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Analyse und Trendabschätzung der Belastung der Umwelt und von Lebensmitteln mit ausgewählten POPs und Erweiterung des Datenbestandes der POP-Dioxin-Datenbank des Bundes und der Länder mit dem Ziel pfadbezogener Ursachenaufklärung

Anhang 6: Tagungsbeiträge auf dem 34th International Symposium on Halogenated Persistent Organic Pollutants

Für Mensch & Umwelt

Umwelt Bundesamt

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Analyse und Trendabschätzung der Belastung der Umwelt und von Lebensmitteln mit ausgewählten POPs und Erweiterung des Datenbestandes der POP-Dioxin-Datenbank des Bundes und der Länder mit dem Ziel pfadbezogener Ursachenaufklärung

Anhang 6: Tagungsbeiträge auf dem „34th International Symposium on Halogenated Persistent Organic Pollutants“, 31.08. bis 05.09.2014, Madrid, Spanien

von

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1 Beitrag 1: Sichere Lebensmittelproduktion aus extensiver Rinderhaltung

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.

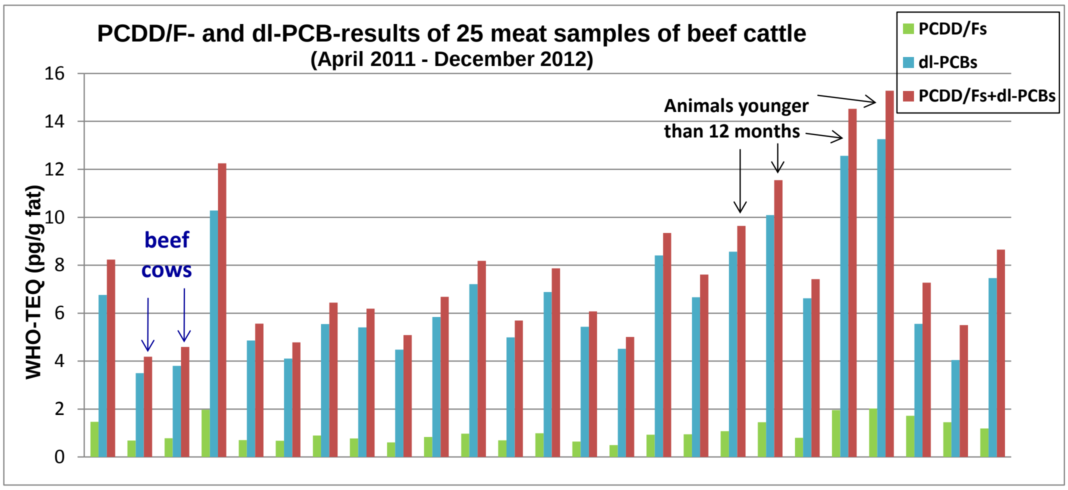

Figure 1: PCDD/F and dl-PCB TEQ-levels in meat of beef cattle on a pasture with elevated PCB-soil levels (mean 2 ng WHO-PCB-TEQ/kg dm, range 0.7 – 5.6).

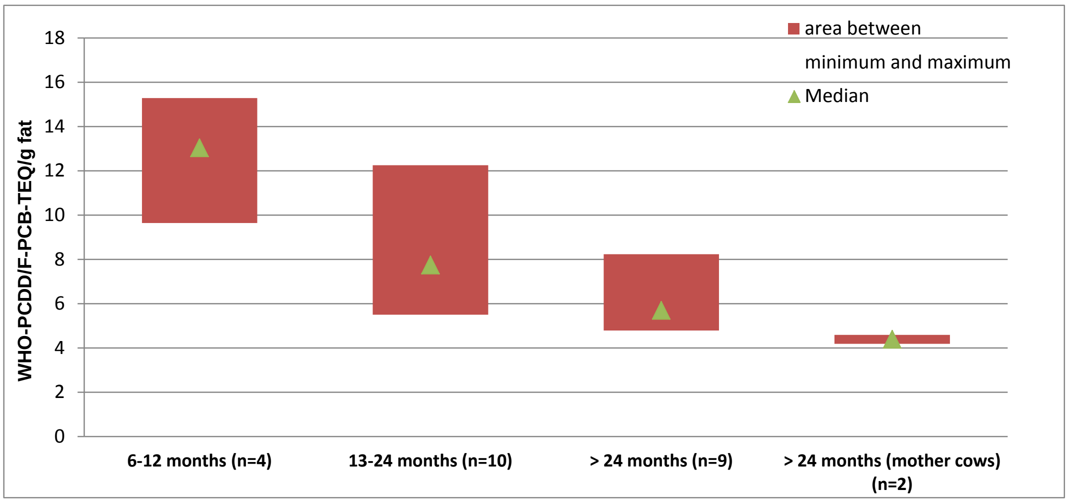

Figure 2: TEQ-levels in meat of cattle within one suckling herd in dependence of age/class at slaughter.

(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]

Acknowledgement

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2 Beitrag 2: PCB-Inventar und Materialfluss

A PCB INVENTORY AND MATERIAL FLOW CONSIDERING PRODUCTION, HISTORIC USES, WASTE MANAGEMENT, SINKS AND SOURCES

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Introduction

The production of polychlorinated biphenyls (PCBs) has stopped about 30 years ago. The global production and use has been compiled and is estimated at around 1.3 million tonnes[1,2]. PCBs have been used in a wide range of closed and open applications which are still present in many countries. The global inventory of PCBs of transformers and capacitors revealed approximately 3 million tonnes of waste PCBs oils and PCBs containing equipment[3]. In the frame of the Stockholm Convention, persistent organic pollutants (POPs) including PCBs are managed and phased out for the protection of human health and the environment. For this aim countries are developing inventories for the individual POPs for assessing their current relevance and managing the remaining stockpiles. PCBs were already included in the 12 initial POPs and were therefore targeted now globally for about a decade. PCB inventories normally only address the PCBs in closed applications in current use and stocks[3]. The PCBs in open applications are not addressed in these inventories and are not directly mentioned in the Stockholm Convention text or the EU POP inventory. Also an inventory of total historic PCBs use is not made and has not been published for any country to our knowledge.

However, it has been revealed that for industrial countries the open applications of PCBs have high relevance for contamination of the environment in particular the application of paints[4] and sealants[5a,b]. Additionally it has been revealed that the application of paint and sealants are a contemporary source for PCBs exposure of cattle[6,7] and chicken/eggs[8]. Furthermore open applications of PCBs in buildings are a contemporary exposure source for humans[9]: for people living in these buildings the PCB exposure via this pathway can considerably exceed their PCB exposure via food[9]. This demonstrates that a PCB inventory and management should also include the PCBs from open applications at least in industrial countries having used these applications in the past.

Furthermore it has been discovered over the last decade that disposed PCBs and PCB contaminated area can result in contamination of the food chain[10a,10b]. Therefore in addition to the PCBs in current use and stocks also these PCB legacies should be considered in a national PCB inventory. This has recently been taken up by the UNEP Dioxin toolkit[11] where PCB reservoirs and contaminated sites are highlighted as potentially PCDF contaminated sites which should be inventoried[11].

During the last 30 years a wide range of information has been compiled in Germany on total PCB use and the distribution in different PCB applications or remaining PCB sources at a certain time. In the current study these informations have been compiled.

Materials and methods

Information on PCB production and use were compiled from a range of studies. Several studies have been initiated by Research & Development project under the German Environmental Ministry12-14. Additional information were extracted from other research studies or national or international reports.

Information on the situation of PCBs in open applications from other countries have been screened and were considered in the compilation and for formulating assessment gaps.

Furthermore the German Environmental Agency is currently conducting a Research & Development project for assessing the impact of Dioxins and PCBs in the environment. Also findings from this project were considered where appropriate.

For air data two additional main information sources were used

a) The German PRTR register was screened for available data and assessed for data gaps.
b) The German POPs inventory having compiled information from PCB emission from thermal sources.

Results and discussion

Total PCB production and use volumes in former East and West Germany

In Germany, from 1930 to 1983, the Bayer AG produced about 159,062 tonnes of PCBs as Clophen in different chlorination grades[2]. After Hillejan & v. Schaaffhausen (1990)[13] a total of approximately 83,000 tonnes PCB were used by West German companies in different applications and products, of which 72,500 tonnes remained in West Germany (Figure 1).

The production and used quantities in East Germany (former German Democratic Republic (GDR)) were far lower. The production of PCBs started in East-Germany in 1955 with highly chlorinated biphenyls[14]. The East-German production volume was 60 t in 1955, until 1964 the total production included about 1,000 tonnes[15]. The PCB production was stopped by a devastating fire and the factory was closed down. The PCB consumption in East Germany was then covered by imports especially from Czechoslovakia. Total PCB imports from 1962 to 1985 for East Germany were 18,600 tonnes[14]. The total amount of PCB in technical applications remained in East Germany is estimated to about 12,300 tonnes for 1955 until 1985 (Figure 1).

Therefore in Germany, approximately 85,000 tonnes of PCB were used as pure products as well as in mixtures with other substances in open and closed systems[12].

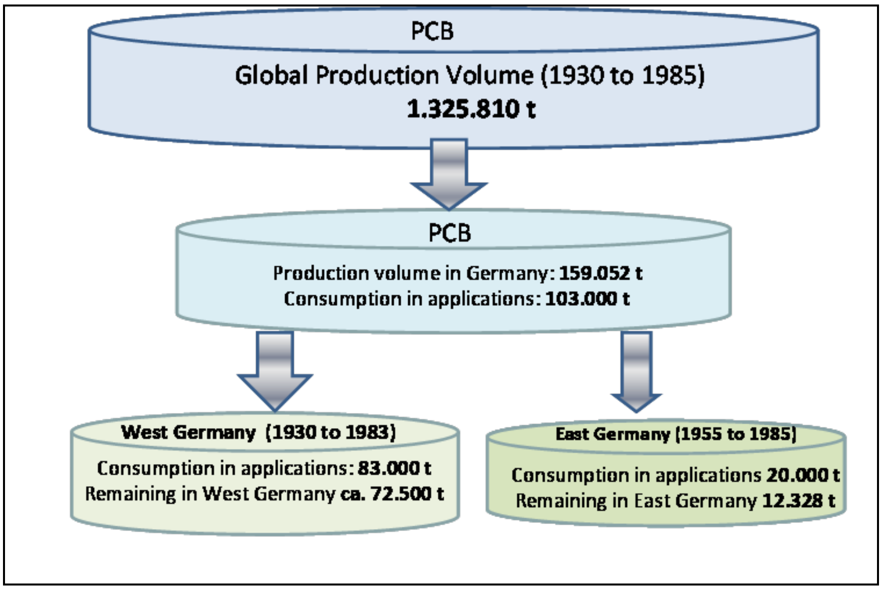

Figure 1: Production and consumption quantities of PCB in former West Germany and East Germany.

Source: Balzer & Rauhut, 1987; de Voogt et al. 1989; Hillejan & Schaaffhausen,1990; Schaaffhausen & Gramenz, 1993; Knetsch, 2011[19]

Use of PCBs within Germany

In West Germany approximately 83,000 t of the approx. 150,000 t produced were brought in applications. The rest were exported as PCBs.

In closed systems, they served as coolants in transformers (fluids), as a dielectric in capacitors and as hydraulic oils. The used volumes in closed applications produced in Germany were[12]:

- Askarel-Transformers (ca. 23,000 t)
- Condensers (ca. 24,000 t)
- Hydraulic oils (ca. 12,500 t).

Approx. 24,000 t of PCB were used in open applications in Germany. In open systems, PCBs were used among other things as a plasticizer in sealants and paints, as a lubricant and as flame retardants in ceiling tiles or cable insulations or specific paper. From this the largest share was used for sealants in buildings and constructions (approx. 20,000 t). The distribution of the remaining 4,000 t for other open applications is not documented. Most probably a large share of this was used in paints. The relevance of PCBs in paints was revealed e.g. in Switzerland where a considerably share of electric posts and high pressure pipes at that time contain PCBs and at least 20% of public swimming pools are contaminated with PCBs.

Waste management of the PCBs

From the PCBs in closed application a considerable share has not been disposed/destroyed appropriately (in particular before legislation for PCB management had been introduced):

- From the approx. 12,500 t PCB of hydraulic oil used in mines it is assumed that most has been released during time of use[12,16].
- For condensers in East Germany approximately 50% (5,000 t) were not appropriately disposed. For West Germany the largest part of the approximately 3,200 t PCB in large condensers is considered to have been appropriately disposed. However from the approximately 10,000 t of PCB in small condensers a considerable share ended in landfills, shredder facilities and secondary metal industries.
- PCBs in transformers were appropriately managed after legislation came in place.

Overall Detzel et al. (1998)[12] estimated that approx. 30 to 50 % of PCBs in closed applications was not appropriately managed (including landfills). Therefore a large share of PCBs from closed applications was released into the environment.

For PCBs in open applications a large share (50 to 80%) is estimated to be still in use in sealants of buildings and other constructions and in paints. Other open uses with shorter life time (paper, cables) have already been disposed to landfills or were treated in incinerators.

Inventory of releases of PCBs

The atmosphere including airborne depositions plays an important role in respect to the influence of PCBs on terrestrial and aquatic ecosystems. It is an essential vector for the transfer of PCBs into the feed and food chain and ultimately into humans. PCBs can be released from primary sources (e.g. building seals, painted constructions or electrical capacitors) from point sources from disposal (e. g. landfills, contaminated sites, construction debris) as well as of secondary sources (e.g. metal industry, incinerators).

The emission of unintentionally produced PCBs from combustion plants and other thermal and diffuse sources is estimated at 200 kg/year in Germany since 2004[17]. Currently only a small share of this is covered by the pollution release transfer register (approximately 24 kg for air for three years) by the metal/steel and cement industry. Releases to further compartments like water and soil are documented by the German Pollutant Release and Transfer Register (ePRTR). It is an online information system for the quantification of the release of such pollutants in the environment. The on-line register of Germany (http://www.thru.de/thrude/) offers the possibility of operation-related emission data to more 90 pollutants. PCB is a category of substances in this list of pollutants. A notification for the register is only required if the thresholds for the release of 0.1 kg / year to air, water and soil are exceeded. Annual report information are available from 2007 to 2012 with detailed information of the branch of industry and their incorporation into the air, water bodies and (via the sewers) in external sewage treatment plants as well as disposed of hazardous and non-hazardous waste into the environment (Table 1).

Table 1: Notification for the German ePRTR of emission for PCB in the air, water and waste water. Source: http://www.thru.de/thrude/

PCB in kg /year	Air	Water	Transfer with waste water	Reporting PRTR
2010	9,7		1,78	Production of iron and steel, cement (air), sewage plant (Transfer with water)
2011	5,6	0,9		Production of iron and steel, urban drainage (water)
2012	9,02	0,8	2,73	Production of iron and steel, cement, incineration (air), sewage plant (Transfer with water), Urban drainage (water)

In environmental matrices inter alia air, soil and vegetation including feed (grass, silage, hay) the PCB congeners stem almost exclusively from commercial legacy PCBs with the exemption of PCB11[18]. This demonstrates the dominance of the industrially produced PCBs as emission and contamination source.

Further considerations

The current release of industrial PCBs and the first rough estimate of these releases highlight the need to further assess these sources and to appropriately manage PCBs in open applications. A more detailed inventory of open applications sealants and paints seems also relevant for feed and food safety considering that several of recent PCB food contaminations (in particular chicken/egg, sheep and beef) in e.g. Germany, Netherlands and Switzerland stem from open PCB applications.

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