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Organophosphate flame retardants and plasticizers in indoor dust, air and window wipes in newly built low-energy preschools

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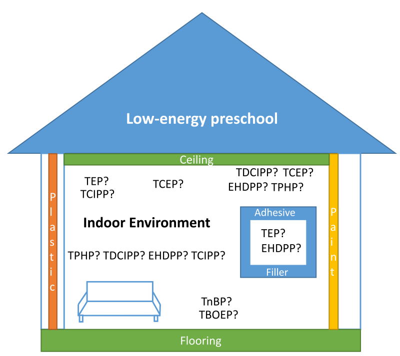
HIGHLIGHTS

- A unique study design to investigate the temporal trends and distribution of OPFRs in preschools.
- OPFRs partition mostly to the indoor dust and surfaces in the preschools.
- Levels of OPFRs were found to be lower in the environmental certified low-energy preschools.
- Changes in the relative composition of OPFRs by time were observed which could be due to introduction of new sources.

GRAPHICAL ABSTRACT



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ABSTRACT

The construction of extremely airtight and energy efficient low-energy buildings is achieved by using functional building materials, such as age-resistant plastics, insulation, adhesives, and sealants. Additives such as organophosphate flame retardants (OPFRs) can be added to some of these building materials as flame retardants and plasticizers. Some OPFRs are considered persistent, bioaccumulative and toxic. Therefore, in this pilot study, the occurrence and distribution of nine OPFRs were determined for dust, air, and window wipe samples collected in newly built low-energy preschools with and without environmental certifications. Tris(1,3-dichloroisopropyl) phosphate (TDCIPP) and triphenyl phosphate (TPHP) were detected in all indoor dust samples at concentrations ranging from 0.014 to 10 μg/g and 0.0069 to 79 μg/g, respectively. Only six OPFRs (predominantly chlorinated OPFRs) were detected in the indoor air. All nine OPFRs were found on the window surfaces and the highest concentrations, which occurred in the reference preschool, were measured for 2-ethylhexyl diphenyl phosphate (EHDPP) (maximum concentration: 1500 ng/m^2). Interestingly, the OPFR levels in the environmental certified low-energy preschools were lower than those in the reference preschool and the non-certified low-energy preschool, probably attributed to the usage of environmental friendly and low-emitting building materials, interior decorations, and consumer products.

1. Introduction

For compliance with the new European Union (EU, 2010) regulations aimed at reducing the energy consumption of buildings, an increasing number of buildings have been constructed based on the

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concept of low-energy housing (SCNH, 2012). The airtight construction proposed by this concept is achieved with efficient insulation, aging-resistant plastic, and functional sealing (Arvela et al., 2014; Langer et al., 2015; SCNH, 2012). However, some of the building materials may contain hazardous compounds and, hence, have a negative effect on the indoor environment and the occupants of the building (Kemmlein et al., 2003; Marklund et al., 2003; van der Veen and de Boer, 2012). These compounds are usually added to the building materials to obtain specific properties and can be subsequently released into the indoor environments via volatilization and abrasion processes (Cao et al., 2014; Liagkouridis et al., 2015; Marklund et al., 2003; Rauert and Harrad, 2015). For example, flame retardants (FRs) are added to building materials and consumer products to prevent the spread of fire (Bergman et al., 2012; Cao et al., 2014; Dishaw et al., 2014; Kemmlein et al., 2003; Stapleton et al., 2014). The use of organophosphate flame retardants (OPFRs) has increased due to increasingly strict regulations and bans on certain brominated FRs (for example, polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD)) (Bergman et al., 2012; Cequier et al., 2014; Dodson et al., 2014; Hartmann et al., 2004; van der Veen and de Boer, 2012). Compared with other FRs, OPFRs release less toxic gases during a fire, owing to the formation of a char layer that shields the material from oxygen (van der Veen and de Boer, 2012). OPFRs are mainly classified into two different groups: non-halogenated OPFRs which e.g. include triphenyl phosphate (TPHP), and halogenated OPFRs such as tris (1, 3-dichloroisopropyl) phosphate (TDCIPP) and tris (2-chloroethyl) phosphate (TCEP) (Brommer and Harrad, 2015; van der Veen and de Boer, 2012). Furthermore, OPFRs are used in various applications and, therefore, the OPFR levels in different microenvironments have increased in recent years. They are also frequently used as plasticizers, stabilizers, anti-foaming compounds, wetting agents, and as additives in lubricants and hydraulic fluids as well as FRs (Hartmann et al., 2004; Marklund et al., 2003; van der Veen and de Boer, 2012). However, some OPFRs are considered persistent, bioaccumulative, and toxic and their usage and subsequent release have therefore been a major concern (van der Veen and de Boer, 2012).

High concentrations ($mg \cdot g^{-1}$) of OPFRs have been found in indoor dust (Cequier et al., 2014; Cao et al., 2014; Langer et al., 2016; Rauert and Harrad, 2015; van den Eede et al., 2012; van den Eede et al., 2011; van der Veen and de Boer, 2012). OPFR concentrations ($ng \cdot m^{-3}$) have also been determined for indoor air (Bergh et al., 2011; Fromme et al., 2014; van der Veen and de Boer, 2012). In general, most OPFRs have relatively low vapor pressure and are hydrophobic (Bergman et al., 2012; Cao et al., 2014; Marklund et al., 2003; van der Veen and de Boer, 2012). Therefore, many OPFRs can partition to indoor dust which can be a significant source of human exposure for these compounds (Dishaw et al., 2014; Liagkouridis et al., 2015; Rauert and Harrad, 2015; van den Eede et al., 2012; van der Veen and de Boer, 2012). TDCIPP and tris(2-chloroisopropyl) phosphate (TCIPP) have been found on hand wipes collected from children and in the indoor dust collected from the indoor environments occupied by the children. However, a significant correlation between these matrices was lacking (Stapleton et al., 2014) and, hence, further studies are warranted to determine the fate of OPFRs in indoor settings and in humans. During their developmental stages, children are especially vulnerable to exposure to endocrine-disrupting chemicals (WHO, 2012) and studies have shown that some OPFRs can have adverse effects on humans. These effects include inhibition of cell growth in liver cells and interruption of neural cell replication and neurodifferentiation (Dishaw et al., 2011; Killilea et al., 2017; Liu et al., 2012; Meeker and Stapleton, 2010; WHO, 1998).

The chemical composition of indoor environments comprising low-energy buildings has rarely been investigated (Derbez et al., 2014; Langer et al., 2015) and the study of semivolatile organic contaminants in these buildings is even more seldom. In this pilot study, nine different OPFRs were analyzed in dust, air, and window wipe samples collected in three newly built low-energy preschools and one reference preschool at different points in time. The objectives of the study were to determine the (i) distribution of OPFRs among the different matrices in the preschools, (ii) influence of environmental certification and the introduction of furnishings and interior decorations on the OPFR levels, and (iii) temporal trends. The results from this study provide new knowledge on the occurrence and distribution of OPFRs in low-energy preschools. This knowledge can be further used to improve the building technique, assess the performance of environmental certifications, and reduce indoor chemical exposure in low-energy buildings.

2. Materials and methods

2.1. Selection of objects and sampling strategy

Three low-energy preschools (LEP A, B, C) and one conventional preschool (reference preschool, RP) were included in this study (see Table S1 for details of the preschools). The RP was built in accordance with the recommendations provided by the Swedish building code BBR 21 (Boverket, 2014). The LEPs were built in accordance with this code and the criteria stipulated by the Swedish Centre for Zero Energy Houses (FEBY 12) (SCNH, 2012). The RP was built with conventional building material available on the market, whereas the three LEP were mainly built with low-emitting building materials (low emission of volatile organic compounds) (see Table S2). Furthermore, two of the LEP (LEP B, C) were respectively designated as environmentally friendly according to the Swan Ecolabel (2016) and Environmental Building Silver Certification (Environmental Building, 2014). The Swan Ecolabel certification stipulates that a low-energy consumption, corresponding to 85% of the recommended value in BBR (Boverket, 2016), of the building must be maintained. Another requirement is that building materials, interior decorations and consumer products must be low-emitting and environmentally friendly, and certain chemical additives cannot exceed 0.01% of the finished product. These chemical additives include the substances in the Candidate List provided by the European Chemical Agency (ECHA). This list includes substances that are classified as (i) persistent, bioaccumulative, and toxic (PBT), (ii) very persistent and very bioaccumulative (vPvB), (iii) carcinogenic, mutagenic, and reprotoxic (CMR), and (iv) endocrine disrupting compounds (EDCs). As a final requirement, the entire construction process should have low impact on the environment, i.e., low levels of CO_2 should be released and low amounts of energy should be consumed during construction (Swan Ecolabel, 2016). Similar to the Swan Ecolabel, the Environmental Silver certification is based on the fulfillment of certain criteria. These criteria set limits on the energy consumption (75% of the value listed in BBR (Boverket, 2016)) and the chemical content of the building materials, interior decorations and consumer products (Environmental Building, 2014). The products should contain (i) <0.1% of carcinogenic and mutagenic compounds, mercury (Hg), lead (Pb), PBT and vPvB compounds, (ii) <0.5% of reprotoxic compounds, and (iii) <0.01% of cadmium (Cd).

For all preschools in our study, the air exchange rate (AER) of the mechanical heat recovery ventilation system was set at maximum speed during the first year of occupancy in order to reduce the emissions from the building materials and eliminate moisture from the building. However, during sampling period 2 (SP2) in the RP, the ventilation system shut down unintentionally due to maintenance of the roof. Moreover, the ventilation system also malfunctioned during sampling periods 3 and 4 (SP3 and 4) in LEP C. This malfunction was due to a wrong electrical connection and, in turn, the ventilation system was shut down for 1–2 h every afternoon during these periods. A full description of the preschools is provided in the supplementary information section.

Sampling was conducted in four sampling periods (SP1, SP2, SP3, and SP4) during the first year of occupancy in all preschools. The sampling was performed every season and the first sampling period (SP1) began directly after the building was completed, but before preschool