RESIDUE LEVEL AND CONTAMINATION SOURCE OF POLYCHLORINATED DIBENZO-P-DIOXIN AND DIBENZOFURANS IN THE KOREAN POPULATION

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Introduction

Chlorinated hydrocarbons, such as polychlorinated biphenyls, polychlorinated dibenzo-p-dioxin and dibenzofurans are ubiquitous contaminants in the global environment. Due to their bioaccumulation potential, organochlorines tend to magnify in the food chain and cause adverse effects on human and wildlife situated at the top of the food chain. Investigators have reported that 2378-tetrachlorodibenzo-p-dioxin (2378-TeCDD) and its congeners produce a variety of toxic effects, such as teratogenicity, carcinogenicity, immunotoxicity and lethality, in some species of experimental animals. Furthermore, these compounds are detected in high concentrations in human tissues from some developed and industrialized countries. Thus, these compounds, namely PCDD/DFs have attracted considerable attention regarding public health. However, in Korea the contamination of PCDD/DFs and other related compounds is not clear in the general population.

In this study measured the levels of PCDD/DFs in human adipose tissue and milk in the Korean population as well as in food samples. The purpose of the present study is to measure the contamination levels and investigate the residue profiles of PCDD/DFs in human for Korea. Furthermore, We will try to estimate the dietary exposure and assess the risks associated with the consumption of fishes and meats.

Materials and Methods

Sample Collection

The human adipose samples analysed in this study were collected at hospital located in Seoul(a large city with many types of PCDD/DFs emission), Masan(a town located in an industrialized area) and Jinju(a rural agriculture area). The human milk samples were only collected from Masan city. Raw foodstuffs were collected from different locations in Masan and Pusan from Korea during July 1994 to March 1995. Table 1 lists the details of food samples used in the present study collected from Korea. Fish samples representing different species, namely; Mackerel (*Scomber australasicus*), Croaker (*Argyrosomus argentatus*), Alaska Pollack (*Theragra chalcogramma*) and Hair tail (*Trichiurus iepturus*) were collected from fish markets in Masan and Pusan. Animal origin food samples such as chicken fat, pork fat and beef meat were obtained from butcher shop in markets, transported to laboratory with dry ice and preserved at -20℃ until analysis.

Determination of PCDDs/DFs and dioxin-like PCBs

Approximately 10g of human samples and 50 g of food samples were homogenized, freeze-dried and extracted using a Soxhlet apparatus with dichloromethane. Fractionation was carried out with an activated silica-gel and an alumina column. In the charcoal-impregnated silica-gel mixture column fractionation step, adsorbed PCDDs/DFs and dioxin-like PCBs were eluted into two fractions. The first fraction, eluted with 25% dichloromethane in hexane, consisted of mono-*ortho*-substituted CBs. The second fraction, eluted with toluene, comprised PCDDs/DFs and non-*ortho*-substituted CBs. Identification and quantification of PCDD/DF homologues and non-*ortho*- and mono-*ortho*- substituted CBs was performed by HRGC-HRMS (R>10000, 10% valley).

Result and Discussion

1. PCDD/DFs contamination statue of Human adipose tissue

The mean concentrations on a lipid weight basis of PCDDs and PCDFs in human adipose samples from Seoul, Masan and Jinju were 413 pg/g (range : 61 -6390 pg/g) and 38 pg/g (3.1 - 148 pg/g), respectively. The mean values of International Dioxin Toxic Equivalents (I-TEQ) of PCDDs and PCDFs detected in these samples from the three regions were calculated as 9 pg TEQ/g and 8 pg TEQ/ g, respectively. The residue levels of PCDD/DFs in adipose tissues taken from the Korean population were generally lower than those of other countries.

The percent contribution of TeCDD to total PCDD-TEQs was 35% followed by 32% for PeCDD, 25% for HxCDD, 4% for HpCDD and OCDD. In the case of PCDF-TEQ, PeCDF occupies 78.3%, with 18.2 % for HxCDF, 2.86% for TeCDF, 0.6% for HpCDF and 0.04% for OCDF. Especially, 23478-PeCDF-TEQ at 37% most greatly contributed to the total dioin-TEQ, among the analyzed 2378-chlorine substituted compounds. Although the contribution of the HxCDF congener in Korean samples was slightly higher than that of Japan, on the whole, the composition pattern of PCDFs resembled to that of Japan. The contribution to TEQ of 23478-PeCDF in total PCDFs in human adipose tissues was markedly higher in samples from Japan (86%) and Korea (76%), than in those from Spain (62%) and the UK (63%). It was estimated that more than 90% of PCDD/DFs uptake by humans was attributed to food consumption. Based on this information, the similarity in the percent contribution of PCDD/DFs between Japanese and Korean tissues was probly due to similar dietary habits.

PCDD/DFs and TEQ concentrations determined in the present study, we could not find statistical significant correlation among the figures obtained from the samples collected at the three regions. Similar results have been reported on human milk and human adipose samples from various countries. The lack of regional differences of PCDD/DFs contamination in Korean was probably due to the uniform food supply. However, the residue levels of PCDDs, especially OCDD, for some donors in Seoul was detected at equal or slightly higher levels than in industrialized countries. Accordingly, the possibility of an

increase of PCDDs residues in the Seoul population can not be excluded.

2. PCDD/DFs contamination statue of Human milk

The distribution profiles of PCDD/DF congeners in human adipose tissue and milk of Korean were slightly different. PCDF congeners were more predominant in human milk than in the adipose tissue, whilst the concentrations of PCDDD congeners were higher than in the adipose tissue. Therefore, although the individual congeners of PCDD/DFs found in milk samples was broadly similar to that of human adipose tissue, PCDF congeners were present at higher concentrations in human milk compared with in adipose tissue samples. These results indicate that less persistent PCDF congeners which have shorter half lives and more readily eliminated from the human body are present in human milk large proportions, whilst more stable PCDDs are more easily retained in the adipose tissue.

Based on the analytical data assuming a daily consumption of 800mL milk with 3% fat, the average daily intake of PCDD/DFs via human milk for a baby weighing 5 Kg could be calculated. The average daily intake of PCDD/DFs for a breast-fed baby has been calculated to me 39pg/kg body weight/day for 2378-TeCDD and 86 pg/kg/day for total TEQ. These levels are far above all virtually safe doses (VSD) or tolerable daily intake (TDI) values proposed by health authorities in various countries, ranging from 0.001 (US EPA) to 4 (WHO) pg/kg/day. It must be mentioned, however, that the VSD and TDI concepts are based on intake during an entire lifetime and therefore should not be applied to the relatively short period of nursing. On the other hand, it should be considered that a breast-feeding infant might be more sensitive to the toxic chemicals than an adult.

3. PCDD/DFs and Dioxin-like PCBs contamination statue of food

The total PCDDs/DFs were detected in all foodstuff samples. The mean concentrations on a wet weight basis of total PCDDs/DFs determined from the various foodstuffs were 0.43 pg/g (n=3) for alaska pollack, 0.85 pg/g (n=2) for chicken fat, 0.9 pg/g (n=2) for pork fat, 1.02 pg/g (n=3) for croaker, 1.69 pg/g (n=2) for beef meat, 1.86 pg/g (n=4) for hair tail and 2.84 pg/g (n=4) for mackerel.

The total dioxin-like PCB concentrations varied from a mean low value of 56 pg/g (on a wet weight basis) in pork fat samples to a mean high value of 175 pg/g in beef. For fish samples, dioxin-like PCB congeners were not analyzed.

On the whole, the contamination levels of total PCDDs/DFs in fish samples were higher than those in meat samples. Furthermore, the contributions of low-chlorinated PCDD/DFs on total PCDD/DF concentrations in fish samples were relatively higher than those in meat samples. This phenomenon was also reflected in the percent composition of PCDD/DFs-TEQ. The total PCDDs/DFs-TEQ concentrations ranged from 0.03 pg TEQ/g for pork fat to 0.89 pg TEQ/g for mackerel. Among the analyzed isomers of PCDDs/DFs in fish samples, percent contribution of TEQ for 2,3,4,7,8-PeCDF on total PCDDs/DFs was shown to be 42%. This suggests that fish consumption is a significant exposure source for low-

chlorinated PCDDs/DFs on the general population in Korea.

The daily intake of PCDDs/DFs via fish and meat consumption was calculated to be 42 pg TEQ/person /day, using the 1994 Korean food consumption rate. The values are equivalent to 0.7pg/kg body weight/day for TEQ, by calculation under consideration of 60 kg for body weight.

In overall, the mean daily intake of PCDDs/DFs via fish consumption was about 10 times higher than those via meat consumption. The average daily intake of PCDDs/DFs-TEQ via fish consumption from various countries was published 0.13pg/kg/day for UK, 0.04 pg/kg/day for USA, 0.28 pg/kg/day for Canada and 0.98 pg/kg/day for Japan, respectively. Although the daily dioxin TEQ intake via fish consumption in Korea (0.63 pg/kg/day) was lower than that in Japan, this value was higher than that for the other countries. It is important to note that fish consumption is one of the main sources of PCDDs/DFs exposure in the Korean population.