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Ecological Risk Assessment of Flame Retardants in WA State

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Introduction

In the EPA framework, ecological risk assessment consists of four major components: problem formulation, exposure assessment, toxicity assessment, and risk characterization (EPA, 1997, 1998). Problem formulation states the purpose of the assessment, defines the problem, and describes the plan for analyzing and characterizing risk. While exposure assessment evaluates environmental data to assess potential exposure to receptors, toxicity assessment reviews ecotoxicology literature to describe the stressor-response profile (e.g., identify concentration or dose threshold where adverse effects may occur). Risk characterization integrates exposure and effects data to estimate ecological risk. Uncertainties are also summarized in risk characterization. Finally, ecological risk assessments are often non-linear and iterative (e.g., lessons learned are incorporated or new data become available for subsequent iterations).

Similar to human health risk assessments, ecological risk assessments can be performed deterministically or probabilistically (EPA, 2001). In a probabilistic analysis, one or more variables (exposure or effects) are defined as a probability distribution, rather than a single point estimate. As a result, the output of a probabilistic analysis is a probability distribution of risk rather than a single number. While probabilistic assessments provide more information and may better characterize uncertainties, deterministic assessments are more easily performed and more suitable when exposure and effects data are limited, as is the case here.

In this analysis, the four components of ecological risk assessment will be briefly reviewed, results for aquatic and terrestrial receptors will be summarized, and conclusions will be presented.

Methods

A) Problem Formulation

The purpose of this assessment is to evaluate six flame retardants, identified in HB-2545 (Table 1), in terms of ecological effects on aquatic and terrestrial receptors. Flame retardants included two non-halogenated/phosphorus-containing chemicals (TPP, IPTPP), two halogenated (chlorine)/phosphorus-containing chemicals (TCPP, V6), and two halogenated (bromine) chemicals without phosphorus (TBPH, TBB). It is uncertain if environmental levels of these flame retardants adversely impact ecological receptors in WA. As such, environmental data (collected in WA or estimated via modeling), together with ecotoxicology data (obtained from several regulatory authorities), were used to estimate ecological risk in this analysis.

B) Exposure Assessment

Environmental data on the six flame retardants in WA state were compiled. Environmental media included stormwater and stormwater sediment in Clark County (Medlen, 2018), surface water, sediment, and fish tissue in 10 WA lakes (Mathieu, in prep), Columbia River surface water (Alvarez et al, 2014), and Columbia River sediment (Counihan et al, 2014). Not all flame retardants were measured in all media (Table 2). For example, only TBB was analyzed in fish tissue, and no IPTPP data were available in any media. Notably, soils were not sampled for these six flame retardants in WA.

Environmental data may be used for exposure assessment. For ecological receptors inhabiting environmental media (e.g., plants, soil invertebrates, aquatic invertebrates, fish), an exposure point concentration (EPC) is estimated for a specific environmental medium (e.g., mg/L water). If data are randomly collected, a 95% upper confidence limit (95UCL) of the arithmetic mean is typically used to estimate EPC. Alternatively, without probability-based sampling, EPC typically defaults to the sample maximum.

For surface water and sediment, stormwater and stormwater sediment concentrations of flame retardants were generally higher than corresponding river or lake concentrations in WA. With the exception of TBB in sediment, stormwater and stormwater sediment were conservatively used in this analysis to estimate flame retardant EPC in surface water and sediment, respectively. When environmental media are not sampled, contaminant concentrations may be modeled. This was the case for TCPP and V6 in soils in this analysis, where a predicted environmental concentration (PEC) was estimated with the EUSES model (EU, 2008a,b; EUSES, 2018). PEC is essentially equivalent to EPC. TOXNET/HSDB (2018) was also searched for soil concentration data on the six flame retardants.

For higher trophic level wildlife receptors (e.g., birds, mammals), exposure is often determined via ingested dose of food and environmental media (mg/kg BW/d). This exposure has been referred to as average daily dose (ADD) (EPA, 1998) or total daily intake (TDI) (EC, 2016a). For example, TDI for TBB has been modeled for piscivorous mammals (e.g., mink, river otter) via dietary exposure (EC, 2016a). This calculation incorporates information on TBB concentration in water, food ingestion rate (FIR), and bioaccumulation factor (BAF) for TBB in prey fish.

C) Toxicity Assessment

Ecotoxicology data may be used for toxicity assessment. Toxicity reference values (TRVs) are specified in terms of concentration or dose for a variety of ecological effects for both acute and chronic scenarios. A stated goal of ecological risk assessment is to protect local populations and communities of plants and animals (EPA, 1999). To achieve this objective, preferred toxicity endpoints are growth, reproduction, and survival (EPA, 1997). These effects are considered ecologically relevant, because they can impact higher levels of ecological organization (e.g., populations). Ideally, these endpoints are conservatively selected, from among the most sensitive species. For our use, preferred ecotoxicology data are from an authoritative scientific and regulatory source (e.g., EPA, ECHA). Data from these sources typically represent a weight of evidence and consensus view.

In terms of concentration (e.g., mg/L water), a no effects concentration (NOEC) or low effects concentration (LOEC) is preferred for ecological risk assessment for receptors who inhabit environmental media (e.g., plants, soil invertebrates, aquatic invertebrates, fish). The European Union calculates a predicted no effects concentration (PNEC) with use of an application factor (AF) to account for uncertainty in the quality and quantity of the toxicity data set (ECHA, 2018a). AF may range from 1-1000. Extrapolations between media are sometimes performed with equilibrium partitioning. For example, equilibrium partitioning was used to derive a sediment PNEC for TCPP and V6 from a water

PNEC (EU, 2008a,b), as well as a soil PNEC for V6 from a water PNEC (EU, 2008b). Wet wt to dry wt conversions for derived sediment PNECs were performed, according to EU (2008c). In terms of dose (mg/kg BW/d), a daily no effects dose from food and environmental media is often calculated in ecological risk assessment for upper trophic level receptors (e.g., birds, mammals).

For aquatic biota (e.g., aquatic invertebrates, fish), EPA/DFE (2015) has compiled acute and chronic toxicity data on several flame retardants. EPA/DFE (2015) also presents results from ECOSAR modeling which uses quantitative structure-activity relationships (QSARs) to predict toxicity for chemicals without toxicity data from structurally similar chemicals with toxicity data. ECOSAR predicts acute and chronic toxicity endpoints for 111 organic chemical classes for freshwater algae, invertebrates, and fish. For a neutral organic chemical class, comprised of lipophilic chemicals, ECOSAR predicts no effects at saturation (NES) when the estimated chronic toxicity value exceeds the water solubility limit or when log Kow>8.0 (e.g., TBPH, TBB). In these cases, dietary uptake may be more relevant than limited uptake across respiratory surfaces (e.g., gills) and/or dermal absorption. However, bioaccumulation of TBPH and TBB appears limited, due to metabolic biotransformation, at least more so for TBB than TBPH (EC, 2016a).

For terrestrial biota (e.g., plants, soil invertebrates, birds, mammals), comparatively less toxicity data on flame retardants were available, relative to aquatic biota. Two databases were searched for terrestrial ecotoxicology data on the six flame retardants. In particular, EPA/ECOTOX (2018) was searched by CAS number, while ORNL/RAIS (2018) was searched by chemical name. For example, ECOTOX listed a dietary dose no observed effect level (NOEL) for TCPP in the American kestrel (Fernie et al, 2015). In addition to these searches, several reports were identified which contain relevant data (e.g., EC, 2016a,b; ECHA, 2013, 2018b; EU, 2008a,b).

D) Risk Characterization

Risk characterization refers to comparison of exposure and effects data. When a deterministic analysis is performed, this can be accomplished with a simple ratio, termed a hazard quotient (HQ). HQ can be expressed in terms of concentration in environmental media (e.g., for plants, soil invertebrates, aquatic invertebrates, fish) or daily dose (e.g., for birds, mammals). Exposure (numerator) and effects (denominator) are expressed as point estimates in the same units, so HQ is a point estimate and unitless. When HQ<1, adverse effects are unlikely, whereas when HQ>1, effects are possible (Table 3). HQ was conservatively calculated with highest exposure (EPC, PEC, or TDI) and lowest (most sensitive) effect (PNEC or TRV) estimates. Cumulative effects can be assessed with hazard index (HI) by summing HQ over chemicals with similar toxic mechanisms ($HI = \Sigma HQ$). Cumulative effects were not assessed in this analysis, since all individual HQs were conservatively calculated and <0.2.

Uncertainty in exposure and effects data included incomplete or nonrandom sampling for exposure data, gaps in effects data from authoritative scientific and regulatory sources, and uncertainties associated with modeling. For example, nonrandom sampling methods preclude generalization to the underlying statistical population of contaminant data, so 95UCL could not be used to represent EPC. Relative to aquatic receptors, few effects data were available for terrestrial receptors. With respect to modeling exposure (e.g., EUSES model, dietary dose) and effects (e.g., ECOSAR, equilibrium partitioning), model uncertainty (conceptual basis) and parameter uncertainty (model inputs) also contribute uncertainty in this analysis (NCRP, 1996).

Results and Discussion

A) Freshwater Aquatic Biota (water, sediment, and fish tissue)

Results for aquatic biota (e.g., aquatic invertebrates, fish), based on concentrations of flame retardants in water, show that effects are unlikely (HQ<1) for all flame retardants, except IPTPP, for which there were no data (Table 4). For EPC, the maximum concentrations of flame retardants in stormwater were used (Medlen, 2018), since stormwater concentrations were higher than corresponding lake and river concentrations in WA data (Mathieu, in prep; Alvarez et al, 2014). The reporting limit (RL) was used for TBPH and TBB, since these data were nondetect. For ecotoxicology data, acute and chronic bioassay data were used with an appropriate AF to estimate PNEC (ECHA, 2018a). Although ECOSAR predicted NES for TBPH and TBB, suitable bioassay data for these flame retardants is available from another source (EC, 2016a). Finally, it should be noted that although EPA/DFE (2015) classified TPP and IPTPP aquatic toxicity as "very high," a relatively low EPC for TPP resulted in HQ<1 and no EPC for IPTPP precluded calculation of HQ.

Results for benthic biota, based on concentrations of flame retardants in sediment, show that effects are unlikely (HQ<1) for TCPP and V6 (Table 5). Again, no relevant sediment data were available for IPTPP. For EPC, maximum concentrations of flame retardants in stormwater sediment were identified (Medlen, 2018), since stormwater sediment concentrations (except for TBB) were higher than corresponding lake and river sediment concentrations in WA data (Mathieu, in prep; Counihan et al, 2014). Both TBPH and TBB were nondetect in sediment. For TCPP and V6, a sediment PNEC was predicted from a water PNEC with equilibrium partitioning methods (EU, 2008a,b). For other flame retardants, however, no ecotoxicology data for sediments could be located.

Fish tissue (fillet) was analyzed for TBB but was not detected at the RL (Table 6). Because no ecotoxicology data were identified for TBB in fish tissue (e.g., ERED, 2018), HQ could not be calculated. Furthermore, with the exception of TBB, no fish tissue data from WA state were located for other flame retardants in our analysis.

B) Terrestrial Biota (soil and dietary dose)

Results for terrestrial biota (e.g., plants, soil invertebrates, birds, mammals), based on modeled concentrations of TCPP and V6 in soil, show that effects are unlikely (HQ<1) (Table 7). However, data for remaining flame retardants were incomplete, precluding calculation of HQ. In particular, the EUSES model (EU, 2008a,b) was used to estimate a soil PEC for TCPP and V6, because soil measurements were unavailable for all six flame retardants. Soil PNEC estimates for TCPP and IPTPP were based on chronic bioassays (EU, 2008a; ECHA, 2018b), while a soil PNEC for V6 was derived via equilibrium partitioning (EU, 2008b). Although soil PNEC estimates were identified for TPP and IPTPP (ECHA, 2013, 2018b), no exposure data were available for these flame retardants. Finally, neither exposure nor effects data could be located for TBPH and TBB in soils.

Results from a dietary dose analysis of TBB for piscivorous mammals (EC, 2016a) showed that effects were unlikely (HQ<1) (Table 8). TDI was modeled for both mink and river otter, considering TBB concentration in water, food ingestion rate (FIR), and a bioaccumulation factor (BAF) for TBB in prey fish. HQ results are conservative, given that the input nondetect TBB concentration in water was the RL (50 ng/L). Dose-based TRVs for the mink and river otter were derived from a two generational rat study on reproduction (EC, 2016a).

In another study, a dietary dose NOEL (based on weight change) was estimated for TCPP for the American kestrel (Table 8). However, no corresponding TDI data were identified, precluding calculation of HQ. Among the six flame retardants, therefore, only TBB data were available to perform a dose-based analysis.

Conclusions

For the six flame retardants specified in HB-2545, there are limited environmental data in WA (chemical concentrations in stormwater and stormwater sediment, river and lake surface water and sediment, and fish tissue), as well as limited ecotoxicology data (acute and chronic toxicity information). These data are used to describe exposure (e.g., EPC, PEC, TDI) and effects (e.g., PNEC, TRV), respectively, in order to estimate ecological risk with HQ methodology. Because soil was not sampled in WA and sediment bioassay data were unavailable for the flame retardants in our analysis, these data gaps were partly filled with modeling methods (e.g., soil PEC estimated with the EUSES model, soil and sediment PNEC estimated with equilibrium partitioning).

Based on the most conservative environmental and ecotoxicology data identified, aquatic populations and communities in WA (e.g., aquatic invertebrates, fish) appear unlikely to be adversely impacted. However, effects are more uncertain for terrestrial populations and communities in WA (e.g., plants, soil invertebrates, birds and mammals), primarily due to sparse data. More extensive environmental and ecotoxicology data are needed to more fully characterize ecological risk to flame retardants in WA.

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Table 1. Flame retardants in HB-2545 listed for evaluation.

CAS Number	**Acronym**	**Chemical Name**
115-86-6	TPP	Triphenyl phosphate
13674-84-5	TCPP	Tris (1-chloro-2-propyl) phosphate
26040-51-7	TBPH	Bis (2-ethylhexyl) tetrabromophthalate
38051-10-4	V6	Bis (chloromethyl) propane-1,3-diyl tetrakis-(2-chloroethyl) bis(phosphate)
68937-41-7	IPTPP	Isopropylated triphenyl phosphate
183658-27-7	TBB	2-ethylhexyl-2,3,4,5-tetrabromobenzoate

Table 2. Environmental media sampled and flame retardants measured in WA state for this analysis.

Flame Retardant	**Stormwater surface water**	**Stormwater sediment**	**Lake surface water**	**Lake sediment**	**Fish tissue**	**Columbia River surface water**	**Columbia River sediment**
	(Medlen, 2018)	(Medlen, 2018)	(Mathieu, in prep)	(Mathieu, in prep)	(Mathieu, in prep)	Alvaraz et al (2014)	Counihan et al (2014)
TPP	X	X	X	X		X	X
TCPP	X	X	X	X		X	X
TBPH	X	X		X			
V6	X	X	X	X			
IPTPP							
TBB	X	X		X	X		

X=analyte measured in sampled media.
Notes: soil not sampled, IPTPP not measured in any media.

Table 3. Hazard quotient (HQ) description and interpretation.

HQ estimate	**Concentration-based HQ**	**Dose-based HQ**
	(EPC/PNEC)	(TDI/TRV)
HQ<1	effects unlikely	effects unlikely
HQ>1	effects possible	effects possible

Concentration-based HQ applicable to receptors inhabiting environmental media (e.g., plants, soil invertebrates, aquatic invertebrates, fish). Dose-based HQ applicable to higher trophic level receptors (e.g., birds and mammals).

Table 4. Results for aquatic biota-freshwater.

Flame Retardant	EPC Summary	EPC	Ecotox Data (EPA/DfE, 2015)	Ecotox Conc	AF	PNEC=Ecotox Conc/AF	HQ=EPC/PNEC
TPP	max, Clark County, WA, stormwater outfall, 2017, Medlen (2018)	83 ng/L	30 d LOEC (chronic), rainbow trout, experimental, ECHA (2013), DFE class-very high tox	37000 ng/L	50	740 ng/L	0.11
TCPP	max, Clark County, WA, stormwater outfall, 2017, Medlen (2018)	857 ng/L ("J" qualified)	static 96 hr LC50 (acute), fathead minnow, experimental, EC (2016b), DFE class-moderate tox	51000000 ng/L	30	1700000 ng/L	0.00050
TBPH	max, Clark County, WA, stormwater outfall, 2017, Medlen (2018)	<50 ng/L (nondetect at RL)	ECOSAR predicts NES due to low water solubility (2E-9 mg/L) and high log Kow (12), DFE class-low tox, dietary uptake may be relevant (EC, 2016a)	79000 ng/L 15 d LC50 (acute), Daphnia carinata, experimental, EC (2016a)	100	790 ng/L	0.063

Table 4. Results for aquatic biota-freshwater (continued).

Flame Retardant	EPC Summary	EPC	Ecotox Data (EPA/DfE, 2015)	Ecotox Conc	AF	PNEC=Ecotox Conc/AF	HQ=EPC/PNEC
V6	max, Clark County, WA, stormwater outfall, 2017, Medlen (2018)	10 ng/L	23 d NOEC (chronic), reproduction, Daphnia magna, experimental, EU (2008b), DFE class-moderate tox	3680000 ng/L	50	73600 ng/L	0.00014
IPTPP	No data		No tox data on IPTPP (only mixture and surrogate data), DFE class-very high tox				
TBB	max, Clark County, WA, stormwater outfall, 2017, Medlen (2018)	<50 ng/L (nondetect at RL)	ECOSAR predicts NES due to low water solubility (1.1E-5 mg/L) and high log Kow (8.8), DFE class-low tox, dietary uptake may be relevant (EC, 2016a)	79000 ng/L 15 d LC50 (acute), Daphnia carinata, experimental, EC (2016a)	100	790 ng/L	0.063

Table 5. Results for aquatic biota-freshwater sediment.

Flame Retardant	**EPC Summary**	**EPC**	**Ecotox Data**	**PNEC**	**HQ=EPC/PNEC**
TPP	max, Clark County, WA, stormwater sediment, 0-12 cm, 2017, Medlen (2018)	36.1 ng/g dw	No data		
TCPP	max, Clark County, WA, stormwater sediment, 0-12 cm, 2017, Medlen (2018)	2040 ng/g dw ("E" qualified)	Equilibrium partitioning method, K[susp/water]=5.25 m3/m3, susp matter density=1150 g/L, PNEC[water]=0.64 mg/L, EU (2008a), sediment biota	PNEC[sed]=2920 ng/g ww (13270 ng/g dw) ww to dw (EU, 2008c)	0.15
TBPH	max, Clark County, WA, stormwater sediment, 0-12 cm, 2017, Medlen (2018)	<43 ng/g dw (nondetect at RL)	No data		

Table 5. Results for aquatic biota-fw sediment (continued).

Flame Retardant	EPC Summary	EPC	Ecotox Data	PNEC	HQ=EPC/PNEC
V6	max, Clark County, WA, stormwater sediment, 0-12 cm, 2017, Medlen (2018)	7.12 ng/g dw	Equilibrium partitioning method, K[susp/water]=7.03 m3/m3, susp matter density=1150 g/L, PNEC[water]=0.0736 mg/L, EU (2008b), sediment biota	PNEC[sed]=455 ng/g ww (2068 ng/g dw) ww to dw (EU, 2008c)	0.0034
IPTPP	No data		No data		
TBB	max, WA lake sediment, 0-2 cm, 2018, Mathieu (in prep)	<25 ng/g dw (nondetect at RL)	No data		

Table 6. Results for aquatic biota-fw fish tissue.

Flame Retardant	EPC Summary	EPC	Ecotox Data	PNEC	HQ=EPC/PNEC
TBB	max, bass, sucker, pikeminnow, fillet, WA lake, 2017, Mathieu (in prep)	<3 ng/g ww (nondetect at RL)	No data		
All other flame retardants in this analysis	No data for WA fish		No data		

Table 7. Results for terrestrial biota-soil.

Flame Retardant	PEC Summary	PEC	Ecotox Summary	Ecotox Conc	AF	PNEC=Ecotox Conc/AF	HQ=PEC/PNEC
TPP	No data		ECHA (2013)			0.218 mg/kg soil dw	
TCPP	PEC regional soil, EUSES model, EU (2008a)	0.00265 mg/kg soil ww	NOEC (chronic), lettuce seedling emergence/growth, experimental, EU (2008a)	17 mg/kg soil dw	10	1.7 mg/kg soil dw (1.5 mg/kg soil ww)	0.0018
TBPH	No data		No data				

Table 7. Results for terrestrial biota-soil (continued).

Flame Retardant	PEC Summary	PEC	Ecotox Summary	Ecotox Conc	AF	PNEC=Ecotox Conc/AF	HQ=PEC/PNEC
V6	PEC regional soil, EUSES model, EU (2008b)	0.0000635 mg/kg soil ww	Equilibrium partitioning method, K[soil/water]=7.55 m3/m3, soil density=1700 g/L, PNEC[water]=0.0736 mg/L, EU (2008b)			PNEC[soil]=0.327 mg/kg soil ww	0.00019
IPTPP	No data		NOEC (chronic), earthworm reproduction, experimental, ECHA (2018b)	250 mg/kg soil dw	50	5 mg/kg soil dw	
TBB	No data		No data				

Table 8. Results for terrestrial wildlife-dose.

Flame Retardant	TDI Summary	TDI	Ecotox Summary	AF	TRV	HQ=TDI/TRV
TBB	mink (piscivorous), BAF=8446 L/kg, FIR-0.22 g/g BW/d, dietary dose model, EC (2016a) water EPC=RL=50 ng/L, nondetect, Medlen (2018)	0.093 mg/kg BW/d	23 mg/kg BW/d, derived from a rat study on reduction in birth wt of second generation pups, EC (2016a)	10	2.3 mg/kg BW/d	0.040
TBB	river otter (piscivorous), BAF=8446 L/kg, FIR=0.16 g/g BW/d, dietary dose model, EC (2016a) water EPC=RL=50 ng/L, nondetect, Medlen (2018)	0.068 mg/kg BW/d	14 mg/kg BW/d, derived from a rat study on reduction in birth wt of second generation pups, EC (2016a)	10	1.4 mg/kg BW/d	0.049
TCPP	No data		22 ng/g BW/d, 21 d NOEL, weight, American kestrel, Fernie et al (2015)			