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Long-term emissions of hexabromocyclododecane as a chemical of concern in products in China

Li Li [a], Roland Weber [b], Jianguo Liu [a,*], Jianxin Hu [a]

[a] College of Environmental Sciences and Engineering, Peking University, Beijing 100871, PR China
[b] POPs Environmental Consulting, Lindenfirststr 23, Schwäbisch Gmünd 73527, Germany

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ABSTRACT

There has been ever-increasing international interest in investigating the long-term emissions of chemicals in products (CiPs) throughout their entire life cycle in the anthroposphere. Hexabromocyclododecane (HBCDD) is a contemporary example of special interest due to the recent listing of this hazardous flame retardant in the Stockholm Convention and the consequent need for parties to take appropriate measures to eliminate this compound. Here, we conducted a scenario-based dynamic substance flow analysis, coupled with interval linear programming, to forecast the future HBCDD emissions in China in order to assist with the implementation of the Stockholm Convention in this current world's predominant HBCDD manufacturing and consuming country. Our results indicate that, under a business-as-usual scenario, the cumulative HBCDD production will amount to 238,000 tonnes before its phase-out, 79% of which will be consumed in domestic market, accumulate as stocks in flame-retarded polystyrene insulation boards, and ultimately end up in demolition waste. While the production is scheduled to end in ca. 2021, emissions of HBCDD would continue until after 2100. For the entire simulation period 2000–2100, 44% of total cumulative emissions will arise from the industrial manufacture of HBCDD-associated end-products, whereas 49% will come from the end-of-life disposals of HBCDD-containing waste. The most effective end-of-life disposal option for minimizing emissions we found was, a pre-demolition screening combined with complete incineration. Our study warns of the huge challenges that China would face in its eliminating HBCDD contamination in the following decades, and provides an effective methodology for a wider range of countries to recognize and tackle their long-term emission problems of hazardous CiPs.

1. Introduction

Recent years have seen an increasing collaboration among academia, consumers and policy makers to disclose, assess and alarm potential risks of chemicals in products (CiPs) on human health and the environment (Kogg and Thidell 2011; UNEP and DTIE 2011). One of the major chemical groups of concern is hazardous flame retardants, e.g. polybrominated diphenylethers (PBDEs) and hexabromcyclododecane (HBCDD) (DiGangi et al. 2010; Shaw et al. 2010). For decades, HBCDD has been used in flame-retarded (FR) expanded and extruded polystyrene (EPS and XPS) insulation boards, textiles and other minor consumer products (UNEP 2015). HBCDD is ubiquitous in the environment (Covaci et al. 2006) with evident adverse environmental and health impacts (Marvin et al. 2011). While it has been listed as a persistent organic pollutant (POP) in the Stockholm Convention in 2013 (UNEP 2013), HBCDD is still in production and uses at a level of >30,000 tonnes a^{-1} worldwide (UNEP 2015). At present, China has become the world's dominant HBCDD producer and consumer (POPRC 2011; POPRC 2012). The absence of HBCDD-specific management strategies in China is in dramatic contrast to the rocketing market demands for FR insulation materials that are resulted from ever-increasing pursuit of both fire safety and energy efficiency in constructions during the past decade (Peking University 2012). While some previous attempts have been made to characterize the emissions of HBCDD in industrialized countries such as Switzerland (Morf et al. 2007), the United Kingdom (Mark et al. 2010), and Germany (Potrykus et al. 2015), these estimates from single countries are of limited relevance to developing countries and countries with economies in transition, in particular China. This irrelevance is because the latter countries lack appropriate waste management frameworks and adequate levels of sophisticated waste destruction capacities for problematic waste such as POPs (Weber et al. 2013). Hence, establishing the emissions in China and exploring viable emission mitigation strategies are essential for the global progress towards an HBCDD-free environment.

There are, however, two major unique features of HBCDD as a CiP which make the emission estimation of this compound more challenging than that of traditional contaminants, e.g., chemicals used in either agriculture or industrial processing. The *first* feature is that, in addition to the immediate and concentrated emissions from production and new industrial uses, there are also long-term, continuous and dispersive emissions of the compound from its in-use and end-of-life waste stocks.

* Corresponding author.
E-mail address: jgliu@pku.edu.cn (J. Liu).

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HBCDD is designed to be stable and durable during its use phase (Kajiwara et al. 2013). After the use phase the bulk of HBCDD in products will end up in waste stocks such as construction and demolition (C&D) waste and obsolete textiles (Morf et al. 2008), thus, the end-of-life disposal is likely to be a major source of HBCDD emissions in the future. For example, 75% of the United Kingdom's total HBCDD emissions in 2030 are anticipated to come from waste landfill (Mark et al. 2010). The *second* feature is the uncertainty that arises from inconsistent, vague and/or inadequate input data on marketing, content, and end-of-life management of products (Vyzinkarova and Brunner 2013). For example, it is difficult to stipulate a typical HBCDD content in products, since the content is substantially variable among products across China given various manufacturing technologies, fireproof endurance rating (FER) requirements and modes of application (Beijing Institute of Technology 2011). Previous studies (Abbasi et al. 2014; Managaki et al. 2009; Morf et al. 2007; Morf et al. 2008) collected information from multiple available sources and assembled individual “crisp” point estimates without considering their uncertainty intervals and fuzziness. Such a patchwork of various information, however, leads to misclosures of estimates (Müller et al. 2014) because these “snapshots” of realistic uses and emissions from individual specific angles are often random.

Recently, dynamic substance flow analysis (DSFA) (Allesch and Brunner 2015; Baccini and Brunner 2012; Müller et al. 2014) has been adopted as an appropriate tool to characterize the time-dependent changes in emissions of the problematic chemicals such as PBDEs and HBCDD (Abbasi et al. 2014; Morf et al. 2008; Morf et al. 2005; Vyzinkarova and Brunner 2013). To describe the long-term emissions of HBCDD under constraints of data uncertainties, we implemented two novel modifications to the traditional DSFA: (i) we established a scenario-based DSFA to enable evaluating the effects of different end-of-life management options on the past and future emissions of HBCDD from anthropogenic in-use and waste stocks; and (ii) we incorporated interval linear programming, which is an uncertainty optimization technique (Huang et al. 1992; Huang et al. 1995), into the scenario-based DSFA to adjust misclosures and to reconcile inconsistent estimates as compatible and reliable input parameters. Our intentions were to respond to the imminent concerns about end-of-life treatment of HBCDD and long-term emissions of a wider range of CiPs. Furthermore, since most of the 179 parties to the Stockholm Convention have just started to develop inventories and management strategies of HBCDD stocks, we hope that the case study provides a methodological foundation for other countries in particular developing and transition countries.

2. Methods and data

2.1. Production and consumption of HBCDD and associated products in China

Historical annual production data of HBCDD in China were compiled and presented in Fig. 1 (Beijing Institute of Technology 2011; Peking University 2012; and statistics courtesy of the China Flame Retardant Society). Given that (i) China is likely to register an at least five-year specific exemption under the Stockholm Convention to continue its HBCDD production, and (ii) market shares of alternative chemistries are projected to increase in the near future, we tentatively assumed that the HBCDD production would continue but linearly decrease in the five years following China's ratification of the HBCDD amendment to the Stockholm Convention that is expected in 2016. The HBCDD stockpile awaiting for sale is negligible nationwide, as most Chinese producers manufacture HBCDD in a build-to-order manner. Customs records demonstrate that HBCDD has never been imported as neither pure chemical nor formulation to China; export volume accounted for on average 37% of the annual HBCDD production before 2012 (Beijing Institute of Technology 2011), which has declined since 2013 to almost zero by 2015 because an increasing number of previous importers have voluntarily restricted their

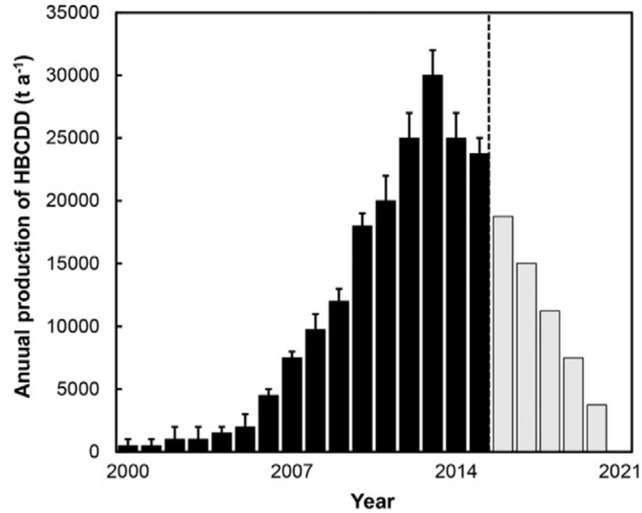

Fig. 1. Historical and assumed future annual production of HBCDD in China.

HBCDD trade (Z. Zhou, personal communication, June 11th, 2015). There are no records of imported HBCDD-containing products such as FR-EPS/XPS insulation boards or textiles, but only 4% exported FR-EPS insulation boards (Peking University 2012), because EPS and XPS have low densities and are normally produced in the country of use.

Previous Chinese market surveys demonstrate that HBCDD has been used in two sectors to produce three major end-products: (i) <2% of annual domestic trade was used in FR polyester fabric or textiles before 2009, but this usage has subsequently been almost completely replaced by other lower-priced flame retardants (Beijing Institute of Technology 2011); (ii) the rest of annual domestic trade went to FR-EPS and FR-XPS insulation boards which were used in external insulated composite systems (EICSs). In China, HBCDD has not been used in (i) furniture or virgin polystyrene packaging materials due to the absence of mandatory FER code for these items, or (ii) high impact polystyrene (HIP) in electrical and electronic equipment due to its price disadvantage over other flame retardants (Beijing Institute of Technology 2011); these situations are in contrast to cases in other countries (e.g., POPRC 2011; Rani et al. 2014).

2.2. Principle equations of DSFA

Fig. 2 illustrates the anthropogenic cradle-to-grave processes of HBCDD and associated end-products, including *Production*, *Industrial Processing* (processing EPS, XPS insulation boards and textiles using HBCDD), *Consumer Use*, as well as *Waste* (including *Demolition* and *Disposal*). Here, we discriminated two categories of processes: in a *transient* process, the apparent residence time of HBCDD is shorter than the calculation time step of a year so that no HBCDD accumulates within the process; while in a *continuous* process, the apparent residence time is longer than a year and we use a time-variant state variable, *stock*, to represent the stopover and storage of HBCDD within the process. Individual processes are connected through dynamic HBCDD *flows* (mass per time) that are visualized as lines with an arrow from the output process to the input process.

The HBCDD stock in a given process i at a certain time t, $M_i(t)$, is determined by related flows using the following differential mass-balance equation (Baccini and Brunner 2012; Brunner and Rechberger 2004; Müller et al. 2014):

$$\frac{\mathrm{d}M_i(t)}{\mathrm{d}t} = \underbrace{I_i(t)}_{\text{inflow}} \underbrace{- I_i(t) \times \sum_{j=1}^{p} TC_{i\to j}(t)}_{\text{outflow}} \underbrace{- M_i(t) \times \sum_{k=1}^{3} EF_{i\to k}(t)}_{\text{emissions}} - \underbrace{WS_i(t)}_{\text{waste}} - \underbrace{DEG_i(t)}_{\text{degradation}} \quad (1)$$

where,