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Concentrations of organophosphate flame retardants and plasticizers in urine from young children in Queensland, Australia and associations with environmental and behavioural factors

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ABSTRACT

In recent years, the production and usage volumes of organophosphate flame retardants (OPFRs) has increased substantially. Certain OPFRs are suspected reproductive toxins, carcinogenic, and neurotoxic. Insufficient information is available on human exposure pathways to these chemicals, particularly in Australia. We aim to assess the association between OPFR concentrations in the urine of children to environmental and behavioural risk factors. Concentrations of eight OPFRs and eleven metabolites were measured in the urine of 51 children, aged 3–29 months, in Southeast Queensland, Australia and compared to their behavioural and environmental risk factor data obtained by an online questionnaire. Of the 11 OPFR metabolites analysed, 55% were frequently detected in the majority (> 80%) of samples. The most frequently detected metabolite was bis(1,3-dichloroisopropyl) phosphate (BDCIPP) (detected in 100% of samples), followed by 1-hydroxy-2-propyl bis(1-chloro-2-propyl) phosphate (BCIPHIPP) (96%), diphenyl phosphate (DPHP) (94%) and bis(1-chloroisopropyl) phosphate (BCIPP) (86%). In multivariable modelling, age was positively associated with concentrations of bis (2-butoxyethyl) phosphate (BBOEP) and negatively associated with concentrations of BCIPP and BCIPHIPP. Other non-age related factors, including vacuuming frequency, hand-washing frequency and presence and number of some electrical appliances in the home were also associated with concentrations of OPFR metabolites.

1. Introduction

The phase-out of polybrominated diphenyl ethers (PBDE), because of environmental and human health concerns, contributed to the development and use of alternative flame retardants including organophosphate-based flame retardants (OPFRs) (Butt et al., 2014; van der Veen and de Boer, 2012). In 2006, the global use of OPFRs was 465,000 t (van der Veen and de Boer, 2012), and is projected to increase to 2.8 million tons in 2018 (Israel Chemicals Ltd, 2015). OPFRs are used in a wide array of consumer products and have numerous applications in transport and the built environment as flame retardants, plasticizer and hydraulic fluids (van der Veen and de Boer, 2012). OPFRs are semi-volatile organic compounds that can contaminate the indoor and outdoor environments via repeated cycles of adsorption (to dust and other media) and volatilisation (Abdallah and Covaci, 2014; Shoeib et al., 2014). There is limited human epidemiological data regarding OPFR exposure and health outcomes (Canbaz et al., 2016; Meeker et al., 2013a; Meeker and Stapleton, 2010; Preston et al., 2017a, 2017b). However, from cell and animal studies some OPFRs have been shown to affect immune, reproductive, endocrine and neurological systems, as well as acting as carcinogens (van der Veen and de Boer, 2012).

Human exposure to these compounds generally occurs via ingestion of contaminated dust, inhalation of volatilised OPFRs and ingestion of food contaminated with OPFRs (Cequier et al., 2015; Hoffman et al., 2015b; Xu et al., 2016). Patterns of exposure are expected to vary amongst specific OPFRs, due to their unique physicochemical properties, as well as their different patterns of application (Table 1). For

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Table 1
Physicochemical properties and applications of OPFRs included in this study.

#	Name	Abbr.	CAS	Vapour pressure[a]	Henry's constant[b]	log K_{OA}	Application	Urinary metabolites measured in this study
1	Tris(2-chloroethyl) phosphate	TCEP	115-96-8	6.67	8.07×10^{-3}	8.42	Flame retardant, plasticizer, lacquer, paint, glue, industrial processes	Bis(2-chloroethyl) phosphate (BCEP)
2	Tris(2-chloroisopropyl) phosphate	TCIPP	13674-84-5	3.30	4.25×10^{-4}	8.84	Flame retardant, plasticizer	Bis(1-chloroisopropyl) phosphate (BCIPP); 1-hydroxy-2-propyl bid(1-chloro-2-propyl) phosphate (BCIPHIPP)
3	Tris(1,3-dichloro-2-propyl) phosphate	TDCPP	13674-87-8	5.60×10^{-6}	1.33×10^{-4}	10.3	Flame retardant, plasticizer, lacquer, paint, glue	Bis(1,3-dichloroisopropyl) phosphate (BDCIPP)
4	Tributyl phosphate	TBP	126-73-8	0.90	0.32	7.87	Plasticizer, hydraulic fluids, floor finish, wax, lacquer, paint, glue, anti-foam agent	Dibutyl phosphate (DBP)
5	Trimethylphenyl phosphate	TMPP	1330-78-5	6.60×10^{-5}	5.42×10^{-3}	11.7	Plasticizer, polyvinylchloride, hydraulic fluids, cellulose, cutting oils, transmission fluids	bismethylphenyl phosphate (BMPP)
6	Triphenyl phosphate	TPHP	115-86-6	4.10×10^{-3}	0.335	10.8	Flame retardant, plasticizer, hydraulic fluids, lacquer, paint, glue	Diphenyl phosphate (DPHP)[c]
7	Tris(2-butoxyethyl) phosphate	TBOEP	78-51-3	2.80×10^{-5}	1.22×10^{-6}	11.7	Flame retardant, plasticizer, floor finish, wax, lacquer, paint, glue, anti-foam agent	Bis(2-butoxyethyl) phosphate (BBOEP); Bis(2-butoxyethyl) hydroxyethyl phosphate (BBOEHEP); Bis(2-butoxyethyl) hydroxyl-2- butoxyethyl phosphate (3OH-TBOEP)
8	Tris(2-ethylhexyl) phosphate	TEHP	78-42-2	2.94×10^{-3}	7.96×10^{-3}	11.7	Flame retardant, plasticizer, fungus resistance	Bis(2-ethylhexyl) phosphate (BEHP)
9	2-Ethylhexyl diphenyl phosphate	EHDPP	1241-94-7	8.67×10^{-5}	2.50×10^{-2}	11.7	Plasticizer, hydraulic fluids	Diphenyl phosphate (DPHP)[c]

[a] Pa at 25 °C.
[b] Pa m^3/mol at 25 °C.
[c] DPHP is a metabolite of both TPHP and EHDPP and potentially other aryl-OPFRs.