DIOXINS AND PCBS IN HOME PRODUCED EGGS IN THE NETHERLANDS

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Introduction

Following the discovery of elevated dioxin levels in eggs from a small farm in 2001[1] and follow-up by various studies on potential measures[2], producers of eggs from free-range hens introduced a strict plan including regular self-control. As a result, levels in free range eggs have strongly declined and are nowadays well below the MLs set in the EU. Only occasionally, farms with contaminated commercial eggs are discovered, due to new sources (e.g. use of building debris in the courtyard or coated asbestos roof plates), and/or the switch of farms from inside to outside husbandry[3]. A different situation may apply to home-produced eggs, only used for own consumption and as such not checked for the presence of PCDD/Fs or PCBs. Based on experience with commercial farms, measures were advised through websites and dedicated magazines, but it is unclear to what extent these were followed-up. In 2013, a small study on home-produced eggs was performed in the vicinity of Harlingen to evaluate the potential release of PCDD/Fs and PCBs from a local waste incinerator. This study showed elevated levels with a highest level for dioxins and dl-PCBs around 10 pg TEQ/g fat.

The question was whether this finding reflected merely the situation for home-produced eggs rather than being caused by the new waste incinerator. This was supported by results from the CONTEG study in Belgium, showing elevated levels in this type of eggs[4]. This possibility caused concern among health organisations in the Netherlands and in order to investigate the current situation, a study was started among private owners. Following a call via adds in local journals all through the Netherlands, more than 200 people volunteered to participate. A selection was made based on various criteria and samples from 62 unique addresses were collected and analysed for PCDD/Fs and PCBs by GC/MS. The study shows that average levels are about ten-fold higher than commercial eggs and may substantially contribute to the intake of this group of consumers. The data confirm that the levels around Harlingen were not unusual for hens from private owners.

Materials and methods

Egg samples were analysed by GC/HRMS for PCDD/Fs and PCBs by routine methods applied at RIKILT. Fat was extracted using the ASE 350 (Dionex, USA) with hexane/acetone (1:1) as extraction solvent. After drying and determining the fat content, the fat was spiked with the ^{13}C labeled standards and purified using the PowerPrep™ system (FMS). The clean-up resulted in the collection of two purified fractions, one with mono-ortho dl-PCBs and non-dioxin-like PCBs and the other with dioxins and non-ortho dl-PCBs. Both fractions were concentrated using the Power-Vap (FMS) and analyzed by GC-HRMS using an Agilent (Wilmington, USA) gas-chromatograph 6890N (GC column DB5 MS 60m, 0.25mm i.d., 0.25µm; J&W, Folson, USA) and an AutoSpec Ultima high resolution mass spectrometer (Waters, Milford, USA) operated in electron impact ionization mode using selected-ion monitoring and controlled by Masslynx data system. GC-HRMS data were processed using Masslynx Targetlynx software to determine the concentrations and subsequently the TEQ levels, based on WHO-TEFs 2005. The performance of the methods is regularly checked by participation in PT-tests.

Results and discussion

Levels in eggs

Levels of PCDD/Fs, dl-PCBs and ndl-PCBs in the various samples from the 62 addresses are presented in Figure 1. There was a large variation in the types of chickens and also the size of the eggs. In two cases the owners collected separately the eggs from relatively small silky fowls (Gallus gallus domesticus brisson) as well as other hens. Overall, the levels were relatively high with median values of 2.8 pg TEQ/g fat for PCDD/Fs, 2.0 pg TEQ/g fat for dl-PCBs and 4.8 pg TEQ/g fat for the sum of both. The average level of 6.1 pg TEQ/g fat for the sum was about 10-fold higher than the average level in eggs in the Netherlands as determined in the monitoring project on samples from the National Residue Control Plan. Although EU-MLs don't apply for these eggs, respectively 33 and 25% of the levels were higher than the MLs for PCDD/Fs and the sum of PCDD/Fs and dl-PCBs of 2.5 and 5 pg TEQ/g fat. Levels of the ndl-PCBs showed a median of 13 ng/g fat and were higher than the ML of 40 ng/g fat in 6% of the cases. The highest observed sum-TEQ level was 19 pg TEQ/g fat, with 13 samples containing more than 10 pg TEQ/g fat. However, also some rather low levels were observed.

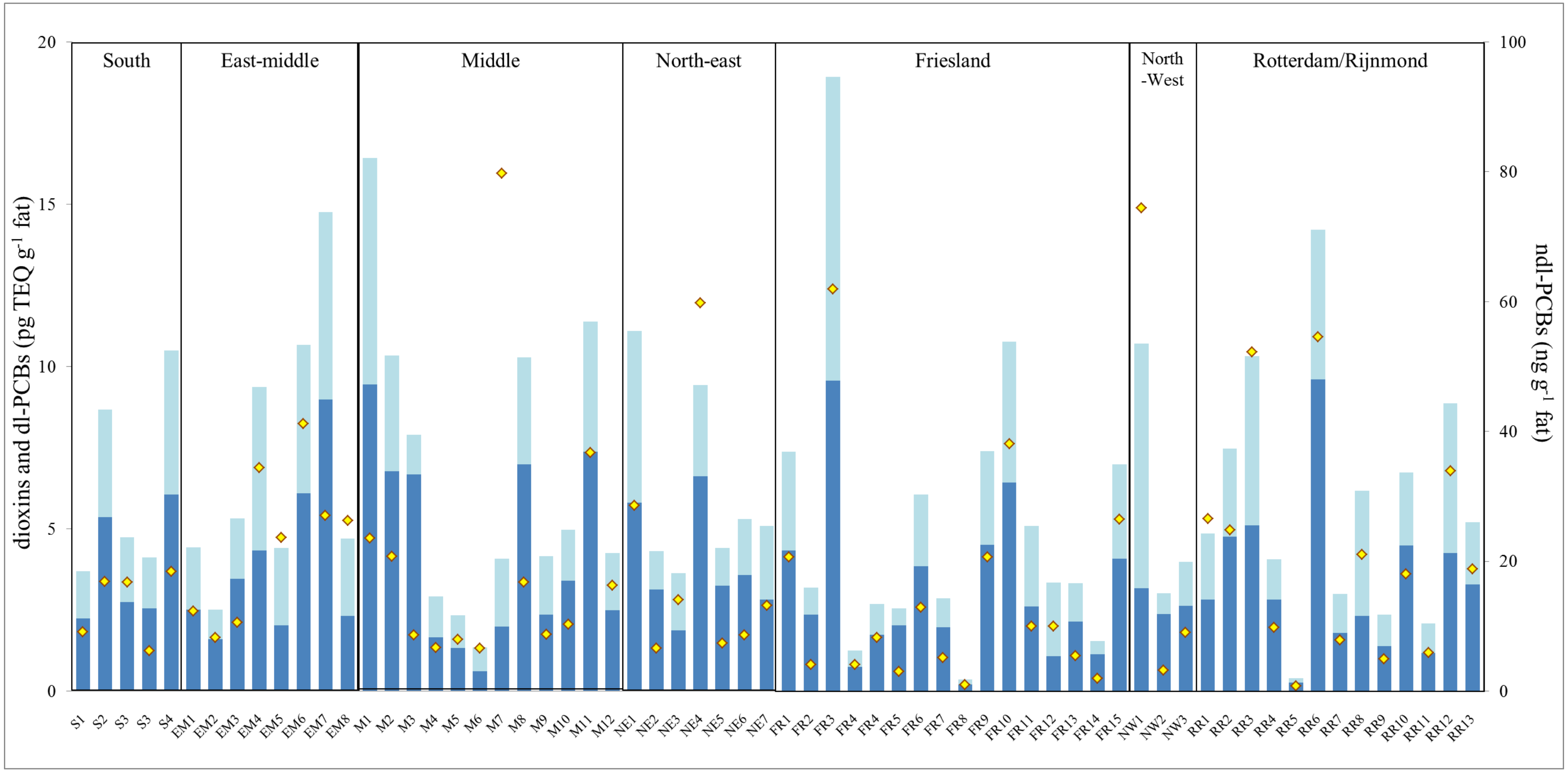


Figure 1. Levels of PCDD/Fs (dark blue), dl-PCBs (light blue) and ndl-PCBs (yellow diamonds) in the egg samples collected in different regions in the

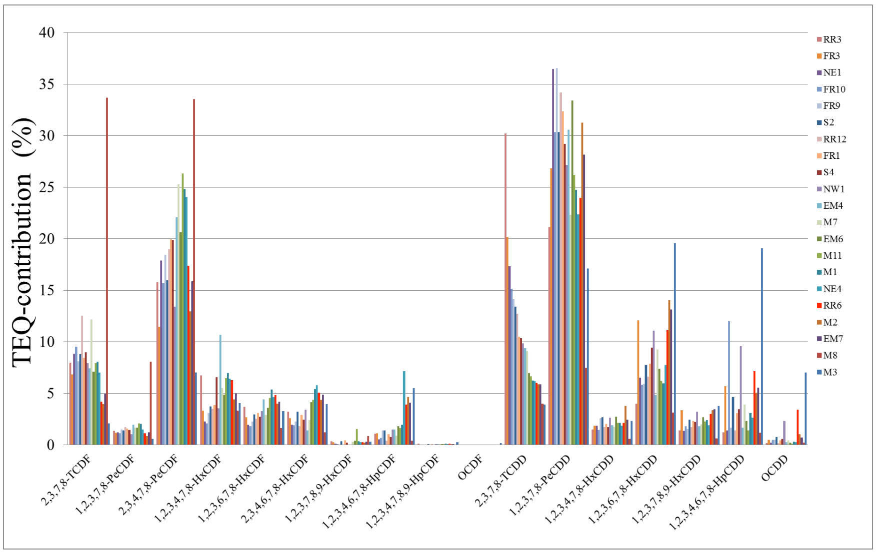
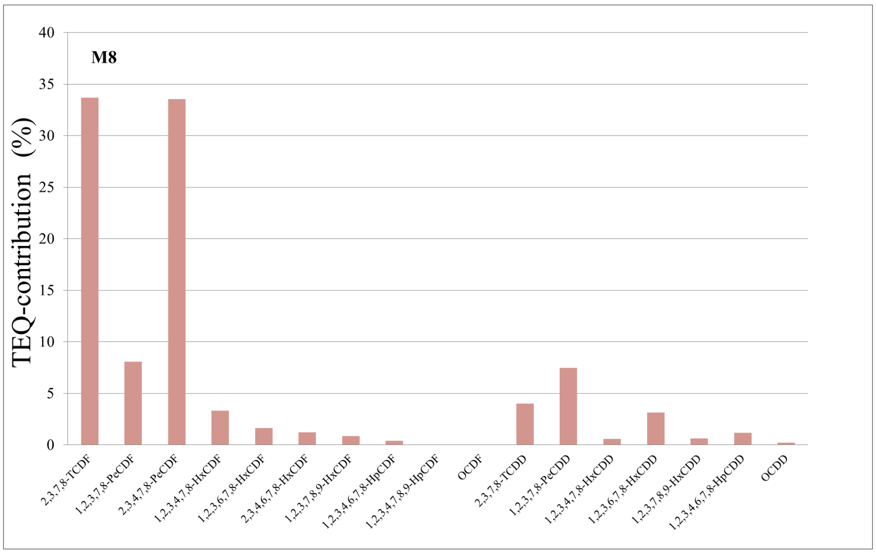
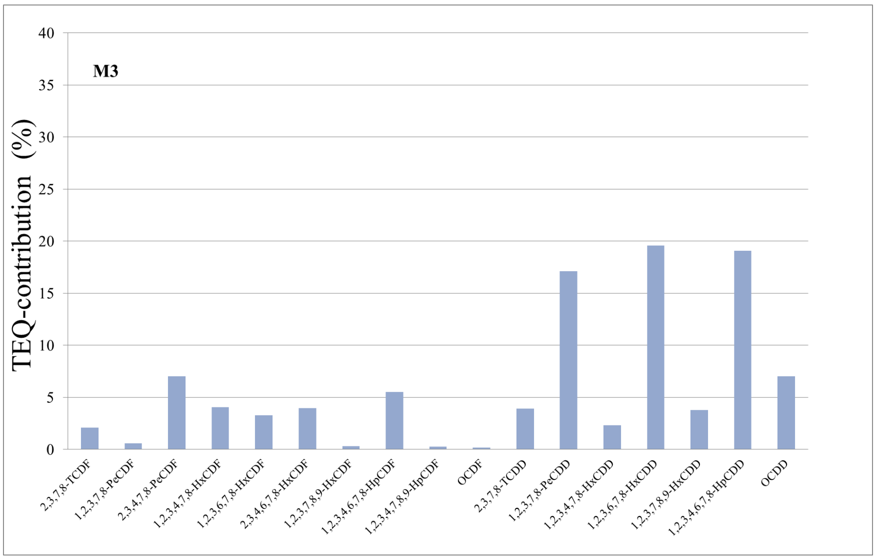


Figure 2. PCDD/F congener patterns observed in the eggs with the highest levels, expressed as relative contribution to the PCDD/F-TEQ. Samples M8 and M3 showed clearly different patterns.




M3
TEQ-contribution (%)
40
35
30
25
20
15
10
5
0
2,3,7,8-TCDF
1,2,3,7,8-PeCDF
2,3,4,7,8-PeCDF
1,2,3,4,7,8-HxCDF
1,2,3,6,7,8-HxCDF
2,3,4,6,7,8-HxCDF
1,2,3,7,8,9-HxCDF
1,2,3,4,6,7,8-HpCDF
1,2,3,4,7,8,9-HpCDF
OCDF
2,3,7,8-TCDD
1,2,3,7,8-PeCDD
1,2,3,4,7,8-HxCDD
1,2,3,6,7,8-HxCDD
1,2,3,7,8,9-HxCDD
1,2,3,4,6,7,8-HpCDD
OCDD

There were no clear differences between the regions, despite the fact that the Rotterdam/Rijnmond area is much more industrial than e.g. Friesland, in the North of the Netherlands. The data confirm that the levels around Harlingen were not unusual for hens from private owners.

Congener patterns

Congener patterns are an important tool for obtaining an indication of the potential source of a contamination. Previously we observed e.g. farms with problems specifically related to PCBs. However, as shown in Figure 1, there were no eggs where dl-PCBs contributed primarily to the TEQ-level. On average the contribution of dl-PCBs was 41% with a range of 38 to 49%. When focusing on the PCDD/F-TEQ, the relative contribution of PCDFs was relatively stable, with a median of 41%. This is also reflected in Figure 2, showing the PCDD/F congener patterns for the samples with the highest levels and two deviating patterns. In most cases the pattern reflects the general background pattern in the Netherlands, and very similar to patterns related to burning of waste. This was confirmed by interviews with owners of some of the most contaminated eggs, stating that the hens were foraging on areas where in former times waste was burnt.

The pattern observed in sample M8 showed a relatively high contribution of PCDFs, pointing to a PCB-contamination, but this was not confirmed by the PCB-levels and contribution to the sum-TEQ. Also the pattern observed in M3, with a relatively high PCDD contribution, was not recognized from our database.

Exposure of consumers eating their own eggs

When asked, participant reported to obtain on average 37 eggs in the week with some seasonal variation during the year. In general, these eggs were consumed by their households and were distributed to family and neighbors. The fat content of the eggs was on average 5.0 gram with a range of 2.0 to 7.2. This implies that consumption of an average egg with 6.1 pg TEQ/g fat amounts to 30 pg TEQ being respectively 11 and 4% of the TWI for a child of 20 kg b.w. and an adult of 60 kg b.w. For the highest observed level of 19 pg TEQ/g fat, these values are about 3-fold higher, i.e.34 and 13%. This comes on top of the background exposure from other sources. The median background exposure to PCDD/Fs and dl-PCBs for the general population (7-69 years) in the Netherlands was recently[5] estimated to be 0.5 pg TEQ/kg bw per day (or 3.5 pg TEQ/kg bw per week) (median) and the high background exposure (95^{th} percentile) 1.0 pg TEQ kg/bw per day (or 7.0 pg TEQ/kg bw per week). For children (2-6 years), these values were 0.9 pg TEQ/kg bw per day (median) and 1.5 pg TEQ/kg bw per day (95^{th} percentile). In these calculations, average egg concentrations of 0.6 pg TEQ/g fat were used and eggs contribute only to a small extent to the background exposure. For adults, weekly consumption of a few home produced eggs does not lead to exceedance of the TWI. But children with a high background exposure could exceed the TWI when consuming one egg with the highest TEQ level or two eggs with an average TEQ level per week. Consumption of own eggs with relatively high TEQ concentrations can substantially contribute to the total exposure to PCDD/Fs and dl PCBs.

Conclusion

Home-produced eggs can be an important source of dioxins and dl-PCBs and owners should attempt to lower the levels by measures to reduce the levels in soil and the uptake of soil by laying hens.

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