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Brominated and organophosphorus flame retardants in body wipes and house dust, and an estimation of house dust hand-loadings in Dutch toddlers

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ABSTRACT

Children generally have higher Flame Retardant (FR) concentrations in serum compared to other age groups. Toddler behavior enhances direct contact with house dust since their frequent presence proximate to the floor, and their mouthing behavior. This study aimed to thoroughly investigate FR levels in body wipes of toddlers 8–18 months old) and in indoor dust using a noninvasive sampling technique. In this cross-sectional study, body wipes from hands, mouth and back, and indoor household dust samples were collected in twenty-one families and analyzed for one brominated- and seven organophosphorus FRs (polybrominated diphenyl ether 209 (BDE209), tris(2-chloroisopropyl) phosphate (TCIPP), tris(chloroethyl) phosphate (TCEP), tris(1,3-dichloroisopropyl) phosphate (TDCIPP), tris(phenyl) phosphate (TPHP), tris(methylphenyl) phosphate (TMPP), resorcinol bis(diphenyl phosphate) and bisphenol A bis(diphenyl phosphate)). Accelerated solvent extraction was used for extraction and the extract was measured with liquid chromatography combined with mass spectrometry. Non-parametric correlation analyses were performed to assess associations. All FRs were detected in body- and indoor dust samples (median range: 1.0 ng/hand wipe (BDE209) to 65 ng/hand wipe (TCIPP)) and were mostly correlated with each other. We estimated that approximately 260 mg dust (range 50–880 mg) accumulated on toddler's hands per day. Hand-to-mouth frequency was negatively associated with FR levels in wipes ($\tau = -0.38$, $p = 0.04$). With increasing age FR concentrations (BDE209, TCEP, TDCIPP, TPHP and TMPP) on hands decreased significantly ($p = 0.01–0.03$). Girls had significantly less FRs (TCEP, TCIPP, TPHP and TMPP) on the hands ($p = 0.01–0.03$) than boys. This is to the best of the authors' knowledge the first study in Europe that measured brominated- as well as organophosphorus FRs in several types of body wipes from toddlers and that estimated the amount of house dust that accumulates on toddler's hands.

1. Introduction

Flame Retardant chemicals (FR)s are added to a wide range of consumer products in order to prevent spread of fire. They are able to leach from products and end up in our environment, for example in house dust and the food chain (e.g. fish) (Bergh et al., 2011; Wensing et al., 2005). Humans are exposed to FRs through inhalation, ingestion, and absorption of indoor house dust and diet, which may affect their health because FRs have endocrine disrupting abilities resulting in e.g. thyroid dysfunction, and neurodevelopmental and reproductive health effects (Jacobson et al., 2016; Kim et al., 2014; Mercier et al., 2011). There are different groups of FRs, but the most often studied groups are polybrominated diphenyl ethers (PBDEs), relatively old FRs, and organophosphorus flame retardants (OPFRs), relatively new FRs. PBDEs are organohalogen compounds, of which the use of some is restricted, including penta-, octa- and deca-BDE. Deca-BDE (BDE209) was mostly used in the Netherlands until restriction in the European Union in 2008 (European Union, 2008). In the USA BDE209 was voluntarily phased out in all applications in 2013 (Environmental Protection Agency, 2014). OPFRs, organophosphorus compounds, are often used worldwide nowadays due to the restrictions of PBDEs, although regulations restrict the use of a few OPFRs. For example tris(chloroethyl) phosphate (TCEP) is on the list of substances of very high concern REACH Annex XIV (ECHA, 2009).

Several studies reported that young children have higher concentrations of PBDEs in serum compared to other age groups. A study from Australia investigated ΣPBDE serum concentrations in several age groups and found that children aged 2–6 years have the highest mean

Abbreviations: FRs, Flame retardant(s); PBDEs, polybrominated diphenyl ether(s); OPFRs, organophosphorus flame retardant(s); BDE209 or deca-BDE, polybrominated diphenyl ether 209; TCIPP, tris(2-chloroisopropyl) phosphate; TCEP, tris(chloroethyl) phosphate; TDCIPP, tris(1,3-dichloroisopropyl) phosphate; TPHP, tris(phenyl) phosphate; TMPP, tris(methylphenyl) phosphate; LOD, limit of detection; HWs, hand wipe(s); MWs, mouth wipe(s); BWs, back wipe(s); sd, standard deviation; IQR, interquartile range

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concentration (mean 41 ng/g lipid, range 33–49), followed by children aged 0–2 years (mean 31 ng/g lipid, range 24–38) compared to respectively 20 ng/g lipid (range 12–28) and 9.4 ng/g lipid (range 7.9–11) for age groups 13–30 years and ≥ 31 years (Toms et al., 2009). In addition, serum PBDE levels of Swedish mothers and their children (11–15 months) were compared and median concentrations ranged from 0.057 to 1.5 ng/g lipid for children and 0.036–0.95 ng/g lipid for their mothers (Sahlström et al., 2014). The same trend was found in a study on OPFRs in urine samples, the highest concentrations were found in the lowest age groups (Van den Eede et al., 2015).

The finding that toddlers are more exposed to FRs than adults can be attributed to two reasons. First, infants and toddlers have a lower body weight compared to their relative surface area: 0.064 m^2/kg for 0–2 years versus 0.024 m^2/kg for 2–18 years and 0.028 m^2/kg for 18 years and older (Environmental Protection Agency, 2011b; Phillips et al., 1992). Second, toddlers and pre-schoolers ingest more dust compared to adults: on average 30 mg/day for 6 week to one year old children and 60 mg/day for 1–6 year old children versus 30 mg/day for adults, as estimated based on literature on soil and dust ingestion by the US Environmental Protection Agency (US EPA) (Environmental Protection Agency, 2011a). Third, young children's behavior enhances direct contact with house dust since their frequent presence proximate to the floor, and their mouthing behavior in which a child touches objects with the mouth or puts objects into the mouth, including chewing, biting, sucking and licking (Groot et al., 1998).

Mouthing behavior has several reasons, sucking to gain nutrients via breast feeding and feeling comfortable (Groot et al., 1998), to alleviate pain and discomfort during teething, and importantly to explore their environment by looking, touching and mouthing (Tulve et al., 2002). Object-to-mouth behavior is greater indoors compared to outdoors and decreases with age (Xue et al., 2010). This exploring behavior also leads to a frequent hand-to-object and hand-to-mouth contact. When hands frequently touch a variety of objects, dust may adhere to the skin. Subsequently, when hands are often mouthed, dust ingestion is likely to increase. Especially mouthing objects or ingesting food that is fallen on the floor, have the potential of enhancing the amount of dust ingestion. FRs in dust on hands might also be absorbed by the skin, and if children do not mouth their hands, but still frequently touch surrounding furniture and objects, we expect that exposure is still increased in these toddlers.

Another exposure pathway considers clothes in contact with the skin. Dust adheres to fabrics which leads to exposure by wearing dust containing clothes. Research on dermal absorption of FRs is limited (Abdallah et al., 2015). Although a low absorption rate of penta-BDE has been demonstrated in rats, constant exposure to FRs may still affect human health (Roper et al., 2006). A recent study on dermal absorption of OPFRs estimated absorption rates in human ex vivo skin models and found higher rates for 2–3 year old toddlers compared to adults and a positive correlation between PFR fraction absorption and the water solubility of the PFR (Abou-Elwafa Abdallah et al., 2016). Dermal absorption of for example TCEP was estimated with a minimum and maximum indoor FR concentration at 0.1–38.6 ng/kg bw day in toddlers versus < 0.1–4.3 ng/kg bw day in adults (Abou-Elwafa Abdallah et al., 2016).

Toddler FR exposure is investigated in two ways. Several studies assess exposure based on FR concentration indoors and estimate minimum and maximum ingestion of dust according to the EPA recommendations (Bradman et al., 2014; Brommer and Harrad, 2015; Jones-Otazo et al., 2005; Toms et al., 2015). Others measure exposure in human samples such as serum, urine or hand wipes. Although it is an invasive sampling method, various studies collect serum at child age. For example, the CHAMACOS study measured FRs in serum of 7-year old children (Erkin-Cakmak et al., 2015), the North Carolina Toddler Cohort study measured FRs in serum of 11–36 months old toddlers (Stapleton et al., 2012), and Sahlström et al. (2014) collected serum of 11–15 months old toddlers (Sahlström et al., 2014). Two less invasive methods use urine and hair and were suggested to be used as measurement for child exposure (Kucharska et al., 2015). However, in order to investigate FR levels in or on toddlers considering toddlers' specific behavior such as mouthing and touching objects and being proximate to the floor, hand wipes seem more appropriate. Stapleton et al. (2012) used hand wipes in respectively 11–36 months and 26–68 months old toddlers (Stapleton et al., 2012, 2014). Other important body parts to assess indoor FR exposure include the mouth, and skin underneath clothes and to our best knowledge, no study has investigated other body parts of toddlers than the hands.

Research on FR levels in or on toddlers in light of their specific behavior is limited. Therefore, this study aims to thoroughly investigate Brominated Flame Retardants (BFRs) and OPFRs levels on toddlers and in indoor dust by means of a noninvasive sampling technique using (body) surface wipes and observation of the toddler's behavior, divided into three aims. First we aim to report FR levels in body wipe samples and house dust samples and to assess associations between FR levels in body wipes and indoor house dust. Second, we aim to estimate the amount of house dust and FRs that accumulate in body wipes in a 30 min unstructured playtime. And third, we aim to assess associations between FR levels in body wipes and behavior and demographic factors age, gender and socio-economic status (SES).

2. Materials and methods

2.1. Study design and participants

A cross-sectional study was conducted among participants of the LINC cohort (LInking EDCs in maternal Nutrition to Child health). The LINC cohort comprises of 300 mother-child pairs, living in both rural and urban areas of Zwolle, Purmerend and Den Helder, the Netherlands (de Cock et al., 2016). Recruitment and home visits took place between August and October 2013. Thirty caregivers with a toddler of eligible age 8–18 months old) at the moment of recruitment were invited to participate by postal mail and a follow-up telephone call. The caregiver (s) and toddler needed to be available for 60–90 min on working days. Twenty-one families participated in this study and reasons for not participating were: not enough time for a home visit (N = 8) and illness at the day of the home visit (N = 1). The study was approved by the medical ethics committee of the VU University medical center.

2.2. Sample collection

Samples were collected during the home visit. Body wipes were caregiver-collected under instruction and supervision of one of the authors (EJS). House dust was collected by of one of the authors (EJS). The researcher and parent were wearing non-latex gloves during sample collection. Collection methods for both body wipes and house dust are described in detail below.

2.2.1. Body samples

Three type of body wipes were collected: a hand wipe (HW), a mouth wipe (MW) and a back wipe (BW). One body wipe was collected at the start of the home visit, in order to gain information on FR levels on the toddlers' hands, mouth and back skin. One body wipe, the so-called accumulation wipe, was collected directly after the 30 min observation to assess accumulation of dust and FRs. Methods for collection of wipes were based on Stapleton et al. (2008). Non-sterile gauze pads (5 cm × 5 cm) were used, wetted with a few drops of 3.0% isopropanol. First, the palm, back, between the fingers and wrist of both hands were wiped. Second, the mouth and surrounding skin, from ear to ear, chin and under the nose was wiped. Third, the back surface of the toddler was wiped between both shoulders and from the cervical vertebrae to the waist belt (diaper). All wipes were placed separately in a sealable sample bag, temporarily stored in a portable cooling device and within 12 h stored under frozen conditions (−20 °C) at the Vrije Universiteit