Receiving environment shapes transport and bioaccumulation of polybrominated diphenyl ethers near two submarine municipal outfalls

by

Pamela Dinn
B.Sc., University of Victoria, 2004

A Thesis Submitted in Partial Fulfillment
of the Requirements for the Degree of

Master of Science

in the School of Earth and Ocean Sciences

Supervisory Committee

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Abstract

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The fate and bioaccumulation of a contaminant entering the marine environment through wastewater outfalls depends on the contaminant's persistence and affinity for particles. The physical characteristics of the receiving environment, e.g. current velocity, sedimentary processes, and the availability of organic carbon are also important. However, these latter effects are not usually evaluated quantitatively. This thesis investigates the near-field accumulation in sediment and biota of particle-reactive polybrominated diphenyl ethers (PBDEs) entering coastal waters via two municipal outfalls: one discharging into a high energy, low sedimentation environment near Victoria, B.C., Canada; the other into a low energy, high sedimentation environment near Vancouver, B.C. We used ^{210}Pb profiles in sediment box cores together with an advection-diffusion model to determine surface mixing and sedimentation rates, and to model the depositional history of PBDEs at these sites. A particularly important finding of this study is that the very high energy environment to the southeast of the Victoria outfall accumulates PBDEs despite not having net sediment accumulation. Although the discharge of PBDEs was much lower from the Victoria outfall than from Vancouver, some sediment PBDE concentrations were higher near Victoria. Most PBDEs were dispersed beyond the near-field at both sites, but a greater proportion was captured in the sediment near the Vancouver outfall where rapid burial was facilitated by inorganic sediment supplied from the nearby Fraser River. Clearly, treating wastewater to the same level, regardless of local oceanographic conditions, will not result in a uniform environmental footprint. Total PBDE concentrations in benthic invertebrate communities were higher near Vancouver than Victoria, despite lower concentrations in sediments, and correlated with organic carbon normalized sediment concentrations. Principal Components Analysis suggested uptake of individual PBDE congeners was determined by sediment properties (TOC, grain size), whereas PCB congener uptake was governed by physico-chemical properties (octanol-water partitioning coefficient). Our results suggest that sediment quality guidelines for PBDEs and likely PCBs may be more relevant if corrected to TOC content in sediment. In addition, where enhanced wastewater treatment increases the ratio of PBDEs to particulate organic carbon in effluent, nearfield benthic invertebrates may face increased PBDE accumulation. This underlines the need for source control of persistent organic contaminants, which cannot be broken down by conventional wastewater treatment.

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Acknowledgments

This research project would not have been possible without the support of many people. First I would like to express thanks to my supervisor, Dr. Sophia Johannessen, for always being available with helpful insights and guidance. I could not have wished for a better supervisor. Thanks are also due to all of the members of my committee who have helped improve this thesis in numerous ways: My co-supervisor, Dr. Michael Whiticar at the University of Victoria, Dr. Robie Macdonald and Dr. Peter Ross at the Institute of Ocean Sciences, and Christopher Lowe at the Capital Regional District.

Funding for this project was provided by NSERC-IPS, the Capital Regional District, Metro Vancouver, Environment Canada and Fisheries and Oceans Canada. Thanks to the CRD for encouraging this project from the beginning and providing ongoing funding and data sharing.

Thanks to all of my friends in the School of Earth and Ocean Sciences graduate studies program for advice and encouragement along the way, and to my family and friends for keeping me cheerful during the rough spots. Special thanks to my husband, David Pugh, for endless patience and understanding during the duration of my studies.

Chapter 1
Introduction

Submarine municipal outfalls

Submarine municipal outfalls release a complex mixture of chemicals into marine environments, and these discharges can lead to impassioned debates about the environmental effects of various methods of treatment and disposal. Wastewater discharges have the potential to degrade coastal oceans through several mechanisms [1]. Excess loading of nitrogen and phosphorus can cause eutrophication [2], and persistent contaminants such as metals can accumulate in sediment and cause toxicity in benthic invertebrates. As well, some organic chemicals can bioaccumulate to higher trophic levels in the marine food web e.g. [3, 4]. Higher levels of sewage treatment will remove more nutrients and persistent contaminants from wastewater before discharge to the oceans, but most persistent contaminants cannot be degraded by conventional wastewater treatment [5, 6]. This means that regulators must often choose between disposing of contaminants on land or in the ocean. Ideally, these choices would depend on the presence or extent of degradation in the receiving environment [7]. It is important to consider the site specific characteristics of the receiving environment itself when assessing the risk of sewage disposal and treatment options in coastal oceans, but this aspect has not been well studied.

The public and regulators usually focus on the concentration and flux of contaminants in wastewater, including nutrients, total organic carbon (TOC), trace metals and persistent and hydrophobic chemicals like polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs). However, the fate of these chemicals and the effects on the near-field area depend strongly on the physical environment into which they are discharged. Current velocity and the local supply of particles determine sediment accumulation rates, grain size distribution and the type of benthic habitat. Particle-active contaminants settle in marine sediment and are then taken up by benthic invertebrates, which can lead to further accumulation in the food web. Benthic invertebrate accumulation of contaminants present in sediment depends on surface concentrations, chemical properties and the species of benthic invertebrates present [8]. Benthic invertebrates themselves further modify the environment in which they live by redistributing contaminants in sediment through biomixing, bioturbation and bioirrigation [9]. Together, physical, chemical and biological factors affect the dispersal, near-field concentrations, and bioaccumulation of particle-active contaminants originating in wastewater outfalls.

Comparison of Vancouver and Victoria receiving environments

The cities of Vancouver and Victoria, B.C., on Canada's west coast discharge municipal wastewater through diffusers on the seafloor into contrasting receiving environments within the Georgia Basin. Metro Vancouver discharges wastewater through five outfalls, including one at Iona Island that provides primary treatment to ~600,000 people and flows directly into the Strait of Georgia at a depth of 80 m [10]. High sedimentation rates in the Strait of Georgia have been measured by [11] and [12] and are confirmed by a high proportion of fine sediment. These sediment accumulation rates are largely supported by the Fraser River [13], which discharges 17 x 10^6 tonnes of terrigenous particles every year, representing 80% of the total particle input to the Strait [14, 15].

The Iona Island outfall discharges near the mouth of the Fraser River, into a low energy/high sedimentation environment that is characterized by weak currents and highly stratified water [15]. The asymmetrical tidal currents here flow mainly in a north-south direction at an average speed of 0.1 m s^{-1} [16]. The benthic invertebrate community near the Iona outfall is influenced by high percent fines (50-94%) and a low proportion of gravel (<1%). Victoria's Macaulay Point outfall is one of eight in the Capital Regional District (CRD). It serves ~120,000 people and discharges screened wastewater at a depth of ~60 m into the high energy/low sedimentation environment of Juan de Fuca Strait.

In contrast to the Iona setting there are few external sources of sediment to the Juan de Fuca Strait, as no major rivers drain directly into the Strait, and little of the sediment from the Fraser River reaches this far [15]. Cliff erosion provides some sediment locally, but very little of this material settles, because Juan de Fuca waters are well mixed by tidal currents [14, 17]. Currents near the Macaulay Point outfall pipe flow in a northwest-southeast direction and average 1.0 m s^{-1} [14]. The low sedimentation rate here is also implied by the high proportion of rock and gravel (1-20%) and little fine-grained material (19-38%), both evidence of erosion and resuspension in the local sediment [18]. For simplicity, I will refer to the Iona Island outfall as the Vancouver outfall, and the outfall at Macaulay Point as the Victoria outfall, throughout this thesis.

Benthic invertebrate communities

Benthic invertebrate communities near the Victoria and Vancouver outfalls have many species in common, particularly those more tolerant to environments with high organic carbon and other loadings from wastewater [10, 18, 19]. However, community composition, indicated by the proportions of groups such as annelids, molluscs, echinoderms and arthropods, are different because of the different physical environments they live in [10, 18]. Benthic invertebrate communities that are closely associated with contaminated sediment may accumulate high concentrations of hydrophobic organic contaminants such as PBDEs and PCBs through direct contact with the sediment particles, or through ingestion of particles and other organisms [4, 8]. Sediment-dwelling organisms are often the first level of the food chain to accumulate highly hydrophobic and lipophilic persistent contaminants, and therefore they can play an important role in the transfer of organic chemicals such as PBDEs to higher trophic levels [8, 20, 21]. The rate at which benthic species accumulate PBDEs from sediment can be species specific [22] as well as congener specific [3, 4, 23].

Sediment Quality Guidelines

In North America, the risk that any particular hydrophobic chemical present in sediment will be toxic to biota is often characterized using sediment quality guidelines (SQGs), which approach the problem of risk management using concentrations in sediment as predictors of toxicity [24]. Sediment quality guidelines are derived either empirically or theoretically. Empirically-derived SQGs are based on correlations of measured total concentration in sediment to direct toxicity to benthic organisms [25, 26]. Theoretical SQGs are based on chemical partitioning of chemicals in sediments and quantify the bio-availability of contaminants in sediment based on pore water concentrations [25]. SQGs have a number of recognized limitations. Empirical SQGs do not consider bio-availability, and theoretical SQGs do not consider sediment ingestion as a route of exposure and therefore underestimate effects [26, 27]. The most widely used SQGs are empirically based, relying on total concentrations of chemicals in sediment, and are most useful when they are used concurrently with multiple assessment approaches [7, 27, 28].

In the case of PBDEs, guidelines have only recently been established [18], and past evaluation of PBDEs relied on SQGs developed for PCBs, which have similar chemical structures and ranges in halogenated congeners and are far better studied [5, 20, 29, 30]. However, this approach to dealing with PBDEs neglects environmental factors that may lead to very different accumulations of PCBs and PBDEs. Sediment chemistry can influence the bio-availability of contaminants, and toxicological sensitivities can vary among species [25]. The Canadian federal government recently developed environmental quality guidelines for PBDE concentrations in a variety of matrices. Guidelines for fish, mammals and birds and are based on laboratory toxicity tests. Water quality guidelines were extrapolated from fish tissue guidelines, and SQGs were developed from water quality guidelines. PBDE SQG concentrations are normalized to 1% TOC. The guidelines are target concentrations that are designed to be used with ongoing monitoring in Canada to assess the environmental effects of legislated changes in PBDE use [24].

Polybrominated diphenyl ethers and polychlorinated biphenyls

Polybrominated diphenyl ethers (PBDEs) provide a proxy for the transport and fate of particle-reactive contaminants discharged with municipal wastewater, because outfalls represent a major route of entry into coastal water [5, 30, 31]. While legacy contaminants such as PCBs have reached widespread distribution in the environment [32], the more recently introduced PBDEs are found in highest concentration close to their entry points [33, 34]. Widely used as flame retardants in furniture, fabrics and electronics [35], PBDEs enter municipal wastewater streams through household dust and landfill disposal. PBDEs and PCBs both exist as many chemical congeners with similar structures, but their histories of use and global distribution are very different [5, 20, 29, 30]. PCBs were banned decades ago and are declining globally. PBDEs were introduced in 1978, and in many locations PBDE burdens now surpass PCBs [33]. Reviews tracking PBDE levels published up to 2004 showed that these chemicals were increasing exponentially in many environmental matrices [33, 36, 37], including Great Lake trout and California peregrine falcons [38, 39]. However, these compounds began to decrease in Europe following a ban [40] and may also have peaked in North America as bans take effect. Total PBDEs are now decreasing in B.C. marine waters and elsewhere [41].

The chemical structures and major physical properties of PBDEs resemble those of PCBs, suggesting similar toxicological properties such as endocrine disruption. The similarity between the structure of PBDEs and that of the thyroid hormone thyroxine means that the artificial compound can disrupt the endocrine system of marine animals [42, 43]. PBDEs have been widely detected in sediment, water and animals [35, 36, 44-46]. They have been shown to bioaccumulate in marine organisms [4, 8, 23, 47], although not in all food chains [46]. PBDEs are even more particle-reactive and hydrophobic than PCBs. Log octanol-water coefficients (log K_{ow}) for PBDEs range from 5.24 to 10.33 versus 5.09 to 8.18 for PCBs. The high K_{ow} values for PBDEs indicate their persistence in sediment [48], and sediments are expected to provide a reservoir of PBDEs that will support continued cycling in the environment for decades to come [33, 36]. The persistence and bioaccumulative potential of PBDEs, coupled with their long range transport potential warrants concern about future toxicity hazards. These traits led to PBDEs being assessed under the Canadian Environmental Protection Act (CEPA) in 2006, as potential chemicals of concern to aquatic ecosystems.

Of the 209 possible congeners of PBDEs, only 40 have been manufactured. They have been produced in three commercial mixtures: pentaBDE, octaBDE and decaBDE. The penta and octa formulations are composed of the lighter congeners, such as BDE-47 and BDE-99, and have faced worldwide restrictions in the past decade due to the bioaccumulative and toxic nature of these congeners [30, 49]. PBDEs have never been manufactured in Canada, but are present in a wide variety of household products imported into the country. The use of pentaBDE and octaBDE was phased out in Canada beginning in 2004 [49]. The manufacture of these mixtures by industry in the United States also ended in 2004, as recommended by the U.S. Environmental Protection Agency (U.S. EPA). Environment Canada will implement a ban on the import of products containing decaBDE beginning in 2013, corresponding with the voluntary phase out of the manufacture of this chemical mixture in the U.S. Due to their widespread presence in consumer goods, PBDEs are expected to continue to enter the environment for years to come. PBDEs and PCBs preferentially partition into sediments due to their hydrophobicity (octonol:water coefficients from 10^5 to 10^{10} [48, 50]), and sediments can be a long-term reservoir and/or a potential source of these chemicals [21, 33]. Sediment depth profiles and surface measurements of PBDEs collected near municipal outfalls can therefore be used to infer the depositional history and fate of contaminants that pass through the outfall and settle on the seafloor [51].

Thesis objective

This thesis presents a case study characterizing the fate of PBDEs discharged into two very different receiving environments. The fate of a contaminant entering the marine environment through wastewater outfalls depends on the contaminant’s persistence and affinity for particles. However, differences in the physical characteristics of the receiving environments, e.g. current velocity, sedimentary processes, are also important, but yet are only infrequently evaluated quantitatively. Chapter 2, entitled "*Effect of receiving environment on the transport and fate of polybrominated diphenyl ethers near two submarine municipal outfalls,*" is a paper with authors Dinn, P.M., Johannessen, S.C., Macdonald, R.W., Lowe, C.J., and Whiticar, M.J.. It has been accepted for publication in the journal Environmental Toxicology and Chemistry. In this paper I investigate the near-field accumulation of PBDEs from the Victoria and Vancouver outfalls using ^{210}Pb-dated sediment cores and surface grab samples collected near the two outfalls. I determine the depositional history, spatial distribution and accumulation of PBDEs in the two regions, and the proportion of total PBDEs from wastewater that is captured in near-field sediment versus that dispersed farther afield, and compare these proportions at the high and low energy sites.

Chapter 3 is a paper entitled "*PBDE and PCB accumulation in benthos near marine wastewater outfalls: The role of sediment organic carbon*" with authors Dinn, P.M., Johannessen, S.C., Ross, P., Macdonald, R.W., Whiticar, M.J., Lowe, C.J., and van Roodselaar, A.. It has been submitted for publication in the journal Environmental Science & Technology. In this paper I evaluate the effects of different physical environments and the availability of organic carbon on the uptake of PBDEs and PCBs from sediment by benthic invertebrates. I compare PBDE and PCB uptake using biota-sediment accumulation factors (BSAFs). I evaluate congener patterns of these compounds in different matrices through congener ratios and principal component analysis (PCA) to determine the factors driving PBDE and PCB congener sorting among matrices and between sites. I use these results to assess the applicability of PCB sediment quality guidelines to PBDE risk assessment. I also draw conclusions about the effects of increased sewage treatment on PBDE uptake in different environments.

In Chapter 4 I put forward overall conclusions drawn from the results of Chapters 2 and 3, presenting the implications of PBDE accumulation and fate for other particle-reactive contaminants in wastewater. Chapter 4 further expands on implications for sewage treatment and sediment quality guidelines.

Chapter 2

Effect of receiving environment on the transport and fate of polybrominated diphenyl ethers near two submarine municipal outfalls

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Total number of words, including table and figure captions, and references: 6,239

EFFECT OF RECEIVING ENVIRONMENT ON THE TRANSPORT AND FATE OF POLYBROMINATED DIPHENYL ETHERS NEAR TWO SUBMARINE MUNICIPAL OUTFALLS

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Abstract

The fate of contaminants entering the marine environment through wastewater outfalls depends on the contaminant's persistence and affinity for particles. However, the physical characteristics of the receiving environment, e.g., current velocity, sedimentary processes, may be even more important. Due to the complexity of natural settings and the lack of appropriate comparative settings, this is not frequently evaluated quantitatively. We investigated the near-field accumulation of particle-reactive polybrominated diphenyl ethers (PBDEs) entering coastal waters via two municipal outfalls: One discharging into a high energy, low sedimentation environment near Victoria, B.C., Canada; the other into a low energy, high sedimentation environment, near Vancouver, B.C. We used ^{210}Pb profiles in box cores together with an advection-diffusion model to determine surface mixing and sedimentation rates, and to model the depositional history of PBDEs at these sites. Surprisingly, 88-99% of PBDEs were dispersed beyond the near-field at both sites, but a greater proportion of PBDEs was captured in the sediment near the Vancouver outfall where rapid burial was facilitated by inorganic sediment supplied from the nearby Fraser River. Although the discharge of PBDEs was much lower from the Victoria outfall than from Vancouver, some sediment PBDE concentrations were higher near Victoria.

Keywords –

Polybrominated diphenyl ethers, Submarine outfalls, Marine sediment, Georgia Strait, Particle-reactive contaminants

INTRODUCTION

The discharge of municipal wastewater into coastal marine environments has led to an impassioned debate over the environmental effects of treatment and disposal. The public and regulators usually focus on the concentration and flux of contaminants, including nutrients, trace metals and persistent organic pollutants like polychlorinated biphenyls and polybrominated diphenyl ethers (PBDEs). Regulations governing the discharge of municipal wastewater generally consider only the chemical composition and quantity of the effluent and not the environment into which the effluent discharges. However, the energy of a marine environment and the local supply of particles determine sediment accumulation rates, grain size distribution and the type of benthic habitat. Together these factors affect the dispersal and near-field concentration of particle-active contaminants.

The cities of Vancouver and Victoria, BC, Canada discharge municipal wastewater into contrasting receiving environments within the Georgia Basin (**Fig. 2.1**) via diffusers on the seafloor. Metro Vancouver discharges wastewater through five outfalls, including one at Iona Island that provides primary treatment to approximately 600,000 people and flows directly into the Strait of Georgia. Sedimentation in the Strait of Georgia is largely supported by the Fraser River, which discharges 17 x 10^6 tonnes of terrigenous particles each year, representing 80% of the total particle input to the Strait [1, 2]. The Iona outfall discharges near the mouth of the Fraser River, into a low energy/high sedimentation environment that is characterized by weak currents and highly stratified water [3]. Direct measurements show high sedimentation rates of fine-grained

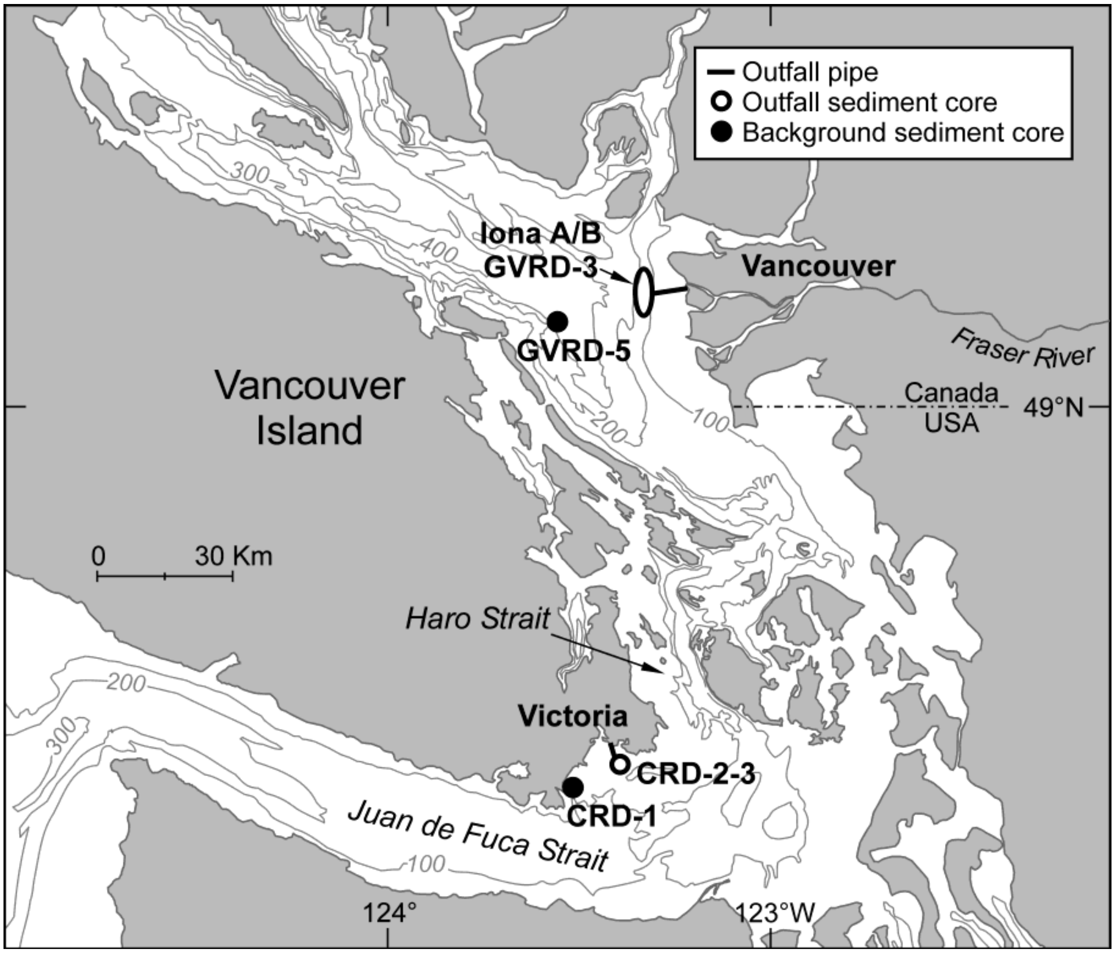

Figure 2.1 Locations near outfall discharges (GVRD-3, Iona A & B, CRD-2 & 3) and in background locations (GVRD-5, CRD-1). Circles represent the near-field receiving environment, and locations of surface sediment samples taken from within these circles are shown in Figure 2.3.

mud and the near absence of gravel in this area [4]. Victoria’s Macaulay Point outfall is one of eight in the Capital Regional District (CRD). It serves approximately 120,000 people and discharges screened wastewater into the high energy/low sedimentation environment of Juan de Fuca Strait. In contrast to the Iona setting there are few external sources of sediment to the Juan de Fuca Strait, as no major rivers drain directly into the strait, and little of the sediment from the Fraser River reaches this far [3]. Cliff erosion provides some sediment locally, but very little of this material settles, because Juan de Fuca waters are well mixed by strong tidal currents [2, 4]. The low sedimentation rate here is also implied by the high proportion of rock and gravel and little fine-grained material, both evidence of erosion and resuspension in the local sediment [5]. Hereafter we will refer to the Iona Island outfall as the Vancouver outfall, and the outfall at Macaulay Point as the Victoria outfall.

Polybrominated diphenyl ethers (PBDEs) provide a proxy for the transport and fate of particle-reactive contaminants discharged with municipal effluent because outfalls represent a major route of entry into coastal water, according to Song et al. [6], and Ross et al. [7]. While legacy contaminants, such as polychlorinated biphenyls, have a widespread distribution in the environment, the more recently introduced PBDEs are found in highest concentration close to their entry points [8-10]. Widely used as flame-retardants in furniture, fabrics and electronics [11], PBDEs enter municipal wastewater streams through household dust and landfill disposal. They are hydrophobic, particle-reactive compounds with high octanol-water coefficients (K_{ow} $10^{5.9}$ to $10^{9.97}$) [12] and are persistent, bioaccumulative and toxic [8, 11, 13]. Previous work [10, 14] compared the depositional histories of polychlorinated biphenyls and PBDEs in the Strait of Georgia and explained their distribution over a wide area in surface sediments.

Here we compare the fate of PBDEs discharged into two very different receiving environments adjacent to the Vancouver and Victoria municipal outfalls. We determine the depositional history, spatial distribution and accumulation rate of PBDEs at the two sites, using ^{210}Pb-dated sediments cores and surface grab samples. We contrast the congener patterns and the proportion of total PBDEs from wastewater that is captured in near-field sediment at the two sites.

MATERIALS AND METHODS

Sample collection

Seven sediment cores were collected (Fig. 2.1) using a Pouliot box corer (20 cm x 30 cm cross section). Core lengths were approximately 50 cm near Vancouver and 15-20 cm near Victoria. Two near the Victoria outfall (Capital Regional District (CRD)-2 and CRD-3), three near the Vancouver outfall (Greater Vancouver Regional District (GVRD)-3, Iona-A and Iona-B), and two at sites far away from the direct influence of outfalls (GVRD-5 in the Strait of Georgia and CRD-1 in Parry Bay, near the Juan de Fuca Strait). The GVRD-3 and GVRD-5 cores have been described previously [10, 15]. The cores collected near the Vancouver outfall were sub-sampled into 1 cm intervals from the sediment surface to 10 cm; at 2 cm intervals from 10 cm to 20 cm; and then at 5 cm intervals from 20 cm to the bottom (~ 50 cm). The cores collected near the Victoria outfall and in Parry Bay were shorter due to the difficulty of coring in coarse sediment found in Juan de Fuca Strait. These cores were sub-sampled into 1 cm intervals over their entire length (15-20 cm). To minimize contamination from smearing by the corer wall, the outermost 1 cm of material was discarded. Each sub-sample was homogenized and then divided between a 500 ml amber glass jar for PBDE analysis and a 120 ml plastic container for ^{210}Pb determination.

Sediment core dating

Radiometric determinations were conducted by Flett Research Ltd. (Winnipeg, Manitoba). ^{210}Pb activity was inferred from the ingrowth of ^{210}Po (counting errors <3%) following the procedures of Eakins and Morrison [16] and then salt-corrected. The activity of ^{226}Ra, determined for subsamples from the top, middle and bottom of each core, was inferred from the ingrowth of ^{222}Rn over 4 d, following the method of Mathieu et al. [17] modified by Flett Research Ltd.

The bottom of the bioturbated surface mixed layer (SML) in each sediment core was identified as the depth at which there was an abrupt change in the slope of log [^{210}Pb] versus sediment depth. Sediment accumulation rate (g cm^{-2} yr^{-1}) and mixing rate (cm^{2} yr^{-1}) were modelled using advective-diffusive equations [18] that incorporated a SML with a high diffusion rate overlying deeper sediments with a slower diffusion rate, where the diffusion rate is the biological mixing rate (see [3]). A constant sediment accumulation rate was assumed in all profiles except GVRD-3, where the sedimentation rate had clearly increased following the installation of the Vancouver deep outfall in 1988.

Chemical analysis

PBDE determinations for the Iona A and Iona B sediment cores were performed by the Fisheries and Oceans Canada Laboratories of Expertise in Aquatic Chemical Analysis at the Institute of Ocean Sciences (BC, Canada). Other sediment cores and surface sediment samples were analyzed by AXYS Analytical Services (BC, Canada). Both laboratories follow United States Environmental Protection Agency method 1614 [19] to measure 40 PBDE congeners by high resolution gas chromatography/high resolution mass spectrometry. Each batch of 10 samples included a duplicate sample and at least one procedural blank. Internal standard recoveries, agreement between duplicates and levels of analytes in procedural blanks all met in-house criteria as per U.S. EPA methods. Detection limits ranged from 12.8 to 105 pg g^{-1} for BDE-209 and 0.05 to 12.4 pg g^{-1} for all other congeners. Procedural blanks showed concentrations above detection limits for seven congeners, but these levels were < 5pg g^{-1} except for BDE-209. Levels of BDE-209 in procedural blanks (76 ± 11 pg g^{-1}) were minor compared to concentrations in sediment and therefore results were not blank corrected. ΣPBDEs are reported as the sum of all PBDE congeners detected in a sample. Organic carbon determination and grain size analysis were performed by ALS Laboratory Group as per United States Environmental Protection Agency Method 9060A [20] and Forestry Canada method NOR-X319 [21].

PBDE deposition

Bioturbation precludes our setting exact dates against sediment horizons. Rather, each depth in the sediment represents a combination of material mixed over a number of years depending on the sedimentation rate and the depth of the mixed layer. We modeled historical PBDE deposition for each core using Matlab code to produce a projected vertical profile of PBDE concentration with depth. Input parameters for the PBDE model included sedimentation velocity, SML depth and diffusion rates (also called mixing rates) within and below the SML (Table S1). Surface mixed layer depth and sedimentation velocity were determined directly from the profiles of ^{210}Pb, while diffusion rates were calculated based on the ^{210}Pb profiles, using advective-diffusive equations [18]. Laboratory background PBDE concentrations were used as initial values because there is no natural background source of PBDEs to provide initial values. The advective-diffusive equations use sedimentation velocity, surface mixed layer (SML) depth, and mixing rates within and below the SML, parameterized from the ^{210}Pb model fits, to re-distribute the accumulation history and produce a modeled vertical profile [10]. This profile is displayed on the same figure as measured PBDE concentrations to allow comparison between model results and data. A contaminant history was determined by testing a variety of linear, exponential and constant increases and varying PBDE dates of entry to optimize the fit to the data.

The distribution of PBDEs in surface sediments surrounding the outfalls was interpolated among measured values by dividing each near-field region into segments based on dominant current direction, distance from the outfall, and organic carbon content and grain size (values previously determined by the CRD and Metro Vancouver [5, 22]). Fluxes into the sediments were calculated from estimated surface concentrations and sedimentation rates.

RESULTS AND DISCUSSION

Sediment accumulation rates and SML

All the cores except CRD-3 show evidence of a benthic mixed layer of 7 to 14 cm (**Fig. 2.2**; **Supplemental Data, Table 2.S1**), in agreement with previous studies in this area [3, 10]. There is a marked decrease of ^{210}Pb activity in the top 5 cm of the CRD-3 core (Fig. 2.2), likely caused by a slump of surficial sediment. Surface data in this core are therefore disregarded, and the sedimentation rate for CRD-3 is modeled using data points below the slump only. Sedimentation velocities range over 0.28 to 2.05 cm yr^{-1}, with associated accumulation rates of 0.21 to 2.04 g cm^{-2} yr^{-1} (Supplemental Data, Table 2.S1), similar to those reported previously for this area [3, 10]. Sedimentation rates are highest near the Vancouver outfall, partly because of the influence of the outfall itself and partly because of the proximity to the Fraser River. The ^{210}Pb profiles indicate approximately constant sedimentation rates over time at both Vancouver and Victoria sites except for site GVRD-3. Site GVRD-3 is at the location most strongly affected by the Vancouver outfall, and a satisfactory model to fit the data can be produced only by including an abrupt increase in sedimentation rate in 1989, plausibly resulting from the installation of the deep outfall in 1988.

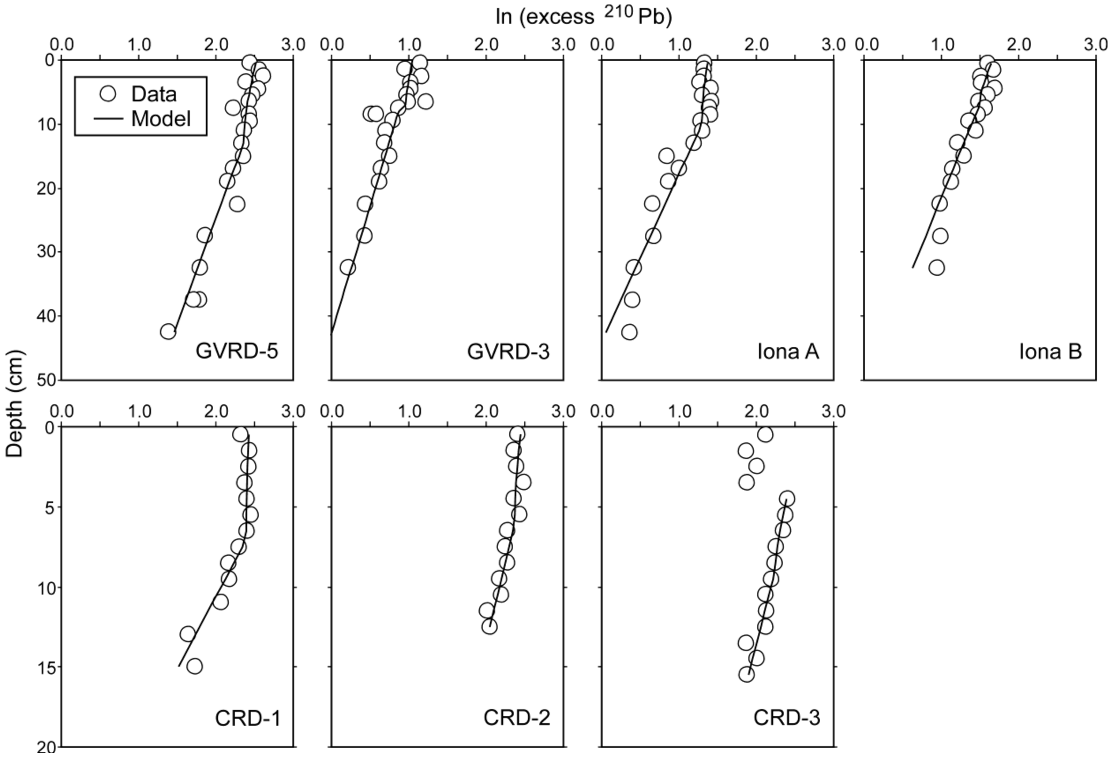

Figure 2.2 Profiles of the natural log of excess ^{210}Pb in sediment cores. Dots represent data and solid lines the accumulation and mixing model. The abrupt change in the GVRD-3 model at about 10 cm corresponds to a change in the sediment accumulation rate with the installation of the Iona Island outfall in 1988. The marked decrease in ^{210}Pb concentrations in the top 5.5 cm of the CRD-3 core represent a surficial slump and only the portion of the core below the slump was modelled.

Sediment accumulation rates near the Victoria outfall are lower, with a maximum of 0.79 g cm^{-2} yr^{-1} to the northwest of the outfall. Attempts to collect sediment cores in the region southeast of the Victoria outfall failed because of the high proportion of gravel (14 – 20%), which reflects the high energy of waters in Juan de Fuca Strait (Fig. 2.1). We conclude that the area southeast of the Victoria outfall is essentially non-depositional, based on the combination of an unusually high proportion of gravel, low percent fines (**Supplemental Data, Table 2.S2**) and very fast currents.

The Vancouver near-field depositional area stretches from 7 km north of the outfall to 4 km south and is approximately 2 km wide with little lateral dispersion [5, 23]. The plume from the Vancouver outfall is carried northwards and southwards by local tidal currents, which range from 0.1 to 0.15 m s^{-1} [24]. The Victoria near-field region of effluent deposition is smaller than Vancouver's and covers a circle of approximately 1 km radius surrounding the outfall pipe. Currents near the Victoria outfall are 1.0 m s^{-1}; [2], i.e., 10 times faster than those near Vancouver.

Surface sediment concentration and distribution of PBDEs

To increase the areal coverage of the distribution of PBDEs in surface sediments around the Victoria and Vancouver outfalls, sediment core data were augmented with PBDE measurements from 41 surface grab samples collected previously by Metro Vancouver and the CRD (Supplemental Data, Table 2.S2). Surface concentrations of PBDE range from 950 to 19,000 pg g^{-1} near Victoria and 1,000 to 10,900 pg g^{-1} near Vancouver. These concentrations fall in the mid to low range of ΣPBDEs measured in marine sediments exposed to municipal discharges worldwide (11,000 to 200,000 pg g^{-1} [25-28]). A map of surface PBDE concentration does not represent a simple snapshot in time in most coastal settings where depth of bioturbation and sediment accumulation rates vary. Benthic organisms mix older, less contaminated material toward the surface thereby making surface sediment less contaminated than it would be without mixing. Furthermore, the span of time represented by a surface sample depends on a combination of sediment accumulation rate and the depth of the surface mixed layer. We discuss spatial patterns in surface concentration first, but then, to calculate contaminant fluxes, we use the sedimentation rates estimated above.

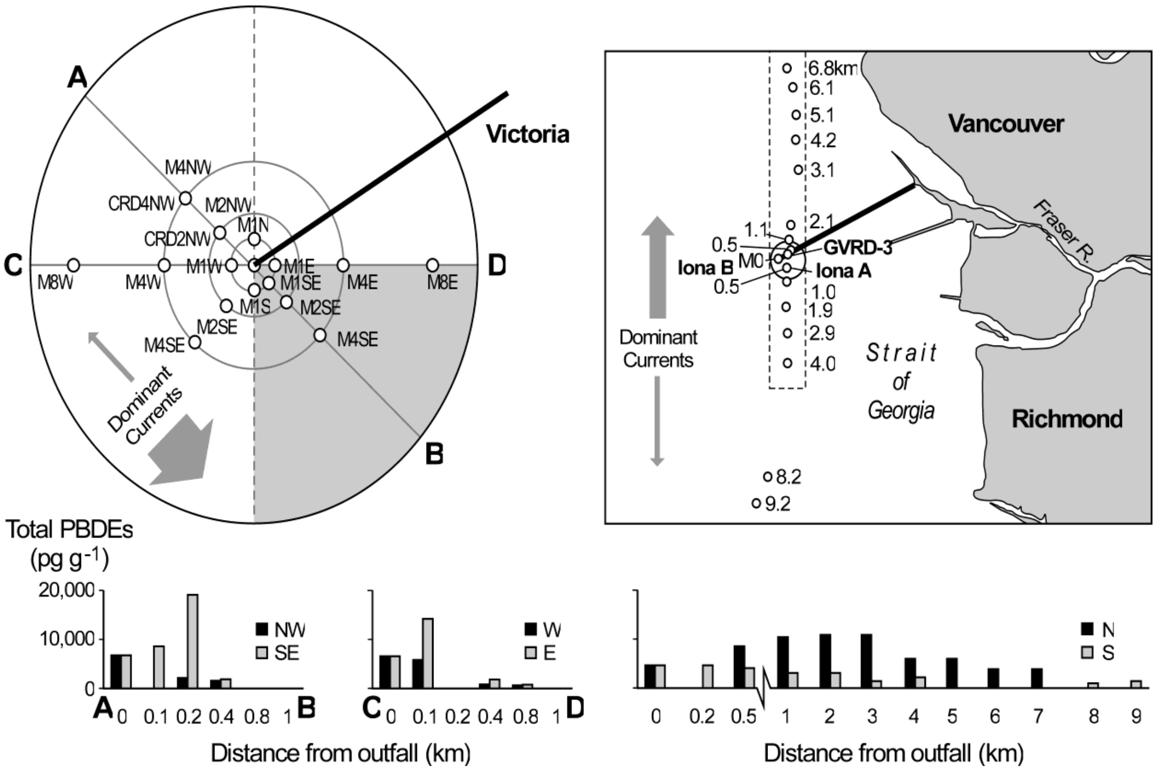

Figure 2.3 Distribution of Total polybrominated diphenyl ethers (PBDEs) in surface sediments surrounding the outfall pipes (thick black lines) in Metro Vancouver and Victoria. Maps shown are blow-ups of circles drawn in Figure 2.1. Samples lie mainly along northwest-southeast and west-east axis in Victoria and a north-south axis in Vancouver. Concentrations of PBDEs with distance from the outfall along these axes are shown in bar graphs. The two graphs have different scales reflecting the different receiving environments. The Victoria receiving environment covers a circle of 1 km radius. A circle of 1 km radius is drawn in the middle of the Metro Vancouver graph for comparison, but this receiving environment stretches from 7 km north to 4 km south (dashed line rectangle). The southeast quadrant of the Victoria receiving environment is likely non-depositional, represented by grey shading.

Dominant transport of particles from the Vancouver outfall and the nearby mouth of the Fraser River is to the north [22, 29], and this is also the direction of highest

ΣPBDE surface sediment concentration (**Fig. 2.3**). Concentrations increase to 3 km north, and then decline with distance from the outfall. Concentrations of PBDEs are lower to the south of the Iona outfall, and decline more quickly with distance (Fig. 2.3). To estimate PBDE distribution, the near-field region (22,000 m^2) is divided into homogenous sectors in the north and south. PBDE concentrations are interpolated among measured surface concentrations in each quadrant within circles 0 to 200 m, 200 to 400 m and 400 to 1000 m from the outfall and then within 1 km ellipses from 1 km to 7 km north, and 1 km to 4 km south (Circle of 1 km radius plus full near-field region are depicted in Fig. 2.3). At the Vancouver outfall, PBDE concentration is positively correlated with percent fines (particles of 0.002 - 0.063 mm) ($r^2 = 0.39$, n=16;) and with total organic carbon ($r^2 = 0.71$, n=16) (**Supplemental Information, Figure 2.S1**), implying that PBDE concentrations here are controlled by particle deposition as expected of such highly hydrophobic compounds. The contribution of BDE-209 to ΣPBDEs is uniform throughout the Vancouver near-field, further indicating consistent partitioning of PBDEs to sediment.

Fast currents near Victoria disperse particles quickly, resulting in dilution of the PBDEs to background concentrations within a 1 km radius of the pipe. Homogenous sectors within this 9,800 m^2 area are created by first dividing the circle into northwest, northeast, southeast, and southwest quadrants (represented by dashed lines in Fig. 2.3) based on grain size and dominant currents. Within each quadrant, PBDE surface concentrations are estimated in concentric circles at 0 to 200 m, 200 to 400 m and 400 to 1000 m distance from the outfall pipe. Sediments in the dominant current direction, to the southeast, have apparently little or no net accumulation. Sediment accumulation occurs mainly to the northwest, as indicated by a greater proportion of fine sediment, but the highest concentrations of PBDEs are found to the southeast and east, with significantly lower concentrations to the west and northwest (Fig. 2.3). This result was unexpected, as PBDE accumulation is generally associated with accumulation of fine particles, and near Vancouver PBDE concentrations correlate positively with percent fines, as mentioned above. Instead, the near-field region of the Victoria outfall shows a negative correlation of PBDEs with percent fines ($r^2 = 0.31$, n=26) while still showing a positive correlation with total organic carbon ($r^2 = 0.55$, n=24). We infer from these circumstances that PBDE concentrations at Victoria are not controlled by sediment deposition, but remain closely associated with organic carbon. The high concentration of PBDEs and total organic carbon in the southeastern quadrant, where there is no sediment accumulation (represented by shaded grey area in Fig. 2.3), implies that PBDE may deposit even where there is no net deposition of sediment. We propose that this is the result of either direct, fugacity-driven diffusion of PBDEs into the sediment caused by the high concentration of PBDEs in the plume passing over the sediment in this area, or biologically-mediated transport into sediments (i.e., by ingesting passing particles and retaining contaminants).

Depositional history of PBDEs

Modeled profiles of PBDEs in the present study indicate that these chemicals initially entered the system between 1973 and 1978, consistent with the known onset of worldwide production [9, 30, 31]. Cores collected at stations distant from the influence of outfalls (CRD-1 and GVRD-5) show a gradual increase in ΣPBDEs towards surface maximum concentrations of 300 to 800 pg g^{-1}, considerably lower than the concentrations measured in cores collected near the outfalls (surface concentrations 2,000 - 12,000 pg g^{-1}). Cores collected within the influence of the Vancouver outfall show a rapid exponential increase with time (**Fig. 2.4**). Although an exponential increase likely also occurred at the Victoria sites given worldwide increases in PBDE use, models using exponential or linear increases are equally capable of fitting the sediment profile data.

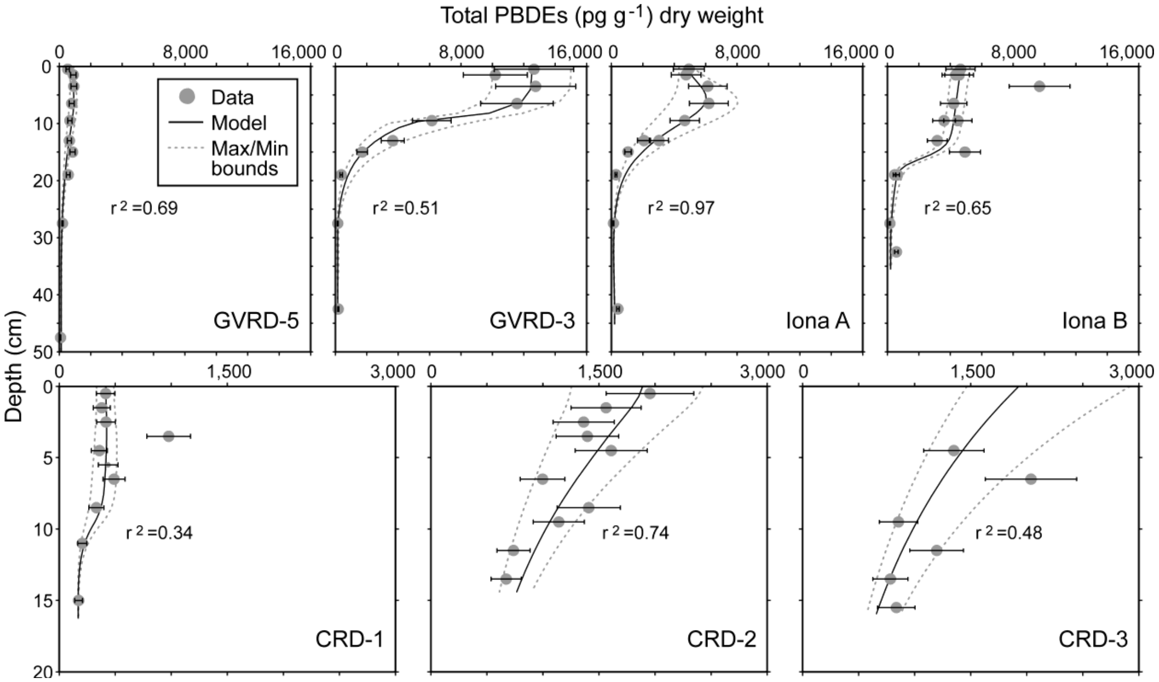

Figure 2.4 Concentrations of total PBDE in sediment cores (dots) and the mixing model (solid lines) show the depositional history of total PBDEs in these cores.

Concentrations of ΣPBDEs measured in effluent between the years 2004-2005 averaged 360,000 pg L^{-1} at the Victoria outfall [32] and 130,000 pg L^{-1} at the Vancouver outfall [22] (Table 2.1). The lower concentrations in Vancouver's effluent reflect primary treatment compared to simple screening of particles >6 mm before discharge at Victoria. Since PBDEs are actively sorbed on particles, they are partially transferred into the sludge that is removed from the outfall train during primary treatment.

Concentrations of ΣPBDEs in effluent at both outfalls is at the low end of the range reported for wastewater discharges across North America (14,000 - 900,000 pg L^{-1}) [6, 33-36] , probably reflecting the limited industrial input to the wastewaters of Victoria and Vancouver. The annual total flux of PBDEs to the ocean from effluent, calculated from the concentration in wastewater and the volume of wastewater discharged each year, is 24 kg yr^{-1} from the Vancouver outfall and 5 kg yr^{-1} from the Victoria outfall. The flux from the Vancouver plant is higher despite lower concentrations in the effluent because the volume of wastewater discharged, including stormwater runoff, averages 498 million L day^{-1} [22] compared to 40 million L day^{-1} in Victoria [32].

Flux of PBDEs to sediments

In all regions except the southeastern quadrant of the Victoria outfall region PBDE flux to sediments was calculated from the product of surface concentration and sediment accumulation rate (as determined by ^{210}Pb modeling). The apparent absence of sedimentation in Victoria's southeastern quadrant means that PBDE flux cannot be calculated, but must be estimated from the PBDE inventory. Since there is no sedimentation to the southeast of the Victoria outfall, we assume that most of the PBDE is concentrated in the surface mixed layer of the sediment, the depth to which it can be mixed by benthic organisms. Given a SML depth of 8 cm (mid-range for sediment cores collected in the Strait of Georgia and Juan de Fuca Strait), and assuming that the concentration of PBDEs measured in surface grab samples represents the entire homogenous mixed layer in this quadrant (Fig. 2.3), we calculate the inventory of PBDEs represented by each grab sample and the average value for the quadrant. To provide a rough estimate of average flux, we divide this inventory by 36 years, the number of years of PBDE deposition as determined by models at nearby stations (Fig. 2.4).

The flux of ΣPBDEs into near-field sediments is much greater at the Vancouver outfall (2,200 g yr^{-1}) than at the Victoria outfall (69 g yr^{-1}). This is also true for individual congeners, e.g., BDE-47, BDE-99 and BDE-209 (**Table 2.1; Supplemental Information, Table 2.S3**). Despite the larger flux of ΣPBDEs into sediments at the Vancouver outfall, surface concentrations are generally higher in Victoria sediments because of the different physical processes in the two receiving environments. Particle loading from the Fraser River and the resulting high sedimentation rates have previously been shown to dilute concentrations of mercury in sediment near Vancouver [37]. Given the high sedimentation rates that we measured near the Vancouver outfall, we conclude that sediment dilution is occurring here, and that organic material and contaminants from this outfall are rapidly buried and diluted by sediment from the Fraser River. Since there are no significant external sources of sediment near Victoria, organic matter is not diluted so rapidly by an influx of inorganic particles, leading to higher concentrations than would be expected from a comparison of flux or effluent concentration alone.

Proportion of discharged PBDE flux captured by local sediments

A higher proportion of ΣPBDE flux from deep sea outfall pipes into the ocean is accounted for in sediments within the near-field region of the Vancouver outfall (9%) than within that of the Victoria outfall (1%) (Table 2.1; Supplemental Data, Table 2.S3). Slower currents near Vancouver allow a greater proportion of PBDEs to accumulate, where they are quickly buried by sediment. Fast currents near the Victoria outfall cause rapid dispersal and resuspension of particles, and there is little or no accumulation of sediment, allowing 99% of PBDEs released via the outfall to be dispersed into the open ocean.

Table 2.1 The flux of PBDEs from wastewater and into sediment, and proportion of PBDEs from wastewater captured in sediment. Because of their different physical properties, the Vancouver receiving environment extends to 7 km while the Victoria receiving environment extends only to 1 km.

		Victoria				Metro Vancouver			
		Flux (g/yr)							
	Distance from outfall	Total PBDEs	BDE47	BDE99	BDE209	Total PBDEs	BDE47	BDE99	BDE209
Effluent		5300	1000	1000	2300	24000	4100	4800	12000
Sediment	0-200m	6	0.5	0.3	3	8	1	0.7	5
	200-400m	4	0.6	0.4	3	28	2	2.1	21
	400m-1km	59	2	6	67	230	13	11	158
	1km-6.8km	NA	NA	NA	NA	1940	106	230	1330
	Total Receiving Environment	69	3	7	73	2207	122	244	1514
Percent of Effluent flux accounted for in Sediment		Percent (%)							
		Total PBDEs	BDE47	BDE99	BDE209	Total PBDEs	BDE47	BDE99	BDE209
	0-200m	0.1	0.05	0.03	0.10	0.03	0.02	0.01	0.04
	200-400m	0.1	0.06	0.04	0.1	0.1	0.04	0.04	0.2
	400m-1km	1.1	0.2	0.6	3	1.0	0.3	0.2	1
	1km-6.8km	NA	NA	NA	NA	8.2	2.6	4.9	10.8
	Total Receiving Environment	1.3	0.3	0.7	3.2	9.3	2.9	5.2	12.4

Congener patterns

Twenty-three congeners were detected in the Vancouver and Victoria sediment samples (**Fig. 2.5**, Supplemental Data, Table 2.S2). At both outfalls the highly hydrophobic and particle-reactive BDE-209 is the dominant congener, contributing 62% of ΣPBDEs on average. Two congeners, BDE-47 (10±5%) and BDE-99 (8±5%), are the next most dominant. Modeled sediment profiles of these three congeners showed that their depositional history tracks the general trend indicated by ΣPBDE deposition (models not shown). The congeners BDE-49, -100, -153, -154, -206, -207 and -208 comprised 0.1-3% of the total, and all other congeners contributed <0.1% each (Fig. 2.5). The congener ratios remained consistent with depth in cores collected near Victoria in 2006, and in cores collected near Vancouver in 2003, consistent with data presented by Johannessen [10]. However, the two cores collected in 2009 (Iona A and Iona B; Supplemental Information Table 2.S1) show a shift in PBDE congener profiles downwards, with the contribution of BDE-209 to total PBDEs decreasing from 60% near the surface to 40% at depth. We attempted to correlate this decrease with the BDE-209 debromination products identified by La Guardia [38]. In the Iona A core, contributions from BDE-207 and -208 increased from 4% near the surface to 14% at depth. No other debromination products were detected. In the Iona B core, the proportion of BDE-207 and -209 did not increase. One other debromination product, BDE-201, was detected, but it only increased slightly with depth, from 0.2% to 0.7% of the total. More evidence is needed before we can assert that debromination is occurring in sediments in this region.

We use the ratios of individual congeners to ΣPBDEs in wastewater at the two sites to compare composition patterns. The greatest contributors were BDE-47 (20–30%), -99 (20–30%) and -209 (20–60%). Other congeners detected include BDE-100, -153, -154, -206, -207 and -208 and comprise 0.05 to 5% of ΣPBDE. The contribution from all other congeners is <0.05%. The overall pattern of congener ratios in wastewater is similar at both outfalls, indicating usage of similar materials containing PBDEs by the residents of both Victoria and Vancouver.

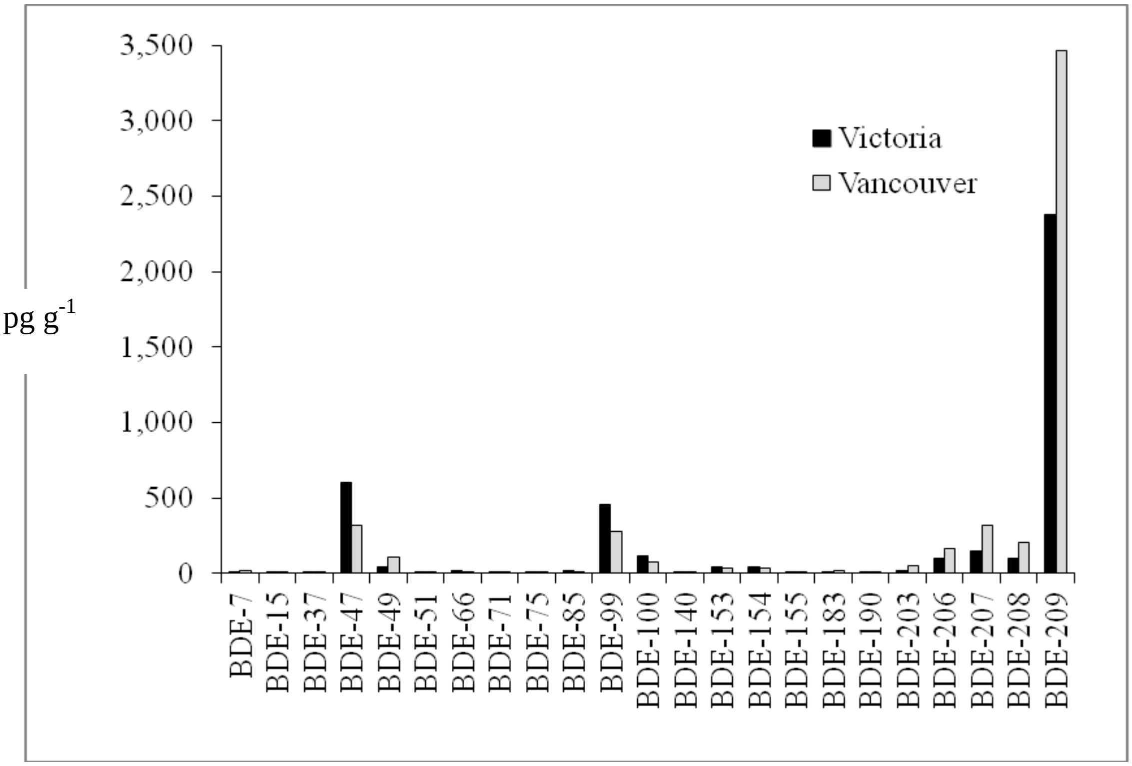

Figure 2.5 Average concentrations of 23 PBDE congeners in sediment near the influence of the Victoria and Vancouver outfalls.

Comparison of congener ratio patterns in wastewater and sediment shows that BDE-209 is present in much higher proportion in sediment than in effluent, while BDE-47 and -99 are present in lower proportion. Ten percent of BDE-209 released into the nearfield is captured in sediment at the Vancouver site and 2% at Victoria, compared to 0.2 to 2.8% of BDE-47 and 0.7 to 2.3% of BDE-99. This suggests that the relatively greater hydrophobocity of BDE-209 leads to a partial separation of congeners during deposition and transport. Despite its tendency to partition strongly to particles, most of this congener is dispersed far from the outfalls, likely in association with suspended fines.

The distribution of BDE-209 in sediment is very different at the two sites, showing a uniform distribution at Vancouver (60–74%) and a widely ranging distribution at Victoria (30–87%). In the southeastern quadrant of the Victoria site, BDE-209 accounts for only ~50% of ΣPBDEs, compared with its 60 to 86% contribution in all other quadrants. The low proportion of 209 in this part of the site is similar to that in the effluent (40%).

CONCLUSION

Clearly, treating all wastewater to the same level, regardless of local oceanographic conditions, will not result in a uniform environmental footprint. The low energy/high sedimentation environment near the Vancouver outfall traps nine times the proportion of effluent PBDEs captured by the high energy/low sedimentation environment near the Victoria outfall. This leads to a higher inventory of PBDEs in sediment near Vancouver, because the volume of PBDE discharged from Vancouver is five times that from Victoria, despite a lower concentration in wastewater. Contaminants discharged through the Vancouver outfall are rapidly diluted by inorganic sediment, while the low sedimentation rate near Victoria results in high surface PBDE concentrations. A particularly important finding of the present study is that the very high-energy environment to the southeast of the Victoria outfall appears to accumulate PBDEs despite not having net sediment accumulation. It seems likely that regions like this have been mistakenly neglected when accounting for particle-reactive sinks in coastal waters. We propose that contaminant accumulation without sedimentation likely occurs via direct fugacity-driven diffusion or by biological uptake either by filter feeders or by animals that forage in the water but reside in sediments part of the time.

Acknowledgement

We thank Cindy Wright, Tara Macdonald, Simon Grant, Amalis Riera, Katleen Robert, and the staff of Biologica Environmental Services Inc. for field assistance and suggestions. We appreciate the assistance at sea of the officers and crews of the *CCGS Vector*, the *Richardson Point* and the *John Strickland*. The present study has benefited from insight offered by Peter Ross and Brenda Burd. Chemical analyses were carried out by AXYS Analytical and the DFO LEACA lab. We thank an anonymous reviewer for providing succinct advice for clarifying a number of issues. Funding was provided by NSERC-IPS, the Capital Regional District, Metro Vancouver, Environment Canada and Fisheries and Oceans Canada.

Supporting Information Available

Table 2.S1 presenting parameters used for sedimentation modeling in sediment cores using 210Pb concentrations and an advective-diffusive mixing model. Table 2.S2 presenting grain size distribution, % TOC, ΣPBDEs, concentrations of the 23 PBDE congeners detected, from surface sediment samples collected and analyzed by Metro Vancouver and the CRD. Table 2.S3 presenting estimated ranges of PBDE flux to sediment for BDE-47, -99 and -209, complementing Table 2.1 in the main text that only contains ranges for ΣPBDEs. Figure 2.S1 presenting correlations of PBDEs in sediment with %TOC and % fines at Victoria and Vancouver.

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Supplementary Information

Table S1 Parameters used for sedimentation modelling in sediment cores using ^{210}Pb concentrations and an advective-diffusive mixing model

Sample	Station relative to outfall	Core Length (cm)	Surface Mixed Layer (cm)	Sedimentation velocity (cm/yr)	Sediment Accumulation rate (g/cm^2/yr)	Mixing rate in upper layer (cm^2/yr)	Mixing rate in lower layer (cm^2/yr)	Date Collected
Iona A	200m S of Iona	45	10	0.82 ± 0.02	0.76 ± 0.46	16	0.01	2009
Iona B	200m W of Iona	35	7	0.91 ± 0.04	0.81 ± 0.04	2	0.01	2009
GVRD-3	500m N of Iona	45	7	2.05 ± 1.53 † 1.24 ± 0.13 ††	2.04 ± 1.81 † 1.24 ± 0.12 ††	5	0.01	2003
GVRD-2	Reference	45	12	0.28 ± 0.004	0.21 ± 0.004	12	0.01	2003
GVRD-5	Reference	50	14	1.03 ± 0.025	0.58 ± 0.11	10	0.01	2003
GVRD-7	Reference	45	0	0.36 ± 0.007	0.36 ± 0.02	0.01	0.01	2003
CRD-2	200m NW of Macaulay	14	7	0.63 ± 0.03	0.80 ± 0.24	5	0.01	2006
CRD-3	400m NW of Macaulay	12	7	0.59 ± 0.04	0.78 ± 0.04	1	0.01	2006
CRD-1	Reference	16	7	0.28 ± 0.005	0.37 ± 0.007	20	0.01	2006

† After outfall installed (post-1996)

†† Before outfall installed (pre-1996)

Supplementary Information Table 2: Sediment Chemistry including percent organic carbon, grain size distribution and concentrations of PBDEs

Station	Distance from outfall pipe	Site	Latitude ^{o}N	Longitude ^{o}E	% TOC	% Gravel	% Sand	% Silt	% Clay	% Fines (Clay and Silt)
CRD pipe	0m	Victoria	48.4026	236.5896	3.8	20.4	60.9	12.2	6.5	18.8
CRDM1N	100m N	Victoria	48.4046	236.5901	1.2	3.6	59.3	26.9	10.4	37.3
CRDM1NE	100m NE	Victoria	48.4039	236.5913	2.4	6.7	61.1	23.0	9.3	32.3
CRDM1E	100m E	Victoria	48.4027	236.5911	4.5	12.2	62.7	17.6	7.5	25.1
CRDM1SE	100m SE	Victoria	48.4020	236.5908	7.4	18.3	60.6	13.9	6.3	20.2
CRDM1S	100m S	Victoria	48.4016	236.5894	1.7	14.2	58.6	19.8	7.5	27.3
CRDM1SW	100m SW	Victoria	48.4021	236.5886	3.0	10.0	63.2	19.1	7.7	26.8
CRDM1W	100m W	Victoria	48.4027	236.5884	2.6	4.9	68.2	18.8	8.3	27.0
CRDM1NW	100m NW	Victoria	48.4039	236.5887	0.8	0.5	61.5	28.9	9.2	38.1
CRDM2N	200m N	Victoria	48.4055	236.5904	1.2	9.0	55.0	26.5	9.6	36.1
CRDM2NE	200m NE	Victoria	48.4035	236.5925	1.6	6.1	60.5	23.5	10.0	33.5
CRDM2E	200m E	Victoria	48.4026	236.5926	2.0	11.7	62.7	18.4	7.3	25.6
CRDM2SE	200m SE	Victoria	48.4017	236.5917	2.8	15.1	62.5	15.9	6.5	22.4
CRDM2S	200m S	Victoria	48.4007	236.5891	1.5	4.1	59.6	27.0	9.3	36.3
CRDM2SW	200m SW	Victoria	48.4018	236.5874	1.1	2.6	62.5	25.7	9.2	34.9
CRDM2W	200m W	Victoria	48.4027	236.5867	4.6	2.1	66.5	23.1	8.4	31.4
CRDM2NW	200m NW	Victoria	48.4035	236.5874	1.0	1.0	61.7	27.8	9.5	37.3
CRDM4E	400m E	Victoria	48.4027	236.5949	2.1	10.5	59.2	21.7	8.7	30.4
CRDM4SE	400m SE	Victoria	48.4007	236.5940	1.9	14.2	59.8	18.3	7.7	26.0
CRDM4SW	400m SW	Victoria	48.4007	236.5850	0.8	2.3	62.1	26.2	9.4	35.6
CRDM4W	400m W	Victoria	48.4027	236.5842	1.0	0.4	62.7	26.6	10.1	36.7
CRDM8E	800m E	Victoria	48.4027	236.6002	2.1	8.9	57.9	22.5	9.8	32.3
CRDM8W	800m W	Victoria	48.4027	236.5785	0.8	0.6	64.5	26.2	8.9	35.1
CRDM8W	800m W	Victoria								
CRD2NW	200m NW	Victoria								

Table S2 cont.

Station	BDE-7	BDE-15	BDE-37	BDE-47	BDE-49	BDE-51	BDE-66	BDE-71	BDE-75	BDE-85	BDE-99	BDE-100
CRD pipe	9.17	2.58	0.77	1570	115	13.3	36.4	22.2	2.88	55.9	1360	321
CRDM1N	1.82	1.93	0.55	129	13.7	1.97	4.14	1.47	<0.32	3.86	96.4	28.0
CRDM1NE												
CRDM1E	9.77	3.70	0.54	695	92.1	9.65	20.10	10.20	1.34	23.50	624	149
CRDM1SE	18.9	6.69	0.80	861	119	11.60	20.20	13.80	1.33	25.30	611	153
CRDM1S	6.81	2.21	0.50	305	28.0	2.90	7.61	2.94	0.59	10.70	318	81.8
CRDM1SW												
CRDM1W	11.0	4.61	1.16	1430	108	9.77	40.50	16.40	3.20	67.80	1450	355
CRDM1NW												
CRDM2N												
CRDM2NE	6.04	2.19	0.29	251	31.6	3.54	6.63	4.08	0.41	6.57	165	45.6
CRDM2E												
CRDM2SE	7.22	4.35	1.29	4420	157	20.5	95.0	21.2	7.36	104	2770	768
CRDM2S												
CRDM2SW	5.32	2.25	0.32	202	20.2	2.28	6.36	2.32	0.33	5.55	142	39.6
CRDM2W												
CRDM2NW	2.75	1.88	0.30	152	14.7	1.90	4.90	2.06	0.38	4.28	103	29.3
CRDM4E	4.48	2.35	0.37	184	21.3	2.58	5.98	2.31	0.35	5.08	136	38.2
CRDM4SE	3.08	2.31	0.33	247	27.7	3.12	6.05	3.36	0.39	7.09	179	48.8
CRDM4SW	2.53	1.85	0.32	128	12.7	1.81	4.17	2.00	0.27	3.67	95.4	28.0
CRDM4W	1.74	1.75	0.40	130	12.8	1.71	4.60	1.75	<0.25	3.01	89.9	24.8
CRDM8E	2.85	2.02	0.30	171	11.5	1.46	4.91	1.83	0.22	3.94	115	32.4
CRDM8W	1.37	1.46	0.28	137	8.67	1.18	4.19	1.37	<0.26	3.99	99.5	27.1
CRDM8W	1.99	1.20	0.23	89	6.61	0.89	2.77	1.26	<0.21	2.42	61.2	17.6
CRD2NW	8.59	3.84	0.39	158	23.2	2.95	5.84	2.54	0.33	4.02	116	29.8

Table S2 cont.

Station	BDE-140	BDE-153	BDE-154	BDE-155	BDE-183	BDE-190	BDE-203	BDE-206	BDE-207	BDE-208	BDE-209	ΣPBDEs
CRD pipe	3.78	137	110	6.16	28.8	<3.04	48.2	146	231	151	2290	6830
CRDM1N	0.60	12.0	10.2	1.07	7.98	0.607	2.67	10.5	18.5	10.9	595	987
CRDM1NE												
CRDM1E	2.28	84.4	60.9	4.11	60.6	<8.88	91.4	642	806	552	10100	14200
CRDM1SE	1.93	62.7	52.4	4.14	28.9	3.98	41.4	170	184	136	4270	6970
CRDM1S	1.02	30.0	29.6	2.42	5.92	<0.52	2.47	13.1	19.0	11.9	956	1880
CRDM1SW												
CRDM1W	4.63	148	128	8.65	17.8	<1.22	6.13	28.9	60.5	34.8	1800	5850
CRDM1NW												
CRDM2N												
CRDM2NE	0.64	17.3	16.4	1.16	6.19	<1.81	8.08	31.2	59.3	39.7	903	1670
CRDM2E												
CRDM2SE	6.80	239	212	16.9	36.1	<8.37	73.1	334	609	463	8410	19000
CRDM2S												
CRDM2SW	0.77	16.4	15.1	1.52	4.59	<7.93	24.9	161	317	164	6980	8150
CRDM2W												
CRDM2NW	<0.10	12.0	10.8	1.06	3.98	<2.18	6.24	15.3	42.4	21.3	610	1070
CRDM4E	0.66	17.2	14.1	1.53	6.86	<2.36	10.9	54.8	116	59.1	2370	3100
CRDM4SE	0.58	19.2	16.1	1.41	11.0	<2.01	8.89	47.1	65.7	45.4	1070	1870
CRDM4SW	0.56	11.7	11.1	1.18	3.73	<0.96	4.80	19.7	32.2	18.9	562	974
CRDM4W	<0.89	10.2	9.42	0.94	4.31	<1.23	4.32	19.7	32.5	17.8	602	999
CRDM8E	0.51	14.7	14.00	1.94	6.15	<2.54	3.45	11.6	22.4	14.0	621	1090
CRDM8W	0.47	11.3	11.60	1.23	9.09	0.523	6.02	17.8	29.0	17.0	535	949
CRDM8W	0.28	8.61	7.97	0.98	3.10	<7.91	4.35	13.2	28.0	17.6	684	972
CRD2NW	0.50	13.0	11.5	1.64	4.91	0.926	5.41	44.5	74.4	45.6	822	1430

Table S2 cont.

Station	Distance from outfall pipe	Site	Latitude ^{o}N	Longitude ^{o}E	% TOC	% Gravel	% Sand	% Silt	% Clay	% Fines (Clay and Silt)
CRD4NW		Victoria								
CRDPB1	Ref Site	Victoria	48.3543	236.4892	0.8	0.2	73.9	18.4	7.6	26.1
CRDPB2	Ref Site	Victoria	48.3603	236.4930	NA	0.3	71.8	20.7	7.5	28.2
CRDPB3	Ref Site	Victoria	48.3605	236.4937	NA	0.3	61.6	28.8	9.7	38.5
Iona 1	6.8K N	Vancouver	49.2654	236.6991	0.7	0.3	72.2	19.3	8.2	27.5
Iona 2	6.1K N	Vancouver	49.2590	236.7021	1.0	0.9	48.4	36.8	14.0	51.0
Iona 3	5.1K N	Vancouver	49.2499	236.7037	1.4	0.0	19.9	60.9	19.2	80.1
Iona 4	4.2K N	Vancouver	49.2416	236.7039	1.4	0.0	13.1	63.6	23.3	86.9
Iona 5	3.1K N	Vancouver	49.2318	236.7050	1.6	0.0	6.3	66.0	27.8	93.8
Iona 6	2.1K N	Vancouver	49.2318	236.7050	1.6	0.0	6.8	66.4	26.8	93.2
Iona 7	1.1K N	Vancouver	49.2135	236.7012	1.6	0.0	10.1	64.0	25.9	89.9
Iona 8	0.5K N	Vancouver	49.2085	236.7000	1.5	0.2	14.1	61.2	24.3	85.0
Iona 9	0K	Vancouver	49.2038	236.6994	1.4	0.0	26.6	54.4	19.0	73.4
Iona 200S	0.2K S	Vancouver								
Iona 200W	0.2K W	Vancouver								
Iona 10	0.5K S	Vancouver	49.1993	236.6991	1.2	0.2	23.8	58.3	17.7	76.0
Iona 11	1.0K S	Vancouver	49.1946	236.6988	1.1	0.3	27.1	56.3	16.3	72.6
Iona 12	1.9K S	Vancouver	49.1865	236.6987	1.1	0.0	26.7	56.1	17.2	73.3
Iona 13	2.9K S	Vancouver	49.1778	236.6994	1.3	0.0	11.6	62.9	25.5	88.4
Iona 14	4.0K S	Vancouver	49.1678	236.6995	1.3	0.0	7.7	64.3	28.1	92.4
Iona 15	8.2K S	Vancouver	49.1304	236.6886	1.2	0.0	23.3	58.3	18.4	77.2
Iona 16	9.2K S	Vancouver	49.1214	236.6826	1.2	0.0	33.4	52.0	14.6	66.6

Table S2 cont.

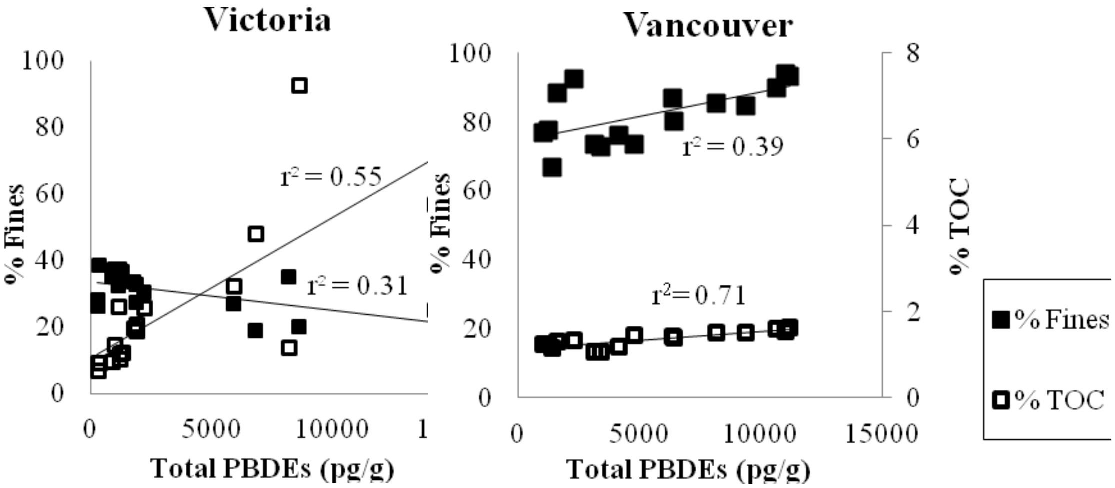
Station	BDE-7	BDE-15	BDE-37	BDE-47	BDE-49	BDE-51	BDE-66	BDE-71	BDE-75	BDE-85	BDE-99	BDE-100
CRD4NW	10.8	5.09	0.39	103	21.2	3.15	4.24	2.61	0.31	2.36	70.8	19.9
CRDPB1	0.261	0.392	0.126	23.6	2.03	<0.12	1.22	<0.21	<0.15	0.499	14.6	4.8
CRDPB2	0.247	0.387	0.179	27.7	2.25	0.309	1.14	0.417	<0.21	0.610	18.3	5.73
CRDPB3	0.55	0.561	0.154	31.8	2.59	0.286	1.16	0.478	<0.18	0.708	22.3	6.88
Iona 1	5.59	1.72	0.45	224	51.8	6.34	8.73	3.86	0.50	9.05	205	52.2
Iona 2	8.57	2.33	0.65	312	75.8	8.99	12.40	5.94	0.67	13.2	282	76.7
Iona 3	19.5	3.54	1.27	391	138	16.60	16.70	10.5	1.18	17.9	427	116
Iona 4	21.1	3.86	1.07	408	147	14.60	17.70	12.3	0.96	13.4	322	88.8
Iona 5	59.0	6.01	1.38	457	216	23.10	37.40	16.8	1.22	17.0	353	106
Iona 6	65.5	5.76	0.61	535	199	20.80	17.60	19.1	<0.18	19.7	424	115
Iona 7	70.6	4.74	0.71	529	203	20.70	23.20	19.0	1.55	18.8	436	122
Iona 8	50.3	4.36	<0.16	501	159	15.20	22.40	14.2	1.09	23.7	502	133
Iona 9	21.8	2.64	<0.10	322	99.6	9.66	13.30	10.4	0.86	11.5	294	77.1
Iona 200S	18.6	6.62	<0.32	438	146	<0.05	14.24	6.39	<0.16	<0.55	286	99
Iona 200W	19.6	8.92	<0.25	422	162	<0.14	14.32	5.88	0.47	<1.2	251	95
Iona 10	9.69	2.02	0.80	273	73.1	9.26	13.00	6.17	0.76	30.80	399	101
Iona 11	7.65	2.49	0.63	273	79.7	9.51	11.90	5.80	<0.08	9.83	238	66.2
Iona 12	6.36	2.06	0.43	163	47.4	7.53	7.15	2.99	<0.06	5.21	126	36.9
Iona 13	4.21	1.69	<0.19	130	36.2	6.04	5.07	2.32	<0.12	3.84	88.0	31.6
Iona 14	2.60	1.46	<0.25	135	29.1	4.44	5.62	2.32	0.27	4.63	107	33.0
Iona 15	1.15	0.774	<0.23	99.9	16.7	2.25	4.62	2.25	<0.15	3.83	88	25
Iona 16	0.865	0.782	0.253	106	16	1.85	4.73	1.8	0.251	4.6	102	26.1

Table S2 cont.

Station	BDE-140	BDE-153	BDE-154	BDE-155	BDE-183	BDE-190	BDE-203	BDE-206	BDE-207	BDE-208	BDE-209	ΣPBDEs
CRD4NW	0.40	8.29	8.67	1.55	3.51	0.615	4.57	35.3	68.4	47.3	973	1450
CRDPB1	<0.18	2.36	2.32	0.294	0.662	<8.53	0.89	4.09	6.65	4.58	199	273
CRDPB2	<0.21	2.68	2.6	0.356	0.859	0.184	1.31	7.20	10.1	8.04	204	300
CRDPB3	<0.20	4.8	4.3	0.553	1.44	<3.09	1.55	5.88	10.1	6.39	216	326
Iona 1	<1.1	21.6	21.0	3.23	<0.45	<3.21	26.4	140	267	149	2940	4140
Iona 2	<0.25	31.0	32.3	<0.16	10.1	3.42	31.8	132	293	181	2270	3780
Iona 3	<0.13	48.3	43.0	7.34	20.5	4.26	71.7	206	383	266	4070	6280
Iona 4	6.04	34.2	36.8	8.63	19.6	<4.22	76.6	231	585	348	3810	6210
Iona 5	8.17	40.3	44.2	<0.51	27.4	<4.35	110	413	914	499	7390	10400
Iona 6	<0.32	50.5	53.3	9.04	17.8	<5.36	44.6	461	779	530	7590	7970
Iona 7	9.68	52.9	52.9	<0.62	29.2	<6.29	84.9	286	469	307	7640	4620
Iona 8	<0.55	56.7	60.8	10.20	28.7	<3.59	77.1	227	386	259	5430	4220
Iona 9	6.26	31.5	35.0	<0.25	14.3	<5.36	43.1	145	227	154	3100	3440
Iona 200S	<0.56	34.6	46.1	9.43	32.4	<3.65	<6.5	29.8	173	141	3560	10960
Iona 200W	<0.62	31.4	44.1	10.01	23.1	<3.25	<4.5	<6.2	175	145	3369	10700
Iona 10	<0.91	54.1	45.1	<0.36	13.4	<4.56	<3.5	110	203	151	2720	3070
Iona 11	<0.85	26.6	26.4	<0.23	23.7	<2.32	30.4	102	166	107	2250	1730
Iona 12	3.29	13.8	14.5	<0.55	<1.2	<3.65	<5.1	119	206	146	2160	2290
Iona 13	<0.15	12.0	13.5	2.73	<1.5	<2.35	<4.2	59.2	105	81	1150	1000
Iona 14	<0.55	12.6	14.5	2.35	6.09	<2.35	25.8	95.1	188	111	1510	1450
Iona 15	0.647	10.5	10.2	1.17	4.33	<1.87	13	34.7	88.2	54	539	1000
Iona 16	<0.54	11.9	14.3	<0.42	5.03	<0.84	15.1	58.6	112	61.6	910	1450

Table S3: Flux of PBDEs in effluent from Vancouver and Victoria outfalls, flux of PBDEs into sediment at intervals within the receiving environment, and proportion of flux from effluent captured in sediment. Because of their different physical properties, the Metro Vancouver receiving environment extends to 6.8 km while the Victoria receiving environment extends only to 1 km.

	Distance from outfall	Victoria				Metro Vancouver			
		Flux (g/yr)							
		Total PBDEs	BDE47	BDE99	BDE209	Total PBDEs	BDE47	BDE99	BDE209
Effluent		5300 (4200-6400)	1000	1000	2300	24000 (19000-29000)	4100	4800	12000
Sediment	0-200m	4.0 (2.9-5.1)	0.4 (0.3-0.5)	0.3 (0.2-0.4)	2.3 (1.6-2.9)	8.1 (4.5-9.8)	0.8 (0.4-0.9)	0.65 (0.37-0.79)	5 (3-7)
	200-400m	3.9 (2.9-4.9)	0.5 (0.3-0.6)	0.4 (0.3-0.5)	2.5 (1.9-3.2)	31 (17-38)	1.8 (1.0-2.2)	2.1 (1.2-2.5)	18 (10-22)
	400m-1km	44 (32-55)	1.8 (1.3-2.3)	6.1 (4.7-7.7)	50 (38-63)	230 (120-280)	13 (7.1-16)	11 (6.0-13)	158 (80-190)
	1km-6.8km	NA	NA	NA	NA	1900 (940-2300)	100 (49-120)	98 (47-118)	1100 (600-1600)
	Total Receiving Environment	52 (38-65)	2.7 (1.9-3.3)	6.9 (5.2-8.6)	55 (42-69)	2200 (1100-2700)	116 (57-140)	111 (55-134)	1300 (770-1900)
		Percent (%)							
		Total PBDEs	BDE47	BDE99	BDE209	Total PBDEs	BDE47	BDE99	BDE209
Percent of Effluent flux accounted for in Sediment	0-200m	0.08 (0.05-0.1)	0.04 (0.03-0.04)	0.03 (0.2-0.4)	0.10 (0.07-0.13)	0.03 (0.02-0.04)	0.02 (0.01-0.02)	0.01 (0.01-0.02)	0.04 (0.02-0.05)
	200-400m	0.07 (0.05-0.09)	0.04 (0.03-0.05)	0.04 (0.2-0.5)	0.11 (0.08-0.14)	0.13 (0.07-0.16)	0.04 (0.02-0.05)	0.04 (0.02-0.05)	0.15 (0.08-0.18)
	400m-1km	0.82 (0.71-1.2)	0.17 (0.12-0.21)	0.6 (0.5-0.8)	2.2 (1.6-2.7)	0.97 (0.50-1.2)	0.32 (0.17-0.39)	0.23 (0.13-0.28)	1.3 (0.7-1.6)
	1km-6.8km	NA	NA	NA	NA	8.2 (4.0-9.9)	2.4 (1.2-2.9)	2.0 (1.0-2.5)	8.8 (6.3-16)
	Total Receiving Environment	0.97 (0.7-1.2)	0.24 (0.18-0.31)	0.7 (0.5-0.8)	2.4 (1.8-3.0)	9.3 (4.6-12)	2.8 (1.4-3.3)	2.3 (1.2-2.8)	10.3 (6.0-15)



Supplementary Figure 2.S1

Correlation of total PBDE concentration in surface sediments with percent fines (<0.06mm) and total organic carbon (TOC) near the Victoria and Metro Vancouver outfalls. Total PBDEs are correlated with % TOC at both locations (r2=0.55 and 0.71). Total PBDEs are positively correlated with % fines at Iona Island outfall (r2=0.39), but show a weak negative correlation at Macaulay Point (r2=0.31).

Chapter 3

PBDE and PCB accumulation in benthos near marine wastewater outfalls: The role of sediment organic carbon

PBDE and PCB accumulation in benthos near marine wastewater outfalls: The role of sediment organic carbon

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Abstract

Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) were measured in sediments and benthic invertebrates near submarine municipal outfalls in Victoria and Vancouver, B.C., Canada, two areas with very different receiving environments. PBDE concentrations in wastewater exceeded those of the legacy PCBs by eight times at Vancouver and 35 times at Victoria. Total PBDE concentrations in benthic invertebrates were higher near Vancouver than Victoria, despite lower concentrations in sediments, and correlated with organic carbon-normalized concentrations in sediment. Principal Components Analysis indicated that the uptake of individual PBDE congeners was largely determined by sediment properties (i.e. organic carbon, grain size), while PCB congener uptake was governed by physico-chemical properties (i.e. octanol-water partitioning coefficient). Our results suggest that the utility of sediment quality guidelines for PBDEs and likely PCBs benefit if they are based on organic carbon–normalized concentrations. In addition, where enhanced wastewater treatment increases the ratio of PBDEs to particulate organic carbon in effluent, nearfield benthic invertebrates may face increased PBDE accumulation.

Keywords

Polybrominated diphenyl ethers, Polychlorinated biphenyls, Organic carbon, Marine benthic invertebrates, Municipal outfalls

Introduction

Submarine municipal wastewater outfalls release a complex mixture of chemicals into the coastal ocean. Persistent and hydrophobic contaminants, such as polybrominated diphenyl ethers (PBDEs), partition strongly onto particles, settle in marine sediments, and may be bioaccumulated by benthic invertebrates [1]. PBDEs were introduced in 1987 and are used as flame retardants in a variety of household products [2]. They presently find their way into the environment mainly through wastewater [3] and are found in high concentration near points of entry [4]. PBDEs, therefore, can be used as tracers of the transport and partitioning of particle-active organic contaminants discharged by marine outfalls [1, 4].

The cities of Vancouver and Victoria on Canada's west coast discharge wastewater into contrasting physical environments, presenting the opportunity to evaluate the effect of the receiving environment on PBDE bioaccumulation. Vancouver's Iona Island outfall provides primary treatment to ~600,000 people and discharges at a depth of 80 m directly into the Strait of Georgia (Fig. 3.1) [5]. The Strait of Georgia is characterized by slow currents of 0.1 m s^{-1} [5] and high sedimentation rates (1–4 cm yr^{-1}) in the Central Strait [6]). The benthic invertebrate community structure in the Strait reflects the high percent fines (50–94%) and low proportion of gravel (<1%) [7]. Victoria's Macaulay Point outfall provides for ~120,000 people, screens wastewater to < 6 mm, and discharges into the Juan de Fuca Strait at 60 m depth [8]. Fast currents of 1.0 m s^{-1} [8] and low sedimentation rates, which characterize the Juan de Fuca Strait [9], lead to low percent fines (19–38%) and high proportions of gravel (1–20%) [8]. Many of the same benthic invertebrate species are found near both outfalls, particularly those tolerant to high

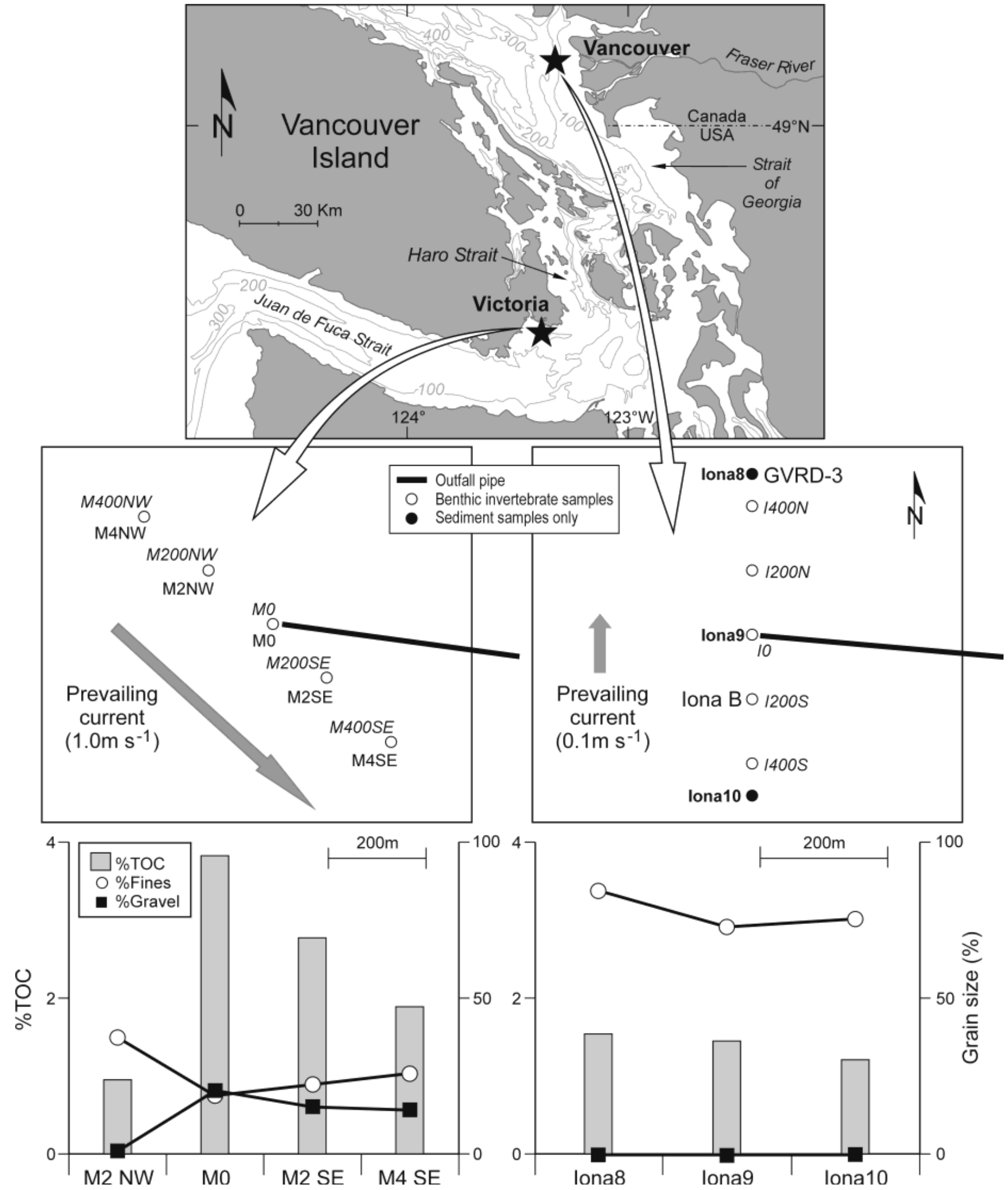

Figure 3.1 Three sets of samples were collected near the Victoria and Vancouver municipal wastewater outfalls. Benthic invertebrates (*italic labels*) in 2010, surface sediment (**bold labels**) 2006 to 2010, and sediment cores (plain labels) 2006 to 2009. Prevailing currents (1 cm = 0.2 m s^{-1}) and sediment property graphs demonstrate differences in the two receiving environments.

concentrations of organic carbon and other loadings from wastewater, but they may be present in different community compositions [5, 8, 10]. Hereafter we refer to the outfalls as the Vancouver and the Victoria outfall.

Wastewater regulations do not consider currents and sedimentation rates in specific receiving environments, although these factors control deposition of particle-active contaminants [11]. We present a case study investigating the role of the receiving environment to the accumulation of wastewater PBDEs by benthos, and compare results with uptake of the more globally-distributed and well-studied PCBs [12]. We also examine the significance of organic carbon in local sediment to PBDE bioaccumulation, and determine if PBDE distributions and sorting are similar to those of PCBs. We discuss implications for sediment quality guidelines (SQGs) and possible effects of different levels of sewage treatment.

Experimental

Sampling.

A 0.1 m^2 stainless steel Van Veen grab was used to collect benthic invertebrates and surface sediments in April, 2010, aboard the *Richardson Point* and the *John Strickland*. Twelve pooled benthic invertebrate samples, including two replicates, were collected at 10 stations along the axis of maximum current velocity and particle deposition from the Victoria and Vancouver wastewater outfalls pipes (Fig. 3.1). Because our goal was to compare contaminant concentrations between two regions with potentially different species assemblages, we combined all benthic invertebrate species found at each site into a pooled sample. Macroinvertebrates were separated from sediment using a 1 mm screen, then combined in glass jars, frozen immediately, and later analyzed for PBDEs, PCBs and % lipid. Identification and counting of species was conducted by Biologica

Environmental Services Ltd. [5, 8]. Biomass was calculated from average species weights for adults and juveniles. The Capital Regional District (CRD) and Metro Vancouver sampled sediment near the Victoria and Vancouver outfalls between 2006 and 2010 (Fig. 3.1), collecting the top 2 cm of sediment for analysis of PCBs, PBDEs, total organic carbon (TOC) and grain size (3 field reps per station) [5, 8]. 24 h composite wastewater samples were collected at the Victoria and Vancouver outfalls prior to discharge by the CRD (2004‒2010) and Metro Vancouver (1997‒2005) and analysed for PCBs and PBDEs [5, 8].

Lab Analysis.

PBDE, PCB and % lipid analyses for benthic invertebrates and sediment cores were conducted by the Laboratory of Expertise in Aquatic Chemical Analysis, Institute of Ocean Sciences, Department of Fisheries and Oceans (Sidney, BC, Canada). PBDEs and PCBs in sediment and wastewater were analyzed by Axys Analytical (Sidney, BC, Canada). Both laboratories followed EPA Method 1614 to measure 47 PBDE congeners by high resolution gas chromatography – mass spectrometry. Data quality objectives for precision were met with matrix spike recoveries (50-150%) and relative standard deviation of duplicate analyses (<50%) in all samples. Method accuracy was demonstrated by the use of certified reference materials. Laboratory blanks were <10% sample concentration for all sample batches except some PBDEs and PCBs in benthic tissues. Therefore a blank correction was applied to all tissue results. Determination of percent organic carbon and grain size in sediment was performed by ALS Group (Vancouver, BC, Canada) as per USEPA Method 9060A [13] and Forestry Canada Method NOR-X319 [14].

Data Analysis.

Congener-specific biota sediment accumulation factors (BSAFs) were calculated at each benthic invertebrate sample site as the ratio of the lipid-normalized contaminant concentration in benthic invertebrates to the organic carbon-normalized concentration in sediments as per [15, 16]. BSAFs were determined for PBDEs and PCBs and are plotted as a function of the log K_{ow} value of each congener. Only congeners detected in both sediment and biota samples were included. At three sites where locations of benthic invertebrate samples were not exactly paired spatially with sediment samples (I400N, I200N and I400S), sediment contaminant concentrations were interpolated from the nearest known values (Fig. 3.1b). Sediment PBDE concentrations at three locations have been previously published (CRD-2, Iona B, GVRD-3; Fig. 3.1) [6, 11]. Principal component analysis (PCA) was performed on Pirouette 4.0 using geometrically-centered, log-transformed PBDE and PCB congener concentrations. PCA pre-treatment included removal of congeners detected in <70% of samples, and replacement of “non-detect” results with detection limit values for congeners detected in 70 to <100% of samples.

Results and Discussion

Wastewater.

The concentrations of total PBDEs (ΣPBDEs) and total suspended solids are higher in Victoria wastewater (350±170 ng L^{-1} and 200±50 mg L^{-1}, respectively [8]), which undergoes only screening, than in Vancouver wastewater (130±115 ng L^{-1} and 83±12 mg L^{-1}) [5], which undergoes primary treatment (Table 3.1). The concentration of ΣPCBs is similar in Vancouver and Victoria wastewater (13±4 ng L^{-1} versus 10±2 ng L^{-1}). The ratio of ΣPBDEs to ΣPCBs is 8:1 at Vancouver and 35:1 at Victoria illustrating, as previously observed (e.g. [4]), that wastewater is presently a major conduit for PBDEs to enter the environment, while the largely relict PCBs are now distributed globally. Given a volumetric outflow of 500,000 m^3 d^{-1} from Vancouver compared with 40,000 m^3 d^{-1} from Victoria, the flux of ΣPBDEs released from the Vancouver outfall is 3.5 times greater than that from Victoria and ΣPCBs 16 times greater (Table 3.2).

Table 3.1 Comparison of receiving environment, wastewater, sediment and benthic invertebrate characteristics at submarine municipal wastewater outfalls in Victoria and Vancouver, B.C., Canada

	Parameter	Unit	Victoria	Vancouver
Receiving Environment	Sediment accumulation rate	g/cm^2 yr	0-0.80	0.76-2.04
	Current	m/s	1	0.1
	Outfall pipe depth	m	60	80
Wastewater	Population served	persons	120,000	600,000
	Flow	m^3/day	40,000	498,000
	TOC	mg/L	50	46
		kg/day	2,000	22,900
	TSS	mg/L	201	83
		kg/day	8,000	41,000
Sediment	TOC	%	2.4	1.4
	Fines	%	29.4	78.1
Benthic Invertebrates	Lipid	%	1.5	0.4
	Moisture	%	71.1	84.5
	Abundance	Individuals /0.1m^2	768	647
	Biomass	g/0.1m^3	18	26
	Annelids	%	56	36
	Arthropods	%	23	0.2
	Molluscs	%	18	62
	Echinoderms	%	0	0

Upon discharge, the initial distribution of wastewater in the receiving environment is dictated by ambient current velocity and direction. Since currents at the Vancouver outfall are relatively slow (0.1 m s^{-1}), more of the discharged particles and associated chemicals settle in nearby sediments. Currents at the Victoria outfall are approximately 10 times greater, particles are transported quickly from the outfall, and fewer particles settle in near-field sediments around the outfall pipe [11].

Sediment.

Dry weight concentrations of PBDEs are similar in sediment near Vancouver and Victoria [5, 8]. However, TOC-normalized concentrations are approximately twice as high near Vancouver, because sediment TOC is lower (Table 3.1). The concentrations of PBDEs, PCBs, and TOC in Victoria sediment are higher than expected compared to Vancouver based on flux from wastewater (Table 3.2). This is because the large flux of inorganic particles from the Fraser River near Vancouver buries wastewater particles quickly and dilutes PBDEs and other contaminants. Little inorganic sediment accumulates near Victoria, so although <1% of wastewater derived contaminants are deposited in nearby sediment, these remain relatively undiluted [11].

Benthic Invertebrates.

The mean concentration of ΣPBDEs in benthic invertebrate communities near the Vancouver outfall is 2.4 times higher than near the Victoria outfall, despite the lower sediment and wastewater concentrations at the former. ΣPCB concentrations in benthos are approximately equal at the two sites (Fig. 3.2). Neither ΣPBDE nor ΣPCB in benthic invertebrate tissue correlates with dry weight contaminant concentration in sediment

(r^2=0.00001, p=0.99, n=9). Instead, we find that ΣPBDEs in benthos near both outfalls correlate with TOC-normalized sediment concentrations (r^2 = 0.63, p=0.033, n=7), except at two sites near the end of the Victoria outfall pipe, M0 and M200SE, where the high energy environment inhibits particle deposition. PCB concentrations in benthos correlate with TOC-normalized sediment concentrations at Vancouver (r^2 = 0.90, p=0.20, n=3) and Victoria (r^2 = 0.69, p=0.17, n=5), but, unlike the case for PBDEs, we could not incorporate samples from the two sites into a single linear relationship.

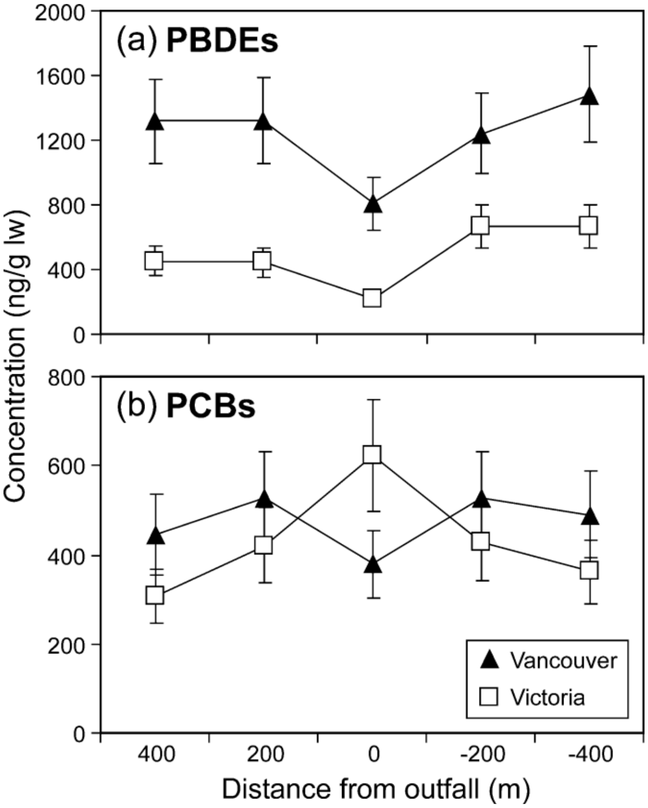

Figure 3.2 Concentrations of PBDEs (a) in benthic invertebrates are higher near submarine outfalls in Vancouver than Victoria. Concentrations of PCBs (b) are similar, and lower than PBDEs. Note different scales for (a) and (b). Sites are along the tidal current axis (north-south in Vancouver, northwest-southeast in Victoria) with distance from the outfalls. Variation is evident across and within sites.

That limits our ability to predict PCB uptake by benthos from TOC-normalized concentration in sediment. PBDE concentration differences in the benthos are largely due to sediment TOC: about half as much organic carbon is available in Vancouver sediment, so despite approximately equal dry weight ΣPBDE sediment concentrations at the two sites, benthic invertebrates near Vancouver may have to ingest twice as much sediment to process the same amount of organic carbon. In the process they will ingest twice the load of particle active contaminants. ΣPBDEs in benthic invertebrates also correlate with percent fines (r^2=0.87, p=0.0022, n=7), but this is not true for ΣPCBs (r^2=0.016, p=0.78, n=7).

Abundance (individuals m^{-2}) of benthic invertebrates is similar near the Victoria and Vancouver outfalls, but biomass is approximately 50% higher near Vancouver (Table 3.1). Many of the same benthic invertebrate species are found in both regions e.g. *Axinopsida serrica*, *Capitella capitata*. However Vancouver's benthic community is dominated by molluscs and animals with larger body sizes [5], while Victoria's benthic community contains more small annelids [8] and a higher average lipid content (Table 3.1). Preliminary data from the Vancouver outfall site (data not shown) indicate that the larger animals are more highly contaminated than the smaller, so the higher PBDE concentration in the community near Vancouver may in part reflect the larger average size at this site. Species-specific variation in accumulation of PBDEs has been reported previously [16, 17] and may contribute to the different degrees of contamination of the benthos near Victoria and Vancouver. Benthic species composition is affected by substrate, likely explaining the correlation between PBDEs in benthos and sediment grain size.

BSAFs.

Despite differences in benthic invertebrate species composition among sites, BSAFs reveal similarities in chemical partitioning between Victoria and Vancouver. BSAFs plotted against octanol-water partition coefficient (K_{ow}) values [18, 19] show a parabolic relationship for both PBDEs and PCBs (Fig. 3.3a-d). PCB BSAFs increase from $\log K_{ow}$ ~5.5 to a maximum at $\log K_{ow}$ ~7, and then decrease (Fig. 3.3c,d), consistent with PCB congener accumulation behaviour reported elsewhere e.g. [20, 21]. PBDE accumulation also displays a parabolic relationship with K_{ow} (Fig. 3.3a,b), but BSAFs are approximately twice as high as for PCBs at all Vancouver and Victoria sites except M0 and M200SE (Fig. 3.3a). The higher BSAFs for PBDEs than for PCBs may be due to the difference in the present supply of these two contaminants: PBDE concentration in sediments is increasing, while PCBs are decreasing [6], such that benthic invertebrates have a greater opportunity to accumulate the currently-discharged PBDEs. It is also possible that some benthic invertebrates are accumulating PBDEs directly from the water column, increasing the apparent PBDE BSAF. Overall, PBDE and PCB BSAFs are different from each other (Fig. 3.3), and this difference is evident at both sites, implying that BSAFs are driven by factors other than just site specific physical or biological differences.

Within site variation.

Benthic invertebrate communities at the end of each outfall pipe (M0 and I0; Fig. 3.1) have lower concentrations of ΣPBDEs and higher lipid content than sites farther from the pipe (Fig. 3.2). In addition, PBDE BSAF values at M0 and M200SE, which likely receives almost the same particle input as M0 due to the prevailing current direction (Fig. 3.1), are lower than at all other sites (Fig. 3.3a). Growth dilution explains the relatively low PBDE concentrations in benthic invertebrate communities at these three sites, something that has previously been proposed to explain the lower BSAFs for mussels living at the end of the nearby Clover Point outfall pipe in Victoria [15].

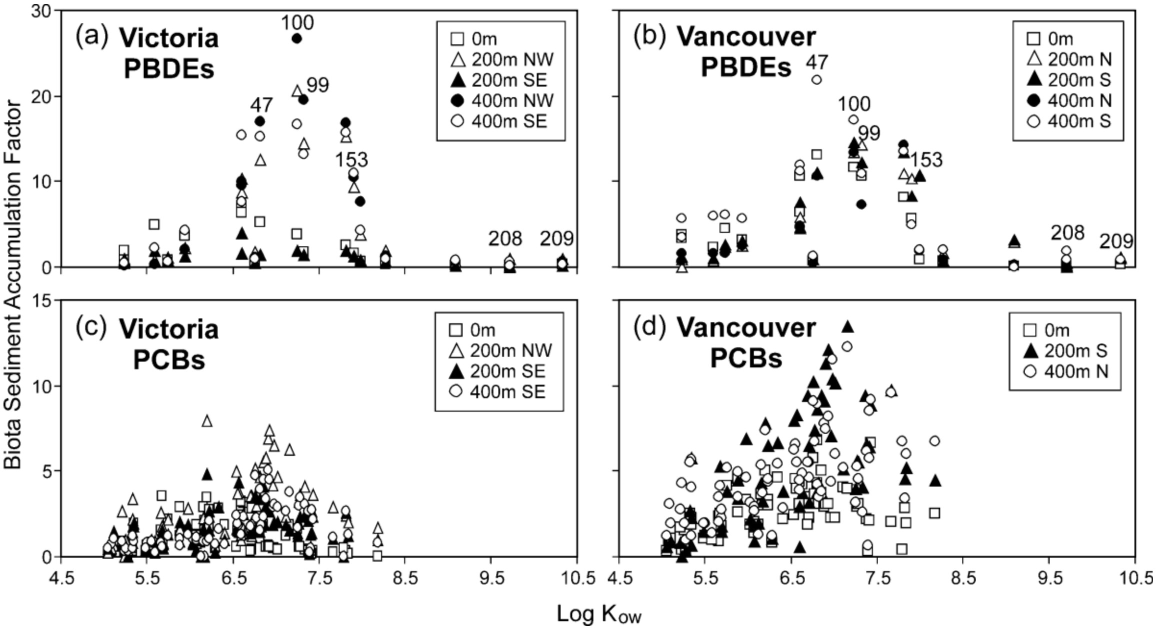

Figure 3.3 Biota sediment accumulation factors for PBDEs (a,b) are higher than for PCBs (c,d), except at M0 and M200SE (a). Sites are along the tidal current axis at the Victoria and Vancouver submarine outfalls.

Unlike all of the other Vancouver and Victoria sites examined, M0 and M200SE exhibited no correlation between ΣPBDEs in benthic invertebrate and TOC-normalized ΣPBDEs in sediment. The rapid transport of lighter congeners from the end of the pipe is a possible explanation. At M0, closest to the pipe, no PBDE congeners are correlated with TOC-normalized levels. However, 200 metres away at M200SE, the heavier BDE-100 and BDE-209 congeners are correlated with TOC-normalized sediment concentrations, while lighter congeners e.g. BDE-47 are not. Sediment at M0 and M200SE sites have higher TOC (3.8 and 2.8%) and lower percent fines (18.8 and 22.4%) than average (Table 3.1), demonstrating their physical difference from all other sites.

Table 3.2: The six highest major PBDE and PCB congeners detected in wastewater from the Vancouver and Victoria municipal wastewater outfalls (B.C., Canada) are presented as mean (standard deviation). Daily loading from wastewater (g day^{-1}) and concentrations in sediment (ng g^{-1} dw^{-1} TOC^{-1}) and benthic invertebrates (ng g^{-1} $lipid^{-1}$) are shown alongside each congener's respective

Matrix		Wastewater (g day^{-1})				Sediment (ng g^{-1} dw^{-1} TOC^{-1})				Benthic Invertebrates (ng g^{-1} $lipid^{-1}$)		
Site		Vancouver outfall		Victoria outfall		Vancouver outfall		Victoria outfall		Vancouver outfall		Victoria outfall
Congener	Rank	(n=17)	Rank	(n=9)	Rank	(n=6)	Rank	(n=6)	Rank	(n=6)	Rank	(n=6)
BDE-209	1	14.3 (10.5)	1	6.02 (4.38)	1	325 (100)	1	108 (96)	3	221 (157)	4	36.1 (23)
BDE-99	2	13.1 (2.71)	3	2.53 (0.96)	3	27.6 (6.4)	3	28.7 (36)	2	247 (60)	2	111 (40)
BDE-47	3	11.4 (2.40)	2	2.78 (1.08)	2	30.9 (5.8)	2	42.0 (58)	1	351 (93)	1	202 (23)
BDE-100	4	2.51 (0.55)	4	0.562 (0.23)	8	7.87 (1.2)	4	7.69 (10.0)	4	101 (28)	3	44.8 (11)
BDE-153	5	1.31 (0.9)	7	0.240 (0.21)	10	3.39 (1.0)	7	2.73 (3.1)	7	18.6 (4.9)	7	8.81 (2.7)
BDE-208	6	0.992 (1.22)	5	0.326 (0.33)	4	17.0 (8.5)	5	5.66 (5.5)	14	4.15 (3.5)	18	0.409 (0.15)
ΣPBDEs	–	49.2 (18.3)	–	14.0 (6.8)	–	456 (129)	–	213 (219)	–	1112 (327)	–	457 (67)
CB-110/115	1	0.367 (0.10)	1	0.017 (0.004)	1	4.13 (0.83)	2	12.6 (8.0)	5	19.8 (6.8)	5	23.4 (12)
CB-52	2	0.298 (0.10)	2	0.015 (0.004)	8	2.39 (1.0)	9	8.24 (4.8)	8	11.5 (4.1)	9	13.3 (7.8)
CB-118	3	0.265 (0.07)	6	0.012 (0.003)	2	4.09 (1.8)	4	11.0 (6.9)	6	18.9 (5.6)	4	24.6 (13)
CB-147/149	4	0.232 (0.04)	5	0.013 (0.005)	10	2.23 (1.5)	7	9.13 (7.3)	3	21.4 (7.9)	6	19.4 (5.3)
CB-153	5	0.227 (0.04)	3	0.014 (0.004)	3	4.06 (1.1)	5	10.5 (8.7)	1	38.0 (13.5)	1	33.9 (4.4)
CB-83/99	6	0.170 (0.04)	9	0.009 (0.003)	18	1.64 (0.9)	11	6.06 (3.1)	9	12.3 (4.7)	8	15.2 (4.1)
ΣPCBs	–	6.31 (1.3)	–	0.415 (0.01)	–	86.8 (13)	–	227 (150)	–	466 (172)	–	428 (119)

PBDE and PCB congener patterns.

Of the 47 PBDE and 209 PCB congeners analyzed, we detected 37 PBDEs in all matrices, 147 PCBs in wastewater and sediment, and 127 PCBs in benthic invertebrates (Supplementary Information, Table 3.S1). More PCB than PBDE congeners are detected, because all of the 209 theoretically possible PCB congeners were produced, historically, compared to only approximately 47 PBDEs. Ranking PBDE congeners in order of their contribution to total concentrations shows that PBDEs are dominated by ~BDE-209>99>47>100, which together contribute >80% of ΣPBDEs in all of the matrices (Table 3.2). PCB rankings vary more widely among matrices than do the PBDEs, with no single PCB congener contributing >10% to the total, and most contributing <5% (Table 3.2).

Despite the dominance of BDE-209, -99, -47 and -100 in all matrices, patterns differ markedly among wastewater, sediment and benthic invertebrate samples (Table 3.2). The ratio of BDE-209 to ΣPBDE is substantially higher in sediment (50–70%) than in wastewater (30–40%), reflecting its strong partitioning onto particles (log K_{ow} = 9.4), but is lower in benthic invertebrates (10–20%), because it is not easily bioaccumulated [22, 23]. The contributions of the less brominated BDE-47, -99 and -100 are higher in benthic invertebrates than in wastewater, reflecting their more bioaccumulative nature [22].

Principal Components Analysis of PBDE congener patterns identify three sample clusters on the scores plot - wastewater, sediment and benthic invertebrates, thereby providing a clear separation on the basis of sample patterns among these matrices (Fig. 3.4a). The lighter, more bioaccumulative, congeners (i.e. BDE-154, -47, -100) plot in the same region as the benthic invertebrate scores, while the heavier BDE-206, -208 and -209 plot far from this matrix (Fig. 3.4b). However, physico-chemical properties (log K_{ow}) are not the primary factors controlling PBDE sorting in benthic invertebrates. Loadings correlate with log K_{ow} (r^2=0.76, p=0.00001, n=18) for the second PC factor but not the first PC factor, indicating that chemical properties secondarily influence PBDE congener sorting. The first PC factor accounts for 38% of the variability and benthic invertebrate scores along this axis correlate with TOC (r^2=0.58, p=0.0040, n=12) and % fines (r^2=0.47, p=0.0047, n=14). The second PC factor explains 33% of the variability and benthic invertebrate scores along this axis correlate with % lipid (r^2=0.69, p=0.00014, n=15) and benthic community structure, based on correlation with the percent of annelids (r^2=0.58, p=0.0027, n=13). Sediment TOC and % fines appear to be the main factors influencing PBDE congener patterns in tissue, adding to our observations that PBDE bioaccumulation is controlled by sediment organic carbon. Biota properties and log K_{ow} are secondary.

Similar to those of the PBDEs, PCB congener patterns also group by matrix and receiving environment (Fig. 3.4c). However, the separation among matrices is not as clear for the PCBs, likely reflecting a more even distribution among reservoirs in the coastal environment for this legacy compound [1, 24]. The first PC factor explains 38% of the variability, and loadings for this factor correlate with K_{ow} (r^2=0.48, p=0.00001, n=68) (Fig. 3.4d). No other parameter correlates with the first factor. The second PC factor explains 18% of the variability and benthic invertebrate scores for this factor correlate with % lipid (r^2=0.87, p=0.00001, n=14), % fines (r^2=0.57, p=0.0018, n=14) and the proportion of annelids in the benthic invertebrate communities (r^2=0.52, p=0.0077, n=12). Physico-chemical properties appear to represent dominant factors influencing

PCB sorting in benthic invertebrates, with sediment and biological properties playing a secondary role (Fig. 3.4c,d).

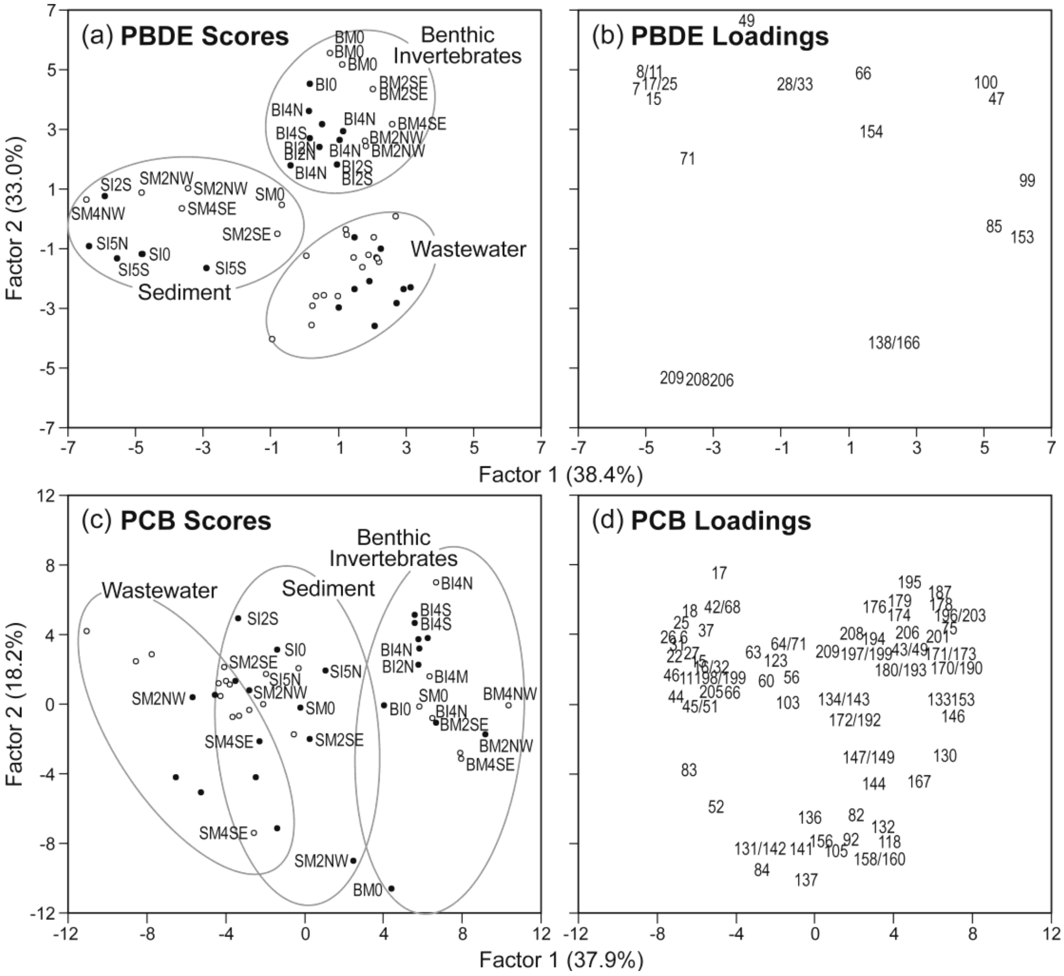

Figure 3.4 Different PBDE (a,b) and PCB (c,d) compositions between matrices are shown by PCA sample scores and loadings. Unlabelled sample points represent wastewater. "B" represents benthic invertebrates and "S" sediments. "I" represents the Iona (Vancouver) outfall (open circles), and "M" the Macaulay (Victoria) outfall (closed circles).

Sediment quality guidelines (SQGs) approach the problem of risk management using contaminant concentrations in sediment as predictors of toxicity [25-27]. Past evaluation of PBDEs relied on predictions made from SQGs developed for PCBs [15]. However, this approach to dealing with PBDEs neglected the environmental factors that result in very different accumulation profiles between the PCBs and PBDEs. Our findings of the important role for TOC in determining biological uptake of PBDEs from sediment, coupled with recent Canadian SQGs for PBDEs which are corrected on the basis of 1% TOC [28], underscores the importance of considering sediment properties when predicting bioaccumulation of this class of chemicals. We find here that although accumulation of PBDEs in sediment is controlled by the receiving environment alone, accumulation in benthic invertebrates is controlled by two factors: the physical environment through its effect on sediment accumulation and species assemblage, and sediment TOC content.

Enhanced sewage treatment at Vancouver compared to Victoria apparently decreases the flux of particulate organic carbon more than that of PBDE [30], which is manifest as a higher ratio of PBDEs to TOC near Vancouver. Since conventional treatment technologies do not break down persistent chemicals, selective removal of TOC then leads, counter-intuitively, to higher benthic bioaccumulation. Source control (i.e. regulating persistent chemicals, or preventing their entry into municipal wastewater by prohibition) or specific treatment to remove these compounds may be required to limit accumulation in marine biota.

Acknowledgements

We thank Tara Macdonald, Brenda Burd, Cindy Wright, Shirley Lyons, Simon Grant, Amalis Riera, Katleen Robert, Farida Bishay and the staff of Biologica Environmental Services Inc. for field assistance and project insights. We appreciate the assistance at sea of the officers and crews of the *CCGS Vector,* the *Richardson Point* and the *John*

Strickland. Chemical analyses were carried out by Axys Analytical and the Laboratory of Expertise for Aquatic Chemical Analysis (LEACA; Fisheries and Oceans Canada). Funding was provided by NSERC-IPS, the Capital Regional District, Metro Vancouver, Environment Canada and Fisheries and Oceans Canada.

Supporting Information Available

Table S1 listing detection rates of PBDE and PCB congeners in wastewater, sediment and benthic invertebrates near the Victoria and Vancouver outfalls in British Columbia, Canada.

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Supplementary Information

Table 3.S1: PCB and PBDE congeners were detected in wastewater, sediment and biota collected in the vicinity of the Victoria and Vancouver wastewater outfalls in British Colombia, Canada

	Victoria			Vancouver		
PBDEs:	Effluent	Sediment	Biota	Effluent	Sediment	Biota
PBDE Congeners Detected	38	35	35	36	36	39
Congeners in 100% of samples	28	27	27	21	19	24
Congeners in 70 - 99% of samples	3	3	0	6	3	6
Congeners in >0 - 70% of samples	7	5	8	9	14	9
PCBs:	Effluent	Sediment	Biota	Effluent	Sediment	Biota
PCB Congeners Detected	149	154	129	146	102	129
Congeners in 100% of samples	109	105	106	123	88	100
Congeners in 70 - 99% of samples	18	34	12	6	9	13
Congeners in >0 - 70% of samples	22	15	11	17	2	16

Chapter 4
Conclusions

Fate and distribution of PBDEs in surface sediments

Comparison of municipal outfall discharges in two contrasting coastal regions demonstrates that the fate of PBDEs, PCBs and possibly other particle-reactive contaminants depends on the local receiving environment. At both the Vancouver and Victoria outfalls (Fig.2.1 and 3.1, Appendix A), PBDEs released into the ocean are predominantly dispersed far from their source. This dispersion is probably in association with the fine particulate fraction. A higher proportion of PBDEs is captured by the low energy-high sedimentation environment at the Vancouver outfall pipe (9%) compared to the high energy-low sedimentation environment at the Victoria outfall pipe (1% captured). PBDE congeners partially separate during transport from outfalls, and the heaviest congener, BDE-209, is captured in highest proportion by near-field sediments (Table 2.1, Appendix B). The lighter congeners e.g. BDE-47 and -99 are subject to the longest range of transport and thus a lower proportion of these congeners are deposited in the near-field adjacent to the outfalls. One potential source of error when comparing PBDE fluxes in sediment to those from wastewater is that the concentration of PBDEs in wastewater shows high seasonal variation [10, 18]. In this study samples were collected during each season, with the average used for flux calculations (Appendix C). More study into the effect of these seasonal variations on sediment and benthic accumulation would be useful.

The distribution of PBDEs within near-field surface sediments surrounding municipal outfall pipes is controlled by sediment deposition near the Vancouver outfall, where, as observed in other locations e.g. [33, 52], ΣPBDEs follow the distribution of fine-grained sediment. The opposite is true near Victoria, where the concentration of ΣPBDEs is inversely correlated with % fines. PBDEs have been previously shown to be associated with organic carbon throughout the sewage treatment process [5], and PBDEs in the Victoria and Vancouver region remain closely associated with organic carbon once settled in sediment. Concentrations of both PBDEs and organic carbon are high to the southeast of Victoria’s outfall pipe, where the substrate contains a large amount of gravel and a small proportion of fine material. Accumulation of a particle-active contaminant in an area with no net sediment accumulation is a novel finding. In general, this means that non-depositional regions should be included when calculating sediment reservoirs of particle-active contaminants in coastal waters.

In the Victoria site, I propose that PBDE deposition without sedimentation is the result of one of two mechanisms. It could be the result of direct, fugacity-driven diffusion of PBDEs into the sediment caused by the high concentration of PBDEs in the overlying plume of wastewater e.g. [53]. This diffusion would be accelerated by benthic invertebrates living in the underlying sediment as they mix high concentrations of PBDEs downwards. Alternatively, the benthic invertebrates could mediate the transport of these contaminants into sediments by ingesting particles, retaining and concentrating the lipophilic PBDEs, and then ultimately passing the PBDEs to surface sediments.

Physical processes such as sedimentation and current velocity have a pronounced effect on PBDE surface sediment concentration, regardless of PBDE flux to the region. Despite the larger flux of ΣPBDEs landing in sediments near the Vancouver outfall (2,200 g yr^{-1}) than near the Victoria outfall (69 g yr^{-1}), surface concentrations are generally lower in Vancouver sediments. Particle loading from the Fraser River leads to the high sedimentation rates measured near the Vancouver outfall, and I conclude that contaminant dilution by inorganic sediment is occurring here. Since there are no significant sources of sediment near Victoria, contaminants from the outfall remain in high concentration in surface sediments, diluted only by benthic mixing to the surface of older, less contaminated material.

Benthic Invertebrate Accumulation

The mean concentrations of PBDEs in benthic invertebrates communities near the Vancouver outfall are higher than near the Victoria outfall (Fig. 3.2, Appendix D), despite the fact that sediment and wastewater concentrations are lower near Vancouver. Sediment TOC content was found to be a major force driving uptake and bioaccumulation near the Victoria and Vancouver outfalls, and explains the different concentrations in biota. Total PBDE concentrations in benthic invertebrates near the Victoria and Vancouver outfalls do not correlate with dry weight sediment concentrations. Instead, PBDE tissue concentrations correlate with TOC normalized sediment concentrations. Dry weight sediment concentrations of ΣPBDEs are similar near Vancouver and Victoria, but twice as much total organic carbon (TOC) is present in Victoria sediment. This indicates that benthic invertebrates need only ingest half as much sediment at Victoria versus Vancouver to receive the same amount of nutrients, and therefore may receive only half the contaminant load. To test this theory, it would be necessary to analyze the types of organic carbon present in sediment at Victoria and Vancouver, because bioavailability and nutrient value can vary with carbon source.

BSAFs for both PCBs and PBDEs exhibit a parabolic relationship with $\log K_{ow}$, reaching a maximum BSAF at $\log K_{ow}$ ~7. However, average BSAFs for PBDEs are twice as high as for PCBs. Studies in other environmental compartments, including sediment, have shown PBDEs to be increasing while PCBs are decreasing e.g. [30, 35, 54]. Benthic invertebrates are the first trophic level exposed to new contaminants entering sediment, and the communities I studied are very near PBDE entry points. It is therefore reasonable to conclude that PBDE concentrations in biota are increasing more quickly relative to PCBs, because PBDE fluxes to the environment at these outfall sources are much higher than PCBs (Table 3.1, Appendix C, E and F).

PBDE and PCB pattern uptake

Analysis of 47 PBDE congeners and 209 PCB congeners showed that PBDEs are dominated by a few congeners. Together BDE-209, -99, -47 and -100 contribute >80% to ΣPBDEs, while most PCB congeners contribute <5% to the total (Appendix I). Principal components analysis (PCA) identifies clear separation of both PBDE and PCB congener patterns between matrices. Despite their similar chemical structures, the partitioning of PCBs and that of PBDEs are not controlled by the same factors. Partitioning of PCB congeners depends mainly on the log K_{ow} value of a particular congener, or in other words, the chemical properties of the PCBs themselves. The physical characteristics of the environment and the benthic invertebrate community structure play secondary roles in the sorting of PCB congeners. In comparison, the partitioning of PBDE congeners depends mainly on the physical environment, which controls grain size distribution and TOC accumulation through currents and sediment accumulation rates. Benthic community structure and the chemical properties of individual congeners play secondary roles in PBDE sorting. The differences in PBDE and PCB sorting are consistent in the very different Vancouver and Victoria study areas, suggesting that these key sorting mechanisms may be transferrable to other marine outfall settings. This would provide a basis for a predictive model for PBDE and PCB distributions into analogous environments.

Contrasting controls over PCB and PBDE congener sorting may reflect the different stages of contaminant distribution in the environment. PCBs have been sorted by the physical environment for decades, and may be entering the last stages of environmental equilibrium. In contrast, PBDEs are relatively new chemicals in the early stages of environmental distribution, which are slowly being transported away from entry points into the wider environment. Their transport currently depends on physical factors in their receiving environment. However, current controls over PBDE sorting may not last forever. As PBDEs become legacy contaminants themselves, partitioning of PBDE congeners may come to be controlled by chemical properties, in the same manner as PCB congeners.

Wider Implications

Our results have implications for the interpretation of sediment quality guidelines, the prediction of the effects of increased levels of sewage treatment, the siting of new municipal outfalls, and the regulation and monitoring of municipal sewage outfalls.

Sediment quality guidelines

Sediment quality guidelines do not exist for all chemicals, because of the impossibility of developing and validating such guidelines for the vast number of chemicals in the environment [24]. Instead, SQGs developed for one chemical may be used to predict risks for others with a similar chemical structure, as has often been the case for PCBs and PBDEs. However, this thesis demonstrates that PBDEs and PCBs do not transport, sort or accumulate equally, despite their very similar structures. Our findings of the importance of TOC content in sediment to the bio-availability of PBDEs is in line with recent PBDE SQGs developed by the Canadian government, which are normalized for 1% TOC [24]. Both recognize the importance of considering sediment properties when predicting the risk to biota of this class of chemicals. The Canadian SQGs do not consider site specific variation in TOC content, and are intended to be used as a guideline for PBDE monitoring, rather than a direct link for assessing risk to higher trophic levels based on sediment concentrations of PBDEs [49]. The source of organic carbon making up TOC content may also affect bioavailability, and further study is required to assess this phenomenon.

The benthic invertebrate species present in a community have an additional effect on PCB and PBDE sorting in this study. Since many of the species were the same in both regions, it is possible that differences in PCB and PBDE accumulation by benthic invertebrates would be more pronounced among sites with more disparate benthic assemblages. Further study would be necessary to determine these differences.

Regulation of outfall discharges

The concentration of contaminants in wastewater is not sufficient to predict the environmental risk of chemical mixtures released through outfalls. The receiving environment plays a primary role in the accumulation of contaminants released from the outfall. An environment with slower currents and higher rates of sedimentation can be expected to capture more hydrophobic contaminants than an environment with fast currents that quickly disperse particles far from the outfall. If the spatial extent of the damaged near-field environment surrounding an outfall pipe is the primary concern, then a high-energy environment can be expected to be the least impacted, because effluent will be quickly diluted in the water column and the area affected will be very small. For example, in the high energy environment surrounding the Victoria outfall, sediment concentrations and benthic invertebrate community diversity shows that effects of the outfall are measurable within a 1 km radius of the outfall pipe [18]. In the low energy environment surrounding the Vancouver outfall, the effects of discharge can be measured from 7 km north to 4 km south of the outfall pipe [10]. However, the high sedimentation environment near the Vancouver outfall dilutes contaminants in the sediment, so that sediment contaminant concentrations are lower. Organic carbon content in the sediment of a local receiving environment also needs to be considered. Because benthic organisms feed on an organic carbon basis, rather than on the basis of total sediment mass, PBDEs and possibly other persistent contaminants will be less bio-available in an environment with higher TOC, whether of natural or anthropogenic origin

Overall, results show that uniform application of wastewater outfall regulations in all coastal environments may not be appropriate. When deciding on the most appropriate wastewater treatment method, the type of marine environment should be considered and compared to the probable effects of biosolid disposal on land. The extreme hydrophobicity of PBDEs leads them to partition into biosolids in all types of wastewater treatment plants [5, 55].

In addition, since increasing wastewater treatment is associated with an increased PBDE/TOC ratio, enhanced wastewater treatment will clearly not reduce the exposure of benthic organisms to PBDEs, leaving source control as the most effective option. These findings are important for outfall monitoring standards, which currently consider only the concentration and flux of contaminants, rather than potential harm to ecosystems at individual sites.

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Appendix

Appendix A
Locations of sampling sites

Station	Distance from outfall pipe	Site	Latitude °N	Longitude °E
CRD pipe	0m	Victoria	48.4026	236.5896
CRDM1N	100m N	Victoria	48.4046	236.5901
CRDM1NE	100m NE	Victoria	48.4039	236.5913
CRDM1E	100m E	Victoria	48.4027	236.5911
CRDM1SE	100m SE	Victoria	48.4020	236.5908
CRDM1S	100m S	Victoria	48.4016	236.5894
CRDM1SW	100m SW	Victoria	48.4021	236.5886
CRDM1W	100m W	Victoria	48.4027	236.5884
CRDM1NW	100m NW	Victoria	48.4039	236.5887
CRDM2N	200m N	Victoria	48.4055	236.5904
CRDM2NE	200m NE	Victoria	48.4035	236.5925
CRDM2E	200m E	Victoria	48.4026	236.5926
CRDM2SE	200m SE	Victoria	48.4017	236.5917
CRDM2S	200m S	Victoria	48.4007	236.5891
CRDM2SW	200m SW	Victoria	48.4018	236.5874
CRDM2W	200m W	Victoria	48.4027	236.5867
CRDM2NW	200m NW	Victoria	48.4035	236.5874
CRDM4E	400m E	Victoria	48.4027	236.5949
CRDM4SE	400m SE	Victoria	48.4007	236.5940
CRDM4SW	400m SW	Victoria	48.4007	236.5850
CRDM4W	400m W	Victoria	48.4027	236.5842
CRDM8E	800m E	Victoria	48.4027	236.6002
CRDM8W	800m W	Victoria	48.4027	236.5785
CRDPB1	Ref Site	Victoria	48.3543	236.4892
CRDPB2	Ref Site	Victoria	48.3603	236.4930
CRDPB3	Ref Site	Victoria	48.3605	236.4937
Iona 1	6.8K N	Vancouver	49.2654	236.6991
Iona 2	6.1K N	Vancouver	49.2590	236.7021
Iona 3	5.1K N	Vancouver	49.2499	236.7037
Iona 4	4.2K N	Vancouver	49.2416	236.7039
Iona 5	3.1K N	Vancouver	49.2318	236.7050

Iona 6	2.1K N	Vancouver	49.2318	236.7050
Iona 7	1.1K N	Vancouver	49.2135	236.7012
Iona 8	0.5K N	Vancouver	49.2085	236.7000
Iona 9	0K	Vancouver	49.2038	236.6994
Iona 10	0.5K S	Vancouver	49.1993	236.6991
Iona 11	1.0K S	Vancouver	49.1946	236.6988
Iona 12	1.9K S	Vancouver	49.1865	236.6987
Iona 13	2.9K S	Vancouver	49.1778	236.6994
Iona 14	4.0K S	Vancouver	49.1678	236.6995
Iona 15	8.2K S	Vancouver	49.1304	236.6886
Iona 16	9.2K S	Vancouver	49.1214	236.6826

Appendix B
PBDE concentrations in sediment (pg g^{-1} dw^{-1})

Sample	7	15	37	47	49	51	66	71	75	85	99
Laboratory Blank	<0.1	<0.1	<0.1	3.51	<0.1	<0.1	<0.1	<0.1	<0.1	0.134	3.05
Laboratory Blank 2	<0.1	<0.1	0.396	2.63	<0.1	<0.1	<0.1	<0.1	<0.1	0.177	2.41
CRD pipe	9.17	2.58	0.77	1570	115	13.3	36.4	22.2	2.88	55.9	1360
CRDM1N	1.82	1.93	0.55	129	13.7	1.97	4.14	1.47	<0.32	3.86	96.4
CRDM1E	9.77	3.70	0.54	695	92.1	9.65	20.10	10.20	1.34	23.50	624
CRDM1SE	18.9	6.69	0.80	861	119	11.60	20.20	13.80	1.33	25.30	611
CRDM1S	6.81	2.21	0.50	305	28.0	2.90	7.61	2.94	0.59	10.70	318
CRDM1W	11.0	4.61	1.16	1430	108	9.77	40.50	16.40	3.20	67.80	1450
CRDM2NE	6.04	2.19	0.29	251	31.6	3.54	6.63	4.08	0.41	6.57	165
CRDM2NW	2.75	1.88	0.30	152	14.7	1.90	4.90	2.06	0.38	4.28	103
CRD200NW	8.59	3.84	0.39	158	23.2	2.95	5.84	2.54	0.33	4.02	116
CRD400NW	10.8	5.09	0.39	103	21.2	3.15	4.24	2.61	0.31	2.36	70.8
CRDM2SE	7.22	4.35	1.29	4420	157	20.5	95.0	21.2	7.36	104	2770
CRDM2SW	5.32	2.25	0.32	202	20.2	2.28	6.36	2.32	0.33	5.55	142
CRDM4E	4.48	2.35	0.37	184	21.3	2.58	5.98	2.31	0.35	5.08	136
CRDM4SE	3.08	2.31	0.33	247	27.7	3.12	6.05	3.36	0.39	7.09	179
CRDM4SW	2.53	1.85	0.32	128	12.7	1.81	4.17	2.00	0.27	3.67	95.4
CRDM4W	1.74	1.75	0.40	130	12.8	1.71	4.60	1.75	<0.25	3.01	89.9
CRDM8E	2.85	2.02	0.30	171	11.5	1.46	4.91	1.83	0.22	3.94	115
CRDM8W	1.37	1.46	0.28	137	8.67	1.18	4.19	1.37	<0.26	3.99	99.5
CRDM8W (Dup)	1.99	1.20	0.23	89	6.61	0.89	2.77	1.26	<0.21	2.42	61.2
CRD Ref 2	0.247	0.387	0.179	27.7	2.25	0.309	1.14	0.417	<0.21	0.610	18.3
CRD Ref 3	0.55	0.561	0.154	31.8	2.59	0.286	1.16	0.478	<0.18	0.708	22.3

Sample	7	15	37	47	49	51	66	71	75	85	99
Iona6.8kmN	5.59	1.72	0.45	224	51.8	6.34	8.73	3.86	0.50	9.05	205
Iona6.1kmN	8.57	2.33	0.65	312	75.8	8.99	12.40	5.94	0.67	13.2	282
Iona5.1kmN	19.5	3.54	1.27	391	138	16.60	16.70	10.5	1.18	17.9	427
Iona4.2kmN	21.1	3.86	1.07	408	147	14.60	17.70	12.3	0.96	13.4	322
Iona3.1kmN	59.0	6.01	1.38	457	216	23.10	37.40	16.8	1.22	17.0	353
Iona2.1kmN	65.5	5.76	0.61	535	199	20.80	17.60	19.1	<0.18	19.7	424
Iona1.1kmN	70.6	4.74	0.71	529	203	20.70	23.20	19.0	1.55	18.8	436
Iona0.5kmN	50.3	4.36	<0.16	501	159	15.20	22.40	14.2	1.09	23.7	502
IonaZero	21.8	2.64	<0.10	322	99.6	9.66	13.30	10.4	0.86	11.5	294
Iona 200S	18.6	6.62	<0.32	438	146	<0.05	14.24	6.39	<0.16	<0.55	286
Iona 200W	19.6	8.92	<0.25	422	162	<0.14	14.32	5.88	0.47	<1.2	251
Iona0.5kmS	9.69	2.02	0.80	273	73.1	9.26	13.00	6.17	0.76	30.80	399
Iona1.0kmS	7.65	2.49	0.63	273	79.7	9.51	11.90	5.80	<0.08	9.83	238
Iona1.9kmS	6.36	2.06	0.43	163	47.4	7.53	7.15	2.99	<0.06	5.21	126
Iona2.9kmS	4.21	1.69	<0.19	130	36.2	6.04	5.07	2.32	<0.12	3.84	88.0
Iona4.0kmS	2.60	1.46	<0.25	135	29.1	4.44	5.62	2.32	0.27	4.63	107
Iona9.2kmS (Ref)	0.865	0.782	0.253	106	16	1.85	4.73	1.8	0.251	4.6	102
Iona8.2kmS (Ref)	1.15	0.774	<0.23	99.9	16.7	2.25	4.62	2.25	<0.15	3.83	88

Sample	**100**	**140**	**153**	**154**	**155**	**183**	**190**	**203**	**206**	**207**	**208**	**209**	**Total**
Laboratory Blank	0.716	<0.1	0.591	0.336	<0.1	0.188	<0.1	0.434	3.14	4.01	3.07	84.4	104
Laboratory Blank 2	0.574	<0.1	0.444	0.279	<0.1	0.368	0.206	0.422	1.97	3.76	2.56	68.6	84.8
CRD pipe	321	3.78	137	110	6.16	28.8	<3.04	48.2	146	231	151	2290	6830
CRDM1N	28.0	0.60	12.0	10.2	1.07	7.98	0.607	2.67	10.5	18.5	10.9	595	987
CRDM1E	149	2.28	84.4	60.9	4.11	60.6	<8.88	91.4	642	806	552	10100	14200
CRDM1SE	153	1.93	62.7	52.4	4.14	28.9	3.98	41.4	170	184	136	4270	6970
CRDM1S	81.8	1.02	30.0	29.6	2.42	5.92	<0.52	2.47	13.1	19.0	11.9	956	1880
CRDM1W	355	4.63	148	128	8.65	17.8	<1.22	6.13	28.9	60.5	34.8	1800	5850
CRDM2NE	45.6	0.64	17.3	16.4	1.16	6.19	<1.81	8.08	31.2	59.3	39.7	903	1670
CRDM2NW	29.3	<0.10	12.0	10.8	1.06	3.98	<2.18	6.24	15.3	42.4	21.3	610	1070
CRD200NW	29.8	0.50	13.0	11.5	1.64	4.91	0.926	5.41	44.5	74.4	45.6	822	1430
CRD400NW	19.9	0.40	8.29	8.67	1.55	3.51	0.615	4.57	35.3	68.4	47.3	973	1450
CRDM2SE	768	6.80	239	212	16.9	36.1	<8.37	73.1	334	609	463	8410	19000
CRDM2SW	39.6	0.77	16.4	15.1	1.52	4.59	<7.93	24.9	161	317	164	6980	8150
CRDM4E	38.2	0.66	17.2	14.1	1.53	6.86	<2.36	10.9	54.8	116	59.1	2370	3100
CRDM4SE	48.8	0.58	19.2	16.1	1.41	11.0	<2.01	8.89	47.1	65.7	45.4	1070	1870
CRDM4SW	28.0	0.56	11.7	11.1	1.18	3.73	<0.96	4.80	19.7	32.2	18.9	562	974
CRDM4W	24.8	<0.89	10.2	9.42	0.94	4.31	<1.23	4.32	19.7	32.5	17.8	602	999
CRDM8E	32.4	0.51	14.7	14.00	1.94	6.15	<2.54	3.45	11.6	22.4	14.0	621	1090
CRDM8W	27.1	0.47	11.3	11.60	1.23	9.09	0.523	6.02	17.8	29.0	17.0	535	949
CRDM8W (Dup)	17.6	0.28	8.61	7.97	0.98	3.10	<7.91	4.35	13.2	28.0	17.6	684	972
CRD Ref 2	5.73	<0.21	2.68	2.6	0.356	0.859	0.184	1.31	7.20	10.1	8.04	204	300
CRD Ref 3	6.88	<0.20	4.8	4.3	0.553	1.44	<3.09	1.55	5.88	10.1	6.39	216	326

Sample	100	140	153	154	155	183	190	203	206	207	208	209	Total
Iona6.8kmN	52.2	<1.1	21.6	21.0	3.23	<0.45	<3.21	26.4	140	267	149	2940	4140
Iona6.1kmN	76.7	<0.25	31.0	32.3	<0.16	10.1	3.42	31.8	132	293	181	2270	3780
Iona5.1kmN	116	<0.13	48.3	43.0	7.34	20.5	4.26	71.7	206	383	266	4070	6280
Iona4.2kmN	88.8	6.04	34.2	36.8	8.63	19.6	<4.22	76.6	231	585	348	3810	6210
Iona3.1kmN	106	8.17	40.3	44.2	<0.51	27.4	<4.35	110	413	914	499	7390	10400
Iona2.1kmN	115	<0.32	50.5	53.3	9.04	17.8	<5.36	44.6	461	779	530	7590	7970
Iona1.1kmN	122	9.68	52.9	52.9	<0.62	29.2	<6.29	84.9	286	469	307	7640	4620
Iona0.5kmN	133	<0.55	56.7	60.8	10.20	28.7	<3.59	77.1	227	386	259	5430	4220
IonaZero	77.1	6.26	31.5	35.0	<0.25	14.3	<5.36	43.1	145	227	154	3100	3440
Iona 200S	99	<0.56	34.6	46.1	9.43	32.4	<3.65	<6.5	29.8	173	141	3560	10960
Iona 200W	95	<0.62	31.4	44.1	10.01	23.1	<3.25	<4.5	<6.2	175	145	3369	10700
Iona0.5kmS	101	<0.91	54.1	45.1	<0.36	13.4	<4.56	<3.5	110	203	151	2720	3070
Iona1.0kmS	66.2	<0.85	26.6	26.4	<0.23	23.7	<2.32	30.4	102	166	107	2250	1730
Iona1.9kmS	36.9	3.29	13.8	14.5	<0.55	<1.2	<3.65	<5.1	119	206	146	2160	2290
Iona2.9kmS	31.6	<0.15	12.0	13.5	2.73	<1.5	<2.35	<4.2	59.2	105	81	1150	1000
Iona4.0kmS	33.0	<0.55	12.6	14.5	2.35	6.09	<2.35	25.8	95.1	188	111	1510	1450
Iona9.2kmS (Ref)	26.1	<0.54	11.9	14.3	<0.42	5.03	<0.84	15.1	58.6	112	61.6	910	1450
Iona8.2kmS (Ref)	25	0.647	10.5	10.2	1.17	4.33	<1.87	13	34.7	88.2	54	539	1000

Appendix C
PBDE concentrations in wastewater (pg L^{-1})

Date Sampled	Outfall	Units	BDE-7	BDE-8/11	BDE-15	BDE-17/25	BDE-28/33	BDE-47	BDE-49	BDE-66	BDE-71	BDE-75
07-Oct-97	Vancouver	pg/L	1.9	3.1	12	120	290	22000	560	470	36	36
23-Oct-97	Vancouver	pg/L	1.9	3.8	12	110	250	19000	480	410	32	30
27-Nov-97	Vancouver	pg/L	2.5	5.9	12	120	290	21000	550	440	38	34
29-Oct-02	Vancouver	pg/L	4.8	14	44	230	510	27000	630	700	88	50
10-Jun-03	Vancouver	pg/L	3.4	7.9	43	162	450	25000	540	710	100	41
10-Sep-03	Vancouver	pg/L	4.7	8.3	33	181	400	21000	500	490	60	37
25-Feb-04	Vancouver	pg/L	1.1	6.9	34	151	390	22000	480	630	66	40
18-Oct-04	Vancouver	pg/L	19	13	16	350	260	17000	430	290	75	26
15-Mar-05	Vancouver	pg/L	3.6	8	26	170	400	29000	600	480	1.03	35
2004	Victoria	pg/L	7.25	12.5	59.3	568	1580	78500	1820	1960	186	141
2004	Victoria	pg/L	12.4	21.3	106	899	2200	76500	2040	1730	236	186
2004	Victoria	pg/L	15.8	28.1	127	863	2100	89900	2370	2040	347	216
2004	Victoria	pg/L	15.9	26	122	896	2260	91600	2410	2100	320	174
2005	Victoria	pg/L	6.42	21.6	97.7	1170	2340	127000	2770	2360	577	174
2005	Victoria	pg/L	7.2	18.9	37	377	479	33600	729	633	89.5	46.1
2005	Victoria	pg/L	26.7	63.5	76.8	502	1290	90000	1740	1450	214	128
2005	Victoria	pg/L	8.02	17.7	103	1080	1850	109000	2400	2540	547	144
2006	Victoria	pg/L	7.96	13.2	49.4	640	872	34800	1110	887	339	71.9
2006	Victoria	pg/L	4.54	9.28	45.2	514	682	28600	856	695	247	63.2
2006	Victoria	pg/L	4.18	13.6	64.3	662	1380	66300	1410	1140	304	107
2006	Victoria	pg/L	3.83	14.4	60.7	449	1310	75300	1830	1550	178	116
2006	Victoria	pg/L	7.5	24.6	47.7	495	1110	61600	1310	1200	271	68.7
2007	Victoria	pg/L	2.84	9.14	65.6	318	677	47200	1100	823	65	63.3
2010	Victoria	pg/L	23.4	18.4	66.6	904	1530	48000	1400	1210	379	92.4
2010	Victoria	pg/L	7.85	14.1	55.7	761	1480	68700	2020	2160	493	142
2010	Victoria	pg/L	12.6	18.5	82.5	582	1100	57000	1670	1120	200	81.6

Date Sampled	Outfall	BDE-85	BDE-99	BDE-100	BDE-138/166	BDE-154	BDE-155	BDE-206	BDE-208	BDE-209	Total
07-Oct-97	Vancouver	1200	27000	5300	340	2200	140	2100	970	24000	122489
23-Oct-97	Vancouver	1000	23000	4600	310	2000	130	2200	1200	21000	111496
27-Nov-97	Vancouver	1300	26000	5100	350	2100	140	4400	2500	51000	151143
29-Oct-02	Vancouver	1200	31000	5400	3060	2200	210	3000	9300	330000	452199
10-Jun-03	Vancouver	1200	29000	5200	2800	2000	200	230	330	70000	175799
10-Sep-03	Vancouver	950	26000	4600	2478	1800	160	820	1600	59000	157996
25-Feb-04	Vancouver	1000	27000	4700	2569	1800	200	500	850	36000	136460
18-Oct-04	Vancouver	770	16000	3400	230	1300	66	960	650	16000	96133
15-Mar-05	Vancouver	1300	27000	5700	330	1.95	140	1500	770	26000	131892
2004	Victoria	2830	71800	16600	499	5910	391	2620	4920	29200	221608
2004	Victoria	2630	65900	14600	997	8560	635	7720	14600	323000	524577
2004	Victoria	3600	81600	18000	1150	8280	597	9200	9370	305000	536808
2004	Victoria	3190	82700	18600	947	8330	616	6780	13800	350000	586891
2005	Victoria	4850	108000	26200	1290	10200	652	6470	6360	143000	445544
2005	Victoria	1490	32200	6630	350	2500	174	1380	586	31600	114932
2005	Victoria	3950	83100	17700	863	5990	391	4450	1190	118000	333130
2005	Victoria	4570	104000	23000	1170	7800	488	9840	8130	137000	415693
2006	Victoria	1540	33300	7680	354	3130	207	2380	2020	44800	136207
2006	Victoria	1280	28000	6370	292	2570	169	1540	1550	41200	116693
2006	Victoria	2880	60700	13800	736	5220	298	7110	5050	176000	345185
2006	Victoria	3130	66700	15200	634	5540	437	5930	2420	91600	274409
2006	Victoria	2320	52800	12100	411	4300	291	3630	3560	90800	238353
2007	Victoria	1720	43300	8480	469	3240	299	1870	785	61500	173994
2010	Victoria	1830	41900	9090	555	2980	159	5980	11100	87700	216928
2010	Victoria	3040	70700	14400	975	5530	533	20600	27300	246000	466922
2010	Victoria	2450	48900	10400	515	3120	252	18900	25600	280000	454014

Appendix D
PBDE concentrations in benthic invertebrate tissue (pg g^{-1} lw^{-1})

Site	Outfall	Units	BDE-7	BDE-8/11	BDE-15	BDE-17/25	BDE-28/33	BDE-47	BDE-49	BDE-66	BDE-71
Iona 400m N DUP Jar1/2	Vancouver	pg/g lw	949.7729	790.5768	278.0683	8360.037	3233.529	228602.1	28636.4	4469.254	362.9715
Iona 400m N DUP Jar1/2	Vancouver	pg/g lw	1406.522	887.031	266.4698	8512.675	3100.338	218330.2	26845.57	4395.354	497.3
Iona 400m N DUP Jar2/2	Vancouver	pg/g lw	2217.17	2330.999	633.2861	20384.92	6246.225	440520.3	53133.87	9091.443	666.6041
IONA Zero	Vancouver	pg/g lw	5707.275	3031.8	424.8253	27817.42	4342.636	291683.8	73248.48	5941.629	524.3922
IONA 200m N	Vancouver	pg/g lw	2273.336	2227.881	595.4183	17967.4	5091.972	385475.8	60067.39	7066.425	436.3276
IONA 400m N	Vancouver	pg/g lw	2003.815	2962.906	678.3916	20131.87	5261.49	401195.6	64436.74	8272.843	506.0987
IONA 400m N DUP	Vancouver	pg/g lw	1466.947	2366.947	529.7446	16957.94	4388.816	327167.9	53007.01	6288.597	453.6243
IONA 200m S	Vancouver	pg/g lw	1332.075	2170.2	501.6287	16894.18	5068.947	369994.5	51727	8179.431	432.9922
IONA 400m S	Vancouver	pg/g lw	2792.223	3937.7	1009.126	28208.76	6975.983	495628.6	72326.6	12062.32	654.7612
MAC 200m SE	Victoria	pg/g lw	264.2031	281.8	299.2457	3292.83	4029.232	230015.2	22277.7	5688.085	345.9881
MAC Zero	Victoria	pg/g lw	302.7754	309.5	347.6377	2785.305	3346.765	220996.1	22711.85	6161.552	299.5735
MAC Zero DUP	Victoria	pg/g lw	195.6902	190.1	316.2111	2344.118	3111.541	212877.1	19922.89	5861.122	271.949
MAC 200m NW	Victoria	pg/g lw	164.3296	275.1	185.5982	2173.843	1856.207	181473.3	17720.81	4374.5	364.6933
MAC 400m SE	Victoria	pg/g lw	80.14167	136.5	272.177	1317.259	2508.184	197948.1	11125.36	4940.185	174.8
MAC 400m NW	Victoria	pg/g lw	203.0011	218.6	169.0748	1898.762	1909.457	171555.8	20071.79	4038.434	292.8746

Site	Outfall	Units	BDE-75	BDE-85	BDE-99	BDE-100	BDE-138/1	BDE-154	BDE-155	BDE-206	BDE-208
Iona 400m N DUP Jar1/2	Vancouver	pg/g lw	843.321	7621.013	180811.8	70250.58	886.2564	27701.86	2603.554	2133.88	1601.454
Iona 400m N DUP Jar1/2	Vancouver	pg/g lw	382.4389	6030.766	171316.7	68192.77	971.0633	24510.99	449.3976	2620.685	1879.424
Iona 400m N DUP Jar2/2	Vancouver	pg/g lw	507.8742	15709.19	261993.9	115528.4	572.5537	40967.46	7223.257	6540.579	9549.606
IONA Zero	Vancouver	pg/g lw	1283.322	7693.43	216033.7	62411.35	1189.672	19627.15	1887.497	1953.509	1064.559
IONA 200m N	Vancouver	pg/g lw	1000.045	9756.131	298715.8	106067.7	1966.819	40206.79	5556.969	6524.254	1982.272
IONA 400m N	Vancouver	pg/g lw	735.3751	10500	252619.6	125336.5	1737.018	60248.31	8359.313	11811.05	4396.539
IONA 400m N DUP	Vancouver	pg/g lw	858.4898	9688.411	217197.6	106729.4	1301.908	51290.29	7571.509	3121.684	2034.849
IONA 200m S	Vancouver	pg/g lw	171.7482	14624.82	264997.6	109895.3	2514.908	46609.21	6822.837	9424.062	4329.485
IONA 400m S	Vancouver	pg/g lw	2045.67	15694.78	362851.9	144123.9	3624.855	51032.4	10295.95	16953.96	10494.83
MAC 200m SE	Victoria	pg/g lw	698.0004	4587.284	136022.3	51284.44	696.2842	15113.61	1393.525	590.2342	442.9314
MAC Zero	Victoria	pg/g lw	502.1952	1712.052	58940.86	31325.16	268.3209	7295.057	573.8218	479.2196	232.5111
MAC Zero DUP	Victoria	pg/g lw	292.3979	1775.194	61753.03	32629.02	246.3209	6794.882	559.6505	424.9692	210.6156
MAC 200m NW	Victoria	pg/g lw	541.9347	3770.869	148943.7	57477.43	553.3712	15916.03	1479.849	2238.554	598.8453
MAC 400m SE	Victoria	pg/g lw	287.7338	3587.522	125237.4	43147.62	525.0916	13365.71	1741.138	1515.211	485.7412
MAC 400m NW	Victoria	pg/g lw	535.7528	3505.992	137577	52668.12	678.2582	14313.82	1394.653	1467.758	481.889

Site	Outfall	Units	BDE-209	Total
Iona 400m N DUP Jar1/2	Vancouver	pg/g lw	113021.6	683158
Iona 400m N DUP Jar1/2	Vancouver	pg/g lw	121971.8	662567.5
Iona 400m N DUP Jar2/2	Vancouver	pg/g lw	128998.4	1122816
IONA Zero	Vancouver	pg/g lw	58264.76	784131.3
IONA 200m N	Vancouver	pg/g lw	321356.3	1274335
IONA 400m N	Vancouver	pg/g lw	573973.3	1555167
IONA 400m N DUP	Vancouver	pg/g lw	195458.4	1007880
IONA 200m S	Vancouver	pg/g lw	285843.6	1201535
IONA 400m S	Vancouver	pg/g lw	189611.6	1430326
MAC 200m SE	Victoria	pg/g lw	30342.18	507665
MAC Zero	Victoria	pg/g lw	14279.19	372869.5
MAC Zero DUP	Victoria	pg/g lw	18544.7	368321.5
MAC 200m NW	Victoria	pg/g lw	67709.09	507818
MAC 400m SE	Victoria	pg/g lw	22378.35	430774.2
MAC 400m NW	Victoria	pg/g lw	63251.15	476232.1

Appendix E
PCB concentrations in wastewater (pg L^{-1})

Year	Outfall	CB-6	CB-11	CB-15	CB-16/32	CB-17	CB-18	CB-22	CB-25	CB-26	CB-27	CB-31	CB-37
2004	Vancouver	21	150	54	72	48	92	57	10	24	6	140	33
2005	Vancouver	22	280	120	80	53	110	64	11	26	6	160	46
2002	Vancouver	31	280	61	150	89	230	110	17	42	11	310	55
2003	Vancouver	32	230	82	163	100	230	120	18	47	14	330	91
2003	Vancouver	24	240	84	107	59	130	97	14	36	8	270	65
2004	Vancouver	20	220	65	70	48	100	55	10	25	7	140	35
2004	Victoria	36	426	64	120	67	141	85	14	38	11	208	47
2004	Victoria	43	339	72	124	77	153	102	17	43	11	262	51
2004	Victoria	85	521	94	164	118	242	122	31	82	19	340	66
2005	Victoria	60	363	89	146	109	170	97	31	58	36	225	54
2005	Victoria	31	149	62	106	78	120	65	25	46	25	172	43
2005	Victoria	71	474	97	196	124	217	133	37	69	35	319	73
2005	Victoria	42	521	77	138	78	159	117	21	48	11	283	61
2006	Victoria	36	300	60	104	64	129	82	17	36	14	182	44
2006	Victoria	57	231	70	148	127	180	84	43	60	41	209	52
2006	Victoria	57	188	71	164	138	188	89	43	59	46	215	50
2006	Victoria	51	147	62	135	116	157	73	39	55	41	182	40
2006	Victoria	28	467	60	90	49	98	76	13	30	7	171	51

Year	Outfall	CB-42/68	CB-43/49	CB-44	CB-45/51	CB-46	CB-52	CB-56	CB-60	CB-63	CB-64/71	CB-66
2004	Vancouver	36	6	290	210	9	360	63	35	6	78	130
2005	Vancouver	46	7	360	240	12	480	81	44	8	100	160
2002	Vancouver	68	10	340	150	17	470	120	66	9	130	220
2003	Vancouver	92	18	600	500	25	980	180	96	14	200	360
2003	Vancouver	90	11	570	440	18	950	140	76	12	190	300
2004	Vancouver	36	7	250	40	8	460	71	37	5	82	150
2004	Victoria	50	10	252	39	12	362	86	53	7	94	178
2004	Victoria	53	8	267	38	12	394	87	49	8	101	190
2004	Victoria	85	18	420	78	23	636	136	76	13	148	292
2005	Victoria	59	8	277	50	16	409	<1.42	<1.42	8	92	160
2005	Victoria	39	6	188	18	12	274	42	26	7	59	99
2005	Victoria	83	12	376	65	23	515	113	63	11	130	213
2005	Victoria	54	8	263	41	12	353	107	58	8	95	192
2006	Victoria	41	7	242	38	11	308	66	39	6	76	125
2006	Victoria	70	10	3060	291	21	356	74	39	8	85	191
2006	Victoria	70	10	2090	235	23	398	63	32	7	89	160
2006	Victoria	57	9	675	94	19	319	52	28	6	73	119
2006	Victoria	36	7	242	30	10	292	66	46	6	67	146

CB-77	Year	Outfall	CB-82	CB-83	CB-84	CB-92	CB-103	CB-105	CB-114	CB-118	CB-123	CB-130
14	2004	Vancouver	51	260	160	100	3	130	8	350	8	25
19	2005	Vancouver	98	350	230	140	4	350	20	800	11	58
18	2002	Vancouver	51	230	130	82	4	130	9	330	7	22
34	2003	Vancouver	100	490	290	180	5	270	18	730	13	43
27	2003	Vancouver	91	460	290	170	5	250	17	640	11	42
15	2004	Vancouver	50	280	150	100	4	170	11	410	7	31
17	2004	Victoria	50	238	116	83	5	121	7	304	26	26
14	2004	Victoria	48	229	115	81	3	132	9	341	16	25
19	2004	Victoria	54	372	174	137	8	143	9	376	26	37
16	2005	Victoria	49	291	144	108	<5.46	165	11	410	16	28
10	2005	Victoria	29	153	77	51	<3.36	89	5	235	11	18
20	2005	Victoria	61	297	162	109	6	156	12	416	23	35
21	2005	Victoria	46	220	110	74	4	113	8	300	20	22
12	2006	Victoria	36	185	87	60	3	100	8	262	15	20
15	2006	Victoria	31	183	79	55	3	76	6	225	9	14
18	2006	Victoria	35	205	91	65	4	88	7	264	9	23
11	2006	Victoria	26	154	67	50	3	65	6	196	8	15
15	2006	Victoria	40	199	92	64	3	113	8	312	<2.23	22

Year	Outfall	CB-131/142	CB-132	CB-133	CB-134/143	CB-136	CB-137	CB-141	CB-144	CB-146	CB-147/149
2004	Vancouver	7	150	6	25	63	22	72	20	52	300
2005	Vancouver	15	290	10	54	98	50	140	38	110	590
2002	Vancouver	6	140	5	22	71	19	77	25	58	360
2003	Vancouver	13	260	10	1	110	38	130	42	95	660
2003	Vancouver	13	250	9	43	110	40	130	40	90	570
2004	Vancouver	8	160	7	27	65	32	88	24	65	360
2004	Victoria	5	131	7	22	62	25	77	20	63	329
2004	Victoria	7	131	6	24	56	24	81	23	62	334
2004	Victoria	9	192	10	36	131	35	123	45	89	640
2005	Victoria	<8.38	173	9	30	71	28	89	20	75	370
2005	Victoria	5	90	5	14	38	18	54	16	39	222
2005	Victoria	8	186	9	31	84	30	104	30	80	429
2005	Victoria	5	124	5	19	56	23	67	21	59	296
2006	Victoria	5	105	5	18	44	19	57	17	51	250
2006	Victoria	3	88	4	13	33	15	43	13	39	204
2006	Victoria	6	113	6	19	44	19	56	15	45	257
2006	Victoria	3	78	4	14	33	15	40	12	34	190
2006	Victoria	6	119	5	22	47	20	60	17	53	278

Year	Outfall	CB-153	CB-156	CB-158/160	CB-167	CB-170/190	CB-171/173	CB-172/192	CB-174	CB-175
2004	Vancouver	330	51	42	16	58	21	11	73	3
2005	Vancouver	550	122	91	41	120	40	21	93	5
2002	Vancouver	360	39	38	11	58	21	11	74	4
2003	Vancouver	580	73	70	20	70	25	14	83	5
2003	Vancouver	540	78	70	23	71	28	14	85	4
2004	Vancouver	410	57	52	18	60	20	11	64	4
2004	Victoria	395	60	43	15	84	25	15	76	5
2004	Victoria	375	60	43	18	82	25	15	76	4
2004	Victoria	571	63	59	19	98	32	18	118	7
2005	Victoria	459	65	54	19	78	23	16	75	4
2005	Victoria	256	37	29	13	50	14	9	51	3
2005	Victoria	497	65	57	20	94	27	15	101	5
2005	Victoria	366	51	36	15	83	22	17	82	4
2006	Victoria	295	46	32	13	62	18	11	56	3
2006	Victoria	239	34	25	11	45	13	7	43	2
2006	Victoria	294	44	33	14	61	18	11	56	3
2006	Victoria	226	32	23	10	48	13	8	44	2
2006	Victoria	331	48	33	14	53	16	9	48	3

Year	Outfall	CB-176	CB-177	CB-178	CB-179	CB-180/193	CB-183/185	CB-187	CB-194	CB-195	CB-196/203
2004	Vancouver	9	38	17	36	150	52	100	44	15	27
2005	Vancouver	14	55	19	47	210	66	110	28	11	14
2002	Vancouver	16	53	21	50	160	59	120	21	8	12
2003	Vancouver	15	51	19	48	170	64	110	28	11	16
2003	Vancouver	15	45	20	47	170	63	110	27	11	16
2004	Vancouver	11	39	17	34	150	53	95	22	8	12
2004	Victoria	14	44	26	48	230	67	133	42	14	24
2004	Victoria	13	46	22	46	201	61	127	41	13	25
2004	Victoria	27	62	38	97	268	99	198	43	15	30
2005	Victoria	13	46	20	49	197	67	127	42	16	22
2005	Victoria	8	28	12	24	114	37	73	27	8	13
2005	Victoria	17	54	30	62	228	85	171	61	21	34
2005	Victoria	14	49	25	47	209	68	141	42	14	23
2006	Victoria	11	33	20	36	156	48	108	34	11	17
2006	Victoria	7	24	13	23	119	37	77	29	9	14
2006	Victoria	9	31	15	31	140	45	86	29	12	16
2006	Victoria	7	26	12	23	118	35	72	28	10	16
2006	Victoria	9	28	14	30	127	43	79	37	16	16

Year	Outfall	CB-197/200	CB-198/199	CB-201	CB-202	CB-205	CB-206	CB-208	CB-209	Total
2004	Vancouver	15	67	9	18	37	6	10	10	17044
2005	Vancouver	7	34	5	9	10	3	5	10	19759
2002	Vancouver	6	30	5	11	12	2	6	61	18064
2003	Vancouver	6	36	5	11	13	12	5	8	21641
2003	Vancouver	7	41	6	11	15	3	4	11	20782
2004	Vancouver	6	34	5	11	20	2	9	20	17272
2004	Victoria	10	59	9	23	27	4	10	23	17680
2004	Victoria	11	71	8	22	274	31	97	294	18384
2004	Victoria	13	76	10	26	43	<9.59	18	45	20588
2005	Victoria	11	57	9	22	47	14	17	33	18248
2005	Victoria	5	30	4	10	28	40	9	24	15853
2005	Victoria	12	91	11	24	60	6	19	66	19699
2005	Victoria	11	65	8	21	49	6	18	40	17915
2006	Victoria	7	51	7	16	58	5	20	57	16650
2006	Victoria	7	58	5	15	43	17	15	19	19641
2006	Victoria	8	44	5	12	26	14	8	20	18991
2006	Victoria	6	39	5	10	21	8	7	15	16471
2006	Victoria	6	40	5	17	33	4	13	32	16722

Appendix F
PCB concentrations in sediment (ng g^{-1})

Site	Outfall	CB-6	CB-11	CB-15	CB-16/32	CB-17	CB-18	CB-22	CB-25	CB-26	CB-27	CB-31	CB-37
Iona 500m N	Vancouver	150	1121	1016	595	423	689	760	185	338	61	1844	816
Iona 500m N DUP	Vancouver	197	1388	900	785	552	791	862	238	374	69	1938	799
Iona Zero	Vancouver	176	907	767	636	415	687	673	171	293	62	1547	628
Iona 200m S	Vancouver	465	2888	3190	133	1146	2333	1286	426	686	169	3529	2411
Mac 200m SE	Victoria	8	30	26	29	22	43	24	5	11	4	59	15
Mac 200m NW	Victoria	2	22	11	7	5	9	10	2	4	1	23	9
Mac Zero	Victoria	8	58	35	43	27	52	49	8	20	4	127	36
Mac 400m SE	Victoria	3	20	14	20	14	30	18	3	7	3	50	12
Mac 200m NW	Victoria	2	18	10	9	5	11	10	2	4	1	23	8
Mac Zero	Victoria	32	70	71	110	71	156	90	15	37	10	222	56
Mac 200m SE	Victoria	10	26	23	62	40	86	66	8	19	6	193	43
Mac 200m NW	Victoria	2	17	10	8	5	10	9	2	4	1	23	8
Mac 400m SE	Victoria	3	17	11	17	12	26	16	3	6	3	42	12

Site	Outfall	CB-42/68	CB-43/49	CB-44	CB-45/51	CB-46	CB-52	CB-56	CB-60	CB-63	CB-64/71	CB-66	CB-77
Iona 500m N	Vancouver	513	59	1803	233	74	1985	1169	656	98	824	2497	304
Iona 500m N DUP	Vancouver	638	95	2200	323	104	2396	1196	654	114	996	2531	283
Iona Zero	Vancouver	488	57	1600	240	81	1493	907	514	75	720	1853	210
Iona 200m S	Vancouver	1187	2959	3274	514	141	3884	2339	2327	226	2213	5299	757
Mac 200m SE	Victoria	14	2	64	8	4	106	26	13	2	24	58	6
Mac 200m NW	Victoria	6	1	38	2	1	71	21	11	2	15	46	4
Mac Zero	Victoria	35	4	145	18	7	215	72	39	6	63	139	13
Mac 400m SE	Victoria	14	2	68	7	3	114	31	15	3	26	65	5
Mac 200m NW	Victoria	7	1	31	3	1	50	16	9	1	13	34	4
Mac Zero	Victoria	55	9	266	36	12	471	109	62	9	108	213	17
Mac 200m SE	Victoria	72	8	287	38	14	364	138	119	12	134	252	31
Mac 200m NW	Victoria	5	1	21	2	1	33	11	7	1	8	25	3
Mac 400m SE	Victoria	19	2	137	9	4	327	34	16	3	44	83	6

Site	Outfall	CB-82	CB-83	CB-84	CB-92	CB-103	CB-105	CB-114	CB-118	CB-123	CB-130	CB-131/142
Iona 500m N	Vancouver	452	2333	761	627	28	1755	91	3957	100	331	59
Iona 500m N DUP	Vancouver	507	2515	838	725	47	1638	93	3735	86	346	64
Iona Zero	Vancouver	399	1660	618	414	20	1087	62	2380	43	205	41
Iona 200m S	Vancouver	833	253	1015	799	41	2874	155	6635	1411	450	58
Mac 200m SE	Victoria	16	74	32	23	1	79	3	164	3	17	3
Mac 200m NW	Victoria	23	93	55	32	1	96	4	195	3	16	4
Mac Zero	Victoria	35	182	76	54	2	118	7	279	5	19	4
Mac 400m SE	Victoria	13	71	31	21	1	45	2	107	2	7	2
Mac 200m NW	Victoria	7	36	14	9	0	24	1	56	1	4	1
Mac Zero	Victoria	91	379	173	148	3	373	19	880	13	96	19
Mac 200m SE	Victoria	51	230	103	66	2	148	9	329	7	23	6
Mac 200m NW	Victoria	4	25	8	7	0	18	1	43	1	2	0
Mac 400m SE	Victoria	34	174	94	55	2	114	6	280	4	16	4

Site	Outfall	CB-132	CB-133	CB-134/143	CB-136	CB-137	CB-141	CB-144	CB-146	CB-147/149	CB-153
Iona 500m N	Vancouver	1547	217	1388	478	232	733	199	732	3479	4423
Iona 500m N DUP	Vancouver	1688	85	1619	569	215	665	217	657	3685	4362
Iona Zero	Vancouver	993	47	940	303	143	375	116	355	2020	2560
Iona 200m S	Vancouver	1557	106	245	438	177	536	188	1032	103	5282
Mac 200m SE	Victoria	80	2	11	17	14	39	8	29	144	183
Mac 200m NW	Victoria	86	2	13	22	13	34	8	27	141	146
Mac Zero	Victoria	103	4	15	39	17	57	16	45	227	265
Mac 400m SE	Victoria	37	2	6	13	6	17	5	17	94	95
Mac 200m NW	Victoria	19	1	3	5	3	8	2	9	40	51
Mac Zero	Victoria	491	18	71	127	61	327	66	181	997	1220
Mac 200m SE	Victoria	133	5	20	42	19	67	7	50	292	326
Mac 200m NW	Victoria	10	1	1	4	2	5	2	6	27	36
Mac 400m SE	Victoria	84	2	13	25	15	35	8	28	152	164

Site	Outfall	CB-156	CB-158/160	CB-167	CB-170/190	CB-171/173	CB-172/192	CB-174	CB-175	CB-176
Iona 500m N	Vancouver	639	460	217	1195	387	206	1184	58	168
Iona 500m N DUP	Vancouver	576	440	206	1123	416	206	1165	64	174
Iona Zero	Vancouver	331	270	120	847	304	201	813	43	113
Iona 200m S	Vancouver	589	534	240	1276	361	192	1005	66	143
Mac 200m SE	Victoria	65	30	17	62	12	7	28	1	5
Mac 200m NW	Victoria	39	25	10	28	7	4	19	1	3
Mac Zero	Victoria	41	34	13	58	19	11	67	3	9
Mac 400m SE	Victoria	13	11	4	16	6	3	16	1	2
Mac 200m NW	Victoria	8	6	3	11	3	2	10	0	1
Mac Zero	Victoria	251	185	80	500	153	84	394	20	47
Mac 200m SE	Victoria	49	39	14	59	22	12	79	4	14
Mac 200m NW	Victoria	5	3	2	6	2	1	5	0	1
Mac 400m SE	Victoria	33	26	10	24	8	4	20	1	3

Site	Outfall	CB-177	CB-178	CB-179	CB-180/193	CB-183/185	CB-187	CB-194	CB-195	CB-196/203	CB-197/200
Iona 500m N	Vancouver	807	307	555	2523	908	1836	629	266	837	123
Iona 500m N DUP	Vancouver	817	340	621	2385	950	2031	498	227	295	95
Iona Zero	Vancouver	621	221	409	1620	673	1367	300	158	209	52
Iona 200m S	Vancouver	1064	348	548	1831	814	1944	512	315	808	79
Mac 200m SE	Victoria	19	7	13	86	21	34	21	6	10	3
Mac 200m NW	Victoria	12	4	9	45	13	23	12	5	6	3
Mac Zero	Victoria	39	17	35	146	54	109	33	11	23	9
Mac 400m SE	Victoria	11	4	9	36	14	28	12	4	7	3
Mac 200m NW	Victoria	7	3	4	23	8	16	6	2	3	1
Mac Zero	Victoria	223	74	127	916	299	427	186	74	114	32
Mac 200m SE	Victoria	41	22	60	153	69	169	34	13	27	14
Mac 200m NW	Victoria	4	2	3	13	4	10	4	1	2	1
Mac 400m SE	Victoria	13	5	10	43	16	32	10	3	7	2

Site	Outfall	CB-198/199	CB-201	CB-202	CB-205	CB-206	CB-208	CB-209	Total
Iona 500m N	Vancouver	860	108	161	373	48	122	264	57375
Iona 500m N DUP	Vancouver	931	119	187	370	64	133	322	59535
Iona Zero	Vancouver	590	77	117	232	29	73	181	39953
Iona 200m S	Vancouver	121	1224	282	551	47	181	545	83594
Mac 200m SE	Victoria	31	3	8	63	6	30	84	2214
Mac 200m NW	Victoria	14	1	3	9	1	3	6	1639
Mac Zero	Victoria	63	9	19	41	5	14	28	3702
Mac 400m SE	Victoria	20	3	6	24	3	9	33	1437
Mac 200m NW	Victoria	8	1	2	7	1	3	9	714
Mac Zero	Victoria	229	27	46	314	46	150	1260	14316
Mac 200m SE	Victoria	87	19	53	89	10	47	113	5297
Mac 200m NW	Victoria	6	1	1	5	0	2	5	509
Mac 400m SE	Victoria	20	2	5	21	2	9	32	2519

Appendix G
PCB concentrations in benthic invertebrates (pg g^{-1} $lipid^{-1}$)

Site	Outfall	CB-6	CB-11	CB-15	CB-16/32	CB-17	CB-18	CB-22	CB-25	CB-26	CB-27	CB-31	CB-37	CB-42/68
Iona 400m N Jar 1/1	Vancouver	71	1104	479	360	730	1686	936	243	468	92	2659	726	1287
Iona 400m N Jar 1/1	Vancouver	137	1396	575	42	912	2106	966	254	531	131	3206	1273	1498
Iona 400m N Jar 2/2	Vancouver	1578	4251	2592	1644	5609	7143	2612	449	1105	491	7506	3210	2210
Iona Zero	Vancouver	151	1053	249	1072	886	1837	775	242	424	123	2923	591	1361
Iona 200m N	Vancouver	532	2385	987	1965	3164	3523	1201	346	747	197	4402	1369	2121
Iona 400m N	Vancouver	349	2386	1109	2247	3016	4451	1510	439	896	203	6040	1357	2205
Iona 400m N DUP	Vancouver	364	2140	882	2034	2465	3647	1383	379	729	144	5113	1189	2146
Iona 200m S	Vancouver	449	2478	1726	3660	6563	6250	1871	552	995	378	6310	2218	3253
Iona 400m S	Vancouver	528	3491	2407	3494	7737	6237	1839	510	1058	306	7166	2028	3043
Mac 200m SE	Victoria	165	1436	432	1023	1652	4031	681	211	374	323	2085	646	1400
Mac Zero	Victoria	158	1097	373	737	443	1255	640	230	459	371	2186	559	1383
NW	Victoria	196	1297	297	802	775	2204	439	147	355	192	1733	483	1117
Mac 400m SE	Victoria	105	1014	296	489	331	736	428	140	262	151	1275	576	887
NW	Victoria	116	1222	190	647	497	1325	344	<60	236	146	1108	301	1008

Site	Outfall	CB-43/49	CB-44	CB-45/51	CB-46	CB-52	CB-56	CB-60	CB-63	CB-64/71	CB-66	CB-77	CB-82
Iona 400m N Jar 1/1	Vancouver	3980	3840	628	185	6744	1905	804	258	2927	3692	180	1401
Iona 400m N Jar 1/1	Vancouver	4209	4428	860	196	7062	3287	3627	283	2824	6168	847	2547
Iona 400m N Jar 2/2	Vancouver	10502	9034	1332	218	13232	5860	1798	367	6018	8764	542	2902
Iona Zero	Vancouver	4508	3692	585	151	7324	1814	776	239	3163	4014	189	1574
Iona 200m N	Vancouver	7931	6678	1226	320	11374	3679	1479	448	5148	6490	376	2533
Iona 400m N	Vancouver	8413	7027	1332	317	12872	3193	1551	490	5351	6518	474	2710
Iona 400m N DUP	Vancouver	7198	5783	1032	227	10631	3302	1214	432	4705	5881	146	2304
Iona 200m S	Vancouver	13269	9778	2035	429	17398	5137	2150	730	8516	9581	824	3557
Iona 400m S	Vancouver	14436	8811	1779	504	17222	5532	2212	723	7615	9959	462	3711
Mac 200m SE	Victoria	6863	6981	679	234	16690	2196	864	<11	3649	5345	166	1924
Mac Zero	Victoria	7925	12587	629	214	25570	3335	1566	400	4888	8342	201	4692
NW	Victoria	4652	5438	885	297	8690	1378	770	173	2684	3075	219	2574
Mac 400m SE	Victoria	3951	3193	505	182	7412	1827	790	<10	2234	4294	216	1084
NW	Victoria	4246	4522	735	235	8188	1651	584	185	2680	2985	143	1628

Site	Outfall	CB-83	CB-84	CB-92	CB-103	CB-105	CB-114	CB-118	CB-123	CB-130	CB-131/142	CB-132	CB-133
Iona 400m N Jar 1/1	Vancouver	462	1976	2381	110	3653	252	10384	143	1397	107	4714	397
Iona 400m N Jar 1/1	Vancouver	853	3162	2908	109	7059	437	19835	217	1948	242	5572	484
Iona 400m N Jar 2/2	Vancouver	954	4186	4391	137	7212	425	21181	577	2768	161	9770	443
Iona Zero	Vancouver	450	1647	2035	111	3580	268	10791	201	797	96	3168	253
Iona 200m N	Vancouver	364	3688	3665	211	6822	425	19618	445	2141	209	6715	595
Iona 400m N	Vancouver	919	3409	4023	167	7327	482	19827	524	2433	183	7784	679
Iona 400m N DUP	Vancouver	749	2731	3426	137	5867	377	16493	471	2039	112	6215	589
Iona 200m S	Vancouver	1661	5354	5300	316	8398	486	24906	826	3330	221	10041	992
Iona 400m S	Vancouver	931	4577	5012	232	8833	497	26828	557	3018	237	9258	727
Mac 200m SE	Victoria	913	3693	4545	263	6746	444	22152	275	1577	149	5716	519
Mac Zero	Victoria	2093	11387	7464	224	18783	944	46997	708	2863	549	13051	512
NW	Victoria	1221	4774	4778	363	4532	274	15951	206	2293	255	9193	715
Mac 400m SE	Victoria	596	1986	3282	58	7063	390	24116	198	1305	95	3273	480
NW	Victoria	954	4232	3940	<35	4002	209	13812	403	2163	86	6038	560

Site	Outfall	CB-134/143	CB-136	CB-137	CB-141	CB-144	CB-146	CB-147/149	CB-153	CB-156	CB-158/160	CB-167
Iona 400m N Jar 1/1	Vancouver	688	1349	565	1555	405	3918	13492	24806	1205	905	701
Iona 400m N Jar 1/1	Vancouver	1121	2015	973	3200	814	4962	20063	28698	1742	1643	896
Iona 400m N Jar 2/2	Vancouver	1743	4032	648	4941	2136	7300	34219	54421	2007	2640	1036
Iona Zero	Vancouver	458	1081	466	1710	499	2594	9840	15894	1387	1089	521
Iona 200m N	Vancouver	905	2028	851	2284	1069	6009	19348	36429	2409	1987	1206
Iona 400m N	Vancouver	1101	2142	841	2303	993	6917	22205	42127	2148	1544	1287
Iona 400m N DUP	Vancouver	871	1759	580	1754	1019	5883	19237	35054	2058	1351	1176
Iona 200m S	Vancouver	1445	3023	761	2054	1561	8897	31203	53469	2336	1997	1218
Iona 400m S	Vancouver	1246	2538	821	2472	1429	8782	28999	50705	3118	2946	1494
Mac 200m SE	Victoria	854	1987	1032	2681	1158	4825	17233	32022	2528	2353	1006
Mac Zero	Victoria	1983	3229	2179	5253	1475	5085	22393	31805	6003	5277	1489
NW	Victoria	1235	2999	1075	3236	1522	6754	26765	41659	2463	2324	1167
Mac 400m SE	Victoria	599	1288	1025	2025	677	4624	12577	32050	2072	2236	969
NW	Victoria	1052	2559	825	2404	1055	6254	21302	31859	1980	2351	967

Site	Outfall	CB-170/19	CB-171/17	CB-172/19	CB-174	CB-175	CB-176	CB-177	CB-178	CB-179	CB-180/193
Iona 400m N Jar 1/1	Vancouver	3273	928	414	2810	149	430	3355	1014	1840	7330
Iona 400m N Jar 1/1	Vancouver	4154	1137	455	3793	239	629	3736	1283	2428	8299
Iona 400m N Jar 2/2	Vancouver	10155	3175	114	13723	711	1925	10188	4557	7497	14233
Iona Zero	Vancouver	2616	732	513	2011	132	227	1926	737	1073	5305
Iona 200m N	Vancouver	5438	1762	934	4207	329	664	5442	2079	2583	9956
Iona 400m N	Vancouver	5831	1659	871	5018	318	771	6669	2293	2774	11561
Iona 400m N DUP	Vancouver	5253	1490	746	4689	260	647	5562	2072	2478	10218
Iona 200m S	Vancouver	7110	2539	778	6535	692	1345	9625	3919	4347	10106
Iona 400m S	Vancouver	7576	2364	818	7579	504	1100	8366	3795	3357	10132
Mac 200m SE	Victoria	4227	1110	805	3047	204	447	3239	1600	2063	9979
Mac Zero	Victoria	4171	1097	505	2356	146	254	1999	720	1142	7436
NW	Victoria	5035	1733	1002	4105	376	645	4652	2108	2790	10203
Mac 400m SE	Victoria	3149	1001	545	1706	167	268	2122	1264	1184	7214
NW	Victoria	5020	1482	684	3526	354	503	4240	1893	2333	10708

Site	Outfall	CB-183/185	CB-187	CB-194	CB-195	CB-196/203	CB-197/200	CB-198/199	CB-201	CB-202	CB-205
Iona 400m N Jar 1/1	Vancouver	2260	7807	1181	755	2082	206	283	2933	724	97
Iona 400m N Jar 1/1	Vancouver	3228	8990	1288	834	2314	437	341	2254	890	106
Iona 400m N Jar 2/2	Vancouver	11884	27961	2438	2424	10222	2804	1095	13348	4618	112
Iona Zero	Vancouver	1661	4481	916	365	1490	119	192	2051	503	53
Iona 200m N	Vancouver	4302	12497	1741	1300	5281	541	663	6867	1385	218
Iona 400m N	Vancouver	4769	15579	2233	1662	5467	549	620	7832	2093	162
Iona 400m N DUP	Vancouver	3998	13280	1966	1407	4881	406	565	7258	1564	157
Iona 200m S	Vancouver	8253	23524	2340	2065	7118	1175	1080	11509	3808	241
Iona 400m S	Vancouver	8869	22449	2592	2300	8936	1354	1154	12981	3461	318
Mac 200m SE	Victoria	3235	9655	2520	717	4403	179	426	5962	1658	103
Mac Zero	Victoria	1877	3751	895	349	1336	1277	158	1434	1573	31
NW	Victoria	4119	12418	1793	766	3359	194	378	4688	1463	96
Mac 400m SE	Victoria	2289	7152	1541	476	2162	<50	251	3139	1101	77
NW	Victoria	3938	11353	2225	1056	3656	752	426	4160	1518	120

Site	Outfall	CB-206	CB-208	CB-209	Total
Iona 400m N Jar 1/1	Vancouver	842	305	431	261724
Iona 400m N Jar 1/1	Vancouver	867	301	448	358852
Iona 400m N Jar 2/2	Vancouver	3978	3111	5099	665056
Iona Zero	Vancouver	474	159	490	221600
Iona 200m N	Vancouver	2137	864	1324	446081
Iona 400m N	Vancouver	2589	898	1353	486240
Iona 400m N DUP	Vancouver	2131	1081	1837	415678
Iona 200m S	Vancouver	2841	1760	2450	666037
Iona 400m S	Vancouver	4365	1929	4814	668382
Mac 200m SE	Victoria	3339	1392	3330	421222
Mac Zero	Victoria	568	497	565	622095
NW	Victoria	1460	746	1219	427099
Mac 400m SE	Victoria	1521	536	1347	305902
NW	Victoria	1876	645	978	361726