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Sources and Distributions of Organic Wastewater Compounds on the Mokpo Coast of Korea

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Surface water and sediment samples collected from the Mokpo coast of Korea were analyzed for molecular markers of organic municipal wastewaters, i.e., 11 fecal sterols including coprostanol (Cop) and nonylphenolic compounds (NPs), to characterize the main routes of these wastewaters to the coast and to assess contamination levels. Concentrations of Cop ranged from 94 to 7,568 ng/L in surface water and from 43 to 38,108 ng/g dry weight in sediments. Concentrations of NPs [nonylphenol (NP) and nonylphenol mono- and di-ethoxylates ($NP_{1-2}EOs$)] ranged from 123 to 4,729 ng/L in surface water and from 4 to 2,119 ng/ng dry weight in sediments. The levels of these compounds were much higher at stations near the rivers that pass through the urban center of Mokpo and the outfall of the wastewater treatment plant (WWTP). The spatial distribution of Cop levels was statistically similar to that of NPs ($r=0.809$ and 0.982 in surface water and sediments, respectively), indicating that these compounds may have similar discharge points, transport, mixing, and deposition in the study area. These results suggest that considerable amounts of organic wastewater compounds are discharged through rivers and WWTP effluent to the Mokpo coast.

Keywords: Coprostanol, Nonylphenol, Mokpo, Organic wastewater compounds, Wastewater treatment plant

Introduction

Municipal effluents are recognized as a major cause of water quality deterioration in rivers, estuaries and coastal areas. The anthropogenic contaminants present in these effluents may include complex mixtures of household, industrial and agricultural compounds and their degradation products such as surfactants, disinfectants, drugs, pesticides, sterols, and polycyclic aromatic hydrocarbons. Some of these organic wastewater compounds can persist in natural water systems and potentially have long-term effects on aquatic biota. They are hormonally active and have been shown in laboratory studies to disrupt the endocrine system of animals (Jobling et al., 1996; Thorpe et al., 2001). Evidence also indicates that the endocrines system of some fish and other vertebrates have been affected by these compounds (Sumpter and Johnson, 2005), although the mechanisms of endocrine modulation and the long-term, sub-lethal effects of low-level exposure remain poorly understood. Therefore, to evaluate the impact of wastewater discharges, it is necessary to understand the contamination levels and behaviors of these compounds in the environment.

Nonylphenolic compounds (NPs) and fecal sterols have been identified as major organic contaminants in municipal wastewaters and have therefore been considered good organic waste-water indicators (Chalaux et al., 1995; Gonzalez-Orefa and Saiz-Salinas, 1998; Li et al., 2007). NPs are a group of nonionic surfactants widely used since the 1940s in a variety of industrial processes, as well as in residential and commercial cleaning products, and are found at considerable levels in effluent from industrial areas (Li et al., 2004; Choi et al., 2005c). Among the NPs, nonylphenol (NP) and nonylphenol mono- and diethoxylates ($NP_{1-2}EOs$) are the most persistent and therefore most prominent in the aquatic environment

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(Isobe et al., 2001; Jonkers et al., 2005).

Of the fecal sterols, coprostanol (Cop) has been widely used as an indicator of domestic wastewater contamination 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). Thus, the transport and fate of organic wastewater compounds can be assessed by using fecal sterols and NPs as molecular markers.

The Mokpo coast, located in southwest Korea, is characterized by muddy flats with wide tidal ranges. Fresh water flows to the area mainly from the Yeongsan River (Fig. 1). The Yeongsan, Yeongam, and Geumho embankments had been constructed since 1981. These embankments reduced the current flow and led to the accumulation of pollutants along the coast. The inner part of the coast includes harbor and industrial complexes, a large residential area, and a wastewater treatment plant (WWTP). The Daebul industrial complex, founded in 1996, produces chemical pro-ducts, nonferrous metals, machinery, and food pro-ducts (Yoon, 2001). The Samho industrial complex, located on the outer part of the coast, comprises shipyards and machine factories. The rapid industrialization of the area around Mokpo may have been accompanied by significant environmental deterioration that has led to a variety of social and health problems. Until now, environmental studies of contamination of the Mokpo coast have focused on water quality (Yoon, 2001) and trace metals (Kim and Sin, 2002). Little is known about the contamination of the coastal area by organic wastewater from inland sources.

In this study, organic wastewater indicators, i.e., fecal sterols and NPs, were analyzed in 29 sediment and 21 surface water samples from the Mokpo coast, to determine the concentrations, distributions, and sources of organic wastewater compounds.

Materials and Methods

Sample collection

Surface sediment (0-4 cm) and surface water samples were collected from 29 and 21 locations, respectively, along the Mokpo coast in February 2006 (Fig. 1). These samples were collected from the inner to the outer parts of the coastal waters (M1-M13), the harbor zone (H1-H4), the Yeongsan River (R1), a location close to the WWTP (WWTP), streams (R2, R3) that flow into the waters off Mokpo, locations close to those streams (R4, R5), locations near the Samho industrial complex (S1-S4), and locations near a small industrial complex (C1-C2) located in the northern part of the city of Mokpo.

Surface sediment samples were collected using a box-corer deployed from a research vessel. The collected samples were individually wrapped in aluminum foil and immediately frozen on the vessel. The samples were transported to the laboratory in a cooler box with ice or dry ice, stored at -20°C and later freeze-dried. Surface water samples were collected using a bucket sampler, transferred to 10-L polyethylene bottles, and immediately filtered using pre-combusted glass fiber filters (GF/F, Whatman). The filtered samples were acidified with 6 N HCl to depress microbial de-gradation and stored at 4°C and the filters were stored at -20°C until analysis.

Analytical procedures

For the nonylphenolic compounds (NP, $NP_{1-2}EOs$; Cambridge Isotope Laboratories, Andover, MA, USA), sediment and surface water samples were analyzed according to the method of Li et al. (2004). Briefly, approximately 2 g of freeze-dried sediment were placed in a 50 mL Teflon centrifuge tube with a Teflon cap. Two surrogate standards, nonylphenol-$^{13}C_6$ (Cambridge Isotope Laboratories) and bisphenol A-d_{16} (Sigma-Aldrich, St. Louis, MO, USA), and 5 ml of 0.1 N HCl (Suprapur, Merck, Hohenbrum, Germany) solution were added, and the samples were extracted with dichloromethane (Pesticide grade; Merck, Darmstadt, Germany). The extracts were concentrated to about 1 mL under a gentle dry nitrogen flow. Water and sulfur were removed from the extract using anhydrous sodium sulfate (Kanto, Tokyo, Japan) and copper (Sigma-Aldrich), respecttively. The extract was concentrated to 0.5 mL, derivatized using N,O-bis [trimethylsilyl] trifluoro-acetaminide (BSTFA, Sigma-Aldrich), and then cleaned by passage through a Florisil (60-100 mesh, reagent grade, Sigma-Aldrich) chromatography column. After the addition of internal standards, the concentrated eluents were analyzed by gas chromatography-mass spectrometry (Agilent, 6890-5973 N, Wilmington, DE, USA) with selected ion monitoring mode.

For the surface water samples, the surrogate standards were added to 1-2 L of filtered sample, and the sample was extracted twice by liquid-liquid extraction using dichloromethane. The preparation and analysis of the filtered water samples were similar to those of the sediment samples, except the sulfur removal procedure was omitted. For the filters, the analytical method was similar to that of the sediment samples, except the sulfur removal procedure was omitted. The surface sediment and surface water samples were analyzed for 11 fecal sterols

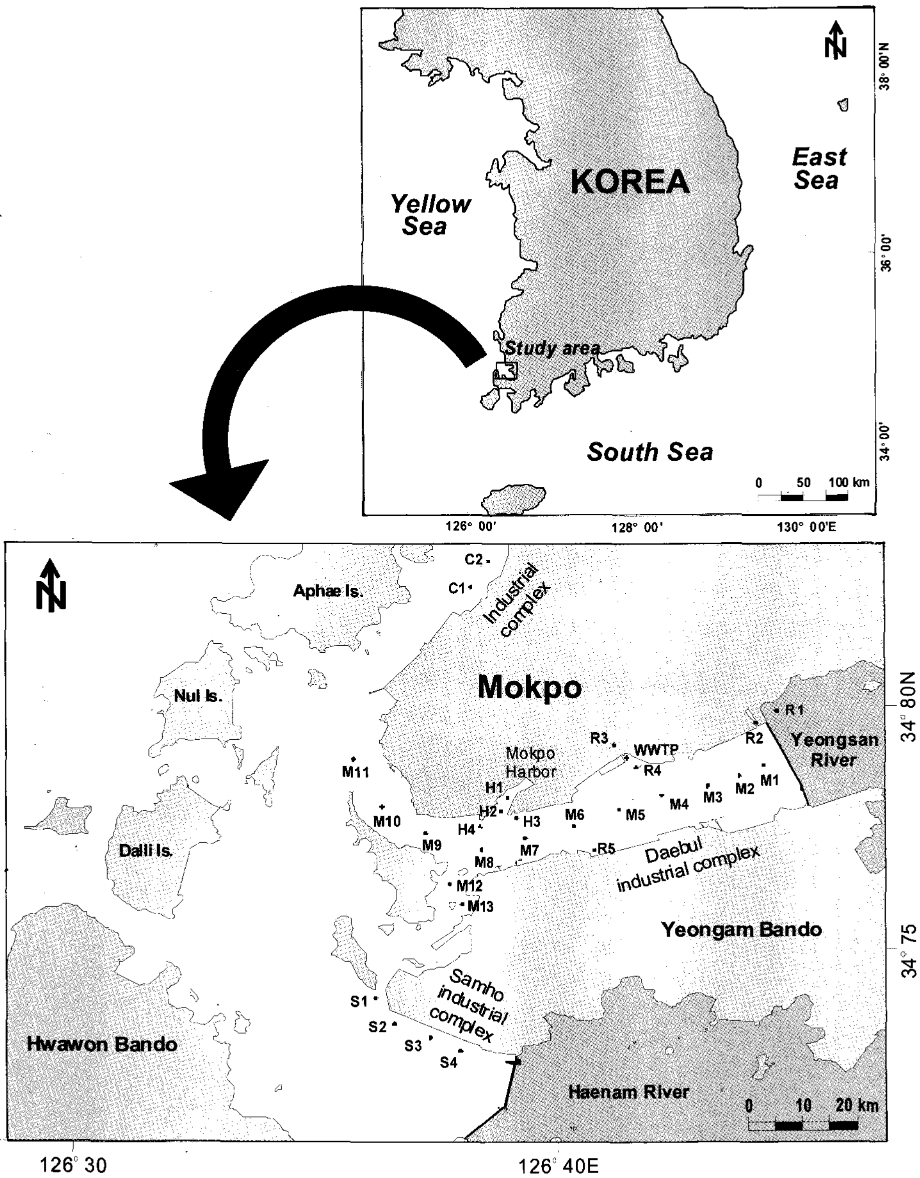

Fig. 1. Sampling locations of surface water and sediments from the Mokpo coast in Korea.

(coprostanol, coprostanone, epicholestanol, epicoprostanol, cholesterol, cholestanol, brassicasterol, campestrol, stigmasterol, stigmastanol, and β-sitostetol) according to the method of Choi et al. (2005a). Briefly, the surface water filter and approximately 5 g of freeze-dried sediment were placed in respective 50 mL Teflon centrifuge tubes with Teflon caps. A surrogate standard, 1-nonadecanol (Dr. Theodor Schuchardt & Co, Hohenbrum, Germany) was added to each, and the samples were extracted twice using dichloromethane-chloroform (Pesticide grade; Merck). The extracts were concentrated to about 1 ml under a gentle dry nitrogen flow and transferred to n-hexane (Pesticide grade; Merck). The extracts were cleaned by passage through a Florisil chromatography column. The eluents were concentrated and derivatized using BSTFA. An internal standard was added to each, and the samples were analyzed by gas chromatography-mass spectrometry with selected ion monitoring.

The total organic carbon (TOC) content in sediments was analyzed using a CHN Elemental Analyzer (Perkin-Elmer, Model 2400; Boston, MA, USA), after removal of calcium carbonate with 1 N HCl.

Quality control (QC)

The recovery of the target compounds was based on five replicate analyses of ultrapure water, filters, or sea sand (Washed sea sand; Fisher Scientific, USA) spiked the 11 fecal sterols or NPs. The recoveries of fecal sterols were 89±5% for the filters and 82±14% for the sea sand, recoveries of NPs were 81±12% for the water samples and 104±15% for sea sand. Procedural •blanks of sediment and surface water were processed in the same way as the samples. The blanks did not contain detectable amounts of the target compounds. The calculated detection limits (S/N=5) of fecal sterols were in the range of 4-14 ng/g dry weight for 5-g sediment samples and 5-18 ng/L for 4-L surface water samples. Those of NPs were about 2 ng/g dry weight for 2-g sediment samples and about 2 ng/L for 2-L surface water samples.

Results and Discussion

Fecal sterols in surface water and sediments

The concentrations of fecal sterols in surface water and sediments from the Mokpo coast are summarized in Table 1. The concentrations of total sterols (coprostanol, coprostanone, epicholestanol, epicoprostanol, cholesterol, cholestanol, brassicasterol, campestrol, stigmasterol, stigmastanol, and β-sitostetol) in surface water ranged from 860 to 84,949 ng/L, with an average of 6,306 ng/L; in sediments, total sterols ranged from 1,651 to 130,514 ng/g dry weight, with an average of 10,416 ng/g dry weight. All of the compounds were detected in Mokpo coastal waters with a 100% frequency, except for coprostanone (Copo). Cholesterol and β-sitostetol were predominant in the target sterols, accounting for 56.1% (23.7-62.7%) and 38.7% (18.4-65.4%) to the total sterols in surface water and sediments, respectively.

The concentrations of Cop in surface water ranged from 94 to 7,568 ng/L. Relatively high Cop concentrations were found at station R2, R3, R4, and WWTP (Fig. 2a). The concentrations of Cop in our study were lower or comparable to those reported for other locations, e.g., Masan Bay and adjacent rivers in Korea (<LOD-23,088 ng/L; Choi et al., 2005a), the Sein River in France (500-56,800 ng/L; Thoumelin et al., 1997), and the Tamaga River entering Tokyo Bay in Japan (400-4,000 ng/L; Chalaux et al., 1995); however, the Cop levels were higher than those reported for the Devlins Creek in Australia (32 ng/L) and those suggested as primary and secondary recreational contact limits for bacteria in Australia and New Zealand (60 and 400 ng/L, respectively; Leeming and Nichols, 1996).

The Cop concentrations in sediments varied from 43 to 38,108 ng/g dry weight. The concentrations of Cop in sediments from all stations were much higher than the background level of 10 ng/g dry weight (Hatcher and McCillivary, 1979), indicating that all stations had been affected by municipal wastewaters. Relatively high Cop concentrations were observed at station R2, R3, R4, and WWTP, whereas low concentrations were observed in outer locations, stations M10-M13, C1, C2, and S1-S4. The highest concentration, 38,108 ng/g dry weight, occurred at station R2 (Samhyang River) and was comparable to those in other areas related to the direct discharge of WWTP effluents, e.g., 35,300 ng/g dry weight in Spain (Jeng et al., 1996) or untreated sewage, e.g., 41,800 ng/g dry weight in Portugal (Mudge and Behianno, 1997). With the exception of the highest concentration, the Cop concentrations (43-7,902 ng/g dry weight) in our study were com-parable to those reported from contaminated areas of Korean coastal waters, e.g., Masan Bay (130-3,964 ng/g dry weight; Choi et al., 2005a), Ulsan Bay (141-8,257 ng/g dry weight; Choi et al., 2005b), and Kyeonggi Bay (3-3,800 ng/g dry weight; Li et al., 2007); and those from other countries, e.g., Hong Kong (380-4,800 ng/g dry weight; Chan et al., 1998), the United States (100-7,530 ng/g dry weight; Writer et al., 1995), and Italy (40-4,410 ng/g dry weight; Fattore et al., 1996). These results suggest

Table 1. Summary of 11 fecal sterols and nonylphenolic compounds in surface water (ng/L) and sediments (ng/g dry weight) from the Mokpo coast in Korea

		Coprostanol	Coprostanone	Cholesterol	Cholestanol	β-sitosterol	Total sterols[a]	NP[b]	NP_{1-2} EOs[c]	NPs[d]	TOC (%)[e]
Sediments	Min	43	< LOD	99	289	200	1,651	4	< LOD	4	0.4
	Max	38,108	25,611	2,700	13,697	10,253	130,514	582	1,797	2,119	2.4
	Mean	2,026	1,517	636	1,858	1,044	10,416	51	146	198	0.8
	SD	7,174	4,712	617	2,632	1,856	23,885	111	428	520	0.4
Surface water	Min	94	< LOD	10	302	204	860	10	94	123	-
	Max	7,568	3,496	979	15,879	4,213	84,949	774	3,956	4,729	-
	Mean	665	197	129	1,420	635	6,306	95	464	559	-
	SD	1,714	758	275	3,460	946	18,468	192	878	1,066	-

[a]the sum of 11 fecal sterols; [b]nonylphenol; [c]nonylphenol mono- & diethoxylates; [d]the sum of NP and NP_{1-2}Eos; [e]total organic carbon.

that municipal wastewaters, particularly domestic wastewaters, have had a considerable impact on Mokpo coastal wasters.

The spatial distributions of Cop in surface water and sediments were statistically similar to those of coprostanone (Copo; r=0.922-0.950, p<0.001), which is an intermediate product of the microbial hydrogenation of cholesterol to Cop within the intestine of humans and in anoxic sediment environments. Thus, Copo could be a good indicator compound, along with Cop.

Nonylphenolic compounds in surface water and sediments

All NPs used in Korea are imported, at 11216 tons in 2004, and are used as surfactant s (60%) such as industrial and domestic cleaners, followed by additives for paints and epoxy resins (12%), copper clad laminates (9%), ink binder (5%), agricultural pesticides (2%), and other uses (ME, 2006a). In Korea, NPs have been banned from kitchen cleaners since 2002 and are scheduled to be banned from all domestic applications in 2007. Thus, the contribution of industrial applications of NPs will increase relative to domestic applications.

In surface water, the concent-rations of NP and $NP_{1-2}EOs$ ranged from 10 to 774 ng/L and from 94 to 3,956 ng/L, respectively. In sediments, NP and NP_{1-2} EOs ranged from 4 to 582 ng/g dry weight and from <LOD to 1,797 ng/g dry weight, respectively (Table 1). The $NP_{1-2}EOs$ level was three times higher than the NP level in sediments and seven times higher than the NP level in surface water, implying that incompletely treated wastewater had been discharged to the study area. This result was consistent with reports from other locations (Ferguson et al., 2001; Isobe et al., 2001). The higher ratios of $NP_{1-2}EOs$ to NP observed in surface water and sediments may be attributable to differences in physicochemical properties such as the log Kow (octanol-water coefficient; 4.48 for NP, 4.17-4.21 for $NP_{1-2}EOs$) or to the in situ transformation of NP_{1-2} EOs to NP (Ying et al., 2002).

The highest concentrations of NP and $NP_{1-2}EOs$ were detected at station WWTP, and relatively high concentrations were measured at stations R2 and R3 (Fig. 2b). The high correlation coefficients between NP and $NP_{1-2}EOs$ (r=0.978, p<0.001 for surface water and r=0.792, p<0.001 for sediments) indicate that these com-pounds may have similar transport, mixing and de-position along the Mokpo coast. The concentrations of NP in surface water and sediments from the Mokpo coast were much lower than those from other locations in Korea known to be highly contaminated with industrial wastewaters; Shihwa Lake (160-26,000 ng/L and 16-5,054 ng/g dry weight; Li et al., 2004), Masan Bay (150-1,680 ng/L, Choi et al., 2005a), Ulsan Bay (<LOD-1,040 ng/g dry weight; Khim et al., 2001), and Kyeonggi Bay (4.7-1,100 ng/g dry weight; Li et al., 2007). These results suggest that the Mokpo coast may be less affected by industrial wastewaters than by domestic wastewaters. The concentrations observed in this study were comparable to those measured in the United States (<LOD-640 ng/L, Nayor et al., 1992) and Canada (<LOD-920 ng/L; Bennie et al., 1997), and were lower than those in Japan (50-1,080 ng/L and 30-13,000 ng/g dry weight; Isobe et al., 2001) and the Yangtze River in China (100-7,300 ng/L; Ding et al., 1999).

However, much lower concentrations of NPs can reportedly disrupt the endocrine system of aquatic organisms (Jobling et al., 1996). Some de-veloped countries have proposed guidelines for NPs in water and sediments to protect aquatic life; the predicted no-effect concentration (PNEC) of NP is 0.608 μg/L in Japan (Furaichi et al., 2004), the maximum permissible concentrations (MPCs) of NP are 0.33 μg/L and 0.105 μg/g dry weight in the Nether-lands (Jonkers et al., 2005), and the water quality guidelines for NPs are 1.0 μg NP-TEQ/L and 1.0 μg NP-TEQ/g dry weight in Canada (Environment Canada, 2002). Based on these guidelines, the NP levels at the stations R2, R3 and WWTP on the Mokpo coast may be associated with effects on sensitive species.

Correlations between nonylphenolic compounds and coprostanol

As shown in Fig. 3a, significant correlations were found between the concentrations of NPs and Cop in surface water (r=0.809, p<0.001) and sediments (r=0.982, p<0.001). The correlations suggest that these compounds may have similar discharge points, transport, mixing and deposition in the Mokpo coastal environment. Li et al. (2007) also reported similar results in Kyeonggi Bay, Korea. The correlation coefficients of NPs and Cop with TOC in sediments were also significant (Fig. 3b), indicating that municipal wastewaters are likely to contribute considerably to the distribution of organic carbons in sediments from the Mokpo coast, although the contributions of NPs and Cop to TOC in sediments were low.

Evaluation of municipal wastewater contamination

The interpretations based on NP and fecal sterol

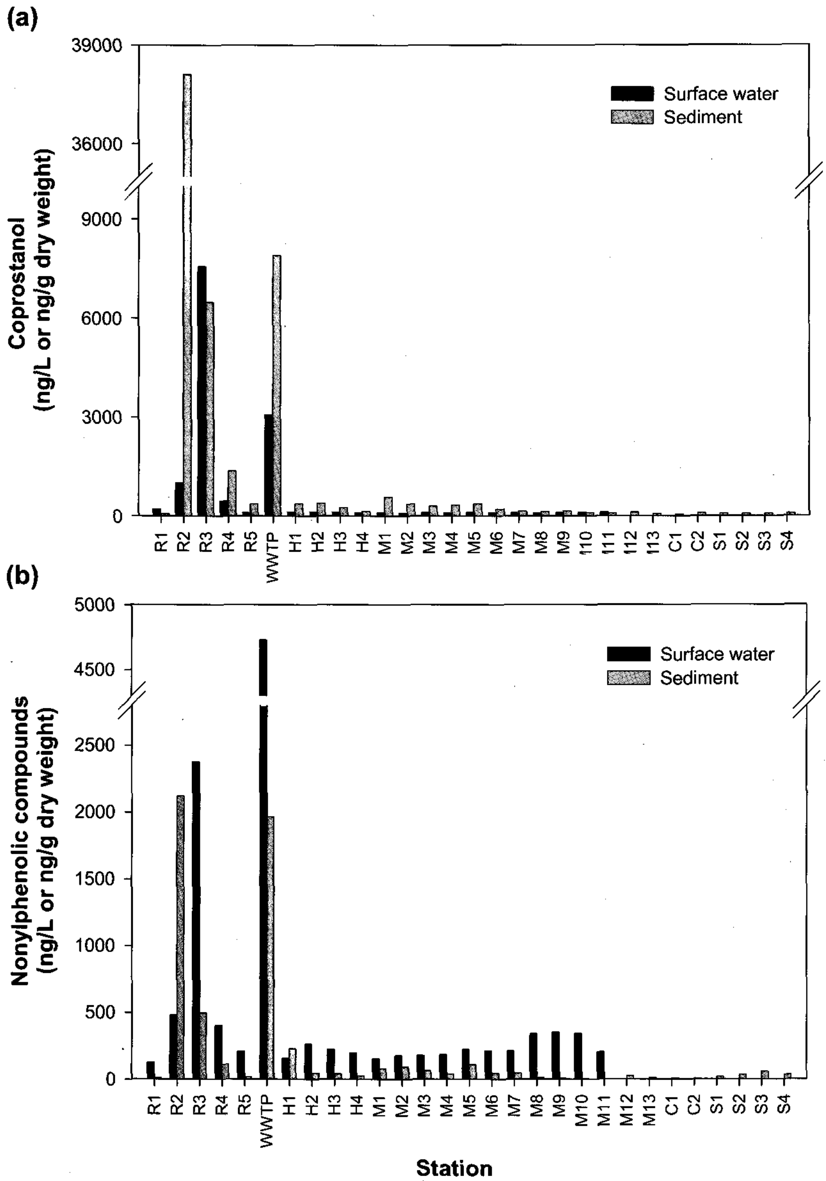

Fig. 2. Distribution of (a) coprostanol and (b) nonylphenolic compounds levels in surface water and sediments from the Mokpo coast in Korea.

concentrations alone may be interfered by other physicochemical factors, such as sediment organic carbon content and particle size distribution. Previous studies have reported various sterol ratios such as Cop/total sterols, Cop/(Cop+cholestanol) and Cop/cholesterol, for indicating domestic wastewater contamination of sediments (Hatcher and McGillivary, 1979; Writer et al., 1995; Chan et al., 1998; Reeves and Patton, 2001).

The ratio of Cop to total sterols varied from 0.02 to 0.31 and the ratio at station R3 was higher than 0.3, suggesting municipal wastewater contamination (Hatcher and McGillivary, 1979; Fattore et al., 1996). The Cop/(Cop+cholestanol) ratio ranged from 0.18 to 0.95, with ratios higher than 0.7 detected at stations R2, R3, and WWTP, indicating municipal wastewater input (Reeves and Patton, 2001). The Cop/cholesterol ratio ranged from 0.04 to 2.78 and values greater than 1.0 (Gonzalez-Orefa and Saiz-Salinas, 1998) were found at stations R2, R3, and WWTP. These three ratios shared similar distributions (Fig. 4), indicating that, of the surveyed sites, stations R2, R3, and WWTP were the most heavily contaminated by municipal wastewaters.

Although the concentrations of individual compounds and concentrations ratios of two or more

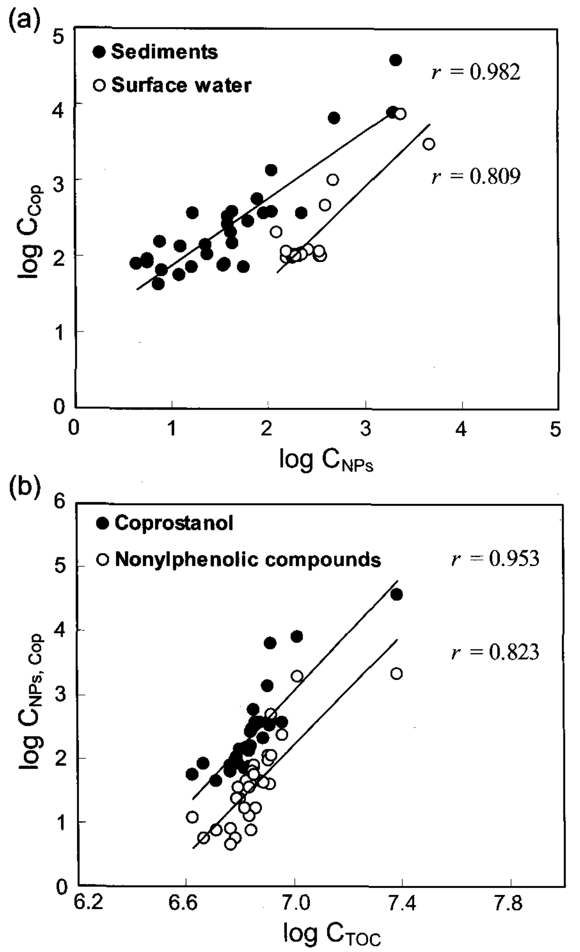

Fig. 3. (a) Correlation between coprostanol and nonylphenolic compounds in surface water and sediments, and (b) correlation of coprostanol and nonylphenolic compounds with total organic carbon in sediments from the Mokpo coast in Korea.

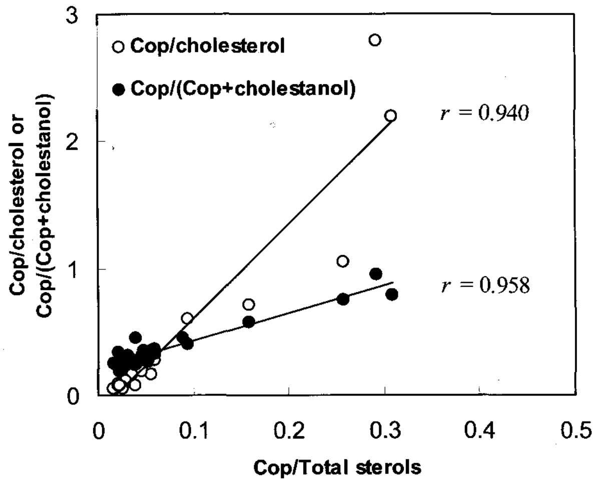

Fig. 4. Correlation among sterols ratios, coprostanol/total sterols, coprostanol/(coprostanol+cholestanol), and coprostanol/cholesterol in sediments from the Mokpo coast in Korea.

sterols can be used to evaluate the extent of municipal waste-water contamination in sediments, multivariate statistical analysis of multiple compounds provides information regarding the origin of organic matter in a single analysis. Several multivariate statistical techniques were investigated; however, correspond-dence analysis (CA) was able to provide the clearest description of the organic matter partitioning among sites (Moon et al., 2004; Choi et al., 2005a, b).

The CA results are shown in Fig. 5. The two axes accounted for 87.4% of the variance (Axis I, 70.5%; Axis II, 16.9%). Plots of the loadings on the first five components (Cop, Copo, epicoprostanol, epicholesterol and NPs) indicated the geochemical source, with municipal wastewater markers loading strongly and positively on Axis I; brassicasterol and campesterol, mainly of phytoplankton origin, were negatively loaded. The classification depended on the size of the molecule according to groups of C_{27}, C_{28}, and C_{29} sterols. Wastewater markers with C_{27} sterols and NPs were clearly separated from phytoplankton with C_{28} sterols, but cholesterol and cholestanol in C_{27} sterols were not apparently separated from the others because of their presence in many marine organisms, from algae to fish, as well as in domestic wastewater discharge (Mudge et al., 1997; Reeves and Patton, 2001). Therefore, Axis I appeared to present a wastewater (positive) and a marine (negative) axis. Sample locations R2 and R3 were the most strongly affected by municipal organic wastewaters, followed by locations WWTP and H1. These results suggest that, despite the high rate of sewage treatment in Mokpo City (97.3%; ME, 2006b), municipal organic wastewaters are still discharged through rivers to the Mokpo coast probably as a result of sewage system mismanagement, and through WWTP effluents, because of incomplete removal processes.

Acknowledgments

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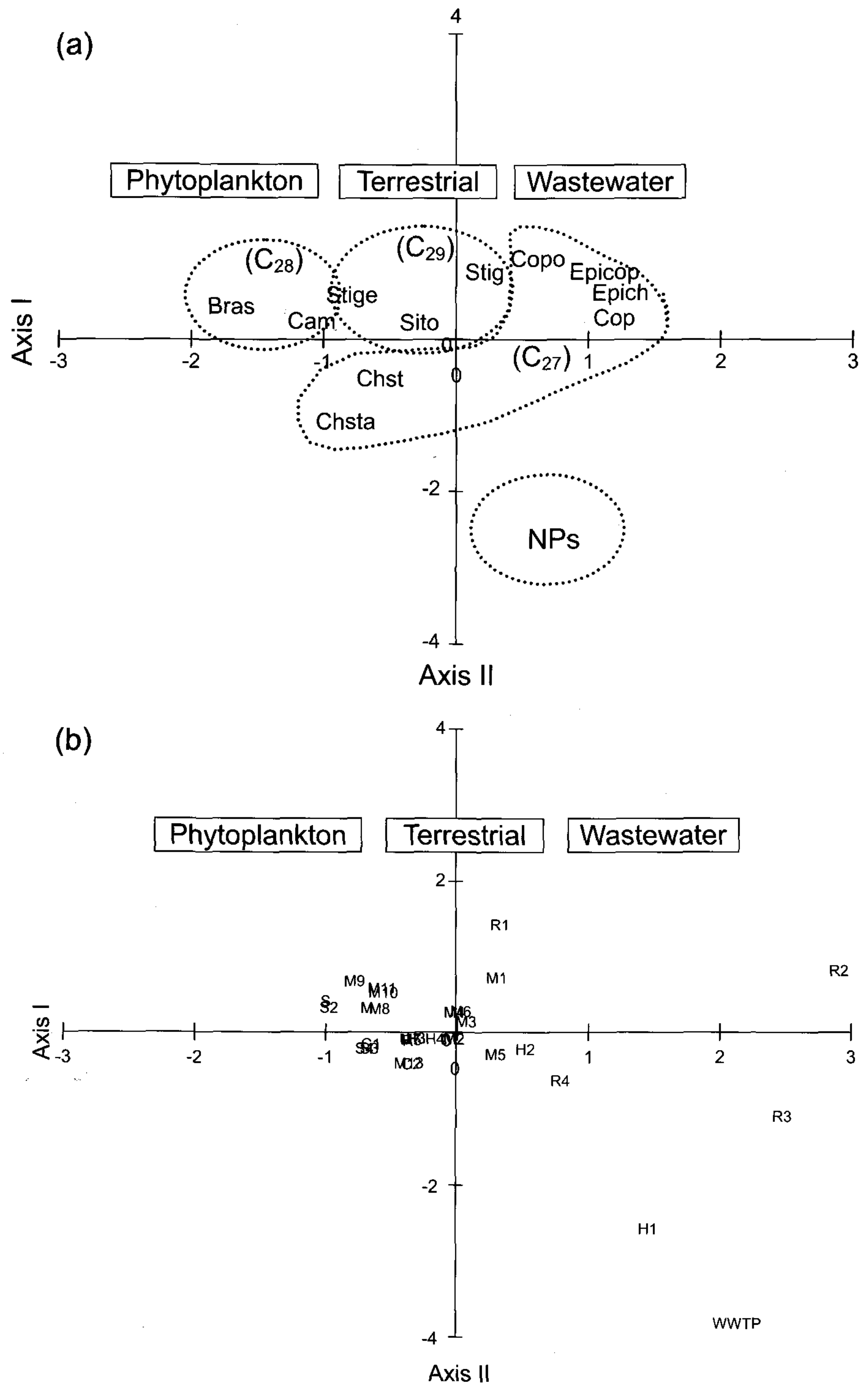

Fig. 5. Correspondence analysis plots for (a) loadings and (b) scores according to composition of fecal sterols and nonylphenolic compounds in sediments from the Mokpo coast in Korea (Cop: coprostanol, Copo: coprostanone, Epichst: epicholestanol, Epicop: epicoprostanol, Chst: cholesterol, Chsta: cholestanol, Bras: brassicasterol, Cam: campestrol, Stige: stigmasterol, Stig: stigmastanol, and Sito: β-sitostetol).

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