TRENDS OF DIOXINS IN GLOBAL BEEF, PORK, CHICKEN, RAW BOVINE MILK, AND DAIRY PRODUCTS FROM 2002 TO 2011

MeeKyung Kim*, Dong-Gyu Kim, Si-Won Choi, Sooyeon Kim, Young Hoon Bong, Seong-Wan Son

Animal, Plant and Fisheries Quarantine and Inspection Agency, 175 Anyangro, Anyang, Gyeonggido 430-757, Republic of Korea

Introduction

Concentrations of dioxins (polychlorinated dibenzo-*p*-dioxins, PCDDs and dibenzofurans, PCDFs) were measured in a variety of foods of animal origin using high resolution gas chromatography/mass spectrometry (HR-GC/MS). Locally-produced beef, pork, chicken and dairy products and imported beef, pork, and chicken were randomly selected for monitoring during the last decade. Overall concentrations of dioxins in foods are gradually decreasing around the world based on a recent report[1]. It seems that the source-reduction policy of dioxins in many countries have resulted in decreasing levels of dioxins through the food chain. Human exposure of dioxins occurs mainly from the intake of food of animal origin included fish. This study presents the average levels of dioxins in global foods of animal origin monitored from 2002 to 2011 in South Korea.

Materials and methods

The monitoring samples of meat were collected from slaughterhouses nationwide for the domestic products and randomly selected from the Korean Automated Import Information System (AIIS) at the port of entries for the import products. Samples of domestic and imported dairy products were purchased from several local markets. Samples of raw bovine milk were collected from several stations representing many farms nationwide. The samples were treated either by extracting fat in a conventional oven at 80℃ or using a Soxhlet extractor with hexane, depending on the fat content[2]. An isotope dilution method was used for the analysis of PCDD/Fs based on U.S EPA Method 1613B. Clean-up was performed with silica, alumina, and carbon columns using a Power-Prep™ (FMS Inc., U.S) automated column clean-up system. The extracts were analyzed by HR-GC/MS (Autospec Ultima, Micromass Co., U.K) equipped with a DB5MS capillary column (50 m x 0.25 mm I.D., 0.25 μm film thickness, J&W Scientific, U.S). Quality controls of the analyses were conducted and the proficiency tests were applied to FAPAS, UK on a regular basis. The laboratory was accredited (ISO/IEC 17025) for the dioxin analysis.

Results and discussion

Table 1 shows the levels of PCDD/Fs in food of animal origin imported from various countries for distribution in South Korea from 2002 to 2011. The average concentrations in samples were 0.158, 0.052, and 0.165 pg TEQ/g fat in 761 beef samples from 6 countries, 1067 pork samples from 17 countries, and 290 chicken samples from 10 countries, respectively. The concentrations were calculated using zero for non-detects. TEQ values calculated with the 1998 WHO-TEF. The average concentrations showed the differences between countries and between types of food. Studies have shown that the concentration of dioxins in beef were relatively higher than pork or chicken[3,4]. This result is expected considering that the life-time of cattle is longer than pig and chicken. However, the average concentration did not show those trends in this sample set. This may be because the environment on which animals are raised and their feedstuffs are different in each country. The level of dioxins in chicken was 5.7 times higher than that in beef from Canada. However, the level of dioxins in beef was 4.6 times and 1.6 times higher than that in chicken from South Korea and the U.S.A, respectively. The levels in pork were higher than in chicken from the samples of most countries except the similar levels in South Korea. Samples of Chilean pork were particularly targeted because of dioxins found (3.9 ~ 8.3 pg TEQ/g fat) over the Korean maximum residue limit (MRL: 2.0 pg TEQ/g fat) in 2003 and 2008 through the inspection service. The products related to this incident were not approved for import. Therefore, the concentrations of these unapproved samples were excluded from the calculation of the average for retailed pork in South Korea. A high concentration (6.26 pg TEQ/g fat) in beef over the MRL (4.0 pg TEQ/ g fat) from the U.S.A in 2006 was also excluded. Relatively

higher dioxins were found in chicken from China followed by Canada, but the number of samples was limited. The dairy products were analyzed included 60 cheese, 22 butter, 16 milk cream, and 12 powdered milk samples from 9 countries. The average concentrations were 0.202, 0.249, 0.258, and 0.510 pg TEQ/g fat in cheese, butter, milk cream, and powdered milk, respectively, and the total average concentration for the 110 dairy samples was 0.356 pg TEQ/g fat. The average concentration was 0.453 pg TEQ/g fat in 180 domestic raw milk samples.

Figure 1 presents the trends of PCDD/Fs in beef, pork and chicken from 2002 to 2011. The levels of dioxins in beef appear to be gradually decreasing although the years 2003, 2004 and 2007 had increasing levels. Samples from Mexico and South Korea contributed to the increasing trend. Pork showed a decreasing trend to less than 0.1 pg TEQ/g fat, except 0.113 pg TEQ/g fat in 2005. It was difficult to interpret the levels of dioxins in chicken. Samples of Brazilian chicken contributed relatively high levels of dioxins in 2006 and 2007. These levels ranged from 0.274 to 3.435 pg TEQ/g fat in 2006 and from nd to 3.210 pg /g fat in 2007. Data on PCDD/Fs in chickens were collected from only two countries in 2006 and 2007; therefore, the results largely reflect the highly contaminated Brazilian samples.

Figure 2 shows the trends and levels of PCDD/Fs in raw milk, powdered milk, milk cream, butter, and cheese. All the samples of raw milk were produced in South Korea. The average concentrations were 0.365, 0.650 and 0.350 pg TEQ/g fat for 40 raw milk samples in 2003, 60 raw milk samples in 2005, and 80 raw milk samples in 2008, respectively. Dairy products were from Australia, Belgium, France, Germany, Italy, South Korea, the Netherland, New Zealand, U.K and U.S.A. The average concentrations of PCDD/Fs were 0.510, 0.258, 0.249, and 0.202 pg TEQ/g fat in 12 samples of powdered milk, 16 samples of milk cream, 22 samples of butter, and 60 samples of cheeses, respectively, purchased in 2009. The levels of PCDD/Fs in raw milk and in powdered milk were similar. The levels of PCDD/Fs in both raw and powdered milk were relatively higher than levels in meat and the other dairy products. Although the absolute levels of PCDD/Fs in milk were low, there may be a concern because children are the primary consumers for milk.

Table 2 presents the estimated dietary human exposure levels based on milk consumption depending on age groups. The milk consumption amounts were taken from the dietary intake survey of infants, children and adolescents from the Korea Health Industry Development Institute, 2008. The estimated dietary intakes of PCDD/Fs were 2.35, 1.80, and 1.62 pg TEQ/person/day assuming a 20 kg body weight in 0-6 year olds, a 30 kg body weight in 7-12 year olds, and a 40 kg body weight in 13-19 year olds, respectively. To put this into perspective, the calculated dietary intake of PCDD/Fs in milk for a 20 kg child account for 11.7% of the lowest WHO-tolerable daily intake (TDI = 1 pg TEQ/ kg body weight) in this study.

In conclusion, the monitoring data indicated that the levels of dioxins in beef, pork, and chicken are gradually decreasing. There were no significant differences in dioxin levels in milk and other dairy products. Each country has different levels of contaminants in the environment and animal feeds resulting in varying levels of dioxins in food products. Milk may be a good indicator for the measurement of dioxin levels in food of animal origin. The level of dioxins in milk and other dairy products may take longer to reflect the decreasing trend of dioxins seen in meats. Estimated dietary exposure of Korean young ages to dioxins showed below the TDI.

References

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Table 1. Levels (pg TEQ/g fat) of PCDD/Fs in food of animal origin from various countries from 2002 to 2011

Country	Beef	Pork	Chicken	Raw bovine milk	Dairy products*
Australia	0.066 (287)	0.191 (9)			
Austria		0.035 (28)			
Belgium		0.044 (58)	0.240 (1)		
Brazil			0.536 (33)		
Canada	0.167 (7)	0.042 (56)	0.959 (2)		
Chile		0.059 (438)			
China			1.263 (1)		Cheese:
Denmark		0.039 (32)	0.091 (19)		0.202 (60)
France		0.015 (41)	0.040 (6)		
Finland		nd (3)			Butter:
Germany		nd (1)			0.249 (22)
Hungary		0.025 (28)			
Korea	0.304 (219)	0.067 (161)	0.066 (85)	0.453 (180)	Milk cream: 0.258 (16)
Mexico	0.370 (8)	0.016 (6)			
Poland		0.094 (22)			Powdered milk:
The Netherland		0.013 (25)			0.510 (12)
New Zealand	0.096 (125)				
Spain		0.008 (17)			
Sweden		nd (2)			
Thailand			0.229 (39)		
U.K			0.005 (2)		
U.S.A	0.159 (115)	0.041 (140)	0.10 (102)		
Average of total	**0.158 (761)**	**0.052 (1067)** **	**0.165 (290)**	**0.453 (180)**	**0.356 (110)**

In parentheses: Number of samples.
nd : not detected.
*Dairy products are from 7 countries.
**The average concentration was 0.047 pg TEQ/g fat when the intended sampling of Chilean pork was excluded.

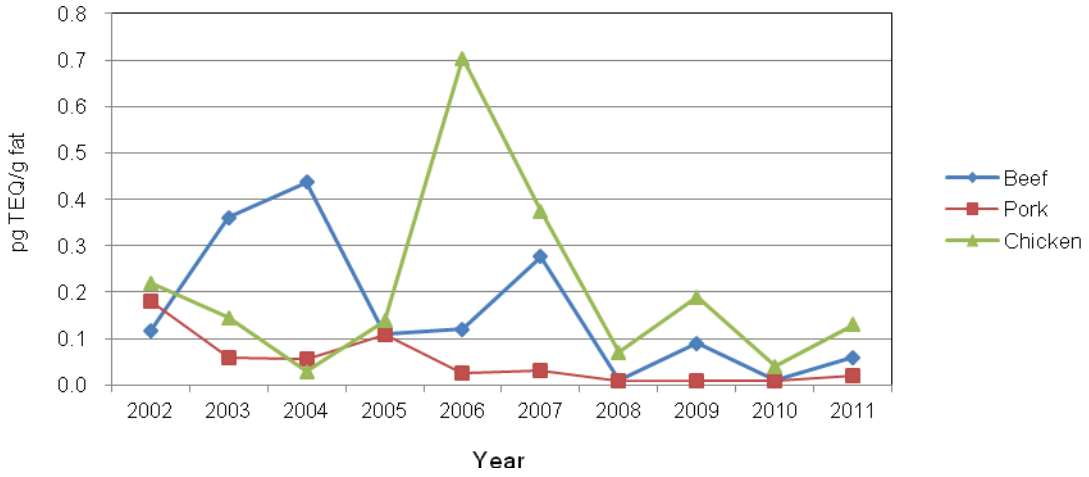

Figure 1. Trends of PCDD/Fs in beef, pork, and chicken in from 2002 to 2011.

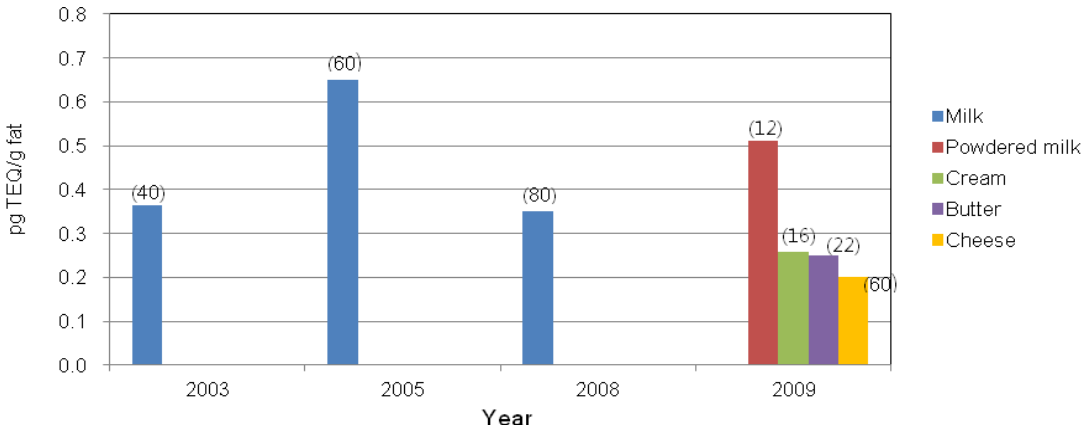

Figure 2. Trends and levels of PCDD/Fs in milk, powdered milk, cream, butter, and cheese from 2003 to 2009. Number of samples in parentheses.

Table 2. Estimated dietary human exposure levels based on milk consumption depending on age groups

Age range in years (body weight)	Consumption of milk (g/person/day)	PCDD/Fs		
		TEQ level in raw bovine milk (pg/g fat)	Exposure level	
			pg/person/day	pg/kg b.w/day
0~6* (20 kg)	148.02 (5.18)**	0.453	2.347	0.117
7~12 (30 kg)	113.44 (3.97)**		1.798	0.060
13~19 (40 kg)	102.12 (3.57)**		1.617	0.040

* Dietary intake survey of infants, children and adolescents (Korea Health Industry Development Institute, 2008).
** Fat content in parentheses.