Environment International 96 (2016) 65–74

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Measurement and health risk assessment of $PM_{2.5}$, flame retardants, carbonyls and black carbon in indoor and outdoor air in kindergartens in Hong Kong

Wen-Jing Deng [a,*], Hai-Long Zheng [b], Anita K.Y. Tsui [c], Xun-Wen Chen [d]

[a] Department of Science and Environmental Studies, The Education University of Hong Kong, Tai Po, N.T., Hong Kong, China
[b] Department of Geography and Resource Management, The Chinese University of Hong Kong, Shatin, N.T., Hong Kong, China
[c] Department of Early Childhood Education, The Hong Kong Institute of Education, Tai Po, N.T., Hong Kong, China
[d] Department of Civil and Environmental Engineering, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, China

ARTICLE INFO

Article history:
Received 16 May 2016
Received in revised form 19 July 2016
Accepted 19 August 2016
Available online xxxx

Keywords:
Indoor air pollution
PBDEs
Dechlorane plus
Particulate matter
Children health

ABSTRACT

Indoor air pollution is closely related to children's health. Polybrominated diphenyl ethers (PBDEs) and dechlorane plus (DP) transmitted through indoor $PM_{2.5}$ and dust, along with carbonyl compounds and black carbon (BC) aerosol were analysed in five Hong Kong kindergartens. The results showed that 60% of the median $PM_{2.5}$ levels (1.3×10^1 to 2.9×10^1 μg/m^3 for indoor; 9.5 to 8.8×10^1 μg/m^3 for outdoor) in the five kindergartens were higher than the guidelines set by the World Health Organization (2.5×10^1 μg/m^3). Indoor $PM_{2.5}$ mass concentrations were correlated with outdoor $PM_{2.5}$ in four of the kindergartens. The PBDEs (0.10–0.64 ng/m^3 in $PM_{2.5}$; 0.30–2.0×10^2 ng/g in dust) and DP (0.05–0.10 ng/m^3 in $PM_{2.5}$; 1.3–8.7 ng/g in dust) were detected in 100% of the $PM_{2.5}$ and dust samples. Fire retardant levels in the air were not correlated with the levels of dust in this study. The median BC concentrations varied by >7-fold from 8.8×10^2 ng/m^{-3} to 6.7×10^3 ng/m^{-3} and cooking events might have caused BC concentrations to rise both indoors and outdoors. The total concentrations of 16 carbonyls ranged from 4.7×10^1 μg/m^3 to 9.3×10^1 μg/m^3 indoors and from 1.9×10^1 μg/m^3 to 4.3×10^1 μg/m^3 outdoors, whilst formaldehyde was the most abundant air carbonyl. Indoor carbonyl concentrations were correlated with outdoor carbonyls in three kindergartens. The health risk assessment showed that hazard indexes (HIs) HIs of non-cancer risks from PBDEs and DPs were all lower than 0.08, whilst non-cancer HIs of carbonyl compounds ranged from 0.77 to 1.85 indoors and from 0.50 to 0.97 outdoors. The human intake of PBDEs and DP through inhalation of $PM_{2.5}$ accounted for 78% to 92% of the total intake. The cancer hazard quotients (HQs) of formaldehyde ranged from 4.5E−05 to 2.1E−04 indoors and from 1.9E−05 to 6.2E−05 outdoors. In general, the indoor air pollution in the five Hong Kong kindergartens might present adverse effects to children, although different schools showed distinct pollution levels, so indoor air quality might be improved through artificial measures. The data will be useful to developing a feasible management protocol for indoor environments.

1. Introduction

Indoor air pollution (IAP) is closely related to human health because most urban citizens spend a large amount of their time in indoor environments (Chithra and Shiva Nagendra, 2012; WHO, 2006). Studies have increasingly found that exposure to IAP may lead to asthma, cardiopulmonary pathologies, neurological impairments, dysfunctional immune systems, reproductive disorders, and even childhood leukaemia (Sohn et al., 2012; Yang et al., 2009). The World Health Organization (WHO) has revealed that IAP is responsible for 2.7% of the global burden of disease (WHO, 2010).

The variety of pollutants detected in indoor air, such as halogenated compounds and volatile organic compounds (VOCs) (Harrad et al., 2006; Hwang et al., 2008; Takigami et al., 2009), may be 2–5 times, and occasionally >100 times, higher than the levels of pollutants found outdoors (Kohler et al., 2005). Polybrominated diphenyl ethers (PBDEs), which belong to halogenated compounds, can be emitted from the polyurethane foam in furniture, mattresses, carpet pads, and electronic equipment (ATSDR, 2004). They are believed to interfere with thyroid hormone balance and contributes to neurodevelopmental deficiencies (Dingemans et al., 2011; Eskenazi et al., 2013; Gascon et al., 2011; Lee et al., 2010). DP has a high production volume but was only recently found in the environment. Hoh et al. (2006) first reported on the environmental occurrence of DP within the North American Great Lakes Basin, where it is manufactured by OxyChem in Niagara Falls, New

* Corresponding author.
E-mail address: wdeng@eduhk.hk (W.-J. Deng).

http://dx.doi.org/10.1016/j.envint.2016.08.013
0160-4120/

York. Since then, many research studies have been carried out in different countries involving different environmental matrixes including soil (Wang et al., 2010), air (Venier and Hites, 2008) and indoor dust (Zhu et al., 2007; Ren et al., 2010), which had led to the prospect that DP is a worldwide contaminant. In addition, DP has the typical characteristics of a persistent organic pollutant: high lipophilicity, resistance to photo- and biodegradation, and accumulation in fish. DP was found to be released from electronic products in the indoor environment (Ren et al., 2009). DP is persistent, bioaccumulative, and subject to long-range transport (Qiu and Hites, 2008; Möller et al., 2010). However, more research is required to better quantify the emissions, exposures, and toxicological effects of DP and its analogs in the environment. In particular, there is a need to obtain more monitoring, and toxicity information, particularly to kindergarten children. VOCs, which are emitted from furniture, decorations and cleaning products (Dodson et al., 2007; Massolo et al., 2010; Wu et al., 2011), are related to allergic effects, lung cancer and childhood leukaemia (Loh et al., 2007; Sax et al., 2006). Among the various air pollutants, the undesirable health effects of particulate matter (PM) have raised considerable concern over indoor air quality (Lin et al., 2002; Pope and Dockery, 2006) because the presence of indoor $PM_{2.5}$ (particles with an aerodynamic diameter <2.5 μm) has been strongly associated with cardiopulmonary disease and lung cancer (Gemenetzis et al., 2006).

IAP in schools may be even more serious than in other areas, because children spend up to eight or more hours per day in school (Branco et al., 2014b). Further, children are more sensitive to the effects of air pollution than adults, mainly due to their underdeveloped respiratory, immune and endocrine systems, comparatively higher amount of air inhalation relative to their body size (Salvi, 2007; Schwartz, 2004; Sousa et al., 2012). Thus, they are more likely to suffer from the risk of exposure to IAP.

Paying attention to the effect of IAP on the health of school children is one of the key focuses of the Healthy Environments for Children Alliance (WHO, 2003). In addition, the significance of IAP in schools has been highlighted by many studies worldwide (Godwin and Batterman, 2007; Hodgson et al., 2004; Yang et al., 2009; Zhang et al., 2006). For example, PBDE compounds have been detected in 100% of the dust samples in California's early childhood education environments (Bradman et al., 2014), where a correlation between pollutant concentrations and the onset of health problems in schoolchildren has been found (Bono et al., 2015; Cartieaux et al., 2011). Further, a high prevalence of asthma has been reported in children attending school in classrooms with $PM_{2.5}$ levels higher than 17.5 $\mu g/m^3$ (Annesi-Maesano et al., 2012).

Most studies have focused on one specific group of pollutants or on thermal conditions (Branco et al., 2014a; Branco et al., 2015; Carreiro-Martins et al., 2014; Fonseca et al., 2014; Rivas et al., 2014). Therefore, measuring a wide range of health-relevant pollutants has become very important for assessing the indoor air pollution levels in schools.

The size distribution of particle-bound fire retardants (FRs) is a critical factor governing their fate in the air. It has also been associated with greater health risk to children because smaller particles can penetrate deeper into the lungs. Yet, few studies have analysed the health risk for kindergarten children exposed to dechlorane plus (DP), which is a halogenated FR used worldwide, and PBDEs transmitted through indoor $PM_{2.5}$ and dust. The present study evaluates the concentrations of these pollutants and their relationships, and additionally explores the health risks to children from FRs (DP, PBDEs) through indoor $PM_{2.5}$ and dust, along with carbonyl compounds and black carbon aerosol, found in our kindergartens and children's homes.

2. Methods

2.1. Description of sampling site

This study was carried out in Hong Kong, which is one of the most developed and urbanised cities in China. Between May 22 and July 23, 2014, indoor and outdoor air samples were obtained from five kindergartens (named: K1, K2, K3, K4, K5) with wide geographical coverage representative of the Hong Kong urban area. Fig. 1 shows the location of these five kindergartens.

Two classrooms from each of the five kindergartens were selected for this study, all of which had wooden flooring, plastic toys, wooden tables and chairs, watercolour pigments, water-based paints and oil paint covering the walls and ceilings. The rooms were ventilated through doors, windows, fans and air conditioning. Data on the characteristics of the classrooms in each school were collected, and they are presented in Table 1. The condition of the school buildings, the furnishing materials and table setting were little different among classrooms in the same kindergarten. As shown in Table 1, the size of the classrooms ranged from 3.3×10^1 to 4.0×10^1 m^2 (median: 3.8×10^1 m^2) and their volume ranged between 1.3×10^2 and 1.9×10^2 m^3 (median: 146.0 m^3). The classrooms were furnished with a computer, a television, an air conditioning system, a fan, small wooden tables, plastic chairs and many plastic toys.

2.2. Sampling and analysis

The indoor air pollutants measured consisted of fine particulate matter ($PM_{2.5}$), black carbon (BC) aerosol, carbonyl chemicals, PBDEs, and DP. The detailed procedures regarding sample preparation, extraction, and instrument analysis are described in the support information, S1.

The sampling equipment was placed 1.0 m above the floor level. One sample of the above referenced pollutants was taken from each classroom. In each site, samples were collected on three non-consecutive days inside the kindergarten and one day outside of the kindergarten.

The real-time level of $PM_{2.5}$ was measured by a Dust-Trak (TSI Model 8532), which is a real-time laser photometer instrument used for determining aerosol mass concentrations at the sampling rate of 1.7 L/min. In addition, $PM_{2.5}$ samples were collected on a quartz filter (47 mm, Whatman Inc., USA), using MiniVol portable air samplers (Airmetrics, USA), as a Dust-Trak calibration system. Membrane filters were conditioned in dry air for at least 48 h and weighed to a precision of 10 mg. Real-time BC concentrations in the indoor air were measured using a microAeth Model AE-51 (Magee Scientific, Berkeley, CA) by drawing ambient air using a quartz filter based strip which then measured BC using a single 880 nm LED.

The carbonyl concentrations were quantified using external calibration curves constructed from the standard solutions of a TO11 carbonyl–DNPH (2,4-Dinitrophenylhydrazine) Mix. The sampling periods for carbonyl compound levels were between five to eight hours depending on the occupation of the classrooms by the students. In-laboratory experiments demonstrated that the collection efficiencies were >93 ± 5% for all target carbonyls under the same flow rate, temperature and RH (relative humidity).

For the analysis of PBDEs and DP in PM2.5 and dust, GC-NCI/MS methods were used. Deionised water was generated in-house using a Super-Q water generation system. Dust and filters with sodium sulphate were weighed in a centrifuge tube. The spiked samples were extracted by a different ratio of solvents. The extracts were analysed by an Agilent gas chromatograph (GC)-mass spectrometer (MS) system (7890B-5977A) (Agilent Technologies, Palo Alto, CA) fitted with a DB-5MS column (15 m long × 0.25 mm i.d. × 0.10 μm or 0.25 μm film thickness), with helium at a rate of 1 ml/min as the carrier gas. Injection of a 2 μL sample was performed with an automatic sampler using the splitless injection mode. Quality assurance/quality control (QA/QC) was conducted by performing field and laboratory blanks, standard spiked recoveries and GC/MS detection limits. Daily multi-level calibrations were also conducted. The positive identification of PBDEs and DP peaks was assured by both their retention times and their mass ratios referenced in the standards. All laboratory glassware, gloves and polyethylene bags used in sample preparation were checked to ensure they were free of POPs. One laboratory control sample and one