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State of Washington

Priority Consumer Products Draft Report to the Legislature

Safer Products for Washington Implementation Phase 2

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Priority Consumer Products
Draft Report to the Legislature

Safer Products for Washington
Implementation Phase 2

Hazardous Waste and Toxics Reduction Program

Washington State Department of Ecology

Olympia, Washington

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Table of Contents

List of Figures and Tables

Page

Figures

Tables

Executive Summary

Legislative requirement

In 2019, the Washington State Legislature directed Washington State Department of Ecology (Ecology), in consultation with Washington State Department of Health (Health), to implement a regulatory program to reduce toxic chemicals in consumer products (Chapter 70.365 RCW).[1]

The law requires Ecology to identify priority consumer products that are significant sources or uses of the five priority chemical classes. The implementation program is called **Safer Products for Washington.**

This report, which identifies our draft priority products and details how they meet the criteria in the law, does not establish regulations or restrictions on these chemical-product combinations. After a 45-day public comment period, we will submit this report to the Legislature by June 1, 2020.

Background

The Safer Products for Washington program includes a regulatory process designed to keep harmful chemicals out of homes, workplaces, schools, and the environment. The law recognizes that small, steady releases of chemicals coming from millions of consumer products are the largest source of toxics entering Washington's environment. These chemical sources pose a significant threat to human health, the environment, and the future of Washington's residents.

Safer Products for Washington creates a systematic approach to reduce exposure to toxic chemicals found in consumer products. The law directs us to take the following actions:

1. Identify priority chemical classes.
2. Identify priority products that are significant sources or uses of those chemicals.
3. Determine if safer alternatives are available and feasible in order to decide whether to restrict or require reporting of priority chemical-product combinations.
4. Implement restrictions or reporting requirements, if any, through a rulemaking process.

The law identified the first set of priority chemical classes. They are:

- Organohalogen flame retardants and flame retardants identified in RCW 70.240.010.[2]
- Perfluoroalkyl and polyfluoroalkyl substances (PFAS).

[1] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365
[2] https://app.leg.wa.gov/RCW/default.aspx?cite=70.240.010

- Polychlorinated biphenyls (PCBs).
- Phenolic compounds.
- Phthalates.

Stakeholder advisory process

Another task specified in statute is to create a stakeholder advisory process by June 2020. While this work is ongoing, we have developed outreach materials[3] and held a public webinar[4] to explain the law and its requirements. Questions and answers from this webinar are available on our website.[5]

Following the webinar, we shared which product categories we were researching. We also invited interested stakeholders to submit information on chemicals and their concentrations in these products, the potential for human or environmental exposure, and possible safer alternatives.

Ecology will conduct further outreach during the 45-day public comment period for this report.

Draft priority products

Ecology, in partnership with Health, consulted peer-reviewed scientific data, government reports, and economic and market information to determine that the products shown below are significant sources or uses of the priority chemicals identified in Chapter 70.365 RCW.[6]

Priority products are significant sources or uses of priority chemicals because they contribute to human exposures or environmental releases. As outlined in the law, at this stage, we included all the chemicals in the class that are reported in a particular priority product. We are concerned about the threat to human and environmental health these chemicals pose when they are used, disposed, and released. Members of all of these priority chemical classes are also listed as contaminants of emerging concern by Governor Inslee's Orca Task Force.

Table 1. Draft priority products and major concerns related to the criteria defined in 70.365.

Priority Chemical or Chemical Class	Draft Priority Product	Major Concerns
Flame retardants	Electric and electronic equipment (device casings)	Human exposure, volume used, recycling

[3] https://fortress.wa.gov/ecy/publications/documents/1904012.pdf
[4] https://www.ezview.wa.gov/Portals/_1962/Documents/saferproducts/Webinar082919slides.pdf
[5] Bit.ly/SaferProductsWA
[6] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365

Priority Chemical or Chemical Class	Draft Priority Product	Major Concerns
PCBs	Printing inks	Environmental release
PFAS	Carpet	Human exposure, volume used
PFAS	Aftermarket carpet treatments	Human exposure, volume used
Phenolic compounds	Food cans (bisphenols)	Human exposure, volume used
Phenolic compounds	Laundry detergent (alkylphenol ethoxylates)	Environmental release
Phenolic compounds	Thermal paper (bisphenols)	Human exposure, volume used
Phthalates	Vinyl flooring	Human exposure
Phthalates	Fragrances in personal care and beauty products	Volume used

Why we chose these priority products

The following sections detail an overview of the nine draft priority products and the chemical each contains. We are concerned about the contribution these products make to the concentrations of priority chemicals found in homes, workplaces, and the environment.

Electric and electronic equipment (device casings)

Priority chemical: Organohalogen flame retardants and flame retardants identified under Chapter 70.240 RCW[7]. Organohalogen flame retardants are chemicals that prevent products from catching fire and contain one or more halogen element(s) bonded to carbon.

Priority product summary: Electric and electronic equipment with plastic device casings are a significant use of flame retardants. Examples of products that might contain plastics encasing electric or electronic components include TVs, printers, game consoles, and audio and video equipment. An average home has 30 items with enclosed electronic circuitry. We are concerned about the potential for exposure to children, workers in occupations involving electronics, and sensitive species.

[7] https://app.leg.wa.gov/RCW/default.aspx?cite=70.240

Printing inks

Priority chemical: Polychlorinated Biphenyls (PCBs)

Priority product summary: Printing inks are a significant source of unintentionally-generated PCBs. Several bodies of water in Washington are contaminated with PCBs, which have many toxic effects, accumulate in people and animals, and contaminate the food supply. Studies suggest that pigment and dye manufacturing is the product group that contributes the most to PCB levels in the environment.

Pretreated carpets and rugs

Priority chemical: Per- and polyfluoroalkyl substances (PFAS)

Priority product summary: Pretreated carpets and rugs are significant sources and uses of PFAS. Data suggests that up to 2,074 metric tons of PFAS enter homes and workplaces through carpet each year, and even more end up in landfills annually. Carpets contribute to human PFAS exposure, particularly for children who spend more time on or near the floor. PFAS are environmentally persistent, meaning after their release into the environment, cleanup can be difficult or impossible in some cases.

Aftermarket treatments for carpets and rugs

Priority chemical: Per- and polyfluoroalkyl substances (PFAS)

Priority product summary: Aftermarket treatments for stain and water resistance for carpets and rugs are significant sources and uses of PFAS. Studies suggest that people in Washington use 2,300 metric tons of aftermarket treatments each year. PFAS in aftermarket treatments are applied as liquid sprays (aerosols), making them more likely to migrate from the carpet to indoor air and dust versus PFAS in pretreated carpet.

Since children spend more time on the floor, they are disproportionately exposed to PFAS. Spills and improper disposal of aftermarket treatment can also release PFAS directly into the environment, and their persistence makes cleanup costly and challenging.

Food and drink cans with linings

Priority chemical: Phenolic Compounds—Bisphenols

Priority product summary: Can linings are a significant use of phenolic compounds. About 2.5 billion cans are sold each year in Washington, and testing shows that a large proportion of those may contain bisphenol-based can liners.

Dietary exposure to Bisphenol A (BPA) is the largest source of exposure to this chemical. Eating canned food leads to higher bodily levels, and while BPA does not persist very long in the body, its widespread detection in people shows that exposures happen often. This is especially

concerning for children, who may be more affected by these chemicals due to their sensitive stage in development.

Laundry detergent

Priority chemical: Phenolic Compounds—Alkylphenol Ethoxylates

Priority product summary: Laundry detergents are a significant source of phenolic compounds. Studies suggest that institutional cleaners—including laundry detergent—are the largest use of alkylphenol ethoxylates. Data suggests that Washingtonians use up to 2 million pounds of laundry detergent per year, which could contain alkylphenol ethoxylates.

Laundry detergents with alkylphenol ethoxylates are discarded down the drain and make their way through wastewater treatment plants to bodies of water. Alkylphenol ethoxylates and their alkylphenol components are already restricted in many other countries, which decreases their presence in those areas.

Thermal paper

Priority Chemical: Phenolic Compounds—Bisphenols

Priority product summary: Thermal paper, which is paper coated with a material that changes color when exposed to heat, is a significant source and use of phenolic compounds. About 3,300 tons of thermal paper are used every year in Washington, and testing shows that much of it contains bisphenols.

People absorb the bisphenols on thermal paper, which raises its concentration in their bodies. Retail workers have higher exposure than the general population due to their frequent contact with thermal paper like receipt tape. Another concern are environmental exposures, especially the recycling of thermal paper—the largest source of BPA to the environment.

Vinyl flooring

Priority chemical: Phthalates

Priority product summary: Vinyl flooring is a significant source and use of phthalates. Vinyl flooring may contain phthalates at concentrations between 9% and 32% by weight. This means flooring can contribute significant amounts of phthalates to homes and the environment.

Multiple studies show the connection between vinyl flooring in the home and higher concentrations of phthalates in both the human body and indoor air and dust. As vinyl flooring ages, phthalates are released into the environment. Data demonstrates that vinyl flooring may contribute 200 pounds of phthalates to Puget Sound each year.

Fragrances in the personal care sector

Priority chemical: Phthalates

Priority product summary: Fragrances used in the personal care and beauty sector include perfumes, colognes, body mists and body sprays. They are a significant source and use of phthalates.

Women have higher exposure to phthalates in fragrances than men. Exposure is higher in women of color, low-income women, and women with lower levels of education. These findings have important implications for environmental justice. Further, phthalates are frequently detected in the environment. Studies suggest that fragrances account for one third of the 34 tons of phthalates released into Puget Sound annually.

In this report: Priority product chapters

The following chapters of this report provide evidence supporting the inclusion of each chemical-product combination listed above based on the criteria in Chapter 70.365 RCW. Each chapter includes a product definition, estimations of the volume used in Washington, potential for human and environmental exposure, and existing regulations. **During the 45-day public comment period, we welcome feedback on the draft product list before March 2, 2020.**

This report will be finalized by June 2020. The next steps include assessing whether safer alternatives are feasible and available, then determining whether a restriction or reporting requirement is appropriate. We will conduct these steps between June 2020 and June 2022.

Acronyms

Table 2. Acronyms with definition and CAS number

Acronym	Definition	Chemical Abstracts Service (CAS) Number
µg/g	micrograms per gram	
µg/L	micrograms per liter	
2,4,6-TBP	2,4,6-Tribromophenol	CAS Number: 118-79-6
ABS	Acrylonitrile Butadiene Styrene polymers	
AP/APE	Alkylphenol/Alkylphenol Ethoxylate	
ATSDR	Agency for Toxic Substances and Disease Registry	
BADGE	Bisphenol A diglycidyl ether	CAS Number: 25085-99-8
BBP	Benzyl butyl phthalate	CAS Number: 85-68-7
BDE	Brominated diphenyl ether	
BPA	Bisphenol A	CAS Number: 80-05-7
BPAF	Bisphenol AF	CAS Number: 1478-61-1
BPAP	Bisphenol AP	CAS Number: 1571-75-1
BPB	Bisphenol B	CAS Number: 77-40-7
BPC	Bisphenol C	CAS Number: 79-97-0
BPF	Bisphenol F	CAS Number: 620-92-8
BPP	Bisphenol P	CAS Number: 2167-51-3
BPS	Bisphenol S	CAS Number: 80-09-1
BPZ	Bisphenol Z	CAS Number: 843-55-0
BTBPE	1,2-Bis(2,4,6-tribromophenoxy)ethane	CAS Number: 37853-59-1
CAP	Chemical action plan	
CDC	Centers for Disease Control	
CPSC	Consumer Product Safety Commission	
DBDPE	Decabromodiphenyl ethane	CAS Number: 84852-53-9
DBP	Di-n-butyl phthalate	CAS Number: 84-74-2
DecaBDE	Decabromodiphenyl ether	CAS Number: 1163-19-5
DEHP	Di(2-ethylhexyl) phthalate	CAS Number: 117-81-7
DEHT	Bis (2-ethylhexyl) terephthalate	CAS Number: 6422-86-2
DEP	Diethyl phthalate	CAS Number: 84-66-2
DIBP	Diisobutyl phthalate	CAS Number: 84-69-5
DIDP	Diisodecyl phthalate	CAS Numbers: 68515-49-1 and 26761-40-0
DINCH	Diisononyl cyclohexandicarboxylate	CAS Number: 474919-59-0
DINP	Diisononyl phthalate	CAS Numbers: 68515-48-0 and 28553-12-0
DMP	Dimethyl phthalate	CAS Number: 131-11-3
DnOP	Di-n-octyl phthalate	CAS Number: 117-84-0

Acronym	Definition	Chemical Abstracts Service (CAS) Number
DPHP	Diphenyl-phosphate	
DTSC	California Department of Toxic Substances Control	
ECHA	European Chemicals Agency	
EDI	estimated daily intake	
EFSA	European Food Safety Authority	
EHDPP	Ethylhexyl diphenyl phosphate	CAS Number: 1240-94-7
EU	European Union	
FOX	Firefighter occupational exposure	
FTOH/FTS	Fluorinated telomer alcohol/sufonates	
GM	geometric mean	
HIPS	high impact polystyrene	
iPCB	Inadvertent Polychlorinated Biphenyl	
IPTPP	Isopropylated triphenyl phosphate	CAS Number: 68937-41-7
ITRC	Interstate Technology & Regulatory Council	
kg	Kilogram	
LOQ	Limit of quantitation	
MBzP	Mono-benzyl phthalate	
MECPP	Mono-(2-ethyl-5-carboxypentyl) phthalate	
MEHHP	Mono (2-ethyl-5-hydroxyhexyl) phthalate	
MEHOP	Mono-(2-ethyl-5-oxohexyl) phthalate	
MEHP	Mono(2-ethylhexyl) phthalate	
MEP	Mono-ethyl phthalate	
Me-PFOSA-AcOH	2-(N-methyl-perfluorooctane sulfonamido) acetate	
mg	Miligram	
MHINP	Mono(hydroxyisononyl) phthalate	
MINP	Monoisononyl phthalate	
MMP	Mono-methyl phthalate	
MnBP	Mono-n-butyl phthalate	
MOINP	Mono(oxoisononyl) phthalate	
ng/kg	nanograms per kilogram	
ng/L	nanograms per liter	
ng/mL	nanograms per milliliter	
NGO	Non-governmental Organization	
NHANES	National Health and Nutrition Examination Survey	
NIEHS	National Institute of Environmental Health Sciences	
NIOSH	National Institute for Occupational Safety and Health	
NP/NPE	Nonylphenol/Nonylphenol Ethoxylate (type of APE)	
NTP	National Toxicology Program	

Acronym	Definition	Chemical Abstracts Service (CAS) Number
OctaBDE	Octabromodiphenyl ether	CAS Number: 32536-52-0
OECD	Organisation for Economic Co-operation and Development	
OP/OPE	Octylphenol/Octylphenol Ethoxylate (type of APE)	
PAP	Polyfluoroalkyl phosphates	
PASF	Perfluoroalkane sulfonyl fluorides	
PBDEs	Polybrominated Diphenyl Ethers	
PC-ABS	Polycarbonate/ABS blends	
PCB	Polychlorinated Biphenyl	
PDFA	Perfluorodecanoate	
PET	Polyethylene terephthalate	CAS Number: 25038-59-9
PFAA	Perfluoroalkyl acid	
PFAS	Per- and polyfluoroalkyl substances	
PFBA	Perfluorobutanoate	
PFBS	Perfluorobutanesulfonic acid	
PFCA	Perfluoroalkyl carboxylic acid	
PFHpA	Perfluoroheptanoate	
PFHxA	Perfluorohexanoate	
PFHxS	Perfluorohexane sulfonate	
PFNA	Perfluorononanoate	
PFOA	Perfluorooctanoate	CAS Number: 335-67-1
PFOS	Perfluorooctane sulfonate	
PFPeA	Perfluoropentanoate	
PFSA	Perfluoroalkyl sulfonate	
RDP	Resorcinol bis (diphenyl-phosphate)	CAS Number: 57583-54-7
RfD	EPA reference dose	
TBBPA	Tetrabromobisphenol A	CAS Number: 79-94-7
TCP	Tricresyl phosphate or Tris(methylphenyl)phosphate (TMPP)	CAS Number: 1330-78-5
TNBP	Tri-n-butyl phosphate	CAS Number: 126-73-8
TOTM	Tris (2-ethylhexyl) trimellitate	
TPP	Triphenyl phosphate	CAS Number: 115-86-6
TTBP-TAZ	2,4,6-tris(2,4,6-tribromophenoxy)-1,3,5-triazine) (CAS 25713-60-4)	
TV	Television	
UL 94	Standard for Tests for Flammability of Plastic Materials for Parts in Devices and Appliances	
WWTP	Wastewater treatment plant	
XRF	X-ray fluorescence	
µg/kg	micrograms per kilogram	

Electric and Electronic Equipment

Overview

Draft priority product

Electric and Electronic equipment (Device Casings)

Examples of electric and electronic equipment that might contain plastics that encase electric or electronic components include:

- TVs, computers and monitors
- Fax machines, printers, shredders and copiers
- Game consoles, audio and video equipment
- Other household electronics such as vacuum cleaners, corded phones, coffee makers
- Remote controls, temperature dials, and adaptors

This priority chemical-product combination only focuses on the device casing. It does not include an inaccessible electronic component, which is a part of an electronic product that is entirely enclosed within another material and is not capable of coming out of the product or being accessed during any reasonably foreseeable use or abuse of the product (RCW 70.365.010(5))[8].

Priority chemical

Organohalogen flame retardants and flame retardants identified under Chapter 70.240 RCW[9].

Organohalogen flame retardants are chemicals which prevent products from catching fire, and that contain one or more halogen elements bonded to carbon.

The non-halogenated flame retardants identified by the department under Chapter 70.240 RCW are: Triphenyl phosphate (TPP), Tri-n-butyl phosphate (TNBP), Ethylhexyl diphenyl phosphate (EHDPP), Tricresyl phosphate (TCP), and Isopropylated triphenyl phosphate (IPTPP).

Priority product summary

Residential and office electric and electronic equipment that contain plastic device casings are a significant use of flame retardants. We considered the criteria in section RCW 70.365.030[10] and are concerned about the contribution this product category makes to the amounts of flame

[8] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.010
[9] https://app.leg.wa.gov/RCW/default.aspx?cite=70.240
[10] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.030

retardants in our homes, workplaces and the environment. We are also concerned with the potential for exposure for infants, young children, occupations that use or dismantle electronics, and sensitive species, such as orcas. Detailed support for our listing of flame retardants in electric and electronic equipment as a priority product is shown below.

Background

Flame retardants are frequently added to plastic components in consumer goods to meet flammability standards. Although these standards don't mandate flame retardants, they do test materials used in products with electrical components to prevent the likelihood of a fire starting from electrical failures. For electric and electronic enclosures, the main flammability standard is UL 94, a standard that provides a method for classifying ignition characteristics of plastic materials (Underwriter's Laboratories Inc., 2007). These enclosures are made from several types of polymer resins such as high impact polystyrene (HIPS), acrylonitrile butadiene styrene polymers (ABS), polycarbonate/ABS blends (PC-ABS) and polyphenylene ether/HIPS blends (Pure Strategies, 2005).

Flame retardants can be broadly categorized into two categories based on how they are incorporated into the material: additive and reactive. **Additive flame retardants** are not chemically bonded to the polymers or chemical materials used in the product but are mixed in the product materials. **Reactive flame retardants** chemically bond to the polymers or chemical materials used in the product and become an integral part of the product structure. In addition to these two categories, there are three main classes of flame retardants: halogenated (also known as organohalogens), non-halogenated (such as organophosphates), and inorganic based. The most common class of flame retardants used in electronic enclosures are the organohalogen flame retardants.

Estimated volume of Flame Retardants in device casings for electric and electronic equipment

Historically, OctaBDE and DecaBDE were flame retardants commonly used in resins for electronic enclosures. But with their phase out, a number of alternatives have replaced their use. There have been a number of studies that have screened or tested electric and electronic enclosures. Common analytes are DecaBDE, DBDPE, TTBP-TAZ, TBBPA and RDP. Table 1 summarizes several flame retardants that have been measured in products with electronic enclosures above 0.1%. The products noted in bold are items in which an individual analyte was found in concentrations above 1% of the total mass.

Table 3. Product testing results with electronic enclosures.

Flame Retardants	Concentration (%)	Product types detected above 0.1% (bold above 1%)	References
Decabrominated Diphenyl Ether (DecaBDE)	N.D. – 8.8	**Computer monitor, TV, power adaptor,** laminator	Ballesteros-Gómez, de Boer, & Leonards, 2014; Gallen, 2014; Latimer, 2013
Decabromodiphenylethane (DBDPE)	N.D. – 16	**TV, power strip**, power adaptor	Ballesteros-Gómez et al., 2014; Jonker 2016
2,4,6-tris(2,4,6-tribromophenoxy)-1,3,5-triazine (TTBP-TAZ)	N.D. – 1.9	**TV, power strip,** electrical adaptors, heat sealer	Ballesteros-Gómez et al., 2014; Jonker, 2016
Tetrabromobisphenol A (TBBPA)	N.D. – 16	**TV, power adaptor, CD player, laminator, heated mattress, foot warming pad controller, shredder, battery charger and car charger, power strip, router, heat sealer,** tower fan	Ballesteros-Gómez et al., 2014; Ecology, 2014; Gallen, 2014; Jonker, 2016; Latimer, 2013
1,2-Bis(2,4,6-tribromophenoxy)ethane (BTBPE)	unknown	Office copier, power strip, TV, heat sealer	Ballesteros-Gómez et al., 2014; Latimer, 2013

There have been additional studies in which total bromine in casings was found at concentrations greater than 1% using XRF, but individual analytes were not identified. The products that were not mentioned in Table 3 but that likely contained a brominated flame retardant were projectors, deep fryers, a clothing iron, a sandwich press, a microwave oven, a coffee maker and space heater (Ecology, 2014; Gallen et al., 2014; Latimer, 2013). In the Ecology study, two of the samples, a flat iron and a clothing iron, had concentrations of total bromine greater that 1% and low levels of TBBPA. This could indicate that the TBBPA was used as a reactive flame retardant. An additional study suggested that halogenated flame retardant TTBP-TAZ is in TV casings at concentrations ranging from less than the limit of detection to 30.7% (Schreder, Peele, & Uding, 2017, not peer reviewed).

The amount of flame retardant needed to meet the flame retardancy function depends on multiple factors including the chemical structure of the polymer, its properties when heated

and when exposed to flames, as well as the type of flame retardants added. Common concentration ranges for halogenated flame retardants are between 2% and 25% (EPA, 2014a; Ministry of Environment and Food of Denmark, 2016).

In addition to chemicals with the function of flame retardants, impurities from these chemicals can also exist. The impurities typically share functional groups and basic structures of the parent compounds. Several such impurities have been identified in electronic enclosures (Ballesteros-Gómez et al., 2014, 2016; Jonker et al., 2016).

Estimated volume used in Washington

Most households contain items with enclosed electronic circuitry, such as computers (which, along with laptops, are in 77% of American homes) monitors, televisions (96% of homes), audio or video equipment, gaming consoles, cameras, clothing irons, vacuum cleaners, etc. (Nielson, 2019; U.S. Census Bureau, 2018). An average household has 30 items with enclosed electronic circuitry (Mars & Nafe, 2016), and the average replacement rate is estimated at two per year (Cleet, 2018). Commercial businesses also own significant (but unknown) numbers of these items.

Washington state's E-Cycle program collects computers, televisions and monitors (Washington Materials Management & Financing Authority, 2018). From 2014 to 2018, between 4 and 6 million pounds of plastic enclosures were collected yearly. These items are a major but unknown fraction of all such enclosures, since not all electronics qualify for acceptance to Washington's E-Cycle program, and not all qualified items are recycled.

Potential for exposure to sensitive populations

There is potential for people, including sensitive populations such as children and workers, to be exposed to flame retardants used in electronics. Additive chemical flame retardants are not chemically bound to the treated materials and can migrate out of products. A large number of flame retardants have been detected in house dust studies. BTBPE (Brown, Whitehead, Park, Metayer, & Petreas, 2014; Dodson et al., 2012; La Guardia & Hale, 2015; Schreder & La Guardia, 2014) and DBDPE (Brown et al., 2014; Johnson, Stapleton, Mukherjee, Hauser, & Meeker, 2013; Stapleton et al., 2009) have been widely detected in house dust. TPP was detected in indoor dust in studies of homes in North Carolina, Boston, California, and Canada (Knudsen, Hughes, Sanders, Hall, & Birnbaum, 2016; La Guardia, Schreder, Uding, & Hale, 2017; Liu, He, Hites, & Salamova, 2016; Ma, Venier, & Hites, 2012; Zhou et al., 2014). TTBP-TAZ and TBBPA have also been found in house dust (Abdallah, Harrad, & Covaci, 2008; Ballesteros-Gómez et al., 2014; Fromme et al., 2014; Guo et al., 2018; Schreder & La Guardia, 2014).

Flame retardants in electronics can contribute to concentrations in house dust. In 2016, Canadian researchers found that surface wipes of home and office electronics had detectable

concentrations of organohalogen and phosphorous based flame retardants. Concentrations in surface wipes of hard polymer casings were correlated with household dust samples, suggesting that the additive flame retardants used in electronics contribute to household dust concentrations (Abbasi, Saini, Goosey, & Diamond, 2016). A more recent study from the Netherlands found that time, age, hours of operation and use of the standby function of electronics had a significant influence on flame retardant concentrations in house dust (Sugeng, de Cock, Leonards, & van de Bor, 2018). This supports an older study that found decaBDE in dust was associated with the presence of televisions in the bedroom (Allen, McClean, Stapleton, & Webster, 2008). TCP and TPP concentrations in house dust have been associated with proximity to electronics (Brandsma, de Boer, van Velzen, & Leonards, 2014). Similar relationships have also been found for DecaBDE and another organohalogen flame retardant, Hexabromocyclododecane (HBCD) (Harrad, Abdallah, & Covaci, 2009; Muehnor & Harrad, 2012).

Exposure to flame retardants is widespread in the U.S. Most of the available studies focused on PBDEs, although their production and use was phased out in 2004 (Jinhui, Yuan, & Wenjing, 2017). PBDEs are environmentally persistent and levels of PBDEs in blood are proportional to a cumulative exposure in the preceding months or even years. The CDC-NHANES study assessed ten PBDE congeners between 2003 and 2004, finding BDE-28, BDE-99, BDE-100, and BDE-153 in more than 60% of blood samples representative of the U.S. general population (CDC, 2009). DecaBDE was not included in that study even though it has been widely used as a flame retardant in electrical and electronic equipment. Levels of PBDEs in children tend to be higher than levels in adults (CDC, 2009; Butt, Congleton, Hoffman, Fang, & Stapleton, 2014; Stapleton et al., 2012).

CDC analyzed stored urine samples for nine organophosphate flame retardants to learn more about PBDE replacements in people six years and older from the 2013 – 2014 NHANES (Ospina, Jayatilaka, Wong, Restrepo, & Calafat, 2018). CDC developed a new method to measure urinary metabolites for several other organophosphate flame retardants and found widespread exposure in the general U.S. population. The study detected the TPP metabolite DPHP in more than 80% of the samples analyzed, suggesting widespread exposure to TPP. In general, women had higher levels than men, and children had higher levels than adults. The study comprised a representative sample of the broader U.S. population, and the results are consistent with previous studies that used only convenience samples.

DBDPE was detected in less than 10% of samples of maternal serum and breast milk collected between 2008 and 2009 in the Sherbrooke region of Canada (Zhou et al., 2014). BTBPE has been detected at low levels in less than 10% of the samples of maternal serum in Norway (Cequier, Marcé, Becher, & Thomsen, 2015).

In U.S. residents, TBBPA has been studied in serum, breast milk and adipose tissue. TBBPA was detected in 35% of the 43 breast milk samples collected in 2004 – 2005 from first-time mothers

in Boston, Massachusetts (Carnigan et al., 2012). TBBPA was detectable at low levels in 31% of human adipose (fat) tissues from people who underwent liposuction surgery in New York City (Johnson-Restrepo, Adams, & Kannan, 2008).

The only widespread detection of TBBPA was 95% of an Inuit population in northern Canada that eats marine mammals (Dallaire et al., 2009). TBBPA has also been studied in other countries and detected in serum (Alberta Health and Wellness, 2008; Kiciński et al., 2012; Kim & Oh, 2014) and breast milk (Cariou et al., 2008; Pratt et al., 2013; Shi, Wu, Li, Zhao, & Feng, 2009; Shi et al., 2013).

Abdallah et al. (2008) estimated human exposure to TBBPA in adults and toddlers (6 – 24 months) via air inhalation, dust, and food ingestion. Dust ingestion appears to be the major pathway of exposure to TBBPA for toddlers in the United Kingdom. A toddler weighing 10 kg and ingesting 200 mg dust per day with TBBPA at the 95th percentile concentration reported in their study would ingest 100 times more TBBPA via house dust than through their normal diet.

Certain occupations that are exposed to electronics have been found to result in relatively high exposure to some flame retardants compared to exposures in the general public. The workers that are most relevant to flame retardants in electronics are office workers, fire fighters, and electronics recyclers. Sjödin et al. (1999) found elevated PBDEs in computer workers compared to cleaning personnel. Upon further evaluation, Jakobsson et al. (2002) found certain PBDE congeners were approximately five times higher in computer technicians compared to other clerks and cleaning personnel. Other studies did not find a clear relationship between exposure to flame retardant-containing equipment and office workers (Watkins, 2011). Workers recycling electronics are exposed to higher levels of flame retardants. Higher levels of PBDEs were found in recycling workers in China (Qu et al., 2007) and Sweden (Sjödin et al., 1999) compared to control groups. Sjödin et al. (1999) found that levels of PBDEs in workers in an electronics dismantling plant were about five times higher than other workers.

Firefighters are exposed to flame retardants during work, and California has a biomonitoring program that includes firefighters: the Firefighter Occupational Exposure (FOX) project. Eight PBDE congeners were measured and firefighters were found to have higher levels of several PBDEs compared to the general California population in NHANES (Park et al., 2015). In the FOX study, nearly one third of the firefighters had particularly high serum levels of DecaBDE, but the levels were lower than in electronic recyclers.

Potential environmental exposure and exposure to sensitive species

Flame retardants used in electronics have been found in Washington's environment. In Washington, PBDE flame retardants have been found in many different environmental media and appear to be a ubiquitous contaminant in aquatic systems (Ecology, 2006, 2011a, 2011b).

Even though some of the most toxic PBDE congeners (pentaBDE and octaBDE) have been restricted since 2004, they are environmentally persistent and can still be found in many environments. DecaBDE may be debrominated to form other congeners (Zhao, Rogers, Ding, & He, 2018). Ecology has detected DecaBDE in WWTP effluent (Ecology & Herrera Environmental Consultants, Inc., 2010), surface water in Puget Sound (Ecology, 2011b), stormwater (Ecology, 2018), sediment (Ecology, 2011a, 2011b, 2016b, 2017, 2018) and freshwater fish (Ecology, 2006, 2012, 2016a, 2019a).

DBDPE is also found in Washington's environment. While it was not detected in WWTP effluent (Schreder & La Guardia, 2014) or sediment (Ecology, 2016b), it has been detected in WWTP influent (Schreder & La Guardia, 2014) and freshwater fish (Ecology, 2016a). TBBPA was not detected in freshwater fish in Washington, but it has been detected in a Swedish river (Gustavsson et al., 2018). BTBPE has been detected in Washington sediment (Ecology, 2016b) and fish (Ecology, 2019b), but was not detected in more recent sediment or stormwater studies (Ecology, 2018). There are no Washington-specific data for TTBP-TAZ.

Flame retardants from electronic enclosures have the potential to contribute to environmental flame retardant concentrations. Flame retardants from electronics can contribute to house dust concentrations, which can be tracked outside (Wild et al., 2015) and released into wastewater when we wash textiles with house dust on them (Saini, Thaysen, Jantunen, McQueen, & Diamond, 2016; Schreder & La Guardia, 2014).

Flame retardants in electronic components can continue to be released after their useful life is over and they are disposed of in landfills (Choi, Lee, & Osako, 2009; Kim, Osako, & Sakai, 2006; Osako, Kim, & Sakai, 2004). Electronic products have a use life of 5 – 15 years (OECD, 2019). In 2012, EPA estimated that electronic waste comprised 1.4% of the municipal solid waste stream in the U.S. (EPA, 2014b). The plastic components of electronics (e.g. electronic enclosures) comprise 25 – 30% of the product by weight (Meyer & Katz, 2016). Based on this and the concentrations noted above, as much as 0.1% of municipal solid waste could be flame retardants from electronic enclosures.

Many electronic components can be recycled, though EPA (2014b) estimated that less than 30% of electronic products are recycled (Meyer & Katz, 2016). Flame retardants can be released into the environment during the recycling process (Matsukami et al., 2015). In fact, TTBP-TAZ has been found in e-waste dust (Guo et al., 2018). When new consumer products are developed from recycled material, there is the potential for flame retardants originally used in electronics to reappear in items such as toys, food packaging and other single use plastic goods (Turner, 2018). This is an additional path by which flame retardants can be released into the environment when discarded, and through which sensitive populations can be exposed.

Exposure to flame retardants in the environment is a concern for sensitive species. PBDEs, BTBPE and other brominated flame retardants are persistent in the environment and can be transported long distances. This property has made them ubiquitous in environmental media

across the globe and as far away as the Arctic, which is an indicator of persistence and bioaccumulation used by the Stockholm Convention on Persistent Organic Pollutants (de Whit, Herzke, & Vorkamp, 2010). Certain PBDE congeners, such as PBDE-99, are highly bioaccumulative, which is concerning for species at higher trophic levels, such as orcas. While studies have shown that decaBDE does not bioaccumulate, lower brominated degradation products (including lower substituted PBDEs) bioaccumulate in organisms and concentrate in the environment (EPA, 2009c). Chemicals that bioaccumulate can increase in concentration in animals at higher trophic levels. This process, termed biomagnification, makes orcas and other predators more vulnerable to bioaccumulative chemicals in the environment. Other brominated flame retardants like TBBPA and BTBPE appear to have lower bioaccumulation potential than PBDEs (La Guardia et al., 2012; Morris et al., 2004). However, modeled K_{ow} values, which predict the way a chemical partitions between water and fat, suggest that many of the replacement brominated flame retardants may still bioaccumulate (Kuramochi, 2014), and many have been detected in wildlife, though at lower concentrations than PBDEs or HBCD (Covaci et al., 2011).

In 2018, Governor Inslee established the Southern Resident Orca Task Force, a 50 member work group charged with developing plans for recovering Puget Sound's struggling southern resident orca population. PBDEs have been identified by the Southern Resident Orca Task Force as a primary contaminant of concern. PBDEs have been found in Southern Resident Orcas (O'Neill et al., 2015; Rayne, Ikonomou, Ross, Ellis, & Barrett-Lennard, 2004; Sloan et al., 2010). Chinook salmon that spend time in Puget Sound, an important orca food source, also have elevated concentrations of PBDEs (O'Neill et al., 2015; Puget Sound Partnership, 2017; Sloan et al., 2010). A similar trend has been observed in Chinook salmon that spend more time near urban areas along the Columbia River (Sloan et al., 2010).

Existing regulations

Currently, no U.S. federal restrictions exist around flame retardants in electric and electronic enclosures. In 2017, the federal Consumer Product Safety Commission (CPSC) granted a petition requiring them to initiate rulemaking to prohibit organohalogen flame retardants in four product categories including plastic casings surrounding electronics. To date, the CPSC has not banned organohalogen flame retardants in these products (CPSC, n.d.).

Regarding flame retardant chemicals that are frequently used in electronic enclosures, in 2019, EPA proposed a rule under TSCA that will ban the processing and distribution in commerce of most uses of decaBDE (EPA, 2019a). TPP and TBBPA are on EPA's proposed list of 20 high priority chemicals to review for the next risk evaluations under TSCA (EPA, 2019b).

In the U.S., there are several state regulations to limit exposures to flame retardant chemicals in electric and electronic enclosures. Washington state banned the use of PBDEs in all nonedible products, including electronic enclosures, under RCW 70.76 Polybrominated

Diphenyl Ethers—Flame Retardants. This regulation bans all PBDEs in these products as of January 2011.

California, Hawaii, Illinois, Maine, Maryland, Michigan, Minnesota, New York, Oregon, and Vermont also have regulations restricting the use of certain flame retardants in products. While products are not well defined, electronics are not exempted (Safer States, 2019). In 2017, Washington restricted additive TBBPA and several other flame retardants commonly found in furniture. This restriction includes furniture with electronic enclosures.

A flame retardant ban recently adopted by the European Union means that after March 1, 2021, the use of halogenated flame retardants in enclosures and stands of electronic displays will be prohibited (European Commission, 2019; European Union, 2019).

Availability of safer alternatives

There are alternative materials and varying types of flame retardants used in plastic electronic enclosures to meet flammability standards. In addition to additive halogenated flame retardants, reactive halogenated flame retardants, and those identified by Ecology under Chapter 70.240 RCW[11], several additive organophosphate flame retardants such as RDP have been used in plastic enclosures.

Other solutions include using metal casings or removing the electronic source from the casing. Several alternatives have been investigated by other organizations (Ecology & Health 2008; EPA, 2014a; Ministry of Environment and Food of Denmark 2016; TCO certified, 2019). As part of our Safer Products for Washington process, we will address the safety and feasibility of these alternatives in phase 3.

[11] http://app.leg.wa.gov/RCW/default.aspx?cite=70.240

Inks and Pigments

Overview

Draft priority product

Printing Inks

Priority chemical

Polychlorinated Biphenyls

Priority product summary

Printing inks are a significant source of inadvertently generated PCBs. Ecology made this determination by considering the criteria in RCW 70.365.030[12] of Safer Products for Washington. Several bodies of water in the state are considered impaired due to PCB contamination. PCBs have a wide variety of toxic effects, accumulate in people and animals, and contaminate our food supply.

Ecology estimates that colored pigments contained in inks are the largest source of inadvertent PCB contamination in consumer goods, and up to 56 million pounds of printing ink are used per year in Washington. Ecology has listed PCBs as a persistent, bioaccumulative, and toxic chemical, and has written a chemical action plan (CAP)[13] to address them. Reduction and targeting iPCBs production was identified as a recommendation both by Ecology's CAP and by Governor Inslee's Orca Task Force. Detailed support for our listing of PCBs in printing inks as a priority product is shown below.

Background

PCBs have historically been used intentionally in a wide range of products, such as electronic equipment, caulking, and carbon copy paper. Most intentional uses, often termed "legacy" uses, have been banned in the U.S. since 1979. Inadvertent PCBs (iPCBs) are PCBs that are not intentionally added to products, but are instead produced as an unintended byproduct of the manufacturing process. Although legacy PCBs are still present in some products currently in use and are still the main source of environmental contamination, iPCBs are the predominant source of new PCBs in consumer goods (Ecology & Health, 2015).

There are 209 distinct PCB compounds (known as congeners) depending on the number and location of chlorination on the biphenyl molecule. Different congeners have different physical properties, toxicity, and environmental fates. The general structure for a PCB is shown below,

[12] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.030
[13] https://fortress.wa.gov/ecy/publications/documents/1507002.pdf

where any of the numbers can contain a chlorine molecule or hydrogen molecule, depending on the congener.

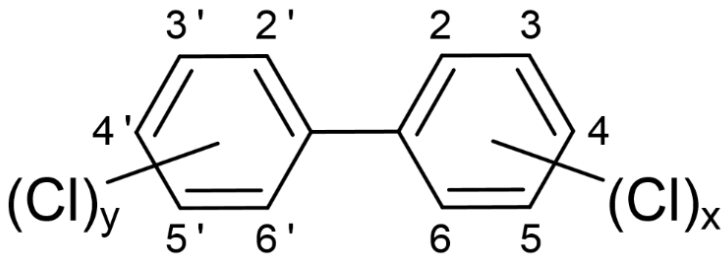

Figure 1. Molecular structure of polychlorinated biphenyls.

Estimated volume of PCBs used in printing inks

iPCBs are widespread in products containing printing inks. Pigments and other compounds can become contaminated with PCBs because of non-specific chlorination processes in many reactions where carbon, chlorine, and heat are involved. Pigments affected include diarylide yellows, phlthalocyanines, and titanium dioxide. Figure 2 shows a list of organic pigments that have been found to contain iPCBs and the PCB congeners that were detected (Heine & Trebilcock, 2018).

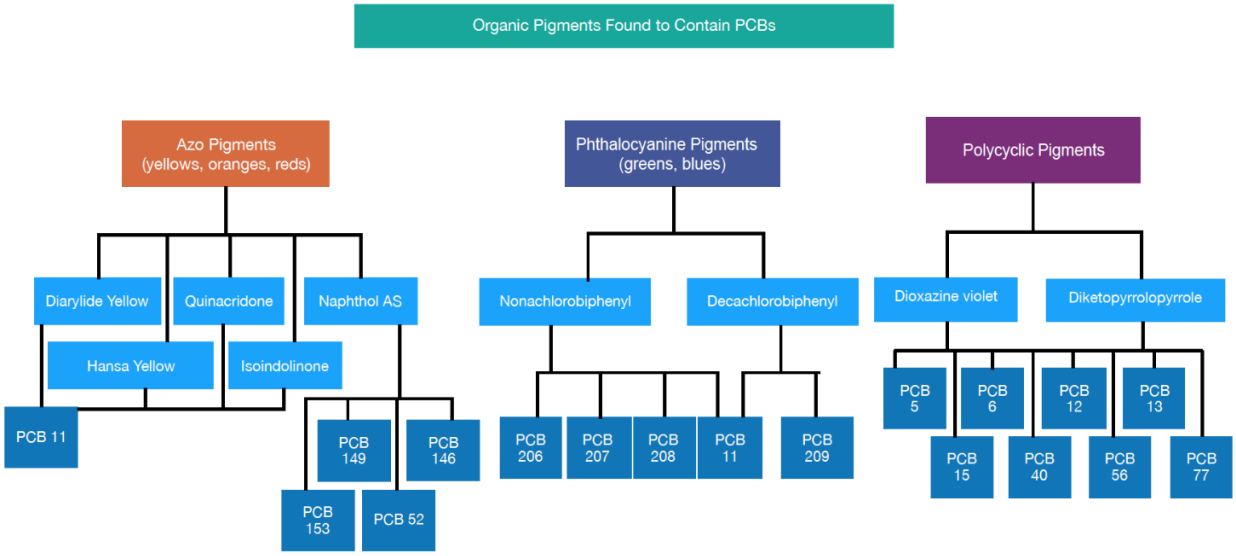

Figure 2. Organic pigments found to contain inadvertent PCBs.

PCB 11 contamination of diarylide yellow pigments is the source that is most associated with pigment contamination, but other congeners have been identified in a variety of pigments.

Many congeners can be produced during pigment production, but PCB 11 was not a component of any legacy PCB products (such as common PCB mixtures sold under the trade name "Aroclor") so it is considered a hallmark of inadvertent production. This means that any detections of PCB 11 in sampling, whether in the environment or of products, likely originated from inadvertent production. However, other PCB congeners can also be produced inadvertently, and processes subject to iPCB contamination often contain a large variety of congeners.

One source of iPCBs is from printing inks. This likely occurs mostly in pigment ingredients in inks, though other ingredients may contribute as well, especially if silicone components are used, as they are in some inks (Ecology & Health, 2015). EPA requires reporting of iPCB generation, and pigment and dye manufacturing are the most commonly reported processes that produce iPCBs. Ecology estimated that pigment and dye manufacturing is the current product group whose manufacture contributes the most to PCB levels (Ecology & Health, 2015).

Printing inks vary widely in components, depending on the intended use, but usually contain pigment or dye (provides color), binder (wets and disperses pigment), solvents (dissolve binder and mix components), and excipients (affect properties of ink). Printing inks are the predominant use of pigments, using an estimated 50% of what is manufactured, with other uses including paints, coatings, and plastics (Nestler, Heine, & Montgomery, 2019).

There are limited studies investigating iPCB concentration in printing inks specifically, but Table 4 shows a summary of studies investigating PCB concentrations in inks, pigments, and consumer products containing printed material. For most printed products, unless there is another ingredient likely to cause PCB contamination, we assume that the majority of iPCB content comes from inks used in their production, since that is the most likely source. These results show that PCB contamination of inks and printed products is widespread and well-documented. They also show that in products containing PCB 11, there are often other congeners present as well, suggesting that the impact of the products is not limited to PCB 11.

Table 4. Product testing results with inks and pigments.

Year	Product	% of Samples with PCB	Total PCB Concentration	PCB 11 Concentration	Other Congeners Detected	Reference
2009	Paint	15/33 (45%)	2 – 200 ppb	0 – 16.4 ppb(13/15 samples)	8, 6, 4, 1, 12/13, 2, 3, 209 all in more than 40% of samples	Hu, Dingfei; Hornbuckle, 2010
2009	Printed paper and plastic	12/12	Not Tested	.11 – 38 ppb	Not Tested	Rodenburg, Guo, Du, & Cavallo, 2010

Year	Product	% of Samples with PCB	Total PCB Concentration	PCB 11 Concentration	Other Congeners Detected	Reference
2012	Pigments	376/588	359 samples <.5ppm 17 samples >50ppm	Not Reported	Not Reported	METI 2013a via Heine & Trebilcock, 2018. Original source no longer online
2013	Yellow Pigments	24/24 (100%)	50 ppb – 919 ppm	42 ppb – 918 ppm	52 (4410 ppb max), 28 (1142 ppb), 77 (670 ppb), 126 (33.5 ppb), 153 (90.6 ppb)	Shang et al., 2014
2014	Printed paper and fabric	Not Reported	Not Tested	1.5 – 86 ppb	Not Tested	Guo, Capozzi, Kraeutler, & Rodenburg, 2014
2015	Municipal Products (products used by the City of Spokane)	39/41 (95%)	0 – 2509 ppb	0 – 63.8 ppb in paint	77, 209, 6, 64/72, 50, 28, 31, 118, 52/69 and others above 100ppb	Spokane, 2015
2016	Consumer Products	193/216 (89.4%)	0 – 2320 ppb	0 – 2290 ppb (64% of samples)	52 (23% of samples), 61/70/74/76 (19%), 31 (15%)	Stone, 2016
2019	Paint Colorants	8/8 (100%)	0.03 – 284 ppb	0.3 – 1.2 ppb (50% of samples)	209 (256 ppb), 208 (3.7 ppb), 206 (4.8 ppb), 146 (1.3 ppb), 52 (2.4 ppb)	Jahnke & Hornbuckle, 2019

Estimated volume used in Washington

Up to 56 million pounds of printing ink are used per year in Washington. One report estimated that the U.S. printing inks market was approximately 2.5 billion pounds in 2017 (Mordor, 2018). This estimate would put Washington's share at 56 million pounds.

This is supported by data from the Color Pigments Manufacturers Association (CPMA), which estimates that the total amount of phthalocyanine and diarylide pigments imported or manufactured in the U.S. is about 90 million pounds per year (Ecology & Health, 2015). This would mean Washington's share (by population) is around 2 million pounds of these pigments. Printing inks contain 5 – 30% pigment by weight (PCC, 2018), so if we only consider these two types of pigments, that would amount to approximately 7 – 40 million pounds of printing ink used.

Ecology's PCB chemical action plan[14] estimated that Washington's share of PCB 11 from yellow pigment is between 0.02 and 31 kg per year, with data from the CPMA pointing toward an annual estimate of 9 – 20 kg (Ecology & Health, 2015).

Potential for exposure to sensitive populations when used

Nearly all people, including infants, are exposed to PCBs. Levels of PCBs in people have declined since the 1980s, but there is still widespread detection of PCBs in the U.S. population, including in infants and children (CDC, 2019; Ecology & Health, 2015). People are mostly exposed to a mixture of PCBs rather than a single PCB compound. PCBs have been detected in human blood serum, adipose tissue, breast milk, and in cord blood (ATSDR, 2019). Some PCBs can remain in the body for years after exposure—varying by type of organism and type of PCB congener—and blood levels generally increase with age (Ecology & Health, 2015). Because PCBs are more readily absorbed than excreted, the levels of individual PCB congeners in the body may vary by exposure source and by differences in how bodies process them (CDC, 2017).

PCBs have been measured and detected in the blood serum of the U.S. general population over age 11 since the 1999 NHANES survey cycle (CDC, 2019). PCB congeners 118, 138, 153, and 180 have been found at higher levels in the environment and in human blood samples than other PCB congeners (EPA, 2015). They were detected in the majority of samples for women aged 16 – 49 from 1999 – 2014 (EPA, 2015). Measured levels of PCBs in human blood decreased by an estimated 87% from 1973 – 2003 (Schecter et al., 2005; Sjodin et al., 2004). However, PCB 153 was detected in both children and adults more recently, with a median level of 7.4 ng/g lipid in 6 – 9-year-old girls, and 0.21 – 55.6 ng/g lipid in the general population (EPA, 2015; CDC, 2019).

In 2013, studies reported the presence of PCB 11, which is associated with iPCB production, in air samples and in the blood of children and mothers (Marek, Thorne, Wang, DeWall, & Hornbuckle, 2013; Zhu et al., 2013). This is an indication that airborne PCBs continue to be an environmental problem affecting large populations. A 2015 study reported PCB congeners 11, 14, 35, 133, and 209 as the most frequently detected non-Aroclor congeners in the serum of

[14] https://fortress.wa.gov/ecy/publications/documents/1507002.pdf

participants (Koh, Hornbuckle, & Thorne, 2015). More information on PCB exposures in people is available in the PCB chemical action plan[15] (Ecology & Health, 2015).

There is the potential for people to be exposed to PCBs in inks and pigments. Food is the main source of exposure for the general population, and we are particularly concerned with levels of PCBs in fish we eat. PCBs occur at the highest concentrations in fatty foods (e.g., dairy products and fish). PCBs from inks may enter fish through stormwater and wastewater effluent, thus leading to human dietary exposure (Ecology & Health, 2015).

Paint colorants—which have similar ingredients as inks—have the potential to emit PCBs, which can increase in the environment to concentrations of more than 500 pg/m^3 within hours of application (Jahnke & Hornbuckle, 2019). PCBs have also been detected in residential environments from indoor air and house dust (Takeuchi, Anezaki, & Kojima, 2017). A study reported concentrations of PCB 11 in indoor air in homes and schools in East Chicago and Columbus Junction (Ampleman et al., 2015). As part of that study, researchers calculated PCB 11 inhalation rates and found adolescents had greater inhalation exposure than mothers due to spending times in schools with high total PCB levels in indoor air (Ampleman et al., 2015).

PCBs in inks and pigments make a significant contribution to exposure. People are exposed to PCBs in air, water, soil, and house dust. Humans can also be exposed to PCBs through direct contact with consumer products that use pigments (Guo, Capozzi, Kraeutler, & Rodenburg, 2014; Rodenburg, Geo, Du, & Cavallo, 2010) as well as from the presence of PCBs in the environment (Vorkamp, 2016).

Biomonitoring studies for PCB congeners unique to dyes and pigments showed that 65% of 85 women in the Midwest had trace levels of PCB 11 in their blood (Ecology & Health, 2015; Marek et al., 2013). On average, about 10% of the total PCBs in study participants came from non-Aroclor PCBs. This study also found that, on average, 50% (but up to 100%) of non-Aroclor PCB concentrations in the participants were likely due to pigment exposure (Koh et al., 2015).

PCB 11 concentrations have not decreased since 2004, and in 2007, PCB 11 was found in 91% of air samples taken near 40 Chicago area elementary schools (Heine & Trebilcock, 2018).

Exposure to PCBs in inks and pigments is particularly relevant for infants, children, women of childbearing age, and tribal members. Although levels of PCBs in blood for young people have declined over the past three decades, there is still potential for exposure through the use of products that contain these chemicals. Due to the presence of PCBs in consumer products, and their release into the air, dust, and food supply, there is potential for infants and young children to be exposed, especially since they have more contact with house dust (Harrad et al., 2009; Rudel et al., 2003, 2008; Takeuchi et al., 2017). Infants are also exposed to PCBs in breast milk, as they accumulate in the mother's body over many years and are stored in the fat in breast

[15] https://fortress.wa.gov/ecy/publications/documents/1507002.pdf

milk. For example, serum levels increased after birth in breastfed infants and then decreased in early adolescence (CDC, 2019).

A study analyzed blood serum levels in children and their mothers from urban and rural U.S. communities (n = 200) for 209 PCBs from 2008 – 2010, and found widespread detection of all congeners, including PCB 11. This study reported variability of all PCBs and major metabolites in two generations of people, which suggests that short-term exposures to PCBs may be a significant component of what is measured in human serum (Marek et al., 2014).

Tribal members may also be more at risk from exposure to PCBs than the general public. Tribal members often consume more fish (Ecology, 2013), and exposure to PCBs has been linked to higher incidence of diabetes and cardiovascular disease in tribal members (Nestler et al., 2019). The National Tribal Toxics Council does not consider the 50ppm allowance for iPCBs in pigments to be sufficiently protective of their population due to increased fish consumption. The Confederated Tribes of the Umatilla Indian Reservation has requested a lower tolerance level, specifically calling out inks and pigments (Nestler et al., 2019).

Potential for exposure to sensitive species when used

PCBs have been found in Washington's environment. PCBs have been found in freshwater, marine water, and sediment in Washington. PCB levels in Washington mussels, an indicator species for detecting local environmental contamination, are found well above national median concentrations (Ecology & Health, 2015). PCBs, including PCB 11, have been detected in atmospheric deposition in the Seattle metro area (Rodenburg, Winstanley, & Wallin, 2019). See Ecology's published chemical action plan[16] (Ecology & Health, 2015) for more details on PCBs in Washington's environment.

The fish tissue equivalent of Washington's human health water quality criterion for PCBs is 5.3 ppb. Washington Department of Fish and Wildlife measured the total PCB concentrations in edible tissues of four marine fish species as compared to this value. All samples of Herring and Chinook, and most (70 – 80%) of the English Sole and Coho have exceeded the criterion (Ecology & Health, 2015). There are 158 303(d) listings for PCBs in Washington's 2012 Water Quality Assessment. This means they are in the polluted water category, and beneficial uses—such as drinking, recreation, aquatic habitat, and industrial use—are impaired by PCB pollution (Ecology & Health, 2015).

PCBs from inks have the potential to contribute to environmental PCB concentrations. PCB 11 is considered a hallmark of iPCB contamination, specifically from pigments and dyes, since it is known to be present in many printed materials, and it is not found in legacy PCB products (Heine & Trebilcock, 2018). PCBs have been shown to leach from printed materials when

[16] https://fortress.wa.gov/ecy/publications/documents/1507002.pdf

exposed to water, such as from washing printed clothing, recycling printed paper, or if printed material is exposed to rain during storm events (Guo et al., 2014).

Limited data is available, but it was estimated that two paper recycling facilities in Washington discharge 28g of PCBs per year, with 3.8g being PCB 11, and that the Spokane River Wastewater Treatment Plant (WWTP) was discharging 71g of PCBs/year (Ecology & Health, 2015). Product testing results suggest that pigments may account for the majority of detected PCB 11 in the environment (Guo et al., 2014), and thus almost certainly contribute other congeners as well.

Exposure to PCBs in the environment is a concern for sensitive species. PCBs bioaccumulate in animals, and increase in concentration higher in the food chain. They have been noted as a chemical of concern by the Southern Resident Orca Task Force, and reducing exposure was one of the four recommendations of the task force (Southern Resident Orca Taskforce, 2018a). PCB contamination reduces the health of aquatic organisms throughout the food chain, including forage fish, salmon, and orcas.

PCBs have been detected in many aquatic organisms at potentially harmful levels throughout Washington. Levels of PCB associated with health impacts are observed in seals in the Strait of Georgia, and most Southern Resident Killer Whales exceed the health effects thresholds for PCB residues (Cullon et al., 2009; Hickie, Ross, Macdonald, & Ford, 2007). PCBs are persistent chemicals, so once released in the environment they can be challenging or impossible to remove, affecting wildlife for years to come. See Ecology's chemical action plan[17] for more details about effects on wildlife health (Ecology & Health, 2015).

Existing regulations

In 2014, RCW 39.26.280[18] established purchasing policies for Washington state with a preference for products and packaging that do not contain PCBs. In 2018, Governor Jay Inslee signed Executive Order 18-02: Southern Resident Killer Whale Recovery and Task Force. Through this executive order, the governor directed state agencies to implement nine immediate actions to benefit Southern Resident Orca Whales. In September of 2018, the task force published draft recommendations, including accelerating the implementation of the 2014 PCB purchasing law "to reduce the PCBs entering Puget Sound from products." Product suppliers to the state will provide information about PCBs in their products, and this information can be shared with other purchasers that want to avoid products containing PCBs (Southern Resident Orca Taskforce, 2018b).

The federal Toxic Substances Control Act (TSCA) bans intentional production and import of intentionally added PCBs and limits iPCBs in pigments to an average of 25 ppm, with a maximum of 50 ppm. This level was set to reduce the impact on industry, except for in the case

[17] https://fortress.wa.gov/ecy/publications/documents/1507002.pdf
[18] https://app.leg.wa.gov/RCW/default.aspx?cite=39.26.280

of diarylide and phthalocyanine pigments, when it was considered feasible to reduce concentrations of iPCBs (Nestler et al., 2019).

The Stockholm Convention does not specify a limit on iPCBs in pigments. However, it dictates that signees develop an action plan to reduce iPCB releases. Some countries party to the Convention, such as Canada, have set a 50 ppm maximum limit and 25 ppm average limit on iPCBs (Nestler et al., 2019).

Availability of safer alternatives

Methods of manufacturing are possible to reduce or eliminate (in the case of titanium dioxide) the amount of inadvertent PCB content in pigments and dyes. In addition, inadvertent PCB production is not a problem for non-chlorinated pigments. Some organizations, including HP and Apple, have a policies in place prohibiting the purchase of ingredients (including inks) with PCBs over 0.1 ppm (Heine & Trebilcock, 2018). Product testing has shown printed material with a wide variety of colors containing PCB levels below the detection limit, indicating that low PCB inks are available (Ecology & Health, 2015; Stone, 2014). As part of our Safer Products for Washington process, we will address the safety and feasibility of printing inks with lower iPCB content in phase 3.

Carpets

Overview

Draft priority product

Pretreated carpets and rugs used in residential and commercial settings

Priority chemical

Perfluoroalkyl and polyfluoroalkyl substances (PFAS)

Priority product summary

Pretreated carpets and rugs used in residential and commercial settings are significant sources and uses of PFAS. Ecology made this determination by considering the criteria in RCW 70.365.030[19] of Safer Products for Washington. We are concerned about the contribution carpet makes to the amounts of PFAS in our homes, workplaces and environment, as well as the potential for exposure for infants, young children and workers. In 2009, EPA estimated that carpet and carpet care products were the second largest use of fluorotelomers, with the broadly defined "textiles" category ranking first.

We estimate that 1,290 – 2,074 metric tons of PFAS are brought into Washington homes and workplaces in carpet each year, and even more (5,376 – 8,640 metric tons) are disposed of in our landfills annually. In addition to making a significant contribution to PFAS use, carpets can contribute to human PFAS exposure, particularly for children and infants who spend more time on or near the floor. Children living in homes with treated carpet have higher exposures to PFAS.

The PFAS from our carpets can also be released into the environment. PFAS are environmentally persistent, making clean-up difficult or impossible in some cases. In 2018, Governor Inslee convened the Southern Orca Task Force to recommend actions to recover the southern orca population. The task force named PFAS as an emerging concern for orcas. Detailed support for our listing of PFAS in carpet as a priority product is shown below.

Background

PFAS have been used in pretreated carpet since the 1970s. The specific PFAS used have changed over time. From 1970 to 2002, the largest use of PFOS-derived substances was for carpet applications (48,000 tons globally from 1970 to 2002) (DEPA, 2013; Paul, Jones, & Sweetman, 2009). Currently, fluoropolymers or fluorotelomer-based acrylate polymers are generally used for carpet stain resistance and carpet care treatments (Bowman, 2018; KEMI

[19] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.030

2015). However, other PFAS can also be present as impurities (for example, PFHxA and PFBA) (Bowman, 2018) and can be formed during environmental degradation (FTOHs and PFCAs) (Washington & Jenkins, 2015).

Estimated volume of PFAS used in carpet

We estimate that PFAS are found in over half of carpets in Washington at concentrations that can make significant contributions to the volume of PFAS in our homes and workplaces. A 2008 survey estimated that 56 – 90% of carpets sold in Denmark were treated with fluorinated substances (DEPA, 2013). PFAS concentrations in carpet vary depending on the specific compound (Table 5). If we assume that there are 50 kg of carpet in the average home, treated carpet could contribute 2,860 – 72,600 µg PFCA and 3,675 – 200,500 µg FTOH/FTS to the average home (Ecology, 2019). The Swedish Chemicals Agency estimates that PFAS, specifically PTFE and PASF, can constitute around 15% of the fiber weight in synthetic carpets (KEMI, 2015), which would contribute 750,000 µg PFAS to the average home.

Table 5. Reported concentrations of PFAS in carpet.

The concentrations vary by PFAS analyte. (^ indicates estimation by KEMI, 2015)

Product Description	Concentration	PFAS Measured	Reference
Pre-treated carpeting	484 µg/m^2	Total PFCA (C5 – C12)	EPA, 2009
Pre-treated carpeting	57.2 µg/kg	Total PFCA (C4 – C14)	Kotthoff, 2015
Carpet	4,010 µg/kg	FTOH/FTS	Liu et al., 2014
Carpet samples	73.5 µg/kg	FTOH/FTS	Kotthoff, 2015
Carpet	15%^	PTFE and PASF	KEMI, 2015

Estimated volume sold and disposed of in Washington

We estimate that 1,290 – 2,074 metric tons of PFAS are found in carpets sold annually in WA. Approximately 15,360 metric tons of carpet (minus their untreated backings) are sold yearly in WA. This estimate is based on California Carpet Stewardship Program data and adjusted for the population of WA. If we use the estimate that 56 – 90% of carpets are pretreated and contain approximately 15% PFAS by weight, there is the potential for 1,290 – 2,074 metric tons of PFAS to be found in carpets sold annually in WA.

We estimate that carpet contributes 5,376 to 8,640 metric tons of PFAS to WA landfills and 7 – 11 metric tons of PFAS to the environment through illegal dumping each year. Ecology's draft chemical action plan[20] on PFAS estimated that over 64,000 metric tons of carpet end up in WA landfills annually (Ecology, 2019). If 56 – 90% of carpets disposed of in WA are treated with 15% PFAS by weight (KEMI, 2015), we estimate that carpets contribute 5,376 – 8,640 metric tons of PFAS to WA landfills each year. Additionally, in 2004, Ecology estimated that 84 metric tons of carpet were illegally dumped on WA roadways (Ecology, 2005), which could contain 7 – 11 metric tons of PFAS.

Potential for exposure to sensitive populations

Nearly all people, including women of childbearing age, infants and young children, are exposed to PFAS. The National Health and Nutrition Examination Survey routinely detects PFOA, PFOS, PFNA and PFHxS in blood serum of nearly all participants (CDC-NHANES, 2015, 2017). PFBS and PFHpA are less commonly detected in serum, likely due to their relatively quick excretion rates in people (Kubwabo, Kosarac, & Lalonde, 2013), but can be found in indoor dust (de la Torre, Navarro, Sanz, & de los Angeles-Martinez, 2019; Karaskova et al., 2016; Strynar & Lindstrom, 2008). Serum levels of some PFAS have declined in the U.S. general population due to the phase out of the use of PFOA- and PFOS-related compounds in U.S. production (CDC-NHANES, 2017). Similar results and trends were reported in a study of 610 American Red Cross blood donors from 2000 through 2015 (Olsen et al., 2017). PFPeA, PFHxA, PFHpA, and PFBS were commonly detected in breast milk among Korean women (Kang et al., 2016) and PAPS (e.g., 4:2 diPAP, 6:2 diPAP, 8:2 diPAP, and 10:2 diPAP) have been detected in Canadian breast milk (Kubwabo et al., 2013). More information about exposure can be found in the PFAS chemical action plan Health Appendix (Ecology, 2019).

People, including infants and young children, are exposed to PFAS from treated carpets. As carpet degrades, PFAS can be released into indoor air and accumulate in dust. People inhale and ingest contaminated air and dust, leading to human exposure to PFAS. Homes and offices with carpet can have higher concentrations of various PFAS in the indoor environment (Fraser et al., 2013; Gewurtz et al., 2009; Kubwabo, Stewart, Zhu, & Marro, 2005). Karaskova et al. (2016) found that the concentrations of the sum of 20 PFAS on carpeted floors was higher than other floor types (Karasova et al., 2016).

Because children spend more time on or near the floor and have relatively high respiration rates and frequent hand-to-mouth activity, they are exposed to more contaminated air, carpet and house dust. Karaskova et al. (2016), Tian et al. (2016), Shoeib et al. (2011) and Trudel et al. (2008) have found that house dust is an important PFAS exposure route for toddlers. Washburn et al. (2005) estimated that the reasonable maximum exposure scenario for PFOS in carpet was two orders of magnitude higher for infants than adults, meaning infants could be exposed to

[20] https://fortress.wa.gov/ecy/publications/documents/1804002.pdf

PFAS at a level that is about 100 times higher than adults. Studies show that children with carpets in their bedrooms have higher concentrations of PFOS, PFHxS, and Me-PFOSA-AcOH in their bodies than children with other types of bedroom flooring (Harris et al., 2017). Boronow et al. (2019) found that women living in homes with treated carpet had higher exposure to PFAS, PFNA and PFDeA. Fraser et al. (2012) found that office workers in buildings with higher concentrations of FTOH in the air had higher concentrations of PFOA in their serum.

Treated carpet makes a significant contribution to exposure. Based on a survey of the concentration of PFAS in 116 consumer articles, EPA concluded that carpet and carpet care liquids were likely significant sources of exposures to PFCAs (EPA, 2009). Trudel et al. (2008) found that treated carpet was the most prominent consumer product exposure pathway, and estimated that between 5 – 64% of PFOS exposure was related to contact with treated carpet.

Potential environmental exposure and exposure to sensitive species

PFAS are contaminants in Washington's environment. In Washington, PFAAs have been detected in drinking water, surface waters, groundwater, wastewater effluent, freshwater, sediments, and wildlife. PFAAs have also been identified in drinking water in Issaquah and in and around three military bases: Joint Base Lewis-McChord, Naval Air Station Whidbey Island, and Fairchild Air Force Base. Environmental monitoring in the state has shown that PFAA concentrations are highest in urban surface water and surface waters receiving minimally diluted WWTP effluent (Ecology, 2016, 2019). PFAAs and PFOSA have also been found in Washington wildlife, such as freshwater fish and osprey eggs (Ecology, 2016, 2019).

PFAS from carpet have the potential to contribute to environmental PFAS concentrations. PFAS from carpet are also released into the environment through disposal of carpet in landfills (Lang, Allred, Peaslee, Field, & Barlaz, 2016). Based on the volume estimates described previously, carpet may contribute up to 8,640 metric tons of PFAS to WA landfills and 11 metric tons of PFAS to the environment through illegal dumping each year.

The persistence of PFAS in the environment heightens concerns about disposal and release. PFAS can be released from landfills into air, groundwater and as leachate (Hamid & Grace, 2018) and PFAS from illegally dumped carpet are released directly into the environment. PFAS from indoor carpet can accumulate in house dust (Gewurtz et al., 2009; Knobeloch et al., 2012; Kubwabo et al., 2005) and can be tracked into the outdoor environment (Wild et al., 2015) or released into wastewater through the laundering process (Shoeib, Harner, Webster, & Lee, 2011).

Exposure to PFAS in the environment is a concern for sensitive species. PFAS are environmentally persistent and some are bioaccumulative (Ecology, 2019). Two PFAS, PDFA and

PFOS, have been detected in salmon in Puget Sound (Meador, Yeh, Young, & Gallagher, 2016). Governor Inslee's Orca Task Force identified PFAS as a chemical of emerging concern.

Existing regulations

PFAS are not currently regulated under established environmental laws including the Resource Conservation and Recovery Act (RCRA), the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), the Clean Water Act (CWA), or the Clean Air Act (CAA) (ITRC, 2019). Several bills are active in Congress to address PFAS, including directing federal agencies to regulate PFAS under the aforementioned laws, but none have been enacted into law (GovTrack, 2019).

In the absence of federal action, states have begun setting drinking water levels and clean up levels to limit exposures to PFAS chemicals (ITRC, 2019). There are 22 adopted laws in 13 states and 63 pending bills in 18 states pertaining to limits, bans, monitoring, or reporting of PFAS chemicals. Of the 22 adopted laws, 11 directly ban PFAS-containing products (Safer States, 2019). California Department of Toxic Substances Control (DTSC) has proposed PFAS in carpets as a priority product under their Safer Consumer Products program. This law asks manufacturers to consider alternatives to chemicals of concern in their products and may lead to regulation (DTSC, 2019).

PFAS may be considered Washington state dangerous waste as halogenated organic compounds (173-303 WAC). Halogenated organic compounds are dangerous waste when present at or above 100 ppm in a waste stream (code WP02) and extremely hazardous waste when present above 1% in a waste stream (code WP01).

Existing recommendations: PFAS in carpet were identified as a priority in our draft chemical action plan on PFAS.

Availability of safer alternatives

There are a number of ways to meet the function of stain and dirt resistance in carpet. This can be achieved by using PFAS chemistries, non-PFAS "drop in" alternatives, as well as fibers that are inherently stain resistant. Non-PFAS "drop in" solutions include siloxane polymers, polyurethanes, sulfonation, and silicate clay-based repellent. Inherently stain resistant fibers include wool, polypropylene, polyethylene terephthalate, and polytrimethylene terephthalate. Several alternatives have been investigated by other agencies, such as DTSC (DTSC, 2018). Manufacturers such as Interface have stopped production of PFAS-containing carpets (Interface, 2019), and vendors such as Home Depot and Lowe's have phased out sale of PFAS-containing carpets (Home Depot, 2019). As part of our Safer Products for Washington process, we will address the safety and feasibility of these alternatives in phase 3.

Aftermarket Carpet Treatments

Overview

Draft priority product

Aftermarket stain and water resistance treatments for carpets and rugs used in residential and commercial settings

Priority chemical

Perfluoroalkyl and polyfluoroalkyl substances (PFAS)

Priority product summary

Aftermarket treatments for stain and water resistance for carpets and rugs used in residential and commercial settings are significant sources and uses of PFAS. Ecology made this determination by considering the criteria in RCW 70.365.030[21] of Safer Products for Washington. We are concerned about the contribution aftermarket carpet treatments make to the amounts of PFAS in our homes, workplaces and environment, as well as the potential for exposure for infants, young children and workers. In 2009, EPA estimated that carpet and carpet care products were the second largest use of some PFAS, with the broadly defined "textiles" category ranking first.

We estimate that 2,300 metric tons of aftermarket treatments are used and disposed of each year in Washington. People can be exposed to PFAS while applying aftermarket treatments and as the product wears off the carpet over time. Since children and babies spend more time on or near the floor, they are disproportionately exposed to PFAS in dust. Epidemiological studies show that children living in homes with treated carpet have higher exposures to PFAS.

The PFAS from aftermarket treatments can also be released into the environment. Spills and improper disposal of aftermarket treatment can release PFAS directly into environmental media. PFAS are environmentally persistent, making clean-up difficult or impossible in some cases. In 2018, Governor Inslee convened the Southern Orca Task Force to recommend actions to recover the southern orca population. The task force named PFAS an emerging concern for orcas. Detailed support for our listing of PFAS in carpet as a priority product is shown below.

Background

PFAS have been used in aftermarket carpet treatments since the 1950s. They are sprayed on carpet in homes and workplaces by the general public and professionals to prevent staining and reduce wear and tear. The specific PFAS used in carpet treatment changed over time. From

[21] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.030

1970 to 2002, the largest use of PFOS-derived substances was for carpet applications (48,000 tons globally from 1970 to 2002) (DEPA, 2013; Paul, Jones, & Sweetman, 2009).

From the 1950s to the early 2000s, PFOS-based fluorochemicals were used in carpet treatments, and in 2003, manufacturers moved to PFBS-based chemistries. According to safety data sheets, carpet treatments contain fluorochemicals at concentrations between 1 and 5%.

There is no publicly available information on what exact fluorochemicals manufacturers use. However, Kothoff et al. (2015), EPA (2009b) and Herzke (2012) detected PFCAs and FTOHs in impregnation and nanosprays, including carpet treatments (Table 6). Fluorotelomer-based side-chain fluorinated polymers degrade into FTOHs and PFCAs in the environment (Washington & Jenkins, 2015).

Estimated volume of PFAS used in aftermarket treatments

Multiple PFAS have been detected in aftermarket treatments at concentrations that can make significant contributions to the volume of PFAS in our homes. Example concentrations of PFAS in aftermarket treatments are shown in Table 6. Concentrations of PFAS in carpet following aftermarket treatment are shown in Table 7. EPA (2009b) estimated that there are about 1 kg of household carpet care products in the home. One kg of residential carpet care treatments could contribute 953 – 14,945 µg PFCAs and 185,730 – 232,387 µg of FTOHs in the home, depending on the concentrations shown in Table 6.

Higher concentrations of PFAS have been found in commercial carpet care products (12,000 µg/kg Total PFCA (C5-12)), relative to household carpet care products (EPA, 2009b). EPA (2009b) estimated that 6 kg of commercial carpet care products are used during one treatment of a home, suggesting that commercial aftermarket treatments may contribute 72,000 µg PFCAs to the home per treatment. Manufacturers recommend reapplying these products to carpet every two years.

A case study from 2012 reported high concentrations of PFAS in carpet in a household that regularly used aftermarket carpet treatments (Table 7, Beeson, Genuis, Benskin, & Martin, 2012). Based on the residual concentrations of PFAS in carpet observed in Beesoon et al. (2012) and EPA's (2009a) estimate of 50 kg carpet in the average home, aftermarket treatments may contribute approximately 8 g PFOA, 59 g PFOS, and 144 g PFHxS to the average home, if reapplied every two years.

Table 6. Reported concentrations of PFAS in aftermarket carpet treatments and related products.

Ecology estimated the volume in Washington based on the concentration and the product volume estimation described below.

Product Description	Concentration	PFAS Measured	Estimated volume of PFAS in WA per year	Reference
Water proofing agents	29,889 µg/L	Total PFCA	69 Kg	Herzke, Olsson, & Posner, 2015
Household carpet/fabric-care liquids and foams	953 µg/kg	Total PFCA (C5 – C12)	2.2 Kg	EPA, 2009b
Water proofing agents	464,774 µg/L	FTOH/FTS	1076 Kg	Herzke et al., 2012
Impregnating sprays (waterproofing)	1,857,300 µg/kg	FTOH/FTS	4307 Kg	Kotthoff, Muller, Jurling, Schlummer, & Fiedler, 2015

Table 7. Concentrations of PFAS in carpet following aftermarket treatment.

Product Description	Concentration	PFAS Measured	Reference
Carpet	112 – 1170 µg/kg	PFOS	Beesoon et al., 2012
Carpet	8 – 153 µg/kg	PFOA	Beesoon et al., 2012
Carpet	12 – 2880 µg/kg	PFHxS	Beesoon et al., 2012

Estimated volume used in Washington

We estimate that up to 2,300 metric tons of aftermarket carpet treatments are used and disposed of in WA each year, potentially contributing 69 kg PFCA and 4307 kg FTOH/FTS to WA's homes and environment. We based our estimate on two approximations. First, there are 15,360 metric tons of carpet sold yearly in WA (17 million square yards—based on California Carpet Stewardship Program, adjusted for the population of WA). Second, there is an aftermarket treatment rate of 1500sq ft/gallon every two years after five years of carpet use. We assume carpet care liquids have the same density as water. It is unclear what percentage of the carpet in WA is retreated every two years. Our estimations follow manufacturers' recommendations.

Potential for exposure to sensitive populations

Nearly all people, including infants, children, men and women of childbearing age, and workers, are exposed to PFAS. The National Health and Nutrition Examination Survey routinely detects PFOA, PFOS, PFNA and PFHxS in blood serum of nearly all participants (CDC-NHANES, 2015, 2017). PFBS and PFHpA are less commonly detected in serum, likely due to their relatively quick excretion rates in people (Kubwabo, Kosara, & Lalonde, 2013), but can be found in indoor dust (de la Torre, Navarro, Sanz, & de los Angeles-Martinez, 2019; Karaskova et al., 2016; Strynar & Lindstrom, 2008).

Serum levels of some PFAS have declined in the U.S. general population due to the phase out of the use of PFOA- and PFOS-related compounds in U.S. production (CDC-NHANES, 2017). Similar results were reported in a study of 610 American Red Cross blood donors from 2000 through 2015 (Olsen et al., 2017). PFPeA, PFHxA, PFHpA, and PFBS have been commonly detected in breast milk among Korean women (Kang et al., 2016) and PAPS (e.g., 4:2 diPAP, 6:2 diPAP, 8:2 diPAP, and 10:2 diPAP) have been detected in Canadian breast milk (Kubwabo et al., 2013). More information about exposure can be found in the PFAS chemical action plan[22] Health Appendix (Ecology, 2019).

People, including infants, children, men and women of childbearing age, and workers are exposed to PFAS from aftermarket carpet treatments. People can be exposed to PFAS from aftermarket carpet treatments during application, before the product dries, and as it degrades over time. During application, dermal contact and inhalation may occur. The exposure pathway during product degradation is similar for aftermarket treatment and pretreated carpet.

However, PFAS in pretreated carpet are polymerized whereas aftermarket treatments are applied as liquid sprays, making them more likely to migrate from the carpet into dust and air and increasing the need for reapplication. As carpet treatment wears off, PFAS can be released into indoor air and accumulate in dust. Beesoon et al. (2012) reported high serum concentrations of PFHxS, PFOS and PFOA in a family that had their carpet commercially treated approximately every two years for 15 years. PFAS were also detected in their carpet, house dust and indoor air. Homes and offices with carpet can have higher concentrations of various PFAS compared to non-carpeted facilities (Fraser et al. 2013; Gewurtz et al., 2009; Kubwabo, Stewart, Zhu, & Marro, 2005). Karaskova et al. (2016) found that the concentrations of the sum of 20 PFAS on carpeted floors was higher than other floor types.

Because children spend more time on or near the floor and have relatively high respiration rates and increased hand-to-mouth activity, they are exposed to more contaminated air, carpet and house dust. Karaskova et al. (2016), Tian et al. (2016), Shoeib et al. (2011), and Trudel et al. (2008) have found that house dust is an important PFAS exposure route for toddlers. Washburn et al. (2005) estimated that the reasonable maximum exposure scenario for PFOS in carpet was

[22] https://fortress.wa.gov/ecy/publications/documents/1804002.pdf

two orders of magnitude higher for infants than adults, meaning infants could be exposed to PFAS at a level that is approximately 100 times higher than adults. Children with carpets in their bedrooms have been found to have higher concentrations of PFOS, PFHxS, and Me-PFOSA-AcOH in their bodies than those with other flooring types (Harris et al., 2017). Boronow et al. (2019) found that women living in homes with treated carpet or upholstery had higher exposure to PFAS, PFNA and PFDeA. Fraser et al. (2012) found that office workers in buildings with higher concentrations of FTOH in the air had higher concentrations of PFOA in their serum.

Treated carpet makes a significant contribution to exposure. Based on a survey of the concentration of PFAS in 116 consumer articles, EPA concluded that carpet and carpet care liquids were likely the most significant sources of exposures to PFCAs out of the 13 consumer product categories studied (EPA, 2009b). Trudel et al. (2008) found high levels of PFOA and PFOS in carpet, and determined that treated carpet was the most prominent exposure pathway from consumer products. They estimated that between 5 and 64% of PFOS exposure was related to contact with treated carpet, and concluded that exposure to these chemicals is high for infants, toddlers and children due to their close contact to carpets (Trudel et al., 2008).

Potential environmental exposure and exposure to sensitive species

PFAS have contaminated Washington's environment. In Washington, PFAAs have been detected in drinking water, surface waters, groundwater, wastewater effluent, freshwater, sediments, and wildlife. PFAAs have also been identified in drinking water in Issaquah and in and around three military bases: Joint Base Lewis-McChord, Naval Air Station Whidbey Island, and Fairchild Air Force Base. Environmental monitoring in the state has shown that PFAA concentrations are highest in urban surface water and surface waters receiving minimally diluted WWTP effluent (Ecology, 2016, 2019). PFAAs and PFOSA have also been found in Washington wildlife, such as freshwater fish and osprey eggs (Ecology, 2016, 2019).

PFAS from aftermarket carpet treatments have the potential to contribute to environmental PFAS concentrations. PFAS in aftermarket treatments can be released into the environment if the product spills or leftovers are improperly disposed. In the volume estimation section of this chapter, we estimate that up to 2,300 metric tons of aftermarket carpet treatment are used each year. If treatments only last two years, we would anticipate that over two years this volume would be released from the carpet into our homes, workplaces and ultimately the environment.

When disposed in landfills, treated carpet can contribute to environmental release as the treatment wears off (Lang, Allred, Peaslee, Field, & Barlaz, 2016). PFAS can be released from landfills into air, groundwater, and as leachate (Hamid & Grace, 2018). The volume of carpet and potential PFAS releases from its disposal are further discussed above, in the chapter for pretreated carpet.

Stain and water resistance for mill-treated carpets and rugs lasts approximately five years. Aftermarket treatments do not last as long, and manufacturers recommend retreatment every two years. This recommendation suggests that PFAS in aftermarket treatments may migrate from the carpet more quickly than PFAS in mill-treated carpets. PFAS from treatment applied to carpet can accumulate in house dust (Beesoon et al., 2008; Kubwabo et al., 2005), and can be tracked into the outdoor environment (Wild et al., 2015) or released into wastewater through the laundering process (Shoeib et al., 2011).

Exposure to PFAS in the environment is a concern for sensitive species. PFAS are environmentally persistent and some are bioaccumulative (Ecology, 2019). Environmental monitoring has detected two PFAS, PDFA and PFOS, in salmon in Puget Sound (Meador, Yeh, Young, & Gallagher, 2016). In March of 2018, Governor Inslee established the Orca Task Force, a 50-member work group charged with developing plans for recovering Puget Sound's struggling southern resident orca population. The task force identified PFAS as a chemical of emerging concern.

Existing regulations

Established environmental laws, including the Resource Conservation and Recovery Act (RCRA), the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), the Clean Water Act (CWA), and the Clean Air Act (CAA), do not currently regulate PFAS (ITRC, 2019). Several bills are active in Congress to address PFAS, including directing federal agencies to regulate PFAS under the aforementioned laws, but none have been enacted into law (GovTrack, 2019).

In the absence of federal action, states have begun setting drinking water levels and clean up levels to limit exposures to PFAS chemicals (ITRC, 2019). There are 22 adopted laws in 13 states and 63 pending bills in 18 states pertaining to limits, bans, monitoring, or reporting of PFAS chemicals. Of the 22 adopted laws, 11 directly ban PFAS-containing products (Safer States, 2019). California Department of Toxic Substances Control (DTSC) has proposed PFAS in carpets as a priority product under their Safer Consumer Products program. This law asks manufacturers to consider alternatives to chemicals of concern in their products and may lead to regulation (DTSC, 2019).

PFAS may be considered Washington state dangerous waste as halogenated organic compounds (WAC 173-303-100(6)). Halogenated organic compounds are dangerous waste when present at or above 100 ppm in a waste stream (code WP02), and extremely hazardous waste when present above 1% (10,000 ppm) in a waste stream (code WP01).

Existing recommendations: Ecology identified PFAS in aftermarket carpet treatments as a priority in our draft chemical action plan[23] on PFAS (Ecology, 2019).

Availability of safer alternatives

There are a number ways to meet the function of stain and dirt resistance in aftermarket carpet treatment. Manufacturers achieve this using PFAS chemistries, non-PFAS drop-in alternatives, and by choosing alternative carpet materials that do not require aftermarket stain treatment, due to the materials' natural stain resistance. Non-PFAS chemical solutions on the market include silicone dioxide (ProtectME, 2019), and proprietary anionic non-fluorinated polymers (Bridgepoint Systems, 2019; Tri-Plex Technical Services, Ltd., 2019).

Inherently stain resistant fibers include wool, polypropylene, polyethylene terephthalate, and polytrimethylene terephthalate. Other agencies, including DTSC, have investigated several alternatives (DTSC, 2018). Manufacturers such as Interface have stopped production of PFAS-containing carpets (Interface, 2019), and vendors including Home Depot and Lowe's have phased out sale of PFAS-containing carpets (Home Depot, 2019, Lowes 2019). As part of our Safer Products for Washington process, we will address the safety and feasibility of these alternatives in phase 3.

[23] https://fortress.wa.gov/ecy/publications/documents/1804002.pdf

Food and Drink Cans

Overview

Draft priority product

Food and drink cans

Priority chemical

Phenolic Compounds—Bisphenols

Priority product summary

Can linings are a significant source and use of phenolic compounds. Ecology made this determination after considering the criteria in RCW 70.365.030[24] of Safer Products for Washington. We are concerned about the contribution metal can linings make to bisphenol concentrations in humans and the environment. Approximately 2.5 billion cans are sold each year in Washington, and testing shows that a large proportion of those may contain bisphenol-based can liners.

Dietary exposure to Bisphenol A (BPA) is the largest source of exposure to this chemical, and consuming canned food leads to higher biological levels. This is particularly concerning for children and infants, who have the largest exposure to BPA, and who may be more affected by these chemicals due to their sensitive stage in development. Bisphenols from can linings may also enter the environment. Governor Inslee's Orca Task Force has declared them a contaminant of emerging concern for our Puget Sound orca population. Detailed support for our listing of bisphenols in can linings as a priority product is shown below.

Background

Bisphenols, and specifically BPA, are commonly used in epoxy resin-based can linings. These linings prevent reaction of the food or beverage with the metal can, maintaining food taste and structural integrity of the can.

Resins containing BPA are formed by reacting epichlorohydrin (ECH) with BPA, to create bisphenol A diglycidyl ether (BADGE). These BADGE monomers then react with BPA monomers to create a polymer (Figure 3). Epoxies become more solid the higher number of monomers that are contained in the polymer. Manufacturers may also use Bisphenol F (BPF) in place of BPA to create the resin. BPA-based resin will be composed of approximately 85% BPA by weight, and the polymer can also contain unreacted BPA monomer.

[24] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.030

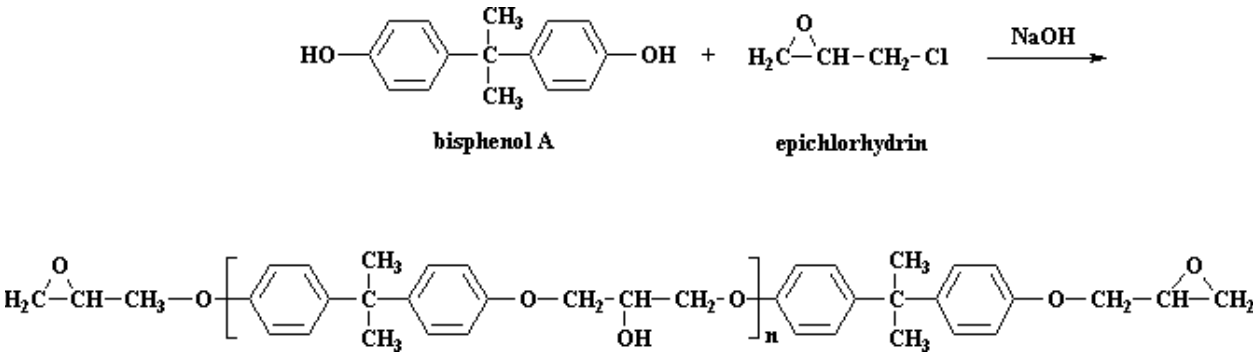

Figure 3. The creation of BPA based epoxy resin from BPA and epichlorhydrin (PSLC, 2019).

Resins may also contain curing agents, which help improve mechanical, chemical, and heat-resistance properties. The addition of heat or chemicals called accelerators may improve the curing process. Other sealants and polymers may also be used to line cans, and the chemicals used depend on what is contained in the can.

Estimated volume of Bisphenols used in can linings

Bisphenols are widely present in can linings and canned food. When studying can linings, researchers often analyze the can contents in order to identify the components of the lining as well as the leaching rate of the lining into its contents. Thus, bisphenol levels in food prior to canning may contribute to testing results, and could provide false positives as to the contents of can linings. However, un-canned food has consistently low levels of BPA—mostly under the limit of detection—suggesting that false positives would be rare (Cao et al., 2011).

Below is a summary of studies investigating bisphenol use in canned food (Table 8). Only two studies were identified that directly investigate can linings for presence of bisphenols. In 2013, EU migration limits for BPA in food contact material were lowered from 600 ng/g to 50 ng/g, so while many of the values below are above the current requirement, they were in compliance when sampled. These results show widespread and well-documented use of bisphenol-based can linings, and that these linings may contaminate the contents of the cans.

Table 8. Summary of studies investigating bisphenol prevalence and concentration in canned food and beverages.

Year	Chemical	Prevalence	Level	Reference
2004	BPA	25/79 (32%)	11 – 191 ng/g in food	Thomson & Grounds, 2005
2006	BPA	55/97 (57%)	Not Disclosed	Environmental Working Group, 2007
2009	BPA	69/69 (100%)	0.032 – 4.5 µg/L in soft drink	Cao, Corriveau, & Popovic, 2009

Year	Chemical	Prevalence	Level	Reference
2010	BPA	19/22 (86%)	0.32 – 106 ng/g in food	Cao et al., 2011
2010	BPA	17/17 (100%)	0.019 – 0.54 µg/L (ppb) in soft drinks and beer	Bureau of Chemical Safety, 2010
2010	BPA	63/105 (60%)	0.23 – 65.0 ng/g in food	Haffner, Smith, Harris, Paepke, & Birnbaum, 2010
2010	BPA	46/50 (92%)	0.7 – 1140 ng/g in food and drink	Chase Wilding et al., 2010
2011	BPA	71/78 (91%)	2.6 – 730 ng/g in food	Noonan, Ackerman, & Begley, 2011
2013	BPA	27/31 (87%)	0.01 – 146 ng/g in food	Liao & Kannan, 2013
2013	BPAF, BPAP, BPB, BPF, BPP, BPS, BPZ	10.5%, 11.2%, 2.62%, 10.1%, 3.37%, 20.9%, 2.25% (food—not all canned)	All with 95th percentile under 0.74 ng/g, suggesting inadvertent contamination. Max of 1130 ng/g BPF found in mustard (which has natural BPF).	Liao & Kannan, 2013
2015	BPA	45% of brands	45% of brands surveyed used BPA-lined cans for some or all of their products	Lunder & Geller, 2015
2015	BPA	26/37 (70%)	0.24 – 149.0 ng/g in food	Lorber, Schecter, Shropshire, & Christensen, 2015
2017	BPA	96/252 (32%) of linings	25 – 140 ng/g in food (4 samples)	Cox, 2017

An article published in 2015 stated that "according to coatings specialists, roughly 80%" of epoxy coatings used in can linings are BPA based (Waldman, 2015). This is supported by a statement from the North American Metal Packaging Alliance which estimated that "75% of canned foods sold in the U.S. are lined with a BPA-based polymer" (Environmental Working Group, 2015). Another article published in 2018 quoted an internal Can Manufacturers Institute survey which found that at least 90% of can linings do not use BPA based linings, would could suggest that the industry may be phasing out BPA use (McTigue Pierce, 2018). However, testing data shows that any phase-out of BPA in can linings is incomplete, and other bisphenols besides BPA have not been extensively analyzed, leaving open the possibility of their use, either currently or as a substitute in the future.

Estimated volume used in Washington

Approximately 2.5 billion cans are sold each year in Washington. As shown above, testing results, and even statements from industry vary widely as to the percentage of cans containing bisphenol-based can liners. According to the 2018 – 2019 Can Manufacturers Institute annual report, about 125 billion cans are produced per year in the U.S. and Canada (CMI, 2019). Taking Washington state's population proportion of that would give 2.5 billion cans sold per year in Washington. Depending on which industry group's statement you use (North American Metal Packaging Alliance or Can Manufacturers Institute), BPA-lined cans could consist of as low as 10% or as much as 75% of this figure. This would amount to between 250 million and 1.9 billion BPA-lined cans sold in Washington per year, or 34 to 253 BPA-lined cans per person. (This estimate does not include the alternate bisphenols besides BPA.)

Potential for exposure to sensitive populations when used

Nearly all people are exposed to bisphenols. Findings in the Fourth National Report on Human Exposure to Environmental Chemicals indicate widespread exposure to BPA. CDC scientists found BPA in more than 90% of the urine samples representative of the U.S. population. NHANES data from 2003 to 2014 show a decreasing trend in the urinary BPA concentration for the general U.S. population (CDC, 2019). Because BPA does not persist for long periods of time in the body, its widespread detection in people indicates that exposures occur frequently. In 2011, the World Health Organization (WHO) estimated the mean dietary daily intake of BPA for adults to be 0.4 – 1.4 µg per kg of body weight (WHO, 2011).

The levels of BPA in humans have changed over time, likely due to their replacement in products. Besides BPA, other bisphenols that may be used in can linings, such as BPS and BPF, have also been detected in the urine of the U.S. general population, according to NHANES 2013 – 2014 survey. Exposure to other bisphenols (e.g., BPS) appears to have increased in the U.S. population due to substitution (Ye et al., 2015). However, their levels appear to be lower compared to BPA, and the detection frequency is higher for BPA than other bisphenols (95.7%, 89.4%, and 66.5% for BPA, BPS, and BPF respectively) (Lehmler, Liu, Gadogbe, & Bao, 2018).

Due to inaccuracies in historical testing methods, most BPA levels in organisms are likely severely underreported. One study found levels in urine 19 times higher using direct analysis versus the indirect analysis used by federal agencies and in the NHANES data set (Gerona, vom Saal, & Hunt, 2019). This has potentially affected the result of risk calculations by suggesting that population exposure levels are much lower than they are in reality.

There is the potential for people to be exposed to bisphenols used in can linings. General population exposure to BPA may occur through the ingestion of foods in contact with BPA-containing materials. According to the FDA, when foods are in direct contact with any packaging

material, small but measurable amounts of the packaging materials can migrate into food and can be consumed with it (FDA, 2014)

The FDA is working with the National Toxicology Program (NTP) to answer questions about BPA migration into food from food contact materials. According to a recent study, migration from packaging into beverages and drinking water is as significant as migration to solid food (Russo et al., 2019). Water has been shown to absorb BPA from liners during heating. For example, low levels of BPA were detected in water from all unheated cans, rising from 0.06 to 32 ng/cm^2 after heating at 100°C (Takao, Lee, Kohra, & Arizono, 2002). Another study found BPA from lined cans leaches into water held at 121°C to a much higher extent than water held at 80°C (Sajiki et al., 2007).

Some studies show elevated migration levels of BPA from canned foods. A significant increase in levels of BPA was observed in tomatoes produced in Italy during canning. This testing also showed that the heating process during canning or damage by denting may increase BPA migration levels (Errico et al., 2014). Most migration of BPA (80 – 100%) seems to occur during the can processing step. Storage (up to 9 months) did not alter the levels of BPA migration (Goodson, Robin, Summerfield, & Cooper, 2004).

Can linings make a significant contribution to exposure to bisphenols. The European Food Safety Authority (EFSA) indicates that eating food and/or drinking water that contains BPA is the largest source of BPA exposure (EFSA, 2015). Consumption of canned food, including some specific types, such as canned vegetables and fruit, canned pasta, and canned soup, is correlated with higher urinary BPA concentrations (Hartle, Navas-Acien, & Lawrence, 2016). In Europe, the consumption of canned meat and vegetables has been identified as the largest source of dietary intake of BPA (Russo, Barbato, Mita, & Grumetto, 2019).

Carwile (2011) characterized exposure and biological processing of dietary BPA in a group of healthy adult humans. The study found that on a controlled diet, canned food items are likely to be significant dietary sources of BPA. Consumption of one serving of canned soup daily over 5 days was associated with a more than 1000% increase in urinary BPA over soup prepared without canned ingredients (Carwile, 2011). In addition, a study found that urinary BPA concentrations were more than 16 times as high after drinking canned beverages versus drinking glass-bottled beverages (Bae & Hong, 2015). The study also showed that exposure to BPA from drinking canned beverages increased blood pressure (Bae & Hong, 2015).

Exposure to bisphenols in can linings is particularly relevant for infants, children, and certain demographic groups. Children and infants are exposed to bisphenols at a time when their development is especially susceptible. EFSA scientists found dietary exposure to BPA is highest among children aged six months to ten years (explained by their higher food consumption on a body weight basis), with estimated BPA dietary intake of up to 0.875 µg per kg of body weight per day (EFSA, 2013, 2015). Concentration of BPA in breastmilk, a possible exposure route to infants, has been correlated with drinking canned beverages (Tateoka, 2015). Women of

childbearing age had dietary exposures comparable to men of the same age (up to 0.388 µg per kg of body weight per day) (EFSA, 2015).

Certain demographic groups may eat more canned food, and thus have higher potential exposure to bisphenols. USDA calculated that in 2004, African Americans spent more per person on canned vegetables than any other ethnicity. In addition, it was found that people from the southern U.S. spend more on canned vegetables than other regions, while people from the Midwest spend the most on canned fruit. The elderly (over 64) spent the most of any age group (USDA, 2008). Those who receive food assistance through the Supplemental Nutrition Assistance Program or Women, Infant and Children program consume an average of 1.3 times as much canned food as the general population (Laatsz, 2012).

Potential environmental exposure and exposure to sensitive species

Bisphenols have been found in Washington's environment. Bisphenols are present throughout Washington in various environmental media. The Lower Columbia River Estuary Partnership found BPA in the Columbia river in 2004 (LCREP, 2007). As part of the Puget Sound Toxics Loading Study,[25] Ecology found BPA in WWTP biosolids (Ecology, 2010a), in 53.3% of effluent (Ecology, 2010b), and in stormwater, with higher levels during rain events (Ecology, 2011). BPA was found in Puget Sound at much higher levels than in the relatively uninhabited Barkley Sound, BC, indicating human contributions to environmental concentrations (Keil, Salemme, Forrest, Neibauer, & Logsdon, 2011).

Outside of Washington, BPA is frequently detected, sometimes at concentrations that exceed levels defined as protective of human and environmental health by governmental agencies (Corrales et al., 2015). Other bisphenols are also commonly found in environmental media, such as sediment, water, soil, dust, and throughout the WWTP process (Chen et al., 2016; Hu, Zhu, Yan, Liao, & Jiang, 2019).

Bisphenols from can linings have the potential to contribute to environmental bisphenol concentrations. A 2008 risk assessment by the EU found that epoxy resin production is one of the largest sources of BPA release to bodies of water (Aschberger et al., 2008). In addition, during the recycling process, cans will be thoroughly washed and water likely released to WWTPs or the environment. Although bisphenols are often removed at greater than 90% efficiency by WWTPs, due to the large volumes of wastewater discharged by recycling facilities, this is still a major source of environmental release (PPRC, 2015).

Sewage sludge has some of the highest environmental levels of BPA detected (Michałowicz, 2014). Discarded cans also have the potential to contribute bisphenols to the environment.

[25] https://fortress.wa.gov/ecy/publications/documents/1103010.pdf

High levels of BPA have been detected in landfill leachate, and groundwater has been contaminated with BPA near waste dumps (Michałowicz, 2014).

Exposure to bisphenols in the environment is a concern for sensitive species. Bisphenols can have a variety of detrimental effects on wildlife (Cioci, Apfelbacher, Strong, & Innes, 2015). There is evidence that BPA is slightly bioaccumulative in some species (Corrales et al., 2015). BPA has been shown to cause embryonic deformities, abnormal behavior, growth inhibition in fish, and reduce the number of offspring in fish (Kang, Aasi, & Katayama, 2007). A wide variety of bisphenols have shown endocrine disrupting, cytotoxic, genotoxic, reprotoxic, and neurotoxic effects (Chen et al., 2016). Growth effects have been shown in fish exposed to water with concentrations as low as 0.078 µg/L (Kang, Aasi, & Katayama, 2007).

While BPA is the most studied bisphenol, there is evidence that other compounds have similar, if not worse, effects on wildlife. For example, BPF, BPS, BPAF, BPB, and BPC have shown estrogenic activity similar to BPA (Chen et al., 2016), and can cause stress effects at low levels in a variety of organisms (Zhou, 2018). Bisphenols were deemed a chemical of emerging concern for the endangered Puget Sound orca population (Southern Resident Orca Taskforce, 2018).

Existing regulations

There are no federal regulations pertaining to bisphenols in can linings. In the U.S. there is a state-driven movement to limit exposure to BPA. There are 31 pending legislative policies in 13 states and 30 laws in 14 states pertaining to limits, bans, monitoring, or reporting of BPA in products such as children's cups and food containers. Of the 30 adopted laws, 6 laws in 6 states directly ban BPA in infant formula cans and baby/child food containers. California Department of Toxic Substances Control (DTSC) has listed BPA in food packaging as a proposed priority product under its workplan (DTSC, 2019). In Washington state, BPA is banned from children's food and beverage containers but metal cans are exempt, under RCW 70.280.020(1)[26] (Safer States, 2019).

BPA has been banned from infant feeding bottles across the EU since 2011. It is also banned in other materials that come into contact with food intended for infants and children under three years. France has banned BPA in all food packaging, containers and utensils (ECHA, 2019). In 2018, the European Commission (EC) reduced the specific migration limit (SML) of BPA from plastics, coatings and varnishes for metals and other contact sources to food from 0.6 mg/kg to 0.05 mg/kg. The reduced SML coincided with a ban on BPA in plastic bottles and packaging intended for babies and children (ECHA, 2019; PackagingLaw, 2018). The European Chemicals Agency (ECHA) has officially declared that exposure to BPA poses a serious human health concern (ECHA, 2019).

[26] https://app.leg.wa.gov/RCW/default.aspx?cite=70.280.020

In 2010, EPA proposed rulemaking under the Toxic Substances Control Act (TSCA) to identify BPA as "a substance that may present an unreasonable risk of injury to the environment on the basis of its potential for long-term adverse effects on growth, reproduction and development in aquatic species at concentrations similar to those found in the environment." This proposed rule has been stalled in the Office of Management and Budget for several years (EPA, 2010).

Availability of safer alternatives

There are a number of ways to store preserved food and beverages, including metal cans with can linings and containers made of different materials. Can linings may be comprised of bisphenols as well as alternative substances such as oleoresins, acrylics, or PET plastic. Other food packaging options include glass, lined cardboard, and PET plastic containers. Several alternatives have been investigated by other agencies, such as the National Resources Defense Council (Singla, 2016), the Berkeley Center for Green Chemistry (Berkeley, 2016), and the Food Packaging Forum (Geueke, 2016). As part of our Safer Products for Washington process, we will address the safety and feasibility of these alternatives in phase 3.

Laundry Detergent

Overview

Draft priority product

Laundry Detergent

Priority chemical

Phenolic Compounds—Alkylphenol Ethoxylates

Priority product summary

Laundry detergents are a significant source and use of phenolic compounds. Ecology made this determination after considering the criteria in RCW 70.365.030[27] of Safer Products for Washington. We are concerned about the contribution laundry detergents make to environmental concentrations of alkylphenol compounds. In 2015, California Department of Toxics Substances Control estimated that institutional cleaners—including laundry detergent—are the largest use of NPEs.

We calculate that as much as 2 million pounds of laundry detergent potentially containing APEs are used in Washington each year. Laundry detergents containing APEs are disposed of down the drain and make their way through wastewater treatment plants to bodies of water. There, the alkylphenol compounds act as toxicants and can disturb the endocrine systems of aquatic life. Alkylphenols have been identified by Governor Inslee's Southern Orca Task Force as a chemical of emerging concern. Alkylphenols and alkylphenol ethoxylates have been heavily restricted in many other countries, leading to lower concentrations in those areas. Detailed support for our listing of APEs in laundry detergent as a priority product is shown below.

Background

Alkylphenol Ethoxylates (APEs) are used as surfactants in laundry detergents to help clean clothing and linens. They consist of an alkylated phenol ring with anywhere from one to over 70 ethoxylate groups in a chain (DTSC, 2018). The most commonly found ethoxylates are nonylphenol ethoxylate (NPE) and octylphenol ethoxylate (OPE).

Manufacturers have largely phased out the production of residential laundry detergents containing APEs, but they are still present in some detergents marketed for commercial or industrial use, especially for use in hospitals, hotels, and nursing homes (on-premises laundries). Large uniform providers, such as Cintas and Aramark, have phased out the use of

[27] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.030

APEs in their detergents. The majority of APE-containing laundry detergents use NPE as the surfactant, so NPE numbers are often used in place of total APEs when considering the class.

Estimated volume of APEs in laundry detergents

Laundry detergents are likely the largest use of APEs in commerce. The percentage of commercial detergents using APEs is unclear, but a market report contracted by California Department of Toxic Substances Control (DTSC) in 2015 estimated that "institutional cleaners" (including laundry detergents and other cleaning products) was the dominant use of NPEs globally at 39% of use by volume (Figure 4). This is similar to a 2002 finding that 41% of household detergents tested contained NPEs, although residential use of NPEs in detergents has been phased out since that time (Cheng & Ding, 2002). Ecology was unable to find a large quantity of non-laundry detergent cleaning products available on the market containing APEs, suggesting this category consists mainly of laundry detergent, which is the most prevalent historical use. DTSC also estimated that cleaning products containing NPEs had concentrations ranging from 10 – 100%, and that over 25% of manufacturers had detergents containing APEs (DTSC, 2018). A search of laundry detergent Safety Data Sheets found NPE ranges from 1 – 40%.

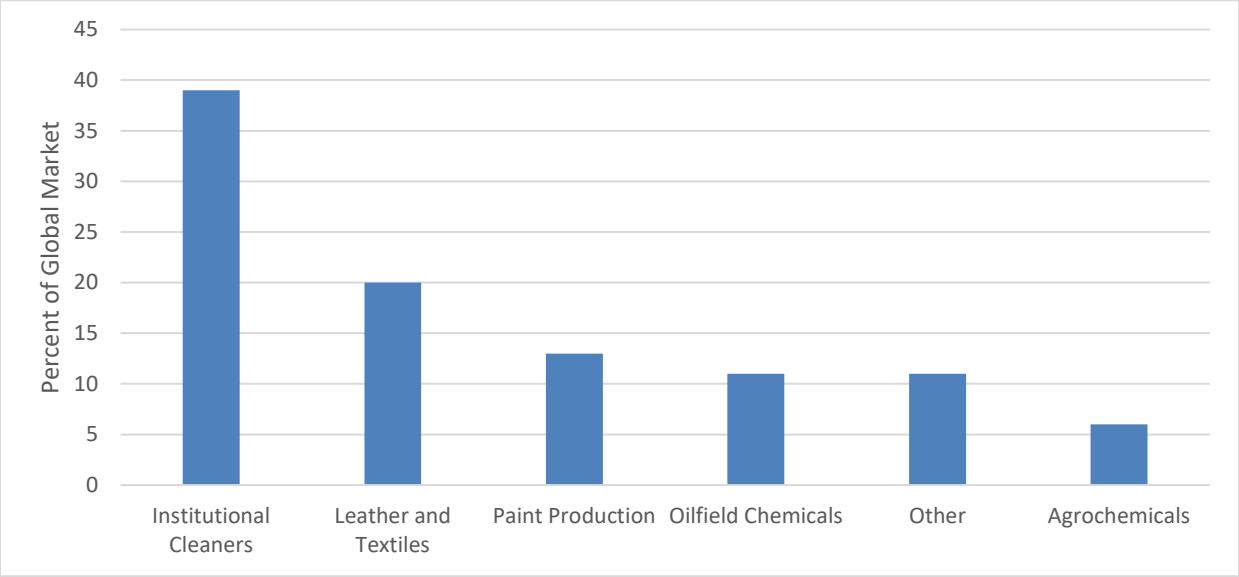

Figure 4. Market use (in %) of nonylphenol ethoxylates globally in 2015, as reported by California DTSC in 2018.

Estimated volume used in Washington

Approximately 2 million pounds of laundry detergent are used per year in Washington state. California DTSC estimated that about 2 billion pounds (lbs) of laundry are generated in on-premises launderers in California (DTSC, 2018). Using a similar methodology, but substituting

Washington state numbers for days occupied at facilities, we estimate that on-premises launderers in Washington generate about 370 million pounds of laundry (Table 9).

DTSC estimated that 8 fluid ounces of 20% NPE-containing liquid is used per 100 lbs of on-premises laundry (DTSC, 2018). If we use these estimations per 100 lbs of laundry—and make the heaviest use assumption that all on-premises launderers use NPE-containing detergent—this would mean that about 2 million pounds of laundry detergent containing 370,000 lbs of NPEs would be discharged per year by Washington on-premises laundries.

Table 9. Estimated amount of laundry generated by on-premises launderers in Washington.

Facility type	Units	Millions of units per year	Generation rate (lbs per unit)[1]	Percentage washed in On-Premises Laundrers[1]	Laundry generated (millions of lbs) per year
Hotels and motels	Occupied room nights	24.5	13.25	100%	325
Hospitals	Inpatient days	3.0	15	10%	5
Nursing facilities	Resident days	5.8	7.1	100%	41
Total					**370**

Potential for exposure to sensitive populations when used

Nearly all people, including infants and women of childbearing age, are exposed to APEs. Alkylphenols are environmental and biological metabolites of APEs. APs are more easily analyzed than APEs due to their varying ethoxylation chain lengths, and they are more persistent. Non-ethoxylated APs are used in much lower quantities than APEs in consumer products, so AP levels are often considered a surrogate of APE presence. In addition, APs have higher aquatic toxicity and endocrine-disrupting activity than APEs, so levels of APs are more relevant from a risk analysis perspective (DTSC, 2018). For these reasons, APs are more often measured in studies than APEs, and can serve as an indicator of APE exposure and hazard (DTSC, 2018).

Detection in the urine of APs indicates recent exposures. In a longitudinal study, 4-tert-octylphenol has not been detected in the urine of the U.S. general population above the 50th percentile for more than three survey cycles (2005 – 2006, 2007 – 2008, and 2009 – 2010), so it was no longer reported after 2010. In 2009 – 2010 it was only detected at the 90th and 95th percentile (CDC, 2019). 4-tert-octylphenol was also detected in the urine of adult Japanese volunteers, though the levels were near or below the detection limit (Inoue et al., 2003; Kawaguchi et al., 2004).

Nonylphenol (NP) has been detected in human breast milk (Ademollo, Ferrara, Delise, Fabietti, & Funari, 2008), showing exposure in women of childbearing age and a pathway for exposure of newborns. It has also been found in maternal blood (Guenther et al., 2002; Li et al., 2013), umbilical cord blood (Chen et al., 2008; Guenther et al, 2002; Huang et al., 2014), urine, and placenta (Calafat et al., 2005). In addition, adipose tissue collected from surgeries in Italy between 2005 and 2007 contained nonylphenol concentrations that ranged from 10 – 226 ng/g (Ferrara et al., 2011).

In 2014, some phenolic compounds, including octylphenol, were measured in the urine of healthy Danish pregnant women. The levels varied significantly among individuals, indicating that there were common exposures associated with a specific occupation or environment (Tefre de Renzy-Martin et al., 2014).

There is the potential for people to be exposed to APEs used in laundry detergent. Monitoring data indicate that the general population contacts nonylphenol via inhalation of ambient air (Rudel et al., 2003; WHO, 2004), ingestion of dust (Lu et al., 2013), food and drinking water, and dermal contact with consumer products containing nonylphenol (HSDB, 2015). They can also be exposed through the end-of-life phase of products containing low amounts of APEs via air, drinking water, and soil (Health Canada, 2001; EC, 2002; EPA, 2010). Alkylphenol and alkylphenol ethoxylates are commonly detected in house and office indoor dust (Kuwabo et al., 2016; Abafe, 2017). This indicates that indoor house or office dust may constitute one of the major routes for human exposure to these compounds. Because laundry detergent is the largest use of APEs, it is likely that use of APE-containing detergents is contributing to all of these exposure pathways.

Laundry detergent makes a significant contribution to exposure. The largest source of exposure to people is thought to be food, especially fish (CDC, 2019; EC, 2008). APEs have been detected in wild fish tissue (Ecology, 2016; Lv et al., 2019). NPEs in laundry detergent are not completely removed by WWTPs, and therefore may accumulate in fish living in receiving waters via contact with contaminated sediment and water.

Another route of APE exposure to humans may be from drinking water. NP has been detected in bottled water and tap water, and is not removed completely by some water treatment options (Mao et al., 2012). APEs and APs from laundry detergent may contaminate drinking water supplies through WWTP effluent or biosolids application.

Exposure to APEs in laundry detergent is particularly relevant for workers. Workers handling laundry detergent have a higher potential for exposure compared to the general population (DTSC, 2018). Occupational exposure to APEs may occur through inhalation and dermal contact with this compound at workplaces where it is produced or used. Workers are exposed to APEs in laundries when they transfer chemicals into washers. These workers can be exposed to either powdered or liquid detergents. Powdered detergents have the greatest potential for inhalation exposure due to breathable dust containing APEs (DTSC, 2018).

Potential environmental exposure and exposure to sensitive species

AP/APEs have been found in Washington's environment. APEs and APs are not naturally occurring, so any detection in the environment is a consequence of human activity. Nonylphenol has been found in almost all environmental media in Washington state. NP was only detected in 1% of samples from freshwater streams during baseflow and storm events as part of data gathering for Ecology's Puget Sound Toxics Loading Study[28] (Ecology, 2011), though this is expected since the majority of environmental NP is coming from wastewater treatment plants (WWTPs). In two other separate phases of this study, NP was detected at up to 400 ng/L in WWTP influent, at up to 200 ng/L in WWTP effluent (Ecology, 2010), and found in 6.3% of samples from WWTP discharges. The Puget Sound Toxics Loading Study also predicted that NPE-containing detergents could emit NP directly into the atmosphere, which would lead to atmospheric deposition (Mackenzie, Mcintyre, Howe, & Israel, 2018).

A 2007 King County survey of endocrine-disrupting compounds found NP in marine water at levels ranging from at or below the limit of detection (ND) up to 0.254 µg/L; in lakes at ND – 0.149 µg/L; in streams at ND – 0.836 µg/L; and in stormwater at up to 44.2 µg/L (KingCounty, 2007). A 2016 study by Ecology found OPE, NP, and NPEs in 48% of freshwater fish tissue, at levels from 445 – 4080 ng/kg (Gottschall et al., 2017).

Globally there is widespread detection of APEs and APs in water, air, and sediment. Detection is often at levels well above the probable no effect concentration (PNEC) for NP—determined by the EU to protect aquatic health—of 0.33 µg/L in water and 0.039 mg/kg in sediment (DTSC, 2018; EC, 2002; Salomon et al., 2019; Selvaraj, Shanmugam, Sampath, Joakim Larsson, & Ramaswamy, 2014).

APEs from laundry detergents have the potential to contribute to environmental APE concentrations. Facilities that use high levels of APE-containing products have higher levels of APEs in their wastewater compared to facilities that do not use APE-containing products (Nagarnaik, Mills, & Boulanger, 2010). In addition, inadequately treated wastewater can release NP in the aquatic environment, further emphasizing that a large proportion of environmental NP comes from uses that are disposed of into wastewater systems, such as detergents (Mao et al., 2012).

NP concentrations are much lower in EU than in Asia. This suggests that NP concentrations in the environment are in part due to NPE/NP use in products in Asian markets that are restricted in the EU. NPE/NP and OPE/OP have been restricted for almost all uses in the EU, including laundry detergent and other cleaning products (Mao et al., 2012). In European WWTPs, NP has

[28] https://fortress.wa.gov/ecy/publications/documents/1103010.pdf

significantly decreased in both influent and effluent concentrations since the implementation of restrictions (Höhne & Püttmann, 2008).

During wastewater treatment, NP from laundry detergents can accumulate in biosolids (Mao et al., 2012). Since biosolids can then be applied to land for agricultural purposes, this allows another route of environmental exposure to APs. See the figure below for a diagram of potential pathways for laundry detergent used in on-premises laundry to enter the environment.

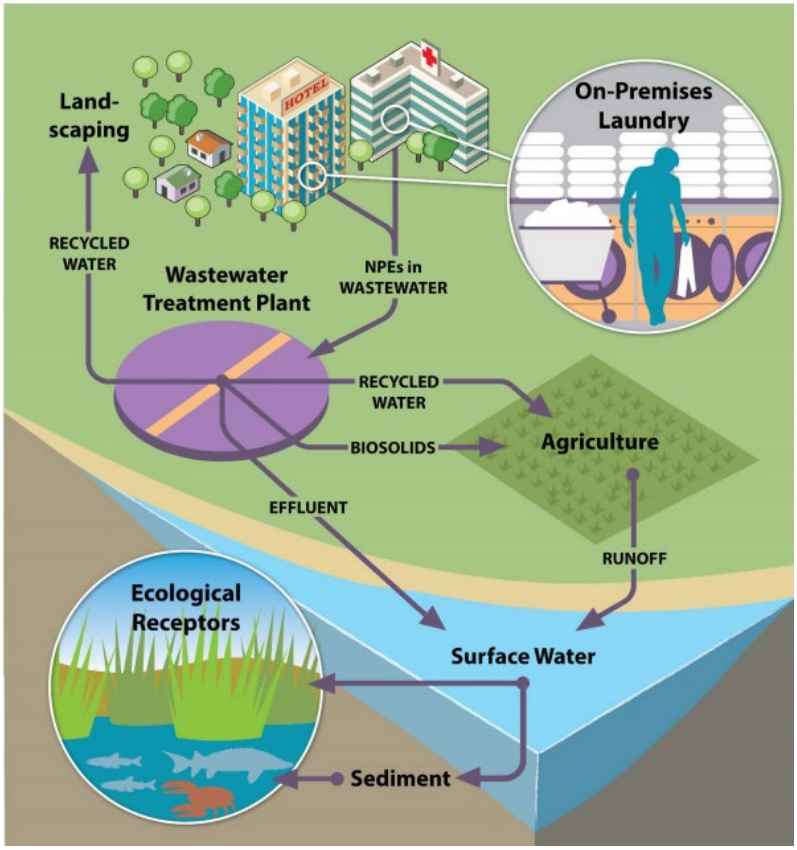

Figure 5. Potential exposure pathways for NPEs from laundry detergents (DTSC, 2018).

(This figure does not include all possible exposure pathways for NPEs from consumer products to the environment.)

Exposure to APEs in the environment is a concern for sensitive species. Using EPA's standard evaluation procedure, APs and APEs are slightly- to very-highly toxic to freshwater fish and invertebrates depending on the ethoxylate chain length. The ability of APEs to damage cell membranes increases with decreasing APE ethoxylation chain length. Consequently, non-ethoxylated NP and OP are the types most toxic to aquatic organisms (DTSC, 2018). In addition to toxicity, NP and OP have shown endocrine-disrupting properties (Celino-Brady, Petro-Sakuma, Breves, Lerner, & Seale, 2019), and therefore are restricted in the EU (ECHA, 2013). NP

causes reproductive and growth impairment in a variety of aquatic organisms, and can be immunotoxic to Pacific oysters, even at low concentrations (DTSC, 2018).

Another concern is the synergistic effect of APEs on toxicity with other compounds, specifically pesticides. This means that when combined with these compounds, such as in stormwater runoff or when applied in agriculture, the resulting mixture is more harmful to wildlife than the sum of the two chemicals separately (DTSC, 2018).

Existing regulations

Currently there are no federal or state regulations specifically addressing APEs in laundry detergents. In 2014, EPA proposed a Significant New Use Rule (SNUR) under the Toxic Substances Control Act (TSCA). The rule would require manufacturers to provide at least 90 days notice to EPA before commencing or resuming any significant new use of the 15 NP/NPEs that are no longer used in commerce. This would give EPA the opportunity to evaluate the intended use and, if warranted, take action to prohibit or limit the activity before it occurs (EPA, 2014). EPA has also added a nonylphenol and nonylphenol ethoxylates category to the Toxics Release Inventory (TRI) Program list of reportable chemicals (EPA, 2019).

The EU restricts nonylphenol and nonylphenol ethoxylates via Annex XVII at greater than 0.1% in multiple products including cleaning products. Other APEs are on the European Substance of Very High Concern (SVHC) candidate list (ECHA, 2012), requiring authorization before use.

South Korea's K-REACH (The Act on the Registration and Evaluation of Chemicals) program recently adopted restrictions similar to the European Commission's to restrict the intentional use of NPEs (greater than or equal to 0.1%) in various products, including domestic, industrial, and institutional cleaning products (DTSC, 2018).

Availability of safer alternatives

There are a number of surfactants available for laundry detergents. In addition to APEs, non-APE chemicals include alcohol ethoxylates, alkyl polyglucosides, and alkyl sulfate esters. Several agencies have investigated these alternatives including EPA (2012), DTSC (2018), BizNGO (2014), and ECHA (2014). As part of our Safer Products for Washington process, we will address the safety and feasibility of these alternatives in phase 3.

Thermal Paper

Overview

Draft priority product

Thermal Paper: Paper coated with a material formulated to change color when exposed to heat

Priority chemical

Phenolic Compounds—Bisphenols

Priority product summary

Thermal Paper is a significant source and use of phenolic compounds. Ecology made this determination by considering the criteria in RCW 70.365.030[29] of Safer Products for Washington. We are concerned about the contribution bisphenol-containing thermal papers make to the amounts of bisphenols in our bodies, especially to workers who are occupationally exposed to thermal paper. Approximately 3,300 tons of thermal paper are used every year in Washington, and testing shows that much of it contains BPS- or BPA-based chemical developers. The European Food Safety Authority (EFSA) estimates that thermal paper use is one of the leading sources of human exposure to BPA.

Multiple industries use thermal paper every day. People absorb the chemicals on thermal paper, significantly raising the concentration of bisphenols in their bodies. Retail workers are especially susceptible to this pathway due to the use of thermal paper for receipt tape. They have much higher exposure than the general population and consistently higher biological levels.

We are also concerned about the contribution thermal paper may make to bisphenol concentration in the environment. Recycling of thermal paper is the largest source of BPA to the environment. Governor Inslee's Orca Task Force has declared bisphenols a contaminant of emerging concern for our orca population. Detailed support for our listing of bisphenols in thermal paper as a priority product is shown below.

Background

Bisphenols, such as Bisphenol A (BPA) and Bisphenol S (BPS) (molecular structures below), are utilized as developers in the chemical reaction that provides color when using thermal paper. Bisphenols are organic acid solids which melt when exposed to sufficient heat. When melted, bisphenols will combine with dyes and alter the dye's pH, causing the dye to change color. Other components such as sensitizers and stabilizers can help with performance and reliability

[29] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.030

of this developer-dye reaction. All of these components are mixed into a thermally reactive layer and are applied to a wide range of base papers as a coating.

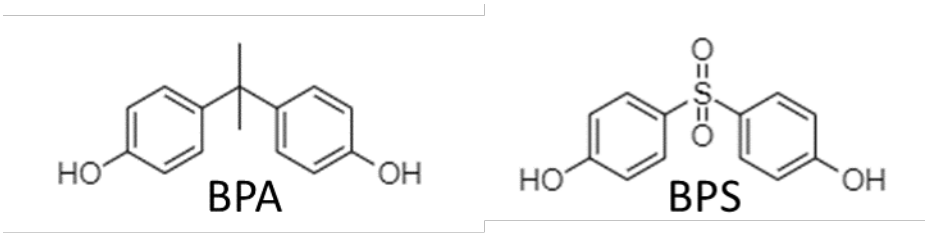

Figure 6. Molecular structures of Bisphenol A (BPA) and Bisphenol S (BPS).

Estimated volume of Bisphenols used in Thermal Paper

Bisphenols are frequently detected in thermal paper at high concentrations. The following is a summary of bisphenol concentrations found in thermal paper where, for the most part, the chemical is assumed to have been added intentionally (Table 10). Other results not included consist of detections at much lower concentrations (usually less than 100ppm), which we assume result from inadvertent contamination during manufacturing (such as due to contaminated recycled paper). Bisphenols found in commercially-available thermal paper include BPA and BPS, though other bisphenols have been proposed as alternatives (EPA, 2015; Pelch et al., 2019).

In the U.S., BPA has been detected in thermal paper at levels as high as 28,000 ppm (Lunder, Andrews, & Houlihan, 2010) (2.8%), and BPS at 71,000 ppm (7.1%). In general, it seems that recently, some thermal paper manufacturers have switched from BPA developers to BPS and other alternatives, such as Pergafast 201 and acetic acid, though bisphenols make up the majority of the developers in receipts (Table 11). These results show the widespread and well-documented use of bisphenols as thermal paper developers.

Table 10. Summary of studies investigating bisphenol concentrations found in thermal paper.

Year	Country	Chemical	Concentration	Reference
2010	U.S.	BPA	300 – 15400 ppm	Mendum, Stoler, Vanbenschoten, & Warner, 2011
2010	U.S. and Japan	BPA	8000 – 28000 ppm	Lunder, Andrews, & Houlihan, 2010
2011	Worldwide	BPA	<LOQ – 13900 ppm (Including Inadvertent)	Liao & Kannan, 2011
2012	Belgium	BPA	9000 – 21000 ppm	Geens, Goeyens, Kannan, Neels, & Covaci, 2012

Year	Country	Chemical	Concentration	Reference
2012	Worldwide	BPS	.014 – 22000 ppm (Including Inadvertent)	Liao, Liu, & Kannan, 2012
2013	China	BPA	2580 – 14700 ppm	Lu, Chang, Sojinu, & Ni, 2013
2014	US	BPA	54 – 79 µg/cm^3	Apfelbachter, Cioci, & Strong, 2014
2014	US	BPS	37 – 75 µg/cm^3	Apfelbachter, Cioci, & Strong, 2014
2015	Brazil	BPA+BPS	1000 – 43000 ppm	Rocha, Azevedo, Gallimberti, Campiglia, & Barbosa, 2015
2015 – 2017	Germany	BPA	4000 – 32400 ppm	Eckardt & Simat, 2017
2015 – 2017	Germany	BPS	5000 – 18000 ppm	Eckardt & Simat, 2017
2017	India	BPA	300 – 6600 ppm	Rajankar, Mohapatra, & Mathur, 2018
2018	US	BPA	14,500 ppm (78.6 µg/cm^3)	Zaharias, Miller & Olson, 2018
2018	US	BPS	71,000 ppm (311 µg/cm^3)	Zaharias, Miller & Olson, 2018
2019	Worldwide	BPA	<LOQ – 20270 ppm (Including Inadvertent)	Molina-Molina et al., 2019
2019	Worldwide	BPS	<LOQ – 13290 ppm (Including Inadvertent)	Molina-Molina et al., 2019

Table 11. Summary of studies investigating bisphenol prevalence in thermal paper.

Year	Country	Chemical	Prevalence in Thermal Paper Tested	Reference
2010	U.S.	BPA	8/10 (70%)	Mendum, Stoler, Vanbenschoten, & Warner, 2011
2010	U.S. and Japan	BPA	12/39 (31%)	Lunder et al., 2010
2011	Worldwide	BPA	97/103 (94%) Including Inadvertent	Liao & Kannan, 2011
2012	Belgium	BPA	32/44 (73%)	Geens, Goeyens, Kannan, Neels, & Covaci, 2012
2012	Worldwide	BPS	111/111 (100%) Including Inadvertent	Liao, Liu, & Kannan, 2012

Year	Country	Chemical	Prevalence in Thermal Paper Tested	Reference
2013	China	BPA	42/42 (100%)	Lu, Chang, Sojinu, & Ni, 2013
2014	US	BPA	9/18 (50%)	Apfelbachter, Cioci, & Strong, 2014
2014	US	BPS	9/18 (50%)	Apfelbachter, Cioci, & Strong, 2014
2015	Brazil	BPA+BPS	186/190 (98%)	Rocha, Azevedo, Gallimberti, Campiglia, & Barbosa, 2015
2015 – 2017	Germany	BPA	46.9% – 52.5%	Eckardt & Simat, 2017
2015 – 2017	Germany	BPS	6.1% – 11.4%	Eckardt & Simat, 2017
2016	US	BPA	5/103 (5%)	Lucia, 2016
2016	US	BPS	98/103 (95%)	Lucia, 2016
2017	India	BPA	12/12 (100%)	Rajankar, Mohapatra, & Mathur, 2018
2018	US	BPA	30/167 (18%)	Zaharias, Miller & Olson, 2018
2018	US	BPS	126/167 (75%)	Zaharias, Miller & Olson, 2018
2019	Worldwide	BPA	85/112 (75.9%)	Molina-Molina et al., 2019
2019	Worldwide	BPS	14/112 (12.5%)	Molina-Molina et al., 2019
2019	Worldwide	BPF	0%	Molina-Molina et al., 2019

Estimated volume used in Washington

Approximately 3,300 tons of thermal paper are used per year in Washington. Minnesota Pollution Control Agency estimated in 2015 that national thermal paper use was about 146,000 imperial tons annually (Cioci, Apfelbacher, Strong, & Innes, 2015). Taking the Washington population share of that would give 3,300 tons (6.6 million pounds) of thermal receipt paper used.

An EU risk assessment estimated that 1,860 metric tons of BPA were used per year for thermal paper in the EU during 2005 – 2006 (Aschberger et al., 2008). Taking the ratio of populations (Washington versus EU) and extrapolating to current time would give an estimated usage of 29 metric tons (64,000 lbs) of BPA in Washington for thermal paper. This does not take into account BPS use or changes in thermal paper components since 2006.

Potential for exposure to sensitive populations when used

Nearly all people are exposed to bisphenols. Findings in the Fourth National Report on Human Exposure to Environmental Chemicals indicate widespread exposure to BPA. CDC scientists found BPA in more than 90% of the urine samples representative of the U.S. population. NHANES data from 2003 to 2014 show a decreasing trend in the urinary BPA concentration for the general U.S. population (CDC, 2017). Because BPA does not persist for long periods of time in the body, its widespread detection in people indicates that exposures occur frequently. In 2011, the World Health Organization (WHO) estimated the mean dietary daily intake of BPA for adults to be 0.4 – 1.4 μg per kg of body weight (Hines et al., 2017).

The levels of BPA in humans have changed over time, likely due to their replacement in products. Besides BPA, other bisphenols that may be used in can linings, such as BPS and BPF, have also been detected in the urine of the U.S. general population, according to NHANES 2013 – 2014 survey (Lehmler, 2018). Exposure to other bisphenols (e.g., BPS) appear to have increased in the U.S. population due to substitution (Ye et al., 2015). However, their levels appear to be lower compared to BPA, and the detection frequency is much higher for BPA than other bisphenols.

Due to inaccuracies in historical testing methods, most BPA levels in organisms are likely severely underreported. One study found levels in urine 19 times higher using direct analysis versus the indirect analysis used by federal agencies and in the NHANES data set (Gerona, vom Saal, & Hunt, 2019). This has potentially affected the result of previous risk calculations by suggesting that population exposure levels are much lower than they are in reality.

Bisphenols in thermal paper make a significant contribution to exposure. Exposure to bisphenols through thermal paper can occur because bisphenols exist as free molecules in the coating layer. These molecules wear off and can easily be transferred to the skin or anything that touches the paper (Liao & Kannon, 2011). Bisphenols can then be absorbed through the skin or transferred from unwashed hands to food and ingested (Hormann et al., 2014).

Thermal paper is a significant source of BPA for the general population above three years of age (accounting for up to 15% of total exposure in some population groups) (EFSA, 2013). EFSA (2013) indicates that thermal paper is the second largest source of external BPA exposure after eating food and/or drinking water that contains BPA.

Prolonged exposure to BPA from single dermal contact leads to detectable urinary levels in people. One study found that after dermal exposure, urine levels remain elevated for up to 9 days, whereas after oral exposure they return to normal after 10 – 20 hours. This contributed to the fact that after exposure, a greater proportion of the BPA was detected in urine for dermal exposure than with oral exposure (Liu & Martin, 2017). Thus, it seems likely that dermal exposure to BPA will lead to a higher proportion of BPA in systemic circulation compared to oral exposure.

Several studies have shown that briefly handling receipt papers leads to significant absorption into the body. One study found that over 88% of BPS exposure for most humans comes from handling thermal receipts (Liao et al., 2012). Another study found that BPA transfers readily from receipts to skin and can penetrate the skin to such a depth that it cannot be washed off. This study determined that absorption of BPA into the body increases as much as ten-fold when thermal paper is handled with moist or greasy fingers (Biedermann, Tschudin, & Grob, 2010).

Exposure to bisphenols in thermal paper is particularly relevant for children and retail workers. Several studies have calculated estimated daily intake (EDI) for exposure to bisphenols from thermal paper. A study measured BPA in 44 thermal papers collected in Belgium. Exposure levels in the Belgian general population from thermal paper were 445 ng BPA/day (Geens et al., 2012). EFSA estimated that BPA exposure through thermal paper averaged 0.071 µg/kg/day, or 4.4 µg/day for an average 62 kg adult, with the highest exposure group being adolescents aged 10 – 18 years, who had exposure averaging 1.6 times higher than adults, at 0.113 µg/kg/day (EFSA, 2015).

The exposure for people who regularly contact thermal paper, such as employees who handle paper receipts, can be much higher. For instance, median intake was found at 1.42 µg/day of BPA and BPS in the Brazilian general population, while intake levels for people who were occupationally exposed was 71 µg/day (Rocha et al., 2015). In Shenzhen, China, EDIs were found at 0.69 µg/day for general population and 40.4 µg/day for workers at a supermarket (Lu et al., 2013). At an Italian market, exposures from thermal paper were 0.0625 µg/day for the general population, and 66.8 µg/day for occupationally exposed individuals (Russo, Barbato, & Grumetto, 2017).

The CDC reports that people working in retail industries have 30% more BPA in their bodies than the average U.S. adult (CDC, 2017). On average, workers in the National Institute for Occupational Safety and Health (NIOSH) study had BPA levels in their urine about 70 times higher than adults in a NHANES general population study (Hines et al., 2017). NHANES data also showed that overall, workers with potential occupational exposure are more likely to have detectable levels of urinary BPA. Notably, females with potential occupational exposure had significantly higher urinary BPA excretion compared with females with unlikely occupational exposure. However, there was no statistically significant association between occupation and urinary BPA in males (Hehn, 2016).

In cashiers who handled bisphenol-containing receipts, bisphenols levels in urine were significantly higher than in non-cashiers. Post-shift levels (0.54 µg/g) of urinary BPS were significantly higher than pre-shift levels (0.23 µg/g) for the 32 cashiers who handled BPS-containing receipts (Thayer et al., 2016). Other studies found a significant increase in urinary BPA levels for cashiers handling thermal paper daily (Ndaw, Remy, Jargot, & Robert, 2016), and that levels remained elevated 48 hours after handling thermal receipts (Liu et al., 2017).

Potential environmental exposure and exposure to sensitive species when used

Bisphenols have been found in Washington's environment. Bisphenols are present throughout Washington in all forms of environmental media. The Lower Columbia River Estuary Partnership found BPA in the Columbia river in 2004 (LCREP, 2007). As part of the Puget Sound Toxics Loading Study,[30] Ecology found BPA in biosolids (Ecology, 2010a), in 53.3% of WWTP Effluent (Ecology, 2010b), and in stormwater, with higher levels during rain events (Ecology, 2011). BPA was found in Puget Sound at much higher levels than in the relatively uninhabited Barkley Sound, BC, indicating human contributions to environmental concentrations (Keil, Salemme, Forrest, Neibauer, & Logsdon, 2011).

Outside of Washington, BPA is frequently detected, sometimes at concentrations exceeding levels protective of human and environmental health as defined by governmental agencies (Corrales et al., 2015). Other bisphenols are also commonly found in environmental media, such as sediment, water, soil, dust, and throughout the WWTP process (Chen et al., 2016; Hu, Zhu, Yan, Liao, & Jiang, 2019).

Bisphenols from thermal paper have the potential to contribute to environmental bisphenol concentrations. A 2008 risk assessment by the EU found that while thermal paper production is one of the smallest industrial uses of BPA, recycling of the paper contributes the largest industrial source of BPA entering the environment (Aschberger et al., 2008). Although BPA is often removed at greater than 90% efficiency by WWTPs, due to the large volumes of wastewater discharged by recycling facilities, this is still a major source of environmental release (PPRC, 2015).

Sewage sludge has some of the highest environmental levels of BPA detected, so biosolids used in agriculture have the potential to be sources of BPA to soil, groundwater, and surface water (Michałowicz, 2014). End-of-life thermal paper also has the potential to contribute bisphenols to the environment, either through landfill, recycling, or littering (Wang, Liu, & Liu, 2019). High levels of BPA have been detected in landfill leachate, and groundwater has been contaminated with BPA near landfills (Michałowicz, 2014).

Exposure to Bisphenols in the environment are a concern for sensitive species. Bisphenols can have a variety of detrimental effects on wildlife (Cioci et al., 2015). There is evidence that BPA is slightly bioaccumulative in some species (Corrales et al., 2015). BPA has been shown to cause embryonic deformities, abnormal behavior, and growth inhibition, and can reduce the number of offspring in fish (Kang, Aasi, & Katayama, 2007). Many bisphenols show endocrine disrupting, cytotoxic, genotoxic, reprotroxic, and neurotoxic effects (Chen et al., 2016). Growth effects

[30] https://fortress.wa.gov/ecy/publications/documents/1103010.pdf

have been shown in fish exposed to water with concentrations as low as 0.078 µg/L (Kang, Aasi, & Katayama, 2007).

While BPA is the most studied bisphenol, there is evidence that other compounds have similar, if not worse, effects on wildlife. For example, BPF, BPS, BPAF, BPB, and BPC have shown estrogenic activity similar to BPA (Chen et al., 2016), and can cause stress effects at low levels in a variety of organisms (Zhou, 2018). Bisphenols were deemed a chemical of concern for the endangered Puget Sound orca population (Southern Resident Orca Taskforce, 2018).

Existing regulations

There are no U.S. federal regulations regarding BPA in thermal paper. In the US, there is a state-driven movement to limit exposures to BPA. There are 31 pending legislative policies in 13 states and 30 laws in 14 states pertaining to limits, bans, monitoring, or reporting of BPA in products. Of the 30 adopted laws, two pertain to paper. In 2011, Connecticut established the first state ban on BPA in thermal receipt paper. In 2019, Illinois prohibited the manufacture, distribution or use of BPA in making banking or business paper (Safer States, 2019). California Department of Toxic Substances Control (DTSC) has proposed bisphenols in office machinery consumables as a priority product under their Safer Consumer Products program. This law asks manufacturers to consider alternatives to chemicals of concern in their products and may lead to regulation (DTSC, 2018).

In 2010, EPA proposed rulemaking under the Toxic Substances Control Act (TSCA) to identify BPA as "a substance that may present an unreasonable risk of injury to the environment on the basis of its potential for long-term adverse effects on growth, reproduction and development in aquatic species at concentrations similar to those found in the environment." This proposed rule has been stalled in the Office of Management and Budget for several years (EPA, 2010).

BPA will be banned in thermal paper in the EU starting in 2020 (ECHA, 2016). BPS and BPA are banned in thermal paper in Switzerland (ChemicalWatch, 2019a).

Availability of safer alternatives

Several alternatives to bisphenol-containing thermal paper are available and in circulation. There are a number of automatic paper documentation options including thermal paper and printed paper. For thermal paper, developers include bisphenols, ascorbic acid, Pergafast 201, and urea compounds. Another thermal paper alternative is heat-voided paper. For some applications, no receipt or a digital receipt may be suitable alternatives. Alternatives have been investigated by other agencies, such as EPA (2014) and MPCA (Cioci et al., 2015). Several retailers have policies in place prohibiting use of bisphenols in thermal receipts (ChemicalWatch, 2019b). As part of our Safer Products for Washington process, we will address the safety and feasibility of alternatives to bisphenols in thermal paper during phase 3.

Flooring

Overview

Draft priority product

Vinyl flooring

Priority chemical

Phthalates

Priority product summary

Vinyl flooring is a significant source and use of phthalates. Ecology made this determination after considering the criteria in RCW 70.365.030.[31] We estimate vinyl flooring may contain phthalates at concentrations ranging from 9 – 32% by weight, contributing significant amounts of phthalates to our homes, workplaces, and environment.

We are particularly concerned with the potential for infants and young children to be exposed to phthalates from vinyl flooring. Multiple studies have reported associations between vinyl flooring in the home and higher concentrations of urinary phthalate metabolites in infants, children and pregnant women.

Vinyl flooring has been associated with higher concentrations of phthalates in air and dust. As vinyl flooring ages, phthalates are released into the environment. In 2011, Ecology estimated that vinyl flooring may contribute 200 pounds of phthalates to Puget Sound each year. Governor Inslee's Southern Orca Task Force named phthalates an emerging chemical of concern for orcas and their food sources. Detailed support for our listing of phthalates in vinyl flooring as a priority product is shown below.

Background

Phthalates are used in vinyl flooring, also known as PVC flooring, to soften plastic and increase flexibility and durability. Historically, ortho-phthalates, such as DEHP and DINP, were the majority of the phthalates used in vinyl flooring. But over time, the industry has moved toward terephthalates. Recently, an NGO study supported retailers' claims that they have stopped selling flooring containing ortho-phthalates (Miller, Belliveau, Walsh, & Shade, 2019, not peer reviewed). Ecology is verifying the extent of ortho-phthalate removal from new flooring sold in Washington.

[31] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.020

Estimated volume of Phthalates used in vinyl flooring

We estimate that over half of vinyl flooring may contain phthalates at concentrations ranging from 9 to 32% by weight. The volume of phthalates used in vinyl flooring has changed over time. In 2011, Washington state estimated that among polyvinyl chloride (PVC) products, including flooring, 30% are composed of DEHP (Ecology 2011). Afshari et al. (2004) found that 17 – 18.5% of the PVC flooring was comprised of DEHP.

In 2014, a study of 16 types of vinyl flooring found concentrations of phthalates ranging from 9 – 23% of the flooring by weight (Liang & Xu, 2014). In 2016, California Office of Environmental Health Hazard Assessment estimated that DINP was present in vinyl floors at up to 18.9% in their safe use determination statement (California Office of Environmental Health Hazard Assessment, 2016). A summary of the percent of ortho-phthalates found in vinyl flooring is shown in Table 12. If we assume a home has 151 square feet of vinyl flooring weighing approximately 0.9 kg/sq ft, vinyl flooring containing 18% DIDP per weight could contribute 24 kg of DIDP to the home.

Table 12. Examples of the percentage of vinyl flooring by weight comprised of phthalates.

(*Based on California's Safe Use Determination)

Phthalate	Major Metabolites	Concentration	Reference
DEHP	MEHOP, MEHHP, MEHP, MECPP	17 – 18.5%	Afshari, Gunnarsen, Clausen, & Hansen, 2004
DINP	MINP, MHINP, MOINP	20%	Liang & Xu, 2014
DEHP	MEHOP, MEHHP, MEHP, MECPP	23%	Liang & Xu, 2014
BBP	MBzP	15%	Liang & Xu, 2014
DnBP	MnBP	9%	Liang & Xu, 2014
DINP	MINP, MHINP, MOINP	18.9%*	California, 2016
DEHP	MEHOP, MEHHP, MEHP, MECPP	0.16 – 32.3%	Noguchi & Yamasaki, 2016

In 2015, ortho-phthalates were commonly sold in vinyl flooring. An NGO study analyzed 65 vinyl flooring tiles and detected phthalates in 58% (Ecology Center, 2015, not peer reviewed). It is important to note that in 2018 the same group reassessed phthalates in vinyl flooring sold from retailers advertising "phthalate-free flooring," and did not detect phthalates (Miller et al., 2019, not peer reviewed).

Estimated volume used in Washington

We estimate that vinyl flooring sold in WA each year contributes 4,536 – 16,783 metric tons of phthalates to our homes and workplaces, and that .17 tons of phthalates are released into the environment from vinyl flooring each year in WA. Recent national estimates of the sales of resilient flooring, a category of flooring comprised largely of types of vinyl flooring, range from $3.68 billion in 2016 (Floor Covering Weekly, 2016) to $4.5 billion in 2019 (Resilient Floor Covering Institute, 2019), the lower amount corresponding to 4.27 billion square feet.

Using Washington's population in proportion to the national population, this translates to approximately 100 million square feet (90,718 metric tons) sold annually in the state. Based on the detection rate of 58% (Ecology Center, 2015, not peer reviewed) and a concentration range of 9 – 32% phthalates by weight (Table 12), we estimate that 4,536 – 16,783 metric tons of phthalates are found in vinyl flooring purchased annually in WA, and that 0.17 tons of phthalates are released to the environment from vinyl flooring.

Potential for exposure to sensitive populations

Nearly all people, including children, men and women of childbearing age, and workers, are exposed to phthalates. Phthalate exposure is widespread in the U.S. general population. The Centers for Disease and Control (CDC) (2019) notes that over 90% of Americans have measurable levels of many phthalate metabolites in their bodies. Ten years of biomonitoring data conducted by the CDC shows widespread exposure to most phthalates in the U.S. population. Between 2001 – 2002 and 2009 – 2010, DEHP exposure has declined about 37%, whereas DINP and DIDP exposure have increased (Zota, Calafat, & Woodruff, 2014). DIDP metabolites were detected in 89% of participants in 2005 – 2006 and 94% of participants in 2009 – 2010 (Zota, Calafat, & Woodruff, 2014). DINP metabolites were detected in 96% of participants in 2005 – 2006 and 98% of participants in 2009 – 2010 (Zota, Calafat, & Woodruff, 2014).

Between 2010 and 2011, the Washington Environmental Biomonitoring Study analyzed urine samples from 422 women of childbearing age and teens for phthalate metabolites. MEOHP, MEHHP, MEHP, and MBZP are some of the major metabolites from phthalates used in vinyl flooring (Table 12). These metabolites were detected in 98% of samples (Health, 2019). This is similar to results from a cohort of 378 pregnant women from Charleston, SC. MBzP, MEHHP, MEOP and MEHP were all detected in greater than 90% of urine samples (Wenzel et al., 2018).

Children show similar exposure patterns as women of childbearing age. MBzP and MEHHP were detected in greater than 96% of samples from children ages 6 to 11 collected between 2007 and 2012 (Odebeatu, Taylor, Fleming, & Osborne, 2018). Human biomonitoring studies have measured parent phthalate in serum (Specht et al., 2014) and their metabolites in human urine (CDC, 2019; Dong et al., 2017; Health Canada, 2013; Silva et al., 2007; Suzuki et al., 2009; Tefre

de Renzy-Martin et al., 2014), semen (Chen et al., 2017; Nassan et al., 2016), saliva (Silva, 2005), and breast milk (Hogberg et al., 2008; Main et al., 2006). Phthalates can cross the placental barrier (Fennel, Krol, Sumner, & Snyder, 2004), and some have been detected in cord blood (Ashley-Martin, 2015) and amniotic fluid (Silva et al., 2004).

These findings demonstrate that exposure to phthalates is widespread and that sensitive populations often have particularly high exposures.

Sensitive populations are exposed to phthalates in vinyl flooring. Epidemiological studies observe higher concentrations of BBP metabolites in people who live in homes with vinyl flooring. Because phthalates are not chemically bound to PVC in vinyl flooring, they can leach out over time, allowing for dermal exposure and contaminating indoor air and house dust (Xu, Cohen-Hubal, Clausen, & Little, 2009). House dust ingestion and inhalation is an important exposure route for infants and young children because they spend more time on or near the ground, have more frequent hand-to-mouth activity, and experience faster respiration rates than adults. A Swedish study of preschool dust observed higher concentrations of DINP in rooms with PVC flooring (Larsson et al., 2017).

Multiple studies have found associations between phthalate metabolites in children's urine and vinyl flooring in the home. A prospective birth cohort of 239 children from New York reported that BBP concentrations in indoor air were significantly higher in rooms with vinyl or linoleum flooring (Just et al., 2015). Children living in homes with vinyl or linoleum floors had higher concentrations of BBP metabolites in their urine (Just et al., 2015). These results are similar to a Swedish study of 110 infants, which found that PVC flooring in the bedroom was associated with higher urinary BBP metabolite concentrations (Carlstedt, Jonsson, & Bornehag, 2012).

Another study shed light on the magnitude of the impact of vinyl flooring on children's urinary BBP metabolites, reporting that children who lived in homes with 100% vinyl flooring had urinary concentrations of BBP metabolites 15 times higher than those of children who lived in homes with no vinyl flooring (Hammel et al., 2019). The relationship between BBP metabolites and vinyl flooring in the home has also been observed in pregnant women (Shu et al., 2019).

Vinyl flooring has also been associated with worsening asthma symptoms, particularly in children. In fact, another study found that children living with vinyl flooring in their bedroom were 1.5 times more likely to develop asthma during the following 10-year period as compared with children who live in homes with other types of flooring. The association was strongest for children whose parents had vinyl flooring in their bedrooms during pregnancy (Shu, Jönsson, Larsson, Nånberg, & Bornehag, 2014). A study of 10,851 children in Sweden found the presence of floor moisture and PVC significantly increased the risk of asthma (Bornehag et al., 2005).

Workers in buildings with vinyl flooring can also have increased exposure. In a Swedish study of personnel in four geriatric hospitals, researchers found asthma symptoms were more common in the two buildings with signs of dampness. They suggested that this was related to phthalate

degradation in PVC flooring (Norbäck, Wieslander, Nordström, & Wålinder, 2000). A similar study in office buildings also implicated PVC flooring. After employees complained of several respiratory, conjunctival and dermal symptoms, researchers identified degradation of the plastic floor coverings as the source of chemicals in the air (Tuomainen, Seuri, & Sieppi, 2004).

Potential environmental exposure and exposure to sensitive species

Releases of phthalates from vinyl flooring can contribute to environmental concentrations. Phthalates found in vinyl flooring can be released from the product into air and dust (Xu et al., 2009). Following their release from flooring, phthalates can be tracked outside where they can contaminate the environment.

Phthalates are frequently detected in the environment. In the Puget Sound area, Commencement Bay and the Lower Duwamish Waterway are EPA superfund sites, partially due to phthalate contamination. Because phthalates are not environmentally persistent, the concentrations observed at these sites are reflective of current release levels. In 2010, a remedial investigation into phthalate levels in the Lower Duwamish Waterway found DEHP in surface sediment, surface water, fish and invertebrates samples and BBP in surface sediment (Lower Duwamish Waterway Group, 2010).

A similar investigation into Commencement Bay showed decreasing concentrations of many contaminants between 1999 and 2008. However, during that same time period, concentrations of DEHP increased. DEHP was detected in all 30 sediment samples and BBP was detected in 20% of sediment samples (Ecology, 2010). The City of Tacoma has been monitoring phthalates in stormwater sediment traps from 2001 to 2016. DEHP and BBP were detected in stormwater sediment traps (City of Tacoma, 2017). King County has conducted additional monitoring in the Duwamish Estuary and still detected DEHP in sediment, and concluded that juvenile Chinook salmon are exposed to phthalates from sediment in the estuary (King County, 2018).

In 2011, Ecology's Puget Sound Toxics Loading Study[32] estimated the environmental release of phthalates to the Puget Sound area from various sources, including vinyl flooring. Twenty percent of phthalates, 7 tons per year, are attributable to PVC products. Of the PVC products, vinyl flooring contributes 1.4% of phthalates or 0.1 ton of phthalates released into Puget Sound each year (Ecology, 2011). Expanding this 0.1 ton estimate from the Puget Sound region only to the entire population in Washington, we expect that 0.17 tons of phthalates are released to the environment from vinyl flooring.

Phthalates are also an emerging and major source of leachate contaminant from landfills. They are primarily emitted from consumer products and building materials, which include flooring

[32] https://fortress.wa.gov/ecy/publications/documents/1103010.pdf

(Ramakrishnan et al., 2015). As phthalates are used as plasticizers, it is likely that PVC materials in the landfill are contributing to the phthalates in leachate (Kalmykova, Bjorklund, Stromvall, & Blom, 2013). There is widespread evidence from worldwide landfill studies that phthalates are leaching, and can become ubiquitous contaminants in the surrounding environment. Disposal of household materials such as flooring is a primary source of phthalates that can contaminate various environmental media (Liu et al., 2010; Paxeus, 2000; Reid, Brougham, Fogarty, & Roche, 2007; Wowkonowicz & Kijenska, 2017). Phthalate-contaminated leachate can seep into surrounding groundwater to a greater extent than surface water or topsoil (Liu et al., 2010).

Phthalates are a concern for sensitive species. Governor Inslee's Orca Task Force named phthalates as a chemical of emerging concern.

Existing regulations

Currently, no U.S. federal regulations exist for phthalates in vinyl flooring. EPA is evaluating five phthalates under the revised Toxic Substances Control Act (TSCA): DBP, BBP, DEHP, DIBP, and dicyclohexyl phthalate. These phthalates are slated for further assessment by EPA (EPA, 2019). Additionally, manufacturers requested that EPA conduct risk evaluations for DIDP and DINP under TSCA Section 6. EPA granted both of these manufacturer requests.

In the U.S., there is a state-driven movement to limit exposures to phthalates. There are 31 proposed bills in 13 states and 30 adopted laws in 14 states pertaining to limits, bans, monitoring, or reporting of phthalates. Of the 30 existing laws, 20 ban phthalates in a variety of food containers, paper products, and children's products. Federal policies (including the federal Consumer Product Safety Improvement Act (CPSIA)) prohibit more than 0.1% of three types of phthalates in children's toys: DEHP, DBP, and BBP (CPSC, 2019; Safer States, 2019).

The European Chemicals Agency (ECHA) maintains a list of 10 phthalates on their Candidate List of Substances of Very High Concern published under the REACH Regulation (ECHA, 2019). Four phthalates (BBP, DBP, DEHP, and DIBP) are restricted to 0.1% by weight (individually or combined) of the plasticized material in any product, including flooring (QIMA, 2019).

Availability of safer alternatives

There are a number of resilient flooring options including cork, tile, vinyl flooring, linoleum, rubber, and polyolefins. Vinyl flooring may use orthophthalate plasticizers or alternatives, such as Diisononyl cyclohexandicarboxylate (DINCH), Tris (2-ethylhexyl) trimellitate (TOTM), and Bis (2-ethylhexyl) terephthalate (DEHT). Several alternatives have been investigated by other agencies, such as Northwest Green Chemistry (Northwest Green Chemistry, 2018), Health Care Without Harm (Lent, Silas, & Vallette, 2009), and Healthy Building Network (Lott, 2014). As part of our Safer Products for Washington process, we will address the safety and feasibility of these alternatives in phase 3.

Fragrances

Overview

Draft priority product

Fragrances in the personal care and beauty sector, which include perfumes, toilet waters, colognes, body mists and body sprays.

Priority chemical

Phthalates: Synthetic chemical esters of phthalic acid

Priority product summary

Fragrances used in the personal care and beauty sector are a significant source and use of phthalates. Ecology made this determination after considering the criteria in RCW 70.365.020.[33] We are concerned about the contribution fragrances make to phthalate exposure in sensitive populations and the environment. Women of childbearing age and pregnant women have higher exposure to the phthalates used in fragrances than men. Among women, exposure is higher in women of color, low-income women and women with lower educational attainment. These findings have important implications for environmental justice.

Phthalates are frequently detected in the environment, particularly in urban areas, such as Commencement Bay and the Duwamish Estuary. In 2011, Ecology estimated that fragrances accounted for 33% of the 34 tons phthalates of released into Puget Sound annually. While phthalates are not environmentally persistent, the large diffuse sources have led to widespread environmental contamination that has the potential to impact sensitive populations. Governor Inslee's Orca Task Force named phthalates a chemical of concern for orcas and their food sources. Detailed support for our listing of phthalates in fragrances as a priority product is shown below.

Background

Phthalates serve two functions in fragrances: 1) solvents to keep ingredients well-blended and 2) fixatives that help extend the scent lifetime. In order to function in fragrances, phthalates must be able to volatize, making lower molecular weight phthalates more useful. The most commonly reported phthalates used in fragrances are diethyl phthalate (DEP), dimethyl phthalate (DMP), di-n-butyl phthalate (DBP), butylbenzyl phthalate (BBP) and di(2-ethylhexyl) phthalate (DEHP).

[33] https://app.leg.wa.gov/RCW/default.aspx?cite=70.365.020

Estimated volume of phthalates used in fragrances

Phthalates are frequently detected in fragrances at concentrations that contribute a significant volume to our homes and environment. Since 2002, FDA has been surveying the concentrations of phthalates in personal care and beauty products. While overall their data show a decline in the use of phthalates, DEP was detected in almost half (11 out of 25) of the fragrances sampled in 2010 (FDA, 2013). The concentrations ranged between 480 – 40,000 ppm.

More recent studies suggest phthalates are still widely found in fragrances (Table 13). DEP is the most frequently detected phthalate in fragrances and it is found at mean concentrations between 0.16 and 0.18%, with the maximum concentration reported as 4.4%. In 2015, a study of 47 fragrances detected at least one phthalate in all products analyzed (Al-Saleh, 2016). Of the five phthalates analyzed (DMP, DEP, DBP, BBP, and DEHP), DEP was found in the greatest concentration and present in all 47 products analyzed. This is consistent with a 2013 study of 12 fragrances that detected DEP in all products tested (Guo and Kannan, 2013a). Table 13 shows detection frequencies and concentrations of phthalates in fragrances. These studies support our 2011 estimation that the total phthalate concentration in fragrances is around 1.2% (Ecology, 2011).

In studies of multiple types of personal care and beauty products, fragrances frequently have the highest concentrations and detection rates for phthalates analyzed. A 2011 study by Koniecki et al. tested 252 products from the Canadian market for 18 phthalates. They detected DEP, DMP, DEBP, DBP, and DEHP. DEP was the most frequently detected phthalate and the highest concentrations were found in fragrances (Koniecki, Wang, Moody, & Zhu, 2011). Guo and Kannan's (2013) study of 170 personal care products also found that fragrances had the highest detection frequency and concentration for DEP.

Table 13. Detection frequencies and concentrations of phthalates in fragrances, such as perfumes and colognes.

Phthalate	Metabolites	Detection Frequency	Mean (and Maximum) Concentration	Reference
DEP	MEP	11 out of 25	7813 ppm (max 44,000 ppm)	FDA, 2013
DEP	MEP	47 out of 47	1622 ppm (max 23,649 ppm)	Al-Saleh, 2016
DMP	MMP	47 out of 47	30 ppm (max 405 ppm)	Al-Saleh, 2016
BBP	MBZP	47 out of 47	8 ppm (max 187 ppm)	Al-Saleh, 2016
DEHP	MEHOP, MEHHP, MEHP, MECPP	46 out of 47	6 ppm (max 147.5 ppm)	Al-Saleh, 2016

Phthalate	Metabolites	Detection Frequency	Mean (and Maximum) Concentration	Reference
DEP	MEP	12 out of 12	3420 ppm (max 7980 ppm)	Guo & Kannan, 2013
DBP	MBP	8 out of 12	0.21 ppm (max 0.97 ppm)	Guo & Kannan, 2013
BBP	MBZP	3 out of 12	6.70 ppm (max 78.3 ppm)	Guo & Kannan, 2013
DEHP	MEHOP, MEHHP, MEHP, MECPP	12 out of 12	2.71 ppm (max 12.2 ppm)	Guo & Kannan, 2013

The EPA exposure factors handbook (2011) estimates that 0.65 grams of colognes and toilet waters are used per application and applied between 0.56 – 0.85 times per day, on average. If we estimate that fragrances contain 3420 ppm DEP and assume an application rate of 0.85 time per day, over the course of a year, a person could contribute 0.7 grams of DEP to their body, home and environment. If the fragrance contained a higher concentration of DEP, such as 44,000 ppm, 8.9 grams of DEP could be released. Based on the studies discussed above, we conclude that phthalates are frequently used in fragrances and contribute to the volume of phthalates found in our bodies, homes and the environment.

Estimated volume used in Washington

We estimate that fragrances are one of the leading contributors of phthalates to Washington's environment and that 17 tons of phthalates are released into Washington's environment each year from fragrance use. In 2011, Ecology published the Puget Sound Toxics Loading Study,[34] which estimated the contribution various consumer products and chemicals make to Puget Sound. Ecology estimated that there were 13 tons of phthalates contained in the cosmetics and personal care products used yearly in the Puget Sound Region, with 11 tons coming from fragrances. Fragrances released more phthalates to Puget Sound than the other consumer product categories considered, and accounted for greater than 30% of total phthalate release (Ecology, 2011). Expanding this estimate beyond the Puget Sound region to the entire state, we estimate that 17 tons of phthalates are released from fragrances in personal care products in Washington each year.

Based on the continued use of phthalates in fragrances, we anticipate that fragrances still contribute a significant volume of phthalates to Washington's environment. Using Washington state's population to estimate its proportion of national sales, in 2018, $213 million was spent on skincare and makeup products in our state (The NPD Group, 2019). These two categories are only part of a larger market for beauty and personal care, on which Washington spends about

[34] https://fortress.wa.gov/ecy/publications/documents/1103010.pdf

two billion dollars yearly (Statista, 2019). One survey by a cosmetics company found that the average woman owns almost 40 makeup products (Stowaway Cosmetics Survey, 2015).

Potential for exposure to sensitive populations when used

Nearly all people, including infants, children, men and women of childbearing age, and workers, are exposed to phthalates. Phthalate exposure is widespread in the U.S., with vulnerable populations, such as women of color, pregnant women, women of childbearing age and low-income populations having higher exposures. The Centers for Disease and Control and Prevention (CDC) notes that over 90% of Americans have measurable levels of phthalate metabolites in their urine. CDC also reports that women have higher levels of urinary metabolites than men for phthalates that are associated with personal care and beauty products (CDC, 2019).

Between 2010 and 2011, the Washington Environmental Biomonitoring Study analyzed urine samples from 422 women of childbearing age and teens for phthalate metabolites. MEP, MBP, MEOHP, MEHHP, MEHP and MBZP are some of the major metabolites from phthalates used in fragrances (Table 13). These metabolites were detected in 98% of samples. Washington's Low Income Survey and Testing Project analyzed phthalate metabolites in 579 low-income women of childbearing age and teenagers, and observed higher concentrations of MEP, the major metabolite from DEP, compared to the general Washington sample (DOH, n.d.).

These findings are particularly relevant for pregnant women and women of childbearing age. A prospective study of 446 pregnant women found that lower educational attainment and income were associated with higher concentrations of DBP metabolites in their urine (Polinski et al., 2018). Similar results were observed in a cohort of 378 pregnant women from Charleston SC. MEP, MMP, MBzP, MBP, MEHHP, MEOP and MEHP were all detected in greater than 90% of urine samples, with MEP found at the highest concentrations (Wenzel et al., 2018).

Children show similar exposure patterns as women of childbearing age. MEP, MBzP, MBP, MEHHP, MEOP were detected in greater than 96% of samples from children ages 6 to 11 collected between 2007 and 2012, with MEP detected at the highest concentration (Odebeatu, Taylor, Flemming, & Osborne, 2019).

These findings demonstrate that exposure to phthalates is widespread, and that sensitive populations often have particularly high exposures.

There is the potential for sensitive populations to be exposed to phthalates in fragrances. Women of childbearing age, pregnant women and children can be exposed to phthalates when applying fragrances. Phthalates in fragrances can be inhaled, absorbed dermally, ingested, or transferred to house dust. Although phthalates are not expected to bioaccumulate and are not stored in our bodies for a long time, we do have repeated daily exposure to them through their presence in consumer products.

A 2013 analysis of phthalates in personal care products found that "leave on" products, such as lotions and face creams, delivered higher doses of phthalates than "rinse off" products, like shampoo. The analysis also found that DEP contributed the largest exposure of the 7 phthalates included in the study (Guo & Kannan, 2013a). (While they did not include fragrances in their study, the results may be applicable to fragrances as they are intended to be "leave-on" products and frequently contain higher concentrations of DEP than other types of personal care products.)

This estimation is supported by epidemiological studies of women of childbearing age. Parlett et al. (2013) found that women's use of fragrances was associated with an increase in the concentration of urinary phthalate metabolites. Similarly, Buckley et al. (2012) found that personal care product use was associated with phthalate metabolites in urine in 50 women age 18 to 38.

Perfume and cologne use was associated with higher levels of DEHP metabolites. This is particularly concerning because women of color have been documented to have higher phthalate levels from exposure attributable to fragrances in beauty products (Helm Nishioka, Brody, Rudel, & Dodson, 2018; Zota & Shamasunder, 2017). Additionally, women who normally tried to buy fragrance-free products had lower concentrations of DEHP metabolites in their urine.

Exposure to phthalates in fragrances is also an occupational concern. A 2017 study of cosmetics and perfume sales clerks found elevated levels of MEP, MBP and MEHP in urine after working shifts (Huang, Liao, Chang, Chan, & Lee, 2018).

Children and adolescents are exposed to phthalates in fragrances. A study of 100 Latina girls developed a community-led intervention to reduce exposure to phthalates from consumer products. By using products labeled as "phthalate-free," they were able to reduce MEP in urine by over 24% (Harley et al., 2016). This suggests that personal care products are a significant source of phthalate exposure for girls. Another study reports that urine phthalate metabolites in infants increase when they are exposed to infant care products that can contain fragrances, such as baby shampoos, baby lotions, and baby powder (Sathyanarayana et al., 2008).

Potential environmental exposure and exposure to sensitive species

Phthalates from fragrances contribute to environmental phthalate concentrations. Phthalates found in fragrances can migrate into various environmental media (Méndez-Díaz, Abel daiem, Rivera-Utrilla, Sanchez-Polo, & Bautista-Toledo, 2012; Wu, Mahmood, Wu, & Zheng, 2008). In the Puget Sound area, Commencement Bay and the Lower Duwamish Waterway are EPA superfund sites, partially due to phthalate contamination. Because phthalates are not

environmentally persistent, the concentrations observed at these sites are reflective of current release levels.

In 2010, a remedial investigation into phthalate levels in the Lower Duwamish Waterway found DEHP in surface sediment, surface water, fish and invertebrates samples and BBP and DMP in surface sediments (Lower Duwamish Waterway Group, 2010). A similar investigation into Commencement Bay showed decreasing concentrations of many contaminants between 1999 and 2008. However, during that same time period, concentrations of DEHP increased. DEHP was detected in all 30 sediment samples and BBP was detected in 20% of sediment samples (Ecology, 2010).

The City of Tacoma has been monitoring phthalates in stormwater sediment traps from 2001 to 2016. DEHP was ubiquitously detected in stormwater sediment traps (City of Tacoma, 2017). King County has conducted additional monitoring in the Duwamish Estuary and still detected DEHP in sediment, and concluded that juvenile Chinook salmon are exposed to phthalates from sediment in the estuary (King County, 2018).

In 2011, Ecology published the Puget Sound Toxics Loading Study,[35] which estimated environmental release of a number of toxic chemicals to the Puget Sound area. This report concludes that fragrances are the single largest source of releases and account for about one third (11 out of 34 tons) of total phthalate releases into the Puget Sound Basin. Sources included in this analysis were personal care products, PVC materials, air emissions, and other materials. We expanded this calculation from the Puget Sound region only to evaluate statewide releases, and estimate that 17 tons of phthalates are released into Washington's environment from fragrances.

Although discharge of phthalates from fragrances is primarily assumed to be through publicly-owned treatment works via sanitary sewers and septic systems (Ecology, 2011), there is the potential for partially-used fragrance containers to be disposed of in landfills. Phthalates are a major source of leachate contaminant from landfills, and are primarily emitted from consumer products and building materials (Ramakrishnan et al., 2015). Phthalates found in fragrances (e.g. DEP, DBP, BBP, DEHP) are consistently detected across landfill leachates (Paxeus, 2000).

The degradation of phthalates depends on many factors, including the length of the carbon chain for each phthalate and the surrounding pH, microbes/nutrients, and temperature (Huang, Nkrumah, Appiah-Sefah, 2013; Liu et al., 2010). Studies have also found that landfill leachate contaminated with phthalates can seep into surrounding groundwater (Liu et al., 2010). Phthalates can escape landfills and enter surrounding environmental media.

[35] https://fortress.wa.gov/ecy/publications/documents/1103010.pdf

Exposure to phthalates in the environment is a concern for sensitive species. Governor Inslee's Orca Task Force, a work group developing plans to recover Puget Sound's orca population, identified phthalates as a chemical of emerging concern.

Existing regulations

Currently, no federal regulations exist regarding phthalates in fragrances. In the U.S. there are a number of state regulations to limit exposures to phthalate chemicals. There are 31 pending legislative bills in 13 states and 30 adopted laws in 14 states pertaining to limits, bans, monitoring, or reporting of phthalate chemicals. Of the 30 existing laws, 20 ban phthalates in a variety of food containers, paper products, and children's products (Safer States, 2019). California is attempting to pass the Toxic-Free Cosmetics Act which would specifically ban phthalates in cosmetics, including fragrances (CA Legislature, 2019; Safer States, 2019).

Federal policies (including the federal Consumer Product Safety Improvement Act (CPSIA)) focus on the removal of phthalates from children's toys (CPSC, 2019; Safer States, 2019). EPA is evaluating five phthalates under the revised Toxic Substances Control Act (TSCA): DBP, BBP, DEHP, DIBP, and dicyclohexyl phthalate, and slated others for further assessment (EPA, 2019). Additionally, manufacturers requested that EPA conduct risk evaluations for DIDP and DINP under TSCA Section 6. EPA granted both of these manufacturer requests.

The European Chemicals Agency maintains a list of 10 phthalates on their Candidate List of substances of Very High Concern published under the REACH Regulation (European Chemicals Agency, 2019). Canada banned the use of DEHP in cosmetics and restricted it (as well as DIDP, DINP, DNOP, DBP, and BBP) in children's toys and some child care products (Canadian Health Services, 2019). Two phthalates, dibutyl phthalate (DBP), and Bis(2-ethylhexyl) phthalate (DEHP) are banned in cosmetics sold in the European Union (CosIng, 2019).

Availability of safer alternatives

There are a number of solvents and fixatives used in cosmetic fragrances in addition to phthalates, such as dipropylene glycol and triethyl citrate. Many fragrances are also dissolved in water and ethanol, though these are not compatible with all compounds. In addition, cosmetics could be formulated to be fragrance-free to avoid the use of solvents. Several alternatives have been investigated by other agencies, such as Northwest Green Chemistry (Northwest Green Chemistry, 2018). As part of our Safer Products for Washington process, we will address the safety and feasibility of these alternatives in Phase 3.

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