

PCDDS, PCDFS, PCBS AND ORGANOCHLORINE INSECTICIDES IN HUMAN BREAST MILK COLLECTED FROM ASIAN DEVELOPING COUNTRIES: RISK ASSESSMENT FOR INFANTS

Tatsuya Kunisue¹, Mafumi Watanabe¹, Masayuki Someya¹, In Monirith¹, Tu Binh Minh¹, Annamalai Subramanian², Touch Seang Tana³, Pham Hung Viet⁴, Maricar Prudente⁵ and Shinsuke Tanabe¹

¹ Center for Marine Environmental Studies (CMES), Ehime University, Tarumi 3-5-7, Matsuyama 790-8566, Japan

² Center of Advanced Studies in Marine Biology, Annamalai University, Parangipettai 608502, Tamil Nadu, India

³ Social and Culture Observation Unit (OBSES) of the Cabinet of the Council of Minister, Kingdom of Cambodia

⁴ Center of Environmental Chemistry, Vietnam National University, 334 Nguyen Trai Str. Thanh Xuân Distr., Hanoi, Vietnam

⁵ Science Education Department, De La Salle University, 2401 Taft Avenue, 1004 Manila, Philippines

Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofurans (PCDFs), biphenyls (PCBs) and organochlorine insecticides are lipophilic stable contaminants, and have been of great concern regarding their toxic effects on humans and wildlife. In developed countries, it is suggested that residue levels of these contaminants in various environmental media and biota have generally decreased in recent decades (1, 2). In Asian developing countries, however, few studies have reported regarding exposure to these contaminants, especially dioxins and related compounds, for humans. In dumping sites of municipal wastes in Asian developing countries, secondary formation of dioxins and related compounds is anticipated, because a lot of various wastes has been dumped daily and continuously burned under low temperature by spontaneous combustion or intentional incineration. In addition, it is doubtful that organochlorine insecticides have been used there for public health purposes. These are suspected that many residents around these dumping sites are exposed to these contaminants. It is especially feared that *in utero* and lactational exposure to dioxins and related compounds may adversely affect brain development and immune systems of infants and children (3-6).

The present study attempted to elucidate the contamination status of PCDDs, PCDFs, PCBs and organochlorine insecticides in human breast milk collected from dumping sites of municipal wastes and reference sites in Asian developing countries, such as India, Cambodia, Vietnam and Philippines, and to assess the risk of exposure in their infants.

Materials and Methods

Human breast milk samples were collected from Perungudi, Chennai in India on Aug. 2000, Meanchey, Phnom Penh in Cambodia on Nov. 1999 and Dec. 2000, Tay Mo, Hanoi in Vietnam on Apr. 2000 and Payatas, Quezon in Philippines on Feb. 2000, which have dumping sites of municipal wastes in the suburbs of urban area. In addition, samples were also collected from reference sites in India, Cambodia and Vietnam, where are located further away than 5 km from dumping sites. Breast milk samples were stored at -20 °C until analysis.

HUMAN EXPOSURE II

Chemicals analyzed in this study were PCDDs, PCDFs, PCBs, DDTs, HCHs, HCB, chlordanes compounds (CHLs). Extraction was conducted using the method reported by Hirai *et al* (7). Cleanup and separation processes were followed by the method previously described (8, 9). Identification and quantification of PCDDs, PCDFs, non- and mono-*ortho* coplanar PCBs was performed using HRGC (Agilent 6890)-HRMS (JEOL JMS-700D and GCmate). Quantification of total PCBs, DDTs, HCHs, HCB, CHLs was performed using GC-ECD (Hewlett-Packard 6890). TEQs (toxic equivalencies) were calculated using WHO-TEFs (10).

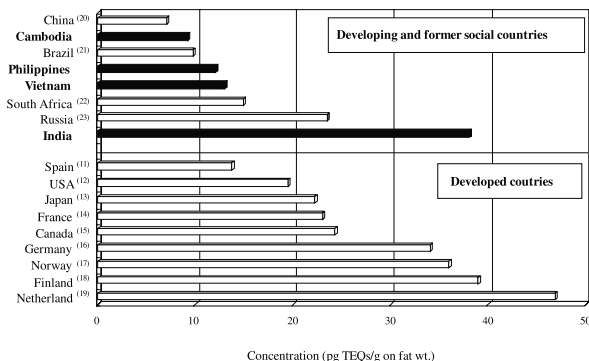
Results and Discussion

PCDDs, PCDFs, PCBs and organochlorine insecticides were detected in all the samples of human breast milk analyzed in this study (Table 1), indicating that the residents around dumping sites of

Table 1. Mean and range concentrations (pg/g or ng/g on fat wt.) of dioxins and related compounds, PCBs and organochlorine insecticides in human breast milk collected from residents around dumping sites of municipal wastes and reference sites in Asian developing countries

	India		Cambodia		Vietnam		Philippines
	dumping site <i>n</i> = 11	reference site <i>n</i> = 8	dumping site <i>n</i> = 19	reference site <i>n</i> = 16	dumping site <i>n</i> = 8	reference site <i>n</i> = 10	dumping site <i>n</i> = 9
PCDDs	290 (150-780)	160 (70-240)	49 (14-170)	55 (20-150)	32 (10-81)	27 (14-41)	190 (29-730)
PCDFs	50 (15-130)	18 (9.4-43)	15 (5.2-55)	11 (4.4-24)	20 (7.3-42)	20 (9.6-45)	21 (5.9-44)
non-ortho PCBs	260 (30-610)	91 (42-340)	51 (29-130)	42 (19-79)	62 (17-100)	56 (29-100)	76 (26-160)
mono-ortho PCBs	38000 (2500-170000)	6100 (2900-13000)	8000 (820-28000)	4900 (1300-12000)	24000 (4200-46000)	15000 (2800-22000)	8800 (1700-28000)
TEQs	38 (8.5-140)	12 (7.0-17)	9.2 (5.2-21)	7.8 (1.9-15)	13 (4.6-24)	12 (6.5-19)	12 (5.0-37)
PCBs	110 (12-240)	30 (9.2-69)	56 (14-170)	38 (11-130)	170 (50-350)	98 (54-210)	72 (17-160)
DDTs	420 (170-830)	430 (160-1000)	1200 (360-3800)	1800 (310-11000)	2800 (640-6900)	1600 (480-3200)	190 (35-570)
HCHs	790 (100-2100)	640 (300-1300)	4.8 (1.5-18)	5.6 (<0.21-21)	74 (15-160)	53 (25-79)	4.7 (<0.56-10)
CHLs	10 (0.51-38)	0.91 (<0.14-3.8)	1.7 (<0.26-5.0)	1.6 (<0.21-5.3)	1.6 (<0.20-3.8)	1.9 (<0.22-1.8)	15 (4.2-37)
HCB	1.5 (<0.38-3.8)	1.0 (<0.14-3.7)	1.8 (0.59-8.1)	1.6 (0.70-3.2)	5.3 (1.4-9.5)	3.5 (1.7-6.4)	<0.56

PCDDs, PCDFs, non- and mono-*ortho* PCBs and TEQs were represented with pg/g on fat wt.
PCBs, DDTs, HCHs, CHLs and HCB were represented with ng/g on fat wt.



Reference data (11-23) were recalculated with WHO-TEFs (10).

Fig. 1 Comparisons of TEQs in human breast milk collected from dumping sites in Asian developing countries (India, Cambodia, Vietnam and Philippines) and those reported from other countries.

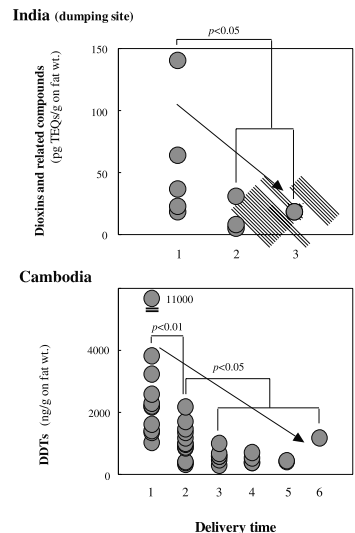


Fig. 2 Relationship between concentrations of TEQs and DDTs in human breast milk collected from India and Cambodia and the number of delivery times.

municipal wastes and reference sites in India, Cambodia, Vietnam and Philippines have been exposed to these contaminants. Concentrations of dioxins and related compounds in human breast milk from dumping sites in India were significantly higher than those from reference sites and other Asian developing countries ($p < 0.05$) and the levels of TEQs were comparable to those (11-19) in general public of developed countries (Fig. 1). This indicates that significant pollution sources of dioxins and related compounds present in dumping sites in India and residents around there have been exposed to relatively high levels of these contaminants. On the other hands, TEQs in human breast milk from Cambodia, Vietnam and Philippines were relatively low and not significantly different between dumping sites and reference sites. The dominant organochlorine insecticides in human breast milk from India, Cambodia, Vietnam and Philippines were HCHs, DDTs, DDTs and DDTs, respectively, and not significantly different between dumping sites and reference sites. Especially, levels of HCHs in Indian and DDTs in Vietnamese and Cambodian were relatively high, indicating these contaminants have been used for public health purposes in these countries.

As examined the relationship between concentrations of TEQs and organochlorines in human breast milk and the number of delivery times, levels of these contaminants tended to decrease with increase of the number of delivery times (Fig. 2). Furthermore, estimated daily intakes (EDIs) of TEQs exceeded 4 pg TEQs/kg/day of TDI (WHO, 1998) for all the Asian developing countries, and EDIs of HCHs and DDTs exceeded 0.3 μg /kg/day of TDI (Health Canada, 1996) for all the Indian and 20 μg /kg/day of TDI (WHO, 1982) for a few of Vietnamese and Cambodian, respectively. These results suggest that first infants have been exposed to higher levels of dioxins and related compounds and organochlorine insecticides from breast milk and might be at higher risk for these contaminants.

Acknowledgments

This study was supported by Waste Management Research Grants on the "Formation and Behavior of Dioxins and Their Related Persistent Organic Pollutants in Uncontaminated Combustion Process" from the Ministry of Environment. This study was also supported by the Toyota Foundation, Sumitomo Foundation and the Japan Fund for Global Environment (Japan Environment Corporation).

References

1. Alcock, R. E., Jones, K. C. (1996) Dioxins in the environment: A review of trend data. *Environ. Sci. Technol.*, 30, 3133-3143.
2. Noren, K., Meironyte, D. (2000). Certain organochlorine and organobromine contaminants in Swedish human milk in perspective of past 20-30 years. *Chemosphere*, 40, 1111-1123.
3. Porterfield, S. P. (1994) Vulnerability of the developing brain to thyroid abnormalities: Environmental insults to the thyroid system. *Environ. Health. Perspect.*, 102 (suppl 2), 962-966.
4. Koopman-Esseboom, C., Morse, D. C., Weisglas-Kuperus, N., Lutkeschipholt, I. J., van der Paauw, C. G., Tuinstra, L. G. M. T., Brouwer, A., Sauer, P. J. J. (1994). Effects of dioxins and polychlorinated biphenyls on thyroid hormone status of pregnant women and their infants. *Pediatr. Res.*, 36, 468-473.
5. Weisglas-Kuperus, N., Sas, T. C. J., Koopman-Esseboom, C., van der Zwan, C. W., de Ridder, M. A. J., Beishuizen, A., Hooijkaas, H., Sauer, P. J. J. (1995). Immunologic effects of background prenatal and postnatal exposure to dioxins and polychlorinated biphenyls in Dutch infants. *Pediatr. Res.*, 38, 404-410.
6. Weisglas-Kuperus, N., Patandin, S., Berbers, G. A. M., Sas, T. C. J., Mulder, P. G. H., Sauer, P. J. J., Hooijkaas, H. (2000) Immunologic effects of background exposure to polychlorinated biphenyls and dioxins in Dutch preschool children. *Environ. Health Perspect.*, 108, 1203-1207.
7. Hirai, T., Fujimine, Y., Kodaira, T., Watanabe, S. (2001) Simple solid-phase lipid extraction of

HUMAN EXPOSURE II

- dioxins from maternal breast milk. *Organohalogen Compd.*, 50, 138-141.
8. Kunisue, T., Watanabe, M., Monirith, I., Subramanian, A., Tana, T. S., Prudente, M., Tanabe, S. (2001) Contamination by dioxin related compounds in human breast milk collected from Asian developing countries. *Organohalogen Compd.*, 52, 282-285.
 9. Watanabe, M., Tanabe, S., Tatsukawa, R., Amano, M., Miyazaki, N., Petrov, E. A., Khuraskin, S. L. (1999) Contamination and specific accumulation of persistent organochlorines in Caspian seal (*Phoca caspica*) from the Caspian Sea, Russia. *Arch. Environ. Contam. Toxicol.*, 37, 396-407.
 10. Van den Berg, M., Brinbaum, L., Bosveld, A. T. C., Brunstrom, B., Cook, P., Feeley, M., Giesy, J. P., Hanberg, A., Hasegawa, R., Kennedy, S. W., Kubiak, T., Larsen, J. C., Rolaf van Leeuwen, F. X., Liem, A. K. D., Nolt, C., Peterson, R. E., Poellinger, L., Safe, S., Schrenk, D., Tillit, D., Tysklind, M., Younes, M., Waern, F., Zacharewski, T. (1998) Toxic equivalency factor (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. *Environ. Health Perspect.*, 106, 775-792.
 11. Schuhmacher, M., Domingo, J. L., Llobet, J. M., Kiviranta, H., Vartiainen, T. (1999) PCDD/F concentrations in milk of nonoccupationally exposed women living in southern Catalonia, Spain. *Chemosphere*, 38, 995-1004.
 12. Schecter, A., Fürst, P., Ryan, J. J., Fürst, C., Meemken, H-A., Groebel, W., Constable, J., Vu, D. (1989). Polychlorinated dioxin and dibenzofuran levels from human milk from several locations in the United States, Germany and Vietnam. *Chemosphere*, 19, 979-984.
 13. MHW. (1998) Investigation on dioxins and related compounds in human breast milk. The Ministry of Health and Welfare, Japan.
 14. Gonzalez, M. J., Jimenez, B., Hernandez, L. M., Gonnord, M. F. (1996) Levels of PCDDs and PCDFs in human milk from populations in Madrid and Paris. *Bull. Environ. Contam. Toxicol.*, 56, 197-204.
 15. Dewailly, E., Nantel, A., Bruneau, S., Laliberte, C., Ferron, L., Gingras, S. (1992) Breast milk contamination by PCDDs, PCDFs and PCBs in arctic Quebec: A preliminary assessment. *Chemosphere*, 25, 1245-1249.
 16. Furst, P., Furst, C., Wilmers, K. (1994) Human milk as a bioindicator for body burden of PCDDs, PCDFs, organochlorine pesticides, and PCBs. *Environ. Health Perspect.*, 102 (suppl 1), 187-193.
 17. Becher, G., Skaare, J. U., Polder, A., Sletten, B., Rosslund, O. J., Hansen, H. K., Ptashkas, J. (1995). PCDDs, PCDFs, and PCBs in human milk from different parts of Norway and Lithuania. *J. Toxicol. Environ. Health*, 46, 133-148.
 18. Kiviranta, H., Purkunen, R., Vartiainen, T. (1999) Levels and trends of PCDD/Fs and PCBs in human milk in Finland. *Chemosphere*, 38, 311-323.
 19. Liem, A. K. D., Albers, J. M. C., Baumann, R. A., Van Beuzekom, A. C., Den Hartog, R. S., Hoogerbrugge, R., Den Jong, A. P. J. M., Marsman, J. A. (1995) PCBs, PCDDs, PCDFs and organochlorine pesticides in human milk in the Netherlands. Levels and trends. *Organohalogen Compd.*, 26, 69-74.
 20. Schecter, A., Jiang, K., Rapke, O., Furst, P., Furst, C. (1994) Comparison of dibenzodioxin levels in blood and milk in agricultural workers and others following pentachlorophenol exposure in China. *Chemosphere*, 29, 2371-2380.
 21. Paumgarten, F. J. R., Cruz, C. M., Chahoud, I., Palavinskas, R., Mathar, W. (2000) PCDDs, PCDFs, PCB, and other organochlorine compounds in human milk from Rio de Janeiro, Brazil. *Environ. Res. Sect. A*, 83, 293-297.
 22. Schecter, A., Startin, J. R., Rose, M., Wright, C., Parker, I., Woods, D., Hansen, H. (1990) Chlorinated dioxin and dibenzofuran levels in human milk from Africa, Pakistan, southern Vietnam, the southern U.S. and England. *Chemosphere*, 20, 919-925.
 23. Schecter, A., Fürst, P., Fürst, C., Groebel, W., Kolesnikov, S., Savchenkov, M., Beim, A., Boldonov, A., Trubitsun, E., Vlasov, B. (1990). Levels of dioxins, dibenzofurans and other chlorinated xenobiotics in human milk from the Soviet Union. *Chemosphere*, 20, 927-934.