DIETARY INTAKE OF DIOXINS AMONG JAPANESE POPULATION ESTIMATED BY DUPLICATE PORTION ANALYSIS: LOW PROPORTION OF ADULTS WHOSE LONG-TERM INTAKE EXCEEDED THE TOLERABLE DAILY INTAKE

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Abstract

Dietary intake of dioxins was estimated by duplicate portion analysis of consecutive three-day food samples among 65 men and 234 women (aged 17-72 years), who were living in 60 different areas of 20 prefectures in Japan. The mean (median) intake of PCDDs+PCDFs, coplanar PCBs (co-PCBs) and total dioxins, expressed on the basis of toxicity equivalents (TEQ), was 0.51 (0.38), 0.64 (0.43) and 1.16 (0.84) pg/kg body weight/day, respectively. In multiple regression analysis, dietary intake of total dioxins was associated with intake amount of fish and shellfish, and milk and dairy products. Proportion of those whose dietary intake exceeded the tolerable daily intake, set by the World Health Organization (4 pg TEQ/kg/day) and European Union (2 pg TEQ/kg/day), was estimated at 2.7% and 12.5%, respectively. However, these proportions were considered to be overestimated because of the effect of day-to-day within-person variation. Therefore, the ratio of within- and between-person variance was estimated by applying random effects one way analysis of variance to repeated measurements for another group of 35 persons. When the effect of within-person variation of dietary intake was excluded, the proportion of subjects exceeding the TDI of WHO and EU decreased to 0.09 % and 4.2 %, respectively.

Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDDs), polychlorinated dibenzo-furans (PCDFs) and coplanar polychlorinated biphenyls (co-PCBs) are lipophilic and persistent environmental contaminants that exhibit a variety of toxic effects[1]. In general populations, over 90% of background exposure to dioxins originates from foods. Therefore, evaluation of dietary intake is crucial in the exposure assessment of dioxins.

There have been a lot of studies on the dietary intake of dioxin-related compounds among general populations of various countries. However, most of these previous studies were based on the market basket method, and the distribution (variance or standard deviation) of the intake remains largely unknown.

In Japan, "A study on the accumulation of dioxin-related compounds in humans" has been carried out under the supervision of the Ministry of Environment since 2002. The purpose of this project was to assess the blood levels and the dietary intake of dioxins and related factors among general Japanese population. In this paper, the authors report the 4-year results of the dioxin intake from foods, estimated by duplicate portion analysis. Furthermore, the authors estimated the proportion of subjects whose long-term dioxin intake exceeded the tolerable daily intake (TDI), taking into account the day-to-day within-person variation.

Materials and methods

The study population consisted of 299 subjects, 65 men and 234 women (aged 17-72 years), who were recruited from 60 different study areas of 20 prefectures in Japan (group 1). They were required to have been aged 15-72 years, living in the same study area for at least 10 years, and to have had no severe anemia. Participation was essentially voluntary. The purpose of the present study was explained thoroughly, and written informed consent was obtained from each participant. The surveys were performed between 2002 and 2005.

All food items or menus and beverages, exactly the same as those consumed during consecutive three days, were collected from each participant. Food samples were weighed, homogenized and mixed, and subjected to analysis for dioxins. In addition, kinds and weights of all food items and drinks consumed were recorded by nutritionists.

All measurements of PCDDs/PCDFs and co-PCBs in food samples were done at the Institute of General Science for the Environment, METOCEAN Environment Inc., Shizuoka, Japan, by isotope dilution high-resolution gas chromatography/mass spectrometry (GC/MS), after liquid/liquid extraction and gel clean up. Calculation of toxicity equivalent (TEQ) was based on toxicity equivalency factors (TEF) of WHO 1998. Zero was assigned to values below the detection limit.

Single measurement of short-term dioxin intake has larger standard deviation (SD) than long-term intake because of random within-person variation[2]. Therefore, to assess the magnitude of the within- and between-person variance of dioxin intake, food samples of consecutive three days were collected three times in 1999 and 2000, from 35 persons (18 men and 17 women, 41-68 years of age, group 2) living in Chugoku region. Random-effects one-way analysis of variance[3] was applied to repeated measurements of dioxin intake in group 2, in order to estimate the ratio of within-person and between-person variance ($k= s_w^2/s_b^2$). Using the following equation, SD with no within-person variation, i.e., SD of long-term dioxin intake, was estimated.

$$\text{Observed } SD^2 = s_b^2 + s_w^2$$
$$= s_b^2 + k\, s_b^2$$

where s_b^2 is the true SD^2. Normal distribution with a mean of 299 subjects (group 1) and an SD with no within-person variation was considered, and the proportions of subjects whose dietary intake exceeded the tolerable daily intake (TDI), set by the WHO (1999) and European Union (EU) (2001), were estimated.

Results and Discussion

The mean (median) TEQ of PCDDs, PCDFs, co-PCBs and total dioxins was 14.97 (11.41), 14.23 (10.54), 36.67 (24.16) and 65.89 (48.63) pg-TEQ/day, respectively. The largest proportion of PCDDs, PCDFs and co-PCBs derived from 1,2,3,7,8-PeCDD, 2,3,4,7,8-PeCDF and PCB126, respectively. Table 1 presents the dietary intake of PCDDs+PCDFs, co-PCBs and total TEQ (pg-TEQ/kg body weight/day) according to sex, age, regional block, residential area and calendar year. For all subjects, the mean (median) intake of PCDDs+PCDFs, co-PCBs and total TEQ was 0.51 (0.38), 0.64 (0.43) and 1.16 (0.84) pg-TEQ/kg body weight/day, respectively. The intake of PCDDs+PCDFs, co-PCBs and total TEQs increased significantly with age, but did not differ according to sex. The intake of co-PCBs differed according to regional block (P=0.06), being somewhat higher in Chugoku and Shikoku, and Kyushu and Okinawa. Fishing villages showed the highest intake of PCDDs+PCDFs, co-PCBs and total TEQs, followed by farming villages and urban areas. In multiple regression analysis, TEQs of PCDDs+PCDFs, co-PCBs and total dioxins were significantly or marginally significantly associated with intake amount of fish and shellfish, and milk and dairy products (data not shown).

The proportion of those whose intake exceeded the TDI set by the WHO (1999, 4 pg-TEQ/kg/day) and EU (2001, 2 pg-TEQ/kg/day) was 2.7% and 12.5%, respectively. These proportions were considered to be overestimated because the results were based on single measurement of 3-day samples. In 35 persons (group 2) with three repeated measurements of dioxin intake (N=105), the mean, median and geometric mean of total TEQ was 1.48, 1.10 and 1.08 pg TEQ/kg/day, respectively. Using random effects one-way analysis of variance, the ratio of s_w^2/s_b^2 for log (total TEQ) was estimated at 1.48, which was much larger than that of log (food weight) (0.50). In 299 persons of group 1, log (total dioxins) followed a normal distribution with a mean of -0.161 and an SD of 0.779. Assuming a normal distribution with a mean of -0.161 and an SD of $0.779/(1+1.48)^{1/2}=0.495$, the proportion of those whose log-term dietary intake exceeded the 4 and 2 pg TEQ/kg/day was estimated at 0.09% and 4.2%, respectively.

To our knowledge, this is the first study to evaluate the distribution of long-term intake of dioxins, using duplicate portion analysis and simultaneously accounting for the within-person variation. Although very low percent of Japanese adults were considered to ingest dioxins exceeding the TDI, it remains unclear whether general populations are not at risk for adverse effects caused by dioxins. An inverse association between prenatal PCBs and dioxin exposure and neurodevelopment of infants has been reported among general population in Europe, U.S. and Japan[4,5]. Therefore, it may be prudent to further decrease the dioxin intake and body burden by reducing the release of dioxins into the environment.

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Table 1. Dietary intake of dioxins (pg-TEQ/kg/day) according to sex, age, region, residential area, and calendar year.

			PCDDs+PCDFs			co-PCBs		Total-TEQ	
			No.	Mean	Median (25%, 75%)	Mean	Median (25%, 75%)	Mean	Median (25%, 75%)
Total			299	0.51	0.38 (0.25, 0.65)	0.64	0.43 (0.21, 0.77)	1.16	0.84 (0.49, 1.4)
Sex	Men		65	0.47	0.38 (0.19, 0.69)	0.63	0.36 (0.17, 0.80)	1.11	0.79 (0.41, 1.4)
	Women		234	0.52	0.39 (0.27, 0.64)	0.65	0.43 (0.23, 0.75)	1.17	0.855 (0.50, 1.4)
					P=0.45[a]		P=0.45[a]		P=0.54[a]
Age	10-29		32	0.45	0.32 (0.175, 0.575)	0.52	0.37 (0.14, 0.775)	0.98	0.72 (0.31, 1.4)
	30-39		54	0.37	0.295 (0.22, 0.50)	0.39	0.28 (0.16, 0.55)	0.77	0.595 (0.40, 0.98)
	40-49		65	0.47	0.41 (0.27, 0.54)	0.56	0.39 (0.22, 0.62)	1.03	0.71 (0.51, 1.2)
	50-59		80	0.56	0.43 (0.28, 0.705)	0.80	0.485 (0.25, 0.925)	1.35	0.905 (0.55, 1.6)
	60-69		68	0.64	0.50 (0.295, 0.79)	0.79	0.63 (0.325, 1.00)	1.44	1.10 (0.745, 1.85)
					P=0.0005[b]		P<0.0001[b]		P<0.0001[b]
Region	Hokkaido, Tohoku		60	0.45	0.37 (0.255, 0.55)	0.52	0.355 (0.195, 0.575)	0.97	0.775 (0.48, 1.2)
	Kanto, Koshin-etsu		59	0.58	0.34 (0.21, 0.70)	0.72	0.39 (0.21, 0.81)	1.30	0.78 (0.47, 1.6)
	Tokai, Hokuriku, Kinki		60	0.43	0.36 (0.235, 0.585)	0.57	0.34 (0.165, 0.74)	1.01	0.665 (0.475, 1.4)
	Chugoku, Shikoku		60	0.54	0.435 (0.23, 0.715)	0.67	0.54 (0.185, 0.965)	1.21	0.985 (0.495, 1.7)
	Kyushu, Okinawa		60	0.56	0.485 (0.29, 0.69)	0.74	0.54 (0.325, 0.84)	1.31	1.00 (0.74, 1.6)
					P=0.22[b]		P=0.06[b]		P=0.11[b]
Residential area		Urban	103	0.42	0.31 (0.21, 0.60)	0.54	0.37 (0.18, 0.68)	0.96	0.77 (0.46, 1.3)
		Farming village	101	0.51	0.37 (0.25, 0.66)	0.64	0.37 (0.21, 0.73)	1.15	0.79 (0.49, 1.2)
		Fishing village	95	0.62	0.49 (0.32, 0.75)	0.75	0.55 (0.29, 0.82)	1.38	1.10 (0.64, 1.6)
					P=0.007[b]		P=0.03		P=0.01
Year	2002		75	0.47	0.41 (0.26, 0.59)	0.80	0.48 (0.23, 0.87)	1.27	0.86 (0.52, 1.5)
	2003		74	0.66	0.535 (0.34, 0.81)	0.67	0.465 (0.21, 0.94)	1.33	1.05 (0.64, 1.8)
	2004		75	0.44	0.35 (0.24, 0.63)	0.58	0.42 (0.22, 0.74)	1.02	0.78 (0.46, 1.4)
	2005		75	0.49	0.30 (0.19, 0.50)	0.53	0.34 (0.20, 0.66)	1.01	0.71 (0.40, 1.1)
					P=0.0001[b]		P=0.31[b]		P=0.02[b]

[a] Wilcoxon rank sum test. [b] Kruskal-Wallis test.