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Estimating European historical production, consumption and atmospheric emissions of decabromodiphenyl ether

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HIGHLIGHTS

- Total DecaBDE consumption in Europe for the period 1970–2010 is estimated to be between 185,000 and 250,000 tonnes.
- European atmospheric emissions of BDE-209 is predicted to peak in 2004 at 10 tonnes/year.
- The waste management phase is responsible for the majority of BDE-209 environmental emissions.
- The volume of BDE-209 present in the anthroposphere is declining and is predicted to fall to negligible levels by 2030.

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ABSTRACT

A European scale production, consumption and environmental emissions inventory is produced for decabromodiphenyl ether (DecaBDE) for the period 1970–2020. A dynamic substance flow analysis model of DecaBDE is developed and emission of the main congener, BDE-209, to environmental compartments is estimated. From 1970 to 2010, it is estimated that a total of 185,000–250,000 tonnes of DecaBDE was consumed in Europe. Consumption peaked in the late 1990s at approximately 9000 tonnes/year and has declined by ~30% in 2010. Predicted BDE-209 atmospheric emissions peak in 2004 at 10 tonnes/year. The waste management phase of the BDE-209 life cycle is responsible for the majority of atmospheric emissions via volatilisation and particle bound emissions from landfills, whilst leakage from Sewerage systems is the major source of emissions to the hydrosphere. Use of sewage sludge from wastewater treatment works as an agricultural fertiliser is the most important pathway of BDE-209 to soil. Although DecaBDE consumption has declined in recent years, the stock in use for 2010 remains considerable (60,000 tonnes) and is likely to act as a source of atmospheric emissions for several decades. Uncertainties exist in these estimations and more field or experimental data is needed to clarify the significance of certain emission pathways, in particular, emissions from landfill sites.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) are brominated flame retardant (BFR) chemicals used in a range of applications to prevent or slow combustion in the event of a fire. PBDEs are added to foams, plastics and textiles to be used in construction, automobiles, electronic and electrical (E&E) equipment (BSEF, 2010). The three most commonly used commercial PBDE mixtures (c-PentaBDE, c-OctaBDE and c-DecaBDE) have been produced in large volumes with recent global production rates of more than 50,000 tonnes/year (BSEF, 2010). Historically, c-DecaBDE has been produced in volumes larger than any other commercial PBDE (BSEF, 2010). The congener BDE-209 is the major constituent of c-DecaBDE (La Guardia et al., 2006). PBDEs have been found to be widespread in the European environment (Arellano et al., 2011; Hale et al., 2006) and biota (de Wit et al., 2006; Munoz-Arnanz et al., 2011) and pose a potential risk to animals and humans (Darnerud, 2003). BDE-209 concentrations in the blood of children aged 9–11 years old of Baden-Württemberg, Germany have shown a slight increase during the 2000s, whilst concentrations of lower brominated PBDEs have decreased (Link et al., 2012). This suggests BDE-209 is becoming as environmentally relevant as some of the other lower brominated PBDE congeners and is an increasing concern for human health.

1.1. European and UK regulation of DecaBDE

Directive 2002/95/EC (RoHS Directive) on the restriction of the use of certain hazardous substances in E&E equipment specifies that from July 2006 new E&E equipment should not contain any PBDEs, although it was decided that c-DecaBDE be added to the list of exemptions in 2005 (European Court of Justice, 2005). That decision was annulled by the European Court of Justice and as of July 2008, c-DecaBDE can no longer be used in E&E equipment (European Court of Justice, 2008) and major producers in the US have announced they will discontinue c-DecaBDE production and use by 2013 (US EPA, 2010).

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C-DecaBDE was exempted from amendments to the Stockholm Convention on Persistent Organic Pollutants (POPs) as it did not meet the persistency criteria for a POP (European Court of Justice, 2008). There is now growing evidence that suggests BDE-209 can degrade by debromination to lower-brominated congeners which are classified as POPs (Zeng et al., 2010). Given the large volumes of c-DecaBDE produced in the last few decades, this could be a significant source of lower brominated congeners after release into the environment as Schenker et al. (2008) estimated by the degradation of BDE-209. The Voluntary Emissions Control Action Programme (VECAP), which is supported by the European Brominated Flame Retardants Industry Panel (EBFRIP) and the Bromine Science and Environmental Forum (BSEF), began in 2004 and aims to reduce production and processing emissions from industry. The programme has achieved significant reductions (~60%) in environmental emissions of c-DecaBDE in just 3 years (VECAP, 2010).

No European scale estimates of c-DecaBDE consumption or emissions have been made to date. Therefore, the aim of this study is to produce an estimate of European consumption, use and environmental emissions of the main congener BDE-209, for 1970–2020 and consider future emissions under various scenarios. Estimating the major sources and emission pathways of BDE-209 to the environment is valuable for assessing risk and can help identify the major uncertainties in the fate and behaviour of the chemical. To achieve this, a discrete dynamic substance flow analysis model is developed and applied to improve on the methods used in similar studies for other POPs (Paul et al., 2009; Prevedouros et al., 2004a; Schenker et al., 2008) which after modelling underestimated environmental concentrations. These estimates will also provide input for a multimedia environmental fate model (EVn-BETR region presented in Fig. A1) in a future study and are specifically, though not exclusively, designed to facilitate this purpose.

2. Methods

2.1. Historical European consumption of c-DecaBDE

No data for the production rates of c-DecaBDE in individual European countries are currently available. Consequently, we use other available data from BSEF (2010), VECAP (2010) and the European Chemicals Bureau (ECB, 2003) on Global and European industrial consumption of c-DecaBDE (presented in Table A1). Global industrial consumption of c-DecaBDE by BSEF members has increased from 30,000 tonnes/year in the early 1990s to a peak of approximately 66,000 tonnes/year in 2002 (BSEF, 2010). European industrial consumption peaked during the 1990s at 9100 tonnes/year and has fallen slightly during the 2000s to between 6900 and 8600 tonnes/year (BSEF, 2010; VECAP, 2010). Future demand is expected to decrease as c-DecaBDE is phased out and manufacturers switch to using alternative flame retardants, a process which has already begun (US EPA, 2010). Using the BSEF and VECAP consumption figures gives a total consumption of approximately 150,000 tonnes for 1991–2010.

2.2. Estimated European consumption

Estimates are derived from global and European industrial consumption data. For 1991–2005 BESF figures are used and for 2008–2010 VECAP figures are used (Table A1). For the missing years, 2006 and 2007, no data is available and linear interpolation is used. The VECAP figure for 2008 is likely an underestimate as coverage was incomplete (VECAP, 2010). The European Commission (ECB, 2003) figures are not used in the estimates as only 2 figures are presented. For the period prior to 1991 no data is currently available. C-DecaBDE production likely started at the same time as the other PBDEs in the mid 1960s. Therefore, we assume c-DecaBDE production and consumption began in 1970 for all estimates (Alcock and Busby, 2006; Prevedouros et al., 2004a). Estimated consumption rates from the following scenarios are presented in Fig. 1.

2.2.1. C-DecaBDE consumption scenarios

2.2.1.1. Scenario C0. The trend in global c-DecaBDE consumption is estimated using the available global data and is used as a surrogate for the trend in European consumption. Scenario C0 uses the available, yet limited, global industrial consumption data from BSEF (2010) (Table A1). Linear interpolation is used between 1969 (0 tonnes/year) and data points for 1991, 1999 and 2001, and consumption is assumed to be constant from 2004 to 2010 at the 2003 figure of 56,000 tonnes/year. This scenario gives a total global c-DecaBDE consumption for 1970–2010 of 1.3 million tonnes. For the years 1999 and 2001–2003 both global and European c-DecaBDE consumption data is available (BSEF, 2010). According to this data, from 1999 to 2003, Europe had a mean 14% (±2%) share of the global c-DecaBDE market. Assuming the European market share was a constant 14% from 1970 to 2010, total European consumption for the period is estimated at 185,000 tonnes in this scenario. However, it is possible that the European market share of c-DecaBDE has been different in the past than estimated in this scenario, although a lack of comparative data prevents confirmation of this.

2.2.1.2. Scenarios C1 and C2. Scenarios C1 and C2 estimate European consumption using the BSEF and VECAP European consumption data. Both scenarios use the combined BSEF and VECAP data post-1991 and use interpolation between 2005 and 2008 for the missing data of 2006 and 2007. C1 uses linear interpolation from 0 tonnes/year in 1969 to 8900 tonnes/year in 1992; C2 uses linear interpolation from 0 tonnes/year in 1969 to 4200 tonnes/year in 1991, calculated from the BSEF global figure for 1991 (30,000 tonnes/year) and assuming European consumption was equal to a constant 14% of the global market. Total European consumption for 1970–2010 is estimated at 250,000 and 200,000 tonnes for scenarios C1 and C2 respectively.

2.2.1.3. Scenario C3. Scenario C3 is derived from global bromine production using the data for 1970–2005 compiled by Schenker et al. (2008). This scenario uses world bromine production data, % BFR production, % individual PBDE production and c-DecaBDE % share of PBDEs to estimate world c-DecaBDE production (see Schenker et al., 2008 for more details). We then assume that the European market share has been a constant 14% to estimate European c-DecaBDE production. For 1970–2005 global consumption is estimated at 0.4 million tonnes and 1 million under scenarios C3 and C0 respectively. For the same period, European consumption is estimated at 56,000 and 145,000 tonnes under scenarios C3 and C0 respectively. Scenario C3 estimates consumption at levels several times lower than the other scenarios. This approach has been used by previous studies (Prevedouros et al., 2004a; Schenker et al., 2008) which underestimated modelled environmental concentrations. In their supporting information Schenker et al. (2008) present an additional high-emission scenario in which PBDE production is increased by a factor of three, leading to better agreement of modelled and measured concentrations. Therefore, estimating consumption using this approach leads us to question the validity of using bromine production as a surrogate for c-DecaBDE consumption without further modification. Of the four c-DecaBDE European consumption scenarios only C1 and C2 are used to derive emission estimates.

2.3. BDE-209 consumption scenarios

Developing scenarios C1 and C2 further, the fraction of the congener BDE-209 in c-DecaBDE mixtures is examined. Early mixtures of c-DecaBDE contained a lower fraction of BDE-209 than those of present. Analysis shows that several different c-DecaBDE mixtures contained BDE-209 at a fraction of >90% w/w, the remainder made up of Nona and OctaBDEs, mainly BDE-206 (La Guardia et al., 2006) (Fig. A2). Only in 1995 did manufacturers commit to a BDE-209 purity of >97% in their c-DecaBDE mixtures (OECD, 1995). The Bromkal mixtures 79-8DE and 82-0DE were produced in Europe and the other mixtures