

TEMPORAL TREND AND SEASONAL VARIATION OF PCDD/F LEVELS IN COW'S MILK IN TAIWAN

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Introduction

Increased health cautiousness among the people of Taiwan in recent years motivated them to adopt western style diet, resulting in increased consumption of fresh cows milk. Because of the common belief among the people that taking cold drinks or milk in winter is not good for health, the price of cow milk is generally lower in winter. Despite such health concern, overall consumption of milk and dairy product is high. To meet the demand in Taiwan most of the dairy products are imported, mainly from Australia and EU. Increased awareness among the people following the Belgium dioxin crisis in 1999 attracted attention of local media, and consequently the issue of dioxin in dairy products and relevant domestic products has become the topic of concern. Considering the heavy consumption of cows milk in Taiwan by infants and children and knowing that dairy products are one probable exposure route of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), we initiated the analysis of cows milk from the year 2000. Statistical data of PCDD/Fs levels in cow milk in the period from 2001 to 2003 is included as part of the national TDS study^{1,2}.

Materials and Methods

Sample collection and preparation: Cows milk of 13 popular brands were collected twice a year from supermarkets islandwide. A compound sample was prepared by mixing 4-8 bottles of milk. A total of 65 compound samples were analyzed during 2000-2005. An aliquot of 100 grams was pre-treated and analyzed, using the Chinese National Standard (CNS) method to test for residual dioxins in foods (CNS 14758)³.

Instrumental analysis: Samples were analyzed for the seventeen 2,3,7,8-substituted PCDD/Fs. The sample spiked with 15 ¹³C-labelled PCDD/Fs were analyzed by using an Agilent 6890N high resolution gas chromatograph coupled with a Micromass AutoSpec Ultima high resolution mass spectrometer (HRGC/HRMS). The analysis of PCDD/Fs congeners was performed with a DB-5 MS capillary column (60 m × 0.25 mm i.d. × 0.25 μm film thickness) with dynamic mass resolution greater than 10000. The concentrations of PCDD/Fs congeners were quantified with isotope dilution method. The LODs for tetra~penta-chlorinated PCDD/Fs, hexa~hepta-chlorinated PCDD/Fs, and octa-chlorinated PCDD/Fs were 0.1 pg, 0.2 pg, and 0.5 pg, respectively. The toxic equivalents (TEQ) were calculated based on WHO-TEF system. The concentrations of the non-detected congeners were calculated with their LODs (upper-bound concentration).

Results and discussion

Temporal trend: Table 1 shows the PCDD/Fs in cow milk samples expressed in pg WHO-TEQ/g lipid weight (lwt). The average PCDD/Fs level was 1.88 pg WHO-TEQ/g lwt in 2000 and showed steady decrease to 1.84, 1.55, and 1.34 pg WHO-TEQ/g lwt in the following three years. Surprisingly, it increased abruptly to 1.73 pg WHO-TEQ/g lwt in 2004, but a sudden drop to 1.13 pg WHO-TEQ/g lwt was noted in 2005. The measured values were well below the 5 pg WHO-TEQ/g lwt tentative maximum limit, which was set specifically to manage the suspected food items during the Belgium dioxin crisis in 1999. An apparent decline of PCDD/Fs levels (exclude the 2004 data) was observed in the temporal trends as shown in Fig. 1. A similar declining trend of PCDD/Fs levels in cow milk was noted by Schmid et al.⁴, who reported that the PCDD/Fs levels (0.51±0.19 ng I-TEQ/kg milk fat) in consumer milk in Switzerland in 2001 were significantly lower than those levels in 1990/1991 (1.3±0.23 ng I-TEQ/kg milk fat) and 1984 (1.9 ng I-TEQ/kg milk fat). Schaum et al.⁵ also reported the national average CDD/CDF levels in 2000-2001 in the US milk supply was half as that in 1996. Similar studies by Baumann et al.⁶ reporting the dioxins and non-ortho PCBs in Dutch consumer milk in the period 1997-2002 indicated a significant decline from about 5 in 1997 to about 0.8 WHO-TEQ (total) pg/g fat in 2002.

Seasonal variation: The national average of PCDD/Fs level in cow milk in Taiwan (for example 1.55 in 2002) was significantly higher than those reported in other countries, for example, 0.51±0.19 in Switzerland⁴, 0.8 in Dutch⁶, and 0.71 in U.S.⁷. The possibility of seasonal and regional differences was first investigated of the milk that the milk processing factories usually collect from distinctive regions. The PCDD/Fs levels in milk samples collected from all regions appeared to be similar. This was ascribed to the feed for the cows, which was from similar sources, irrespective of the regions. We have noticed the possible seasonal variation of PCDD/Fs levels in cow milk in Taiwan collected in 2001¹. Similar results were obtained during the following 2 years, but to a less extent, as shown in Fig. 2. Preliminary results indicate that the cause might be the difference in PCDD/Fs levels in feed used in summer and winter. The decreasing PCDD/Fs levels were observed in imported Bermuda grass (*Cynodon doctylon*) and alfalfa (*Medicago sativa*), domestic Napier grass (*Pennisetum americanum*), and domestic corn silage. The unit price of imported feed is more expensive than domestic corn silage and Napier grass. The feed in summer mainly consisted of a mixture of both imported and domestic grass, while the feed in winter was mostly corn silage. To confirm this hypothesis, a collection of more detailed and comprehensive data is under process. Nevertheless, the importance of regulating PCDD/Fs levels in feed is demonstrated.

Acknowledgements

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Table 1: The PCDD/Fs levels (upperbound) in cow's milk in Taiwan from 2000 to 2005

Year	Number of Samples	PCDD/Fs (pg WHO-TEQ/g lw)			
		Min.	Max.	Mean	S.D.
2000	9	1.40	2.40	1.88	0.39
2001	14	0.608	3.18	1.84	0.76
2002	10	0.902	2.64	1.55	0.51
2003	16	0.193	3.28	1.34	0.84
2004	10	0.286	3.68	1.73	0.96
2005	9	0.269	1.79	1.13	0.45

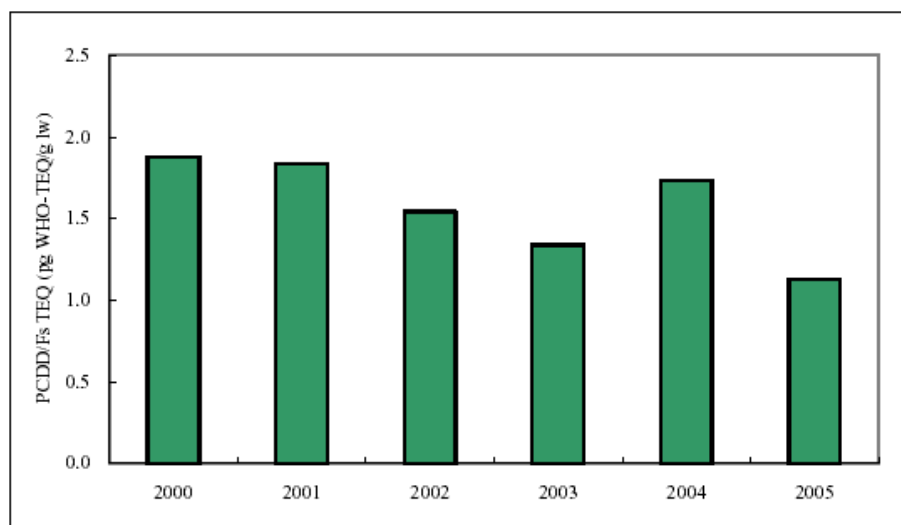


Figure 1: The temporal trend of PCDD/Fs levels in cow's milk in Taiwan from year 2000 to 2005.

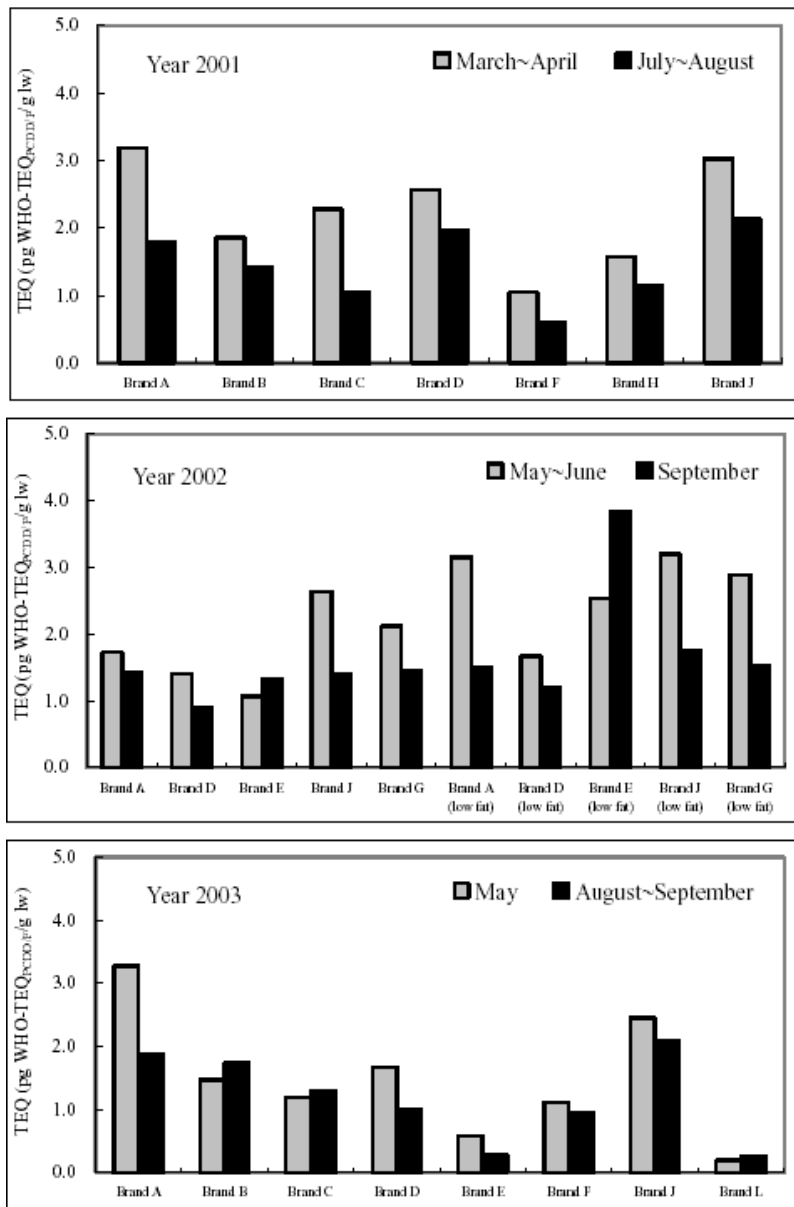


Figure2: The seasonal variation of PCDD/Fs levels in cow's milk in Taiwan.