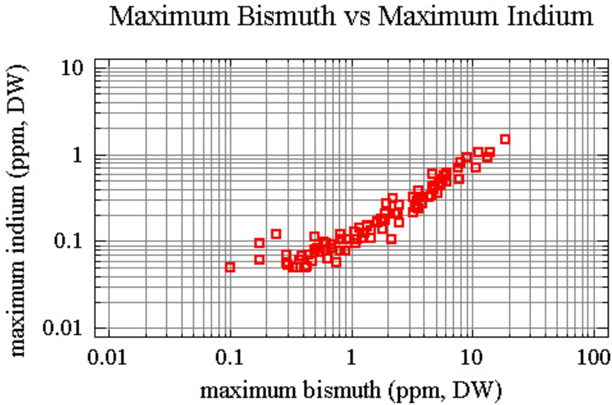
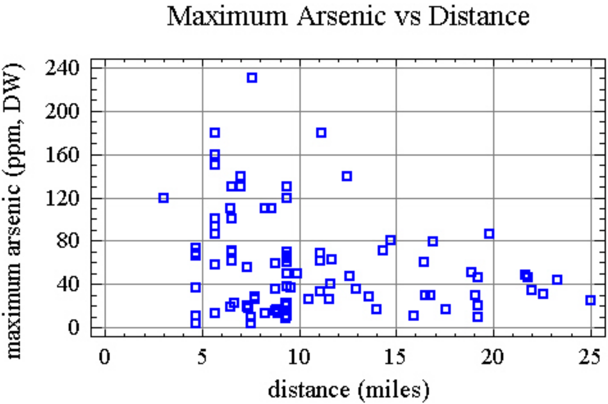


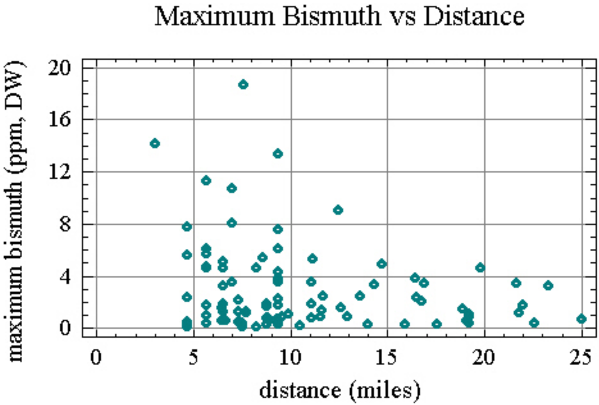
Maximum Arsenic Concentrations versus Distance


Maximum Lead Concentrations versus Distance
Lead Concentration (in ppm, DW)
4000
3000
2000
1000
0
-20
-10
0
10
20
30
Distance (in miles)
Location Type
△ residential
□ forest


Maximum Arsenic vs Maximum Antimony
maximum antimony (ppm, DW)
100
10
1
0.1
1
10
100
1000
maximum arsenic (ppm, DW)


Maximum Bismuth vs Maximum Indium
maximum indium (ppm, DW)
10
1
0.1
0.01
0.01
0.1
1
10
100
maximum bismuth (ppm, DW)


Maximum Arsenic vs Distance
maximum arsenic (ppm, DW)
240
200
160
120
80
40
0
0
5
10
15
20
25
distance (miles)


Maximum Bismuth vs Distance

Tacoma Smelter Plume Site
Credible Evidence Report

Attachment A
Maximum Arsenic and Lead Concentrations
for Footprint Study Sampling Locations

Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
South Vashon-Maury Island					
1	18.12	9.70	67	79	2
2	21.66	9.67	120	370	2
3	16.46	8.88	120	520	2
4	16.54	9.08	54	170	2
5	11.12	8.10	85	140	2
7	15.12	8.38	48	140	2
8	19.55	8.39	90	330	2
9	21.67	8.49	110	260	2
10	23.67	8.19	67	88	2
11	19.94	8.14	41	60	2
12	18.65	8.00	51	110	2
15	11.87	7.62	39	210	2
16	11.71	7.46	99	210	2
17	8.04	7.51	64	110	2
18	8.75	6.74	25	67	2
19	10.81	6.67	39	48	2
21	16.01	7.29	210	530	2
22	23.93	6.99	88	430	2
23	23.93	6.99	150	170	2
24	23.93	6.99	100	410	2
25	14.45	6.60	45	57	2
27	8.95	6.55	54	140	2
29	0.40	6.44	110	73	2
30	0.61	6.17	110	300	2
31	2.62	6.21	52	99	2
32	6.16	6.20	63	53	2
33	8.44	6.26	200	290	2
34	15.43	6.47	100	650	2
36	6.76	5.83	120	190	2
37	2.06	5.94	76	86	2
40	6.68	5.57	130	180	2
41	8.77	5.56	80	290	2
43	6.82	5.26	58	320	2
44	4.26	5.31	75	180	2
46	-2.47	4.71	79	270	2
47	-2.47	4.71	36	90	2
48	-2.47	4.71	260	570	2
49	10.98	4.81	14	13	2

Tacoma Smelter Plume Site
Credible Evidence Report

Attachment A
Maximum Arsenic and Lead Concentrations
for Footprint Study Sampling Locations

Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
50	9.18	4.33	160	320	2
51	-1.15	4.23	120	380	2
52	-2.47	4.71	130	220	2
53	-1.12	3.98	170	730	2
54	2.24	4.01	130	620	2
55	8.49	4.07	180	860	2
57	-2.00	3.79	180	230	2
58	-11.18	3.63	13	16	2
59	-9.30	3.52	160	820	2
60	-4.90	3.46	120	330	2
62	5.70	3.52	360	1300	2
63	3.64	3.19	130	400	2
65	-6.30	3.06	130	430	2
66	-8.08	3.20	220	460	2
67	-1.08	2.76	110	200	2
68	3.57	2.82	140	340	2
69	14.53	2.49	85	280	2
70	25.32	8.14	190	250	2
71	36.29	8.23	110	190	2
73	29.87	7.63	130	150	2
74	31.24	8.01	170	710	2
75	33.20	7.95	65	82	2
76	35.52	8.55	160	720	2
77	36.29	8.23	39	41	2
82	45.39	8.83	84	130	2
83	41.68	8.08	140	240	2
84	41.39	7.61	140	490	2
85	41.39	7.61	430	670	2
86	34.67	8.11	290	250	2
87	36.55	7.64	110	220	2
88	35.53	7.32	200	450	2
89	33.35	7.24	63	100	2
90	33.71	6.79	220	310	2
91	35.42	7.00	340	920	2
92	41.39	7.61	150	290	2
93	37.56	6.69	210	490	2
94	33.96	6.63	200	860	2
95	32.14	6.49	74	200	2
96	28.75	6.35	140	690	2

Tacoma Smelter Plume Site
Credible Evidence Report

Attachment A
Maximum Arsenic and Lead Concentrations
for Footprint Study Sampling Locations

Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
98	30.09	5.93	130	770	2
99	33.71	6.44	150	400	2
100	31.10	5.73	150	250	2
101	31.10	5.73	180	370	2
102	31.10	5.73	370	370	2
103	31.10	5.73	230	400	2
104	28.76	5.68	200	220	2
105	24.48	5.05	170	130	2
106	28.64	4.92	230	940	2
107	26.34	4.72	240	500	2
108	29.45	5.32	30	41	2
109	25.50	4.41	190	460	2
110	28.39	4.46	280	850	2
111	30.16	4.59	270	920	2
112	34.14	4.38	460	1000	2
113	33.00	4.10	250	900	2
114	28.03	4.16	140	160	2
King County Mainland, Shoreline					
1	72.96	4.76	79	170	2
2	70.80	7.08	39	62	2
3	78.35	7.20	54	190	2
4	45.64	11.21	160	230	2
5	40.62	12.06	260	460	2
6	45.64	11.21	79	180	2
7	45.64	11.21	21	21	2
8	40.62	12.06	24	41	2
9	60.24	10.32	160	790	2
10	60.24	10.32	50	85	2
11	60.24	10.32	120	580	2
12	36.31	12.31	170	290	2
13	36.31	12.31	42	47	2
14	29.85	14.63	52	110	2
15	29.85	14.63	28	66	2
16	29.85	14.63	9.6	11	2
North Vashon Island					
1	7.66	14.37	11	21	2
2	7.75	14.01	6.5	17	2

Tacoma Smelter Plume Site
Credible Evidence Report

Attachment A
Maximum Arsenic and Lead Concentrations
for Footprint Study Sampling Locations

Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
3	9.44	13.88	9.9	41	2
4	10.16	14.00	19	36	2
5	8.17	13.04	7.2	21	2
6	9.13	12.73	99	170	2
7	11.04	12.75	14	42	2
8	6.52	12.54	11	30	2
9	12.48	12.56	10	45	2
10	9.73	12.37	16	91	2
11	8.70	12.38	56	220	2
12	6.34	12.34	3.7	46	2
13	3.95	12.19	10	23	2
14	8.27	11.82	60	160	2
15	10.00	11.95	14	21	2
16	15.74	12.02	32	75	2
17	15.74	12.02	37	220	2
18	14.49	11.73	27	44	2
19	10.66	11.67	35	79	2
21	5.63	11.45	26	77	2
22	3.94	11.42	18	32	2
24	6.56	11.12	20	81	2
26	10.61	11.13	32	120	2
27	14.92	11.23	25	45	2
28	15.16	10.89	64	140	2
29	17.16	10.65	62	320	2
30	16.61	10.64	140	410	2
31	6.45	10.55	33	170	2
32	4.65	10.61	11	41	2
33	3.06	10.51	27	150	2
34	1.89	10.29	31	96	2
35	4.17	10.20	20	120	2
36	6.96	10.30	7.3	11	2
37	9.43	10.27	33	210	2
38	14.34	10.18	25	41	2
39	12.25	9.94	23	39	2
40	11.62	9.86	41	56	2
41	4.87	9.65	45	180	2
42	4.87	9.65	39	85	2
43	2.25	9.82	40	64	2
44	1.90	9.32	43	170	2

Tacoma Smelter Plume Site
Credible Evidence Report

Attachment A
Maximum Arsenic and Lead Concentrations
for Footprint Study Sampling Locations

Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
45	4.80	9.29	32	69	2
46	7.66	9.41	34	58	2
47	9.63	9.36	85	280	2
48	17.13	9.78	68	300	2
49	16.45	9.32	50	81	2
50	10.00	8.99	63	170	2
51	7.87	9.00	25	22	2
52	5.65	8.69	56	130	2
53	0.30	8.85	69	95	2
54	1.92	8.57	29	130	2
55	5.04	8.71	59	330	2
56	8.06	8.56	42	140	2
57	10.31	8.63	1.2	51	2
58	13.30	8.68	23	58	2
59	14.95	8.91	30	39	2
60	10.74	8.25	29	190	2
61	9.41	7.89	110	370	2
62	5.53	8.10	39	96	2
63	3.50	8.33	73	120	2
64	2.34	7.84	53	440	2
65	2.32	7.56	79	130	2
66	6.28	7.69	89	250	2
67	9.41	7.89	22	35	2
68	6.04	7.15	28	58	2
69	-0.82	6.84	47	210	2
70	-1.81	6.54	38	140	2
King County Mainland					
2	39.25	21.62	49	300	2
3	38.45	22.01	35	170	2
11	36.19	24.99	25	94	2
14	43.12	26.69	6.8	29	2
18	54.36	22.47	17	110	2
19	49.98	20.08	26	100	2
20	55.01	21.38	26	73	2
21	60.82	15.92	11	67	2
23	69.58	15.59	24	62	2
28	67.45	16.88	29	54	2
30	59.17	18.85	51	200	2

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Attachment A
Maximum Arsenic and Lead Concentrations
for Footprint Study Sampling Locations

Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
31	51.04	19.24	9.6	32	2
34	57.99	19.20	47	81	2
35	34.99	21.37	34	79	2
36	19.56	16.87	79	220	2
37	16.16	19.79	86	530	2
38	20.98	19.10	30	130	2
46	22.15	16.43	60	180	2
54	28.70	15.30	41	180	2
55	30.49	14.74	81	280	2
56	43.22	14.05	26	71	2
57	38.69	16.50	30	200	2
58	37.09	12.92	36	130	2
60	42.48	12.47	140	670	2
62	49.93	12.59	48	100	2
64	55.51	10.79	45	150	2
65	56.54	11.07	69	100	2
68	95.78	8.46	18	65	2
70	91.41	7.73	29	110	2
74	86.28	8.85	20	64	2
76	80.16	11.20	28	120	2
77	82.08	11.64	41	130	2
79	76.52	11.05	62	410	2
80	74.44	9.92	24	150	2
84	67.50	11.15	180	400	2
85	65.79	11.67	63	170	2
88	36.82	15.71	24	150	2
89	37.14	13.62	34	170	2
97	31.82	14.31	71	270	2
101	63.91	9.89	50	110	2
103	76.98	9.91	40	140	2
104	37.45	16.50	21	69	2
107	28.07	17.47	28	53	2
228	64.79	19.51	36	210	2
229	67.61	19.19	20	85	2
367	98.77	10.23	20	220	2
408	44.12	22.61	31	56	2
501	80.10	9.93	14	45	2
502	81.77	7.90	38	170	2
503	70.52	16.78	30	620	2

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504	90.37	7.28	32	220	2
505	49.48	23.28	44	190	2
506	69.18	14.81	14	75	2
507	23.07	17.53	17	37	2
508	99.63	10.74	24	180	2
509	27.35	20.22	23	110	2
510	62.98	15.84	7.5	18	2
511	52.86	17.92	27	99	2
512	47.77	21.79	47	160	2
Pierce County					
102	257.70	-0.89	142	240	1
103	224.50	-0.87	114	107	1
105	136.50	-1.38	35.5	262	1
106	149.30	-1.22	136	390	1
107	161.60	-0.99	122	787	1
108	188.30	-1.03	233	875	1
110	221.40	-1.25	440	900	1
112	143.30	-1.72	30.2	509	1
113	156.20	-1.56	140	578	1
114	170.70	-1.33	61.3	673	1
115	186.90	-1.30	78.3	120	1
118	129.20	-2.68	21.8	259	1
119	133.60	-2.51	27.7	181	1
120	140.70	-2.23	21	129	1
121	148.80	-1.94	22.1	316	1
122	162.10	-1.77	56.1	194	1
123	171.40	-1.73	72.7	254	1
124	182.60	-1.73	81.1	144	1
125	198.50	-1.81	122	171	1
127	129.70	-3.30	27.1	928	1
128	133.30	-2.99	10.4	83.3	1
130	148.00	-2.56	48.8	662	1
131	156.30	-2.38	42.6	170	1
132	163.70	-2.26	26.1	197	1
133	175.70	-2.14	93.6	353	1
134	185.80	-2.15	28.8	75.2	1
135	196.10	-2.21	475	947	2
136	204.80	-2.31	105	138	1

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Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
138	216.40	-2.68	112	286	1
139	134.70	-3.53	22.5	551	1
140	140.80	-3.28	20.1	594	1
142	151.80	-2.85	19.4	3050	1
143	159.80	-2.66	18.7	62.1	1
144	164.30	-2.59	23.9	205	1
145	173.20	-2.50	40.9	194	1
146	184.20	-2.58	25	47.9	1
148	201.70	-2.63	115	233	1
149	206.40	-2.77	16.1	26	1
150	148.50	-3.29	13.3	273	1
151	153.10	-3.18	17.3	6670	1
152	160.60	-3.00	21.1	288	1
153	167.10	-2.92	19.5	131	1
154	175.40	-2.85	37.6	461	1
155	198.70	-3.06	92.1	279	1
157	208.70	-3.32	58	338	1
159	151.90	-3.65	13.1	305	1
163	175.10	-3.22	13.6	272	1
165	187.80	-3.34	21.5	49.6	1
168	206.20	-3.66	58.5	108	1
169	211.00	-3.87	52.2	103	1
171	159.90	-3.86	12.3	56	1
173	170.00	-3.65	16.6	127	1
174	177.20	-3.58	18.5	94.8	1
175	181.60	-3.68	104	402	2
176	188.10	-3.70	74.5	227	1
178	200.20	-3.82	26.3	35.1	1
179	205.20	-4.02	50	103	1
180	208.10	-4.15	47.4	83.3	1
181	213.90	-1.32	1050	3990	2
203	200.90	-3.23	133	622	1
215	136.00	-2.12	39	276	1
241	144.00	-3.60	14.4	2820	1
242	137.90	-3.80	11.7	833	1
243	143.20	-4.14	16.6	792	1
244	145.60	-3.86	14.1	445	1
245	148.50	-4.23	10	136	1
247	164.50	-1.17	129	417	1

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Attachment A
Maximum Arsenic and Lead Concentrations
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Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
299	209.30	-2.04	204	156	1
304	202.30	-4.38	70.1	425	1
306	162.50	-5.03	10.3	122	1
308	185.90	-4.80	25	88	1
309	193.40	-4.97	79	75	1
310	199.80	-4.97	52.9	144	1
311	205.60	-5.20	88	257	1
313	184.80	-5.39	17.5	37.5	1
314	189.60	-5.54	34.4	126	1
315	196.80	-5.77	19	80.4	1
316	201.80	-5.79	25.9	42.1	1
317	208.00	-6.09	29.1	48.3	1
319	177.80	-5.94	30.6	80.3	2
321	190.50	-6.18	46.4	298	1
323	200.40	-6.42	204	301	2
324	205.00	-6.58	15.6	82.7	1
327	183.70	-6.60	13.4	225	1
329	195.90	-6.65	76.8	410	2
331	201.60	-6.88	177	411	2
341	99.30	5.90	16.4	84	1
346	87.30	3.83	19.8	60.2	1
349	77.70	4.06	44.9	132	2
351	71.80	4.20	15.8	69.8	1
353	114.30	8.41	7.45	58.6	1
355	111.40	8.03	20.1	69.8	1
359	104.10	7.30	8.47	58.3	1
361	193.20	-4.66	88.3	176	2
362	202.50	-5.34	223	474	2
383	82.70	3.00	74.1	129	1
384	80.10	3.77	21.9	49.1	1
388	81.80	3.86	18.2	62.6	1
390	82.10	3.92	13.8	39.4	1
393	184.40	-4.16	20.3	98.1	1
396	151.60	-4.54	11.1	200	1
397	146.20	-4.87	20.7	443	1
401	170.20	-7.09	28.5	214	1
403	191.50	-7.38	69.1	108	1
409	189.10	-8.13	18.2	68.8	1
411	201.10	-8.21	90.1	209	2

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Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
415	182.40	-8.80	19.8	333	1
418	197.20	-9.40	36.7	91.2	2
419	204.50	-9.62	51.9	268	2
420	207.70	-9.92	34.5	141	1
422	182.70	-9.82	12.9	54.3	1
423	186.70	-9.70	30	100	1
424	192.20	-9.94	33.4	102	1
425	198.20	-10.21	17.1	86.9	1
426	201.80	-10.29	30.4	68.7	1
441	177.00	-7.13	33.9	159	1
442	187.30	-9.27	10.9	172	1
444	196.10	-9.35	27.9	53.8	1
503	314.40	-3.92	64.5	111	2
504	297.00	-3.98	60.7	482	2
505	288.40	-4.54	25.5	83.6	2
509	288.90	-4.74	40.7	74.3	2
516	274.90	-4.16	25.1	58.6	2
518	265.20	-2.25	171	310	2
521	266.40	-4.13	56	122	2
524	255.70	-3.07	135	306	2
525	263.80	-3.20	71.8	187	2
527	244.50	-3.23	128	184	2
530	234.30	-3.58	163	577	2
531	239.50	-4.09	53.7	114	2
540	231.90	-5.91	97	248	2
542	237.80	-6.76	80.8	290	2
545	231.40	-6.81	88.9	152	2
546	225.20	-6.66	117	208	2
547	226.30	-7.06	182	336	2
550	247.40	-6.59	12.8	22.3	2
565	237.70	-4.33	69.2	358	2
570	298.40	-5.30	34	137	2
599	291.00	-3.25	25.1	49.4	2
601	291.00	-5.99	23.2	96.3	2
602	290.50	-7.18	8.72	23.5	2
604	278.40	-6.83	24.9	73	2
605	276.16	-7.99	11.8	37.7	2
606	273.10	-5.46	7.9	22.2	2
607	272.30	-6.56	34.3	84	2

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Sampling Location Code	Direction from smelter (bearing, degrees)	Distance from smelter (in miles)	Maximum Arsenic (in ppm, DW)	Maximum Lead (in ppm, DW)	Location Type (see notes)
608	269.60	-7.96	14.7	43.6	2
609	256.40	-4.72	29.7	50.2	2
613	249.90	-7.21	20.7	57.2	2
614	258.10	-11.51	9.24	13.5	2
615	259.30	-12.79	3.76	15.5	2
616	260.10	-13.84	8.21	34.2	2
617	260.00	-14.84	7.18	33.9	2
618	253.50	-11.91	12.9	43	2
620	256.60	-13.93	11.5	40	2
621	257.50	-15.01	10.1	45.3	2
622	243.40	-10.04	9.53	31.3	2
624	249.80	-13.18	8.08	34.7	2
626	230.70	-9.05	28.4	61.5	2
628	237.80	-10.55	31.4	153	2
631	245.90	-13.71	10.3	46.9	2
632	247.70	-15.09	11.8	41.9	2
633	231.90	-10.80	18.1	51.1	2
635	243.60	-15.14	6.06	35.2	2
637	231.80	-12.76	10.6	36.2	2
638	237.10	-14.61	12	45.9	2
639	224.70	-12.76	62.5	91.8	2
640	227.60	-13.33	35.3	123	2
642	204.50	-11.17	51.7	202	2
644	225.80	-14.45	43.1	213	2
655	205.40	-15.11	91.6	390	2
657	210.70	-15.99	43.8	194	2
683	253.60	-7.30	11.3	35.4	2
687	229.30	-12.61	36.2	75.3	2
690	264.80	-6.47	29.6	136	2
696	201.20	-10.77	13.9	19.4	2
697	206.20	-11.84	99.2	199	2
698	207.20	-11.24	62.1	254	2
699	236.80	-11.20	18.6	137	2
703	320.50	-9.44	10.3	23	2
706	306.00	-12.89	4.43	17.4	2
707	302.10	-13.73	12.8	67.5	2
708	297.40	-15.29	8.53	65.8	2
710	326.90	-7.13	17	36.2	2
711	309.30	-9.50	2.99	8.9	2

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713	299.70	-11.97	6.23	30.6	2
715	291.90	-14.65	5.35	33.9	2
717	315.60	-6.28	19.5	46.3	2
718	303.90	-7.22	12.5	37.4	2
724	276.40	-11.58	7.96	33.9	2
725	269.90	-12.02	4.98	10.8	2
801	233.40	-10.95	8.83	18.8	2
802	233.40	-10.20	30.6	89.7	2
803	226.80	-9.28	96.1	188	2
804	210.70	-16.26	59.4	67.6	2

NOTES:

Locations in Pierce County from south through northwest are arbitrarily assigned negative distances for plotting purposes.

Arsenic and lead concentrations are in parts per million (ppm), dry weight basis.

Location type codes are 1 = residential (disturbed), 2 = forested (undisturbed).