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Environmental Pollution 129 (2004) 431–441

ENVIRONMENTAL POLLUTION

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Persistent organochlorine residues in human breast milk from Hanoi and Hochiminh city, Vietnam: contamination, accumulation kinetics and risk assessment for infants

Nguyen Hung Minh[a], Masayuki Someya[a], Tu Binh Minh[a], Tatsuya Kunisue[a], Hisato Iwata[a], Mafumi Watanabe[a], Shinsuke Tanabe[a,*], Pham Hung Viet[b], Bui Cach Tuyen[c]

[a]*Center for Marine Environmental Studies, Ehime University, Bunkyo-cho 2-5, Matsuyama 790-8577, Japan*
[b]*Center for Environmental Technology and Sustainable Development, Hanoi National University, 334 Nguyen Trai Street, Hanoi, Vietnam*
[c]*University of Agriculture and Forestry, Hochiminh City, Vietnam*

Received 6 June 2003; accepted 21 November 2003

"Capsule": *It is suggested that daily intake rates of persistent organochlorines in mothers in Vietnam may result in health risk for nursing children.*

Abstract

Despite the ban on persistent organochlorines (OCs) in most of the developed nations, their usage continued until recently in many Asian developing countries including Vietnam, for agricultural purposes and vector-borne disease eradication programs. In this study, we collected human breast milk samples from the two big cities in Vietnam: Hanoi ($n = 42$) and Hochiminh ($n = 44$) and determined the concentrations of persistent OCs such as PCBs, DDT and its metabolites (DDTs), hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), chlordane compounds (CHLs) and *tris*-4-chlorophenyl-methane (TCPMe). The contamination pattern of OCs was in the order of DDTs > PCBs > HCHs > CHLs≈HCB≈TCPMe. Compilation of available data indicated that DDT residue levels in human breast milk from Vietnam were among the highest values reported for Asian developing countries as well as developed nations. This result suggests recent usage of DDTs in both north and south Vietnam. Interestingly, in both cities, the *p,p'*-DDT portion was higher in multiparas than those in primiparas. Considering the fact that the interval between the first and the second child of a mother in Vietnam is usually short, this result probably indicates continuous intake of DDTs in the population. Analysis of infant exposure to DDTs via breast milk suggested that the daily intake rates for number of individuals are close to or above the threshold for adverse effects which may raise concern on children health.

Keywords: DDTs; PCBs; TCPMe; Human breast milk; Vietnam; Infant health

1. Introduction

Widespread contamination and toxic effects of persistent organic pollutants in humans and wildlife have been of great concern and received considerable attention during the past four decades. Despite the ban on persistent organochlorines (OCs) in most of the developed nations since the early 1970s, their usage continued until very recently in many developing countries for agricultural and public health purposes. As these compounds are highly lipophilic and persistent, human chronic exposure via food chain has led to the accumulation of both parent compounds and their metabolites in lipid rich tissues such as adipose tissues and human breast milk. Among human tissues, breast milk is a convenient sampling matrix for measuring residue concentrations of persistent OCs. The samples are easy to collect and highly suitable for estimating body burdens of persistent OCs, and thus may provide useful information about their accumulation kinetics in humans. In addition, The OC residue concentrations in human breast milk are a key factor for evaluating the toxic potential of contaminants in infants.

* Corresponding author. Tel./fax: +81-89-927-8171.
E-mail address: shinsuke@agr.ehime-u.ac.jp (S. Tanabe).

0269-7491/$ - see front matter
doi:10.1016/j.envpol.2003.11.012

Recently, hormone-like activities of some persistent OCs such as PCBs, DDTs, and HCHs have been suggested (Cheek et al., 1999; Colborn et al., 1993; Kelce, 1995; Vos et al., 2000). These facts have raised public concern towards the adverse effects of such OCs to human health, especially for infants due to their susceptibility to environmental impacts. Walkowiak et al. (2001) observed association between postnatal exposure to PCBs and mental/motor development of children from 30 months onward. In the Netherlands, Vreugdenhil et al. (2002) reported a considerable association between prenatal exposure to PCBs and changing playbehavior of children. In the United States, Longnecker et al. (2001) reported a strong association between *p,p'*-DDE levels in mother's serum and the likelihood of premature birth and baby's weight at birth. These findings highlighted the role of persistent OCs, particularly PCBs and DDTs in the neuropsychological development in children.

Despite the fact that great efforts have been done worldwide to phase out the usage of OC insecticides such as DDT, HCHs and CHLs, some recent inputs of DDTs have been recorded in many developing countries like Mexico, India, Thailand and Vietnam (Pandit et al., 2002; Stuetz et al., 2001; Minh et al., 2002; Nhan et al., 1998). In Vietnam, recent input of DDTs to environment has been suggested in both northern and southern parts of the country. For example, relatively high levels of DDTs have been found in various environmental compartments such as surface waters, sediment (Hung et al., 2002; Phuong et al., 1998) as well as fish, mollusks and birds (Nhan et al., 1998; Minh et al., 2002). These elevated concentrations of DDTs could be a result of recent application of DDT for malaria control and, to a lesser extent, for sanitary purposes (Nhan et al., 2001). However, comprehensive studies examining the contamination of persistent OCs in Vietnamese human as a result of bioaccumulation through food chains have not been made in recent years. Therefore, appropriate perspectives of the authorities and public to the possible adverse impacts of DDTs as well as other OCs on human health are still obscure.

Over the last few years, in the frame-work of the Asia-Pacific Mussel Watch Program, our laboratory has conducted comprehensive investigations on the distribution, behavior and fate of persistent OCs in various environmental compartments such as air, water, soils, sediments and biological samples (fish, mussels and birds) and foodstuff from Vietnam (Kannan et al., 1992, 1995; Iwata et al., 1994; Thao et al., 1993; Monirith et al., 2000; Minh et al., 2002). Recently, through the Core University Program supported by the Japan Society for the Promotion of Science (JSPS) in which we are also involved, we collected human breast milk from the two big cities in Vietnam, Hanoi and Hochiminh and determined the concentrations of persistent OCs such as PCBs, DDTs, HCHs, CHLs, HCB and TCPMe to evaluate the status of contamination in Vietnamese population in comparison to other countries in the region. In addition, accumulation kinetics of OCs in Vietnamese human were studied. Potential risk for breastfed infants due to OC exposure was also evaluated.

2. Materials and methods

2.1. Sample collection

Human breast milk samples were collected from two big cities in Vietnam; Hanoi ($n=42$) and Hochiminh ($n=54$) during the years 2000 and 2001. These milk samples were kept in ice immediately after collection, shipped to our laboratory in Japan with dry ice and preserved there at −20 °C until analysis. We randomly selected the breast milk donors from two communities in Hochiminh city, Vinh Loc and Dong Thanh; and two communities in Hanoi, Me Tri and Tu Liem. Data on the biological characteristics of the donors and relevant information of sampling sites are given in Table 1. The biological characteristics show similarity between such cohorts. The informed consents were obtained from all the donors. Questionnaires on dietary aspects were recorded. Years of residence of the donors in their area varied widely from 1 to 37 years indicating that the cohorts actually consist of women from various places and not entirely represent only resident communities. However, the migrations were believed as regular and also within the regions of each city.

2.2. Chemical analysis

A portion of 10 grams of milk samples was applied to extraction column (2 cm diameter) packed with 10 g of pre-cleaned diatomite earth (Merck, Damstadt, Germany). The samples were then kept in the columns for 30 min allowing maximum absorption of the samples onto the material before they were eluted by 200 ml of diethyl ether at a flow rate of 1 ml/min. The eluates were dried by anhydrous Na_2SO_4 and concentrated to 8 ml. An aliquot of 2 ml was used for fat content determination by gravimetric method and the remaining volume was evaporated under gentle nitrogen stream down to 5 ml, which was then mixed with 5 ml of dichloromethane (DCM) to obtain 10 ml sample in hexane/DCM (1:1). The sample was then subjected to gel permeation chromatography (GPC) for fat removal and a same mixture of hexane/DCM (1:1) was used as eluting solvent in GPC system at a flow rate of 5 ml/min. The first fraction eluted with 120 ml solvent containing lipids was discarded and the following 100 ml eluate containing OCs was collected and concentrated to 3 ml. The concentrate was then applied to a chromatography column

Table 1
Relation information of breast milk donors from Hanoi and Hochiminh city, Vietnam

Location		Age	Weight (kg)	Height (cm)	Number of children	Number of pregnant	Occupation
Hanoi	Mean	29	50	157	1.6	1.6	Housewife: 58%
(2000, $n=42$)	Range	20–44	40–63	150–166	1–7	1–7	Others: 42%
Hochiminh	Mean	27	49	155	1.6	1.7	Housewife: 61%
(2001, $n=44$)	Range	18–37	31–64	140–170	1–5	1–7	Others: 39%

packed with 12 g activated Florisil for separation of PCBs and OC insecticides as described in our previous report (Minh et al., 2000). A procedural blank was run for every batch of five samples to verify cross-contamination.

Quantification method was similar to those reported previously (Minh et al., 2000). Briefly, TCPMe and *tris*-4-chlorophenyl-metanol (TCPMOH) were quantified by GC-MSD (Hewlett-Packard series 6890). Cluster ions were monitored at m/z 311, 313, 346, 348 for TCPMe and 139, 251, 253, 362, 364 for TCPMOH. The other organochlorines (PCBs, DDTs, HCHs, etc.) were quantified by GC-ECD (Hewlett Packard series 6890) using DB-1 fused silica capillary column (30 m length). The column oven temperature was programmed from 60 to 160 °C at a rate of 20 °C/min, held for 10 min, then increased to 260 °C at a rate of 20 °C/min and held for 20 min. PCB standard used for quantification was an equivalent mixture of Kanechlor preparations with identified PCB composition and content. Concentrations of individually resolved peaks of almost sixty PCB isomers and congeners were summed up to obtain total PCB concentrations. Recovery rates of the target chemicals through this analytical method ($n=6$) were 99.5% ± 3.4 for DDTs; 101% ± 4.9 for PCBs, 99% ± 2.5 for HCHs, 98% ± 2.1 for HCB, 101% ± 2.2 for CHLs, 108% ± 2.9 for TCPMe and 90% ± 5.9 for TCPMOH. Concentrations were not corrected for recovery rate. DDTs represents the sum of *p,p'*-DDT, *p,p'*-DDD and *p,p'*-DDE, and CHLs include *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, and oxychlordane. HCHs include α,-β and γ-isomers. In this study, concentrations of α- and γ-HCHs were mostly below the detection limit (0.1 ng/g). Therefore, HCHs represent mainly concentrations of β-HCH. Similarly, TCPMOH was found only in less than 30% of the examined samples at levels above the detection limit of 1 ng/g lipid wt. Concentrations of OCs were expressed as ng/g on a lipid wt basis, unless otherwise specified.

Regarding quality assurance and quality control, our laboratory participated in the Intercomparison Exercise for Persistent Organochlorine Contaminants in Marine Mammal Blubber organized by the National Institute of Standards and Technology (1999, Gaithersburg, MD, USA) and Marine Mammal Health and Stranding Responses Program of the National Oceanic and Atmospheric Administration's National Marine Fisheries Service (Silver Spring, MD. USA). We analyzed Standard Reference Material SRM 1945 for selected PCB congeners and persistent OC insecticides. Our results were in good agreement with the standard reference values. However, this analytical method applied Soxhlet extraction, which is little different with solid phase extraction used for the human breast milk in the present study. The method for human breast milk was qualified by cross-checking analysis using corresponding real samples to compare with earlier result reported by Kunisue et al. (2002b) and standard deviations of approximately 15% for PCBs and chlorinated pesticides was considered as satisfactory result.

2.3. Statistical analysis

Test for significant difference and correlation ($P < 0.05$) were performed using, respectively, Mann-Whitney U test and Spearman test, which are available in StatView version 5 (SAS Inc., 1998).

3. Results and discussions

3.1. Residue levels and accumulation pattern

Lipid-normalized concentrations of OCs in human breast milk from Hanoi and Hochiminh city, Vietnam are given in Table 2. We considered the accumulation of OCs in 2 groups: primiparas and multiparas. In general, the residue pattern of OCs in human breast milk in Vietnam followed the order of DDTs > PCBs > HCHs > CHLs≈HCB≈TCPMe. Significant differences in OC levels between two cities were found only for HCHs. The pattern of OCs in human breast milk observed in this study is different to that found in Vietnamese foodstuff analyzed 10 years ago (Kannan et al., 1992) or in human breast milk from Hochiminh city (Schecter et al., 1989), which showed higher level of HCHs compared to PCBs. However, the present pattern is in accordance with those recently observed in birds collected from Red river estuary, (Minh et al., 2002), mollusks from Hanoi region (Nhan et al., 2001), and

Table 2
Concentrations of organochlorides in human breast milk (ng/g lipid wt.) in Hanoi and Hochiminh city, Vietnam[a]

	Age	Fat content	PCBs	DDT compound				CHL compounds				β-HCH	HCB	TCOMe
	(years)	(%)		*p,p'*-DDE	*p,p'*-DDD	*p,p'* = DDT	DDTs	oxy	t-nona	c-nona	CHLs			
Hanoi														
Primiparas	27	2.5	76	2200	13	190	2400^{+}	0.88	2.2	1.9	2.5	69	4.2	3.7
Multiparas	31	2.1	72	1500	8.9	150	1700^{+}	0.92	0.86	0.3	1.4	46	3.5	3.9
Overall[b]	29	2.3	74	1900	11	170	2100	0.90	1.53	1.1	2.0	58	3.9	3.8
Range	20–44	0.7–5.7	26–210	420–6300	3–50	34–6900	480–6900	0.25–1.6	<0.21–8.1	<0.21–3.2	<0.72–13	11–160	0.62–9.5	1.4–8.3
Hochiminh														
Primiparas	24	3.3	88	2700*	8.3	310	3020*	2.7	4.2	0.8	7.8	14	2.8	7.2
Multiparas	29	2.9	70	1300*	5.7	220	1500*	2.1	3.2	0.8	6.0	13	2.1	7.2
Overall	27	3.1	79	2000	7.0	265	2300	2.4	3.7	0.8	6.9	13.5	2.5	7.2
Range	18–37	1.5–6.9	29–200	340–16,000	2.7–18	100–1000	440–17,000	<0.25–9.3	1.3–15	<0.21–2.1	1.3–26	4.1–35	1.3–10	1.1–26

oxy: oxychlordane, *t*-nona: *trans*-nonachlor, *c*-nona: *cis*-nonachlor. DDTs = *p,p'*-DDE + *p,p'*-DDT, CHLs = oxy + *t*-nona + *c*-nona.
[a] Arithmetic means were given.
[b] Average concentration from primiparas and multiparas; $^{+}P = 0.056$; $^{*}P < 0.05$.

in biotic samples from Red river delta (Nhan et al., 1998).

The concentrations of PCBs are rather uniform in both the cities in accordance with those observed in foodstuffs from different parts of Vietnam (Kannan et al., 1992). The global comparison of PCB residues in human breast milk is given in Table 3. We cited data from recent studies which used high resolution gas chromatography for quantification of individual PCB congeners. Although the cited data may differ between laboratories, it is possible to draw some relevant comparison to understand the magnitude of contamination. In comparison to other developing countries like Cambodia, India and the Philippines, residue levels of PCBs in human breast milk from Vietnam are slightly higher. However, these PCBs levels are still below those reported for developed countries (Table 3). Recent global inventory of PCBs production and consumption has indicated that common applications of PCBs (i.e. for industrial purposes) in Vietnam during the past years were not higher than those in China, Hong Kong, India and the Philippines (Breivik et al., 2002). Hence, the higher PCB residues observed in human breast milk from Vietnam suggest additional sources of PCBs besides industrial sources like transformers, capacitors, etc. A likely source of PCBs in Vietnam could be the release from different kinds of military weapons used extensively during the Vietnam War as suggested earlier (Thao et al., 1993).

Mean concentrations of DDTs in human breast milk were 2100 ng/g and 2300 ng/g in Hanoi and Hochiminh city, respectively, and the levels in primiparas group were higher than those in multiparas group (Table 2). This difference indicates that excretion via milk during lactation is an important factor reducing DDT burden in nursing mothers. Further examination of the DDTs composition revealed that *p,p'*-DDE is the predominant compound accounting for 85–90% of the total DDT concentrations. Interestingly, in both cities, the proportion of *p,p*'-DDT was higher in multiparas than those in primiparas. We observed that time interval between the first and the second child of a mother in both the cohorts is usually short around 3 years. Therefore, the higher proportion of *p,p'*-DDT accumulated in multiparas group could be interpreted as an evidence for continuous intake of DDTs.

Elevated concentrations of DDTs in various environmental compartments in Vietnam have also been reported in a number of recent investigations. Results of the Asia-Pacific Mussel Watch Program indicated that DDT concentrations in mussels and fish from Vietnamese coastal waters are among the highest values reported for the countries in this region (Kannan et al., 1995; Monirith et al., 2000; Minh et al., 2002). Interestingly, Nhan et al. (2001) reported higher levels of DDTs in sediments from populated locations in Hanoi as

Table 3
Comparison of organochlorine residues in human breast milk from various countries (ng/g lipid wt.)

Country	Year of Sampling	*n*	PCBs	DDTs	CHLs	HCHs	HCB	Reference
Industrialized countries								
Australia	1995	60	500	1200	–	350	–	Quinsey et al., 1995
Czech Republic	1996	17	1160	1050	–	70	–	Schoula et al., 1996
Germany	1995–1997	246	550	240	–	40	80	Shade et al., 1998
Turkey	1995–1997	104	–	2100	–	457	50	Cok et al., 1997
Russia	1996–1997	140	380	1040	37	280	91	Polder et al., 2003
Sweden	1997	40	324	143	–	–	12	Noren et al., 2000
Ukraine	1993–1994	197	594	2700	38	730	168	Gladen et al., 1999
UK	1997–1998	168		470	–	103	43	Harris, 1999
US (Massachusets)	1993	122	320	–	–	–	–	Korrick et al., 1998
Japan	1998	49	200	290	85	210	14	Konishi et al., 2001
Developing countries								
Cambodia	2000	28	42	1600	1.8	5.5	1.7	Kunisue et al, 2002b
China (Guangzhou)	2000	54	33	3550	–	1110	–	Wong et al., 2002
China (Hongkong)	1999	132	42	2870	–	950	–	Wong et al., 2002
India	2000	8	30	420	0.9	650	1.0	Kunisue et al., 2002b
Philippines	2000	10	72	190	15	4.7	–	Kunisue et al., 2002b
Vietnam (North)	2000	42	74	2100	2.0	58	3.9	Present study
Vietnam (South)	2001	54	79	2300	6.9	14	2.5	Present study
Mexico	1998	60	–	4100	–	60	30	Waliszewski et al., 2001

– data not available.

compared to those from paddy fields in Red river delta. This evidence indicates recent applications of DDTs for other purposes such as sanitary and malaria control rather than for agriculture. The widespread and elevated contamination of DDTs in the environment and food chains may explain the high residue levels found in human breast milk from Vietnam.

To understand the magnitude of DDT contamination in Vietnamese population, residue levels of DDTs in human breast milk in different countries were compiled (Table 3). Interestingly, DDT concentrations in Vietnamese human breast milk were among the highest values reported for the countries surveyed. This observation again suggests the recent use of DDTs in Vietnam and that Vietnam may be a potential source of DDTs in the south Asian region. In general, DDT levels in human breast milk were observed to be higher in developing countries than those in developed nations (Table 3), which could be due to the recent use of this insecticide for malaria control and, to a lesser extent, for sanitary purposes in developing countries. In this context, the role of the south Asian region as an emission source to pristine areas in higher latitudes deserves further monitoring studies. Our recent study on seasonal variations of persistent OCs in migratory birds from Lake Baikal wintering in south eastern Asian region has indicated elevated exposure to DDTs and HCHs in the southern wintering sites, suggesting the south Asian region as a potential source of OC insecticide contamination for higher latitude areas (Kunisue et al., 2002).

The spatial distribution of HCHs in our study are somewhat in agreement with those reported in earlier studies showing higher HCHs levels in Hanoi compared to Hochiminh (Thao et al., 1993; Iwata et al., 1994; Kannan et al., 1995). There are two possible reasons for this spatial pattern: (i) due to the lower latitudinal position, Hochiminh city has typical tropical climate which perhaps facilitates more volatilization of HCHs; (ii) on the other hand, Hanoi city is located at higher latitude toward the north and close to China, the world's largest producer/user of HCHs (Li et al., 1998). Thus, higher export of HCHs to these areas leading to higher concentrations in environment and biota could be expected. In the global comparison, human breast milk from North Vietnam showed intermediate levels of HCHs, which are lower than those in China, Hong Kong, India and Japan but higher than those in other Asian developing countries like Cambodia and the Philippines (Table 3). Besides, our recent study on birds demonstrated that HCH concentrations in migratory birds collected from Vietnam were significantly higher than those in resident birds, which could be due to the accumulation in stopover sites in India and South China (Minh et al., 2002).

Total chlordane concentrations were 2.0 and 6.9 ng/g in Hanoi and Hochiminh city, respectively. The most abundant compound was *trans*-nonachlor, accounting for about 50 percent of the total CHLs. In general, CHL residues in Vietnamese human breast milk were considerably lower than those in industrialized countries such as Japan and Russia etc. (Table 3). Similarly, levels

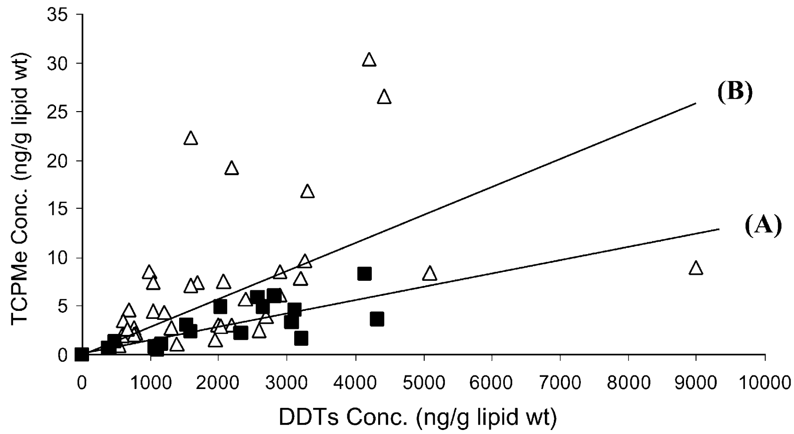

Fig. 1. Correlation between concentrations (ng/g lipid wt.) of TCPMe and DDTs observed in human breast milk from Hanoi ($n=18$, $P<0.001$) and Hochiminh city ($n=39$, $P<0.001$). The squares and line A are for Hanoi city. The triangles and the line B are for Hochiminh city.

of HCB in human breast milk were rather low when compared to global levels, which indicate minimal exposure of the general population to this chemical throughout the country.

Concentrations of TCPMe in human breast milk were 3.8 and 7.5 ng/g in Hanoi and Hochiminh city, respectively. In contrast, only trace levels of TCPMOH were found in less than 30% of the total samples. The TCPMe concentrations in human breast milk from Vietnam are comparable to those of CHLs suggesting widespread contamination of TCPMe in Vietnamese humans. Besides, it is also recognized that the TCPMe and DDT levels in all samples have significant correlation ($P<0.001$; Spearman test; Fig. 1). This result probably supports the early hypothesis that TCPMe is a byproduct in production of technical DDTs (Buser, 1995). A good correlation between TCPMe and DDT concentrations was also observed earlier in Japanese human adipose tissues (Minh et al., 2000). To date, data on contamination of TCPMe in human breast milk is rather scant. The only available data is from Sweden (Rahman et al., 1993), which reported slightly lower TCPMe residues compared to those in Vietnam. To our knowledge, data from the present study probably comprises the most recent and extensive analyses of TCPMe in human breast milk samples.

3.2. Specific accumulation according to parity and age

It is known that adult female excrete lipophilic contaminants such as OCs via lactation and thus reducing the body burden of such contaminants. In this study, we observed higher concentrations of OCs in human breast milk of primiparous mothers as compared to multiparas. Fig. 2 shows the relationship between parity and DDTs concentrations in human breast milk from Hanoi and Hochiminh cities. The relationship however, is less pronounced for other compounds like PCBs and HCHs

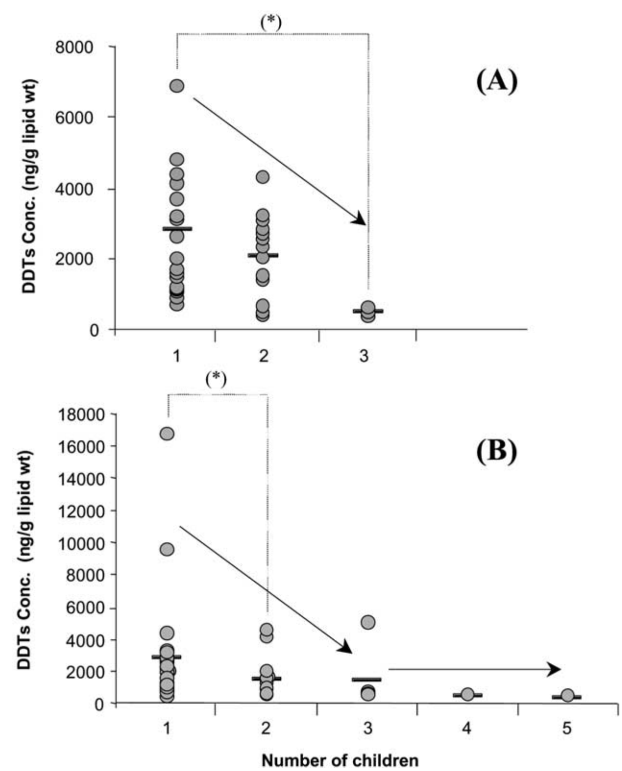

Fig. 2. Concentrations of DDTs in maternal milk versus the corresponding number of children (A: Hanoi, B: Hochiminh). The short horizontal bars indicate mean concentration in each group and the stars (*) indicate significant difference ($P<0.05$).

(data not shown), probably due to the lower background levels. Indeed, the present result provides another evidence for the influence of lactation on OC burden of nursing women (Albers et al., 1996).

Because of the potential influence of parity on OCs levels, we have examined the correlation between residue levels and mother's age independently for the groups of primiparas and multiparas. In both groups, strong correlations of contaminant levels and age were observed. However, only DDTs levels of multiparas group in Hochiminh city significantly correlated with age ($P < 0.05$; Spearman test). Nevertheless, slopes of the regression lines for DDTs and mother's age (Fig. 3) are quite similar regardless of the differences in groups and areas. This result would probably be interpreted as an evidence of similar intake rates of DDTs in Vietnamese mothers. Besides, it should be noted that other factors such as length of lactation, time of sampling during breast-feeding etc. could also influence the OC levels in women. For instance, Schecter et al. (1998) reported that DDE and HCB levels in breast milk of a mother nursing twins might reduce approximately 80 and 90 percent respectively, during thirty-eight month period. This lactation period is probably rather long compared to those in Vietnamese cohort which is six months, typically. However, lack of information regarding length of lactation and time of sampling during breast-feeding, etc in the Vietnamese cohort did not allow us to further examine their influence to the OC levels. Further investigation towards these effects would be necessary in future study. Nevertheless, the available data from this study probably indicates that parity and age play important roles in controlling the OC burden in humans.

3.3. Decreasing trend of OC in human breast milk in Vietnam

Although DDTs levels in human breast milk from Vietnam are still in the highest range in our worldwide comparison, it should be noted that their concentrations have declined over the past 10 years. The DDT levels in this study are perhaps one fourth of those reported in 1989 (Schecter et al., 1989). Assessment of decreasing trend of DDTs and other OCs in Vietnamese women would give a further insight into human exposure in the past, present and future.

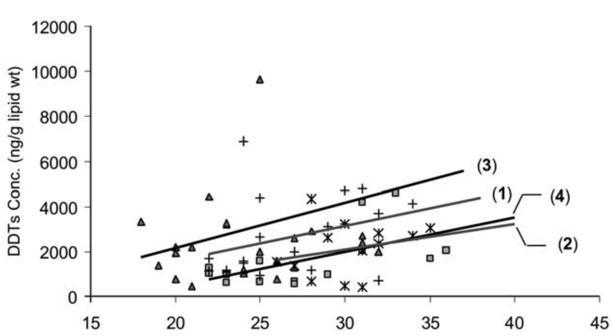

Fig. 3. Relationship between DDT concentrations and mother's age. The cross mark (+) and line 1 indicate primiparas group in Hanoi; the stars (∗) and line 2 the multiparas in Hanoi. Similarly, the triangles (△) and line 3 indicate primiparas group in Hochiminh; the squares (□) and line 4 the multiparas in Hochiminh.

The decrease of persistent organic pollutants such as DDTs, PCBs and HCHs in human breast milk was suggested to follow first-order kinetic (Noren et al., 2000) according to the equation:

$$-dC/dt = k \times C_t \quad \text{or} \quad C_t = C_O \times e^{-k \times t} \tag{1}$$

where C_0 and C_t are the concentrations at the first and the last investigation, respectively; k: the rate constant and t: the time interval between investigations (year). Another important factor for the assessment is half-life time ($t_{dec1/2}$) defined as the duration in which initial concentrations decrease to a half. Based on the residue concentrations of OCs in 1989 reported by Schecter et al. (1989) and the levels in 2001 obtained in this study, the rate constant and $t_{dec1/2}$ were calculated for the group in Hochiminh city and given in Table 4.

Residue levels of *p,p'*-DDT have decreased from 4700 to 2700 ng/g lipid wt. in over 10 years with $t_{dec1/2}$ of around 3 years. On the other hand, *p,p'*-DDE decreased rather slowly with a $t_{dec1/2}$ of 6 years. The results of the present study are somewhat in agreement with those in Sweden showing half-life time 4 and 6 years for *p,p'*-DDT and *p,p'*-DDE, respectively (Noren and Meironyte, 2000). The slightly shorter of half-life time observed here could be due to the tropical climate that exist in Vietnam may have facilitated volatilization of *p,p'*-DDT in the environment leading to its faster decrease in food chains (thus in humans). The half-life time of DDT compounds in this study was estimated basing on first-order kinetic with regard to steady state condition. Given that DDTs exposure in Vietnam may still occurs to some extent, the calculation might slightly underestimate values of half-life time. Nevertheless, the estimated half-life time is fairly beneficial for assessing the trend of DDT contamination in Vietnamese human breast milk. Assuming that the decrease trend of DDTs remains more or less constant, we can estimate the DDTs levels reaching approximately 700 ng/g lipid wt in the year 2011 (Fig. 4). However, lower residue levels in future can be expected if the use of DDTs is completely phased out from now. The decreasing trend of

Table 4
Estimation of time-trend factors from 1989 to 2001

Compounds	Mean concentration[a]		Trend factor	
	Year 1989[b]	Year 2001	k value (yr^{-1})	$t_{dec1/2}$ (yr)
p,p'-DDT	4700	270	0.26	3
p,p'-DDE	6700	2000	0.11	6
PCB-138	30	15	0.06	11
PCB-153	31	18	0.05	13
PCB-180	23	15	0.04	18

[a] ng/g lipid wt.
[b] Schecter et al., (1989; $n = 7$).

PCBs was lower compared to those of DDTs (11–18 years for some major congeners such as CB-138, −153 and −180). This result is somehow in agreement with those reported in Sweden showing the half-life time for some PCB congeners varying from 11 to 17 years (Noren and Meironyte, 2000).

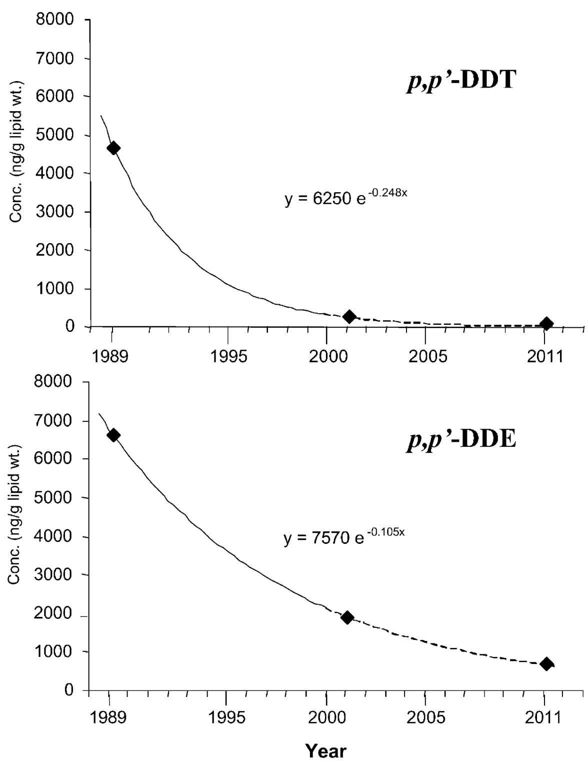

Fig. 4. Estimation of time-trend curve of *p,p'*-DDT and *p,p'*-DDE residues in human breast milk in Vietnam (see text for further details of the estimation of declining trend).

3.4. Levels of OCs related to adverse health outcomes on children

The potential effects of PCBs on the neurological development in children relevant to environmental concentrations have received particular attention. Extensive studies were recently conducted in Germany (Walkowiak et al., 2001) and the Netherlands (Vreugdenhil et al., 2002) suggesting an association between elevated PCB exposure and effects on the neuropsychological function in children. The results from those studies are shown in Fig. 5 in comparison with the PCB concentrations in human breast milk reported in different countries. As can be seen in that figure, PCBs concentrations above 400 ng/g is apparently associated with some adverse health effects in children such as change in playing behavior and impairment of mental and motor development. However, it should also be noted that because toxic potency of individual PCB congeners are different, human health risk caused by PCBs exposure could vary depending on levels as well as pattern PCBs contamination in each population and thus the above comparison may not provide a definite health risk assessment for a certain population unless more comprehensive data could be obtained. Nevertheless, the observed PCB levels in Vietnamese women seem rather below this harmful threshold indicating lesser health risk imposed by PCBs contamination.

DDTs which are predominant contaminants in developing countries compared to PCBs, may also be of concern with respect to mother and children health. Some recent studies suggest that *p,p'*-DDE, the major metabolite of *p,p'*-DDT may associate with adverse effects such as pre-term birth, lower weight and small baby size at birth, however it may not relate to breast

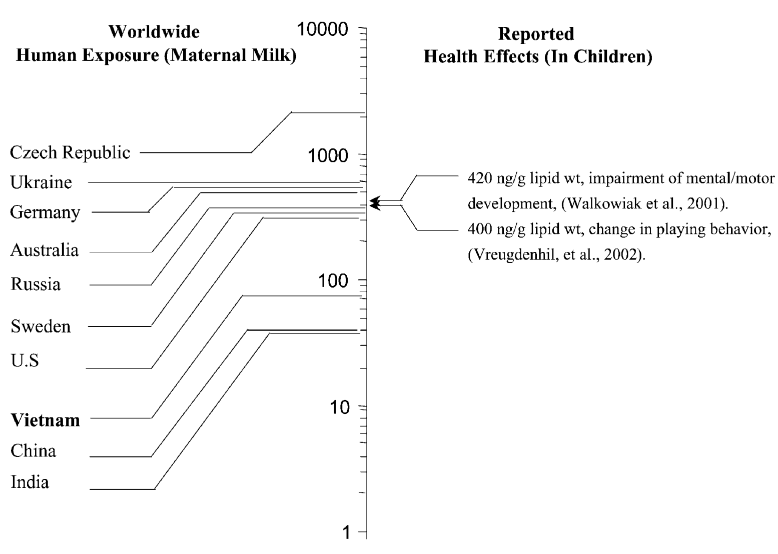

Fig. 5. Comparison of PCBs residue levels in human breast milk from various countries including Vietnam (corresponding to Table 3) with the reported levels found to cause adverse effects on child health.

cancer in women (Longnecker et al., 2001; Heuyer et al., 2000; Gammon et al., 2002). Our present research demonstrated that the current DDT contamination in Vietnamese women is relatively high with regard to the global comparison (Table 3). Moreover, approximately 15% of the women in this study have DDT levels above 4000 ng/g lipid wt which is comparable to average levels in the United States in the early 1960s (Daniel Smith, 1999). It would be important to note that the DDE levels in blood samples collected from nursing women in the US during this period, apparently associated with some adverse effects like pre-term birth and small baby size at birth (Longnecker et al., 2001). Regardless differences in racial and epidemiological characteristics, etc which may cause different toxicological response in the cohort in Vietnam and those in the United States, the above result may still raise concern regarding mother and infant health in Vietnam because it is known that pre-term birth is a great contributor to infant mortality. In the viewpoint of environmental toxicology, elevated DDT concentrations in Vietnamese human breast milk perhaps underlines higher risk to both mother and infant health and deserve stricter regulation to phase out completely the use of DDTs.

3.5. Tolerable daily intake

Daily intake of OCs by infants was calculated based on the assumption that the average milk consumption of a 5 kg infant is 700 g/day (Oostdam et al., 1999). The mean values of daily intake of organochlorines were estimated by using Eq. (2):

$$\mathrm{DI} = \frac{C_{\mathrm{milk}} \times 700g \times C_{\mathrm{lipid}}}{5} \qquad (2)$$

where DI is daily intake (µg/kg body wt./day); C_{milk}: concentration of the chemical in milk (µg/g lipid wt); C_{lipid}: lipid content in milk (%).

The estimated daily intakes are given in Table 5 and individual intakes are shown in Fig. 6. It was recognized

Table 5
Estimated daily intake (µg/kg body wt/day) of OCs by infants in Vietnam

		PCBs	DDTs	CHLs	HCHs	HCB
Hanoi		0.25	7.0	0.1	0.17	0.01
	range	0.30–1.2	0.8–27	<0.003–0.05	0.05–0.43	<0.003–0.04
Hochiminh		0.34	11.0[a]	0.03	0.06	0.01
	range	0.11–0.94	1.3–110	<0.003–0.08	0.01–0.31	<0.003–0.04
TDIs[b]		1.0	20	0.5	0.3	0.27

[a] Medial value 7.2.
[b] Cited from Oostdam et al., 1999 (Health Canada guideline).

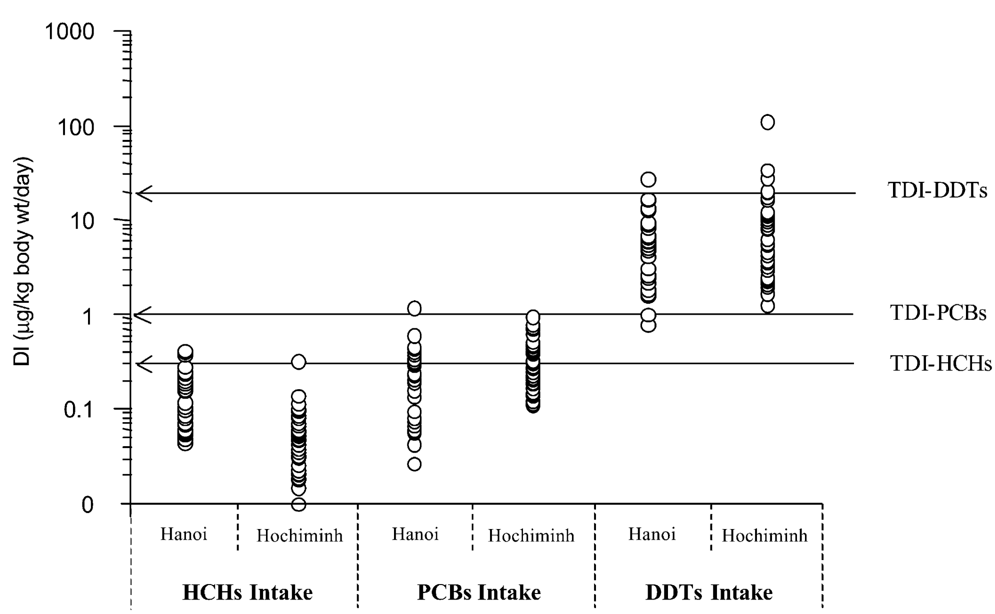

Fig. 6. Estimated daily intake of PCBs and DDTs by infants in Hanoi and Hochiminh city in comparison with the Tolerable Daily Intake (TDI) proposed by Health Canada (Oostdam et al., 1999).

that although intake of DDTs by most infants is below the guideline proposed by Health Canada (Oostdam et al., 1999) in average, intake by some individuals is close to or exceeds this guideline. This fact may raise greater concerns on infant health because children are highly susceptible to effects from environmental contaminants.

4. Conclusions

To our knowledge, this is the most recent and extensive studies on the contamination by persistent OCs in human breast milk from Vietnam. Our data clearly indicate the recent usage of DDTs in both North and South of Vietnam. Number of childbirth and age of mothers played an important role in influencing the OC burdens in lactating women. The current status of DDT contamination in Vietnamese population is still high with a number of those even being close to the threshold level reported to cause incidences of pre-term birth. Estimated breast-fed children intakes of some chemicals exceeded the guideline proposed by Health Canada. Comprehensive studies on the temporal trends of OCs in Vietnam are therefore necessary to understand the source and evaluate possible long-term impacts of OCs in tropical ecosystem.

Acknowledgements

The authors wish to thank the staff of the Center for Environmental Technology and Sustainable Development, Hanoi National University, Hanoi, and University of Agriculture and Forestry, Hochiminh City, Vietnam for their valuable support during our sampling surveys. This study was supported by a Grant-in-Aid from the Scientific Research on Priority Areas (Project Nos. 13027101) of the Japanese Ministry of Education, Science, Sports, Culture and Technology and for Scientific Research (Project No. 12308030) of Japan Society for the Promotion of Science. Financial assistance was also provided by "Formation and Behavior of Dioxins and their Related Persistent Organic Pollutants in Uncontrolled Combustion Processes" from the Waste Management Research Grants of the Ministry of the Environment; the Sumitomo Foundation and the Core University Program between Japan Society for the Promotion of Science (JSPS) and National Center for Natural Science and Technology, Vietnam (NCST). This study was also supported by "21st Century COE Program" from the Japanese Ministry of Education, Science, Sports, Culture and Technology and Research Revolution 2002 (RR 2002) Project for sustainable Coexistence of Human, Nature and the Earth (FY 2002). The awards of the JSPS Postdoctoral Fellowship for Researchers in Japan to Dr. M. Watanabe (No. 04166) are acknowledged.

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