Estimated intake of PCDD/Fs from infant formulas marketed in Spain

Susana Lorán[1], Susana Bayarri[1], Pilar Conchello[1], Antonio Herrera[1]

[1]Veterinary Faculty, University of Zaragoza

Introduction

Dietary intake is the major route of PCDD/Fs exposure, contributing more than 90% of the daily exposure. Available data indicate a reduction in exposures and body burden in the past decades[(1,2,3,4)] due to measures introduced to control the emissions of PCDD/Fs. A considerable amount data exists for concentrations of these compounds in human milkindicating that children ingest higher PCDD/F doses than adults due to their higher food consumption in relation to the body weight[(3,4,5,6,7)]. However, information about substituted breast milk at this concern is scarce.

In 1998, World Health Organization (WHO) recommended a tolerable daily intake (TDI) of PCDD/F in a range of 1 to 4 pg WHO-TEQ/kg b.w./day[8]. In later revisions, a tolerable weekly intake of 14 pg WHO-TEQ/kg b.w./week[9] and a tolerable monthly intake of 70 pg WHO-TEQ/kg b.w./month[10] have been stablished. For practical reasons these maximum intakes can be handled as if it were a tolerable daily intake of 2 pg TEQ per kg bw per day.

Congener-specific analyses of PCDDs and PCDFs were performed on a number of milk based infant formulas collected in Spain in 2004. The main objective of this study was to estimate the dietary intake of dioxins by children up to one year of age through the comsumption of infant formulas.

Methods and Materials

Samples of infant formulas were acquired from local markets, big supermarkets and chemists. The 10 different trades studied were selected among the best well-known in Spain. Totally, 70 samples (5 different batches from the same manufacturer) divided in three groups were collected, 25 infant formula, 25 follow-on formula and 20 special lactose-free formulas.

A composite (1 kg) was made by mixing proportional pooled samples of 5 different batches from the same commercial trade. Lastly, 14 analytical samples were made up, 5 of infant formula, 5 of follow-on formula and 4 of special lactose-free formulas.

About 20 grams test portions of infant formula were mixed with sodium sulphate anhydrous and extracted with acetone-n-hexane (1:1) in an automated Soxhlet extractor. Prior extraction, samples were spiked with 13C-PCDD/Fs labeled internal standards. Clean-up was carried out in a sequence as follows: decomposition with concentrated sulfuric acid, followed by multilayer column and alumina column chromatography. All solvents employed were of Pestiscan grade (Lab-Scan, Dublin, Ireland) and hexane was also glass distilled.

The eluting fraction containing the compounds of interest were analyzed by high resolution gas chromatography (HRGC) coupled to Ion Trap MS/MS (Varian, Saturn 2000 MS/MS). Chromatographic separation was achieved with a Varian Factor Four fused-silica capillary column (60m x 0.25 mm ID, 0.25 µm film thickness) with helium as the carrier gas at a linear velocity of 1ml/min in the splitless injection mode (2µl). The quantification was carried out by the isotopic dilution method. The methodology was validated according to EPA Method 1613[11].

Results and Discussion

Average results of PCDD/PCDFs in pooled samples of infant formula, follow-on formula and lactose-free formula analyzed, are summarized in Table 1.The toxic equivalents (TEQ) were calculated using WHO-TEF (1998) system[2]. The use of Toxic Equivalents allows an assessment of the toxicological significance of the complex mixtures of dioxin. For this analysis, data were derived assuming that levels for non-detected congeners were half the limit of determination (LOD).

The concentrations of dioxins in all of the baby food sampled were generally well within the EU limit for milk and milk products (3 pg WHO-TEQ/g fat). It's important to take into account that mixtures of vegetable oils are added to these skimmed-milk based products in order to meet the nutrient requirements for children at this age. The EU limit applicable to vegetable oils is 0.75 pg WHO-TEQ/g fat.

Table1. Average results of PCDDs/PCDFs (pg-WHO-TEQ/ g fat) in infant formulas.

Baby Food	Mean ±SD (pg WHO-TEQ/g fat)		
	PCDD	PCDF	Σ PCDD/PCDF
Initial formula n = 5	0.15 ± 0.09	0.08 ± 0.03	0.24 ± 0.11
Follow-on formula n = 5	0.38 ± 0.41	0.39 ± 0.48	0.77 ± 0.89
Lactosa-free formula n = 4	0.36 ± 0.18	0.25 ± 0.15	0.61 ± 0.33

PCDD/Fs levels found in infant formulas were similar to those reported in different countries as shown in Table 2. However, the information published concerning the levels of PCDD/PCDFs in this kind of dairy-like products is scarce. These results are also compared with those found in breast milk and dairy products, not considering dioxin-like PCBs, with higher levels of this organohalogen compounds than infant formulas have.

Table 2. Levels of Σ PCDD/PCDFs reported for several countries for breast milk, dairy products and related food.

Country	Food Analyzed	PCDD/PCDF (pg WHO-TEQ/g fat)	Reference
EU	Breast Milk	8-16*	EU Task 3.2.5[1]
Belgium	Breast Milk	16-52.1	Focant et al[6]., 2002
Portugal	Breast Milk	4.8-19.9	Reis et al[7]., 2002
Spain	Milk and dairy products	0.80	Llobet et al[2]., 2003
Spain	Powdered full fat milk	1.87**	Ramos et al[12]., 1999
United Kingdom	Infant formulas	0.2-0.4	F.S.A. U.K.[4]
Brazil	Infant Formulas	0.22-0.45***	Päpke and Tritscher[13], 2000
Australia	Infant Formulas	0.0036-0.018****	Australian Government[14].

* National averages in EU for the period 1995-1999. Toal pg I-TEQ/g fat.

** Total I-TEQ pg/g fat basis.

*** Range found in n = 4 samples analyzed. Total I-TEQ pg/g fat basis.

**** Mean range of dioxin concentratios in food, in pgTEQ/g fresh weight.

Assessment of dietary exposure is vital to obtain fundamental data concerning the safety of foods, problems and trends in the intake of chemicals and to identify the sources of unusual residues. The average daily intake of PCDD/PCDFs was estimated based on the assumption of different body weight and daily milk consumption in children at birth, 6 months and 12 months of age as recomended for nursing infants[15].

In the first month of life the estimated dietary intake of PCDD/Fs is about 1,8 pg-TEQ/kg bw/day. This level keeps constant up to 6 months of age where an estimated calculated intake of 1,7 pg-TEQ/kg bw/day has been measured. Since 6 months of age, a lower dietary intake of PCDD/Fs through the consumption of infant formulas proportionally to the highest body weight is expected. However, it doesn't occurs as it has been calculated in 1,45 pg-TEQ/kg

bw/day. This is due to the mayor contamination of follow-on formulae than in those intended for infants from birth.

In the special case of lactosa-free formulae the estimated daily intake of PCDD/Fs ranged from 0,98 pg-TEQ/kg bw/day at birth to 1,55 pg-TEQ/kg bw/day at 12 months of age.

There have been few estimates of dietary intakes of dioxins and PCBs by formula-fed infants in other countries and there is a wide variation in these studies[15]. The presented results suggest that nursing infants doesn't exceed the TDI of 2pg/kg bw/day, on the basis of PCDD/F exposure alone, through the consumption of infant formulas. Besides, these estimations consist with those estimated in the UK[15] for children up to 6 months, milk substituted fed, that were also below these recommendations.It's important to take into account that the estimated intake of toxic equivalentes (TEQ) through infant formulas in the first year of life could be increased with the introduction of dioxin-like PCBs and other food that an infant receives from the age of four months.

Besides, the average values calculated for infant formulas suggest a lower intake of PCDD/Fs by newborns from commercials breast milk substitutes than those calculated for human feeding, exceeding the TDI derived by WHO during this short period of life. Intakes of PCDD/Fs in breast feeding-children have been measured in the EU[1] (1998): 66.1 pg PCDD/F-WHO-TEQ/kg body weight; Germany[3] (1998): 24-145 pg I-TEQ/kg bw/day; Portugal[7] (2002): 51 pg/kg bw/day; Belgium[6] (2002): 103 pg TEQ/kg bw/day. All results show higher intakes of PCDD/Fs in breastfed children than in infant formula fed. Furthermore, cumulative dose of exclusively breastfed infants in Taiwan were found significantly greater compared to that of formula-fed infants[16].

According to the results of the current study, estimated intakes of PCDD/Fs through the consumption of infant formulas represent a 50% of the TDI (2 pg-TEQ/kg bw/day) for children fed with special lactosa-free formulas during the first month of life up to 90% of the TDI for children fed with initial formula at the same age.

Acknowledgment

This work has been supported by the Spanish Ministry of Science and Technology (Project AGL 2001-2533) and Diputación General de Aragón (Grupo Investigación Consolidado. Project AO1)

References.

1. European Comission (2000), Assessment of dietary intake of dioxins and related PCBs by the population of EU Member States. Report of expert participating in Task 3.2.5

2. Llobet, J.M., Domingo, J.L., Bocio, A., Casas C., Teixidó, Müller, L. (2003) Human exposure to dioxins through the diet in Catalonia, Spain: carcinogenic and non-carcinogenic risk. Chemosphere 50, 1193-1200.

3. Päpke O. (1998). PCDD/PCDF: Human Background Data for Germany, a 10-Year Experience. Environmental Health Perspectives Supplements 106, S2.

4. Food Standards Agency UK (2004). Dioxin and dioxin-like PCBs in infant formulae. Food Survey Information Sheet 49/04.

5. Schrey, P., Wittsiepe, J., Selenca, F., 1996. Dietary intake of PCDD/F by small children measured by the duplicate method. Organohalogen Comp. 9, 261-268

6. Focant, J.-F., Pirard, C., Thielen, C., and De Pauw, E. (2002) Levels and profiles of PCDDs, PCDFs and cPCBs in Belgian breast milk. Estimation of infant intake. Chemosphere 48, 763-770.

7. Reis, F., Pereira Miguel, J.M., Pisarra, M.I., Sampaio, C., Calheiros, J. (2002) Infant exposure to PCDD/Fs in Portugal: First results from an environmental health survey program near Lisbon. Organohalogen Compounds Vol. 55. 247-250

8. WHO 1998: Assessment of the health risk of dioxins: re-evaluation of the Tolerable Daily Intake (TDI)

9. SFC 2001. European Commision, Scientific Commitee of Food, Brussels, Belgium. Opinion of the SFC on the risk assessment of dioxins and dioxin-like PCBs in food. (22 November 2000); Opinion on the risk assessment of dioxins and dioxins-like PCBs in food (update based on the new scientific information available since the adoption of the SFC opinion on 22 November 2000, 30 May 2001.

10. JECFA Summary of fifty-seventh meeting of the Joint FAO/WHO Expert Committee on Food Additives, Rome, 5-14 June 2001. 24-40, 2001.

11. EPA: U.S. Environmental Protection Agency (1994). Method 1613 Tetra through Octa-chlorinated dioxins and furans by isotope dilution HRGC/HRMS.

12. Ramos, L., Eljarrat, E., Hernández, L.M., Rivera, J., González, M.J. (1999) Comparative study of methodologies for the análisis of PCDDs and PCDFs in powdered full-fat milk, PCB, PCDD and PCDF levels in commercial samples from Spain. Chemosphere 38, 2577-2589.

13. Päpke O., Tritscher, A. (2000) Organohalogen Compounds 47, 389-392.

14. Australian Government. Department of the Environment and Heritage (2004). National dioxins program. Dioxins in aurstralia: a summary of the findings of studies conducted from 2001 to 2004.

15. Cervera, P., Clapés, J., Rigolfas, J. (1999). Alimentación y Dietoterapia. Edit. McCgraw-Hill Interamericana. 3th edition.

16. Chao, H-R., Wang, S.-L., Lee, C.-C, Yu, H.-Y., Lu, Y.-K., Päpke, O. (2004) Level of polychlorinated dibenzo –*p*-dioxins, dibenzofurans and biphenyls (PCDD/Fs, PCBs) in human milk and the input to infant body burden. Food and Chemical Toxicology 42, 1299-1308.