HEALTH RISK ASSESSMENT OF EXPOSURE TO DIOXIN-LIKE PCBS AND DIOXINS IN THE CITY OF MENEN (BELGIUM)

Johan Nouwen[1], Jeroen Provoost[1], Christa Cornelis[1], Jan Bronders[1], Raf De Fré[1], Rudy Van Cleuvenbergen[1]

[1]Flemish Institute for Technological Research (Vito)

Introduction

The City of Menen is a well-outlined residential area located in the neighbourhood of two waste incinerators. The waste incinerators (Menen (Belgium) and Roncq (France)) are in full operation since the eighties. Emission measurements indicate that they fulfil the European Union dioxin emission standard of 0.1 ng TEQ/m³. Despite this, new deposition measurements and analysis of milk in this region indicate a high burden of the local environment with dioxin-like polychlorinated biphenyls (dioxin-like PCBs) and polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs).

It should be noted that formerly the City of Menen was also surrounded by other potential dioxin sources, among them a dye factory, some small illegal cable burning houses, a pressed board manufacturer, and a metal recycling plant. Before 1984, fly ashes of the waste incinerator were locally used as road materials and transported. This could be a secondary source. Additionally the Environmental Inspection has regularly noticed some large open waste fires in this region and follows up the situation.

On request of the Public Waste Agency of Flanders (OVAM) an inventory of all dioxin measurements within the city area was made. The ratio of the dioxin-like PCBs compared to the PCDD/Fs in these measurement results was unexpectedly high. As a consequence of this and in order to achieve an optimal risk assessment for the people living in the City of Menen an additional sampling of soil, vegetables and eggs was carried out. This article focuses on the relative amounts of dioxin-like PCBs and PCDD/Fs in the different media and the consequences for the outcome of the risk assessment.

Methods and materials

In order to carry out the risk assessment as accurately as possible, measurements in different media and biota originating from the City of Menen were used instead of modelling results. Soil, vegetables, eggs (on behalf of OVAM) and milk (on behalf of the Belgian Food Agency FAVV) were analysed for dioxin-like PCBs and PCDD/Fs. Topsoil samples (0-2 cm) were collected at six different locations in the City of Menen. For reasons of sensitivity and in order to reduce the costs for analysis, three pooled samples of six free-range eggs corresponding with three different locations, were analysed. Also twenty samples of vegetables among them endive, beans, lettuce, pumpkins, cucumber, carrots and leek ready for consumption were analysed. The reported results are upper bound levels. Deposition measurements were carried out in this region on behalf of the Flemish Environment Agency (VMM)[1] and reported as medium bound levels.

Three scenarios were examined based on different human exposure patterns. The most likely situation, a lowly exposed scenario (scenario I), was comprised of individuals whose exposure pattern would be representative for the general population. These individuals would consume products sold commercially and would mainly be exposed via the diet as is the case for the general population in Flanders. These people only reside in the impact area. A medium exposed scenario (scenario II) considered individuals living in the City of Menen and consuming 25 % vegetables originating from the gardens at this location. The highly exposed case (scenario III) was applied for individuals who lived in the impact area consuming 25 % home grown vegetables and consuming 1 to 7 eggs a week from free-range chickens in this area. This scenario was considered since consumption of free-range eggs in residential areas is not unusual in Flanders. Additionally chickens peck most of the time in the top soil layer, which contains the highest concentrations of dioxin-like PCBs and PCDD/Fs[2]. Consequently, consumption of free-range eggs could be an important exposure pathway and should be considered for the City of Menen.

There is only limited information on the composition and the levels of dioxin-like PCBs and PCDD/Fs in a normal Flemish diet. The dietary background exposure was estimated at 2.7-3.1 pg WHO-TEQ/(kg bw day)[3]. This is substantially higher than the estimation by Focant et al.[4] arriving at an average daily dietary background exposure apart from the mono-ortho PCBs of 2,0 pg WHO-TEQ/(kg bw day). The upper limit of the average daily dietary background exposure was chosen for the calculations of risk indices since some subpopulations are even

more exposed thereby exceeding the upper limit of the tolerable daily intake (TDI) as defined by the World Health Organization (WHO). Dietary background exposure is taken into account proportionally in each exposure scenario.

The human exposure to dioxin-like PCBs and PCDD/Fs was estimated based on 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) equivalents (WHO-TEQ) in various environmental media and biota. The exposure pathways considered in this assessment include inhalation of air and particles, dermal contact with particles, ingestion of soil, vegetables, water and free-range eggs. Exposure was calculated as the average daily intake of TCDD equivalents per unit body weight. Details on the equations and the calculation of the time-fraction can be obtained from the literature[5]. In Flanders the risk assessment for non-carcinogens is based on the most sensitive subpopulation, mostly children, whereas for carcinogens the risk assessment is based on the unit risk factor of 1:100000 exposed individuals. TCDD is considered as a carcinogen by the WHO[6]. Despite a TDI is given instead of unit risk. Consequently risk indices were calculated for children, adults and lifelong exposure separately. It is assumed that the receptor population is exposed for a 70 year lifetime, divided into two age groups: children (up to 6 years old, 15 kg average body weight) and adults (15-70 years old, 70 kg body weight). Exposure to dioxin-like PCBs and PCDD/Fs was estimated in each group and a time-weighted average daily dose was calculated from the results. The total exposure is divided by the TDI resulting in a risk index (RI). The WHO proposed 1-4 pg WHO-TEQ/kg bw day as the TDI. The upper limit corresponds with a level which should not be exceeded whereas the 1 pg WHO-TEQ/kg bw day is the target value to be met in the long run[6]. Since the average daily dietary background exposure in Flanders is already relatively high, the upper limit was used as the TDI in the modelling. The risk assessment was carried out for the highest concentration in the topsoil layer (42.18 ng WHO-TEQ/kg dm) and in vegetables (0.15 ng WHO-TEQ/kg fw). For the free-range eggs an average concentration of 33.13 pg WHO-TEQ/g fat was used. In order to have an idea about the impact of the variation in the measurements on the RIs, calculations in medium and highly exposed scenarios were also carried out for the minimum and maximum concentrations in vegetables and eggs.

Results and discussion

Table 1 gives an overview of the relative contributions of the dioxin-like PCBs and PCDD/Fs to the total concentrations in soil, vegetables, eggs and milk. The observed soil concentrations are elevated compared to normal background concentrations (±3 ng WHO-TEQ/kg dm) in unpolluted areas. The proposed soil clean-up value for residential areas in Flanders (65 ng WHO-TEQ/kg dm)[7] is not exceeded. Of particular interest is the high contribution of the dioxin-like PCBs (non-ortho and mono-ortho PCBs) compared to PCDD/Fs for the soil sample of the Wervikstraat (dioxin-like PCBs: 56.4%; PCDD/Fs: 43.6%) and to a lesser extent the soil sample of the Lage Weg (dioxin-like PCBs: 18.6%; PCDD/Fs: 81.4%). For other soil samples the contribution of the dioxin-like PCBs was 11% or less.

The observed concentrations in vegetables are very low and much lower than the level for further action (at the moment PCDD/Fs only) as defined by the European Union[8] although some highly accumulating species like pumpkins were analysed. Consequently, there is no indication that consumption of home grown vegetables in the residential area of Menen should be avoided. The results of the Wervikstraat again show a relatively high contribution of the concentration of dioxin-like PCBs (54.5%-71.9%) to the total concentration in vegetables compared to other locations as witnessed by the results for pumpkins, cucumber and lettuce.

The observed concentrations of PCDD/Fs in the free-range eggs exceed the threshold values of the European Union[8] on the average by more than a factor 6. One should note that this threshold value includes presently only PCDD/Fs and is not applicable for free-range eggs from hens owned by private persons. Free-range bio-eggs elsewhere in Flanders usually contain less than 12 pg WHO-TEQ/g fat[9]. The contribution of the PCDD/F-congeners to this amount is 10 pg WHO-TEQ/g fat or less as is the case for two out of the three locations. However, there is an unexpectedly high contribution of dioxin-like PCBs to the total concentration for these locations (68.4 % and 79.3%).

Also the contribution of dioxin-like PCBs in cow's milk is unexpectedly high for the locations Ropswalle and Lage Weg compared to the location Cardijnlaan and other measurements[10]. Additionally, the level for further action (at the moment PCDD/Fs only) as defined by the European Union[8] is exceeded on two locations (Ropswalle and Cardijnlaan).

An overview of the measured deposition of dioxin-like PCBs and PCDD/Fs in the neighbourhood of Menen[1] is given in table 2. Measurement campaigns of the Flemish Environment Agency (VMM) in the spring of 2002 and the autumn of

2002 showed that generally deposition of PCB126 in Flanders is below 10 pg WHO-TEQ/m².day and usually fluctuating around 2 pg WHO-TEQ/m².day. From table 2 it is clear that in the vicinity of Menen significantly higher PCB126-levels are found in the Wervikstraat, Sluizenkaai and close to the French border. The monthly measurements for the Wervikstraat are continuously elevated (not shown here). Also the deposition of PCDD/Fs is highly elevated in the Wervikstraat and close to the French border, suggesting that a local source is responsible for the contamination of this region. The sum of the measured PCB126 en PCDD/F-deposition exceeds the Flemish guideline value of 26 pg WHO-TEQ/m².day (French border: 36 pg WHO-TEQ/m².day; Wervikstraat: 66 pg WHO-TEQ/m².day).

The measurements for soil, food and deposition in the vicinity of Menen clearly indicate that a considerable contamination by a local PCB-source is not widespread and restricted to smaller parts of this city. As a consequence of this additional deposition measurements were carried out in the neighourhood of a metal recycling plant.

Table 1: Overview of the measured concentrations in the different environmental media

Location	matrix	non-ortho PCBs	mono-ortho PCBs	PCDD/Fs	Total
	Soil (ng WHO-TEQ/kg dm)				
Murisson		0.77 (3.6 %)	0.27 (1.2 %)	20.54 (95.2 %)	21.59 (100 %)
Murisson		1.64 (4.1 %)	0.32 (0.8 %)	38.32 (95.1 %)	40.27 (100 %)
Coulonstraat		0.91 (5.6%)	0.31 (1.9 %)	15.04 (92.5 %)	16.26 (100 %)
Lage Weg		1.94 (14.5 %)	0.54 (4.1 %)	10.83 (81.4 %)	13.31 (100 %)
Wervikstraat		17.75 (42.1 %)	6.06 (14.3 %)	18.38 (43.6 %)	42.18 (100 %)
Rozenlaan		1.01 (8.3 %)	0.33 (2.7 %)	10.80 (89.0 %)	12.14 (100 %)
	Vegetables (ng WHO-TEQ/kg fw)				
Murisson	pumpkin	0.0024 (1.9 %)	0.012 (9.7 %)	0.110 (88.4 %)	0.12 (100 %)
	cucumber	0.0053 (4.8 %)	0.012 (10.8 %)	0.094 (84.4 %)	0.11 (100 %)
	lettuce	0.0042 (6.8 %)	0.012 (19.2 %)	0.046 (74.0 %)	0.06 (100 %)
	chinese lettuce	0.0082 (19.9 %)	0.012 (29.1 %)	0.021 (51.0 %)	0.04 (100 %)
	carrot	0.0044 (7.4 %)	0.012 (20.2 %)	0.043 (72.4 %)	0.06 (100 %)
Murisson	cucumber	0.0090 (8.0 %)	0.041 (36.2 %)	0.063 (55.8 %)	0.11 (100 %)
Coulonstraat	pumpkin	0.0011 (3.9 %)	0.012 (42.7 %)	0.015 (53.4 %)	0.03 (100 %)
	lettuce	0.0018 (8.3 %)	0.012 (55.3 %)	0.008 (36.4 %)	0.02 (100 %)
	endive	0.0027 (5.6 %)	0.012 (24.6 %)	0.034 (69.8 %)	0.05 (100 %)
Lage Weg	cucumber	0.0082 (15.7 %)	0.015 (28.7 %)	0.029 (55.6 %)	0.05 (100 %)
	lettuce	0.0050 (11.6 %)	0.012 (27.9 %)	0.026 (60.5 %)	0.04 (100 %)
	endive	0.0094 (17.6 %)	0.015 (28.1 %)	0.029 (54.3 %)	0.05 (100 %)
	carrot	0.0150 (15.6 %)	0.013 (13.6 %)	0.068 (70.8 %)	0.10 (100 %)
	cucumber	0.0032 (6.8 %)	0.012 (25.4 %)	0.032 (67.8 %)	0.05 (100 %)
Wervikstraat	pumpkin	0.0026 (9.4 %)	0.012 (45.1 %)	0.012 (45.5 %)	0.03 (100 %)
	cucumber	0.0200 (14.8 %)	0.059 (43.7 %)	0.056 (41.5 %)	0.14 (100 %)
	lettuce	0.0600 (52.6 %)	0.022 (19.3 %)	0.032 (28.1 %)	0.11 (100 %)
Rozenlaan	lettuce	0.0033 (5.6 %)	0.012 (20.6 %)	0.043 (73.8 %)	0.06 (100 %)
	carrot	0.0065 (8.2 %)	0.012 (15.1 %)	0.061 (76.7 %)	0.08 (100 %)
	leek	0.0320 (22.1 %)	0.013 (9.0 %)	0.100 (68.9 %)	0.15 (100 %)
	beans	0.0027 (4.3 %)	0.012 (19.1 %)	0.048 (76.6 %)	0.06 (100 %)
	Eggs (ng WHO-TEQ/kg fat)				
Murisson		8.8 (31.0 %)	1.6 (5.6 %)	18.0 (63.4 %)	28.4 (100 %)
Lage Weg		16.0 (51.1 %)	5.4 (17.3 %)	9.9 (31.6 %)	31.3 (100 %)
Wervikstraat		22.0 (55.4 %)	9.5 (23.9 %)	8.2 (20.7 %)	39.7 (100 %)
	Milk (ng WHO-TEQ/kg fat)				
Ropswalle		8.6 (63.5 %)	2.5 (18.5 %)	2.4 (18.0 %)	13.5 (100 %)
Lage Weg		25 (66.4 %)	11 (29.3 %)	1.6 (4.3 %)	37.6 (100 %)
Cardijnlaan		4.2 (50.6 %)	1.1 (13.3 %)	3 (36.1 %)	8.3 (100 %)

Table 2: Measured deposition (ng WHO-TEQ/m².day) for PCB126 and PCDD/Fs in the neighbourhood of Menen in 2002.

Location	PCB126		PCDD/Fs			
Time period	spring 2002	autumn 2002	5-6/01	11-1/02	4-6/02	10-12/02
Menen-Rekkem	2.1	< 1	4.4	2.2	2.6	2.6
Wervikstraat	41	38	12	4.5	25	24
Ter Berken	2.5	1.5	3.9	3.6	4.5	4.7
Menen French border	17	3.4	12	5.1	19	5
Menen Sluizenkaai	13	2.7	6.4	4.7	6.2	6.2

Close to a nearby metal recycling plant the Flemish guideline value is regularly exceeded by more than a factor 10 (> 300 pg WHO-TEQ/m².day for the sum of PCB126 and PCDD/Fs; see table 3). Also the ratio of the PCB126-deposition and the PCDD/F-deposition is unexpectedly high since for normal air concentrations the contribution of dioxin-like PCBs compared to PCDD/Fs is usually less than 20 %. Presently, it is unclear if the observed deposition in the immediate surrounding of this metal recycling plant originates from other sources or from blown up dust particles from the metal recycling site. The relevance of the potential contribution of shredder activities and blown up dust particles caused by local on-site and off-site heavy traffic is currently investigated. The observed deposition of dioxin-like PCBs and PCDD/Fs combined with the relatively high concentrations of these persistent pollutants in soil, eggs and milk indicate that the region of Menen is heavily polluted. One or more local sources other than both nearby waste incinerators pollute the environment significantly requiring additional emission reducing measures. Consequently, a risk assessment was carried out on behalf of the OVAM.

Table 3: Measured deposition (ng WHO-TEQ/m².day) of PCB126 and PCDD/Fs close to a metal recycling plant in the neighbourhood of Menen in 2002-2003.

Location	PCB126	PCDD/Fs	PCB126	PCDD/Fs	PCB126	PCDD/Fs
Time period	10-11/02	10-11/02	11-12/02	11-12/02	1-2/03	1-2/03
Flanders	174	156	258	131	194	191
France metal recycling plant	69	38	101	46	100	73
France Leie	23	18	21	12.5	32	40

The risk-index was calculated by dividing the summation of the background and lifelong exposure by the upper limit of the TDI as defined by the WHO. A RI < 1 means that there is no indication for a health risk. On the contrary, a RI $\geq$ 1 indicates a risk for human health. The calculated exposure is always higher for

children than for adults (see table 4). This has to be at least partially attributed to the different consumption behaviour and lower bodyweight of a child. The lifelong average exposure is hardly different from the exposure of adults as witnessed by the resulting RIs except for the highly exposed scenario (scenario III). Comparison of all exposure scenarios shows an increase of the RIs with increasing consumption of locally produced food. Just residing in the impact area (scenario I) does not result in a meaningful risk. The resulting RI for this scenario is mainly due to the dietary background exposure. The exposure and consequently resulting RI due to consumption of vegetables (scenario II) is only slightly higher than in scenario I but still the RI < 1. Consequently there is no reason to discourage the consumption of locally grown vegetables. Additional consumption of one or more free-range eggs results in a RI >1. From the point of view of lifelong exposure a limited consumption of free-range eggs (1 egg/week or less) might be acceptable although it should be dissuaded for vulnerable populations like children since the risk assessment for the most sensitive group results in a RI =1.39 > 1. The resulting ranges for the minimum and maximum concentrations in vegetables (scenario II) and eggs (scenario III) is also given between brackets. Based on the measurements of dioxin-like PCBs and PCDD/Fs in locally produced food the human exposure in addition to the general background exposure could be due for more than 70 % to dioxin-like PCBs in extreme cases (Wervikstraat). Consequently, the upcoming European Union food safety standards which will come into force by the end of 2004 are very useful. Free-range eggs from hens owned by private persons are beyond the scope of this legislation but can have serious consequences on the outcome of the risk assessment.

Table 4: Overview of the calculated RIs.

Scenario*	Child + background	Adult + background	Lifelong + background
I	0.79	0.75	0.76
II	0.89 (0.80-0.89)	0.80 (0.76-0.80)	0.81 (0.76-0.81)
IIIa	1.39 (1.32-1.49)	0.91 (0.89-0.93)	0.95 (0.93-0.98)
IIIb	2.44 (2.21-2.73)	1.14 (1.09-1.21)	1.26 (1.18-1.34)
IIIc	4.50 (3.98-5.22)	1.62 (1.50-1.78)	1.86 (1.71-2.08)

* Scenario I: residents consuming only commercially sold food, Scenario II: consumption of 25 % home grown vegetables, Scenario III: consumption of 25 % home grown vegetables and 1 (IIIa), 3 (IIIb) and 7 (IIIc) eggs/week respectively.

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References

1. Vlaamse Milieumaatschappij (VMM) (2003) Persnota Vlaamse Milieumaatschappij Depositie van dioxines en PCB126 in 2002, communication to the press (in Dutch).
2. Schuler F., Schmid P., Schlatter Ch., Chemosphere, 34, 711-718, 1997.
3. De Fré R., Cornelis C., Lewyckyj N., Mensink C., Nouwen J., Schoeters G., Swaans W., Wevers M. (2001) Proposal of environmental quality objectives for dioxin deposition. Vito-report 2001/MIM/R163, Mol Belgium (in Dutch).
4. Focant J.F., Eppe G., Pirard C., Massart A.C., André J.E., De Pauw E., Chemosphere, 48, 167-179 (2002)..
5. Nouwen, J., Cornelis, C., De Fré, R., Wevers, M., Viaene, P., Mensink, C. Patyn J., Verschaeve, L., Hooghe R., Maes, A., Collier, M., Schoeters, G., Van Cleuvenbergen, R., Geuzens, P. , Chemosphere 43, 909-923 (2001).
6. World Health Organization, (1999) Assessment of the health risk of dioxins: Re-evaluation of the tolerable daily intake (TDI). WHO report EHBI 010201, Geneva, Switzerland.
7. J. Nouwen, C. Cornelis, J. Provoost, Organohalogen Compounds, 62, 411-414, 2003.
8. Official Journal of the European Communities Council Directive 2735/2001/EC of 29 November 2001 amending Commission Regulation 466/2001/EC setting maximum levels for certain contaminants in foodstuffs (2001).
9. http://www.favv-afsca.fgov.be
10. Van Cleuvenbergen R., Mannaert P., Van Durme N., Vinkx C., Goeyens, Organohalogen Compounds, 56, 441-444, 2002.