

LEVELS OF POLYCHLORINATED DIBENZO-*p*-DIOXINS, DIBENZOFURANS, PCBs, DDTs AND HCHs IN HUMAN ADIPOSE TISSUE AND BREAST MILK FROM THE SOUTH OF VIETNAM

Hirofumi Nakamura^A, Muneaki Matsuda^A, Hoang Trong Quynh^B, Haong Dinh Cau^B, Huynh Thi Kim Chi^C, Tadaaki Wakimoto^A

A: Department of Environment Conservation, Ehime University, Tarumi 3-5-7, Matsuyama 790, JAPAN

B: College of Medicine, Hanoi, VIETNAM

C: Song Be Province Center for Mother and Child Health Protection, Thu Dau Mot, VIETNAM

ABSTRACT

Nineteen human adipose tissue samples and sixteen human breast milk samples were collected from Tay Ninh province in the south of Vietnam in 1991. These samples were analyzed simultaneously for PCDDs/DFs, PCBs, HCHs and DDTs. The mean values of 2,3,7,8-TeCDD were 4.3pg/g fat basis and 2.7pg/g fat basis for adipose tissue and breast milk, respectively. DDT and its metabolites (DDE) were found in considerable level in adipose tissue and breast milk 5.2ppm and 5ppm, respectively. The result showed the heavy DDTs contamination in the south of Vietnam. While PCBs contamination was found in all samples.

KEY WORDS

2,3,7,8-TeCDD, PCDDs/DFs, Agent Orange, The South of Vietnam, Adipose Tissue, Breast Milk, DDTs, PCBs, HCHs

INTRODUCTION

Our purpose in this study is to determine the levels of organochlorine compounds in human adipose tissue and then to consider the countermeasure for human health. The south of Vietnam was polluted by 2,3,7,8-TeCDD, a contaminant of 2,4,5-T herbicide used in Agent Orange, Purple, Pink, Green during the Second Indochina War (1961-72). The samples were collected from provincial hospital in Tay Ninh at May in 1991, in cooperation with 10.80 Committee of Vietnam government. Tay Ninh prov. was one of among the worst sprayed areas. The levels of 2,3,7,8-TeCDD in human adipose tissue and breast milk in Vietnam had been reported.¹⁻⁵⁾ Recent investigations by A. Schecter et al. documented that the levels of 2,3,7,8-TeCDD in Vietnamese inhabited in the south had been decreased slightly in 1984-90.¹⁻⁴⁾

In earlier study⁶⁾, we have reported that high concentrations of DDTs were detected in soil samples from the south of Vietnam and PCBs contamination in a few soil was noticeable in northern districts of the Tay Ninh prov.. There was little contamination of

HUTOX

soils with HCHs in Tay Ninh prov.⁶⁾ Consequently, many agricultural products have been noted to be contaminated by DDTs, HCHs and PCBs.⁷⁾ The average daily intakes of these chemicals by Vietnamese were higher than those observed in developed countries.⁷⁾ It is important to analyse these compounds in human adipose tissues and breast milk for their health effect.

ANALYTICAL METHOD

Samples

Human adipose tissue samples (2 males and 17 females) and breast milk samples (16) were collected from a provincial hospital in Tay Ninh. They were placed in chemically clean containers with 10% formalin, and transported to Japan. The samples were stored at -30°C until analyse.

Extraction and clean-up

Analytical procedure⁸⁾ was performed by a simultaneous method developed in our laboratory for PCBs, HCHs and DDTs with PCDDs/DFs in human sample. Samples were ground with anhydrous sodium sulfate and extracted with methylene chloride by soxhlet extraction. The extract was concentrated and determined the weight of fat. Subsequently, whole extractable fat was subjected to clean up procedures. Scheme of the analytical procedure is shown in Fig.1.

Equipment

Analysis of PCDDs/DFs was performed with JMS-SX-102A mass spectrometer (JEOL, Japan) connected to a Hewlett Packard HP 5890 II gas chromatograph. Analysis of organochlorine compounds (pesticides and PCBs) was performed with a ^{63}Ni electron caputer detector (ECD) connected to a Hewlett Packard HP 5890 II gas chromatograph.

RESULTS AND DISCUSSION

2,3,7,8-TeCDD and other PCDDs/DFs

Data of the individual PCDD and PCDF isomers identified in the adipose tissues and breast milk are shown in Table 1. The recovery of internal standard (^{13}C -labelled 2,3,7,8-TeCDD/DF and OCDD/DF) for all samples generally ranged between 40-90%. The mean value of 2,3,7,8-TeCDD for 19 of human adipose tissues and 16 of human breast milk were 4.3pg/g and 2.7pg/g, respectively. These levels were much lower than the data reported recently by A.Schecter et al. (TeCDD levels in adipose tissue from the south of Vietnam, 11.6pg/g⁴⁾, 1990). This reason may be because adipose tissues were almost collected from relatively young women (from 13 to 43 years old, mean=28). Those levels in this study were lower than that of industrialized countries. We couldn't find out the correlation with between 2,3,7,8-TeCDD level and an increasing age in

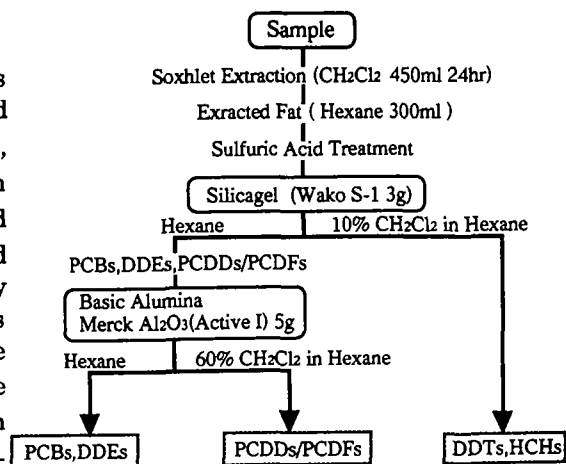


Fig.1 Simultaneous Determination of Organochlorine Compounds for human adipose tissue and breast milk

adipose tissue samples. (Fig.2) M.Matsuda et al. (1993) reported⁹⁾ that 2,3,7,8-TeCDD was detected in the south of Vietnam, but the level was very low. For these reason , we concluded that 2,3,7,8-TeCDD sprayed as defoliant no longer does not effect directly to human health.

For adipose tissues,the mean level of PCDFs was higher than that reported recently⁴⁾. Especially, the mean level of 1,2,3,4,6,7,8-HpCDF (46pg/g) was very high among of PCDFs. In this study, the level of each of PCDDs isomers was less than that reported recently⁴⁾, nevertheless the level of each of PCDFs isomers was higher than that. (Fig. 3) We consider that PCDFs contamination is in progress in Tay Ninh prov. In fact, high concentration of PCDFs in some soil samples from that area was reported by M.Matsuda et al.(1993)⁹⁾. It is well known that PCBs is one source of PCDFs pollution, but there was no correlation between PCDFs and PCBs in adipose tissues. In this reason, other sources except for PCBs might be considered for PCDFs pollution in Tay Ninh prov.

Data of 2,3,7,8-TeCDD toxic equivalents by age are shown in Fig.4. The mean of TEQ levels was 18pg TEQ/g fat basis and 12pg TEQ/g fat basis for adipose tissue and breast milk, respectively. For adipose tissue,TEQ level found in this study is lower than that in other industrialized countries; the United Kingdom (57pg TEQ/g)¹⁰⁾, Ehime prefecture;rural area in Japan (17pg TEQ/g,1993)¹¹⁾, Osaka prefecture;urban area in Japan,(26pg TEQ/g,1993)¹¹⁾. For breast milk, TEQ level found in this study is also lower than that in other industrialized countries; the Federal Republic of Germany (32 pg TEQ/g)¹²⁾, the Japan (24 pg TEQ/g)¹²⁾,the United State (17pg TEQ/g)¹²⁾ and Netherlands (30pg TEQ/g)¹²⁾. Comparing with either TEQ or 2,3,7,8-TeCDD levels,the present level of PCDDs/DFs in Vietnamese is lower than that of industrialize countries.

Organochlorine pesticides and PCBs

Date of organochlorine pesticides and PCBs are shown in Table 2. The mean of PCBs levels (310ng/g fat basis and 300ng/g fat basis, for adipose tissues and breast milk, respectively) was lower than that of other countries; Poland (860ng/g, in adipose tissue,1994)¹³⁾, Japan (1100ng/g, in breast milk,1977)¹⁴⁾, Spain (900ng/g, in breast milk,1991)¹⁵⁾. While, it was higher than that of Indian (120ng/g, in breast milk,1990)¹⁶⁾. During Second Indochina War, PCBs could have been use in military activities. Tay Ninh prov. was one of the worst hit area by such activities.¹⁷⁻¹⁸⁾ V.D.THAO et al. reported⁶⁾ that the high level PCBs (92 ng/g dry wt in soil) was found in former U.S.Air Force base grounds in Tay Ninh prov.. That data was much higher than Japanese soils (37ng/g wt)¹⁹⁾. Investigation of level of PCBs may be necessarily to continue for human being in the south of Vietnam.

The levels of DDTs in adipose tissues (*p,p'*-DDE 3200ng/g fat basis, *p,p'*-DDT 1700ng/g fat basis) and breast milk (*p,p'*-DDE 3400ng/g fat basis, *p,p'*-DDT 1400ng/g fat basis) in the present study were considerably. The continuing usage of DDTs in agriculture and public health operations is explicable from their isomer or metabolite pattern in adipose tissues and breast milk.

K.Kannan et al.⁷⁾ (1992) reported that elevated concentration of PCBs, DDTs and HCHs were found in animal fat, meat, butter and seafood from Vietnam.(Fig.5) Therefore, we concluded that the predominant sources of PCBs and DDTs contamination in human body were daily products.

HUTOX

CONCLUSIONS

- As a result of analyzed 19 of adipose tissue samples and 16 of breast milk samples in Tay Ninh prov., levels of 2,3,7,8-TeCDD from defoliant were very low.
- Human adipose tissues and breast milk from the south of Vietnam received high contamination by DDTs which is using as Maralaria control. Research efforts in appropriate technology and safety devices are critically needed in Vietnam.

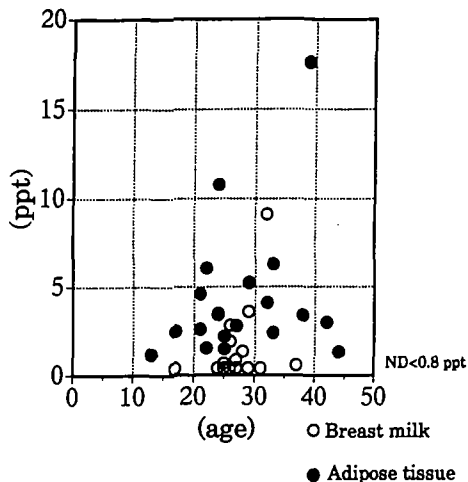


Fig.2 2,3,7,8-TeCDD levels by age in Tay Ninh (1991)

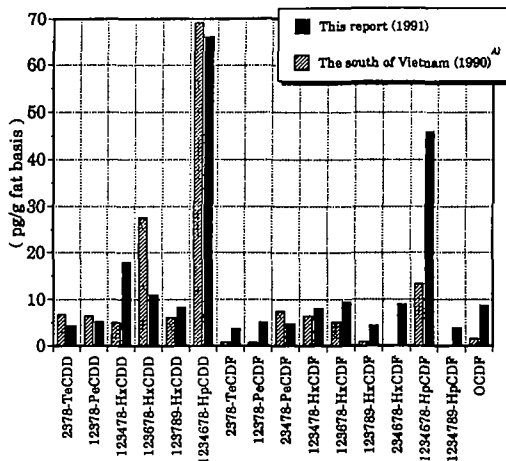


Fig.3 Levels of PCDDs/DFs (except for OCDF) in adipose tissues from the south of Vietnam

A) A.Schecter et al. (1990) ⁶⁾

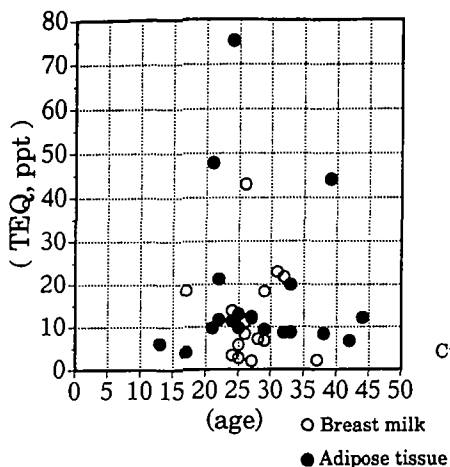


Fig.4 2,3,7,8-TeCDD toxic equivalents by age in Tay Ninh (1991)

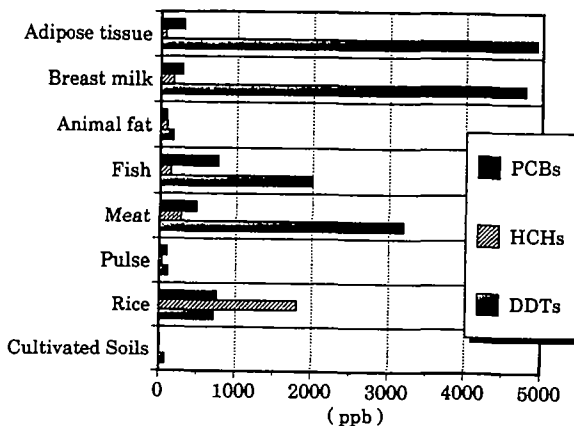


Fig.5 Levels of organochlorine pesticides and PCBs in foodstuffs, soil and human samples from the south of Vietnam

References: Animal fat, Fish, Meat, Pulse and Rice, by fat basis (K.Kanan et al. 1992) ⁷⁾
Cultivated Soil, by dry wt (V.D.Thao et al., 1992) ⁸⁾

Table 1 Levels (pg/g, fat basis) of PCDDs/DFs in human adipose tissue and breast milk from the south of Vietnam (Tay Ninh),1991

Age (n=)	Human adipose tissue				Breast milk	
	13~21	22~29	30~44	mean	17~37	mean
	4	7	8		16	
2378-TeCDD	1.1 — 4.6	1.5 — 11	1.3 — 18	4.3	<0.8 — 9.1	2.7
12378-PeCDD	<1.3 — 8.6	1.7 — 13	<1.1 — 17	5.2	<0.5 — 9.7	4.1
123478-HxCDD	<0.1 — 89	1 — 140	0.9 — 54	18	<0.8 — 16	4.6
123678-HxCDD	<0.1 — 30	3.3 — 57	<1 — 44	11	<0.8 — 32	12
123789-HxCDD	<0.1 — 24	<1 — 37	<1 — 53	8.2	<0.8 — 11	3.8
1234678-HpCDD	1.7 — 280	8.6 — 460	1 — 90	66	11 — 120	39
OCDD	20 — 37	99 — 230	72 — 500	180	120 — 220	190
2378-TeCDF	1.0 — 11	2.1 — 8.7	0.8 — 5.7	3.7	0.1 — 27	6.1
12378-PeCDF	1.7 — 10	1.7 — 21	2.0 — 4.5	5.1	1.8 — 13	5.4
23478-PeCDF	2.4 — 9.4	1.7 — 14	3.1 — 22	4.8	<0.5 — 46	7.4
123478-HxCDF	<0.9 — 28	<0.7 — 44	0.8 — 41	8.0	<0.8 — 10	4.7
123678-HxCDF	<0.9 — 30	1.7 — 49	<1.2 — 38	9.4	<0.8 — 12	4.6
123789-HxCDF	<0.9 — 31	<0.6 — 21	<0.8 — 4.5	4.4	<0.8 — 41	2.5
234678-HxCDF	<0.9 — 33	1.7 — 50	<1 — 41	8.9	<0.8 — 40	5.3
1234678-HpCDF	<1.6 — 260	3.2 — 420	<1.6 — 78	46	<1 — 63	14
1234789-HpCDF	1.5 — 18	<1.6 — 27	<0.2 — 4.5	3.9	<0.4 — 1.3	0.9
OCDF	<0.9 — 61	<0.7 — 50	<0.6 — 15	8.7	<0.9 — 5.2	2.8
PCDDs	20 — 680	56 — 360	76 — 570	290	15 — 300	77
PCDFs	7.8 — 500	23 — 710	18 — 150	100	3.2 — 152	50
TEQ	4.1 — 48	9.8 — 75	6.8 — 44	18	2.1 — 42	12

Table 2 Levels (ng/g, fat basis) of organochlorine compounds in human adipose tissue and breast milk from the south of Vietnam (Tay Ninh),1991

Age n	Human adipose tissue				Breast milk	
	13~21	22~29	30~44	mean	17~37	mean
	4	7	8		16	
p,p'-DDE	470 — 4000	1500 — 8500	760 — 8200	3200	420 — 7600	3400
p,p'-DDT	640 — 3000	100 — 9000	400 — 2400	1700	13 — 3100	1400
p,p'-DDD	30 — 44	9.1 — 1800	16 — 48	140	14 — 240	50
o,p'-DDE	1 — 9	<0.1 — 16	1 — 6.2	4.1	1 — 9.8	4.9
o,p'-DDT	43 — 150	5.6 — 630	10 — 250	180	26 — 340	130
o,p'-DDD	<0.1 — 1	<0.1 — 34	<0.1 — 1	3.4	<0.1 — 1	0.2
α -HCH	1 — 6	<0.1 — 31	1.3 — 30	6.8	<0.1 — 0.5	0.3
β -HCH	3 — 16	<0.1 — 400	1.1 — 88	42	17 — 330	140
γ -HCH	6 — 24	3 — 88	2.9 — 13	14	1.2 — 142	21
δ -HCH	<0.1 — 1	<0.1 — 6	<0.1 — 1	1.5	<0.1 — 14	12
PCBs	80 — 400	120 — 1200	130 — 510	310	90 — 630	300

HUTOX

REFERENCES

- 1) A.Schecter, J.Ryan and J.Constable(1986):CHLORINATED DIBENZO-P-DIOXIN AND DIBENZOFURAN LEVELS IN HUMAN ADIPOSE TISSUE AND MILK SAMPLES FROM THE NORTH AND SOUTH OF VIETNAM:Chemosphere,15,1613-1620
- 2) A.Schecter, H.Y.Tong, S.J.Monson, M.L.Gross and J.Constable (1989):ADIPOSE TISSUE LEVELS OF 2,3,7,8-TCDD IN VIETNAMESE ADULTS LIVING IN VIETNAM,1984-87 : Chemosphere,18,1057-1062
- 3) A.Schecter, Dan Vu, H.Y.Tong, S.J.Monson, M.L.Gross, J.D.Constable(1989):LEVELS OF 2,3,7,8-TCDD AND 2,3,7,8-TCDF IN HUMAN ADIPOSE TISSUE FROM HOSPITALIZED PERSONS IN NORTH AND SOUTH OF VIETNAM 1984-88:Chemosphere,19,1001-1004
- 4) A.Schecter, H.Y.Tong, S.J.Monson, M.L.Goss, S.Raisanen, T.Karhunen , E.K. Osterlund , J.D.Constable, Hoang Dinh Cau, Le Gao Dai, Hoang Tri Quynh, Ton Duc Lang, Nguyen Thi Phuong, Phan Hoang Phiet, Dau Vu (1990):HUMAN ADIPOSE TISSUE DIOXIN AND DIBENZOFURAN LEVELS AND "DIOXIN TOXIC EQUIVALENTS"IN PATIENTS FROM THE NORTH AND SOUTH OF VIETNAM:Chemosphere,20,943-950
- 5) A.Schecter, Peter Furst,Christiane Furst, Olaf Papke, Michael Ball, Le Cao Dai, Hoang Tri Quynh ,Nguyen Thi Ngoc Phoung, Albert Beim, Boris Vlasov, Vassant Chongchet, John D.Constable , Karan Charles (1991):DIOXIN,DIBENZOFURANS AND SELECTED CHLORINATED ORGANIC COMPOUNDS IN HUMAN MILK AND BLOOD FROM CAMBODIA ,GERMANY,THAILAND,THE U.S.A., THE U.S.S.R.,AND VIETNAM.:Chemosphere,23 , 1903-1912
- 6) V.D.THAO, M.KAWANO, M.MATSUDA, T.WAKIMOTO, R.TATSUKAWA, H.D.CAU, H.T.QUYNH (1993):CHLORINATED HYDROCARBON INSECTICIDE AND POLYCHLORINATED BIPHENYL RESIDUES IN SOILS FROM SOUTHERN PROVINCES OF VIETNAM:Inter J.Environ.Anal.Chem,50,147-159
- 7) K.Kannan, S.Tanabe, H.T.Quynh, N.D.Hue,and R.Tatsukawa (1992):Residue Pattern and Dietary Intake of Persistent Organochlorine Compounds in Foodstuffs from Vietnam :Arch. Environ. Contam.Toxicol,22,367-374
- 8) H.NAKAMURA, M.MATSUDA, T.WAKIMOTO (1993):Simultaneous Determination of Several Organochlorine Compounds(PCDDs/DFs,PCBs,DDTs,HCHs)in Limited Human Samples :Journal of Environmental Chemistry,3,450-451
- 9) M.MATSUDA,H.FUNEMO AND T.WAKIMOTO (1993):PCDDs/PCDFs Pollution in Vietnam Soils :Journal of Environmental Chemistry,3,418-419
- 10) Raquel Duarte-Davidson, Stuart J.Harrad,Susan Allen, Andrew S.Sewart,and Kevin C.Jones (1993):The Relative Contribution of Individual Polychlorinated Biphenyls (PCBs), Polychlorinated Dibenzo-p-dioxins (PCDDs) and Polychlorinated-p-furans(PCDFs) to Toxic Equivalent Values Derived for Bulked Human Adipose Tissue Samples from Wales,United Kingdom:Arch. Environ. Contam.Toxicol,24,100-107
- 11) N.Sawamoto, M.Matsuda, T.Wakimoto, S.Moriwaki, K.Mandai (1994):The transition of PCDDs/DFs in human adipose tissues in Japan: Journal of Environmental Chemistry,4,406-408
- 12) World Health Organisation .Levels of PCBs, PCDDs,and PCDFs in breast milk: results of WHO-coordinated interlaboratory quality control studies and analytical field studies .FADL, Copenhagen.Environmental Health series 1989;34.
- 13) J.K.Ludwicki, K.Goralczyk(1994):Organochlorine Pesticides and PCBs in Human Adipose Tissues in Poland(1994): Bull.Environ.Contam.Toxicol,52,400-403
- 14)Yakushiji, T.;Watanabe,I.;Kuwabara,K.;Yoshida,S.;Koyama,K.;Kunita,N(1979):Levels of polychlorinated biphenyls (PCBs) and organochlorine pesticides in human milk and blood collected in Osaka Prefecture from 1972-1977. Int. Arch. Occup. Environ.Health,43,1-15
- 15)C.Conde, C.Maluenda and C.Arrabal (1993):Organochlorine Residues in Human Milk in Spain. Polychlorinated Biphenyls (PCBs)from 1988 to 1991 :Bull.Environ.Contam. Toxicol,51 ,832-837
- 16)S.Tanabe, F.Gondaira, A.Subramanian, A.Ramesh, D.Mohan, P.Kumaran, V.K.Venugopalan and R.Tatsukawa (1990):Specific Pattern of Persistent Organochlorine Residues in Human Breast Milk from South India:J.Agric.Food Chem.,38,899-903
- 17)A.H.Westing, Ecotoxicology and Climate, (John Wiley and Sons Ltd.,1989) pp237-257.
- 18) H.T.Quynh, L.C.Dai, L.Thanh, L.B.Van, L.T.Tien, L.N.Thanh and L.B.Thuy,Proceedings of National Symposium:"Long-term Effects of Chemical War in Vietnam"(Committee 10-80, Hanoi ,in Vietnamese1986)pp 19-23
- 19) Yakushiji, Masters Thesis (Ehime University, Japan 1972),85pp