Distribution of Fecal Sterols, Nonylphenol, and Polycyclic Aromatic Hydrocarbons in Surface Water from Masan Bay, Korea

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Fecal sterols, nonylphenolic compounds (NPs), and polycyclic aromatic hydrocarbons (PAHs) were determined in surface water from Masan Bay and its adjacent rivers in February 2005. Concentrations of coporstanol (Cop), an indicator of fecal pollution, in surface water ranged from <10 to 13,853 ng/L, and concentrations of nonylphenol, the most toxic of the NPs, ranged from 10.2 to 481 ng/L, and concentrations of PAHs ranged from 8.61 to 223 ng/L. The concentrations of the compounds measured in this study were lower than or comparable to those at other locations in Korea and other countries. The contamination of Cop and PAHs in surface water was associated with the discharge from rivers passing through cities and/or industrial complexes. The NP contamination was associated with wastewater treatment plant (WWTP) effluents through outfalls as well as riverine discharge. Compared to ecotoxicological values, the concentrations of NPs from rivers, the mouths of rivers, and WWTP outfall areas exceeded guidelines, suggesting that hot spot areas may pose a potential risk to sensitive species.

Key words: Coprostanol, Nonylphenol, Riverine discharge, Wastewater treatment plant

Introduction

Masan Bay, one of the most heavily contaminated estuarine systems in Korea, was designated as part of the special management coastal area in 1983. The area is surrounded by the cities of Masan, Changwon and Jinhae cities which are highly urbanized (>1 million people in the drainage basin, with an average population density of 2,682 people/km^2) and heavily industrialized (operation of about 1,300 industrial plants). The rapid urbanization and industrialization of this area was accompanied by considerable environmental pollution and social problems. Many studies have reported on the substantial contamination in Masan Bay from toxic organic contaminants (Khim et al., 1999; Hong et al., 2003; Yim et al., 2005; Moon et al., 2008; Choi et al., 2009a, b). High concentrations of contaminants in the Masan Bay sediments were reported to be associated with

anthropogenic activities. Thus, the pathways and fates of contaminants from anthropogenic activities must be investigated to design strategies for environmental management of risks due to toxic contaminants.

Coprostanol (Cop), a fecal sterol, has been widely used as an indicator of fecal and sewage contamination in aquatic environments because it exists in high concentrations in human feces (40-60% of total fecal sterols excreted) (Chan et al., 1998). Distribution profiles of fecal sterols together with other related sterols can provide useful information on source identification in the marine environment (Leeming et al., 1996; Mudge and Lintern, 1999). Nonylphenolic compounds (NPs) are a group of nonionic surfactants widely used in a variety of industrial processes, and residential and commercial cleaning products since the 1940s. NPs act as endocrine disrupters and are frequently found in effluents from industrial complexes (Bennie et al., 1997; Li et al., 2004). Cop and NPs have regularly

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used to investigate the degree of contamination of surface water and sediments from wastewater (Leeming and Nichols, 1996; Elhmmali et al., 2000; Noblet et al., 2004; Diez et al., 2006; Lara-Martin et al., 2008).

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental pollutants that primarily result from anthropogenic activities. PAHs are generated from incomplete fuel combustion, domestic and industrial wastewaters, and spillage of crude oil and its refined products (Hites et al., 1997; Pettersen et al., 1997). Because PAHs are toxic, carcinogenic, and mutagenic to all organisms, including humans, PAHs have been extensively studied in various environmental and biological compartments (Yim et al., 2005; Moon et al., 2007; Moon et al., 2010).

Most contaminants are transported to the coastal areas by water and eventually disappear. Investigation of surface water can provide information on present pollution and pathways of contaminants. However, little attention has focused on occurrence and distribution of organic contaminants in freshwater and saltwater from Korean coastal waters including Masan Bay. We assessed water contamination from anthropogenic impacts using the major organic contaminants (Cop, NPs, and PAHs) in urban and industrial effluents. The present study investigated fecal sterols [Cop, cholesterol (Chst), and cholestanol (Chsta)], NPs [nonylphenol (NP), nonylphenol mono-ethoxylate (NP_1EO), and nonylphenol diethoxylate (NP_2EO)], and 16 PAHs in surface water from Masan Bay and its adjacent rivers.

Materials and Methods

Sample collection

Water samples were collected in February 2005 from six rivers (Hoewon, Sanho, Changwon, Nam, Guidong and Sini rivers) that discharge to Masan Bay and from 40 stations in Masan Bay (Fig. 1). Samples, collected by water-sampler, were separated into particulate and liquid phases. Particulate samples were isolated by filtering the water through pre-combusted glass fiber filters (GF/F, Whatman, Florham Park, NJ, USA). Particulate samples were stored at -30°C until extraction. Liquid samples were acidified with 6 N HCl to remove an effect of microbial degradation and stored at 4°C.

Analytical method

Detailed descriptions of water sample extraction and cleanup procedures for fecal sterols, NPs and PAHs have been reported previously (Choi et al.,
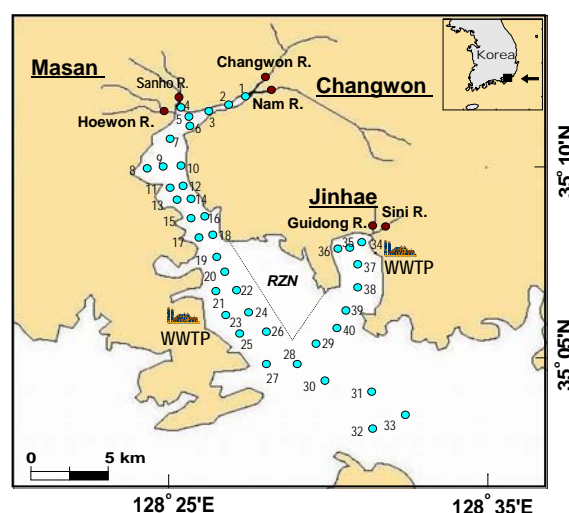

Fig. 1. Map showing the sampling locations of six rivers and seawater collected from Masan Bay, Korea. WWTP indicates the wastewater treatment plant, and RZN indicates the restricted zone for the navy.

2007; Moon et al., 2007). For fecal sterols, briefly, filter samples were placed in respective 50 mL Teflon centrifuge tubes with Teflon caps. A surrogate standard, 1-nonadecanol (Dr. Theodor Schuchardt & Co, Hohenbrunn, Germany), was added to each tube, and the samples were extracted twice using dichloromethane-chloroform (ultra residue analysis; J.T. Baker, Phillipsburg, NJ). The extracts were then passed through a Florisil (60-100 mesh, reagent grade; Sigma-Aldrich, St. Louis, MO, USA) chromate-graphy column. The eluents were concentrated and derivatized using N,O-bis [trimethylsilyl] trifluo-roacetaminide (BSTFA; Sigma-Aldrich). After the addition of internal standards, the concentrated eluents were transferred to vials for instrumental analysis.

For NPs (NP, NP_1EO, and NP_2EO; Cambridge Isotope Laboratories, Andover, MA, USA), the filtered water samples were extracted twice by liquid-liquid extraction, and the filter samples were extracted by a mechanical shaker using dichloro-methane (ultra residue analysis; J.T. Baker). A surrogate standard, nonylphenol-$^{13}C_6$ (Cambridge Isotope Laboratories) was added to the matrix before extraction. The extracts were concentrated to about 1 mL under a gentle stream of dry nitrogen. The extract was concentrated to 0.5 mL, derivatized using BSTFA, and then cleaned by passage through a Florisil chromatography column. After the addition of internal standards, the concentrated eluents were transferred to vials for instrumental analysis.

For PAHs, briefly, the filtered water samples were

extracted twice by liquid-liquid extraction, and the filter samples were extracted for 5 h by reflux extraction. The extract was cleaned by passage through an activated silica gel (70-230 mesh, Merck, Whitehouse Station, NJ) column. The eluents were concentrated to approximately 1 mL and were evaporated at room temperature to 50 to 100 µL. The residues were dissolved with 50 µL n-nonane (pesticide grade, Fluka, Switzerland) for instrumental analysis. This study analyzed 16 non-alkylated PAHs (48755-U, Supelco, Bellefonte, PA, USA) that are recognized as priority pollutants by the United States Environmental Protection Agency were analyzed. These include naphthalene (NaP), acenaphthylene (AcPy), acenaphthene (AcP), fluorene (Flu), phenanthrene (PhA), anthracene (AnT), fluoranthene (FluA), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-c,d]pyrene (InP), dibenzo[a,h]anthracene (DbA) and benzo[g,h,i]perylene (BgihP). Surrogate stan-dards (NaP-d8, AcPy-d8, FluA-d12, PhA-d10, Pyr-d10, BaP-d12, and BghiP-d12; Cambridge Isotope Laboratories) were spiked to verify the recovery.

Fecal sterols, NPs, and PAHs were quantified using a gas chromatograph (GC-6890 series: Agilent, Wilmington, DE, USA) equipped with a mass spectrometer (5973N; Agilent). The capillary column used was a DB-5MS (30 m, 0.25 mm i.d., and 0.25 m film thickness, J&W Scientific, Palo Alto, CA, USA). The MS was operated in selected ion monitoring mode, using the molecular ions of the compounds mentioned.

QA/QC and statistical analysis

All of the surrogate standards were detected with no interferences. Solvents injected before and after the injection of standards showed negligible contamination or carryover. Blanks did not contain quantifiable amounts of the target compounds. The recovery of the target compounds was based on five replicate analyses of filters and purified water (HPLC grade; J.T. Baker) spiked with the target compounds. The recoveries of fecal sterols, NPs, and PAHs were $89 \pm 5\%$, $81 \pm 12\%$, and $75 \pm 17\%$, respectively.

Statistics were calculated using Excel 2007 software (Microsoft, Redmond, WA, USA) for Windows, and a Pearson correlation analysis was performed to investigate the relationships between chemical compounds after data transformation using SPSS software for Windows 10.0 (SPSS 2000).

Results and Discussion

Fecal sterols

Concentrations of fecal sterols, NPs and PAHs in surface water samples are summarized in Tables 1 and 2. Concentrations of fecal sterols (Cop, Chst, and Chsta) were measured in the particulate phase because of their strong affinities to particulate matter (log $Kow = 8.42$) (Mudge and Ball, 2006). LeBlac et al. (1992) reported that the particulate faction of Cop was 94% of the total dissolved and particulate Cop in point source, river, and seawater. Fecal sterols in the present study were detected in almost all surface water samples (39 of 46 samples for Cop, 46 samples for Chst, and 38 samples for Chsta). Concentrations of Cop, an indicator of fecal pollution, in surface water ranged from <10 (less than the limit of detection) to 13,853 ng/L (mean: 1,324 ng/L). Concentrations of Chst and Chsta ranged from 133 to 18,010 ng/L (mean: 2,113 ng/L), and <10 to 1,992 ng/L (mean: 247 ng/L), respectively. Chst was predominant in the fecal sterols, accounting for 75.5% (16.0-100%) of the total sterols. It may be because Chst has various sources in the marine environment. In fact, Chst is present in many marine organisms

Table 1. Summary of fecal sterols, and nonylphenolic compounds in surface water from Masan Bay in Korea (ng/L)

		Coprostanol	Cholesterol	Cholestanol	Nonylphenol	NP1EO[a]	NP2EO[b]	ΣNP[c]
Seawater (n = 40)	Min	<10[d]	133	<10	10.2	23.6	<LOD	55.9
	Max	4,099	7,603	538	110	680	3,631	4,396
	Mean	310	749	66.2	33.2	130	415	578
	Stdev	837	1,408	113	21	121	640	774
River water (n = 6)	Min	4,222	2,536	1,153	95.5	587	784	1,467
	Max	13,853	18,010	1,992	481	1,084	2,386	3,910
	Mean	8,080	11,205	1,450	299	836	1,913	3,048
	Stdev	3,942	5,384	373	168	169	601	893

[a]nonylphenol monoethoxylate, [b]nonylphenol diethoxylate, [c]sum of NP, NP1EO and NP2EO, [d]less than limit of detection.

Table 2. Summary of PAHs in seawater from Masan Bay in Korea (ng/L)

	Min	Max	Mean	Stdev
NaP	0.01	33.6	6.95	5.85
AcPy	0.06	0.83	0.34	0.17
AcP	0.05	1.65	0.42	0.38
Flu	0.16	1.77	0.40	0.27
PhA	0.50	6.02	1.78	1.16
AnT	0.03	2.75	0.48	0.49
FluA	0.43	4.98	1.23	1.08
Pyr	0.29	3.72	0.93	0.78
BaA	0.09	4.82	0.80	1.06
Chr	0.22	9.63	1.77	2.32
BbF	1.30	29.6	5.13	5.05
BkF	0.27	6.98	1.21	1.48
BaP	0.12	15.0	1.79	2.77
InP	1.17	77.4	9.31	15.43
DbA	0.44	54.1	5.94	10.62
BghiP	0.44	21.2	2.59	4.07
$\Sigma CPAH^{a}$	5.66	165	24.2	33.8
ΣPAH^{b}	8.61	223	41.1	46.8

[a]Sum of the six carcinogenic PAHs of IARC recommendation.
[b]Sum of total sixteen PAHs.

from plankton to fish as well as domestic sewage discharge (Mudge and Seguel, 1999; Peng et al., 2002).

Cop concentrations in the present study were lower than or comparable to those reported for other locations; Mokpo coast (94 to 7,568 ng/L) (Choi et al., 2007) and Masan Bay, Korea (<LOD to 23,088 ng/L) (Choi et al., 2005), Mekong delta, Vietnam (<ND to 97,100 ng/L) (Isobe et al., 2004), and the Po River, Italy (8,300 to 9,200 ng/L) (Gilli et al., 2006).

Cop concentrations decreased with increasing distance from land (Fig. 2). This distribution is consistent with a representative trend of toxic organic contaminants observed in Masan Bay (Khim et al., 1999; Yim et al., 2005; Choi et al., 2009b). The distribution may have been impacted by land-based sources and an outside-bound net transport/advection. Cop concentrations greater than 1,000 ng/L were found in all rivers and the mouths of rivers (stations M1, M2, and M4). The concentrations of Cop in the six rivers (mean ± standard deviation; $8,080 \pm 3,942$ ng/L) were about 25 times higher than those (310 ± 837 ng/L) observed in the seawater from Masan Bay. Profiles of fecal sterols such as Cop/(Cop+Chsta) and Cop/Chst, were provided. Cop/(Cop+Chsta) and Cop/Chst ratios, which are useful indicators of urban sewage input showed higher values in sewage (Leeming et al., 1996; Mudge and Lintern, 1999; Mudge and Seguel, 1999). Cop/(Cop+Chsta) ratios ranged from 0.16 to 0.88, and Cop/Chst ratios were -
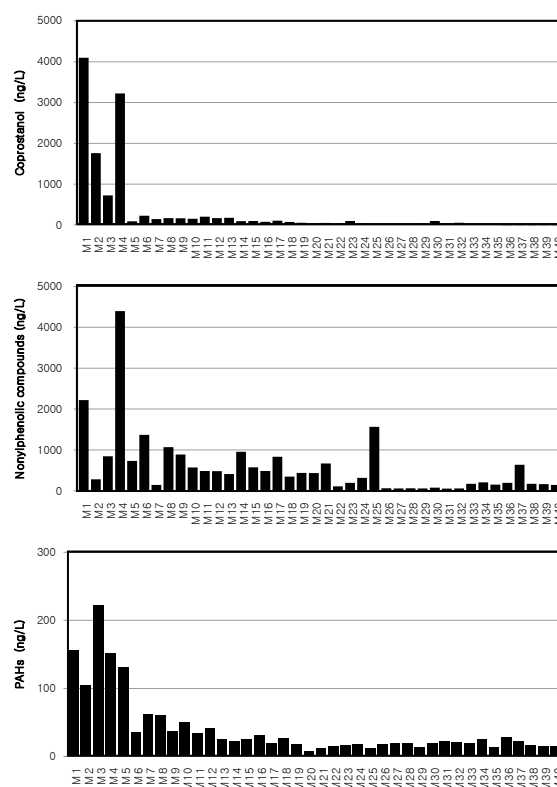

Fig. 2. Spatial distribution of coprostanol, ΣNP (the sum of NP, NP_1EO and NP_2EO), and ΣPAH (sum of 16 PAHs) in seawater from Masan Bay, Korea.

4.54. High values were found in rivers and the mouths of rivers. Spatial distribution of the ratios in seawater was similar to that of Cop concentrations ($r=0.878$-0.881). These results indicate that riverine discharge is an important source of fecal pollution in Masan Bay.

Riverine mass discharge of Cop into Masan Bay can be estimated by using the flow rates reported by Choi et al. (2005). The results showed that the input of Cop from the six rivers was 880 mg/day, and the input from Sanho, Changwon, and Nam rivers contributed to 88% of the total riverine discharge, suggesting that the three rivers are main routes of domestic effluents to Masan Bay.

Nonylphenolic compounds

Measurable levels of NPs were detected in 46 of 46 samples for NP, 46 samples for NP_1EO, and 41 samples for NP_2EO. Concentrations of NP, which is the most toxic of the NPs and has the greatest potential for bioaccumulation, ranged from 10.2 to 481 ng/L (mean: 67.9 ng/L). NP_1EO and NP_2EO were found at concentrations ranging from 23.6 to 1,084 ng/L (mean: 222 ng/L) and from <20 to 3,631 ng/L

(mean: 608 ng/L), respectively. The composition of NPs in surface water was $NP_2EO > NP_1EO > NP$. The composition patterns of NPEOs can be divided into three categories; (i) $NP_2EO > NP_1EO > NP$, (ii) $NP2EO \approx NP_1EO < NP$, and (iii) $NP_2EO > NP > NP_1EO$ (Hong et al., 2010). The composition of case (i) indicates that the sources of NPs are closer than the sources of pollution in case (ii). For case (iii), the composition pattern can be explained by differences in degradation and supply rates between long chain NPEOs that degrade to NP_2EO, and NP_2EO that degrades to NP_1EO and/or NP. The composition patterns in the present study reflect the introduction of fewer degradation products of NPEOs (nonyl-phenol polyethoxylates).

Compared to NP concentrations reported in Korea, the concentrations of NP in the present study were much lower than that in the Shihwa Lake and its surrounding creeks (up to 26,000 ng/L) (Li et al., 2004), and comparable to those in Mokpo coast, Korea (10-770 ng/L) (Choi et al. 2007). This result was within the ranges reported for different countries; Japan (50-1100 ng/L) (Hayakawa et al., 2001; Isobe et al., 2001), China (100-7,300 ng/L) (Shao et al., 2005), Taiwan (1800-10,000 ng/L) (Ding et al., 1999), the United States (<LOD to 640 ng/L) (Naylor et al., 1992), Canada (<LOD to 920 ng/L) (Bennie et al., 1997), Germany (10-400 ng/L) (Zellenr and Kalbfus, 1997), and Switzerland (<LOD to 480 ng/L) (Ahel et al., 2000).

Concentrations of ΣNP (the sum of NP, NP_1EO, and NP_2EO) decreased with increasing distance from land (Fig. 2). Concentrations of ΣNP greater than 1,000 ng/L were found at six rivers, the mouths of the rivers (M1 and M4), and the wastewater treatment plant (WWTP) outfall area (M25). The mean concentration of ΣNP in the rivers ($3,048 \pm 893$ ng/L) was about five times higher than that (578 ± 74 ng/L) in the seawater from Masan Bay, indicating that an important source for NPs is riverine discharge.

Relatively high concentrations in the WWTP effluent outfall areas (M25 and M37) imply that WWTP effluents containing NPs are discharged into the bay. Although NPs are highly treatable in WWTPs with biological treatment facilities, effluent from WWTPs is still considered one of the major sources of NP and short-chain NPEOs due to incomplete removal and degradation of these surfactants (Ferguson et al., 2001; Isobe et al., 2001). Moon et al. (2008) reported heavy contamination of WWTP outfall areas by chemicals associated with industrialized effluents (PCDDs/Fs, doxins-like PCBs, NPs, and PBDEs). The major sources of industrial

wastewater contamination in Masan Bay may be riverine discharge and WWTP effluents.

In the riverine mass discharge of ΣNP, the input of ΣNP from the six rivers was 420 mg/day, and the input from Sanho, Changwon, and Nam rivers contributed to 81% of the total riverine discharge. The mass discharge pattern of ΣNP was similar to that of Cop, indicating that the three rivers are the main routes of industrial and domestic effluents.

Polycyclic aromatic hydrocarbons

All of the PAHs compounds were detected in all the seawater samples (40 of 40 samples). ΣPAH (the sum of 16 PAH compounds) in seawater ranged from 8.61 to 223 ng/L (mean: 40.6 ng/L). The sum of the six potential carcinogenic PAHs (the sum of BaA, BbF, BkF, BaF, InP, and DbA) (IARC, 1984), ranged from 5.66 to 165 ng/L (mean 23.8 ng/L), accounting for 52% of ΣPAH in seawater. The predominant compounds were NaP, BbF, InP, DbA, and BghiP, collectively accounting for $75 \pm 7.6\%$ of ΣPAH.

Concentrations of ΣPAH in the present study were comparable to those in Mokpo coasts (mean 28 ng/L) (Moon et al., 2007). However, the ΣPAH concentrations in the study were lower than those reported in the following coastal regions: Deep Bay, South China (mean: 69 ng/L) (Qiu et al., 2009), Yangtze estuary (mean: 183 ng/L) (Sicre et al., 1993), Seine River (mean: 204 ng/L) (Fernandes et al., 1997), and Bahia Blanca estuary, Argentina (mean: 700 ng/L) (Arias et al., 2009).

Spatial distribution of ΣPAH in seawater showed a clear decreasing trend from inner to outer locations (Fig. 2). Concentrations of ΣPAH greater than 100ng/L were found in the mouths of rivers (stations M1, M2, M3, M4, and M5), and con-centrations greater than 50 ng/L were in harbor areas (stations M7, M8, and M10). Thus, the major source of PAHs in seawater is riverine discharge and shipping traffic is a minor source.

Correlations between contaminants

The results of correlation analysis for Cop, ΣNP, and ΣPAH concentrations in seawater are shown in Fig. 3. The correlations between contaminants showed positive coefficients ($r = 0.498$-0.704, $p < 0.005$), indicating that they may have similar discharge points, transport, mixing and deposition in the Masan Bay. A highly significant correlation was found between the concentrations of Cop and ΣPAH ($r = 0.704$), indicating that these contaminants primarily originated from riverine discharge. The relationships of ΣNP with Cop and ΣPAH were
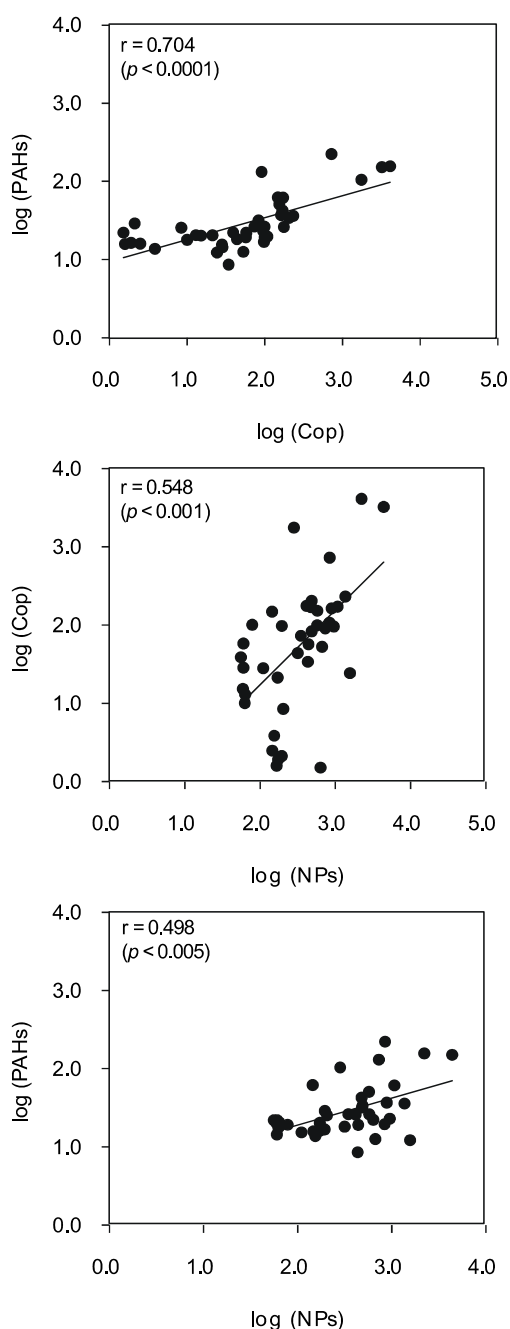
Fig. 3. Correlation between coprostanol, ΣNP, and Σ PAH concentrations in seawater from Masan Bay, Korea.

organisms. Concentrations of NPs and PAHs analyzed in the present study were compared to screening and ecotoxicological values, to evaluate the ecotoxicological risk on aquatic organisms.

Concentrations of PAHs (AcP, Flu, PhA, AnT, FluA, Pyr, BaA, BaP, InP, and DbA) in the present study were two or three orders of magnitude lower than marine screening benchmarks of PAHs for ecological risk assessment (US EPA, 2009), suggesting that contamination of PAHs along Masan Bay is categorized in the low range.

Guidelines and ecotoxicological values for NPs in water have been proposed in various countries to protect aquatic life. The US and European regulatory standards for NP are 1,000 ng/L. The Ministry of the Environment of Japan also suggested that the predicted no effect concentration of NP is 608 ng/L (Furarchi et al., 2004). The Netherlands proposed maximum permissible concentrations (MPCs) for NP (330 ng/L) and NP1EO plus NP2EO (120 ng/L) in water (Jonkers et al., 2005). Environment Canada (2002) proposed the water quality guidelines for ΣNP (1,000 ng toxic equivalency (TEQ)/L for freshwater and 700 ng TEQ/L for seawater) using toxic equivalency factors like dioxins and furans, to consider the additive effects of NP and NPEOs.

NP concentrations measured in the present study did not exceed water quality guidelines, and showed that 7% of 46 water samples exceeded the MPC (330 g/L) (Fig. 4). However, for NPEOs, the NP1EO plus NP2EO concentrations were greater than MPC (120
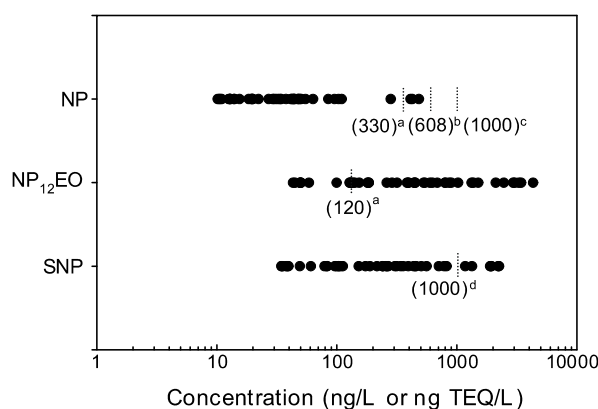
relatively low ($r = 0.548$ and 0.498). It may be because the NP contamination was associated with WWTP effluents through outfalls as well as river discharge.

Ecological concerns

Trace levels of NPs and PAHs can cause chronic effects and disrupt the endocrine systems of aquatic

Fig. 4. Concentrations of nonylphenolic compounds with comparison of screening values reported elsewhere. [a]maximum permissible concentrations for (NP1EO+NP2EO) and NP in the Netherlands (Jonkers et al., 2005), [b]predicted no effect concentration of NP in Japan (Furachi et al., 2004), [c]regulatory standard for NP in USA and Europe, and [d]water quality guideline of NPs in Canada (Environment Canada, 2002).

ng/L) for 83% of 46 samples, and the ΣNP concentrations were greater than the Canadian guidelines for 83% of the six river samples and 10% of 40 seawater samples. These results indicate that hot spot areas such as rivers, the mouths of rivers, and WWTP outfall areas may pose a potential risk to aquatic organisms. Therefore, continuous monitoring of NPs in Korean coastal waters including urbanized, industryalized and sewage affected estuaries may be necessary.

Acknowledgments

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