Environ. Sci. Technol. **2005**, *39,* 4714−4720

Organochlorine Contaminants in Human Adipose Tissues from China: Mass Balance Approach for Estimating Historical Chinese Exposure To DDTs

HARUHIKO NAKATA,*,† TETSUYA NASU,† SHIN-ICHI ABE,‡ TAKESHI KITANO,‡ QIYUAN FAN,§ WEIHUA LI,∥ AND XUCHENG DING∥

Graduate School of Science and Technology, Kumamoto University 2-39-1 Kurokami, Kumamoto 860-8555 Japan, Faculty of Science, Kumamoto University 2-39-1 Kurokami, Kumamoto 860-8555 Japan, Zunyi Medical College, 143 Da Lian Road, Guizhou 563003, P. R. China, and Shanghai Institute of Planned Parenthood Research, 2140 Xie Tu road, Shanghai, 200032, P. R. China

Concentrations of persistent organochlorines (OCs), such as DDTs, hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), and chlordane compounds (CHLs) were determined in 34 human adipose tissues collected from Guizhou Province, southern China, during 2002. DDT was the predominant contaminant among OCs analyzed; concentrations ranged from 420 to 20 000 ng/g on a lipid wt basis (average ± sd: 5700 ± 4100 ng/g). Concentrations of DDTs and the ratio of *p,p′*-DDT/ΣDDT in humans in China were significantly higher than those reported for developed countries. DDT levels in humans in Guizhou Province were comparable to those from Shanghai City, implying the presence of significant sources of DDTs in inland and coastal areas in China. Age-dependent accumulation of HCH concentration was found in this study, possibly because of the considerable reduction in average dairy intake (ADI) of HCHs by Chinese during the recent two decades. On the basis of the information of the ADI rates, half-lives, and body burdens of DDTs, the magnitude of historical DDT exposures by Chinese was estimated. This suggested that approximately 80% of DDTs deposited in Chinese adipose tissues was accumulated before the 1990s. The monitoring of OC levels and epidemiological studies are needed in China to understand the status of contamination and the risks to humans.

Introduction

Environmental contamination by organochlorines (OCs) has been a major concern over the past four decades in the world because of their persistence, long-range transport, and biological effects. Most of the developed nations have already banned or restricted the production and usage of OCs, such as DDTs, hexachlorocyclohexanes (HCHs), hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), and chlordane compounds (CHLs). However, recent studies from Asian developing countries have reported the elevated concentrations of organochlorine pesticides in various environmental media, suggesting the presence of significant sources of these compounds (*1*−*3*). For example, several evidences point that DDTs are still being used in China (*4*, *5*).

It is well-known that organochlorine pesticides had been produced and widely used in China between the 1950s and the 1980s (*6*). The amount of DDT and HCH production in China was 0.4 and 4.9 million tonnes, respectively, accounting for 33 and 20% of the total world production (*7*). In recent years, several studies have examined the status of OCs contamination in water, sediment, soil, and wildlife in China. The results showed that DDT concentrations in mussels from coastal waters were extremely high, about several tens μg/g (lipid wt basis), which was 1−3 orders of magnitude greater than those found from other Asian countries (*2*, *8*). The ratios of *p,p′*-DDT/ΣDDT were also elevated in marine fish from Hangzhou Bay (20−55%) (*9*) and Xiamen coastal waters (40−65%) (*10*). In terms of human samples, it has been reported that DDT concentrations were high in breast milk collected from Hong Kong, Guangzhou (*11*), Beijing (*12*), and Dalian (*13*). These observations strongly suggest continuing use of DDTs in China, although there are reports of banning of production of this compound in 1983 (*14*). Earlier investigations were concentrated around coastal areas in eastern China, with little information available for inland regions. Monitoring OC residue levels in inland areas of China will provide better indications of contamination and distribution of OCs in this country. Furthermore, there has been a lack of relevant data on OC levels in human adipose tissues in China. Data on OCs in Chinese adipose tissues may provide the basis for estimating the amounts of historical exposures to OCs and also for assessing their human health risks in China.

In a previous study, we reported OC levels in 39 foodstuffs and 5 human mammary glands collected from Shanghai city during 2000 and 2001 (*8*). The results showed that the ADI of DDTs has significantly decreased, from 42 μg/person/day in 1973 (*15*) to 2.16 μg/person/day in 2001 (*8*). Surprisingly, the ADI of DDTs in China was comparable to that in developed nations, such as Japan (1.30 μg/person/day), the United States (1.30 μg/person/day), and Australia (1.95 μg/person/day) in the late 1980s (*16*). Nevertheless, average DDT concentration in humans in Shanghai city (7600 ng/g lipid wt) was approximately 1 order of magnitude greater than the levels in Korea (780 ng/g) (*17*) and the United States (677 ng/g) (*18*). These findings suggest that Chinese were exposed to elevated quantities of DDTs in the past, some of which still continue to be present in their bodies. However, no investigation has been conducted to estimate the historical exposure to DDTs by Chinese, possibly because of the lack of concentration data in human adipose tissues in China.

In this study, we determined OCs, such as DDTs, HCHs, HCB, PCBs, and CHLs, in 34 human adipose tissues collected from Guizhou Province, an inland province in southern China, during 2002. We examined levels and patterns of OC accumulation in Chinese. Furthermore, the amount of historical exposure to DDTs was estimated on the basis of the rate of decline in ADI, the half-lives of DDT in humans, and the body burden of DDTs obtained in this study.

* Corresponding author phone: +81-96-342-3380; fax: +81-96-342-3380; e-mail: nakata@sci.kumamoto-u.ac.jp.
† Graduate School of Science and Technology, Kumamoto University.
‡ Faculty of Science, Kumamoto University.
§ Zunyi Medical College.
∥ Shanghai Institute of Planned Parenthood Research.

10.1021/es050493d CCC: $30.25

Published on Web 05/24/2005

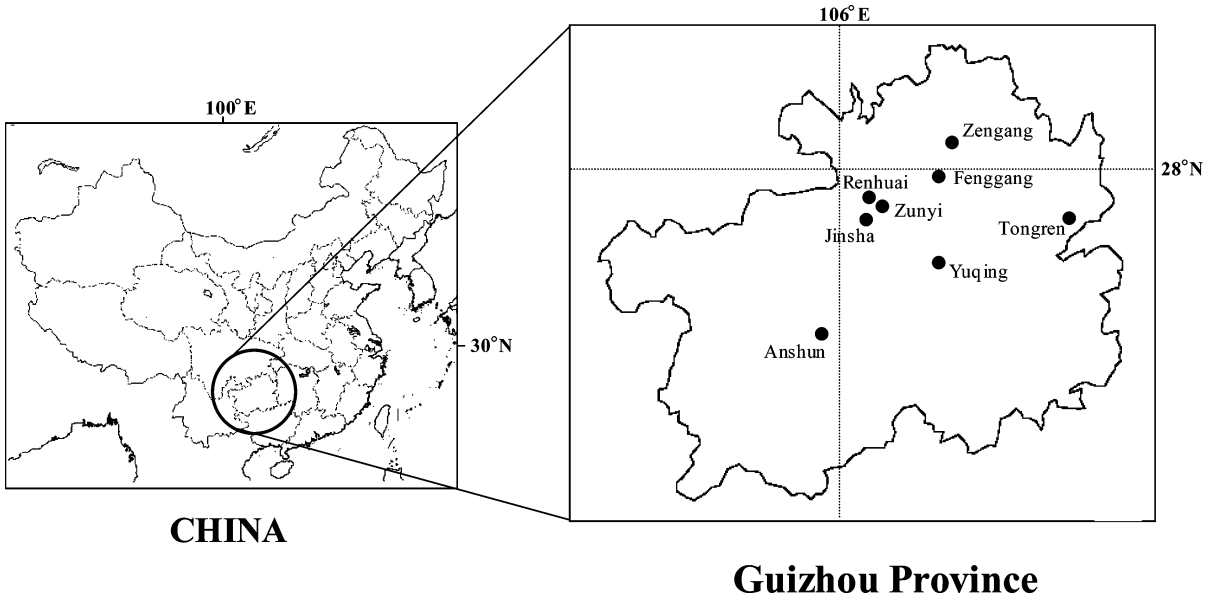

FIGURE 1. Map showing sampling sites of human adipose tissues analyzed in this study.

Experimental Section

Samples. Thirty-four human adipose tissues (17 males and 17 females) were obtained in hospitals in eight locations of Guizhou Province, southern China during 2002 (Figure 1). These samples were taken from randomly selected patients. Informed consent was obtained from donors for use of adipose samples for scientific investigations. All the samples were sealed in plastic bags and stored at −20 °C until chemical analysis.

Chemical Analysis. OCs were analyzed according to the method described previously (*8*). Briefly, approximately 1−2 g of adipose tissue samples were ground with sodium sulfate and extracted with mixed solvents of dichloromethane and hexane (8:1) using a Soxhlet apparatus. The lipid content was determined gravimetrically by using an aliquot of the extract. Seven ^{13}C labeled tri- through decachlorinated biphenyls were spiked into the extracts as surrogate standards. The extract was added into dry florisil column to remove lipids, followed by elution with acetonitrile containing 20% hexane-washed water. The eluates were collected in a separatory funnel containing hexane and hexane-washed water. After shaking and partitioning, the hexane layer was concentrated and passed through a florisil-packed glass column for fractionation. The first fraction eluted with hexane contained HCB, PCB congeners, *p,p′*-DDE, and *trans*-nonachlor, while the second fraction eluted with 20% dichloromethane in hexane contained chlordane compounds (*trans*- and *cis*-nonachlors and chlordanes and oxychlordane), *p,p′*-DDD, *p,p′*-DDT, and HCHs (α-, *β*-, and *γ*-HCH isomers). Each fraction was concentrated and injected into GC-MS (Hewlett-Packard 6890 GC coupled with a 5973 mass selective detector) and GC-ECD (Hewlett-Packard 6890 GC coupled with a ^{63}Ni electron capture detector) for quantification. A DB-1 capillary column (30 m × 0.25 mm i.d., J&W Scientific Inc., U.S.) was used in this study. The oven temperature was set from 70 °C to 160 °C at a rate of 10 °C/min and held for 10 min. Then, the temperature was increased to 260 °C at a rate of 2 °C/min with a final hold time of 20 min. The temperatures of injector and detector were set at 270 and 300 °C, respectively. Helium and nitrogen were used as carrier and makeup gases, respectively. Recovery percentages of PCBs congeners ranged from 79 to 118%. As for organochlorine pesticides, the recoveries of analytes ranged from 88 to 107%, and their concentrations were not corrected for recovery percentages. The detection limit was 0.04 ng/g for DDTs, 0.03 ng/g for HCHs, 0.02 ng/g for CHLs, and 0.01 ng/g for HCB and PCBs.

For quality assurance and quality control, our laboratory participated in Inter Laboratory Comparison Exercise for Persistent Organochlorines in Marine Mammal Tissues, organized by the National Institute of Standards and Technology (NIST) and National Oceanic and Atmospheric Administration (NOAA). Standard reference material (SRM 1945) was analyzed for selected PCB congeners and organochlorine pesticides. Reliable results were obtained by comparison of generated data from our laboratory with those of standard reference values (*19*).

Statistical Analysis. The significant difference of OC concentrations between samples was confirmed by employing Wilcoxon *U*-test. Spearman's rank test was also used to examine a significant correlation. A software from Excel Statistics (Esumi, Tokyo) was used in this study.

Results and Discussion

Levels. Concentrations of OCs and lipid content in human adipose tissues analyzed are shown in Table 1. Residue levels in samples were in the order of DDTs > HCHs > HCB = PCBs > CHLs. Mean concentration and standard deviation (sd) of DDTs was 5700 ± 4100 ng/g (lipid wt basis), and the highest concentration was found in a 68-year-old male (20 000 ng/g; Table 1). The major component of DDTs was *p,p′*-DDE, followed by *p,p′*-DDT and *p,p′*-DDD. HCH concentrations in humans showed a large variation among samples, ranging from 47 and 3300 ng/g, lipid wt (mean ± sd = 630 ± 720 ng/g; Table 1). *β*-HCH was the dominant isomer, accounting for 97−99% of total HCH concentrations. HCB and PCBs levels in this study were almost similar, ranging from 7.6 to 100 ng/g (40 ± 19 ng/g) and from 1.1 to 110 ng/g (31 ± 32 ng/g), respectively. Hexachlorobipnenyls, such as CB-153 and 138, were the predominant PCB congeners. CHL concentrations in adipose tissues were generally low, within the range of <0.02 to 13 ng/g (mean ± SD: 4.4 ± 3.2 ng/g). Oxychlordane and *trans*-nonachlor were the major components of ΣCHL.

OC concentrations measured in this study were compared with those in human adipose tissues reported for other countries (Table 2). DDTs levels in humans in China were apparently higher than those reported in Japan (*20*, *21*), the United States (*18*), and Korea (*17*) and were comparable to those in Mexico (*22*). The ratio of *p,p′*-DDT to ΣDDT in

TABLE 1. Concentrations of Organochlorine Contaminants (ng/g Lipid Wt Basis) in Human Adipose Tissues Collected from Guizhou Province, China

sample no.	sex	age (yr)	lipid (%)	*p,p′* -DDE	*p,p′* -DDD	*p,p′*-DDT	DDTs	a-HCH	b-HCH	g-HCH	HCHs	HCB	PCBs	t-nona	c-nona	t-CA	c-CA	OXY	CHLs
1	M	19	82	7900	33	410	8300	<0.03	330	<0.03	330	45	16	<0.02	<0.02	<0.02	<0.02	<0.02	<0. 02
2	M	24	68	1500	21	240	1800	1.1	86	<0.03	86	39	27	2.2	<0.02	<0.02	<0.02	2.0	4.2
3	M	28	84	8800	290	1200	10 000	1.9	470	<0.03	470	29	18	1.2	<0.02	1.5	<0.02	<0.02	2.8
4	M	30	88	3000	15	400	3400	1.0	210	<0.03	210	31	8.5	0.59	<0.02	<0.02	<0.02	1.2	1.8
5	M	32	88	3700	34	790	4500	1.4	140	<0.03	140	42	13	1.2	<0.02	<0.02	<0.02	0.37	1.5
6	M	35	90	9900	120	1400	11 000	4.4	1800	<0.03	1800	69	92	1.3	<0.02	<0.02	<0.02	1.6	2.8
7	M	40	59	6400	30	290	6700	2.7	710	<0.03	710	24	65	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
8	M	42	88	3100	23	430	3600	1.1	270	<0.03	270	39	21	1.2	<0.02	<0.02	<0.02	1.5	2.7
9	M	44	87	5200	61	690	6000	1.3	110	<0.03	110	32	35	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
10	M	45	82	2000	35	280	2300	1.6	570	<0.03	570	35	12	3.6	<0.02	<0.02	<0.02	2.6	6.1
11	M	50	82	4900	32	320	5300	4.7	420	<0.03	420	16	10	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
12	M	55	80	3900	24	400	4300	1.0	260	<0.03	260	100	110	1.4	<0.02	<0.02	<0.02	1.6	3.0
13	M	56	78	2500	120	640	3300	1.6	220	<0.03	220	52	9.2	1.1	<0.02	1.5	<0.02	<0.02	2.6
14	M	60	85	9200	100	3300	13 000	4.1	830	<0.03	830	67	98	3.6	<0.02	<0.02	1.5	2.9	8.0
15	M	64	89	1700	16	160	1900	7.6	340	<0.03	340	17	3.9	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
16	M	68	83	16 000	180	3400	20 000	7.7	3300	<0.03	3300	69	25	3.1	<0.02	<0.02	<0.02	3.9	7.0
17	M	72	83	7700	27	590	8300	3.3	2000	<0.03	2000	41	94	7.1	<0.02	<0.02	<0.02	3.8	11
18	F	11	81	3200	88	3500	6800	0.93	85	<0.03	85	43	19	1.9	<0.02	<0.02	2.1	<0.02	4.0
19	F	12	82	2000	23	390	2400	0.89	130	<0.03	130	65	16	<0.02	<0.02	<0.02	2.0	<0.02	2.0
20	F	17	77	380	4.7	32	420	0.89	47	<0.03	47	7.6	1.1	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
21	F	27	87	2700	13	290	3000	0.78	220	<0.03	220	20	3.1	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
22	F	28	75	4100	50	1100	5300	<0.03	170	<0.03	170	32	15	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
23	F	29	77	2100	42	200	2300	<0.03	510	<0.03	510	73	8.3	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
24	F	30	88	2700	19	510	3200	1.0	210	<0.03	210	21	75	3.8	<0.02	3.3	3.5	<0.02	11
25	F	35	78	1900	32	330	2300	0.93	200	<0.03	200	32	7.8	<0.02	<0.02	2.5	<0.02	<0.02	2.5
26	F	38	78	3000	110	980	4100	1.4	400	<0.03	400	43	5.7	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
27	F	45	84	3500	28	640	4200	1.7	660	<0.03	660	35	25	1.8	<0.02	<0.02	<0.02	0.70	2.5
28	F	46	85	210 0	31	400	250 0	1.1	380	<0. 03	380	17	6.1	<0. 02	<0. 02	1.7	<0. 02	<0. 02	1.7
29	F	48	85	8500	38	1400	9900	1.9	1000	<0.03	1000	43	50	2.2	<0.02	<0.02	0.54	2.2	4.9
30	F	50	86	2900	15	370	3300	2.6	1400	<0.03	1400	30	24	1.5	<0.02	<0.02	<0.02	1.5	3.0
31	F	51	85	9000	70	970	10 000	2.9	1200	<0.03	1200	29	10	<0.02	<0.02	<0.02	<0.02	2.2	2.2
32	F	55	82	5600	31	890	6500	2.4	400	<0.03	400	50	23	1.2	<0.02	<0.02	1.6	<0.02	2.8
33	F	59	83	3300	36	550	3900	1.0	190	<0.03	190	28	11	0.70	<0.02	<0.02	<0.02	1.5	2.2
34	F	74	84	8900	57	2300	11 000	2.9	2100	<0.03	2100	37	87	4.0	<0.02	4.3	3.1	1.4	13
average ± SD (total)		42 ± 17	82 ± 6.1	4800 ± 3400	54 ± 57	880 ± 920	5700 ± 4100	2.3 ± 1.8	630 ± 720	<0.03	630 ± 720	40 ± 19	31 ± 32	2.2 ± 1.6	<0.02	2.5 ± 1.1	2.0 ± 1.0	1.9 ± 1.0	4.4 ± 3.2
range (t otal)		11−74	59−90	380−16 000	4.7−29 0	32−3500	420−20 000	<0.03−7.7	47−3300	na[b]	47−3300	7.6−100	1.1−110	<0.02−7.1	na	<0.02−4.3	<0.02−3.5	<0.02−3.9	<0.02−13
average ± SD (male)		45 ± 16	82 ± 7.9	5700 ± 3800	68 ± 74	880 ± 990	6700 ± 4800	2.9 ± 2.2[a]	710 ± 870	<0.03	710 ± 870	44 ± 22	39 ± 37	2.3 ± 1.8	<0.02	1.5 ± 0.0	1.5	2.1 ± 1.1	4.5 ± 2.9
range (male)		19−7	59−90	1500−16 000	15−290	160−3400	1800−20 000	<0.03−7.7	86−3300	na	86−3300	16−100	3.9−110	<0.02−7.1	na	<0.02−1.5	na	<0.02−3.9	<0.02−11
average ± SD (female)		39 ± 17	82 ± 3.9	3900 ± 2600	40 ± 28	870 ± 870	4800 ± 3100	1.6 ± 0.8[a]	550 ± 570	<0.03	550 ± 570	36 ± 17	23 ± 25	2.1 ± 1.2	<0.02	2.9 ± 1.1	2.1 ± 1.1	1.6 ± 0.6	4.3 ± 3.6
range (f emale)		11−74	75−88	380−9000	4.7−110	32−3500	420−11 000	0.78−2.9	47−2100	na	47−2100	7.6−73	1.1−87	<0.02−4.0	na	<0.02−4.3	<0.02−3.5	<0.02−2.2	<0.02−13

[a] p <0.05. [b] na: not available.

TABLE 2. Concentrations of Organochlorine Compounds in Human Adipose Tissues (ng/g Lipid Wt) Collected from Various Countries in Recent 10 Years

country	year	n	DDTs[c]	HCHs[f]	HCB	PCBs	CHLs[h]	reference
China (Guizhou Province)	2002	34	5700 ± 4100	630 ± 720	40 ± 19	31 ± 32	4.4 ± 3.2	this study
China (Shanghai City)[a]	2001	5	7600 ± 5800	7400 ± 6100	64 ± 37	70 ± 39	42 ± 31	8
Japan	1999	20	2200 ± 2000	680 ± 770	60 ± 38	2100 ± 1300	310 ± 180	20
Japan	1998	22	780 ± 470	330 ± 180	41 ± 17	1700 ± 820	230 ± 160	21
Mexico	1997−1998	60	5661 ± 5020[d]	156 ± 118	58 ± 29	NA[j]	NA	22
Jordan	1996	50	3200 ± 1100	1400 ± 450	120 ± 60	NA	NA	23
Turkey	1995−1996	56	2130 ± 1026[e]	520 ± 339	33 ± 36	NA	NA	24
United States (NY state)[b]	1994−1996	194	677	45[g]	20	361	116[i]	18
Korea	1994−1995	32	1100	190	20	400	NA	17

[a] The mammary glands were analyzed. [b] Samples were collected from patients of breast cancer with no recurrence. [c] Sum of *p,p′*-DDE, *p,p′*-DDT, and *p,p*′-DDD. [d] Sum of *p,p′*-DDE, *p,p′*-DDT, *p,p′*-DDD, and *o,p′*-DDT. [e] *p,p′*-DDE, *p,p′*-DDT. [f] Sum of α-HCH, *β*-HCH, and *γ*-HCH. [g] *β*-HCH. [h] Sum of *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor, *trans*-nonachlor, and oxychlordane. [i] *trans*-Nonachlor and oxychlordane. [j] NA: not available.

humans in China was 14 ± 8%; this value was significantly higher than that in Japan (8.2 ± 1.9%) (*20*), the United States (3.0%) (*18*), and Korea (6.1%) (*17*). These results suggest that the Chinese population has been exposed to DDTs more recently than the populations in developed nations. DDT levels in humans in Guizhou Province were somewhat similar to that in Shanghai City (Table 2). This may suggest the presence of significant sources of DDTs in inland areas, similar to that in coastal regions in China. Distribution of OCs in pine needles in China also suggested new inputs of *p,p′*-DDT and use of dicofol in southwest inland areas in this country (*25*).

HCH concentrations in humans in this study were higher than those in the United States (*18*), Korea (*17*), and Mexico (*22*) and comparable to those in Japan (*20*, *21*) and Turkey (*24*) (Table 2). HCH levels in humans in Shanghai City were 1 order of magnitude greater than those in Guizhou Province (Table 2). It has been reported that use of technical HCHs was concentrated in the southeastern part of China (*26*). Especially, Jiangsu and Zhenjian Provinces, near Shanghai City, were areas with the highest usage of HCHs in China, whereas the HCH use in Guizhou Province was relatively small (*26*). Geographical variation in HCHs usage in China might explain the differences in concentrations among human tissue samples from different locations. In addition, ages of humans analyzed may be attributed to the observed differences in HCH concentrations. In this study, it was found that HCH levels significantly increased with age. It has been reported that HCH residues in humans in Shanghai City were anomalously high in older women, aged >80 years (*8*). The age of humans from Shanghai City analyzed (65 ± 19 years old) was approximately 20 years older than that from Guizhou Province (42 ± 17 years old). This difference may partly influence the variations in HCH levels in samples collected between the two locations. The profiles of age- and sex-dependent accumulation of OCs in Chinese will be discussed later.

HCB concentrations were comparable to those in Shanghai City and other countries (Table 2). Regarding PCBs, the levels in humans in China were low, approximately 1−2 orders of magnitude lower than those in Japan (*20*, *21*), the United States (*18*), and Korea (*17*) (Table 2). In general, low PCB residues were also found in surface water, sediment (*27*), foodstuffs (*8*), mussel (*2*), fish, and birds (*9*) collected in China, in comparison with those in other Asian countries. These results indicate small PCB usage in this country. CHL levels in Chinese were significantly lower than those in Japanese (*20*, *21*) and the U.S. people (*18*) (Table 2). Similar to HCHs, CHL levels in humans in Guizhou Province were 1 order of magnitude lower than those in Shanghai City, implying spatial difference in CHL use in China. It has been reported that large variation in CHL levels was found in fish samples collected between marine and freshwater environments in China (*9*), although CHLs are still being used for termite control in this country (*25*). Further investigations are needed to understand the geographical distribution of OC contamination in China.

Sex- and Age-Dependent Accumulation. In general, OC concentrations in males were higher than those in females (Table 1). However, there was no significant sex-related difference in all OC concentrations, with an exception of α-HCH, which showed higher residues in males than in females ($p < 0.05$).

Significant relationship was found between age and HCH concentrations in this study, while there was no such correlation found for other contaminants analyzed (Figure 2). It is well-known that age-dependent accumulation of OCs in humans have linked to various factors, such as the biological status of the samples, the locations where samples are collected, the variation of intake, the historical usage, and the number of samples. While it is difficult to identify what is the major contributor to decide the accumulation profile in this study, we understand that this difference might be due to the variation in historical intakes of OCs. Our previous study has examined OC concentrations in 39 food items obtained in China during 2000 and 2001 (*8*). The results suggested that the ADI of HCHs in Chinese has dramatically decreased from 532 μg/person/day in 1980 (*6*) to 0.13 μg/person/day in 2001 (*8*). Such a great reduction of ADI of HCHs during the past 20 years might contribute to the wide range of exposures between young and old generations, which could result in the age-dependent accumulation observed in this study.

On the other hand, the ADI of DDTs has decreased in the past, but the reduction rate is smaller than that of HCHs. The estimated ADI of DDTs was 42 μg/person/day in 1973 (*15*), and it has decreased to 2.16 μg/person/day in 2001 (only $^1/_{20}$ reduction during the past 30 years) (*8*). This might be due to the more persistent nature of DDTs than HCHs in the environment. In addition, there are many evidences that DDT has been used recently in China; nevertheless, little data are available about the present use of HCHs in this country. The less significant correlation between age and DDT residues in Chinese might partly be due to the continued and present usage of this pesticide even now.

Mass balance Approach for Estimating Historical Exposure to DDTs by Chinese. On the basis of the available ADI value of DDTs in Chinese and the DDT concentration we measured in this study, we estimated the historical exposures to DDT by Chinese. First, to estimate the total amount of DDT intake and body burdens during 1973 and 2001, we used the data of the reduction rate of ADI during these periods and the half-life of DDTs. However, this estimation involves uncertainties, especially the half-life of

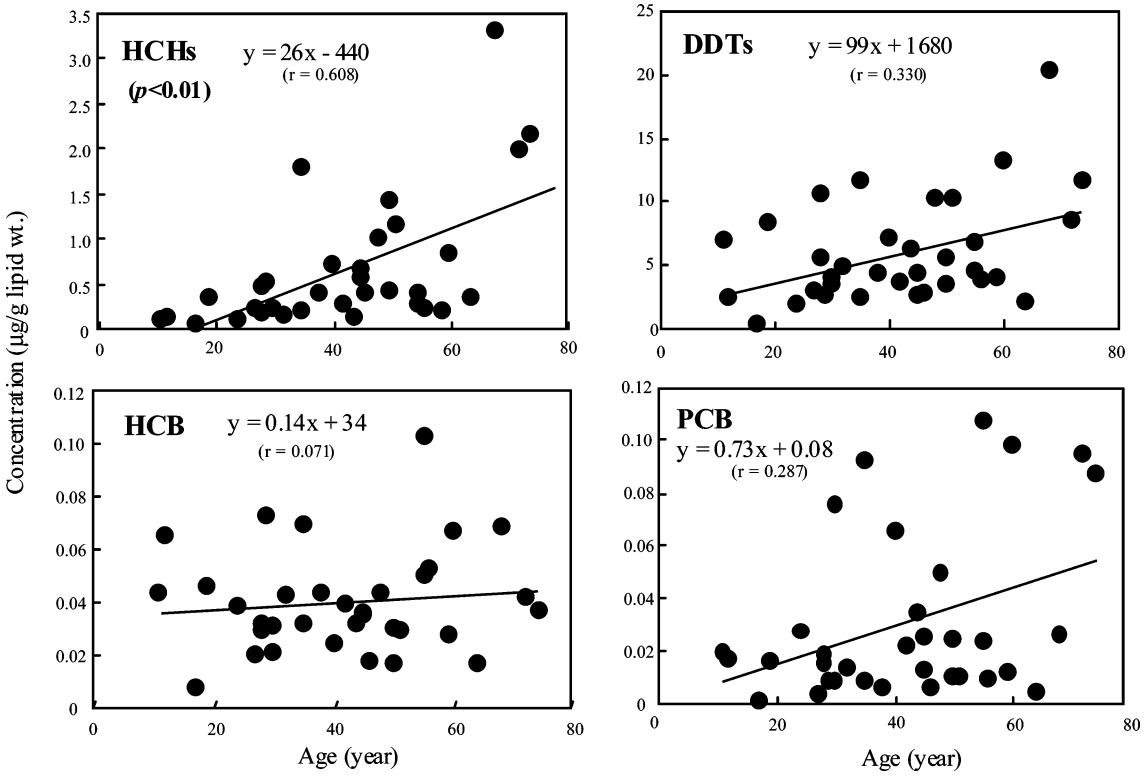

FIGURE 2. Age-dependent accumulation of organochlorines in human adipose tissues from Guizhou Province, China.

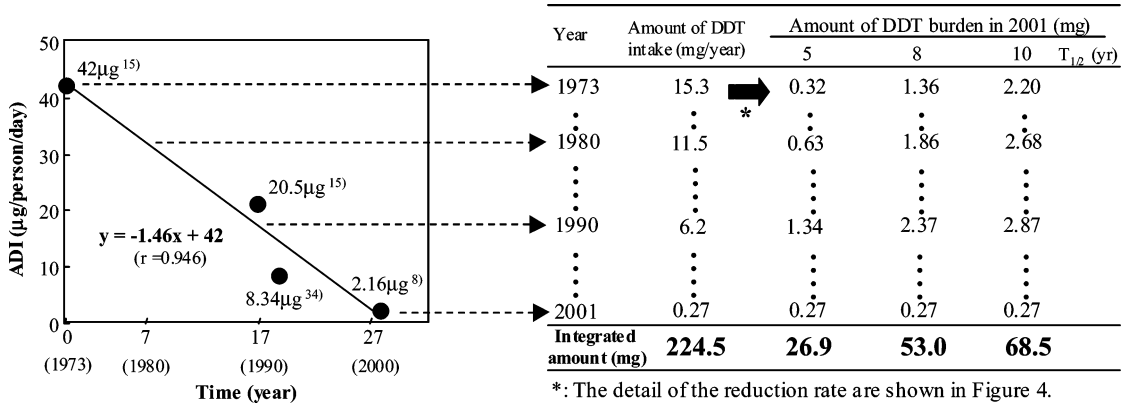

FIGURE 3. Estimation of the amounts of DDTs intake rates by Chinese during 1973 and 2001. Reduction rates of ADI of DDTs in Chinese and half-lives of DDTs in humans were used for this estimation.

DDTs, because this is not available for humans. To obtain DDT half-life, we examined the relationship between the half-lives and lipophilicity of environmental contaminants.

It has been reported that the half-life of a compound generally increases with the *n*-octanol/water partitioning coefficients (log *K*ow) in rats and humans (*27*). For 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD), with a log *K*ow of 6.8 (*29*), the half-lives in humans range from 5.8 to 9.7 years (average: 7.78 yr) (*30*). While the reported half-lives of PCBs showed large variations in humans, more lipophilic congeners, such as hexa- and heptachlorinated biphenyls (Log *K*ow: 6.7−7.1) (*29*), had relatively longer half-lives, almost 5 years or more (*31*). In the DDTs, the log *K*ows are 6.9, 7.0, and 6.2 for *p,p′*-DDT, *p,p′*-DDE, and *p,p′*-DDD (*32*), which are similar to those of TCDD and highly chlorinated biphenyls. These facts imply that the half-lives of DDT compounds are also long, probably in the range of 5−10 years in humans. The metabolic half-life of DDTs in marine mammals has been estimated as 6.3 years (*33*). In this context, we used three different half-life values of DDTs, such as 5, 8, and 10 years, to calculate the amount of DDT intake and body burden during 1973 and 2001.

The correlation between time (year) and ADI of DDTs can be expressed as follows (Figure 3):

$$y = -1.46x + 42 \ (r = 0.946)$$

where x and y are time (year) and ADI of DDT (μg/person/day), respectively. On the basis of this equation, the amount of DDT intake in 1973 was calculated as 15.3 mg. However, this value has decreased constantly, and the reduction rate depended on the DDT half-life. When the half-life of DDT in humans is defined as 5 years, the DDT reduction rate can be expressed as follows (Figure 4):

$$y = 15.3e^{-0.139x}$$

where x and y are time difference between year and 1973 (year) and DDT body burden (mg) in adipose tissues, respectively. On the basis of this equation, it was calculated that the amount of DDTs decreased from 15.3 mg in 1973 to

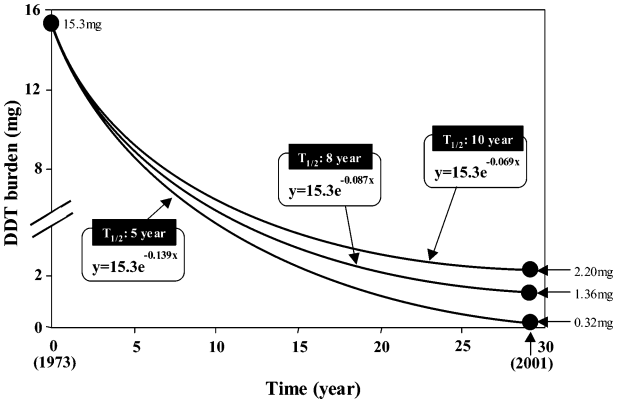

FIGURE 4. Estimation of the reduction rates of DDTs ingested in 1973 in 2001. Three different scenarios of half-lives of DDTs in humans, such as 5, 8, and 10 years, were used for calculation.

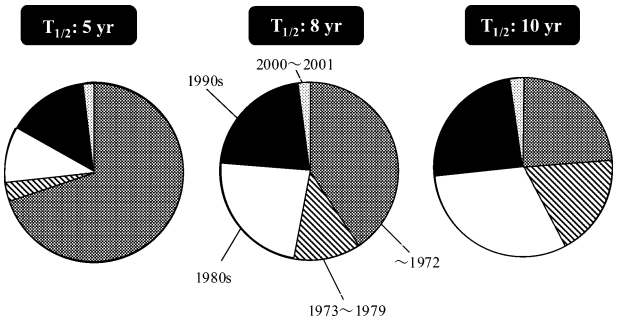

FIGURE 5. Percentage of DDT body burdens in Chinese fat in five different periods: pre-1972, 1973−1979, 1980s, 1990s, and 2000−2001. The half-lives of DDTs, such as 5, 8, and 10 years, were used in the calculation.

0.32 mg in 2001 (Figure 4). Similarly, DDT intake and burden during 1974 and 2001 were estimated. Consequently, the DDT burden in Chinese during 1973 and 2001 was estimated as 26.9 mg (Figure 3). Because total amount of DDT intake during 1973 and 2001 was calculated as 224.5 mg (Figure 3), this suggests that approximately 90% of DDTs that have accumulated since 1973 have been excreted until 2001.

We have conducted similar calculations by using different DDT half-lives, 8 and 10 years. The body burden and the reduction rates of DDTs are shown in Figures 3 and 4, respectively. When DDT half-lives were defined as 8 and 10 years, the estimated body burdens of DDT during the past 30 years were 53.0 mg and 68.5 mg, respectively (Figure 3). In this case, the body burdens in the 1980s were relatively higher than that in other periods, such as 1973−1979, 1990s, and 2000−2001.

A recent clinical study examined 130 adults in China (63 males and 67 females), aged between 35 and 49 years old, and reported that average body weight and fat percentages were 74.6 kg and 27.1% in males and 63.9 kg and 36.9% in females (*35*). On the basis of these values, we calculated DDT body burden in Chinese adipose tissues by multiplying DDT concentrations analyzed in this study for the ages 35−50 years (male: 6, female: 6) and the average weight of body fat (male: 20.2 kg, female: 23.5 kg). The DDT body burden in Chinese was calculated as 96.5 mg in males and 86.3 mg in females, and the average body burden (91.4 mg) is higher than the amount of DDT burdens estimated previously (Figure 3). It is reasonable to explain the differences in DDT amounts ($T_{1/2}$: 5 years, 64.5 mg; $T_{1/2}$: 8 years, 38.4 mg; $T_{1/2}$: 10 years, 22.9 mg) for accumulations before 1972 in Chinese.

To understand the historical trends in DDT accumulation in Chinese, the percentage of DDT burdens from five different periods, such as pre-1972, 1973−1979, 1980s, 1990s, and 2000−2001, to total body burden (91.4 mg of DDTs) is shown in Figure 5. When DDT half-life was set as 5 years, the percentage of DDT depots that accumulated before 1972 was apparently higher than that of other periods. However, the accumulation rates were higher for 1973−1979 and the 1980s when the half-lives of DDTs increasd (Figure 5). Interestingly, in all of the cases, DDT accumulated in the 1990s occupied only 17−26% to the total amounts. These results suggest that approximately 80% of DDT burdens have been accumulated before the 1990s, at least 10 years ago in human tissues in China. Probably, this is explained by the persistent profile of DDTs in organisms as well as a long life span of humans. In addition, this study implies that contamination of DDTs continues, while the ADI has decreased in recent years. Additional research on monitoring of OCs, especially DDTs, and epidemiological studies are needed to understand the status of contamination and the human health risks of these compounds in China.

Literature Cited

(1) Tanabe, S.; Iwata, H.; Tatsukawa, R. Global contamination by persistent organochlorines and their ecotoxicological impact on marine mammals. *Sci. Total. Environ.* **1994**, *154*, 163−177.

(2) Monirith, I.; Ueno, D.; Takahashi, S.; Nakata, H.; Sudaryanto, A.; Subramanian, A.; Karuppiah, S.; Ismail, A.; Muchtar, M.; Zheng, J.; Richardson, B. J.; Prudente, M.; Hue N. D.; Tana, T. S.; Tkalin, A. V.; Tanabe, S. Asia-Pacific mussel watch: monitoring contamination of persistent organochlorine compounds in coastal waters of Asian countries. *Mar. Pollut. Bull.* **2003**, *46*, 281−300.

(3) Kannan, K.; Tanabe, S.; Giesy, J. P.; Tatsukawa, R. Organochlorine pesticides and polychlorinated biphenyls in foodstuffs from Asian and Oceanic countries. *Rev. Environ. Contam. Toxicol.* **1997**, *152*, 1−55.

(4) Zhou, J. L.; Maskaoui, K.; Qiu, Y. W.; Hong, H. S.; Wang, Z. D. Polychlorinated biphenyl congeners and organochlorine insecticides in the water column and sediments of Daya Bay, China. *Environ. Pollut.* **2001**, *113*, 373−384.

(5) Gong, Z. M.; Tao, S.; Xu, F. L.; Dawson, R.; Liu, W. X.; Cui, Y. H.; Cao, J.; Wang, X. J.; Shen, W. R.; Zhang, W. J.; Qing, B. P.; Sun, R. Level and distribution of DDT in surface soils from Tianjin, China. *Chemosphere* **2004**, *54*, 1247−1253.

(6) Li, Y. F.; Cai, D. J.; Singh, A. Technical hexachlorocyclohexane use trends in China and their impact on the environment. *Arch. Environ. Contam. Toxicol.* **1998**, *35*, 688−697.

(7) Zhang, G.; Parker, A.; House, A.; Mai, B.; Li, X.; Kang, Y.; Wang, Z. Sedimentary records of DDT and HCH in the Pearl River Delta, South China. *Environ. Sci. Technol.* **2002**, *36*, 3671−3677.

(8) Nakata, H.; Kawazoe, M.; Arizono, K.; Abe, S.; Kitano, T.; Shimada, M.; Li, W.; Ding, X. Organochlorine pesticides and polychlorinated biphenyl residues in foodstuffs and human tissues from China: Status of contamination, historical trend and human dietary exposure. *Arch. Environ. Contam. Toxicol.* **2002**, *43*, 473−480.

(9) Nakata, H.; Hirakawa, Y.; Kawazoe, M.; Nakabo, T.; Arizono, K.; Abe, S.; Kitano, T.; Shimada, H.; Watanabe, I.; Li, W.; Ding, X. Concentrations and composition of organochlorine contaminants in sediments, soils, crustaceans, fishes, and birds collected from Lake Tai, Hangzhou Bay, and Shanghai city region, China. *Environ. Pollut.* **2005**, *133*, 415−429.

(10) Klumpp, D. W.: Huasheng, H.; Humphrey, C.; Xinhong, W.; Codi, S. Toxic contaminants and their biological effects in coastal waters of Xiamen, China. I. Organic pollutants in mussel and fish tissues. *Mar. Pollut. Bull.* **2002**, *44*, 752−760.

(11) Wong, C. K. C.; Leung, K. M.; Poon, B. H. T.; Lan, C. Y.; Wong, M. H. Organochlorine hydrocarbons in human breast milk collected in Hong Kong and Guangzhou. *Arch. Environ. Contam. Toxicol.* **2002**, *43*, 364−372.

(12) Yu, H.; Zhu, Z.; Zhao, X.; Zhang, X.; Wang, D. Levels of organochlorine pesticides in Beijing human milk, 1998. *Bull. Environ. Contam. Toxicol.* **2003**, *70*, 193−197.

(13) Kunisue, T.; Someya, M.; Kayama, F.; Jin, Y.; Tanabe, S. Persistent organochlorines in human breast milk collected from primiparae in Dalian and Shenyang, China. *Environ. Pollut.* **2004**, *131*, 381−392.

(14) Wolfe, D. A.; Champ, M. A.; Cross, F. A.; Kester, D. R.; Park, P. K.; Swanson, R. L. Marine pollution research facilities in the People's Republic of China. *Mar. Pollut. Bull.* **1984**, *15*, 207−212.
(15) Chen, J.; Gao, J. The Chinese total diet study in 1990. Part I. Chemical contaminants. *J. AOAC Int.* **1993**, *76*, 1193−1205.
(16) FAO/UNEP/WHO. Experiment of GEME/food in coordinating dietary intake studies. In *Monitoring dietary intakes*; Macdonald, I., Ed.; Springer-Verlag: Berlin, 1991; pp 126−135.
(17) Kang, Y. S.; Matsuda, M.; Kawano, M.; Wakimoto, T.; Min, B. Y. Organochlorine pesticides, polychlorinated biphenyls, polychlorinated dibenzo-*p*-dioxins and dibenzofurans in human adipose tissue from western Kyungnam, Korea. *Chemosphere* **1997**, *35*, 2107−2117.
(18) Muscat, J. E.; Britton, J. A.; Djordjevic, M. V.; Citron, M. L.; Kemeny, M.; Busch-Devereaux, E.; Pittman, B.; Stellman, S. D. Adipose concentrations of organochlorine compounds and breast cancer recurrence in Long Island, New York. *Cancer Epidemiol., Biomarkers Prev.* **2003**, *12*, 1474−1478.
(19) Kucklick, J. R. In *Preliminary results from the third annual NIST/NOAA inter laboratory comparison exercise for organochlorines in marine mammal tissues.* 2001; p 106.
(20) Minh, T. B.; Watanabe, M.; Tanabe, S.; Yamada, T.; Hata, J.; Watanabe, S. Specific accumulation and elimination kinetics of Tris(4-chlorobiphenyl)methane, Tris(4-chlorophenyl)methanol, and other persistent organochlorines in humans from Japan. *Environ. Health Perspect.* **2001**, *109*, 927−935.
(21) Minh, T. B.; Watanabe, M.; Tanabe, S.; Yamada, T.; Hata, J.; Watanabe, S. Occurrence of Tris(4-chlorophenyl)methane, Tris-(4-chlorophenyl)methanol and some other persistent organochlorines in Japanese human adipose tissues. *Environ. Health Perspect.* **2000**, *108*, 599−603.
(22) Waliszewski, S. M.; Aguirre, A. A.; Infanzon, R. M.; Benitez, A.; Rivera, J. Comparison of organochlorine pesticides levels in adipose tissue and human milk of mother living in Veracruz, Mexico. *Bull. Environ. Contam. Toxicol.* **1999**, *62*, 685−690.
(23) Alawi, M. A.; Tamimi, S.; Jaghabir, M. Storage of organochlorine pesticides in human adipose tissues of Jordanian males and females. *Chemosphere* **1999**, *38*, 2865−2873.
(24) Cok, I.; Bilgili, A.; Yarsan, E.; Burgaz, S. Organochlorine pesticide residue levels in human adipose tissue of residents of Manisa (Turkey). *Bull. Environ. Contam. Toxicol.* **1998**, *61*, 311−316.
(25) Xu, D.; Zhong, W.; Deng, L.; Chai, Z.; Mao, X. Regional distribution of organochlorinated pesticides in pine needles and its indication for socioeconomic development. *Chemosphere* **2004**, *54*, 743−752.
(26) Li, Y. F.; Cai, D. J.; Shan, Z. J.; Zhu, Z. L. Gridded usage inventories of technical hexachlorocyclohexane and lindane for China with 1/6° latitude by 1/4° longitude resolution. *Arch. Environ. Contam. Toxicol.* **2001**, *41*, 261−266.
(27) Zhou, J. L.; Hong, H.; Zhang, Z.; Maskaoui, K.; Chen, W. Multiphase distribution of organic micropollutants in Xiamen harbour, China. *Water Res.* **2001**, *34*, 2132−2150.
(28) Saver, J. G.; White, D.; Erhardt, P.; Bachmann, K. Estimating xenobiotic half-lives in humans from rat data: influence of log P. *Environ. Health Perspect.* **1997**, *105*, 1204−1209.
(29) Mackay, D.; Shiu, W. Y.; Ma, K. C. In *Illustrated Handbook of Physical-Chemical Properties and Environmental Fate for Organic Chemicals. Vol II: Polynuclear Aromatic Hydrocarbons, Polychlorinated Dioxins, and Dibenzofurans*; Lewis Publisher: Boca Raton, FL, 1992.
(30) Geyer, H. J.; Schramm, K.-W.; Feicht, E. A.; Behechti, A.; Steinberg, C.; Bruggemann, R.; Poiger, H.; Henkelmann, B.; Kettrup, A. Half-lives of tetra-, penta-, hexa-, hepta-, and octachlorodibenzo-*p*-dioxin in rats, monkey, and humans − a critical review. *Chemosphere* **2002**, *48*, 631−644.
(31) Shirai, J. H.; Kissel, J. C. Uncertainty in estimated half-lives of PCBs in humans: impact on exposure assessment. *Sci. Total Environ.* **1996**, *187*, 199−210.
(32) Finizio, A.; Vighi, M.; Sandroni, D. Determination of n-octanol/water coefficient (Kow) of pesticide. Critical review and comparison of methods. *Chemosphere* **1997**, *34*, 131−161.
(33) Fraser, A. J.; Burkow, I. C.; Wolkers, H.; Mackay, D. Modeling biomagnification and metabolism of contaminants in harp seals of the Barents Sea. *Environ. Toxicol. Chem.* **2002**, *21*, 55−61.
(34) Zhang, M.; Yang, D.; Fang, C.; Wei, K. Monitoring of organochlorine pesticide residues − the GEMS/Food program in China. *Biomed. Environ. Sci.* **1997**, *10*, 102−106.
(35) Yao, M.; McCrory, M. A.; Ma, G.; Tucker, K. L.; Gao, S.; Fuss, P.; Roberts, S. B. Relative influence of diet and physical activity on body composition in urban Chinese adults. *Am. J. Clin. Nutr.* **2003**, *77*, 1409−1416.

Received for review March 14, 2005. Revised manuscript received April 15, 2005. Accepted April 19, 2005.

ES050493D