Environment International 117 (2018) 57–68

Contents lists available at ScienceDirect

Environment International

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

# Towards a systematic understanding of the dynamic fate of polychlorinated biphenyls in indoor, urban and rural environments

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ARTICLE INFO

*Keywords:*
Polychlorinated biphenyls
Indoor
Rural
Near-field
Far-field
Model

ABSTRACT

Indoor environments and urban areas are hubs of chemical stocks and emissions, which contaminate those indoor and urban areas as well as the surrounding rural areas. Here, we introduce a newly developed nested multimedia indoor-urban-rural chemical fate model, coupled with a substance flow analysis, aiming to provide an integrated and dynamic understanding of the mass distribution, concentrations, and major pathways of contaminants within and between indoor, urban and rural environments. The model is applied to simulate the emissions, transport and fate of polychlorinated biphenyl (PCB) congeners 28 and 153 in the Western Baltic drainage basin over time. Whereas > 90% of PCBs were used in the urban outdoor environment, the model indicates that ~80% of emissions occurred indoors because of higher emission factors in open-end usage. Atmospheric advection is highly effective in transporting the bulk of the PCBs emitted indoors to urban (> 85%) and rural (> 75%) environments. The rural environment is identified as the main locale for accommodating (> 80%) and removing (> 50%) the emitted PCBs. Contamination of exposure-relevant compartments in the rural environment is anticipated to decrease slower than, and thus outlast, that in the indoor environment, which implies an increasing importance of the food chain accumulation in overall human exposure to PCBs over time. Our model demonstrates that, whereas the indoor environment contains an insignificant fraction of the total emissions remaining in the regional environment, it experiences orders of magnitude higher concentrations than the rural environment. Therefore, while including indoor and urban environments in modeling influences little the modeled overall chemical fate on a regional scale, it strongly affects modeling the human exposure associated with multimedia concentrations.

## 1. Introduction

More than half of the world's population lives in urban areas (United Nations, 2014); urban residents spend on average 90% of their lifetime indoors (U.S. Environmental Protection Agency, 2011); > 90% of urban activities are nourished by goods, energy and information supplied from outlying rural areas (Baccini and Brunner, 2012). As a result of these three facts, indoor, urban and rural environments constitute an interconnected and interdependent system. Numerous anthropogenic chemicals are synthetized and processed at industrial sites located within urban environments. Chemicals can accumulate in in-use industrial and consumer products, as well as in waste disposal facilities such as landfills and dumps. These anthropogenic chemical stocks constitute part of the "*technosphere*" (Baccini and Brunner, 2012), which encompasses all human activities within the socioeconomic system and geographically overlaps with urban and indoor areas of the *physical environment*. Chemicals are emitted from the technosphere into the physical environments of different scale during the entire product lifespan via, e.g., passive volatilization, material abrasion, and material-dust partitioning (Liagkouridis et al., 2014). After entering indoor and urban environments, some chemicals undergo subsequent transport to rural environments and even to remote areas like the Arctic, with moving air and water (i.e., the "urban halo" effect) (Diamond and Hodge, 2007).

Notorious examples of such chemicals are polychlorinated biphenyls (PCBs). While PCB usage is largely confined to indoor and urban outdoor environments (Erickson and Kaley, 2011), the occurrence of PCBs is ubiquitous in various environmental matrices around the world (Iwata et al., 1993; Kalantzi et al., 2001; Meijer et al., 2003). Humans can take up these chemicals from environments of different scale via different routes, e.g., inhalation of contaminated air from the indoor environment ("near-field") and ingestion of contaminated food originating in the rural environment ("far-field") (Fantke et al., 2016; Norström et al., 2010). Once entering human bodies, PCBs are able to

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https://doi.org/10.1016/j.envint.2018.04.038
Received 2 March 2018; Received in revised form 20 April 2018; Accepted 20 April 2018

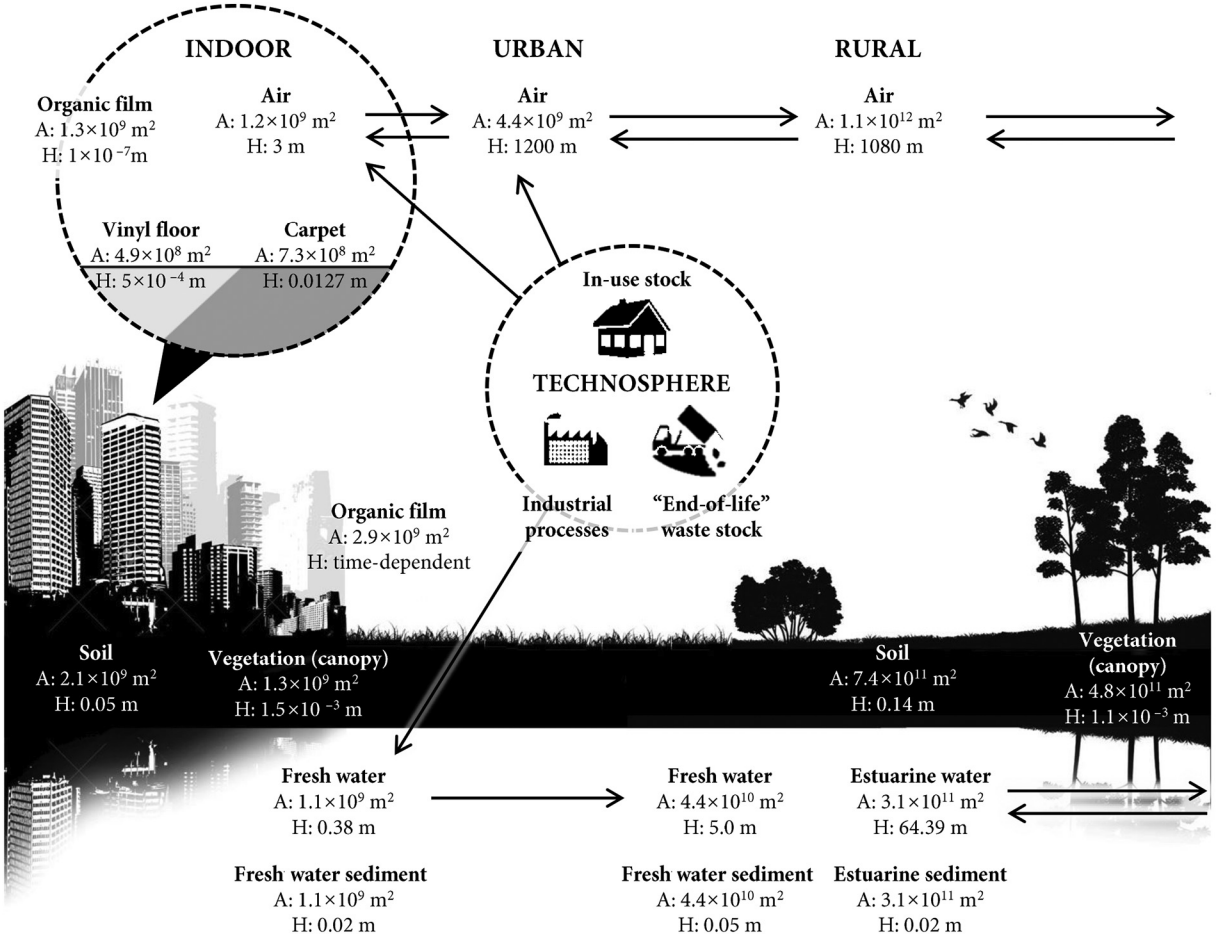


**Fig. 1.** Model representation of the technosphere, and the indoor, urban and rural environments of the Western Baltic drainage basin. The area (A) and height/depth/thickness (H) of each environmental compartment are also given. The indoor area is calculated based on the fraction (27.7%) of total indoor floor area in the Stockholm urban area (Cousins, 2012). The urban area is the sum of the reported areas of "urban agglomerations" (defined as areas with > 500,000 population) within the modeled region (Demographia, 2017); the rural area is calculated by subtracting the urban area from the total regional area [the default in CoZMo-POP (Wania et al., 2006)]. Arrows indicate mass exchanges among the three environments of different scale.

exert detrimental health effects such as endocrine disruption, neurotoxicity and carcinogenicity (Kimbrough, 1995). Therefore, in order to protect humans from adverse health outcomes, it is of vital importance first to understand the fate of chemicals in both the technosphere and environments of different scale.

A number of pioneering modeling studies have addressed separately the fate of chemicals in either indoor (Liagkouridis et al., 2014; Shin et al., 2013; Zhang et al., 2009), urban (Csiszar et al., 2012a; Csiszar et al., 2012b; Diamond et al., 2001), and rural (Glüge et al., 2016; Wania et al., 2006) environments. What is still missing is a holistic, comparative picture of chemical fate along the entire indoor-urban-rural continuum, which would equip us to answer the following important questions:

(I) An environment can be contaminated due to releases within that environment or due to releases elsewhere. What is the relative importance of these contributions? For example, what fraction of the contamination of the rural environment is due to releases in the indoor or the urban environment? The answer to this question unravels to what extent a chemical emitted indoors is capable of migrating to rural areas, contaminating the food chain and thus causing human exposure beyond that occurring in indoor environments. However, a few earlier attempts drew contradictory conclusions. For example, based on field evidence, the ventilation of indoor emissions is estimated to contribute ~90% of the polybrominated diphenyl ethers (PBDEs) in Stockholm outdoor air (Björklund et al., 2012). By contrast, follow-up modeling work asserts that ventilation explains only 27% of the observed outdoor contamination and, instead, atmospheric advective input from outside Stockholm is hypothesized to be the major source (Cousins et al., 2014);

(II) What are the respective roles of environments of different scale in shaping the overall fate of chemicals emitted to the physical environment? For example, can we infer from the urban–rural concentration gradients apparent for many anthropogenic chemicals, e.g., PCBs and PBDEs (Harner et al., 2006; Harner et al., 2004; Harrad and Hunter, 2006), that the bulk of these chemicals resides in urban environments? Similarly, a few anthropogenic removal processes, e.g., removal of indoor dust via vacuuming and mopping (Shin et al., 2013; Zhang et al., 2009), have been identified as efficient, if not dominant, pathways removing certain chemicals from indoor and urban environments. How do these anthropogenic removal processes compare quantitatively to natural ones, e.g., degradation and sediment burial (Wania and Daly, 2002), and what are their contributions to the overall permanent loss of a chemical on a regional scale? With identification of the most relevant processes, decision makers can take pertinent actions to minimize the environmental and health risk of chemicals;

(III) How does the contamination in indoor, urban and rural environments co-evolve with time? If the rural environment receives PCBs originally emitted into indoor or urban environments, are there differences in the temporal concentration trends between