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Current nonylphenol pollution and the past 30 years record in an artificial Lake Shihwa, Korea

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Nonylphenol (NP) is a degradation product of nonylphenol polyethoxylates (NPEOs) which are used as detergents, wetting agents, dispersing agents and emulsifiers in various commercial, industrial and domestic applications (Renner, 1997; Ferguson et al., 2003). NP has more severe aquatic toxicity effects than NPEOs and also more persistent and lipophilic chemical properties (Servos, 1999). NP has been recognized as an endocrine-disrupting chemical that causes estrogenic effects in fish and other aquatic organisms (Routledge and Sumpter, 1997). High NP concentrations found in surface sediment samples from an artificial Lake Shihwa originate from sewage and wastewater from industrial complexes and cities that flow into the lake via adjacent creeks (Li et al., 2004a,b). High estrogenic activity was observed in lake sediments, which could have adverse effects on benthic organisms (Khim et al., 1999).

Polychaetes are well known bioindicators of marine sediment quality (Pocklington and Wells, 1992; Fattorini et al., 2005). Some polychaetes, such as *Polydora cornuta* and *Neanthes succinea*, are opportunistic species that are tolerant of hypoxic sediment conditions (Giangrande et al., 2005). These species play an important role in the marine food chain, serving as food for organisms at higher trophic levels and recycling organic matter. Most of these polychaete species are deposit feeders that ingest particle-adsorbed contaminants in the sediment (Elias et al., 2001; Jørgensen et al., 2008). Organic pollutants are readily adsorbed by sediment particles and therefore accumulated in marine sediments, as well as in polychaetes, due to high hydrophobicities and correspondingly low solubilities in water (Ferguson and Chandler, 1998; Jørgensen et al., 2008).

Historical organic pollution trends in marine environments have been investigated using dated sediment cores (Alexander et al., 1999; Yamashita et al., 2000; Wei et al., 2008; Moon et al., 2009; Hong and Shin, 2009). The vertical profiles of NPs detected in dated sediment cores are often reviewed to detect historical pollution events resulting from wastewater treatment plant discharge (Shang et al., 1999; Isobe et al., 2001; Ferguson et al., 2003; Peng et al., 2007). In this study, we investigated current contamination levels of nonylphenolic chemicals in sediment, water and polychaete samples collected from the inner and outer regions of Lake Shihwa and its surrounding creeks. Bioaccumulation factors (BAF) were calculated for nonylphenol in polychaete samples. In addition, a sediment core sample was collected to assess historical records of nonylphenol pollution in Lake Shihwa and a sediment chronology was estimated using radioactive isotopes (^{210}Pb and ^{137}Cs). Organic carbon (OC) contents and carbon stable isotope ratios (δ^{13}C) were determined to clarify the influx patterns and origins of organic matter in Lake Shihwa.

Sampling sites are shown in Fig. 1. Four liters of water per sample were collected in six creeks feeding into Lake Shihwa during May 2008. The samples were filtered in the laboratory within 2 days after collection through GF/F filters (0.7 μm, Whatman, Maidstone, England). Surface sediment samples (upper 3 cm) were collected using a Van Veen grab from seven sites (st.1–st.7) in the inner region of the lake and seven sites (Y1–Y7) along the outer regions of Lake Shihwa in May and August 2008, respectively. A sediment core sample was collected from st.1 and transferred into a 100 mL glass bottle with a 2 cm slice intervals (total 66 cm). Polychaete samples were obtained using a grab sampler and then rinsed through a sieve with 1 mm mesh and sorted in the laboratory. The sediment and polychaete samples were immediately frozen at −20 °C, freeze-dried, homogenized by grinding, and stored in a vacuum desiccator before extraction.

The extraction and purification procedures used for nonylphenolic analysis of water and sediment samples are described else-

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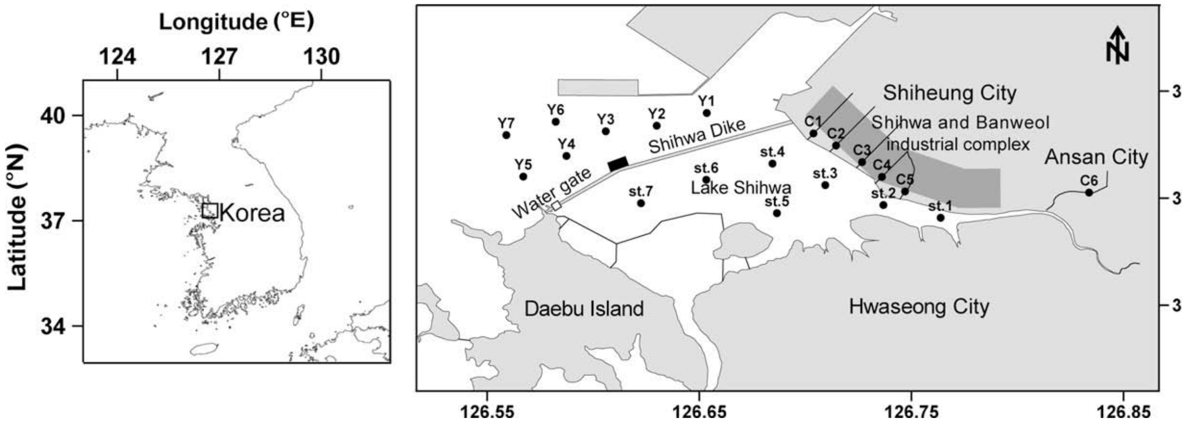

Fig. 1. Sampling locations in the inner (st.1–st.7) and outer (Y1–Y7) regions of Lake Shihwa and surrounding creeks (C1: Okku; C2: Kunja; C3: Jungwang; C4: Shingil; C5: Shihueng; and C6: Hwajung and Ansan; black square: tidal power plant site).

where (Li et al., 2001, 2003; European Standard, 2007). Briefly, 1 L of each filtered water sample was transferred into a 2 L separate funnel and then 100 μL of 1 mg L^{-1} surrogate standards (bisphenol-A-*d16*, Isotec, Miamisburg, OH, USA and *n*-NP1EO ring-$^{13}C_6$, CIL, Andover, MA, USA) were added for quality control. Sixty milliliters of methylene chloride (J.T. Baker, Phillipsburg, NJ, USA) were added and the funnels were shaken vigorously for 10 min by a mechanical shaker. Five grams of freeze-dried sediment sample were placed in a 50 mL Teflon tube, spiked with surrogate standards, and 5 mL of dilute water (Milli-Q system). Five milliliters of acetone (J.T. Baker) and 5 mL of hexane (J.T. Baker) were also added and then the mixture was placed on a mechanical shaker. The organic phase was transferred to a 20 mL vial and concentrated under a gentle stream of nitrogen gas. All extraction procedures were repeated twice. The residual water was removed from the extracting solvent through a funnel filled with fine sodium sulfate powder (Fluka, Buchs, Switzerland) and concentrated to below 1 mL. It was then substituted to hexane under a gentle stream of nitrogen gas. The extracts were derivatized with *N*,*O*-bis-(trimetylsilyl) trifluoroacetamide (BSTFA with 1% TMCS, Sigma–Aldrich, Saint Louis, MO, USA) using a silylation treatment kit and cleaned up using 1 g of activated Florisil (60–100 mesh, Sigma–Aldrich) with 7 mL of hexane (Li et al., 2001).

The analysis of nonylphenol in biotic samples was performed following a method described elsewhere (Wang et al., 2007). Approximately 0.1 g of a homogenized dry polychaete sample was weighed and surrogate standards were added. Then the nonylphenol was extracted using 200 mL of methylene chloride for 16 h on a Soxhlet extractor. The extract was concentrated to 1 mL on a rotary evaporator and exchanged to hexane following the clean-up procedure. The extract from the biotic samples was first cleaned by passing it through a Florisil (2 g, deactivated with 5% H_2O) column in order to remove low and high polar lipids. The first fraction (elution with 18 mL of hexane) was discarded and the second fraction (elution with 10 mL of methylene chloride) was collected. The second fraction was concentrated under a gentle stream of nitrogen gas and then exchanged to 200 μL of hexane. The procedures for derivatization and the clean-up steps for removal of middle polar lipids were the same as those used for the treatment of the water and sediment samples (Li et al., 2001; Wang et al., 2007). GC internal standard (phenanthrene-*d10*, Isotec) was added and the sample was concentrated to 1 mL for GC/MS analysis.

GC/MS analyses were performed for nonylphenolic chemicals using a gas chromatograph (Shimadzu GC-2010, Tokyo, Japan) coupled with a mass spectrometer (Shimadzu GCMS-QP2010 plus). A capillary column DB-5MS (30 m long × 0.25 mm i.d.; film thickness: 0.25 μm, J&W Scientific, Folsom, CA, USA) was utilized for the separation. The mass spectrometer was operated in electron impact ionization (EI) mode at 70 eV with the selected ion monitoring (SIM) method. The method detection limits (MDLs) for NP, NP1EO and NP2EO were 2.11, 12.2 and 22.3 ng g^{-1}, respectively. The surrogate recoveries of bisphenol-A-*d16* and *n*-NP1EO ring-$^{13}C_6$ were 81.4–97.1% and 75.4–118.1%, respectively.

Freeze-dried bulk sediment samples were treated for 24 h with 1 N HCl to remove calcium carbonate. Following neutralization with distilled water, the sediments were again dried (Cifuentes et al., 1988). Four to eight milligrams of sediment were packed in tin foil capsules for instrumental analysis. The organic carbon contents and $\delta^{13}C$ values of the sediment samples were measured by an elemental analyzer-isotope ratio mass spectrometer (Euro EA-Isoprime IRMS, GV instruments, UK). The $\delta^{13}C$ values were expressed in per-mil (‰) notation relative to the Pee Dee Belemnite (PDB) standard and their analytical precision was ±0.15‰.

The sedimentation rate and sediment chronology represented by the sediment core (st.1) were estimated by ^{210}Pb and ^{137}Cs dating techniques at the Korea Basic Science Institute. The radioisotope activities of ^{210}Pb, ^{226}Ra and ^{137}Cs were measured for one sediment core at 2 cm intervals using a well-type HPGe gamma detector (GCW3523, Canberra Inc., USA) calibrated with International Atomic Energy Agency (IAEA) certified reference materials (RGU-1, RGTh-1 and RGK-1).

Water discharged from local municipal and industrial complexes flows via six surrounding creeks into Lake Shihwa, which are the main sources of NP pollution in the lake (Li et al., 2004a,b). The concentrations of NP, NP1EO and NP2EO in the water samples from the surrounding creeks are shown in Fig. 2. The highest concentrations of NP were found in Okku Creek (C1, 16 598 ng L^{-1}) and the lowest in Shingil Creek (C4, 311.9 ng L^{-1}). In April 2000 the NP concentrations in the creeks ranged from 100 to 41 300 ng L^{-1} (Li et al., 2004a) and in June 2002 the NP concentrations ranged from 118.1 to 4 324.1 ng L^{-1} (Li et al., 2004b). Although there have been many efforts to improve the water quality in Lake Shihwa, high concentrations of nonylphenolic chemicals are still present in the surrounding creeks.

The composition patterns of these nonylphenolic chemicals can be divided into three categories including (i) NP2EO ≈ NP1EO < NP, (ii) NP2EO > NP1EO > NP and (iii) NP2EO > NP > NP1EO. The composition of case (i) (for example: C1) indicates that the sources of the nonylphenolic chemicals are more distant than the sources of pollution in case (ii) (for example: C2). Composition patterns in regions represented by case (ii) reflect the introduction of fewer degradation products of NPEOs (Li et al., 2008). NPEOs are degraded to NP2EO via sequential elimination of the ethoxy chain, which then accumulates in the environment and/or is further degraded to

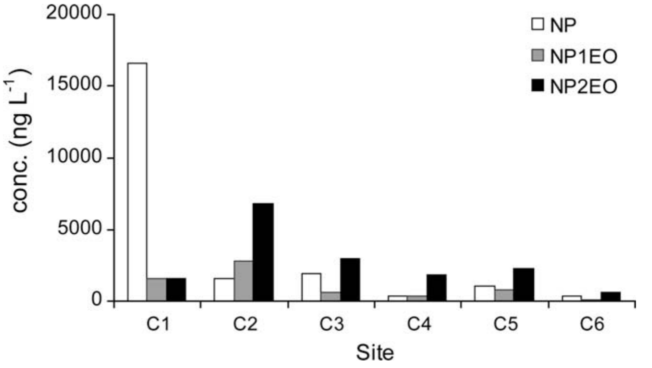

Fig. 2. Concentrations of nonylphenolic chemicals in water samples from six creeks.

NP1EO and NP by microbial activity. Long-chain NPEOs (ethoxy chain >2) are more rapidly degraded than short-chain NPEOs (ethoxy chain $\leqslant 2$) in aquatic environments (John and White, 1998; Ferguson et al., 2003; Liu et al., 2006; Lu et al., 2009). In case (iii) (for example: C3–C6), the composition pattern may be explained by disparities in degradation and supply rates between long-chain NPEOs that degrade to NP2EO, and NP2EO that degrades to NP1EO and/or NP.

The analytical results for nonylphenolic chemicals in surface sediment collected from inner and outer regions of Lake Shihwa during May and August 2008 are reported in Table 1. High concentrations of NP were found in the inner regions, approximately three to 10-fold higher levels than in outer regions. There are significant seasonal variations in concentrations of nonylphenolics, with higher NP concentrations and lower NP1EO and NP2EO concentrations in August than in May. These results are likely explained by increased levels of NPEO degradation under the stronger microbial activity that is associated with increasing water temperature (Li et al., 2007).

Current concentrations of NP in surface sediments from the inner and outer regions of Lake Shihwa were compared with data from the past 10 years as shown in Fig. 3. The current concentrations of NP detected in Lake Shihwa sediments are relatively low compared to the concentrations noted in previous reports. These results may be attributed to the many efforts directed toward environmental improvement that have been enacted during the intervening years, such as a ban on household uses of NPEOs that has been in place since 2002 and water exchange that was initiated through the installation and operation of a water gate at the end of the Lake Shihwa dike in 1999. However, NP concentrations in the surface sediments from the inner regions are still much higher than those from the outer regions as shown in Table 1. The world's largest tidal power plant is currently under construction in the middle of the dike at Lake Shihwa, and is scheduled for completion in 2010. When the tidal plant is being operated, 60 billion tones of seawater will be circulated annually between the inner and outer regions of Lake Shihwa. Therefore, the water quality of Lake Shihwa will be improved, but the benthic biota in the outer mudflat regions could be affected by the outflow of polluted sediments from the inner regions of Lake Shihwa due to critical changes in the physical environment.

Polychaetes are used as pollution indicator species in hypoxic and polluted sediments (Giangrande et al., 2005). We found polychaetes (*N. succinea*) in sediments from st.1, st.2, st.3 and st.5 in May, but not in August (Table 1). The disappearance of polychaetes from Lake Shihwa by late summer may be a result of insufficient sediment oxygen due to water stratification which occurs during the summer season (Han and Park, 1999; Sagasti et al., 2001) and it also indicates that the effects of water exchange via the water gate are restricted to the region near the water gate in the Lake Shihwa dike.

The NP concentrations found in the polychaetes we collected were higher (1156–2757 ng g^{-1} dry weight (dw)) than those previously reported for other organisms in other polluted regions. NP concentrations in mussels collected from Masan Bay in Korea ranged from 50.5 to 289.2 ng g^{-1} dw (Wang et al., 2007), and NP concentrations in snails collected along the coast of Taiwan ranged from 130 to 1560 ng g^{-1} dw (Cheng et al., 2006). NP concentrations in fish from the Kalamazoo River in Michigan, USA ranged from <D.L. to 29.1 ng g^{-1} wet weight (Kannan et al., 2003). David et al. (2009) recently reviewed the alkylphenol concentrations in various marine organisms and found that they were higher in bivalves (mussels and oysters) and gastropods (snails) than in fishes. These differences in bioaccumulation levels may be related to the lipid contents, biological cycles, trophic levels and feeding behaviors of these organisms. The bioaccumulated NP concentrations in the polychaetes we collected from Lake Shihwa were strongly corre-

Table 1
Concentrations (ng g^{-1} dw) of nonylphenolic chemicals during May and August in surface sediments and polychaetes from the inner and outer regions of Lake Shihwa.

Site	May				August			
	Sediment			Polychaetes	Sediment			Polychaetes
	NP	NP1EO	NP2EO	NP	NP	NP1EO	NP2EO	NP
st.1	48.1	29.0	75.1	1894.5	66.2	9.4	<D.L.[a]	–[b]
st.2	55.5	49.0	164.8	1857.7	166.2	27.5	<D.L.	–
st.3	67.2	29.2	96.2	2757.1	66.2	12.4	<D.L.	–
st.4	135.9	70.2	145.9	–	1028.1	8.3	9.9	–
st.5	26.0	5.5	2.1	1155.6	13.1	<D.L.	<D.L.	–
st.6	72.3	15.5	3.8	–	59.0	5.3	<D.L.	–
st.7	15.2	7.0	1.3	–	<D.L.	<D.L.	<D.L.	–
Mean ± SD	60.0 ± 39	29.3 ± 24	69.9 ± 70	1916 ± 656	199.8 ± 393	9.0 ± 9	1.4 ± 4	
Y1	26.5	23.7	24.0	–	36.9	<D.L.	<D.L.	–
Y2	39.5	7.3	0.8	–	18.4	<D.L.	<D.L.	–
Y3	25.5	19.6	51.8	–	12.4	<D.L.	<D.L.	–
Y4	17.1	7.3	13.5	–	6.5	<D.L.	<D.L.	–
Y5	12.7	9.8	8.7	–	6.5	<D.L.	<D.L.	–
Y6	18.6	5.6	<D.L.	–	16.4	<D.L.	<D.L.	–
Y7	10.8	3.6	<D.L.	–	15.9	<D.L.	<D.L.	–
Mean ± SD	21.5 ± 10	11.0 ± 8	14.1 ± 19		16.1 ± 10			

[a] Below detection limits.
[b] Not found.

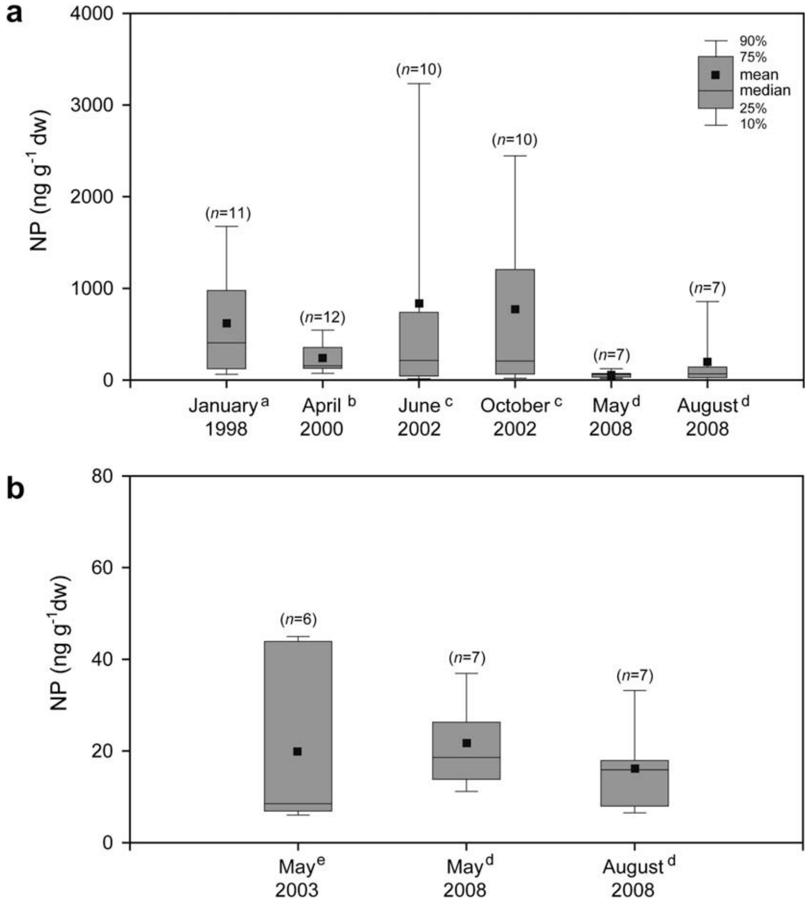

Fig. 3. Box plot (mean, median and the percentile values (10%, 25%, 75% and 90%)) for the quantification levels of NP from surface sediments in the (a) inner and (b) outer regions of Lake Shihwa ([a]Khim et al., 1999; [b]Li et al., 2004a; [c]Li et al., 2004b; [d]this study; [e]Li et al., 2007).

lated ($r^2 > 0.9$) with NP concentrations in surface sediments from the same sites. This relationship may be explained by polychaete feeding behavior, because polychaetes, as non-selective deposit feeders, directly ingest sedimentary organic matter (Jørgensen et al., 2008).

The average BAF values for NP in polychaetes were estimated to be 40 ± 5 using the NP concentrations in the biota and sediment obtained in this study. Polychaetes are important food sources for fish and other predators, and therefore the bioaccumulated NP in polychaetes may transfer to higher trophic levels in the marine ecosystem (Pruell et al., 2000; Magni et al., 2008). The establishment of a fishery in the inner regions of Lake Shihwa has been prohibited since 1998 they are highly polluted, but many species of fishes move in and out of the lake through the water gate. Therefore, it is still possible that the bioaccumulation of pollutants in Lake Shihwa poses risks to the marine ecosystem and eventually to human health.

The average sedimentation rate was 2.41 cm y^{-1} as estimated by a constant excess of ^{210}Pb detected in the core samples from st.1 in Lake Shihwa (Fig. 4). ^{137}Cs had penetrated to the full sediment core depth, but the activity levels of ^{137}Cs were too low for quantitative analysis. Vertical profiles of the nonylphenolics, organic carbon contents and δ^{13}C values in the core samples are shown in Fig. 4. According to the ^{210}Pb dating, NP concentrations have gradually increased since 1980, peaked in the late 1980s and then decreased sharply until 1990. The trend in the 1980s may be related to increases in NP use caused by industrialization and urbanization associated with the development of the Banweol industrial complex from 1977 to 1987. The decrease after 1990 may be due to the construction and operation of the wastewater treatment plant (WWTP) at Banweol in 1990. NP concentrations also peaked in the early 1990s, indicating that untreated wastewater was directly discharged into Lake Shihwa because the adjacent Ansan and Shiheung cities lacked adequate WWTPs.

High organic carbon contents and low δ^{13}C values could indicate large influxes of land-derived organic matter during the corresponding periods (Won et al., 2007). The Shihwa dike was completed in 1994, and after its construction Lake Shihwa was completely isolated from Gyeonggi Bay and the Yellow Sea. The increased sediment NP concentrations detected from the mid to late 1990s may reflect the isolation of Lake Shihwa until 1999. Lake Shihwa was designated as a special management coastal zone in 2000 by the Korean government. NP concentrations declined after 2000 due to environmental improvement efforts such as construction of a water gate and tighter water quality control in the surrounding creeks. Despite these efforts, high concentrations of NP chemicals and the highest organic carbon contents and lowest values of δ^{13}C were detected in the mid-2000s. These trends may reflect the insufficiency of the WWTP in proportion to the expanding local human population coinciding with the construction of massive apartment complexes near Lake Shihwa after 2004. However, the amounts of nonylphenolics deposited recently have rapidly declined, which may reflect reduced NP inflows caused by expansion of the WWTP facilities in the surrounding areas and a ban on the domestic uses of NP chemicals.

High organic carbon contents and low δ^{13}C values in the sediments corresponded to high NP concentrations at the same sediment core depths. A strong correlation between low δ^{13}C values

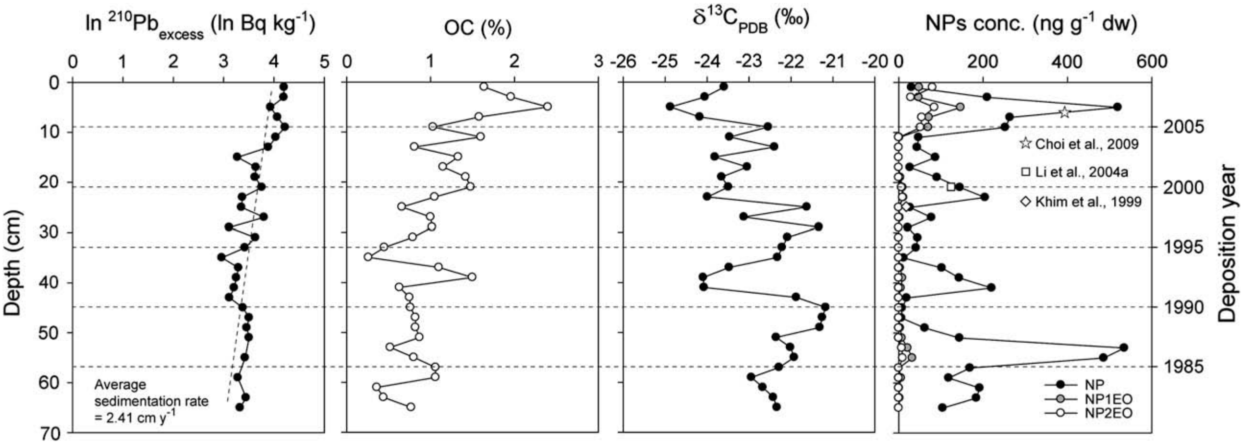

Fig. 4. Vertical profiles of ^{210}Pb$_{excess}$ activity, OC, δ^{13}C and nonylphenolic chemicals from sediment cores in Lake Shihwa (dotted line: deposition year by ^{210}Pb dating).

and high NP concentrations and organic carbon contents in the lake sediments suggests that land-derived organic matter can carry NP to Lake Shihwa.

The data obtained from the st.1 core sediment in this study is well matched to previously reported data which used surface sediments collected within a 5 km radius zone from st.1 in January 1998 (Khim et al., 1999), in April 2000 (Li et al., 2004a) and in 2006 (Choi et al., 2009) as shown in Fig. 4. This correlation between the NP concentrations during the study period and previously reported data suggests that the sediment chronology data is reasonable and also that NP can be considerably persistent under strong hypoxic environments such as that found in the Lake Shihwa sediments. The relative contribution of NP among nonylphenolics increased with increasing sediment core depths, possibly due to *in situ* biotransformation by microbial activity (Ferguson et al., 2003). Historical NP pollution trends recorded in the sediment core can be interpreted with the sedimentation rates and sediment age determined by radioactive isotope techniques. In addition, organic carbon contents and carbon stable isotope ratios in the sediments can be useful tools to understand coastal marine pollution.

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