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Analysis of biomagnification of persistent organic pollutants in the aquatic food web of the Mekong Delta, South Vietnam using stable carbon and nitrogen isotopes

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Abstract

The present study elucidated the biomagnification profiles of persistent organic pollutants (POPs) through a tropical aquatic food web of Vietnam based on trophic characterization using stable nitrogen analysis. Various biological samples collected from the main stream of the Mekong Delta were provided for the analysis for both POPs, and stable nitrogen and carbon isotope ratios. Of the POPs analyzed, dichlorodiphenyltrichloroethane and its metabolites (DDTs) were the predominant contaminants with concentrations ranging from 0.058 to 12 ng/g wet weight, followed by polychlorinated biphenyls (PCBs) at 0.017-8.9 ng/g, chlordane compounds (CHLs) at 0.0043-0.76 ng/g, tris-4-chlorophenyl methane (TCPMe) at N.D.-0.26 ng/g, hexachlorocyclohexane isomers (HCHs) at N.D.-0.20 ng/g and hexachlorobenzene (HCB) at 0.0021-0.096 ng/g. Significant positive increases of concentrations in DDTs, CHLs, and TCPMe against the stable nitrogen ratio ($\delta^{15}N$) were detected, while, concentrations of HCHs and HCB showed no significant increase. The slopes of the regression equations between the log-transformed concentrations of these POPs and $\delta^{15}N$ were used as indices of biomagnification. The slopes of the POPs for which positive biomagnification was detected ranged from 0.149 to 0.177 on a wet weight basis. The slopes of DDTs and CHLs were less than those reported for a marine food web of the Arctic Ocean, indicating that less biomagnification had occurred in the tropical food web. Of the isomers of CHLs, unlike the studies of the Arctic Ocean, oxychlordane did not undergo significant biomagnification through the food web of the Mekong Delta. This difference is considered to be due to a lack of marine mammals, which might metabolize cis- and trans-chlordane to oxychlordane, in the Mekong Delta ecosystem. The biomagnification profile of TCPMe is reported for the first time in the present study. © 2008 Elsevier Ltd. All rights reserved.

Keywords: Biomagnification; Food web; Mekong Delta; Persistent organic pollutants; Stable nitrogen isotope

1. Introduction

Estimation of contaminant biomagnification through a food web has been traditionally based on comparisons of tissue contaminant levels obtained from laboratory experiments on members of particular trophic levels with pub-

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lished aquatic food web models, and data on feeding behavior or stomach contents (Suedel et al., 1994). However, the modeling of biomagnification profiles of anthropogenic chemicals using stable isotope ratios of bio-elements such as carbon and nitrogen has rapidly advanced in this decade (Kidd et al., 1995; Hoekstra et al., 2003; Muir et al., 2003). In general, ¹⁵N/¹⁴N (δ^{15} N) is reported to increase 3–5‰ on average per trophic level (DeNiro and Epstein, 1981; Minagawa and Wada, 1984; Hobson and Welch, 1992; Hobson et al., 2002), while

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 δ^{13} C is reported to increase about only 1‰ per trophic level (DeNiro and Epstein, 1978; Peterson et al., 1985; Peterson and Fry. 1987). Thus, the measurement of the $\delta^{15}N$ is useful to elucidate the trophic positions of organisms in the food web. Consequently, the measurement of $\delta^{15}N$ associated with trace metals or anthropogenic chemicals in the same samples leads to the estimation of the biomagnification of these trace elements and anthropogenic chemicals through marine and freshwater food webs. However, most published studies on the biomagnification profiles of persistent organic pollutants (POPs) using $\delta^{15}N$ have been conducted in the temperate to boreal regions of the Northern Hemisphere. Analysis of the biomagnification profiles of POPs in tropical aquatic food webs has seldom been conducted except for DDTs in the freshwater ecosystem of Malawi, East Africa (Kidd et al., 2001).

The Mekong Delta and its adjacent areas contribute greatly to Vietnamese agriculture and, recently, it became one of the most agriculturally productive areas in Southeast Asia. Can Tho City, which is situated in the middle of the Mekong Delta, has grown rapidly over the last decade with a population now of ca. one million. An increase in environmental contamination by various anthropogenic chemicals such as POPs and trace elements is anticipated in areas close to Can Tho City. In fact, a recent study conducted in the Mekong Delta revealed the presence of POPs including polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethane and its metabolites (DDTs) in sediment and biota (Iwata et al., 2004; Minh et al., 2006). Thus, understanding the trophic transfer of environmental contaminants in the biota of the Mekong Delta ecosystem is critical to evaluating the influence of these contaminants on the ecosystems and human life. Recently, Ikemoto et al. (2008) reported the biomagnification profiles of 21 trace elements through the aquatic food web of the Mekong Delta near Can Tho City. Of these trace elements, a significant trophic level-dependent increase with increasing δ^{15} N was found in concentrations of only three elements, i.e., Se, Rb and Hg.

In the present study, the trophic transfer of several POPs, i.e., PCBs, DDTs, hexachlorocyclohexane isomers (HCHs), hexachlorobenzene (HCB), chlordane compounds (CHLs), and tris-4-chlorophenyl methane (TCPMe), was elucidated using stable isotope analysis of biota samples collected from the ecosystem of the main stream of the Mekong Delta near Can Tho City in South Vietnam.

2. Materials and methods

2.1. Sample collection

Sampling was conducted in the Mekong Delta near Can Tho City, South Vietnam (Fig. 1). Because baseline δ^{15} N values may vary from ecosystem to ecosystem, owing to differences in anthropogenic nutrient inputs (Cabana and Rasmussen, 1996; Cole et al., 2004), it was crucial that all analyzed biota were collected from one ecosystem. Therefore, all sampling was conducted in the main stream of the Mekong Delta near Can Tho City (45°10'N, 141°15'E) on April 23 and 24, 2004. Fish and crustacean samples were purchased directly from a local fishing boat that was operating in the main stream of the Mekong Delta at the time. Phytoplankton was collected by horizontal tow of a NORPAC plankton net (0.10 mm in mesh size) using a



Fig. 1. Sampling location of organisms in the Mekong Delta, South Vietnam.

small boat. In the laboratory, organisms collected by the plankton net were divided into phytoplankton (those at the surface) and other material. These samples were kept frozen at -20 °C until dissection and chemical analysis. The biological samples collected for the present study were also used in the analysis of the biomagnification of trace elements (Ikemoto et al., 2008).

2.2. Stable carbon and nitrogen isotope analysis

Stable carbon and nitrogen isotope ratios were determined using the procedure described in Okuda et al. (2004). Several 10 mg samples of muscle tissues of fish and crustaceans and aggregations of phytoplanton were used as the stable isotope samples. All samples were dried for 24 h at 60 °C, and ground to a powder and then immersed in a chloroform:methanol (2:1) solution for 24 h to remove lipids. The ratios of stable carbon and nitrogen isotopes were analyzed using a gas chromatography-combustion-isotope ratio mass spectrometer (GC-C-IRMS) (PDZ Europa Ltd. ANCA-SL), and presented as per thousand deviations from the standards, calculated as δ^{13} C and δ^{15} N by the following equation: $\delta X(\%) = (R_{\text{sample}}/R_{\text{standard}}-1) \times 1000$, where X is ^{13}C (or ^{15}N) and R is the corresponding ratio $^{13}C/^{12}C$ (or ¹⁵N/¹⁴N). Pee Dee belemnite (PDB) limestone carbonate and atmospheric nitrogen (N_2) were used as the standards for carbon and nitrogen isotope ratios, respectively.

2.3. POPs analysis

POPs including PCBs (60 major congeners, i.e., PCB #4 (with #10), #8, #19, #15 (with #18), #22, #28, #33, # 37, #44, #49, #52, #54, #70, #74, #77, #81, #87, #95, #99, #101, #104, #105, #110, #114, #118 (with #149), #119, #123, #126, #138, #151, #153, #155, #156 (with #171), #157 (with #202), #158, #167, #168, #169, #170, #177. #178, #180, #183, #187 (with #128), #188, #191, #194, #199, #201, #189, #205, #206, #208 and #209), \(\sum DDT\) $(p,p'-DDT, p,p'-DDD \text{ and } p,p'-DDE), \sum CHL (trans-chlor$ dane, cis-chlordane, trans-nonachlor, cis-nonachlor and oxychlordane), \sum HCH (α -HCH, β -HCH and γ -HCH), HCB and TCPMe were analyzed following the method published in Kajiwara et al. (2003). Approximately 100 g of phytoplankton was lyophilized before extraction. The freeze-dried phytoplankton and about 10 g of whole homogenized crustaceans and fish were ground with anhydrous Na₂SO₄ and extracted in a Soxhlet apparatus with a mixture of diethyl ether and hexane for 7-8 h. An aliquot of extract was added to a gel permeation chromatography column (GPC; Bio-Beads S-X3, Bio-Rad Laboratories, CA, 2 cm internal diameter and 50 cm length) for lipid removal. Each extracted sample was concentrated and passed through an activated Florisil column for clean up and fractionation. Quantification of most of POPs was performed using a GC (Agilent 6890N) equipped with a micro-electron capture detector (ECD) and an auto-injection system (Agilent 7683 Series Injector). Identification

and quantification of TCPMe was performed using a GC with a mass selective detector (MSD: Agilent 5973 N). The concentration of individual POPs was quantified from the peak area from the sample that corresponded to an external standard. The PCB standard used for quantification was a mixture of 62 PCB isomers and congeners (BP-MS) obtained from Wellington Laboratories Inc., Ontario, Canada. Concentrations of individually resolved peaks of PCB isomer and congeners were summed to obtain total PCB concentrations.

Procedural blanks were analyzed simultaneously with every batch of five samples to check for interference or contamination from solvents and glassware. Lipid contents were determined by measuring the total non-volatile solvent-extractable material of sub-samples taken from the original extracts.

2.4. Statistical analyses

One-half of the respective limits of detection was substituted for those values below the limit of detection and used in statistical analyses. When >50% of the observations were below the detection limit, further, statistical analyses were not conducted. Single regression analysis was conducted between the stable isotopes values and POP concentrations. The slope of the regression between the log-transformed concentrations of POPs (on a wet weight, dry weight or lipid weight basis) and δ^{15} N and was used as the index of the trophic level-dependent accumulation of POPs through the food web. Food web magnification factors (FWMFs) have been used as indices of trophic level-dependent accumulation in the studies conducted on the Artic Ocean (Hoekstra et al., 2003). FWMFs correspond to "the slope" of the present study divided by 3.8, which is the difference of $\delta^{15}N$ resulting from the increase of one trophic level in an Arctic marine food web (based on Hobson et al. (2002)). However, as mentioned above, the trophic enrichment factors for δ^{15} N range from 3% to 5% (DeNiro and Epstein, 1981; Minagawa and Wada, 1984; Hobson and Welch, 1992; Hobson et al., 2002), indicating the difficulty of applying 3.8‰ as the trophic enrichment factor for δ^{15} N commonly used over the world. Thus, the slope of the regression equation between the log-transformed concentrations of POPs and δ^{15} N was used as the index of biomagnification in this study. A p value of less than 0.05 was considered to indicate statistical significance. These analyses were performed using programs of StatView (version 5.0, SAS Institute).

3. Results

3.1. Food web structure

The values of the stable carbon isotope ratio (δ^{13} C) and the stable nitrogen isotope ratio (δ^{15} N) in the biota ranged from -28% to -22% and 9% to 18%, respectively (Fig. 2). The "map" resulting from plotting δ^{15} N versus δ^{13} C had a triangular distribution, which indicated the presence of several other primary producers in addition to phytoplankton (δ^{13} C of -27%). The distributions of δ^{15} N values indicated that the food web of the main stream of the Mekong Delta consisted of three trophic trophic levels, although a clear 3–4‰ interval in the δ^{15} N distribution was not detected. Two species of decapod crustaceans, *Macrobrachium rosenbergii* and sp. 3, and two species of fish, *Parambassis wolffii* and *Glossogobius aureus*, were situated at the second production level assuming the increase of 3.4‰ per trophic level (Minagawa and Wada, 1984); one species of decapod crustacean, *M. equidens*, and one species of fish, *Puntioplites proctozysron*, were situated at the third production level; and two species of fish, *Cynoglossus* sp. 2 and *Polynemus paradiseus*, were situated at the third to fourth production level of the food web.

3.2. Concentrations of POPs in the biota

Phytoplankton, four species of crustaceans and six species of fish were used for contaminant analysis of POPs (Table 1). POPs were detected in all the samples of organisms collected from the Mekong Delta (Table 2). \sum DDT were the predominant contaminants with concentrations

ranging from 0.058 to 12 ng/g wet weight, followed by \sum PCB (0.017–8.9 ng/g), \sum CHL (0.0043–0.76 ng/g), TCPMe (N.D.–0.26 ng/g), \sum HCH (N.D.–0.20 ng/g), and HCB (0.0021–0.096 ng/g). Relatively, low concentrations of POPs were observed in phytoplankton except for HCB. Concentrations of POPs were not significantly different between crustaceans and fish (analysis of covariance (ANCOVA)).

The examination of the DDT composition revealed that p,p'-DDE, which is a metabolite of p,p'-DDT, was the predominant compound, accounting for 44–98% of the total DDT concentration (Fig. 3). The proportion of p,p'-DDT and p,p'-DDD was about half of the DDTs in the phytoplankton. The proportion of p,p'-DDT and p,p'-DDD in fishes was higher than it was in crustaceans, although no significant difference between the $\delta^{15}N$ values of these two groups of biota was found.

Among \sum CHL, *cis*-chlordane was the most abundant compound (38%) followed by *trans*-nonachlor, oxychlordane, and *cis*-nonachlor at ca. 20% (Fig. 4). In most crustaceans, except for *M. rosenbergii*, oxychlordane and *trans*-nonachlor predominated, being more than 90% of \sum CHL, while in *M. rosenbergii cis*-chlordane and



Fig. 2. Stable isotopes of the members of the aquatic food web in the Mekong Delta: $\delta^{15}N$ (%) versus $\delta^{13}C$ (%). Data for $\delta^{15}N$ and $\delta^{13}C$ were taken from Ikemoto et al. (2008). Bars indicate the standard deviation (SD). Open diamond, phytoplankton; grey square, fish; open circle, crustaceans.

Table 1

Sample size of organisms from the Mekong Delta, South Vietnam used for the present study

Species	n ^a	n ^b
Phytoplankton	6	1
Crustaceans		
Macrobrachium rosenbergii	5	2
Macrobrachium equidens	37	2 pools $(n = 3, n = 20)$
Macrobrachium sp. 3	6	1
Metapenaeus tenuis	7	1 pool ($n = 7$)
Fish		
Pisodonophis boro	6	2
Polynemus paradiseus	6	1 pool $(n = 4)$
Glossogobius aureus	3	1
Puntioplites proctozysron	4	1
Parambassis wolffii	1	1
Cynoglossus sp. 2	3	1 pool ($n = 9$)

^a Number of samples used in stable isotope analysis.

^b Number of samples used in organochlorine contaminants analysis.

trans-chlordane were detected. In all species of fishes, *trans*nonachlor was the most predominant of the CHL compounds, unlike in crustaceans, followed by *trans*-chlordane, *cis*-chlordane and *cis*-nonachlor. The proportions of oxychlordane were lower in fishes. No particular trend in the composition of CHLs against the increase of δ^{15} N values was detected.

Of three isomers of HCHs, only b-HCH was detected from most crustacean and fish samples (Fig. 5). Nevertheless, in phytoplankton, *M. rosenbergii* and *P. boro*, γ -HCH was the predominant HCH.

3.3. Biomagnification of POPs

 \sum DDT and the all *p*,*p*' isomers of DDTs, i.e., *p*,*p*'-DDT, *p*,*p*'-DDD, *p*,*p*'-DDE, showed a significant increase with

increasing δ^{15} N on wet weight and dry weight bases except for *p,p'*-DDD; the slopes of the regression equations were 0.116–0.141 (dry weight basis) and 0.152–0.177 (wet weight basis), respectively. The positive relationship between δ^{15} N and the concentration of *p,p'*-DDD (dry weight basis), however, was marginally significant (*p* = 0.072) (Table 3).

The concentration of \sum CHL indicated a significant increase of δ^{15} N on dry-, wet- and lipid-weight bases. Of the CHL compounds, a significant or almost significant increase was found for only *trans*-nonachlor and *cis*-nonachlor. The slopes of the regression equations for *trans*-nonachlor and *cis*-nonachlor were 0.139 and 0.140 (dry weight basis), 0.176 and 0.177 (wet weight basis) and 0.115 and 0.113 (p > 0.05) (lipid weight basis), respectively.

The concentration of TCPMe increased with increasing δ^{15} N on wet- and dry- and lipid-weight bases (Table 3, Fig. 6). The slope of the regression equation for TCPMe was 0.105–0.170. The concentration of PCBs showed an almost significant increase with increases of δ^{15} N, with a slope of 0.117 (dry weight basis) (p = 0.062) and 0.154 (wet weight basis) (p = 0.068), respectively.

None of HCHs, or its isomers, and HCB were significantly correlated with an increase of $\delta^{15}N$ (Table 3).

4. Discussion

Of the POPs analyzed in this study, significant positive increases of concentrations in DDTs, CHLs and TCPMe with an increase of δ^{15} N values through the Mekong Delta food web were detected, while no significant biomagnification of HCHs and HCB was detected. While PCB analysis indicated an increase with an increase in trophic level, a significant relationship was not found in this study.

An overall biomagnification of \sum DDT had occurred, with a slope of 0.152 on a wet weight basis. This indicates

Table 2

Concentrations of organochlorines (ng/g wet weight) in whole organisms of the Mekong Delta, South Vietnam

Species	n ^a	Lipid (%)	MC (%) ^b	∑PCB	∑DDT	∑CHL	∑HCH	HCB	TCPM
Phytoplankton	1	0.068	96.2	0.017	0.058	0.0043	0.00055	0.0021	N.D.
Crustaceans									
M. rosenbergii-1	1	3.9	70.3	0.53	1.7	0.10	0.010	0.032	0.019
M. rosenbergii-2	1	3.9	70.7	0.51	2.1	0.16	0.0050	0.010	0.019
M. equidens-1	3	1.9	71.4	2.5	11	0.23	0.019	0.039	0.072
M. equidens-2	20	2.4	73.2	3.4	6.0	0.21	N.D.	0.042	0.039
Macrobrachium sp. 3	1	3.5	74.9	3.0	7.8	0.18	0.011	0.070	0.049
M. tenuis	7	2.2	75.2	2.8	3.3	0.070	0.021	0.022	0.014
Fish									
P. boro-1	1	6.3	71.0	8.9	9.9	0.76	0.20	0.096	0.20
P. boro-2	1	2.1	77.3	8.6	2.6	0.51	0.13	0.032	0.26
P. paradiseus	4	1.2	78.8	3.2	8.7	0.23	0.0040	0.012	0.054
G. aureus	1	1.4	77.4	6.2	12	0.26	N.D.	0.035	0.054
P. proctozysron	1	3.8	74.7	2.6	12	0.25	0.012	0.049	0.042
P. wolffii	1	2.0	76.9	2.2	3.5	0.086	0.025	0.019	0.019
Cynoglossus sp. 2	9	1.6	79.5	2.7	9.2	0.27	0.0021	0.0064	0.067

The detection limit for \sum HCH and TCPMe was 0.014 ng/g wet weight and 0.015 ng/g wet weight, respectively.

^a Number of individuals homogenized.

^b Moisture content.



Fig. 3. Composition of DDT and its major metabolites in biota from the Mekong Delta, South Vietnam.

biomagnification of 3.3 times per trophic level, assuming that $\delta^{15}N$ increases 3.4% on average per trophic level (Minagawa and Wada, 1984). Significant biomagnification of \sum DDT through an aquatic food web has also been reported from the Barents Sea (Hop et al., 2002) and the southern Beaufort-Chukchi Seas, Arctica (Hoekstra et al., 2003), southeastern Norway (Ruus et al., 2002), the White Sea (Muir et al., 2003) in the boreal region, the Gulf of the Farallones, California in the temperate region (Jarman et al., 1996) and Malawi. East Africa in the tropical region (Kidd et al., 2001). Of the above studies conducted in the boreal regions of the Northern Hemisphere, that of Hoekstra et al. (2003) reviewed the food web magnification factors (FWMFs) of POPs in the ecosystem of Artic Ocean: p,p'-DDE and p,p'-DDT ranged 3.62–14.5 (corresponding to 13.76-55.1 of the slope) and 1.31-2.78 (corresponding to 4.97-10.56 of the slope), respectively. Ruus et al. (2002) also reported FWMFs of \sum DDT as 0.95 (corresponding to 3.61 of the slope). Compared with these data, the biomagnification of the Mekong Delta is estimated to be less than 1/10 for both p,p'-DDE and p,p'-DDT. Kidd et al. (2001) also reported regression equation slopes of 0.20 ± 0.03 for \sum DDT and 0.26 ± 0.03 for p,p'-DDE (with increases of $\delta^{15}N$) from Malawi, East Africa, which are closer to our results than those from Arctic regions. Thus, it is concluded that the degree of biomagnification of DDTs through the food web as determined by either $\sum DDT$ or the isomers in the tropical regions is much less than in the boreal regions.

Of the isomers of \sum CHL, which are organochlorine pesticides, *trans*-nonachlor and *cis*-nonachlor also underwent significant biomagnification through the Mekong Delta food web, while oxychlordane did not. Although Hoekstra et al. (2003) also reported a significant increase of \sum CHL through the marine Arctic food web lower FWMFs were found for *cis*-chlordane and *trans*-chlordane, such that the FWMF of oxychlordane (which is the metabolized compound of *cis*- chlordane and *trans*-chlordane) was more than 10 times that of *cis*-chlordane and *trans*chlordane. Their result of a higher biomagnification of oxychlordane was contrary to our results, which revealed no significant increase of oxychlordane concentrations with increases in trophic level.

One could consider that the difference in biomagnification between our study and that of Hoekstra et al. (2003) could be attributed to differences in the food web



Fig. 4. Composition of CHL compounds in biota from the Mekong Delta, South Vietnam. Oxy, oxychlordane; t-CA, trans-chlordane; c-CA, cis-chlordane; t-nona, trans-nonachlor; c-nona, cis-nonachlor.

components. The food web of the Arctic Ocean which Hoekstra et al. (2003) analyzed included seals (marine mammals) as the top predator, while the top predators of the Mekong Delta were fish, Cynoglossus sp. 2 and Polynemus paradiseus. Oxychlordane and heptachlor exo-epoxide have been reported to be the most abundant isomers of Σ CHL in all marine mammals (Hoekstra et al., 2003). Tashiro and Matsumura (1977) revealed that the major route of metabolism for both cis-chlordane and trans-chlordane is via dichlorochlordane and oxychlordane in rats. It is likely that the same mechanism exists in Arctic marine mammals, which is consistent with the higher accumulation of oxychlordane in marine mammals. If fish lacked the ability to metabolize both cis- and trans-chlordane, they would not accumulate high levels of oxychlordane, which explains the fact that no significant magnification of oxychlordane occurred through the food web of the Mekong Delta.

As far as we know, this is the first study to report that TCPMe undergoes positive biomagnification through a food web. Unlike the other POPs analyzed in the present study, TCPMe together with tris(4-chlorophenyl) methanol (TCPMOH) are two of the most recently detected environmental contaminants (Buser, 1995). These pesticide compounds, which have structures similar to DDT and dicofol, respectively, are thought to be derived mainly from technical DDT, dicofol and other agrochemicals. Minh et al. (2000) found that elevated residue levels of TCPMe and TCPMOH were observed in both offshore cetaceans of North Pacific Ocean and coastal ones of several Asian countries, suggesting widespread contamination of these compounds in the marine environment of the Indo-Pacific regions.

Although contrary to the above results for DDTs, CHLs and TCPMe, no significant positive biomagnification of \sum HCH and HCB through the Mekong Delta food web was detected. The concentration of \sum HCH was almost the same through the food web, while β -HCH showed an increasing trend. Similarly, no positive biomagnification of \sum HCH and HCB has been reported from southeastern Norway (Ruus et al., 2002). On the other hand, an increase of \sum HCH concentration was reported for the food web of California, but \sum HCH exhibited a lower bioaccumulation than did DDTs (Jarman et al., 1996). The same trends were found in the Arctic (Hoekstra et al., 2003). FWMFs of



Fig. 5. Composition of HCH isomers in biota from the Mekong Delta, South Vietnam.

Table 3 Statistics for the regression between the organochlorine contaminants and $\delta^{15}N$ values of the biota of the Mekong Delta, South Vietnam

Elements	п	Wet weight basis			Dry weight basis				Lipid weight basis				
		Slope	Intercept	r	<i>p</i> -Value	Slope	Intercept	r	p-Value	Slope	Intercept	r	<i>p</i> -value
$Log_{10} \sum DDT$	14	0.152	-1.488	0.576	0.031	0.116	-0.323	0.599	0.024	0.087	1.133	0.491	0.075
$Log_{10} p, p'-DDT$	14	0.177	-3.139	0.731	0.003	0.141	-1.974	0.638	0.014	0.114	-0.551	0.460	0.098
$Log_{10} p, p'-DDD$	14	0.174	-2.887	0.569	0.034	0.137	-1.722	0.495	0.072	0.109	-0.268	0.375	0.187
$Log_{10} p, p'-DDE$	14	0.160	-1.695	0.549	0.042	0.123	-0.530	0.576	0.031	0.096	0.899	0.517	0.058
$Log_{10} \Sigma HCH$	14	0.025	-2.231	0.083	0.779	-0.011	-1.066	0.044	0.880	-0.040	0.383	0.180	0.538
Log_{10} β -HCH	14	0.075	-3.088	0.221	0.449	0.038	-1.924	0.145	0.620	0.010	-0.474	0.048	0.871
$Log_{10} \sum CHL$	14	0.149	-2.852	0.647	0.012	0.112	-1.687	0.700	0.005	0.085	-0.257	0.613	0.020
Log ₁₀ oxychlordane	14	0.077	-2.698	0.303	0.293	0.040	-1.533	0.218	0.453	0.011	-0.079	0.069	0.816
Log ₁₀ cis-chlordane	14	0.109	-3.256	0.445	0.111	0.073	-2.092	0.331	0.248	0.044	-0.642	0.191	0.514
Log ₁₀ trans-nonachlor	14	0.177	-3.601	0.662	0.010	0.140	-2.436	0.698	0.006	0.115	-1.029	0.631	0.016
Log ₁₀ cis-nonachlor	14	0.176	-4.232	0.603	0.023	0.139	-3.067	0.53	0.051	0.113	-1.657	0.427	0.128
Log ₁₀ HCB	14	0.038	-2.152	0.196	0.503	0.001	-0.987	0.006	0.983	-0.028	0.463	0.215	0.460
Log ₁₀ TCPMe	14	0.170	-3.795	0.644	0.013	0.133	-2.630	0.661	0.010	0.105	-1.193	0.576	0.031
$Log_{10} \sum PCB$	14	0.154	-1.839	0.510	0.062	0.117	-0.674	0.501	0.068	0.088	0.781	0.420	0.135

Slops with the significant difference (p < 0.05) are indicated in bold.

α-HCH, β-HCH, γ-HCH through the Arctic food web ranged from 1.19 to 2.19 (corresponding to 4.52–8.32 of the slope), 0.61 to 4.21 (corresponding to 2.32–16.00 of the slope), 0.62 to 1.68 (corresponding to 2.36–6.34 of the slope), respectively, all which indicate higher biomagnification than the present study. The biomagnification of HCB was also reported to be lower than that of DDTs in Jarman et al. (1996) and Hoekstra et al. (2003): FWMFs of HCB



Fig. 6. Relationships between log-transformed organochlorine contaminants (ng/g wet weight) and $\delta^{15}N$ in biota from the Mekong Delta, South Vietnam.

ranged from 1.36 to 2.27 (corresponding to 5.17–8.63 of the slope), which are higher than those of the present study. These studies suggest that \sum HCH and HCB exhibit the same geographic trend of decreasing biomagnification in the tropical regions as seen in \sum DDT. However, further studies on ecosystems with high accumulations of \sum HCH and HCB in various geographical areas, including tropical ecosystems, are necessary, since in addition to the report of Ruus et al. (2002) of no significant accumulation, the concentration of \sum HCH in the present study was close to the detection limit.

The log-transformed octanol-water partition coefficient (log Kow) is considered to be one of the most useful indices for estimating biomagnification through a food web. In general, a chemical with a log Kow of more than 5–6 is considered to undergo biomagnification through a food

web. Suedel et al. (1994), in their review, found that organic compounds with log Kow values <5.0 showed little potential for food-chain biomagnification, and organic compounds with log Kow values between 5 and 7 showed the greatest potential for biomagnification.

The slopes of the regression equations, which indicate the degree of biomagnification through a food web, showed a high correlation with an increase of log Kow (Fig. 7). The slopes of the regression equations for these POPs, which underwent significant increase, ranged from 0.149 to 0.177 (wet weight basis). These slopes are greater than those of the three trace elements that exhibit significant increases with increases of trophic level (wet weight basis) based on samples from the same ecosystem of the Mekong Delta, i.e., 0.114 for Hg, 0.066 for Se and 0.088 for Rb (Ikemoto et al., 2008).



Fig. 7. Relationships between log Kow of persistent organic pollutants (POPs) and the regression equation slope of δ^{15} N and log-transformed POPs levels (µg/g wet weight) in biota from the Mekong Delta. Log Kow of POPs are taken from Mortimer and Connell (1995) and Simpson et al. (1995).

Log Kow of TCPMe was highest at 7.8 followed by that of the three congeners of DDT, i.e., p,p'-DDT, p,p'-DDD, p,p'-DDE, and two isomers of CHLs, i.e., *cis*-nonachlor and *trans*-nonachlor. The slope of these POPs were in the range of 0.160–0.177. In contrast, isomers of HCHs and HCB showed a relatively shallow slope, less than 0.10 (wet weight basis). Of the latter group of POPs, log Kow of the isomers of HCHs and HCB exceeded 5.0, a value that Suedel et al. (1994) consider to be the threshold for biomagnification. However, as mentioned above, it is possible that the low concentration of HCHs and HCB drive down the slope. Further studies, therefore, are necessary to elucidate why HCHs and HCB did not undergo biomagnification through the food web even when the log Kow was more than 5.

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References

- Buser, H.-R., 1995. DDT, a potential source of environmental tris(4chlorophenyl)methane and tris(4-chlorophenyl)methanol. Environ. Sci. Technol. 29, 2133–2139.
- Cabana, G., Rasmussen, J.B., 1996. Comparison of aquatic food chains using nitrogen isotopes. Proc. Natl. Acad. Sci. USA 93, 10844–10847.
- Cole, M.L., Valiela, I., Kroeger, K.D., Tomasky, G.L., Cebian, J., Wigand, C., McKinney, R.A., Grady, S.P., Carvavlho da Silva, M.H., 2004. Assessment of a δ¹⁵N isotopic method to indicate anthropogenic eutrophication in aquatic ecosystems. J. Environ. Qual. 33, 124–132.
- DeNiro, M.J., Epstein, S., 1978. Influence of diet on the distribution of carbon isotopes in animals. Geochim. Cosmochim. Acta 42, 495–506.
- DeNiro, M.J., Epstein, S., 1981. Influence of diet on the distribution of nitrogen isotopes in animals. Geochim. Cosmochim. Acta 45, 341–351.
- Hobson, K.A., Fisk, A., Karnovsky, N., Holst, M., Gagnon, J.-M., Fortier, M.A., 2002. A stable isotope (δ¹³C, δ¹⁵N) model for the North Water food web: Implications for evaluating trophodynamics and the flow of energy and contaminants. Deep-Sea Res. Part II 49, 5131–5150.
- Hobson, K.A., Welch, H.E., 1992. Determinatin of trophic relationships within a high Arctic marine food web using $\delta^{13}C$ and $\delta^{15}N$ analysis. Mar. Ecol. Prog. Ser. 84, 9–18.
- Hoekstra, P.F., O'Hara, T.M., Fisk, A.T., Borgå, K., Solomon, K.R., Muir, D.C.G., 2003. Trophic transfer of persistent organochlorine contaminants (OCs) within an Arctic marine food web from the southern Beaufort-Chukchi Seas. Environ. Pollut. 124, 509–522.
- Hop, H., Borgå, K., Gabrielsen, G.W., Kleivane, L., Skaare, J.U., 2002. Food web magnification of persistent organic pollutants in

poikilotherms and homeotherms from the Barents Sea. Environ. Sci. Technol. 36, 2589–2597.

- Ikemoto, T., Tu, N.P.C., Okuda, N., Iwata, A., Omori, K., Tanabe, S., Tuyen, B.C., Takeuchi, I. 2008. Biomagnification of trace elements in the aquatic food web in the Mekong Delta, South Vietnam using stable carbon and nitrogen isotope analysis. Arch. Environ. Contamin. Toxicol., doi:10.1007/s00244-007-9058-5.
- Iwata, H., Agusa, T., Inoue, S., Kobota, R., Minh, N.H., Minh, T.B., Tu, N.P.C., Kajiwara. N., Kunisue, T., Subramanian, A., Tanabe, S., Viet, P.H., Tuyen, B.C., 2004. Contamination of trace elements in groundwater and persistent organochlorines in sediment from Mekong Delta, South Vietnam. In: Proceeding. International Symposium on the Development of Water Resource Management System in Mekong Watershed. Hanoi, Vietnam, December 3rd–4th 2004, pp. 25–31.
- Jarman, W.M., Hobson, K.A., Sydeman, W.J., Bacon, C.E., McLaren, E.B., 1996. Influence of trophic position and feeding location on contaminant levels in the Gulf of the Farallones food web revealed by stable isotope analysis. Environ. Sci. Technol. 30, 654–660.
- Kajiwara, N., Ueno, D., Monirith, I., Tanabe, S., Pourkazemi, M., Aubrey, D.G., 2003. Contamination by organochlorine compounds in sturgeons from Caspian Sea during 2001 and 2002. Mar. Pollut. Bull. 46, 741–747.
- Kidd, K.A., Schindler, D.W., Muir, D.C.G., Lockhart, W.L., Hesslein, R.H., 1995. High concentrations of toxaphene in fishes from a subarctic lake. Science 269, 240–242.
- Kidd, K.A., Bootsma, H.A., Hesslein, R.H., Muir, D.C.G., Hecky, R.E., 2001. Biomagnification of DDT through the benthic and pelagic food webs of Lake Malawi, East Africa: importance of trophic level and carbon source. Environ. Sci. Technol. 35, 14–20.
- Minh, T.M., Watanabe, M., Tanabe, S., Miyazaki, N., Jefferson, T.A., Prudente, M.S., Subramanian, A., Karuppiah, S., 2000. Widespread contamination by tris(4-chlorophenyl)methane and tris(4-chlorophenyl)methanol in cetaceans from the North Pacific and Asian coastal waters. Environ. Pollut. 110, 459–468.
- Minh, N.H., Minh, T.B., Kajiwara, N., Kunisue, T., Iwata, H., Viet, P.H., Tu, N.P.C., Tuyen, B.C., Tanabe, S., 2006. Contamination by

polybrominated diphenyl ethers and persistent organochlorines in catfish and feed from Mekong River Delta, Vietnam. Environ. Toxicol. Chem. 25, 2700–2709.

- Minagawa, M., Wada, E., 1984. Stepwise enrichment of δ^{15} N along food chains: further evidence and the relation between δ^{15} N and animal age. Geochim. Cosmochim. Acta 48, 1135–1140.
- Mortimer, M.R., Connell, D.W., 1995. A model of the environmental fate of chlorohydrocarbon contaminants associated with Sydney sewage discharges. Chemosphere 30, 2021–2038.
- Muir, D., Savinova, T., Savinov, V., Alexeeva, L., Potelov, V., Svetochev, V., 2003. Bioaccumulation of PCBs and chlorinated pesticides in seals, fishes and invertebrates from the White Sea, Russia. Sci. Total Environ. 306, 111–131.
- Okuda, N., Hamaoka, H., Fukumoto, T., Miyasaka, H., Omori, K., 2004. How to monitor the coastal food-web dynamics using stable-isotope analysis of higher consumers. In: Proceedings of International Symposium on Long-term Variations in the Coastal Environments and Ecosystems, 27–28 September 2004, Matsuyama, Japan, pp. 200–205.
- Peterson, B.J., Howarth, R.W., Garritt, R.H., 1985. Multiple stable isotopes used to trace the flow of organic matter in estuarine food webs. Science 227, 1361–1363.
- Peterson, B.J., Fry, B., 1987. Stable isotopes in ecosystem studies. Ann. Rev. Ecol. Syst. 18, 293–320.
- Ruus, A., Ugland, K.I., Skaare, J.U., 2002. Influence of trophic position on organochlorine concentrations and compositional patterns in a marine food web. Environ. Toxicol. Chem. 21, 2356–2364.
- Simpson, C.D., Wilcock, R.J., Smith, T.J., Wilkins, A.L., Langdon, A.G., 1995. Determination of octanol-water partition coefficients for the major components of technical chlordane. Bull. Environ. Contam. Toxicol. 55, 149–153.
- Suedel, B.C., Boraczek, J.A., Peddicord, R.K., Clifford, P.A., Dillon, T.M., 1994. Trophic transfer and biomagnification potential of contaminants in aquatic ecosystems. Rev. Environ. Contam. Toxicol. 136, 21–89.
- Tashiro, S., Matsumura, F., 1977. Metabolic routes of *cis* and *trans*chlordane in rats. J. Agric. Food Chem. 25, 872–880.