The bioaccessibility of polychlorinated biphenyls (PCBs) and polychlorinated dibenzo-p-dioxins/furans (PCDD/Fs) in cooked plant and animal origin foods

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Introduction

For the general human population, intake of PCBs/PCDD/Fs sorbed to foodstuffs is regarded as the main route of exposure and it has been reported that approximately 90% of the human exposure to PCBs/PCDD/Fs originates from foods, especially those of animal origin (e.g. meat, egg, milk and their products) [1]. The extent to which compounds desorb from a matrix and become accessible for uptake by other organisms has been termed bioaccessibility. For sorbed compounds post ingestion bioaccessibility has been defined as "the maximal amount of contaminants released from the test matrix in a synthetic gastrointestinal system" [2]. When quantifying human exposure to POPs for risk assessment calculations, it is conservatively assumed that POPs sorbed to foods completely desorb in the gastrointestinal tract and are therefore 100% bioaccessible [3]. However, only a fraction of these chemicals is actually soluble in the digestive juice (bioaccessible) and therefore, total exposure/dose estimates likely overestimate the POPs bioaccessibility. To make a more accurate exposure assessment, bioaccessibility of contaminants in matrices, especially in foods, is critical.

Published bioaccessibility data for POPs in foods are very limited. Wang et al [4] reported that the average bioaccessibilities for DDT in market fish were 5.48% and 17.6% in the gastric and intestinal compartments, respectively. Xing et al [5] reported that the bioaccessibility of PCBs in freshwater fish (3%) was much lower than that in leafy vegetables (25%), concluding that the high lipid content of fish resulted in lower bioaccessibility. In contrast, Yu et al [6] reported a positive correlation ($P<0.001$) between the bioaccessibility of polybrominated diphenyl ethers (PBDEs) and fat content of 299 food items, including both vegetables and animal-based foods. Generally, more observations are needed to resolve this discrepancy. The effect of food preparation on the bioaccessibility of POPs is unknown and differences in preparation techniques might also play an important role in the bioaccessibility of sorbed PCBs/PCDD/Fs because they can change the structure and composition of food. Previous bioaccessibility studies of food-sorbed organics have used freeze-drying of raw materials to prepare the samples for in vitro testing [5-6]. However, foods are more frequently cooked prior to consumption. Although the culinary process might vary due to regional, cultural, or personal preferences, preparation methods like boiling in water or frying in cooking oil are universal processes which may influence the physical-chemical behavior of sorbed pollutants. For example, boiling significantly decreased the bioaccessibility of cadmium and lead in vegetables, but increased that of arsenic in rice.

The purpose of this study was to: 1) determine the bioaccessibility of PCBs and PCDD/Fs in boiled and fried foods, 2) discuss possible influencing factors therein, and 3) calculate the estimated daily intake (EDI) of selected PCBs/PCDD/Fs toxic equivalent (TEQ) from foods on a bioaccessibility-corrected basis, and compare these results with published data from the total dietary study (TDS) conducted by the Chinese government.

Materials and methods

Cooking procedure

Each of the six types of food samples was assigned to one of two groups, boiled or fried. Triplicates of each food type were prepared by boiling, and three by frying. For samples to be prepared by boiling; 1.5 g subsamples were spiked with 2000 pg of 13C12-labeled PCDD/Fs (EDF-8999), dioxin-like (mono and non-ortho) PCBs (WP-LCS), and indicator PCBs (EC-9605 SS) to serve as surrogate standards (sample processing controls). The samples were then rehydrated with 4.5 mL ultrapure water. Following hydration 4.0 mL ultrapure water was added to the homogenate, and the samples were boiled at 100 °C on a hot plate for 5 min. The 1.5 g samples in the fried group were similarly spiked with 2000 pg of the surrogates for the PCBs and PCDD/Fs, then rehydrated with 4.5 mL ultrapure water. Following rehydration, the mixtures were fried in 1.0 g of a cooking oil made from corn and soybeans (negligible dioxin/PCB concentration) at 200–300 °C on a hot plate for 5 min.

In vitro digestion procedure

The process for determining the bioaccessibility of PCDD/Fs and PCBs in foods was similar to the method developed by Oomen et al [7] which describes a three-step procedure simulating digestive processes in the mouth, stomach, and small intestine. In brief, each cooked meal (with the cooking matrix, either water or oil) was transferred into a 50mLcentrifuge tube, to which 6mL of simulated saliva was added and the samples were vortexed for 2 min. Then, 13mL of synthetic gastric juice were added, the pH was adjusted to 1–2, and the tube was incubated in a reciprocating water bath for 2 h (37 °C, 60 rpm). In the next step, 12 mL of duodenal juice and 6 mL of bile juice were added and the pH was adjusted to 7.5–8.0 using 1 M $NaHCO_3$ and 1 M HCl. The tube was incubated in the water bath for another 2 h (37 °C, 60 rpm) then centrifuged for 5 min at 18,000 ×g. The supernatant (chyme) was transferred into a separatory funnel containing 100 mL hexane to perform liquid-liquid separation.

Results and discussion

When the foods were prepared by boiling, the mean bioaccessibility (%) in rice (PCBs: 16.5±1.0, PCDD/Fs: 4.9±0.3) and cabbage (PCBs: 4.2 ± 0.9, PCDD/Fs: 1.9 ± 0.7) were lower than in animal origin foods (beef, PCBs: 49.0 ± 3.3, PCDD/Fs: 7.8 ± 0.9; egg, PCBs: 29.7 ± 3.1, PCDD/Fs: 8.6 ± 1.3; fish, PCBs: 26.9 ± 2.5, PCDD/Fs: 7.9 ± 1.3; milk powder, PCBs: 72.3 ± 1.6, PCDD/Fs: 28.4 ± 1.2). When fried in cooking oil, the bioaccessibilities of all analytes in all foods increased, but the increase in plant based foods (rice, PCBs: 3.4×, PCDD/Fs: 3.6×; cabbage, PCBs: 10.3×, PCDD/Fs: 7.9×) was greater than that of animal origin foods (beef, PCBs: 1.6×, PCDD/Fs: 3.4×; egg, PCBs: 2.1×, PCDD/Fs: 1.8×; fish, PCBs: 2.8, PCDD/Fs: 3.2×). Comparison of PCBs/PCDD/Fs bioaccessibility in rice and cabbage showed that bioaccessibility was greater in the low fat, high carbohydrate/protein content food (rice) than in the low carbohydrate/protein, low fat content food (cabbage), regardless of the method used to prepare the food. Adjusting for bioaccessibility reduced the gross estimated daily intake (EDI) of 112 pg WHO-TEQ/day, by 88% and 63% respectively for foods prepared by boiling and frying.

Our results indicate that: 1) The method used for cooking is an important determinant of PCBs/PCDD/Fs bioaccessibility, especially for plant origin foods, 2) there might be a joint fat, carbohydrate and protein effect that influences the bioaccessibilities of PCBs/PCDD/Fs in foods, and 3) use of bioaccessibility estimates would reduce the uncertainty in TEQ calculations

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Table 1 Average bioaccessibility (%) of PCDD/F and PCB in cooked foods obtained from simulated gastrointestinal juice

Food items	Fat (%) in raw	Fat (%) after frying	Carbohydrate (%)	Protein (%)	PCDD/F [a] Boiled (water)	Fried (oil)	PCB [b] Boiled (water)	Fried (oil)
Rice *(n=3)	0.2#	16.8	79.0#	8.1#	4.9±0.33	17.7±0.45	16.5±0.96	55.7±2.50
cabbage *(n=3)	0.2#	17.0	0.3#	1.5#	1.9±0.65	15.2±1.30	4.15±0.89	39.5±3.84
Milk powder *(n=3)	3.8	/	35.6#	15-20#	28.4±1.16	/	72.3±1.62	/
egg *(n=3)	3.1	28.3	0.5#	12.8#	8.6±1.25	15.7±2.03	29.7±3.11	61.2±4.77
beef *(n=3)	4.8	21.5	0.5#	18.6#	7.8±0.88	26.6±2.47	49.0±3.28	77.0±3.46
Fish *(n=3)	3.1	19.8	0.2#	18.6#	7.9±1.29	25.3±3.50	27.0±2.46	77.0±5.76

*: fortified foods (spiked concentration: 2000 pg/g for PCDD/F(4 Cl and 5-7 Cl) and PCB, 4000 pg/g for PCDD/F (8 Cl))，#: data source: China Food Composition (2009), Book 1, 2nd edition, Peking University Medical Press, ISBN: 9787811167276，a: average of 17 toxic PCDD/F congeners，b: average of 12 dioxin-like PCB congeners and 6 indicator PCB congeners

References

1, Zhang L., Li J. and Liu X. (2013) Chemosphere **90**, 1625–1630.

2, Collins CD., Craggs M., Garcia S., Kademoglou K., and Lowe S. (2015) Environ. Int. **78**, 24–31.

3, Rostami I. and Juhasz A.L., (2011) Crit. Rev. Environ. Sci. Technol. **41**, 623–656.

4, Wang H, Zhao Y, Man Y, Wong C. and Wong M, (2011) Food Chem. 127, 1673–1679.

5, Xing GH, Yang Y, Yan Chan, JK., Tao S, and Wong MH., (2008) Environ. Pollut. 156, 1218–1226.

6, Yu Y, Huang N, Zhang X, Li J, Yu Z, Han S, Lu M, Wu M, Sheng G, and Fu J, (2011) Chemosphere 83, 152–160.

7, Oomen A, Rompelberg C, Bruil M, Dobbe C, Pereboom D, and Sips A, (2003) Arch. Environ. Contam. Toxicol. 44, 281–287.