

ASSESSMENT OF HUMAN EXPOSURE TO PCDD/F AT CONTAMINATED SITES COMPARED TO BACKGROUND EXPOSURE

Åberg A A¹, Tysklind M¹, Wiberg K¹, Hanberg A², Ask I³, MacLeod M⁴

¹ Department of Chemistry, Environmental Chemistry, Umeå University, SE-901 87 Umeå, Sweden

² Institute of Environmental Medicine, Karolinska Institutet, SE-171 77 Stockholm, Sweden

³ SWECO VIAK AB, Västra Norrlandsgatan 10 B, SE-903 27 Umeå, Sweden

⁴ Institute for Chemical and Bioengineering, Safety and Environmental Technology Group, Swiss Federal Institute of Technology, ETH Zürich, HCI G129, CH-8093 Zürich, Switzerland

Introduction

In Sweden, nearly 370 former chlorophenol (CP) wood impregnation sites have been identified in the national inventory. Many of these sites are contaminated with PCDD/Fs and due to the high toxicity of PCDD/Fs, they are therefore prioritised for further investigations of contamination level and for risk assessments. It is generally agreed that most of the human exposure to PCDD/F is due to food intake¹, but local point sources, such as municipal waste incinerators, have been proved to contribute to exposure at a local scale². Increased levels of PCDD/Fs in environmental media close to point sources may affect human exposure through several pathways, such as production and intake of locally grown food, inhalation of dust and particles, dermal contact and ingestion of soil. Many investigators have shown that PCDD/F in air is the general contributor to contamination of the food chain at background levels^{3,4}. A locally elevated background level of PCDD/F in air at contaminated sites may thus affect humans both via the food chain pathway as well as via direct exposure by inhalation. At high soil concentrations, adsorption of soil particles to plant surfaces can also add to the total concentration in plants⁵ as well as contribute to high levels of PCDD/Fs in eggs⁶ and pose a health risk to humans⁷.

Usually, site specific investigations of contaminated sites in Sweden focus on environmental media concentrations in soil, sediment and water. The cost for PCDD/F analysis usually limits the possibilities to thoroughly investigate the fate and exposure at such sites. Also, the samples taken only reveal the situation today and not the accumulated effect over several years. To improve our understanding of the extent to which contaminated soil can contribute to increments in human background exposure we therefore used a multi-media fate- and exposure-modelling approach. Population defined scenarios at different environmental concentrations then allows a quantitative comparison of exposure by several pathways.

Materials and Methods

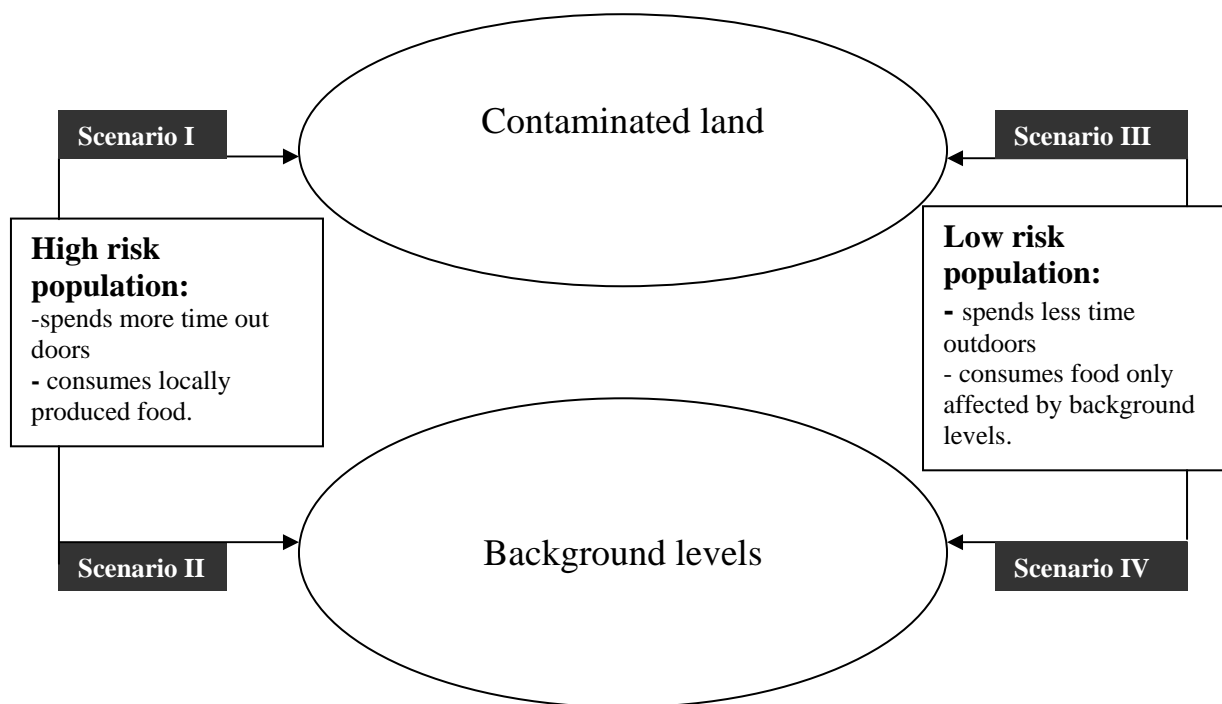
Parameterization of model The multi media fate and exposure model CalTOX⁸ was parameterized to represent the southern Swedish climate with an annual mean temperature of ~7°C, annual mean precipitation of 570 mm and annual mean wind speed of 4.6 m/s. The regional-scale landscape consists of both land (90%) and water (10%). Degradation rates appropriate to the range of temperatures in southern Sweden were taken from the study by Sinkkonen and Paasivirta⁹. Physico-chemical properties of selected PCDD/F congeners were mainly taken from Mackay et al.¹⁰ and temperature adjusted according to Beyer et al.¹¹. The values are presented in Table 1. The congeners examined in this study were 2,3,7,8-TCDD, 1,2,3,4,7,8- HxCDD, 1,2,3,4,6,7,8-HpCDD, OCDD, 2,3,4,7,8-PeCDF and 1,2,3,4,6,7,8-HpCDF. The congeners TCDD and OCDD were chosen since they represent the full interval of physico- chemical properties and thus the expected behaviour of PCDD/Fs in the environment. The congeners HxCDD, HpCDD and HpCDF represent congeners which usually contribute significantly to the total TEQ-value at contaminated sites. PeCDF is commonly found in food products.

Table 1. *PhysiCo-chemical properties of selected PCDD/Fs.*

Physico-chemical properties	2,3,7,8-TCDD	1,2,3,6,7,8-HxCDD	1,2,3,4,6,7,8-HpCDD	OCDD	2,3,4,7,8-PeCDF	1,2,3,4,6,7,8-HpCDF
log V^s (Pa)	-4.92	-6.95	-7.88	-7.18	-5.82	-7.34
log S_w^s (mol/m ³)	-4.61	-5.46	-5.79	-6.68	-4.1	-6.29
H (Pa·m ³ /mol)	3.34	1.08	1.27	0.69	0.51	1.43
log Kow	7.01	7.76	7.78	7.84	6.63	7.18
log Koc	6.64	7.90	8.86	9.11	7.6	7.63

Exposure Scenarios

The degree of exposure is not solely dependent on the source strength of the contaminant. Human factors, such as rates of food intake contact rates with contaminated media, contribute as well. Therefore, a set of four exposure scenarios were defined which allow a comparison of exposure at different source strength as well for different population behaviour. The scenarios are schematically presented in Figure 1.

**Figure 1.** *A schematic representation of the two populations at different source strengths combined to four modelling scenarios.*

The high risk population represents people who produce and consume their own food. The food products consist of vegetables, eggs and locally caught fish. In addition, the population spends more time outdoors, i. e. in their gardens, which also affect the dermal contact with contaminated material as well as soil ingestion and inhalation. The low risk population consume only commercially available food products obtained from regions with background concentrations. Compared to the high risk population, they also spend less time outdoors, which lowers

Risk assessment

their contact with the contaminated area. Food consumption characteristics of both populations were as far as possible estimated from a Swedish study of intake of PCDD/F from food products¹². Besides, the Human Exposure Factors Handbook^{13, 14, 15} was used to select values for parameters relevant to the other included exposure pathways (soil ingestion, inhalation and dermal contact). Table 2 summarises the food consumption characteristics of the populations.

Table 2. Food consumption characteristics of adult individuals in high and low risk populations.

Parameter	Low risk population	High risk population
Body weight (kg)	70	70
Fruit and vegetable intake (kg/kg-d)	0.0035	0.0035
Milk intake (kg/kg-d)	0.0053	0.0053
Meat intake (kg/kg-d)	0.0013	0.0013
Egg intake (kg/kg-d)	0.00013	0.00013
Fish intake (kg/kg-d)	0.0005	0.0005
Fraction fruits & vegetables that are exposed produce	0.47	0.47
Fraction of fruits and vegetables local	0	0.33
Fraction of eggs local	0	0.5
Fraction of fish local	0	0.6
Fraction of milk local	0	0
Fraction of meat local	0	0

A survey of PCDD/F contaminated soil investigations was done to estimate the in-place contamination in the contaminated land scenarios (I and III). The range of contamination level varies widely from place to place and between different congeners. The contamination levels in the exposure scenarios have therefore been set near the high end of a realistic range of contamination levels, but without approaching a true "worst-case" level. Few measurements on background levels of PCDD/Fs in Sweden have been done recently. Matscheko et al.¹⁶ analysed background concentrations in agricultural soil and measurements of PCDD/Fs in air was earlier done by Tysklind et al.¹⁷. Even though atmospheric deposition has decreased over the last ten years, these data were used to define the levels of PCDD/Fs in the background scenarios (II and IV). Modelled PCDD/F concentrations are shown in Table 3. For the contaminated land scenario, it is assumed that only the contaminated site contributes to PCDD/F levels in other compartments.

Table 3. Environmental concentrations in contaminated land and background scenarios (n d = not detected, d w = dry weight).

Environmental concentrations	2,3,7,8-TCDD	1,2,3,6,7,8-HxCDD	1,2,3,4,6,7,8-HpCDD	OCDD	2,3,4,7,8-PeCDF	1,2,3,4,6,7,8-HpCDF
Contaminated soil (pg/g d w)	20	20 000	200 000	200 000	200	200 000
Background soil (pg/g d w)	n d	0.23	2.8	15	0.45	3.05
Background air, rural (pg/m ³)	0.002	0.002	0.03	0.24	0.002	0.01

Results and Discussion

Preliminary results from a sensitivity analysis indicate that both K_{oc} and degradation rates in soil have a great influence when modelling the total exposure of PCDD/F to humans. Due to the high bioconcentration potential of

Risk assessment

fish, intake of locally caught fish may be an important exposure pathway to humans. Landscape parameters such as surface water flow and depth of the surface water should therefore resemble site specific conditions as far as possible to avoid misleading results. One major disadvantage of modelling exposure from intake of fish in CalTOX is that the model does not take biomagnification into account. Uptake of hydrophobic compounds in fish is not only due to bioconcentration but also to feed intake. Therefore, refined modeling efforts should consider both bioconcentration and biomagnification.

The use of a population with production of own food on a site with high levels of contamination in our scenarios is probably not a truly realistic scenario. Still, we used the approach to model a worst-case scenario to compare a high risk scenario to the background scenario. The final results will allow relative comparison of total intake and contributions from different exposure pathways from the contaminated soil to the total background exposure from food consumption. In future work, the model will be validated with field measurements of plant uptake and air concentrations at a real contaminated site. After validation different remediation options of the site will be evaluated.

Acknowledgements

We are grateful to the Swedish Environmental Protection Agency who financed the project “ Evaluation of Fate and Transport: A Comprehensive View”.

References

1. Buckley-Golder D. *Compilation of EU dioxin exposure and health data*. 1999. European Commission DG Environment, UK Department of the Environment Transport and the Regions (DETR). AEAT/EEQC/0016.
2. Pirard C, Eppe G, Massart A-C, Fierens S, De Pauw E, and Focant J-F. *Environ Sci Tech* 2005;39:4721-4728.
3. McLachlan M. *Chemosphere* 1997;34:1236-1276.
4. Schuhmacher M, Jones KC, Domingo JL. *Environ Pollut* 2006;142:143-150.
5. Smith KEC, Jones KC. *Sci Total Environ* 2000;246:207-236.
6. Schuler F, Schmid P, Schlatter C. *Chemosphere* 1997;34:711-718.
7. Dahlgren J, Warshaw R, Horsak RD, Parker III FM, Takhar H. *Environ Res* 2003;92:99-109.
8. McKone TE. CalTOX: A Multimedia Total Exposure Model for Hazardous Waste Sites, Part I-III. 1993. Department of toxic substances control, Lawrence Livermore National laboratory, Livermore.
9. Sinkkonen S, Paasivirta J. *Chemosphere* 2000;40:943-949.
10. Mackay D, Shiu W-Y, Ma KC. *Illustrated handbook of physical-chemical properties and environmental fate for organic chemicals. Volume II. Polynuclear aromatic hydrocarbons, polychlorinated dioxins and dibenzofurans*, LEWIS Publishers, Michigan, USA, 1992.
11. Beyer A, Wania F, Gouin T, Mackay D, Matthies M. *Environ Toxicol Chem* 2002;21:941-953.
12. Lind Y, Darnerud PO, Aune M, Becker W. *Exponering för organiska miljökontaminanter via livsmedel*. SLV Report 26, 2002, National Food Administration, Uppsala, Sweden.
13. *Exposure Factors Handbook, vol I*. 1996, EPA/600/P-95/002Ba. Office of Research and Development, National Center for Environmental Assessment, U S Environmental Protection Agency, Washington.
14. *Exposure Factors Handbook, vol II*. 1997, EPA/600/P-95/002Fb. Office of Research and Development, National Center for Environmental Assessment, U S Environmental Protection Agency, Washington.
15. *Exposure Factors Handbook, vol III*. 1997, EPA/600/P-95/002Fc. Office of Research and Development, National Center for Environmental Assessment, U S Environmental Protection Agency, Washington.
16. Matscheko N, Tysklind M, de Wit C, Bergek S, Andersson R, Sellstrom U. 2002. *Environ Toxicol Chem* 2002; 21:2515-2525.
17. Tysklind M, Fängmark I, Marklund S, Lindskog A, Thaning L, and Rappe C. *Environ Sci Tech* 1993; 27:2190-2197.