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Generic methodology to prevent food contamination by soil born legacy POPs in free range livestock

Farida Amutova[a,b,c,*], Matthieu Delannoy[a], Araylym Akhatzhanova[b,c], Nurlan Akhmetsadykov[b], Gaukhar Konuspayeva[b,c], Stefan Jurjanz[a]

[a] *URAFPA, University de Lorraine-INRAE, 54000, Nancy, France*
[b] *Antigen LLP, Scientific and Production Enterprise 040905, Almaty region, Kazakhstan*
[c] *Faculty of Biology and Biotechnology, Al-Farabi Kazakh National University, 050040, Almaty, Kazakhstan*

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ABSTRACT

Government monitoring commonly includes regulating POPs in animal feed and products of animal origin, with many countries setting Maximum Residue Levels (MRLs) to ensure safe tolerable concentrations. However, these MRLs do not address the presence of most POP families in soil, where concentrations can be much higher due to the contaminants' strong affinity and persistence in comparison to other environmental matrices. Extensive damage to food and production systems during a pollution incident causing soil contamination by POPs lead to severe economic and social consequences for the affected area. To mitigate these effects, it is crucial to implement necessary measures for consumer protection while also focusing on rehabilitating conditions for food production, tailored to both commercial farms and private holders. In this context, the present work aims to develop and test a methodology for assessing the tolerable concentration of the most cancerogenic legacy POPs in soil for various livestock animals in diverse rearing systems ensuring the safety of food of animal origin. Therefore, we summarize existing knowledge about the risk of POP transfer in different livestock breeding systems via soil exposure, and modeling via a backward calculation from the MRLs the corresponding tolerable quantity of POPs that may be ingested by animals in the considered rearing system. Results of these simulations showed that soil ingestion is a predominant contamination pathway, which is a central factor in the risk assessment of POP exposure on livestock farms, especially in free-range systems. In field conditions of POP exposure, low productive animals may be more susceptible to uptake through soil than high-yielding animals, even if the feed respected MRLs. Results show that PCDD/Fs revealed the lowest security ratio for low productive dairy cows (1.5) compared to high productive ones (52). Laying hens with a productivity of 45% show also as a high sensitivity to POPs exposure via soil ingestion. Indeed, their security ratio for PCDD/Fs, lindane and DDT were 3, 2 and 1, respectively. In perspective, proposed methodology can be adapted for assessing the risk of industrial POPs newly listed in the Stockholm Convention. In practice, it could be useful for food producers to apprehend their own risk of chemical contamination.

* Corresponding author. URAFPA, University de Lorraine-INRAE, 54000, Nancy, France.
E-mail address: amutovafb@gmail.com (F. Amutova).

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1. Introduction

Nowadays, it is admitted that 90% of chronic human exposure to some legacy POPs occurs through consumption of contaminated food, especially products of animal origin [1,2]. These types of products contain generally more fat than food of plant origin and therefore tend to accumulate more easily the lipophilic POPs [3–10]. Consequently, the most frequent cases of POP contaminated food appear among them. Food of animal origin is indeed a predominant source of human exposure to POPs [11] especially for such compound families as polychlorinated dibenzo-p-dioxins and polychlorinated dibenzo-p-furans (PCDD/Fs), polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) due to their high bioaccumulation in fatty tissues and high persistency [12–16].

Recent reports indicate that the contamination of large areas with dioxins, dioxin-like PCBs, OCPs, and other POPs in various countries has led to the transfer of these chemicals from contaminated environmental matter (water, plants and soil) to farm animals grazing in such territories [17–20]. Raising livestock on soils that have been historically contaminated due to past applications of PCBs [19,21,22], PCDD/Fs releases [23–25], PCBs emitted from previously used building materials but also from cooling oils of electrical transformers [20,26,27], and OCPs historically applied in agriculture [3–5,7–10,28] could lead to the consumption of contaminated food, especially of animal origin [1,2].

The contamination of food of animal origin is directly related to the daily ingestion of different contaminated environmental matters as water, plants and especially contaminated soil by free-range animals. Soil could retain POPs for several decades due to its strong affinity to soil organic matter (SOM), and the concentrations of these contaminants are higher in several order of magnitude than in water or feed. Moreover, literature reported that free-range animals can ingest consequent amounts of soil. According to previous studies [29–32] dairy cattle may ingest 2–5% of dry soil but in extreme conditions up to 10% of dry soil depending on the grazing conditions. Free-range poultry could also consume soil in highly variable proportions: from 5 to 30% of soil in the totally ingested dry matter [19]. These huge variations can be due to the quality of the soil cover, the surface available per chicken in the outdoor run, the age of the birds and the nutritional balance of the diet [33–36]. When such high soil intakes would take place on plots with contaminated surface soil, the high transfer and bioaccumulation of the main families of POPs [37] would lead to significant contaminations of the food produced by these animals. Therefore, the intake of soil must be considered as the main exposure factor for livestock, especially in areas with a historical contamination [17,19,38].

The control of POP presence in feed for animals and food of animal origin is usually included in governmental monitoring, and legislation defines maximum tolerable concentrations of the concerned compounds in these matters. Indeed, numerous countries defined maximum residue levels (MRLs), which established a maximum tolerable concentration of POPs, which has shown not to endanger human health. However, such MRLs do not cover presence most of the POPs in soil (except PCDD/Fs), which could reach much higher concentrations compared to feed due to its strong affinity and persistence of these contaminants in comparison to other environmental matrices [39–43]. Several papers [37,44–49] reported a high transfer of soil borne POPs to different body tissues, suggesting a considerable bioavailability of these compounds. This has been confirmed by some studies on hens, goats, and piglets [46,49–51]. These data show a real need to estimate the potential of a soil surface to produce their safe food with free-range livestock.

Indeed, a MRL is a threshold concentration beyond which regulations prohibit any commercialization of such food products in order to protect consumers. In cases of exceeding thresholds, investigations are undertaken to identify and eliminate the contamination source on the farm level and to launch expensive remediation programs.

Unacceptable concentrations of POPs in soil can be assessed by calculating backwards from the MRLs in animal foodstuffs (starting point) to determine the tolerable POP intake by the animal. Some well-known indicators of POP transfer are used for this purpose, i.e. transfer rates (TRs) and bioconcentration factors (BCFs). In addition to soil intake, other sources of oral exposure (e.g. feed) must be integrated to calculate the tolerable residual POP supply in order to not exceeding the safety level in animal food production.

In these calculations, the application of an additional security ratio may be necessary depending on the uncertainty of the used parameters, as usually done in risk assessments. The knowledge of the exact quantity of totally ingested pollutants (i.e. from all ingested matrices), which would transfer to the final food product, allows classifying farm production systems depending on their sensitivity to a potential situation of contamination. Not available or imprecise parameters of such calculations, such as the analytical method of detection [52], source and type of contamination [53] animal species, grazing conditions, and animal productivity [54–56] would enhance the application of such security factors. The latter would lead by their accumulation to an overestimation of the real risks.

The resulting massive destruction of foodstuffs and production systems in a situation of a pollution incident enhances heavy economic and social consequences for the concerned area [57]. However, they can and should be limited to what is strictly necessary while ensuring a perfect consumer protection. Rehabilitation of conditions allowing food production in contaminated areas is required for commercial farms, but also for private holders producing food for their households. The large surface areas of commercial farms, especially with large animals such as cattle, in comparison to generally restraint surface area sizes of private holders need to adapt management and restoring strategies to each case. Therefore, the precise establishment of the transfer of pollutants but also its efficient reduction can be considered as the main way to evaluate and manage the risks in the case of a contaminated area.

Based on the foregoing, the **goal of this work is** to develop and to test an approach to evaluate the tolerable POP concentrations in soil for different livestock animals in different rearing systems, allowing the safety of the produced food of animal origin. We therefore summarize existing knowledge about the risks of POP transfer in different livestock breeding systems via soil exposure and modeling via a backward calculation from the MRLs the corresponding and tolerable quantity of POPs, which may be ingested by the animals in the considered rearing system.

2. Material and methods

2.1. General scheme of the approach

Contrarily to food, nearly no regulation exists about the use of soil depending on its concentration of POPs. However, some countries have regulations specifying the use of soil/area depending on their contents in PCDD/Fs, [58–64]. According to the last critical review [17] these limits are currently not appropriate for safe free range livestock productions since they are mostly much over 10 ng TEQ/kg. We therefore built an assessment as a backward calculation starting from the maximum residue limit (MRL) generally fixed by authorities, evaluate then the transfer across the different animals to get the tolerable exposure via the supply of different vector matrices to finally come to the concentration in the soil on which these animals explore or pasture. Fig. 1 summarizes this approach.

For this reason, the approach started with the maximum tolerable concentration of the different POPs in foodstuffs. Indeed, such concentrations are fixed for PCDD/Fs and PCBs [65–67], OCPs [68] and have also been established recently for perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) [69] in food products. The absence of such regulations may reflect that the oral exposure of consumers is considered as a minor pathway, or the existing knowledge does not allow yet fixing such thresholds (for ex. residues of some OCPs). In the second case, toxicological reference values as for example the tolerable daily intake (TDI) could provide approximate concentration to build an approach.

The regulatory threshold concentration is indeed a ratio between the quantity of the considered compound and that of the foodstuff. By consequence, the productivity of the producing animal may influence this ratio by the dilution-concentration effect, especially for excreted foodstuffs like milk and eggs. The transfer rates (TRs) allow deduction of the daily tolerable exposure amount (DTEA), an TDE equivalent for food producing animals, preventing exceedance of the regulatory threshold. In the case of possibly consumed tissues (muscle, fat, offal), the rhythm of growth and the duration of bioaccumulation of the compound in the tissue must be calibrated to forecast the concentration of POPs. The DTEA can be calculated for these cases using the corresponding bioconcentration factor (BCF).

The DTEA can be sliced by the compound supply of each intake matter (feed, soil, water and sometimes air) and the bioavailability of the compound in the considered matter. The main route of POP exposure to ruminants and poultry is the oral consumption of contaminated feed and soil [70,71]. In context of legacy POPs (PCDD/Fs, PCBs and OCPs), water and air are generally minor vectors and can therefore be neglected. Feedstuffs are often regulated [72] and cannot be sold if their POP concentrations exceeded the regulatory thresholds. Only pastured grass or self-produced feeds (silage, hay, and cereal grains) could carry a locally deposited contamination, but POPs are poorly transferred in the aqueous plant sap reducing the contamination of such feeds to (mainly) soiling. By consequence, the main variation factor in the supply is the vector soil, by its intake rate and by its concentration of the compound. The soil intake can be evaluated today in numerous rearing systems. Finally, the maximum tolerable concentration of the considered POP in soil is the target of this approach.

The parameter setting of this approach is not necessarily simple, due to the interactions between parameters what will be developed here below.

2.2. Parameter settings

Highly productive cows or hens will dilute the bioavailable amount of POPs in a bigger amount of product (milk or eggs) enhancing by consequence a higher sensitivity of low productive animals in extensive rearing systems. However, very productive animals have also higher nutritional needs and by consequence will ingest more feed and – if grass on pasture is low – more soil.

The transfer rates can be extracted from literature [37,73–77]. Nevertheless, it has to be checked that the duration of exposure to the studied compound allowed to reach a steady state in order to get valuable TRs. The settings for meat producing animals is more

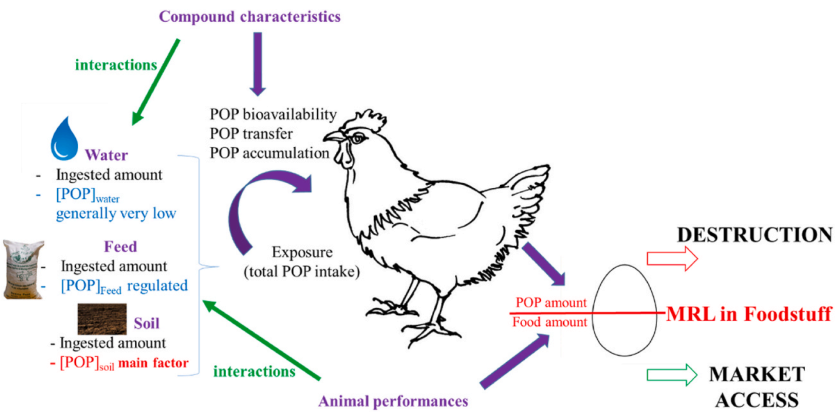

Fig. 1. Global approach to determine the maximum tolerable concentration of POPs in soil via a backward calculation from the MRL in foodstuff.

complicated since such animals will accumulate their whole life, and tissue concentrations will be fixed only after slaughter. The growth rhythm and by consequence the duration of the rearing period are very important parameters that would disadvantage extensively (and slowly) raised animals. Here too, BCFs are available in the literature but studies should have a duration consistent with the life expectancy of the considered animals [37,76,78–80].

The bioavailability of the ingested POPs is generally (very) high in water and feedstuffs [71,81–83] but can be reduced in soil [45,84]. Although the literature gives some indications for reduced bioavailability depending on the characteristics of soil and the considered compound, no consensual methodology is available today to test it in a routine manner. Therefore, available data cover a very limited number of cases. Otherwise, bioavailability is generally set at 100% [82,85].

The intake of feed (and water) is generally well documented in agricultural recommendations. Some difficulties may occur when animals do not ingest optimal feeds in very extensive systems such as the grass intake for free-ranged poultry or pigs. Literature offers some rare data whose extrapolations to the studied rearing system warrants caution. Intensively reared animals are mainly raised indoors on a concrete floor and by consequence would hardly have direct access to soil reducing their soil intake only to dust or mud deposited on the feed. Numerous data for soil intake of outside raised animals have been published in the last decades allowing a determination or a reliable extrapolation for the majority of rearing systems [29–32,86,87]. Here again, extensively reared animals would be more exposed to consequent soil intakes due to the use of generally poorer surfaces or free-range rearing even when vegetation does not offer large grass swards (drought, flood lands, winter grazing, …).

All these settings seem to indicate that extensive systems may be much more exposed to POP accumulation in the produced food than intensively raised animals on which we will focus our investigations.

2.3. Choice of scenarios of animal models and their corresponding rearing system

Firstly, three POP representatives have been chosen due to their persistency and their clearly established noxious effects on human health and their high persistency, especially in soil: PCDD/Fs, DDT and lindane.

Two models have been chosen for the dairy production: a high yielding cow (50 L/day) and a low yielding cow (5 L/day). Both types of animals may be on pasture, i.e., raised outdoor enhancing a risk to produce contaminated milk via soil intake but in contrasting rearing conditions. High yielding cows need an abundant grass sward to face nutritional requirements, which would by consequence limit soil ingestion on background levels. By contrast, low yielding cows, frequently raised in extensive holding systems, have to cope with sparse grass cover increasing significantly the risk of soil ingestion. Moreover, the dilution effect of absorbed pollutants in the excreted milk amount will strongly differ between both animal types. Nevertheless, climatic disorders, more and more frequent in the actual climate change situation, would modify this model situations by dust ventilation in a drought situation or soil mudding after heavy rainfalls. The egg production has been modeled only for extensively raised laying hens, as intensive systems would hardly use outdoor plots. Consistent with previous models, only extensively and slowly reared meat animals have been modeled: beef cattle raised up grass based during 3 years, broiler chicken raised up in alternative systems as for example organic as well as free-ranged pigs. Table 1 summarize the chosen animals as well as their zootechnical parameters used for modeling.

3. Results

3.1. Modeling of POPs transfer risk in different livestock breeding systems

Simulations of transfer risks of different POPs to different livestock species were performed in respect to European MRLs [67,68,88] for milk, eggs and several edible animal tissues as presented in Tables 2–4. Highly toxic POPs such as 17 congeners of PCDD/Fs, DDT and lindane (i.e. gamma-HCH) have been chosen as examples. Main rearing systems integrated in the modeling contain 2 groups of

Table 1
Rearing systems used in our simulations as well as the zootechnical parameters of these animals.

	Productivity	Daily feed intake [a]	Daily soil intake
Dairy cow	50 L/day	25 kg of dry matter	0,3 kg [b]
	5 L/day	10 kg dry matter	1 kg [b]
Laying hen	45 % [c]	150 g of commercial feed	0,01 kg [d]
	Life expectancy (months)		
Beef cattle	36	10 kg of dry matter	1 kg [e]
Broiler chicken	6	70 g of commercial feed	0,005 kg [f]
Growing pigs	12	2,5 kg of commercial feed	0,5 kg [g]

[a] following current feeding recommendations.
[b] according to Jurjanz et al., 2012.
[c] meaning on average 45 eggs in 100 days for each hen.
[d] according to Jondreville et al., 2010.
[e] according to Collas et al., 2020.
[f] according to Jurjanz et al., 2015.
[g] according to Collas et al., 2023.

Table 2
Simulation scenarios of transfer risk for different livestock animals grazing on soil contaminated by PCDD/Fs using transfer rate approach.

Type of rearing system	Food product	MRLs, in fat of food product	MRLs, in fresh matter of food product[a]	TR (%)[b]	Ingested amount of dioxins	DTEA of TCDD via soil respecting MRL[c]	Acceptable concentrations of dioxins in dry soil[d]	Literature derived concentrations of dioxins in dry background soil[e]	Security ratio[f]
Dairy cows (50 L/day)	Milk	2.5 ng WHO TEQ/kg	0.1 ng WHO TEQ/kg	32	0.31 ng/kg milk	15.5 ng/day	51.7 ng/kg	1 ng/kg	52
Dairy cows (5 L/day)	Milk			32	0.31 ng/kg milk	1.55 ng/day	1.55 ng/kg		**1.5**
Laying hens (45% or 1.6 g yolk fat/day)	Eggs	1.75 ng WHO TEQ/kg	0.009 ng WHO TEQ/kg	38	0.02 ng/egg	0.032 ng/day	3.2 ng/kg		**3**

[a] MRLs fresh matter was achieved through multiplying MRLs on 0.04 kg fat (per 1 kg cow milk) or on 0.0054 kg fat per 1 egg. Values of fat milk and fat yolk were derived from Jensen et al., 1991 and Amutova et al., 2021, respectively.
[b] TR (%) - Transfer rate corresponds to transfer of POP from ingested matter to food products and derived from Amutova et al., 2021. This TRs is based on mean values of 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF as they represent the major contribution to total TEQ of all PCDD/Fs.
[c] DTEA - Daily tolerable exposure amount. It was calculated by multiplying the ingested amount of pollutants (achieved through TR) on animal productivity (L milk/day or yolk fat/day) for cattle and hens.
[d] Acceptable concentrations of PCDD/Fs in soil were achieved from the ratio of DTEA of pollutants via soil to classical soil intake (indicated in Table 1).
[e] Literature derived concentrations of PCDD/Fs (WHO TEQ ng/kg) in dry background farmland soils based on 500 samples analyzed by German EPA(Bussian B. et al., 2013).
[f] Security ratio was calculated as the multiple between the acceptable POP concentration in soil before overpassing MRLs in the produced food products. Bold values of security ratio express a high risk.

Table 3
Simulation scenarios of transfer risk for different livestock animals grazing on soil contaminated by DDT and lindane using transfer rate approach.

Pesticide	Type of rearing system	Food product	MRLs, in fat of food product	MRLs, in fresh matter of 1 hen egg[a]	TR (%)[b]	Ingested amount of pesticide[c]	DTEA of pesticide via soil respecting MRL[c]	Acceptable concentrations of pesticide in dry soil[d]	Literature-derived concentrations of pesticide in dry soil[e]	Security ratio[f]
DDT	Laying hens (45% or 1.6 g yolk fat/day)	Eggs	50 μg/kg	0.27 μg	14	1.9 μg	3.04 μg/day	304 μg/kg	250 μg/kg	**1**
Lindane	Laying hens (45% or 1.6 g yolk fat/day)	Eggs	10 μg/kg	0.054 μg	22	0.24 μg	0.38 μg/day	38 μg/kg	24 μg/kg	**2**

[a] MRLs fresh matter was achieved through multiplying MRLs on 0.0054 kg fat per 1 egg. Values of fat yolk were derived from Amutova et al., 2021.
[b] TR (%) - Transfer rate corresponds to transfer of OCPs from ingested matter to food products and derived from Amutova et al., 2021.
[c] DTEA - Daily tolerable exposure amount. It was calculated by multiplying the ingested amount of OCPs (achieved through TR) on animal productivity (yolk fat/day).
[d] Acceptable concentrations of OCPs in soil was achieved from the ratio of DTEA of OCPs via soil to classical soil intake (indicated in Table 1).
[e] Literature derived concentrations in dry soil of OCPs (μg/kg) from Sailaukhanuly et al., 2016.
[f] Security ratio was calculated by the ratio of acceptable concentration of OCPs in soil to literature derived concentrations of OCPs in soil. Security ratio expresses a number by which OCP concentration in soil is multiplied in order to increase safety for consumed eggs. Bold values of security ratio express a high risk.

livestock animals depending on their raising purposes.

Group 1 - excreting animals: high productive dairy cows (50 L/day), low productive dairy cows (5L/day), and laying hens with a productivity of 45% (Tables 2 and 3).
Group 2 – meat producing animals: extensively growing beef cattle, extensively growing pigs and extensively growing chicken (Table 4).

Table 4
Risk assessment of livestock exposed to POPs through soil ingestion using EU MRLs and BCFs (bioconcentration factors).

POP	Animal	Edible tissues	EU MRLs in animal tissue, fat basis	EU MRLs in animal tissue, in fresh matter[a]	BCF[b]	Acceptable concentrations of POP in dry soil[c]	Literature-derived concentrations of POP in dry soil[d]	Security ratio[e]
DDT	Broilers	Muscle	1 μg/kg	0.03 μg/kg	17	0.18 μg/kg	250 μg/kg	**0.001**
		Liver		0.06 μg/kg	18	0.33 μg/kg		**0.001**
Lindane	Broilers	Muscle	0.01 μg/kg	0.0003 μg/kg	2	0.015 μg/kg	24 μg/kg	**0.001**
		Liver		0.0006 μg/kg	2	0.03 μg/kg		**0.001**
PCDD/Fs	Broilers	Liver	–	0.3 ng/kg	43	0.7 ng/kg	1 ng/kg	**1**
	Pigs	Fat	1 ng/kg	0.15 ng/kg	2	7.5 ng/kg	1 ng/kg	**7.5**
	Lamb	fat	2.5 ng/kg	0.4 ng/kg	2	20 ng/kg	1 ng/kg	20
	Cattle	liver	–	0.3 ng/kg	11	2.7 ng/kg	1 ng/kg	**2.7**

[a] - EU MRLs in animal tissue, in fresh matter calculate by multiplying of EU MRL in tissue, fat basis on fat mass (%) of correspondent tissue. Fat content in muscle, liver and fat were taken as 3%, 6 % and 15%, respectively.
[b] BCF – bioconcentration factor (unitless) derived from Amutova et al., 2021.
[c] Acceptable concentration of POP in soil was estimated by proportion of MRL in fresh tissue and BCF
[d] - Literature derived mean concentration of total PCDD/Fs (WHO TEQ ng/kg) in background farmland soils based on 500 samples analyzed by German EPA(Bussian B. et al., 2013), concentration for DDT and lindane were taken from (Sailaukhanuly et al., 2016).
[e] - Security ratio was calculated by the ratio of permissible concentrations of POPs in soil to literature derived concentrations of POPs in soil. Security ratio expresses a number by which POP concentrations in soil is multiplied in order to increase safety for consumed food products (edible tissue). Bold values of security ratio express a high risk.

Different zootechnical parameters of these animals were taken into account for reliable estimations (Table 1) such as classical daily feed intake [89–91] and daily soil intake [34]. In addition, simulations were based on TR for milk and eggs as well as BCFs for body tissues derived from elsewhere [37,75]. Subsequently, DTEA of the pollutants via soil was calculated in correspondence to established MRLs in the foodstuff (Tables 2–4).

Simulation scenarios of transfer risk for different livestock animals grazing on soil contaminated by PCDD/Fs, DDT and lindane (Tables 2–4) using TR and BCF-based approaches allowed to:

1) Express quantitatively the ingested amount of POPs per kg fresh milk or per 1 whole hen egg. This value has been calculated through the proportion of MRLs in a given foodstuff and corresponding TRs derived from our latest meta-analysis study [37].
2) Quantitatively estimate the DTEA of POPs via soil and acceptable concentrations of POPs in dry soil based on soil intake of studied farm animals.
3) Calculate a security ratio indicating the multiple between the POP concentration acceptable in soil before exceeding MRLs in the produced food products (i.e. the higher the value of security ratio the lower the risk to reach or overpass MRLs). The security ratio was classified as follows: >100 safe, 10 to 100 possible risk; 0 to 10 high risk.

Results of these simulations showed that soil ingestion is a predominant contamination pathway, which is a central factor in the risk assessment of POP exposure on livestock farms, especially in free-range systems. Indeed, extrapolation of established EU MRLs of PCDD/Fs, DDT and lindane for food and estimated TRs (Tables 2 and 3) or BCFs (Table 4) showed several observations.

In field conditions with POP exposure, low productive animals may be more susceptible to uptake through soil than high-yielding animals (Table 2), even if the feed respected MRLs. Indeed, animals in more extensive systems are often ranged on poorer surfaces increasing the risk of soil intake – by weaker vegetation cover of the soil or by a stronger exploring activity of such animals in order to cover their nutritional requirements. Lower values of acceptable concentrations of POPs in soil as well as security ratio indicate the vulnerability of low-producing livestock (dairy cows 5L/day vs dairy cows 50L/day as well as laying hens 45%), and therefore the health risk to consume contaminated milk or eggs (Table 2). This could be explained by higher bioaccumulation of pollutants in their body due to slow depuration processes as consequence of low excretion [92]. PCDD/Fs as one of the most toxic cancerogenic POP family revealed the lowest security ratio for low productive dairy cows (1.5) compared to high productive ones (52). Laying hens with a productivity of 45% (or 3 eggs/week) were also shown to be highly sensitive to POPs exposure via soil ingestion. Indeed, their security ratio for PCDD/Fs, lindane and DDT and were 3, 2 and 1, respectively (Tables 2 and 3).

Risk assessment of meat-producing animals (broilers, pigs, lamb, cattle) showed that the most sensitive edible tissues and organs are liver, followed by muscle and fat (Table 4). In case of daily soil ingestion and raising on contaminated territories, security ratio would vary from highly risky to exceed MRL (0.001 for broilers) to possible risky (between 7.5 and 20 for pigs and beef cattle).

Results (Tables 2–4) demonstrated that even a relatively low concentration of POPs in soil may lead to contamination close to MRLs in food of animal origin. Therefore, livestock animals raised on outdoor plots are under a high risk of POP contamination and therefore may produce contaminated edible tissues, milk or eggs. Soil should be considered as the main POPs carrier in the case of contamination situation in a farm. Indeed, water is a weak vector of chlorinated POPs and contaminated vegetation could be easily removed from such areas by simple cutting. By consequence, the management of contaminated soil would be the main problem: soil may accumulate a lot of POPs over a long exposure time and is hardly possible to be removed from the areas.

4. Discussion

4.1. Methodological approach based on transfer rate and bioconcentration factor

The considered approach to assess the risks of POPs transfer from contaminated soil to farm animals and food products can be used in the assessment and monitoring of contaminated areas. However, several important nuances must be added before the practical application of this approach. The correctly calculated pollutant transfer coefficients (TR and BCF), the zootechnical parameters of the animal on the farm, the animal's diet, and daily soil consumption should be considered.

Simulation scenarios of transfer risk for different livestock animals grazing on soil contaminated by selected POPs (Tables 2–4) showed that daily soil ingestion may be considerably impacted on obtained acceptable concentrations in soil and therefore enhancing risk to exceed MRL. For example, in our suggested risk simulations, laying hens with a productivity of 45%, ingesting 10 g of soil per day (dry matter), should not exceed the calculated acceptable concentration 3.2 ng/kg (Table 2). However, there are cases where chickens might ingest up to 30 g of soil per day [19]; in this scenario, applying the proposed approach, the acceptable concentration in soil would be considerably reduced. Consequently, in conditions of severe soil contamination, when for example soil concentration of PCDD/Fs achieve 2 ng TEQ/kg as mentioned in studies [19,20] and even higher (5–15 ng TEQ/kg) as in a study of a German governmental agency [93], the risk of exceeding the threshold in the final food product could increase in several order of magnitudes.

Equations for calculating the transfer rate and bioconcentration factor can be found in the literature [37,77,94,95]. There is, however, no standardized protocol that includes all the required initial parameters. The transfer coefficient is used to calculate the transfer of a contaminant from the ingested matrix to excreted animal food, such as milk and eggs. The matrix consumed could be contaminated feed, grass, silage, or soil. Equation (1) is used to calculate TR:

$$\mathrm{TR}_{product} = \frac{[pollutant]_{fat\ of\ product} \times \text{Daily fat excretion of product}}{[pollutant]_{intake\ matter} \times \text{Daily intake}} \times 100\% \qquad \text{(Eq.1)}$$

The [pollutant]fat of product corresponds to the concentration of pollutant in the fat of the food product (pg/g fat) and [pollutant] intake matter to concentration in the intake matrix feed or in soil (pg/g dry matter). Daily fat excretion of product is amount of fat (kg) of the product (milk or egg) produced by the given farm animal per day. Daily intake is the amount of feed or soil (kg) ingested by animal per day.

A recent meta-analysis of data of POP transfer [37] revealed that the critical factor for accurately calculating the TR is the concentration of pollutants obtained when the animal reaches a state of equilibrium or steady state. The definition of steady state can be found elsewhere [37,96]. The achievement of a steady state can be reasonably predicted by the half-life of the specific compound, and it is generally accepted that the steady state condition is achieved after an exposure period of 3.3 half-lives [97]. This approach is thoroughly explained elsewhere [95]. Hence, the estimation of the steady state of the given animal for a given pollutant needs to focus on the half-life of the target compound and the productivity of the animal. For example, Table 5 provides half-lives for TCDD, DDT, and lindane in milk and eggs, the animal production cycle and demonstrates how to predict exposure time to reach steady state. According to the literature [97] 3.3 half-lives of the compound ensures that 90% steady state is achieved. By multiplying the half-life of the compound by 3.3, it is therefore possible to determine the exposure time on the animal and estimate the steady-state concentration of the pollutant.

Exposure time of the animals should be consistent with animal production cycles. Indeed, dairy cows (6 years) and meat cattle (2 years) or laying hens (1–2 years) and broilers (2 months) have different animal production cycles (Table 5). Dairy animals and laying birds have a longer lifetime period than meat-producing animals, which could impact the achievement of steady-state. In contrast, meat producing animals could hardly achieve steady-state due to dilution of pollutant during intensive body growth and due to their relatively short lifetime. Therefore, POP transfer to meat of growing animals estimated by distribution of pollutant in different animal tissues (liver, fat, offal) is classically performed using BCFs. equation (2) of BCF is given below:

$$BCF = \frac{[\text{pollutant}]_{tissue}}{[\text{pollutant}]_{intake\ matter}} \qquad \text{(Eq.2)}$$

Where pollutant in brackets mean concentration of pollutant in tissue or in intake matter (soil or feed).

4.2. A preventive supporting decision tool

As a result of anthropogenic chemical activities over the last two centuries [98,99] animals are exposed to many distinct molecules [100] derived from the environment, and especially from soil. Recent awareness of this diversified exposure is addressed by the fairly

Table 5
Predicted exposure time of POPs in milk and eggs ensuring steady state condition.

	TCDD-eggs	TCDD - milk	DDT- milk	Lindane - milk
Half-life TCDD	30 days	40 days	10 days	18 days
Animal production cycle	1–2 years	6–10 years	6–10 years	6–10 years
Predicted exposure time to achieve steady state	99 days (~3 months)	132 days (~4 months)	33 days	60 days

recent concept of the exposome [101]. However, for livestock systems this global exposure is little considered, unlike human exposure, which is constantly being improved. As this study has shown, TRs and BCFs are two interesting concepts for describing and predicting the transfer of contaminants into food products, as they enable contaminant concentrations in the soil to be directly linked to levels in the produced food. In this sense, it allows us to rethink the management of chemical safety in food products of animal origin. It adds to the current strategy to control food products of animal origin a necessary lever in chemical safety: prevention.

Indeed, current chemical safety controls raise concerns because they make producers responsible for contamination of commercialized food production, whereas they are also victims of contamination in their immediate environment. Indeed, the expenses related to harm, including affected animals, contaminated areas, and management costs, should be borne by the polluters in line with the polluter pays principle and extended producer responsibility. Farmers should not be held responsible for historical PCB application, industrial emissions of PCDD/F, and dioxin-like compounds (except, for instance, cases of open burning in the backyard) [20]. Furthermore, current chemical safety controls are mainly focused on conventional farming systems and do not consider self-production and self-consumption, which represent a major source of exposure when producing on contaminated soils [102]. In addition, the realization of such controls is known to require a certain delay before the identification of the contaminated food producing systems. Generally, consumers will be in contact with these contaminated food during this time period that could otherwise be avoided by preventing approaches. This led to different crisis as the 1999 ″dioxin broiler crisis" [103], in which tons of contaminated animals were consumed before anyone was aware of their non-compliance with dioxin and PCB MRLs, with potential repercussions on the health of the population.

To implement this prevention layer, a generic and simple decision tool can be theorized to help food producers to apprehend their own risk of chemical contamination (Fig. 2). The starting point should be a chemical analysis of representative soil samples from their land where free-range animals are reared. Additionally, on farms with PCDD/Fs or PCBs contamination from specific sources affecting food, livestock management recommendations suggested by the study [20] could be implemented. Then, the found chemical substances should be evaluated regarding the ability to be bioaccumulated in target foodstuff based on bibliographical data and chemical characteristics.

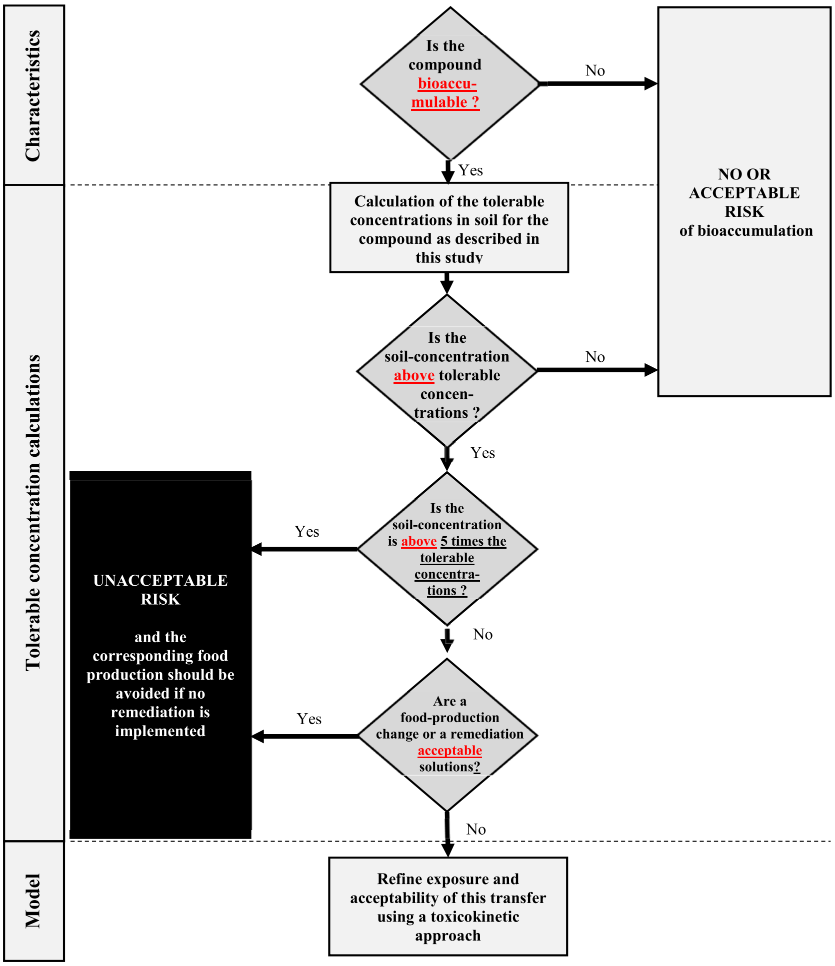

Fig. 2. Flow chart as a preventive supporting decision tool of food product contamination.

Indeed, bioaccumulation is a prerequisite for the concentration of contaminants in edible tissues, and therefore in food products of animal origin. In the case of the presence of bioaccumulable pollutants in soil, proposed methodology of determining soil limits in respect to TR and BCF could be applied to assess the risk of POP transfer to livestock animals. Soil concentrations of PCDD/Fs and OCPs, as demonstrated in this study, along with other families of POPs (PCBs, PFOS, PFOA, SCCP, PBDEs), can be incorporated into the proposed methodology if it becomes necessary to conduct a risk assessment in the event of contamination with these substances. Soil concentrations of selected pollutants should therefore be compared to acceptable soil concentrations calculated in the present study. If soil levels are greater than 5 times of these tolerable concentrations, animal production should clearly be avoided. If soil concentrations are below tolerable levels, food production can be carried out on this soil. In other cases, the tool highlights the risk of such production, but calculation uncertainties may lead to overestimations. If maintaining a production is needed according to a particular context, further investigations can refine the risk to accurately assess which cases of production can be allowed. In this case, other models should be developed, involving a toxicokinetic models as a function of farming practices to highlight the most at-risk conditions and avoid food production under these conditions.

One of the added values of implementing such a decision-support tool is its genericity and its ability to be updated for emerging pollutants. It requires a relatively simple database including bioaccumulation status, BCF and TR for food products of animal origin, and the calculations as described in the current study. In this context, such a methodology relies on a limited set of animal experiments to obtain the necessary BCFs and TRs [33,37,74]. In addition, chemo-monitoring can be implemented thanks to non-targeted approaches [104]. The data from a soil sample could today be reprocessed to find emerging compounds. Then, food producers can be quickly informed and may carry out necessary quantification and risk assessments. Moreover, this preventive methodology can also be implemented in non-conventional farming systems such as self-production and self-consumption.

5. Conclusion

In the context of contamination of large areas by different POPs, there is a compelling need to formulate and implement appropriate regulatory thresholds for soil to guarantee the safety of food production. In these areas, safeguarding human health considering both MRLs and TDI becomes crucial. This is particularly pertinent for subsistence farming and locally produced food, where MRLs are tailored for commercial food sales and may not fully address the concerns of those relying on generally over-exposed self-grown produce.

This study outlined a generic methodology aiming the prevention of food contamination by various soil-borne legacy POPs in free-range livestock. The methodology integrates transfer coefficients (TR and BCF) to assess the risk of POPs contamination in different livestock rearing systems, including dairy cows, laying hens, and meat-producing animals. This approach could be adapted and applied for assessment of risk of new industrial POPs such as PFOS, PFOA, Perfluorohexanesulfonic acid (PFHxS), polybrominated diphenyl ethers (PBDE), short-chain chlorinated paraffins (SCCPs), listed in Stockholm convention. Simulations of various contamination scenarios showed that soil ingestion should be considered as a central factor in the risk assessment of POP exposure on livestock farms, especially in free-range systems. Results demonstrated that even a relatively low concentration of POPs in soil may lead to contamination close to MRLs in food of animal origin. In perspective, proposed methodology could be useful for food producers to apprehend their own risk of chemical contamination.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable requests.

CRediT authorship contribution statement

Farida Amutova: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Conceptualization. **Matthieu Delannoy:** Writing – review & editing, Validation, Data curation, Conceptualization. **Araylym Akhatzhanova:** Writing – review & editing, Conceptualization. **Nurlan Akhmetsadykov:** Project administration, Funding acquisition. **Gaukhar Konuspayeva:** Writing – review & editing, Formal analysis. **Stefan Jurjanz:** Writing – review & editing, Validation, Supervision, Data curation, Conceptualization.

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.

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