



## Nonylphenol in bivalves and sediments in the northeast coast of China

Juan Wang<sup>1</sup>, Won-joon Shim<sup>2</sup>, Un-hyuk Yim<sup>2</sup>, Narayanan Kannan<sup>2</sup>, Donghao Li<sup>1,\*</sup>

1. Key Laboratory of Nature Resources of Changbai Mountain and Functional Molecular, Ministry of Education of China, Yanbian University, Yanji 133002, China. E-mail: [wangjuan@ybu.edu.cn](mailto:wangjuan@ybu.edu.cn)

2. Oil and POPs Research Group, Korea Ocean Research and Development Institute, Geoje-shi 656-834, Korea

Received 03 December 2009; revised 28 February 2010; accepted 04 March 2010

### Abstract

To understand nonylphenol (NP) contamination in the northeast coastal environment in China, NP was determined in 19 sediment and 20 bivalve samples collected during November 2005. NP was identified in all sediment samples in the range of 8.8 to  $1.0 \times 10^3$  ng/g dw (dry weight), with an average of  $0.12 \times 10^3$  ng/g dw. Most of the bivalve samples (oysters and mussels) contained detectable amounts of NP that ranged from ND (not detected) to  $7.6 \times 10^3$  ng/g dw, with an average value of  $0.79 \times 10^3$  ng/g dw. These results indicated that NP is ubiquitous in sediments and bivalves from the northeast coast of China, and are even present in bivalves sold as seafood. Serious contamination with NP was recorded off the coast of Bohai Strait and in semi-enclosed coastal environment such as Jiaozhou Bay and Liaodong Bay. Possible environmental and human health implications were understood in this study.

**Key words:** nonylphenol; Yellow Sea; Bohai Sea; sediment; bivalve

**DOI:** 10.1016/S1001-0742(09)60313-2

### Introduction

Nonylphenol (NP) is a known endocrine disruptor and its presence in the environment has drawn considerable concern (Soto et al., 1991; Jobling and Sumpter, 1993). NP is a biodegradation product of nonylphenol polyethoxylates (NPEs) surfactants which are widely used in household detergents, pesticide formulations, and other applications (Renner, 1997). NPEs accounted for 80% of alkylphenol polyethoxylates (APEs) productions with an estimated 500,000 tons of world annual production in 1997 (Renner, 1997). In China, the amount of APEs production is about 50,000 tons annually. There is increasing concern on the environmental behavior and fate of NP due to its estrogenicity and large production globally.

NP is ubiquitous in aquatic environment (Ying et al., 2002). Since the  $\log K_{ow}$  value of NP is 4.48, it is adsorbed onto sediments easily. And its half-life is approximately 60 years in the stratified sediment (Shang et al., 1999). Reported levels ranged from 0.1 to 13,700 ng/g for NP in sediment (Bennie et al., 1997; Blackburn et al., 1999; Ferguson et al., 2001; Isobe et al., 2001; Li et al., 2004). In China, levels of NP were from 18.4 to 7,808 ng/g (Chen et al., 2006; Hou et al., 2005). NP is lipophilic, which would lead to a bioaccumulation in organisms (Ekelund et al., 1990; Ishibashi et al., 2006). Concentration of NP in crustaceans and fish from the Adriatic Sea of Italy ranged from 118 to 399 ng/g (fresh weight) and from 9.5 to 1431

ng/g (fresh weight), respectively. It was reported that NP intake was estimated to be 12.2  $\mu\text{g/day}$  per person in Italian (Ferrara et al., 2005).

According to previous studies (Chen et al., 2006; Hou et al., 2005; Hu et al., 2005a, 2005b), NP was also founded in sediments and biota from rivers and bays such as Pearl River and Bohai Bay in China. While, there is little information available on NP in the coastal environment of China, especially NP in organisms. Jin et al. (2008) have reported the presence of persistent organic pollutants such as PCBs and organochlorine pesticides in bivalves from the northeast coast of China, whereas there are few reports on the occurrence of NP. This study, therefore, focused on the contamination level and distribution pattern of NP in sediments and bivalves from the northeast coast of China.

## 1 Materials and methods

### 1.1 Chemicals

Nonylphenol standard, BSTFA, gas chromatographic internal standards (GCIS) <sup>2</sup>H<sub>10</sub>-phenanthrene, and surrogate standard (*n*-heptylphenol) were obtained from Chem. Service, USA. Dichloromethane, hexane, acetone were from Aldrich, USA. All solvents were pesticide grade. HCl was from Merck, Germany. Florisil obtained from Supelco, USA, was used for column cleanup after activation at 400°C for 12 hr. Anhydrous sodium sulfate and all glassware were heated at 400°C for 12 hr and washed with solvent before using. The copper was obtained from

\* Corresponding author. E-mail: [dhli@ybu.edu.cn](mailto:dhli@ybu.edu.cn)

[www.jesc.ac.cn](http://www.jesc.ac.cn)

Merck, Germany; it was used to remove sulfur from the environmental sample extract. A stock solution of 300 mg/L in acetone was prepared. It was diluted in acetone to prepare appropriate calibration standards, surrogate standard (0.5 mg/L) and GCIS (0.5 mg/L).

### 1.2 Sample collection

Sediment samples were collected from 19 sites using a van Veen grab sampler in November 2005 along the northeast coast of China (Fig. 1). The surface sediment samples were placed in glass bottles with Teflon-lined cap, and stored at  $-20^{\circ}\text{C}$  until analysis. Mussel (*Mytilus edulis*) or oyster (*Ostrea edulis*) samples from 20 sites were collected at the same time except for seven sites (10, 11, 19, 21, 22, 23 and 24) where organisms were purchased in local coastal markets. Bivalve samples were homogenized with homogenizer and kept at  $-20^{\circ}\text{C}$  until use.

### 1.3 Extraction and clean up

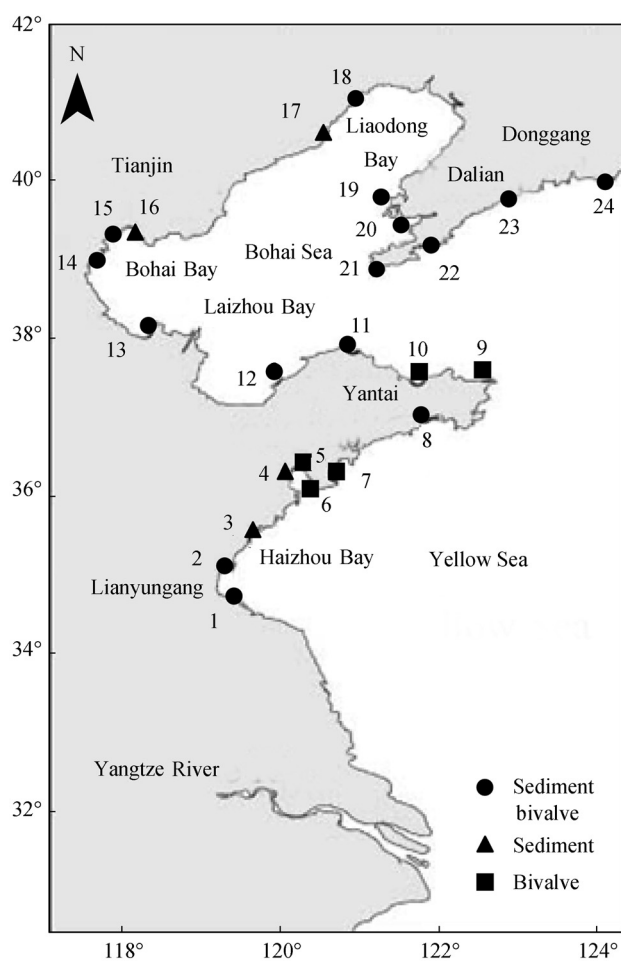
NP in sediment samples was extracted using acid-digested sediment suspension extraction (ADSSE) method (Li et al., 2003). Briefly, 3 g of wet sediment sample were transferred into a 50 mL Teflon centrifuge tubes with Teflon-lined cap. Surrogate standard (*n*-heptylphenol) was added for quality control. Five milliliter of HCl solution

was added for digestion and shaken vigorously using a vortex mixer, then, 5 mL of dichloromethane was added to the suspension. The mixture was shaken vigorously within 20 min using a vortex mixer followed by centrifugation at 3000 r/min. The dichloromethane extract was transferred into a glass tube. The extraction was repeated twice and the extracts were added to the glass tube. The extract was concentrated to 1 mL with gentle flow of dry nitrogen gas. The extract was subjected to silyl derivatization and clean-up using a silyl-derivatization kit after removal of water and sulfur with anhydrous sodium sulfate and copper, respectively (Li et al., 2001). A brief description of the kit and derivatization course is as follows: the column contains 1 g of activated Florisil powder between an anhydrous sodium sulfate layers in a 10-mL glass syringe. The sample was added on the column after silyl derivatization with BSTFA. Phenolic silyl derivatives were eluted with 7 mL of hexane. The elution was concentrated to 0.2 mL with a gentle flow of nitrogen gas followed by the addition of GCIS and then it was subjected to GC-MS analysis after the final volume was set to 0.5 mL.

NP in bivalve samples was extracted and prepared as described by Wang et al. (2007). Three grams of wet or 0.5 g of homogenized dry sample were transferred into a Soxhlet extractor and extracted with 200 mL of dichloromethane for 12 hr after the addition of surrogate standard. The dichloromethane extract was collected and volume reduced to about 1 mL at  $37^{\circ}\text{C}$  using rotary-vacuum evaporator. To remove water from the extract, 0.5–2 g of anhydrous sodium sulfate was added to every extract. Then the extract was transferred into a glass vial, and concentrated to below 200  $\mu\text{L}$  under a gentle flow of nitrogen gas at room temperature. Solvent was exchanged to approximately 200  $\mu\text{L}$  of hexane. The cleanup course contained two steps. First step was performed in column I which contained 2 g of hexane-rinsed Florisil powder (deactivated with 5%  $\text{H}_2\text{O}$ ) between 1.5 g of anhydrous sodium sulfate layers in a 10-mL glass syringe. The sample was added to the column. Eighteen milliliter of hexane was eluted first and discarded, and then phenolic analytes were eluted with 10 mL dichloromethane. The first 2 mL was discarded, and the next 8 mL was collected and concentrated with a gentle flow of nitrogen gas, and then the solvent was exchanged to acetone for derivatization with BSTFA. The second step was performed in column II which contained 1 g of hexane-rinsed Florisil powder in between 1.5 g anhydrous sodium sulfate layers in a 10-mL glass syringe. Seven milliliter of hexane was used to elute phenolic silyl derivatives. This fraction was further concentrated with a gentle flow of nitrogen gas. The final volume was set to 0.5 mL, it was subjected to GC-MS analysis.

### 1.4 NP determination

NP was analyzed using GC-MS-QP2010 (AOC-5000, Shimadzu, Japan) with a DB-5MS capillary column (30 m  $\times$  0.32 mm i.d.  $\times$  0.25  $\mu\text{m}$  film thickness). Helium gas was used as carrier gas at a flow rate of 1.0 mL/min. The temperature program was as follows: 80 to  $100^{\circ}\text{C}$  at



**Fig. 1** Locations of sediment and bivalve sampling stations along the northeast coast of China.

20°C/min, hold for 2 min, 100 to 200°C at 10°C/min, 200 to 380°C at 20°C/min, hold 20 min. The injector temperature was maintained at 280°C. Two microliter of sample was injected. Ion source temperature was 200°C. The ionization potential of the mass spectrometer was set at 70 eV. The analytes were analyzed in selected ion mode.

### 1.5 Quality assurance and quality control (QA/QC)

Procedural blank values were below the method detection limits and hence data have not been corrected with blank values. The GC internal standards were added to the extract before the instrumental analysis. To control the experimental quality, *n*-heptylphenol was used as a surrogate standard in real sample analysis. It was spiked into the samples before extraction. The procedure described above was validated for recoveries of NP (ranging from 75% to 78% for sediments and from 93% to 108% for bivalves). For duplicate samples, the relative percent differences were less than 19% for sediments and 2% for bivalves. The recoveries of surrogates were 94% ± 22% for sediments and 97% ± 20% for bivalves. Method detection limits for NP was 0.4 and 3 ng/g in sediment and bivalve samples, respectively. NP detected in sediment and bivalve samples was qualitatively and quantitatively determined based on retention time as well as relative intensities of quantification and confirmation ions (Li et al., 2001).

## 2 Results and discussion

### 2.1 NP in sediments

Surface sediments were collected from 19 sites in the coastal regions of the Yellow Sea and Bohai Sea of China

from Lianyungang to Donggang in November 2005. NP was detected in all the sediment samples at concentrations ranged from 8.8 to  $1.0 \times 10^3$  ng/g dry weight (dw), with an average value was  $0.12 \times 10^3$  ng/g dw (Table 1). The concentrations of NP in sediments from Jiaozhou Bay of northern Yellow Sea, Penglai along the coast of Bohai Strait, Tanggu in Bohai Bay and Lushun along the coast of Bohai Strait were higher than the average value ( $0.12 \times 10^3$  ng/g dw). Overall, approximately 20% of the sediment samples contained concentrations of NP that were above average. The highest concentration of NP was detected in sediments from Penglai along the coast of Bohai Strait. The Bohai Strait passes through Bohai Sea and Yellow Sea, and Penglai and Lushun are two terminals in the strait. The geographical position and water movement in this region differed from that of the other regions sampled in this study, which may explain the higher concentrations of NP that were found in this area. Jiaozhou Bay and Bohai Bay are the typical inner sea areas that are semi-enclosed by a coastal water body. Because they are semi-enclosed, particulates from land are easily deposited in the bay. In addition, there are two large estuaries (Dagu Estuary in Jiaozhou Bay and Haihe Estuary of Bohai Bay) that may bring pollution to the bay. These are likely reasons for the high concentrations of NP found in these bays.

NP concentrations have previously been evaluated in sediments from coastal environments in different countries and locations in China. A comparison of the NP concentrations observed in the present study with those recently reported for other Asian coastal regions is summarized in Table 2. The concentrations of NP observed in the present study were lower than those reported for Tokyo Bay, Japan,

**Table 1** Sampling information and concentrations of NP in sediment and bivalve samples from the northeast coast of China

Sampling site	Location	Region	Sediment NP (ng/g dw)	Bivalve		
				Lipid content (%)	NP (ng/g dw)	NP (ng/g lipid)
1	Lianyungang	Northern Yellow Sea	29	9.5	$1.8 \times 10^3$	$1.89 \times 10^4$
2	Fengshan	Northern Yellow Sea	30	5.0	88	$1.75 \times 10^3$
3	Rizhao	Northern Yellow Sea	10	–	–	–
4	Jiaozhou	Northern Yellow Sea	$0.13 \times 10^3$	–	–	–
5	Chengyang	Northern Yellow Sea	–	8.1	$0.82 \times 10^3$	$1.01 \times 10^4$
6	Lichang	Northern Yellow Sea	–	10	95	$0.945 \times 10^3$
7	Qingshan	Northern Yellow Sea	–	9.5	$0.21 \times 10^3$	$2.21 \times 10^3$
8	Rushan	Northern Yellow Sea	15	13	$0.15 \times 10^3$	$1.15 \times 10^3$
9	Weihai	Northern Yellow Sea	–	8.4	71	$0.845 \times 10^3$
10	Yantai	Northern Yellow Sea	–	12	76	$0.636 \times 10^3$
11	Penglai	Bohai Straits	$1.0 \times 10^3$	10	$0.10 \times 10^3$	$1.00 \times 10^3$
12	Longkou	Laizhou Bay	13	7.8	$0.15 \times 10^3$	$1.92 \times 10^3$
13	Fengjiabao	Laizhou Bay	32	7.8	69	$0.891 \times 10^3$
14	Tianjin	Bohai Bay	11	14	$0.26 \times 10^3$	$1.86 \times 10^3$
15	Tanggu	Bohai Bay	$0.26 \times 10^3$	8.6	67	$0.777 \times 10^3$
16	Hangu	Bohai Bay	17	–	–	–
17	Huludao	Bohai Bay	61	–	–	–
18	Jinzhou	Liaodong Bay	8.8	12	$7.6 \times 10^3$	$6.33 \times 10^4$
19	Fuzhou Bay	Liaodong Bay	12	12	$3.3 \times 10^3$	$2.75 \times 10^4$
20	Jinzhou, Dalian	Liaodong Bay	74	4.2	$0.84 \times 10^3$	$2.00 \times 10^4$
21	Lushun	Bohai Straits	$0.51 \times 10^3$	12	$0.13 \times 10^3$	$1.08 \times 10^3$
22	Dalian Bay	Northern Yellow Sea	66	15	44	$0.290 \times 10^3$
23	Zhuanghe	Northern Yellow Sea	29	15	ND	ND
24	Donggang	Northern Yellow Sea	12	13	ND	ND
Mean			$0.12 \times 10^3$	10	$0.79 \times 10^3$	$7.76 \times 10^3$
SD			$0.24 \times 10^3$	3.0	$1.8 \times 10^3$	$1.53 \times 10^4$

–: no data; ND: not detected.

**Table 2** Comparison of NP concentrations in sediment samples from Asian coastal regions

Location	Concentration (ng/g dw)	Reference
Yeongil Bay, Korea	89.1 <sup>a</sup>	Koh et al., 2006
Masan Bay, Korea	92–557	Li et al., 2008
Tokyo Bay, Japan	2.2–4560	Hashimoto et al., 2005
Tokyo Bay, Japan	20,700 <sup>b</sup>	Kurihara et al., 2007
Lingding Bay, China	60–571	Chen et al., 2006
Macao coast, China	293–1208	Chen et al., 2006
South China Sea, China	59–163	Chen et al., 2006
Jiaozhou Bay, China	3.6–299	Fu et al., 2007
Northeast coast, China	8.8–1.0 × 10 <sup>3</sup> (0.12 × 10 <sup>3</sup> ) <sup>a</sup>	This study

<sup>a</sup> Average value; <sup>b</sup> maximum value.

but similar to coastal regions of China and Korea.

The results of some studies have indicated a decreasing trend of NP in water and sediments from the rivers to the estuary and further to the sea in systems throughout China, including the Pearl River and adjacent region of the South China Sea, Bohai Bay and adjacent Nanpaiwu River, and Jiaozhou Bay and adjacent rivers (Chen et al., 2006; Fu et al., 2007; Hu et al., 2005b). Specifically, the concentrations of NP ranged from 110 to 7808 ng/g dw in rivers along the Pearl River Delta and from 48 to 31,704 ng/g dw in rivers adjacent to Jiaozhou Bay (Chen et al., 2006; Fu et al., 2007). Furthermore, the maximum concentration of NP was 970 ng/g dw in Nanpaiwu River adjacent to Bohai Bay (Hu et al., 2005b). Taken together, these findings suggest that rivers are the major source of NP in coastal environments in China. When comparing with the NP concentrations in Chinese rivers, the concentrations of NP observed in the present study were low. These findings correspond with those of previous studies and further confirm that river is a major source of NP in coastal environment of China.

A predicted no effect concentration (PNEC) of 39 ng/g can be derived from the aquatic PNEC assuming equilibrium partitioning for the sediment compartment (European Commission, 2002). Approximately 40% of the samples collected in the present study had concentrations of NP greater than the PNEC for sediment. The results suggest that NP may have adverse effects on sediment-dwelling species.

## 2.2 NP in bivalves

To further estimate the level of NP contamination in the northeast coastal environment of China, bivalves (oysters and mussels) were collected from 20 sites along the coast of the Bohai and Yellow Sea of China from Lianyungang to Donggang in November, 2005 (Fig. 1). The results and supporting information are listed in Table 1. NP was detected in all of the bivalve samples except for one sample collected from Zhuanghe and one from Donggang in the northern Yellow Sea. The concentrations of NP in the bivalves ranged from ND to 7.6 × 10<sup>3</sup> ng/g dw (average, 0.79 × 10<sup>3</sup> ng/g dw) or from ND to 6.33 × 10<sup>4</sup> ng/g lipid (average, 7.76 × 10<sup>3</sup> ng/g lipid). The level of NP differed obviously among sampled coastal regions and the standard

deviation of NP was 1.8 × 10<sup>3</sup>. These results suggest that there were different sources of NP in the areas evaluated in this study. Indeed, the input amount and water quality of wastewater discharged differed greatly as a result of variations in the industry and population of coastal cities. Jin et al. (2008) reported that PCBs and organochlorine pesticides were present in bivalves collected from the same sites evaluated in this study. In comparison, the standard deviation in the concentrations of NP (SD = 1.8 × 10<sup>3</sup>) in all bivalves samples was higher than that of PCBs (SD = 6.24) and organochlorine pesticides (SD: 0.63–681). These results demonstrate that variations in sampling area have a greater impact on NP than PCBs and organochlorine pesticides. In other words, local pollution source is largely responsible for the level of NP. Additionally, the concentrations of NP in bivalves from 25% of sampling sites were greater than the average value (0.79 × 10<sup>3</sup> ng/g dw). A high concentration of NP in sediments was detected in Jiaozhou Bay. Similarly, high concentration of NP was observed in bivalves collected from Jiaozhou Bay (Chengyang site). These results further confirmed that Jiaozhou Bay has been seriously impacted by NP pollution. The highest concentration of NP in bivalves was detected in Jinzhou of Liaodong Bay; although high concentrations were also observed in bivalves collected from other sampling sites in Liaodong Bay (Fuzhou Bay and the Jinzhou site of Dalian). Liaodong Bay is a semi-enclosed bay similar to Bohai Bay and Jiaozhou Bay. Therefore, these results suggest that the semi-enclosed bays may be more prone to the accumulation of NP than other kinds of areas. A high concentration of NP (1.8 × 10<sup>3</sup> ng/g dw) was also observed in bivalves from the Lianyungang site. Lianyungang is an important harbor in China which is in half-moon shape. The frequent harbor activity is likely responsible for the high concentration of NP in this area. There was no correlation between the concentration of NP in sediments and bivalves based on lipid ( $R^2 = 0.050$ ). Several factors may be responsible for this poor correlation, including the total organic carbon content (TOC) of sediments, characteristics of the water body, estuary deposition, harbor activity and differences in the ecosystem characteristics of the coastal environment.

There were several studies conducted to evaluate the levels of NP in bivalves in oceans. For example, the concentrations of NP were found to range from 51 to 289 ng/g dw in mussels collected from Masan Bay, Korea (Li et al., 2008). Hu et al. (2005a) reported that 4-NP was detected in all biota samples from Bohai Bay, with concentrations ranging from 142.6 to 677.8 ng/g lipid weight. In the Adriatic Sea, the concentrations of NP detected in clams and mussels ranged from 243–265 ng/g lipid weight (Ferrara et al., 2001). In the present study, higher concentrations were observed in Lianyungang and Jiaozhou Bay of northern Yellow Sea and in Liaodong Bay (Jinzhou Bay, Fuzhou Bay and Jinzhou of Dalian). The concentrations of NP in bivalves from Laizhou Bay, Bohai Bay and most of the coastal regions in the northern Yellow Sea observed in the present study were similar to those observed elsewhere (Ferrara et al., 2001; Hu et al., 2005a;

Li et al., 2008).

NP has already impacted most coastal bivalves in China, even those sold as seafood. In the present study, bivalves in 7 sites were bought from several coastal markets and analyzed to determine if they contained levels of NP that posed a health risk. Only one sample purchased from Fuzhou Bay market in Liaodong Bay region was found to have a relatively high concentration of NP ( $3.3 \times 10^3$  ng/g dw), the concentrations of NP in the other samples were in the range of ND to  $0.13 \times 10^3$  ng/g dw. The results of the present study indicated that there was serious NP pollution in bivalves from Liaodong Bay; therefore, it is possible that farms used to cultivate bivalve in the bay have been contaminated.

According to Food and Agriculture Organization of the United Nation (FAO), approximately 26 kg of seafood is consumed annually per person in China (Lindkvist et al., 2008). Because the average concentration of NP in bivalves from the northeast coast of China was  $0.14 \times 10^3$  ng/g wet weight in the present study, it is possible that there is a NP intake of about 10  $\mu$ g/day per person in China. This intake, which is based only on bivalve consumption, is slightly lower than the intake of NP of 12  $\mu$ g/day per person based on the consumption of 12 types of seafood in Italy (Ferrara et al., 2005); and much lower than the doses that causes effects in laboratory animals (9  $\mu$ g/g body weight) (Chapin et al., 1999). The domestic demand for seafood in China is expected to increase greatly in the future as the Chinese seafood industry grows which will pose global challenges and have widespread policy implications (Lindkvist et al., 2008). Therefore, additional studies should be conducted to clarify the risk that contamination of seafood with NP poses to human health.

### 2.3 POPs pollution in the northeast coast of China

Among the various types of organic pollutants identified in the study area by Jin (2008) and in the present study, NP, PCBs, DDTs, HCHs and total chlordane were detectable in most bivalve species. NP was the predominant contaminant with an average of  $0.79 \times 10^3$  ng/g dw, followed by DDTs (average, 600 ng/g dw), PCBs (average, 10 ng/g dw), HCHs (average, 10 ng/g dw) and total chlordane (average, 1 ng/g dw) in bivalve. There may be a risk of synergistic effects of various pollutants, including those mentioned in this study which is of particular concern in seafood.

## 3 Conclusions

NP was detected in sediments and bivalves collected from the northeast coast of China. Furthermore, the levels of NP differed greatly among sampling regions. Specifically, areas along the coast of the Bohai straits such as Penglai and Lushun and the semi-enclosed coastal environments such as Jiaozhou Bay and Liaodong Bay had higher concentrations of NP than other areas. Although the levels of NP observed in the present study did not pose a high risk to human health, there are still reasons

for concern because the distribution of NP observed in this study indicates an overall contamination along the northeast coast of China. Synergistic effects of various pollutants noted in the present and previous study may occur in areas observed. Overall, the results of this study indicate that there is a cause for concern regarding organic pollution in the northeastern coastal environment in China.

### Acknowledgments

This study was supported by the National Natural Science Foundation of China (No. 20767006) and the National High Technology Research and Development Program (863) of China (No. 2006AA09Z175).

### References

- Bennie D T, Sullivan C A, Lee H B, Peart T E, Maguire R J, 1997. Occurrence of alkylphenols and alkylphenol mono- and diethoxylates in natural waters of the Laurentian Great Lakes basin and the upper St. Lawrence River. *Science of the Total Environment*, 193: 263–275.
- Blackburn M A, Kirby S J, Waldock M J, 1999. Concentrations of alkylphenol polyethoxylates entering UK estuaries. *Marine Pollution Bulletin*, 38: 109–118.
- Chapin R E, Delaney J, Wang Y, Lanning L, Davis B, Collins B et al., 1999. The effects of 4-nonylphenol in rats: a multigeneration reproduction study. *Toxicological Sciences*, 52: 80–91.
- Chen B, Duan J C, Mai B X, Luo X J, Yang Q S, Sheng G Y et al., 2006. Distribution of alkylphenols in the Pearl River Delta and adjacent northern South China Sea, China. *Chemosphere*, 63: 652–661.
- Ekelund R, Bergman Å, Granmo Å, Berggren M, 1990. Bioaccumulation of 4-nonylphenol in marine animals – a re-evaluation. *Environmental Pollution*, 64: 107–120.
- European Commission, 2002. European Chemicals Bureau, European Union Risk Assessment Report. 4-Nonylphenol (branched) and Nonylphenol. Environment and Quality of Life Series. EUR-20387-EN.
- Ferguson P L, Iden C R, Brownwell B J, 2001. Distribution and fate of neutral alkylphenol ethoxylate metabolites in a sewage-impacted urban estuary. *Environmental Science & Technology*, 35: 2428–2435.
- Ferrara F, Fabietti F, Delise M, Bocca A P, Funari E, 2001. Alkylphenolic compounds in edible molluscs of the Adriatic Sea (Italy). *Environmental Science & Technology*, 35: 3109–3112.
- Ferrara F, Fabietti F, Delise M, Funari E, 2005. Alkylphenols and alkylphenol ethoxylates contamination of crustaceans and fishes from the Adriatic Sea (Italy). *Chemosphere*, 59: 1145–1150.
- Fu M Z, Li Z Y, Gao H W, 2007. Distribution characteristics of nonylphenol in Jiaozhou Bay of Qingdao and its adjacent rivers. *Chemosphere*, 69: 1009–1016.
- Hashimoto S, Horiuchi A, Yoshimoto T, Nakao M, Omura H, Kato Y et al., 2005. Horizontal and vertical distribution of estrogenic activities in sediments and waters from Tokyo Bay, Japan. *Archives of Environmental Contamination and Toxicology*, 48: 209–216.
- Hou S G, Xu J, Wang L, Dai S G, Liu X Y, 2005. Primary study on nonylphenol and nonylphenol polyethoxylates in aquatic environment at Lanzhou reach of Yellow River. *Environmental Chemistry*, 24: 250–254.

- Hu J Y, Jin F, Wan Y, Yang M, An L, An W et al., 2005a. Trophodynamic behavior of 4-nonylphenol and nonylphenol polyethoxylate in a marine aquatic food web from Bohai Bay, North China: Comparison to DDTs. *Environmental Science & Technology*, 39: 4801–4807.
- Hu J Y, Wan Y, Shao B, Jin X, An W, Jin F et al., 2005b. Occurrence of trace organic contaminants in Bohai Bay and its adjacent Nanpaiwu River, North China. *Marine Chemistry*, 95: 1–13.
- Ishibashi H, Hirano M, Matsumura N, Watanabe N, Takao Y, Arizono K, 2006. Reproductive effects and bioconcentration of 4-nonylphenol in medaka fish (*Oryzias latipes*). *Chemosphere*, 65: 1019–1026.
- Isobe T, Nishiyama H, Nakashima A, Takada H, 2001. Distribution and behaviour of nonylphenol, octylphenol, and nonylphenol monoethoxylate in Tokyo metropolitan area: their association with aquatic particles and sedimentary distributions. *Environmental Science & Technology*, 35: 1041–1049.
- Jin Y, Hong S H, Li D, Shim W J, Lee S S, 2008. Distribution of persistent organic pollutants in bivalves from the northeast of China. *Marine Pollution Bulletin*, 57: 775–781.
- Jobling S, Sumpter J P, 1993. Detergent components in sewage effluent are weakly estrogenic to fish: An *in vitro* study using rainbow trout (*Oncorhynchus mykiss*) hepatocytes. *Aquatic Toxicology*, 27: 361–372.
- Koh C H, Khim J S, Villeneuve D L, Kannan K, Giesy J P, 2006. Characterization of trace organic contaminants in marine sediment from Yeongil Bay, Korea: 1. Instrumental analyses. *Environmental Pollution*, 142: 39–47.
- Kurihara R, Watanabe E, Ueda Y, Kakuno A, Fujii K, Shiraishi F et al., 2007. Estrogenic activity in sediments contaminated by nonylphenol in Tokyo Bay (Japan) evaluated by vitellogenin induction in male mummichogs (*Fundulus heteroclitus*). *Marine Pollution Bulletin*, 54: 1315–1320.
- Li D, Dong M, Shim W J, Yim U H, Hong S H, Kannan N, 2008. Distribution characteristics of nonylphenolic chemicals in Masan Bay environments, Korea. *Chemosphere*, 71: 1162–1172.
- Li D, Oh J R, Park J, 2003. Direct extraction of alkylphenols, chlorophenols and bisphenol A from acid-digested sediment suspension for simultaneous gas chromatographic-mass spectrometric analysis. *Journal of Chromatography A*, 1012: 207–214.
- Li D, Park J, Oh J R, 2001. Silyl derivatization of alkylphenol, chlorophenols, and bisphenol A for simultaneous GC/MS determination. *Analytical Chemistry*, 73: 3089–3095.
- Lindkvist K B, Trondsen T, Xie J, 2008. Restructuring the Chinese seafood industry, global challenges and policy implications. *Marine Policy*, 32: 432–441.
- Li Z Y, Li D, Oh J R, Je J G, 2004. Seasonal and spatial distribution of nonylphenol in Shihwa Lake, Korea. *Chemosphere*, 56: 611–618.
- Renner R, 1997. European bans on surfactant trigger transatlantic debate. *Environmental Science & Technology*, 31: 316A–320A.
- Shang D Y, Macdonald R W, Ikononou M G, 1999. Persistence of nonylphenol ethoxylate surfactants and their primary degradation products in sediments from near a municipal outfall in the strait of Georgia, British Columbia, Canada. *Environmental Science & Technology*, 33: 1366–1372.
- Soto A M, Justicia H, Wray J W, Sonnenschein C, 1991. p-Nonylphenol, an estrogenic xenobiotic released from 'modified' polystyrene. *Environmental Health Perspectives*, 92: 167–173.
- Wang J, Dong M, Shim W J, Kannan N, Li D, 2007. Improved cleanup technique for gas chromatographic-mass spectrometric determination of alkylphenols from biota extract. *Journal of Chromatography A*, 1171: 15–21.
- Ying G G, Williams B, Kookana R, 2002. Environmental fate of alkylphenols and alkylphenol ethoxylates – a review. *Environmental International*, 28: 215–226.