# Potential Effects of PBDEs on Puget Sound and Southern Resident Killer Whales

## A Report on the Technical Workgroups and Policy Forum

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*The Environmental Protection Agency (EPA) Region 10 and the National Marine Fisheries Service (NMFS) Northwest Region are concerned about the potential effects of polybrominated diphenyl ethers (PBDEs) on Puget Sound and Southern Resident killer whales. In coordination with NMFS, EPA Region 10's Office of Water and Watersheds hosted a series of technical workgroups during spring 2013 on the following topics:*

- *PBDE removal efficiency in wastewater treatment plants*
- *PBDE modeling in Puget Sound (fate, transport, and bioaccumulation)*
- *The need to establish a PBDE toxicological threshold for Southern Resident killer whales*
- *No Observed Effect Concentration (NOEC) levels of mixtures of persistent organic pollutants (including PCB and PBDE congeners)*

*Subsequent to the technical workgroups, EPA Region 10 hosted a policy forum on PBDEs and Southern Resident killer whales on June 6th, 2013. The policy forum was the culmination of technical workgroups and provided an opportunity for senior level staff at the Washington State Department of Ecology (Ecology), NMFS, the Puget Sound Partnership, and EPA to learn about and discuss the issues surrounding PBDEs in Puget Sound, especially as they relate to killer whales. An impressive group of people participated in the workgroups - from national PBDE experts to local wastewater treatment plant operators (King County and LOTT Clean Water Alliance) to key staff from Ecology, NMFS, EPA, as well as Canadian researchers. See the agendas and participant lists for more details (Appendices A and B).*

*This report provides a summary of the lessons learned from each workgroup and is a first step in addressing emerging contaminants like PBDEs. This report also provides a brief summary of the priority actions that will be implemented following the discussions from the policy forum. EPA Region 10 and NMFS Northwest Region are very appreciative of the time and expertise that workgroup members shared during this process.*

## Introduction to PBDEs

PBDEs are a class of flame retardants called polybrominated diphenyl ethers. PBDEs have been used as flame retardants since the 1970s in products including fabrics, plastics, polyurethane foam, wire insulation, cushions, mattresses, and cars. Since PBDEs are not chemically bound to products, they are more likely to leach out into the environment. PBDEs are a complex mixture of 209 congeners that vary by the number of bromine atoms and location of the molecule. Certain PBDE congeners are persistent, bioaccumulative, and toxic to both humans and wildlife. They are endocrine disruptors and the critical endpoint of concern for human health is neurobehavioral effects.

Phase-out regulations[1] will prevent new PBDEs from being produced, which will help protect the environment from further contamination. However, PBDEs cannot be fully eliminated from the environment because they will continue to be released from in-use products.

[1] In January, 2006, Ecology developed a final Chemical Action Plan (CAP) for PBDEs: https://fortress.wa.gov/ecy/publications/summarypages/0507048.html

## PBDE Pathways to Puget Sound

According to Ecology and King County's *Puget Sound Toxic Loadings Analysis*[2]:

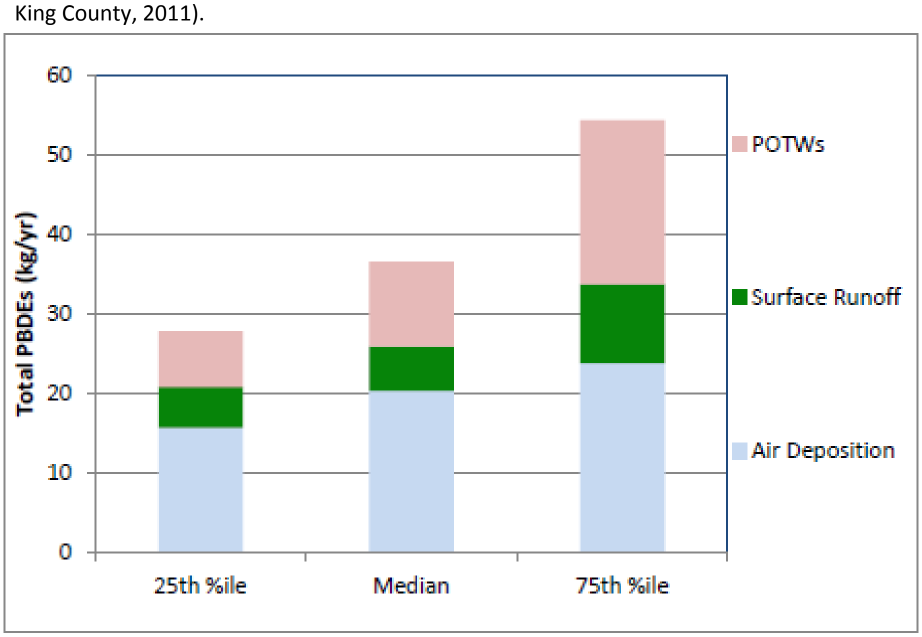
- PBDEs are released from indoor consumer and office products, become attached to dust particles, and are subsequently delivered to the sanitary sewer through washing machine rinse water during the washing of fabrics with the attached PBDE-enriched dust, and rinsing other materials with attached dust particles.

- PBDEs are ubiquitous environmental contaminants, and although voluntary actions and bans have removed major PBDE formulations from new consumer products, much of the PBDEs produced historically may remain in consumer products and commercial office products and these potentially represent substantial diffuse ongoing sources.

- The conceptual model of PBDE transport and fate following release suggests that much of the PBDEs will be initially released to air and atmospheric transport will deliver comparatively high loads directly to Puget Sound.

- PBDEs deposited on land will also be mobilized during storm events and delivered to surface waters, but in quantities lower than for direct atmospheric deposition. Some of the PBDEs deposited to land are also likely to be transported in storm sewers and delivered directly to Puget Sound or indirectly by way of wastewater treatment plants.

Fig. 1 Total PBDE Loads to Puget Sound as the Sum of Major Delivery Pathways (Ecology and King County, 2011).




[2] Ecology and King County, 2011. Control of Toxic Chemicals in Puget Sound: Assessment of Selected Toxic Chemicals in the Puget Sound Basin, 2007-2011. Washington State Department of Ecology, Olympia, WA and King County Department of Natural Resources, Seattle, WA. Ecology Publication No. 11-03-055. www.ecy.wa.gov/biblio/1103055.html

- Atmospheric deposition accounts for the largest pathway (44-56%), followed by Publicly Owned Treatment Works (POTWs) (25-38%), and surface runoff (18%).

- No PBDE loading data are available for groundwater.

- There appears to be a net export of 11kg total PBDEs at the ocean boundary.

## PBDEs and Killer Whales in Puget Sound

The Southern Resident killer whales are listed as endangered under the U.S. Endangered Species Act (ESA). ESA section 7 consultations occur for any federal action that may affect the killer whales, such as permits, research, etc. Some federal actions include an increase in persistent pollutants, such as PBDEs, into the whales' critical habitat and there is potential for bioaccumulation in the whales.

The Southern Resident killer whales carry high concentrations of persistent pollutants, making them by far the most contaminated resident killer whale community in the northeastern Pacific. PBDEs are considered to be an emerging threat to the Southern Residents because they are endocrine disruptors that can affect thyroid hormone levels, they may cause subtle neurobehavioral effects, mimic or offset reproductive processes, and alter immune response in many species.

Although it is not clear if PBDEs levels in the Southern Residents are at or near a health-effects threshold, their body burdens are above PBDE concentrations associated with altered thyroid hormones in grey seals. Based on known mixture effects between PBDEs and PCBs, it is reasonable to assume the whales' are susceptible to synergistic effects between these two pollutants, such that the whales' current levels of both may be sufficient to produce adverse health effects.

## Workgroup 1: PBDE Removal Effectiveness in Wastewater Treatment Plants

This workgroup included experts from the EPA, NMFS, Ecology, US Geological Survey (USGS), King County, LOTT Clean Water Alliance, and nationally renowned experts in PBDEs and other endocrine disruptors. See Appendix A for agenda and participant list.

Since wastewater treatment plants (WWTPs) are not designed to treat PBDEs, their presence can present a challenge for wastewater engineers. PBDEs have low water solubility, and are very hydrophobic and lipophilic.

*PBDE Removal Efficiency via Wastewater Treatment*

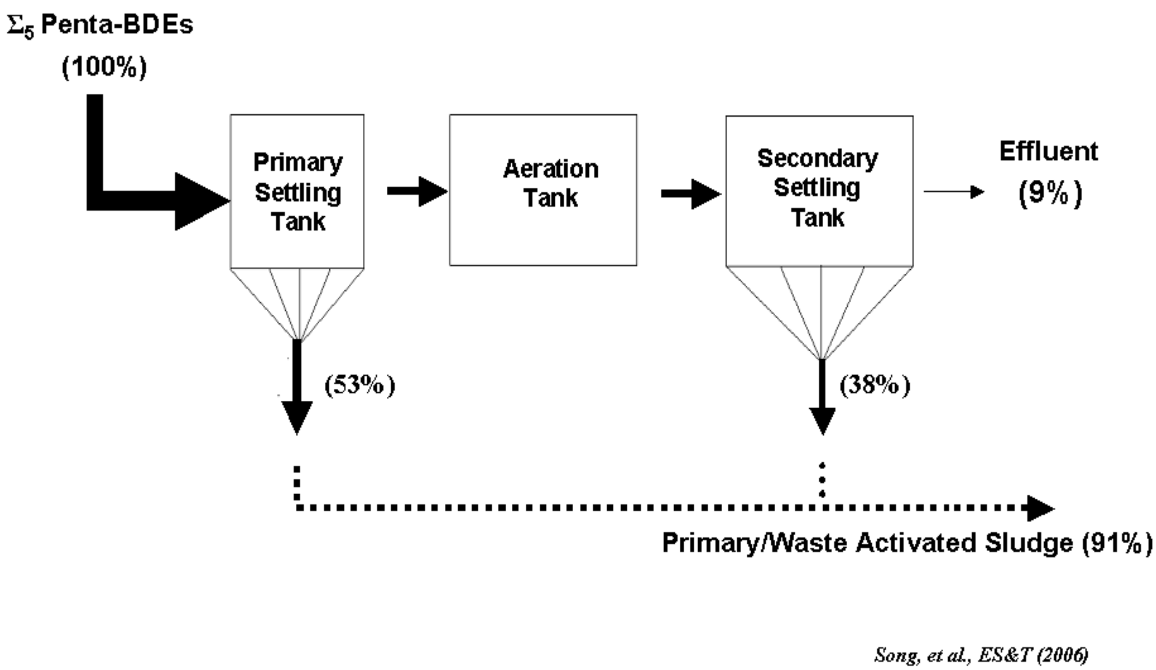
**There is general agreement that sorption to solids is the primary removal mechanism of PBDEs in wastewater.**

- PBDEs are generally not removed via biodegradation or volatilization during the wastewater treatment process.

- A 2013 Canadian study[3] "…determined PBDE levels in influent, primary effluent, and final effluent collected from diverse treatment processes including four aerated lagoons, two facultative lagoons, four primary treatments, eight secondary biological treatments and two advanced treatments. Parameters examined for correlation included seasonal temperature, community sizes, industrial inputs, and operational conditions. … Median removal efficiencies for all process types exceeded 90% except primary treatment at 70%. PBDE levels and removals were correlated to the levels and removals of conventional parameters that represent wastewater strength, such as chemical oxygen demand and total suspended solids. The role of the primary clarifier was significant (82% removal) and removal was associated with hydraulic retention time (HRT) and surface loading rate. Best removal of PBDEs was achieved at greater than 2000 mg/L mixed liquor suspended solids (MLSS), longer than 10 h of HRT, and 9 days of solids retention time."

- Other researchers estimate 91% PBDE removal by activated sludge treatment, leaving 9% in the effluent[4]. See Figure 2 below.

Figure 2. PBDE Removal efficiency by activated sludge treatment (Song et al., 2006).




[3] Kim, M, P Guerra, M. Theocharides, K. Barclay, S.A. Smyth and M. Alaee, 2013. "Parameters affecting the occurrence and removal of polybrominated diphenyl ethers in twenty Canadian wastewater treatment plants." *Water Research*, 47, 2213-2221.

[4] Song, M., S. Chu, R. J. Letcher, and R. Seth. 2006 Fate, partitioning, and mass loading of polybrominated diphenyl ethers (PBDEs) during the treatment processing of municipal sewage. *Environmental Science and Technology*, 40, 6241-6246.

*Degrading/Debrominating PBDEs*

- Ultraviolet radiation causes some debromination of PBDEs, but most of these studies have focused on solids, not water.

*Biosolids*

- 85-95% of the PBDEs in wastewater are transferred to biosolids produced during wastewater treatment[5].
- The main environmental risk of PBDE contamination after wastewater treatment is via sludge-disposal routes[6].
- Since PBDEs concentrate in the solids stream, PBDEs may concentrate in sewage sludge (biosolids), which is increasingly being applied as a soil amendment[7]. Disposal or reuse of biosolids generated at treatment plants is a potential pathway for the release of PBDEs into the environment.

*Combined Sewer Overflows (CSOs)*

- Large amounts of PBDEs can be discharged from wastewater treatment plants during peak CSO storm events. Stormwater and discharges from WWTPs with periodic "pass through" could contribute disproportionately to the release of PBDEs.

*Workgroup Findings*

- Reducing particulates/Total Suspended Solids (TSS) removes PBDEs from the effluent.
- Higher sludge age provides for greater PBDE removal[8]. The longer the sludge is held in residence time, the better for sorbing out hydrophobic contaminants like PBDEs.
- Release may or may not be proportional to WWTP capacity. PBDE concentrations in the influent of some WWTPs may be much greater than others due to contributions from certain industries (e.g. textile, plastics, electronic/vehicular salvage, landfill leachate operations). Thus, some WWTPs may discharge greater quantities of PBDEs even if removal rates remain constant.

*Individual Recommendations from the PBDE Removal Effectiveness Workgroup*

- Optimize existing technologies to improve contact time between liquids and solids. Maximize the entrainment of solids to reduce the amount of suspended material being discharged.
- Encourage advanced treatment to remove suspended solids from effluent. (Membrane Bioreactors (MBR) or tertiary treatment involving filtration can reduce TSS to less than 2 mg/l).
- Educate the public about PBDE sources and pathways, as well as ways to reduce loading.

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[5] Arnold, R.G., S. Teske, M. Tomanek, J. Engstrom, C. Leung, J. Zhang, Q. Banhani, D. Quanrud, W.P. Ela and A.E. Saez, 2008. "Fate of Polybrominated Diphenyl Ethers during Wastewater Treatment/Polishing and Sludge Stabilization/Disposal." *New York Academy of Sciences Annals*, 1140, 394-411.

[6] Langford, K., M. Scrimshaw and J. Lester. 2007. "The Impact of Process Variables on the Removal of PBDEs and NPEOs During Simulated Activated Sludge Treatment." *Environmental Contamination and Toxicology*, 53, 1-7.

[7] La Guardia, M.J., R.C. Hale, E. Harvey and D. Chen. 2010. "Flame-Retardants and Other Organohalogens Detected in Sewage Sludge by Electron Capture Negative Ion Mass Spectrometry." *Environmental Science & Technology*, 44(12), 4658-4664.

[8] Langford, K., M. Scrimshaw and J. Lester. 2007. "The Impact of Process Variables on the Removal of PBDEs and NPEOs During Simulated Activated Sludge Treatment." *Environmental Contamination and Toxicology*, 53, 1-7.

- New replacements for PBDEs are also potentially hazardous (e.g. HBCD). Some of these alternative flame retardants are showing up in biota.
- Source control- Since brominated flame retardants are so persistent, reducing or eliminating their use is the most effective action to reduce their presence in our wastewater.
- Green stormwater infrastructure has the potential to reduce the input of PBDEs to WWTPs in combined sewer conveyance systems.
- Work to reduce/eliminate pass through events, storm water releases, and operational upsets.

*Monitoring*

- Consider alternative brominated and non-brominated flame retardants in future monitoring and discussions.
- Be as inclusive as possible in terms of the PBDE congener list examined via monitoring. Including less abundant congeners (particularly the octa- and nona- brominated) in a high concentration media like biosolids may provide evidence of degradation.
- As PBDEs strongly associate with particulates, review monitoring data on the release of suspended solids from WWTPs.
- Because PBDEs are hydrophobic and tend to composite in sewage sludges, monitoring schemes could include flame retardant concentrations in WWTP sewage sludges.

## Workgroup 2: PBDE Modeling in Puget Sound

This workgroup included experts from the EPA, NMFS, Ecology, the University of Washington, and Canadian universities. See Appendix A for agenda and participant list.

*Greg Pelletier (Ecology) presented on Ecology's PBDE Model for Puget Sound*

- Ecology is working to develop computer models to predict concentrations of toxic chemicals in water, sediment, and biota of Puget Sound and how they respond to changes in loading.
- Load estimates are from 2007 and are an initial estimate of the loading of selected toxic chemicals from all sources in the watersheds and atmospheric deposition.
- Greg Pelletier developed a Puget Sound Regional Toxics Model in 2009.
- PBDEs behave similarly to PCBs in the environment, so Ecology is applying the PCB model to PBDEs. Now Ecology is working to update the 2009 model for PCBs and is adding PBDEs, PAHs, and selected metals. The Puget Sound Regional Toxics Model consists of three models used concurrently:
    - Circulation and transport of water. A model to predict transport of water between regions of Puget Sound and between surface and deep layers of the water column[9] .
    - Contaminant fate and transport. A model to predict water and sediment concentrations of PCBs in response to external loading and internal processes[10].
    - Food web bioaccumulation. A model to predict PCBs in Puget Sound biota in response to water and sediment concentrations [11].

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[9] A. L. Babson, M. Kawase and P. MacCready. 2006. Seasonal and interannual variability in the circulation of Puget Sound, Washington: A box model study. *Atmosphere-Ocean*. 44 (1), 29-45.

[10] Davis, J.A. 2004. The long-term fate of polychlorinated biphenyls in San Francisco Bay (USA). *Environmental Toxicology and Chemistry*. 23 (10), 2396-2409.

[11] Arnot, J.A. and F.A.P.C. Gobas. 2004. A food web bioaccumulation model for organic chemicals in aquatic ecosystems. *Environmental Toxicology and Chemistry*. 23 (10), 2343-2355.

- Modeling results will be available in April, 2014.

*Brendan Hickie (Trent University, Canada) presented on modeling bioaccumulation from prey to killer whales*

- Because many marine mammals eat high on the food chain, have relatively long lives, have large fat stores, have limited reproduction, and a high maternal investment, they are vulnerable to accumulation of high levels of endocrine disrupting contaminants from their prey. Killer whales are one of the most contaminated marine mammals in the world.
- The Southern Residents come into a highly urbanized area and can pick up lots of contaminants. However, the whales are highly mobile and feed on Chinook which are also highly mobile making it challenging to assess contaminants in the killer whales.
- Hickie has created several lifetime accumulation models which take into account the growth of the animal, day to day energy requirements, estimates contaminant intake based on prey, tracks contaminants in blubber, and allows for elimination pathways.
- Once persistent pollutants (e.g., PCBs) are ingested, they are absorbed into the bloodstream. Some pass through the feces; diffusion occurs from the bloodstream back into the gut contents. In juvenile killer whales, growth dilution occurs until they reach about 15-18 years of age. For pollutants not eliminated by biotransformation, they will increase in the body. Concentrations in females that stop reproducing will start to increase.
- Because of the high amount of pollutants offloaded from mother to calf during gestation and lactation, the biomagnification factor in calves approaches 350x relative to prey. Adult killer whales have a potential biomagnification factor closer to 100x that of what is in their prey.
- Biotransformation can differ in many marine mammals. If a congener metabolizes, you do not see a rise in concentration with age in males (this may apply to PBDEs). These congeners that readily metabolize can pose health problems (e.g., some can bind to thyroid receptors), whereas some are eliminated quickly.
- There are a few studies that have estimated PCB toxicity threshold levels in marine mammals. These range from 1.3mg/kg to 78 mg/kg with endpoints ranging from immunotoxicity, endocrine disruption, reduced population growth rates, vitamin A metabolism, and reproductive failure.
- The entire Southern Resident killer whale population exceeds the 17 mg/kg PCB effects threshold. For killer whales to have levels below this threshold, their prey would need to be less than 8 ng/g of total PCBs.
- Toxicity data for PBDEs are very thin in the literature. However, dealing with PBDEs is likely simpler than other pollutants such as PCBs because you are dealing with only a couple of congeners that accumulate in upper trophic level species.

*Teresa Mongillo (NMFS) presented on estimating PBDE exposure from Solo Point wastewater effluent*

- Teresa developed an incremental increase model to assess the degree to which PBDE loadings from the Solo Point wastewater treatment facility affected killer whale body burdens.
- The cumulative difference in the predicted PBDE loadings specific to Solo Point from the 5 years worth of discharge will add to the long-term accumulation that the whales will experience.
- NMFS anticipates that Southern Resident killer whales would incur adverse health effects over a shorter period of time than would otherwise occur absent of the discharge.

- Increasing PBDE levels in the whales only further exacerbates their current susceptibility to adverse health effects including effects to the whales' reproductive, endocrine, and immune systems.

*Jessica Lundin (University of Washington) presented on using scat to measure PBDEs and other toxicants in killer whales*

- Killer whale scat samples (approximately 300 samples) are being analyzed for toxicants, hormones (thyroid, stress, and pregnancy), and genotype.
- The lower molecular weighted PCBs are found more often in scat and the higher molecular weighted congeners are found in the blubber. There is a good comparison between levels in scat versus levels in blubber in a specific individual in J pod.
- Resampling similar individuals each year will provide trend information.
- Mean PCB levels in J pod are statistically higher than mean levels in K or L pods. Mean DDT levels are statistically higher in K and L pods than mean levels in J pod. PBDE levels are higher in J pod than K and L pods (however, not statistically). These preliminary results are likely due to differences in the pods' ranges.
- Jessica is currently examining PBDE congeners by age and sex class. Reproductive age females who have had a calf had lower levels of all compounds. There are higher PBDE levels in post reproductive females and reproductive age males. BDE-47 was found in the highest concentration in the scat.
- They have developed a fecal pregnancy test for killer whales using testosterone and progesterone levels measured in the scat. The preliminary results indicate that all the known pregnant females had similar levels of these hormones. In one individual, hormone levels measured in the scat indicated pregnancy, but the calf was found washed ashore later in the year.

## Workgroup 3: Toxics Thresholds and Mixtures

This workgroup included experts from NMFS, EPA, Ecology, the University of Washington, Trent University and Simon Fraser University, Canada, and the British Columbia Ministry of the Environment. See Appendix A for the agenda and participant list.

*Sam Wasser (University of Washington) presented on sampling for temporal patterns in toxicant exposure and physiological health in killer whales*

- A challenge in assessing environmental impacts on population recovery is the long time span between an event and its population impacts.
- The issue with matching toxicants in prey and in killer whale blubber is the time lag. Similarly, there is a time lag matching toxicants in biopsies with gene expression, endocrine response, morbidity and mortality.
- Sam Wasser et al. are currently using non-invasive tools to simultaneously sample for temporal patterns of toxicant exposure and physiological health using scat detection dogs.
- Scat can measure: reproductive failure and/or success (P4, T), toxicants, nutritional health (GC, T3), genotypes.
- Advantages to this method include: increased sample size, tight temporal sampling (no time lag, which means they may tie physiological measures to changes in fish availabilities and other

stressors, and assess endocrine disruption), may better reflect what is in circulation at any given time; they are finding a good correspondence between fecal and blubber toxicants.

- Glucocorticoids (or GCs) increase with psychological and nutritional stress, whereas the thyroid hormone, $T_3$, decreases with nutritional stress.
- For GCs: since prey abundance and the number of vessels in close proximity to the whales peak around the same time, GCs should be highest at the peak if boats were the primary influential factor, but lowest if prey availability is. They[12] found that when the whales first arrive in Haro Strait (when boats and fish are low) GCs are high. When fish availability is peaking, GCs are low, therefore it appears fish availability is driving these levels.
- $T_3$ controls metabolism and changes on a slower time scale. Therefore, the thyroid hormone levels at time of arrival to the Salish Sea should reflect conditions prior to transitioning to the new conditions.
- If prey ingestion is the greatest source of circulating toxicants, than the toxicants in the scat will be highest when prey abundance is greatest. Furthermore, it is hypothesized that these relatively high levels of toxicants will suppress $T_3$, GCs, and pregnancy concentrations at that time. However, if fat metabolism is the greatest source of circulating toxicants, toxicants in scat will be highest when food deprivation is greatest and these relatively high levels of toxicants will suppress $T_3$, GCs, and pregnancy concentrations at that time.

*Dawn Noren (NMFS) presented on the dynamics of persistent organic pollutant (POP) transfer from female dolphins to their offspring during lactation*

- Little is known about POP dynamics in marine mammals, specifically in whales.
- Reproductive history and body condition may impact the relative quantity of POPs transferred as well as circulating POP levels in females.
- An influx of toxicants at a young age may interfere with developmental processes.
- The objectives of this study were to 1) quantify the dynamics of POP transfer from female bottlenose dolphins to calves during gestation and lactation (quantify POP levels in placenta, milk, and blood of females and calves); 2) quantify total lipids and lipid classes in all samples; 3) determine how female age and reproductive history influence POP transfer dynamics; and 4) assess relationships between female and calf body condition indices (body mass, blubber thickness) and POP levels in milk and blood.
- *Preliminary Conclusions*: 1) POP levels were greatest in milk, followed by calf blood, placenta, and female blood; 2) PCB transfer dynamics during lactation in dolphins differ from grey seals. In dolphins, levels in milk and female blood decrease overtime, and increase in calf blood; 3) maternal age and reproductive history appear to influence POP levels in placenta, milk, female blood, and calf blood; 4) mobilization of PCBs in milk is not solely related to milk lipid content in dolphins.

[12] Ayres, K.L., R.K. Booth, J.A. Hempelmann, K.L. Koski, C.K. Emmons, R.W. Baird, K. Balcomb-Bartok, M.B. Hanson, M.J. Ford, and S.K. Wasser. 2012. Distinguishing the Impacts of Inadequate Prey and Vessel Traffic on an Endangered Killer Whale (*Orcinus orca*) Population. PLoS ONE 7(6):

*Lizzy Mos (British Columbia Ministry of the Environment) presented on risk assessment of marine mammals*

- Risk assessments of marine mammals are difficult for several reasons: they are exposed to complex mixtures, there are few in captivity, and traditional endpoints (mortality and growth) cannot be assessed.
- One way to assess risk is to use non-invasive biomarker-derived toxicity reference values (TRVs).
- Mos et al.[13] sampled harbor seal pup blubber from five locations (along a naturally existing gradient of POPs) and collected two food baskets in Georgia Strait and Puget Sound.
- Biomarkers of immunotoxicity and endocrine disruption indicate that exposure due to nursing leads to sub lethal adverse effects.
- Using information on exposure and effect in seals, they derived a PCB TRV (PCBs represented the greatest risk; the TRV is 1.3 mg/kg lipid weight).
- Similar mechanisms of toxicity among species suggest that TRVs can be used in protecting all marine mammal species.
- Biomarker-based guidelines can be used in risk assessment, but population consequences are unknown.

*Brendan Hickie (Trent University) presented on Bioaccumulation Modeling Perspective on PBDEs in Killer Whales- Lessons Learned from PCBs. This was a similar talk given in the Modeling workgroup. See above for summary bullet points.*

*Teresa Mongillo (NMFS) presented on exposure to a mixture of persistent pollutants and the health implications to the Southern Resident Killer Whales*

- The interactions of persistent pollutants (e.g., PBDEs and PCBs) are primarily additive and synergistic, and sometimes antagonistic.
- Exposure to a mixture of contaminants containing contrasting toxicities (e.g., a mixture of estrogenic and anti-estrogenic compounds) may mediate the effects.
- Mixture interactions between PCB and PBDE congeners were associated with reduced thyroid hormone levels, cytotoxicity, enhanced developmental neurotoxicity and cytogenotoxicity, enhanced neurobehavioral defects, and induced enzyme activity.
- Endocrine disruptors (such as PBDEs) produce nonmonotonic dose-response effects. Some compounds can also interact at doses below the no-observed-effect-concentrations (NOEC) and produce significant effects.
- Disregarding synergistic interactions may underestimate risk to an individual or to the population.
- Because additive or synergistic mixture effects can occur from a wide range of doses, low concentrations of the pollutants have the potential to cause adverse health effects in Southern Resident killer whales and hinder recovery.

[13] Mos, Lizzy, M. Cameron, S.J. Jeffries, B.F. Koop, and P.S. Ross. 2010. Risk-Based Analysis of Polychlorinated Biphenyl Toxicity in Harbor Seals. Integrated Environmental Assessment and Management. 6(4): 631-640.

*Individual Recommendations from the Toxics and Mixtures Workgroup*

- Fecal sample collection from the Southern Residents should occur concurrently with the Northern Resident population, as they are genetically similar but their range is in an area with far fewer of the environmental stressors than the Southern Residents (including lower toxicant loads). Performing a parallel study and analysis would help better understand the true relevance of the fecal measures and associated health outcomes (such as pregnancy success and immune status) on the Southern Resident killer whales.
- Using a single scat sample, we can associate PBDE (and other toxicant) levels with reproductive success. We need to evaluate the toxicant measures of these groups of females for associated differences in toxicant level. In the case these levels are different, the measures will serve as a guide for determining a toxicological threshold for reproduction.
- Adopt a toxicity threshold for a complex mixture derived from a reliable field study of another species (not subject to confounding factors such as sex, age, and diet).
- Monitoring efforts should continue to establish and identify the levels at which health effects occur in Chinook salmon.
- Given that a controlled toxicology experiment with killer whales is highly unlikely, using surrogate organisms and a weight of evidence approach may be the most realistic means of defining a toxicological threshold.
- In addition to laboratory studies, we should try to use the data that are collected from wild killer whales on toxicant levels and population effects.
- A truly protective PBDE threshold must be confirmed that the exposure concentration does not cause an adverse effect and is not simply a 100 or 1000-fold less than a published 'no effect' concentration.
- We need an approach that couples a review of all available toxicological effects data on killer whales (and closely associated marine mammals) with actual effects data from salmon, i.e. their primary prey base. If salmon populations are found to be affected at lower concentrations than killer whales, then a threshold must be established that is protective for salmon in order to maintain a healthy food source for killer whales.
- We need to address immune system toxicity by evaluating other hormone measures, genetic markers that indicate a compromised immune system, or by measuring immuneglobulins in the scat samples and associating those measures with PBDE levels.
- There should be a continued effort to collect blubber and fecal samples from captive whale populations, Southern Resident killer whales, and Northern Resident killer whales to measure contaminant levels in combination with health indices.
- Blubber sampling is currently the best method of sampling for toxicants; fecal sampling should continue to see what proportion of the toxicants in the scat are from the whale and what proportion are from the prey.
- In addition to collecting samples to assess contaminant levels in killer whales, we need to collect samples to assess health (e.g., hormone levels, immune response indices such as white blood cell counts, etc.) from a captive cetacean population, such as the Navy dolphins.

*Data Gaps and Uncertainties*

- The effects of PBDEs in the presence of other contaminants with similar opposing mechanisms of action.
- The contribution of factors other than PBDE exposure on killer whale health.

- PBDE levels that will cause deleterious population effects in killer whales and whether threshold levels determined for other animal systems would apply to killer whales.
- Additive or other interacting effects when other contaminants (e.g., PCBs, DDTs) are present or when there is another major issue, such as food limitation.
- Interactive effects of PBDEs with each other and with other chemicals present in killer whales.
- Effects of PBDE exposure during embryonic and early life-stage development.
- Health effects and bioaccumulation capabilities of these compounds on killer whales, including how the effects vary by age-sex class.
- Association of blubber, blood, and fecal toxicant levels in killer whales.
- Extent of mobilization of toxicants during food shortage in killer whales.
- Health consequence of these short term bursts of high toxicants loads.
- Winter foraging and behavior of this whale population.

## Policy Forum

On June 6th, 2013, EPA Region 10 convened senior level staff from EPA, NMFS, Ecology, the Puget Sound Partnership, and the Puget Sound Institute at a policy forum to discuss PBDEs in Puget Sound and its Southern Resident killer whales. See Appendix B for the policy forum agenda and participant list. Staff presented on EPA and Ecology's respective phase out plans for PBDEs, and briefed the group on the discussions and recommendations from the technical workgroups. Participants acknowledged the need for additional monitoring of PBDEs and other flame retardants/persistent bioaccumulative toxics in Puget Sound.

There was general agreement among participants that the Puget Sound Ecosystem Monitoring Program (PSEMP) is the appropriate venue through which to continue discussions regarding a Puget Sound-wide monitoring program for PBDEs (https://sites.google.com/a/psemp.org/psemp/home). PSEMP is a collaboration of state, federal, tribal and local government agencies, non-governmental organizations, watershed groups, businesses, academic researchers, local integrating organizations, and other private and volunteer groups and organizations - all dedicated to monitoring environmental conditions in Puget Sound. In June, 2013, Catherine Gockel of EPA briefed the PSEMP toxics workgroup on the results of the technical workgroups and policy forum. Lynne Barre of NMFS provided a similar briefing to the PSEMP marine mammals workgroup at their June, 2013 meeting. With leadership from EPA and NMFS, PSEMP will convene a sub-workgroup to continue this discussion during Fall 2013.

# Appendix A: Workgroup Agendas & Participant Lists

## Group 1: PBDE Removal Effectiveness in Wastewater Treatment Plants

**Agenda: First meeting: *April 9: 10 am - noon***

**10:00-10:10 - Introductions and Logistics:** Catherine Gockel, EPA

**10:10-10:15 - Behavior of PBDEs in the environment**: Catherine Gockel, EPA

**10:15-10:30 - PBDEs and killer whales in Puget Sound**: Teresa Mongillo, NMFS

**10:30-11:00 - Hear from Puget Sound Wastewater Treatment Utilities:** LOTT and King County

- Ben McConkey- LOTT's facilities, treatment processes, removal efficiency, etc. Discuss PBDE monitoring that LOTT has performed.
    - LOTT- Budd Inlet WWTP
- Bob Bucher- King County's facilities, treatment processes, removal efficiency, etc.
    - King County- West Point WWTP
    - King County- Brightwater Membrane Bioreactor (MBR) WWTP

**11:00-12:00 - Discussion: How are PBDEs changed/removed by various types of treatment?**

- Biological treatment
- Chemical-physical treatment
- Membrane Bioreactor (MBR)
- Advanced oxidation
- Other?

**Second Meeting: April 16: 10 am - noon**

**10:00-10:30 - Discussion (cont'd): How are PBDEs changed/removed by various types of treatment? *What processes are available to treat PBDEs through wastewater treatment plants?***

- Focus on certain congeners (BDE-047, BDE-099, BDE-209)
- Discuss engineering realities and challenges

**10:30-11:00- Discussion: Degrading/Debrominating PBDEs**

- Debromination pathways for PBDEs
- Degradation via UV light
- Degradation via microorganisms in wastewater treatment processes
- Some congeners get distorted in a WWTP and others pass through the plant

**11:00-11:30 - Discussion: Monitoring for PBDEs**

- Major WWTPs that discharge to Puget Sound are not monitoring for PBDEs
- No EPA approved methods for PBDEs
- Cost of PBDE monitoring
- Challenges associated with monitoring for PBDEs

**11:30-12:00 – Discussion and Conclusion**

- What realistic actions can we take?
- What are the data gaps and uncertainties?
- Next steps:
    - Compile recommendations
    - Follow up meeting to prepare for policy forum
    - Report back to senior level managers at policy forum

## Participants

1. Catherine Gockel, EPA Region 10 Seattle
2. John Palmer, EPA Region 10 Seattle
3. David Ragsdale, EPA Washington Operations Office
4. Erin Seyfried, EPA Region 10 Seattle
5. Ed Furlong, USGS Toxic Substances Hydrology Program
6. Dana Kolpin, USGS Toxic Substances Hydrology Program
7. Rob Hale, Virginia Institute of Marine Science
8. Foroozan Labib, Washington State Department of Ecology
9. Bob Bucher, King County
10. Deb Lester, King County
11. Ben McConkey, Lacey, Olympia, Tumwater and Thurston County (LOTT) Clean Water Alliance
12. Teresa Mongillo, National Marine Fisheries Service
13. Lynne Barre, National Marine Fisheries Service
14. Jeff Fisher, National Marine Fisheries Service

### Group 2: PBDE Modeling Technical Workgroup

**Agenda: First meeting: Wednesday, April 24   10 am- noon**

10:00-10:10 - Introductions and Logistics: Catherine Gockel, EPA

10: 10-11:00 - Puget Sound Regional Toxics Model: Greg Pelletier, Washington State Department of Ecology

11:05-11:25 - Modeling Bioaccumulation from Prey to Killer Whales – Lessons Learned from PCBs: Brendan Hickie, Trent University, Ontario, Canada

11:25-12:00 – Discussion

**Second meeting: Thursday, May 16   10 am – noon**

10:00-10:10 - Introductions and Logistics: Catherine Gockel, EPA

10:10-10:25 - Estimating PBDE Exposure from Solo Point Wastewater Effluent: Teresa Mongillo, NMFS

10:25-10:40 – Using Scat to Measure PBDEs and Other Toxicants in Killer Whales: Jessica Lundin, University of Washington

10-40-11:50 – Discussion  (5 minute break at 11 am)

11:50-12:00 – Wrap up

### Participants

1. Catherine Gockel, EPA Region 10 Seattle
2. John Palmer, EPA Region 10 Seattle
3. Ben Cope, EPA Region 10 Seattle
4. Greg Pelletier, Washington State Department of Ecology
5. Frank Gobas, Simon Fraser University, Canada
6. Brendan Hickie, Trent University, Canada
7. Lynne Barre, National Marine Fisheries Service
8. Teresa Mongillo, National Marine Fisheries Service
9. Lyndal Johnson, National Marine Fisheries Service
10. Mary Arkoosh, National Marine Fisheries Service
11. Joe Dietrich, National Marine Fisheries Service
12. Sandie O'Neill, National Marine Fisheries Service
13. Gina Ylitalo, National Marine Fisheries Service
14. Jeff Fisher, National Marine Fisheries Service
15. Jessica Lundin, University of Washington

## Group 3: Toxicological Thresholds for the Protection of Southern Resident Killer Whales

**Agenda: First Meeting: April 23rd**

1:00 – 1:10 **Introductions and Logistics-** Catherine Gockel, EPA

1:10 – 1:20 **The Need for Toxicological Threshold Levels in Killer Whales-** Teresa Mongillo, NMFS

1:20 – 1:50 **Current work on toxics:**

- Sam Wasser, UW –Sampling for temporal patterns in toxicant exposure and physiological health in killer whales
- Dawn Noren, NMFS – The dynamics of persistent organic pollutant (POP) transfer from female dolphins to their offspring during lactation

1:50 – 2:05 **Risk Assessment of Marine Mammals-** Lizzy Mos, BC Ministry of Environment

2:10 – 3:00 **Discussion: Exposure to PBDEs- Past and Future Studies**

- What do we know about dynamics of PBDEs in the body? What do we need to know?
- What are the threshold levels for PCBs in harbor seals and how were they derived?
- What are the best methods to analyze PBDEs in the body? (blubber, fecal, modeling)
- What biomarker studies, monitoring studies, or other, should be performed? What has worked in other species?

**Second Meeting: May 7th**

1:00 – 1:30 **(Continued Discussion): Exposure to PBDEs**

- Teresa Mongillo, NMFS- Summary of last meeting's presentations and open discussion on questions below:
- What are the best methods to analyze PBDEs in the body? (blubber, fecal, modeling)
- What biomarker studies, monitoring studies, or other, should be performed? What has worked in other species?
- What do we know about dynamics of PBDEs in the body? What do we need to know?
- Can we use threshold levels in harbor seals to estimate risk in killer whales?

| | | |
|---|---|---|
| 1:30 – 2:00 | **Discussion: Can we use toxicity reference values (TRVs) or tissue residue guidelines (TRGs) in fish to protect killer whales?** | |

- Brendan Hickie, Trent University- Bioaccumulation Modeling Perspective on PBDEs in Killer Whales – Lessons Learned from PCBs
- Is it feasible to establish PBDE toxicological thresholds for killer whales, or can we use guidelines or reference values in surrogate species (harbor seals, fish)?

2:05 – 2:30 **Discussion: Mixture Effects and Threshold Levels**

- Teresa Mongillo, NMFS- Exposure to a Mixture of Persistent Pollutants: Health Implications to the Southern Resident Killer Whales
- Mixture interactions may enhance adverse health effects, should we focus on a total pollutant threshold (PCBs + PBDEs + DDTs etc.) instead of single compound thresholds?

2:30 – 3:00 **Discussion: Conclusion**

- What are realistic actions we can take?
- What are the data gaps and uncertainties?
- Next Steps:
    - Compile Recommendations
    - Follow up meeting to prepare for policy forum
    - Report back to senior level managers at policy forum

### Participants

1. Catherine Gockel, EPA Region 10 Seattle
2. John Palmer, EPA Region 10 Seattle
3. Teresa Mongillo, National Marine Fisheries Service
4. Lynne Barre, National Marine Fisheries Service
5. Joe Dietrich, National Marine Fisheries Service
6. Sandie O'Neill, National Marine Fisheries Service
7. Gina Ylitalo, National Marine Fisheries Service
8. Dawn Noren, National Marine Fisheries Service
9. Jeff Fisher, National Marine Fisheries Service
10. Tim Rymer, National Marine Fisheries Service
11. Randy Marshall, Washington State Department of Ecology
12. Holly Davies, Washington State Department of Ecology
13. Lizzy Mos, British Columbia Ministry of the Environment, Canada
14. Brendan Hickie, Trent University, Canada
15. Jessica Lundin, University of Washington
16. Samuel Wasser, University of Washington
17. Frank Gobas, Simon Fraser University, Canada

# Appendix B: Policy Forum Agenda & Participant List

## AGENDA - PBDEs & Puget Sound Killer Whales Policy Forum

**June 6th, 2013 9 am – noon**

Location: EPA Region 10 (1200 Sixth Avenue, Seattle) - 12th floor Bitterroot Room

**9-9:30 am: Welcome & Introduction to PBDEs in Puget Sound**

- Dan Opalski, EPA Region 10's Office of Water and Watersheds
- Donna Darm, National Marine Fisheries Service
- Catherine Gockel, Puget Sound Team & NPDES Permits Unit
  - Introduction to PBDEs, PBDE sources and loadings to Puget Sound
- Kirsten Hesla, EPA Headquarters
  - Action Plan/SNUR for PBDEs
- Holly Davies, Ecology Waste 2 Resources
  - Ecology's Chemical Action Plan for PBDEs

**9:30-10:00 am: Toxics & Mixtures Workgroup**

- Catherine Gockel, Puget Sound Team & NPDES Permits Unit
  - Overview of group members and approach
- Teresa Mongillo, National Marine Fisheries Service
  - PBDEs and Southern Resident Killer Whales
  - Summary of workgroup

**10:00-10:30 am: Modeling PBDEs in Puget Sound**

- Catherine Gockel, Puget Sound Team & NPDES Permits Unit
  - Overview of workgroup, members, and what we learned

-5 minute Break-

**10:35-11:30 am: PBDEs and Wastewater Effluent**

- Mike Lidgard, EPA Region 10 NPDES Branch Chief
- Catherine Gockel, Puget Sound Team & NPDES Permits Unit
  - Overview of workgroup and members
  - Wastewater Treatment and PBDEs
- Recommendations
- Discussion

**11:30-noon: Discussion &Conclusion**

**Participants**

1. Dan Opalski, EPA Region 10 Seattle
2. Tom Eaton, EPA Region 10 Washington Operations Office
3. Michael Lidgard, EPA Region 10 Seattle
4. Karen Burgess, EPA Region 10 Seattle
5. John Palmer, EPA Region 10 Seattle
6. Catherine Gockel, EPA Region 10 Seattle
7. Donna Darm, National Marine Fisheries Service
8. Teresa Mongillo, National Marine Fisheries Service
9. Lynne Barre, National Marine Fisheries Service
10. Jeff Fisher, National Marine Fisheries Service
11. John Stein, NOAA Northwest Fisheries Science Center
12. Bill Moore, Ecology Headquarters
13. Greg Zentner, Ecology Southwest Regional Office
14. Kevin Fitzpatrick, Ecology Northwest Regional Office
15. Mark Henley, Ecology Northwest Regional Office
16. Tracy Collier, Puget Sound Partnership
17. Joel Baker, Puget Sound Institute
18. Andy James, Puget Sound Institute