

## Oral Bioaccessibility of PBDEs in Dust Using an In Vitro Gastrointestinal Model

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### Introduction

Recent studies indicated that sources other than the diet, including indoor dust, were important contributors of human exposure to PBDEs (e.g., Wu et al. 2007, Allen et al. 2008; Stapleton et al. 2008, Johnson-Restrepo & Kannan 2009, Zhu et al. 2009, Toms et al. 2009). PBDE exposure through indoor dust is significant, in particular, for children (Toms et al. 2008). Whether observed indoor dust PBDE concentrations actually pose a risk to humans has been controversially discussed (Banasik & Hardy 2009, Sjoedin et al. 2009). Until now, a maximum daily dust ingestion of 100-200 mg/day and complete absorption of PBDEs from dust were assumed in exposure assessment for children. The figures on maximum daily indoor dust ingestion have recently been revised by the USEPA and were estimate to be 30 mg/d for age group 6-12 month and 60 mg/d for age groups 1-<6 years and 6-<21 years (USEPA 2008). There is no data available on what percentage of PBDEs adsorbed to dust is solubilised in the gastrointestinal tract and, thus, available for absorption. However, the understanding of the oral bioavailability of PBDEs in dust is important when trying to establish the extent to which humans may be at risk from exposure to these compounds.

Physiologically based extraction tests have been applied to assess the release of chemicals from sample matrices (soil, dust) and to estimate their bioaccessibility as rapid screening methods. Various gastrointestinal digestion models have been developed to evaluate the oral bioaccessibility of organic compounds, such as PAHs, PCBs, organochlorine pesticides, PCDD/Fs (Review: Dean & Ma 2007), mainly in soil. When using physiologically based extraction tests, it is often assumed implicitly that the contaminants released into the digestive fluid, which can be separated from the residue by centrifugation or filtration, are completely available for absorption.

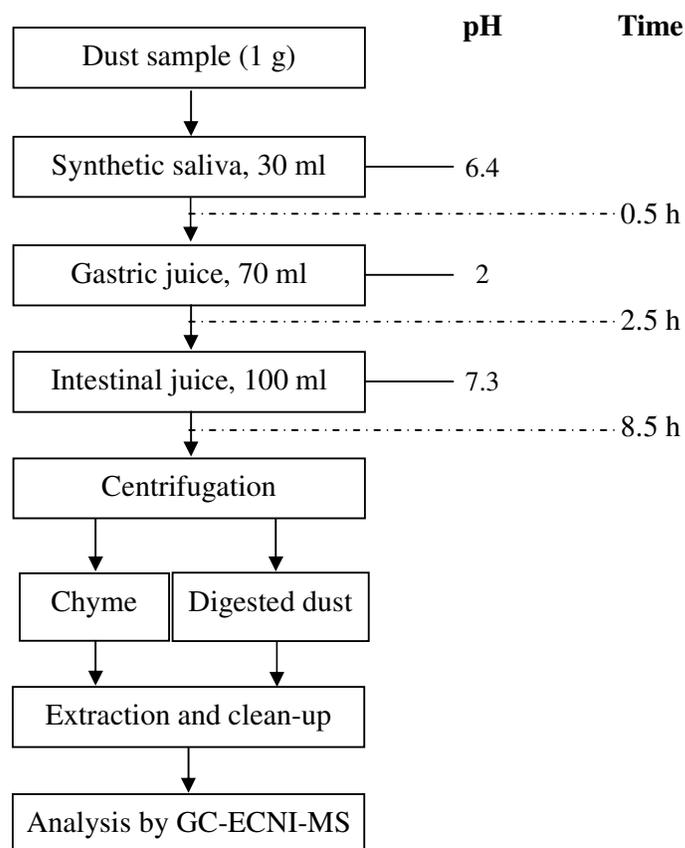
In the present study, an artificial digestion system was used to determine the PBDE fraction in dust available for absorption from the gastrointestinal tract in order to assess the relevance of indoor dust ingestion for human exposure to PBDEs. Oral bioaccessibility of PBDEs is discussed on the basis of results obtained with a house dust standard reference material (NIST SRM 2585) certified for its PBDE content.

### Materials and Methods

For the present study, the indoor dust standard reference material NIST 2585 recently certified for its content of a number of BDE congeners (Stapleton et al. 2006) was chosen as test sample. Certified concentration for BDE47, BDE99, BDE100, BDE153, BDE154, BDE183 and BDE209 were 498, 892, 145, 119, 84, 43, and 2510 ng/g dry weight, respectively.

The in vitro gastrointestinal model described in the German Standard DIN 19738, 2004, was applied. This procedure uses synthetic saliva, synthetic gastric and intestinal juices at physiological pH values

and physiological residence times for fasting conditions (Figure 1). It measures the fraction of a pollutant that would be solubilised in the gastrointestinal tract (i.e., that would be bioaccessible) and therefore available for absorption.



**Figure 1.** Flowchart of the in vitro gastrointestinal model

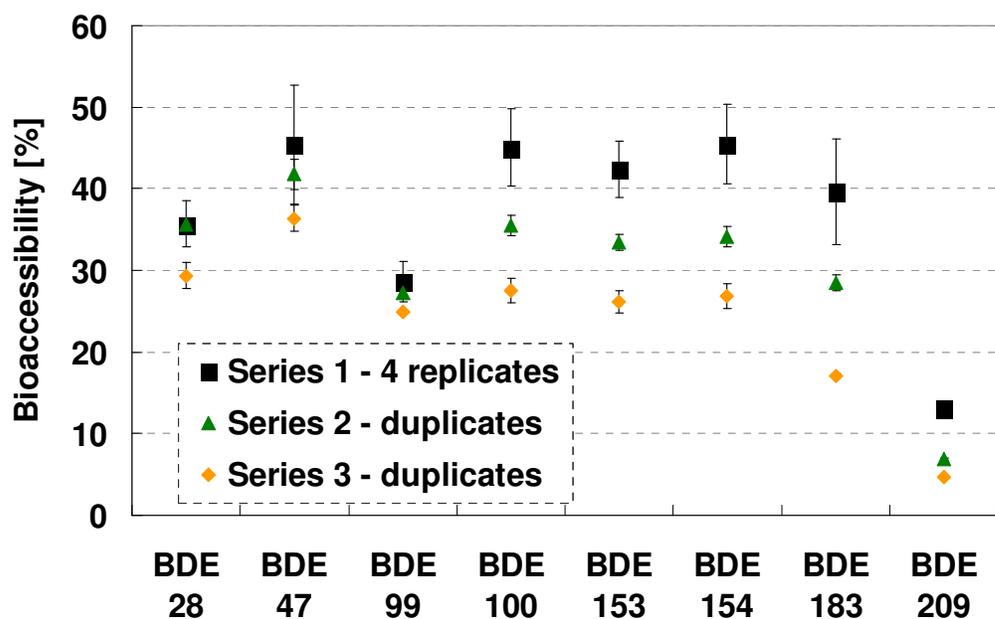
The digestion experiment was performed at 37 °C. It was started by suspending 1 g of dry material in 30 mL of synthetic saliva and stirring for 30 min. Then 70 mL of gastric juice was added. The pH of the gastric solution is adjusted to 2.0 with an automated titrator adding HCl for a period of 2 h. This was followed by a 6 h intestinal phase. 100 mL of synthetic intestinal juice were added and the pH increased to 7.3 by addition of solid NaHCO<sub>3</sub>. At the end of the digestive process, the mixture was centrifuged at 7000 g for 10 min and the supernatant was decanted. The residue was re-suspended in 30 mL of de-ionized water and stirred for 10 min, then centrifuged, and the supernatant was combined with the decanted chyme solution.

The original dust, chyme and dried dust residues were analysed for BDE28, BDE47, BDE49, BDE66, BDE85, BDE99, BDE100, BDE138, BDE153, BDE154, BDE183, BDE196, BDE197, BDE203, BDE206, BDE207 and BDE209 using GC-ECNI-MS. PBDEs were extracted with toluene and the obtained extracts clean-up by GPC and multi-layer silica gel column chromatography (Stiehl et al., 2008). From the determined concentrations, mass balances were calculated for the individual BDE

congeners. Analytical quality control included procedural blanks, recovery tests, regular analysis of a laboratory reference material (Sediment, Quasimeme QBC003MS) as well as participation in international interlaboratory exercises on the determination of PBDEs in biota, sediment and dust.

### Results and Discussion

The in vitro digestion model was adapted where necessary and optimised by analysing the indoor dust standard reference material NIST SRM 2585 under repeatability and within-laboratory reproducibility conditions. On average  $96 \pm 10$  % of the test compounds were recovered from the chyme and digested dust in total. This indicates no significant losses during artificial digestion, sample preparation and analysis. The repeatability standard deviation of PBDE concentrations in chyme ranged from 6 to 16% (n=4) while reproducibility standard deviation calculated from samples series conducted with freshly prepared digestion juices in different weeks indicated a much higher variability (Figure 2). All operations were carried out according to the same protocol and by the same person. The observed variance between the series indicates that the physiologically based extraction test underlies certain variations, the causes of which have not yet fully been understood. Nevertheless, as shown in Figure 2, all three series showed similar patterns in bioaccessibility even though a fairly high variation in the percentages of the individual BDE congeners was observed.



**Figure 2.** Bioaccessibility of PBDEs in NIST SRM 2585 house dust standard reference material

Average bioaccessibility for the tri- to hepta-BDEs ranged from 27 to 42 % while that observed for BDE209 was about 10% (range 7 to 14 %). It seems that BDE209 is significantly less bioaccessible than the other investigated congeners. For tri- to hepta-BDEs, bioaccessibility of individual congeners

did not appear to be correlated with degree of bromination. This observation was in line with results of Ruby et al. 2002 who did not find any correlation between bioaccessibility of PCDD/Fs in soil and the degree of chlorination.

The results of the in vitro digestion experiments imply that less than 50 % of the PBDEs present in the studied house dust might become bioaccessible. The bioaccessible fraction of a compound forms the maximum percentage that is available for crossing biological membranes for absorption but does not provide any information on the fraction, which is actually absorbed via intestine. However, the results demonstrated that a substantial part of the PBDEs ingested via dust cannot be absorbed in the gastrointestinal tract since it is not bioaccessible. Further experiments including real world samples covering a realistic range of concentrations are needed to better understand bioaccessibility and bioavailability of PBDEs in humans.

### **Acknowledgement**

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