Environmental Pollution 181 (2013) 75–80

Contents lists available at SciVerse ScienceDirect

Environmental Pollution

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

Dioxin-related compounds in house dust from New York State: Occurrence, *in vitro* toxic evaluation and implications for indoor exposure

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ARTICLE INFO

Article history:
Received 7 April 2013
Received in revised form
3 June 2013
Accepted 5 June 2013

Keywords:
CALUX
Brominated dioxins
House dust
PBDEs
TIE

ABSTRACT

This study analysed sulphuric-acid-treated extracts of house dust from New York State with DR-CALUX assay and HRGC-HRMS to elucidate the total dioxin-like (DL) activities, the occurrence of various dioxin-related compounds (DRCs), including PBDD/Fs, and their toxic contribution. The DL activities were 30–8000, median 210 pg CALUX-TEQ/g. PCDD/Fs, PBDD/Fs and DL-PCBs were detected with a large variation in concentrations (0.12–80, 0.33–150, 0.46–35, medians 1.7, 2.1 and 5.6 ng/g, respectively) and profiles, indicating the existence of multiple contamination sources in homes. PCDD/Fs, PBDD/Fs and DL-PCBs with known potency theoretically contributed <1%–130%, <1%–21% and <1%–6.8%, respectively, of the measured CALUX-TEQs. These results and those from DR-CALUX assays with fractionated dust extracts indicated that a substantial portion of the CALUX-TEQs could be caused by unknown dust contaminants. Considering that the DRC intake from indoor dust ingestion can be significant, identification of unknown DL contaminants in indoor dust is necessary.

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1. Introduction

Indoor dust can be a sink of many contaminants (Butte and Heinzow, 2002), including those that are released during weathering of polymeric and textile materials in consumer electronics and other household products (Suzuki et al., 2009). Over the past decade, occurrence of additive flame retardants (FRs) has gained considerable interest, as they were found at notable concentrations in house dust (Abdallah et al., 2008; Sjödin et al., 2008; Stapleton et al., 2009). These findings also indicated that house dust was a major source of polybrominated diphenyl ether (PBDE) exposure (Jones-Otazo et al., 2005; Wu et al., 2007). The importance of indoor exposure to PBDEs is most evident in the U.S., where elevated contamination levels in house dust were proposed as a plausible explanation for the much higher human residue levels of PBDEs than for populations of other countries (Johnson-Restrepo and Kannan, 2009; Lorber, 2008).

Indoor exposure to dioxin-related compounds (DRCs) has rarely been studied because conventional DRCs, namely polychlorinated dibenzo-*p*-dioxins/dibenzofurans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (DL-PCBs), are released mainly from combustion processes and industrial activities (UNEP, 1999) and do not have obvious sources in the indoor environment. Exposure to DRCs *via* non-dietary pathways was regarded as minor in comparison to diet, and was reported to contribute less than 10% of the total daily intake for Americans (Lorber et al., 2009). However, exposure through dust ingestion becomes more relevant when non-regulated DRCs are considered. Polybrominated dibenzo-*p*-dioxins/dibenzofurans (PBDD/Fs) can be generated from brominated FRs, most favourably from the ubiquitous PBDEs (Kannan et al., 2012; Weber and Kuch, 2003). The formation of PBDFs from PBDEs requires only mild thermal stress (<300 °C, Mandalakis et al., 2008; Weber and Kuch, 2003) or natural light exposure (Kajiwara et al., 2008). PBDFs were detected in technical PBDE formulations as impurities (257 ng/g in PentaBDE up to 30–48 μg/g

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http://dx.doi.org/10.1016/j.envpol.2013.06.010

in DecaBDE; Hanari et al., 2006) and also in TV cases (up to 500 μg/g; Watanabe and Sakai, 2003). Recent studies in Japan and Vietnam have found PBDFs in indoor dust at concentrations on the order of hundreds to tens of thousands of picogram per gram (Takigami et al., 2009; Suzuki et al., 2010; Tue et al., 2010). Indoor dust PBDF levels correlated strongly with those of PBDEs (Ma et al., 2009; Tue et al., 2010).

PBDD/Fs can bind to the aryl hydrocarbon receptor (AhR) and induce the expression of related genes with potencies similar to those of PCDD/Fs (Behnisch et al., 2003; Kannan et al., 2012; Olsman et al., 2007). These non-regulated DRCs are thus expected to exert dioxin-like (DL) toxic effects, including reproductive and developmental abnormalities, immune deficiency, tumour promotion and endocrine disruption (van Leeuwen et al., 2000). In our previous reports (Suzuki et al., 2010; Tue et al., 2010), PBDFs and unidentified, but persistent, AhR agonists were found to contribute significantly to the total DL activities in indoor dust from Japan and Vietnam, based on studies that involved a combination of Dioxin-Related Chemical-Activated LUciferase gene eXpression (DR-CALUX) assay and instrumental chemical analysis. These results highlight the need for the reassessment of DRC exposure *via* indoor dust ingestion.

Considering the high concentrations of PBDEs reported in the U.S. indoor environment, the occurrence and potential risk of DRCs including PBDD/Fs need to be investigated. Using a combined *in vitro* bioassay/instrumental analysis approach, we aimed at elucidating the DL activities in house dust from New York State to evaluate the significance of various DRCs in indoor dust.

2. Material and methods

2.1. Sample collection

Dust samples were obtained from vacuum cleaner bags in 21 houses in Albany (AL) and its surrounding areas including Slingerlands (SL), Loudonville (LV), Troy (TR), Fultonville (FV), Rensselaer (RE), Castleton (CA), Ballston Spa (BS), Schenectady (SC) and Niskayuna (NI) in New York State, USA between August 2005 and July 2006 (see Supplementary data for further details). Compared with house dust samples collected using standardised vacuum cleaners and protocols (e.g. with predefined criteria for room and surface selection, area and time for vacuuming), those from donor-provided vacuum bags can be subject to variation in vacuuming methods and habits but are more easily available and may provide a measure of dust concentrations integrated over the home for a longer period (Allen et al., 2008). The samples were transferred to pre-cleaned glass jars, transported to the laboratory, sieved through a 500-μm sieve for removal of large debris, and stored at −20 °C until analysis.

2.2. Sample extraction and clean-up

A portion of each dust sample (0.5–2.0 g) was extracted using a rapid solvent extractor (SE100, Mitsubishi Chemical Analytech, Japan) with an acetone/hexane mixture (1:1 v/v, 35 °C) and subsequently with toluene (80 °C) for 1 h each at a flow rate of 6 ml/min. The combined extract was solvent-exchanged into hexane, and a portion was used for bioanalytical and chemical analyses of DRCs. To target only contaminants with potential metabolic persistence—a criterion for dioxin-related compounds (van den Berg et al., 2006), non-persistent compounds such as polyaromatic hydrocarbons (PAHs) were removed from the extract by successive treatment with concentrated sulphuric acid, sulphuric acid-impregnated silica gel column and gel-permeation chromatography, according to a previously reported method (Tue et al., 2010). The extract was then concentrated, solvent-exchanged into 0.1 mL biochemical-grade dimethyl sulphoxide (DMSO) and stored at 4 °C for subsequent analyses. Every set of seven samples was accompanied with a procedural blank. The repeatability of the above procedure was determined by the analysis of NIST SRM2585 reference dust material with DR-CALUX.

2.3. DR-CALUX assay

Dioxin-like activity was measured using DR-CALUX assay with a rat hepatoma cell line with an AhR-regulated luciferase gene construct (H4IIE-luc, BioDetection Systems b.v., The Netherlands). The culture conditions, assay procedures and data analysis followed the protocol described elsewhere (Suzuki et al., 2007). Throughout the study, the calculated EC_{50} of the 2,3,7,8-CDD (TCDD) standard was 7.7 ± 1.1 pM ($n = 27$) and the maximum induction relative to DMSO control ranged from 6.1 to 15.0 (10.6 ± 2.3), satisfying the data quality objectives described in the standard operating procedure. The limit of quantification ranged from 0.07 to 0.80 pM TCDD per well. Results were expressed in picogram CALUX TCDD-equivalent (CALUX-TEQ) per gram dust.

2.4. Chemical analyses of DRCs and evaluation of toxic equivalents (TEQs) for the identified compounds

Sulphuric acid-treated extracts containing at least 100 pg CALUX-TEQ ($n = 16$) were analysed for individual PCDD/Fs, PBDD/Fs, Mo–DiBPCDD/Fs and DL-PCBs using gas chromatography–high resolution mass spectrometry (HRGC–HRMS) after spiking of $^{13}C_{12}$-labelled standards and additional clean-up with multilayer silica gel column containing copper chips and activated carbon-impregnated silica gel column. The concentration of each homologue group was calculated from the areas of peaks identified with authentic standards and those of unidentified peaks. Recoveries of the labelled standards were between 64% and 110%. Other details on the analytical method and quality assurance/quality control have been reported elsewhere (Tue et al., 2010).

The DL activity of an individual DRC was calculated as WHO-TEQ or theoretical CALUX-TEQ, product of its concentration and toxic equivalency factor (TEF) or CALUX-derived relative potency factors (REP). The WHO-TEQs, used for comparison with other studies on DRCs, were calculated using TEFs assigned by the World Health Organization for 2,3,7,8-substituted PCDD/Fs and DL-PCBs (van den Berg et al., 2006). The WHO-TEQs of PBDD/Fs were calculated using TEFs of PCDD/Fs with the corresponding substitution patterns as interim TEFs (van den Berg et al., 2013). Theoretical CALUX-TEQs were used to evaluate the contribution of the known DRCs to the experimental DL activities obtained with DR-CALUX, and were calculated using EC_{20}- or EC_{25}-based REPs compiled in a previous report (Tue et al., 2010).

2.5. Fractionation scheme for characterisation of unexplained dioxin-like activities

In order to evaluate the DL activities of unknown compounds, including brominated DRCs, five sulphuric acid-treated extracts (obtained in 2.2) were subjected to the following fractionation procedure for separation of (a) compounds in the fraction used in conventional dioxin analysis from those with higher polarity, and (b) PBDD/Fs from PCDD/Fs and DL-PCBs. Each extract was loaded on a multilayer silica cartridge connected to an alumina cartridge at the outlet (Miura Aquatech, Japan). Elution with hexane yielded a non-polar fraction containing PCBs (fraction H). Then halogenated dibenzo-*p*-dioxins/dibenzofurans trapped in the alumina cartridge during the first elution were eluted with toluene (fraction T). Finally a mixture of dichloromethane/hexane (1:3 v/v) was used to elute more polar compounds in the multilayer silica cartridge (fraction D). In a parallel experiment, the multilayer cartridge was loaded with silver nitrate-impregnated silica gel which shifted the elution of PBDD/Fs into fraction D instead of fraction T. All fractions were solvent-exchanged into DMSO and the DL activity of each fraction was evaluated with DR-CALUX.

3. Results and discussion

3.1. DR-CALUX-derived dioxin-like activities

All sulphuric acid-treated dust extracts exhibited AhR agonism as evidenced by dose-dependent induction of luciferase activities with response slopes similar to that of the TCDD standard. DR-CALUX measurement of the extracts obtained from a triplicate sample of SRM2585 indicated that the first extraction step with acetone/hexane yielded 3370 ± 99 pg CALUX-TEQ/g and the second step with toluene 300 ± 10 pg CALUX-TEQ/g, indicating good extraction repeatability. The total CALUX-TEQs obtained with this extraction scheme was slightly higher than the CALUX-TEQs obtained for extracts with only toluene (3000 ± 150 pg/g), the common extraction solvent for dioxin analysis.

The CALUX-TEQ levels in house dust from Albany, NY were in the range of 30–8000 pg/g (median 210), similar to the ranges reported in a limited number of studies on *in vitro* DL activity in indoor dust: 38–1400 pg/g (median 110–220) in house/office dust from Japan (Suzuki et al., 2007) and 49–1000 pg/g (median 140–520) in house dust from Vietnam (Tue et al., 2010). However, the CALUX-TEQ levels in this study varied over a wide range of up to two orders of magnitude, and the upper quartile (550 pg/g) was higher than those reported for common houses in previous studies and comparable to that reported for house dusts in e-waste recycling areas

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