RELATIONSHIP BETWEEN FOOD PREFERENCE IN WOMEN AND PCDDs/DFs LEVELS IN BREAST MILK IN THE NORTHERN DISTRICT OF JAPAN

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Abstract

We analyzed PCDDs/DFs in breast milk samples donated by healthy mothers in Ishikawa Prefecture in the northern district of Japan, and found out that food preference during pregnancy contributed to the levels of specific isomers in breast milk. In this paper we emphasized the necessity to concentrate our effort not only on estimating TEQ intake via ingestion, but also on understanding the relationship between food preference and isomer profiles for elucidation of exposure sources of DXNs, in order to facilitate risk management against subtle adverse health effect.

Introduction

It is generally accepted that intakes of PCDDs/DFs (= DXNs) from food account for the majority of human exposure to such polychlorinated compounds, other than occupational and accidental exposure. In the last decade, numerous studies have been performed nationwide in Japan, in relation to the Ministry of Health, Labor and Welfare, Government of Japan, and reported food contamination with DXNs and human intake of DXNs via ingestion. Efforts of such studies were preferably expended to indicate a daily intake of DXNs and establish a tolerable daily intake, using the TEF scheme and TEQ methodology. In this field, however, informations derived from a relationship between food habit and DXN profiles in human tissue samples, are not involved. Therefore the scope of discussion does not extend exposure sources of DXNs. We consider that such details seem to be quite important for risk management in a public health aspect. Accordingly taking it into consideration, that dietary intake may relate to levels and profiles of DXNs in breast milk, we focused our studies on mothers living in a prefecture in the northern district of Japan, where the traditional Japanese diet based on rice supplemented seafood is still rooted, to find out a relationship between food preference in women and DXN levels of in breast milk.

Materials and Methods

The study included 35 healthy mothers (15 primiparae and 20 multiparae; mean age 29.3±4.00 yr) who delivered in Ishikawa Prefecture which is located in the northern part of Japan, and faces to the Sea of Japan.

10 ml of breast milk was collected in 2003 from each volunteer during 5-7 days after parturition. After a range of pretreatment procedures for DXNs, quantitation was performed by a high resolution mass spectrometer (HRMS; JEOL MStation- JMS700) equipped with a gas chromatograph (GC; HP-6890).

The nutritional data including frequency of food ingesta during pregnancy were gathered using self-administered questionnaire which modified a semi-quantitative food frequency questionnaire designed by Katagiri et al[1]. The daily/weekly average food intake for each food group was calculated on the Classification of the National Nutrition Survey.

Data were statistically analyzed using the SPSS version 11.0 series for Windows, and logarithmetic transformation of the measured values of DXNs was performed to improve normality. Focusing on rich protein foods like meat, egg, fish, pulse and milk, simple correlations between every intake of these foods and the DXN levels in breast milk were tested by Spearman's rho to determine the relationship between them. Additionally Student's *t*-test was used in the analysis of data to determine significant bivariate differences between food preference and DXN levels in breast milk.

Results and Discussion

DXN levels in breast milk were shown in Table 1. Specifically high level in PCDDs were observed in 1,2,3,6,7,8-HxCDD and OCDD, while only 2,3,4,7,8-PCDF was a specific high level isomer in PCDFs. Additionally ∑PCDDs-TEQ and ∑PCDFs-TEQ were 2.76 and 2.04 pg TEQ/ g lipid, respectively. As shown in Table 2, significant positive correlations were found between fish intake and TCDD (r = 0.359, $P < 0.05$), 1,2,3,7,8,9-HxCDF ($r = 0.412$, $P < 0.05$), and 1,2,3,4,6,7,8-HpCDF ($r = 0.408$, $P < 0.05$) levels in breast milk. Table 3 shows that DXN level difference between two groups, namely; those who ingested green-yellow vegetables more than 200g per day, and the rest who ingested those vegetables 200g or below per day. It is noticeable that the DXN levels of the subjects with comparatively high ingestion of green-yellow vegetables were generally higher than those of the rest. Especially level difference of 1,2,3,6,7,8-HxCDD was statistically significant ($P < 0.05$); 10.24 pg/g-lipid for the subjects with higher ingestion, and 5.48 pg/g-lipid for the rest. Similarly, as shown in Table 4, the DXN levels of the subjects who ate fruits once or more everyday were generally higher than those of the rest. Such a tendency was remarkably observed in PCDFs, and ∑PCDFs-TEQ level (2.58 pg TEQ /g-lipid) of the subjects who ate fruits once or more everyday was significantly higher than that of the rest (1.77 pg/ g-lipid). In response to this, statistically significant level differences were recognized in three isomers of PCDFs; 1,2,3,7,8-PCDF, 2,3,4,7,8-PCDF and 2,3,4,6,7,8-HxCDF ($P < 0.05$). On the other hand, statistically significant level difference in PCDDs was recognized in only 1,2,3,4,7,8- HxCDD ($P < 0.05$).

The analytical results of DXN levels in breast milk showed that the concentration levels were very low in comparison with those in Japan's urban areas, though the isomer profiles were very similar. It is generally acknowledged that the isomer profiles reflect the characteristics of exposure sources. Therefore major exposure sources to DXNs in Ishikawa Prefecture seem to be similar to other areas in Japan.

Because of the high lipophilic properties, DXNs can be highly accumulated in fish and other aquatic animals[2]. According to a national report of the Ministry of Health, Labor & Welfare[3], 52% of the total intake of DXNs in Japan is derived from food, 70.5% of which is sea food. In addition 15.5% and 8.5% are meat and egg, and milk and dairy products, respectively. It should be noticed that these portions are based on the calculation using the TEF scheme and TEQ methodology.

Apart from this issue, our studies demonstrated that the isomers performing a statistically significant relationship with food intake, varied depending on food frequency of each subject. Higher intake of green-yellow vegetables and fruits was significantly related to higher levels of 1,2,3,6,7,8-HxCDD and 2,3,4,7,8-PCDF, respectively. It is noted that both isomers existed at specifically high level in breast milk. The human TEFs for 1,2,3,6,7,8-HxCDD and 2,3,4,7,8-PCDF are given 0.1 and 0.5, respectively. As a result, concern to these isomers for direct application to human risk assessment is probably degraded. On the other hand, our study showed that significantly higher TCDD level was recognized in the subjects ingesting fish more than the ordinary. That is; higher fish intake is significantly related to higher level of TCDD featuring the most toxic potency. This phenomenon might be the reason to explain that higher human risk assessment is attributed to higher fish intake. In fact, however, the TCDD level in breast milk was not so high that it appeared remarkably in the isomer profiles, and TCDD level proportion expressed in percentage of ∑PCDDs/DFs-TEQ was within 10 %. In contrast, level proportions of 1,2,3,6,7,8-HxCDD and 2,3,4,7,8-PCDF in ∑PCDDs/DFs-TEQ were 12 % and 35 %, respectively, which occupied approximately 50% of the total TEQ, even if evaluation of TEF for these isomers is low. Therefore, in our view, it is necessary to concentrate our effort not only on estimating TEQ intake via ingestion, but also on understanding the relationship between food habit and isomer profiles for elucidation of exposure sources of DXNs, in order to facilitate total risk management against subtle adverse health effect.

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References

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Table 1 PCDDs/DFs levels in 44 breast milk samples from Ishikawa Prefecture, Japan

PCDDs	MEAN pg/g-lipid	S D	PCDFs	MEAN pg/g-lipid	S D
2,3,7,8-TCDD	0.35	(1.75)	2,3,7,8-TCDF	0.57	(1.61)
1,2,3,7,8-PCDD	1.62	(1.67)	1,2,3,7,8-PCDF	0.15	(1.75)
1,2,3,4,7,8-HxCDD	0.61	(1.67)	2,3,4,7,8-PCDF	3.34	(1.71)
1,2,3,6,7,8-HxCDD	5.80	(1.72)	1,2,3,4,7,8-HxCDF	1.01	(1.87)
1,2,3,7,8,9-HxCDD	0.91	(1.68)	1,2,3,6,7,8-HxCDF	0.95	(1.76)
1,2,3,4,6,7,8-HpCDD	2.89	(1.60)	2,3,4,6,7,8-HxCDF	0.60	(1.84)
OCDD	25.57	(1.64)	1,2,3,7,8,9-HxCDF	0.01	(2.55)
			1,2,3,4,6,7,8-HpCDF	0.60	(1.83)
			1,2,3,4,7,8,9-HpCDF	0.12	(1.78)
			OCDF	0.19	(2.12
∑PCDDs -TEQ	2.76	(1.66)	∑PCDFs -TEQ	2.04	(1.68)

MEAN = geometric mean. SD = geometric SD

Table 2 Correlation coefficients[1] between rich protein food intakes[2] during pregnancy and PCDDs/DFs levels in breast milk

PCDDs/DFs	Meat	Egg	Fish		Pulse	Milk
2,3,7,8-TCDD	0.240	-0.027	0.359	*	-0.005	0.088
1,2,3,7,8-PCDD	0.221	0.102	0.131		-0.024	0.233
1,2,3,4,7,8-HxCDD	0.240	0.039	0.174		0.028	0.147
1,2,3,6,7,8-HxCDD	0.222	0.186	-0.077		0.182	0.073
1,2,3,7,8,9-HxCDD	0.162	0.133	-0.028		0.161	0.081
1,2,3,4,6,7,8-HpCDD	0.086	0.224	0.224		0.065	0.120
OCDD	0.178	0.076	0.178		-0.192	0.081
2,3,7,8-TCDF	0.012	0.352	0.145		0.027	-0.005
1,2,3,7,8-PCDF	-0.184	0.105	0.326		-0.016	-0.007
2,3,4,7,8-PCDF	0.034	-0.024	0.232		-0.054	0.194
1,2,3,4,7,8-HxCDF	-0.011	0.047	0.161		0.086	0.252
1,2,3,6,7,8-HxCDF	0.038	-0.016	0.194		0.008	0.171
2,3,4,6,7,8-HxCDF	0.001	0.141	0.142		0.002	0.122
1,2,3,7,8,9-HxCDF	-0.037	-0.077	0.412	*	-0.174	0.218
1,2,3,4,6,7,8-HpCDF	-0.006	-0.054	0.408	*	-0.005	0.251
1,2,3,4,7,8,9-HpCDF	-0.044	0.106	0.079		-0.021	0.218
OCDF	-0.056	0.109	-0.045		-0.059	0.108

[1]: Correlation coefficients were tested by Spearman's rho. [2]: Food intakes express the estimated total amount of food every week. *: $P < 0.05$

Table 3 Comparison of PCDDs/DFs levels in breast milk by intake of green-yellow vegetables

PCDDsDFs (pg/g-lipid)	Normal intake[1] or below N = 30		Plenty intake N = 5	
	MEAN	SD	MEAN	SD
2,3,7,8-TCDD	0.35	(1.83)	0.43	(1.48)
1,2,3,7,8-PCDD	1.58	(1.73)	2.25	(1.36)
1,2,3,4,7,8-HxCDD	0.59	(1.75)	0.80	(1.27)

1,2,3,6,7,8-HxCDD	5.48	(1.73)	10.24	(1.36)	*
1,2,3,7,8,9-HxCDD	0.87	(1.74)	1.34	(1.18)	
1,2,3,4,6,7,8-HpCDD	2.87	(1.64)	3.48	(1.53)	
OCDD	24.34	(1.66)	39.01	(1.35)	
2,3,7,8-TCDF	0.57	(1.55)	0.97	(2.10)	
1,2,3,7,8-PCDF	0.15	(1.71)	0.17	(2.12)	
2,3,4,7,8-PCDF	3.24	(1.72)	4.91	(1.80)	
1,2,3,4,7,8-HxCDF	0.97	(1.86)	1.60	(2.13)	
1,2,3,6,7,8-HxCDF	0.95	(1.87)	1.01	(1.43)	
2,3,4,6,7,8-HxCDF	0.58	(1.73)	0.85	(2.82)	
1,2,3,7,8,9-HxCDF	0.09	(1.91)	0.22	(6.29)	
1,2,3,4,6,7,8-HpCDF	0.62	(1.92)	0.64	(1.54)	
1,2,3,4,7,8,9-HpCDF	0.12	(1.79)	0.12	(1.93)	
OCDF	0.18	(1.64)	0.34	(5.04)	
∑PCDDs -TEQ	2.68	(1.71)	3.99	(1.33)	
∑PCDFs -TEQ	1.98	(1.67)	3.02	(1.84)	
∑PCDDs/ DFs -TEQ	4.68	(1.69)	7.14	(1.50)	

[1]: 200g intake per day is to be used as a measure of normal intake.
MEAN = geometric mean. SD = geometric SD *: $P < 0.05$

Table 4 Comparison of PCDDs/DFs levels in breast milk by intake of fruits

PCDDsDFs (pg/g-lipid)	Intake of fruits less than once every day		Intake of fruits once or more every day		
	MEAN	SD	MEAN	SD	
2,3,7,8-TCDD	0.34	(1.93)	0.38	(1.63)	
1,2,3,7,8-PCDD	1.44	(1.85)	1.97	(1.43)	
1,2,3,4,7,8-HxCDD	0.52	(1.82)	0.76	(1.45)	*
1,2,3,6,7,8-HxCDD	5.41	(1.97)	6.76	(1.45)	
1,2,3,7,8,9-HxCDD	0.91	(1.92)	0.97	(1.47)	
1,2,3,4,6,7,8-HpCDD	2.76	(1.78)	3.18	(1.41)	
OCDD	26.29	(1.82)	25.79	(1.47)	
2,3,7,8-TCDF	0.55	(1.68)	0.63	(1.55)	
1,2,3,7,8-PCDF	0.12	(1.72)	0.19	(1.65)	*
2,3,4,7,8-PCDF	2.85	(1.83)	4.29	(1.52)	*
1,2,3,4,7,8-HxCDF	0.93	(2.10)	1.19	(1.67	
1,2,3,6,7,8-HxCDF	0.88	(2.09)	1.08	(1.41)	
2,3,4,6,7,8-HxCDF	0.50	(1.87)	0.79	(1.76)	*
1,2,3,7,8,9-HxCDF	0.08	(2.03)	0.13	(2.99)	
1,2,3,4,6,7,8-HpCDF	0.59	(2.03)	0.67	(1.67)	
1,2,3,4,7,8,9-HpCDF	0.13	(1.87)	0.11	(1.71)	
OCDF	0.19	(1.62)	0.21	(2.80)	
∑PCDDs -TEQ	2.52	(1.87)	3.27	(1.41)	
∑PCDFs -TEQ	1.77	(1.78)	2.58	(1.53)	*
∑PCDDs/ DFs -TEQ	4.31	(1.82)	5.89	(1.44)	

MEAN = geometric mean. SD = geometric SD *: $P < 0.05$