Human Exposure III

Dioxins and PCBs in The Netherlands

Carin E.J. Cuijpers^a, Harry J. Bremmer^a, Niels B. Lucas Luijckx^b, Job A. van Zorge^c and A.K. Djien Liem^a ^a National Institute of Public Health and the Environment (RIVM), P.O. Box 1, NL-3720 BA Bilthoven, The Netherlands ^b Public Health Department, Ministry of Health, Welfare and Sport, P.O.Box 5406, 2280 HK Rijswijk, The Netherlands ^c Directorate-General for Environmental Protection, Ministry of Housing, Spatial Planning

and the Environment, P.O. Box 30945, 2500 GX The Hague, The Netherlands

Introduction

The general situation with respect to dioxin related compounds in The Netherlands has been extensively investigated in the beginning of the 1990s (1). The major categories of dioxin sources could be identified and their emissions quantified. Municipal waste incineration was found to contribute for about 80% of all dioxin emissions into air (Fig. 1). Field studies revealed a distribution and concentration of dioxins in ambient air and soils similar to that found in other industrialised nations, and a median dietary exposure for the general population of 1 pg (i)-TEQ per kg body weight per day (pg/kg.day) resulted from a comprehensive study on levels of dioxin related compounds in foodstuffs. In 1991 the Dutch government adopted the TDI of 10 pg 2,3,7,8-TCDD/kg.day, as proposed by WHO in 1990 (2) and decided to interpret this value as 10 pg (i)-TEQ/kg.day (3). This policy was (in 1993) supported by the results of the Integrated Criteria Document of the National Institute of Public health and the Environment (RIVM) (1).

In 1996 the Health Council of the Netherlands advised the Dutch government to lower the Tolerable Daily Intake (TDI) from 10 to 1 pg (i)-TEQ/kg.day (4). Ever since in 1989, the detection of dioxins in the milk of cows grazing near the most important municipal solid waste incinerator in the country, at levels that raised concern for public health, the dioxin issue has been on and of a subject of political interest and debate (3). With the advise of the Health Council the debate again arose and therefore, in 1997 the Dutch government asked the RIVM to provide them with the most recent information on dioxins, in order to design a plan. This plan should illustrate the effect of measures which have been taken to reduce human exposure and identify additional measures to further reduce exposure. In the present paper this new information will be summarised.

Sources and emissions

An update of estimates of emissions of dioxin-like compounds into air is presented in table 1. The estimates for 1991 are based on results from a national measurement campaign on emissions of PCDDs, PCDFs and dioxin-like PCBs as reported by Bremmer et al (5,6). Recent information as published by RIVM (7) combined with information obtained from local authorities and representative companies, has been used to provide the estimates for 1996 and 1998. Besides, the recent literature was reviewed. It should be noted that all estimates have been based on a limited number of emission measurements for each process category. The

majority of measurements originate from the period 1989-1992. The situation for 1996 and 1998 is based on the best available information and should be considered with more caution. It is however obvious that the emissions of dioxin related compounds decreased substantially between 1991 and 1996. This decline is even more pronounced if an older estimate for 1989 is taken into account. As a result of improvements in waste incineration technology, the dioxin emission could be reduced from about one kg (i)-TEQ in 1989, down to 50-80 grams of (i)-TEQ in 1996-1998. Emissions caused by the former use of wood preservatives (pentachlorophenol) and those resulting from the combustion of wood have (relatively spoken) become the major contributors to the total amount emitted into air (table 1).

Table 1

Estimates of emissions of dioxins (PCDD/F) and dioxin-like PCBs into air in the Netherlands. The estimates are presented in g TEQ/year, using the International TEFs according to NATO/CCMS (8) and the WHO-TEFs for PCBs (9).

Process category	1991		1996	1998		
	PCDD/F PCDD/F+PCE	PCDDF+PCB	PCDD/F	PCDD/F+PCB	PCDD/F	
Municipal waste incineration	382	401	3.6	3.8	1.6	1.7
Hazardous waste incineration	16	16.8	0.5	0.5	0.5	0.5
Incin. of landfill, biogas, sludge	0.3	0.4	0.3	0.4	0.3	0.4
Burning of cables & electromotors	1.5	2.0	0.3	0.4	0.3	0.4
Hospital waste incineration	2.1	2.7	1.5	2.0	1.5	2.0
Asphalt-mixing installations	0.3	0.4	0.3	0.4	0.3	0.4
Oil combustion	1	1.3	1	1.3	1	1.3
Combustion of coal and lignite	3.7	4.8	4	5.2	4	5.2
Wood combustion	12	15.6	14	18.2	14	18.2
Crematoria	0.2	0.3	0.2	0.3	0.2	0.3
Fires	4	5.2	4	5.2	4	5.2
Various high temperature processes	2.7	4.7	1.8	3.1	1.8	3.1
Traffic	7	9.1	1.2	1.6	0.8	1.0
Sintering processes	26	32.2	18	22.3	2.5	3.1
Metal industry	4	5.2	4	5.2	3.3	4.3
Chemical production processes	0.5	0.7	0.5	0.7	0.5	0.7
Pesticides	0.0	5 0.06	0.04	4 0.04	0.04	0.04
Former use of wood preservatives	25	25	22	22	21	21
Total	488	527	77	93	57	69

Levels in the environment

Information on levels in the Dutch environment has been compiled in the Integrated Criteria Document (1). Since the publication of this document, some additional information has become available. Based on measurements in ambient air covering the period 1991-1993, longterm background concentrations between 20 and 60 fg (i)-TEQ/m³ have been estimated for four different locations (10). The concentrations of dioxin-like PCBs in an urban area appeared to be in the order of 4 fg TEQ/m³ (11, TEFs according to 9). In samples of top soils from 32 background areas collected in August 1991, dioxin levels were found to range

60

between 1.8 and 16.4 ng (i)-TEQ/kg dry soil (12). Previous studies on sediment cores and suspended matter emphasised the importance of riverine fluxes entering the Netherlands through the rivers Rhine, Meuse and Scheldt. From the sediment core studies it appeared that the maximum input of dioxin and PCBs occurred in the period 1950-1980. Between 1975 and 1994 TEQ levels in Rhine related sediments have dropped by 70-80% (13). However, no significant decline in levels of suspended matter in the rivers Rhine and Scheldt could be found between 1985 and 1993, while levels in the river Meuse decreased by twenty percent (14).

Levels in foodstuffs

Since the publication of the results from the comprehensive study on foodstuffs, additional information has been obtained from surveys of consumer milk, fish oils and various primary agricultural products (beef, pig, poultry, mutton, milk, eggs, various fish) sampled in the period 1992-1996. Results have been reported extensively elsewhere (15,16). The study on levels in primary agricultural products indicates a significant decline in the levels of dioxins and PCBs in important animal foods, except for eggs, in the period 1990-1996 (see table 2).

Table 2

Mean dioxin level (in pg (i)-TEQ/g fat) in various primary agricultural products in 1990-1991 and 1996.

Product	1990-1991 (17)	1996 (16)
Beef	1.75	1.25
Pork	0.43	0.25
Chicken	1.65	0.66
Mutton	1.85	0.95
Milk	1.49	0.38
Egg	2.00	2.03

Human exposure

At the DIOXIN''97 symposium in Indianapolis, the results were presented from a study on trends in the dietary exposure to dioxins and PCBs in the Netherlands. In this study the duplicate diet approach was used and results indicated a substantial decline in the average dietary intake of PCDDs, PCDFs and PCBs for the Dutch adult population in the period between 1978 and 1994 (18). In a more recent study (16), available information on levels in foodstuffs (1990-1996) and food consumption patterns (obtained from the Dutch National Food Consumption Surveys; 1987/88 and 1992) has been used to evaluate time trends in the dietary intake of dioxins and non-ortho PCBs. Comparing the intake levels of dioxins and PCBs between 1987/88 and 1992 (food consumption data) using the levels in foodstuffs of 1990/91 showed a decline of 15%, which is ascribed to the reduced fat consumption of the Dutch population in this time period. A further decrease of 40% was observed if the food consumption data of 1992 were combined with the levels in foodstuffs from 1990/91 and from 1994-1996. The median Dutch intake range over this time period was calculated to be 1.2-2.4 pg (total) TEQ/kg.day (16).

Levels in human tissues

In the Netherlands human tissue levels are examined by monitoring polychlorinated compounds, at five years intervals, in human milk. As reported in 1995 (19), there is a clear downward trend of Organochlorine pesticides (OCPs) in the period between 1972-1993. In 1993 only small amounts of HCB, β -HCH and p,p'-DDE still were detectable. On the contrary, between 1983-1993, no significant changes were observed for the most dominating PCB congeners with IUPAC nos. 118,138,153 and 180. Levels of PCDDs and PCDFs have been measured in 1988 and in 1993 in ten pooled respectively 103 individual samples. If the (i)-TEQ levels of the individual congeners in 1993 are expressed as percentage of their mean in 1988, the decrease varies from 0 to 90%, with an average decrease of approximately 30% (average body burden in 1993: 23.5 ± 8.9 (i)-TEQ/g fat).

In the autumn of 1998, in the Netherlands, the next mother's milk survey will start. Of 300 mothers breast milk samples will be obtained and analysed for dioxins, PCBs and OCPs. The results, which will be available in 1999, will demonstrate the further development of the Dutch body burdens. These data will also participate in the third round of the WHO co-ordinated Exposure study.

Recently, in the Netherlands the dioxin and furan body burden for 1998 was estimated using a physiologically based pharmaco-kinetic (PBPK)-modelling approach (20). Levels in mothers milk ((i)-TEQ) are used as an index for the total body burden. If the new information on historical levels in food is used in the input calculation, the model simulation predict a reduction of the dioxin levels of approximately 35% for 1998 compared to the levels of 1993 (preliminary results, van der Molen).

Human health risk

The health risks which are associated with the PCDD/F and PCB exposures is still subject of discussion. In the Netherlands the exposure of nursed infants to PCDD/Fs and PCBs via mother's milk approximately amounts to 200 pg TEQ/kg.day, for the period of breast feeding. This exposure (of short duration) is substantially higher than the current Tolerable Daily Intake (TDI) for these compounds (10 pg TEQ/kg.day, for life-time exposure).

As mentioned in the introduction, in 1996 the Health Council of the Netherlands derived a recommended limit of human exposure to dioxin-like compounds (TDI) of 1 pg (total) TEQ/kg.day, by using a classical risk assessment approach (animal data; LOAEL, safety factors) (4). The critical effects in these animal studies were cognitive development (in young) and endometriosis (in the mother) Rhesus monkeys, and changes in white blood cells of Marmoset monkeys, at a minimum level of 100 pg/kg.day. According to the Health Council there is no need for limitation of breast feeding, since breast feeding has overall a positive effect on the infants development.

At request of the Health Inspectorate the RIVM, recently, performed a quantitative risk assessment (20). The question addressed was: 'In how many infants adverse health effects will occur if exposed to current (1993) levels of PCDD/Fs and PCBs?' To answer this question four epidemiological studies, which addressed the effects of the background exposure to PCDD/Fs and PCBs on the thyroid hormone status of young children and their cognitive and psychomotor development, were reviewed. In these studies no adverse effects of the postnatal exposure (via breast milk) to dioxins, furans and PCBs on the neurological and the psychomotor development of children were convincingly demonstrated. The results with prenatal exposure were considered inconclusive. Since, from none of the studies a clear exposure-effect relationship could be derived, the results are (at present) considered of limited value for quantitative risk assessment purposes. Thus, based on the present knowledge no quantitative risk could be calculated.

62

Risk management and regulatory issues

In April 1998 the Dutch government sent the report (Stappenplan dioxinen) to the parliament. The main conclusions were:

- Emissions of dioxins have already been reduced, almost as far as possible. Marginal improvements may be possible but will only be implemented on the basis of cost-effectiveness.
- A rough calculation, using European Union System (21) for the evaluation of substances and data of emissions abroad, showed that the contribution of foreign sources to human exposure in the Netherlands is in the range of 68 to 81%
- For the time being the government upholds the TDI of 10 pg/kg.day (WHO, 1990), which is interpreted as total TEQ for PCDD/Fs and dioxin-like PCBs, but is striving at a maximum exposure of 1 pg/kg.day
- In 1999 this position will be evaluated taking into account the re-evaluation of the TDI by WHO, new data on trends in exposure and body burden and model calculations for future developments. This will include the desirability of setting new standards and the lowering of existing standards.

Acknowledgement

The authors would like to thank Sandra Ciere, Gijs Kleter, Matthieu Rikken, Wout Slob, and Marco Zeilmaker for their comments and contributions.

References

- 1. Liem, A.K.D., Berg, R.van den, Bremmer, H.J., Hesse, J.M., Slooff, W., Report no. 710401032, **1993**, RIVM, Bilthoven, the Netherlands.
- Ahlborg, U.G., Kimbrough, R.D., and Yrjänheikki, E.J. Eds. Toxic Substances Journal 12, 101-331 (1992).
- 3. Zorge, van J.A. Proceedings of the 1st International Workshop on Risk Evaluation and Management of Chemicals, Japan, 1998.
- 4. Health Council of the Netherlands. Committee on Risk Evaluation of Substances/ Dioxins. Dioxins. The Hague, 1996; publication no. 1996/10, the Netherlands.
- 5. Bremmer, H.J., Troost, L.M., Kuipers, G., Koning, J. de, Sein, A.A., Report number 770501018, **1994**, RIVM, Bilthoven, the Netherlands.
- 6. Bremmer, H.J., Booy H. Report number 601014005 1995, RIVM, Bilthoven, the Netherlands.
- 7. RIVM. Achtergronden bij: Milieubalans 97. RIVM, 1997, Bilthoven
- 8. NATO/CCMS. International toxicity equivalency factors (I-TEF) method of risk assessment for complex mixtures of dioxins and related compounds. North Atlantic Treaty Organization, report no. 176, 1988, Brussels
- Ahlborg, U.G., Becking, G.C., Birnbaum, L.S., Brouwer, A., Derks, H.J.G.M., Feeley, M., Golor, G., Hanberg, A., Larsen, J.C., Liem, A.K.D., Safe, S.H., Schlatter, C., Wærn, F., Younes, M., Yrjänheikki, E., *Chemosphere*, 28, 1994, 1049
- 10. De Jong, A.P.J.M., Van Jaarsveld, J.A., Bolt-Moekoet, A., Report number 770501019, 1996, RIVM, Bilthoven, the Nethelands.
- 11. Lopez Garcia, A., Boer, A.C. den, Jong, A.P.J.M. de, Environ. Sci. Technol., 30, 1996, 1032
- 12. Berg, R. van den, Hoogerbrugge, R., Groenemeijer, G.S., Gast, L.F.L., Liem, A.K.D., Report number 770501014, **1994**, RIVM, Bilthoven, the Nethelands.

- Evers, E.H.G., Laane, R.W.P.M., Groeneveld, G.J.J., Olie, K., Organohalogen Compounds, 28, 1996, 117
- 14. Beurskens, J.E.M. Microbial transformation of chlorinated aromatics in sediments. Thesis, Landbouwuniversiteit Wageningen, 1995
- 15. Liem, A.K.D., Theelen, R.M.C. Dioxins: Chemical Analysis, Exposure and Risk Assessment. Thesis, Utrecht University, 1997, ISBN 90-393-2012-8
- 16. Dooren-Flipsen, M.M.H. van, Klaveren, J.D. van, Liem, A.K.D., Report number 97.36, 1997, RIKILT-DLO, Wageningen
- 17. Liem AKD, Theelen RMC, Slob W, Wijnen JH. Report number. 730501034, 1991, RIVM, Bilthoven, the Netherlands.
- Liem, A.K.D., Hoogerbrugge, R., Cuijpers, C.E.J., Hartog, R.S. den, Hijman, W.C., Linders, S.H.M.A., Marsman, J.A., Velde, E.G. van der, Zomer, B., Organohalogen Compounds, 33, 1997, 112
- Liem, A.K.D., Albers, J.M.C., Baumann, R.A., Beuzekom, A.C. van, Hartog, R.S. den, Hoogerbrugge, R., Jong, A.P.J.M. de, Marsman, J.A., Organohalogen Compounds, 26, 1995, 69
- 20. C.E.J. Cuijpers, M.J. Zeilmaker, G.W. van der Molen, W. Slob, E. Lebret. Report number 529102007, **1997**, RIVM, Bilthoven, the Netherlands.
- European Chemical Bureau, 1996, EUSES, European Union System for the Evaluation of Substances. Inspra, Italy. EUR 17308 EN.

64