Contamination levels of PCDDs, PCDFs and non-ortho coplanar PCBs in blood samples collected from residents in high cancer-causing area close to Batch-type Municipal solid waste incinerator in Japan

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Introduction

There is a batch-type MSW incinerator with a capacity of 60 tons/day in Ryugasaki city of Ibaraki prefecture, Japan. In the residents within 1.2 km under the downwind from the incinerator, their death rate from cancer was epidemiologically revealed to be remarkably lifted at a high level of 42%, ca twice times of 21% in the outside area. It is well known that dioxin analogues composing of polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and non-ortho coplanar PCBs (Co-PCBs) have promortive activity for carcinogenesis. This insists that these dioxin analogues might be the causal agent for the high cancer death rate. Therefore, we determined these compounds in the soil around the incinerator. Consequently, it was found the residential area of high cancer death rate was highly polluted by dioxin analogues emitted from the incineration facility[1].

In this study, we first developed an analytical manhood for dioxin analogues in the blood, and analyzed blood samples from the resident in order to evaluate risk assessment for human pollution in the concern area.

Materials and Methods

Blood sample: About 60 ml of peripheral blood samples were individually collected from 13 men (mean age: 45 years and range: 23 - 63 years) and 5 women (mean age: 46 years and range: 30 - 72) in the high cancer death rate area within 2 km from the incinerator, Ryugasaki city, Ibaraki prefecture in March, 1996. After sampling, serum was separeted from the each sample and stored in-20℃ until analysis.

Analytical method: After spiking of internal standards (five $^{13}C^{12}$-PCDDs, five $^{13}C^{12}$-PCDFs and three $^{13}C^{12}$-Co-PCBs, each 100 pg), lipid was extracted from ca. 20 g of serum sample

according to a method by Patterson et al.[2]. The lipid sample was cleaned up on a multi-layer column and alumina column according to our previous report[3]. The purified extract was dissolved in 10 μl of n-decane and analyzed for PCDFs, PCDDs and Co-PCBs in EI-SIM mode at a resolution of 10000 using a Hewlett・Packard 6890 gas chromatograph-JEOL 700M mass spectrometer according our report described elsewhere[3].
The concentrations of PCDDs, PCDFs and Co-PCBs were corrected with the recoveries of their respective internal standards. Finally, the 2,3,7,8-TCDD toxicity equivalency quantity (TEQ) were calculated for PCDDs and PCDFs using I-TEFs[4] and for Co-PCBs using TEFs[5].

Results and discussion

In Japan, there is no standard analytical method for dioxin analogues in human blood. Therefore, we tried to establish a suitable blood analytical method. In this study, we compared four lipid extraction methods from serum and/or plasma reported by Schecter et al.[6], Pattern et al.[3], Nygren et al.[7] and Japanese standard method[8]. After addition of $^{13}C_{12}$-labeled internal standards, lipid was extracted from cattle serum according to above four methods. As illustrated in Fig. 1, the method by Patternson et al. gave the most effective lipid extraction, showing the extracted lipid weight to be equivalent to 1.5 to 2.3 times greater than those of other methods. In addition, we analyzed dioxin analogues in cattle serum using a combination of these four extraction method and our purification method including multi-layer silica gel column and alumina column chromatography. Consequently, the combination of method by Shecter et al. or by Patterson et al. showed higher recoveries of internal standards than did others (Fig. 2). From these two results, we decided an assortment of Pattersons' and our methods to be more excellent analytical method for serum.
As shown in Table 1[9-14], the level of PCDDs/PCDFs in blood from general population in various countries is in a range of 15 to 29 pgTEQ/g lipid, on the average. In general, the value in Japan, USA and Germany was higher than that of Russia and North Vietnam. Compared to these, the blood level of residents in area around a batch a batch-type MSW incinerator in Ryugasaki city of Ibaraki prefecture, Japan showed remarkably higher with a level of 81 pgTEQ/g lipid, on the average in men and 149 pgTEQ/g lipid in women. In addition, their level was much higher than that (36 pgTEQ/g lipid) of south Vietnamese who had been exposed to Agent Orange composing mainly of 2,4,5-T during a long period of 1962 to 1970. As well as a case of PCDDs/PCDFs, the residents gave a lifted level of 37 pgTEQ/g lipid in men and 23 pgTEQ/g lipid in women, than did general person in Fukuoka, Japan (5.6 and 11 pgTEQ/g lipid) and Germany (9.1 pgTEQ/g lipid). Since 1986, the amount of MSW exceeded the combustion capacity (60 tons/day) of the incinerator, because of a rapid population increase by a construction of new towns in the surrounding regions, resulting in the incineration under an incomplete condition to have been continued until recent years. Therefore, this heavily human pollution in area close to the facility is considered to be brought by direct inhalation of dioxin analogues from flue gas , and by indirect intake of vegetables contaminated by the flu gas.

Now, we are going to analyze additional 20 blood samples, and will present the data in this symposium.

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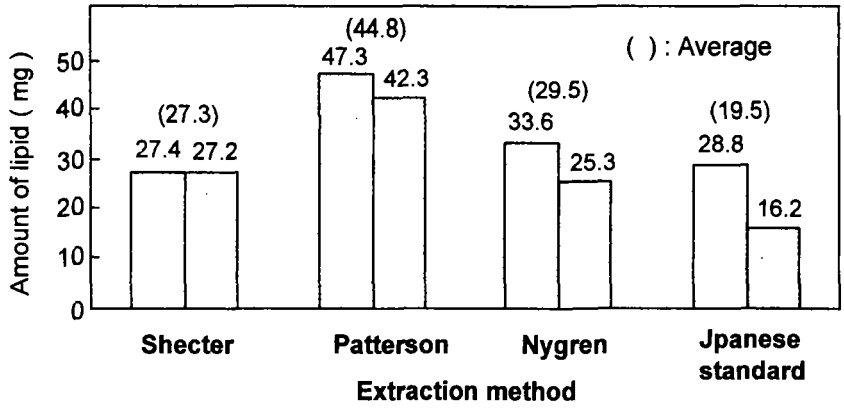

Fig. 1. Comparison of lipid amount extracted from bovine serum (15 ml) by four extraction methods

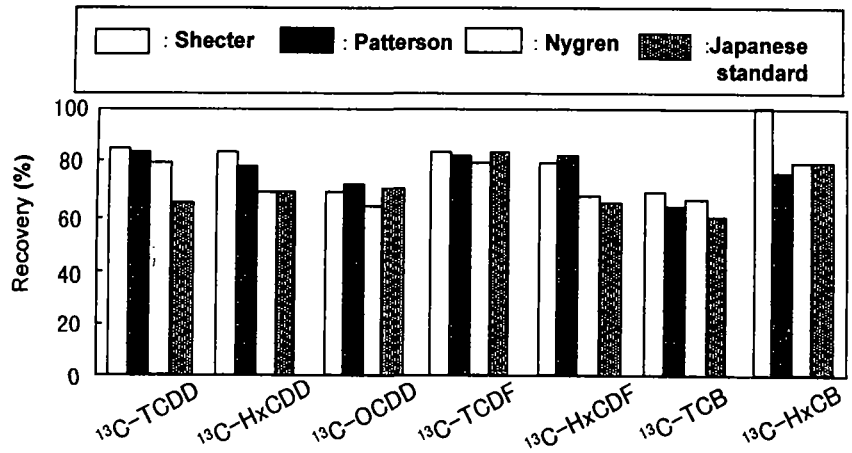

Fig. 2. Comparison of recovery of $^{13}C_{12}$-labeled internal standards in bovine serum by four combinated analytical methods

Table 1. PCDDs, PCDFs and Co-PCBs in human blood from various countries

(ppt, blood fat)

Country Location	Japan* Ibaraki	Japan* Ibaraki	Japan[9] Fukuoka	Japan[10] Fukuoka	Japan[11] Fukuoka	Germany[12] Duisuburg Simmerath
Year	1996	1996	1996	1993	1991-92	1996
Number	13	5	4	60	3	61
Sex	Man	Woman	Man	Woman		Woman
Age	45 (23-69)	46 (30-72)	40-46	20 (18-29)	Adult	31 (25-35)
PCDDs	57 (20-174)	135 (11-443)	15	10 (4.8-19)	14 (11-15)	12 (3.5-28)
PCDFs	23 (13-49)	14 (7-21)	14	7.1 (2.9-12)	11 (8-12)	11 (5.0-24)
Co-PCBs	37 (10-82)	23 (13-37)	11	5.6 (1.4-12)	-	-
PCDDs+PCDFs	81 (34-200)	149 (22-463)	29	17 (7.7-32)	24 (20-27)	23(8.6-53)

Country Location	Germany[13]	USA[14]	USA[15] Guam	Russia[15] St. Petersburug	Vietnam[15] North	Vietnam[15] South
Year	1994	1996	1991-92	1980-91	1980-91	1980-91
Number	134	100**	10	60**	82	383
Sex						
Age	40 (22-69)	Adult	>40	>40	>40	>40
PCDDs	11 (2.6-279	20.1	21	11	7	26
PCDFs	8.5 ((11-26)	7.5	7	6	8	10
Co-PCBs	9.1 (1.8-33)	-	-	-	-	-
PCDDs+PCDFs	19 (14-53)	27.6	28	17	15	36

Figures in the parenthesis show minimum to maximum.
*: this study * *: pooled sample -: No data