



Baseline

Distribution, accumulation, and potential risk of polybrominated diphenyl ethers in the marine environment receiving effluents from a sewage treatment plant



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ABSTRACT

We investigated the effect of sewage treatment plant effluent on the distribution and accumulation of polybrominated diphenyl ethers (PBDEs) in the marine environment. PBDEs concentrations in seawater and surface sediments were within the ranges 1.58–6.94 ng/L and 2.18–307 ng/g dw, respectively. PBDE concentrations in sediments gradually decreased with increasing distance from inner bay including the sewage outfall. The contribution of effluent to PBDE distributions was very limited. The concentrations of PBDEs in sediment core decreased exponentially with increasing depth. PBDE concentrations in oysters were 4.7–37 ng/g lw. BDE-209 was the dominant congener in marine environmental samples receiving effluent. Estimated dietary exposures of PBDEs by seafood were 0.01–0.08 ng/kg body weight/day, which were more than six orders of magnitude lower than the reported levels of PBDEs with the no observable adverse effects. Hazard quotients demonstrated that, at present, PBDEs posed no potential risk to benthic organisms in the study area.

Since the late 1970s, polybrominated diphenyl ethers (PBDEs) have been added to a wide range of household products, including textiles, furniture, and electronic equipment to reduce fire hazards (de Wit, 2002; Peng et al., 2009; Xiang et al., 2013; Lee and Kim, 2017). Although the use of these compounds can reduce fire accidents and save lives and property, they have had unforeseen environmental consequences (Wu et al., 2013). Because of their potential bioaccumulation, persistence, and the detrimental health effects associated with exposure, manufacturing of two of three PBDE formulations (i.e. penta- and octa-BDE) were discontinued in the U.S. in December 2004 (Renner, 2004; La Guardia et al., 2012) and these compounds were added to the Persistent Organic Pollutants (POPs) list of the Stockholm Convention in May 2009 (Stockholm Convention, 2009). In July 2008, use in electrical equipment of the third and most widely used PBDE formulation, deca-BDE, was banned in the European Union (Betts, 2008; La Guardia et al., 2012) and it was phased out of production in the USA by the end of 2013 (US EPA, 2010). However, it is still widely used in Asia, including in China and Korea (BSEF, 2010; Clarke et al., 2010; Lee and Kim, 2017). In most cases, PBDEs are simply mixed into the polymer products without chemical bonding. They can leach out from these products during production, usage, or disposal (de Wit, 2002; Song et al., 2006). Despite restrictions on their use, PBDEs are

incorporated in many products that are currently in common use, and they will likely be cycled through the environment for some time to come (Lee and Kim, 2017).

Sewage treatment plants (STPs) have been considered a major point source of many POPs, including PBDEs and polychlorinated biphenyls (Wang et al., 2007; Zeng et al., 2011). A large proportion of these contaminants can be partitioned or adsorbed into sewage sludge, but there may still be residues in the effluent that is released into the receiving water bodies (North, 2004; Zeng et al., 2011). High levels of PBDEs have been found in STP effluent and sludge and may be widely dispersed via the use of sludge as fertilizer on agricultural lands (Hale et al., 2003). Because of their low vapour pressure and high hydrophobicity, PBDEs are strongly adsorbed onto soil, sediments, and suspended organic material in the water column, thus facilitating their transfer to aquatic organisms (Shaw and Kannan, 2009; Loganathan and Lam, 2011). This can result in detrimental effects on aquatic ecosystems (Wakelin et al., 2008). Although it accounts for only a small fraction of the total release of PBDEs from STPs (North, 2004; Song et al., 2006; Lee and Kim, 2017), the mass loading of PBDEs via effluent should not be ignored (La Guardia et al., 2007), and recent studies have demonstrated that this is a point source of environmental PBDE contamination (Toms et al., 2006, 2008; Lee and Kim, 2017).

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A great deal of data is available about the presence of PBDEs in a wide variety of environmental compartments in Korea (Moon et al., 2007, 2010; Hong et al., 2010; Choi et al., 2014; Byun et al., 2013; Lee et al., 2014a). However, only a few studies of the distribution of PBDEs in sludge have been conducted (Lee et al., 2014b; S. Lee et al., 2014), and studies on PBDEs in marine environments around STPs in Korea are scarce. To our knowledge, the occurrence of PBDEs in STPs, and their potential subsequent release into coastal water have not been reported for the Korean coast. In our previous study (Lee and Kim, 2017), we determined the levels of PBDEs in STP influent, effluent, and sludge as a point source of PBDEs. For a follow-up study, in the present study we evaluated the occurrence and distribution of PBDEs in the marine environment where Tongyeong STP effluents flowed to. The hazard quotient (HQ) was estimated to assess the potential hazard posed by PBDEs in Tongyeong Bay.

Tongyeong STP is located in south coast of Korea and its sewage treatment capacity is 54,000 t of wastewater per day and serves a population of about 116,000. STP consists of primary settlement followed by bioreactor treatment and secondary clarifier. UV disinfection employ before release into the bay. About 80% of influent is from municipal wastewater sources and the effluent is directly discharged into Tongyeong Bay (TSY, 2013; Lee and Kim, 2017).

To assess the effect of seasonal variation on the distribution of PBDEs, seawater ($n = 12$) was sampled in May and August 2012 and January 2013 (Fig. 1). The surface sediment ($n = 12$) and oysters (*Crassostrea gigas*) ($n = 5$) samples were collected in August 2012 and March 2013, respectively. Sediment core ($n = 1$) was collected from near Tongyeong STP sewage outfall from Tongyeong Bay in August 2012. Detailed information on sample collection is available in the Supplementary information (SI).

The accuracy of the instrumental and analytical methods was validated as according to our previous work (Lee et al., 2014a, 2014b). Particulate and dissolved phase of water samples was separated by glass fiber filter (GFF) and solid phase extraction (SPE) disks, respectively. The filtered GFF, SPE disk, sediment and biota (about 15–20 g wet weight) samples were mixed with anhydrous sodium sulfate to remove water. The dried samples were spiked surrogate standard ($^{13}\text{C}_{12}\text{BDE-139}$, 10 ng and $^{13}\text{C}_{12}\text{BDE-209}$, 20 ng) and extracted using a Soxhlet extractor for 16 h with 200 mL of dichloromethane. Activated copper granules were used to remove elemental sulfur. The extracts were finally cleaned up for PBDE analysis using a method described by Lee and Kim (2017). $^{13}\text{C}_{12}\text{PCB-189}$ (4 ng) was used as a GC internal standard.

PBDE congeners were analyzed following the method of Lee and Kim (2017) using an Agilent 6890 gas chromatography coupled with an Agilent 5975 mass spectrometry with helium as the carrier gas. The mass spectrometer was operated in an electron-capture negative ionization mode using methane as the reagent gas. Using the internal standard, calibration curves were produced and quantified for each sample solution of 19 BDE congeners. The 2 μL injections were made in the splitless mode, with a purge time of 0.2 min. For the determination of all PBDEs, the GC column was a 15 m \times 0.25 mm (i.d) fused-silica capillary tube coated with DB-1 (0.25 μm film thickness; Agilent J&W Scientific) column. The injection port was held at 285 $^{\circ}\text{C}$. The temperature program of the GC oven was as follows: initial temperature 2 min at 50 $^{\circ}\text{C}$; first rate 20 $^{\circ}\text{C}/\text{min}$ to 200 $^{\circ}\text{C}$, final rate 10 $^{\circ}\text{C}/\text{min}$ 320 $^{\circ}\text{C}$ (hold 5 min). For identification and quantitation of each PBDE, two and more target and confirmation ions were selected.

Nineteen PBDE congeners representing the most abundant PBDE congeners in the marine environment were investigated in all samples: BDE-17, -28, -47, -66, -99, -100, -138, -153, -154, -183, -196, -197, -201, -202, -203, -206, -207, -208, and -209. The limit of detection (LOD) was set to a signal-to-noise ratio of 3, and LODs were from 0.01 to 0.05 ng/g for each PBDE congener (Lee and Kim, 2017). A method blank (sodium sulfate) was included with every seven samples to check for secondary contamination. Average recoveries of $^{13}\text{C}_{12}\text{BDE-139}$ and $^{13}\text{C}_{12}\text{BDE-209}$ were $83 \pm 21\%$ and $103 \pm 25\%$, which satisfied the guidelines of the standard method of the United States Environmental Protection Agency (US EPA, 2007; Lee and Kim, 2017). The standard reference material (SRM 1944; New York/New Jersey waterway sediment) was purchased from NIST (Gaithersburg, MD, USA) and analyzed three times. Recoveries of eight PBDE congeners (BDE-47, -99, -100, -153, -154, -183, -206, and -209) in the standard reference materials ranged from 46% to 119% (Lee and Kim, 2017). BDE-209 had lower recoveries ($46 \pm 1.5\%$) than the other PBDE congeners. In this study, all samples were treated with the size exclusion chromatography for clean up. The low recoveries could be caused by the excessive clean-up procedure. Therefore the concentration of BDE-209 in our study had a relative uncertainty compared with other PBDE congeners. In this study, the reported concentrations were not corrected for the recoveries of surrogate standard (Lee and Kim, 2017).

The hazard quotient (HQ) approach based on the measured contaminant concentrations in sediments was used to assess the potential ecological risk in study area. HQ provides a quantitative estimate of the hazard associated with contaminants (Lemly, 1996; Wu et al., 2013).

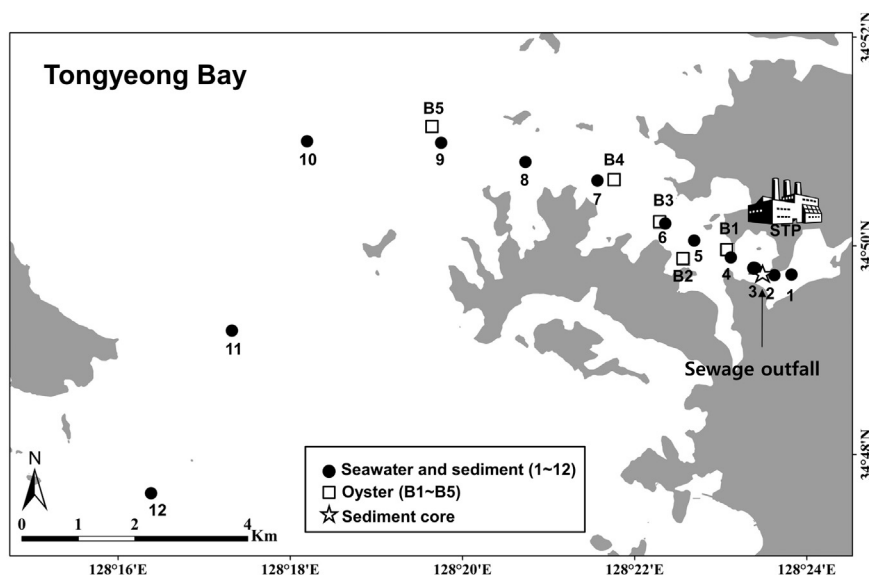


Fig. 1. Location map of sampling sites in Tongyeong Bay for seawater, surface sediment and sediment cores (1–12), and oysters (B1–B5).

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