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Assessing infant exposure to persistent organic pollutants via dietary intake in Australia

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ABSTRACT

Persistent organic pollutants (POPs) including polybrominated diphenyl ethers (PBDEs); organochlorine pesticides (OCPs); and polychlorinated biphenyls (PCBs) persist in the environment, bioaccumulate, and pose a risk of causing adverse human health effects. Typically, exposure assessments undertaken by modeling existing intake data underestimate the concentrations of these chemicals in infants. This study aimed to determine concentrations of POPs in infant foods, assess exposure via dietary intake and compare this to historical exposure. Fruit purees, meat and vegetables, dairy desserts, cereals and jelly foods (n = 33) purchased in 2013 in Brisbane, Australia were analyzed.

For OCPs and PCBs, concentrations ranged up to 95 pg/g fw and for PBDEs up to 32 pg/g fw with most analytes below the limit of detection. Daily intake is dependent on type and quantity of foods consumed. Consumption of a 140 g meal would result in intake ranging from 0 to 4.2 ng/day, 4.4 ng/day and 13.3 ng/day, for OCPs, PBDEs and PCBs, respectively. PBDEs were detected in 3/33 samples, OCPs in 9/33 samples and PCBs in 13/33 samples. Results from this study indicate exposure for infants via dietary (in contrast to dust and breast milk) intake in Australia contribute only a minor component to total exposure.

1. Introduction

Environmental pollutants such as persistent organic pollutants (POPs) are chemicals that persist in the environment, bioaccumulate through the food web, and pose a risk of causing adverse effects to human health and the environment. POPs include brominated flame retardants such as polybrominated diphenyl ethers (PBDEs); organochlorine pesticides (OCPs); and polychlorinated biphenyls (PCBs). These chemicals enter the environment, resulting in exposure and bioaccumulation in biota, including humans, presenting potential health risks (Law et al., 2014).

PBDEs are a group of halogenated organic compounds termed brominated flame retardants (BFRs). Addition of these chemicals to electrical and electronic equipment, building materials, carpet and textiles reduces flammability and therefore harm and destruction caused by unwanted fire. Their use in such materials for this purpose has been widespread since the 1970s. Toxicological studies have found these compounds may interfere with thyroid hormone homeostasis (Kim et al., 2009), reproductive development (Blake et al., 2011) and neurodevelopment (Birnbaum and Staskal, 2004). The human health impacts associated with BFR exposure include diabetes, neurobehavioral and developmental disorders, reproductive health effects and alteration in thyroid function (as summarised in Kim et al., 2014).

Organochlorine pesticides (OCPs) such as dichlorodiphenyltrichloroethane (DDT), and hexachlorobenzene (HCB) are lipophilic chemicals that are known to accumulate in biota including humans (Laug et al., 1951; Egan et al., 1965). They are persistent and subject to long-range transport (Shen et al., 2005; Kang et al., 2012; Mrema et al., 2012). DDT has been used as an insecticide for malaria control and HCB was used as a fungicide for seed treatment (Australian Pesticides and Veterinary Medicines Authority, 2013). Human exposure to these chemicals may result in reproductive effects (Longnecker et al., 2005; Mahalingaiah et al., 2012), cancer (as reviewed in Mrema et al., 2012), reduced childhood growth in boys (Burns et al., 2012) and development of obesity, dyslipidaemia and insulin resistance (Lee et al., 2011). PCBs were compounds used as coolants and lubricants in transformers and capacitors and other

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electrical equipment as they were good insulators. Exposure to PCBs has been linked to health effects including neurobehavioral and immunological changes in children, cancers (including prostate and testicular) and diabetes (Faroon and Ruiz, 2015).

HCB and PCBs are classed as "probably carcinogenic" to humans by the Agency for Toxic Substances and Disease Registry (ATSDR, 2000, 2004). These chemicals became ubiquitous in the environment and humans due to their high usage in the past, physico-chemical properties of persistence, potential volatility and lipophilicity. The PBDEs, OCPs and PCBs investigated in this work are among the chemicals covered under the Stockholm Convention on persistent organic pollutants (POPs) to which Australia is a party. As such there is an obligation as part of Article 16 to contribute to global monitoring of these chemicals (Stockholm Convention on POPs, 2010).

Evidence is increasing that low dose chronic exposure to environmental toxicants, especially POPs, may contribute to the risk of chronic non-communicable diseases (World Health Organization, 2010). Children are thought to be especially vulnerable to POP exposures as their developmental physiology can result in receiving a higher dose of a chemical toxicant for a given level of environmental exposure.

Over the last decade many studies have been carried out to assess the concentration of these chemicals in human serum and breast milk samples in Australia. Such biomonitoring studies have shown that for PBDEs, concentrations are highest in the youngest age groups of the population. In contrast, PCBs and OCPs are found at highest concentrations in older age groups (Toms et al., 2014 submitted). Infants are exposed to all three chemical groups through placental transfer (Toms et al., 2014 submitted) as well as via breast milk (Harden et al., 2007; Toms et al., 2007; Mueller et al., 2008).

The major exposure pathway for POPs in the general population is diet (Fraser et al., 2009). Infants may be more exposed to POPs via their diets because they consume more food and water per unit of body weight and may favor particular foods that could result in higher exposure via those foods such as dairy products (American Academy of Pediatrics, 2003). In babies, additional exposures may occur through breastfeeding (Kannan et al., 1994, 1997) and placental transfer (Siddiqui et al., 1981; Dewan et al., 2013). Infants are likely to experience greater exposure to POPs found in indoors, such as PBDEs, because of crawling and mouthing behavior, which results in greater dust ingestion and therefore higher chemical exposure (Cohen Hubal et al., 2000).

Exposure assessments undertaken by modeling existing intake data underestimate the serum concentrations of these chemicals in children (Toms et al., 2008; Gyalpo et al., 2015). Estimates from applying models suggested there is a level of inconsistency between measured and modeled POP data in infants suggesting that intake in infants is much higher than predicted. This may stem from a lack of data on POPs intake via food. This study aimed to determine concentrations of POPs in baby foods and assess the contribution of this pathway to estimated total exposures. This data was used to determine current intake for the general population and compare to estimated intake one decade ago.

2. Materials and methods

2.1. Sample collection

Baby and toddler foods (n = 33) were purchased in April 2013 from three supermarkets in Brisbane, Queensland, Australia. The different foods represented fruit-, vegetable-, meat/vegetable- and dairy-based foods. This selection was chosen to represent foods which may potentially have high (meat, fish and dairy/egg) and low (fruit and vegetable) concentrations of POPs based on that found in market basket studies (Domingo, 2012; Kim et al., 2013; Mihats et al., 2015; Perello et al., 2015). Both organic (n = 10) and conventional (n = 23) foods were purchased. The weight in each pack ranged from 90 to 220 g and all were indicated to contain one serving per package, inferring that an infant would consume the entire pack in one meal. Samples were purchased to include those from glass jars (n = 8), tins (n = 4), plastic containers (n = 2) and pouches (n = 19), however the majority of foods sold were in pouches at almost 60% of all samples (Table 2). All samples were purchased and stored in their own packaging until transfer to solvent washed glass jars which were frozen at −20 °C prior to analysis. Samples were analyzed at the National Research Centre for Environmental Toxicology, Brisbane, Australia. Most baby foods were made in Australia although some ingredient lists indicated they were produced from local (which means ingredients were produced in Australia) and imported (which means ingredients were produced in countries other than Australia) products. A small number of foods were made in France, New Zealand or United Kingdom. Breast milk intake is calculated using concentrations of POPs in Australian breast milk from previous studies (Harden et al., 2007; Mueller et al., 2008; Chen et al., 2015).

2.2. Chemical analysis

2.2.1. Extraction and clean-up of baby food samples

Baby food samples were left out overnight in sealed glass containers to defrost prior to being weighed out (average 12.7 ± 1.9 g) and placed in a prepared accelerated solvent extractor (ASE) cell. ASE cells were pre-extracted using hexane and acetone (80/20 v/v) and then loaded with a filter paper followed by florisil (5 g), hydromatrix (3 g), filter paper, acid silica (40%) (8 g), filter paper, hydromatrix (2 g) and topped with Na_2SO_4. Samples were placed on top and spiked with internal standards ($^{13}C_{12}$ BDE-77 and $^{13}C_{12}$ PCB-153). ASE conditions for extraction were hexane and DCM (80/20 v/v), 1500 psi, 90 °C, 4 min static time, 60 s purge time and 3 static cycles. Extracts were blown down to dryness and reconstituted into hexane. Prior to analysis samples were spiked with a recovery/instrument standard ($^{13}C_{12}$ BDE-138). Recoveries of $^{13}C_{12}$ BDE-77 and $^{13}C_{12}$ PCB-153 were 66 and 59% respectively. A prepared ASE cell without any sample was used as the method blank (n = 4) with each batch of 10 food samples with relatively low levels of HCB and PCB-153 observed in blanks. PCB-153 was detected in 2 of 4 blanks and HCB was detected in 1 of 4 blanks. The average concentration of HCB and PCB-153 in these blanks was approximately 30% and 44% of the average concentration in food samples. All samples were blank corrected for HCB and PCB-153 by subtracting the average value in blanks from the level in food items. In both cases the average value in food items exceeded the average level in blanks by a factor of 2. The reproducibility of data was assessed through extraction and clean-up of a replicate food sample. The only POP detected in the replicate samples was HCB (% RSD = 28) (see Table S1).

2.2.2. Analysis of baby food samples

All extracts were analyzed using a gas chromatograph (Thermo Scientific Trace 1300) coupled to a high resolution mass spectrometer (Thermo Scientific DFS) operated in electron impact (EI) mode. A 0.18 mm (i.d.) × 30 m fused silica capillary column coated with a 5% phenyl methylpolysiloxane (0.2 μm film thickness) was used for the separation of analytes. The injection port and transfer line temperatures were maintained at 280 °C and the oven temperature program was 100 °C for 1 min, then 10 °C min^{-1} to 280 °C and held for 10 min; total run − time 29 min. The mass spectrometer operating conditions were as follows: ion source 290 °C;