

## HEALTH RISK ASSESSMENT OF DIOXIN EXPOSURE IN THE CITY OF MENEN (BELGIUM)

Johan Nouwen<sup>1</sup>, Jeroen Provoost<sup>1</sup>, Christa Cornelis<sup>1</sup>, Jan Bronders<sup>1</sup>

<sup>1</sup>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)) were in full operation since the eighties. Both waste incinerators are still operational and emission measurements indicate that they fulfil the European Union dioxin emission standard of 0.1 ng TEQ/m<sup>3</sup>. Despite of this, new deposition measurements and analysis of milk in this region indicate a high burdening 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. In the past, before 1984, fly ashes of the waste incinerator were locally valorized as road materials and transported. This could be a secondary source. Additionally the Environmental Inspection has regularly noticed some large open wastefires in this region and follows up the situation.

On request of the Public Waste Agency of Flanders (OVAM) an inventory of all measurements in this area was made. An additional sampling of soil, vegetables and eggs was carried out in order to achieve an optimal human risk assessment for the City of Menen.

At first instance this risk assessment was carried out by using a deterministic approach. The used human exposure model calculates point estimates based on a combination of unique parameter values. This, however, gives no indication of the variation of the model output. The value of a model parameter is chosen such that a conservative point estimate is obtained for the risk index. Due to variation, it is often unclear how conservative this estimate really is. Hence the variation of the calculated risk index is not known. The calculations of the risk index are subjected

to two sources of variation: uncertainty and variability. This article focuses on the quantification of the variation in the risk indices using a probabilistic approach.

### **Methods and materials**

In order to carry out the risk assessment as accurately as possible preference was given to measurements in different media and biota originating from the City of Menen instead of modelling results. Soil, eggs and vegetables were analysed for dioxin-like PCBs and PCDD/Fs. Topsoil samples (0-2 cm) were collected at six different locations. Concentrations were ranging from 12.14 to 42.18 ng WHO-TEQ/kg dm. Three pooled samples of six free-range eggs each contained 28.4, 31.3 and 39.7 pg WHO-TEQ/g fat respectively. Also twenty samples of vegetables among them endive, beans, lettuce, pumpkins, cucumber, carrots and leek ready for consumption were analysed. Concentrations varied from 0.02 to 0.15 pg WHO-TEQ/g fw.

Three scenarios were examined based on different exposure patterns. The common case, 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 Flemish population. These people only reside in the impact area. A medium exposed scenario (scenario II) considers 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 to individuals who lived in the contaminated area consuming 25 % home grown crops 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<sup>1</sup>. One should also take into account that free-range eggs regularly exceed the European Union guidelines for food<sup>2</sup>. 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/F's in a normal Flemish diet. The dietary background exposure was estimated based on a official list of the average daily consumption<sup>3</sup>, a calculation of the daily consumption of well defined food categories using data of the National Institute of Statistics<sup>4</sup> and a survey of food<sup>5</sup>. This information was

combined with recent analytical data from the Belgian Federal Agency for the Safety of the Food Chain (FAVV) for the reference year 2000. When a variation of the fat content of some food products and the assumption that dioxin-like PCBs contribute for 50 % to the total dietary background exposure are taken into account, this results in an average daily background exposure of 2.7-3.1 pg WHO-TEQ/(kg bw day)<sup>6</sup>. The upper limit of the average daily dietary background exposure was chosen for the calculations since some subpopulations are even more exposed thereby exceeding the upper limit of the tolerable daily intake (TDI). 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 and was estimated by the models presented in VLIER-HUMAAN<sup>7</sup>. Details on the equations and the calculation of the time-fraction can be obtained from the literature<sup>7,8</sup>. 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 World Health Organization (WHO)<sup>9</sup>. 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<sup>9</sup>. Since the average daily dietary background exposure is already relatively high, the upper limit was used as the TDI in the modelling.

The deterministic 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. The probabilistic approach includes variation in the modelling. The first step involves the computation of probability distribution functions for the relevant input parameters from the literature and measured data. The distributions are given in table 1.

*Table 1 : Input parameters and probability distributions.*

Model parameters	Symbol	Unit	Distributions	
			Adult	Child
<i>Location</i>				
Concentration soil*	C <sub>s</sub>	ng TEQ/kg.dm	T (12.14; 24.29; 42.18)	
Time human inside winter	T <sub>iw</sub>	h/d	U (13; 15)	U (9; 11)
Time human outside winter	T <sub>ow</sub>	h/d	U (1; 3)	U (1.5; 2.5)
Time human inside summer	T <sub>is</sub>	h/d	U (8; 12)	U (4; 8)
Time human outside summer	T <sub>os</sub>	h/d	U (4; 8)	U (4; 8)
Length of the site	L	m		U (10; 100)
Width of the site	W	m		U (10; 100)
Surface roughness	Sr	m		U (0.2; 0.7)
Build-on or paved surface	f <sub>h</sub>	-		U (0.15; 1.00)
<i>Soil</i>				
Soil temperature	T <sub>soil</sub>	K		N (283; 0.70)
Fraction organic carbon	f <sub>oc</sub>	-		LN (-4.36; 0.58)
Water filled porosity	SN <sub>w</sub>	-		U (0.09; 0.39)
Air filled porosity	SN <sub>a</sub>	-		U (0.10; 0.35)
<i>Climate</i>				
Suspended particles outside	TSPo	kg/m <sup>3</sup>	LN (50%: 7.55E-8; 90%: 1.01E-7)	
Fraction soil dust in indoor air	FSDia	-	U (0.20; 0.85)	
<i>Plant and animals</i>				
Concentration plants*	C <sub>pt</sub>	mg/kg fw	T (6.99E-9; 4.00E-8; 1.81E-7)	
Crop yield	Yv	kg/m <sup>2</sup>	T (0.11; 0.28; 0.44)	
Dry matter root	DM <sub>r</sub>	-	U (0.1; 0.2)	
Dry matter stem	DM <sub>s</sub>	-	U (0.1; 0.2)	
Amount of eggs per week	Ae	-	T (0; 2; 7)	
Concentration eggs*	Cegg	pg TEQ/g fat	T (28.4; 31.3; 39.7)	
<i>Human</i>				
Body weight	BW	kg	LN (73; 7)	LN (15; 4)
Volume inhaled	V <sub>i</sub>	m <sup>3</sup> /d	LN (16.2; 3.8)	LN (9.3; 2.4)
Total surface area body	TSA <sub>b</sub>	m <sup>2</sup>	LN (10%: 1.61; 95%:2.19 )	LN (10%: 0.72; 95%: 0.88 )
Surface area hands	SA <sub>h</sub>	m <sup>2</sup>	LN (10%: 0.08; 95%: 0.11)	T (0.039; 0.04; 0.043)
Surface area body covert by dust, inside	SAD <sub>i</sub>	kg/m <sup>2</sup>	U (1.50E-5; 4.30E-5)	U (3.10E-5; 1.50E-3)
Surface area body covert by dust, outside	SAD <sub>o</sub>	kg/m <sup>2</sup>	U (8.00E-6; 6.60E-3)	U (9.00E-3; 1.50E-2)
Surface area forearms + hands	SA <sub>ah</sub>	m <sup>2</sup>	LN (10%: 0.20; 95%: 0.28)	0,1
Surface area arms + hands	SA <sub>ah</sub>	m <sup>2</sup>	N (0.34; 0.02)	T (0.133; 0.137; 0.143)
Surface legs and feet	Alf	m <sup>2</sup>		T (0.21; 0.22; 0.24)
Ingestion soil	I <sub>s</sub>	kg/d	A (2,60E-5)	LN (1.27E-4; 1.27E-4)
Fraction of dust restrained in long	FD <sub>l</sub>	-	N (0.75; 0.05)	N (0.75; 0.05)
Consumption home grown vegetables	C <sub>vegetables</sub>	kg/d	A ( 0,369)	A (0,216)
Fraction contaminated fruit and vegetables	f <sub>fv</sub>	-		U (0,14; 0,57)
<i>Contaminant</i>				
Octanol-water partition coefficient	Kow	g/g	LN (6.80; 0.729)	

N: normal; LN: log-normal; U: uniform; T: triangular; A: average

\*measurements in the framework of this investigation

The second step implies the random selection of values from the probability distributions by using a Latin-Hypercube sampling method<sup>10</sup>. Calculations of the RI were repeated for 5000 combinations of parameter values using Monte-Carlo simulations. The third step involves displaying the results as a ranking of the input parameters according to the correlation between these input parameters and the model output<sup>10</sup> on the one hand and a frequency distribution of calculated RIs on

the other. The Monte Carlo simulations and sensitivity analysis were performed by means of the Crystal Ball®<sup>11</sup> simulation software.

### Results and discussion

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. Table 2 shows the calculated RIs for all exposure scenarios using the deterministic approach. The calculated exposure is always higher for children than for adults. This has to be at least partially attributed to the different consumption behaviour and lower bodyweight of a child. The lifelong average exposure is not significantly 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 as expected 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 in 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  $RI < 1$ . Based on this evaluation and the observed concentrations in vegetables which are much lower than the level for further action as defined by the European Union<sup>12</sup> there is no indication that consumption of home grown vegetables should be avoided. Additionally the observed concentrations are very low although some highly accumulating vegetables were analysed. Additional consumption of 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 just 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 observed concentrations of PCDD/Fs in the free-range eggs exceed the threshold values of the European Union<sup>12</sup> 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 chickens kept by private persons. The observed concentrations in free-range eggs originating from the City of Menen are also substantially higher compared to these originating from other locations in Flanders<sup>2</sup>.

*Table 2: Overview of the calculated RIs using a deterministic approach.*

Scenario*	Child + background	Adult + background	Lifelong + background
I	0.79	0.75	0.76
II	0.89	0.80	0.81
IIIa	1.39	0.91	0.95
IIIb	2.44	1.14	1.26
IIIc	4.50	1.62	1.86

\* 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.

Table 3 shows the frequency distributions of the RIs. These were narrow in the lowly and medium exposed scenarios in contrast to the highly exposed scenario. Most of the variance in the lowly exposed scenario is due to the soil ingestion by children (44 %), inhaled volume by adults (17 %), body weight of children (13 %), soil concentration (12 %) and the fraction soil dust in indoor air (6 %). In the medium exposed scenario 71 % of the variance is explained by the concentration of dioxin-like PCBs and PCDD/Fs in vegetables and the fraction of consumed contaminated vegetables. Consumption of free-range eggs determines the variation in the highly exposed scenario (94 %). Contribution of other parameters in each of these scenarios was negligible.

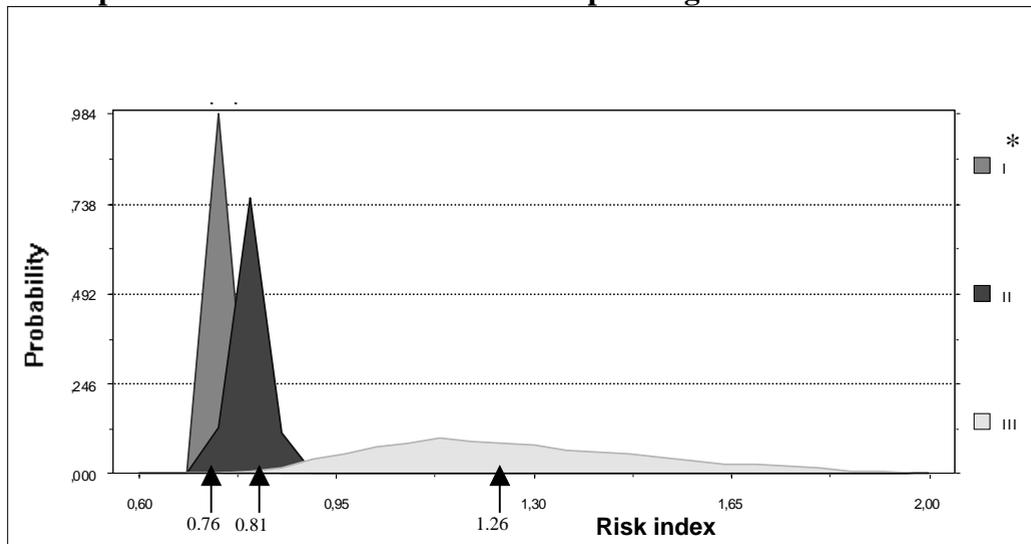
*Table 3: Overview of the calculated RIs for lifelong exposure using a probabilistic approach.*

Scenario*	Minimum	Maximum	Mean	Median
I	0.75	0.82	0.75	0.75
II	0.75	0.92	0.79	0.79
III	0.79	2.20	1.27	1.24

\* 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 to 7 eggs/week.

The model calculations were evaluated by comparing the calculated RI in the deterministic approach to the calculated distribution of RIs for the lifelong exposure in each scenario. Figure 1 shows the frequency distribution diagram for the 5000 calculated RIs. Each of these RIs originates from a random parameter set selected from the ranges as given in table 1. The probability resulting from the frequency divided by the number of trials (5000) is given in the same figure. All RIs < 1 in scenario I and scenario II. In scenario III more than 87 % of the calculated RIs > 1 which suggests that a limited consumption of free-range eggs from this area could be tolerated but that that it is not advisable.

**Figure 1: Frequency and probability distribution diagram of 5000 random parameter combinations and corresponding deterministic RIs.**



\*I = scenario I; II = scenario II; III = scenario III

These conclusions are in agreement with those of the deterministic approach. However, a significant number of calculated RIs in the probabilistic approach exceeds the RIs of the corresponding exposure scenarios in the deterministic approach as witnessed by comparison of the average and maximum RIs from table 3 and the RIs for lifelong exposure from table 2. Consequently, the deterministic approach is not as conservative as initially assumed.

Uncertainties related to the model formulas itself, the toxicology (TDI), background exposure and the measurements were not considered. One has to take into account that other variables not related to the uncertainty of the modelling also play an important role, among them uncertainty of sampling, seasonality of the measurements and analytical measurement errors.

The observed variation on the RIs should be interpreted carefully. One should keep in mind that the measurements are only representative of the residential area of Menen in which the sampling was carried out. Also the choice of the parameter distributions is discussable. Only limited information for some of the parameters is available. A survey of location specific data with regard to consumption behaviour is missing. Information on consumption of products from animal origin other than

eggs (milk, meat) produced in this area could possibly place the risk assessment in another perspective.

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