Concentrations of Polychlorinated Biphenyls in Indoor Air and Polybrominated Diphenyl Ethers in Indoor Air and Dust in Birmingham, United Kingdom: Implications for Human Exposure

STUART HARRAD,* SADEGH HAZRATI, AND CATALINA IBARRA

Division of Environmental Health and Risk Management, Public Health Building, School of Geography, Earth, and Environmental Sciences, University of Birmingham, Birmingham, B15 2TT, United Kingdom

Polybrominated diphenyl ethers (PBDEs) and polychlorinated biphenyls (PCBs) were measured in air (using PUF disk passive samplers) in 31 homes, 33 offices, 25 cars, and 3 public microenvironments. Average concentrations of $\Sigma$BDE (273 pg m$^{-3}$) and $\Sigma$PCB (8920 pg m$^{-3}$) were an order of magnitude higher than those previously reported for outdoor air. Cars were the most contaminated microenvironment for $\Sigma$BDE (average = 709 pg m$^{-3}$), but the least for $\Sigma$PCB (average = 1391 pg m$^{-3}$). Comparison with data from a previous spatially consistent study, revealed no significant decline in concentrations of $\Sigma$PCB in indoor air since 1997–98. Concentrations in indoor dust from 8 homes were on average 215.2 ng $\Sigma$BDE g$^{-1}$, slightly higher than other European dust samples, but twenty times lower than Canadian samples. Inhalation makes an important contribution (between 42 and 63% for adults) to overall UK exposure to $\Sigma$PCB. For $\Sigma$BDE, dust ingestion makes a significant but—in contrast to Canada—a not overwhelming contribution (up to 37% for adults, and 69% for toddlers). Comparison of UK and Canadian estimates of absolute exposure to $\Sigma$BDE suggest that differences in dust contamination are the likely cause of higher PBDE body burdens in North Americans compared to Europeans.

Introduction

Polychlorinated biphenyls (PCBs) have found widespread use in a diverse range of applications, with around 1.2 million tonnes produced worldwide (1). Of this, approximately 67 000 and 40 000 t were produced and used, respectively, in the UK (1). Owing to concerns about their adverse effects on humans and wildlife, their production—but not their use—ceased in the UK and throughout most of the industrialized world in the late 1970s. Although UK human exposure to PCBs via the diet has fallen in recent years in response to the cessation of their production (2, 3), human health concerns remain: currently a substantial proportion of UK school-children and toddlers are exposed to dioxins and dioxin-like PCBs via the diet at levels that exceed the UK government’s recommended tolerable daily intake (3). Although the majority of nonoccupational exposure to PCBs has been widely considered to occur via the diet (4), there have been increasing indications that indoor air remains contaminated by PCBs remaining in use in applications such as permanent elastic sealants and acoustic ceiling tiles (5–7). As a result, inhalation of indoor air could constitute a significant exposure pathway; a fact compounded by the downward trend in dietary exposure.

Polybrominated diphenyl ethers (PBDEs) have found wide use as flame retardants. In recent years, production and use of PBDEs has been in the guise of three formulations: penta (consisting primarily of BDEs 47 and 99, 37% each, alongside smaller amounts of other tetra-, penta-, and hexa-BDEs), octa (a mixture of hexa-(10–12%), hepta- (44–46%), octa-(33–35%), and nona-(10–11%)), and deca (98% decabromodiphenyl ether—BDE 209—and 2% various nona-BDEs) (8, 9). Worldwide, PBDE production is dominated by the deca commercial formulation, with global demand in 2001 estimated 56 100 t (10). This is similar to the 1999 estimate of 54 800 t (11). By comparison, 2001 global demand for the penta-product was 7500 t (10), down slightly from 8500 t in 1999 (11). Production and use of commercial PBDE formulations in Europe was considerably less than that in North America, for example, in 2001, 7100 t of penta-product was used in North America, compared to just 150 t in Europe (10). The uses for these commercial formulations are myriad: the penta-product was employed principally to flame-retard polyurethane foams in carpet underlay, vehicle interiors, furniture, and bedding; the octa-formulation was used to flame-retard thermoplastics such as high-impact polystyrene, and the deca-product was used principally in plastic housings for electrical goods such as TVs and computers, as well as in textiles (8). As a result of concerns surrounding these contaminants owing to their presence in the diet and indoor air and dust (12–14), and human tissues (15), coupled with evidence relating to their potential adverse effects on human health (9, 16), several jurisdictions have banned the marketing and use of penta- and octa-BDEs. Furthermore, the main U.S. producer and the U.S. EPA have reached a voluntary agreement to discontinue production of the penta- and octa-BDE mixtures. Despite this, there remain strong concerns that the existing (largely indoor) reservoir of PBDEs (and PCBs) associated with treated goods represents a substantial source of current and future exposure to these compounds, both via direct inhalation and ingestion of contaminated indoor air and dust, and in time via dietary exposure following their emission, transport, and incorporation into the diet (17).

This study reports concentrations of a number of individual PCB and PBDE congeners in indoor air (sampled using PUF disk passive air samplers) taken from 31 homes, 33 offices, 25 cars, and 3 public microenvironments within the West Midlands, the UK’s second largest conurbation (population 2.5 million). These data are combined and compared with existing data on UK dietary exposure to PCBs to provide a preliminary indication of the relative significance of inhalation and diet to overall human exposure to PCBs for UK adults and toddlers. Further, given recent indications that, for Canadians, ingestion of indoor dust may constitute the most important exposure pathway to PBDEs for many individuals (13), concentrations of PBDEs in dust samples taken from 8 West Midlands homes are reported, and used in conjunction with estimates of inhalation and dietary intakes to provide a preliminary indication of the relative

* Corresponding author e-mail: S.J.Harrad@bham.ac.uk; tel: +44 121 414 7298; fax: +44 121 414 3678.