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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 analysed.

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.

Keywords: baby foods, persistent organic pollutants, exposure, polybrominated diphenyl ethers, polychlorinated biphenyls, organochlorine pesticides
**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 Mrma 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 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, physicochemical 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 favour 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; Kannan et al. 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.

**Materials and Methods**

*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 220g 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 analysed 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).

Chemical analysis

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 florisol (5 g), hydromatrix (3 g), filter paper, acid silica (40%) (8 g), filter paper, hydromatrix (2 g) and topped with Na2SO4. Samples were placed on top and spiked with internal standards (13C12 BDE-77 and 13C12 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 (13C12 BDE-138). Recoveries of 13C12 BDE-77 and 13C12 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).

**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; ionisation energy 70 eV; electron multiplier voltage set to 1E6. Resolution was maintained at 5000 (10% valley definition) throughout the sample sequence. Multiple ion detection (MID) experiments were performed with monitoring of the exact masses of appropriate ions for native and selected labeled internal standard. Quantification of PBDEs was performed using \textsuperscript{13}C\textsubscript{12} BDE-77 with PCBs and HCB levels determined using \textsuperscript{13}C\textsubscript{12} PCB-153. Identification was by the GC retention time and ion abundance ratios with reference to internal standards. Criteria used for positive identification and quantification were: (i) retention time (ii) ion ratio ± 20% of the theoretical ion ratio; (iii) signal to noise ratio greater than 5:1.
Baby food samples were analysed for the following chemicals: OCPs - hexachlorobenzene (HCB), o,p'-DDE, p,p'-DDE, o,p-'DDD, p,p,'DDD, o,p'-DDT, p,p'-DDT; PBDEs – BDEs – 17, -28, -47, -49, -66, -71, -77, -85, -99, -100, -119, -126, -138, -153, -154 and -183; PCBs –44, -52, -66, -87, -101, -110, -118, -138, -141, -149, -151, -153, -170, -180, -183. Results are reported as pg/g fresh weight (fw), those with values below the LOD (which ranged from 0.1 to 0.5 pg/g fw (Table 1)) are reported as zero.

Results and discussion

A total of 9 POPs were detected in baby foods tested in this study. PCB concentrations were on average highest followed by PBDEs and OCPs. The individual congeners of PCBs -118, -153, -138, -149, -170 and -180 ranged from below LOD to 95 pg/g fw; BDEs – 47 and -100 were detected ranging from below the LOD to 32pg/g fw; and the only OCP detected, HCB ranged from below <LOD to 30pg/g fw (Table 1). Approximately 58% of baby food samples had some detectable levels of POPs. Food groups where POPs were detected included beef/lamb, poultry, fish, fruit/vegetables and dairy/eggs. The majority of POPs (29/38 analytes) were not detected in these Australian baby food samples. Overall for those detected, concentrations were < 100 pg/g fw. Those not detected were: OCPs -o,p’-DDE, p,p’-DDE, o,p,’DDD, p,p,’DDD, o,p’-DDT, p,p’-DDT; BDEs – 17, -28, -49, -66, -71, -77, -85, -99, -119, -126, -138, -153, -154 and -183; and PCBs – 44, -52, -66, -87, -101, -110, -141, -151, and -183. BDE-209 was not included in the analysis.

Table 1. Concentrations (pg/g fw) of OCPs, PCBs and PBDEs (frequency of detection, mean, median and range) in baby food samples
<table>
<thead>
<tr>
<th></th>
<th>Frequency of detection</th>
<th>Minimum LOR (pg/g fw)</th>
<th>Maximum (pg/g fw)</th>
<th>Mean (pg/g fw)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>OCPs</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>HCB</td>
<td>27%</td>
<td>0.1</td>
<td>29.9</td>
<td>2.0</td>
</tr>
<tr>
<td>BDE-47</td>
<td>3%</td>
<td>0.1</td>
<td>22.8</td>
<td>n/a</td>
</tr>
<tr>
<td>BDE-100</td>
<td>6%</td>
<td>0.1</td>
<td>31.7</td>
<td>1.4</td>
</tr>
<tr>
<td>PCB-118</td>
<td>3%</td>
<td>0.5</td>
<td>9.8</td>
<td>n/a</td>
</tr>
<tr>
<td>PCB-153</td>
<td>36%</td>
<td>0.5</td>
<td>95.4</td>
<td>8.8</td>
</tr>
<tr>
<td>PCB-138</td>
<td>33%</td>
<td>0.5</td>
<td>76.3</td>
<td>8.7</td>
</tr>
<tr>
<td>PCB-149</td>
<td>27%</td>
<td>0.5</td>
<td>53.4</td>
<td>5.2</td>
</tr>
<tr>
<td>PCB-180</td>
<td>3%</td>
<td>0.5</td>
<td>54.5</td>
<td>n/a</td>
</tr>
<tr>
<td>PCB-170</td>
<td>3%</td>
<td>0.5</td>
<td>15.6</td>
<td>n/a</td>
</tr>
</tbody>
</table>

*median not reported due to low detection frequency which results in a zero median value for all analytes; n/a – not applicable due to small frequency of analyte detection; LOR – limit of reporting

It should be noted that these results are interpreted with caution due to the small number of detectable concentrations. The highest concentration of OCPs (HCB 29.9 pg/g fw) was found in a poultry based food, that of PBDEs (BDE-100 at 31.7 pg/g fw) was also in poultry based food while for PCBs (PCB-153 at 95.4 pg/g fw) was in a fruit based food (Table 2).

POPs were detected in samples across all packaging types (glass, tin, plastic container and pouches) and all food groups (fruit-based, vegetable based, meat-vegetable based and dairy-based foods) as well as organic/non-organic products. The highest PCB and PBDE concentrations were detected in foods labeled as “organic”. It should be noted that no “homemade” baby foods, that is, foods made from raw ingredients in a domestic kitchen, were analysed which could have been a way to investigate if commercial packaging and processing (which is not used in the domestic kitchen) affected POP concentrations in foods. The analytes determined in this assessment are not chemicals that would be used purposely in present day in either an organic or non-organic food production environment. Rather these OCP, PCB and PBDE analytes are part of the food chain due to past exposure from industries using these chemicals.
or historical use of pesticides (Mrema et al. 2013; Fromme et al. 2015; Wang et al. 2015). Contamination could occur from biosolids used as fertilizer or from residues in soil or sediment. Xia et al. (2010) investigated long term and heavy applications of biosolids on soil and found an accumulation of PBDEs in the surface layer of biosolids-amended soils. Food specific contamination can be considered also. Poultry and meat, especially free-ranged, may be exposed by eating from contaminated soils (Schoeters and Hoogenboom 2006), while fruit and vegetables may be contaminated via the atmosphere-plant-food chain pathway (Yang et al. 2007) and/or via soil uptake (Muller et al. 1994).
Table 2. Concentrations (pg/g fresh weight) of POPs by individual food sample.

<table>
<thead>
<tr>
<th>Container Type</th>
<th>Container ID</th>
<th>PCB-118</th>
<th>PCB-153</th>
<th>PCB-138</th>
<th>PCB-149</th>
<th>PCB-180</th>
<th>PCB-170</th>
<th>BDE-47</th>
<th>BDE-100</th>
<th>HCB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Beef/lamb</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pasta and Beef Bolognese</td>
<td>1</td>
<td>&lt;0.5</td>
<td>12.9</td>
<td>23.0</td>
<td>15.2</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>22.8</td>
<td>&lt;0.1</td>
<td>7.5</td>
</tr>
<tr>
<td>Spaghetti Bolognese</td>
<td>3</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Ravioli Bolognese</td>
<td>4</td>
<td>&lt;0.5</td>
<td>23.8</td>
<td>33.0</td>
<td>12.8</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>2.8</td>
</tr>
<tr>
<td>Beef and Vegetable</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Beef, apple, beetroot and pumpkin</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Beef with Vegetables and Rice</td>
<td>2</td>
<td>&lt;0.5</td>
<td>7.3</td>
<td>6.2</td>
<td>3.2</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Pasta Bolognese</td>
<td>1</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
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<tr>
<td>Shepherds’ Pie</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Baby's Bolognese with Macaroni</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
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<tr>
<td>Beef and Veggie Casserole</td>
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<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Lamb, Carrot and Sweet Potato</td>
<td>2</td>
<td>&lt;0.5</td>
<td>12.2</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Lamb with Sweet Potato</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>8.4</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Creamy Chicken and Vegetable</td>
<td>2</td>
<td>&lt;0.5</td>
<td>27.3</td>
<td>24.7</td>
<td>25.6</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>31.7</td>
<td>3.2</td>
</tr>
<tr>
<td>Chicken Casserole with Rice</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Chicken and Pumpkin Rice</td>
<td>3</td>
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<td>27.1</td>
<td>17.5</td>
<td>13.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
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<tr>
<td>Mild Butter Chicken</td>
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<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>6.4</td>
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<tr>
<td>Chicken Pasta and Vege Casserole</td>
<td>1</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>0.3</td>
</tr>
<tr>
<td>Creamy Chicken and Vege Puree</td>
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<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>29.9</td>
</tr>
<tr>
<td>Chicken with Sweetcorn and Pasta</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Apricot Chicken</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>10.3</td>
</tr>
<tr>
<td>Chicken with Orange and Honey</td>
<td>2</td>
<td>&lt;0.5</td>
<td>&lt;LOR</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Chicken, Basil and Tomato Pasta</td>
<td>2</td>
<td>&lt;0.5</td>
<td>10.0</td>
<td>10.4</td>
<td>5.0</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>1.6</td>
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<tr>
<td>Golden Sweetcorn and Chicken</td>
<td>1</td>
<td>&lt;0.5</td>
<td>29.2</td>
<td>26.4</td>
<td>21.0</td>
<td>&lt;0.5</td>
<td>&lt;0.5</td>
<td>&lt;0.1</td>
<td>&lt;0.1</td>
<td>&lt;LOR</td>
</tr>
<tr>
<td>Category</td>
<td>Description</td>
<td>Container Type</td>
<td>Minimum</td>
<td>Maximum</td>
<td>Mean</td>
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<td>Fish</td>
<td>Tuna and Rice Vegetable Puree</td>
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<td>4.3</td>
<td>20.5</td>
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<tr>
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<td>&lt;LOR</td>
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<td>Fruit/Veg</td>
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<td>&lt;LOR</td>
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<td>9.8</td>
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<tr>
<td></td>
<td>Apple and Oatmeal</td>
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<td>Rissoni Pasta and Garden Vegetables</td>
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<td>&lt;LOR</td>
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<tr>
<td>Dairy/Egg</td>
<td>Smooth Chocolate Custard</td>
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<td>76.3</td>
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</table>

* container type - 1 glass, 2 pouch, 3 tin, 4 plastic container

*<LOR-less than limit of reporting*
Australian comparisons

The only other Australian study to report PBDE levels in infant foods (n=4) was conducted in 2004 and reported slightly higher levels with BDEs – 17, 28, 49, 66, 85, 99, 153 and 154 ranging from 0.22 – 27 pg/g fw and BDE-47 and -100 ranging from 25-34 pg/g fw and not detected – 3.5 pg/g fw, respectively. These foods were dairy based infant dessert, fruit infant dessert, mixed infant cereal and infant dinner (Food Standards Australia and New Zealand 2007). Due to the small sample size of infant foods from the 2004 study, results from adult foods were investigated. BDE-47 and -100 concentrations ranged from LOD to 230 pg/g fw and LOD to 100 pg/g fw, respectively with the highest concentrations in boiled eggs, pork chops, bacon and cream (Food Standards Australia and New Zealand 2007). Overall, PBDE concentrations in baby foods from 2013 have concentrations lower than found in 2004 although due to the small sample size in the 2004 study, comparisons are made with caution. Concentrations of PCBs have not been measured in Australian baby food, but the most recent report found highest concentrations in fish up to 4 pg/g fw (Food Standards Australia and New Zealand 2004) while HCB was not detected in foods (Food Standards Australia and New Zealand 2003).

Concentrations are related to usage and in Australia, HCB usage stopped in 1985 (Australian Pesticides and Veterinary Medicines Authority 2013), PCBs were banned in 1975 (Australian Government Department of the Environment accessed 15 July 2015) and PBDEs in 2007, although due to the persistence (with PBDE half-lives ranging from 1.6 – 6.5 years (Geyer et al. 2004; Thuresson et al. 2006); and PCB half-
lives ranging from 10 – 15 years (Ritter et al. 2011) and longevity of products containing PBDEs/PCBs, exposure has continued well past these times.

In contrast to the food concentrations, POP concentrations in blood serum from Australian children (0-4 years old) in 2012/13 are highest for OCPs – HCB mean 5.8 ng/g lipid followed by BDE-47 at 4.4 ng/g lipid and lowest in PCBs with the mean ranging from non detected to 3.5 ng/g lipid (Toms et al. 2014 submitted). While pharmacokinetic modeling considering absorption, metabolism and excretion were not considered here, the reason for this disparity may be that food intake is not the dominant exposure route for infants and toddlers and rather placental transfer, breastfeeding and dust ingestion are important (Gyalpo et al. 2015).

*International comparisons*

For OCPs and PCBs, Australian baby food ranged up to 95 pg/g fw and for PBDEs up to 32 pg/g fw with most analytes below the limit of detection. In Ireland, baby food samples (n=41) had no detectable concentrations of the OCPs and PCBs that were analysed in the current study (Food Safety Authority of Ireland 2004). In contrast, baby foods including fruit, meat, fish and cereal based foods from Romania did have detectable concentrations of DDTs (Dobrinas et al. 2011). In Korea, Jeong et al. (2014) report mean PBDE concentrations in homemade baby food (but not commercial baby foods) of 5.9 and 0.54 pg/g fw for BDE-47 and BDE-100, respectively which are lower than found in Australian baby food. BDE-47 and -100 were determined in baby foods from the US and China and were higher than in Australia with mean concentrations of 182 and 91 pg/g fw in the US and 12 and 4.6 pg/g fw in China, respectively (Liu et al. 2014). There is limited data on POPs in
baby food. A review of market basket studies of PBDEs for countries across the world found that PBDE intake via food is dependent on the diet of that region as well as the contribution of the different congeners (Domingo 2012). The differences in POP concentrations in food are likely due to the points in time when these chemicals were banned and subsequently withdrawn from usage in different countries.

**Intake**

Intake is dependent on the quantity and type of purchased baby food an infant or toddler is fed. An exposure intake (in ng/day) is calculated by multiplying the concentration in ng/g fw by the actual fresh weight of the baby meal. Using an example weight of 140 g per purchased baby meal, one meal per day would result in OCP, PBDE and PCB exposure ranging from 0 to 4.2 ng/day, 4.4 ng/day and 13.3 ng/day, respectively.

Another source of dietary intake of POPs for infants is breast milk. The rate for children 0 – 3 years “ever breastfed” in Australia was 92% in 2011/12 (Australian Bureau of Statistics 2007). For this reason intake via this pathway is described here. Previous work in Australia has estimated PBDE exposure intake rates for infants via breast milk to be approximately 160 ng/day. This exposure intake rate (breast milk consumption in grams/day by lipid content (percentage) of breast milk by POP concentration in breast milk (ng/g lipid)) via breast milk is based on a milk concentration of 5.6 ng/g lw (BDE -47 and -100 combined) and milk (3.7% lipid) consumption of 770 g/day (Toms et al. 2009). For HCB and PCB 138 with mean concentrations in breast milk of 4.8 ng/g lipid and 7.0 ng/g lipid, respectively (Chen et al. 2015), intake is 140 ng/day and 200 ng/day, respectively. Infant intake of PBDEs
but not OCPs or PCBs via breast milk, dust and air was assessed and found the dominant pathway to be breast milk, followed by dust and then air (Toms et al. 2009). In addition, Domingo (2012) reports that based on the lowest observable effect level (LOAEL) of 1 mg/kg/day (based on effect of the PBDE group -pentaBDEs) determined by Darnerud et al. (2001), human health risks associated with dietary exposure to PBDEs is not of great concern. Comparison of the dietary exposure intake routes of baby food and breast milk suggests even at the highest POP levels found in these food items, exposure for infants via dietary intake in Australia is relatively minor.

Conclusions

The results of this study suggest limited contamination of baby foods in Australia with the POPs investigated here. The lower concentration of POPs in baby foods collected in 2013 compared to baby and adult foods in 2004 suggest background levels of these chemicals in Australia are also decreasing. This may be reflective of the global production changes, both recent and historical, depending on the POP, as well as manufacturers’ and regulatory bodies’ efforts to limit emissions to the environment. Despite these low levels there is increasing concern about exposure to environmental pollutants in young children and further work should focus on the analysis of frequently used pesticides.

Acknowledgements

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

- OCPs, PCBs, and PBDEs detected in low concentrations in Australian baby foods.
- Daily dietary POPs intake is dependent on type and quantity of foods consumed.
- Infant dietary intake of POPs in Australia a minor component to total exposure.