Framework and baseline application of a screening methodology for multimedia assessment of human exposures at dioxin-contaminated sites

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Introduction

Polychlorodibenzo-p-dioxins and polychlorofurans (PCDD/Fs), mainly hepta- to octachlorinated furans, have been formed in large amounts in chlorophenolic (CP) wood preservatives used at sawmills in Finland (1-3). Tens of sawmill sites have been initially remediated. However, the residual problem of associated PCDD/Fs has been gradually recognised. In such cases there has been a great need for environmental exposure and risk assessment and for the requisite information and methodologies.

There are few site-specific multimedia models available addressing exposures and risks to PCDD/Fs. General dioxin exposure and risk models (e.g., 4-6) have to be modified to account for site-specific factors, e.g. for the areal distribution of contamination and for the relative contribution of a site to overall exposures and risks. On the other hand, common models for contaminated site assessment are not directly applicable to PCDD/Fs due e.g. to their mixture identity. Only a few models have been applied to wood preservation sites (7).

The aim of the present work is to delineate a systematic framework for quantitative assessment of human multimedia exposures to site-derived PCDD/Fs for screening purposes.

Methods

A framework is presented for the assessment of toxicological risks to man caused by local contamination of soil, sediment and related materials, specifically at sawmill sites. The framework is initially tested in a case of a contaminated sawmill area projected for housing. Emphasis is put on risk identification, spatial scales of risks, risk comparisons, data and model uncertainties, and relationships with cleanup criteria definitions.

The framework is a modification of previous approaches to site-specific dioxin risk assessment (7, 8). Novel features include additional consideration of variation, the account for background risks, linked with the definition of the scope (boundaries) of assessment, and the attention to the interface of risk analysis with risk management criteria. The approach does not include formal quantitative uncertainty analysis or more extensive modeling of site exposures and risks.

PCDD/F intake values in the various exposure routes are calculated for sensitive land use by using environmental concentrations at the site or at analoguous cases (after modification); common transfer models (Table 1); data and estimates of the rate, frequency and duration of exposure; and assumptions of the fraction of contaminated media. Specifically, it is assumed as a worst case that soil heavily contaminated and treated for CPs is spread in site vicinity (not on cultivation areas) with no breakdown. Exposures are estimated separately for children (averaging ages 2-12 yrs) and adults. In the baseline application, point estimates and ranges are used. Effect levels are accounted for by published data and models of tumorigenicity, immunotoxicity and reproductive/developmental toxicity (initially by TDI based on the latter).

Results and discussion

Starting with observed concentration levels of up to 3 ng ITEQ g^{-1} (dw) in remaining sawmill soil and nearby lake sediment, and 2 000 ng ITEQ g^{-1} (dw) in heavily contaminated soil assumed to be spread on site after treatment of CPs, we get rough conservative estimates of total exposures (time-averaged chronic daily intake values) of around 3, 80 and 2 pg ITEQ kg^{-1} d^{-1} for adult residents, children and workers, respectively (Table 1). Estimates based on central statistics are much lower, reflecting the effects of 'cascading' variation and uncertainty.

Using the models inversely, case-specific options to protect human receptors from exposures above a specified level may be identified, and analysed quantitatively and comprehensively in a simple way. To reduce exposures of all receptors with high certainty (based on maximum values) to a TDI of 5 pg kg^{-1} d^{-1}, soils with PCDD/F > above 1 ng ITEQ g^{-1} may be treated (or removed/isolated). This soil concentration is twice the recent health-based proposals for trigger (intervention) values of PCDD/Fs in residential soil (9). With stricter TDI values the cleanup criteria and different model assumptions would be correspondingly lower.

General conclusions of the methodology and results of the assessment at this stage include:

1 By *maximum* estimates and conservative assumptions, *intake values for children exceed TDIs; plausible values are orders of magnitude lower* due to compounding uncertainty.
2 The framework is useful for gauging the importance of exposure routes and mechanisms and for pinpointing data and modeling needs.
3 The framework can be used to *derive health-protective soil quality criteria* (intervention).

Specific conclusions pertaining to the significance of and information on the various exposure routes include:

1 *Soil ingestion is among the dominant exposure routes* in the residential scenario considered.
2 *Dermal absorption* is a notable route when high skin permeation is assumed, but model validity is questionable for the isomers and forms of occurrence (in particles) (cf. 26).
3 *Airborne PCDD/Fs may be a notable source* for cleanup workers only, but data and models for quantitative assessment of this route are as yet insuffient.
4 The rough estimates of site-derived exposure via homegrown biota are relatively small.

Development needs and options of the assessment approach include:

1 Consideration of *additional factors* may increase exposure estimates (e.g., new routes and transfer mechanisms) or decrease them (e.g., breakdown, translocation in cleanup).
2 The *temporal and spatial distributions of exposures may be considered more extensively* by exposed time fraction and contaminated matter fraction, to assess site contributions.
3 *Isomer-specific assessment* is desirable particularly for the HpCDFs making up most of the ITEQs, but would require accounting for the environmental fate properties of these isomers.

4 Additional *measurements* are needed of PCDD/F levels in targeted exposure media and resultant body burdens to *verify* and develop the approach and models.
5 The *approach may be extended* to effects models, to probabilistic risk and uncertainty assessment, and analysis of risks in different cleanup options.
6 The assessment approach can be implemented e.g. as a spreadsheet model.

Table 1. Estimates of plausible worst-case and median levels of human chronic exposures to PCDD/Fs at a sawmill site contaminated by chlorophenolic wood preservatives in a sensitive residential scenario (small houses, gardens). (In parentheses, estimates for children 2-12 yrs).

Media (exposure route or receptor)	PCDD/F Med-max conc. at site[1] (normally ng ITEQ g^{-1} dw)	Intake of contaminated media by exposure route[2]									Total chronic daily intake[5]
		oral			inhalation			dermal contact			
		intake of media g d^{-1} fw)	expo. time[3] (%)	con-tam.[4] (%)	intake of media (m^3 d^{-1})	expo. time[3] (%)	con-tam.[4] (%)	intake of PCDD/Fs (pg d^{-1})	expo. time[3] (%)	con-tam.[4] (%)	CDI_{Max} (pg ITEQ $kg^{-1}d^{-1}$)
- soil left	.02-3.7	.002-.02[6]	40[7]	30				.0004-.4[8]	40[7]	30	1.6
at site		(.01-.2)[6]	(80)[7]	(50)				(.005-.02)[8]	(80)[7]	(50)	(8.5)
- soil to treat	300-2000	.0002-.002[9]	0.1[10]	90				.001-.004[11]	.1[10]	100	.15
(worker expo.)											
- treated soil	(30-200)[12]	.002-.02	4[13]	20				.01-.04[8]	4[13]	20	.65
(applied)		(0.01-0.2)	(20)[13]	(30)				(.005-.02)[8]	(20)[13]	(30)	(66)
- edible plants	(.02-3	200-400[15]	30[16]	30							.2
from site	pg g^{-1})[14]	(100-200)	(30)[16]	(30)							(1.4)
- air/ambient	(.001-.2				4-5[18]	40[19]	20				.001
(dust)	pg m^{-3})[17]				(3-9)[18]	(80)[19]	(50)				(.025)
- air/occupat.	(3-6 ng m^3)[20]				18-22[21]	.1[10]	80				1.8
(solids treat.)											
- lake sed.	.02-3							.01-.04[8]	.1[22]	30	.2
(bathing)								(.005-.02)[8]	(.2)[22]	(50)	(1)
- lake water	(.2-30	1	.5[24]	30				.1-.4[8]	.5[22]	30	.001
(bathing)	ng l^{-1})[23]	(10)	(.5)[24]	(50)				(.05-.2)[8]	(.5)[22]	(50)	(.04)
- edible fish	(.07-.12	45-90[26]	100	30							.5
(pike/burbot)	pg g^{-1}fw)[25]	(22-45)[26]	(100)	(30)							(.8)
Total, adult											3.1
total, child											(78)
total, worker											2.0
Background[27]											1.5
TDI[28]											5

[1]From (10) if not stated otherwise; [2]Cumulative exposure during the respective age period; [3]Fraction of days exposed (a function of exposure frequency and duration); [4]Fraction of media contaminated (a function of the spatial range and type of receptor activity); [5]Converting dw to fw based values with a water content of 90 % for plants and 50 % for soil, and assuming a body weight of 60 kg for an adult and 20 kg for a child; [6]From (11), (12) and (13); [7]Exposure daily of children and bidaily of adults during snowless period; [8]Based on an exposed skin area of 0.05 m^2, soil loading of 0.02-0.2 g m^{-2} (14, 15) and skin permeation models in (16) and (17); [9]90 % protection (10 % of normal intake); [10]0.1 % of adult life (1 mo) spent handling the compost; [11]90 % protection of exposed skin; [12]90 % dilution with clean materials of the soil to be composted for CPs (no PCDD/F removal assumed); [13]Exposure twice a week (of children) and twice a month (of adults) during snowless period to compost spread in near-site soils; [14]Transfer to vegetables, using a transfer factor of 0.1 % cited by (5), and concentrations in remaining soil; [15]From Finnish data on average vegetable intake of adults (18), assuming 50 % rate for children; [16]Homegrown fraction, mean of the range given by (19); [17]From data in (20) and from soil-vapor and soil-aerosol model of Jury applied to TCDD in (21), assuming proportional total air and soil concentrations; [18]From weight-specific active breathing rates for children and adults given in (22), based on 6- and 4-hr daily stay outdoors, respectively; [19]Estimated time outdoors (as in footnote 7); [20]Based on air dust data from cleanup works in (22) and (7), assuming proportional soil (compost) and air concentrations; [21]Twice the normal active breathing rate for adults (cf. footnote 20) and 8-hr workday; [22]Contact during 20 % (for adult) and 50 % (for child) of bathing time (cf. footnote 24); [23]1 % of sediment PCDD/Fs is distributed by resuspension to corresponding volume of surface water swallowed or bathed in; [24]1-hr bathing bidaily for adults and children in summer months; [25]Based on 10 % of the muscle concentrations measured in a river contaminated by PCDD/Fs from CP production (23); [26]From Finnish average daily fish consumption (includes frequency of consumption) (18), assuming 50 % values for children and 4-fold values for active recreational fishers' households and 20 % cooking loss given by (24); [27]Based on Finnish food basket and food PCDD/F level surveys (18); [28]Nordic TDI guideline value (based on weekly value of 35 pg^{-1} kg^{-1} d^{-1}).

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