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Polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs) soil contamination in Lausanne, Switzerland: Combining pollution mapping and human exposure assessment for targeted risk management☆

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A R T I C L E I N F O

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A B S T R A C T

In December 2020, high soil concentrations of polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs) were discovered across large parts of Lausanne, Switzerland. Concentrations reached up to 640 ng $TEQ_{WHO-2005}$/kg dry weight. The most likely source was a former municipal waste incinerator. A three-step, multidisciplinary approach to human health risk assessment was conducted to determine the potential population exposure to PCDD/Fs and identify appropriate preventive measures. First, exposure scenarios were developed based on contaminated land uses. Second, the toxicological risks of different scenarios were evaluated using a toxicokinetic model estimating increases in blood serum PCDD/F concentrations over background concentrations from the general population's food consumption. Third, a detailed geostatistical mapping of PCDD/F soil contamination was performed. Stochastic simulations with an external drift and an anisotropic model of the variogram were generated to incorporate the effects of distance from emission source, topography, and main wind directions on the spatial distribution of PCDD/Fs in topsoil. Three main scenarios were assessed: i) direct ingestion of soil by children in playgrounds; ii) consumption of vegetables from private gardens by children and adults; and iii) consumption of food from livestock and poultry raised on contaminated soil. The worst exposure scenario involved the consumption of eggs from private hen houses, resulting in PCDD/F concentrations in serum an order of magnitude higher than might normally be expected. No relevant increases in serum concentrations were calculated for direct soil ingestion and vegetable consumption, except for cucurbitaceous vegetables. Combining mapping and exposure scenario assessment resulted in targeted protective measures for land users, especially concerning food consumption. The results also raised concerns about the potential unsafe consumption of products derived from animals raised on land with PCDD/F concentrations only moderately over environmental background levels.

1. Introduction

Dioxins (polychlorinated-dibenzo-*p*-dioxins, PCDDs) and furans (polychlorinated-dibenzofurans, PCDFs), abbreviated together as PCDD/Fs, are a family of 210 aromatic organochlorine congeners. Exposure to dioxins and furans first attracted public attention after the 1976 Seveso incident, when more than 30 kg of 2,3,7,8-tetrachlorodibenzo-p-dioxin (TCDDs) were released into the environment

☆ This paper has been recommended for acceptance by Charles Wong.

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(Mocarelli, 2001). PCDD/Fs are ubiquitous in the environment, mainly originating from the unintentional emissions of anthropogenic processes, particularly older-generation municipal solid waste (MSW) incinerators, household or hospital incinerators, paper-pulp chlorine bleaching processes, pesticide production, and some natural phenomena (e.g., forest fires, eruptions) (Schmid et al., 2005). PCDD/Fs are formed during combustion and thermal processes involving carbon and chlorine and are released into the atmosphere and transported by combustion fumes. They are then deposited on topsoil, where they tend to adsorb on organic matter due to their low mobility and lipophilicity. In soil, PCDD/F half-lives are estimated at between 25 and 100 years, depending on the congeners and the soil's physicochemical properties. Their presence in soil results in terrestrial food-chain contamination—the main source of PCCD/F exposure in the general population (Lorber et al., 2000). Several studies have reported on soils contaminated by PCDD/Fs and the subsequent exposure of nearby populations, such as in Sweden (Aberg et al., 2010), Italy (Di Lorenzo et al., 2015), and the USA (Kruse et al., 2014).

The main human contamination route for PCCD/Fs is oral (Knutsen et al., 2018). Following absorption, PCDD/Fs mainly accumulate in the liver and adipose tissue due to their binding to hepatic enzymes and their high lipophilicity, respectively. Known human health effects include transient liver damage (affecting detoxification enzymes such as cytochromes P450: CYP1A1, CYP1A2, and CYP1B1), peripheral nerve damage, reproductive and developmental toxicity (Linda S. Birnbaum, 2017), and cancer (ATSDR, 2006). Other effects are suspected, namely on the immune system (affecting cytokines and interleukins 1 and 6) and type 2 diabetes (Knutsen et al., 2018). All these health effects may be due to PCDD/Fs' persistent activation of the aryl hydrocarbon receptor (AHR) and the consequent homeostasis imbalance (L. S. Birnbaum, 2017; Safe et al., 2018).

Two congeners are currently recognized as class I carcinogens (carcinogenic to humans) by the International Agency for Research on Cancer (IARC, 2012): 2,3,7,8-TCDD and 2,3,4,7,8-PeCDF. Suspected cancers include respiratory tract cancers, prostate cancer, and certain types of lymphomas and sarcomas (IARC, 2012). A recent meta-analysis observed significant associations between environmental TCDD concentrations and certain cancers (esophageal, larynx, and kidney cancer, non-Hodgkin's lymphoma, myeloma, and soft tissue sarcoma) (Xu et al., 2016). TCDD's carcinogenic effects are due to its binding to the intranuclear AHR, which alters the transcription of messenger RNA encoding cellular response enzymes (Popp et al., 2006). Thus, TCDD is an indirect (non-genotoxic) threshold carcinogen that primarily promotes tumorigenesis by inhibiting cell communication and apoptosis (Knerr and Schrenk, 2006). The carcinogenicity of other congeners remains suspected but has not yet been demonstrated due to the complexity of exposure scenarios and the latency period before the onset of symptoms associated with low-dose chronic exposure. However, the thresholds identified so far for cancer are significantly higher than for reprotoxic effects. Based on a non-linear dose-response model, Simon et al. suggested a threshold dose for cancer of 100 pg TEQ/kg bodyweight (bw)/day, which is 300 times the current tolerable daily intake (TDI) (Simon et al., 2009).

The most sensitive critical effect in humans is reproductive toxicity (Knutsen et al., 2018), specifically sperm quality. Several studies have reported PCDD/Fs' endocrine and reproductive disruption during childhood and pre-adolescence. Mocarelli et al. evidenced the significant effect of TCDD concentrations in the blood serum of 1-to-9-year-old boys on their adult sperm quality (Mocarelli et al., 2008). More recently, Minguez-Alarcón et al. (Mínguez-Alarcón et al., 2017) showed associations between lower sperm concentrations and higher blood serum levels of TCDD alone and PCDDs. Indeed, this was the reference study used by the European Food Safety Authority (EFSA) in 2018 to establish a No Observed Adverse Effect Level (NOAEL) for PCDD/Fs of 7.0 pg TEQ/g serum lipid in 9-year-olds and to derive a tolerable weekly intake of 2 pg TEQ/kg bw (Knutsen et al., 2018).

In December 2020, soil surveys were carried out before the redevelopment of a plot of land in the city of Lausanne, Switzerland. In addition to common pollutants (metallic trace elements, polycyclic-aromatic hydrocarbons), PCDDs, PCDFs, and polychlorinated biphenyls (PCBs) were analyzed in two composite samples. Unexpectedly high PCDD/F concentrations of 96 and 107 ng $TEQ_{WHO\text{-}2005}$/kg dry weight (dw) were measured, alerting authorities and leading to successive measurement campaigns in 2021.

An environmental consulting firm researched putative historical sources of this contamination, identifying two industrial plants: a former MSW incineration plant and a thermoelectric plant. Both were able to generate emissions consistent with the observed congener concentration profiles. However, analysis of the potential emission scenarios showed that the thermoelectric plant's different fuel sources (including coke/coal and fuel oils) were insufficient to explain the observed soil contamination. Ruling out the thermoelectric plant confirmed the MSW incineration plant as the main source of contamination.

The incineration plant treated approximately 48,000 tonnes of waste/year from its commissioning in 1958 to its decommissioning in 2006. Several transformations were carried out over the years, including installing a wet-flue gas cleaning system (electro-dynamic Venturi) in 1982 and beginning electrostatic-precipitator ash treatment in 1997. It was, therefore, likely that PCDD/F emissions occurred primarily before the late 1990s. The incineration plant was located in a narrow valley in the upper part of the city, probably contributing to limited atmospheric dispersion and resulting in local deposition of the combustion particles.

The elevated PCDD/F concentrations measured in soils, with several exceeding regulatory values, raised questions about local inhabitants' exposure to PCDD/Fs and their potential deleterious health effects. This highlighted the need for a detailed risk assessment, which was duly commissioned by the regional environment and public health authorities. Access to local playgrounds in contaminated areas and the consumption of meat from contaminated land were restricted pending the risk assessment's results.

In performing and documenting our health risk assessment to support the authorities' decisions, we aimed to prioritize risk situations regarding topsoil PCDD/F contamination and identify appropriate protective measures to communicate to Lausanne's population. The rationale was to combine an exposure assessment based on expected soil uses with geostatistical mapping of PCDD/F contamination levels.

2. Materials and methods

2.1. Measurement of contaminated soils

Soil samples were progressively collected working outwards from the initial contamination area by selecting plots: (1) without soil disturbances since the presumed end of PCDD/F emissions by the MSW incinerator (i.e., no excavations or soil inputs) and (2) on public sites, especially with high potential for exposure (e.g., playgrounds). A composite sample of the first 5 to 20-cm layer of topsoil was obtained by mixing 5 to 16 grab samples taken across each plot. After their arrival at the laboratory, the soil samples were dried at 40 °C until their weight remained constant (2 days maximum), they were then crushed and sieved with a 2 mm mesh sieve. Overall, 126 undisturbed soil samples were selected for subsequent mapping of PCDD/F levels. These levels ranged from 1.72 to 640 ng TEQ/kg dw, with an average of 93 ng TEQ/kg dw and a median of 47 ng TEQ/kg dw. Samples were taken and analyzed by a private laboratory using the DIN 38414-24 German standard methods for soil samples. Briefly, these underwent accelerated solvent extraction and were analyzed using High-Resolution Gas Chromatography coupled with High-Resolution Mass Spectrometry (Zhou and Liu, 2018). Soil concentrations were calculated using the World Health Organizatio n's 2005 toxic equivalent (TEQ) system ($TEQ_{WHO\text{-}05}$). All PCDD/F concentrations are expressed in TEQ in this manuscript.

2.2. Exposure assessment

Exposure assessments were based on expected soil uses that might lead to higher dioxin uptake than a regular diet, available on the market and meeting regulatory standards. We focused on food from contaminated soils and the oral ingestion of soil, with the latter being the exposure scenario used by Swiss regulations to determine PCDD/F limit values in soil. Accordingly, PCDD/F uptake via the skin, inhalation, and water consumption was considered negligible due to their low volatility and high hydrophobicity (Knutsen et al., 2018). Three exposure scenarios were considered relevant to the Lausanne context:

- direct ingestion of soil by children (hand-to-mouth reflex) (scenario S1);
- consumption of vegetables grown on contaminated soils (S2); and
- consumption of animal products reared on and/or fed with forage harvested on contaminated soil (S3).

PCDD/Fs readily accumulate in animal fat, and concentrations above recommended food consumption thresholds have been reported for soil concentrations ranging from 5 to 20 ng TEQ/kg dw (Amutova et al., 2021; Weber et al., 2018; Weber et al., 2019). The consumption of animal products originating from Lausanne mainly concerned eggs from private hen houses, which could have been consumed daily by the hens' owners and their families. Additionally, Lausanne municipality owns around 100 suckling ewes and their lambs (*Roux du Valais* and *Miroir* breeds) that ensure the maintenance of 30 ha of grassland plots throughout the city and a specific population of *Mangalica* pigs bred locally for slaughter. Due to the minimal consumption of local chicken and *Mangalica* pork, the exposure assessment related to animal products focused solely on egg and mutton consumption.

Predicted doses for the different exposure scenarios (S1–3) were computed as a function of soil concentration levels and activity intensities (e.g., days in playground/year, frequency of vegetable consumption). Appendices A to C detail the models and parameters used to calculate the exposure doses for scenarios S1–S3, respectively.

2.3. Estimation of human PCDD/F levels in blood serum

The EFSA's current recommended TDI is 0.3 pg TEQ/kg bw/day (Knutsen et al., 2018). This is the theoretical value to make a 9-year-old's blood serum concentration correspond to the NOAEL reported for a reprotoxic effect in young adolescents (Mínguez-Alarcón et al., 2017). In Switzerland and most European countries, average dietary intake is above this TDI: in 2008, it was estimated at 0.6 pg TEQ/kg bw/day (BAG, 2008). Although intake from regular food is probably slightly lower today—due to the slow decrease in environmental PCDD/F concentrations—it is unlikely to be lower than EFSA's TDI. We therefore decided to compare our exposure scenarios using the relative increases in expected blood concentrations obtained via toxicokinetic modeling (versus the level expected from regular food consumption).

A concentration and age-dependent model (CADM) was used to predict PCDD/F blood concentrations. This was initially developed by Carrier et al., (1995) to predict TCDD levels in the body and was successively adapted by Aylward et al., (2005) and Ruiz et al., (2014). The model accounts for age-related changes in body weight, PCDD/F sequestration in the liver, and includes PCDD/F intake via breastfeeding. The two endpoints considered were PCDD/F levels after chronic exposure up to 9 and 35 years old, corresponding to the age at which the lowest NOAEL was observed and the maximum age in the CADM, respectively. The reference situation involving regular food consumption, adjusted for the Swiss population's estimated dietary intake and concentration levels in breast milk, gave expected blood concentrations of 7.0 and 10.4 pg TEQ/g lipid at 9 and 35 years old, respectively. Table 1 presents the simulation parameters. Unless specified otherwise, the default parameters were those used in EFSA's TDI calculation (Knutsen et al., 2018).

Table 1
CADM model simulation parameters.

Parameter	Swiss population	EFSA simulation
Breastfeeding duration	12 months	12 months
Breastmilk concentration	5 pg/g fat	6 pg/g fat
Dietary intake	0.6 pg/bw day[a]	0.5 pg/bw day[a]
Body weight	Adjusted for age according to CADM	

[a] To be doubled for children, who consume more in relation to their body weight.

2.4. Soil contamination mapping

Exposure assessments and corresponding blood level estimates were used to determine soil concentration intervals of interest, defined as the range of concentrations for which similar protective measures are deemed appropriate (e.g., restricting the use of private garden plots of the same interval). PCDD/F soil contamination samples (n = 126) were then mapped to the soil dataset using a non-linear geostatistical method (Chilès and Delfinger, 2012; Cressie, 2015; Goovaerts, 1997). A set of 1000-block conditional stochastic simulations of PCDD/F concentrations were generated using Geovariances' Isatis© software and then post-processed to calculate the local probability (for each model grid cell) that a PCDD/F grade belongs to each of the concentration intervals considered. The highest probability calculated was assigned to the targeted grid cell, and calculations were repeated for each grid cell to produce a map of the most probable concentration interval over the whole study area.

Exploratory data analysis showed that PCDD/F grades (after Gaussian transformation) were lower the further their soil sampling site was from the location of the former MSW incinerator. Similarly, soil data highlighted that the smaller the altitude difference between the top of the incinerator's chimney and the soil sampling plot, the higher the soil contamination level. Simulations with an external drift were thus generated using a variable combining the horizontal and vertical distances from the chimney top to the sampling point, providing exhaustive secondary information on the whole study area. These simulations were carried out using an anisotropic model of the residue variogram, with a longer range (2300 m) in the north-westerly direction (45°) of dominant winds than in the south-westerly direction (135°; 1500 m), which was consistent with the incinerator's airborne dispersion of gaseous and particulate PCDD/Fs.

3. Results

3.1. Soil measurements

The 126 soil PCDD/F measurements performed across the Lausanne area, between January and August 2021, showed that the varying soil contamination concentrations were not localized but extended to large parts of this city of 140,000 inhabitants. 29% of samples were collected on unused grass borders, 28% near sports fields or playgrounds, 12% in forest areas, 10% near vegetable gardens, 9% in parks, and 12% in other areas (e.g. schoolyard, agricultural area). PCDD/Fs concentrations found ranged between 0.1 and 640 ng TEQ/kg dw (mean: 89.8, median: 46.6; interquartile range: 97).

As Fig. 1 shows, pentachlorinated-dioxin (1,2,3,7,8-PeCDD) and 2,3,4,7,8-pentachlorodibenzofuran (2,3,4,7,8-PeCDF) were the most abundant of the 17 congeners measured, with TEQ contributions of 39% and 11%, respectively.

3.2. Exposure assessment

Average daily PCDD/F intakes (absolute doses) for exposure

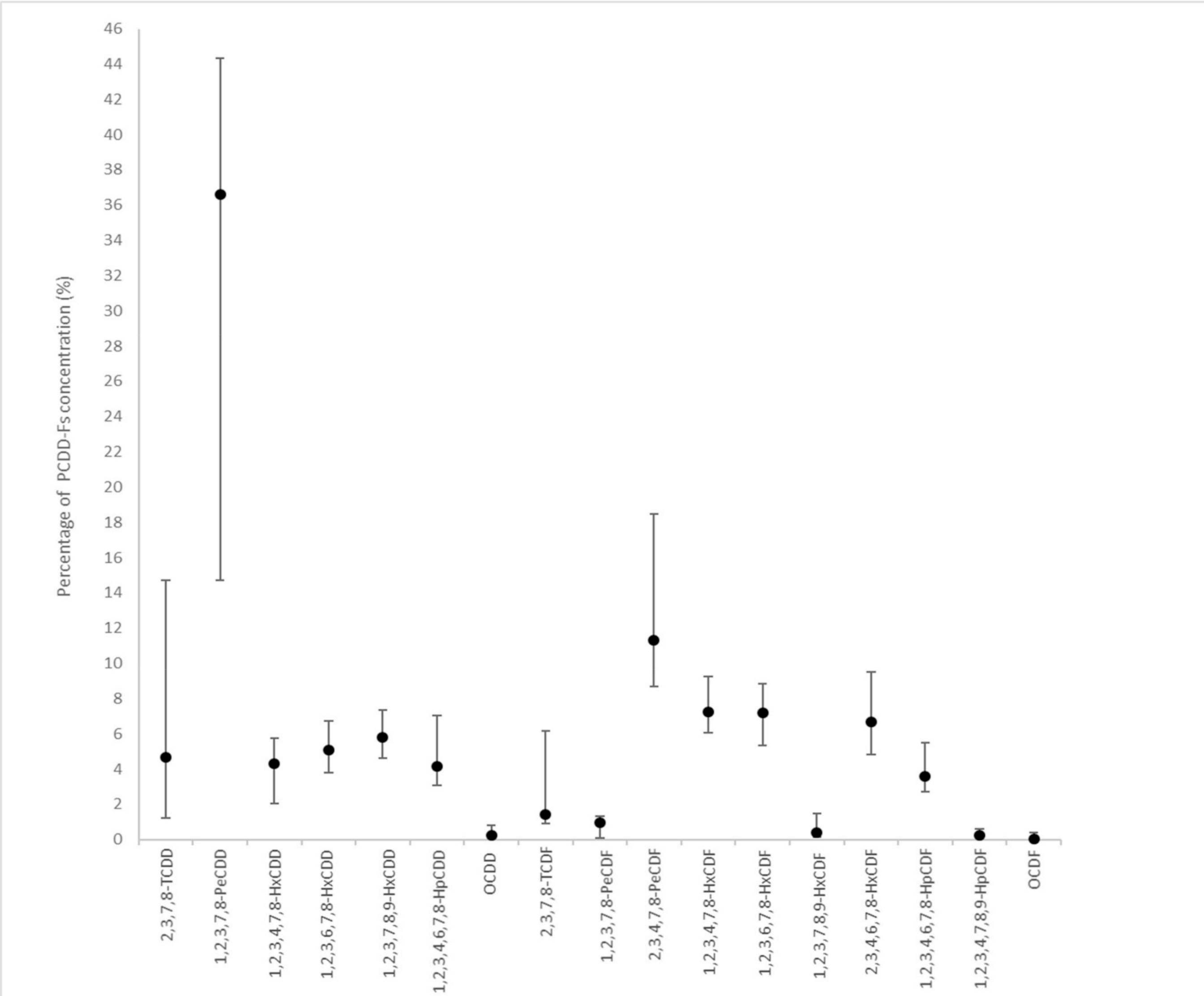

Fig. 1. Congener profiles (%) and variation among the soil samples according to their concentrations in $TEQ_{WHO-2005}$/kg dry weight. Black circles are mean values, and the error bars represent minimum and maximum values.

scenarios S1–S3 were computed for different soil concentration intervals of interest. Appendices A–C detail these results for concentrations of 20 and 100 ng TEQ/kg dw, corresponding to the current Swiss investigation and remediation limits, respectively.

For direct soil ingestion by children (exposure scenario S1), Fig. A1, the TDI on contaminated soils was exceeded at PCDD/F concentrations ≥100 ng TEQ/kg dw. At these concentrations in soil, PCDD/F levels above twice the TDI were calculated for infants frequenting parks and gardens more than 100–150 days/year. For the other exposure scenarios, TDIs were calculated at less than twice the target value, except when soil concentrations were ≥300 ng TEQ/kg dw.

Regarding vegetable consumption (exposure scenario S2), frequently eating cucurbitaceous vegetables led to PCDD/F TDIs (Figs. B1 and B.2) being exceeded for infants and children, even at soil concentrations of 20 ng TEQ/kg dw with consumption above 50 portions of 100 g/year. In adults, TDIs were exceeded at soil concentrations above 50 ng TEQ/kg dw and consumption frequencies >100 portions of 100 g/year. At concentrations >100 ng TEQ/kg dw and consumption frequencies above 200 days/year, TDI values could be exceeded by more than 3 and 10 times in adults and children under 4 years old, respectively.

Tubers (e.g., potatoes) and root vegetables (e.g., carrots) are in direct contact with soil, resulting in PCDD/F loading, but to a lesser extent than cucurbitaceous vegetables: unpeeled carrots contain about one-third of cucurbitaceous PCDD/F levels. Hence, calculated TDIs were exceeded for infants and children consuming over 90 portions of 100 g of unpeeled carrots/year growing in PCDD/F soil concentrations above 20 ng TEQ/kg dw (Fig. B3). TDIs were exceeded for adults consuming over 100 servings/year of unpeeled carrots growing in PCDD/F soil concentrations >100 ng TEQ/kg dw (Fig. B4). Peeled carrots were calculated to contain about one-tenth the PCDD/F level of unpeeled carrots. TDIs were only calculated to be exceeded for young children at soil concentrations of 200 ng TEQ/kg dw (Fig. B4). Leafy vegetables (lettuce) and beans accumulate negligible amounts of PCDD/Fs compared to root vegetables (ADEME and INERIS, 2017); therefore, these vegetables were not investigated further.

Our animal product consumption (exposure scenario S3) exposure assessment suggested that average egg and mutton consumption might induce contamination levels significantly beyond TDIs for children and adults. In terms of yearly chronic doses, however, calculated exposure remained negligible with local mutton because of the expected low frequency of consumption (Fig. C4). On the contrary, with eggs, TDIs were surpassed for both children and adults as soon as consumption was more than a few dozen eggs/year at PCDD/F soil concentrations of 20 ng TEQ/kg dw (Fig. C1). Levels ten times the TDI were calculated at PCDD/F soil concentrations of 50 ng TEQ/kg dw for consumption frequencies of ≥50 eggs/year for young children and >250 eggs/year for adults.

3.3. Estimation of PCDD/F levels in human blood serum

An estimate of the expected relative increase in PCDD/F level (C_1–C_0/C_0, expressed as a %) in serum was calculated for the three exposure scenarios (S1–S3). The direct soil ingestion scenario (S1) led to a slight increase in calculated blood serum concentrations compared to the general population. Indeed, as Fig. 2 shows, a 10%–20% increase in serum concentration was expected at soil concentrations >100 ng TEQ/kg dw and exposure frequencies >100 days/year. Increases >20% were only calculated at concentrations >250 ng TEQ/kg dw and almost-daily exposure frequencies (>200 days/year).

Regarding vegetables, consuming cucurbitaceous vegetables presented the greatest increase in concentrations in blood serum. In terms of relative increases in body burden, Fig. 3a and b shows very significant increases (>100%) in expected PCDD/F concentrations in blood serum. At soil concentrations >150 ng TEQ/kg dw and consumption >60 portions of 100 g/year, expected concentrations could be multiplied by 2–3 in children and by 2 in adults. At consumption above 30 portions of 100 g/year, a significant increase (>50%) in PCDD/F concentrations in blood serum was expected at soil concentrations between 20 and 100 ng TEQ/kg dw, corresponding to Swiss legislation's soil investigation and remediation thresholds, respectively (SwissFederalCouncil, 1998).

Regarding other vegetables, only the consumption of unpeeled root vegetables was calculated to lead to a possible notable increase in the expected PCDD/F blood serum concentration for the greatest consumption levels (Fig. 3c–f). At PCDD/F soil concentration levels >150 ng TEQ/kg dw and consumption >80 portions of 100 g/year, expected concentrations could increase by about 50% in children and adults (Fig. 3c and d). At soil concentrations <100 ng TEQ/kg dw, the expected increase in serum concentration remained low (<20%), even when consuming >40 unpeeled carrot portions of 100 g/year. For peeled carrots, the expected increase in PCDD/F blood serum concentrations was low, even at consumption frequencies >120 portions of 100 g/year (Fig. 3e and f). At PCDD/F soil concentrations >150 ng TEQ/kg dw and a peeled carrot consumption >60 portions of 100 g/year, we only calculated an expected increase of 10% in children and adults.

Of all the scenarios studied, egg consumption led to the highest estimated PCDD/F exposure. This was due to the bioaccumulation of PCDD/Fs in animal lipids (i.e., egg yolk) and the high egg consumption rate expected of private poultry owners (who consume most of their production). As Fig. 4 shows, egg consumption can lead to significant increases in PCDD/F blood serum concentrations. At PCDD/F soil concentrations >150 ng TEQ/kg dw and almost-daily egg consumption, expected concentrations were calculated at 10 times greater in children and 6 times greater in adults. For consumption of more than 200 eggs/year, a moderate-to-very-significant increase (50%–400%) in PCDD/F levels was expected at soil concentrations between 20 and 100 ng TEQ/kg dw, corresponding to Swiss legislation's investigation and remediation threshold values, respectively (SwissFederalCouncil, 1998). Due to its infrequent consumption (≤5 times/year), the mutton scenario would not lead to any relevant increase in exposure levels compared to the TDI or the regular food consumption scenario (Fig. C7). Nonetheless, according to sheep toxicokinetic model simulations, consuming mutton would exceed regulatory limits (2.5 pg TEQ/g fat, Commission Regulation (EU) No 1259/2011) if sheep were reared on Lausanne's contaminated soils (on average > 100 ng TEQ/kg dw; see Fig. C6).

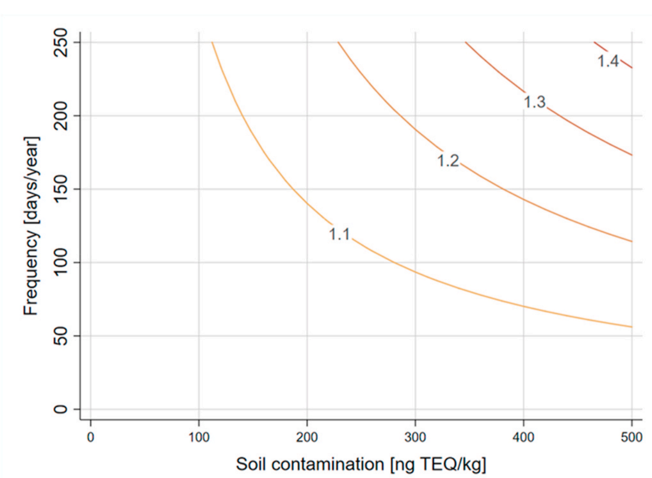

Fig. 2. Relative increases in expected PCDD/F concentrations in blood serum at 9 years old relative to normal dietary intake for the direct soil ingestion scenario (e.g., a ratio of 1.2 means that a 20% increase in the serum concentration is expected in this situation).

3.4. Risk management and protective measures

Interpreting relative increases in PCDD/F blood serum levels must consider existing biological variability. Biomonitoring studies among the general population reveal high variability, with age, dietary practices, and body mass index being the main influencing factors. A review of PCDD/F biomonitoring data covering more than 26,000 individuals in Italy, from 1989 to 2010, showed a coefficient of variation of 0.72, with a factor of about 6 between the 5th and 95th percentile (Consonni et al., 2012). Body burdens differing by a factor of two or more between two individuals in the general population are therefore common. To include the uncertainties in the Swiss population's current PCDD/F blood serum levels, we used thresholds of 20% and 100% increases in blood serum levels to determine protective strategies. The following categories were used:

- Low increase (<20%). This increase should not be noticeable in the general population. Only general hygiene recommendations are proposed below this threshold, mainly to prevent 'peak' exposure situations (e.g., accidental soil ingestion by children).
- Significant increase (20%–100%). Preventive measures are required; technical or organizational measures are implemented.
- Important increase (>100%). This level of exposure must be avoided. Restrictive or prohibitive measures should be implemented.

We used these evaluation categories and PCCD/F soil concentrations at different intervals of interest to define recommendations for the different land-use patterns identified, as Table 2 shows.

These general recommendations, combined with the soil contamination mapping, are intended to provide practical guidance accessible to affected inhabitants. Fig. 5a shows a map of the most-probable soil contamination categories as per the following PCDD/F intervals: 20–50, 50–100, 100–200, and >200 ng TEQ/kg dw. Only long-undisturbed top soils were mapped over the whole study area. Lausanne's authorities used this mapping to develop an interactive map of the contaminated area, allowing landowners to consult the targeted recommendations for their plots of land.

Fig. 5b shows the map of the probabilities of belonging to the assigned intervals. These two maps provide indissociable information on each plot within the study perimeter: the most probable interval of the plot's real pollutant level, and the probability of belonging to that class, i.e., the class estimate's level of accuracy. Confidence levels ranged from 0% (certainty that it does not belong to the interval) to 100% (certainty that it does), dependent notably on the conditioning data's quality and quantity and the spatial heterogeneity in the neighborhood of the attribute. Fig. 5b shows that soils located at the center of the maps, close

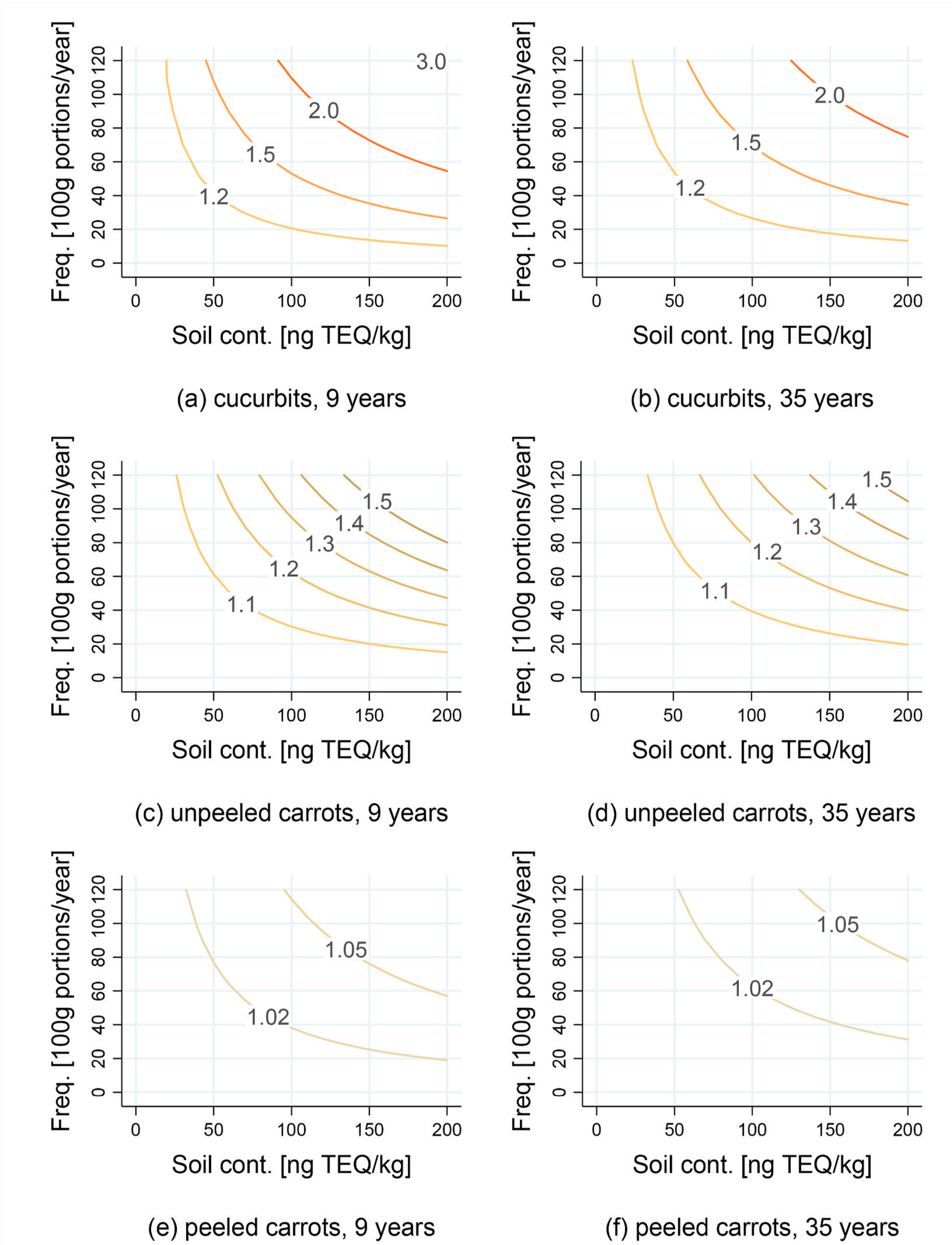

Fig. 3. Increases in expected PCDD/F concentrations in blood serum relative to normal dietary intake for three vegetable consumption scenarios and at 9 and 35 years old, expressed as a function of soil contamination and consumption frequency.

to the former incinerator, are categorized in the >200 ng TEQ/kg dw class, with high probabilities ranging from 80% to 100%. The greatest categorization uncertainties, with probabilities close to 50%, were located in areas where data density was low (with great heterogeneity in the spatial distribution of pollution) and/or in the transition zone between two intervals.

4. Discussion

The present study mapped PCDD/F soil contamination in the Lausanne area and considered exposure scenarios relating to different land uses. Important variations in potential exposure were calculated, leading to blood serum concentrations ranging from double to more than 10 times that expected in the general population. At similar soil pollution concentrations, the studied scenarios were categorized in order of increasing exposure: peeled vegetable root consumption < direct soil ingestion < non-peeled root vegetable consumption < cucurbitaceous vegetable consumption < egg consumption. Several essential elements for risk assessment and prioritizing protective measures could be highlighted based on these results.

– Vegetables' modest contribution to dietary intake (except for cucurbitaceous vegetables) was consistent with reports in the literature (Grassi et al., 2010). Meat and milk are frequently cited as

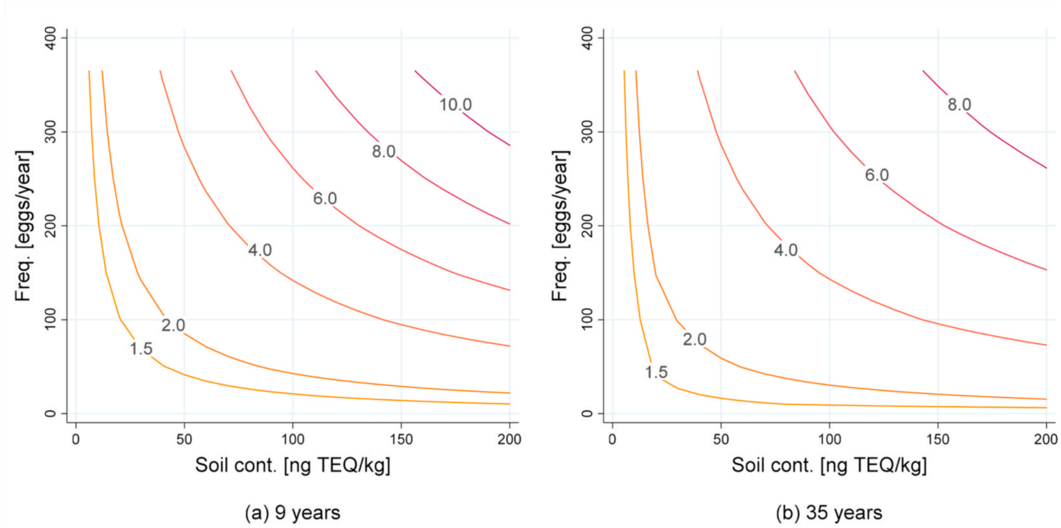

Fig. 4. Increases in expected PCDD/F concentrations in blood serum relative to normal dietary intake for egg consumption at 9 and 35 years old, expressed as a function of soil contamination and consumption frequency.

Table 2
Recommendations by soil contamination level and use pattern:

☐ **no measures,**

☐ **technical or organizational measures, and**

☐ **restrictive or prohibitive measures.**

		Intervals of soil concentration [ng TEQ/kg dw]			
		20–50	50–100	100–200	> 200
Home gardening	Consumption of root vegetables	Yes	Washed and peeled only		
	Consumption of cucurbitaceous vegetables	≤ 100 g of vegetables/person/week		No	
	Consumption of other fruits and vegetables (washed)	Yes			
Use of parks and gardens[1]	Visiting/playing in parks and gardens	Yes			≤ 3 times/week
Chicken owners	Consumption of eggs	≤ 1 egg/person/week	No		
	Offering or selling eggs	No			
	Consumption of chickens	No			

[1]*direct soil ingestion by children*

major sources of contamination due to the bioaccumulation of PCDD/Fs from feed and soil into lipid-rich animal tissues (Amutova et al., 2021), especially in suckling ruminants (Driesen et al., 2022). Accordingly, mutton and pork from animals raised on Lausanne's contaminated soils showed contamination levels above the maximum regulatory levels for animal food products (2.5 pg TEQ/g fat). This means that this meat cannot be marketed and jeopardizes the continued use of eco-pastoralism to maintain 30 ha of grassland across the city. Nevertheless, considering the low frequency of consumption of these food items, this will have a negligible impact on PCCD/F serum concentrations and human health. This is not true of private egg consumption. The consumption of home-produced eggs falls outside the scope of food safety regulations and may have a high frequency (e.g., daily consumption), thus leading to potentially high dietary exposure.

– The increase in blood serum concentrations expected for 35-year-olds was slightly lower than for 9-year-olds. This trend is the opposite of the increase in blood concentrations observed with age in the general population (related to the biopersistence of PCDD/Fs) (Antignac et al., 2016). It is also presumably due to children's lower body mass, which creates a relative increase in concentrations at equal exposure doses. This reinforces the need to protect the youngest children from risky situations.
– PCDD/F concentrations in soils appear to be only a very crude proxy for human exposure. The presence of contaminated soil does not necessarily mean that local residents will be exposed too. Residents

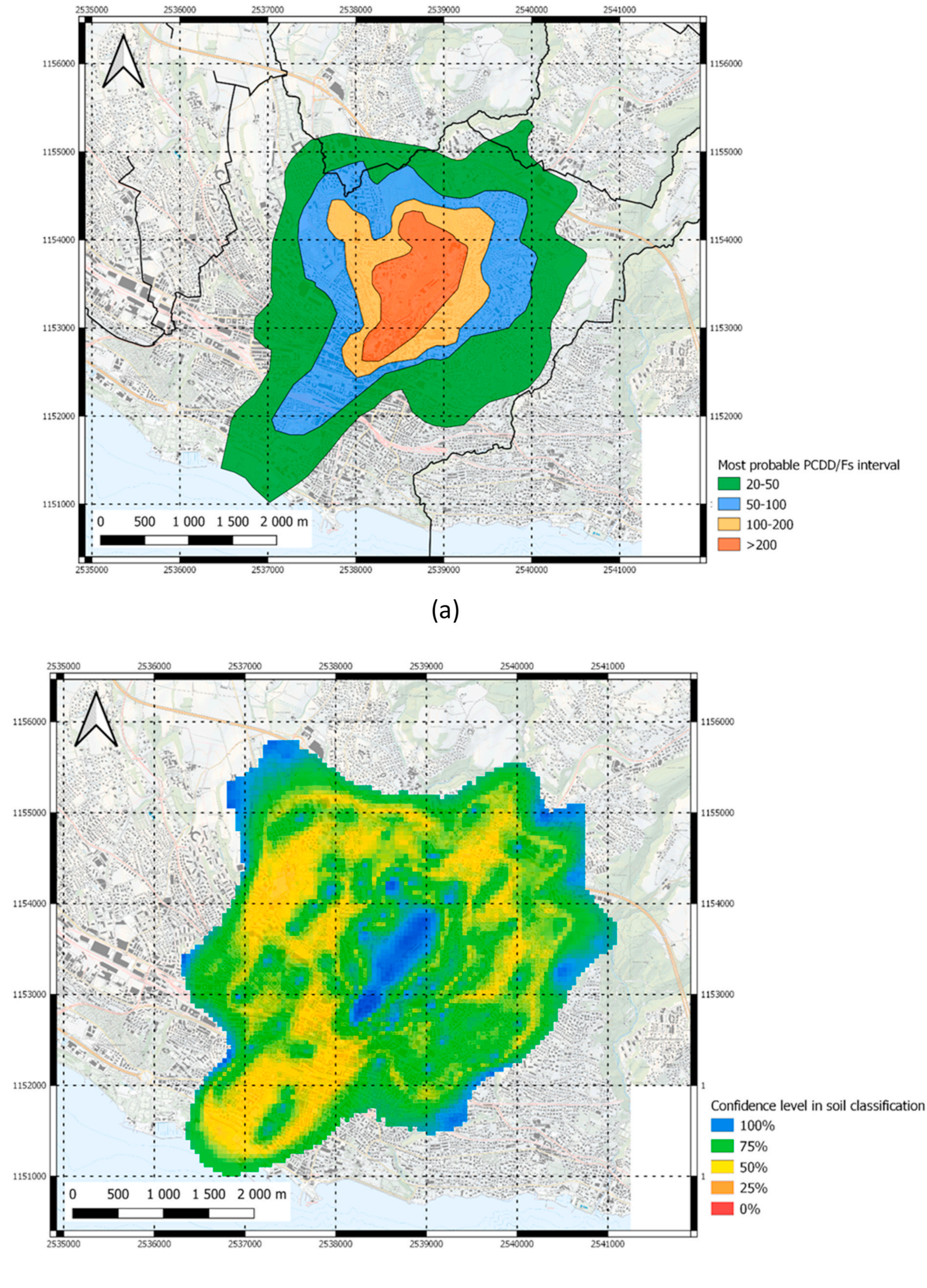

Fig. 5. Map of the PCDD/F intervals of interest: 20–50, 50–100, 100–200, and >200 ng TEQ/kg dw, with (a) the most probable soil classification and (b) probabilities of belonging to the assigned class.

without access to a garden and not consuming food produced on contaminated soils face negligible exposure compared to the intake of PCDD/Fs they might expect from their regular dietary exposure. In addition, many plots of land had been redeveloped (e.g., new constructions, landscaping) since the incineration plant ceased operations. Such soil disturbance leads to a significant dilution effect since PCDD/Fs are deposited and mostly persist in the first 10-cm layer of soil. Conversely, significant or important exposure may occur via the frequent consumption of eggs and, although to a lesser extent, cucurbitaceous vegetables grown in contaminated soil. Although soil mapping was an essential component of our risk assessment, it only indicated potential exposure: effective preventive measures or useful data for research purposes (e.g., epidemiological studies) requires considering land use conditions.
– Comparing different exposure scenarios by dose and expected increases in PCDD/F blood serum levels also highlighted an imbalance in the regulatory thresholds tolerated in soils and food. Soil regulations protect against direct soil ingestion, but little consideration is given to transfers along the food chain. On the contrary, food regulations focus on animal-fat-rich items and target the most potentially contaminated foods (eggs, milk, meat, fish), but they do not consider PCDD/F-absorbing vegetables. While PCDD/F levels in cucurbitaceous vegetables are much lower than in meat or eggs at similar soil concentrations, their consumption frequency can be high in individuals' private vegetable gardens.
– Finally, the relative dose (higher concentrations in serum than the reference scenario of normal diet in the general population) rather than the absolute dose seems to be the most appropriate metric because it allows us to: (1) situate a specific exposure in relation to other sources of contamination (i.e., general diet) and (2) consider the progressive accumulation of a pollutant up to the age at which the critical effect is determinant (i.e., the decrease in fertility of 9-year-olds).

Analysis of MSW incineration activities has shown that the 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF congeners frequently dominate TEQ emissions, but 1,2,3,7,8-PeCDD's contributions are generally lower than 2,3,4,7,8-PeCDF's (Xiong et al., 2022), in contrast to Lausanne's contamination (Fig. 1). Likewise, several studies have shown different dominant congeners to those in the Lausanne area's contaminated soils. Aberg et al. reported that 2,3,4,6,7,8-HpCDF was the most abundant congener in polluted soils in Sweden (Aberg et al., 2010), and Die et al. reported that 1,2,3,4,6,7,8-HpCDF, OCDD, and OCDF were the major congeners in the soils of urban green spaces in China (Die et al., 2021). Nonetheless, the PCDD/F congener profile in Lausanne was similar across all the soil samples, whatever their contamination levels (from 20 to > 200 ng TEQ/kg dw, data not shown), suggesting a single source of contamination. Levels of dioxin or furan releases in the MSW fumes at the time are unknown. However, it is noteworthy that the same pattern of 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF concentrations was also observed in spot wastewater measurements from the incinerator in 1996. Interestingly, the total TEQ soil contamination in Lausanne came mainly from PCDD/Fs, whereas levels of dioxin-like PCBs remained low, in the order of 2%–5% of the total contribution of TEQ PCDD/Fs and dl-PCBs.

The present study had some limitations. First, there were uncertainties related to the development of the mapping and exposure scenarios. Soil mapping was based on point measurements ($n = 126$) on undisturbed soils and did not reflect actual variability in the field. The assessment of PCDD/Fs concentration intervals in soils is carried out using an interpolation method in the areas where no data is available, but there is an inevitable uncertainty in estimation, which is quantified with the geostatistical approach. Moreover, the exposure model's parameters (e.g., human bioavailability, vegetation cover, ingested masses) were often conservative average values. To reflect realistic chronic exposure, we considered the central values of parameters found in the literature, when available. In cases of uncertainty or missing data, we used a conservative estimate. These choices were made to enable the comparison of different exposure situations and provide general recommendations to the public; they did not fully reflect the variability and uncertainties associated with individual situations. Secondly, current soil contamination is probably slightly lower than in the 1980s or 1990s. Because of PCDD/Fs' long half-lives in soils, however, it was estimated that these concentrations had only slightly decreased since the plant ceased emissions. Finally, the estimated PCDD/F contamination for Switzerland's general population was based on breast milk data from 2008. This baseline level was probably overestimated as the literature suggests that population contamination has progressively decreased in many countries since the introduction of emission restrictions. This decrease—very marked between the 1990s and 2000s—seemed to plateau after 2010 (Ae et al., 2018).

Overall, despite the uncertainties inherent in exposure modeling, coupling soil contamination mapping with prevention recommendations appears to be an interesting decision-making and communication approach: it helps explain measures to restrict or limit access to public spaces or certain activities. This is particularly useful for private landowners (several thousand land plots in the area) to whom the authorities can only issue recommendations, not restrictions. However, the study highlighted a lack of available data, firstly, on the historical exposure conditions of the region's inhabitants (when the incinerator was operating), particularly regarding inhalation exposure to particles, and secondly, on the Swiss population's current PCDD/F contamination levels. A biomonitoring case-control study will have to be conducted in the Lausanne area to answer these questions.

5. Conclusion

This interdisciplinary study performed a human health risk assessment by combining the mapping of PCDD/F soil contamination with exposure scenarios based on different land uses and types of food production and a human toxicokinetic model. Results emphasized the important levels of PCDD/F exposure possible from consuming cucurbitaceous vegetables or animal food products farmed on soils with PCDD/F concentrations as low as 20 ng TEQ/kg dw. They also confirmed the great relevance of integrating consumption frequencies and land uses in health risk assessment, especially from a regulatory perspective. The consumption of animal products farmed on soils with relatively low levels of PCDD/Fs, even below the regulatory norms, was particularly concerning.

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Author contribution

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Direct Soil ingestion (scenario S1)

Exposure scenario

The direct ingestion of soil by children was estimated using a relatively simple model. It uses similar parameters than those considered in the environmental Swiss authorities' regulatory assessment (Mailander and Hammann, 2005). The absorbed dose of PCDD/Fs, D_{soil}, was estimated by the following equation [A.1]:

$$D_{soil} = \frac{1}{10^3} \bullet \frac{C_{soil} \bullet M_{soil} \bullet F \bullet B \bullet \ C_{veg}}{BW} \quad \text{[A.1]}$$

•D_{soil} is the daily intake [pg TEQ $_{WHO\text{-}05}$/kg bw/day]
•C_{soil} is the concentration of PCDD/Fs in soil [ng TEQ $_{WHO\text{-}05}$/kg dw]

The soil concentration Csoil ranges used (20–400 ng TEQ WHO-05/kg dw) reflects the range of concentrations detected in areas where children may play (parks and gardens).

•M_{soil} is the mass of soil ingested per day [mg soil/d]

M_{soil} reflects the involuntary ingestion of soil by children (hand to mouth contact). A high variability is reported for this parameter, notably because of the evolution of the hand-mouth reflex and the surface area of the hands with age. A central value of 80 mg soil, corresponding to the ingested amount of soil for children aged 1–6 years, was used here (EPA, 2017). This value is conservative for older age groups, whose soil intake is expected to decrease and reach a minimum by age 12 (mean value of 30 mg soil).

•F is the frequency of exposure [-] (nb day/year)

A range of frequency F, up to 250 days/year, was used to reflect the different possible uses of the land. The value of 250 was considered a reasonable maximum by the working group, typically corresponding to a child living in a house with a garden who will go out as soon as the weather permits (approx. 4–5 days per week throughout the year).

•BW is the child's body weight [kg]

The evolution of body mass with age used here is that of the Concentration and Age Dependent Model (CADM), as published by EFSA (Knutsen et al., 2018b).

•B is the bioavailability of the PCDD/Fs in the soil ingested []

B should reflect both the bioaccessibility and absorbability of PCDD/Fs in soil, and is unknown in the case of Lausanne soils. Published data suggest that it is well below 100% and probably closer to 50% (Kerger et al., 2007). In the absence of precise data, a value of 75% was chosen by the working group as being reasonably conservative (EPA, 2011).

•C_{veg} is the vegetation cover [-] (0 fully covered, 1 uncovered)

*Vegetation cover Cveg is generally accepted as a protective factor, limiting direct access to contaminated soil (*EPA 2005*). To our knowledge, there is however no consensus over its quantitative value. A conservative value of 100% (no protection from vegetation cover) was therefore used. The evolution of body weight BW with age considered in this study is that of the CADM (concentration- and age-dependent model), allowing consistency with the toxicokinetic model used later* (Knutsen et al. 2018b).

The parameters used for the assessment of direct ingested soil doses are summarized in Table A1 below.

Table A.1
Direct soil ingestion – simulation parameters

Parameter	Value
Soil concentration, C_{soil}	0–400 ng TEQ $_{WHO\text{-}05}$/kg dw
Mass of soil ingested per day, M_{soil}	80 mg soil/d
Frequency of exposure, F	0–250 d/y
Bioavailability of the PCDD/Fs, B	0.75 (75%)
Vegetation cover, C_{veg}	1 (uncovered)
Body weight, BW	5–70 kg

Exposure dose estimates

The results for the average daily intake of PCDD/Fs for the direct soil ingestion scenarios are illustrated for concentrations of 20 and 100 ng TEQ $_{WHO\text{-}05}$/kg dw (corresponding to the investigation and remediation limit, respectively). No exceedance of the TDI is observed at 20 ng TEQ $_{WHO\text{-}05}$/kg dw for direct soil ingestion, only the results for 100 ng TEQ $_{WHO\text{-}05}$/kg dw are shown in Fig. A1. To simplify reading, only results above the EFSA TDI of 0.3 pg TEQ $_{WHO\text{-}05}$/kg bw/day displayed.

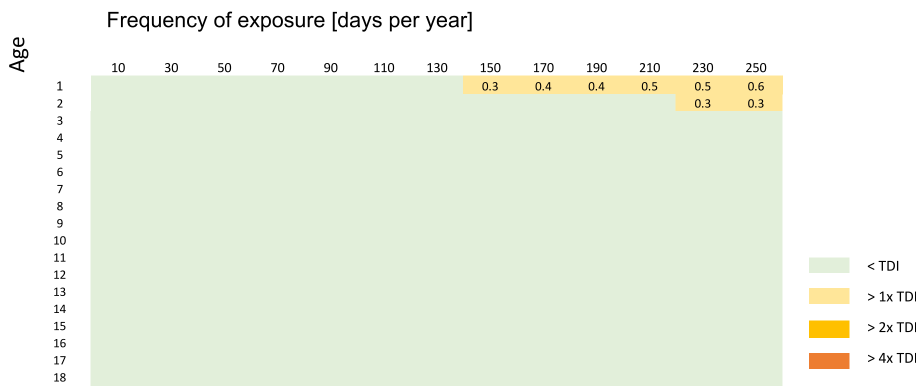

Fig. A.1. PCDD/Fs intake from direct soil in pg TEQ $_{WHO\text{-}05}$/kg bw/day at soil concentrations of 100 ng TEQ $_{WHO\text{-}05}$/kg dw for different age and frequencies of use of parks and gardens.

Appendix B. Vegetables consumption (scenario S2)

Exposure scenario

The PCDD/Fs uptake through vegetable is assessed according to the expected consumption, the PCDD/Fs soil concentrations and the plant species grown. For most vegetables PCDD/Fs assimilation is negligible and contamination is mostly at the surface of the unwashed or unpeeled vegetable. This is not true however for all of them. Bioconcentration factor (BCF), describing the ability of the compound to migrate from organic matter in the soil to the vegetable organs, have to be considered. The BCF is a dimensionless factor calculated according to formula [B.1]

$$BCF = \frac{C_{veg}}{R_{S/F} \bullet C_{soil}} \quad \text{[B.1]}$$

- •C_{veg} is the concentration in the edible crop organ in [ng/g Fresh Matter]
- •C_{soil} is the concentration in the soil [ng TEQ $_{WHO\text{-}05}$/kg dw]
- •$R_{S/F}$ the ratio between dry mass and fresh mass for the crop [-].

RS/F values used are those of the TROPHé study (ADEME, 2017).

Little intra-species variability is expected in the assimilation of PCDD/Fs, BCFs are however congener dependent. Average BCFs values were estimated on the basis of the TROPHé study, which investigated experimentally PCDD/Fs assimilation in zucchini, carrots, and potatoes over a wide range of soil concentrations (ADEME, 2017). Thanks to the relative uniformity of congener profiles observed in Lausanne soils, it was feasible to assess the vegetable concentration using average BCFs for each PCDD/Fs congener and convert it into TEQ $_{WHO\text{-}05}$ values.

Following the TROPHé approach, Zucchini exhibits BCFs about 4 and 22 times higher than unpeeled carrots and peeled carrots, respectively. Zucchini belong to the cucurbit family, which includes also squash, cucumbers, gherkins, melons, chayotes, pumpkins, pumpkins, watermelons and patties. Indeed, the species of this vegetable family have the ability to assimilate persistent organic pollutants (POPs). While the precise mechanism is not yet well known, it has been hypothesized that cucurbits' assimilation is enhanced by: (1) the mobilization of PCDD/Fs attached to soil organic matter through root exudate (Hulster and Marschner, 1993; Inui et al., 2008), and (2) the active assimilation of hydrophobic compounds through proteins binding (Inui et al., 2008).

In the absence of a defined use profile and consumption patterns of fruits and vegetables grown in private gardens in Switzerland, a field visit was made to two typical gardens. This allowed to define realistic production and consumption ranges to build exposure scenarios. The main vegetables grown were tomatoes, lettuce, beans, berries, zucchini and cucumbers. Some plots also grew radishes, carrots, eggplant, peppers, leeks, cabbage and chard. Each plot had between 2 and 5 cucurbit plants depending on its size. Considering that a plant produces on average 7 fruits of 800 g, and that plots produce food for household of 2–6 members, cucurbits' consumption up to 130 annual servings (100 g portions) per person were considered.

The parameters used for the assessment of direct vegetables consumption are summarized in Table B1 below.

Table B.1Vegetables consumption – simulation parameters

Parameter	Value range
Soil concentration, C_{soil}	0–200 ng TEQ $_{WHO\text{-}05}$/kg dw
Frequency of consumption of 100g portions	0-120 port./year
BCF for cucurbits	0–0.35 (median 0.13)[1]
BCF for unpeeled carrots	0–0.094 (median 0.037) [1]
BCF for peeled carrots	0–0.033 (median 0.0048) [1]

1 depending on the PCDD/F congener (ADEME, 2017).

As an example, the PCDD/F concentrations estimated in cucurbitaceous vegetables for different values of soil concentrations are presented in Table B2. The table compares the concentrations estimated in TROPHé study (ADEME, 2017) to the concentrations measured in cucurbits cultivated in a soil at 41–76 ng TEQ$_{WHO\text{-}05}$/kg in Lausanne. The concentrations in the cucurbits cultivated in Lausanne are lower than those expected based on the TROPHé study. Due to the small sample number, it is unclear whether this difference comes from the cucurbit species considered (zucchini vs. squash) or from the variability in soil PCDD/F concentrations.

Table B.2Estimated PCDD/Fs concentrations in cucurbits versus soil concentrations in *TROPHé study* (ADEME, 2017) compared to PCDD/Fs quantified in cucurbits

Soil concentration (ng TEQ $_{WHO\text{-}05}$/kg dw)	3	22	50	41–76	100	211
PCDD/F [pg TEQ $_{WHO\text{-}05}$/g fresh matter]	0.03	0.37	0.79	–	1.66	3.48
PCDD/F [pg TEQ $_{WHO\text{-}05}$/per 100g portion]	2.8	37.3	79.1	–	165.8	348.3
Cucurbit 1 (Lausanne) [pg TEQ $_{WHO\text{-}05}$/g fresh matter]	–	–	–	0.10	–	–
[pg TEQ $_{WHO\text{-}05}$/per 100g portion]				10		
Cucurbit 2 (Lausanne) [pg TEQ $_{WHO\text{-}05}$/g fresh matter]	–	–	–	0.04	–	–
[pg TEQ $_{WHO\text{-}05}$/per 100g portion]				4		
Cucurbit 3 (Lausanne) [pg TEQ $_{WHO\text{-}05}$/g fresh matter]	–	–	–	0.30	–	–
[pg TEQ $_{WHO\text{-}05}$/per 100g portion]				30		

Exposure dose estimates

The results for the average daily intake of PCDD/Fs for the vegetables consumption scenarios are illustrated for concentrations of 20 and 100 ng TEQ $_{WHO\text{-}05}$/kg dw (corresponding to the investigation and remediation limit, respectively). To simplify reading, only results above the EFSA TDI of 0.3 pg TEQ $_{WHO\text{-}05}$/kg bw/day are displayed.

Cucurbits

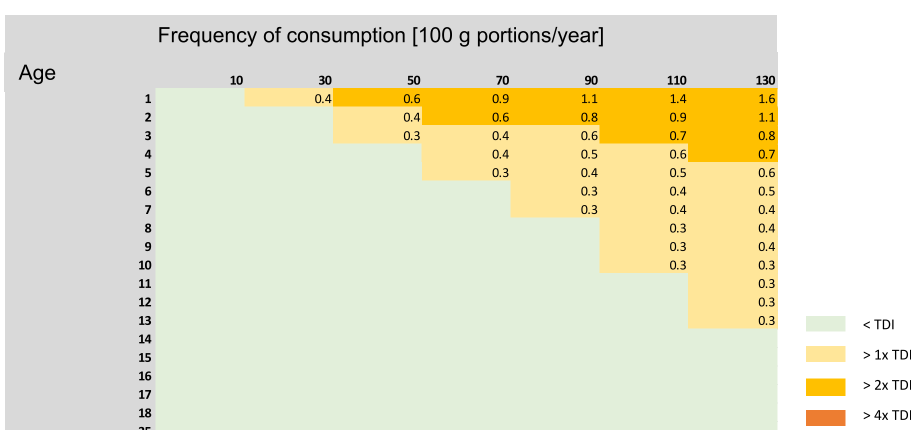

Fig. B.1. PCDD/Fs intake from cucurbits in pg TEQ $_{WHO\text{-}05}$/kg bw/day at soil concentrations of 20 ng TEQ $_{WHO\text{-}05}$/kg dw for different age and consumption frequencies (100 g portions per year).

Age	Frequency of consumption [100 g portions/year]						
	10	30	50	70	90	110	130
1	0.6	1.7	2.8	3.9	5.0	6.1	7.2
2	0.4	1.1	1.9	2.6	3.3	4.1	4.8
3		0.8	1.4	2.0	2.5	3.1	3.7
4		0.7	1.1	1.6	2.1	2.5	3.0
5		0.6	1.0	1.4	1.8	2.1	2.5
6		0.5	0.9	1.2	1.5	1.9	2.2
7		0.5	0.8	1.1	1.4	1.7	2.0
8		0.4	0.7	1.0	1.2	1.5	1.8
9		0.4	0.6	0.9	1.1	1.4	1.6
10		0.4	0.6	0.8	1.1	1.3	1.5
11		0.3	0.6	0.8	1.0	1.2	1.4
12		0.3	0.5	0.7	0.9	1.1	1.4
13		0.3	0.5	0.7	0.9	1.1	1.3
14			0.5	0.7	0.9	1.0	1.2
15			0.5	0.6	0.8	1.0	1.2
16			0.4	0.6	0.8	1.0	1.1
17			0.4	0.6	0.8	0.9	1.1
18			0.4	0.6	0.7	0.9	1.1
35			0.3	0.5	0.6	0.7	0.9

< TDI
> 1x TDI
> 2x TDI
> 4x TDI

Fig. B.2. PCDD/Fs intake from cucurbits in pg TEQ $_{WHO-05}$/kg bw/day at soil concentrations of 100 ng TEQ $_{WHO-05}$/kg dw for different age and consumption frequencies (100 g portions per year).

Unpeeled carrots

Age	Frequency of consumption [100 g portions/year]						
	10	30	50	70	90	110	130
1					0.4	0.4	0.5
2						0.3	0.4
3							
4							
5							
6							
7							
8							
9							
10							
11							
12							
13							
14							
15							
16							
17							
18							
35							

< TDI
> 1x TDI
> 2x TDI
> 4x TDI

Fig. B.3. PCDD/Fs intake from unpeeled carrots in pg TEQ $_{WHO-05}$/kg bw/day at soil concentrations of 20 ng TEQ $_{WHO-05}$/kg dw for different age and consumption frequencies (100 g portions per year).

Age	Frequency of consumption [100 g portions/year]						
	10	30	50	70	90	110	130
1		0.6	0.9	1.3	1.7	2.0	2.4
2		0.4	0.6	0.9	1.1	1.4	1.6
3			0.5	0.7	0.8	1.0	1.2
4			0.4	0.5	0.7	0.8	1.0
5			0.3	0.5	0.6	0.7	0.8
6				0.4	0.5	0.6	0.7
7				0.4	0.5	0.6	0.7
8				0.3	0.4	0.5	0.6
9				0.3	0.4	0.5	0.5
10					0.4	0.4	0.5
11					0.3	0.4	0.5
12					0.3	0.4	0.5
13					0.3	0.4	0.4
14						0.3	0.4
15						0.3	0.4
16						0.3	0.4
17						0.3	0.4
18						0.3	0.4
35							

< TDI
> 1x TDI
> 2x TDI
> 4x TDI

Fig. B.4. PCDD/Fs intake from unpeeled carrots in pg TEQ $_{WHO-05}$/kg bw/day at soil concentrations of 100 ng TEQ $_{WHO-05}$/kg dw for different age and consumption frequencies (100 g portions per year).

Peeled carrots

PCDD/Fs intake from unpeeled carrots is below TDI (results not shown).

Appendix C. Consumption of food of animal origin (scenario S3)

Egg consumption – exposure scenario

The online physiologically-based toxicokinetic (PBTK) model of the National Institute of Public Health and the Environment (RIVM) was used to estimate PCDD/Fs concentrations in the egg yolk lipids (RIVM; Van Eijkeren et al., 2006).

According to the literature, a hen consumes an average of 140 g of feed per day (Van Der Meulen et al., 2008) and when having access to an outdoor field may ingests between 2 and 10 g dw of soil per day (Kijlstra 2004), which corresponds to 1–7% of her diet. An egg weighs approximately 60 g and contains 32% yolk and 30% lipids in the yolk (Van Eijkeren et al., 2006). The fraction absorbed of PCDD/Fs following ingestion of contaminated soil was estimated to be 40–60% (Van Eijkeren et al., 2006).

The parameters used for the simulation are summarized in Table C1. The simulations are conducted until a stationary situation is reached (after about 200 days). The contamination of the background due to residues in the feed is neglected.

Table C.1
Eggs consumption – simulation parameters.

Parameter	Value
Hen's daily soil ingestion	10 g dw
Absorbed fraction of dioxin from soil	50%
Average weight of eggs	60 g
Proportion of yolk in eggs	30%
Proportions of lipids in egg's yolk	30%

As an example, PCDD/F concentrations obtained in eggs at different soil concentrations are presented in table C2. This table compares PCDD/F concentrations in eggs reported by the literature to those measured in eggs produced in two local and private henhouses (i.e., soil concentrations were 91 ng TEQ WHO-05/kg, and the measured PCDD/F concentrations were 27–29 pg TEQWHO-05/g lipid).

Table C.2
Estimated PCDD/Fs concentrations in egg versus soil concentrations from the model of Van Eijkeren et al. (2006).

Soil concentration ng TEQ $_{WHO\text{-}05}$/kg	Calculated egg concentration (data from literature) pg TEQ $_{WHO\text{-}05}$/egg	Calculated egg concentration Private henhouse 1 (Lausanne) pg TEQ $_{WHO\text{-}05}$/egg	Measured egg concentration Private henhouse 2 (Lausanne) pg TEQ $_{WHO\text{-}05}$/egg
1	4	–	–
20	85	–	–
50	213	–	–
100	425	311	311
200	850	–	–

Egg consumption – Exposure dose estimates

The results for the average daily intake of PCDD/Fs for the egg consumption scenarios are shown for soil concentrations of 20 and 100 ng TEQ $_{WHO\text{-}05}$/kg dw (corresponding to the investigation and remediation limit, respectively).

Frequency of consumption [100 g portions/year]

Age	10	30	50	70	90	110	130	150	170	190	210	230	365	400
1	0.3	0.8	1.4	2.0	2.5	3.1	3.7	4.2	4.8	5.4	5.9	6.5	10.3	11.3
2	0.2	0.6	0.9	1.3	1.7	2.1	2.5	2.8	3.2	3.6	4.0	4.4	6.9	7.6
3	0.1	0.4	0.7	1.0	1.3	1.6	1.9	2.2	2.5	2.7	3.0	3.3	5.3	5.8
4	0.1	0.4	0.6	0.8	1.1	1.3	1.5	1.8	2.0	2.2	2.5	2.7	4.3	4.7
5	0.1	0.3	0.5	0.7	0.9	1.1	1.3	1.5	1.7	1.9	2.1	2.3	3.6	4.0
6	0.1	0.3	0.4	0.6	0.8	1.0	1.1	1.3	1.5	1.7	1.8	2.0	3.2	3.5
7	0.1	0.2	0.4	0.5	0.7	0.9	1.0	1.2	1.3	1.5	1.6	1.8	2.8	3.1
8	0.1	0.2	0.4	0.5	0.6	0.8	0.9	1.1	1.2	1.3	1.5	1.6	2.6	2.8
9	0.1	0.2	0.3	0.5	0.6	0.7	0.8	1.0	1.1	1.2	1.4	1.5	2.4	2.6
10	0.1	0.2	0.3	0.4	0.5	0.7	0.8	0.9	1.0	1.1	1.3	1.4	2.2	2.4
11	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	1.0	1.1	1.2	1.3	2.1	2.3
12	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0	1.1	1.2	1.9	2.1
13	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0	1.1	1.2	1.9	2.0
14	0.0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0	1.1	1.8	1.9
15	0.0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0	1.1	1.7	1.9
16	0.0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	0.9	1.0	1.6	1.8
17	0.0	0.1	0.2	0.3	0.4	0.5	0.6	0.6	0.7	0.8	0.9	1.0	1.6	1.7
18	0.0	0.1	0.2	0.3	0.4	0.5	0.5	0.6	0.7	0.8	0.9	1.0	1.5	1.7
35	0.0	0.1	0.2	0.2	0.3	0.4	0.4	0.5	0.6	0.6	0.7	0.8	1.2	1.4

> 1x TDI
> 2x TDI
> 4x TDI
> 10x TDI

Fig. C.1. PCDD/Fs intake from eggs in pg TEQ $_{WHO\text{-}05}$/kg bw/day at soil concentrations of 20 ng TEQ $_{WHO\text{-}05}$/kg dw for different age and consumption frequency (nb. of eggs per year).

Age	Frequency of consumption [100 g portions/year]													
	10	30	50	70	90	110	130	150	170	190	210	250	365	400
1	1.4	4.2	7.1	9.9	12.7	15.6	18.4	21.2	24.1	26.9	29.7	35.4	51.7	56.6
2	0.9	2.8	4.7	6.6	8.5	10.4	12.3	14.2	16.1	18.0	19.9	23.7	34.7	38.0
3	0.7	2.2	3.6	5.1	6.5	7.9	9.4	10.8	12.3	13.7	15.2	18.1	26.4	28.9
4	0.6	1.8	2.9	4.1	5.3	6.5	7.6	8.8	10.0	11.2	12.3	14.7	21.5	23.5
5	0.5	1.5	2.5	3.5	4.5	5.5	6.5	7.5	8.5	9.5	10.5	12.5	18.2	20.0
6	0.4	1.3	2.2	3.1	3.9	4.8	5.7	6.5	7.4	8.3	9.2	10.9	15.9	17.4
7	0.4	1.2	1.9	2.7	3.5	4.3	5.1	5.8	6.6	7.4	8.2	9.7	14.2	15.6
8	0.4	1.1	1.8	2.5	3.2	3.9	4.6	5.3	6.0	6.7	7.4	8.8	12.9	14.1
9	0.3	1.0	1.6	2.3	2.9	3.6	4.2	4.9	5.5	6.2	6.8	8.1	11.9	13.0
10	0.3	0.9	1.5	2.1	2.7	3.3	3.9	4.5	5.1	5.7	6.3	7.6	11.0	12.1
11	0.3	0.8	1.4	2.0	2.5	3.1	3.7	4.2	4.8	5.4	5.9	7.1	10.3	11.3
12	0.3	0.8	1.3	1.9	2.4	2.9	3.5	4.0	4.5	5.1	5.6	6.7	9.7	10.7
13	0.3	0.8	1.3	1.8	2.3	2.8	3.3	3.8	4.3	4.8	5.3	6.3	9.3	10.1
14	0.2	0.7	1.2	1.7	2.2	2.7	3.1	3.6	4.1	4.6	5.1	6.1	8.8	9.7
15	0.2	0.7	1.2	1.6	2.1	2.6	3.0	3.5	3.9	4.4	4.9	5.8	8.5	9.3
16	0.2	0.7	1.1	1.6	2.0	2.5	2.9	3.4	3.8	4.3	4.7	5.6	8.2	9.0
17	0.2	0.6	1.1	1.5	1.9	2.4	2.8	3.2	3.7	4.1	4.5	5.4	7.9	8.7
18	0.2	0.6	1.0	1.5	1.9	2.3	2.7	3.1	3.6	4.0	4.4	5.2	7.7	8.4
35	0.2	0.5	0.8	1.2	1.5	1.9	2.2	2.5	2.9	3.2	3.6	4.2	6.2	6.8

> 1x TDI
> 2x TDI
> 4x TDI
> 10x TDI

Fig. C.2. PCDD/Fs intake from eggs in pg TEQ $_{\text{WHO-05}}$/kg bw/day at soil concentrations of 100 ng TEQ $_{\text{WHO-05}}$/kg dw for different age and consumption frequency (nb. of eggs per year).

Sheep meat consumption

Breeding system of the sheep herd of the Lausanne municipality

The sheep herd of the Lausanne municipality is composed of around 90 “Roux du Valais” and “Miroir” suckling ewes. Ewe reproduction follows ram introduction in September–November and lambing period further occurred in February–April. Lambs are weaned around 100 days in milk (May–July). Sheep are divided on flocks of 8–13 ewes suited with lambs. According to their flock, sheep grazed from April to October over differential sets of 6–11 grassland paddocks (total of 9.1 ha split into 53 paddocks) located all around the city area. Overall, each flock pastured twice per paddock over the grazing season, thus being exposed to wide range of soil contamination levels. Over the wintering period (November to March), sheep are housed in free-stall barn and fed with first-cut hay harvested on several grassland paddocks (total of 18.7 ha split into 67 paddocks) showing also contrasted soil PCDD/F concentrations (5–25% from lands with soil PCDD/Fs concentrations ≥200 ng TEQ $_{\text{WHO-05}}$/kg dw).

Scaling and calibration of suckling ewe and growing lamb physiologically-based toxicokinetic models

In order to handle the complexity of PCDD/Fs exposure pattern along time according to sheep folk moving along pasture paddock, dynamic simulations were performed using physiologically-based toxicokinetic (PBTK) models. Models initially developed for lipophilic contaminants fate into lactating (Lerch et al. 2018) or growing cattle (Albechaalany et al., 2022) were adjusted by interspecies scaling and recalibration to describe PCDD/Fs fate into suckling ewe and growing lamb, respectively.

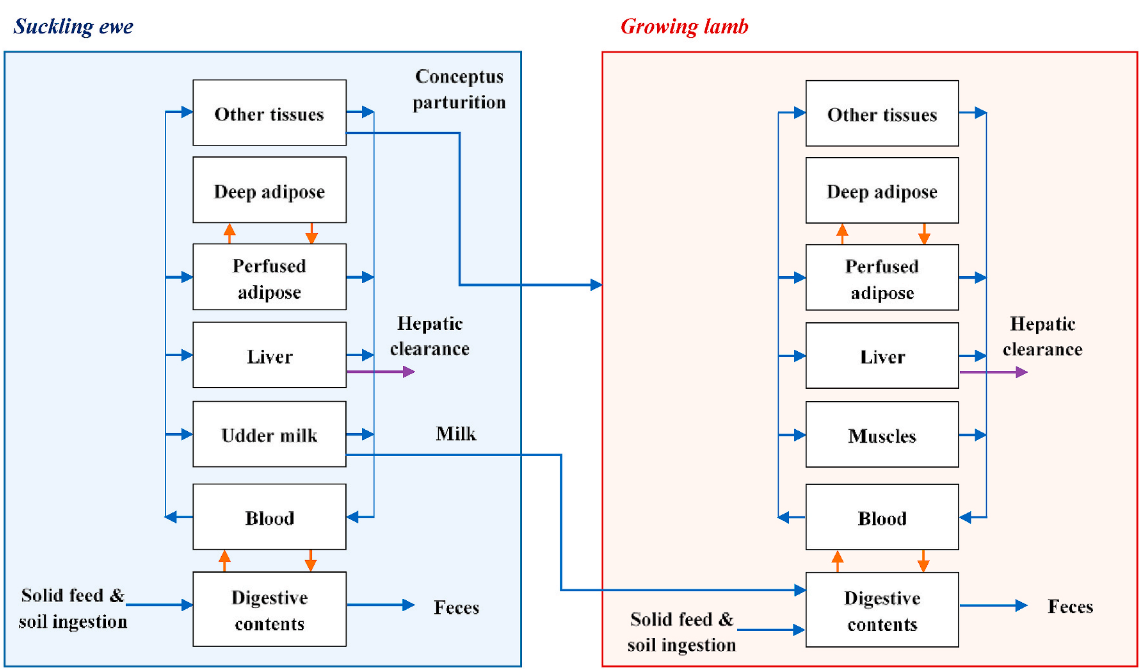

Fig. C.3. Conceptual diagrams of the physiologically-based toxicokinetic (PBTK) models describing the fate of PCDD/Fs into suckling ewe and growing lamb.

Rationale and framework

Each of the two PBTK models (i.e. lactating or growing animal) consists of three coupled sub-models, one describing the PCDD/F absorption, distribution, metabolism and excretion (ADME), the second feed intake and lipid digestion, and the last lactation or growth physiology. Such coupling offers a generic framework to explore the respective effects of contaminant physicochemical properties, lipid nutrition, and animal physiology on the feed-to-food toxicokinetic.

Conceptual diagrams of the ewe and lamb models are illustrated in Fig. C3. Into the ADME sub-model, mechanistic formalisms of PCDD/F adjective, diffusive, and degradation flows are hybridized from previous dairy cow fugacity (McLachlan, 1994) and PBTK (MacLachlan, 2009) models. The PCDD/F congener flows from feed and milk (only for lamb) intakes to the digestive tract, where it is excreted into feces or absorbed into blood. From blood, PCDD/F is either excreted back to the digesta or distributed between liver, adipose tissues (first to blood-perfused, later to deep sub-compartments), udder milk (later excreted through milk, only for ewe), muscles (only for lamb) and other tissues compartments. Liver is figured as the sole site for contaminant degradation (i.e. metabolism; purple arrows, Fig. C3). Advective flows (blue arrows) represent where the PCDD/F is transferred from one compartment to another together with an advective medium (digesta, blood or milk). Diffusive flows (orange arrows) represent where the PCDD/F crosses the interface between two compartments (digestive tract/blood and perfused/deep adipose tissues interfaces) by passive diffusion along the concentration gradient (Kelly et al., 2004). Further details about the initial bovine models set-up are provided in Lerch et al. (Lerch et al. 12–15 September 2022; Lerch et al. 2018).

Liver metabolic rate calibration

Absorption, distribution and excretion flows are computed from either the contaminant lipophilicity (i.e. coefficient of partition between octanol and water, K_{ow} harvest (Amutova et al., 2021), the specific tissue blood-perfusion rates (McLachlan, 1994) and the diffusive parameters (MacLachlan, 2009). Those parameters were keep unchanged when scaling the models from bovine to ovine. Conversely, the first-order metabolism rate constant into the liver (k_{met}) was specifically fitted for every single 17 2,3,7,8-chlorosubstituted PCDD/F congeners by adjustment against *in vivo* data describing PCDD/F toxicokinetic in growing lamb (Hoogenboom et al., 2015). The resultant k_{met} together with K_{ow} values are listed Table C1.

Table C.3
Coefficient of partition between octanol and water (K_{ow}) and first-order metabolism rate constant into the liver (k_{met}) calibrated for the 17 2,3,7,8-chlorosubstituted PCDD/F congeners used for model simulations.

PCDD/F congener	log K_{ow}^{1} (unitless)	k_{met} (d^{-1})
2,3,7,8-TCDD	6.6	5.2
1,2,3,7,8-PeCDD	7.2	5.4
1,2,3,4,7,8-HxCDD	7.6	2.0
1,2,3,6,7,8-HxCDD	7.6	0.8
1,2,3,7,8,9-HxCDD	7.6	3.2
1,2,3,4,6,7,8-HpCDD	8.0	4.0
OCDD	8.4	4.0
2,3,7,8-TCDF	6.5	800
1,2,3,7,8-PeCDF	7.0	160
2,3,4,7,8-PeCDF	7.1	4.0
1,2,3,4,7,8-HxCDF	7.5	4.0
1,2,3,6,7,8-HxCDF	7.6	2.8
1,2,3,7,8,9-HxCDF	7.7	32.0
2,3,4,6,7,8-HxCDF	7.6	2.8
1,2,3,4,6,7,8-HpCDF	8.0	6.0
1,2,3,4,7,8,9-HpCDF	8.2	8.0
OCDF	8.6	8.0

[1]From Amutova et al. (2021).
[2]Calibrated against biological data describing PCDD/Fs toxicokinetics in growing lamb (Hoogenboom et al., 2015).

Digestion and physiological sub-models adjustments

In order to further resolve the several differential equations of the ADME sub-model, the kinetics of body weight (BW), and of fresh and lipid masses in feed, digesta, feces, body tissues and milk compartments should be described. This was initially accomplished for bovine thanks to the coupling with lipid digestion (INRA, 2018), and lactation (Martin and Sauvant, 2007) or growth (Hoch and Agabriel, 2004) sub-models. For the present study, in both case (lactating female and growing offspring) the digestion and physiological sub-models were first adjusted in size by dividing by 10-fold diet intake level and every body compartments fresh and lipid masses in order to scale a bovine into an ovine. Additionally, for the ewe model, time adjustment of gestation and lactation lengths were performed (286 d and 300 d for cow to 152 d and 100 d for ewe, respectively). Accordingly, parameters of the equations for conceptus growth and lactation kinetics were kept unchanged, with only bovine-to-ovine time adjustment of 1.88 and 3.0-fold shortening for gestation and lactation lengths, respectively. Moreover, 10-fold lower conceptus weight (10.7 kg at parturition for the sum of lamb foetus, foetal membranes and fluids) and milk yield (1 kg at the peak of lactation for a suckling ewe) from cow to ewe were considered. Lastly, milk fat content was increased from bovine to ovine by a constant 1.7-fold factor (i.e. whole lactation average of 4.3% and 7.4% for cow and ewe milk, respectively). Output simulations of physiological traits are illustrated over a whole gestation-lactation cycle in Fig. C4. Those are in broad agreement with productive levels and classical patterns of extensive grass-based suckling sheep husbandry (chapter 20 into INRA, 2018).

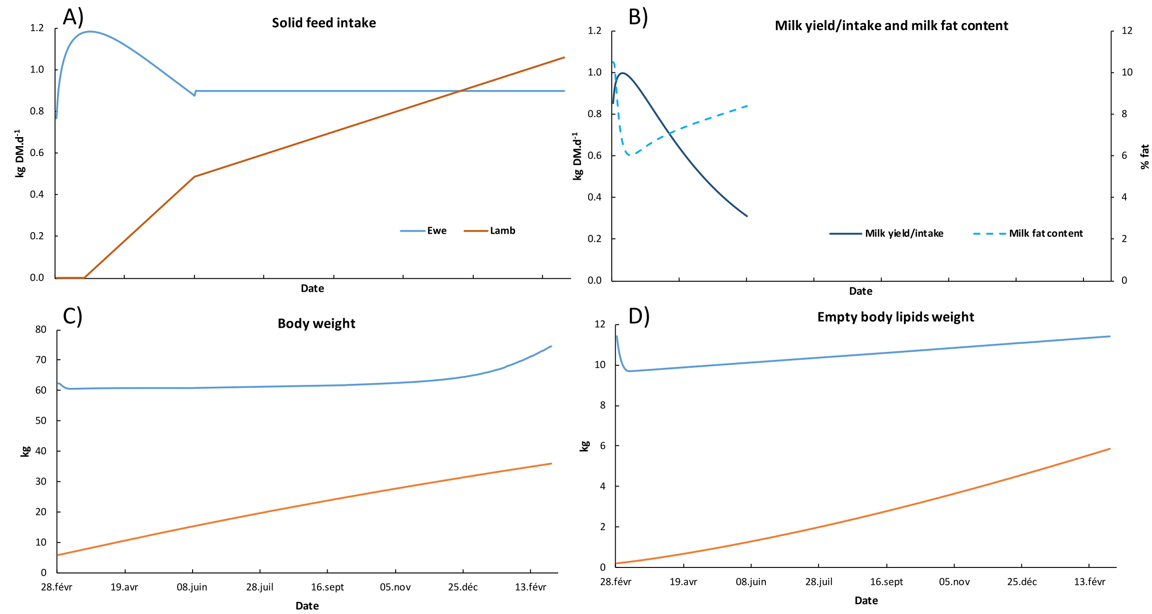

Fig. C.4. Simulations of productive and physiological traits of suckling ewe and growing lamb. A) solid feed intake, B) milk yield or intake and milk fat content, C) body weight and D) empty body lipids weight.

Scenario of ewe pasture calendar and exposure to PCDD/Fs

A ewe and lamb PCDD/F exposure scenario was further simulated over three successive gestation-lactation cycles. The case of the sheep folk which was the most often raised at pasture on the more contaminated area of Lausanne over the 2019–2021 years was selected. For the grazing season, the specific pasture calendar of this folk was used, by fixing the soil PCDD/F contamination level of every paddocks grazed to either direct measurements when available, or alternatively according to the paddock localization on the soil contamination map (see main text for details). Soil ingestion was considered as the only exposure route to PCDD/Fs. Indeed, in ruminants consumption of contaminated soil is one of the most relevant source of diffusive exposure to PCDD/Fs (Chatelet et al., 2015). In the present scenario, a constant soil intake level of 1% of total dry matter intake was assumed based on measurements performed on-site in September 2021 (S. Lerch, personal communication) using acid-insoluble ashes as marker of soiling (Van Keulen and Young, 1977). Over the wintering period, the concentration in hay offered to sheep was set to 0.10 ng $TEQ_{WHO-2005}$/kg DM according to direct hay measurement performed in December 2021. Intake PCDD/F concentration against time and physiological events corresponding to such exposure scenario over one year are summarized in Fig. C5. Accordingly, the simulated sum of PCDD/Fs TEQ concentration in adipose tissue of ewe and lamb are reported for the third lactation in Fig. C6. Direct *post mortem* adipose tissue PCDD/F measurements performed on three ewes and one lamb of the corresponding folk slaughtered in summer 2021 are also reported, and are remarkably in agreement with model simulations.

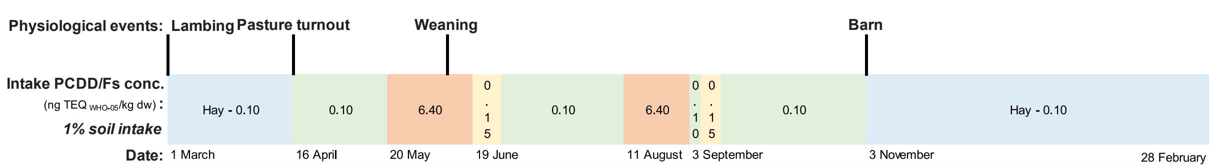

Fig. C.5. One-year lactation-gestation cycle and intake PCDD/F concentration of the exposure scenario for the sheep folk most often raised at pasture on the more contaminated area of Lausanne.

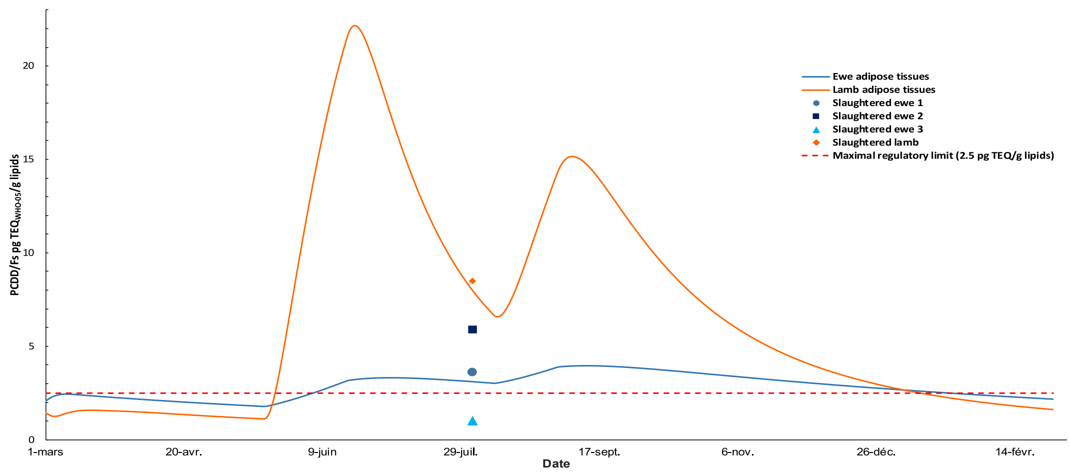

Fig. C.6. Model simulations of ewe and lamb adipose tissues PCDD/Fs total $TEQ_{WHO\text{-}05}$ concentrations for the sheep folk most often raised at pasture on the more contaminated area of Lausanne.

Human sheep meat consumption exposure scenario

Fig. C.7reports the results for the average human daily intake of PCDD/Fs for the sheep meat consumption scenarios, taking an average level of 10 pg/g lipids and a7% fat content in meat.

Age	Frequency of consumption [100 g portions/year]					
	1	2	5	10	15	20
1					0.3	0.5
2						0.3
3						
4						
5						
6						
7						
8						
9						
10						
11						
12						
13						
14						
15						
16						
17						
18						
35						

Fig. C.7. PCDD/Fs intake from sheep meat in pg TEQ $_{WHO\text{-}05}$/kg bw/day according to the pasture calendar of the most PCDD/Fs exposed ewe group along the third lactation for different age and consumption frequency (nb. of sheep meat portions per year).

Appendix

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