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Emission inventory and environmental distribution of decabromodiphenyl ether in China

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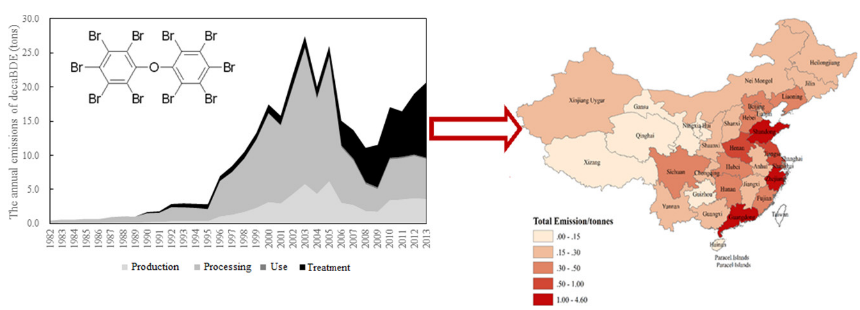
HIGHLIGHTS

- The annual total emissions showed a fluctuating trend, with a steady growth from 1982 to 2003.
- Emissions were mainly concentrated in Guangdong, Shandong and Zhejiang provinces, with emissions of 4.5, 4.3 and 4.1 t in 2013.
- Soil and sediment were the main sinks of decaBDE, accounting for 99.8% of the total concentration.

GRAPHICAL ABSTRACT



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ABSTRACT

Decabromodiphenyl ether (decaBDE) is a highly brominated flame retardant that recent studies have identified as a potential persistent organic pollutant. Large amounts of decaBDE have been consumed and released in the environment in China, while no emission inventory has been available until now. In this study, a substance flow analysis was applied to establish the emission inventory of decaBDE in China from 1982 (the first year of decaBDE production in China) until 2013 based on activity data, transfer coefficients, and emission factors. The results show that the stock of decaBDE continually increased, reaching a peak of 290,000 tons in 2007. The annual processing capacity of decaBDE also increased, and the processing capacity in 2013 was 49,000 tons. Historical accumulative emissions were estimated to be 313.3 tons from 1982 to 2013, and the annual emissions peaked in 2003 at 27.5 tons. On average, decaBDE processing was the major source (58.4%) of total emissions, followed by treatment, production, and usage processes. From 1982 to 2013, decaBDE was released mainly into water sources, accounting for 50.7% of the accumulative emissions. At the provincial level, Guangdong, Shandong, and Zhejiang provinces were the largest producers in China. Simulations produced by the level III fugacity model showed that the projected concentration was very consistent with the measured value. The stock of decaBDE in the soil and sediment phases accounted for 99.8% of the total stock, and the transfer among the four environmental phases occurred mainly at the atmosphere–soil interface.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are a group of bromine-containing organic compounds often used as brominated flame retardants (BFRs). Among BFR mixtures, the most widely used commercial additives are pentabromodiphenyl ether (pentaBDE), octabromodiphenyl ether (octaBDE), and decabromodiphenyl ether (decaBDE). Due to their persistence and toxicity, commercial pentaBDE and octaBDE were listed in Annex A (i.e., prohibition of production) of the Stockholm Convention on Persistent Organic Pollutants (POPs) (COP.4, 2009a; COP.4, 2009b) in 2009, but decaBDE is still widely used. Recent studies have found that decaBDE has several characteristics of POPs, and can be degraded into pentaBDE and

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octaBDE. The European Union, United States, and several other countries have introduced restrictions or prohibitions of the use of decaBDE (VECAP, 2007).

There is currently much ongoing research regarding environmental concentrations of decaBDE, and several countries have already established emission inventories. Lassen et al. (1999) used a substantial flow analysis (SFA) to estimate the annual emissions of BFRs in Denmark in 1997. Based on this preliminary study, Palm et al. (2002) calculated PBDE emissions and the per capita annual emissions of decaBDE. Sakai et al. (2006) established the decaBDE annual atmospheric emission inventory in Japan (Sakai et al., 2006). In the same year, Yamaguchi et al. (2006) calculated decaBDE emissions to the atmosphere, soil, and water in Japan from 1978 to 2015. Since then, decaBDE emissions inventories have been established for Switzerland (Morf et al., 2007), Sweden (Björklund et al., 2012), and the United States (US EPA, 2010).

In recent years, studies conducted in China have also examined decaBDE emissions. For example, Ni et al. (2013) estimated the mass flow of decaBDE during the treatment of waste electrical and electronic equipment (WEEE). In addition, Tien et al. (2012) used an SFA to analyze the flow of PBDEs during the informal processing of WEEE in China in 2005 (Tien et al., 2012). However, these studies have mainly focused on decaBDE emissions from WEEE processing, with no research at the national level from a full life cycle perspective.

In October 2015, decaBDE was added to Annex F (risk management assessment) by the Persistent Organic Pollutants Review Committee, resulting in a recommendation for the Convention's Conference of Parties to consider listing it in Annex A in 2017. China is one of the largest producers of decaBDE in the world. Once decaBDE is listed in Annex A, as a party of the convention, China will be required to restrict its production and use, identify decaBDE stocks and waste, and reduce emissions from the existing stock. Therefore, there is an urgent need for the world to establish a decaBDE emission inventory for China.

In this study, based on activity data, transfer coefficients, and emission factors, SFA was applied to establish China's emission inventory of decaBDE from 1982 (the first year of decaBDE production in China) until 2013. The stock and main release processes were identified, and the emissions were allocated at the provincial level. The Mackay level III fugacity model was then applied to simulate the environmental distribution of decaBDE in Beijing. The predicted concentration was compared to measured environmental data from the literature. In addition, the environmental transfer and transformation behavior of decaBDE was simulated to provide a foundation for environmental and human health risk evaluations.

2. Methods

2.1. Substance flow analysis

DecaBDE is a potential POP. Most POPs are used as active ingredients in end products, meaning that emissions occur not only from the production process, but also from the use and disposal of the products. Therefore, in this study, an SFA was used to calculate decaBDE emissions. SFA is a method to describe or analyze the input, output, and flow processes of a substance in a system and is an important tool for quantitatively analyzing the environmental effects of a substance during its life cycle (Udo de Haes et al., 1988). The SFA method is widely used in the circular economy, industrial ecology, and other fields. In recent years, the SFA has also been used to establish POP emission inventories.

In this study, the system was defined as the entire life cycle (production, processing, use, and treatment) of decaBDE in China from 1982 until 2013 and was divided into 10 process units: production, processing, use, WEEE formal processing, WEEE informal processing, recycling, incineration, landfill, wastewater, and sewage treatment plants (labeled from 1 to 10, respectively). In the environmental system, atmosphere is represented as a, water is w, soil is s, and 0 is defined as outside the boundary. Fig. 1 shows the flow of decaBDE between the various process units and emissions into the environmental media. The output of the process unit consists of emissions into the environment and transfer to the next unit. The emission factors are represented as $E_{i\text{-}a}$, $E_{i\text{-}w}$, and $E_{i\text{-}s}$ (where $i = 1$–10); transfer coefficients between the process units are represented as $f_{i\text{-}j}$ (where $i, j = 0$–10).

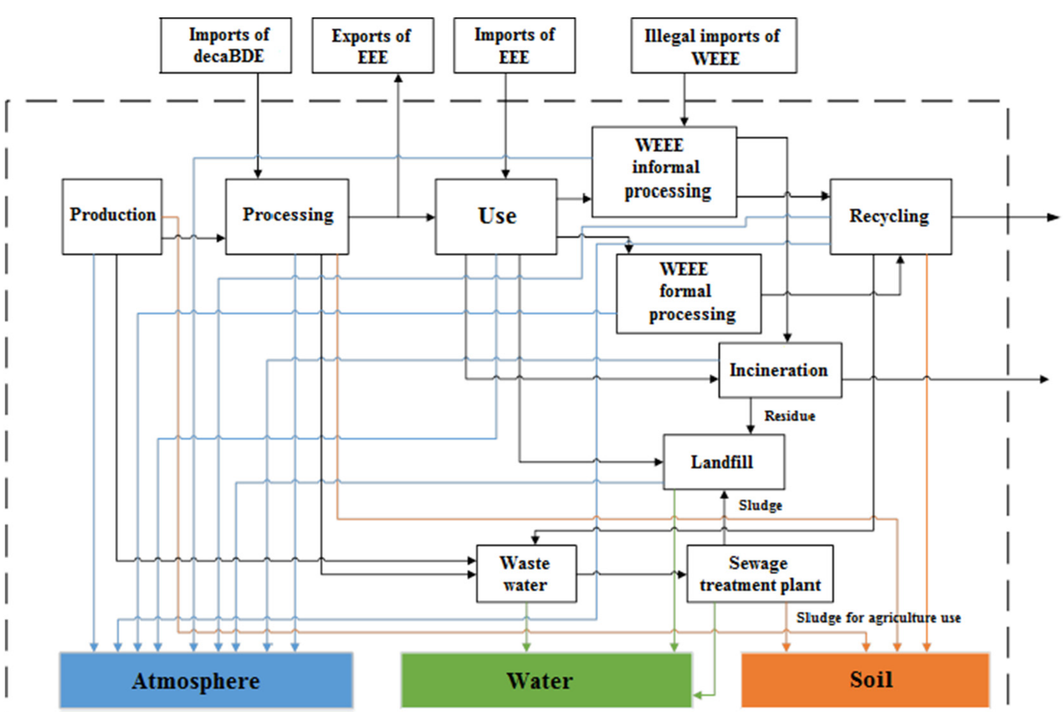

Fig. 1. Substantial flow analysis (SFA) system of decabromodiphenyl ether (decaBDE) in China.