

1 **Supporting Information**

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3 **Biomagnification of mercury in mollusks from coastal
4 areas of the Chinese Bohai Sea**

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24 **Analysis of Hg.** For THg analysis, approximately 0.05-0.1 g of the mollusk tissue was
25 weighed into a nickel boat, completely covered with an appropriate amount of diatomite and
26 directly detected using the Hydra-C mercury analyzer (Teledyne Leeman Labs, USA)
27 following USEPA method 7473.

28 For MeHg analysis, KOH/CH₃OH solution was used for MeHg extraction. Approximately
29 0.1 g of soft tissue was transferred into a 8-mL brown glass container and extracted with 4 mL
30 of 25% (m/v) KOH/CH₃OH solution by shaking for 4 h at 37 °C and 240 rpm with a shaker.
31 The digested solution was then diluted with CH₃OH to the appropriate concentration for
32 determination. The concentrations of MeHg were determined with a MERX Automatic
33 Methylmercury System (Brooks Rand Laboratories, Seattle, WA) following USEPA method
34 1630.

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36 **Table S1. Lipid Content, Trophic Level (TL), THg and MeHg Concentrations of**37 **Mollusks Collected from Bohai Sea in 2009 and 2012 (Mean \pm SD)**

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Species	Class	Order	Lipid content		TL	THg ($\mu\text{g kg}^{-1}$ d.w.)	MeHg ($\mu\text{g kg}^{-1}$ d.w.)
			N	(% d.w.)			
Amu	Bivalvia	Pterioida	9	11.01 \pm 3.05	15	3.13 \pm 0.16	70.0 \pm 29.7
Chl	Bivalvia	Pterioida	7	14.03 \pm 2.55	16	3.07 \pm 0.22	104.0 \pm 53.2
Cyc	Bivalvia	Veneroida	7	7.51 \pm 1.99	14	3.02 \pm 0.25	56.8 \pm 21.1
Mac	Bivalvia	Veneroida	4	7.93 \pm 0.64	8	2.95 \pm 0.54	67.6 \pm 51.9
Mer	Bivalvia	Veneroida	9	13.55 \pm 1.80	20	2.61 \pm 0.74	46.5 \pm 13.2
Mya	Bivalvia	Myoida	6	11.34 \pm 0.89	8	2.74 \pm 0.29	83.9 \pm 45.8
Myt	Bivalvia	Mytioida	4	15.50 \pm 0.53	10	2.72 \pm 0.35	79.9 \pm 45.5
Ost	Bivalvia	Veneroida	7	15.71 \pm 2.22	10	2.97 \pm 0.51	96.2 \pm 63.7
Sca	Bivalvia	Arcoida	8	13.21 \pm 4.54	17	3.08 \pm 0.49	72.8 \pm 43.0
Nev	Prosobranchia	Gastropoda	10	6.18 \pm 1.72	18	3.28 \pm 0.19	107.4 \pm 57.4
Rap	Prosobranchia	Gastropoda	21	6.99 \pm 1.76	39	3.33 \pm 0.28	130.7 \pm 64.5
Total			92	10.41 \pm 4.05	175	3.05 \pm 0.45	89.6 \pm 55.7
							43.8 \pm 46.4

39 Amu, *Amusium veneriformis*; Chl, *Chlamys farreri*; Cyc, *Cyclina sinensis*; Mac, *Mactra*
 40 *veneriformis*; Mer, *Meretrix meretrix*; Mya, *Mya arenaria*; Myt, *Mytilus edulis*; Ost,
 41 *Crassostrea talienwhanensis*; Sca, *Scapharca subcrenata*; Nev, *Neverita didyma*; Rap,
 42 *Rapana venosa*.

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44 Table S2. Results for the Analysis of Certified Reference Materials (CRMs) ($\mu\text{g kg}^{-1}$)

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CRM	Source	Description	N	THg		MeHg	
				Obtained value	Certified value	Obtained value	Certified value
TORT-2	NRCC ¹	Lobster Hepatopancreas	4	269.2±13.2	270±60	126.9±5.3	152±13
DORM-3	NRCC	Fish Protein	5	369.4±14.9	382±60	329.6±17.9	355±56
DORM-4	NRCC	Fish Protein	4	394.2±6.1	410±55	339.5±10.4	354±31

46 ¹NRCC—National Research Council Canada

48 **Table S3. Concentrations of THg and MeHg and %MeHg in the Mollusk Samples from
49 Bohai Sea during 2007-2012**

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Mollusks	N	THg ($\mu\text{g kg}^{-1}$)				MeHg ($\mu\text{g kg}^{-1}$)				%MeHg			
		Mean	Median	Min	Max	Mean	Median	Min	Max	Mean	Median	Min	Max
Amu	38	89.7	89.7	44.6	226.4	35.8	30.1	9.1	121.2	40.1	36.9	8.3	92.2
Chl	33	108.9	93.7	35.9	244.4	38.7	34.4	9.6	90.6	38.3	37.1	11.9	70.7
Cyc	32	62.9	59.4	32.0	132.6	20.3	19.2	5.5	57.0	33.8	32.8	8.8	74.9
Mac	25	86.8	61.9	35.7	203.7	32.6	16.9	5.5	169.8	35.3	25.7	11.6	92.6
Mer	56	59.4	54.1	27.2	140.5	22.3	17.4	3.5	59.7	37.6	29.7	6.6	94.2
Mya	16	96.4	81.6	42.0	217.7	34.1	25.6	7.4	95.4	32.5	32.9	16.3	48.4
Myt	30	100.8	80.9	27.7	274.5	20.0	14.4	2.6	64.4	21.3	20.0	4.2	59.1
Ost	40	105.8	103.5	32.4	242.3	46.1	42.7	12.7	97.5	49.5	42.8	12.7	92.5
Sca	43	88.1	76.4	36.9	211.8	19.2	15.3	2.1	104.5	21.1	17.2	5.7	57.0
Nev	35	91.4	79.6	36.5	261.2	50.2	44.1	12.4	138.7	57.3	59.4	15.6	95.1
Rap	83	151.1	125.2	53.9	461.1	102.6	83.6	14.3	295.5	65.8	68.7	21.7	95.8
Total	431	99.4	80.9	27.2	461.1	45.1	29.4	2.1	295.5	42.6	37.2	4.2	95.8

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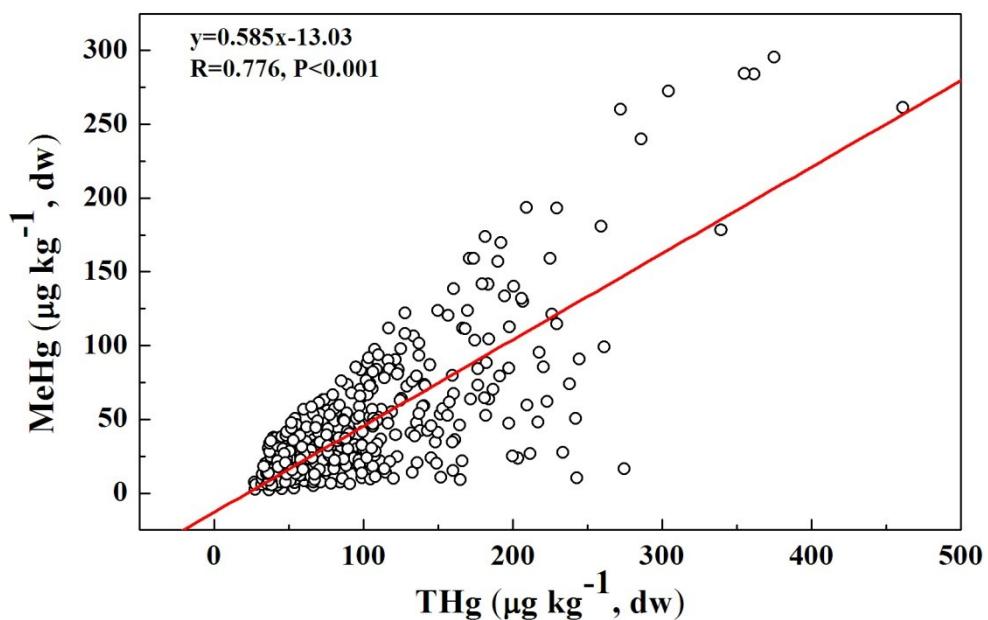
53 **Table S4. Concentrations of Hg species ($\mu\text{g kg}^{-1}$) in Mollusks from Different Areas in the**
 54 **World**

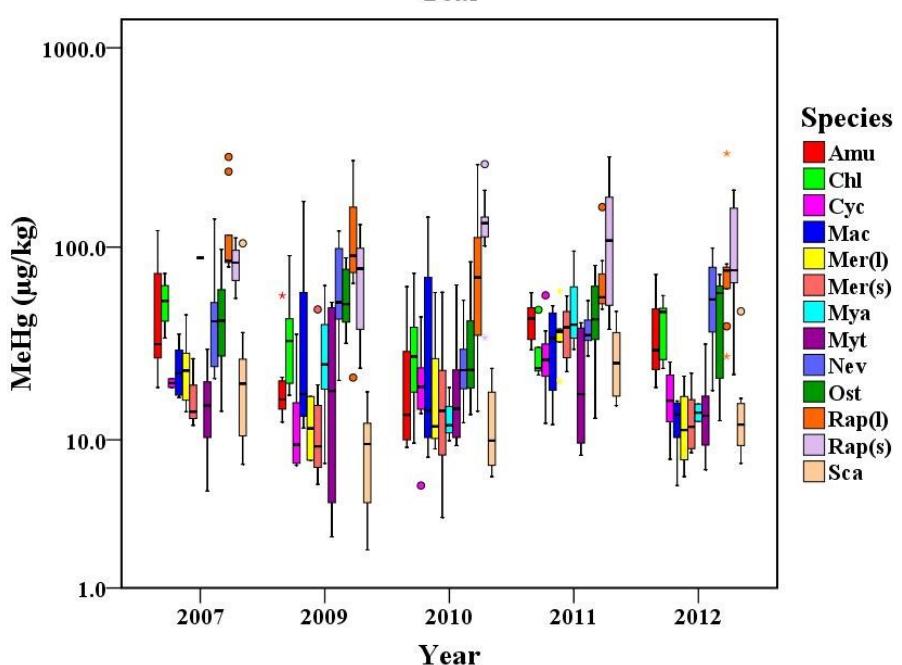
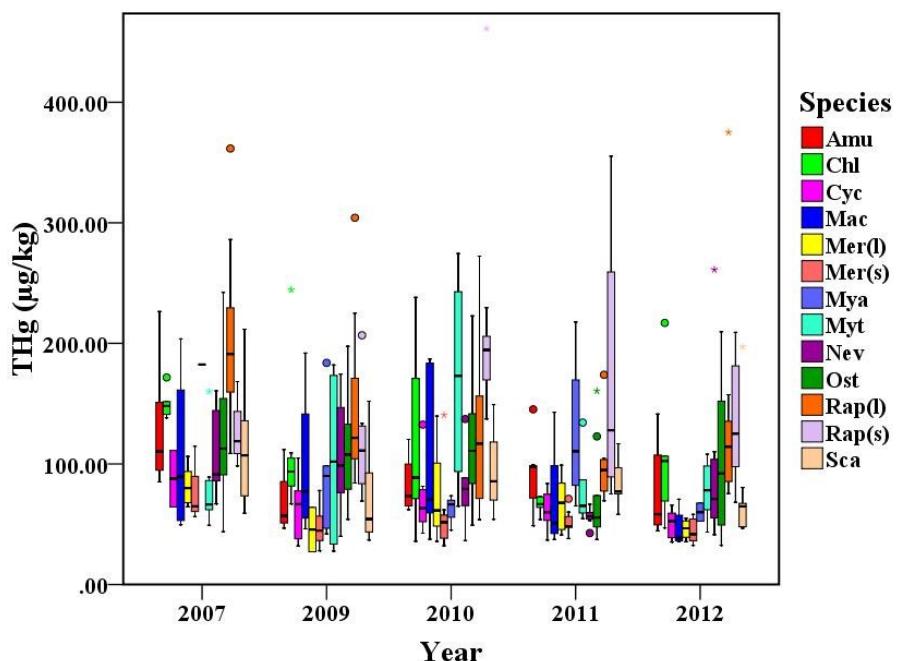
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Species	Location		THg	MeHg	Reference
Zebra mussel	Lakes in the USA	d.w.	529.0±78.0		1
Oyster	Te'rminos Lagoon, Mexico	d.w.	880.0(400.0-2000.0)		2
Oyster	Coastal lagoons, SE Gulf of California, Mexico	d.w.	700.0(60.0-910.0)		3
Oyster	Gulf of Mexico	d.w.	30.0-500.0	10.0-200.0	4
Mussel/Oyster	French Coast	d.w.	147.0±62.0 (39.0-437.0)	64.0±35.0 (8.0-238.0)	5
Mollusk	Gulf of Taranto, Italy	d.w.	626.0(21.0-1403.0) 1019.0(224.0-1867.0)	176.4(n.d.-377.0) 524.4(37.0-1321.0)	6
Mangrove Oyster	North Coast of Villa Clara, Cuba	d.w.	190.0-690.0		7
Common mussel	Sepetiba Bay, Brazil	d.w.	76.0±7.1	48.0±5.1	8
Mangrove Oyster		d.w.	19.0±3.6	6.9±0.4	
Oyster	Ceara River, Brazil	d.w.	154.0±60.0 (56.0-300.0)		9
Oyster/Clam /Mussel	Todos os Santos Bay, Bahia, Brazil	d.w.	<30.0-350.0		10
Mollusk	Kastela Bay, Adriatic Sea	d.w.	138.0-325.0	30.3-59.3	11
Mollusk	Mar Piccolo, Ionian Sea	d.w.	200.0(110.0-290.0)		12
Mussel	Southern Baltic Sea	d.w.	110.0±10.0		13
Cupped oyster		w.w.	32.0(5.0-90.0)	13.0(2.0-36.0)	
European flat oyster	Mediterranean Sea	w.w.	30.0(5.0-470.0)	12.0(2.0-188.0)	14
Mediterranean mussel		.	40.0(20.0-60.0)	16.0(8.0-24.0)	
Blue mussel	West Greenland	d.w.	60.0±30.0	11.0±2.0	15
Mussel	Krka Estuary, Croatia	w.w.	14.0-33.0	3.8-9.1	16
Bivalve	Turkey	w.w.	<LOD-120.0		17
Rocky oyster	Coast of Qeshm Island, Persian Gulf, Iran	d.w.	50.7		18
Blue Mussel	Masan Bay, Korea	d.w.	20.0±7.0	8.8±5.4	19
Washington clam		d.w.	34.0±4.5	16.0±3.5	
Mollusk	Markets throughout Korea	d.w.	51.0		20
Mollusk	Coastal area of Guangdong, China	w.w.	13.9±9.3 (1.8-53.3)		21
Marine bivalve	East Coasts of Hong Kong, China	d.w.	30.2-91.9	8.9-18.8	22

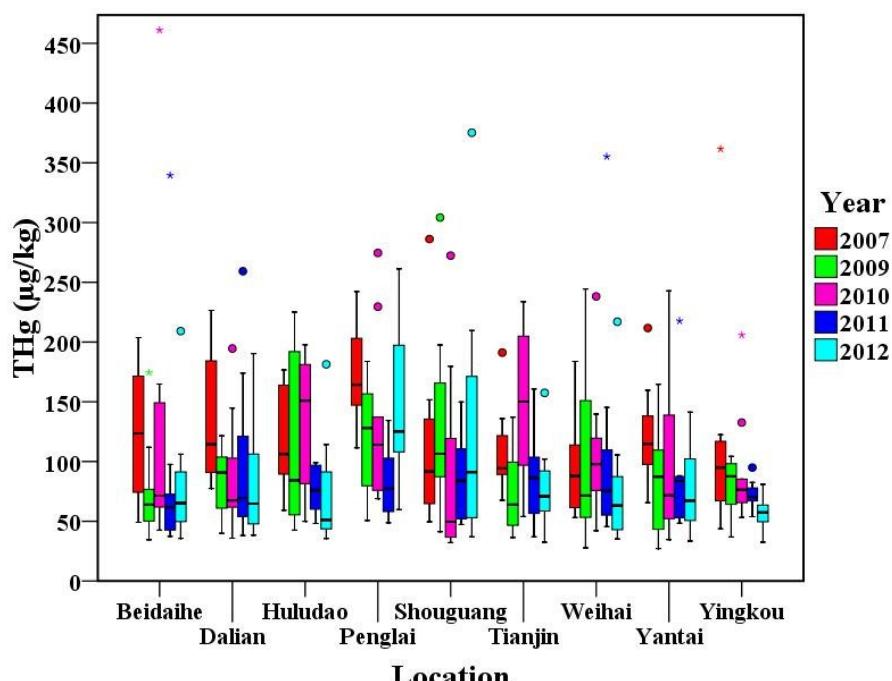
Mussel/Clam/Oyster	Bohai Sea, China	W.W .	6.7-453.0	4.8-168.4	23
Marine bivalve	Dalian City, China	d.w.	70.0-350.0		24
Mollusk	Bohai Sea, China	d.w.	99.4(27.2-461.1)	45.1(2.1-295.5)	This work

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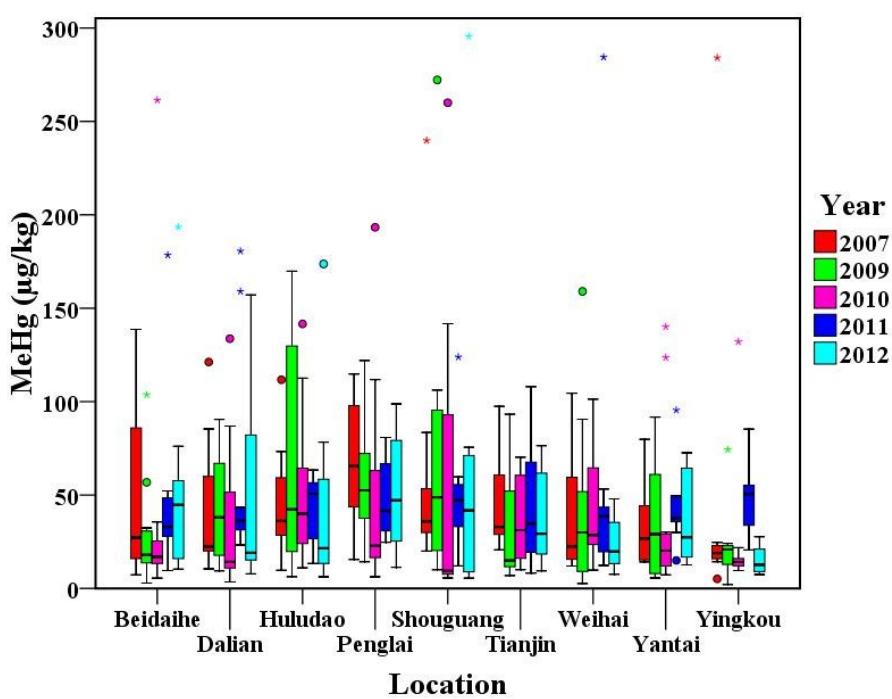




70 **Figure S2.** Boxplots of THg and MeHg in mollusks collected from the year 2007 to 2012.



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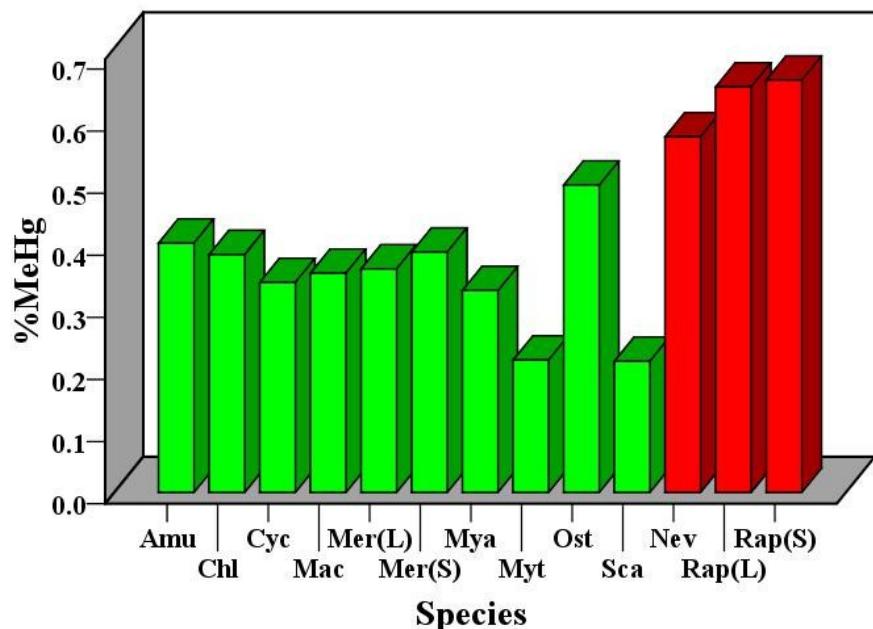
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77 **Figure S3.** Boxplots of THg and MeHg in mollusks from the nine coastal areas along Bohai

78 Sea.

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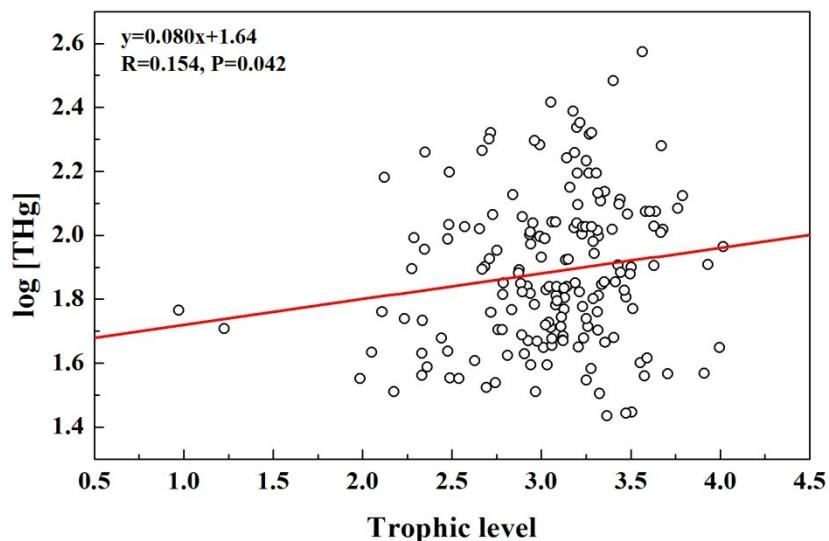


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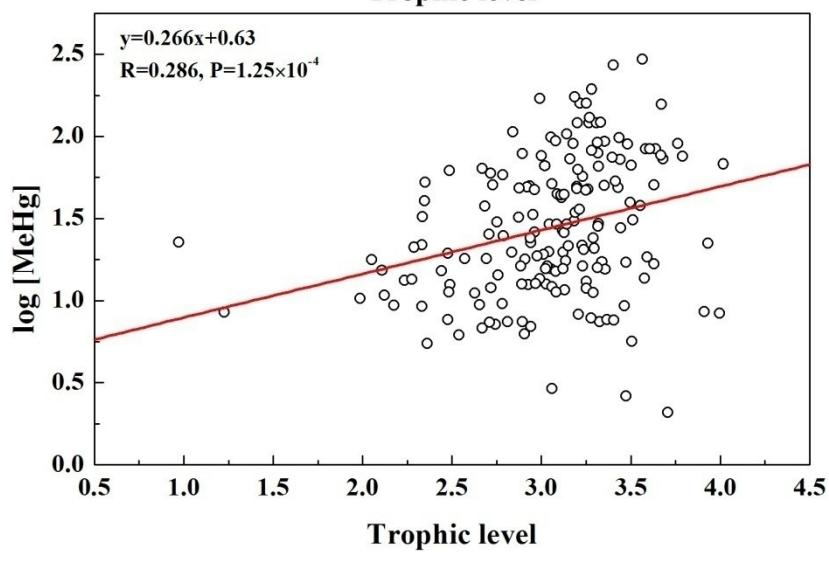
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83 **Figure S4.** Percentages of MeHg in THg in 11 mollusk species.

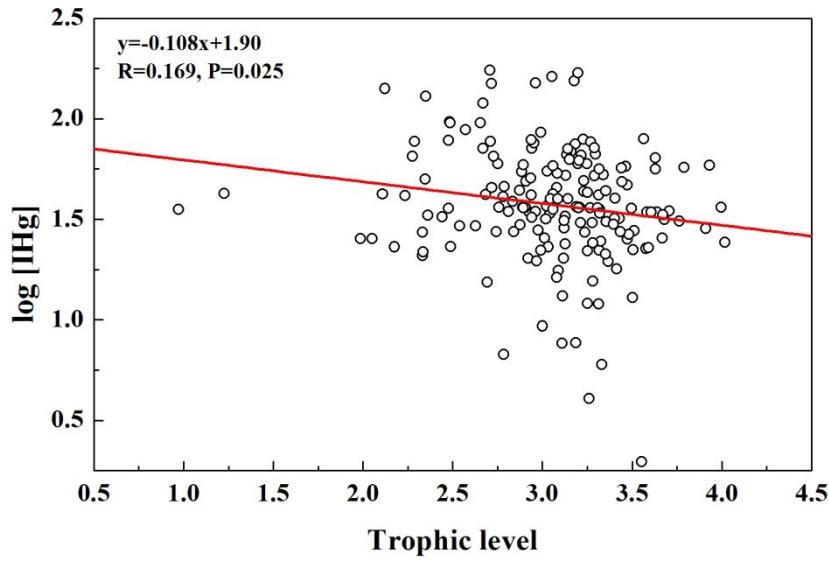
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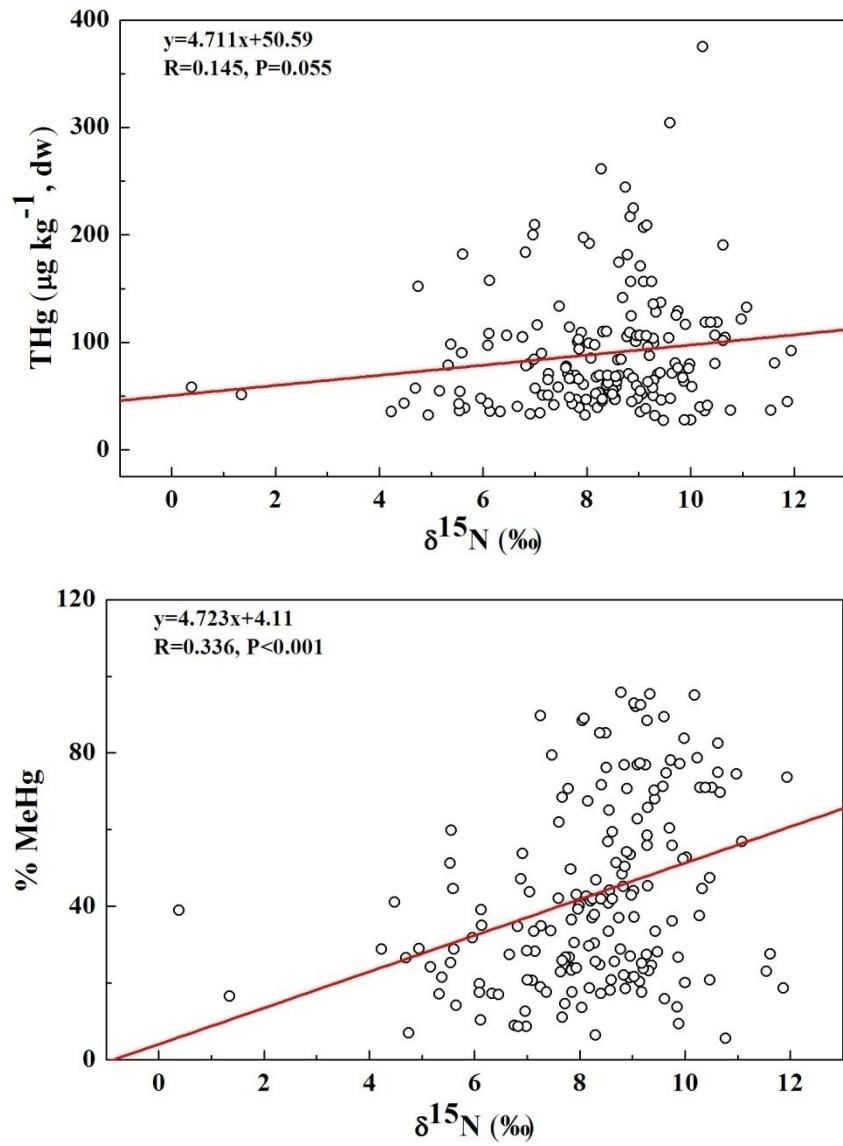


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88 **Figure S5.** Relations between $\log[\text{Hg}]$ and trophic level (TL).

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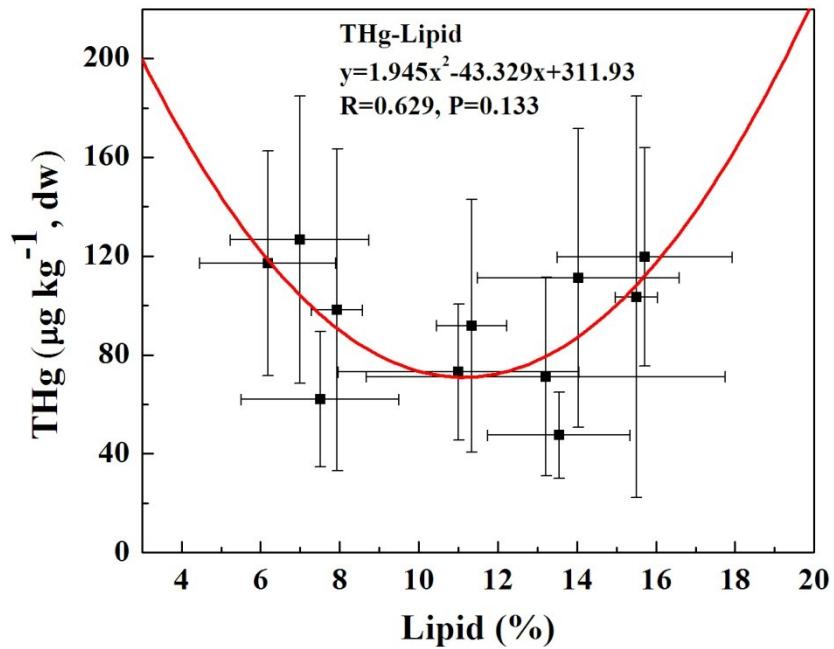
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95 **Figure S6.** Relations between concentrations of THg and %MeHg and $\delta^{15}\text{N}$.



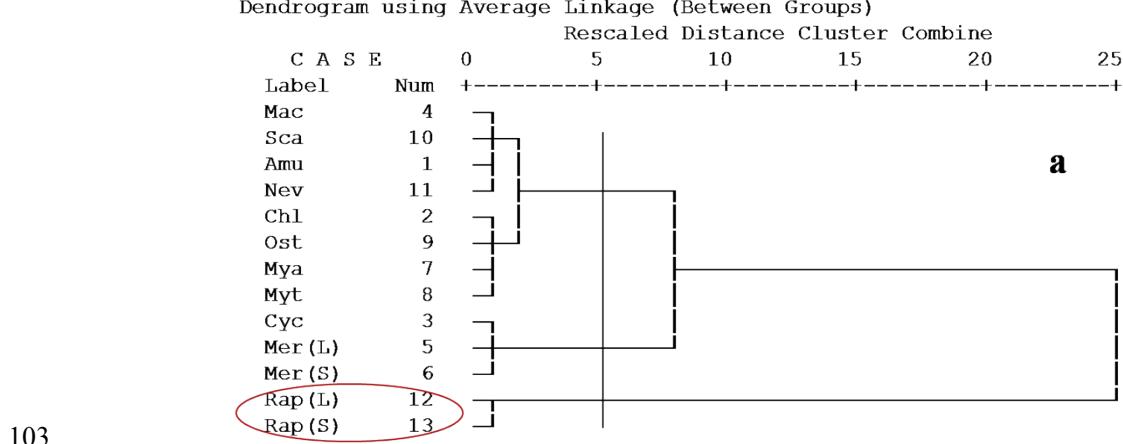
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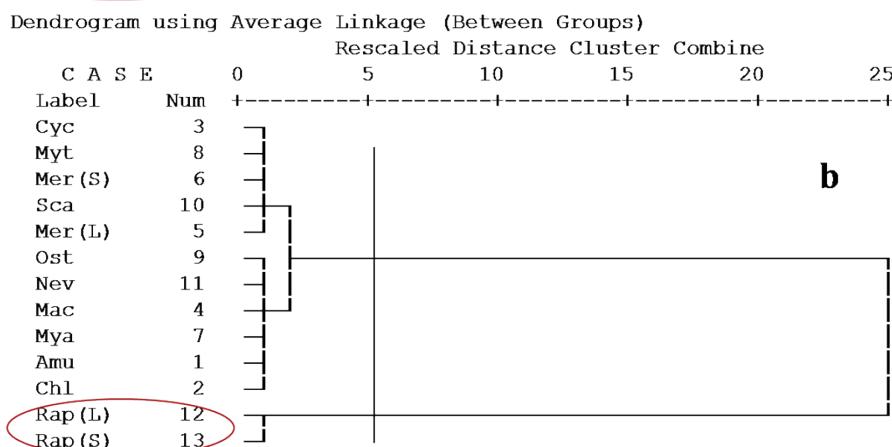
101 **Figure S7.** Relations between concentrations of THg and lipid content.



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108 **Figure S8.** Dendrogram derived from the hierarchical cluster analysis by using the data
109 matrix of THg (a) and MeHg (b) in mollusks.

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References

- 113 1 B. D. Blackwell, C. T. Driscoll, M. E. Spada, S. G. Todorova, M. R. Montesdeoca, *Environ. Toxicol. Chem.*, 2013, **32** (3), 638-643.
- 114 2 C. A. Aguilar, C. Montalvo, L. Rodríguez, J. G. Cerón, R. M. Cerón, *Int. J. Environ. Sci. Technol.*, 2012, **9**, 579-586.
- 115 3 C. C. Osuna-Martínez, F. Páez-Osuna, R. Alonso-Rodríguez, *Bull. Environ. Contam. Toxicol.*, 2010, **85**, 339-343.
- 116 4 D. A. Apeti, G. G. Lauenstein, D. W. Evans, *Mar. Pollut. Bull.*, 2012, **64**, 2399-2408.
- 117 5 D. Claisse, D. Cossa, J. Bretaudeau-Sanjuan, G. Touchard, B. Bomblé, *Mar. Pollut. Bull.*, 2001, **42**, 329-332.
- 118 6 L. Spada, C. Annicchiarico, N. Cardellicchio, S. Giandomenico, A. Di Leo, *Int. J. Hyg. Environ. Health.*, 2012, **215**, 418-426.
- 119 7 S. Olivares-Rieumont, L. Lima, S. Rivero, D. W. Graham, C. Alonso-Hernandez, Y. Bolaño, *Bull. Environ. Contam. Toxicol.*, 2012, **88**, 589-593.
- 120 8 H. A. Kehrig, M. Costa, I. Moreira, O. Malm, *J. Braz. Chem. Soc.*, 2006, **17**, 1409-1418.
- 121 9 A. Vaisman, R. Marins, L. Lacerda, *Bull. Environ. Contam. Toxicol.*, 2005, **74**, 582-588.
- 122 10 M. M. Souza, C. C. Windmöller, V. Hatje, *Mar. Pollut. Bull.*, 2011, **62**, 2254-2263.
- 123 11 Z. Kljaković-Gašpić, N. Odžak, I. Ujević, T. Zvonarić, M. Horvat, A. Barić, *Sci. Total Environ.*, 2006, **368**, 199-209.
- 124 12 N. Cardellicchio, C. Annicchiarico, G. Assennato, M. Blonda, A. Di Leo, S. Giandomenico, L. Lopez, L. Spada, N. Ungaro, *Fresenius Environ. Bull.*, 2010, **19** (9), 1841-1847.
- 125 13 H. Dabrowska, O. Kopko, R. Turja, K. K. Lehtonen, A. Góra, L. Polak-Juszczak, J. Warzocha, S. Kholodkevich, *Mar. Environ. Res.*, 2013, **84**, 1-9.
- 126 14 G. Brambilla, M. C. Abete, G. Binato, E. Chiaravalle, M. Cossu, E. Dellatte, R. Miniero, R. Orletti, P. Piras, A. Roncarati, A. Ubaldi, G. Chessa, *Regul. Toxicol. Pharmacol.*, 2013, **65**, 269-277.
- 127 15 F. Rigét, P. Møller, R. Dietz, T. G. Nielsen, G. Asmund, J. Strand, M. M. Larsen, K. A. Hobson, *J. Environ. Monit.*, 2007, **9**, 877-883.
- 128 16 N. Mikac, Ž. Kwokal, D. Martinčić, M. Branica, *Sci. Total Environ.*, 1996, **184**, 173-182.
- 129 17 S. Colakoglu, G. Ulukoy, H. B. Ormancı, F. A. Colakoglu, *Food Addit. Contam: Part B.*, 2012, **5** (4),

- 141 272-278.
- 142 18 G. Shirneshan, A. R. Bakhtiari, A. Kazemi, M. Mohamadi, N. Kheirabadi, *Bull. Environ. Contam. Toxicol.*, 2012, **88**, 962-966.
- 143 19 E. Kim, H. Kim, K. Shin, M. Kim, S. R. Kundu, B. Lee, S. Han, *Environ. Toxicol. Chem.*, 2012, **31** (6), 1254-1263.
- 144 20 J. H. Kim, J. Y. Lee, J. E. Seo, J. Y. Jeong, K. K. Jung, H. J. Yoon, K. S. Park, *Food Addit. Contam: Part B.*, 2012, **5** (4), 260-264.
- 145 21 P. Li, X. B. Feng, P. Liang, H. M. Chan, H. Y. Yan, L. G. Chen, *Environ. Toxicol. Chem.*, 2013, **32** (3), 541-547.
- 146 22 K. Pan, W. X. Wang, *Environ. Pollut.*, 2011, **159**, 2500-2506.
- 147 23 L. N. Liang, J. B. Shi, B. He, G. B. Jiang, C. G. Yuan, *J. Agric. Food Chem.*, 2003, **51**, 7373-7378.
- 148 24 L. Q. Zhao, F. Yang, X. W. Yan, *Hum. Ecol. Risk Assess.*, 2013, **19**, 145-150.