SOIL CLEAN-UP VALUES FOR POLYCHLORINATED BIPHENYLS (PCBs) AND DIOXINS (PCDD/Fs) USING HUMAN EXPOSURE MODELLING

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Introduction

Clean-up standards for soil pollution were promulgated on the 27th of April 1996 by the implementing order of the Flemish soil remediation decree. The soil clean-up standards differentiate between five land-use categories: natural areas, agricultural areas, residential areas, recreational areas and industrial areas. Generally, the clean-up standards for the last four categories are obtained by human exposure modelling. Soil clean-up standards for polychlorinated biphenyls (PCBs) and dioxins (PCDD/Fs) were derived in this framework.

The procedure for derivation of soil clean-up standards for PCBs and PCDD/Fs included following steps:

- Determination of background concentrations in soils of unpolluted areas;
- Information collection of the considered pollutants: toxicology, physico-chemical properties, occurrence in the environment, human exposure and behaviour in the soil;
- Modelling using the human exposure model VLIER-HUMAAN;
- Application of additional adjustments criteria;

Because there are a significant number of PCBs and PCDD/Fs impacted sites across Flanders resulting from a wide range of industrial activities, agricultural practices and usage practices, there is a real need for appropriate soil clean-up standards for PCBs and PCDD/Fs. However setting suitable soil quality objectives for PCBs and PCDD/Fs is a very complex matter.

From toxicological studies with individual PCB congeners it is known that some PCBs elicit a different toxicological mode of action and metabolic behaviour. PCBs and PCDD/Fs show different physico-chemical properties resulting in a different environmental behaviour. If one observes the ratios of dioxin-like PCBs and PCDD/Fs through the food chain one will notice a remarkable shift: while PCDD/Fs are dominant in the different environmental compartments, dioxin-like PCBs are in biological matrices[1]. Nevertheless PCBs and PCDD/Fs show very similar behaviour in the soil. Both were found to be quite immobile in soils showing strong sorption to organic matter and therefore, would not be leached out by rainfall and infiltration, though lateral transport during surface erosion of the soil can occur. Due to their strong sorption to organic matter, diffusion processes and evaporation from the soil are limited. Soil is a typical accumulating matrix with long memory: Inputs received in the past will remain. Due to the very long half-life of PCBs and PCDD/Fs in subsurface soils, there is hardly any clearance. In surface soils half-life of those persistent chemicals can be substantially shorter as a consequence of photochemical decomposition immediately following the contamination[2].

Methods and Materials

The human exposure multimedia model VLIER-HUMAAN is used in the framework of the Flemish soil decree for the derivation of soil clean-up values for the indicator-PCBs, dioxin-like PCBs and PCDD/Fs in the solid phase. It is based on the formulas used in the Dutch HESP-model[3] with some additions and modifications. These changes relate mainly to chemical-specific parameters and to land-use scenarios (agricultural, residential, recreational and industrial areas).

For each land-use type soil clean-up values are derived. The calculations of the exposure model are grouped in three layers. The first layer calculates the distribution of the contamination in the soil. The second layer covers the migration to other compartments and the concentrations in these compartments and corresponding media (air, water, vegetables …). In the third layer exposure of human beings (children, adults) in contact with the considered environmental compartments is calculated.

Total exposure for non-carcinogenic compounds is divided by the TDI resulting in a risk index (RI), accounting for differences between oral and respiratory intake. Children and adults are considered separately. The soil clean-up value is primarily derived on the level in the soil corresponding with a calculated RI equal to 1. Total exposure comprises exposure from the polluted site together with background exposure from undefined sources. Dietary background exposure is taken into account proportionally in each exposure scenario.

Dioxin-like PCBs and PCDD/Fs show a rather specific and complex behaviour. The atmosphere is an important transport medium. Atmospheric background deposition of dioxin-like PCBs and PCDD/Fs contributes significantly to the concentration in vegetables and grass. Hence, consumption of contaminated fodder by cattle results in transfer of dioxin-like PCBs and PCDD/Fs to meat and milk. Since consumption of vegetables, meat and milk are important for human exposure to dioxin-like PCBs and PCDD/Fs in agricultural (vegetables, meat, milk) and residential areas (vegetables), the model VLIER-HUMAAN was modified for those two land-use categories:

- In order to accomplish appropriate estimations a mathematical correction was carried out with equal contributions of background deposition and blown up dust particles to the resulting concentrations in vegetables and grass. At normal soil concentrations, soil related processes like suspended soil particles and blown up dust contribute only slightly to the final concentration of dioxin-like PCBs and PCDD/Fs of contaminated vegetables and subsequent contamination of meat and milk. For relatively higher soil concentrations the soil related processes are becoming increasingly important.
- The concentrations of dioxin-like PCBs and PCDD/Fs in vegetables, meat and milk as calculated by the original model VLIER-HUMAAN differed significantly from the field observations. Consequently, in order to improve estimation of the concentrations of the dioxin-like PCBs and PCDD/Fs in these media, transfer factors in its simplest form, based on experimental data from the literature were used [4].

A similar approach was followed for calculating the soil clean-up values of the indicator-PCBs. For recreational and industrial areas human exposure via consumption of dairy products and meat of locally grazing cattle and consumption of home grown vegetables is not considered. So, mathematical corrections are not relevant since they do not have any impact on the other exposure pathways.

A number of PCB congeners show 'dioxin-like' toxicity (PCB77, PCB81, PCB126, PCB169, PCB105, PCB114, PCB118, PCB123, PCB156, PCB157, PCB167 and PCB189). The toxicological potency of these PCB congeners is similar to the PCDD/F congeners and related to that of 2,3,7,8-TCDD using Toxic Equivalency Factors (TEFs). In 1998, WHO established a revised TDI range of 1-4 pg WHO-TEQ/(kg bw day), which included dioxins, furans and the 12 dioxin-like PCBs. This approach obviously covers dioxin-like activities only, and does not address non-dioxin-like effects of PCB-congeners usually represented by the 7 indicator-PCBs (PCB28, PCB52, PCB101, PCB118, PCB138, PCB153, PCB180). Whether or not this approach is protective of non-dioxin-like PCB effects remains unclear. Unlike for the dioxin-like PCBs, a corresponding TEF concept cannot be applied to the non-dioxin-like PCB-congeners due the lack of major criteria. The most important shortcoming is the lack of a common mechanism of action that results in all different types of toxic outcomes. Additionally, analysis of indicator-PCBs in soils is much cheaper than that of dioxin-like PCBs and PCDD/Fs. A possible relationship between both in soils is currently under investigation. For these pragmatical and practical reasons it was opted to calculate soil clean-up values for dioxin-like PCBs and PCDD/Fs using a TDI of 4 pg WHO-TEQ/(kg bw day) as well as for indicator-PCBs using a TDI of 0,01µg/(kg bw day)[5].

An average daily dietary background exposure of 3,1 pg WHO-TEQ/(kg bw day) was taken into account for the derivation of dioxin-like PCBs and PCDD/Fs. For the indicator-PCBs the background exposure was ignored in the calculations since normal background exposure in Flanders is already at the same level or higher than the TDI. This implies that additional soil contamination could not be tolerated[6]. The average background deposition for dioxin-like PCBs and PCDD/Fs amounts to 6,37 pg WHO-TEQ/(m².day) and 7,23 pg WHO-TEQ/(m².day) for respectively agricultural and residential areas. For the indicator-PCBs an average background deposition of 2,59 ng/(m².day) was used for agricultural and residential areas[6].

Calculations for dioxin-like PCBs and PCDD/Fs were carried out using the average physico-chemical properties of 2,3,7,8-TCDD. PCB153 was selected as the most representative congener for the indicator-PCBs. Consequently, soil-clean-up values for the sum of the indicator-PCBs were derived taking average physico-chemical properties of this congener. Physico-chemical data (solubility (25 °C), vapour pressure (25°C), Henry coefficient, octanol-water partition coefficients, organic carbon-water partition coefficients, permeation through polyethylene and polyvinyl-chloride water pipeline, diffusion coefficients air and water) were calculated from a compilation of data collected from the literature.

Results and Discussion

The modelling results served as the basis for the final proposal for soil clean-up values. Table 1 gives an overview of the background values and clean-up values for indicator-PCBs, dioxin-like PCBs and PCDD/Fs in soil after rounding off. For the indicator-PCBs the initial modelling result for land-use agriculture (0,044 mg/kg dm) was below the background value. In order to avoid that sites would be assessed as polluted and included in the inventory of contaminated sites, this value had to be increased. For that reason the requirement that the lowest soil clean-up value is at least 5 times the detection limit and sufficient different from the normal background value according to standard measurement methods is used as general rule. This results in a soil clean-up value of 0,24 mg/kg dm for agricultural areas.

The initial calculations for the dioxin-like PCBs and PCDD/Fs in the land-use category agriculture were significant different from the normal soil background concentration. For the residential area one has to take into account increased transfer from soil to vegetables in case of soil concentrations comparable to the proposed soil clean-up values. Consequently, cultivation of home-grown vegetables with known elevated transfer of dioxin-like PCBs and PCDD/Fs should be avoided. Only cultivation of vegetables with known reduced transfer is advisable as far as measurements do not prove the opposite[7].

Table 1: Background values and clean-up values for indicator-PCBs, dioxin-like PCBs and PCDD/Fs in soil.

	Indicator-PCB's (total)	dioxine-like PCB's + PCDD/F's (total)
Background value	0,12 mg/kg dm	3 ng WHO-TEQ/kg dm
Clean-up values		
Agricultural area	0,24 mg/kg dm	9 ng WHO-TEQ/kg dm
Residential area	0,91 mg/kg dm	65 ng WHO-TEQ/kg dm
Recreational area	2,57 mg/kg dm	260 ng WHO-TEQ/kg dm
Industrial area	10,44 mg/kg dm	1600 ng WHO-TEQ/kg dm

The soil clean-up values are derived for a standard soil containing 2% organic material and 10 % clay. For soils with a different organic matter content, correction of the listed soil clean-up values is required. For that reason, the impact of the organic matter on the calculation results was also checked. A change of the organic matter content from 1 to 10 % did hardly change the calculated soil clean-up values neither for the indicator-PCBs nor for the dioxin-like PCBs and PCDD/Fs. This is mainly due to the strong preference and sorption of those chemicals to the organic material in the soil. Consequently, corrections for the organic matter content of the soil were not proposed.

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