Environ Sci Pollut Res (2014) 21:6440–6447
DOI 10.1007/s11356-013-1882-8

Exposure of ruminants to persistent organic pollutants and potential of decontamination

Guido Rychen · Stefan Jurjanz · Agnès Fournier · Hervé Toussaint · Cyril Feidt

Received: 22 December 2012 / Accepted: 29 May 2013 / Published online: 14 June 2013

Abstract Human activities are emitting persistent organic pollutants (POPs) to the environment. These compounds have raised concerns about the risk of transfer through the food chain via animal products. They are characterized by a strong persistence in environmental matrices and a lipophilicity which may lead to their accumulation in fat tissues. In EU Regulations (no. 1881/2006, 1259/2011), maximum acceptable levels for polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans (PCDD/Fs), and dioxin-like or nondioxin-like polychlorinated biphenyls (PCBs) in food of animal origin have been set. Transfer rates from contaminated fodder to milk have been established: for PCBs, the rate of transfer varies from 5 to 90 % and for PCDD/Fs from 1 to 40 %. The differential transfer of the compounds towards milk is related to the hydrophobicity of the pollutants and to their metabolic susceptibility. According to numerous authors, soil is the major reservoir for POPs, and its involuntary ingestion by farm animals reared outdoors may be the main cause of animal product contamination (meat, milk, or eggs). Recent studies seem to indicate that soil is a real risk matrix in terms of transfer of pollutants to the food chain. A POP crisis management is extremely difficult, since it impacts many farmers located in the contaminated area. The question arising is to know if livestock contaminated by POPs may be decontaminated and further used for their initial purpose. Recent data demonstrate that the decontamination process appear feasible and depends on initial level of contamination or the physiological status of the animals.

Keywords Pollutant · Ruminant · Decontamination · Milk · Meat

Introduction

Human activities are at the origin of persistent organic pollutants (POPs) emissions in the atmosphere resulting in pollutant deposition over the entire Earth's surface. These deposits affect agricultural areas near or far away from emitting sources (Beyer et al. 2000; Lohman and Seigneur 2001) and suggest potential concerns when considering the transfer risk of these compounds in the food chain (Rychen et al. 2005; Antignac et al. 2006). Defined in the Stockholm Convention, POPs are considered toxic to human health and ecosystems. They include either pesticides, compounds of industrial origin such as DL and NDL PCBs (dioxin-like or non dioxin-like polychlorinated biphenyls), or unintentional byproducts of industrial processes such as dioxins/furans (PCDD/Fs). In recent decades, dioxins and PCBs have been the most problematic POPs in terms of transfer to animal products.

The European Union has set maximum levels for polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans (PCDD/Fs), and polychlorinated biphenyls (PCBs) in foodstuffs of animal origin (EU Regulations nos. 1881/2006, 1259/2011). When animal products exceed regulatory limits of POPs, marketing of animal products is prohibited and herds have to be slaughtered. Thus, in recent POP crises in France (defectuous Incinerator in Gilly-sur-Isère, 2001), accidental fire in St. Cyprien (2008), defectuous industrial processes in Grez Bouère (2011), more than 8,000 cattle have been slaughtered. Such major crises cause human, social, and economic disasters. From these practical crises situations, several scientific questions have been raised and concern (1) the identification of the exposure levels of ruminants in contaminated areas, (2) the characterization of the bioavailability of POP from environmental matrices (fodder and soil), and (3)

Responsible editor: Philippe Garrigues

G. Rychen (✉) · S. Jurjanz · A. Fournier · H. Toussaint · C. Feidt
Unité de Recherche Animal et Fonctionnalités des Produits Animaux (UR AFPA), Université de Lorraine, INRA, ENSAIA, 2 avenue de la Forêt de Haye, BP 172, 54505, Vandoeuvre-lès-Nancy, France
e-mail: guido.rychen@univ-lorraine.fr

the evaluation of the potential of decontamination of ruminants whose levels exceed regulatory standards.

POP exposure of ruminants

Ruminant exposure via soil and forage

Upon issue, POPs are directed towards the Earth's surface by gaseous or particulate deposits depending on environmental conditions. Atmospheric transport of these compounds can result in contamination of sites away from any source of emissions (Lohman and Seigneur 2001; Garban et al. 2002). According to Bennett et al. (1998), Van Pul et al. (1998), and Beyer et al. (2000), TCDD dioxin congeners and OCDD can travel hundreds of kilometers. Volatility and transport over long distances is a phenomenon also known for polychlorinated biphenyls (PCBs). These compounds are stable and can cover distances greater than 1,000 km from the location of emissions and be found in isolated areas (Teil et al. 2004). Thus, contamination of vegetation in a given site is not exclusively linked to the nearest source. Generally, less volatile compounds are deposited predominantly in a restricted area around the emission source, while volatiles are spread more widely. Thus, contaminants are transported in the atmosphere down to the laminar layers circulating around the plant allowing the interaction between contaminants and leaf surfaces (Bakker et al. 2001). Gaseous deposition, dry or wet deposition of particles, and root absorption are at the origin of contact between POPs and plants (Welsch-Pausch et al. 1995; McLachlan 1999; Bakker et al. 2000; Bakker et al. 2001; Teil et al. 2004). Since POPs are highly lipophilic and thus poorly soluble in the sap of plants (Simonich and Hites 1994), contamination by root absorption is considered negligible in fodder (Wild and Jones 1992; Welsch-Pausch et al. 1995; Kipopoulou et al. 1999). Fodder contamination is mainly driven by atmospheric deposition (Smith and Jones 2000; Thomas et al. 2002). In contrast, wet deposition (solubilization of pollutants in rain or fog) is limited due to the hydrophobicity of the molecules.

Many factors like environmental conditions, characteristics of plants, or physicochemical properties of compounds, influence the type of deposit and the amount of pollutants adsorbed on the leaves. Plant characteristics such as hairiness of the leaf, the composition of the cuticle or the plant architecture affect the POP concentrations from one plant to another (Howsam et al. 2000; Bakker et al. 2001; Wild and Jones 1992). The cuticle, wax rich, contributes to the accumulation of lipophilic molecules (Müller et al. 2001; Smith et al. 2001) via cutin (Thomas et al. 1998). The outside temperature is also determining the form in which POPs will be present in the atmosphere (Howsam et al. 2000; Bakker et al. 2001; Blais et al. 2003) as well as wind direction (Bakker et al. 2001; Lohman and Seigneur 2001; Teil et al. 2004; Smith et al. 2001). Finally, the rain can modulate the deposit through leaching or increased wet deposition.

Performance and density of forage production are also explanatory factors of pollutant concentrations in fodder via the exchange surface with the gas phase (Smith et al. 2001). For example, the deposition surface of the plant is 6 to 14 times higher than that of the ground on which they are planted (Simonich and Hites 1994). When contaminants are deposited on the ground, they tend to accumulate in the surface horizon, i.e., the first 5 cm (Fries 1982; Stevens and Gerbec 1988; Jones et al. 1989). Due to their lipophilicity and low aqueous solubility, POPs are generally strongly adsorbed to soil components. Their adherence to soil components depends on their physico-chemical properties as vapor pressure (Henry constant), K_{ow} (partition coefficient octanol water), and climatic factors (Duarte-Davidson and Jones 1996). Soil characteristics such as organic matter composition, organic carbon content, acidity, or redox potential, control the processes of absorption, and desorption of contaminants and their distribution in the liquid solid and gaseous phases (Billeret et al. 2000; Chiou et al. 2000; Huang et al. 2003).

Under certain circumstances, POP concentrations in soil can reach 1,000 ng/kg dry matter for PCDD/Fs and 50 μg/kg for PCBs (Krauss and Wilcke 2003). In plants, contamination levels are generally lower and do not exceed values of a few tens of nanograms for PCDD/F and 1–2 μg PCB per kg of forage dry matter (Costera-Pastor et al. 2006; Welsch-Pausch et al. 1995). Such values are of course very fluctuating and are dependent on the type of contamination. Thus, a chronic contamination at a low level will result in a contaminant accumulation in the soil (memory effect), while the forage may be slightly contaminated (permanent renewal of the plant mass). In contrast, when the contamination is severe but with a short duration, the fodder may be more contaminated than soil. Thus, both soil and fodder matrices may be risk matrices for the food chain.

If fodder intake by ruminants is well-known and controlled, it has to be recalled that involuntary soil ingestion by grazing animals has not been really studied in the past decades. The following section summarizes the current knowledge on soil ingestion in ruminants reared outdoors.

Soil ingestion by grazing ruminants

Soil may be ingested by grazing ruminants, either inadvertently or by ingestion of plants contaminated with soil or feed (hay and salt licks) deposited on the ground. Thus, in contaminated areas soil is a risk matrix in terms of entry of contaminants into the food chain (Mamontova et al. 2007). Indeed, even if soil ingestion will always be limited and much lower than the ingestion of forage, the differential contamination of soils and forages can lead to exposure via both matrices. Therefore, it appears necessary to characterize soil ingestion by grazing cattle in various rearing systems.

Soil ingestion by wild animals (Beyer et al. 1994), sheep (Healy 1967), or cattle (Healy 1968; Fries 1982) has been estimated using indigestible markers such as insoluble ash in hydrochloric acid or titanium. However, the available data on soil ingestion by grazing livestock have been obtained under conditions that do not correspond to today's practices. Thus, these data are not easily transferable to the current grazing systems (Healy 1968; Mayland et al. 1975; Thornton and Abrahams 1983). It should also be noted that soil ingestion is necessarily variable depending on rearing and feeding practices. Fries (1982) described soil ingestion levels ranging from 1 to 3 % of total dry matter. For Healy (1968) and Thornton and Abrahams (1983), daily ingestion of soil could reach 1 kg of dry matter in the worst conditions of pasture. More recently, Jurjanz et al. (2012) sought to determine the amounts of soil ingested in dairy cows under intensive grazing. These authors have focused their attention on grazing pressure and the daily allowance of fodder. Figures 1 and 2 indicate an inverse relationship between the amount of grass available for cattle and soil ingestion. They highlight that when the fodder is abundant (potential ingestion of more than 15 kg of dry matter per cow per day), soil ingestion is very limited and does not exceed 2.5 % of the total dry matter intake (Fig. 1). In situations where forage supply is reduced, the amounts of ingested soil rapidly increase exceeding 1 kg of dry matter per cow per day. Figure 2 also shows that grass height less than 50 mm resulted in an increased risk of soil ingestion. This study provided an interesting perspective on soil ingestion by ruminants and revealed a certain number of factors of variation. It should also be noted that the rate of soil ingestion may also be linked to the animal species and the mode of grasping forage (sheep shearing shaves for example). Being aware that in contaminated areas soil may be more contaminated than fodder, the contribution of soil to the POP exposure of ruminants needs to be taken into account when assessing the risk of POPs transfer in the food chain.

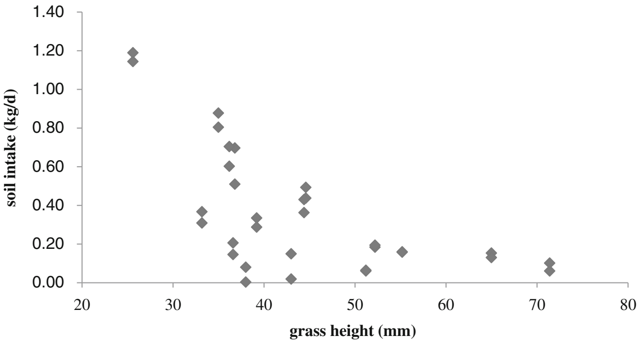


Fig. 2 Relationship between grass height and soil intake during grazing

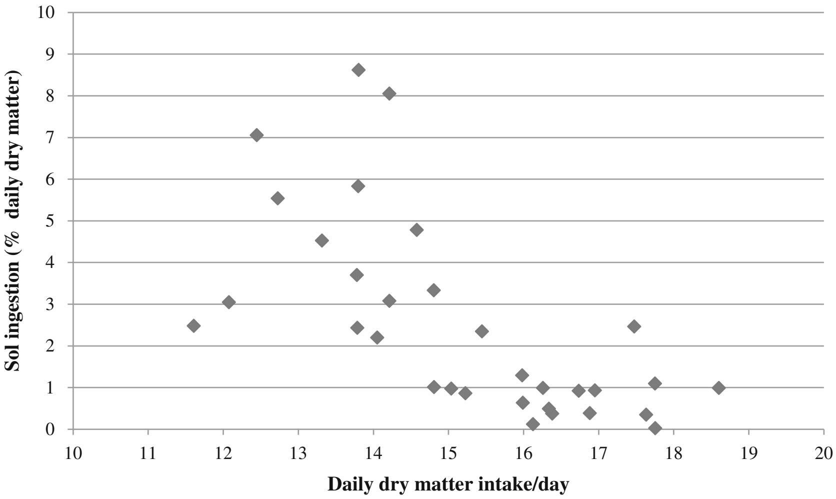
Bioavailability of POPs from fodder and soil and transfer to animal products

Several studies in ruminants have been performed in order to characterize the contamination process in ruminants (Fries et al. 1973; Thomas et al. 1999; Rossi et al. 2010; McLachlan 1994; Huwe and Smith 2005). This process is based on two successive steps:

- An initial phase where the pollutants accumulate in adipose tissue and liver
- An equilibrium phase where the contaminant concentrations in the target tissues remain constant

POP bioavailability, including absorption, distribution, metabolism, and excretion, is depending on the physical and chemical properties of the different compounds. Thus,

Fig. 1 Relationship between daily grass intake and soil ingestion



for low chlorinated compounds like PCB 74, Thomas et al. (1999) showed that intestinal absorption was high and close to 81.6 %, while only 18.4 % was excreted in feces. After absorption and distribution in various tissues and organs via the blood stream, 51 % of the amount absorbed was metabolized. The remaining part was stored in the adipose tissue and contributed to the milk excretion by 36.8 %. For highly chlorinated PCB congeners, Thomas et al. (1999) demonstrated that the absorption of PCB 138 for example was 54 % and a main part was excreted in feces. Once absorbed, PCB 138 was stored in the fatty tissues of animals (not metabolized) and excreted in the milk by 74 %. McLachlan (1994) observed that the “excretion/ingestion” ratio may even be higher than 100 % (106.5 %) for a given compound in case of mobilization and excretion of PCBs previously stored in the body fat of the animals.

Since ruminants are potentially exposed to contaminated soil and plant matrices, there is a need to estimate the bioavailability of POP from each of these matrices.

Transfer rate of POPs from fodder to milk

Transfer rates of pollutants from contaminated fodder need to be determined at equilibrium when: “POP excretion/POP ingestion=constant=transfer rate” (Jilg et al. 1992; McLachlan and Richter 1998; Fries et al. 1999; Winters et al. 2000; Richter and McLachlan 2001; Costera-Pastor et al. 2006). In the dairy goat, Costera-Pastor et al. (2006) found for example that 2,3,7,8-TCDD from “naturally” contaminated hay (area polluted by a defective incinerator) was transferred at 40 % to milk. For DL-PCBs, transfer rates exceeded 80 % for PCB 105, 118, and 157. Transfer rates were more variable for NDL PCBs, 5 % (PCB 101) to over 40 % (PCB 118, 153, and 180). Fournier et al. (2012) studied the transfer rates of POP from contaminated corn silage collected in the PCB contaminated area of St. Cyprien (fire of a wood industry in 2009). In this study, milk contamination increased very fast and after the second week of exposure to corn silage (4.65 pg toxic equivalent quantities (TEQ)/g) the regulatory threshold of 6 pg TEQ/g fat (MG) was exceeded. At plateau, contamination levels were around 20 pg TEQ per g of fat and average transfer rates were estimated at 53 % for DL-PCBs and 47 % for the sum DL-PCBs+PCDD/Fs. These two examples indicate that POPs adsorbed to contaminated fodder are rapidly and highly transferred to milk, suggesting high bioavailability of these organic pollutants. These results also point out that animal reared in contaminated areas result in animal products which may quickly exceed the regulatory thresholds defined in EU regulations 1881/2006 and 1259/2011, and therefore become unfit for human consumption.

Relative bioavailability of soil bound POPs

Ounnas et al. (2010) established the transfer rates of soil bound PCBs to milk under controlled conditions. The animals were exposed for 80 days to an experimental feed containing 5 % of PCB contaminated soil. Transfer rates of PCBs were found between 6 and 62 % depending on the compounds, with an average value of 44 %. These transfer rate levels appeared similar to those obtained with POP-contaminated fodder (Fournier et al. 2012; Costera-Pastor et al. 2006), suggesting that soil-bound POPs are also highly bioavailable. Although, several authors suggested that organic pollutants would be less available from soil matrices due to their high affinity for organic matter (Pu et al. 2006; Kookana 2011). In order to properly characterize the bioavailability of soil bound POPs, the relative bioavailability (RB) approach (Littell et al. 1997) has been used with the aim to estimate the soil’s ability to retain contaminants or not during the digestive process. These approaches involve two groups of animals subjected to increasing doses of either the “contaminated soil” matrix or a reference matrix (usually oil). This approach is based on the assumption that the concentration of a compound in the tissues or products of excretion is directly proportional to the absorbed dose. If linear responses are obtained, the relative bioavailability is the ratio between the slopes of the dose–response curves for both matrices.

This approach has been applied to evaluate the relative bioavailability of soil bound I-PCBs in lactating goats (Jondreville et al. 2011). RB values varied from 36 to 50 % for PCB 118, 138 and 153, and 73 % for PCB 180. Mean NDL PCB RB was 50 % (Fig. 3), suggesting that half of the soil pollutants were extracted from the soil during the digestive process. Since a significant part of soil contaminants may be extracted during the digestive phase, soil needs to be considered as a risk matrix.

Overall, the recent published data indicate that in case of ruminant exposure to POPs via contaminated fodder or soil

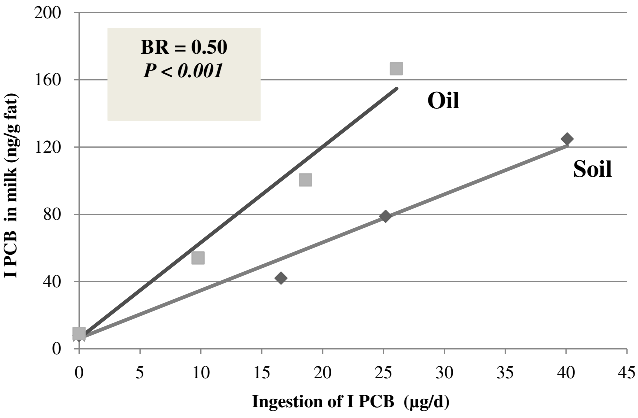


Fig. 3 Relative bioavailability of soil bound PCBs in the lactating goat

matrices, a significant part of these pollutants will be available to the animal and will result in the contamination of animal products. Furthermore, Costera-Pastor et al. (2006) and Fournier et al. (2012) clearly demonstrated that exposure to contaminated fodder rapidly results in animals products unfit for human consumption. In both studies, the threshold values were exceeded in less than 20 days. The seasonal and spatial variability of polychlorinated biphenyls (PCBs) in vegetation and cow milk was also studied in a high altitude pasture in the Alps (1900 m a.s.l.). PCB contamination in vegetation shows a concentration peak in June, which was mainly interpreted as the consequence of a temporary PCB enrichment of the air layer above the ground due to net emission fluxes from the soil (Tato et al. 2011). Lake et al. (2013) also observed seasonal variation in the levels of PCDD/Fs, PCBs, and PBDEs in cow milk. Changes in contaminant inputs from grass and silage were identified as being the most important source of these fluctuations.

Decontamination potential in ruminants

Several studies were aimed at assessing the decontamination process in ruminants (Fries et al. 1973; Thomas et al. 1999; Rossi et al. 2010; McLachlan 1994; Huwe and Smith 2005; Rychen et al. 2012). These authors suggested two distinct phases including (1) a rapid decontamination phase, characterized by a high flow of contaminants from the peripheral compartment to the blood, and (2) a slow decontamination phase, during which the exchange between the blood and the peripheral compartment is more limited. The two paragraphs below reveal that the decontamination process is related to the physiological status of the animals.

Decontamination of ruminants via excretion in milk

Milk excretion is considered as the major route of elimination of POPs in ruminants (Glynn et al. 2009; Rossi et al. 2010): it is linked to the physiological status of the animal (Gill et al. 1992; Thomas et al. 1999; Chamberland et al. 1994; Glynn et al. 2009) and to the nature of the compounds (McLachlan et al. 1990; Rossi et al. 2010). In a study reported by Fries et al. (1973), nine cows were exposed daily to 200 mg/day of Aroclor 1254 (a commercial mixture of PCBs) for 60 days. After cessation of exposure, these authors demonstrated a 50 % decrease in the concentration of PCBs in the milk during the first 15 days (phase 1), after 15 days the concentrations decreased less rapidly (phase 2). Thomas et al. (1999) have observed that the burden of PCBs in the body decreases by 25 % during the lactating period. According to Rossi et al. (2010), the energy balance of the animal is the main factor that affects the excretion rate of PCBs in milk. Indeed, the body fat mobilization that occurs after parturition increases the excretion of PCBs in milk. In contrast, reduced fat mobilization when the energy balance is positive helps to lower the concentration of PCBs in milk.

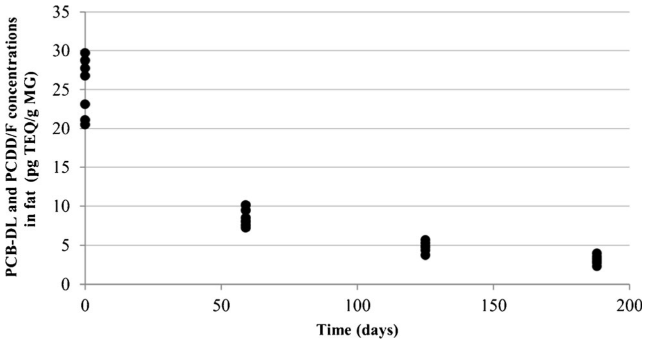


Fig. 4 Decontamination kinetics of DL PCB in growing limousine heifers (*n*=7; threshold value, 4 pg TEQ/g fat matter)

Thus, milk production directly results in the decontamination of dairy cows (Gill et al. 1992; Glynn et al. 2009; Rossi et al. 2010). Recently, in the lactating goat, Fournier et al. (2012) showed that the level of PCB in milk may drop from 20 pg TEQ/g fat to less than 6 pg TEQ/g fat (maximal acceptable level) in less than 21 days after stopping the exposure of dairy ruminants to contaminated corn silage. Thus, as soon as dairy ruminant exposure to organic pollutants is stopped, these animals undergo a decontamination phase which is linked to the pollutant excretion in milk. The time necessary to reach acceptable levels in milk is depending on three major parameters: the initial level of contamination, the level of milk production and the adipose status of the animals. Overall, according to the available literature, lactating ruminants may be decontaminated in a few weeks.

Decontamination of ruminants via increase of the body mass

It is established that pregnant females transfer part of the pollutant load in utero. Furthermore, after birth, the calf will be exposed to pollutants from mother milk. Thus, calves from contaminated herds generally present higher POP levels than

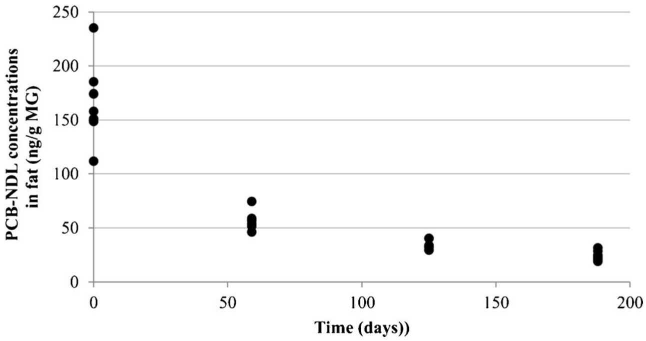


Fig. 5 Decontamination kinetics of NDL PCB in growing limousine heifers (*n*=7; threshold value, 40 ng/g fat matter)

their mother (Rychen et al. 2011). The question of farmers owning such contaminated young bovines is to apply adequate decontamination practices in order to put on the market bovines with POP levels below the regulatory threshold values (EU Regulations No. 1881/2006, 1259/2011).

Gill et al. (1992) have studied the PCB depuration kinetics in calves initially contaminated during suckling. Eight growing females calves needed a 13-month period safe feeding to lower the PCB load from 0.42 to 0.07 μg/g fat. As part of a national program aimed at controlling organochlorine contaminants levels in adipose tissue of farm animals in Sweden, Glynn et al. (2009) established correlations between the levels of POPs, the weight gain of the animals and the volume of their adipose tissue. These authors observed a “growth dilution” and demonstrated an inverse relationship between the pollutants levels and the volume of adipose tissue of the animals.

Chamberland et al. (1994) reported an experiment where 65 heifers were removed from a PCB contaminated area and housed in an experimental farm for a period of 10 months to evaluate their potential of decontamination. Pericaudal biopsies were carried out at day “0, 117, 203” in order to determine PCB levels in the body fat. These authors showed a decrease in the concentration of PCBs in body fat, from 0.52 μg/g fat the first day of the study to 0.09 μg/g fat at the end of the study. According to these authors this result was due either to the compound metabolism of to the pollutant dilution in the organism (increased volume of adipose tissue) of animals during their growth. Chamberland et al. (1994) favored the second hypothesis by observing a decrease in the concentration of PCB 118, 138, 153, 180, compounds known to be highly resistant to metabolism.

Such observations were confirmed by Rychen et al. (2012) who characterized the decontamination process of cattle through a 6-month growing period. Eight 10-month-old Limousine heifers with an initial PCB contamination level higher than 20 pg/g TEQ fat (DL PCB) and 100 ng/g fat (NDL PCB) were moved from a contaminated area to an experimental farm located in an area free from any PCB contamination. The heifers were reared under controlled conditions and fed a standard diet based on grass silage and hay in order to achieve a daily weight gain of 800 g/day. These animals were subjected to a pericaudal biopsy every 2 months. Figures 4 and 5 show the evolution of the concentrations of DL PCBs and NDL PCBs. At the end of the study, all animals had reached levels of PCBs below the threshold of regulatory 4 pg TEQ/g fat for DL PCBs and 40 ng/g for NDL PCBs. The time needed to reach these threshold values was depending on the compounds: about 6 months for DL PCBs (Fig. 4) and less than 4 months for NDL PCBs (Fig. 5). To achieve such a level of decontamination, a mean weight gain of more than 150 kg was necessary. These results suggest a growth dilution and a significant increase in fat mass as suggested by Glynn et al. (2009) and Chamberland et al. (1994).

Conclusion

In POP-contaminated areas, forage and involuntary ingestion of soil are risk matrices for livestock and their food products. The most recent results clearly demonstrate that the pollutants from these matrices are highly bioavailable, and therefore a significant part of contaminants from fodder or soil will be absorbed, distributed, metabolized, and/or excreted by ruminants. If the published results are often incomplete and limited, i.e., limited number of molecules, animal replicates, fodder, type of soil, they represent a sufficient basis to clearly identify main risks of transfer of POPs into the food chain. In historical contaminated areas, feeding practices shall strictly avoid soil ingestion. A particular attention has to be paid to young animals whose contamination level may be much higher than their parent’s one.

Decontamination of cattle appears realistic for lactating and growing animals. In contaminated areas, the main challenge will be to stop the exposure of animals through feeding uncontaminated fodder and avoiding exposure to contaminated soil. Such recommendations appear valid in areas where the contamination process is stopped and where newly produced fodder is demonstrated to be uncontaminated (absence of significant transfer from soil to plants). It has to be recalled that decontamination is linked to excretion of POP in milk fat or dilution process which requires an important weight gain of the animals, i.e., several months needed to fatten young bovines.

Further research need be conducted to precisely identify (1) risk feeding practices, (2) distribution of POPs in the body, and (3) the kinetic parameters of the contamination and decontamination in relation with fat deposition or release. In contaminated areas, livestock producers are expecting scientific data to help them to anticipate, predict, and manage POP crises.

References

Antignac JP, Marchand P, Gade C, Matayron G, Qannari EM, Le Bizec B, André F (2006) Studying variations in the PCDD/PCDF profile across various food products using multivariate statistical analysis. Anal Bioanal Chem 384:271–279

Bakker MI, Casado B, Koerselman JW, Tolls J, Kollöffel C (2000) Polycyclic aromatic hydrocarbons in soil and plant samples from the vicinity of an oil refinery. The Sci Tot Environ 263:91–100

Bakker MI, Tolls J, Kollöffel C (2001) Deposition of atmospheric semivolatile organic compound to vegetation. American Chemical Society Chapter 16:218–236

Bennett DH, McKone TE, Matthies M, Kastenberg WE (1998) General formulation of characteristic travel distance for semivolatile chemicals in a multimedia environment. Environ Sci Technol 32:4023–4030

Beyer WN, Connor EE, Gerould S (1994) Estimates of soil ingestion by wildlife. J Wildlife Manage 58:375–382

Beyer A, Mackay D, Matthies M, Wania F, Webster E (2000) Assessing long-range transport potential of persistent organic pollutants. Environ Sci Technol 34:699–703

Billeret M, Berny P, Mazallon M, Buronfosse T (2000) Bioavailability of polycyclic aromatic hydrocarbons and polychlorinated biphenyls in rats from naturally contaminated soils—preliminary evaluation of the influence of soil parameters. Env Tox Chem 19:2614–2620

Blais JM, Kimpe LE, Backus S, Comba M, Schindler DW (2003) Assessment and characterization of PCB near a hazardous waste incinerator: analysis of vegetation, snow and sediments. Environ Toxicol Chem 22:126–133

Chamberland G, Tremblay A, Lamothe P, Gignac M (1994) Clinical chemistry, growth and PCB levels in beef cattle exposed to a PCB fire. Toxicol Environ Chem 44:177–187

Chiou CT, Kile DE, Rutherford DW, Sheng G, Boyd SA (2000) Sorption of selected organic compounds from water to a peat soil and its humic-acid and humin fractions: potential sources of the sorption nonlinearity. Environ Sci Technol 34:1254–1258

Costera-Pastor A, Feidt C, Marchand P, Le Bizec B, Rychen G (2006) PCDD/F and PCB transfer to milk in goats exposed to long-term intake of contaminated hay. Chemosphere 64:650–657

Duarte-Davidson R, Jones KC (1996) Screening the environmental fate of organic contaminants in sewage sludges applied to agricultural soils: II. The potential for transfers to plants and grazing animals. Sci. Total Environ 185:59–70

Fournier A, Feidt C, Marchand P, Le Bizec B, Rychen G (2012) PCB contamination and decontamination of ruminants accidentally exposed to contaminated corn silage. 7th International PCB Workshop, 27–31 May 2012, Arcachon, France

Fries GF (1982) Potential polychlorinated biphenyl residue in animal products from application of contaminated sewage sludge to land. J Environ Qual 11:14–20

Fries GF, Marrow GS, Gordon GH (1973) Long-term studies of residue retention and excretion by cows fed a polychlorinated biphenyl (Aroclor 1254). J Agric Food Chem 21(1):117–121

Fries GF, Paustenbach DJ, Mather DB, Luksembur WJ (1999) A congener specific evaluation of transfer of chlorinated dibenzo-*p*-dioxins and dibenzofurans to milk of cows following ingestion of pentachlorophenol-treated wood. Environ Sci Technol 33:1165–1170

Garban B, Ollivon D, Teil MJ, Blanchard M, Blanchoud H, Moteley-Massei A, Chesterikoff C, Hanselin L, Rolet J, Le Genti L, Chevreuil M (2002) Rapport PIREN-Seine 31p.

Gill I, Roberts G, Galvin J (1992) Management of land and livestock contaminated with polychlorinated biphenyls. Aust Vet J 69:155–157

Glynn A, Aune M, Nilsson I, Darnerud P, Bignert A, Nordlander I (2009) Declining levels of PCB, HCB and p,p′-DDE in adipose-tissue from food producing bovines and swine in Sweden 1991–2004. Chemosphere 74:1457–1462

Healy WB (1967) Ingestion of soil by sheep. Proc N Z Soc Anim Prod 27:109–120

Healy WB (1968) Ingestion of soil by dairy cows. New Zeal J Agr Res 11:487–499

Howsam M, Jones KC, Ineson P (2000) PAHs associated with the leaves of three deciduous tree species. I. Concentrations and profiles. Environ Pollut 108:413–424

Huang W, Peng P, Yu Z, Fu J (2003) Effects of organic matter heterogeneity on sorption and desorption of organic contaminants by soils and sediments. Appl Geochem 18:955–972

Huwe JK, Smith DJ (2005) Laboratory and on-farm studies on the bioaccumulation and elimination of dioxins from a contaminated mineral supplement fed to dairy cows. J Agric Food Chem 53:2362–2370

Jilg T, Müller W, Hagenmaier H, Päpke O (1992) Carry-over of polychlorinated dibenzodioxins (PCDD) and dibenzofurans (PCDF) from feed into the milk of the dairy cow. Agribiol Res 45:303–310

Jondreville C, Fournier A, Ounnas F, Julien-David D, Marchand P, Venisseau A, Le Bizec B, Travel A, Rychen G, Feidt C (2011) Relative bioavailability of indicator polychlorobiphenyls present in soil to farm animals: laying hens and lactating goats. Congress Dioxin 2011, Brussels

Jones KC, Stratford JA, Tidridge P, Waterhouse KS, Johnston AE (1989) Polynuclear aromatic hydrocarbons in an agricultural soil: long-term changes in profile distribution. Environ Pollut 56:337–351

Jurjanz S, Feidt C, Pérez-Prieto LA, Ribeiro Filho HMN, Rychen G, Delagarde R (2012) Soil intake of lactating dairy cows in intensive strip grazing systems. Animal 6:1350–1359

Kipopoulou AM, Manoli E, Samara C (1999) Bioconcentration of polycyclic aromatic hydrocarbons (PAHs) in vegetables grown in an industrial area. Environ Pollut 106:369–380

Kookana RS (2011) Fate of Persistent Organic Pollutants (POPs) in Soils and Their Effect on Food Safety. http://www.agnet.org/library.php?func=viewid=20110804154803&type_id=4. Accessed August 2012

Krauss M, Wilcke W (2003) Polychlorinated naphthalenes in urban soils: analysis, concentrations, and relation to other persistent organic pollutants. Environ Pollut 122:75–89

Lake IR, Foxal CD, Fernandes A, Lewis M, Rose M, White O, Dowding A (2013) Seasonal variations in the levels of PCDD/Fs, PCBs and PBDEs in cows' milk. Chemosphere 90:72–79

Littell RC, Henry PR, Lewis AJ, Ammerman CB (1997) Estimation of relative bioavailability of nutrients using SAS procedures. J Anim Sci 75:2672–2683

Lohman K, Seigneur C (2001) Atmospheric fate and transport of dioxins: local impacts. Chemosphere 45:161–171

Mamontova EA, Tarasova EN, Momontov AA, Kusmin MI, McLachlan MS, Khomutova MI (2007) The influence of soil contamination on the concentrations of PCBs in milk in Siberia. Chemosphere 67:S71–S78

Mayland HF, Florence AR, Rosenau RC, Lazar VA, Turner HA (1975) Soil ingestion by cattle on semiarid range as reflected by titanium analysis of feces. J Range Manage 28:448–452

McLachlan MS (1994) Model of the fate of hydrophobic contaminants in cows. Environ Sci Technol 28:2407–2414

McLachlan MS (1999) A framework for the interpretation of measurements of SOCs in plants. Environ Sci Technol 33:1799–1804

McLachlan MS, Richter W (1998) Uptake and transfer of PCDD/Fs by cattle fed naturally contaminated feedstuffs and feed contaminated as a result of sewage sludge application. 1. Lactating cows. J Agric Food Chem 46:1166–1172

McLachlan MS, Thoma H, Reissinger M, Hutzinger O (1990) PCDD/F in an agricultural food chain Part 1: PCDD/F mass balance in a lactating cow. Chemosphere 20:1013–1020

Müller JF, Hawker DW, McLachlan MS, Connel DW (2001) PAHs, PCDD/Fs, PCBs and HCB in leaves from Brisbane, Australia. Chemosphere 43:507–515

Ounnas F, Feidt C, Toussaint H, Rychen G, Marchand P, Le Bizec P, Jurjanz S (2010) Polychlorinated biphenyl and low polybrominated diphenyl ether transfer to milk in lactating goats chronically exposed to contaminated soil. Environ Sci Technol 44(7):2682–2688

Pu X, Lee LS, Galinsky RE, Carlson GP (2006) Bioavailability of 2,3',4,4',5-pentachlorobiphenyl (PCB118) and 2,2',5,5' - tetrachlorobiphenyl (PCB52) from soils using a rat model and a physiologically based extraction test. Toxicology 217:14–21

Richter W, McLachlan MS (2001) Uptake and transfer of PCDD/Fs by cattle fed naturally contaminated feedstuffs and feed contaminated as a result of sewage sludge application. 2. Non-lactating cows. J Agric Food Chem 49:5857–5865

Rossi F, Bertuzzi T, Vitali A, Rubini A, Masoero F, Morlacchini M (2010) Monitoring of the declining trend of polychlorobifenyls concentration in milk of contaminated dairy cows. Ital J Anim Sci 9:88–92

Rychen G, Ducoulombier C, Grova N, Jurjanz S, Feidt C (2005) Terms and risk of transfer of persistent organic pollutants into milk. Inra Prod Anim 18:355–366

Rychen G, Fournier A, Pinte J, Marchand P, Venisseau A, Toussaint H, Le Bizec B, Jurjanz S, Feidt C (2011) Decontamination of accidentally PCB contaminated heifers and transfer of PCB from mother to calves. 30th International Symposium on Halogenated Environmental Organic Pollutants and POPs, 21–25 August 2011, Brussels, Belgium. Organohalogen Compounds 73:280–283

Rychen G, Fournier A, Pinte J, Marchand P, Venisseau A, Toussaint H, Le Bizec B, Jurjanz S, Feidt C (2012) Lifestock exposure to polychlorobiphenyls and potential of decontamination. 7th International PCB Workshop, 27–31 May 2012, Arcachon, France

Simonich SL, Hites RA (1994) Vegetation-atmosphere partitioning of polycyclic aromatic hydrocarbons. Environ Sci Technol 28:939–943

Smith KEC, Jones KC (2000) Particles and vegetation: implications for the transfer of particle-bound organic contaminants to plants. Sci Total Environ 246:207–236

Smith KEC, Thomas GO, Jones KC (2001) Seasonal and species differences in the air-pasture transfer of PAHs. Environ Sci Technol 35:2156–2165

Stevens JL, Gerbec EN (1988) Dioxin in the agricultural food chain. Risk Anal 8:329–335

Tato L, Tremolada P, Ballabio C, Guazzoni N, Parolini M, Caccianiga M, Binelli A (2011) Seasonal and spatial variability of polychlorinated biphenyls (PCBs) in vegetation and cow milk from a high altitude pasture in the Italian Alps. Environ Pollut 159:2656–2664

Teil MJ, Blanchard M, Chevreuil M (2004) Atmospheric deposition of organochlorines (PCBs and pesticides) in northern France. Chemosphere 55:501–514

Thomas GO, Smith KEC, Sweetman AJ, Jones KC (1998) Further studies of the air-pasture transfer of polychlorinated biphenyls. Environ Pollut 102:119–128

Thomas GO, Sweetman AJ, Jones KC (1999) Input–output balance of polychlorinated biphenyls in a long-term study of lactating daily cows. Environ Sci Technol 33:104–112

Thomas GO, Jones JL, Jones KC (2002) Polychlorinated dibenzo-*p*-dioxin and furan (PCDD/F) uptake by pasture. Environ Sci Technol 36:2372–2378

Thornton I, Abrahams P (1983) Soil ingestion—a major pathway of heavy metals into livestock grazing contaminated land. The Sci Tot Environ 28:287–294

Van Pul WAJ, de Leeuw FAAM, Van Jaarsveld JA, Van der Gag MA, Sliggers CJ (1998) The potential for long-range transboundary atmospheric transport. Chemosphere 37:113–141

Welsch-Pausch K, McLachlan MS, Umlauf G (1995) Determination of the principal pathways of polychlorinated dibenzo-*p*-dioxins and dibenzofurans to *Lolium multiflorum* (Welsh Ray Grass). Environ Sci Technol 29:1090–1098

Wild SR, Jones KC (1992) The polynuclear aromatic hydrocarbon (PAH) content of herbage from a long-term grassland experiment. Atmosph Environ 26a(7):1299–1307

Winters D, Fries GF, Lorber M, Ferrario J, Byrne C (2000) A study of the mass balance of dioxins and furans in lactating cows in background conditions. Part 1: study design and analysis of feed. Organohalogen Compd 46:534–537