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Contamination of nonylphenolic compounds in creek water, wastewater treatment plant effluents, and sediments from Lake Shihwa and vicinity, Korea: Comparison with fecal pollution

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ABSTRACT

Nonylphenolic compounds (NPs), coprostanol (COP), and cholestanol, major contaminants in industrial and domestic wastewaters, were analyzed in creek water, wastewater treatment plant (WWTP) effluent, and sediment samples from artificial Lake Shihwa and its vicinity, one of the most industrialized regions in Korea. We also determined mass discharge of NPs and COP, a fecal sterol, into the lake, to understand the linkage between discharge and sediment contamination. Total NP (the sum of nonylphenol, and nonylphenol mono- and di-ethoxylates) were 0.32–875 μg L^{-1} in creeks, 0.61–87.0 μg L^{-1} in WWTP effluents, and 29.3–230 μg g^{-1} TOC in sediments. Concentrations of COP were 0.09–19.0 μg L^{-1} in creeks, 0.11–44.0 μg L^{-1} in WWTP effluents, and 2.51–438 μg g^{-1} TOC in sediments. The spatial distributions of NPs in creeks and sediments from the inshore region were different from those of COP, suggesting that Lake Shihwa contamination patterns from industrial effluents differ from those from domestic effluents. The mass discharge from the combined outfall of the WWTPs, located in the offshore region, was 2.27 kg d^{-1} for NPs and 1.00 kg d^{-1} for COP, accounting for 91% and 95% of the total discharge into Lake Shihwa, respectively. The highest concentrations of NPs and COP in sediments were found in samples at sites near the submarine outfall of the WWTPs, indicating that the submarine outfall is an important point source of wastewater pollution in Lake Shihwa.

1. Introduction

Lake Shihwa, an artificial saltwater lake (surface area = 56.5 km^2, drainage basin = 476.5 km^2), is located on the west coast of South Korea (Fig. 1) where the tidal flats are developed and the lake provides habitat for aquatic animals and birds. There are large industrial complexes in the Lake Shihwa region (total industrial area = 31 km^2), where more than 8500 companies produce metal products, machinery, equipment for industry, and chemicals and chemical products. The cities of Shiheung and Ansan (total population exceeds 1 million) are located on Lake Shihwa. Rapid growth of the population and industrial development has led to deterioration of water quality and biodiversity in this region (Li et al., 2004a,b; Kim et al., 2009; Yoo et al., 2009).

In 2000, the South Korean government designated Lake Shihwa as a special management coastal zone and constructed a water gate to promote exchange of water with the adjacent Yellow Sea. However, many studies continue to report substantial contamination in Lake Shihwa water, sediment, and biota from trace metals and toxic organic contaminants such as polychlorinated biphenyls, polycyclic aromatic hydrocarbons, perfluorinated compounds, and nonylphenols (NPs) (Li et al., 2004a,b; Koh et al., 2005; Kim et al., 2009; Yoo et al., 2009; Hong et al., 2010). In particular, it was reported that Lake Shihwa was one of the most contaminated areas in Korea for nonylphenol (Li et al., 2004a,b; Koh et al., 2005; Choi et al., 2009). Although many studies have investigated the contamination of Lake Shihwa by chemicals from industrial wastewater pollution, little attention has been focused on pollution from wastewater treatment plants (WWTPs). WWTPs treat approximately 5×10^8 kg d^{-1} of wastewater and discharge the effluent and any associated chemical or fecal contamination through an underground pipeline to Lake Shihwa (MOE, 2009). We assessed contamination from wastewater discharge by using chemical wastewater indicators such as NPs and comparing with fecal sterols. To our knowledge, this is the first report on the contribution of these WWTP-derived compounds to wastewater pollution in the coastal industrialized region of Korea.

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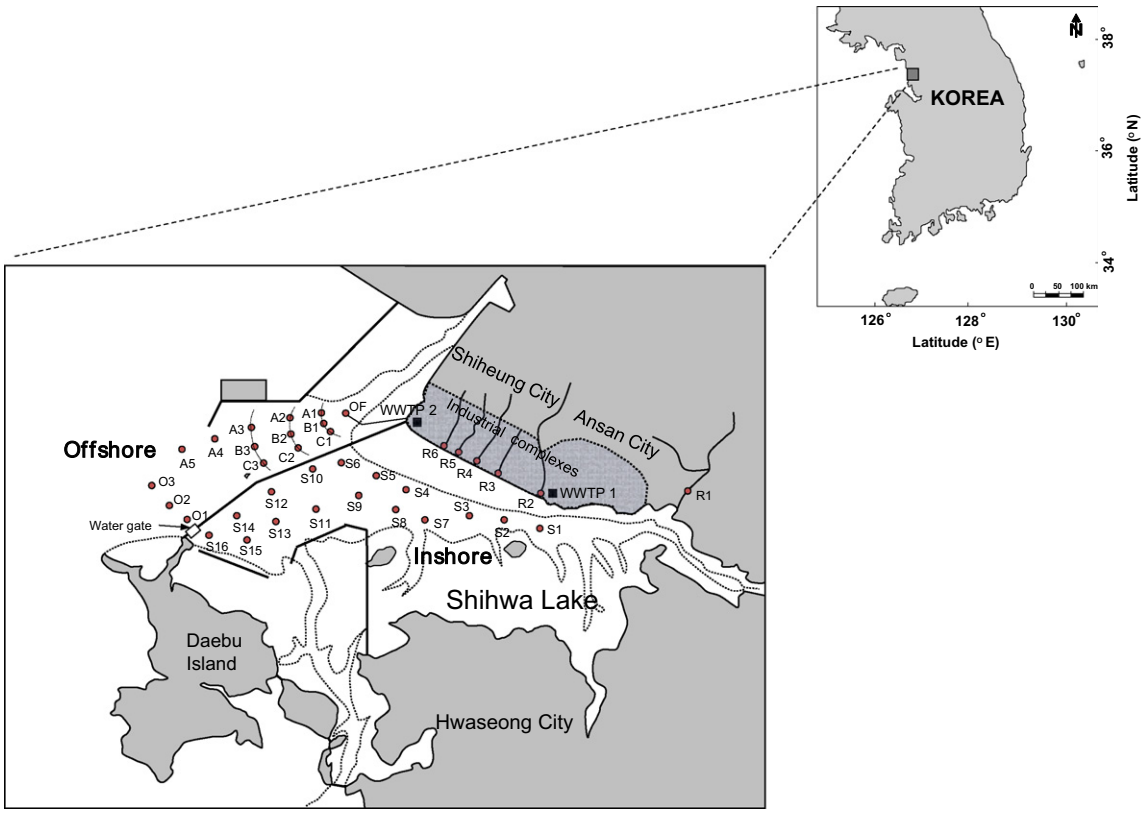

Fig. 1. Sampling locations of creek water (R1–R6), wastewater treatment plant effluent (WWTP1 and WWTP2), and surface sediment samples [S1–S16, OF (the outfall of WWTPs), A1–C3, O1–O3] from the artificial seawater Lake Shihwa, Korea.

NPs are metabolites of nonionic surfactants widely used in a variety of industrial processes and residential and commercial cleaning products since the 1940s. These contaminants are commonly found in effluents from industrialized areas (Bennie et al., 1997; Kannan et al., 2003; Li et al., 2004a,b; Choi et al., 2009) and have been used as markers for industrial contamination (Diez et al., 2006; Lara-Martin et al., 2008). Coprostanol (COP), a fecal sterol, has been widely used as an indicator of fecal pollution in the aquatic environment because of its occurrence at high concentrations in human feces (40–60% of total fecal sterols excreted; Chan et al., 1998). Distribution profiles of COP together with other fecal sterols such as cholestanol (CHOA) can provide useful information on urban sewage pollution in the marine environment. Grimalt et al. (1990) suggested a range of 0.7–1.0 for COP/(COP + CHOA) as characteristic of urban sewage polluted sediments and <0.3 as unpolluted sediments. High COP/(COP + CHOA) ratios of approximately 0.6 also were found in locations adjacent to a sewage outfall (Jeong and Han, 1994) and a sewage sludge disposal site (Chan et al., 1998).

The present study investigated current contamination of NPs in sediments from Lake Shihwa region compared to the distribution of fecal sterols. We also investigated creek surface water and wastewater treatment plant effluents (WWTPEs) and their mass discharge into the Lake Shihwa, to understand the linkage between discharge and sediment contamination.

2. Materials and methods

2.1. Sample collection

The sampling locations for creek water, WWTPEs, and sediment from Lake Shihwa in Korea are presented in Fig. 1. Surface sediment samples (0–4 cm depth) were collected using a box core sampler at 16 stations (S1–S16) in the inshore region and 15 stations in the offshore region of the lake in March 2008 (Fig. 1). The collected samples were individually wrapped in aluminum foil and immediately frozen. The sediment samples were transported to the laboratory, stored at −20 °C, and subsequently freeze dried.

Water samples were collected monthly between June 2008 and May 2009 from six creeks (R1–R6) discharging from the inshore industrialized region and from two WWTPs (WWTP1 and WWTP2) whose outfalls discharge at depth further offshore in Lake Shihwa. WWTP1 and WWTP2 treat 2.77×10^8 kg d^{-1} and 2.14×10^8 kg d^{-1} of wastewaters, respectively (MOE, 2009). Influent volumes received by the plants are derived from both industrial (55%) and domestic (45%) sources. Both WWTPs use an activated sludge for biological treatment, followed by a biological aerated filtration for WWTP1 and sand filtration for WWTP2 (MOE, 2009). The creek water sample collection was conducted at low tide in the surveyed area by using a bucket sampler and flowmeter (BFM001, Valeport, England) to estimate flow rate and mass discharge. Samples were filtered in the laboratory using GFF filters (0.7 μm, Whatman, Maidstone, England) within 2 d of collection.

2.2. Analytical procedures

The water and sediment samples were analyzed for NPs [nonylphenol (NP) and nonylphenol mono- and di-ethoxylates ($NP_{1+2}EO$)] and two fecal sterols (COP and CHOA) according to the method of Choi et al. (2007, 2009) and Li et al. (2007). For NPs (NP and $NP_{1+2}EO$; pure standards from Cambridge Isotope Laboratories, Andover, MA, USA), the filtered water samples were extracted three times by liquid–liquid extraction, and the filter and freeze-dried sediment samples were extracted twice with a mechanical