MEASUREMENT OF HUMAN DIETARY INTAKE OF DIOXIN-LIKE COMPOUNDS (DLCs) BY THE DUPLICATE DIET METHOD

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Introduction

Previous estimates of UK human dietary exposure to dioxin-like chemicals (DLCs) – *viz* PCDD/Fs and PCBs - have been indirect, *i.e. via* total diet studies. As part of a study of the absorption of human dietary intake of DLCs[1)], we obtained direct estimates of human exposure of 14 male subjects *via* the duplicate diet approach. Subjects were divided into two age groups: one aged 23.9±4.5 years and the other aged 49.4±5.2 years. All subjects were male, with BMI values in the range 23.1±2.2 (younger group) and 23.2±2.5 (older group). In addition to studying the subjects' normal omnivorous diets, we also studied intakes resulting from ingestion of a vegan diet. We anticipated that such intakes would be low, as the general consensus is that the principal vector of non-occupational exposure to DLCs is *via* the ingestion of animal fats. The following DLCs were studied: PCB congeners 18, 28, 31, 37, 47, 49, 51, 52, 77, 81, 99, 101, 105, 114, 118, 123, 126, 128, 138, 153, 156, 157, 167, 169, 180, and 189; and all seventeen 2, 3, 7, 8-chlorinated PCDD/Fs.

Materials and Methods

Full details of sampling and analytical protocols are contained in another presentation to this conference[1)]. In summary, exposure of each subject was measured via the duplicate diet approach over two separate 1 week periods (one for the normal and one for the vegan diet). Aliquots of homogenised pooled diet samples were freeze-dried and analysed *via* GC/MS for DLC content.

Results and Discussion

A summary of the results is as follows:

- Reassuringly, the mean exposure of all subjects to PCDD/Fs and PCBs both separately and combined (expressed as WHO-TEQ) are slightly lower than most recent TDS-based exposure estimates; are within the WHO' s proposed TDI of 1-4 pg WHO-TEQ/kg bw/d; and – although not strictly comparable with previous TDS data - are consistent with a decreasing temporal trend in the UK – see Table 1.
- As shown in Table 2, for all subjects, the mean upper bound daily exposure to PCDD/Fs and PCBs combined during the normal diet trials was 1.29 pg WHO-TEQ/kg bw (range 0.45-2.33). Mean upper bound ΣPCB daily exposure during the normal diet trials was 2.85 μg ΣPCB/person

(range 0.13-8.85). Lower bound estimates for the normal diet trials (all subjects) were: for PCDD/Fs and PCBs combined, daily mean exposure was 0.95 pg WHO-TEQ/kg bw (range 0.03-2.25), while mean ΣPCB daily exposure was 2.84 μg ΣPCB/person (range 0.13-8.85). Clearly, even within a group of just 14 individuals, there is a wide range of exposures.

- By comparison, the mean ΣPCB exposure is higher than the most recent TDS-based estimate of 0.34 μg ΣPCB/person/d (which excluded fruit, vegetables and other non-fatty foodstuffs), but is consistent with recent estimates for other industrialised countries that did include contributions from fruit and vegetables– see Table 3.
- Average daily exposures to PCDD/Fs *and* PCBs combined during the normal diet trials for younger subjects (1.56 pg WHO-TEQ/kg bw upper end; 1.43 pg WHO-TEQ/kg bw lower end) exceeded those for the older group (1.15 pg WHO-TEQ/kg bw upper end; 0.53 pg WHO-TEQ/kg bw lower end). While this is partly attributable to the different food consumption rates (on average 0.066 kg dry food per week per kg body weight for the younger group *c.f* 0.057 kg dry food per week per kg body weight for the older group), the younger group's diet had a higher DLC content – expressed as WHO-TEQ.
- Average daily exposures to ΣPCB during the normal diet trials for younger subjects (5.01 μg ΣPCB/person upper and lower bound) exceeded those for the older group (0.99 μg ΣPCB/person upper and lower bound). Although this is partly attributable to the higher food consumption rates of the younger subjects, the younger group's diet appears to have a far higher ΣPCB content. Detailed scrutiny of the exposure data for each age group, showed that while exposures to penta, hexa, and heptachlorinated PCBs were similar for the two groups, exposures to the tri- and tetrachlorinated congeners were much higher for the younger group. While we cannot be certain, the marked elevation of the tri- and tetrachlorinated PCBs - which are the predominant congeners in herbage – suggests that the diets prepared for the younger subjects were prepared using an unusually contaminated batch of vegetable oil.
- We hypothesised that administering a vegan diet would minimise DLC intakes, on the basis that previous research had shown comestibles based on animal fats to constitute the principal vector of DLC exposure. Comparison of DLC exposures on a pg WHO-TEQ/kg bw/d basis, revealed that average intakes of PCDD/Fs and PCBs – both separately and combined – were appreciably lower during the vegan diet trials for both age groups (Table 2). Interestingly, while ΣPCB exposures during the normal diet trials for the older group far exceeded those for the corresponding vegan diet trials; a similar comparison for the younger group, showed ΣPCB exposures during the normal diet trials to be comparable to those in the corresponding vegan diet trials. In fact, for some subjects, exposures to some tri- and tetrachlorinated PCBs were higher in the vegan diet trial. This greater exposure during the vegan diet trials cannot be attributed to a higher food consumption rate (on average 4.66 kg dry food per week during the normal diet trials *c.f* 4.33 kg dry food per week during the vegan diet trials), and is instead attributable to the fact that the vegan diet has higher concentrations of these congeners. In order to minimise weight loss during the vegan diet trial, it was necessary to prepare the vegan diet in such a way that its calorific value as far as possible matched those of normal omnivorous diets. To achieve this, the vegan diet contained a higher than normal content of vegetable oils. As discussed above, we consider it

possible that the oil used during the younger group's trials (both diets) was from an unusually contaminated batch.

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Table 1: Temporal Trend in Average Adult UK Dietary Exposure (pg WHO TEQ /kg bw/d) to PCDD/Fs and PCBs

Year	PCDD/Fs	PCBs	PCDD/Fs + PCBs	Reference
1982	4.6	2.6	7.2	2
1992	1.6	0.9	2.5	2
1997	0.8	0.9	1.7	2
1999	0.64	0.65	1.29	This study (normal diet – all subjects; upper bound estimates)

Table 2: Summary of Daily Dietary Intake Data for this Study (Expressed as pg WHO TEQ / kg bw/ d unless otherwise stated)

Subject Group/ Statistical Parameter	PCDD/Fs (Normal Diet)	PCDD/Fs (Vegan Diet)	PCBs (Normal Diet)	PCBs (Vegan Diet)	PCDD/Fs + PCBs (Normal Diet)	PCDD/Fs + PCBs (Vegan Diet)	PCBs (Normal Diet – µg ΣPCB/ person/ d)	PCBs (Vegan Diet – µg ΣPCB/ person/ d)
[1]Average ± σ_{n-1}	1.00±0.74	0.046±0.109	0.43±0.23	0.21±0.11	1.43±0.83	0.25±0.18	5.01±2.72	3.30±1.07
[1]Range	0.03-1.91	0.001-0.268	0.16-0.71	0.07-0.36	0.32-2.25	0.07-0.55	1.69-8.85	1.85-5.05
[2]Average ± σ_{n-1}	1.01±0.74	0.059±0.108	0.54±0.22	0.29±0.14	1.56±0.83	0.35±0.21	5.01±2.72	3.30±1.07
[2]Range	0.04-1.92	0.011-0.280	0.30-0.78	0.07-0.49	0.45-2.33	0.08-0.67	1.69-8.85	1.85-5.05
[3]Average ± σ_{n-1}	0.13±0.22	0.02±0.01	0.40±0.49	0.02±0.02	0.53±0.67	0.04±0.03	0.99±1.01	0.18±0.16
[3]Range	0.01-0.63	0.01-0.03	0.02-1.25	0.00-0.07	0.03-1.88	0.02-0.09	0.15-2.59	0.04-0.51
[4]Average ± σ_{n-1}	0.34±0.19	0.23±0.10	0.81±0.32	0.46±0.12	1.15±0.47	0.70±0.20	0.99±1.02	0.18±0.16
[4]Range	0.20-0.72	0.13-0.37	0.47-1.28	0.33-0.63	0.70-2.00	0.45-0.96	0.13-2.59	0.05-0.51
[5]Average ± σ_{n-1}	0.53±0.68	0.031±0.072	0.41±0.38	0.108±0.117	0.95±0.86	0.14±0.16	2.84±2.82	1.62±1.76
[5]Range	0.01-1.91	0.001-0.268	0.02-1.25	0.003-0.360	0.03-2.25	0.02-0.55	0.15-8.85	0.04-5.05
[6]Average ± σ_{n-1}	0.64±0.61	0.15±0.13	0.65±0.30	0.38±0.15	1.29±0.68	0.53±0.25	2.85±2.82	1.62±1.76
[6]Range	0.04-1.92	0.01-0.37	0.30-1.28	0.07-0.63	0.45-2.33	0.08-0.96	0.13-8.85	0.05-5.05

[1]Younger subjects (lower bound); [2]Younger subjects (upper bound); [3]Older subjects (lower bound); [4]Older subjects (upper bound); [5]All subjects (lower bound); [6]All subjects (upper bound); lower bound means where concentration of a DLC was below the detection limit. concentration was assumed to be zero; upper bound means that where concentration was below detection limits, concentration was assumed to equal the detection limit

Table 3: Average Daily Adult Dietary Exposures to PCDD/Fs and PCBs (Based on WHO TEFs unless otherwise stated)

Type of sample analysed	Country	Year	PCDD/Fs (pg TEQkg bw)	PCBs (pg TEQ/kg bw)	ΣPCB (µg/person)	Reference/Comments
Duplicate normal diet	UK	1999	0.64±0.15	0.65±0.38	2.85 ± 2 .82[a]	This study (all subjects. upper bound)
Total diet study	UK	1997	0.8*	0.9*	-	2
Duplicate diet study	Italy	ns	-	-	3.5 ± 1.2[b]	3
Duplicate diet study	Netherlands	1994	0.53±0.23	0.92±0.24	1.2 ± 0.25[c]	4 –i-TEFs
Duplicate diet study	Italy	ns	-	0.66±0.36*	3.72 ± 1.51[a]	5 –i-TEFs
Estimate	UK	ns	-	-	0.56[a]	6
Total diet study	UK	1992	1.6	0.9	0.34[b]	2, 7
Total diet study	Spain	1996	3.5	-	-	8 –i-TEFs
Total diet study	New Zealand	ns	0.18	0.15	0.13[b]	9 – upper bound. adult males - i-TEFs
Duplicate diet study	Germany	ns	0.7±0.2	-	0.7±0.25[e]	10 – i-TEFs

Note: *Converted for comparison by assuming 60 kg bodyweight for subjects studied.; ns Not stated. [a] Sum of 26 congeners; [b] Sum of 25 congeners: [c] Sum of 29 congeners: [d] Sum of 29 congeners; [e] Sum of 10 congeners