PROPOSED LIMIT VALUES FOR DIOXIN DEPOSITION IN FLANDERS

Raf De Fré, Christa Cornelis, Clemens Mensink, Johan Nouwen, Greet Schoeters[1] and Edward Roekens[2]

[1] Vito, Flemish Institute for Technological Research, Boeretang 200, B-2400 Mol, Belgium
[2] VMM, Vlaamse Milieumaatschappij, Kronenburgstraat 45 bus 3, B-2000 Antwerpen

Introduction

Emission into the atmosphere followed by deposition on plants and animal uptake is the main pathway of human exposure to dioxins. Dioxin deposition is monitored in Flanders since 1993 by Vito on behalf of the VMM. At present every year two measurement campaigns with 70 to 80 sampling points are organised. Advantages of deposition monitoring are the integration of samples over 1 month and the possibility for application on a large scale with low cost equipment. Reference values for measured data were initially taken from Prinz et al.[1] who proposed guidelines for air concentration and deposition in North-Rhine Westfalia, Germany. In 1998 VMM and Vito collaborated in this study to formulate adapted limit and guide values for dioxins in air and deposition in Flanders.

Emission and immission levels

The emissions of dioxins in Flanders are well known for municipal waste incineration, which is assumed to account for more than 50% up to 1990. For other industrial sectors, except for sinter plants, data are less abundant and therefore less accurate. Air concentrations and deposition of dioxins from waste incinerators were calculated with the OPS model for the Flemish territory as annual average values in 5 x 5 km squares for the years 1985, 1990 and 1996 (table 1 and 2). The results illustrate the decreasing contribution from waste incinerators to environmental levels. Experimentally determined concentrations in air ranged from 100 to 255 fg TEQ/m^3 in 1992 as averages of 3 to 4 samples at 6 different sites, and are considerably higher than model results, indicating the presence of important unknown sources, if not poor model performance. Deposition measurements in Flanders since 1993 show that there is a gradual decrease in levels and in the number of places with high values. In 1994 the majority of samples were higher than 10 ng TEQ/m^3/y, with a maximum of 374, while in 1997 less than 30 % of the samples were higher than 10 ng TEQ/m^3/y. From 1996 onwards dioxin emissions from municipal waste combustion were virtually eliminated, but deposition levels have not declined to the same extent as the emissions. Fewer data are available on the evolution of air concentrations. In 1999 on a rural site in Mol still values from 50 to 150 fg TEQ/m^3 were measured. This persistence of atmospheric dioxin levels illustrates the need to monitor and to set quality objectives in order to reduce human exposure.

Table 1: Calculated dioxin air concentrations from waste incineration in Flanders

fg TEQ/m	1985	1990	1996
Territorial average	7.1	7.5	0.82
Maximum	40	52	5.8
Minimum	1.1	1.2	0.15

Table 2: Calculated deposition of dioxins from waste incineration in Flanders

	Cumulative 1985-1996	1996	
	ng TEQ/m3	pg TEQ/m3/d	ng TEQ/m3/y
Territorial average	21	0.43	0.16
Maximum	149	3.4	1.24
Minimum	3.3	0.15	0.05

Criteria for guidelines

Human exposure to dioxins via dietary intake in Belgium has been among the highest in the world, as shown by the 1988 mothers milk analysis by WHO/EURO, where the Belgian pooled sample contained 39.5 pg TEQ/g milk fat. Levels of 34.4 pg TEQ/g fat in 1993 in Flanders, and 25.5 pg TEQ/g fat for the Brussels and Walloon region indicate a decreasing trend (Liem et al.[3]). The actual total uptake of dioxins in Flanders for 1997 was estimated to be around 1 pg TEQ/kg for adults and 2.4 pg TEQ/kg/d for children. Data on human toxicity and ecological effects of dioxins were reviewed to derive the guidelines of air concentration and deposition. Relying mostly on data compiled by the U.S. EPA and the Dutch Health Council the following criteria were selected as a basis for environmental quality levels:

- a daily intake of 3 pg TEQ/kg bodyweight to define the limit value in air and deposition,
- 1 pg TEQ/kg/d to define the guide values,
- 0.01 pg TEQ/kg/day as a long term objective to minimize cancer risk and infant exposure.

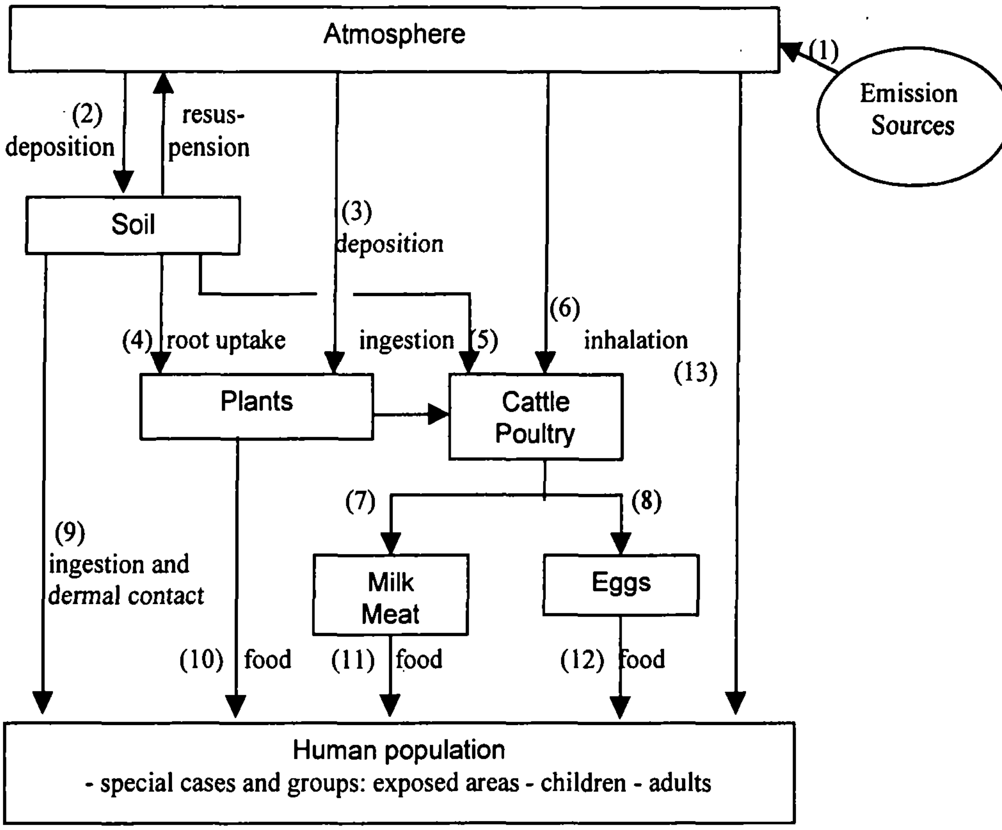
The selection of the first two criteria was supported by the estimates of actual daily intake and decided shortly before WHO lowered the acceptable daily intake value from 10 to 4 - 1 pg TEQ/kg/day in 1998. From analysis of the exposure pathways it is clear that the last objective cannot be met by setting environmental standards alone.

Transfer models

Figure 1 represents the fluxes of dioxins that are relevant for human exposure, starting from emissions into the atmosphere. The transfers represented by arrows are calculated by mathematical models, or a transfer coefficient in its simplest form. For most matrices in figure 1, results of dioxin analysis are available, allowing to estimate the model coefficients. The main route of uptake through cow's milk and meat is well documented, as analytical data from the Belgian federal ministry of agriculture are available since 1991. Dioxin concentrations for most other diet ingredients were taken from the neighbouring countries the Netherlands (Liem[3]) and Germany (Fürst[4]). With an average concentration in 1997 of 1.4 pg TEQ/g fat in cow's milk (2.3 in 1996), the intake of dioxins in Flanders for 1997 was estimated at 1 pg TEQ/kg/d for adults and 2.4 pg TEQ/kg/d for children, and declining. Dioxin intake by people living near sources such as a waste incinerator with uncontrolled emissions depends on the fraction of locally produced food and may be e.g. more than 6 times higher in a scenario where 100 % of the milk, 50 % of the meat and 25% of the vegetables consumed are taken from local production.

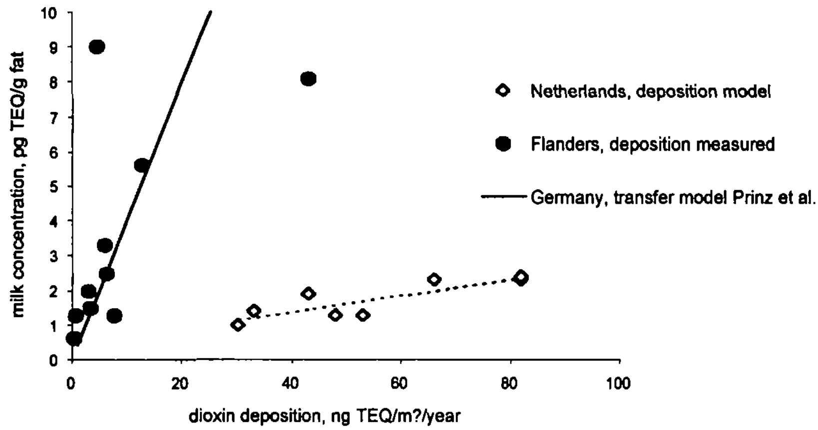
For the definition of air concentration and deposition quality standards, initially we used the chain model by Slob et al.[5] to describe the transfers from emissions to cow's milk. The chain model consists of the OPS atmospheric dispersion model, the prairy model to calculate concentrations in grass and soil, and the cow model to calculate milk concentration from uptake of grass and soil. Unknown parameters in the chain model were calibrated in the Netherlands using milk concentration data near MSW incinerators, resulting in a predictive error of 21%. To quantify the uptake via root and leaf vegetables, meat, ingestion of soil particles, inhalation, dermal contact and drinking water we used the VLIER-HUMAAN model.

Figure 1: Pathways of dioxins from atmosphere to human intake



However when we compared experimental deposition data and milk analysis, the results from the chain model did not fit (figure 2). Reasons for this may be that either the model is biased to overestimate deposition, or that the deposition measurement by the Bergerhoff method is not representative for steps (2) and (3) in figure 1. A further complication is that the model does not take into account the relatively high fraction of background deposition caused by unknown sources. For the purpose of our work it was essential to have a calibrated relation between measured deposition and milk contamination. As the chain model was not validated for the intermediate deposition results, transfer coefficients from Prinz et al.[1] were used, since this model takes into account experimentally determined deposition data by the Bergerhoff method and shows good agreement with experimental data from Flanders. At this point it was clear that due to the lack of experimental data, method bias and uncertainty in the relation with deposition, there was not a sufficient basis to formulate guidelines for air concentration of dioxins.

Figure 2: Relation between dioxin deposition and levels in milk fat



By means of the selected transfer models daily intake values were translated into quality objectives for dioxin deposition as given in table 3. Corresponding concentrations in cow's milk are given as a reference only, since there is a legal limit of 5 pg TEQ/g fat in force for milk products since 1998.

Table 3: Proposed limit and guide values for dioxin deposition in Flanders

	Daily intake	Milk concentration	Dioxin deposition	
	pg TEQ/kg/d	pg TEQ/g fat	ng TEQ/m^3/y	pg TEQ/m^3/d
Limit value	3	4.1	3.8	10
Guide value	1	1.4	1.3	3.4

To account for seasonality of the measurements the limit and guide values for monthly averages were taken a factor two higher.

References

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