A PRELIMINARY COMPARISON OF CANADIAN PBDE EXPOSURES FROM ORAL AND INHALATION ROUTES

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Introduction

Polybrominated diphenyl ethers (PBDEs) have garnered growing interest with regards to their observed biomagnification, and recently, with regards to increasing levels in human tissue and breast milk and their potential for human exposures exceeding "safe" levels of toxicity.

Typically, exposure assessments of PBDEs have focused solely on the dietary exposure pathway[1,2,3,4] due to their tendency to biomagnify and due to lack of available data on other exposure pathways, and few have concurrently quantified exposure via inhalation[5], other oral pathways, or dermal routes[6]. Wenning[7] employed modeling techniques to estimate the relative exposure contributions from diet versus inhalation, but modelled diet concentrations were 2-3 orders of magnitude higher than some measured concentrations, and thus may have underestimated the significance of the inhalation route[5]. Harrad[5] demonstrated that the inhalation route contributed 7% towards combined dietary and inhalation exposure in the U.K. Harrad[5] also noted a wide range of PBDE concentrations in indoor air, suggesting that the significance of the inhalation route may vary, depending on an individual's exposure factors. Furthermore, the potential for dermal exposure has been recommended for future research[5] but has not been quantified, and soil exposure, to which children are particularly vulnerable due to their higher intakes, has not been investigated to our knowledge.

Fate and transport modelling has been used to estimate multimedia fate of PBDEs[8,9,10]. These results can be tied to an exposure assessment to surmount the problem of gaps in measured concentrations in exposure pathways. A challenge with this approach is knowledge of an emission rate, which is seldom available. In this case, the multimedia model can be used to back-calculate emission rates and concentrations in all media for which data are not available. In urban areas, surface films are a convenient passive sampler for atmospheric concentrations. We are using surface films for back-calculation because they respond rapidly to changes in emissions and because of their ease of sampling, can cover a broad geographic range[11,12,13]. Moreover, recent work by Butt *et al.*[11] demonstrated an urban-rural gradient in PBDE concentrations in surface films,

suggesting the potential for an urban-rural gradient in inhalation and soil ingestion exposures.

This study aimed to quantify and explore the relative importance of routes of Canadian exposures to PBDEs, including soil and dietary ingestion and indoor and outdoor inhalation pathways. Research objectives included 1) To quantify and compare the relative contributions of dietary, soil and indoor and outdoor inhalation routes of exposure towards Canadians' total PBDE exposure for different life stages; 2) To determine whether inhalation exposure in the GTA varies along an urban-rural gradient, including exposures from both indoor and outdoor sources; 3) To quantify differences in indoor inhalation exposure between occupational versus ambient exposure scenarios; 4) To qualitatively discuss factors affecting the relative uptake of PBDEs across inhalation versus oral routes of exposure, and the potential contribution of the dermal exposure route.

Materials and Methods

Dietary concentrations were based on a combination of measured and modelled concentrations of PBDEs, assumed to be representative of dietary exposure across Canada. Measured dietary concentrations were from an ongoing and thus partially completed Canadian total diet market basket samples from Whitehorse, Yukon[4], which consist of a range of imported foods typically sampled in total diet surveys. However, the Whitehorse survey excludes any produce or grain products (except frozen french fries), requiring the use of modeling techniques to estimate their concentrations. Atmospheric wet and dry deposition onto produce was modelled using MUM-fate, a multimedia fugacity-based fate and transport model[14]. Root uptake into produce and grain was modelled using biotransfer factors derived based on a screening-level Kow-based correlation equation[15] frequently used to estimate uptake of organic contaminants into produce and grain[16]. Soil concentrations were modelled using MUM-fate.

Dietary intake and inhalation rates were taken from Richardson[17], while soil intake rates came from CCME[18]. Dietary intake and inhalation rates included those for different life stages, including the infant, toddler, child, teen, adult and senior, while soil intake rates include toddler and adult. Exposure was assessed for different life stages.

Indoor and outdoor total air concentrations of PBDEs were modelled using MUM-fate. MUM-fate estimated both indoor and outdoor vapour-phase concentrations by back-calculating from surface film concentrations collected across an urban-rural transect of the GTA from 2001[11], while particle-phase concentrations were derived based on a Koa relationship. Inhalation exposure accounted for both indoor and outdoor exposures at rates of approximately 22.8 hours indoors and 1.2 hours outdoors per day, based on Canadian time-activity patterns[17].

Indoor and outdoor surface film sampling sites included nine sites from an urban-rural transect across the GTA[11], allowing for the determination of the presence or absence of an urban-rural gradient in inhalation and soil exposures. Fate and transport modelling using

observed concentrations in surface films, was used to determine whether or not inhalation and soil ingestion exposures varied across an urban-rural gradient. An urban-rural exposure transect was assessed using an adult receptor.

Differences between occupational and indoor air exposures were modelled using MUM-fate, based on surface film PBDE concentrations collected from inside an electronics recycling facility in Richmond Hill, Ontario[11], along with the range of indoor air exposures estimated along an urban-rural transect.

Factors affecting the relative uptake of PBDEs via inhalation versus oral routes of exposure were explored qualitatively. Relative uptake was discussed in terms of observed human milk and tissue concentrations available in the literature. The potential contribution from the dermal route of exposure was also discussed.

Results and Discussion

Exposure results showed that inhalation exposure was greatest under occupational conditions, followed by downtown and rural scenarios. Dietary contributions to exposure were greater than inhalation; however the relative importance of these routes to internal body burden is not clear. Emission rates for dermal exposure sources have not been quantified, and dermal exposures remain unknown but potentially significant.

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