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CARRY-OVER OF DIOXINS AND PCBS FROM FEED TO EGGS AT LOW CONTAMINATION LEVELS

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Introduction

In 2001, monitoring of animal derived food products resulted in the discovery that eggs from free-range chickens in the Netherlands regularly exceeded the existing limit of 5 ng TEQ/kg fat[1]. Follow-up studies strongly suggested that the source of the contamination was not the feed but the outdoor environment. When the new EU food and feed limits became official, an exception was made for eggs from free-ranging chickens until 1-1-2004. Studies were started to investigate the carry-over rates of dioxins and PCBs from feed and soil to eggs at relatively low levels, and to investigate possible ways for lowering the intake and carry-over rates. This paper describes the results of the first study, being the relationship between dioxin and PCB levels in feed and eggs.

Methods and materials

Feed preparation

Soy oil was spiked with standard mixes of dioxins, non-ortho PCBs and mono-ortho PCBs purchased from CIL. The dioxin mix, added at 29 µl/kg, contained all congeners at a level of 6.25 µg/ml, with the exception of TCDD and TCDF (2.5 µg/ml) and OCDD and OCDF (12.5 µg/ml). The no-PCB mixture, added at 272 µl/kg, contained equal concentrations of PCBs 77, 81, 126 and 169 (10 µg/ml). The non-ortho mix, added at 524 µl/kg, contained equal amounts of the 8 mono-ortho PCBs (10 µg/ml). In addition, the oil was spiked with stock solutions of 100 µg/ml of PCBs 105 and 28 (5 ml/kg) , PCBs 118, 52 and 180 (10 ml/kg), PCBs 101, 138 and 153 (20 ml/kg). This should result in a stock oil containing 1000 ng TEQ/kg, with relative contributions of dioxins, non-ortho and mono-ortho PCBs of 50, 30 and 20%. This oil was subsequently diluted with clean oil to solutions containing 20, 40, 75, 150 and 300 ng TEQ/kg. Based on the analysis of the highest oil, the different oils were mixed at 0.9% into a standard chicken feed, resulting in 6 different feeds with intended dioxin levels of 0, 0.1, 0.2, 0.4, 0.75 and 1.5 ng TEQ/kg and equal amounts of dioxin-like PCBs. Intended levels of indicator PCBs were 0, 1.8, 3.6, 6.8, 13.5 and 27 µg/kg.

Chicken experiment

Laying hens were fed on clean feed for 1 week followed by a period of eight weeks on the different feeds and another period of 8 weeks on clean feed. Eggs were collected and pooled per dosage group and day. At different time intervals chickens were slaughtered and the abdominal fat and livers collected.

GC/MS analysis

GC/HRMS analysis on dioxins and dioxin-like PCBs was performed as described by Tuinstra *et al.*[2], with the exception that 3 grams fat or 50 grams of feed was used. Indicator PCBs were analysed by GC/MS using a large volume injector.

Results and discussion

Analysis of feed

Samples were taken from different bags of each feed and analysed for indicator PCBs. This showed that the feed was of good homogeneity (%CV for 3-5 analyses 1-7%). Indicator PCB levels were respectively 0.1, 2.3, 4.3, 6.0, 14.2, 31.7 µg/kg. The feeds were subsequently analysed for dioxins and dioxin-like PCBs (Table 1). Dioxin levels in the lower contaminated feeds were below the limits of quantification, resulting in major differences between upper and lower bound values. Since the different soy oil stocks and therefore feeds were prepared from the same stock solution of soy oil, the levels of the mono- and non-ortho PCBs were used to estimate the dioxin levels in lower contaminated feeds. Based on extrapolation, the blanc feed did not contain significant amounts of dioxins and dioxin-like PCBs. The actual dioxin levels in the feeds were in general 19 to 53% higher than intended. The relative contribution of the different groups was as planned, being 51, 31 and 19% for respectively dioxins, non-ortho and mono-ortho PCBs.

Figure 1 shows the levels of dioxins, non-ortho, mono-ortho and indicator PCBs in egg fat. Even with the lowest contaminated feed, levels increased above background, reaching a maximum around day 58. Maximum dioxin levels obtained with the different feeds were 0.1, 1.4, 2.7, 3.9, 9.4 and 21.5 pg TEQ/g. Steady state conditions were not obtained even after this relatively long exposure period of 8 weeks. Fat levels exceeded the EU limit of 3 pg TEQ/g when chickens were fed the feed with 2 and 0.9 ng TEQ/kg, both higher than the EU limit for feed, but also in the case

Table 1. Levels of dioxins, non-ortho and mono-ortho PCBs expressed in ng TEQ/kg and based on both lowerbound (lb) and upperbound (ub) principles. In addition, detectable congeners were used to estimate levels of non-detectable congeners (profile).

Feed	PCDD/Fs			no-PCBs			mo-PCBs			Total
	lb	ub	profile	lb	ub	profile	lb	ub	Profile	Profile
0	0.00	0.29	0.00	0.00	0.01	0.00	0.00	0.02	0.00	0.00
0.1	0.00	0.29	0.16	0.10	0.10	0.10	0.06	0.07	0.06	0.31
0.2	0.20	0.31	0.30	0.18	0.18	0.18	0.11	0.12	0.11	0.60
0.38	0.42	0.43	0.42	0.25	0.25	0.25	0.16	0.17	0.16	0.83
0.75	0.89	0.90	0.89	0.54	0.54	0.54	0.32	0.32	0.32	1.75
1.5	2.02	2.02	2.02	1.17	1.17	1.17	0.68	0.68	0.68	3.87

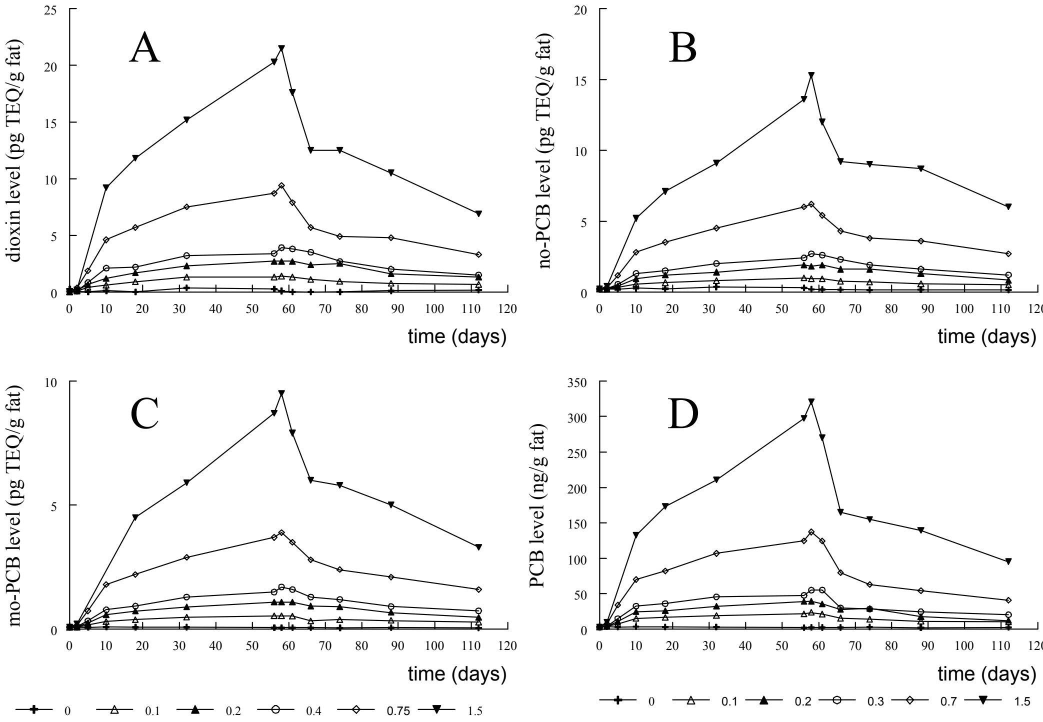

Figure 1. Levels of dioxins (A), non-ortho (B), mono-ortho (C) and indicator (D) PCBs in eggs of chickens fed contaminated feed for 56 days, followed by 56 days on clean feed. Feed levels in the legend are the intended levels.

of the feed with 0.4 ng TEQ/kg. The EU action limit of 2 pg TEQ/g fat was already exceeded with the feed spiked at 0.2 ng TEQ/kg. Figure 2A shows the correlation between dioxin levels in feed and egg fat at day 58. It can be calculated that limits and action levels of 3 and 2 pg TEQ/g in egg fat correlate with feed levels of respectively 0.3 and 0.2 ng TEQ/kg. Based on dioxin levels determined in many samples of Dutch feed, normal levels in feed are well below 0.2 ng TEQ/kg. This is also indicated by the very low levels (<1 pg TEQ/g) in eggs from chickens kept inside.

Maximum total TEQ levels were 0.3, 2.9, 5.6, 8.3, 19.5 and 46.3 for the six different feeds. Indicator PCB levels in eggs followed a similar pattern as dioxins and reached maximum levels of respectively 2, 24, 39, 55, 137 and 321 ng/g fat at day 58 (Figure 1D). Again good correlations were obtained between feed and eggs for both maximum TEQ (Figure 2B) and PCB levels (correlation coefficient 0.999, graph not shown). The ratio [indicator PCB/dioxin TEQ] in the eggs at day 58 was very similar to the ratio in the feed, being about 15,000.

Following replacement of the contaminated feed by clean feed, there appeared to be an initial rapid decrease of the levels, followed by a very slow decrease. This might be due to the fact that the eggs up to one week after the switch were produced during the exposure period, regarding the

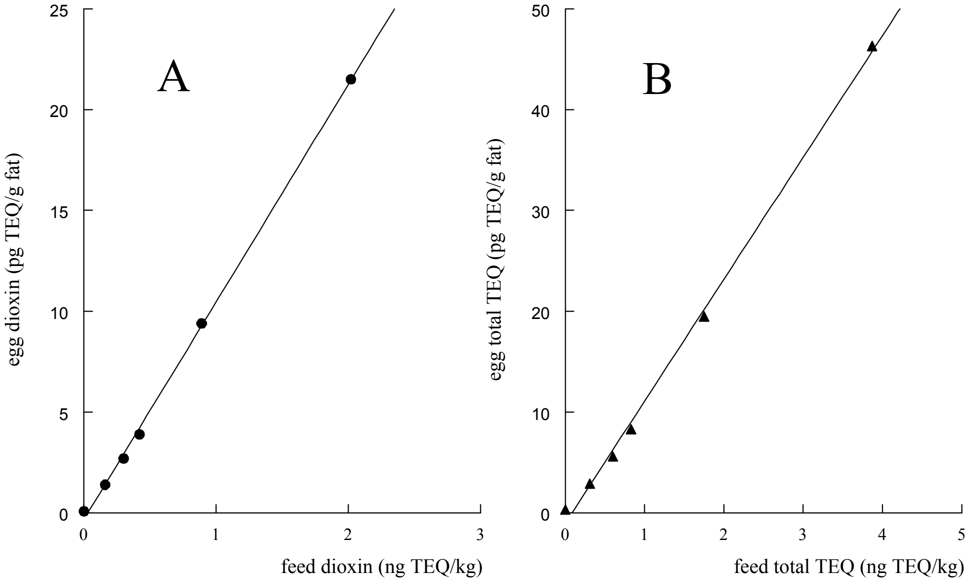

Figure 2. Relation between feed levels of dioxins (A) and total TEQ (B), and maximum dioxin and total TEQ levels in egg fat. Correlation coefficients were respectively 0.999 and 0.998.

10-day production period of an egg. The dioxins and PCBs in eggs produced after the first week must originate from the body fat only, and the transfer of contaminants from this compartment to the egg appears to be much slower. Kinetics in laying hens are presently being modeled.

Conclusions

The new EU limit of 0.75 ng TEQ/kg, established in 2002, is insufficient to guarantee that levels in eggs will not exceed the limit of 3 ng TEQ/kg fat.

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