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Serum polychlorinated biphenyls and their hydroxylated metabolites are associated with demographic and behavioral factors in children and mothers

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ABSTRACT

Factors contributing to the inter-individual variation in body burden of polychlorinated biphenyls (PCBs) and their hydroxylated metabolites (OH-PCBs) have not been fully elucidated. We examined associations between total serum concentrations of 209 PCBs, 64 OH-PCBs, and frequently detected individual congeners with demographic characteristics (age, gender, ethnicity and community of residence), body mass index (BMI or BMI percentile), and breastfeeding history in children and their mothers from 83 U.S. households. There was a significant positive association between age and concentrations of total PCBs and OH-PCBs in mothers. Non-Hispanics had significantly higher concentrations of total PCBs in mothers and OH-PCBs in children than Hispanics. Concentrations of total PCBs were significantly lower in mothers who had longer breastfeeding duration. Living in the Columbus Junction, Iowa community as compared to East Chicago, Indiana was associated with higher total PCBs in children, probably attributable to higher exposures at school. Lower concentrations of OH-PCBs were significantly associated with a higher BMI percentile in children. Congener-specific associations were observed for 30 PCB and 12 OH-PCB congeners and followed comparable trends. To our knowledge, this is the first study to examine factors contributing to variations in serum concentrations of total 209 PCBs and total OH-PCBs in children, as well as to examine ethnic differences in OH-PCB levels. Results from this study revealed that demographic characteristics, body mass index and breastfeeding history are factors that should be considered for human exposure and risk assessment of PCBs and OH-PCBs.

1. Introduction

Polychlorinated biphenyls (PCBs) are a group of synthetic organic compounds consisting of 209 unique congeners with varying numbers and positions of chlorine molecules on the biphenyl backbone. Regardless of origin, PCBs are highly persistent in the environment, bioaccumulate in biota and humans, and induce adverse health effects (U.S. Department of Health and Human Services, 2000). Hydroxylated polychlorinated biphenyls (OH-PCBs) are major PCB metabolites; they appear in the human body not only because of metabolism of PCBs (Grimm et al., 2015), but also because of other origins such as intake of foods containing OH-PCBs, especially contaminated fish and other seafood capable of PCB metabolism (Rylander et al., 2012a), prenatal exposure via placental transfer (Soechitram et al., 2004; Kawashiro et al., 2008; Park et al., 2008; Meijer et al., 2008), and possibly inhalation, as PCBs can react with OH radicals in the troposphere, leading to formation of OH-PCBs in the atmosphere (Totten et al., 2002; Mandalakis et al., 2003).

Prior studies in highly-exposed cohorts have shown that variations in the body burden of PCBs and OH-PCBs from one individual to another may be explained by demographic and behavioral factors. For example, many studies have reported that age is a determinant of human body burden of PCBs (Schaeffer et al., 2006; Axelrad et al., 2009; Bergkvist et al., 2012; Whitehead et al., 2015; Ibarluzea et al., 2011; Schade and Heinzow, 1998; Hardell et al., 2010) and OH-PCBs (Sandau et al., 2000; Nomiyama et al., 2010). Gender and race/ethnicity have also been shown to be associated with serum levels of PCBs (Hardell et al., 2010; Dhooge et al., 2010; Xue et al., 2014; Petrik et al., 2006; Tang-Peronard et al., 2014) or OH-PCBs (Hovander et al., 2006). Community of residence is another factor associated with PCBs and OH-PCBs levels such that residents living in a highly PCB-contaminated location had higher serum concentrations of PCBs and OH-PCBs than residents of a lesser contaminated community (Hovander et al., 2006). Body mass index (BMI) of adults or BMI percentile of children may also predict an individual's body burden of PCBs or OH-PCBs: several

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studies have found a significant association of BMI with PCB (Schade and Heinzow, 1998; Dirinck et al., 2014; Morck et al., 2014) or OH-PCB levels in serum (Weiss et al., 2006). Moreover, breastfeeding history may affect body burden of PCBs or OH-PCBs; women who previously breastfed were found to have a lower body burden of PCBs (Ibarluzea et al., 2011; Schade and Heinzow, 1998; Hardell et al., 2010) and OH-PCBs (Rylander et al., 2012a), whereas children who had been breastfed during infancy were found to have significantly higher concentrations of PCBs in their serum (Gallo et al., 2011; Nawrot et al., 2002).

Importantly, most of the above studies did not examine all of these factors using a multivariate analysis; therefore, their findings could be explained by confounding factors; for example, associations between PCBs and ethnicity may be confounded by ethnic-specific differences in breastfeeding practice. Moreover, studies that examine factors contributing to PCBs and OH-PCBs levels in children have been very limited, despite the emerging evidence supporting the negative health impact of PCB exposure in children (Hisada et al., 2014; Berghuis et al., 2013; Berghuis et al., 2014; Park et al., 2009). In addition, none of the previous studies examined total PCBs concentration as the sum of concentrations of all 209 congeners; instead, focusing on common Aroclor PCB congeners. In our prior work, we demonstrated that as much as 50% of PCBs in human serum were PCB congeners not found in commercial mixtures (Koh et al., 2015). This suggests that total PCBs concentration in human serum could be underestimated by up to 50% if only based on common Aroclor PCB congeners. Furthermore, the role of individual congeners, such as mono-chlorinated PCBs and di-hydroxylated PCBs, has been overlooked in the previous literature.

This study was conducted to fill these research gaps and to investigate associations between total serum concentration of 209 PCBs (Σ_{209} PCBs), 64 OH-PCBs (Σ_{64} OH-PCBs), and frequently detected individual congeners with demographic characteristics (age, gender, ethnicity, community of residence), BMI or BMI percentile, and breastfeeding duration in children and their mothers.

2. Materials and methods

2.1. Study locations, participants and sample collection

The present study used data from the Airborne Exposure to Semivolatile Organic Pollutants (AESOP) Study. Detailed information related to study locations, participants, and sample collection of the AESOP study has been described elsewhere (Koh et al., 2015; Marek et al., 2014; Koh et al., 2016; Ampleman et al., 2015). Briefly, mothers and children were recruited from 86 households in East Chicago, Indiana and the rural area in and around Columbus Junction, Iowa through middle schools. Their serum samples were collected in 2010–2011 and assayed for 209 PCBs and 64 OH-PCBs. We excluded the younger child if there was more than one enrolled child from the same household. Three households were excluded because both PCBs and OH-PCBs data were not available due to poor surrogate standard recovery, giving a final sample size of 83 mother-child pairs. The AESOP study protocols were reviewed and approved by the University of Iowa Institutional Review Board. Written informed consent and assent were obtained from all participants in English or in Spanish.

2.2. Quantification of PCBs and OH-PCBs

Details for quantification and quality control of PCBs and OH-PCBs analysis were presented previously (Koh et al., 2015; Koh et al., 2016). In brief, PCBs were quantified using a modified version of U.S. EPA method 1668 with gas chromatography-tandem mass spectrometry (GC–MS/MS). A total of 174 chromatographic peaks were observed, which represented all 209 PCB congeners as single or co-eluting congeners. Many of the detected PCBs were non-Aroclors (Koh et al., 2015). The OH-PCBs selected for analysis were determined by the availability of commercial calibration standards. A total of 64 OH-PCBs were quantified as single or co-eluting MeO-PCBs in 57 chromatographic peaks (Koh et al., 2016). Among the 83 households included in the present study, PCBs data were available from 82 households and OH-PCBs data were available in 74 households, respectively.

2.3. Quality control

Method blanks (1% potassium chloride, KCl), a mixture of ^{13}C-labelled PCB surrogate standards and standard reference materials were used as quality control samples. Full details of quality control results were presented in our previous studies (Koh et al., 2015; Koh et al., 2016). Limit of quantification (LOQ) was determined based on the mean plus twice the standard deviation of method blanks. Each congener had its own LOQ value and samples below the LOQ value were assigned a value of zero (Hisada et al., 2014; Koh et al., 2015). The list of all 209 PCB congener LOQ values is presented in Supporting Information (SI), Table S1. All 64 OH-PCB congener LOQ values were published previously (Koh et al., 2016).

2.4. Dependent variables

$\sum_{209}$ PCBs and $\sum_{64}$ OH-PCBs were calculated as the sum of concentrations of 209 PCB congeners and 64 OH-PCB congeners (ng/g f.w.), respectively. Individual PCBs and OH-PCBs were selected for statistical analysis in the present study if their detection frequency was ≥65%. Based on this criterion, 30 PCB congeners and co-eluting congeners and 12 OH-PCB congeners and co-eluting congeners were included. Concentration and detection frequency of PCBs are available in SI, Table S2 while those for OH-PCBs were previously published (Koh et al., 2016).

2.5. Independent variables

Age was calculated from the sample collection date and participant's date of birth. Body weight and height were measured by trained field staff and used for calculation of BMI. For children, age- and gender-specific BMI percentiles were calculated by comparison with the 2000 Centers for Disease Control and Prevention Growth Chart (Kuczmarski et al., 2002). Breastfeeding duration in mothers was calculated as the sum of self-reported breastfeeding duration for each child that they had breastfed, whereas breastfeeding duration in children was obtained from breastfeeding duration of the enrolled child reported by his/her mother. Zero month was assigned for a mother if she had never breastfed any child, and for a child if he/she was not breastfed. In addition, ethnicity was included in the list of independent variables and coded as 0 for Hispanic or 1 for non-Hispanic. Community of residence was examined with the Columbus Junction community being coded as 0 and East Chicago as 1. Children's gender was also coded as a binary variable (0 for male and 1 for female).

2.6. Statistical analysis

SAS (version 9.3, SAS Institute, Cary, NC) was used for statistical analysis. The statistical significance level was set as 0.05. Data for mothers and children were analyzed separately. Levels of $\sum_{209}$ PCBs and $\sum_{64}$ OH-PCBs, as well as selected individual congeners were log-transformed to achieve normality. Pearson correlation coefficient was estimated for the relationship between log-transformed $\sum_{209}$ PCBs and $\sum_{64}$ OH-PCBs in 73 children and 73 mothers who had both PCB and OH-PCB data available. In multiple linear regression analyses, log-transformed $\sum_{209}$ PCBs and $\sum_{64}$ OH-PCBs were used as the dependent variables, whereas age, gender (for children), ethnicity, community of residence, BMI (for mothers) or BMI percentile (for children), and breastfeeding duration were independent variables. Log-transformed levels of selected congeners with a detection frequency of 100% were analyzed in the same manner. For congeners that were not detected in all participants, data were analyzed using Tobit regression analysis by