

is interesting to consider that CHLs were relatively scarcely reported in human adipose tissue samples. Greater CHL residues were found in humans from Japan as compared to those from Canada, the United States and Poland. Relatively high concentrations of CHLs were also reported in human breast milk from Japan (37). In Japan, CHLs were mainly used for termite control (38), and the use of these compounds was banned in 1986. Time trend monitoring revealed that CHL residues exhibited increasing trend until 1985 (22). The late restriction or ban of CHLs in Japan may account for the high levels found in human adipose tissues. In view of these facts, continuous monitoring of CHLs in humans from Japan are required.

Conclusions

This study presents current residue levels of two newly detected contaminants, TCPMe and TCPMOH, as well as other classic persistent OCs in human adipose tissue from Japan. To our knowledge, this is the first report indicating the occurrence of TCPMe and TCPMOH in human adipose tissue samples. Our results provide a basis for human exposure to TCPMe and TCPMOH and subsequently for risk assessment. Widespread occurrence of TCPMe and TCPMOH in various kinds of marine mammals and their presence in human samples imply the expansion of contamination by these compounds in wide range of animals including humans. In view of these observations, further comprehensive information regarding contamination status, sources of exposure and toxicokinetics of TCPMe and TCPMOH is necessary to understand bioaccumulation and to evaluate possible risks of these compounds to humans and wildlife.

References and Notes

1. Jarman WM, Simon M, Nostrom RJ, Bruns SA, Bacon CA, Simaret BRT, Riseborough, RW. Global distribution of *tris*(4-chlorophenyl)methanol in high trophic level birds and mammals. *Environ Sci Technol* 26: 1770-1774 (1992).
2. Falandysz J, Strandberg B, Strandberd L, Rappe C. *Tris*(4-chlorophenyl)methane and *tris*(4-chlorophenyl)methanol in sediment and food webs from the Baltic south coast. *Environ Sci Technol* 33: 517-521 (1999).
3. Watanabe M, Tanabe S, Miyazaki N, Petrov EA, Jarman WM. Contamination of *tris*(4-chlorophenyl)methane and *tris*(4-chlorophenyl)methanol in marine mammals from Russia and Japan : body distribution, bioaccumulation and contamination status. *Mar Pollut Bull* (in press) (1999).
4. Poon R, Lecavalier P, Bergman A, Yaminas A, Chu I, Valli VE. Effect of *tris*(4-chlorophenyl)methanol on the rat following short-term oral exposure. *Chemosphere* 34: 1-12 (1997).
5. Korner W, Hanf V, Oettling G, Schuller W, Hangenmaier H. Effects of *tris*(4-chlorophenyl)methanol on proliferation of human breast cancer cells. *Organohalogen compounds* 34: 364-369 (1997).
6. Aguilar JS, Castro V, Bergman A. Effects of four organohalogen environmental contaminants on cytochrome P450 forms that catalyze 4- and 2-hydroxylation of estradiol in the rat liver. *Biochem Mol Med* 60: 149-154 (1997).
7. Foster WG, Desaulnier D, Leingartner K, Wade MG, Poon R, Chu I. Reproductive effects of *tris*(4-chlorophenyl)methanol in the rat. *Chemosphere* 39: 709-724 (1999).
8. Rahman MS, Montanarella L, Johansson B, Larsen BR. Trace levels of *tris*(4-chlorophenyl)methanol and methane in human milk. *Chemosphere* 27: 1487-1497 (1993).
9. Minh TB, Watanabe M, Nakata H, Tanabe S, Jefferson TA. Contamination by persistent organochlorines in small cetaceans from Hong Kong coastal waters. *Mar Pollut Bull* (in press) (1999).

10. Buser HR. DDT, a potential source of environmental *tris*(4-chlorophenyl)methane and *tris*(4-chlorophenyl)methanol. *Environ Sci Tech* 29: 2133-2139 (1995).
11. de Boer J. Environmental distribution and toxicity of *tris*(4-chlorophenyl)methanol and *tris*(4-chlorophenyl)methane. *Rev Environ Contam Toxicol* 150: 95-106 (1997).
12. de Boer J, Wester PG, Evers EHG, Brinkman UAT. Determination of *tris*(4-chlorophenyl)methanol and *tris*(4-chlorophenyl)methane in fish, marine mammals and sediment. *Environ Pollut* 93: 39-47 (1996).
13. Tanabe S, Watanabe S, Kan H, Tatsukawa R. Capacity and mode of PCB metabolism in small cetaceans. *Mar Mamm Sci* 4: 103-124 (1988).
14. Watanabe S, Shimada T, Nakamura S, Nishiyama N, Yamashita N, Tanabe S, Tatsukawa R. Specific profile of liver microsomal cytochrome P-450 in dolphin and whales. *Mar Environ Res* 27: 51-65 (1989).
15. Kannan N, Tanabe S, Ono M, Tatsukawa R. Critical evaluation of polychlorinated biphenyls toxicity in terrestrial and marine mammals : increasing impact of non-*ortho* and mono-*ortho* coplanar polychlorinated biphenyls from land to ocean. *Arch Environ Contam Toxicol* 18: 850-857 (1989).
16. Nakata H, Tanabe S, Tatsukawa R, Amano M, Miyazaki N, Petrov EA. Persistent organochlorine residues and their accumulation kinetics in Baikal seal (*Phoca sibirica*) from Lake Baikal, Russia. *Environ Sci Technol* 29: 2877-2885 (1995).
17. Ferrer A, Bona MA, Castellano M, To-Figueras J, Brunet M. Organochlorine residues in human adipose tissue of the population of Zaragora (Spain). *Bull Environ Contam Toxicol* 48: 561-566 (1992).
18. Greve PA, van Zoonen P. Organochlorine pesticides and PCBs in tissues from Dutch citizens (1968-1986). *Inter J Environ Anal Chem* 38: 265-277 (1990).
19. Focardi S, Fossi C, Leonzio C, Romei R. PCB congeners, hexachlorobenzene, and organochlorine insecticides in human fat in Italy. *Bull Environ Contam Toxicol* 36, 644-650 (1986).
20. Kang Y, Matsuda M, Kawano M, Wakimoto T, Min B. Organochlorine pesticides, polychlorinated biphenyls, polychlorinated dibenzo-*p*-dioxins and dibenzofurans in

- human adipose tissue from western Kyungnam, Korea. *Chemosphere* 35: 2107-2117 (1997).
21. Walker W, Riseborough RW, Jarman W M, Lappe BW, Lappe JA, Tefft JA, de Long RL. Identification of *tris*(4-chlorophenyl)methanol in blubber of harbour seals from Puget Sound. *Chemosphere* 18: 1799-1804 (1989).
 22. Longanathan BG, Tanabe S, Hidaka Y, Kawano M, Hidaka H, Tatsukawa R. Temporal trends of persistent organochlorine residues in human adipose tissue from Japan, 1928-1985. *Environ Pollut* 81: 31-39 (1993).
 23. Williams DV, Lebel GL, Junkins E. Organohalogen residues in human adipose autopsy samples from six Ontario municipalities. *J Assoc Off Anal Chem* 71: 410-414 (1988).
 24. Mes J. Organochlorine residues in human blood and biopsy fat and their relationship. *Bull Environ Contam Toxicol* 48: 815-820 (1992).
 25. Holt RL, Cruse S, Greer ES. Pesticide and polychlorinated biphenyl residues in human adipose tissue from northern Louisiana. *Bull Environ Contam Toxicol* 36: 651-655 (1986).
 26. Archibeque-Engle S, Tessari JD, Winn DT, Keefe TJ, Nett TM, Zheng T. Comparison of organochlorine pesticide and polychlorinated biphenyl residues in human breast adipose tissue and serum. *J Toxicol Environ Health Part A* 52: 285-293 (1997).
 27. Waliszewski SM, Aguirre AA, Infanzon RM, Benitez A, Rivera J. Comparison of organochlorine pesticide levels in adipose tissue and human milk of mothers living in Veracruz, Mexico. *Bull Environ Contam Toxicol* 62: 685-690 (1999).
 28. Ludwicki JK, Goralczyk K. Organochlorine pesticides and PCBs in human adipose tissue in Poland. *Bull Environ Contam Toxicol* 52: 400-403 (1994).
 29. Tanabe S, Falandysz J, Higaki T, Kannan K, Tatsukawa R. Polychlorinated biphenyl and organochlorine insecticide residues in human adipose tissue in Poland. *Environ Pollut* 79: 45-49 (1993).

30. Gomez-Catalan J, Lezaun M, To-Figueras J, Corbella J. Organochlorine residues in the adipose tissue of the population of Navarra (Spain). Bull Environ Contam Toxicol 54: 534-540 (1995).
31. Weistrand C, Noren K. Polychlorinated naphthalenes and other organochlorine contaminants in human adipose and liver tissue. J Toxicol Environ Health Part A 53: 293-311 (1998).
32. Cok I, Bilgili A, Yarsan E, Bagci C, Burgaz S. Organochlorine pesticide residue levels in human adipose tissue of residents of Manisa (Turkey), 1995-1996. Bull Environ Contam Toxicol 61: 311-316 (1998).
33. Burgaz S, Afkham BL, Karakaya AE. Organochlorine pesticide contaminants in human adipose tissue collected in Tebriz (Iran). Bull Environ Contam Toxicol 54: 546-553 (1995).
34. Alawi MA, Tamimi S, Jaghabir M. Storage of organochlorine pesticides in human adipose tissue of Jordanian males and females. Chemosphere 38: 2865-2873 (1999).
35. Nakamura H, Matsuda M, Quynh HT, Cau HD, Chi HTK, Wakimoto T. Levels of polychlorinated dibenzo-*p*-dioxins, dibenzofurans, PCBs, DDTs and HCHs in human adipose tissue and breast milk from the south of Vietnam. Organohalogen Compounds 21: 71-76 (1994).
36. Kutz F W, Wood PH, Bottimore DP. Organochlorine pesticides and polychlorinated biphenyls in human adipose tissue. Rev Environ Contam Toxicol 120: 1-82 (1991).
37. Tojo Y, Wariishi M, Suzuki Y, Nishiyama K. Quantitation of chlordane residues in mother's milk. Arch Environ Contam Toxicol 15: 327-332 (1986).
38. Kawano M, Inoue T, Hidaka H, Tatsukawa R. Chlordane residue in krill, fish and weddel seal from the Antarctic. Toxicol Environ Chem 11: 137-145 (1986).
39. Minh TB, Watanabe M, Tanabe S, Miyazaki N, Jefferson TA, Prudente M, Subramanian A. Widespread contamination by *tris*(4-chlorophenyl)methane and *tris*(4-chlorophenyl)methanol in cetaceans from the North Pacific and Asian coastal waters. Environ Pollut (in press).

Table 1. Information of Japanese human adipose tissue samples analysed in this study

Sample No.	Sex	Age	Residence	Occupation	Cause of death
1	M	50	Kawasaki	Pharmacist	Brain tumor
2	M	55	Tokyo	Officer	Hepatocellular carcinoma
3	M	63	Yokohama	Independent	Myocardial infarction
4	M	65	Chiba	Officer	Pulmonary fibrosis
5	M	68	Tokyo	Officer	Bronchopneumonia
6	M	73	Yokohama	Officer	Lung cancer
7	M	79	Tokyo	Officer	Urinary bladder carcinoma
8	M	87	Tokyo	Without occupation	Bronchopneumonia
9	F	29	Tokyo	Without occupation	Acute myelocytic leukemia
10	F	33	Tokyo	Housewife	Spinal cord tumor
11	F	53	Tokyo	Housewife	Takayasu's arteritis
12	F	75	Tokyo	Housewife	Cerebral hemorrhage
13	F	76	Tokyo	Without occupation	Acute myelocytic leukemia
14	F	85	Tokyo	Housewife	Ovarian carcinoma

Abbreviations : M, male; F, female.

Table 2. Concentrations (ng/g lipid wt) of TCPMe, TCPMOH and other organochlorines in human adipose tissue from Japan

Sample No.	Sex	Age	Fat (%)	TCPMe	TCPMOH	PCBs	DDTs ^a	HCHs ^b	CHLs ^c	HCB
1	M	50	80	11	5.5	1800	350	140	280	30
2	M	55	80	8.9	7.5	1800	1300	150	180	31
3	M	63	74	11	8.1	2600	950	360	260	64
4	M	65	86	6.2	5.8	1300	600	600	110	22
5	M	68	85	3.1	1.1	520	110	60	65	22
6	M	73	66	15	8.5	3200	710	260	710	44
7	M	79	57	11	4.7	1700	660	160	210	30
8	M	87	63	17	16	2500	1900	320	220	27
9	F	29	57	2.5	2.8	560	400	210	110	33
10	F	33	56	3.0	2.9	930	820	430	96	38
11	F	53	57	21	18	2300	820	540	250	68
12	F	75	66	6.7	4.5	2300	1300	560	270	68
13	F	76	78	8.5	8.2	1700	420	510	210	60
14	F	85	74	4.9	2.4	720	620	300	260	38
Mean		70	9.3	6.9	1700	780	330	230	41	
Range		56 - 86	2.5 - 21	1.1 - 18	560 - 3200	110 - 1900	60 - 600	96 - 710	30 - 60	

Abbreviations : M, Male; F, Female.

^aDDTs = sum of *p,p'*-DDE, *p,p'*-DDD and *p,p'*-DDT. ^bHCHs = sum of α -HCH, β -HCH and γ -HCH. ^cCHLs = sum of oxy chlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor and *cis*-nonachlor.

Table 3. Comparison of recent organochlorine residue levels (ng/g lipid wt) in human adipose tissue from various countries

Country	Survey year	PCBs	DDTs	HCHs	HCB	CHLs	Reference
Canada	1984	2000	3400	80 ^a	80	160 ^a	Williams et al. (23)
Canada	1992	nd	600 ^b	25 ^c	25	56	Mes (24)
USA	1984	1200	4100	620 ^a	nd	nd	Holt et al. (25)
USA	1997	196	72 ^e	34 ^a	36	72	Archibeque-Engle et al. (26)
Mexico	1997 - 1998	nd	5660 ^e	156	58	nd	Waliszewski et al. (27)
Poland	1989 - 1992	860	6300 ^e	320	310	nd	Ludwicki and Goralczyk (28)
Poland	1990	1500	15000	250	260	70	Tanabe et al. (29)
Navarra, Spain	1991	2400	4300 ^e	1530 ^a	3400	nd	Gomez-Catalan et al. (30)
Zaragoza, Spain	1988 - 1989	1500	2960 ^b	530 ^a	2950	nd	Ferrer et al. (17)
Netherlands	1986	3400	2500	280	380	nd	Greve and van Zoonen (18)
Italy	1984	1800	8200 ^e	130 ⁱ	2300	nd	Forcadi et al. (19)
Sweden	-	1180	788 ^b	nd	56	nd	Weistrand and Noren (31)
Turkey	1995 - 1996	nd	2130 ^e	520	33	nd	Cok et al. (32)
Iran	1991 - 1992	nd	2900 ^e	770	55	nd	Burgaz et al. (33)
Jordan	1996	nd	3900 ^a	1550	120	nd	Alawi et al. (34)
South Vietnam	1991	300	4900	30	nd	nd	Nakamura et al. (35)
Korea	1994 - 1995	400	1100	190	20	nd	Kang et al. (20)
Japan	1998	1700	780	330	41	230	Present study

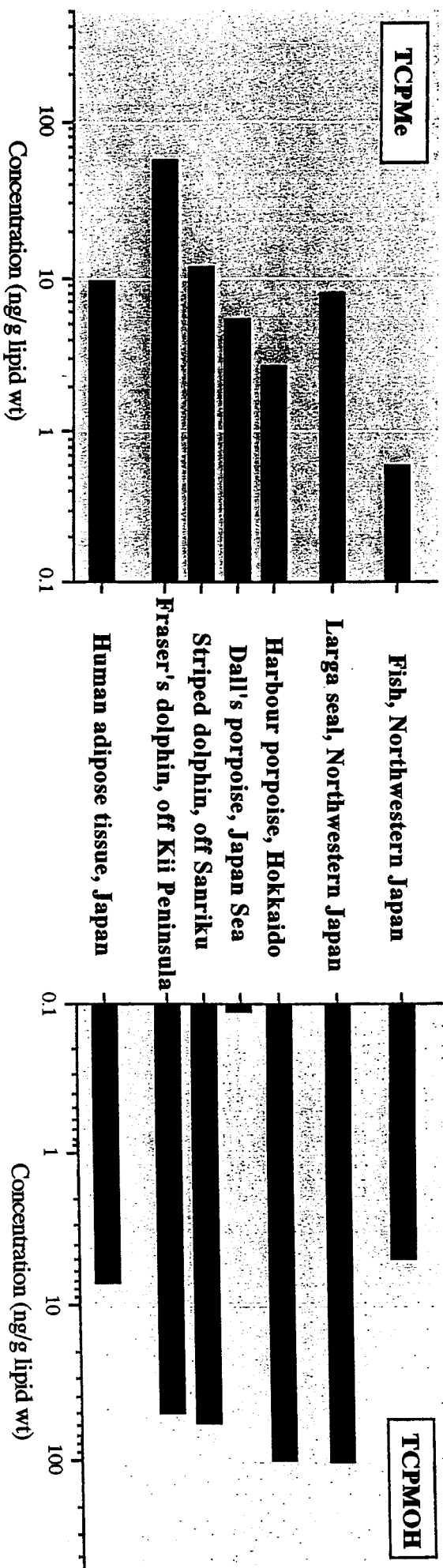
Abbreviation : nd, not determined.

^aβ-HCH only. ^b*p,p'*-DDE + *o,p'*-DDT + *p,p'*-DDT. ^cα-HCH + β-HCH. ^doxy chlordane + trans-nonachlor + cis-nonachlor. ^e*p,p'*-DDE + *p,p'*-DDT. ^f*p,p'*-DDE + *p,p'*-DDD + *p,p'*-DDT + *o,p'*-DDT. ^g*p,p'*-DDT. ^h*p,p'*-DDE only. ⁱγ-HCH only. ^j*p,p'*-DDE + *p,p'*-DDD + *p,p'*-DDT + *o,p'*-DDE + *o,p'*-DDD + *o,p'*-DDT.

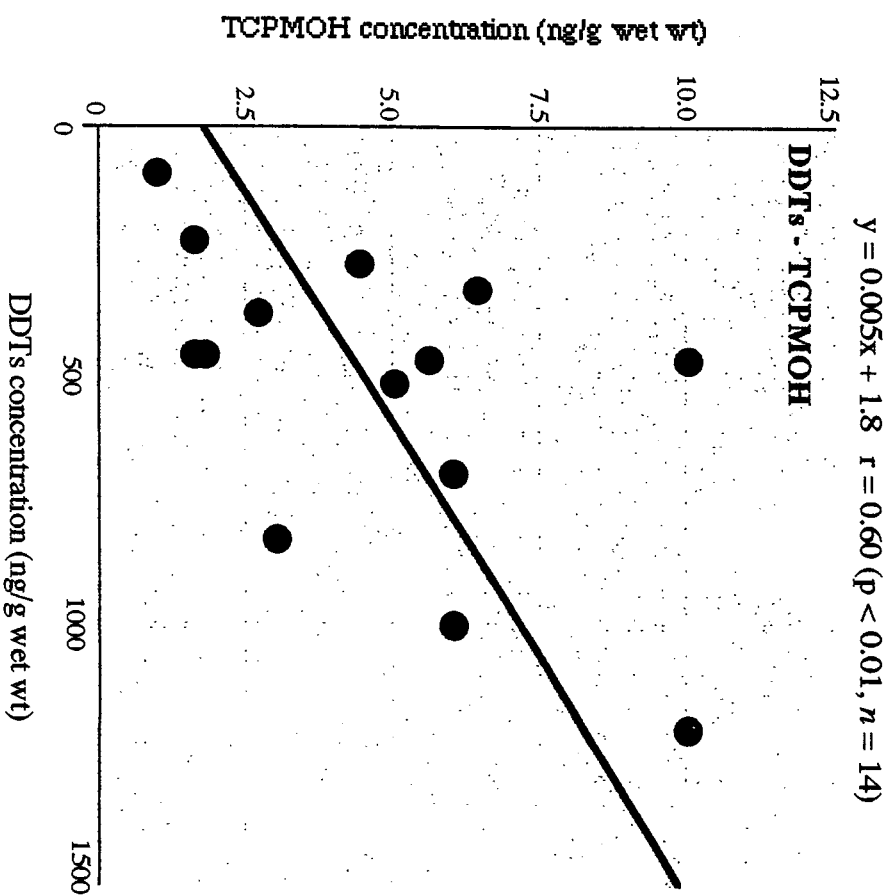
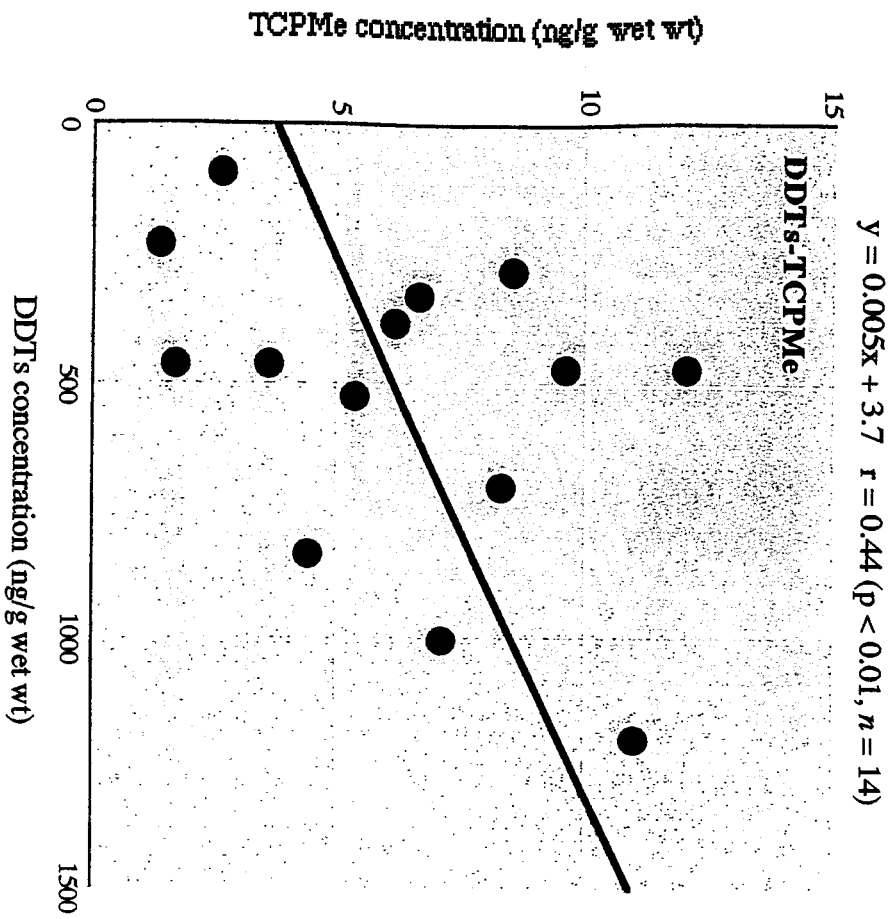
FIGURE LEGENDS

Figure 1. Comparison of TCPMe and TCPMOH residue levels in fishes, marine mammals and humans from Japan. Data on fishes and large seals were cited from Watanabe et al. (3), cetaceans from Minh et al. (39).

Figure 2. Correlation between TCPMe/TCPMOH and DDTs concentrations in Japanese human adipose tissue.



Minh et al., Figure 1



Minh et al., Figure 2