JOURNAL OF AGRICULTURAL AND FOOD CHEMISTRY

Investigation of Methylmercury and Total Mercury Contamination in Mollusk Samples Collected from Coastal Sites along the Chinese Bohai Sea

LI-NA LIANG, JIAN-BO SHI, BIN HE, GUI-BIN JIANG,* AND CHUN-GANG YUAN

Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, P.O. Box 2871, 100085 Beijing, China

This paper presents the investigation results of methylmercury and total mercury in gastropod and bivalve species collected from eight coastal sites along the Chinese Bohai Sea. The total mercury was directly determined by atomic fluorescence spectrometry (AFS), while the methylmercury was measured by a laboratory established high performance liquid chromatography-atomic fluorescence spectrometry system (HPLC-AFS). Certified reference material DORM-2 (Dogfish muscle) was used to validate the two methods and the obtained results proved to be in good agreement with the certified values. It was demonstrated that the mercury contamination was commonly existed in all selected mollusks, with methylmercury and total mercury concentration in the range of 4.8−168.4 and 6.7−453.0 ng Hg g^{-1}, respectively. Mollusks from HuLuDao were the most mercury contaminated, and those from PengLai took the second place. The species-dependent bioaccumulation capacity was observed in this study. Gastropods showed more capacity to bioaccumulate mercury than bivalves, and mercury contents in two kinds of gastropods (*Rapana venosa* and *Neverita didyma*) presented some uplifted trends with the dimensions increasing of the gastropods. *Rapana venosa* was found to be a potential biomarker to monitor mercury pollution in oceans. Evaluations were also made concerning about the ratio of methylmercury to total mercury.

KEYWORDS: Methylmercury; total mercury; HPLC-AFS; mollusks

INTRODUCTION

Mercury is an important but toxic element. Both the direct drainage of industrial wastewater and rainwater runoff leads to mercury contamination in oceans. Mercury is first absorbed by phytoplankton and then by various consumers within the oceanic ecosystem. Mollusks lie in the second trophic level in this ecosystem and accumulate less methylmercury than predatory fish. However, mollusks are popular seafood, and long-term consumption of them may result in methylmercury accumulation in the human body.

The toxicity of mercury is well known since the notorious poisoning accident at Minamata Bay in southern Japan during the 1950s and 1960s. This tragic event was due to the consumption of methylmercury-contaminated fish, and 48 persons died. Acute exposure to methylmercury is lethal, and chronic low-dose consumption of methylmercury-contaminated fish can cause severe adverse effects to organs, the central nervous system, and the immune system. Consumption of mercury-contaminated fish is particularly hazardous for pregnant women because of its heredity effect on the unborn fetus. Its virulence effect cannot be eradicated for many years.

Because methylmercury is the most toxic form of mercury, it is now more appropriate to determine not only the total mercury content of seafood but also the methylmercury content. Gas chromatography (GC) separation coupled with electron capture detection (ECD) proposed by Westöö (*1*) was the commonly used method for the determination of methylmercury. More recently, high performance liquid chromatography (HPLC) hyphenated with atomic fluorescence detector (AFS) has become popular (*2*−*4*).

In this present investigation, the contents of methylmercury and total mercury of 88 mollusks samples collected from eight coastal sites were analyzed. The methylmercury was determined with our previously proposed HPLC-AFS method (*4*), but the sample preparation was slightly modified in order to shorten the preparation time. The AFS method was adopted for the total mercury determination. It was found that the ratio of methylmercury to total mercury varied slightly between gastropods and bivalves, and *Rapana venosa* was found to be a potential biomarker to monitor the mercury pollution in ocean.

MATERIALS AND METHODS

Instrumentation. The HPLC (LC-10AT vp, Shimadzu, Japan) and AFS (AF-610A, Beijing Raileigh Analytical Instrument Co., China) hyphenation system was as described previously (*4*). The AFS peristaltic

* Corresponding author. Fax: 8610−62849179. E-mail: gbjiang@mail.rcees.ac.cn.

10.1021/jf034199l CCC: $25.00
Published on Web 10/31/2003

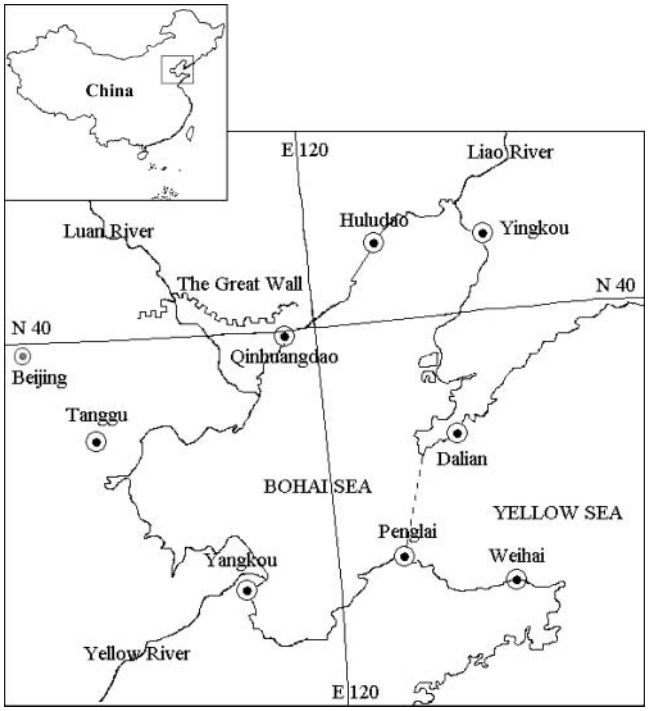

Figure 1. Sampling sites of mollusk samples along the Chinese Bohai Sea.

pump was substituted with a more competent peristaltic pump on the FIA-3100 analyzer (Beijing Wantuo Instrument Co., China), which was used to pump the $K_2S_2O_8$ oxidant solution and KBH_4 reducing agent. The total mercury determination was made with reference to the previous method (*5*).

Reagents and Standards. All chemicals were of guaranteed reagent grade except where specified, and Milli-Q water was used throughout. Stock solutions of standard methylmercury chloride (CH_3HgCl, Merck) and mercury chloride ($HgCl_2$, Merck) (1 mg ml^{-1} as Hg) were prepared by dissolving appropriate amounts in methanol and 5% v/v HNO_3, respectively. The mercury working solutions were obtained by dilution with methanol or 10% v/v HNO_3 and prepared daily before use. All solutions were stored at 4 °C.

Aqueous solutions, 0.1 mol l^{-1} tetrabutylammonium bromide (TBA), 1 mol l^{-1} NaCl, 25% m/v KOH/CH_3OH, and 10 mmol l^{-1} $Na_2S_2O_3$ solution, were prepared weekly and stored at 4 °C. Daily prepared HPLC mobile phase was a mixture of appropriate TBA, NaCl solution, water, and CH_3OH and filtered through a 0.45-μm membrane filter before use.

KBH_4 solutions (0.01% m/v and 0.2% m/v) were prepared daily by dissolving the required amount in 0.2% m/v KOH. An oxidant solution of 1% m/v $K_2S_2O_8$ solution was prepared in 10% v/v HCl.

Sampling. Figure 1 shows the eight sampling sites of mollusks. These sites spread out around the Bohai Sea. Mollusks with different dimensions were divided into several size groups, and the dimensions and numbers of mollusks were listed in **Table 1** according to sampling sites and species. The soft tissues of mollusks were excised with stainless steel scalpel blades, thoroughly rinsed with Milli-Q water to remove extraneous impurities, and homogenized using a blender. The homogenized samples were kept at −18 °C until analysis.

Different species of mollusks were identified according to the catalog of marine mollusks in reference books (*6*).

Procedures. For methylmercury analysis, samples were prepared according to the published method with slight modification (*7*). Briefly, 2 mL of 25% m/v KOH/CH_3OH was added to 1.0−2.0-g homogenized wet samples in a 50-mL centrifuge tube and shaken mechanically overnight. Then 6 mL of CH_2Cl_2 was added, and 1.5 mL of concentrated HCl was dropped in sequence, followed by shaking for 10 minutes to extract organic mercury into the CH_2Cl_2 phase. After centrifuging at 2000 rpm for 10 min, the 4 mL of CH_2Cl_2 phase was transferred into a 10-mL glass tube and extracted with 1 mL of sodium thiosulfate. Shaking for 45 min was needed to hasten the extraction speed. Another centrifugation at 5000 rpm, 4 °C for 15 min, was necessary to separate the fat layer. The water phase was injected directly into the HPLC-AFS system.

For total mercury analysis, approximately 1 g (wet weight) of soft tissues were directly weighed into a PTFE digestion container. Concentrated nitric acid (3 mL) was added, and the containers were sealed and left to predigest overnight on an electrothermal hotplate at 40 °C. After cooling, 2 mL of hydrogen peroxide was added into the containers, which were placed in stainless steel bombs, sealed with a screw closure to avoid any acid leakage, and placed in an oven. The oven temperature was first raised to 50 °C and kept for 1 h, then increased to 160 °C for another 4 h. After cooling to room temperature, the solutions were completely transferred into a 50-mL PET bottle and diluted with Milli-Q water, then determined by AFS method. Reagent blanks were processed simultaneously.

RESULTS AND DISCUSION

Evaluation of the Methods. Certified reference material DORM-2 (Dogfish muscle) was analyzed to validate the two analytical methods. As shown in **Table 2**, the experimental results were in good agreement with the certified values.

Mercury Contents in Mollusks. Contents of methylmercury and total mercury and the ratio of methylmercury to total mercury in gastropods and bivalves at each sampling site are shown in **Tables 3** and **4**, respectively. Mercury was detected in all samples. Methylmercury levels ranged from 4.8 to 168.4 ng Hg g^{-1}, while total mercury contents ranged from 6.7 to 453.0 ng Hg g^{-1}. It was found that mollusks collected from different places contained different levels of methylmercury and total mercury. Roughly, samples collected from HuLuDao contained the highest level, those from PengLai took the second place, and those from the other six sampling sites contained the lowest level. On the basis of the analysis of **Tables 3** and **4**, gastropods showed higher bioaccumulation capacity for mercury than bivalves.

Methylmercury in Mollusks. Methylmercury contents in bivalves varied from 4.8 to 47.5 ng Hg g^{-1}. The methylmercury and total mercury contents in bivalves at the eight sampling sites are given in **Table 4**. Methylmercury levels in bivalves from HuLuDao and PengLai were similar and higher than those from the other sites. As for gastropods, the methylmercury contents were 4.9−168.4 ng Hg g^{-1}, which was similar to those in bivalves. From **Table 3**, it was obvious that gastropods from HuLuDao contained the highest levels of methylmercury, while methylmercury contents in those from other sites differed slightly. For all the three-size group of *Rapana venosa* from HuLuDao, the methylmercury levels ranged from 40.9 to 168.4 ng g^{-1}, and the large *Rapana venosa* contained methylmercury content exceeding the maximum permissible levels (MPLs) of 0.16 mg kg^{-1} wet weight, which was established by the Ministry of Health of the State of Minnesota (USA). Comparing with the similar dimensions of *Rapana venosa* in other sampling sites, those from HuLuDao were the most seriously polluted.

Total Mercury in Mollusks. Total mercury contents in bivalves varied from 6.7 to 194.2 ng Hg g^{-1}. According to **Table 4**, the total mercury contamination levels of bivalves from HuLuDao and PengLai were similar and higher than those from the other sites. Considering the maximum permissible levels (MPLs) of total mercury in fish and shellfish set up by China (0.3 mg kg^{-1}), Europe (EEC decision 93/351, 0.5 mg kg^{-1}), and WHO (0.5 mg kg^{-1}), all the bivalves could be eaten with no worry about the hazards to human health. *Mytilus edulis* had medium mercury contents, but the one from PengLai showed extraor-

Table 1. Dimension and Number for Three Size Classes (S-small, M-middle, L-large) of the Mollusks in Eight Sampling Sites: Bivalves (length) and Gastropods (height)

species	denomination	dimension (mm)	no.
WeiHai			
gastropods	Rapana venosa, Valenciennes, 1846	95 ± 1	2
	Neverita didyma, Röding, 1798	41 ± 3	5
bivalves	Ruditapes philippinarum, Adams & Reeve, 1850	34 ± 2	22
	Meretix meretrix, Linnaeus, 1758	54 ± 2	7
	Sinonovacula constricta, Lamarck, 1818	57 ± 2	14
	Amusium, Röding, 1798	80 ± 6	6
	Chlamys (Azumapecten) farreri, Jones & Preston, 1904	51 ± 4	8
	Crassostrea talienwhanensis, Crosse, 1862	64 ± 11	4
	Mytilus edulis, Linnaeus, 1758	58 ± 4	12
	Mya arenaria, Linnaeus, 1758	66 ± 5	3
PengLai			
gastropods	Neptunea arthritica cumingii, Crosse, 1862	87 ± 3	4
	Rapana venosa, Valenciennes, 1846(L)	95 ± 0	2
	Rapana venosa, Valenciennes, 1846(S)	57 ± 2	7
	Neverita didyma, Röding, 1798(L)	46 ± 4	2
	Neverita didyma, Röding, 1798(S)	21 ± 1	12
bivalves	Mactra (Mactra) veneriformis, Reeve, 1854	35 ± 1	14
	Ruditapes philippinarum, Adams & Reeve, 1850	33 ± 2	20
	Meretix meretrix, Linnaeus, 1758	77 ± 1	7
	Scapharca subcrenata, Lischke, 1869	33 ± 2	15
	Amusium, Röding, 1798	106 ± 2	2
	Chlamys (Azumapecten) farreri, Jones & Preston, 1904	51 ± 4	13
	Crassostrea talienwhanensis, Crosse, 1862	49 ± 4	6
	Mytilus edulis, Linnaeus, 1758	59 ± 5	8
	Mya arenaria, Linnaeus, 1758	74 ± 1	3
YangKou			
gastropods	Rapana venosa, Valenciennes, 1846	96 ± 6	2
	Neverita didyma, Röding, 1798(L)	31 ± 0	11
	Neverita didyma, Röding, 1798(S)	18 ± 1	23
bivalves	Mactra (Mactra) veneriformis, Reeve, 1854	32 ± 3	12
	Ruditapes philippinarum, Adams & Reeve, 1850	43 ± 4	9
	Meretix meretrix, Linnaeus, 1758	50 ± 3	10
	Sinonovacula constricta, Lamarck, 1818	54 ± 2	11
	Scapharca subcrenata, Lischke, 1869(L)	54 ± 6	3
	Scapharca subcrenata, Lischke, 1869(S)	35 ± 2	6
TangGu			
gastropods	Rapana venosa, Valenciennes, 1846(L)	119	1
	Rapana venosa, Valenciennes, 1846(M)	68 ± 4	3
	Rapana venosa, Valenciennes, 1846(S)	40 ± 4	11
	Neverita didyma, Röding, 1798(L)	26 ± 2	12
	Neverita didyma, Röding, 1798(S)	16 ± 2	43
bivalves	Mactra (Mactra) veneriformis, Reeve, 1854	30 ± 2	27
	Meretix meretrix, Linnaeus, 1758	42 ± 3	10
	Sinonovacula constricta, Lamarck, 1818	55 ± 2	17
	Scapharca subcrenata, Lischke, 1869	49 ± 2	7
	Amusium, Röding, 1798	108 ± 1	2
	Crassostrea talienwhanensis, Crosse, 1862	63 ± 4	5
	Mya arenaria, Linnaeus, 1758(L)	100 ± 5	3
	Mya arenaria, Linnaeus, 1758(S)	68 ± 2	4
QinHuangDao			
gastropods	Rapana venosa, Valenciennes, 1846(L)	81 ± 2	3
	Rapana venosa, Valenciennes, 1846(S)	54 ± 3	5
	Neverita didyma, Röding, 1798	45 ± 1	4
bivalves	Mactra (Mactra) veneriformis, Reeve, 1854	32 ± 2	18
	Ruditapes philippinarum, Adams & Reeve, 1850	28 ± 2	63
	Meretix meretrix, Linnaeus, 1758	55 ± 3	7
	Sinonovacula constricta, Lamarck, 1818	57 ± 2	14
	Scapharca subcrenata, Lischke, 1869	49 ± 1	4
	Amusium, Röding, 1798	40 ± 2	18
	Chlamys (Azumapecten) farreri, Jones & Preston, 1904	68 ± 3	7
	Crassostrea talienwhanensis, Crosse, 1862	49 ± 2	7
	Mytilus edulis, Linnaeus, 1758	68 ± 5	16
HuLuDao			
gastropods	Rapana venosa, Valenciennes, 1846(L)	84 ± 5	2
	Rapana venosa, Valenciennes, 1846(M)	66 ± 2	4
	Rapana venosa, Valenciennes, 1846(S)	44 ± 2	11
	Neverita didyma, Röding, 1798(L)	56 ± 1	3
	Neverita didyma, Röding, 1798(M)	38 ± 4	4
	Neverita didyma, Röding, 1798(S)	24 ± 2	8
bivalves	Mactra (Mactra) veneriformis, Reeve, 1854	34 ± 2	18
	Ruditapes philippinarum, Adams & Reeve, 1850	32 ± 2	42
	Meretix meretrix, Linnaeus, 1758	59 ± 2	5
	Scapharca subcrenata, Lischke, 1869	40 ± 5	5

Table 1. Cont.

species	denomination	dimension (mm)	no.
YingKou			
gastropods	Rapana venosa, Valenciennes, 1846	69 ± 5	4
bivalves	Mactra (Mactra) veneriformis, Reeve, 1854	30 ± 2	22
	Ruditapes philippinarum, Adams & Reeve, 1850	37 ± 2	18
	Meretix meretrix, Linnaeus, 1758	52 ± 3	7
	Sinonovacula constricta, Lamarck, 1818	48 ± 2	33
	Amusium, Röding, 1798	68 ± 3	5
	Mytilus edulis, Linnaeus, 1758	71 ± 7	6
DaLian			
gastropods	Neptunea arthritica cumingii, Crosse, 1862(L)	122 ± 1	2
	Neptunea arthritica cumingii, Crosse, 1862(M)	70 ± 0	5
	Neptunea arthritica cumingii, Crosse, 1862(S)	42 ± 3	14
	Neverita didyma, Röding, 1798	38 ± 2	10
bivalves	Mactra (Mactra) veneriformis, Reeve, 1854	32 ± 1	12
	Ruditapes philippinarum, Adams & Reeve, 1850	30 ± 3	46
	Meretix meretrix, Linnaeus, 1758	50 ± 3	9
	Sinonovacula constricta, Lamarck, 1818	52 ± 2	29
	Scapharca subcrenata, Lischke, 1869	34 ± 2	16
	Amusium, Röding, 1798	72 ± 5	5
	Chlamys (Azumapecten) farreri, Jones & Preston, 1904	50 ± 4	11
	Crassostrea talienwhanensis, Crosse, 1862	59 ± 7	6
	Mytilus edulis, Linnaeus, 1758	61 ± 5	21

Table 2. Results of Methylmercury (MeHg) and Total Mercury (HgT) Contents in DORM-2 (Dogfish Muscle)

sample	determination values (mean[a] ± S. D.[b], ng g^{-1})	certified values (mean[a] ± S. D.[b], ng g^{-1})
MeHg concentration (mean[a] ± S. D.[b], ng g^{-1})[c]	4237 ± 143	4470 ± 320
HgT concentration (mean[a] ± S. D.[b], ng g^{-1})[d]	4560 ± 460	4640 ± 260

[a] $N = 3$. [b] Standard deviation. [c] Determined by HPLC-AFS. [d] Determined by AFS.

dinarily high mercury content. It was supposed that this sample was collected from an extremely mercury-polluted place, and this was also supported by the more severe contamination statues in bivalves from PengLai. As for gastropods, the total mercury contents ranged from 11.3 to 453.0 ng Hg g^{-1}. **Table 3** indicates that mercury contamination level in gastropods from HuLuDao was at least twice that of those from PengLai and other sites. Among the 88 samples, only large *Rapana venosa* in HuLuDao contained high levels of total mercury that went beyond the MPLs set up by China. However, much attention should be paid when consuming a large amount of this species of seafood in a short period. When the total mercury contents in bivalves and gastropods were compared, it was easy to find the great difference between them, which just illuminated the distinct mercury bioaccumulation capacity of them. Maybe this is because the bivalves are grass-eating mollusks, while the three collected gastropod species are all predatory flesh-eating mollusks, the main food of which are bivalves. So from the view of trophic

Table 3. Methylmercury (MeHg), Total Mercury (HgT), and the MeHg/HgT Ratio in Gastropods Collected from Eight Coastal Cities in China (Wet Weight, ng g^{-1})

sites	denomination	MeHg	HgT	MeHg/HgT (%)
WeiHai	Rapana venosa, Valenciennes, 1846	18.6	42.4	44
	Neverita didyma, Röding, 1798	14.1	33.0	43
PengLai	Neptunea arthritica cumingii, Crosse, 1862	27.6	94.1	29
	Rapana venosa, Valenciennes, 1846(L)	19.1	97.9	19
	Rapana venosa, Valenciennes, 1846(S)	18.1	63.0	29
	Neverita didyma, Röding, 1798(L)	44.2	138.7	32
	Neverita didyma, Röding, 1798(S)	38.5	62.2	62
YangKou	Rapana venosa, Valenciennes, 1846	18.8	59.1	32
	Neverita didyma, Röding, 1798(L)	23.2	96.1	24
	Neverita didyma, Röding, 1798(S)	16.1	32.0	50
TangGu	Rapana venosa, Valenciennes, 1846(L)	14.9	37.8	40
	Rapana venosa, Valenciennes, 1846(M)	10.5	24.0	44
	Rapana venosa, Valenciennes, 1846(S)	4.9	11.3	44
	Neverita didyma, Röding, 1798(L)	10.4	26.6	39
	Neverita didyma, Röding, 1798(S)	13.0	42.7	30
QinHuangDao	Rapana venosa, Valenciennes, 1846(L)	13.8	41.7	33
	Rapana venosa, Valenciennes, 1846(S)	16.1	53.4	30
	Neverita didyma, Röding, 1798	17.8	44.0	40
HuLuDao	Rapana venosa, Valenciennes, 1846(L)	168.4	453.0	37
	Rapana venosa, Valenciennes, 1846(M)	55.9	276.7	20
	Rapana venosa, Valenciennes, 1846(S)	40.9	199.2	21
	Neverita didyma, Röding, 1798(L)	49.9	128.0	39
	Neverita didyma, Röding, 1798(M)	34.3	77.2	44
	Neverita didyma, Röding, 1798(S)	29.7	53.1	56
YingKou	Rapana venosa, Valenciennes, 1846	24.4	60.5	40
DaLian	Neptunea arthritica cumingii, Crosse, 1862(L)	25.2	109.5	23
	Neptunea arthritica cumingii, Crosse, 1862(M)	17.5	67.2	26
	Neptunea arthritica cumingii, Crosse, 1862(S)	20.2	83.3	24
	Neverita didyma, Röding, 1798	14.2	39.3	36

Table 4. Methylmercury (MeHg), Total Mercury (HgT), and the MeHg/HgT Ratio in Bivalves Collected from Eight Coastal Cities in China (Wet Weight, ng g^{-1})

sites	denomination	MeHg	HgT	MeHg/HgT (%)
WeiHai	Ruditapes philippinarum, Adams & Reeve, 1850	8.4	14.8	57
	Meretix meretrix, Linnaeus, 1758	8.0	9.1	87
	Sinonovacula constricta, Lamarck, 1818	7.3	10.4	70
	Amusium, Röding, 1798	13.9	20.2	69
	Chlamys (Azumapecten) farreri, Jones & Preston, 1904	11.6	17.1	68
	Crassostrea talienwhanensis, Crosse, 1862	8.2	15.7	52
	Mytilus edulis, Linnaeus, 1758	9.0	11.2	81
	Mya arenaria, Linnaeus, 1758	13.8	18.1	76
PengLai	Mactra (Mactra) veneriformis, Reeve, 1854	15.6	19.7	80
	Ruditapes philippinarum, Adams & Reeve, 1850	21.3	26.9	79
	Meretix meretrix, Linnaeus, 1758	47.5	92.1	52
	Scapharca subcrenata, Lischke, 1869	26.5	44.1	60
	Amusium, Röding, 1798	16.6	23.3	71
	Chlamys (Azumapecten) farreri, Jones & Preston, 1904	22.8	34.0	67
	Crassostrea talienwhanensis, Crosse, 1862	37.0	64.4	58
	Mytilus edulis, Linnaeus, 1758	40.6	194.2	21
	Mya arenaria, Linnaeus, 1758	16.6	24.4	68
YangKou	Mactra (Mactra) veneriformis, Reeve, 1854	12.3	20.7	59
	Ruditapes philippinarum, Adams & Reeve, 1850	12.5	32.5	38
	Meretix meretrix, Linnaeus, 1758	10.4	16.1	65
	Sinonovacula constricta, Lamarck, 1818	8.4	10.7	79
	Scapharca subcrenata, Lischke, 1869(L)	18.0	45.4	40
	Scapharca subcrenata, Lischke, 1869(S)	27.7	68.4	41
TangGu	Mactra (Mactra) veneriformis, Reeve, 1854	5.6	7.8	72
	Meretix meretrix, Linnaeus, 1758	4.8	6.7	72
	Sinonovacula constricta, Lamarck, 1818	8.7	9.8	89
	Scapharca subcrenata, Lischke, 1869	10.4	16.4	63
	Amusium, Röding, 1798	10.8	22.4	48
	Crassostrea talienwhanensis, Crosse, 1862	11.7	15.6	75
	Mya arenaria, Linnaeus, 1758(L)	11.6	16.1	72
	Mya arenaria, Linnaeus, 1758(S)	21.8	51.3	42
QinHuangDao	Mactra (Mactra) veneriformis, Reeve, 1854	13.0	16.9	77
	Ruditapes philippinarum, Adams & Reeve, 1850	13.9	18.6	75
	Meretix meretrix, Linnaeus, 1758	7.5	12.6	59
	Sinonovacula constricta, Lamarck, 1818	14.4	19.6	74
	Scapharca subcrenata, Lischke, 1869	11.2	13.9	81
	Amusium, Röding, 1798	9.7	13.4	73
	Chlamys (Azumapecten) farreri, Jones & Preston, 1904	14.5	19.7	74
	Crassostrea talienwhanensis, Crosse, 1862	14.8	18.1	82
	Mytilus edulis, Linnaeus, 1758	8.9	11.0	80
HuLuDao	Mactra (Mactra) veneriformis, Reeve, 1854	25.2	77.8	32
	Ruditapes philippinarum, Adams & Reeve, 1850	24.4	38.2	64
	Meretix meretrix, Linnaeus, 1758	15.8	19.9	80
	Scapharca subcrenata, Lischke, 1869	38.4	99.3	39
YingKou	Mactra (Mactra) veneriformis, Reeve, 1854	18.7	43.2	43
	Ruditapes philippinarum, Adams & Reeve, 1850	15.7	27.8	56
	Meretix meretrix, Linnaeus, 1758	8.6	15.0	57
	Sinonovacula constricta, Lamarck, 1818	23.6	28.4	83
	Amusium, Röding, 1798	18.8	27.3	69
	Mytilus edulis, Linnaeus, 1758	19.6	29.4	67
DaLian	Mactra (Mactra) veneriformis, Reeve, 1854	12.6	30.4	41
	Ruditapes philippinarum, Adams & Reeve, 1850	6.4	11.1	58
	Meretix meretrix, Linnaeus, 1758	16.8	25.1	67
	Sinonovacula constricta, Lamarck, 1818	15.6	24.7	63
	Scapharca subcrenata, Lischke, 1869	14.4	26.2	55
	Amusium, Röding, 1798	15.3	28.2	54
	Chlamys (Azumapecten) farreri, Jones & Preston, 1904	17.1	30.5	56
	Crassostrea talienwhanensis, Crosse, 1862	13.0	24.1	54
	Mytilus edulis, Linnaeus, 1758	17.1	30.9	55

levels, the gastropods are superior to bivalves and can bioaccumulate mercury effectively.

Combining with the above-mentioned comparison of methylmercury and total mercury content in mollusks from the eight sampling sites, we could easily find the serious mercury contamination levels in mollusks from HuLuDao. In HuLuDao city, the biggest zinc plant in Asia, built in the 1930s', continually pours wastewater and waste residue containing heavy metals into the Bohai Sea. It causes the excess Hg, Zn, Cu, Cd, and Pb deposition in the sediments and halobios. Furthermore, the wastewater from the oil refining plant, chemical plant, and petrochemical industry in JinXi city, on the west of HuLuDao city, contribute to the mercury pollution in the mollusks. The wastewater flows through the WuLi River and finally pours into the Bohai Sea. It was reported that, from the entrance of the WuLi River to the Bohai Sea, the deposited mercury had reached 90 tons. Many halobios have been killed out, and the living halobios cannot be eaten any more. As for PengLai, the mercury pollution in mollusks attribute to the paper mill and the gold mines in the adjacent area. Wastewater from

the paper mill is dumped into the Bohai Sea with no disposal. Also, the private industrial owners extract gold by the gold−mercury amalgam method, which causes the local mercury pollution in rivers and soils, and all the mercury converges to the Bohai Sea at last.

Bioaccumulation of Mercury in Mollusks. Three species of gastropods and two species of bivalves were collected with different sizes. According to the different dimensions in each sampling site, the three gastropods were roughly divided into three-size classes (i.e., the L (large), M (middle), and S (small) classes). Two bivalve samples, *Scapharca subcrenata* and *Mya arenaria*, were divided into L (large) and S (small) size classes. The results listed in **Table 3** showed that mercury bioaccumulation in different sizes of gastropods differed evidently. Mercury in *Rapana venosa* showed uplifted trends with the increasing of the shells' size. *Rapana venosa* from three sites presented the elevated Hg content, especially in the more severely contaminated samples collected from HuLuDao. However, *Rapana venosa* from QinHuangDao did not agree with this trend, probably because the growth dilution effect played an important role. *Neverita didyma* also showed similar variability to *Rapana venosa*. Three dimensions of *Neptunea arthritica cumingii* were only collected in DaLian and the accretion trend was not very clear. Other than *Rapana venosa* and *Neverita didyma*, *Scapharca subcrenata* and *Mya arenaria* did not show the uplifted trends. However, this conclusion was unilateral for lacking of sufficient data. The highest mercury level and the infinite bioaccumulation in the *Rapana venosa* soft tissue might indicate that it is a potential biomarker to monitor the mercury pollution in oceanic ecosystems.

The Ratio of Methylmercury to Total Mercury. The proportions of methylmercury to total mercury in bivalves ranged from 20.9 to 89.3%, which was in accord with the previous reports. Claisse et al. reported a methylmercury/total mercury ratio from 21 to 74% in mussels and oysters along the French Coast (*8*). Mikac et al. reported a methylmercury/total mercury ratio of about 40% in mussels from the Krka Estuary (Croatia) (*9*) in 1996. While for gastropod species, the ratio of methylmercury to total mercury was in the range of 19.5 to 62.0%. However, no literature has reported the ratio of methylmercury to total mercury in gastropods. This ratio in gastropods was lower than that in bivalves, according to our results. The different proportions of methylmercury to total mercury in bivalves and gastropods may correlate with their different absorption mechanism and metabolism. Gastropods accumulated more mercury than bivalves, and to protect themselves from hazards, there might be an automatic biotransformation process, in which methylmercury was converted into less toxic inorganic forms (*10*−*11*).

The different ratios among different sampling sites might correspond to the local methylation conditions in seawaters and sediments, such as temperature, salinity, pH, content of organic matters, numbers, and species of bacteria, which might lead to the different conversion percentage from inorganic mercury to methylmercury.

The contamination levels of methylmercury and total mercury in 13 species of mollusks and gastropods sampled from eight coastal sites along the Bohai Sea were investigated. Mercury were detected in all samples, and gastropods showed more capacity to bioaccumulate mercury than bivalves. The results also indicated that mercury contents in two gastropods presented uplifted trends with the increasing of the shells' dimensions. Due to the Hg drainage from the chemical industries in the adjacent area, mollusks collected from HuLuDao were obviously the most severely polluted samples. Comparing with the Hg contents in *Rapana venosa* in seven sampling sites, we found that *Rapana venosa* could not regulate Hg contents in its body and might be used as a potential biomarker to monitor Hg pollution in oceans.

ACKNOWLEDGMENT

We acknowledge Professor F. C. Adams for his kindly offering CRM DORM-2.

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Received for review February 27, 2003. Revised manuscript received September 25, 2003. Accepted September 30, 2003. This work was jointly supported by National Natural Science Foundation of China-(20137010, 20205008) and Chinese Academy of Sciences (KZCX2-414).

JF034199L