HIGH PCB CONTAMINATION DETECTED IN CATTLE FROM EXTENSIVE FARMING IN SWTZERLAND

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Introduction

The Federal Food Safety and Veterinary Office FSVO (formerly Federal Office of Public Health) regularly examines levels of polychlorinated dibenzo-*p*-dioxins (PCDD), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs) in different food categories from the Swiss market. Three monitoring programs carried out in the years 2003, 2006 and 2008 revealed that cattle meat from extensive farming occasionally exceeded the maximum levels (ML) of 4 pg WHO-TEQ₀₅/g lipid weight (lw) set by the European Union and the Swiss authorities^{1,2}. In extensive keeping animals are fed by grazing outdoor and only short stable residence. Calves are raised by suckler cows. During the winter months the cattle are fed with hay and silage, and only marginal concentrated feeding is permitted. Triggered by the above mentioned observations, the dioxin and PCB monitoring program in 2012 was focused on cattle meat from extensive farming.

Materials and methods

60 meat samples originating from 53 different farms were gathered at an industrial slaughterhouse in Switzerland. The meat samples could easily be traced back to the respective farms by the official labelling system used in Switzerland. The meat was vacuum-packed, frozen and sent to Eurofins Hamburg for analyses of PCBs, PCDDs and PCDFs. Retained samples were stored at the Swiss Federal Office of Public Health. Additional analyses were carried out by the Swiss Federal Laboratories for Materials Testing and Research (Empa) by gas chromatography/high resolution mass spectrometry (GC/HRMS).

Results and discussion

Out of the 60 meat samples analyzed three samples (5%) exceeded the ML. Total WHO-TEQ₀₅ levels in veal from farms A and B slightly exceeded the ML with of 7.3 and 5.3 pg/g lw. In contrast, veal from farm C revealed a level of 19 pg/g lw exceeding the ML by almost a factor of five. Based on these results the three concerned producers of the veal were traced back and additional samples from the farms were taken and analyzed to make sure that the contaminated samples were representative for the whole herd and not outliers. The elevated levels could be confirmed in two out of the three farms as shown in Figure 1. In all samples the total WHO-TEQ₀₅ was dominated by dl-PCB (83 to 97%) vs. PCDD/F. PCB congener 126 with a relatively high toxicity equivalent factor (TEF) of 0.1 contributes between 77 and 89% to the total WHO-TEQ₀₅. The average content (based on 10 animals) expressed as WHO₀₅-TEQ in case of farm A was 5.1 pg/g lw and 26 ng/g lw for the sum of the six indicator PCBs (iPCBs). Seven out of the eleven analyzed meat samples exceeded the ML of 4 pg WHO₀₅-TEQ/g lw while levels of all samples were below the ML of 40 ng/g lw for the sum of the six iPCB. As levels of four samples were only slightly below the ML of 4 pg WHO_{05} -TEQ/g lw further investigations were initiated to investigate possible reasons for the high PCB levels in cattle from farm A. In contrast, eight out of eleven veal samples from farm B revealed concentrations below the ML, and only three samples exceeded the ML of 4 pg WHO₀₅-TEQ/g lw. Two out of the three samples exceeding the ML were very close to 4 pg WHO₀₅-TEQ/g lw. The average PCB content of eleven samples was 3.5 pg WHO₀₅-TEQ/g lw and 15 ng/g lw for the sum of the iPCB. As the meat produced at farm B fulfilled the legal requirements no further measures were taken. The situation of farm C is completely different (see Figure 1): The very high levels of almost 20 pg WHO₀₅-TEQ/g lw were confirmed by additional analyses. The average content of the five samples shown in Figure 1 is 17.5 pg WHO_{05} -TEQ/g lw.



Figure 1: WHO₀₅-TEQ levels in meat samples. The red dashed line indicates the ML of 4 pg WHO₀₅-TEQ/g lw. (VE: veal extensive; BE: beef extensive; UB: upper bound; LB: lower bound).

If calculation of the TEQ is based on WHO₉₈-TEFs the resulting average value is considerably higher (27 pg WHO₉₈-TEQ/g lw). This pronounced difference is due to relatively high amounts of highly chlorinated dl-PCB congeners (see Figure 3) of which PCB 156 and 157 exhibit distinctly higher TEFs in the WHO₉₈-TEF scheme than in the WHO₀₅-TEF scheme (TEFs of both congeners 0.0005 instead of 0.00003). Furthermore, in these samples the average sum of the six iPCB of 480 ng/g lw exceeds the permitted ML of 40 ng/g lw by more than a factor of 12. Similarly, the iPCB pattern is dominated by the highly chlorinated congeners 138, 153 and 180 with lower chlorinated congeners PCB 28, 52 and 101 at vanishingly low concentrations. These findings suggested a highly chlorinated PCB source present in farm C.

Analyses of ca. 20 feed and material samples taken in the stable eventually revealed two wall coatings with PCB contents of 16% and 3%. These paints were applied after construction of the stable more than 40 years ago. Confirmation of this primary source was also based on the perfect matching of the congener patterns of the meat samples and the paint. Further investigations to detect possible secondary PCB sources and uptake pathways into the cattle are running. Due to the very high levels veal and beef from farm C were blocked.

In Figure 2 the ratios sum iPCB/sum dl-PCB are given for veal, beef, milk and material samples for the farms A, B and C. The difference between the ratios of samples originating from farm C to farms A and B is evident. Veal and beef from farm C exhibit an average ratio sum iPCB/sum dl-PCB of 8.4 and milk shows a similar average ratio of 9.2. Meanwhile, the ratio of the feed samples is slightly higher with an average of 11. The slightly higher ratio in case of the feed samples may be due to the presence of higher contents of lower chlorinated PCB congeners e.g. PCB 28, 52 and 101, thus increasing the sum of the iPCB and the ratio. The latter congeners are

almost not present in veal and beef, probably due to faster elimination and metabolism. Furthermore, with an average WHO_{05} -TEQ of 0.30 ng/kg the levels in feed samples are too low to explain the high PCB contents in the meat. Within the material samples a black paint from a metal post as well as the green and grey wall paints with ratios of 11, 13 and 12 respectively, come close to the ratio found in the meat samples. The two oil samples (gear oil and hydraulic oil from machines) exhibit completely different ratios of around 4, also a black vessel that was partially used to administrate salt doesn't fit with the ratio observed in veal and beef. Additionally, with the exception of the green and grey wall paints, with the extremely high sum iPCB contents of 6 and 1% respectively, the other material samples had clearly too low PCB contents to be responsible for the contamination of the animals.

As mentioned above the ratios sum iPCB to sum dl-PCB found in veal, beef and milk from farm A and B are with average values of 3.5 and 3.1 for veal and beef, respectively 4.8 and 3.6 for milk clearly diverging from farm C.



Figure 2: Ratios sum iPCB/sum dl-PCB in veal, beef, milk, feed, and material samples (VE: veal extensive; BE: beef extensive).

Figure 3 shows the dl-PCB congener patterns of the samples from the three farms. Veal, beef and milk samples from farm C fit the best with the dl-PCB pattern of the green and grey wall paints from the stable of this farm. The dl-PCB patterns at farm A and B are clearly different from farm C and reflect a general dl-PCB distribution pattern for veal, beef and milk. Intriguingly, grass silage, silage juice, feed residues from the moving belt, and the black vessel show similar dl-PCB patterns as meat samples. This may be due to the fact that these samples were exposed in the contaminated area and were contaminated via air, dust and paint particles. Analyses of straw samples, used as bedding in the stable, revealed higher PCB contents in samples which were collected close to the walls with the contaminated paint. The maximum level in straw samples was 3.6 mg iPCB/kg dry weight (dw) and 18 ng WHO₀₅-TEQ/kg dw. This sample was collected directly at the bottom of the wall and was partially contaminated with cow dung. This level exceeds the ML for feed set by the EU of 1.25 ng/kg by more than a factor of 14 and the ML for iPCB in feed by more than a factor of 360. The most contaminated straw sample from the middle of the stable contained 0.058 mg iPCB/kg dw and 0.62 ng WHO₀₅-TEQ/kg dw and was thus distinctly less polluted but the iPCB level still exceeds the ML by a factor of six. Certainly, straw used as bedding is not the primary feed for the animals, but during the sampling campaigns it could be observed that cows and the calves ingested straw. Therefore this material can be considered as a possible uptake pathway of PCB. Furthermore, the 40 year old wall paint showed strong signs of erosion and chipping off. A small particle of typically 25 mg (a paint sliver) with an iPCB content of 6% can easily be ingested and would correspond to a maximal uptake of 1.5 mg iPCB.

Active and passive air samples collected in the stable revealed very low average PCB air concentrations of 8.7 and 4.8 ng/m^3 for the sum of the six iPCB, respectively. These very low indoor air concentrations are most likely the result of the effective draft ventilation in the stable. Indoor air can therefore be excluded as a main uptake pathway of PCB.

Possibly, the stable of farm C with the wall paint containing high amounts of PCB is not unique and it is well possible that other farms of the 40'000 cattle farms in Switzerland are exposed to similar PCB sources. Instead of an extensive analytical study a nationwide questionnaire could help to identify affected farms.



Figure 3: dl-PCB congener patterns of meat, milk, feed, and material samples (VE: veal extensive, BE: beef extensive).

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