SAFE FOOD PRODUCTION FROM FREE RANGE BEEF – MINIMIZING TEQ-LEVELS IN MEAT BY TRACKING PCB-SOURCES

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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[1] sets maximum levels for PCDD/Fs and the sum of PCDD/Fs and dioxin-like PCBs (dl-PCBs) in certain foodstuffs. The regulation was amended by Commission Regulation (EU) No 1259/2011 of 2 December 2011[2] 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).

Most of the meat and milk samples on the European market meet the regulatory limits[3]. In the past often feed incidents were responsible for exceeding maximum levels of PCDD/Fs and PCBs in food of animal origin[4-5]. In recent years also sheep (in particular liver)[6] and beef[7,8] from free range production exceeded the existing maximum limits. Floodplains of rivers with historic industrial inputs were discovered as possible exposure pathway in Germany[7a,b,c] and UK[8]. Depending on the source, PCDD/Fs or PCBs can contribute in various ratios to TEQ with dl-PCB often as main contributor. The German Environmental Ministry has therefore published a guidance on environmental protection as a basis for safe food production[9].

In the current study monitoring results of dl-PCB in beef and sheep from some federal states in Germany are presented with particular discussion of the exposure sources and critical PCB levels in soil and feed.

Materials and methods

Samples:

Meat samples of beef and sheep were collected as part of the official food control in different German federal states between 2009 and 2012. Soil and feed samples were collected for identification of possible contamination sources. In addition other potential materials of PCB sources were sampled at locations where meat exceeded EU-limits.

Analysis:

The food samples were analyzed by the competent laboratories for the official food control in the federal states according to Commission Regulation (EC) No 1883/2006 (until 2011)[10a] and Commission Regulation (EU) No 252/2012 (since 2012)[10b]. Feed samples were analyzed by the competent laboratories according to Commission Regulation (EU) No 278/2012[11].

Soil sampling and analysis were performed according to the Federal Soil Protection and Contaminated Sites Ordinance (BBodSchV) and analysed according to German guideline DIN 38414-24 or DIN ISO 10382. For specific materials like paints from silo or rubber belt, in-house methods were applied. Analyses of soil and materials were also performed with HRGC/HRMS at a resolution of 8000 – 10,000 with ^{13}C-isotope dilution method.

Results and discussion

(1) Animals raised on flood plains

Alluvial soil in flood plains along 400 km of the river Elbe was contaminated with high levels of PCDD/Fs (several 100 ng TEQ/kg dry matter (dm) in top layer[7a] and up to 7000 ng TEQ/kg in core layers[12]) from former magnesium production and organochlorine production in the Bitterfeld-Wolfen region[12]. Several research projects were carried out to assess what kind of feed harvest and cattle production is still possible on the contaminated flood plains[7,8]. A guidance document was developed for agricultural use of the floodplain areas addressing e.g. cutting height for grass (for silage or hay)[13].

Further studies were conducted in flood plains of some other German rivers. Also meat and liver of cows and sheep which were raised on some floodplain areas of rivers which were/are industrially influenced exceeded the maximum limits with dl-PCBs as main TEQ-contributor in meat. Comparing PCDD/F and PCB contribution to TEQ in soil, feed and meat, a strong shift towards a higher TEQ-contribution of dl-PCBs in meat was observed[14].

(2) Herds with other exposure sources

In areas other than floodplains, monitoring programes showed exceedance of maximum and/or action levels mainly by free range cattle (mostly as offspring from suckling cows) held in non-flooded areas. In most cases (>90%) the elevated levels were caused by dl-PCBs. In the assessment of cases from 2009 to 2013 a number of contamination sources of the individual herds were revealed:

- Sediments from a dredged water reservoir with elevated PCB and PCDD/F levels
- The application of sewage sludge to agricultural soil in the 1960s/1970s with higher loads of sludge.
- Construction debris scattered and incorporated into soil of a pasture area
- Long term deposition from industrial facilities
- Use of former PCB-contaminated scrap yard as storage area for dung
- Former use as military area
- Area of a former railway line with railway sleepers
- Impregnation of silos with PCB paint. These silos were constructed in the 1960s and 1970s
- Rubber belt from the 1980s with elevated PCB levels (used in the feeding trough for calves)

The fact that some point sources on the farms (PCB in silo painting or PCB in rubber belt) resulted in exceedance of the WHO-PCDD/F-PCB-TEQ food limit showed that not only free range cattle are exposed to PCBs (other than contaminated feed) but that PCBs in open application such as paints or sealants can still contribute to exposure of cattle. Painting of silos already caused a PCB-contamination of milk exceeding the former German regulatory limit for ndl-PCB in the 1980s. At that time a great number of farms were screened and the respective silos were removed. In the current case the WHO-PCDD/F-PCB-TEQ levels in meat of beef cattle exceeded the European maximum limit for meat (due to dl-PCBs) while the levels in milk from the same farm with same feed did not exceed the European maximum limit for milk. This shows that the WHO-PCDD/F-PCB-TEQ limit in meat is more sensitive towards dl-PCB-exposure compared to milk.

In the case of the rubber belt the source of PCBs was not clear. The rubber belt was manufactured in the 1980s and thus PCBs were unlikely added as flame retardant or softeners. The contamination of rubber by PCB was recently reported from Japan[15]. As source, high levels of unintentionally formed PCBs were identified contained in chlorinated paraffins which were added to the rubber as flame retardant[15]. Further assessment is needed here.

In some farms with affected cattle, sources were discovered such as construction debris or demolished buildings. This shows that also open PCB applications like sealants/caulking are contemporary PCB-sources.

For herds where no point source was found a detailed assessment of the PCDD/F and (dl-)PCB levels in soil and grass/hay was made to investigate if the levels in the meat could be explained by these routes of exposure. In one case 25 meat samples were analyzed for PCDD/Fs and dl-PCBs (Fig. 1). In most cases the main TEQ-contribution results from dl-PCBs (average contribution of 86 % to TEQ). Meat samples of beef cows (number of samples: 2) had WHO-PCDD/F-PCB-TEQ levels in the range of the maximum level of 4 pg WHO-TEQ/g fat. Samples from calves and other beef cattle were in most cases clearly above the maximum level while beef cows were in the range of the limit (Fig. 1). As conclusion, similar to humans, PCDD/F and PCB levels in beef cows are reduced by lactation and these contaminants are transferred into the calf. Figure 2 show the WHO-PCDD/F-PCB-TEQ levels in meat depending on the age class of the slaughtered animal. Samples from suckled calves (age 6 – 12 months) had about 2 - 3 times the levels of beef cattle after weaning and feeding on grass for several months (Figure 2). Slightly elevated levels of dl-PCBs in the soil as a result of former military use of the area were discussed as possible source (n = 34 samples, mean value 2 ng WHO-PCB-TEQ/kg dm, range 0.7 – 5.6 ng WHO-PCB-TEQ/kg dm). These levels exceeded the dl-PCB concentration in soil of grassland without suspected

contamination (approx. 0.3 ng TEQ/kg dm)[16]. Levels in grass and hay (n = 8) ranged from 0.11 to 0.20 ng WHO-PCB-TEQ/kg (88 % dm) with a mean value of 0.14 ng/kg. In another case of a pasture land, dredged sediments were used to meliorate soil resulting in dl-PCB-levels between 3.9 and 6.4 ng WHO-PCB-TEQ/kg dm. Feed used on the farm had a median WHO-PCB-TEQ level of 0.12 ng/kg (88 % dm). From 11 meat samples, 9 exceeded the maximum level for WHO-PCDD/F-PCB-TEQ with dl-PCB as major TEQ-contributor.

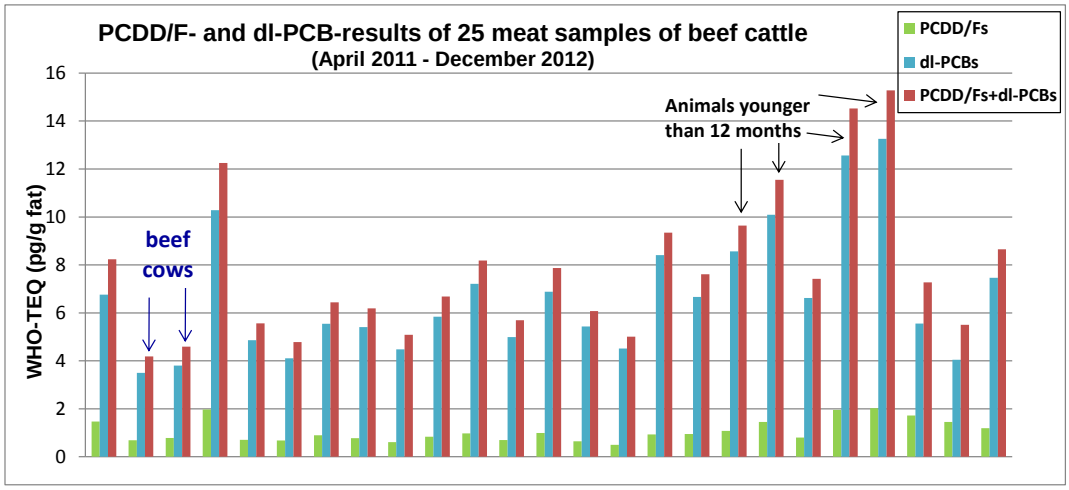

Figure 1: PCDD/F and dl-PCB TEQ-levels in meat of beef cattle on a pasture with elevated PCB-soil levels (mean 2 ng WHO-PCB-TEQ/kg dm, range 0.7 – 5.6).

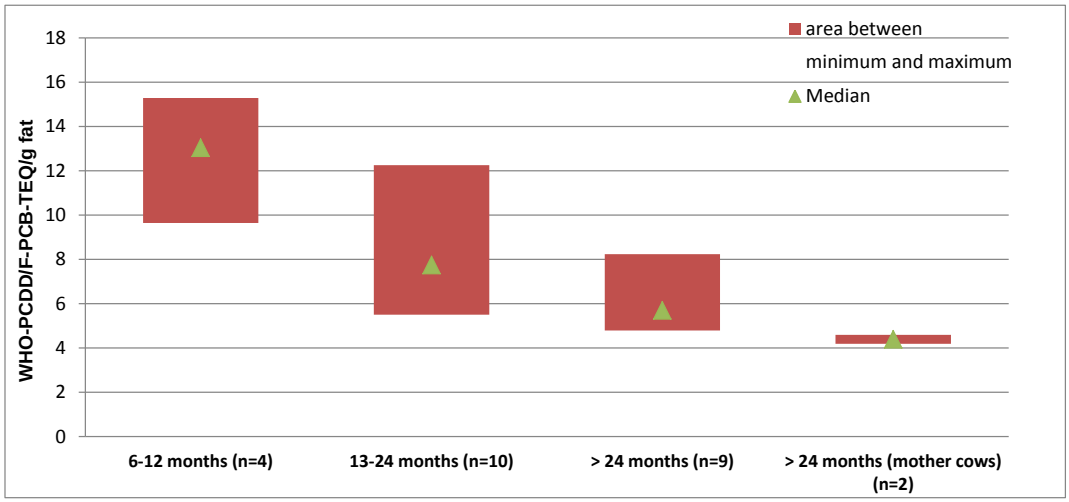

Figure 2: TEQ-levels in meat of cattle within one suckling herd in dependence of age/class at slaughter.

(3) Problematic dl-PCB levels in soil and feed

On flood plains with 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. Also in other herds with dl-PCB levels resulting in exceedance of TEQ-limits in meat, where no point sources could be identified, relatively low soil levels (below 5 ng WHO-PCB-TEQ/kg dm) in combination with feed levels around 0.15 ng WHO-PCB-TEQ/kg dm seem to be responsible for the exceedance of the stringent EU limits for WHO-PCD/F-PCB-TEQ in meat.

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; Figure 1) in combination with grass/feed levels around 0.15 ng TEQ/kg dm considerably below the EU-regulatory limits. 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.

The decrease of TEQ values after weaning (Fig. 2) indicates options to make sure that meat is below maximum levels if cattle are raised on pasture land with dl-PCB levels of approx. 2 to 5 ng TEQ/kg dm. More research is needed to conclude on critical dl-PCB (and PCDD/F) levels in soil and feed with regard to resulting levels in milk and meat. Also the role of open application of PCBs (paints, sealants, PVC-coatings) and of areas with historic PCB impact need further systematic assessment of exposure of cattle and risk to humans.
Based on the findings on exposure sources of beef to PCBs (and PCDD/Fs) a specific leaflet for cattle breeders was developed for the safe production of meat from beef cattle[17] in support to the national guidance "on environmental protection as a basis for safe food production" of the German Environmental Ministry.[9]

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References:

1. Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs, Official Journal of the European Union L 364/5.
2. Commission Regulation (EU) No 1259/2011 of 2 December 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for dioxins, dioxin-like PCBs and non dioxin-like PCBs in foodstuffs, Official Journal of the European Union, L 320/18.
3. European Food Safety Authority (EFSA) (2012) Update of the monitoring of levels of dioxins and PCBs in food and feed, EFSA Journal 2012;10(7):2832.
4. Tlustos C, Anderson W and Evans R "Responding to food contamination incidents: principles and examples from cases involving dioxins", chapter 5 in "Persistent organic pollutants and toxic metals in foods", Rose, M., and Fernandes, A. (editors) (2013), Woodhead Publishing Ltd.
5. Malisch R and Kotz A (2014) Sci Total Environ (2014), http://dx.doi.org/10.1016/j.scitotenv.2014.03.022
6. European Food Safety Authority (2011) EFSA Journal 9: 2297, 1-71.

7a Kamphues J, Schulz AJ, Gude K, Bruns-Weller E, Severin K, Appuhn H,Taube V (2011) Organohalogen Compounds 73, 2040-2041.

7bUngemach LC (2013) Die Dioxinbelastung von Schlachtrindern aus Färsenvornutzung auf exponiertem Grünland nach einer Ausmast mit unbelastetem Futter. PhD, Tierärztliche Hochschule Hannover.

7c Schulz A-J, Wiesmüller T, Appuhn H, Stehr D, Severin K, Landmann D, Kamphues J (2005) Journal of Animal Physiology and Animal Nutrition 89 (2005) 72-78.

8. Lake IR, Foxall CD, Fernandes A, Lewis M, White O, Mortimer D, Dowding A, Rose M (2014) Sci Total Environ. doi: 10.1016/j.scitotenv.2014.01.080.
9. Bundesministerium für Umwelt, Naturschutz, und Reaktorsicherheit (2013) Umweltschutz - Standbein der Lebensmittelsicherheit – Dioxin- und PCB-Einträge vermeiden. 5. aktualisierte Auflage, Januar 2013.

10a. Commission Regulation (EC) 1883/2006 of 19 December 2006; Official Journal of the European Union L364/32, 20.12.2006.

10b. Commission Regulation (EU) No 252/2012 of 21 March 2012 laying down methods of sampling and analysis for the official control of levels of dioxins, dioxin- like PCBs and non-dioxin-like PCBs in certain foodstuffs and repealing Regulation (EC) No 1883/2006. Official Journal of the European Union, L 84/1.

11. Commission Regulation (EU) No 278/2012 of 28 March 2012 amending Regulation (EC) No 152/2009 as regards the determination of the levels of dioxins and polychlorinated biphenyls. Official Journal of the European Union, L 91/87.
12. Götz R, Bauer O-H, Friesel P, Herrmann T, Jantzen E, Kutzke M, et al. (2007) Chemosphere 67: 592–603.
13. Landwirtschaftskammer Niedersachsen (2011) Grünlandbewirtschaftung von Überschwemmungsflächen im Bereich der Elbtalniederung der Bundesländer Mecklenburg-Vorpommern, Niedersachsen, Sachsen-Anhalt, Schleswig-Holstein. Oldenburg, Stand: Februar 2010.
14. Hembrock Heger A (2011) Dioxine und PCB in Böden, Pflanzen, Futter- und Lebensmitteln in Überschwemmungsgebieten in NRW. UBA Fachgespräch Belastung der terrestrischen Umwelt mit Dioxinen und PCB; 13./14. Oktober 2011, Berlin.
15. Takasuga T, Nakano T, Shibata Y (2012) *Organohalogen Compounds* 74, 1437-1440.
16. Bussian B, Schmidt S, Utermann J (2013) Typische Gehalte von Dioxinen und dl-PCB in Böden, Fachgespräch „Dioxine und PCB: Bessere Daten – Schnellere Aufklärung" Bonn 28.10.2013.
17. Merkblatt PCB und Dioxine in Rindfleisch Haltungsempfehlungen für Rinderhalter Stand 22.01.2014.