Learning from Dioxin & PCBs in meat – problems ahead?

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Abstract. Persistent organic pollutants (POPs) including polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs; “Dioxins”), or polychlorinated biphenyls (PCBs) are widely recognized environmental and food contaminants. More than 90% of PCDD/Fs and PCB exposure of the average population stem from animal based food including meat. While average PCDD/F and PCB levels have decreased compared to levels 1980s, still contamination above regulatory limits are observed and a share of the population is above the tolerable daily intake recommended by the WHO. For PCBs the contamination of feed and food along the life cycle from production, use, recycling, end of life and related contaminated sites has been documented and can be seen as a model. Furthermore, it has been recently discovered that levels of PCBs in feed and soil below regulatory limits can result in meat contamination above EU regulatory limits. In particular, beef meat and chicken meat/eggs have been found very sensitive towards PCB contamination in the environment (soil and feed) but also in stables (paints and sealants). For PCDD/Fs, the major exposure pathways are feed, feed additives and contaminated sites. Chlorinated paraffins have substituted PCBs the last 40 years in open application and short chain chlorinated paraffins (SCCPs) were recently (05/2017) listed in the Stockholm Convention. Furthermore, brominated and fluorinated POPs have been listed in the Convention. All these POPs groups can accumulate in meat animals. For these new listed POPs no regulatory limits in food including meat has been established yet. Initial information on presence and risk of new listed POPs to food animals is compiled. A more systematic assessment of exposure and risks of POPs to food animals/meat is needed.

1. Introduction
Polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans (PCDD/Fs), and polychlorinated biphenyls (PCBs) are widely recognized environmental and food contaminants. In the EU, the Commission Regulation (EC) No 1881/2006 of 19 December 2006 sets maximum levels for PCDD/Fs and the sum of PCDD/Fs and dioxin-like PCBs (dl-PCBs) in certain foodstuffs (European Commission 2006). The regulation was amended by Commission Regulation (EU) No 1259/2011 of 2 December 2011 introducing new EU maximum levels for PCDD/Fs, for the sum of PCDD/Fs and dl-PCBs (based on WHO toxicity equivalency factors established in 2005, TEF2005) and for non-dioxin-like PCBs (ndl-PCBs) (European Commission 2011).

Most of the meat and milk samples on the European market meet the regulatory limits (EFSA 2012). Average PCDD/F and PCB levels have decreased compared to levels 1990s which is also reflected in decreasing PCB and PCDD/F levels in human milk (UNEP 2013). However, in recent 10 years more frequent contamination of meat and eggs with PCBs were detected (Weber et al. 2015). This was trigged by the inclusion of PCBs in the regulation since 2006.

In the past often feed incidents were responsible for exceeding maximum levels of PCDD/Fs and PCBs in food of animal origin (Tlustos et al. 2013; Malisch & Kotz 2014). In recent years also sheep (in particular liver) (EFSA 2011) and beef (Kamphues et al. 2011; Weber et al. 2014) from free-range production exceeded the existing maximum limits. Depending on the source, PCDD/Fs or dioxin-like PCBs can contribute in various ratios to TEQ with dl-PCB often as main contributor.

In addition to these chlorinated persistent organic pollutants in the last 8 years the first brominated and fluorinated POPs have been listed in the Stockholm Convention.
In the current paper experiences with PCBs, PCDD/Fs and selected new Stockholm Convention listed POPs and contamination of food animals/meat is compiled.

2. Samples of food contamination resulting from POPs
2.1. Experiences and lessons learnt from PCBs
PCB is the best assessed industrial POP in food and feed due to regulatory requirements and related monitoring and control. Food animals and related meat and other food products can become PCB contaminated along the life cycle of PCBs (Weber et al. 2015):

Exposure from PCB production sites via food animals
Animals and humans were exposed to PCBs around production sites. For the PCB production sites the contamination and exposure via animals/cattle in Anniston (ATSDR 2015) and Brescia/Italy have been documented (Turrio-Baldassarri et al. 2009). For the former PCB production site in Slovakia elevated human exposure was found up to 70 km from the plant with food animals as likely major human exposure pathway (Wimmerová et al. 2015).

Exposure of food animals from PCB use
PCBs have been used in “closed applications” (e.g. transformers, capacitors) and “open applications” (e.g. sealants and paints). Already in the 1970s/80s it had been discovered that PCB-paints used in fodder silos had contaminated cattle (Willett & Hass 1975; Deutscher Bundestag 1989). The recent cases of meat or egg contamination with PCB-paints on farms in Germany (Weber et al. 2015; Winkler 2016), France (Marchant 2017), Netherlands (Hoogenboom et al. 2014) and Switzerland (Zennegg et al. 2014) demonstrate that still PCB in open applications in particular paints exist and need further assessment and management.

PCB exposure of food animals from recycling of PCB containing oils and wastes
Although the recycling of PCB containing oils is not allowed, it can enters recycling cycles and has in some cases contaminated feed and related meat and other animal products. The largest and most costly (approx. 1 billion US$) PCB/Dioxin food scandal occurred in Belgium in 1999 where PCB oil were mixed with food fat impacting meat and eggs from more than 1500 farms (Fiedler et al. 2000; Larebeke et al. 2001). Another large meat contamination case with associated 100 million $ damage cost resulted from use/recycling of PCB-contaminating oil for drying of animal feed contaminating the Irish pork/meat with related recalls (Heres et al. 2010; Marnane 2012). Today in particular in developing countries still 14 million tonnes of PCB contaminated oil and equipment exist which is partly recycled in an uncontrolled manner. However, no monitoring of PCBs in food is established in developing countries and meat or other food contamination is normally not discovered.

PCB exposure of food animals from waste treatment and from contaminated soils
Free-range cattle are ingesting a considerable amount of soil when grazing. In recent survey in Germany contamination of beef meat from a range of sources mainly related to soil contamination were discovered:
- Flood plains of industrially impacted rivers
- Application of sewage sludge from 1960s and 1970s
- Sediment deposits on agricultural land
- Construction debris scattered and incorporated into soil of a pasture area
- Long term emission/deposition from industrial facilities (metal industry; incinerators)
- Areas where (agricultural) machinery was parked (dripping machinery/hydraulic oil)
- Use of former PCB-contaminated scrap yard as storage area for dung

1 If an egg is above the EU regulatory limit, also the chicken meat is above the regulatory limit.
The cases showed that in particular for suckling herds the dl-PCB levels of soils were mostly below 5 ng WHO-PCB-TEQ/kg dm and the feed in average below 0.2 ng WHO-PCB-TEQ/kg dm. As conclusion, meat of free range cattle in particular when calves are fed by milk of grazing cows for a longer period can exceed the EU-regulatory limits at relatively low soil levels (below 5 ng WHO-PCB-TEQ/kg dm) in combination with grass/feed levels around 0.15 ng TEQ/kg dm considerably below the EU-regulatory limits for feed (Weber et al. 2015). When calculating the total intake of the cows (consumption of 10 kg dm of grass/hay containing approx. 3 wt-% soil), a total intake of approx. 2 ng WHO-PCB-TEQ/day from soil and feed might be critical with regard to possible exceedance of the maximum limits for meat from beef in these cases. A systematic screening of PCB contaminated sites is needed for promoting safe feed and food (including meat) production.

2.2. Experience with polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)
PCDD/Fs are not produced intentionally but are emitted from a wide range of sources. The exposure of cattle and chicken and related meat can result from (Fiedler et al. 2000; Huwe et al. 2004; Malisch & Kotz 2014; Tlustos et al. 2013; Weber et al. 2015):
- PCDD/F contamination of feed
- PCDD/F contamination of feed additives (e.g. ZnO or CuSO₄)
- PCDD/F contaminated in stables and in animal bedding (e.g. PCP treated wood)
- PCDD/F contamination of soils and exposure from grazing (+ feed contamination)

All four exposure routes have resulted in meat contamination above regulatory limits. Since the largest amount of PCDD/Fs are from historic releases, the contamination of soils and related exposure of feed and cattle is likely the most relevant PCDD/F exposure route (Weber et al 2008, 2015).

2.3. New listed POPs of emerging concern
Perfluorooctanesulfonic acid (PFOS) and other per-/polyfluorinated alkylated substances (PFAS)
Since 2009 the first fluorinated POPs (PFOS and related precursor chemicals) have been listed in the Stockholm Convention and wide environmental pollution is discovered. Also these POPs can bio-accumulate in meat (Lupton et al. 2014). For PFOS transfer factors from environmental contamination to food animals have been established (Brambilla et al. 2014). Meat and other animal food can become major human exposure pathways for PFOS (Brambilla et al. 2014). Assessments of a PFOS/PFAS contaminated site impacting the groundwater of a farm has shown highest PFOS/PFAS levels in cattle of the farms up to ppm level in blood (Bräuning et al. 2017). Currently approx. 3000 PFAS are in use which are highly persistent or have highly persistent degradation products. There are currently no limit values in food including meat but only limit values for PFOS-selected PFAS in drinking water.

Polybrominated flame retardants
Since 2009 also some brominated flame retardants have been listed as POPs in the Stockholm Convention including polybrominated biphenyl ethers (PBDEs), polybrominated biphenyls (PBBs) and hexabromocyclododecane (HBCD). The most known cattle/meat incident with BFRs occurred 1973 when the Michigan Chemical Company producing PBB and magnesium oxide (a cattle feed supplement) by mistake sent 230 to 500 kg of PBB instead of MgO accidentally to Michigan Farm Bureau Services (Michigan Department of Community Health (2011). As a result of this incident, over 500 contaminated Michigan farms were quarantined, and approximately 30,000 cattle, 4,500 swine, 1,500 sheep, and 1.5 million chickens were destroyed, along with over 800 tons of animal feed, 8165 kg of cheese, 1134 kg butter, 5 million eggs, and 15400 kg of dried milk products (Michigan Department of Community Health 2011). 60% of the Michigan population still have high PBB levels 40 years after the incident (Department of Epidemiology - The Michigan PBB Registry 2017).
For PBDEs it has been shown that poultry and red meat consumption is a major contributor for PBDEs burden of the population in the US. Blood levels of five major PBDE congeners were associated with consumption of poultry fat and red meat fat (Fraser et al. 2009). \( \Sigma \text{PBDE} \) serum concentrations among vegetarians were approx. 25% lower than among omnivores (Frazer et al. 2009). In North America where the largest amount of PBDEs have been used, PBDE exposure to cattle/meat resulted from application of sewage sludge/biosolids (Rawn et al. 2017).

**Chlorinated paraffins**

Chlorinated paraffins are the chlorinated semivolatile organic compounds with highest production volume (>1 million tonne/year) having substituted PCBs in most open applications (Glüge et al. 2016). Most recently short chain chlorinated paraffins (SCCP) have been listed as POPs in the Stockholm Convention. Chlorinated paraffins can bioaccumulate in meat/fat (Ueberschär & Matthes 2004) and might pose a risk for future safe meat production considering the large use volumes in open applications.

2.4 Problematic chemicals in meat production with lower persistence not considered here

There are a wide range of problematic chemicals which are a threat for meat production including pesticides/biocides (e.g. recently fipronil, glyphosate), antibiotics and other veterinary drugs, PAHs or heavy metals. These chemicals are not addressed in this abstract with the focus on POPs but need to be controlled and reduced for healthy and environmental friendly meat production.

3. Governmental guidance documents and support

To support safe food production and to avoid dioxin and PCB contamination of food of animal origin (meat, milk and eggs) different governmental institutions have published guidance documents.

On international level the “Code of Practice for the Prevention and Reduction of Dioxin and Dioxin-like PCB Contamination in Food and Feeds” have been published 2006 (FAO 2006). Several documents were developed on EU E.g. “Evaluation of the Occurrence of PCDD/PCDF and POPs in Wastes and Their Potential to Enter the Food Chain” (Fiedler et al. 2000) or “Guidelines for the enforcement of provisions on dioxins in the event non-compliance with the maximum levels for dioxins in food (DG Sanco 2004).

Also guidance documents have been developed on national level. E.g. the German Environmental Ministry has published a guidance on “Environmental protection – pillar for food safety to avoiding dioxin and PCB entry” (BMU 2013). The Chamber of Agriculture of the federal state of Lower Saxony developed leaflet for cattle breeder to control Dioxin and PCBs input or on cultivation on contaminated land (Landwirtschaftskammer Niedersachsen 2011; 2014).

In the frame of the Stockholm Convention inventory guidance documents for individual POPs have been developed include a chapter for contaminated sites for PFOS, PBDEs, HBCD, PCNs, PCP and HCBD (Secretariat of the Stockholm Convention 2017). The development of inventories of POPs contaminated sites and the securing and remediation of these sites can improve feed and food/meat safety contributing to several Sustainable Development Goals (SDGs) (Bell et al. 2016).

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