FLAME RETARDANTS IN INDOOR AIR AND DUST OF A HOTEL IN JAPAN

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Introduction

For the purpose of fire prevention, fire safety regulations are essential for various products and applications used in all areas of our life, such as buildings, interior decoration, furnishings, upholsteries, electrical/electronic appliances and transport vehicles.[1] Use of flame-resistant or flame-retarded materials are general countermeasures and especially flame retardants (FRs) have been added to the materials in order to achieve the necessary safety level by passing the standardized fire test.[1] However, it is generally unknown to users or residents that which FRs and which amount of FRs are being used in our indoor environment.

Attentions have been paid to brominated flame retardants (BFRs) in indoor air and dust from homes as important exposure media to humans.[2-4] On the other hand, occurrence of BFRs in the indoor environment of highly flame-retarded public facilities (e.g., hotels, schools, hospitals, public transports, *etc*.) is also our concerns from the viewpoint of BFR exposure, because it appears very likely that BFRs are used with larger extent in these facilities compared to homes.[5]

In this study, a Japanese commercial hotel was chosen as an anti-flaming facility and concentrations of FRs {i.e., BFRs and organophosphate triesters (OPTs)} in floor dust samples were investigated. In addition, passive air sampling was conducted to monitor BFRs in indoor air of hotel rooms and performance of an air cleaner placed in the room was evaluated in terms of reduction of BFR concentration in the air.

Materials and Methods

Hotel. One commercial hotel in Osaka, Japan was selected for this case study. The hotel is a ten-story building and has 57 rooms. Prior to air and dust sampling, a handheld X-ray fluorescence (XRF) analyzer (Element tester innovxα6500, Innov-X systems) was used for non-destructive analysis of bromine in interior decoration, furnishings, upholsteries and electronic/electrical appliances aiming at screening of BFR-containing products. As examples of screening results, bromine contents higher than 100 ppm were found in television (TV) back casings (9.2 - 10%, n = 3), thick curtains (1.4 - 2.0%, n = 2), race curtain (1.0%, n = 1), bed spread (0.35%, n = 1), wallpaper (750 ppm - 0.29%, n = 2) and partition wall (0.25%, n = 1).

Dust sampling. Eight dust samples were collected by a vacuum cleaner equipped and used for daily cleaning on each floor in November 2006. After dust collection, impurities were removed and the whole sample amount (7 - 35 g) was used for the subsequent analysis.

Dust analysis. The target substances were extracted from dust samples with acetone and toluene in a soxhlet extractor, respectively. The combined extracts were cleaned up for the determination of brominated compounds {i.e., polybrominated diphenyl ethers (PBDEs, 1-10 brominated), decabromodiphenylethane (DBDPE), hexabromocyclododecanes (HBCDs), tetrabromobisphenol A (TBBPA), tribromophenols (TBPs) and polybrominated dibenzo-*p*-dioxins/furans (PBDD/Fs, 4-8 brominated)} by individual adsorption column chromatography with sample derivatization (ethylation) in the case of TBBPA and TBPs. For analysis of those compounds a GC/HRMS system was used. The purification of OPTs from the same combined extracts was conducted by silica gel column chromatography and eleven OPTs (see Fig. 2) were analyzed by GC/QMS.

Passive air sampling and performance evaluation of an air cleaner. In both of two twin rooms (rooms A and B) with the identical area and symmetrical layout of furnishings, polyurethane foam (PUF) passive air samplers were deployed at the height of 180 cm for three to four weeks from December 2006 to January 2007. In this device, PUF disks (150 mm diameter, 15 mm thickness) were sheltered by two different size stainless steel housings during air sampling.[6] In room B, an air cleaner (ventilation rate: 0.4 - 4.3 m^3/h, Duskin Co. Ltd., Japan) equipped with HEPA and activated carbon filters was placed and operated. Then, performance of the air cleaner was evaluated in terms of reduction of BFR concentrations in room air by comparing collecting amount of BFRs trapped by each sampler in two rooms. Polyflon filter (PF-050, ADVANTEC) was attached on the front surface of air intake of the cleaner for sampling of suspended dust. After experiment, the PUF and polyflon filter were respectively treated for extraction, clean-up and determination of BFRs according to the same protocol with dust samples mentioned above. During experiment, guests were actually staying in both rooms and use of the air cleaner in room B was at the option of guests.

Results and Discussion

Concentrations of BFRs in dust. The concentrations of BFRs and PBDD/Fs measured in each floor dust sample are provided in Fig. 1. The dust levels of PBDEs and HBCDs varied over about two orders of magnitude, from 9.8 – 1,700 ng/g, with a median of 1,200 ng/g for PBDEs, and from 72 – 1,300 ng/g, with a median of 740 ng/g for HBCDs. The concentrations of the above two types of BFRs were most dominant among the investigated BFRs in the dust samples and a significant correlation was observed between PBDE and HBCD concentrations. DecaBDE was most abundant among PBDE congeners in the dust samples. The concentrations of PBDEs are consistent with our previous results of house and office dust samples (median concentrations: house dust 700 ng/g, n = 19; office dust 1,800 ng/g, n = 14) in Japan.[7] Compared to the reported HBCD concentrations (390 -730 ng/g as a median concentration) in house and office dust from Canada, UK and US,[8] our results are in good accordance with them. Although diastereomer patterns of HBCDs were not investigated in this study, it is of our interest to know the contribution ratio of each diastereomer with different behavior and toxicities. PBDD/F concentrations (range: 740 - 6,300 pg/g, median concentration: 3,800 pg/g) were in the same order of magnitude, however, at slightly higher levels compared to those in Japanese house and office dust (house dust: median 1,800

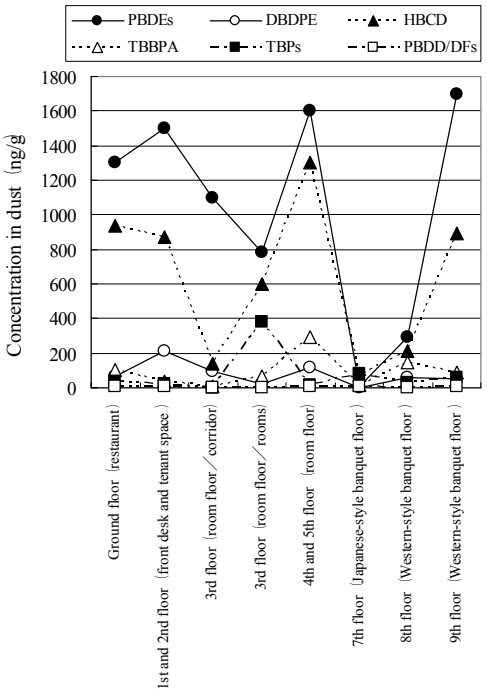

Fig. 1 Concentrations of BFRs and PBDD/Fs in hotel dust samples.

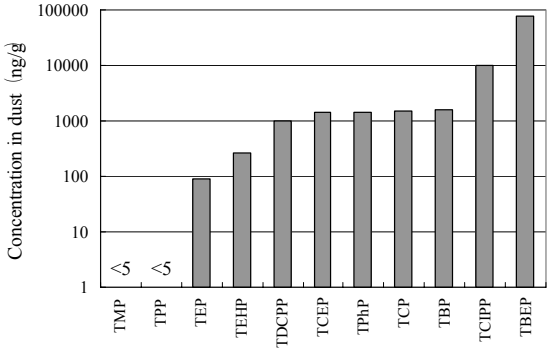

Fig. 2 Concentrations of OPTs in a hotel dust sample from the 4th and 5th floors.

TMP: trimethylphosphate, TPP: tripropylphosphate, TEP: triethylphosphate, TEHP: tris(2-ethylhexyl phosphate), TDCPP: tris(1,3-dichloro-2-propyl)phosphate, TCEP: tris(2-chloroethyl)phosphate, TPhP: triphenylphosphate, TCP: tricrecylphosphate, TBP: tributhylphosphate, TCIPP: tris(2-chloroisopropyl)phosphate, TBEP: tris(butoxyethyl)phosphate

pg/g, n = 19; office dust: median 2,500 pg/g, n = 14).[7] Regarding other BFRs, DBDPE being marketed as an alternative to PBDEs[9] was detected broadly from almost all the dust samples, though the concentrations were one to two orders of magnitude smaller than those of PBDEs and HBCDs.

As a summary on the occurrence of BFRs in the investigated hotel dust samples, it can be derived that BFR and PBDD/F concentrations are on the same level or relatively (not extravagantly) higher compared to those in house and office dust reported by the past studies.[7, 10-12] BFR concentrations were not uniform in every dust in the hotel but different with floors, suggesting localization of source products such as TV sets and furniture are strongly associated with the BFR concentrations in dust.[13]

Concentrations of OPTs in dust. The concentrations of eleven OPTs, additive flame retardants or plasticizers were measured for all the dust samples. As a representative result, data of the dust collected from the 4th and 5th floors were shown in Fig. 2. From this sample, seven OPTs were detected in the order of μg/g, which are equivalent to or exceed the BFR concentrations such as PBDEs and HBCDs. Such results implicate that OPTs are used diversely and intensively in the hotel facility. Especially, tris(butoxyethyl)phosphate (TBEP) and tris(2-chloroisopropyl)phosphate (TCIPP) were detected with high concentrations (77 and 9.8 μg/g, respectively). TBEP is widely used as a plasticizer in polymer dispersion based floor polishes, while TCIPP is added to plastics as FRs.[14] These compounds are generally used in amounts of % order, relative to the whole products. The OPT concentrations varied two orders of magnitude at maximum with samples from each floor, however, there observed a similar tendency in rank order of OPT concentrations among dust samples. As emerging contaminants, occurrence and behavior of OPTs (including condensed type[15] with increasing use) should be taken into account as a risk factor as well as BFRs.

Passive air sampler monitoring results and effect of an air cleaner on the reduction of airborne BFRs. The amount of airborne BFRs collected by passive air samplers (deployed in rooms A and B) and a polyflon filter in the air cleaner (placed in room B) was shown in Fig. 3. The present sheltered PUF devise could sample ng amount of low brominated PBDEs (MonoBDEs to PentaBDEs) and TBPs existing primarily in the vapor phase during a period of four weeks, while it was insufficient for the collection of high brominated PBDEs, HBCDs, TBBPA and PBDD/Fs that exist mainly in the particle phase. On the other hand, ng amount of DecaBDE and HBCDs could be detected from a polyflon filter of the air cleaner set in room B, which was due to the capture of airborne particles. Furthermore, it was suggested that gaseous PBDEs and TBPs were also reduced by the air cleaner from the comparison of amount of those compounds collected by PUF in rooms A and B. This reduction could be explained by the performance of HEPA and activated carbon filters in the cleaner. It is implied that operation of an appropriate air cleaner can reduce both of gaseous and particulate BFRs in indoor air.

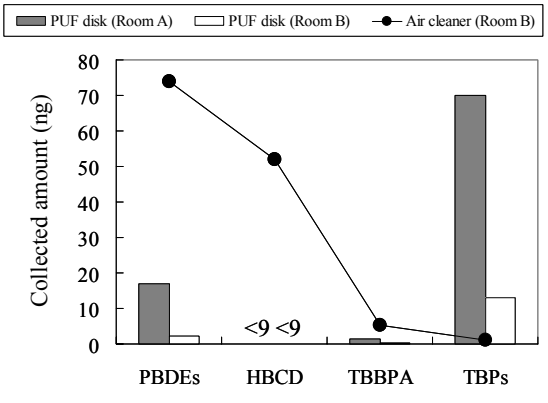

Fig. 3 Collected amount of BFRs trapped by passive PUF disk samplers and a polyflon filter in the air cleaner (equivalents to four week collection).

For many people, a hotel stay is short so that exposure level in a hotel might not be at risk. However, comparatively high level of BFRs in hotel should be associated with exposure concerns during long-term stay and occupational works. Our study results provided information on BFR levels in a hotel and will be useful for planning indoor BFR exposure avoidance as part of general control and management strategy to the public.

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