

POLYCHLORINATED DIBENZODIOXINS, DIBENZOFURANS AND BIPHENYLS IN PAIRED SAMPLES OF LAMB MEAT AND LIVER: LEVELS AND TRENDS

Mortimer DN*

Food Standards Agency, Aviation House, 125 Kingsway, London WC2B 6NH, United Kingdom

Introduction

A survey for dioxins and PCBs in offal, carried out by the Food Standards Agency in 2005, found a rate of non-compliance with the existing European limit for dioxins of almost 50% in the case of sheep liver.¹ These data were presented at the Dioxin 2008 symposium in Birmingham and, at the time, it was suggested that the limit established by the European Commission in 2002 had been set at too low a level, due to a lack of representative data.² Risk assessment indicated that the levels actually found were unlikely to have any significant impact on overall dietary exposure to dioxins. It was noted that the dioxin/PCB ratios for the liver were higher than those normally seen in carcass meat and it was postulated that this may be as a result of preferential sequestration in the liver of certain congeners, notably furans, through protein binding. The samples that had been tested were randomly-purchased retail samples, for which there was very little additional information. There was no evidence that the non-compliant samples had come from animals raised in localized contamination hotspots. In response to the request for a revision of the limit, the Commission called for further data, on the basis of which the European Food Safety Authority (EFSA) would conduct a review. As part of the response, the UK Food Standards Agency carried out this further study, taking samples of both meat and liver from animals of known age, gender and origin, in order to gain a better understanding of the contamination trends.

Table 1. Sample breakdown

Type	Age (years)	Number
Spring lamb	3-4 months	4 (2M, 2F)
Hogget	1 (approx.)	5
Ewe	2	3
	3.5	1
	4	3
	5	5
	6	3
Total		24

Materials and methods

Samples of lamb shoulder meat and liver were collected at slaughter by the Official Veterinarian Surgeon on duty at the plant. They were supplied to the UK Food and Environment Research Agency (Fera, formerly Central Science Laboratory) with details of the age, gender and origin of the animal. They were analysed by high resolution gas chromatography coupled with high or low resolution mass spectrometry, for the 17 dioxin and furan congeners and 12 dioxin-like PCBs assigned Toxic Equivalency Factors by the World Health Organisation. The full methodology has been described previously.³

Results and discussion

A summary of the animals sampled is provided in Table 1. In the case of hoggets (approximately one year-old), the gender was not given. All animals older than 1 year were female. Origins included farms in Northwest Wales (6), Anglesey (5), North Wales (4), Northwest England (3), Southwest Wales (2), Northeast England (2) and Scotland (2).

The analytical results are summarised in Table 2. The whole weight results for shoulder meat are not included as these are heavily influenced by the amount of fat on an individual sample. The data in Table 2 are ordered from lowest to highest on the basis of the total TEQ for the shoulder fat. Samples that would fail to comply with the most recent limits of 4.5 and 10.0 pg WHO₂₀₀₅-TEQ/g fat for dioxins and dioxins plus PCBs respectively in liver, which are set out in Commission Regulation 1259/2011 and apply the WHO-2005 TEFs,⁴ are highlighted. In all, there are ten non-compliant samples out of a total of 24 i.e. 42%. None of the shoulder meat samples were

above the revised limits of 2.5 and 4.0 pg/g fat for dioxins and dioxins plus PCBs respectively. In fact, none were even approaching the revised action level of 1.75 pg/g (applicable to both dioxins and dioxin-like PCBs) set out in Commission Recommendation 2011/516 on the reduction of the presence of dioxins, furans and PCBs in feed and food.⁵ This is highly significant, as it represents strong evidence that none of the animals from which samples were taken had been exposed to atypical levels of dioxins or PCBs.

Table 2. Upper bound results (pg WHO₂₀₀₅-TEQ/g)

Age	Shoulder, fat			Liver, fat			Liver, whole		
	Dioxin	PCB	Total	Dioxin	PCB	Total	Dioxin	PCB	Total
4	0.10	0.08	0.19	3.62	1.77	5.38	0.18	0.09	0.27
1	0.12	0.07	0.19	4.31	1.07	5.38	0.28	0.07	0.35
1	0.09	0.11	0.21	0.65	0.47	1.12	0.06	0.07	0.13
6	0.09	0.12	0.21	1.62	1.04	2.65	0.09	0.05	0.13
5	0.09	0.15	0.25	2.01	1.49	3.50	0.10	0.06	0.16
2	0.17	0.08	0.25	3.13	1.13	4.26	0.20	0.07	0.27
5	0.13	0.14	0.28	1.74	1.04	2.77	0.10	0.06	0.15
5	0.18	0.16	0.34	3.05	1.77	4.81	0.19	0.11	0.30
5	0.21	0.14	0.35	3.04	1.39	4.43	0.27	0.12	0.39
1	0.20	0.16	0.36	1.80	1.28	3.09	0.10	0.07	0.17
1	0.20	0.22	0.42	4.87	3.42	8.29	0.26	0.18	0.43
2	0.26	0.18	0.45	5.79	1.86	7.65	0.24	0.07	0.31
3.5	0.23	0.23	0.46	2.32	1.79	4.11	0.24	0.19	0.43
4	0.28	0.18	0.46	2.03	0.74	2.77	0.18	0.07	0.25
6	0.28	0.18	0.47	5.07	1.72	6.79	0.27	0.09	0.36
4	0.27	0.29	0.55	5.06	2.81	7.86	0.25	0.14	0.39
1	0.36	0.25	0.61	2.08	0.89	2.97	0.14	0.06	0.21
0.33	0.41	0.23	0.64	3.41	0.89	4.30	0.25	0.07	0.31
5	0.47	0.30	0.77	5.24	2.48	7.72	0.32	0.15	0.46
0.33	0.53	0.25	0.78	6.14	1.50	7.64	0.32	0.08	0.40
2	0.57	0.34	0.91	9.35	2.84	12.2	0.63	0.19	0.81
0.33	0.68	0.42	1.10	6.04	2.45	8.48	0.27	0.11	0.38
6	0.82	0.37	1.19	28.9	7.66	36.5	1.40	0.37	1.77
0.25	0.94	0.49	1.43	15.2	4.61	19.9	0.83	0.25	1.08
Min	0.09	0.07	0.19	0.65	0.47	1.12	0.06	0.05	0.13
Mean	0.33	0.21	0.54	5.75	2.09	7.84	0.32	0.11	0.43
Max	0.94	0.49	1.43	28.9	7.66	36.5	1.40	0.37	1.77

Commission Regulation 1259/2011 saw the introduction of limits for non dioxin-like PCBs for the first time at a European level. The limits for both meat and liver are 40 ng/g fat for the sum of CBs 28, 52, 101, 138, 153 and 180. The levels found in this work are not presented as they were all low, with maximum observed levels of 4.6 ng/g fat for shoulder meat and 14.6 ng/g fat for liver (maximum 0.8 ng/g on a whole weight basis). There were no apparent trends in the data about which any conclusion could be drawn and it is assumed that any significant differences in absorption and binding behaviour between meat and liver would only become apparent if animals were exposed to atypically high levels of non dioxin-like PCBs.

Recent work carried out by the European Union Reference Laboratory for Dioxins has demonstrated that the solvent extraction process used for liver analysis can have a significant influence on the concentration of dioxins and PCBs reported on a *fat* basis, although this same influence is not seen for results reported on a whole weight basis.⁶ The implication of this is that the dioxins and PCBs are extracted efficiently from the liver whichever

solvent is used, whilst fat extraction efficiency is more variable. The point at which internal standards are added to the sample extract may also have an influence. For the current work, the same solvent system was used throughout and the correlation between fat concentration and whole weight concentration in the liver samples was very high ($R^2 = 0.97$). In any event, subsequent discussion will focus on shoulder fat and liver whole weight concentrations.

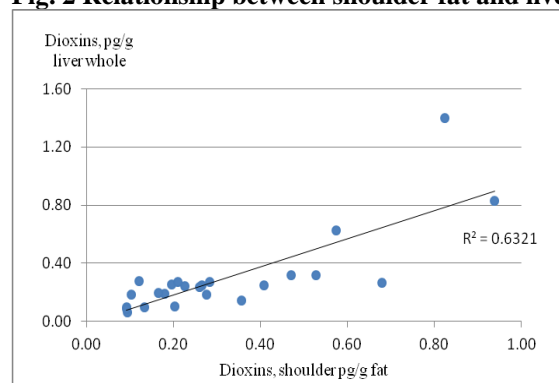
In considering possible trends in the data, the age of the animals was expected to have some influence. Dioxins and PCBs are bioaccumulative and an upward trend in level of contamination with age would be the expected outcome. On the contrary, however, age appeared to have surprisingly little influence on levels of contamination with, if anything, a very slight downward trend for the levels in shoulder fat ($R^2 < 0.1$) and no trend at all for liver ($R^2 < 0.01$, Figure 1). There did not appear to be any correlation between age and the fat content of the livers. This is not to say that time has no influence on the level of contamination in an individual animal, particularly since all of the animals greater than one year old were female, and it is possible that repeated birthing would influence dioxins levels in the animal. A better understanding could only be achieved through long-term studies and bio-monitoring. On the other hand, it does eliminate the option of controlling levels of contamination in meat and liver entering the food chain by restricting the age of sheep at slaughter.

The data were examined to see if there was any evidence for a relationship between shoulder fat and liver concentrations and, indeed, for both dioxins and total TEQ there appears to be a reasonable correlation ($R^2 = 0.55$ - 0.63). This suggests that the overall level of exposure of an animal is reflected in both the shoulder fat and liver. Figure 2 provides an illustration of this relationship in the case of dioxins.

Fig.1 Time trend for liver



Fig. 2 Relationship between shoulder fat and liver



There are two indications that dioxins and PCBs accumulate differently in shoulder fat and liver. Firstly, the dioxin/PCB ratios are different for the two tissue types (Table 3). The ratios are consistently higher in liver, indicating the preferential binding of dioxin/furan congeners. Furthermore, there are distinct differences in the congener profiles between shoulder fat and liver. Figure 3 shows the absolute levels of dioxins and furans in the two samples from a five year-old ewe. The dominance of the furans, which are significant contributors to the TEQ, is very apparent and appears to be characteristic of all sheep liver and probably liver in other species. Results from work recently carried out on behalf of the Food Standards Agency to investigate the impact of flooding events shows a similar distinction between meat and liver in the case of beef cattle.⁸ Changes to profile associated with differences in the contamination source seem to be reflected more in the shoulder fat samples.

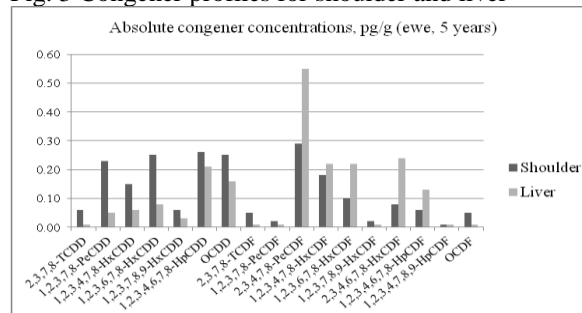
The TEQ levels reported for sheep liver in the current work are consistent with those reported previously and the conclusion that there was no additional concern for health remains valid.² Both the previous and current data were included in the review carried out by the EFSA Panel on Contaminants in the Food Chain, which published its opinion on the risk to public health related to the presence of high levels of dioxins

and dioxin-like PCBs in liver from sheep and deer in July 2011. EFSA concluded that the frequent consumption of sheep liver by women of child-bearing age and children might be a health concern, although this was based on liver with an average TEQ concentration of 26.1 pg/g fat, which is considerably higher than the mean value found in this work (7.8 pg/g fat, Table 2). It is possible that a large proportion of the liver data reviewed by

Table 3. Dioxin / PCB ratios

	Shoulder ratio	Liver ratio	Liver ratio / shoulder ratio
Min	0.62	1.30	1.09
Median	1.46	2.26	1.70
Mean	1.40	2.44	1.75
Max	2.25	4.09	2.33

Fig. 3 Congener profiles for shoulder and liver



EFSA came from animals that had been more highly exposed (over 60% of the results in the EFSA dataset were submitted by Germany) although, as noted above, the fat-based concentration is heavily influenced by the extraction method used. It is also worth noting that excessive consumption of liver is inadvisable due to concerns about vitamin A toxicity.

In conclusion, this work has demonstrated that sheep liver exceeding the current European regulatory limits for dioxins and dioxins plus dioxin-like PCBs, in some cases by a significant margin, did not come from animals exposed to high levels of contamination, since levels in the fat of shoulder meat were well within compliance. Age appears to have little influence on the levels of contamination. Congener profiles for liver are dominated by penta- and hexachloro-dibenzofuran congeners whereas profiles for meat fat are more variable and, it is assumed, reflect the profile of background exposure.

Acknowledgements

This work was funded by the UK government. The author would like to thank Welsh Country Foods for providing the samples and the Food and Environment Research Agency for the sample preparation and analysis.

References:

1. Fernandes A, Mortimer DN, Rose MD, Gem M. (2010) *Chemosphere* 81(4): 536-540.
2. Mortimer DN, Gem M, Rose, MD, Fernandes A, White S. (2008) *Organohalogen Compounds* 70: 883-886.
3. Fernandes A, White S, Dsilva K, Rose M. (2004) *Talanta* 63: 1147-1155.
4. Commission Regulation (EC) No. 1259/2011. *Official Journal of the European Union* L320: 18-23.
5. Commission Recommendation 2011/516/EU. *Official Journal of the European Union* L218: 23-25.
6. European Union Reference Laboratory for Dioxins and PCBs in Feed and Food. Reference 5477.10-15 EURL-Dioxin. Available at http://ec.europa.eu/food/food/chemicalsafety/contaminants/emergency_analyses_en.pdf
7. EFSA Panel on Contaminants in the Food Chain (CONTAM); Scientific Opinion on the risk to public health related to the presence of high levels of dioxins and dioxin-like PCBs in liver from sheep and deer. *EFSA Journal* 2011;9(7):2297
8. Rose M, Lake I, Foxall C, Fernandes A, Lewis M, White O, Mortimer D, Dowding A (2012). PCDD/F and PCB levels in beef from cattle raised on flood plains. Submitted to *Organohalogen Compounds*.