Human absorption of PCDDs, PCDFs, and PCBs from food

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Introduction

Diet is the main source of human exposure to many highly lipophilic pollutants such as PCDD/Fs and PCBs. However, little is known about the degree of absorption of these contaminants from the human digestive tract. While several studies have reported that absorption of PCDD/Fs in nursing infants is nearly complete (> 90 %, e.g. ref.[1]), there has been very little work published on uptake in adults. In one experiment in which a scientist ingested radiolabeled 2,3,7,8-Cl_4DD the absorption was reported to be at least 87 %[2]. In another study contaminant excretion was measured in two individuals[3]. Although uptake was not quantified, the author concluded that for some congeners the theoretical uptake was much less than excretion. Hence the literature provides virtually no basis for estimating PCDD/F uptake in risk assessment.

As a first step towards a better understanding of this process, absorption of PCDDs, PCDFs, PCBs, and hexachlorobenzene (HCB) was determined in eight adults using a mass balance approach, where net absorption was measured as the difference between the daily intake of the contaminants via food and their daily excretion via feces.

Methods

Eight volunteers with no history of an occupational exposure to chlorinated organics were recruited for the study. For each volunteer 2 to 3 mass balances lasting 3 days were conducted.

Contaminant intake was determined with the duplicate method[4,5]: Identical portions of all food consumed by the volunteers including milk-containing beverages (the duplicates) were collected in precleaned glass and aluminium containers. After homogenizing with a mixer the samples were divided and one third was stored at -20 °C for later analysis. Feces were collected using vessels lined with aluminium foil. The connection between the feces and the food samples was established using Fe-capsules as tracers. Volunteers took one with the first and a second with the last meal of the collecting period. The passage times were determined from the colouring of the stool. All of the feces originating from the 3 day study period were stored at -20 °C. Prior to extraction all samples were freeze dried and homogenized.

80 g of food and 15 g of feces, respectively, were soxhlet-extracted for 20 hours with toluene containing ^{13}C-labelled internal standard. The clean up consisted of treatment with H_2SO_4-silica and two chromatographic columns of Alumina B Super 1 and resulted in three fractions: *PCDD/Fs, coplanar PCBs, and other PCBs/HCB.*

The HRGC/HRMS-analysis was carried out using a VG-Autospec Ultima mass spectrometer coupled to a DB5MS column for the PCBs and a RTX2330 column for the PCDD/Fs. Recoveries for 2,3,7,8-Cl_4DD were 80 to 90 % with a few exceptions of 70 %. 1,2,3,7,8,9-Cl_6DF could not be quantified because of interferences.

Results

Here we present the initial results for four volunteers, namely three males aged 25 to 28 years (M1, M2, M3) and one female of 24 years (F1). They were all non-smokers, of normal weight, took no medication, and had "typical" eating habits. For three of the volunteers data from one mass balance are presented, while for M1 two have been analysed. All samples for M1 were also worked up and analysed in duplicate. The results showed good precision for the analytical procedure and good agreement between the relative absorption for the two periods.

Table 1 shows the daily food intake and feces output on a dry weight basis. The measured values lie within reported ranges of 330 to 610 g d^{-1} for food uptake[4] and 20 to 50 g d^{-1} for feces excretion[6]. The I-TEQ intake in Table 1 (NATO-CCMS for PCDDs and PCDFs only) is in the range of 23 to 96 pg d^{-1} reported for 14 persons in North Rhine-Westfalia[4].

Net absorption was calculated as the difference between the amount ingested with diet and the amount excreted with feces. If the latter was greater than the former, the difference was defined as net excretion. The results are presented in Figure 1 as a percentage of the daily intake.

The absorption behaviour for the PCDD/F varied widely between the volunteers. M1 differed strongly from the other three volunteers showing net excretion for most congeners. Net absorption was highest for the lower chlorinated furans, reaching up to 89 % for 2,3,7,8-Cl_4DF. The greatest net absorption for 2,3,7,8-Cl_4DD was 73 %. On the other hand, net excretion was highest for the higher chlorinated dioxins, in particular Cl_8DD (up to 260 % of daily intake) and 1,2,3,6,7,8-Cl_6DD (up to 240 %). With the exception of M1 this was not observed for the higher chlorinated furans. Although high levels of 1,2,3,4,6,7,8-Cl_7DD in feces were obtained in agreement with previously reported results[3], two volunteers showed net absorption because of the high input levels. For F1 and M1 net excretion of this congener took place.

The results for the PCBs were completely different. The tri- to pentachlorinated congeners were absorbed at 80 to near 100 %, whereas the hexa- and hepta-congeners reached only 60 to 70 % and the octachloro-PCB 202 was taken up to an even lesser extent. The results for HCB were similar to these for the hexachlorobiphenyls. M1 showed lower absorption, especially for the higher chlorinated compounds, but no net excretion was observed.

Table 1: Daily intake and output: dry weights and I-TEQs (calculated from PCDD/Fs)

volunteer	daily intake		daily output		I-TEQ balance	
	dry weight [g d^{-1}]	I-TEQ [pg d^{-1}]	dry weight [g d^{-1}]	I-TEQ [pg d^{-1}]	absolute [pg d^{-1}]	relative to intake
M1 a	444	49.4	45	55.6	- 6.2	- 12 %
M1 b	456	47.0	42	49.9	- 2.9	- 6 %
M2	572	65.8	38	41.5	+ 24.3	+ 37 %
M3	417	50.1	34	38.6	+ 11.5	+ 23 %
F1	399	43.6	18	22.2	+ 21.4	+ 49 %

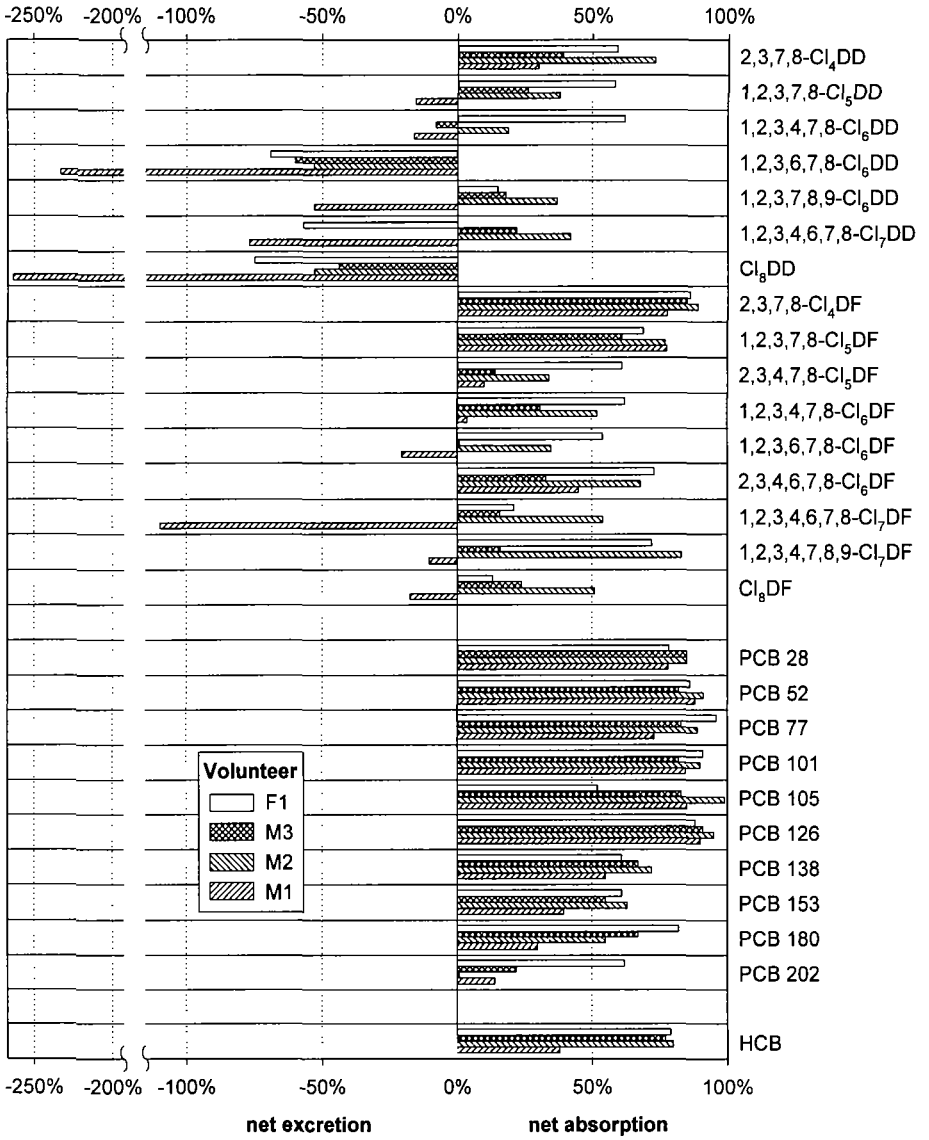

Figure 1: Net absorption of detected contaminants relative to daily intake; net excretion as a fraction of input where output exceeds intake (the values for M1 are means of two mass balances in both of which parallel samples were analysed).

Net absorption of I-TEQs (see Table 1) did not exceed 50 % of daily intake and was negative for M1, thus resulting in a net excretion of I-TEQs for this volunteer. Of the congeners, mainly 2,3,7,8-Cl_4DD and 2,3,4,7,8-Cl_5DF contributed to an I-TEQ net absorption, whereas I-TEQ excretion was dominated by 1,2,3,6,7,8-Cl_6DD. The inclusion of PCBs in the calculation would lead to a higher net absorption of TEQ because of the high net absorption for these compounds. For instance, PCB 126 with a TEF of 0.1[7] causes an additional net absorption of 31 pg TEQ d^{-1} for M1 in balance a.

Discussion and Conclusions

In no case was any PCDD/F congener completely absorbed. For several compounds a net excretion was observed, in particular for M1, the best studied volunteer. Cl_8DD and 1,2,3,6,7,8-Cl_6DD in particular were excreted to an extent that cannot be explained by food ingestion. In one volunteer, this resulted in a net excretion of I-TEQs. The sources of these compounds in the feces are as yet unknown and need to be investigated.

There are considerable differences between the volunteers in the absorption/excretion of PCDD/Fs. They seem not to be caused by sex or different food consumption, but by individual conditions. We hypothesize that absorption is a passive diffusion process, and that differences in body levels lead to different absorption and excretion behaviours at the same level of dietary uptake. In order to investigate this hypothesis, the volunteers' blood will be analysed to determine their body levels.

There is better agreement for the absorption of PCBs and HCB. Absorption was high compared to the PCDD/F and decreased with increasing chlorination. However, differences in absorption behaviour were observed for the higher chlorinated compounds. Because of their high net absorption, inclusion of PCBs in TEQ calculation leads to notable increases in TEQ net absorption.

These results raise doubts about the current methods of risk assessment that presume absorption of 100 % for all of these compounds. Some of them are clearly not absorbed at all. Further investigation is necessary to gain an understanding of the factors determining absorption of PCDD/Fs and PCBs in man.

References

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