GEO PARTNER AG RESOURCE MANAGEMENT

NRP50

Endocrine Disruptors: Relevance for Humans, Animals and Ecosystems

Dynamic Substance Flow Analysis Model for Selected Brominated Flame Retardants as a Base for Decision Making on Risk Reduction Measures (FABRO)

Final Report

Authors

Dr. Leo Morf, Andreas Buser, Ruedi Taverna

GEO Partner AG Resource Management, Zurich

In collaboration with:

Dr. Hans-Peter Bader, Ruth Scheidegger

Department of System Analysis, Integrated Assessment and Modelling, Eawag, Dübendorf

Table of contents

1 Introduction

1.1 Initial situation and aims

Up to now, limited research on the fate, occurrence and emissions of brominated flame retardants (BFRs) during use and disposal activities was done. But the knowledge of anthropogenic substance flows and stocks, such as potential stocks in private households, in waste management and of potential hazardous residual flows into the environment, is essential for early recognition of environmental impact and effective chemicals management. Dynamic substance flow analysis (SFA) based on field data served as a valuable tool to estimate the flows, stocks and emissions.

This project aimed to determine the substance flows inside the anthroposphere and from the anthroposphere to the environment. Therefore, a dynamic model to investigate the behaviour of the anthropogenic metabolism was developed. The model was based on an existing SFA for Switzerland covering the whole life cycle of BFR (Morf et al., 2002). Up-to-date research results and information about the occurrence and fate of the selected substances in the anthroposphere and emissions into the environment have been incorporated. The results of the dynamic SFA have been compared to measurements in environmental compartments and outcomes of other investigations. One of the major goals of this project was to elaborate recommendations for future regulatory, technical and other measures to reduce the risk for humans and the environment regarding these potential endocrine disrupters.

1.2 Objectives

The first main objective was to determine the substance flows and stocks in the anthroposphere and emissions from the anthroposphere to the environment for selected BFRs applying a dynamic substance flow analysis model. The goal was to improve the anthropogenic system part in the SFA by Morf and co-workers (2002) and to include all suitable data that are available in order to get a most accurate system and to appropriately model temporal trends of BFR-flows and stocks in the anthropogenic system and substance transfers into the environment.

The second main objective was to link the results of the anthropogenic metabolism with environmental occurrence and fate results. In order to get access to new results and to benefit from their know-how, this task has been carried out in cooperation with other NRP50 projects and other research groups focusing on the investigation of the environmental fate.

The goal was to find explanations for the occurrence and the fate of BFRs in the environment and to determine relevant emission sources. Thus, it has been tested if the data available on emissions from different processes are appropriate to estimate the emissions of the BFRs to the environment or if there are still major data gaps.

2 Methodical procedure

2.1 System analysis

In the system analysis the system is defined, namely its borders, the balance volumes and the flows in between.

System analysis involved three steps:

- limitation of the system in a geographical boarder and the investigated time period
- establishment of the internal structure of the system: choice and definition of processes and their input and output goods, and determination of flows
- definition of the substances to be studied

2.2 System definition

The system comprises the transport and transformation processes of three selected BFRs in the Swiss anthroposphere.

2.2.1 Selected application areas

The different goods containing BFRs have been split up in four application areas:

- Electrical and electronic equipment (E&E)

  E&E compromises all kind of electrical and electronic equipment such as consumer electronics, household appliances, industrial appliances and machines.

- Transport

  Transport covers vehicles such as passenger cars, buses, lorries, ships and airplanes as well as parts of vehicles.

- Textiles/furniture

  Textiles/furniture (sometimes abbreviated as *textile* or *tex*) compromises flame retarded textiles and furniture such as curtains, carpets, sofas and other upholstered furniture.

- Construction

  Construction contains all plastic materials used in the construction sector, e.g. insulation panels, plastic sheeting or vapour barriers.

2.2.2 Selected subsystems

The system was divided into the three subsystems production and use, waste management and environment. In chapter 3.2, the process production is treated separately in the emission figures that are split up by the different sources.

Table 1 shows the three subsystems comprising the selected processes. For construction applications, the processes construction and deconstruction are part of the system, too. The environmental compartments atmosphere, hydrosphere and soil were outside the system boundary.

Table 1 Subsystems of the model

		E&E	Transport	Textile	Construction
Production and use	Production	x	x	x	x
	Trade	x	x	x	x
	Construction				x
	Use (Stock)	x	x	x	x
	Deconstruction				x
Waste management	Recycling	x	x		x
	Incineration	x	x	x	x
	Sewerage	x	x	x	x
	WWTP	x	x	x	x
	Landfill (Stock)	x	x	x	x
Environment	Atmosphere	x	x	x	x
	Hydrosphere	x	x	x	x
	Soil	x	x	x	x

2.2.3 Definition of the processes and flows

2.2.3.1 Processes

In table 2, the ten selected processes in the system are defined. The processes *use* and *landfill* include a stock where the substances may accumulate. It has been assumed that the other processes did not include a stock, because the residence times are much shorter and can be neglected in a first-order approximation.

Table 2 Definition of the processes inside the system boundary, the environmental compartments and the export

Production	1	The process comprises the production of semi-finished (e.g. moulded) parts and finished products (i.e. construction materials, textiles, E&E and transport appliances) that contains the BFRs under study.
Trade	2	The process comprises the trade in finished products (e.g. E&E products, construction materials, vehicles) that contains the BFRs under study. Sales divisions of the industry are counted among, too.
Construction	3	The process includes the construction operations where plastic sheeting, insulation boards or foams containing BFRs are used.
Use (Stock)	4	The process comprised the consumption and the stock of consumer products, household and industrial appliances as well as vehicles, textiles, furniture and construction materials that contained the BFRs under study.
Deconstruction	5	The process includes the mechanical and manual deconstruction operations on site.
Recycling	6	The process comprises the mechanical and manual treatment of goods in recycling facilities (e.g. e-waste recycling, car recycling …).
Incineration	7	The process comprises the incineration processes in municipal waste incineration plants, sludge incineration plants, fluid bed incineration and cement works.

Sewerage	8	This process comprises the sewer system in which wastewater is collected, transported and delivered to the WWTP.
WWTP	9	The process comprises treatment of municipal wastewater in wastewater treatment plants in Switzerland. It contains wastewater from private households as well as from production sites and surface runoff.
Landfill (Stock)	10	The process comprises landfills where waste that contains BFRs under study is disposed.
Atmosphere	11	The compartment comprises an air volume of a height of 1000 m and the area of Switzerland.
Hydrosphere	12	The compartment comprises the surface water in surface water in rivers and lakes including sediment and aquatic organisms.
Soil	13	The compartment comprises the soil in Switzerland with a mean thickness of 10 cm.
Export	14	The export is defined as countries abroad to which new and waste goods are exported.

2.2.3.2 Flows

The flows used in the model can be split up in three groups. The input flows, where the substances were either imported from abroad or deposited from the atmosphere, are described in table 3. The internal flows are described in table 4, whereas the output flows (i.e. either export to abroad or emissions into the environment) are defined in table 5. The variables are labelled according to the process number of the starting and the end point, e.g. f_{1-2} for the substance flow from *production* to *trade*. A superscript number is added in case of multiple flows with the same starting and end point.

Table 3 Definitions of input flows

From	To	Variable	Description
Abroad	Production	i_1	Import of substances, masterbatches and semi-finished goods to be finalised in Switzerland
Abroad	Trade	i_2	Import of finished goods to Switzerland
Atmosphere	Sewerage	i_8	Atmospheric deposition of BFRs entering the sewer system with the storm water surface runoff

Table 4 Definitions of the internal flows

From	To	Variable	Description
Production	Trade	f_{1-2}	Delivery of semi-finished and finished products to retail market
Production	Incineration	f_{1-7}	Production waste being incinerated
Production	Sewerage	f_{1-8}	Production wastewater entering the sewer system
Trade	Construction	f_{2-3}	Delivery of construction materials to construction sites
Trade	Use	f_{2-4}	Delivery finished products to private households, enterprises and industry
Construction	Use	f_{3-4}	Construction material being built-in to buildings

Construction	Incineration	f_{3-7}	Construction waste being incinerated
Construction	Landfill	f_{3-10}	Construction waste being disposed on landfills
Use	Deconstruction	f_{4-5}	Construction materials in buildings reaching end of service life and being deconstructed
Use	Recycling	f_{4-6}	Materials reaching end of service life and being recycled
Use	Incineration	$f_{4-7}^{(1)}$	Vacuum cleaner dust being incinerated
Use	Incineration	$f_{4-7}^{(2)}$	Materials reaching end of service life and being incinerated
Use	Sewerage	$f_{4-8}^{(1)}$	Mop and cleaning water entering the sewer system
Use	Sewerage	$f_{4-8}^{(2)}$	Wash water originating from textiles washing entering the sewer system
Use	Landfill	f_{4-10}	Materials reaching end of service life and being disposed on landfills
Deconstruction	Recycling	f_{5-6}	Demolition waste being recycled
Deconstruction	Incineration	f_{5-7}	Demolition waste being incinerated
Deconstruction	Landfill	f_{5-10}	Demolition waste being disposed on landfills
Recycling	Production	f_{6-1}	Reuse of recycled material in production
Recycling	Incineration	f_{6-7}	Fractions from appliance recycling being incinerated
Recycling	Sewerage	f_{6-8}	Wastewater from recycling facilities entering the sewer system
Recycling	Landfill	f_{6-10}	Fractions from appliance recycling being disposed on landfills
Incineration	Landfill	$f_{7-10}^{(1)}$	Waste incineration plant air pollution control residues being disposed on landfills
Incineration	Landfill	$f_{7-10}^{(2)}$	Waste incineration plant bottom ash being disposed on landfills
Sewerage	WWTP	f_{8-9}	Wastewater transport through the sewer system to the WWTP
WWTP	Incineration	f_{9-7}	Sludge from WWTP being incinerated
WWTP	Landfill	f_{9-10}	Sludge from WWTP being disposed on landfills
Landfill	Sewerage	f_{10-8}	Leachates from landfill being caught in the sewer system

Table 5 Definitions of output flows

From	To	Variable	Description
Production	Atmosphere	o_{1-11}	Gaseous, dust bound or particulate atmospheric emissions from production processes
Trade	Export	o_{2-14}	Export of finished goods
Construction	Atmosphere	o_{3-11}	Gaseous, dust bound or particulate atmospheric emissions during construction operations
Use	Atmosphere	$o_{4-11}^{(1)}$	Gaseous or dust bound atmospheric emissions during use
Use	Atmosphere	$o_{4-11}^{(2)}$	Particulate atmospheric emissions during use

Use	Atmosphere	$o_{4-11}^{(3)}$	Atmospheric emissions caused by building or vehicles fires
Use	Hydrosphere	$o_{4-12}^{(1)}$	Particle loss and leaching from use entering the hydrosphere
Use	Hydrosphere	$o_{4-12}^{(2)}$	Fire water entering the hydrosphere
Use	Soil	$o_{4-13}^{(1)}$	Particle loss and leaching from use entering the soil compartment
Use	Soil	$o_{4-13}^{(2)}$	Fire water entering the soil compartment
Use	Export	o_{4-14}	Export of used goods (e.g. cars)
Deconstruction	Atmosphere	o_{5-11}	Gaseous, dust bound or particulate atmospheric emissions during deconstruction operations
Deconstruction	Soil	o_{5-13}	Particles loss to soil during deconstruction operations
Recycling	Atmosphere	o_{6-11}	Particulate or dust bound atmospheric emissions from recycling operations
Recycling	Export	o_{6-14}	Export of recycled material
Incineration	Atmosphere	o_{7-11}	Flue gas emissions from incineration
Incineration	Hydrosphere	o_{7-12}	Wash water from incineration entering the runoff ditch
Incineration	Export	o_{7-14}	Export of APC residues
Sewerage	Hydrosphere	o_{8-12}	Sewerage overflow entering the hydrosphere
Sewerage	Soil	o_{8-13}	Sewerage leaking into the soil
WWTP	Hydrosphere	o_{9-12}	Water from the WWTP entering the runoff ditch
WWTP	Soil	o_{9-13}	Sludge from WWTP being applied on farmland
Landfill	Atmosphere	$o_{10-11}^{(1)}$	Gaseous emissions from landfills
Landfill	Atmosphere	$o_{10-11}^{(2)}$	Particulate or dust bound atmospheric emissions during unloading of waste and operations on landfills
Landfill	Soil	o_{10-13}	Leachates from landfills not being caught in the sewer system

The processes and the flows used in the substance flow analysis are illustrated in the schemes in figure 1.

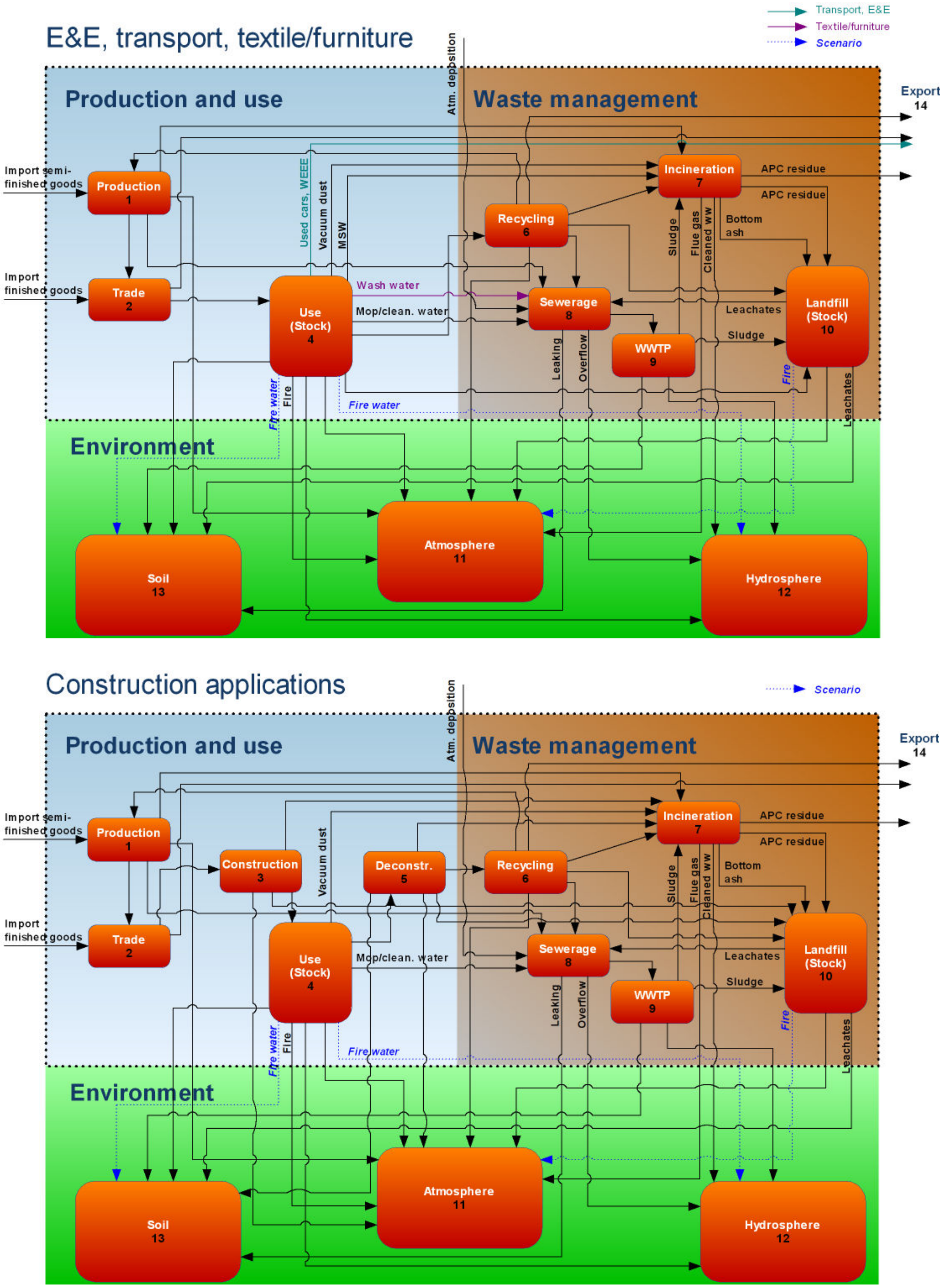


Figure 1 System for application areas E&E, transport and textile (above) und construction applications, including the processes *construction* and *deconstruction* (below). Note that due to space constraints and for purposes of clarity, not all flows are labelled in the figure. The description of the flows can be found in the section 2.2.3.2.

2.2.4 Geographical border and investigated time period

The geographical boundary of the system was identical to the political border of Switzerland. The temporal boundary of the system was the period from 1980 to 2020.

2.2.5 Selection of the substances to be studied

The selection was made in accordance to the risk for the humans, animals and the environment. DecaBDE, which has not been banned like the other polybromodiphenyl ethers OctaBDE and PentaBDE, was selected with the highest priority. DecaBDE has been exempted from the EU RoHS (Restriction of Hazardous Substances) Directive. The EU Parliament has strongly opposed the Commission's decision and has taken its case to the European Court of Justice to annul the DecaBDE exemption. The European Court of Justice is expected to issue a ruling by the end of 2007. The PBDEs are the most discussed BFRs in the scientific community and most studies focus on this group. From the lower brominated PBDEs, the congener BDE-47 was considered to be of highest importance. Firstly, this substance is normally found in the highest concentrations in the different environmental compartments (except of BDE-209). Secondly, the PentaBDE technical mixture containing BDE-47 was considered more interesting than the OctaBDE technical mixture, because of its more diverse use. The priority of HBCD was heightened, as increasing levels were found in the environment (e.g. in sediment cores of Lake Greifen and Lake Thun) and it has been the second most discussed BFR in the last year. On the other hand, TBBPA has been lowered in priority. According to the ongoing risk assessment reports, a lower risk from TBBPA has been assumed compared to HBCD. For TBBPA the data on levels in environment is scarce.

Overview of the selection of substances for quantitative modelling:

- **DecaBDE**: equated with the congener BDE-209 that is the main component of the DecaBDE technical mixture
- **HBCD**: sum of the α, β and γ isomers
- **BDE-47**: congener of most importance in the PentaBDE technical mixture. The consumption trend has been estimated for PentaBDE. As the emission factors are different for the individual congeners, the modelling has done with a single congener selected (i.e. BDE-47). As the percentage distribution of the congeners in the products are believed to remain approximately constant in the substance flows and the stocks in the anthroposphere, the flows and stocks of PentaBDE can be calculated from the results for BDE-47.

No modelling has been done for OctaBDE and TBBPA. However, some data that have been collected for these substances are presented in the report. The temporal trend of OctaBDE might be quite similar to the PentaBDE used in *E&E*, but with a higher consumption and a lower atmospheric emission factor from the use phase. The emissions of polybrominated dibenzo-*p*-dioxins and dibenzofurans resulting from thermal processes and impurities of BFRs (Weber and Kuch, 2003; Hanari et al., 2006), are not subject of the present study.

2.2.6 Modifications compared to the system in Morf et al., 2002

There have been major improvements of the system compared to the 2002 study. The main difference was that a dynamic approach was elaborated in this project in order to get a more comprehensive model to estimate time-dependent substance flows, stocks and emissions for the period from 1980 to 2020. The BFR flows have been split up in four application areas. This splitting allowed using different residence times, transfer coefficients and emission factors for each application area. Another advantage was that substance flows and emissions could be attributed to the different uses and scenarios could be applied for individual application areas.

The process *use* was modelled in more details than in the previous study. In order to account for waste and emissions generated during construction and deconstruction operations the two processes *construction* and *deconstruction* have been added for the corresponding application area. New transfers to the environment included to the model for these processes were atmospheric emissions for both and emissions to soil for *deconstruction*. A fraction was allocated as waste during construction. The same internal flows have been used from *deconstruction* as from *use* after end of life.

Also more detailed output flows from the process *use* have been considered. While in the previous study only outputs at end of life as well as releases to wastewater and atmospheric emissions have been taken into account, the following outputs during service life have been included:

- Atmospheric emissions (gaseous and dust bound)
- Atmospheric emissions (particulates, lost caused by deterioration)
- Atmospheric emissions (unintended fires)
- Emissions to hydrosphere (particulates, lost caused by deterioration)
- Emissions to hydrosphere (unintended fires, scenario)
- Emissions to soil (particulates, lost caused by deterioration)
- Emissions to soil (unintended fires, scenario)
- Releases to wastewater (washing of textiles)
- Releases to wastewater (cleaning and mop water as well as release of dust sticking at textiles during washing)
- Transport to MSWIPs (see glossary; dust in vacuum cleaner bags)

In waste management, the process *sewerage* was added in order to model overflows and leakages in the sewer system. From *recycling* a new flow was added to *production* to take into account a direct reuse of recycled materials containing BFRs for the production of new goods. This additional flow was used in the application area *construction*.

2.3 Mathematical model

A dynamic model to describe the system has been investigated. The model is based on the mathematical material flow analysis (MMFA). MMFA is the extension of the traditional material flow analysis (MFA) with modelling concepts. The MFA was introduced and adapted to regions in the 1980s by Brunner et al. (1990) and Baccini and Brunner (1991). MFA has been extended to MMFA by Baccini and Bader (1996). In the past ten years MMFA has been applied in many different studies in various fields. The model describes mathematically the phenomenological knowledge of the system. It is a generalisation of the models used to describe systems characterised by long residence times. This approach has been discussed intensively in the literature (e.g. Baccini and Bader, 1996). Mathematically, the dynamic of the system is buried in the structure and the type of equations and the so-called parameter functions. The model has been implemented in SIMBOX, a software that was developed at Eawag. The core of the model is as follows: The substances entering the *use* are described by their residence time. In use phase, a fraction of the substance is emitted to the environment, while the rest is distributed after the residence time to different output flows such as waste incineration or recycling. The other processes are described by time dependent transfer coefficients, related to the whole or part of the input. If possible, emission factors have been determined by using experimentally determined data or measured emission values (field data) including their uncertainty. In case of missing data, the emission factors have been calculated with balancing in- and output flows and stock changes in the concerned processes or best guess estimates have been applied.

2.3.1 Equations

The balance equations that served as a base for the system of coupled integro-differential equations with (time-dependent) parameter functions to be used in SIMBOX are listed in the appendix (section 7.1). In order to ensure that the model is not over-determined, some flows are defined as remainders of the sum of inflows minus the outflows from a specific process where no stock is defined. Parameters used in the equations are substance flows, transfer coefficients, proportion coefficients and emission factors (see section 2.4.2).

2.3.2 Uncertainty estimation

For most of the parameters used in the model, there were no data series, from which it would have been possible to calculate a domain of uncertainty. Therefore, best guess estimates had to be applied. The parameters used in the model can be split up in two groups in terms of uncertainty of data. The consumption figures, the transfer coefficients used in the anthroposphere and proportion coefficients have relatively low domains of uncertainty. Most of them range between 30% and 50% (expressed as relative width of a 90% confidence interval). On the other hand, the domains of uncertainty of emission factors and diffuse releases are very high. The reasons of these high uncertainties are highly variable or non available field data. There are for example emission chamber experiments carried out for TVs with PBDEs (Kemmlein et al., 2003; Hirai et al., 2006). The results range between different studies and also different runs by more than two orders of magnitude. Additionally, as no measurements were carried out e.g. for construction materials, assumptions had to be made. The largest proportion of emissions was found on chamber walls, on TV screens and on dust. The proportion of these emissions getting to atmosphere, vacuum cleaner bags, wastewater and washing water had to be estimated as well. Also emission measurements of point sources exhibited a high variance. The emission factors from plastics processing industry measured by the Japanese Ministry of the Environment ranged by two orders of magnitude (Sakai et al., 2006). Therefore, best guess estimates have been applied for the system parameters. These uncertainties do not have a normal distribution, but a right-skewed distribution. The 90% confidence intervals of these parameters have been assumed as $p / 10 < p < p \cdot 10$ for all emission factors and diffuse releases in order to achieve a better traceability. Therefore, a lognormal distribution has been expected to be most appropriate.

Due to the complex mathematical model based on coupled integro-differential equations, normal runs of the model lasted several hours. Therefore, a calculation of uncertainty has not been possible for the dynamic case, as the runs needed for executing Monte Carlo simulations would have taken approximately one month on the available PC. The uncertainty estimations have been carried out for the stationary case for that reason. As the domain of uncertainty is much larger for the emission factors and the transfer coefficients to wastewater compared with the domain of uncertainty for the large internal substance flows in the anthroposphere, it has been expected that the influence on the result would be acceptable. The uncertainty estimations have been carried out with @RISK software for the year 2005.

2.4 Data acquisition

Some of the parameters used in the model are different for every substance, each of the four application areas and varied as well between 1980 and 2020. Therefore, a great deal of data was required for this study. The data was gathered by reviewing the literature published. The literature search has been carried out mainly in the *Web of Science*, with *Google Scholar*, *Google* and in collaboration with researchers from Empa. Contacts have been established to research groups from all over the world, with the industry, organisations and authorities.

2.4.1 Contacts

As the study was highly depending on the amount and the quality of data, a large number of research groups, industrial research, organisations, industry and national authorities have been contacted. The topics information was asked for were:

- Consumption and trends (BFRs, plastics, consumer goods, E&E, construction materials, textiles and furniture)
- Emissions
- Waste management
- Fire statistics
- Levels in the environment
- Modelling

Unfortunately, it was not possible to get a reply of all contacts. An information would have been a big benefit, especially from the EPS Verband Schweiz (HBCD use in EPS and XPS, recycling of EPS and XPS) or from D. Borgnes and B. Rikheim (SFT Norway) concerning their measurements on emissions from MWSIP and a few others. A comprehensive list of all contacts is included in the appendix in section 7.2.

2.4.2 Data input for the model

2.4.2.1 Use figures

In order to determine use figures and trends, the following methods have been applied:

- Import and export statistics of different products such as TVs, computers, cars, sofas etc.
- Plastics consumption
- BFR usage statistics from the industry
- Measurements in waste streams (WEEE and ASR)
- Estimates for other countries
- Future trends

2.4.2.1.1 Import and export statistics

In collaboration with the Swiss customs authority, import and export statistics of more than hundred products have been compiled for each year from 1988 to 2005 (not available for earlier years). The estimation of the amounts of the different BFRs has been carried out using the following parameters and assumptions based on the work by Leisewitz and Schwarz

(2001) and Morf et al. (2002) and newer data available. The mass of the potentially BFR containing material has been estimated using these parameters for the different products:

- Weight per unit
- Printed circuit board area per unit
- Mass fraction of casing, cushioning or textiles per unit
- Mass fraction of small components in electrical und electronic appliances

Estimates for the mean concentrations of the different BFRs at the beginning and at the end of the 1990s have been made:

- Concentration of the individual BFRs in printed circuit boards (g/cm^2)
- Concentration of the individual BFRs in casings, cushioning, textiles and in small components

The flows of BFRs in products imported to Switzerland and from Switzerland to abroad have been estimated as described above. The domestic use of BFRs in production has been estimated relative to import and export, because the import and export figures of the chemical substances as such were not listed separately. The net import and Swiss production for the domestic market have been summed up to the consumption of BFRs in Switzerland. This approach was used for all applications except construction materials, where estimates based on plastics consumption was considered to be more precise.

The BFR contents in the consumer goods have been estimated based on estimates in Morf et al. (2002) and updated according to new knowledge available. They have been estimated for two points in time: the beginning and the end of the nineties. For the time in-between these two points, a continuous changeover of BFR contents has been assumed. As the contents and the types of products containing DecaBDE, OctaBDE and PentaBDE became lesser from the first to the second point, the consumption was not increasing, even though the volume of products containing it showed a small increase over this period (figure 2).

The domestic production has been estimated relative to the import figures (see also section 2.4.2.4.1). For IT, communications technology and consumer electronics, the fraction of the production in Switzerland has been estimated to be negligible, while for small and large household appliances, special appliances and small E&E components the domestic production has been estimated to be half of the import. As most of the vehicles and vehicle components are imported, a domestic production of 3% has been assumed.

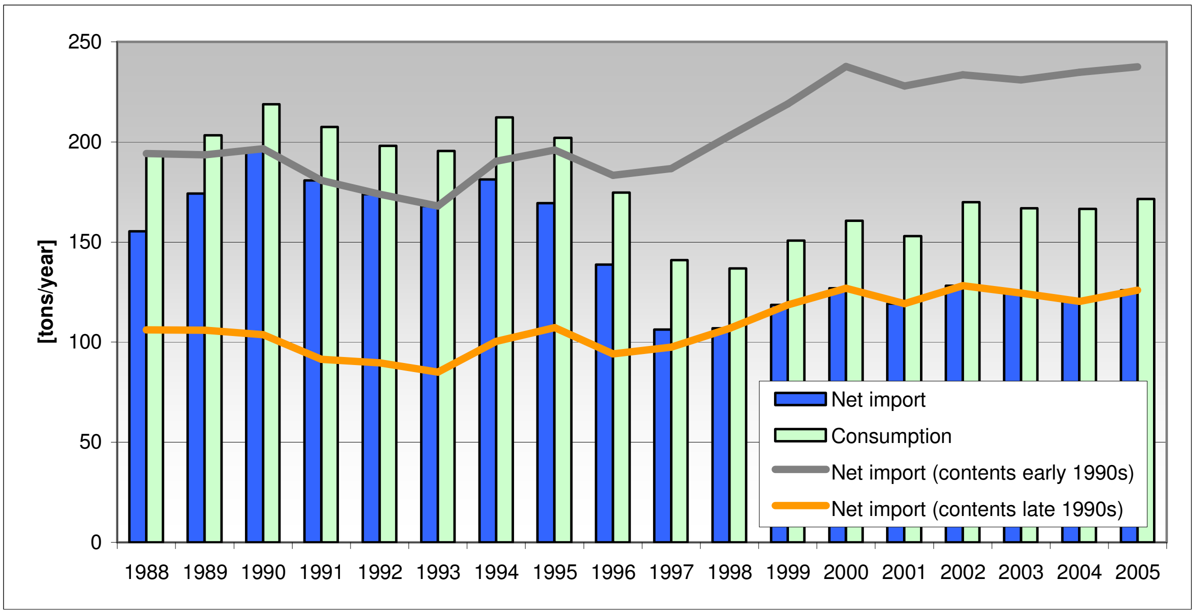


Figure 2 Net import and consumption of DecaBDE in consumer products (excluding construction materials)

2.4.2.1.2 Plastics consumption

An attempt was made to model the trend of BFR consumption in Switzerland. Data on plastics consumption were found in reports written by PlasticsEurope, APME or VKE. As figures for individual plastic types were only available for the years 2001–2004, the data of years 1985[1] to 2000 and future trends have been extrapolated by means of two methods.

In the first method, the expected world future trends for individual plastic types from 2004 to 2010 (PlasticsEurope, 2005) adapted for Europe and the annual growth rates calculated thereof have been used to estimate the consumption until the year 2020 starting from the figure of 2004. For the estimation of the historical consumption, only the total European consumption in 1990 was available. Hence, it has been assumed that the shares of the individual plastic types had remained constant, which was certainly not exactly true. The historical consumption back to 1985 has been calculated with the annual growth rate derived.

In the second method, the future trend and the historical consumption was estimated using the same annual growth rates as before, except for PP where data (Scheidl, 2006) were available. In contrast to the first method, the extrapolation has been made starting from the European consumption figures of the years 2001 to 2004 given by PlasticsEurope.

The Swiss plastics consumption has been calculated by multiplying the European consumption (i.e. EU15, Norway and Switzerland) of each plastic type by the factor 0.0188 according to the Swiss population. Then, each plastic type was allocated to the application areas *construction*, *transport* and *E&E* using the partition given in VKE (2002b) and PlasticsEurope (2004). The two methods to estimate the consumption described above and the two partition alternatives made up four series. Thereof, the minimum and maximum trend curves were considered for the individual plastic types and the different application areas. In the

---

[1] In the proposal, it was intended to include the period from 1985 to 2020 to the model at the time carrying out these trends. After starting the project an extension of the time period to 1980 has been thought to make sense. For these five years, the consumption has not been estimated based on plastics consumption, but on own assumptions.

application area *construction*, these figures could be compared with figures from a study on plastics in the Swiss building and construction industry (Schneeberger, 1999). Figures of plastics consumption in Western European automotive industry, electrical industry and in construction were available from APME and VKE (APME, 1995; APME, 2001; VKE, 2002a; VKE, 2003). They corresponded well with the figures calculated, however they were closer to the maximum estimates (upper trend line) for E&E and transport. In figure 3, the trends in the consumption of expanded and extruded polystyrene in the Swiss construction sector are shown as an example. As the trends derived from PlasticsEurope's data also contained other polystyrene polymers such as HIPS and there was no splitting in EPS and XPS possible, the trends derived from Schneeberger have been used (see figure 3). The figures for other plastic types in this sector are included in the appendix (section 7.3).

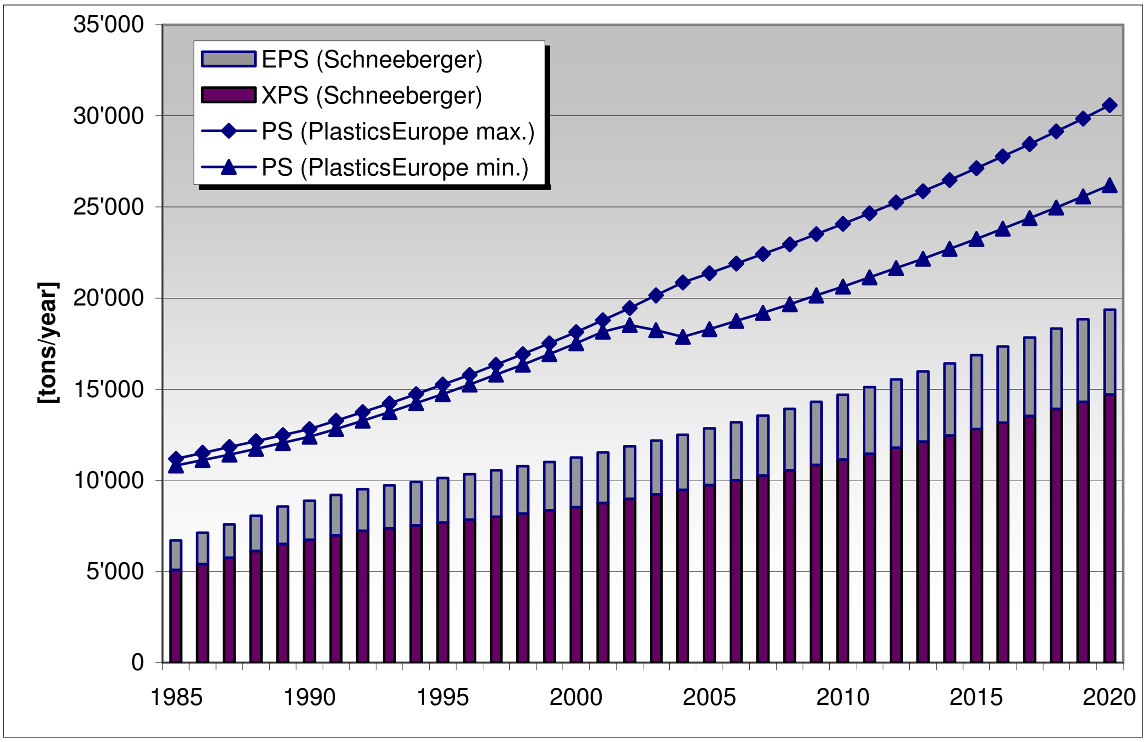


Figure 3 Consumption of expanded and extruded polystyrene in the Swiss construction sector (PS includes different polystyrene plastic types such as EPS, XPS and HIPS)

The Swiss consumption of brominated flame retardants has been estimated using data on

- the consumption of the individual plastic type,
- the fraction that was flame-retarded,
- the fraction of a specific flame retardant therein
- and the mean content of the single substances in the polymer.

The data on percentages and temporal trends for the second and the third factor were very limited and the domain of uncertainty was large. Hence, the resulting uncertainty was too large to get consumption figures with an acceptable uncertainty, except for construction materials where the uncertainty was lower. Therefore, the consumption estimates based on this approach have only been used for the application area *construction*. Additionally, the domestic production was important for construction materials, so that import and export figures were a less appropriate method for the estimation of consumption.

HBCD

The mean content of HBCD in EPS has been assumed to be 0.7% over the whole period (CEFIC, 2006). As DecaBDE was used in XPS earlier instead of HBCD due to its higher thermal stability (Scherzer, 1996 cited in Leisewitz and Schwarz, 2001), no usage of HBCD in XPS has been assumed in the eighties and the beginning of the nineties. In more recent years, a mean content of 2.5% was assumed (von Arx, 1999). In an alternative scenario, the mean contents in EPS and XPS have been assumed as 1.0% and 1.5% (Danish EPA, 1999), respectively. The difference in the HBCD consumption in these two scenarios is only around 2%. Based on the data by the Danish EPA, 68% of the present HBCD consumption in the construction sector have been estimated to be in EPS and 32% in XPS. For the 1980s, a lower consumption has been estimated, as the usage has not been assumed to be comparably common as in the 1990s and these days. Figure 4 illustrates the consumption trend of HBCD in construction materials that has been used in the model. The grey and the orange line show the hypothetical trends, if the old or the new contents were applicable for the whole period.

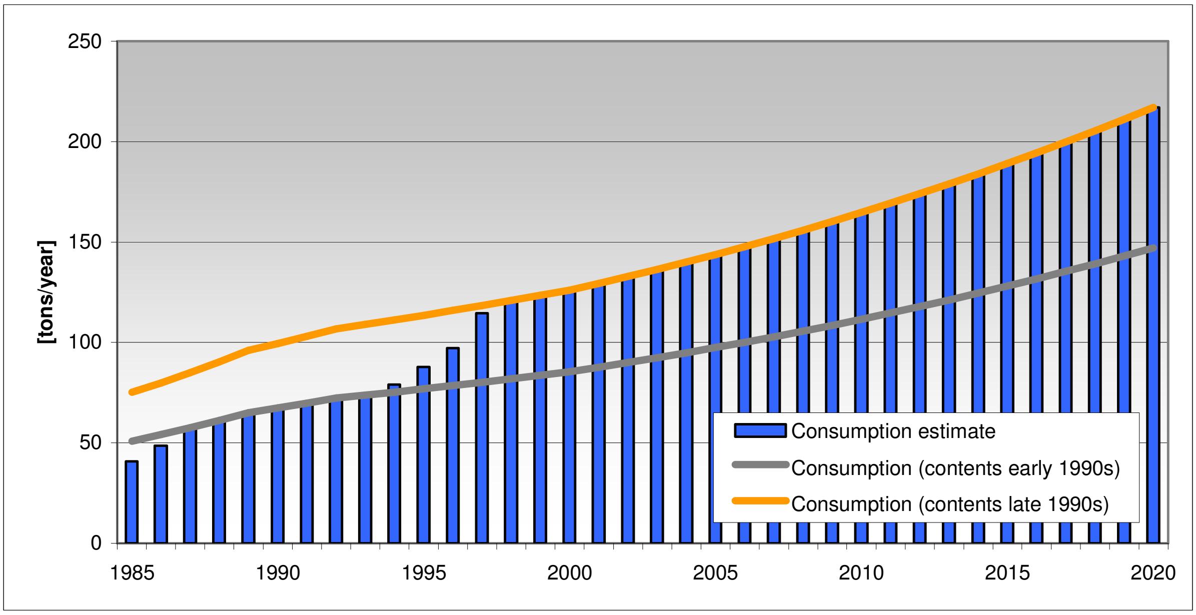


Figure 4 Consumption of HBCD in the Swiss construction sector (in expanded and extruded polystyrene)

DecaBDE

The main use of DecaBDE in construction materials has been estimated to be in polyethylene (PE) and polypropylene (PP) sheeting. The fractions of PE and PP used in the construction sector in sheeting have been estimated based on data given in Schneeberger (1999). These fractions were about 19% for PE and about 63% for PP. The fraction of flame-retarded sheeting and their DecaBDE concentrations have been estimated based on personal communications from Swiss manufacturers (Folag AG, IMS Kunststoffe AG and Sarnafil AG). The mean contents in all (including non flame-retarded) PE and PP sheeting have been estimated as 2.5 g/kg.

An additional use of DecaBDE in the early nineties was in XPS as stated above. This is the reason for the decrease in the second half of the nineties. It has to be emphasized that none of the Swiss manufacturers of construction materials contacted could confirm this use and responsible industry organisation (EPS Schweiz) was not willing to collaborate. Like for HBCD, a lower consumption has been estimated during the 1980s (see figure 5). However, in contrast to HBCD, no further increase for the future has been assumed (see section 2.4.2.1.6).

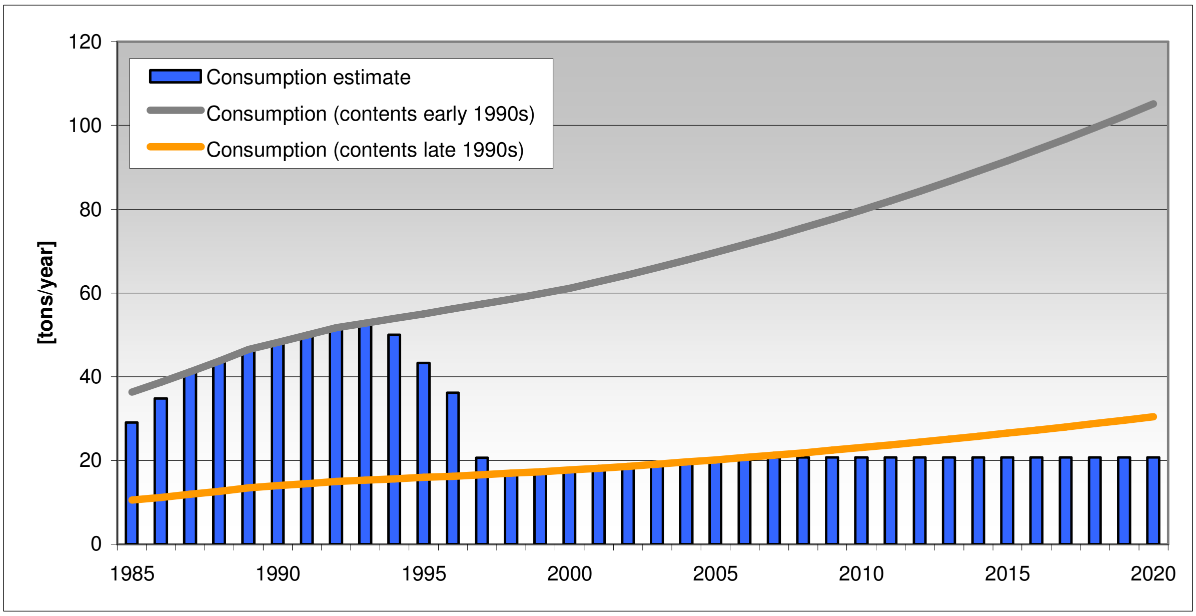


Figure 5 Consumption of DecaBDE in the Swiss construction sector

PentaBDE

According to the European Flame Retardants Association (EFRA, 1996 cited in Leisewitz and Schwarz, 2001) and the UK Department of the Environment, Transport and the Regions (DETR, 2002 cited in Prevedouros et al., 2004a) PentaBDE was used in rigid polyurethane (PUR) foam in the construction sector. A use of PentaBDE in polyvinylchloride (PVC) is given in an old plastics handbook (Gächter and Müller, 1987), where the content is specified as 4.9%. The Swiss PVC industry working committee was not aware of a use of PentaBDE in PVC (N. Helminiak, Arbeitsgemeinschaft der Schweizerischen PVC-Industrie (PVCH), personal communication). Only a small percentage of PVC has to be flame-retarded, as the material is weakly combustible. If flame-retarded, normally aluminium hydroxide is used. However, the use of BFRs could not be ruled out by Helminiak. The situation remained fairly unclear and as also no measurements in waste streams were available, the domain of uncertainty was high in the construction sector.

The main use of PentaBDE in construction materials has therefore been estimated to be in PUR foam and a smaller use in polyvinylchloride sheeting. According to Leisewitz and Schwarz (2001) approximately 21% of the PUR accounts to fitting foam glue (“Montageschaum”) of which 14% are flame-retarded with additive flame retardants. As flame retardants also tris-(2-chlorethyl)-phosphate and TBBPA had been used besides PentaBDE. Contents had usually been around 20% (Leisewitz and Schwarz, 2001). A mean content of 3% in all flame-retarded foam glue has been assumed. These three factors have been multiplied with the estimated PUR consumption in the construction sector.

For PVC, the fraction of sheeting in the construction sector has been estimated as 13% according to Schneeberger (1999). The concentration mentioned above (4.9%) has been considered and a market share of 30% has been assumed. For the late 1990s, no usage of PentaBDE in both PUR and PVC has been assumed (see figure 6).

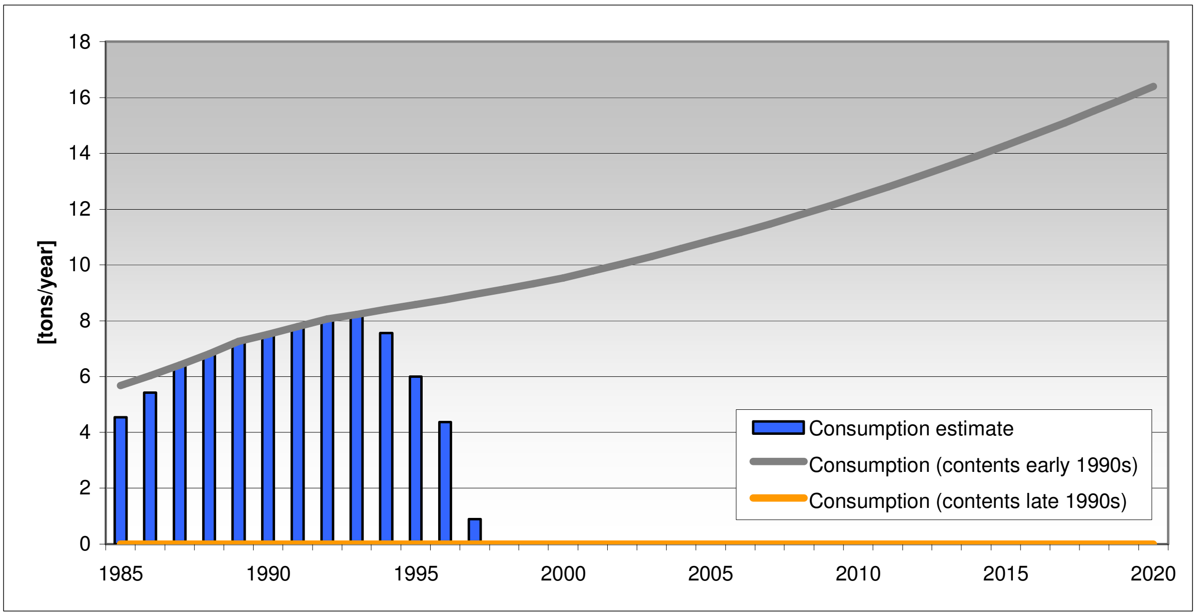


Figure 6 Consumption of PentaBDE in the Swiss construction sector

2.4.2.1.3 Data from bromine industry

The Bromine Science and Environmental Forum (BSEF) published the total market demand by region for the years 1999 and 2001 (BSEF, 2003). A spreadsheet on the worldwide and European consumption with these data and a few other on TBBPA (EBFRIP, 2006) as well as from an OECD survey filled in was sent to the BSEF and EBFRIP for complement. Unfortunately, BSEF, EBFRIP and CEFIC could only deliver a few more data on the temporal trend of the consumption. Especially for PentaBDE, OctaBDE and HBCD, the data obtained was scarce. At least for DecaBDE, a complete time trend from 1991 to 2005 was available. Contradictory to a market study, which forecasted an annual growth rate of 3% in Europe between 2004 and 2009 for BFRs (Townsend Polymer Services & Information, 2005, “Plastic Additives VI”; E. Kramer, FH Nordwestschweiz, personal communication), but in line with Alaee et al. (2003), the consumption varied but remained stable for more than one decade. An increase in the DecaBDE usage in Europe can not be seen in table 7, whereas the worldwide consumption of TBBPA and HBCD was increasing in the last decade (table 6). TBBPA showed even a striking decrease in Europe at the beginning of the current century.

Table 6 Industrial consumption worldwide [kilotons/year] (source: e-mail from BSEF)

	DecaBDE	OctaBDE	PentaBDE	TBBPA	HBCD
1991	30.0	6.00	4.0	41	
1992					
1993					
1994					
1995				109	
1996				111	
1997				122	
1998				120	
1999	54.8	3.82	8.5	127	15.9
2000				139	
2001	56.1	3.79	7.5	104	16.7
2002	65.6			129	21.4
2003	56.4			135	21.9
2004				170	
2005					

Table 7 Industrial consumption in Europe [kilotons/year] (source: e-mail from BSEF)

	DecaBDE	OctaBDE	PentaBDE	TBBPA	HBCD
1991	8.9				
1992	8.0				
1993	7.6				
1994	8.8				
1995	8.6				
1996	8.7				
1997	9.1				
1998	8.0				
1999	8.5	0.45	0.21	13.8	8.9
2000	8.6				
2001	7.7	0.60	0.15	11.6	9.5
2002	7.7				10.0
2003	7.7			7.0	9.6
2004	8.0			7.2	9.8
2005	6.9			5.4	10.6

The data on industrial consumption in Europe have been used to crosscheck the estimates derived from either import and export statistics or plastic consumption data (construction applications). The proportion of the Swiss consumption has been estimated based on a per capita approach. The population of Switzerland makes up about 1.88% of Western Europe

(EU 15 including Norway, Iceland and Switzerland itself). The percentage compared to the whole European population (excluding Russia) is approximately 1.44%. For two reasons, these calculations might not be applicable very well: Firstly, the figures given by BSEF/CEFIC are on industrial consumption in Europe. As probably significant amounts of BFRs are imported in finished products (primarily E&E) from Asia and other regions, the figures might be too low for the European consumption. Secondly, the fire safety regulations are different in the individual countries depending of the application area. It is known that due to more stringent fire regulations consumption in the UK is higher than average (especially for textiles and furniture), while it was lower in Scandinavian countries. For insulation materials, Austria, Germany and Switzerland had strict legal requirements on all building materials concerning the reaction to fire (figure 7). In Eastern Europe, authorities and fire brigades have very strict requirements on fire safety and almost 100% flame-retarded insulation material is used (D. Lausberg, BASF, cited in ECB, 2006a).

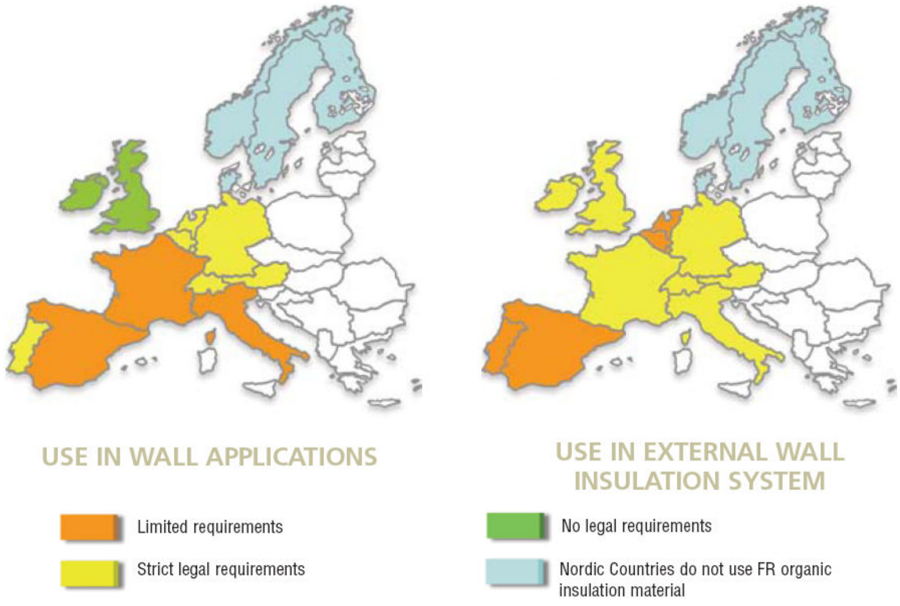


Figure 7 National fire standards for building insulation material (source: BSEF, 2006a)

Another reason for not primarily relying on the data supplied by BSEF and CEFIC has been that these figures were not split up in the application areas used in the substance flow analysis. This allocation has not been possible using the first two methods for DecaBDE and HBCD.

2.4.2.1.4 Measurements in waste streams

Representative measurements in waste streams may be used as a reliable source to estimate past consumption. The gross sample size of the prepared small waste electrical and electronic equipment (sWEEE) mixture processed during the experiment by Morf et al. (2005) was 230 metric tons, representing approximately 0.6% of the annual Swiss sWEEE. It was considered that this was a representative sample for the sWEEE that was disposed of in Switzerland in 2003. There were two studies determining the contents of PBDEs in automobile shredder residues carried out in California and in Japan (Petreas and Oros, 2006; Sakai et al., 2006b). The content determined have been multiplied with the relevant mass flow for *E&E* and *trans-*

port in order to estimate the consumption in these application areas for the time these goods entered the market.

2.4.2.1.5 Estimates for Europe and Japan

Estimates for other countries and regions have also considered being a valuable source for the estimation of the BFR consumption in Switzerland. Prevedouros et al. (2004a) used seven scenarios to estimate the historical consumption of PentaBDE in Europe (figure 8). In scenario 1 to 4, the consumption consisted of production and imports. The parameters used to estimate the production have been (a) reported world bromine production, (b) percentage of the world bromine market attributable to BFRs, (c) percentage of European consumption, (d) fraction of the European BFR market comprised by PBDEs and (e) fraction of the European PBDE market comprised by PentaBDE. For the imports, statistics by the WHO, as well as estimates by the UK Environmental Agency and the UK Department of Environment, Transport, and the Regions have been used. Scenario 5 was provided by WHO data, whereas scenarios 6 and 7 have been generated by means of PUR production and import statistics. Where no data was available, data was estimated for intermediate years by linear interpolation.

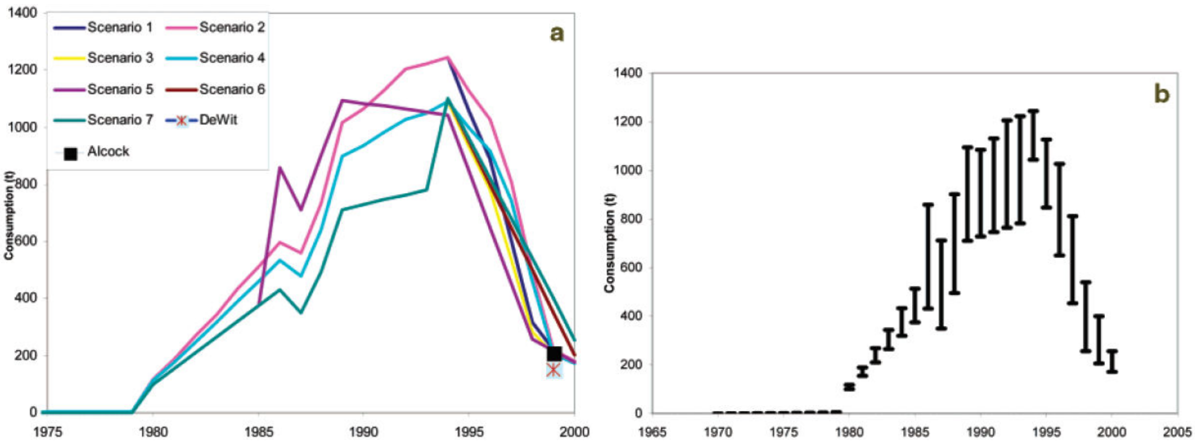


Figure 8 Estimated consumption of PentaBDE in Europe (Prevedouros et. al, 2004)

Due to the ban on PentaBDE that came into force in 2004 in Europe a further decrease has been assumed. Only a small amount of PentaBDE in imported articles would still reach the European market, until PentaBDE would be banned globally. For the model, it has been assumed that from 2008 onwards, there is no import of PentaBDE to Switzerland.

The scenarios 1 to 4 have been adapted for DecaBDE to estimate the European consumption. According to KEMI (1994, cited in ECB, 2002), 75% of the polybrominated diphenyl ethers used in Europe are made up by DecaBDE. The contribution of DecaBDE in 1999 and 2001 could be calculated using consumption figures from BSEF. In 2005, after the ban on PentaBDE and OctaBDE, a fraction of 98% DecaBDE has been assumed. The missing figures on imports of PBDE between 1990 and 2003 have been assumed with a linear interpolation. The missing import figures for the years previous to 1986 have been estimated according to Prevedouros for PentaBDE. The trends of the different scenarios have been compared with the data on the European industrial demand supplied by BSEF and CEFIC. The figures were comparable during the nineties, but lower in the beginning of the current decade. The modelled trends and the data on the European industrial demand, where the net imports of DecaBDE in consumer goods were not included, have been considered to be somewhat too low. Due to these problems, the trends of the European consumption (figure 9) have not been adapted for Switzerland and are not used in the model, but are shown here for comparative purposes.

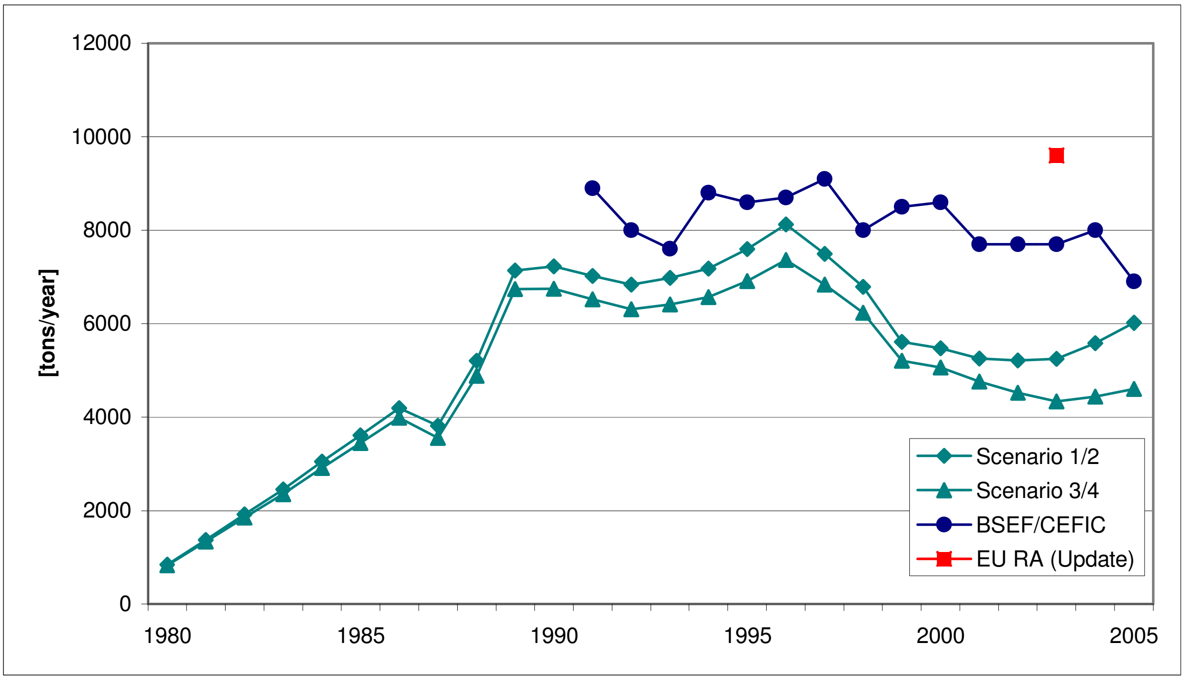


Figure 9 Modelled consumption of DecaBDE in Europe and comparison with data provided by BSEF/CEFIC and the estimate in the updated EU risk assessment

Tokai et al. (2004) modelled the trend of DecaBDE consumption in Japan based on the investigation made by Kagaku Kogyo Nippo-Sha. The authors estimated a strong increase in the 1980s and a peak consumption in the early 1990s. Starting in 1993, the consumption decreased and they predicted that DecaBDE would have completely phased out by the beginning of the second decade in the century.

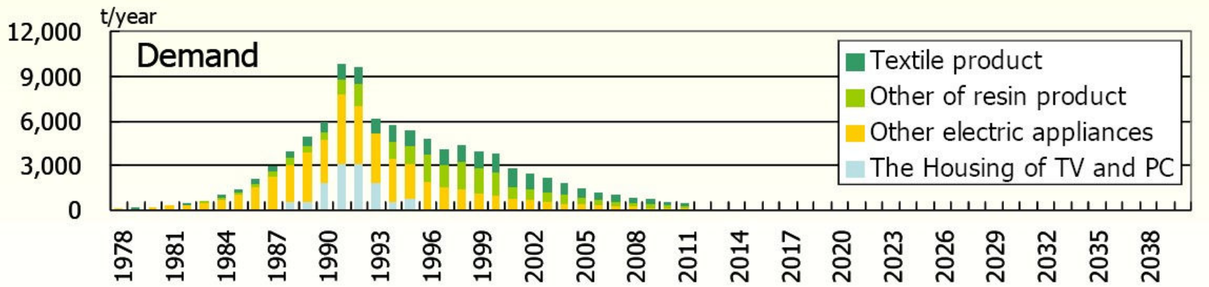


Figure 10 Consumption trend of DecaBDE in Japan (Tokai et al., 2004)

The situation for DecaBDE in Japan is not comparable to the situation in Europe. Especially, there was no peak consumption in Europe in the early 1990s followed by a strong decrease. The consumption of HBCD is notably lower in Asia than in Europe. However, the consumption trend in Japan investigated by Kagaku Kogyo Nippo-Sha (cited in Watanabe and Sakai, 2003) was comparable with Europe (see figure 25 on page 81).

The model by Prevedouros et al. was also used to estimate the trend of the European HBCD consumption. The parameters a, b and c were the same as for PentaBDE and DecaBDE. The percentage of HBCD out of BFRs has been assumed to be the same as for the industrial demand figures for Europe, which was 28% in 1999 and 32% in 2001. As illustrated in figure 11, these estimates fitted pretty well with the data supplied by BSEF/CEFIC as well as with the estimate in the EU risk assessment draft (ECB, 2006a). The amounts are also comparable on a per capita approach put side by side with the use figures generated for Switzerland mainly based on the estimated consumption of HBCD in EPS and XPS, but also measurements in waste streams (WEEE) (section 2.4.2.1.4) and estimates based on import and export statistics (section 2.4.2.1.1). The HBCD consumption has been estimated to be somewhat to increase more gently in Switzerland compared to the trends derived with the approach based on the work by Prevedouros. However, the consumption trend of HBCD could be cross-checked with that approach and the domain of uncertainty hence is smaller compared to e.g. PentaBDE.

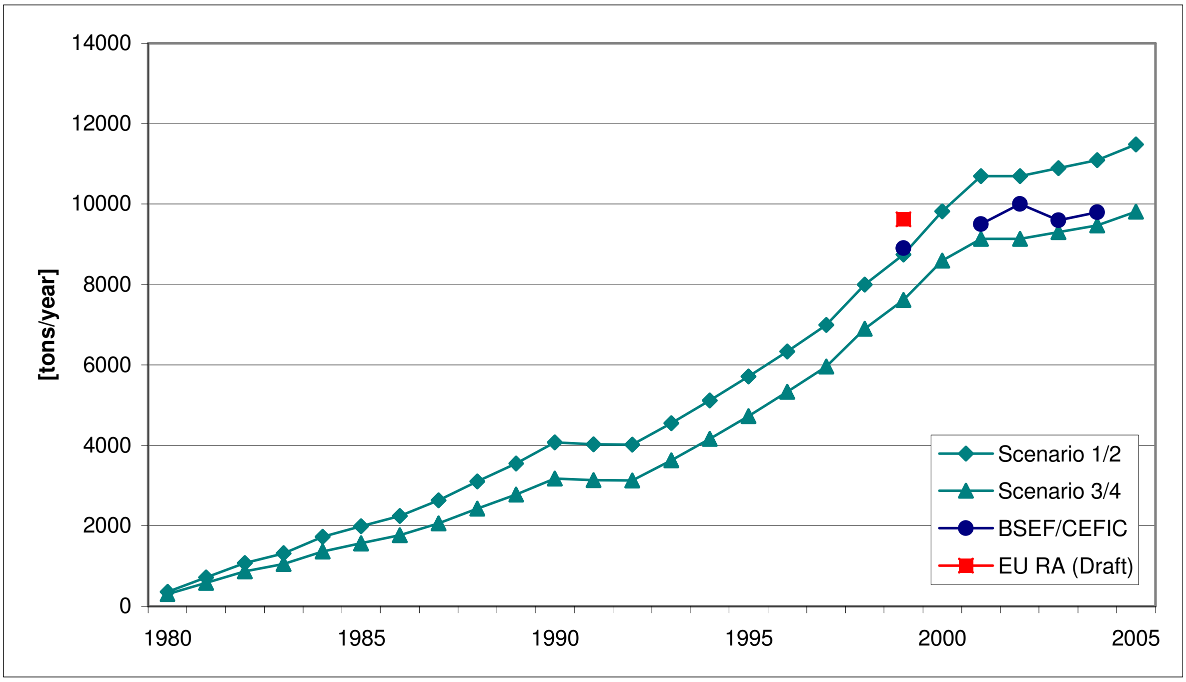


Figure 11 Modelled consumption of HBCD in Europe and comparison with data provided by BSEF/CEFIC and the estimate in the EU risk draft

The historic consumption trend of PentaBDE has been estimated based on mean contents derived from literature, measurements in waste streams (see section 2.4.2.1.4) and own assumptions. As the peer-reviewed consumption trend modelled by Prevedouros et al. (2004b) has been assumed to be a good base for the estimation of the Swiss figures, the trends have been estimated to be pretty similar. The European consumption trend has been converted for Switzerland on a per capita approach in order to get a base for the overall consumption. The consumption for the period between 2000 and 2005 (not included in the model by Prevedouros et al.) has been assumed to further decrease with a large step in the year 2004, when the use of PentaBDE was banned in the European Union. The consumption trend in construction materials estimated is described in section 2.4.2.1.2. The consumption in the application areas *E&E* and *transport* has been estimated based on measurements in waste streams (WEEE and ASR) that were available for one specific year, taking into account residence times in the use phase. This proportion of the consumption in these application areas compared to the overall consumption has been assumed to remain constant over several years to get the consumption trends. As for the application area *textile/furniture* no measurements were available, the estimates by Prevedouros have been converted on a per capita level to Switzerland. Packaging that might also have been a small consumer in past, has not been considered as a fifth application area would have been needed. However, the fraction of the total consumption in this application area has been assumed to be only around 3% and the residence times to be short. The overall consumption trend estimates are illustrated in figure 12.

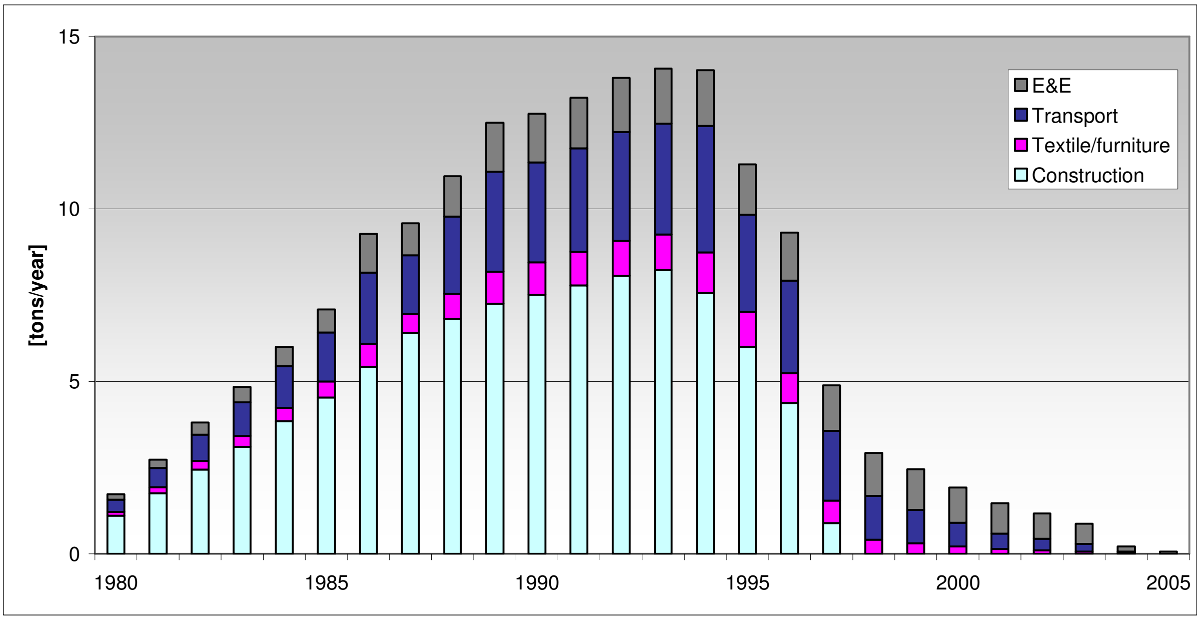


Figure 12 Consumption trend of PentaBDE in Switzerland used in the model

2.4.2.1.6 Future trends of use figures

The future trends of the HBCD and DecaBDE consumption have been estimated according literature available and own assumptions. As HBCD is used in HIPS housings, the consumption is highly dependent on the HIPS demand. The consumer product enclosure market is exclusively made up by HIPS with little change foreseen in the immediate future. Between 1998 and 2003 there was an increase in resin demand for electronics housings of 3.3% annually (2.4% for IT, 4.3% for consumer electronics). In terms of resin competition, it was estimated that HIPS can hold its share against the competing ABS (Dufton, 2003). Contradictory to this market report, in the 2005 market report by Townsend the material used in TV sets, computer housings and mobile phone is switched from HIPS to PC/ABS or PPO, which can use non-halogenated flame retardants (E. Kramer, FH Nordwestschweiz, personal communication). It is presumed that the trend in consumption is highly dependent on the conclusions in the final EU risk assessment report. If a risk would be found, a strong decrease within a few years is expected. But even if the RA would conclude with “no need for risk reduction measures”, no further increase is expected for the application areas *E&E*, *transport* and *textile/furniture*. According to several studies (e.g. Cahill et al., 2005; Morose, 2006; Posner, 2006), there are halogen free alternatives for polymer types such as HIPS and textiles. The concept of green product design has become an important consideration within the flame retardant industry in recent years. The consumer is more educated in environmental issues and has become increasingly demanding when it comes to product and label acceptability. The market for environmentally considerate products is growing steadily. A number of voluntary eco-labels have been developed in order to increase market pressure with regards to product design (e.g. TCO, Blue Angel, Swan and Flower). According to Greenpeace more and more companies declare to phase out the use of BFRs in E&E. In May 2007 also apple that previously was the worst rated company decided to do so (Greenpeace, 2007). On the other hand, there are no alternatives for EPS and XPS except the brominated flame retardants SAYTEX BC-48 (tetrabromocyclooctane) and SAYTEX BCL-462 (dibromoethyl dibromocyclohexane). According to Clariant (de Boysere et al., 2007), there are halogen-free alternatives for all BFRs except for HBCD in EPS and XPS, for additive TBBPA in ABS and for DecaBDE in some HIPS applications. Therefore, it is expected that substitution will at least compensate for the market growth for HBCD in all application areas except construction. For the same reasons, no increase in the consumption of DecaBDE is expected (see figures in section 7.6.1). In order to include possible bans, different scenarios have been modelled to illustrate the influence on substance flows, stocks and emissions (see section 2.5).

2.4.2.2 Residence times in the process *use*

Lifetimes of goods containing brominated flame retardants have been used for the estimation on the residence times in the process *use*. It was operated with mean residence times and an uncertainty range whose extend was dependant on the heterogeneity or homogeneity of the application area.

2.4.2.2.1 E&E

The actual lifetime of office and consumer electronics consists of a service life of 5–6 years and an additional period of 3–4 years where the appliances are stored until they are disposed. Hence, the lifetime in the use phase is approximately 9 years at present. In the mid 1990s, the mean lifetime was 11 years and a decrease to 7 years is expected in future (P. Bornard, SWICO, personal communication). Large household appliances like freezers or washing machines have a lifetime of 15 years and more, whereas small household appliances like vacuum cleaners or coffee percolators are in use for about 5 to 10 years (VKE, 2003). As residence times decreased over the years, a variable residence time – with the result of a more computationally intensive calculation in SIMBOX – was indispensable for this application area. A residence time of 12 years was assumed for 1982 and earlier, 9 years for 1997 and 7 years for 2009 and later. Due to these highly diverse lifetimes, a high range (standard deviation of 5 years) has been assumed.

2.4.2.2.2 Transport

The service life of passenger cars was estimated as 10 years and seemed to remain constant (Leidner, 1981; Hirai and Sakai, 2004). In another study, the mean service life until disposal as approximately 14 years (Reinhardt and Richers, 2004). Other vehicle types may have slightly different service lives. Lorries, buses or airplanes have longer service lives, whereas it is shorter for e.g. motorbikes. As passenger cars make up the by far largest fraction of BFR consumption, the mean lifetime has been assumed as 12 years with a standard deviation of 3 years.

2.4.2.2.3 Textiles

Leidner (1981) estimated the service life of furniture and articles of clothing as 10 and 4 years, respectively. As in Europe only a very small fraction of clothing contain BFRs, 10 years seemed to be an appropriate estimate. The standard deviation has been assumed as 3 years.

2.4.2.2.4 Construction

APME (1995) estimated the number of years after which a product would become waste for the construction sector (see table 8). The main use of brominated flame retardants is in insulation and lining, with some minor usage in other fields. Therefore, it has been assumed that the mean lifetime of plastics in the construction sector is 30 years (standard deviation of 15 years) for construction materials containing BFRs except HBCD.

Table 8 Lifetime distribution of plastics in construction applications [years] (APME, 1995)

	< 2	2–5	5–10	10–20	20–40	> 40
Pipes and ducts		1%	1%	3%	20%	75%
Windows			1%	2%	32%	65%
Insulation	2%			10%	50%	38%
Lining	5%		10%	25%	40%	20%
Profiles	3%		5%	30%	50%	12%
Fitted furniture	1%		25%	49%	20%	5%
Fixed floor coverings	5%	2%	20%	68%	5%	
Wall coverings	2%	8%	50%	30%	10%	

The lifetimes seem to remain stable for the last two decades. In 1981, a mean lifetime of all construction applications including products with a shorter lifetime like coverings was estimated as 25 years (Leidner, 1981). It was also assumed that the lifetime would not change much in the future decade. According to a recent study in Switzerland, the mean service life of EPS insulation panels is about 40 to 50 years (Carbotech AG, 2004), while in the EU risk assessment draft report, the mean service life of EPS and XPS insulation panels was estimated as 50 years. Hence, a lifetime of 50 years (standard deviation of 25 years) seems to be most appropriate for HBCD.

2.4.2.3 Input of atmospheric deposition to sewer system

The substance flow to the sewer system caused by the atmospheric deposition has been treated as an input flow in the model. The reason is that the environment including the atmosphere has been defined to be outside the system boundary. Therefore, the atmospheric deposition has been estimated separately. Instead of a modelled atmospheric deposition, field data have been taken into account. The flow has been estimated as follows: The deposition rate in urban areas that has been supposed to be most appropriate has been multiplied with the area of Switzerland and the proportion of the area that attributes to stromwater in sewerage. It has been assumed that the substances deposited on sealed ground can not be attributed entirely to sewerage system due to effects such as photodegradation or adsorption on dust blown away. A factor of 80% of the total atmospheric deposition on sealed ground has been assumed to enter the sewerage system. The area of Switzerland is 41,285 km², the sealed area drained to sewer has been estimated by adding building area (1.1%), half of the adjacent areas (2.4%) and road area (1.9%) which adds up to 4.2% (Swiss Federal Statistical Office, 2002). The variability of atmospheric deposition measurements is quite large. Measurements of PBDEs deposition rates in Sweden in 2002 and 2004, both made in urban area, differed by approximately a factor of 30 (ter Schure and Larsson, 2002; ter Schure et al., 2004a). The wet-only precipitation rates measured in the Great Lakes basin were between these two studies (Backus et al., 2005). As shown in table 9 the highest deposition rates were measured on the campus of the Kyoto University. However, the atmospheric deposition of PBDEs was more than two orders of magnitude lower in September 2001 compared to August 2000 demonstrating a high variability of that flow (Hayakawa et al., 2004). Recently, the atmospheric deposition was measured in Dübendorf, a community in the agglomeration of Zurich. The wet and dry deposition rate measured with Bergerhoff samplers over a period of 50 days was 9.83 ng/m^2/day for BDE-209 and 0.82 ng/m^2/day for BDE-47 in February 2006 (C. Bogdal, Empa, personal communication). As these rates were measured in Switzerland and it is within the range determined in other studies, it has been assumed to be most appropriate to be included to the model.

For HBCD, two studies measuring the atmospheric deposition rates are known. The deposition rates ranged from 0.02 ng/m^2/day at the Swedish west coast in winter to 366 ng/m^2/day near Stockholm in summer (Remberger et al., 2004). The two measurements in Stockholm (5.5 and 366 ng/m^2/day) differed by nearly a factor of 70. In a study carried out in the Great Lakes basin, wet-only deposition rates between 0.36 and 10 ng/m^2/day were found. As opposite to the Swedish study, the results peaked in winter, which was reckoned to be caused by the increased scavenging efficiency of snow compared to rain and higher concentrations in the particle phase during the winter by the authors (Backus et al., 2005). As a consequence of the lower usage of HBCD in the North America compared to Europe and the larger area, the consumption per area in North America is approximately 20 lower than in Europe. Assuming that the lower consumption would also result in a 20 times lower emission, the measurements in the Great Lakes basin are comparable to the deposition rates in the Swedish urban area. The atmospheric deposition has been assumed to rather have a lognormal than a normal distribution. Therefore the geometric mean of the two measurements near Stockholm, 44.9 ng/m^2/day, seemed to be the most appropriate value. This value might seem somewhat high compared with background or remote areas, but it has to be noted that the measurements did not include the dry deposition flux.

The actual input from atmospheric deposition to sewer system has been estimated according to the assumptions described above. For DecaBDE, the estimated current input is 5.0 kg/year, while it is 22.7 kg/year for HBCD and 0.4 kg/year for BDE-47. The domains of uncertainty are one order of magnitude, because of the high variations in deposition rates. The trend of this flow has been not modelled in SIMBOX, as it is an input flow. Therefore, it has been assumed that the relative change is equal to the consumption trend. The percentages accounted to each application area have also been assumed to be relative of the consumption share.

Table 9 Literature data on atmospheric deposition (ng/m^2/day)

Data source	PBDEs	BDE-209	BDE-47	BDE-99	BDE-183	HBCD	TBBPA
ter Schure, 2004 [b]	7.0/6.8 [a]	10.9/32.6	2.9/3.7	1.9/1.9	1.2/0.7		
ter Schure, 2002 [c]	2.0	0.5/0.08	0.09/0.04	0.2/0.05	0.08/0.15		
Hayakawa, 2004 [d]	1600/28/11	1500/24/8.3	9.7/1.0/0.56	12/0.92/0.52			
Bogdal, 2006		9.83	0.82	0.19	0.27		
Backus, 2005 [e]	0.39–1.3 [a]	2.8–5.3				0.36–10	
Remberger, 2004 [f]						5.5–366	
Remberger, 2004 [g]						0.02–13	

[a] PBDEs excluding BDE-209, [b] urban reference site (dry flux/wet flux), [c] particulate flux/dissolved flux (read from logarithmic graphic), [d] campus of Kyoto University (August/January/September), [e] precipitation, [f] urban site, [g] background and remote sites

2.4.2.4 Transfer coefficients and proportion coefficients in the anthroposphere

Transfer coefficients (TC) and proportion coefficients (PC) are both parameters used in the dynamic model. TCs describe the distribution of BFRs from the total input into a process to a selected output as a fraction of 1. Hence, their range of values is between 0 and 1. A mathematical definition is $TC_i = Outflow_i / \sum Outflows$ except for transfer coefficients in processes having a stock. For flows entering directly into environment compartments, the TCs have been denoted as emission factors (see section 2.4.2.5).

In addition to the transfer coefficients, proportion coefficients have been used in order not to merge all information in the transfer coefficients. They describe which proportion of a good was attributable to a flow (i.e. only the textiles being washed were of importance for the flow from the use into the sewerage). Or as another illustrative example, the fraction of the amounts of BFRs in wastewater reaching WWTP and transferred to sewage sludge and effluents are defined by transfer coefficients. The flow in sewage sludge is further split up to flows to incineration, landfill and soil by dint of proportion coefficients (see figure 13).

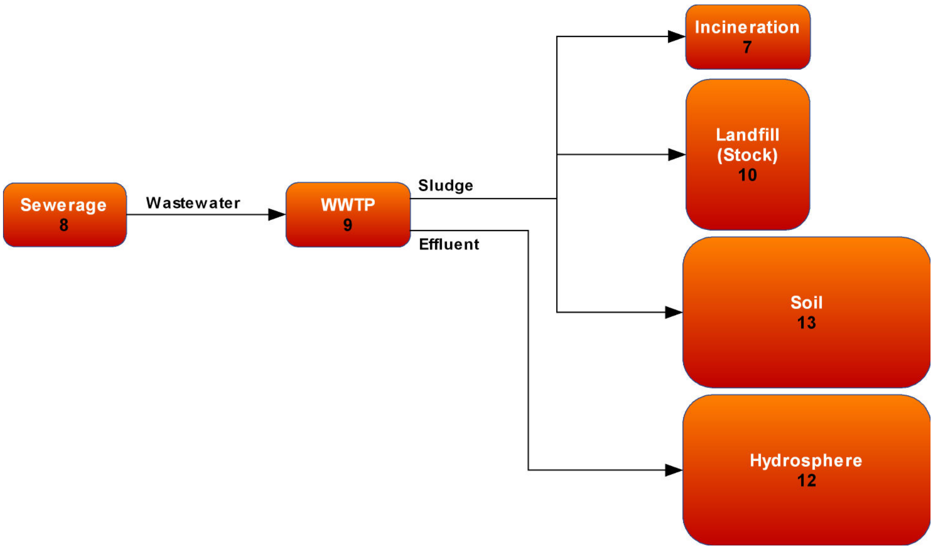


Figure 13 Input in and outputs from the process *WWTP*: transfer coefficients define the fraction of the input in the two outputs sludge and effluent, proportion coefficients define the fraction of the sludge that is incinerated, dumped on landfills and used in the agriculture as a fertilizer.

In the following subsections, the way the different transfer coefficients and proportion coefficients have been determined is described. The values of the coefficients selected for the model are illustrated in lists or figures. As some of the coefficients are different for each application area, each substance and for individual years, not the full list is given in the tables. For periods with changing coefficients either the range or the values at the beginning and at the end are listed. In many cases, there is a linear increase of decrease during this period.

2.4.2.4.1 Production

The domestic production share, the proportion of the production going to the domestic market and to export as well as the releases to wastewater and waste from production processes have been estimated in this study. There is no domestic production of brominated flame retardants and to the authors' knowledge there never was. For HBCD, there is only one production plant in Europe that is situated in the Netherlands (BSEF, 2007). The amount of imported chemicals was considered to be small compared to imports of semi-finished and finished products. Due to the fact, that the substances under study do not have customs tariff number of their own, it was not possible to figure out the amounts imported. However, there is a production of flame retarded masterbatches and resins. According to information from the Kunststoff Verband Schweiz (P. Stauffer, personal communication), there are two primary producers of flame retarded polymers. EMS Grivory manufactures polyamides; Huntsman Advanced Materials produces epoxy resins. Especially for construction materials, there are several domestic manufacturers using flame retarded masterbatches in their products. Otherwise, the Swiss industry processes imported semi-finished products like resins, textile fibres or printed circuit boards. The proportion coefficient *PC(P,domestic)*, which denotes the percentage of the BFR flows imported in semi-finished and finished in Switzerland and the transfer coefficients of substances into waste *TC(P,inc)* and into wastewater *TC(P,sew)* are used in the process *production*. The TC into products was defined as the remainder minus the emissions into the environment.

E&E

According to SWICO (P. Bornard, personal communication), the domestic production share of IT equipment, communication technology and consumer electronics is low. About every fifth PC is assembled in Switzerland these days, while every fourth was assembled in past. The different parts were almost exclusively imported from abroad. The domestic production share of communication technology has been estimated virtually zero, whereas there was a production in the 1980s (e.g. by Ascom). Also for consumer electronics, the production share of Switzerland is expected very low and estimated to be clearly below 5% over the whole period starting in the 1980s.

On the other hand, the domestic production share of small and large household appliances, special appliances, medical appliances and instruments and automatic issuing systems are much higher. A constant share of 50% has been estimated. Over the whole application area, the domestic production share is thus estimated as approximately 30% (*PC(P,domestic)*).

Transport

There is no notable production of passenger cars in Switzerland, but there is a supplier industry. On the other hand, there is a domestic production of trains (e.g. Adtranz, Bombardier, Alstom, Rieter). There is also a production of e.g. agricultural vehicles and airplanes, but as personal cars make up by far the largest amount of BFRs, it has been estimated that the Swiss suppliers account for about 3% of the Swiss consumption (*PC(P,domestic)*).

Textile/furniture

According to EBFRIP (2003, cited in ECB, 2004), the United Kingdom and Ireland are the only EU countries that currently have regulations specifying a level of flame retardancy for domestic upholstery fabrics. As a result, the large majority of upholstered fabrics containing flame retardants are supplied to these markets. According to one of the biggest Swiss mattresses manufacturers (that did not want to be named), no flame retardants are used at present. A minor use for special purposes in the past could not be ruled out. The same was true for textile formulation and back-coating. To the authors' knowledge, there is presently no production of textiles treated with BFRs in Switzerland. The industry associations (Textilverband Schweiz, Verband Schweizer Filialunternehmer, Swiss Retail) and the large furniture retailers (e.g. IKEA, Interio, Pfister) asked were either not able or not willing to supply information on that topic. According to Interieursuisse (P. Platzer, personal communication), either the flame-retarded fibres or the tissues are imported at present. A very common flame-retarded fibre is the polyester fibre Trevira CS, which uses a phosphororganic flame retardant. As it is not known, if the Swiss textile industry manufactured flame retarded fibres in past, it has been assumed that 10% of the fibres were produced domestically between 1980 and 1995. After this period, a linear decrease to 0% in 2005 has been estimated.

Construction

The domestic production share of construction materials must be separated in two groups. For plastics foils and similar materials, the share has been estimated as approximately 80% (M. Schneider, Sarnafil, personal communication), while it is smaller for insulating materials. This share has been assumed as 30%. There was no information on time trends available. As HBCD is used in EPS and XPS foams, a share of 30% has been estimated to be appropriate. Also PentaBDE was mainly used in polyurethane foams and the same share is thus estimated. DecaBDE is used in both plastics foils and insulating materials. The domestic production share was therefore estimated as 70% to present. As DecaBDE was used in XPS insulation panels instead of HBCD in the 1980s and 1990s, a share of 30% in 1980 followed by a linear increase until 1998 has been estimated.

Production wastewater and solid waste

A survey was carried out in the UK textile industry that allowed to calculate actual TCs for DecaBDE (Wragg, 2005). In connection with the Voluntary Emissions Control Action Programme (VECAP), there was a large reduction of the unwanted flows into waste and wastewater between 2004 and 2005. Loss 4.1% and 1.7% to waste could be calculated from the data given for these two years. No measured data on losses of DecaBDE to waste was available for other application areas that could be used to calculate TCs. In the Emission Scenario Document on Plastics Additives (OECD, 2004), losses to waste were estimated. During raw materials' handling, 0.1% is lost to solid waste with another 0.01% in residue bags. No losses to solid waste are given for compounding and conversion. The waste off-cuts in semi-finished goods have been estimated to amount to <5–20% for panels and 5–10% for textiles. The loss calculated from the measured data was considered to be most appropriate to cover the heterogeneous production process. Therefore, it has been assumed that *TC(P,inc)* was 4.1E–02 for all application areas for 2004 and before. It was expected that until 2008 virtually all companies would participate the VECAP program or would have started own initiatives to reduce emissions and losses. Hence, the 1.7% stated above was taken starting from the year 2008 with a linear decrease in the intermediate period (table 10).

The losses to wastewater calculated from the UK textile industry survey were 0.56% and 0.18% for 2004 and 2005, respectively. There were no data for the other application areas. The losses to wastewater given Emission Scenario Document on Plastics Additives are 0.1% during raw materials' handling, 0.011% during compounding and 0.005% (open processes, solid articles), 0.01% (open processes foamed articles), 0.003% (partially open processes) and 0.001% (closed processes) during conversion. According to VECAP (BSEF, 2006b), the losses to wastewater in the plastics sector are by at least a factor of 10 smaller than in the textile industry. The transfer coefficient *TC(P,sew)* from production to sewerage in the sectors E&E and construction was thus assumed as a tenth of the TC in the textile industry. As vehicles contain both, flame-retarded plastic and textile parts, the mean of the TCs in these two application areas has been assumed to be most appropriate for *transport*. For textiles, the TC has been assumed to be 5.6E–03 from 1995 to 2004. Due to the Federal Law on the protection of waters (Gewässerschutzgesetz, GSchG) that was implemented in 1992 a reduction of BFR release to wastewater of 20% has been assumed, as these chemicals were not emission reduction targets. Hence, the TC was 7.0E–04 during the 1980s. The same assumption was made analogous for the other application areas.

For HBCD, TCs are provided in EU risk assessment draft report of HBCD (ECB, 2006a). The 2007 version of the draft report (ECB, 2007b) could not be taken into account any more. In contrast to this risk assessment, it has been assumed for Switzerland that all wastewater is lead into the sewerage system. A TC into wastewater of 5.0E–06 on the formulation of polystyrene beads containing HBCD for the manufacture of EPS or the production of HIPS has been calculated. For the formulation of polystyrene compounds containing HBCD for the manufacture of XPS, a TC of 3.2E–05 has been derived from the data given in the risk assessment. The TC for the formulation of polymer dispersions for textile back-coating was 5.0E–06. For the manufacture of HIPS as well as EPS and XPS the TCs were 3.0E–05 and 1.2E–05, respectively. At last, the TCs for the industrial use of HBCD powder for flame-retarded XPS and textile back-coating have been calculated as 1.0E–05 and 1.7E–03. These TC have been summed up for the individual application areas according to their relative importance. The same relative changes in the TCs as for DecaBDE to the Federal Law on the protection of waters and the VECAP program have been presumed. As no data on losses to waste were available, the same values as for DecaBDE have been assumed.

A major use of PentaBDE was in flexible PUR foam that was used in upholstered furniture and automobiles (ECB, 2000). PentaBDE may have been used in some textile applications in past, but it have had a small market share and the use was thought to be discontinued in the early 1990s at the latest. There was also a use of PentaBDE in rigid PUR foam used in the construction sector and in printed circuit boards. The TC into wastewater estimated in EU Risk Assessment was 5E–04 for the production of flexible PUR foam. The relative changes and the TCs for the other application areas than *textile/furniture* have been estimated as above. As no data on losses to waste were available, the same values as for DecaBDE have been assumed.

Summary

The TCs and PCs used as parameters from the model for the process *production* are summarized in the table below.

Table 10 Transfer and proportion coefficients applied in the dynamic SFA model for the process *production*

	Substance	Application area	Year	Value	Uncertainty
PC(P,domestic)	all	E&E	independent	3.0E–01	±30%
PC(P,domestic)	all	transport	independent	3.0E–02	±30%
PC(P,domestic)	all	textile	1980–1995	1.0E–01	±50%
PC(P,domestic)	all	textile	2005–2020	0.0	-
PC(P,domestic)	DecaBDE	construction	1980	3.0E–01	±30%
PC(P,domestic)	DecaBDE	construction	1998–2020	7.0E–01	±30%
PC(P,domestic)	HBCD	construction	independent	3.0E–01	±30%
PC(P,domestic)	PentaBDE	construction	independent	3.0E–01	±30%
TC(P,inc)	all	all	1980–2004	4.1E–02	±50%
TC(P,inc)	all	all	2008–2020	1.7E–02	±50%
TC(P,sew)	DecaBDE	E&E, construction	1980–1989	7.0E–04	TC/10 – 10·TC
TC(P,sew)	DecaBDE	E&E, construction	1995–2004	5.6E–04	TC/10 – 10·TC
TC(P,sew)	DecaBDE	E&E, construction	2008–2020	1.8E–04	TC/10 – 10·TC
TC(P,sew)	DecaBDE	transport	1980–1989	3.9E–03	TC/10 – 10·TC
TC(P,sew)	DecaBDE	transport	1995–2004	3.1E–03	TC/10 – 10·TC
TC(P,sew)	DecaBDE	transport	2008–2020	9.9E–04	TC/10 – 10·TC
TC(P,sew)	DecaBDE	textile	1980–1989	7.0E–03	TC/10 – 10·TC
TC(P,sew)	DecaBDE	textile	1995–2004	5.6E–03	TC/10 – 10·TC
TC(P,sew)	DecaBDE	textile	2005–2020	-	-
TC(P,sew)	HBCD	E&E	1980–1989	4.1E–05	TC/10 – 10·TC
TC(P,sew)	HBCD	E&E	1995–2004	3.2E–05	TC/10 – 10·TC
TC(P,sew)	HBCD	E&E	2008–2020	1.1E–05	TC/10 – 10·TC
TC(P,sew)	HBCD	transport	1980–1989	1.1E–03	TC/10 – 10·TC
TC(P,sew)	HBCD	transport	1995–2004	8.8E–04	TC/10 – 10·TC

TC(P,sew)	HBCD	transport	2008–2020	2.9E–04	TC/10 – 10·TC
TC(P,sew)	HBCD	textile	1980–1989	2.2E–03	TC/10 – 10·TC
TC(P,sew)	HBCD	textile	1995–2004	1.7E–03	TC/10 – 10·TC
TC(P,sew)	HBCD	textile	2005–2020	-	-
TC(P,sew)	HBCD	construction	1980–1989	4.3E–05	TC/10 – 10·TC
TC(P,sew)	HBCD	construction	1995–2004	3.4E–05	TC/10 – 10·TC
TC(P,sew)	HBCD	construction	2008–2020	1.1E–05	TC/10 – 10·TC
TC(P,sew)	PentaBDE	textile	1980–1989	7.5E–04	TC/10 – 10·TC
TC(P,sew)	PentaBDE	textile	1995–disuse	6.0E–04	TC/10 – 10·TC
TC(P,sew)	PentaBDE	transport	1980–1989	4.1E–04	TC/10 – 10·TC
TC(P,sew)	PentaBDE	transport	1995–disuse	3.3E–04	TC/10 – 10·TC
TC(P,sew)	PentaBDE	E&E, construction	1980–1989	7.5E–05	TC/10 – 10·TC
TC(P,sew)	PentaBDE	E&E, construction	1995–disuse	6.0E–05	TC/10 – 10·TC

2.4.2.4.2 Trade

The flow from *trade* to *use* or to *construction* (construction materials) has been estimated using import and export figures, literature data and own assumptions (see chapter 2.4.2.1). The resulting consumption trends were fluctuating because of differences in the import and export figures between individual years. Firstly, this was thought to be an artefact of the methodology applied and secondly, fluctuations in consumption trends would have resulted in fluctuations throughout the model, which would have made interpretations difficult. Therefore, the consumption trends have been smoothed by means of two consecutive simple moving averages. Depending on the extent of fluctuation simple moving average calculations over 3 or 4 data points have been applied.

The proportion of the flows to *trade*, which are exported to abroad *TC(T,exp)*, have been estimated from import and export figures by dividing the export by the sum of import of semi-finished and finished products. For construction materials, information from industry was used to estimate the TCs. For foils and sheets the domestic production is high and there is also a significant amount exported to abroad, while for insulation panels Swiss production is much less important (M. Schneider, Sarnafil, personal communication). As DecaBDE is used in foils and sheets as well as insulation panels, *TC(T,exp)* has been estimated as 0.3, whereas for HBCD, used in EPS and XPS panels, the coefficient has been estimated as 0.1. As the TCs were also fluctuating for the other three application areas, they have been smoothed for the same reason and in the way as the consumption trends.

The transfer coefficient *TC(T,const)* is defined for the purpose to be able to switch the model to either construction or E&E, transport and textile applications. If the value is 1, the BFRs in construction materials pass through the process *construction* first before they reach the stock of the process *use*. As for all the other application areas this detour is not necessary, the value there is 0.

Table 11 Transfer coefficients applied in the dynamic SFA model for the process *trade*

	Substance	Application area	Year	Value	Uncertainty
TC(T,const)	all	E&E, transport, tex	independent	0	-
TC(T,const)	all	construction	independent	1	-
TC(T,exp)	all	E&E	dependent	3.7E−01 – 4.7E−01	±30%
TC(T,exp)	all	transport	dependent	8.0E−02 – 3.6E−01	±30%
TC(T,exp)	all	textiles	dependent	1.9E−01 – 3.5E−01	±30%
TC(T,exp)	DecaBDE	construction	independent	3.0E−01	±30%
TC(T,exp)	HBCD	construction	independent	1.0E−01	±30%
TC(T,exp)	PentaBDE	construction	independent	3.0E−01	±30%

2.4.2.4.3 Construction

The transfer coefficient *TC(C,waste)* describes the percentage of substances that get into waste during construction operations. The remainder minus the emissions into the environment characterizes the proportion of the flow that enters into *use*. The proportion coefficient *PC(C,landfill)* denotes the distribution of the waste generated between landfill and incinerator, whereas the remaining fraction is incinerated. The fraction of combustible building waste that was disposed on landfills has been assumed to be equal to the data from the process *deconstruction* (see section 2.4.2.4.5).

Based on information from the line of business it has been estimated that 2–5% of the used EPS and XPS is cut off during mounting and disposed of with combustible building waste. The loss of PUR during construction operations is depending on the application. According to suppliers, the loss is normally low and was estimated to be 2% of the used volume. For PE pipe insulation, foils and panels the waste generation during use has been assumed on average to correspond to 5% of the used volume (Danish EPA, 1999). As HBCD is mainly used in EPS and XPS, while PUR and PE is treated with PBDEs, a mean loss of 3.5% and a range of 2–5% seemed to be adequate for both substances. As PentaBDE is only used in PUR foam, 2% have been considered.

Table 12 Transfer and proportion coefficients applied in the dynamic SFA model for the process *construction*

	Substance	Application area	Year	Value	Uncertainty
TC(C,waste)	DecaBDE	construction	independent	3.5E−02	±30%
TC(C,waste)	HBCD	construction	independent	3.5E−02	±30%
TC(C,waste)	PentaBDE	construction	independent	2.0E−02	±30%
PC(C,landfill)	all	construction	dependent	see figure 16	±30%

2.4.2.4.4 Use

Outputs from the use phase of end-of-life products are either directed to a recycling process, to a waste incinerator, directly to a landfill or are exported abroad. For each application area the TCs are determined for the time period investigated according to available data and expert knowledge. As the historical waste management data are very limited, extrapolations between known data points and own assumptions had to be applied. Emissions and releases during the use phase are discussed in section 2.4.2.5.

E&E

More than one decade ago, efforts began to establish separate collection of disused E&E equipment in Switzerland. In 1990, the Swiss Waste Disposal Foundation (SENS) was established with the single purpose of managing the disposal of refrigeration equipment and waste electrical and electronic equipment (WEEE). After a short interval, SWICO introduced a recycling and disposal scheme for office electronics and IT equipment. In 1998, the Swiss Ordinance on the Return, Acceptance and Disposal of Electrical and Electronic Appliances was established (VREG, 1998). Another disposal route that has been considered as recycling (metal recycling) is the collection of small WEEE (sWEEE) together with metal scrap and the treatment in large automobile recycling plants. According to estimates in the waste concept for Switzerland (BUWAL, 1992), 75% of the consumer electronics and IT equipment were disposed as municipal solid waste in 1990. For the years 1980–1990, a recycling rate of 0.25 has been estimated for this route. It has been assumed that at that time this proportion of sWEEE was disposed off via waste incineration or landfills (municipal solid waste). The share between waste incineration and landfill is according the statistics for burnable municipal solid waste. It has been estimated that the recycling rate for the relevant fractions of sWEEE containing BFRs via the SENS-SWICO collection system was 0.1 in 1991. Before this year, there was no recycling via this disposal route. In Morf et al. (2002) a rate of 0.5 in the year 1999 has been estimated according to information from SENS. In between the years 1999 to 2005 the SENS-SWICO collection system was established more and more (mobile phones, phones, etc. were collected, too). In a more detailed study (Waber, 2001), the recycling rate for the SENS-SWICO collection system was estimated for sWEEE with 0.64 in 2001. Export figures for sWEEE exist for the year 2001 (Waber, 2001). In that year, approx. 2% of the entire disposed sWEEE was exported only. Before 1999 it has been assumed that no export took place, and for the years 2000 to 2005 the export rate has been estimated with 2%. In Waber (2001), the amount of sWEEE going to large ASR treatment plants was estimated as 6700 tons/year (5200 tons/year consumer electronics, 1500 tons/year small household appliances). This figure represented a proportion of approximately 0.17. A steady decrease of this rate has been assumed until 2005 (0.02). For year 2005, the recycling rate via the SENS-SWICO collection system has been estimated with 0.93 (remaining routes: 0.02 through recycling in larger ASR plants, 0.03 still disposed via municipal solid waste and 0.02 leaves the country via export). In-between a steady increase of the recycling rate has been assumed due to the optimization of the SENS-SWICO collection system.

Large appliances, measuring instruments and similar devices are another important part of the BFR flow in e-waste. Based on the data generated from import and export statistics as well as from literature data they represent roughly 25% of the DecaBDE flow in WEEE. The share for the other BFRs under study has been estimated to be pretty similar. According to estimates in the waste concept for Switzerland (BUWAL, 1992), 50% of the large household appliances (e.g. fridges, cooking stoves, washing machines and dish washers) were disposed as municipal solid waste in 1990. It has been assumed that this figure was constant in the 1980s throughout 1990. Starting that year, a steady decrease to 0.01 in 2005 has been estimated. No export has been assumed and the share between waste incineration and landfill is supposed to be according the statistics for burnable municipal solid waste. The proportion of appliances deposited as illegal dumping in e.g. forests has been estimated to be small and therefore assumed to be included in the dumping on controlled landfills. The mean TCs in the application area *E&E* calculated from the TCs of sWEEE (75% of the BFR flow in E&E) and large WEEE (25%) are shown in figure 14.

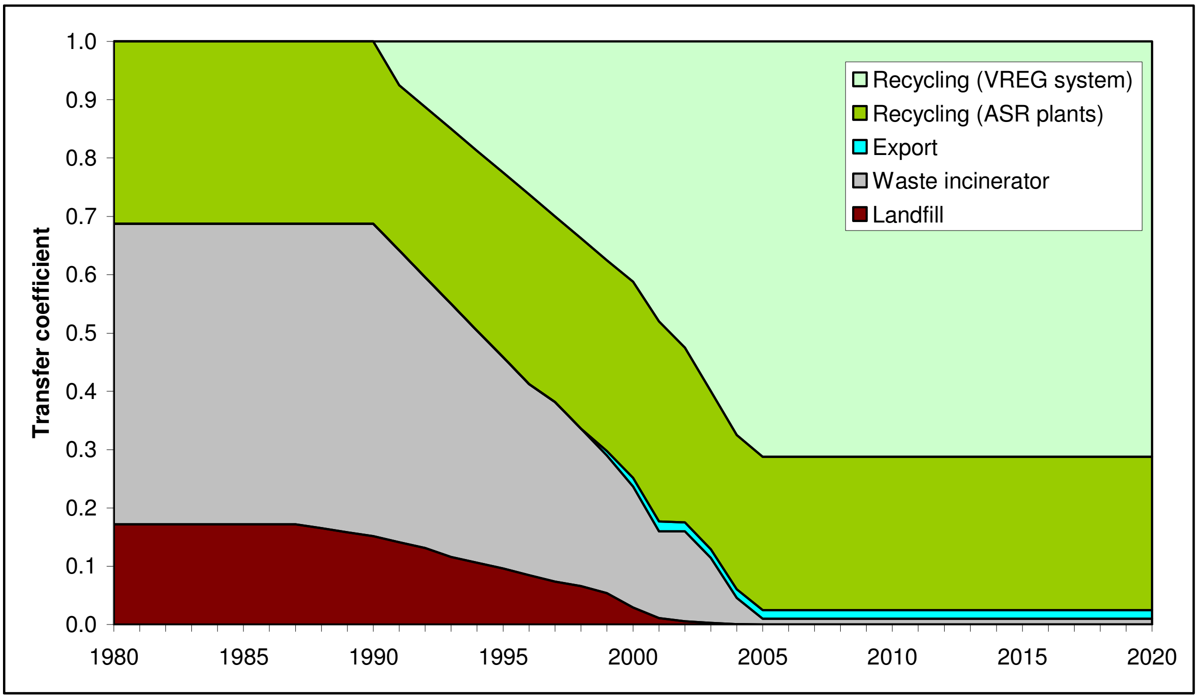


Figure 14 Transfer coefficients of e-waste at end of life in Switzerland (small and large appliances)

Transport

Cars and other vehicles at the end of their service life are either recycled or exported. According to Stiftung Auto Recycling Schweiz, the share of export on the entire vehicles (passenger cars) at end of life was strongly increasing from roughly 6% in 1988 to 50% in 2004. For 2005, a share of 44% was given. According to (Stiftung Auto Recycling, personal communication) most of the lorries at end of life are exported. Disused trains are about 40 years old and are recycled in Switzerland (SBB, personal communication). Because of the long residence time, only a small percentage of disused wagons have contained BFRs up to present. As opposite to trains, a large proportion of tramways and buses are exported at the end of life. Airplanes have been assumed to be mostly exported. Due to the fact that the BFR flows in passenger cars are by far bigger than the flows in the other vehicle types like lorries, buses, agricultural vehicles, caravans, motorbikes, trains, ships, airplanes and helicopters, the same share of export has been applied as for passenger cars for simplification reasons. For the future, a constant TC(U,recycling) of 0.56 has been assumed (see figure 15 and table 13).

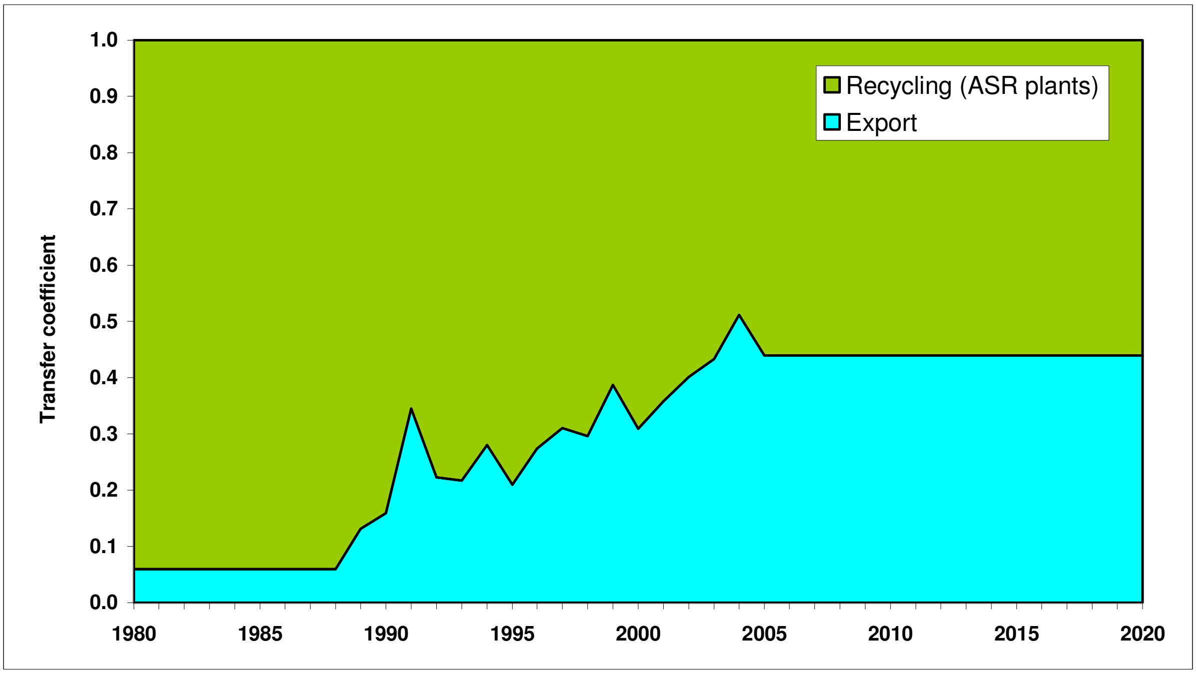


Figure 15 Share of export of entire vehicles at end of life in Switzerland (source 1988–2005: Stiftung Auto Recycling Schweiz)

Textile/furniture

The by far largest part of textiles such as upholstered furniture, furniture covers, mattresses, curtains, carpets, flexible foam components and protective clothing treated with flame retardants are either applied in combination with furniture or material for interior decoration. At the end of life, these products are collected as cumbersome waste from building demolition (coated textiles for interior decoration), or by the municipal solid waste collection system (furniture upholstery and interior decoration). The textiles in the interior decoration of vehicles make part of the application area *transport* and go to ASR treatment plants. The other textile products have been assumed to be included to 100% in the cumbersome waste. This waste fraction is disposed in the same manner as burnable waste fraction according to the national waste statistic from FOEN (BUWAL, 1998; BUWAL, 2003): 1990: 22% to landfill / 78% to waste incineration; 1996: 20% to landfill / 80% to incineration. Before 1990 the ratio was approximately ¼ to landfill, ¾ to incineration. At the end of the nineties the ratio decreased drastically, so that in 2005 there was no more direct landfilling of untreated waste (see figure 16). It has been assumed that for this kind of products there is no recycling at all during the time period of 1995 up to 2005. Textile recycling activities that have started at the beginning of the nineties may concern protective clothing to some extend. But as these applications are not of particular importance, the recycling path for this part was neglected.

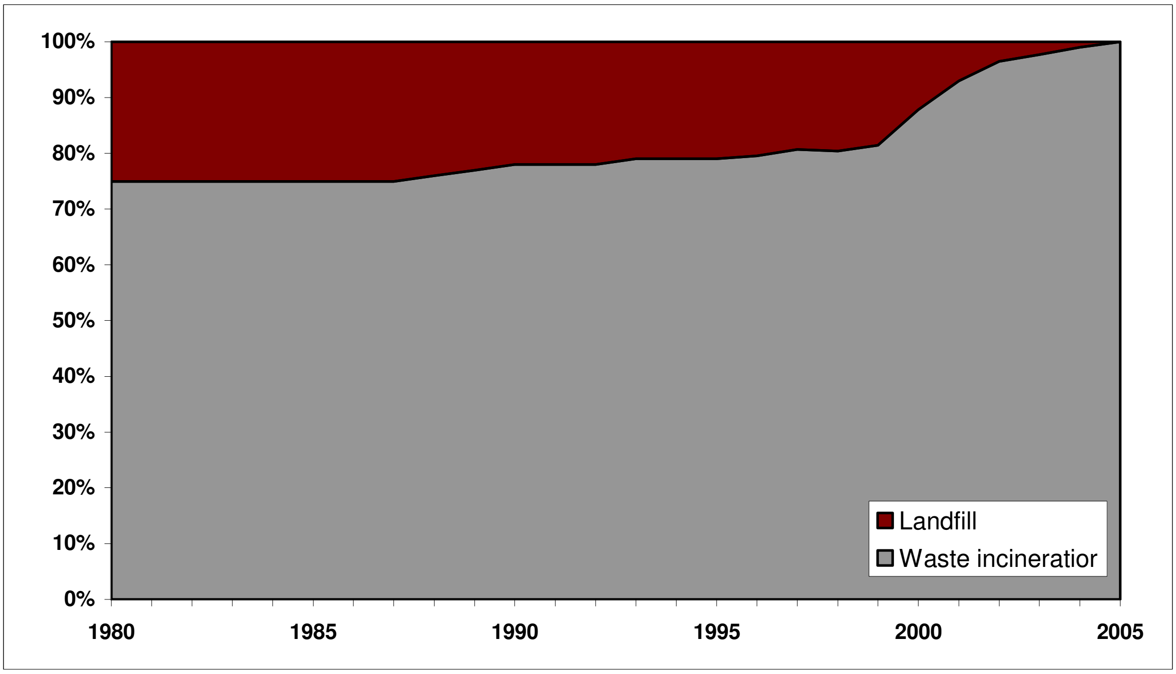


Figure 16 Trend of the share of landfilled and incinerated waste of total burnable solid waste in Switzerland from 1980 to 2005

Construction

After their residence time in the process *use*, the construction materials are being deconstructed. Hence, the total of this substance flow is directed to the process *deconstruction*.

Summary

The transfer coefficients used as parameters from the model for the process *recycling* are summarized in the table below. The TCs to export have been defined as the remainder to 1 and are not listed. Note that there is not necessarily a linear trend for the intermediate years not listed in the table. The releases to sewerage are given in section 2.4.2.5.8.

Table 13 Transfer coefficients applied in the dynamic SFA model for the process *use*

	Substance	Application area	Year	Value	Uncertainty
TC(U,deconst)	all	E&E, trans, tex.	independent	0	-
TC(U,deconst)	all	construction	independent	1	-
TC(U,recycling)	all	E&E	1980–1990	0.31	±30%
TC(U,recycling)	all	E&E	2005–2020	0.98	±30%
TC(U,incineration)	all	E&E	1980–1987	0.52	±30%
TC(U,incineration)	all	E&E	2005–2020	0.01	±30%
TC(U,landfill)	all	E&E	1980–1987	0.17	±30%
TC(U,landfill)	all	E&E	2005–2020	0	±30%
TC(U,recycling)	all	transport	1980–1987	0.95	±30%
TC(U,recycling)	all	transport	2006–2020	0.50	±30%

TC(U,incineration)	all	textile	1980–1987	0.75	±30%
TC(U,incineration)	all	textile	2004–2020	0.99	±30%
TC(U,landfill)	all	textile	1980–1987	0.25	±30%
TC(U,landfill)	all	textile	2004–2020	0.01	±30%

2.4.2.4.5 Deconstruction

The transfer coefficients used in the process *deconstruction* are *TC(D,landfill)* and *TC(D,recycling)*, which describe the proportion of substances in demolition waste that get disposed on landfills and to recycling facility, respectively. The remainder minus the emissions to the environment characterizes the proportion that is incinerated.

Figure 17 Construction waste containing different fractions such as concrete or XPS boards (source: Empa)

Construction material that is treated with flame retardants is mainly insulating foams and panels, plastic sheeting, vapour barriers as well as epoxy resins and polycarbonates. These construction materials are assigned to the burnable construction waste fraction. The disposal routes of burnable construction waste fraction in Switzerland from 1980 to 2010 are shown in table 14. It demonstrates the shift from landfilling to incineration. It also shows the rather high amounts that have been expected to be disposed uncontrolled during the eighties until the mid-nineties, according to BUWAL (1998). For 2002 this share has been estimated to be only 1%.

Table 14 Waste disposal routes of the burnable construction waste fraction in Switzerland from 1980 to 2010. The recycling rates of EPS and XPS insulation panels are expected to be higher (see text). *own estimate for illegal disposal recycling results in waste incinerator share of 93% instead of 98%

	Data source	Recycling	Waste incinerator	(Controlled) landfill	Uncontrolled/ illegal disposal	Uncertainty
1980	Own estimate	0%	5%	48%	47%	±50%
1992	BUWAL, 1998	-	14%	43%	43%	±30%
1994	BUWAL, 1998	-	33%	10%	47%	±30%
1996	BUWAL, 1998	-	42%	6%	46%	±30%
2002	BUWAL, 2002*	4%	93%	2%	1%	±30%
2005	Own estimate	10%	88%	1%	1%	±30%
2010	Own estimate	20%	78%	1%	1%	±30%

No intended recycling of this waste fraction did occur up to this century. The percentage of plastic construction materials being recycled could not be determined accurately. The competent authority supposes that the biggest part of plastic construction materials accruing during renovation or deconstruction operations is thrown in troughs and incinerated consecutively. Recycling may occur to some extent (R. Quartier, FOEN, personal communication). According to Kunststoff-Recycling Lenzburg (personal communication), there is a recycling of plastic construction waste (also when treated with flame retardants) taking place, if the material is pure enough and there is a customer of the recycled material. Especially on large construction sites, some recycling activities have been expected to occur. It was therefore assumed that from the beginning of this century recycling occurs to some small extend (1%) and reaching a proportion of 20% in 2010. The proportion of HBCD recycled is higher than for material containing other BFRs. The recycling percentage of HBCD used in EPS insulation panels was expected to be 30% in 2005. A further increase to 60% in 2010 was supposed (Carbotech AG, 2000). As EPS-Verband Schweiz (including EPS-Recycling Schweiz) was reluctant to answer questions, it has been assumed that this expectation made five years ago is still appropriate and the same recycling rate also applies for XPS panels.

For simplification reasons, the illegal disposal was accounted to (controlled) landfills. The influence of this simplification has been estimated to be small, as the material illegally disposed is normally underground (below the construction) or at least covered with soil. A difference to regular landfills might be the fact that the leachates are not captured to the same extent.

2.4.2.4.6 Recycling

Due to data gaps, rough estimates have been done to determine this TC for the different application areas. Some electronic appliance recycling facilities operate without generating wastewater (e.g. like Immark AG), while others discharge wastewater into the sewerage system. Some work steps of scrapped car recycling are done outdoors, including the intermediate storage of certain goods. The sites are drained into the sewer system, after the particles have been removed in sludge traps (Mr. Wilde, Metso Lindemann GmbH, Düsseldorf, personal communication). It is known that there are losses to sewerage, but the amounts are not known and there is no data that allows estimating appropriately. It was thus assumed that losses were the same as to the air, where data is available (Sakai et al., 2006c). In addition to site drainage, it was though that releases occur from cleaning the facilities, washing the worker's clothes and hands. The loss from recycling facilities into sewerage system is denoted in *TC(R,sew)*. This TC also decreases with technical innovation and the reduction of atmospheric emissions (see section 2.4.2.5.6).

There are four different directions in the anthroposphere where substances can be led after leaving the recycling facility. Three of them are listed in table 16. The fourth, the proportion of the substances leaving the recycling facility to be incinerated was defined as the remainder. There are output fractions that are reused in the production, *TC(R,reuse)*, while contaminated metal fractions from demolition waste recycling have been considered as export to abroad. A proportion of the fractions were also dumped on landfills.

E&E

For the application area *E&E*, there are two different recycling disposal routes: in specialised appliance recycling facilities and together with cars in ASR plants (see section 2.4.2.4.4). To avoid introducing a second recycling process, the TCs have been averaged according to the substance flows. As stated above, it has been estimated that approximately 75% of the substance flows in e-waste are recycled in small WEEE and the remaining 25% in large appliances. The TCs in the specialised recycling plants for small WEEE have been determined by Morf et al. (2005). As illustrated in table 15, the TCs to plastic and metal fractions were found to be different for the individual BFRs, the TCs into the different flows in the model are also different for each BFR.

Table 15 Transfer coefficients determined in a small WEEE recycling facility by Morf et al. (2005)

	DecaBDE	OctaBDE	PentaBDE	HBCD	TBBPA (additive)
Plastic fractions	0.91	0.79	0.84	0.85	0.92
Metal fractions	0.09	0.21	0.16	0.15	0.08

The total plastic fraction is incinerated, while the metal fraction is used in the steel industry to recover the metal. The latter substance flow was treated as an export to abroad (metal trade) in the model, even though a substantial part was thought to be recycled in Switzerland. The reason is to avoid introducing an additional process, as metal recycling does not have similar TCs as WEEE recycling or as incineration. As the amounts of BFRs in metal fractions recycled in Switzerland have been expected to be rather small, this simplification has not been supposed to have a big influence on the result. There has been no landfilling or export of the plastic fraction from the small WEEE recycling plants since the start in 1991. The total of this fraction has been incinerated.

It has been assumed that the TCs in large electronic appliances recycling plants are similar as for automobile recycling taking place at the same facilities (see next section). The TCs of small and large e-waste are averaged based on the substance flows to these two recycling processes (see figure 18).

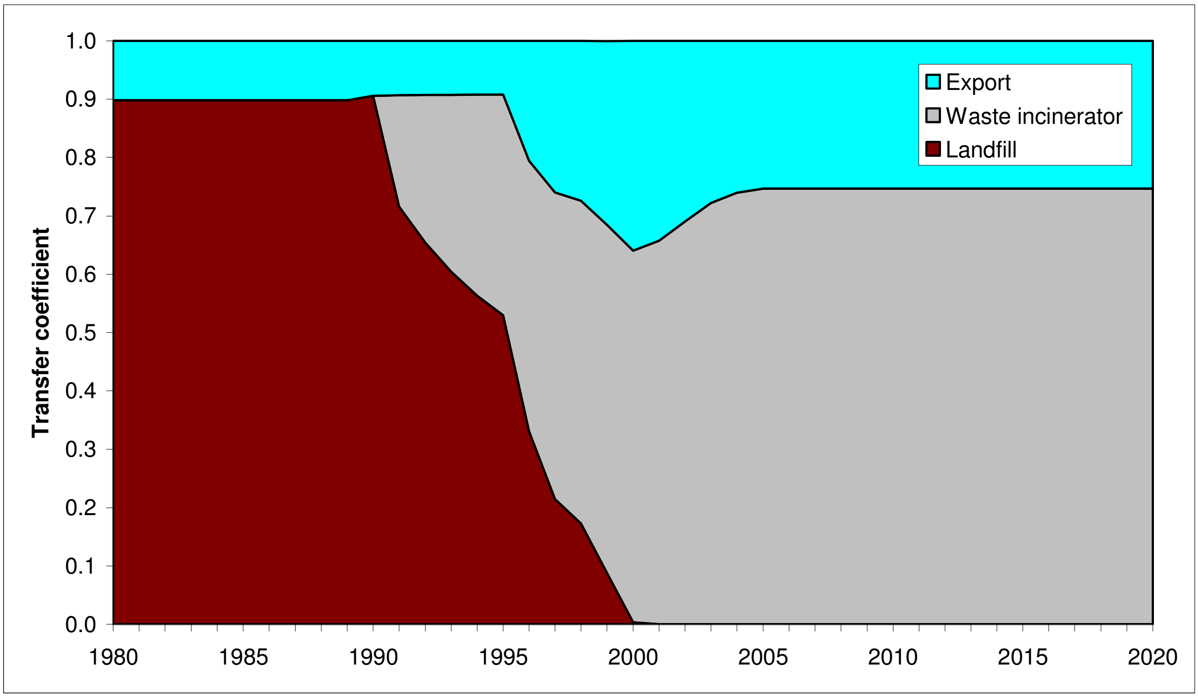


Figure 18 Transfer coefficients in small and large e-waste recycling plants for DecaBDE (averaged over both types)

The figures for HBCD and PentaBDE are not shown, as they are comparable. The transfer coefficient *TC(R,exp)* for HBCD was 0.08 higher during the 1980s, which was reduced to 0.05 since the beginning of the present decade. The other two TCs were proportionally lower. For PentaBDE, the TCs estimated are almost identical to the ones for HBCD.

Transport

Stiftung Auto Recycling Schweiz has supplied data about disposal routes of automobile shredder residue (ASR) back to 1994. Automobile shredder residue is waste material left after automobile and major appliance (refrigerators, ovens etc.) have been crushed and shredded to recover metals and non-metals. According to Stiftung Auto Recycling Schweiz, the share of automobile waste in ASR is about 50% in the 21st century. In the 1990s and 1980s the fraction is supposed to be slightly higher (it has been assumed to be 55% in the 1990s and 60% in the 1980s). The data show that in 1994 56,200 tons ASR was disposed 100% in landfills. In the year 2005, a total amount of 61,000 tons ASR was disposed. The total was transferred to incineration processes (1/3 in Switzerland and 2/3 abroad). The export share was about constant back to the year 2000. Before 2000 most ASR was disposed in Switzerland and it has been assumed that export was zero before 2000. The disposal routes between 1996 and 2001 change from 100% disposal in landfills towards 100% disposal in incineration processes. Further it has been assumed that before 1994 all ASR was disposed in landfills in Switzerland.

According to Stiftung Auto Recycling Schweiz, the ASR fraction of end of life automobiles was about 26% in 2000. Since the plastic fraction in automobiles has approximately doubled from 8% to 16% in the last 10 to 15 years, it has been assumed in a first-order approximation that the ASR fraction in the 1980s was roughly 13%, in the 1990s roughly 20% and in this century roughly 26% of the entire automobile amount at the end of life. The other parts (difference to 100%) are recycled in steel industry (metal fraction). This substance flow was treated as an export to abroad (metal trade) in the model, even though a substantial part was thought to be recycled in Switzerland. The reason was to avoid introducing an additional process, as metal recycling does not have similar TCs as automobile recycling or as incineration (see previous section).

The TCs have been estimated from the TCs determined with measurements of concentrations in different e-waste fractions carried out in a Swiss small e-waste recycling plant by Morf et al. (2005). The behaviour of BFRs in automobile shredder plants might not be the same as in a modern e-waste recycling facility, but it has been estimated that the substance flows are still comparable and there is no study for the substance flows in automobile shredder plants available. The transfer of BFRs in metal fractions has been weighted with the trend of percentage w/w of the metal fraction (100% minus ASR fraction) and treated as an export as stated above. As shown in figure 19, the transfer into burnable waste fractions has been split up into transfer coefficients to landfills *TC(R,landfill)*, incinerator plants *TC(R,inc)* and export *TC(R,exp)*. The latter thus contains both metal fractions and the proportion of the burnable waste exported.

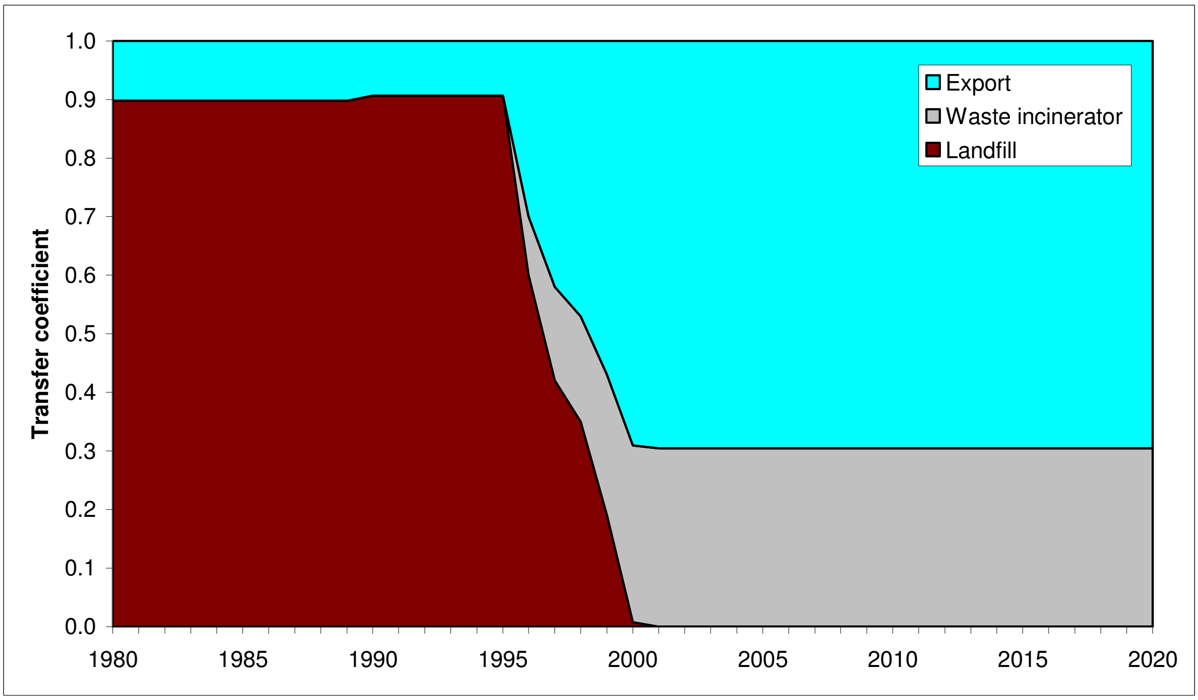


Figure 19 Transfer coefficients in automobile recycling plants for DecaBDE

The figures for HBCD and PentaBDE are not shown, as they are comparable. The transfer coefficient *TC(R,exp)* for HBCD was 0.08 higher during the 1980s, which was reduced to 0.02 since the beginning of the present decade. The other two TCs were proportionally lower. For PentaBDE, the TCs estimated are almost identical to the ones for DecaBDE.

Textile/furniture

No recycling of textiles and furniture has been considered in the model.

Construction

Material recycling started at the beginning of this century. It has been assumed that 20% of the BFRs in construction material collected for recycling could not be reused in production and are therefore incinerated. The remaining 80% (minus small proportions of emission to atmosphere and losses to sewer) were regranulated and reused in production processes. The losses to sewer have been estimated to be equal as in the process *production*.

Summary

The transfer coefficients used as parameters from the model for the process *recycling* are summarized in the table below.

Table 16 Transfer coefficients applied in the dynamic SFA model for the process *recycling*

	Substance	Application area	Year	Value	Uncertainty
TC(R,sew)	all	construction	dependent	Equal TC(P,sew)	TC/10 – TC·10
TC(R,sew)	all	E&E	dependent	see sect. 2.4.2.5.6	-
TC(R,sew)	all	transport	dependent	see sect. 2.4.2.4.6	-
TC(R,sew)	all	textile	-	-	-
TC(R,reuse)	DecaBDE, HBCD	construction	independent	8.0E–01	±50%
TC(R,reuse)	PentaBDE	construction	independent	1.0E–01	±50%
TC(R,reuse)	all	E&E	independent	0	-
TC(R,reuse)	all	transport	independent	0	-
TC(R,reuse)	all	textile	-	-	-
TC(R,landfill)	all	construction	independent	0	-
TC(R,landfill)	all	E&E	dependent	see figure 18	±30%
TC(R,landfill)	all	transport	dependent	see figure 19	±30%
TC(R,landfill)	all	textile	-	-	-
TC(R,export)	all	construction	independent	0	-
TC(R,export)	all	E&E	dependent	see figure 18	±30%
TC(R,export)	all	transport	dependent	see figure 19	±30%
TC(R,export)	all	textile	-	-	-

2.4.2.4.7 Incineration

In municipal solid waste incineration plants (MSWIP), two output goods are produced apart from emissions into the environment: bottom ash and air pollution control (APC) residues. According to Frey and Frischknecht (1989) approximately half of the bottom ash (including a small percentage of fly ash) was used for road construction in 1998. No new data on this issue could be found in the literature. Criteria for the residues were defined in an ordinance coming into operation in 1988. Therefore, the amounts of bottom ash used for road construction have been considered as if they would have been dumped on landfills. Residues of MSWIP air pollution control systems have been landfilled in Switzerland and exported to Germany as the most common end-of-life options. In 1989, approximately 3000 tons were expected to be exported (Frey and Frischknecht, 1989). This is roughly 18 weight-% of the total APC residue amount produced each year in Switzerland's MSWIP at that time. In 1996, the export share was roughly 50% (BUWAL, 1998). In the years 2001 and 2002 the export share was 60% and 62% (for sum of fly ash and APC residues).

From a study in Japan (Tamade et al., 2002), the transfer coefficients of PBDEs and TBBPA into bottom ash *TC(I,bot.ash)* and APC residues *TC(I,APC res.)* have been calculated. As the input substance flow was not known exactly, large uncertainties were associated with the results. For the sum of PBDE, the TC into bottom ash ranged from 2.8E–04 to 5.5E–03, whereas the range was from 3.3E–04 to 6.6E–03 for APC residues. For TBBPA, the ranges were 1.1E–03 to 7.1E–05 and 5.5E–04 to 3.6E–05. Much lower TCs have been calculated from results of a Norwegian study (Borgnes and Rikheim, 2004a; Borgnes and Rikheim, 2004b). The TC into bottom ash for the sum of BFRs derived ranged from 2.4E–06 to 8.0E–06, while the TC into APC residues was 2.0E–07.

In 2006, own measurements were commissioned in a Swiss incineration plant. Nearly all of the concentrations were below the limit of quantification. Only in one of four pooled bottom ash samples, the concentration of DecaBDE (80 µg/kg), OctaBDE (12 µg/kg) and TBBPA (12 µg/kg) could be quantified. The TCs of DecaBDE derived thereof was 7.4E–04. As the concentration was below the limit of quantification in the three other samples, the TC there was less than 9.3E–05. For PentaBDE and HBCD the TCs into bottom ash were less than 5.4E–04 and 7.6E–05, respectively. The derived TCs into APC residues for DecaBDE, PentaBDE and HBCD were less than 1.7E–05, 1.0E–04 and 1.4E–05, respectively.

As the TCs from the studies in Japan and Norway differed by at least one (bottom ash) to more than three orders of magnitude (APC residues) and only the sums of PBDEs and BFRs, respectively, were investigated, the maximum TCs of the Swiss incineration plants (8.0E–06 and 2.0E–07) seem to be the most appropriate estimates (see table 17). The TCs have been considered as constant over the whole period, as (a) no big changes were expected and (b) the impact of these flows for emissions to environment was expected to be rather small.

Table 17 Transfer and proportion coefficients applied in the dynamic SFA model for the process *incineration*

	Substance	Application area	Year	Value	Uncertainty
TC(I,bot.ash)	all	all	all	8.0E–06	TC/10 – TC·10
TC(I,APC res.)	all	all	all	2.0E–07	TC/10 – TC·10
PC(I,landfill)	all	all	1989 and before	0.82	±30%
PC(I,landfill)	all	all	1996	0.50	±30%
PC(I,landfill)	all	all	2001	0.40	±30%
PC(I,landfill)	all	all	2002	0.38	±30%
PC(I,landfill)	all	all	2005	0.25	±30%
PC(I,landfill)	all	all	2020	1.00	–30%

2.4.2.4.8 Sewerage

Neither transfer coefficients, nor proportion coefficients are used to model the process *sewerage*. The estimation of the atmospheric deposition that enters the sewer system is described in section 2.4.2.3. For the emission factors, please refer to section 2.4.2.5.8.

2.4.2.4.9 WWTP

There are three ways to dispose of the sewage sludge: usage as a fertilizer in the agriculture (called "biosolid" in the USA), dumping on landfills and incineration in MSWIPs, sludge incineration plants or in cement works. According to data by Külling (2002), the share of agriculture in Switzerland was 65% in 1980. It decreased to 50% in 1985 and 38% in 2000 (Laube and Vonplon, 2004). After that year a strong decrease took place. Beginning October 1st 2006, the usage of sludge in the agriculture was prohibited in principle. If a canton extends the transitional period, there is the possibility of a further usage in the agriculture (vegetable and feed crops excluded; appendix 2.6 paragraph 5.3 ChemRRV). Therefore, the share has been supposed to be very small in 2010, when just some exceptions of small WWTP in rural areas contribute to this share (A. Laube, FOEN, personal communication). The fraction of sludge dumped on landfills was 17% in 1994, decreased to 1% in 2004 and will stay on a very low level in future due to the ban on dumping burnable waste. There are no data available for the period before 1994 that distinguish between the fraction dumped on landfills and incinerated. As burning the sludge in MSWIPs, sewage incineration plants or cement works is not profitable, a decrease to a very small share back to 1980 has been estimated. Sewage incineration plants were put into operation around this year (e.g. Pro Rheno Basel in 1982). Therefore, the biggest part of the sludge not used in the agriculture was dumped on landfills. A linear increase starting from 1994 back to the mid 1980s has been assumed (see figure 20).

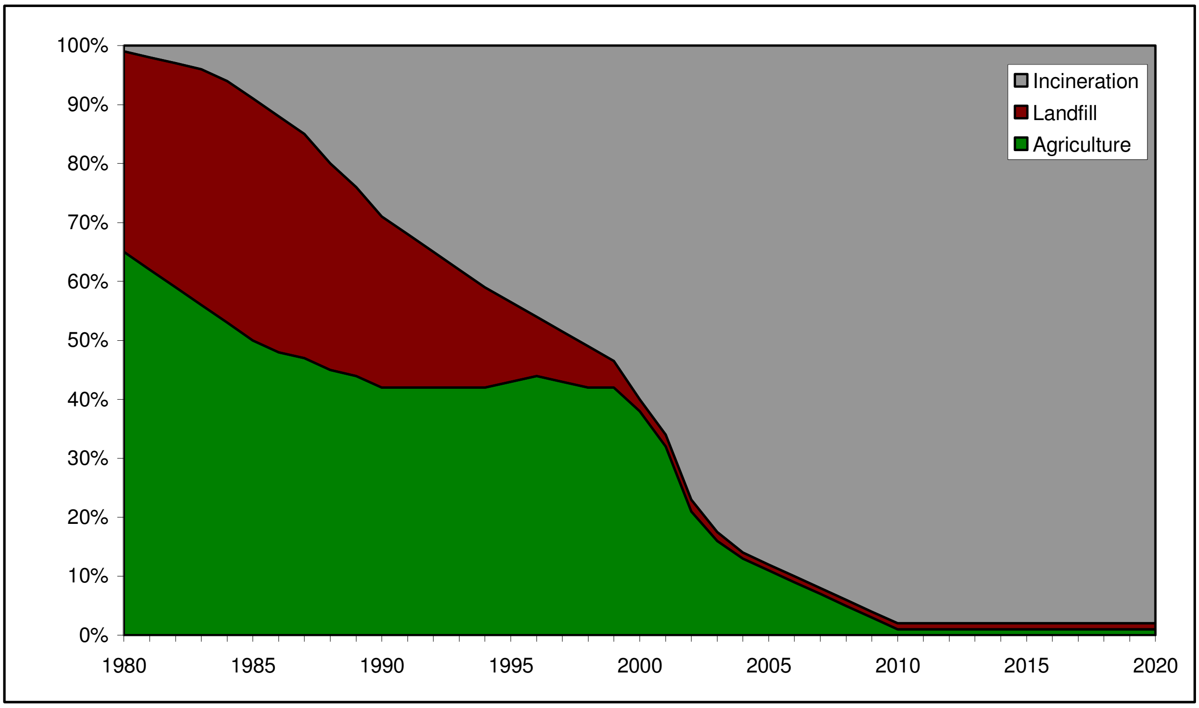


Figure 20 Percentage of sewage sludge usage in agriculture, deposition in landfills and incineration (sources: Külling (2002), Laube and Vonplon (2004), Laube (personal communication) and own estimates)

The transfer coefficient *TC(W,sludge)* expresses which proportion of a substance entering the WWTP will adsorb to the sludge (see table 18). During the sewage treatment process, loss of PBDEs by either volatilization or degradation during treatment is expected to be negligible. Partitioning to and removal with solids is, therefore, expected to be the only significant removal process. Rayne and Ikonomou (2005) measured that DecaBDE partitions to an extent of 97.3% into sludge and as the biological degradation was negligible, the rest enters the effluent. For the congeners BDE-47, BDE-99 and BDE-183, the TCs were measured as 89%, 95% and 92.4%, respectively. EPIWIN calculates for PentaBDE, BDE-183 and DecaBDE as well as for HBCD the same coefficient (93.2%), if no K_{OW} is put in manually. In a study published recently (Song et al., 2006), the proportion of the congeners BDE-47 and BDE-99 partitioning into sludge (sum of primary and waste activated sludge) were found to be 89.7% and 91.2%. Anderson and MacRae (2006) estimated that about 94% of the PBDEs in the influent were removed with the sludge. But due to the use of grab samples and heterogeneity, the mass balance did not match well. Therefore, the results from the two other studies have been taken into account. Where available, the mean of these two numbers has been used for the model, even though the fact that the coefficient is slightly lower for the higher brominated BDE-183 as compared to BDE-99 was surprising.

For HBCD and TBBPA, no measured data on partitioning was available. The only publication with both influent and effluent concentrations of HBCD and TBBPA is Morris et al. (2004). But due to the large variations and thus the big standard deviations, it is not possible to estimate an appropriate TC from these data. When a log K_{OW} of 5.625 (MacGregor and Nixon, 1997) used in the EU Risk Assessment draft is put in, EPIWIN calculates a total removal 89.7% for HBCD. This value was taken for *TC(W,sludge)* for HBCD. The proportion of TBBPA that is found in the effluents seems to be higher than for PBDEs and HBCD. This is reasonable as TBBPA partitions more into the water phase (log K_{OW} = 5.9) und has two conjugate bases (pK_{a1} = 7.5, pK_{a2} = 8.5) (ECB, 2006b), what increase the water solubility.

The domain of uncertainty has been assumed to ±50% of the fraction released into the effluent.

Table 18 Transfer coefficients applied in the dynamic SFA model for the process *WWTP*. For the proportion coefficients see figure 20. The uncertainty is defined relative to the transfer coefficient into the effluent.

	Substance	Application area	Year	Value	Uncertainty
TC(W,sludge)	DecaBDE	all	independent	9.7E–01	±50% (1 – TC)
TC(W,sludge)	HBCD	all	independent	9.0E–01	±50% (1 – TC)
TC(W,sludge)	BDE-47	all	independent	8.9E–01	±50% (1 – TC)
TC(W,sludge)	BDE-99	all	independent	9.3E–01	not modelled
TC(W,sludge)	BDE-183	all	independent	9.2E–01	not modelled
TC(W,sludge)	TBBPA	all	independent	lower (pK_a)	not modelled
PC(W,agri)	all	all	dependent	see figure 20	±30%
PC(W,landfill)	all	all	dependent	see figure 20	±30%

2.4.2.4.10 Landfill

In several investigations around the world, concentrations of BFRs in landfill leachates have been measured. Regional differences, which are in line with the BFR consumption, waste management type and measurements in other environmental media, seem to exist. But as the results are highly variable, there is no clear evidence. The variability might also be due to the type and age of the landfill, the waste dumped, climate, seasonal changes and the concentration of suspended solids and dissolved humic matter. Osako et al. (2004; 2005) determined the proportion of PBDEs in leachates that were freely dissolved, adsorbed to suspended solids and in a complex with dissolved humic matter. The proportion of PBDEs with four and more bromine atoms freely dissolved was less than 10%. On the other hand, more than half of TBBPA was freely dissolved. Therefore, the concentration of organics in the leachate strongly influences the leaching of BFRs, what was also demonstrated in lab scale experiments (Hamre, 2004; Kim et al., 2006).

Intermediate results of an investigation with an interesting experimental carried out in Canada were presented on the DIOXIN Conference (Danon-Schaffer et al., 2006). Leachates were collected from different disposal cells of a landfill dedicated to handling waste during five time periods: 1980–1984, 1985–1989, 1990–1994, 1995–1999 and 2000–2005. The concentration of PBDEs in the leachate was found to be increased from the first to the last disposal period by more than two orders of magnitude. This finding could be due to a lower mean content in older E&E appliances or the fact that the fraction of available PBDEs had already leached out in the older disposal cells. Waste electric and electronic equipment items representing the same periods were analyzed for the content of PBDEs. In contrast to the leachates, the concentration in e-waste provided by a recycling company decreased by almost six orders of magnitude from the second to the last period. The concentration of PBDEs in e-waste from the period 2000–2005 was 14 ng/g, which is comparable to concentrations in alpine soils (Knoth et al., 2007), while it was 1.2% w/w for the period 1985–1989. Hence, it seems that either the representativeness of the e-waste samples was not given.

For DecaBDE, the value most applicable to Switzerland was thought to be the 4.6 ng/l measured in Japan (Osako et al., 2005). In addition, this value corresponds well with the geometric mean (4.7 ng/l) of leachtates measured in Canada (Chiu et al., 2006). In another study carried out in North America, higher concentrations of PBDE were found (Oliaei, 2005). As DecaBDE consumption and the waste management in Japan are more similar to Switzerland than to North America, the result from the latter study was not taken into account. For HBCD, there are only two studies. Concentrations found in Swedish landfill leachates were 3 and 9 ng/l (Remberger et al., 2004). In a study investigating the particulate phase of landfill leachates from Ireland, England and the Netherlands, concentrations between 2.5 and 36,000 ng/g d.w. were found (Morris et al., 2004). Because of the large range and the fact that only the particulate phase (unit ng/g d.w.) was measured, the mean of the measurements carried out in Sweden have been used for estimating the TC. For the two PentaBDE congeners BDE-47 and BDE-99, the concentration in leachates obtained in Austria (Moche et al., 2004) seemed to be most applicable for Switzerland. For BDE-99, the concentration range is approximately similar to measurements in Canada, but higher than in Japan, where PentaBDE usage stopped earlier than in Europe. For BDE-47, the concentration range is somewhat higher than in Canadian and Japanese studies. As the measurements do rather have a log-normal than a normal distribution, it has been assumed that the geometric mean is most appropriate to estimate the releases from landfills. The geometric means of the concentrations obtained in Austria, 16 and 9 ng/l for BDE-47 and BDE-99, have been used for estimating the TC.

Table 19 Concentrations in landfill leachates (ng/l)

Data source	PBDEs	BDE-209	BDE-47	BDE-99	BDE-183	HBCD	TBBPA
Osako, 2004		<0.2–<50	<0.002–0.017	<0.002–<0.5	<0.012–0.065		<0.5–620
Osako, 2005		4.6	2.9[a]	1.0[a]	2.4[a]		25
Oliaei, 2005	29–248	60–98%					
Chiu, 2006		0.6–37	0.5–121	0.06–240	<LQ–7.6		
Danon, 2006	2.7–1470	2.7–57	0.04–6.5[a]	0.3–740[a]	0.5–17.8		
Moche, 2004			1.9–360	0.66–410	0.5–96		
Remberger, 2004						3–9	
Morris, 2004						2.5–36000[b]	<0.3–320[b]

[a] TetraBDE, PentaBDE and OctaBDE, respectively, [b] particulate phase (ng/g dry weight)

As a transfer coefficient *TC(L,leach)* was needed for the model, the concentrations in landfill leachates have been calibrated with the volume of leachates per year and the stock of BFRs on landfills. The volume estimate was done as follows: The area of sanitary landfills was estimated as 300 ha, the area of other types of landfills such as domestic refuse dumps and multifills was estimated as 10,000 ha. The leachate flow rate was estimated as 0.16 l/s/ha (von Arx, 2006). The annual leachate volume thus is approximately 5.0E10 l/year. This volume is about 3% of the wastewater volume. The best estimate BFR substance flows are therefore 0.23 kg/year DecaBDE, 0.30 kg/year HBCD, 1.3 kg/year BDE-47 and 0.81 kg/year BDE-99. The domain of uncertainty is approximately one order of magnitude for all substances, but the substance flows are relatively small, compared to others.

In a first run of the model, the current stocks on landfills have been calculated. The DecaBDE stock in 2005 amounts for 360 tons, whereas it was approximately 87 tons for HBCD and 5.5 tons for BDE-47. The TCs calculated thereof are 6.8E–07/year, 3.4E–06/year and 1.4E–04/year, respectively. The proportion coefficient *PC(L,sew)* denotes the percentage of landfill leachates caught and introduced to sewer system. A value of 0.9 was estimated for the beginning of this century (D. Wirz, Meier & Partner AG, personal communication; von Arx, 2006). It has been assumed that the connection proportion was 0.2 in 1980, followed by a linear increase to 0.9 in 2001.

Table 20 Transfer and proportion coefficients applied in the dynamic SFA model for the process *landfill*

	Substance	Application area	Year	Value	Uncertainty
TC(L,leach)	DecaBDE	all	independent	6.8E–07/year	TC/10 – TC·10
TC(L,leach)	HBCD	all	independent	3.4E–06/year	TC/10 – TC·10
TC(L,leach)	BDE-47	all	independent	1.3E–04/year	TC/10 – TC·10
PC(L,sew)	all	all	1980	2.0E–01	±50%
PC(L,sew)	all	all	2001–2020	9.0E–01	±50% (truncated at 1)

2.4.2.5 Emission factors in the anthroposphere

As transfer coefficients, emission factors (EFs) are parameters used in the dynamic SFA model. EFs describe the proportion of an inflow that is emitted to atmosphere, hydrosphere or soil. In the processes *use* and *landfill*, EFs are defined as the mass fraction of the stock that is annually released.

2.4.2.5.1 Production

As stated earlier, no production of BFRs took or takes place in Switzerland. Hence, EFs from this process have not been used in the model for Switzerland. Direct emissions to hydrosphere and soil have been considered zero. The releases to wastewater are denoted as TCs (see 2.4.2.4). There was no information on the temporal trend of atmospheric EFs from production processes. Therefore, the recently determined EFs have been assumed to be appropriate from the beginning of the present decade until the end of the period under study. It has been estimated that the EFs were higher in the 1980s and 1990s. The Ordinance on Air Pollution Control that was enacted in 1986 (LRV, 1985) probably also had an effect on the emissions of BFRs, even though they were not target substances. It has therefore been assumed that the EFs were reduced from 1980 until 1986 by 50%. A further linear decrease by 50% has been assumed between 1986 and the beginning of the present decade. The EFs for the year 1980 have thus been assumed four times higher than at present.

DecaBDE

Using the specifications given in the *Usage Guide Document* and the *Technical Guidance Document*, atmospheric EFs between 2E−04 and 5E−04 from polymer processing were estimated for BFRs (Sakai et al., 2005). The Japanese Ministry of Environment has investigated nine plastics processing facilities (Sakai et al., 2006c). The EFs measured were in a range between 1.6E−09 and 1.4E−07, which is three to five orders of magnitude lower than the values estimated based on the *Usage Guide Document* and the *Technical Guidance Document*. As the sample size was low and the EFs from the different manufacturers showed a large variation and it was not feasible to distinguish between the acrylonitrile butadiene styrene (ABS) and the polystyrene facilities. The median of the measured values, 4.0E−08, therefore seems to be the best estimate for the application areas *E&E* and *construction*. The Japanese Ministry of Environment has also investigated the emissions from a textile processing facility (Sakai et al., 2006c). The EF derived from the measurements is 9.0E−07. As BFRs are in plastic parts and in textiles or foams in vehicles, the EF for the application area *transport* has been assumed as the mean of the EFs for *E&E* and *textiles/furniture* (see table 21).

HBCD

Atmospheric emissions of HBCD during formulation and industrial use are given in the 2006 draft version of the EU risk assessment report (ECB, 2006a). From these data EFs have been for the individual processes and subsequently for the four application areas for Switzerland. The calculated EFs of the formulation of beads for manufacture of EPS/HIPS and XPS production are 5.8E−05 and 6.1E−06, respectively. The emissions during the manufacturing process of EPS and HIPS have been derived as 3.0E−05. For XPS, the EF has been calculated as 5.8E−05. The calculated factor of the formulation of polymer dispersions for textile back-coating amounts to 1.9E−05. EFs of the industrial use of powder for XPS and for textile back-coating agents have been calculated as 5.4E−05 and 3.4E−07. It has been assumed that a fraction of 50% of the HIPS, EPS and XPS manufactured in Switzerland or HBCD for the domestic textile back-coating has also been formulated here. The EF from *E&E* has therefore been estimated as the sum of the emission factors from the formulation of beads for the manufacture of EPS/HIPS (50%) and the production of HIPS as calculated from the data in the RA draft report. The same has been done in an analogous way for the other application areas. The estimated emission factors are listed in table 21.

BDE-47

In a DIOXIN short paper (Sakai et al., 2006a), EFs of lower brominated PBDEs are given. However, as they are expressed in relation to the DecaBDE mass, they are not applicable for production steps, where polymers containing an OctaBDE or PentaBDE technical mixture are processed. In the EU risk assessment, an atmospheric EF of PentaBDE of 5.0E–04 (congener not specified) from polyurethane foam production was estimated using the guidelines of the *Use Category Document* (ECB, 2000). This value has been estimated to be possibly too high, because measurements for HBCD have shown that such methods might overestimate the emissions (Klatt, 2004), but it has nonetheless been used for the model. For *E&E* and *construction*, lower EFs have been estimated. Because of missing data, values between the EFs for DecaBDE and HBCD for the same applications have been assumed. As for DecaBDE, the EF for *transport* has been estimated as the mean of the EFs for *textile/furniture* and *E&E*.

Summary

The emission factors for the process *production* are summarized in the table below. A linear decrease has been assumed for the EFs of the years that are not included in the list.

Table 21 Emission factors applied in the dynamic SFA model for the process *production*

	Substance	Application area	Year	Value	Uncertainty
EF(P,atm)	DecaBDE	E&E, construction	1980	1.6E–07	EF/10 – 10·EF
EF(P,atm)	DecaBDE	E&E, construction	1986	8.0E–08	EF/10 – 10·EF
EF(P,atm)	DecaBDE	E&E, construction	2001–2020	4.0E–08	EF/10 – 10·EF
EF(P,atm)	DecaBDE	transport	1980	1.9E–06	EF/10 – 10·EF
EF(P,atm)	DecaBDE	transport	1986	9.4E–07	EF/10 – 10·EF
EF(P,atm)	DecaBDE	transport	2001–2020	4.7E–07	EF/10 – 10·EF
EF(P,atm)	DecaBDE	textile	1980	3.6E–06	EF/10 – 10·EF
EF(P,atm)	DecaBDE	textile	1986	1.8E–06	EF/10 – 10·EF
EF(P,atm)	DecaBDE	textile	2001–2020	9.0E–07	EF/10 – 10·EF
EF(P,atm)	HBCD	E&E	1980	2.4E–04	EF/10 – 10·EF
EF(P,atm)	HBCD	E&E	1986	1.2E–04	EF/10 – 10·EF
EF(P,atm)	HBCD	E&E	2001–2020	5.9E–05	EF/10 – 10·EF
EF(P,atm)	HBCD	transport	1980	1.4E–04	EF/10 – 10·EF
EF(P,atm)	HBCD	transport	1986	6.9E–05	EF/10 – 10·EF
EF(P,atm)	HBCD	transport	2001–2020	3.5E–05	EF/10 – 10·EF
EF(P,atm)	HBCD	textile	1980	3.9E–05	EF/10 – 10·EF
EF(P,atm)	HBCD	textile	1986	2.0E–05	EF/10 – 10·EF
EF(P,atm)	HBCD	textile	2001–2020	9.8E–06	EF/10 – 10·EF
EF(P,atm)	HBCD	construction	1980	3.1E–04	EF/10 – 10·EF

EF(P,atm)	HBCD	construction	1986	1.5E–04	EF/10 – 10·EF
EF(P,atm)	HBCD	construction	2001–2020	7.7E–05	EF/10 – 10·EF
EF(P,atm)	BDE-47	E&E, construction	1980	2.0E–05	EF/10 – 10·EF
EF(P,atm)	BDE-47	E&E, construction	1986	1.0E–05	EF/10 – 10·EF
EF(P,atm)	BDE-47	E&E, construction	2001–2020	5.0E–06	EF/10 – 10·EF
EF(P,atm)	BDE-47	transport	1980	1.0E–03	EF/10 – 10·EF
EF(P,atm)	BDE-47	transport	1986	5.1E–04	EF/10 – 10·EF
EF(P,atm)	BDE-47	transport	2001–2020	2.5E–04	EF/10 – 10·EF
EF(P,atm)	BDE-47	textile	1980	2.0E–03	EF/10 – 10·EF
EF(P,atm)	BDE-47	textile	1986	1.0E–03	EF/10 – 10·EF
EF(P,atm)	BDE-47	textile	2001–2020	5.0E–04	EF/10 – 10·EF

2.4.2.5.2 Trade

No emissions from the process *trade* have been included in the model.

2.4.2.5.3 Construction

Emissions to atmosphere from construction operations (i.e. installing of flame-retarded insulation material and sheeting) have been included to the model. As most of DecaBDE has been used in plastic sheeting or vapour barriers that are not assumed to generate any relevant emissions, the emission factor has been defined as zero. Estimates on the emissions of HBCD during *construction* were provided in the EU risk assessment draft report (ECB, 2006a). These activities were assumed to take place outdoors or at open building sites. Therefore, the particles that are small and relatively light are released to the surrounding environment. The weight of the particles being formed at sawing of XPS and EPS boards was measured. The release of particles from sawing of XPS boards was estimated to be 5.8 g particles per ton XPS. The corresponding figures for sawing EPS boards were 445 g particles per ton. However, it was emphasized that it is more common to make a cut in the board with a sharp knife and then to break the board or – at large sites – to cut the boards with a hot wire. Both methods of cutting EPS boards result in much less particles being formed as compared to sawing. Based on these data, an atmospheric emission of 50 g particles per ton EPS board has been estimated. For XPS boards, it has been assumed that half of the generated particles are emitted to the atmosphere. The EF shown in table 22 has been estimated by weighting the emissions from EPS and XPS boards by their contribution to the HBCD consumption (i.e. 68% and 32%, see section 2.4.2.1.2). There are no data on emissions of BDE-47 from PUR foams during construction processes available. It has been assumed that the installation of fitting foam glue generates somewhat higher emissions than the estimates for HBCD. An EF of 5.0E–05 has been selected for the modelling.

Table 22 Emission factors applied in the dynamic SFA model for the process *construction*

	Substance	Application area	Year	Value	Uncertainty
EF(C,atm)	DecaBDE	construction	independent	0	-
EF(C,atm)	HBCD	construction	independent	3.5E–05	EF/10 – 10·EF
EF(C,atm)	BDE-47	construction	independent	5.0E–05	EF/10 – 10·EF

2.4.2.5.4 Use

It has been assumed that the emissions from products in the use phase decrease over time, as the BFRs that are closer to the surface and that are strongly bound to the structure are probably emitted in the early phase in the process *use*. This effect has been considered in the model. However, in order not to get too complicated in the report and due to comparison reasons with other studies, these parameters are outlined as emission factors. There are also TCs for diffuse releases from cleaning and washing to wastewater, for the dust in vacuum cleaner bags being directed to MSWIPs as well as for the fraction being burnt in fires that are described in this section.

Emissions from outdoor applications

In the EU risk assessments, it was assumed that 2% of the polymer products containing PBDEs with outdoor uses are lost to the environment during service life. 75% of these particles were assumed to be released to the soil compartment, 24.9% to hydrosphere and 0.1% to air. It was to be pointed out that it was assumed that 0.1% or less of the total use of plastics containing BFRs are incorporated in outdoor applications that exhibit a loss to environment. Palm et al. (2002b) estimated EFs based on the *Usage Category Document for Plastic Additives*. The derived EF for the loss of XPS to soil was 7.0E–03/year. It has to be emphasized that this value was highly questioned by the bromine industry. Therefore, this value has not been included in the model. Hence, the EFs over the residence time of a product in the use phase have been considered as 1.5E–05 to soil, 5.0E–06 to hydrosphere and 2.0E–08 to air (see table 24).

Releases from washing of textiles

Only the release from the textile itself has been accounted to this TC. The release caused to dust adsorbed to the textile has been accounted to the individual application areas.

In the domestic situation, DecaBDE is most likely to be present as a back-coating on fixed upholstery (i.e. it is nailed/stapled onto the furniture and is not intended to be removed and washed), although it may also be present on some types of removable seat cushions. Few other textiles present in homes will contain DecaBDE (Texconsul, 2003 cited in ECB, 2004). The same is probably true for HBCD. A figure of around 2% was suggested as a reasonable estimate of the percentage of the current textiles that contain DecaBDE that may be subject to washing during use. A release factor of 0.05% during the residence time was assumed. In the EU risk assessment draft report (ECB, 2007b), the same assumption were made for HBCD in the EU risk assessment draft report. Hence, TCs of 1.0E–05/residence time have been used for DecaBDE and HBCD in the SFA model. As PentaBDE is used in PUR foam that is not likely to be washed, the TC for BDE-47 has been defined as zero.

Diffuse emissions and their distribution to atmosphere, sewerage and incineration

The atmospheric emission factor *EF(U,atm.g/d)* and the transfer coefficients to wastewater *TC(U,sew)* and to MSWIPs *TC(U,inc)* are described in this section. A large proportion of diffuse emissions from products in the process *use* are believed to adsorb quickly on dust and on surfaces. A fraction of the substances occurs in the gaseous phase and is emitted directly to atmosphere. Another fraction is bound to dust and is also released from indoor environments to atmosphere. Due to cleaning/mopping and vacuum cleaning, dust is released to wastewater and to waste, respectively. Also substances that are adsorbed to surfaces can be released to wastewater as a result of e.g. window cleaning. In this context, also vehicles or construction materials that are not exposed to outdoor weathering have been considered as being indoors. There is not much known on emission and release pathways of dust and on the partitioning of chemicals in indoor environments (T. Reemtsma, TU Berlin, personal communication).

Emissions to indoor environments

There are two different methods to derive EFs to air and dust: emission chamber measurements and estimates using models based on physical properties and known emission rates of other substances. With the latter method and the plasticizer DIDP (diisodecyl phthalate) as a reference substance, the volatile loss of BFRs from products in indoor as well as outdoor service was estimated to be 5.0E–04 over the service-life in the *Emission Scenario Document* (OECD, 2004). However, it has been supposed that EFs determined in experiments might be more appropriate than based on model calculations. Emission chamber studies were carried out with different electronic appliances including printed circuit boards, televisions and computers. For DecaBDE, an area specific atmospheric EF of 0.3 $ng/m^2/h$ from a TV casing and no detectable emissions from printed circuit boards were reported (Kemmlein et al., 2003b; Kemmlein et al., 2003a). The TV was old (year of manufacture 1979 or before) and the ABS material of the casing contained a technical OctaBDE mixture casing studied. Therefore, it has not been possible to estimate appropriate EFs for DecaBDE using this measurement. However, this value was used in the update of the EU risk assessment report on DecaBDE (ECB, 2004) to calculate the European emissions during service-life of polymer products. Hence, this estimate is not believed to be appropriate. Sakai et al. (2006c) estimated migration rates from concentrations measured in dust inside TVs earlier (Tamade et al., 2002). The migration rates range between 2.1E–07/year and 8.9E–07/year. The amount of dust lost during service-life of the TVs (5, 10 and 13 years old) is unknown. However, this value is essential in order to correctly estimate EFs. Under the assumption made by the authors that half of the dust was lost during service-life the EFs have to be doubled. In an emission chamber study carried out by the same authors, three waste TVs were tested for 24 h and 144 h (Hirai et al., 2006). The amounts of PBDEs in the air as well as on the chamber wall and on the TV screen were added. The amounts in the chamber air were more than two orders of magnitude lower than adsorbed to surfaces. The EFs found were 7.1E–07/year and 4.8E–06/year for PBDEs as well as 5.8E–07/year and 4.3E–06/year for DecaBDE. These values are quite similar or somewhat higher than the EFs based on the amount of dust inside the TVs and the years after production. Hence, there is a relatively good agreement between results obtained by the two different methods. As the loss of dust in the first method is unknown, the mean of the emission chamber study (2.4E–06/year) has been estimated to be the most appropriate EF. This value has been selected for the application area *E&E* as a whole, even though it is clear that EFs are dependent on the material incorporated in as well as the type of appliance and the conditions. A strong dependence on the temperature was shown in an experiment with a printed circuit board. The raise in temperature from 23 °C to 60 °C resulted in a 500 times magnified emission for some congeners (Kemmlein et al., 2003a). It has been assumed that temperature effects are included in the estimate based on the contents in TV dust, which is comparable to the estimate based on the emission chamber experiment. As there were no data on the emissions from construction materials available, the same EF as for *E&E* has been assumed. The emissions from textiles to indoor environments have been assumed to be 5.0E–05/year, which is one order of magnitude higher than the EF from E&E, but one order of magnitude lower than the estimate based on the *Usage Category Document for Plastic Additives* (see below). For the application area *transport*, the mean of *E&E* and *textile/furniture* (2.6E–05/year) has been assumed as BFRs in vehicles are in both types of materials.

Palm et al. (2002b) estimated atmospheric EFs of HBCD based on the *Usage Category Document for Plastic Additives*. For EPS and XPS materials as well as for car inventories and textiles, the estimate was 5.0E–04/year. Emissions from polymer matrix in indoor service to air were estimated to the same value as Palm et al. (2002b) in the EU risk assessment draft report (ECB, 2005b). Data on volatile loss from EPS and XPS were available from emission cell experiments (Kemmlein et al., 2003b; Klatt, 2004). The area specific EFs of EPS derived were in a range of 1.0–4.0 ng/m^2/h, depending on the emission cell size. The values gathered for XPS were 0.1 and 29 ng/m^2/h (Kemmlein et al., 2003b), whereas Klatt (2004) measured an EF of 0.6 ng/m^2/h. The area specific EFs from different sources varied by more than two orders of magnitude (especially for XPS), which made it difficult to choose an appropriate value. As the measured data have been supposed to be more reliable than estimates based on the *Usage Category Document for Plastic Additives*, the area specific EFs have been converted in mass-related EFs. The parameters used (i.e. HBCD content, density and thickness of the samples) were either given in the specific publication or have been taken from the literature. That way mean EFs of 5.5E–06/year for EPS and 2.6E–05/year for XPS have been estimated. The EF for both materials has been derived by adding the two individual EFs according their share in the consumption of HBCD (see section 2.4.2.1.2) as 1.2E–05/year. As no experiment on the emissions from E&E was carried out, the same EF as for DecaBDE has been considered for this application area. Data for the emissions from textiles due to wearing and ageing were provided in the EU risk assessment draft report (Thomas and Stevens, 2006 cited in ECB, 2006a). An EF could be derived from these data that were given as a worst-case scenario. Therefore, 10% of that (i.e. 4.2E–04/year) have been taken into account. For the application area *transport* again the mean of *E&E* and *textile/furniture* has been taken into account.

In the same experiment with the old TV casing mentioned above (Kemmlein et al., 2003a), also emissions of PentaBDE congeners have been measured, even though a technical OctaBDE mixture was detected in the casing. The area specific EFs determined were 1.7 ng/m^2/h for BDE-99, 0.5 ng/m^2/h for BDE-100 and 6.6 ng/m^2/h for BDE-47, while no emissions of BDE-85 could be detected. The contents of these congeners in the TV casing are not known. Normally, these congeners are not contained in the technical OctaBDE mixture in relevant quantities (La Guardia et al., 2006), but they might have been formed in the ageing process. In the printed circuit boards experiment carried out at 60 °C emissions of 0.1 ng/unit/h BDE-85, 2.6 ng/unit/h BDE-99, 1.3 ng/unit/h BDE-100 and 14.2 ng/unit/h BDE-47 were determined. No EFs could be calculated from these two experiments as the BFR content in the samples was entirely unknown.

In an emission chamber test with rigid PUR foam, an area specific EF of 92 ng/m^2/h for BDE-47 and 28 ng/m^2/h for BDE-99 were found (Kemmlein et al., 2006). Based on the BFR content of 1–2%, a density of 80 kg/m^3 and an approximate thickness of 3 cm, EFs of 2.5E–05/year and 7.7E–06/year for BDE-47 and BDE-99 have been calculated.

Concentrations of PBDEs in dust and on interior window films sampled from passenger cars were determined (Gearhart and Posselt, 2006). The mean concentrations of BDE-47 in dust and in window films were 600 ng/g and 65 ng/m^2, respectively. The indoor air concentrations were back-calculated based on the equation given in Butt et al. (2004): $K_{FA} = f_{oc} \cdot K_{OA}$ The fraction of the organic carbon (f_{oc}) has been assumed as 5% (Lam et al., 2005), the film thickness as 200 nm (Diamond et al., 2005). The K_{OA} values have been calculated with the *Least-Squares Adjustment Spreadsheet v1.1* (Schenker et al., 2005). The mean interior volume of the cars sampled was 3.4 m^3 and the air exchange rates have been taken from a car exposure model (www.simsmoke.org) and the temporal fraction of the different conditions (e.g. “sealed stationary car” or “moving car vent open”) have been estimated. The fraction of the substance that is adsorbed to particles has been estimated (see section 2.6.1) to include the emission of dust from vehicles. Finally, the mass of PBDEs in cars has been estimated based on literature data (e.g. Petreas and Oros, 2006) and own assumptions. The estimated atmospheric EF for BDE-47 based on these data and assumptions is 2.6E–05/year. This estimated EF is similar to the one calculated for rigid PUR foam (2.5E–05/year).

Prevedouros et al. (2004a) compared atmospheric EFs from flexible PUR foam of the congeners BDE-47 and BDE-99 modelled based on the partial vapour pressure versus values estimated by dint of the K_{OA}. Values of 8.1E–03/year and 7.0E–04/year were calculated with the former approach, whereas with the latter EFs of 5.6E–05/year and 1.3E–05/year were determined. The EFs derived with the latter approach were 145 and 55 times lower. The EF of BDE-47 was also estimated based on the relationships between the evaporative loss of PCBs from materials such as sealants and physicochemical properties (i.e. K_{OA}) developed by Breivik et al. (2002) resulting in a factor of 1.0E–04/year (Alcock et al., 2003). The volatilisation of PentaBDE (congeners not specified) from PUR foam was estimated as 3.9E–03/year in EU risk assessment report (ECB, 2000). Emission cell measurements were carried out by Wilford et al. (2003). The volatile loss to the air from PUR foam determined was 360 ng/m^3/g foam for BDE-47 and 85 ng/m^3/g foam for BDE-99, respectively. Prevedouros et al. (2004a) calculated the emissions from PU foam based on the work of Wilford et al. (2003). The area specific emission rates were multiplied with the time of the experiment and the surface area of the foam and divided by the BFR content. For the congeners BDE-47 and BDE-99, the determined atmospheric EFs were 9.2E–02/year and 8.0E–02/year during a period of 130 days. In an unpublished study carried out by Wilford et al., even a loss of 97% of the BDE-47 content was measured in a long-term experiment. However, these results experiments might not be applicable for real situations. If such high losses would also happen in real situations, the flame retardancy of the treated foams would decrease rapidly.

By a simple calculation, it could be demonstrated that these (worst case) EFs are probably too high: By multiplying the Swiss population, an average live and work area (44 and 15 m^2/person), a room height of 2.5 m, an average air exchange rate of 1 per hour and measured indoor concentrations, the emission from indoor environments to atmosphere could be estimated. Mean concentrations of BDE-47 in indoor air found in different studies were 18.4 pg/m^3 in homes, 77.0 pg/m^3 in offices and 25.9 pg/m^3 in public microenvironments in the UK (Harrad et al., 2006) and 160 pg/m^3 in Canadian homes (Wilford et al., 2004). As the PentaBDE consumption of Canada was probably higher than in Europe, the data for UK offices was taken for this estimation. Using this number, the releases from indoor environments derived are 0.74 kg/year. For vehicles, the estimate has been done with cars only: By 2005, there were 3.8 million cars in Switzerland with an average interior volume of 4 m^3 and an estimated air exchange rate of 2.7 per hour. The mean BDE-47 concentration found in UK cars was 383 pg/m^3 (Harrad et al., 2006). Using this number, the estimated emissions amounted to 0.14 kg/year. Even if taking other vehicle types like trucks, buses or planes into account, the estimate would probably not change much, because their number is considerably lower than for personal cars. In a worst-case estimate with the highest indoor air concentration measured being 7140 pg/m^3 (Harrad et al., 2004), which is even higher than the 1200 pg/m^3 measured in a dismantling hall of a WEEE recycling plant (Sjödin et al., 2001), the estimated emission would amount to 70 kg/year for buildings and vehicles together. On a per capita base, the emission for EU15 would amount to 47 kg/year in the realistic and 3800 kg/year in the worst-case scenario. The realistic figure is significantly lower than the 4300 kg/year or 43000 kg/year (worst-case scenario) estimated in the EU risk assessment. However, the worst-case estimate based on the highest indoor air concentration is comparable with the non-worst-case estimate from the EU risk assessment. Considering that not the total of the substance in the indoor air is likely to be emitted to atmosphere, due to the removal dust with vacuum cleaners, wet cleaning, washing or degradation, the estimate in the EU risk assessment could be corroborated to be too high. To have emissions of 4300 kg/year from PUR foams in indoor environments, the indoor air concentration would need to be around 8500 pg/m^3 in average. For the worst-case estimate of 43000 kg/year, it would need to be over one order of magnitude higher than the highest concentration ever measured.

A similar estimation as with indoor air concentration has been made based on concentrations found in dust: Dust deposition rates of 3.7±1.3 mg/m^2/day in summer and 2.2±1.3 mg/m^2/day in winter were determined in an American study (Edwards et al., 1998). In a study carried out in Germany, a dust deposition rate of 6.2 mg/m^2/day was measured (Krause, 1991). The higher rate from Germany has been selected in order not to underestimate the generation of dust. Taking into account the average live and work area (see above) and the Swiss population, approximately 1000 tons/year of house dust are annually generated in Switzerland. Typical concentrations of DecaBDE and HBCD in house dust are about 1000 ng/g, while concentrations of PentaBDE are about one order of magnitude lower (A. Gerecke, unpublished data; different international studies). Hence, the loads of DecaBDE and HBCD in Swiss house dust are in the order of magnitude of 1 kg/year. The load of PentaBDE in house dust might be about 0.1 kg/year. As the concentrations of PBDEs in office dust were shown to be about three times higher than in house dust (Suzuki et al., 2006), the loads might be somewhat higher.

These two estimate methods (based on indoor air and dust concentrations) as well as the EFs estimated based on the vapour pressure by Prevedouros et al. (2004a) and the K_{OA} support the EFs of 2.6E–05/year and 2.6E–05/year that have been estimated based on measurements in cars and emission chamber experiment with rigid PUR foam. As it has been assumed that BDE-47 is present in vehicles in both PUR foam and E&E, whereas it is only used in PUR foam in textile/furniture, an EF of 5.0E–05/year has been assumed for this application area.

Pathways in indoor environments

The relative distribution of the emissions to the indoor environment have been roughly estimated based on own considerations. As shown by in the emission chamber experiment (Hirai et al., 2006), the largest fraction of the emitted PBDEs have adsorbed quickly to the chamber wall or the TV screen. Therefore, it has been assumed that half of the substances emitted from E&E get into wastewater by cleaning of surfaces (see table 23). 25% have been estimated to be emitted to atmosphere. Another 25% have been assumed to be split to vacuum cleaner bags, mopping water and washing water (as a result of dust adsorbed to clothes). Due to the higher air exchange in vehicles compared to houses, a larger proportion has been estimated to be emitted to atmosphere and no mopping has been assumed. A large proportion of emissions from *textile/furniture* into indoor environment have been estimated to be adsorbed to particles rubbed off from textiles. The total off the emissions have been assumed to be bound to dust that can be emitted to atmosphere, mopped, vacuumed or washed away from clothes. A large fraction of the emissions from construction materials has been estimated to be emitted directly to atmosphere, while a smaller fraction might get into the indoor environment. Due to the higher volatility of BDE-47, the proportion of the atmospheric emission has been estimated higher than for DecaBDE and HBCD. The relative contribution to the other pathways has been left constant.

Table 23 Assumption of the further pathways of DecaBDE and HBCD emissions into indoor environments

Pathway	E&E	transport	textile/furniture	construction
Atmosphere	25.0%	66.7%	50.0%	75.0%
Mopping water	6.3%	0.0%	12.5%	6.3%
Cleaning water	50.0%	8.3%	0.0%	6.3%
Washing water	6.3%	8.3%	12.5%	6.3%
Vacuum cleaner bags	12.5%	16.7%	25.0%	6.3%

The releases to mopping, cleaning an washing water are summarized in the transfer coefficient *TC(U,sew)*, while the atmospheric emission factor and the transfer coefficient to the process *incineration* are denoted as *EF(U,atm.g/d)* and *TC(U,inc)*, respectively (see table 24 below).

Emissions from fires

For the estimate of emissions from building and vehicles fires in Switzerland, two parameters are required. The percentage of the stock in the process *use* that gets burned in fires *TC(U,fire)* and the emission factors of the BFRs during the fire *EF(U,atm.fire)*. As the combined domain of uncertainty has been estimated as a factor of 10, no own uncertainty has been attributed to *TC(U,fire)* (see table 24).

Hirai et al. (2005) conducted landfill fire simulation experiments in the laboratory. Refuse derived fuel (RDF) or RDF with BFRs added was placed in a stainless steel bowl packed with soil and burnt. The EF of PBDEs to the flue-gas per amount of DecaBDE input was 9.1E–02. Using this atmospheric EF, the estimated emissions of PBDEs from open fires and open burning were estimated as high as 640 kg/year and 40 kg/year in Japan (Sakai et al., 2006a). However, for DecaBDE emissions only, an EF of 5.0E–04 was determined. The OctaBDEs were the dominant congeners in the flue-gas with an EF of 7.4E–02 per amount of DecaBDE burnt. For PentaBDEs and TetraBDEs EFs of 2.5E–04, 1.2E–04 per amount of DecaBDE were found. A high proportion of DecaBDE seemed to be debrominated during the experiment, mainly to OctaBDE. If looking at DecaBDE only, the stated emissions can therefore be reduced to 3.5 kg/year and 0.25 kg/year in Japan. On the other hand, the metabolites formed and being equally or even more toxic are of large relevance and need more study. The EFs of HBCD and TBBPA measured in the experiment were 1.1E–04 and 7.0E–03, respectively. As there was no experiment burning RDF with BDE-47 as the sole BFR, the EF for TetraBDEs (relative to the amount of DecaBDE) measured by Hirai et al. have been selected for the model.

In a Swedish study (Simonson et al., 2000), emissions from both simulated TV and room fires were measured. The content of DecaBDE and TBBPA in the two TVs allotted to the Swedish and the US market was not known. The emissions were therefore quantified to the mass of material burned. The DecaBDE emissions from the US TV were 8.9 mg/kg burned material, while the emissions from the Swedish TV could not be quantified. For TBBPA, the emissions were 100.42 and 27.26 mg/kg burned material. Assuming that only the back cover (2.1 kg HIPS plastic) of the V0 rated US TV was flame-retarded and a typical DecaBDE loading of 12%, approximately 250 g DecaBDE were incorporated in the TV. As only additive TBBPA can contribute to emissions, it has been assumed that only the ABS plastic parts contained this flame retardant. The mass of ABS in the TV was 850 g and typical loadings are 20–26% (Albemarle Corporation, 2006). Thus, the US TV has been estimated to contain up to 200 g of additive TBBPA. The absolute and the relative amount of combustible material combusted were 3.14 kg and 48%. Assuming the percentage of the back cover and ABS plastics combusted was also 48%, the EF has been estimated as approximately 2.1E–04 for DecaBDE and 3.3E–03 for additive TBBPA. The DecaBDE emissions estimated that way are a factor two compared to the EF from the landfill fire simulation experiments. In the same study, also large-scale room experiments have been conducted. The 16 m^2 rooms contained a sofa, an armchair, a coffee table, bookshelves, curtains, a carpet and a TV. The furniture was chosen as typical for mainland Europe. Thus, the sofa was resistant to cigarette ignition but not ignition from a small flame. The total weight was approximately 500 kg. The emissions measured were 0.17–6.8 mg for DecaBDE and not detected to 9.0 mg for TBBPA. Applying these emissions to total floor area burnt out by fires in Japan that was 1.574 million m^2 in 2004, maximum emissions of 0.67 kg/year and 0.89 kg/year for DecaBDE and TBBPA can be estimated.

It has to be emphasized that in the Swedish study, the chemical analyses were not conducted with isotope labelled internal standards. The internal standards were 3,3',4,4'-tetrabromodiphenylether for DecaBDE and pentabromophenol for TBBPA. Especially for DecaBDE due to its chemical properties, the use of isotope labelled internal standards is evident (A. Gerecke, personal communication). Therefore, the EF derived in the landfill fire simulation experiments have been supposed to be most appropriate for the model. However, the EFs derived from the Japanese and the Swedish studies are quite comparable for DecaBDE and TBBPA. For HBCD, the EF from the landfill fire experiment has been considered for the SFA model, too.

The number of building fires remained fairly stable over the last two decades, ranging from 13,000 to 22,000 incidents in the 19 member cantons of the Vereinigung kantonaler Feuerversicherungen (figure 21). As the member cantons make up approximately 80% of the buildings in Switzerland, the total number of building fires is about 25% higher (E. Kozlowski, VKE, personal communication). 70% of the claims of fire originate from residential buildings, of which there were 1.46 million in Switzerland in 2000 (Swiss Federal Statistical Office, 2006). Taking the 2000 fire statistics into account, the percentage of residential buildings with a fire incident was approximately 1.1%. In the period from 1986 to 1995, a percentage of 0.93% was determined (Fontana et al., 1999).

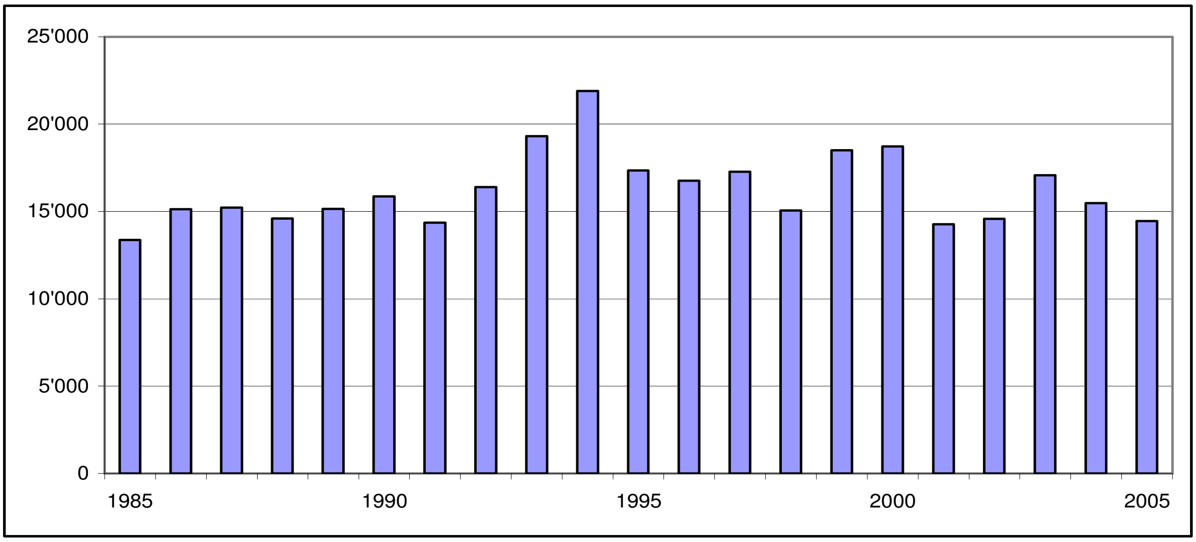


Figure 21 Number of building fires in the 19 member cantons of the Vereinigung kantonaler Feuerversicherungen (source: E. Kozlowski, VKF, personal communication)

From these fire incidents, approximately 0.5–1.0% were total losses (>¾ of a building destroyed). The proportion of room fires is by far bigger (P. Ridolfi, Gebäudeversicherung Zürich, personal communication). About 72% of the fires were self-terminated or extinguished by occupants and caused losses smaller than 2000 CHF (Fontana et al., 1999). A further 26% of the fires were extinguished in the room of origin by the fire brigade and caused losses smaller than 50,000 CHF. In the period from 1986 to 1995, building losses amounted to a fraction of 2.3E–04/year of the value insured in average. Contents, furniture and fixtures are estimated as follows: In 0.75% of the cases, there is a total loss, in further 1.25% of the cases, half is lost, while in the 26% room fires 10% of the residence contents are lost. In the 72% small fires, a loss of 1% is estimated. The mean loss of contents, furniture and fixtures in cases of fire is therefore estimated as 4.7%. Hence, the total loss is estimated as 4.7E–04/year for Switzerland. This is about double of the building loss in Switzerland and the floor space loss estimated in Japan (Sakai et al., 2006a). In Austria, there are almost 2000 car fires every year. 700 of them are officially reported, because of the higher loss (Österreichische Länderversicherer, 2006). It is assumed that half of the BFR containing plastics and textiles are burnt in average of these vehicles and no loss on the non-reported fires. Taking the 10% smaller Swiss population and the 3.9 million cars in Switzerland (Swiss Federal Statistical Office) into consideration, approximately 8.0E–05/year of the BFR content in cars is burnt per year. As cars make up the by far biggest amount of BFRs in vehicles and data for other vehicle types are not available, the same fraction has been supposed to be appropriate for all vehicle types.

It has not been possible to estimate EFs of BFRs to soil and surface water. In consequence of the destruction of the World Trade Center, elevated levels of PBDEs were found in sewage sludge (Litten et al., 2003). The authors assumed that ash and fire-fighting water from the WTC would have contained large amounts of PBDEs. Unfortunately, due to sampling strategy and the limited samples of different locations and dates, it was not possible to estimate emissions to soil or hydrosphere properly (S. Litten, personal communication). There are currently experiments running to quantify such emissions in the Swedish National Testing and Research Institute.

On May 25th 2007, there was a major fire at the Swisspor production facility in Steinhausen, Switzerland. As this company is a major producer of construction materials (mainly EPS, XPS and PUR insulation panels), high amounts of flame retardants could be burnt, too. The amount of HBCD contained in the destroyed buildings has been difficult to estimate. Considering the annual consumption of HBCD in construction materials of 150 t/year and the turnover period and the market share of Swisspor, an amount of 1–10 tons might be realistic. The stock of HBCD masterbatches might have been higher than average at that time, because Chemtura announced an off-list price increase of 10% for its HBCD based flame retardants effective June 15th 2007. Based on the EF of 1.1E–04, emissions from that major fire event could have been in the order of magnitude of 0.1–1 kg. The emissions released to the environment during the fire have not been expected to be of large significance for the level of Switzerland, but they might be relevant for the local contamination of soil.

Figure 22 Major fire at Swisspor in Steinhausen on May 25th 2007 (source: Keystone)

Summary

The emission factors and transfer coefficients in the process *use* are listed in table 24. Remark: "/res. time" means over the whole residence time of a product in the process *use*.

Table 24 Emission factors and transfer coefficients applied in the dynamic SFA model for the process *use*

	Substance	Application area	Year	Value	Uncertainty
EF(U,atm.p)	all	all	independent	2.0E–08/res. time	EF/10 – 10·EF
EF(U,hyd)	all	all	independent	5.0E–06/res. time	EF/10 – 10·EF
EF(U,soil)	all	all	independent	1.5E–05/res. time	EF/10 – 10·EF
TC(U,wash)	DecaBDE	textile	independent	1.0E–05/res. time	EF/10 – 10·EF
TC(U,wash)	HBCD	textile	independent	1.0E–05/res. time	EF/10 – 10·EF
TC(U,wash)	BDE-47	textile	independent	0	-
EF(U,atm.g/d)	DecaBDE	E&E	independent	6.1E–07/year	EF/10 – 10·EF
EF(U,atm.g/d)	DecaBDE	transport	independent	1.7E–05/year	EF/10 – 10·EF
EF(U,atm.g/d)	DecaBDE	textile	independent	2.5E–05/year	EF/10 – 10·EF
EF(U,atm.g/d)	DecaBDE	construction	independent	1.8E–06/year	EF/10 – 10·EF
EF(U,atm.g/d)	HBCD	E&E	independent	6.1E–07/year	EF/10 – 10·EF
EF(U,atm.g/d)	HBCD	transport	independent	1.9E–05/year	EF/10 – 10·EF
EF(U,atm.g/d)	HBCD	textile	independent	3.2E–05/year	EF/10 – 10·EF
EF(U,atm.g/d)	HBCD	construction	independent	9.0E–06/year	EF/10 – 10·EF
EF(U,atm.g/d)	BDE-47	E&E	independent	2.1E–05/year	EF/10 – 10·EF
EF(U,atm.g/d)	BDE-47	transport	independent	5.8E–05/year	EF/10 – 10·EF
EF(U,atm.g/d)	BDE-47	textile	independent	5.3E–05/year	EF/10 – 10·EF
EF(U,atm.g/d)	BDE-47	construction	independent	2.3E–05/year	EF/10 – 10·EF
TC(U,sew)	DecaBDE	E&E	independent	1.9E–06/year	EF/10 – 10·EF
TC(U,sew)	DecaBDE	transport	independent	4.8E–06/year	EF/10 – 10·EF
TC(U,sew)	DecaBDE	textile	independent	1.3E–05/year	EF/10 – 10·EF
TC(U,sew)	DecaBDE	construction	independent	8.6E–07/year	EF/10 – 10·EF
TC(U,sew)	HBCD	E&E	independent	1.5E–06/year	EF/10 – 10·EF
TC(U,sew)	HBCD	transport	independent	1.7E–05/year	EF/10 – 10·EF
TC(U,sew)	HBCD	textile	independent	4.0E–05/year	EF/10 – 10·EF

TC(U,sew)	HBCD	construction	independent	2.2E–06/year	EF/10 – 10·EF
TC(U,sew)	BDE-47	E&E	independent	1.9E–06/year	EF/10 – 10·EF
TC(U,sew)	BDE-47	transport	independent	2.4E–06/year	EF/10 – 10·EF
TC(U,sew)	BDE-47	textile	independent	4.7E–06/year	EF/10 – 10·EF
TC(U,sew)	BDE-47	construction	independent	9.4E–07/year	EF/10 – 10·EF
TC(U,inc)	DecaBDE	E&E	independent	3.0E–07/year	EF/10 – 10·EF
TC(U,inc)	DecaBDE	transport	independent	4.4E–06/year	EF/10 – 10·EF
TC(U,inc)	DecaBDE	textile	independent	1.3E–05/year	EF/10 – 10·EF
TC(U,inc)	DecaBDE	construction	independent	1.5E–07/year	EF/10 – 10·EF
TC(U,inc)	HBCD	E&E	independent	3.0E–07/year	EF/10 – 10·EF
TC(U,inc)	HBCD	transport	independent	2.4E–05/year	EF/10 – 10·EF
TC(U,inc)	HBCD	textile	independent	5.6E–05/year	EF/10 – 10·EF
TC(U,inc)	HBCD	construction	independent	7.5E–07/year	EF/10 – 10·EF
TC(U,inc)	BDE-47	E&E	independent	1.9E–06/year	EF/10 – 10·EF
TC(U,inc)	BDE-47	transport	independent	2.4E–06/year	EF/10 – 10·EF
TC(U,inc)	BDE-47	textile	independent	4.7E–06/year	EF/10 – 10·EF
TC(U,inc)	BDE-47	construction	independent	9.4E–07/year	EF/10 – 10·EF
EF(U,atm.fire)	DecaBDE	all	independent	5.0E–04	EF/10 – 10·EF
EF(U,atm.fire)	HBCD	all	independent	1.1E–04	EF/10 – 10·EF
EF(U,atm.fire)	BDE-47	all	independent	1.2E–03	EF/10 – 10·EF
TC(U,fire)	all	E&E, textile	independent	4.7E–04/year	no own uncert.
TC(U,fire)	all	transport	independent	8.0E–05/year	no own uncert.
TC(U,fire)	all	construction	independent	2.3E–04/year	no own uncert.

2.4.2.5.5 Deconstruction

Deconstruction of buildings and constructions containing flame-retarded materials generates a release of minor particles and fragments to the environment. The BFRs might be released from these particles to the environmental compartments subsequently. The emissions during *deconstruction* depend, among other things, on how the deconstruction is performed. Current methods include imploding a structure with explosives, using a crane and wrecking ball technique or deconstructing the structure.

Estimates on the emissions of HBCD were provided in the EU risk assessment draft report (ECB, 2006a). Manual deconstruction was mimicked by manual breaking of boards. The release was measured by weighing of particles being formed. No particles were formed by two breaks of a 456 g XPS board, whereas two breaks of a 287 g EPS board was estimated to generate 0.026 g particles (i.e. 90 g/ton). It has been assumed that the proportion of the EPS that are recycled subsequently is deconstructed manually. For the remaining proportion of demolition material not being recycled, the lack of information on how much the PS-boards are being broken during demolition makes it difficult to calculate the potential emissions. Therefore, the number of 1.0E–03 given in the *Use Category Document on Plastic Additives* was taken into account for the mechanical demolition. As this number was classified as a worst-case estimate in the EU risk assessment, half of it has been assumed to be more appropriate. The EFs have been estimated by multiplying the emissions estimated to happen during manual deconstruction by the relative contribution of EPS and XPS in the consumption of HBCD and by the percentage being recycled (see section 2.4.2.4.5). The remaining proportion not being recycled has been multiplied by the emission estimate for mechanical demolition. These two EFs have added and split up to the environmental compartments atmosphere and soil (50% each). As the EFs for HBCD depend on the proportion of the insulation panels being recycled, they vary over time (see table 25).

There are no data for DecaBDE and BDE-47. DecaBDE is not used in materials that are estimated to generate many particles during deconstruction. It has therefore been assumed that the EFs to both atmosphere and soil are a fifth of the emission generated during manually deconstruction of EPS boards. As rigid PUR foam has been estimated to generate less and less lightweight particles than EPS, intermediate EFs between manual and mechanical deconstruction have been assumed for BDE-47.

Table 25 Emission factors applied in the dynamic SFA model for the process *deconstruction*

	Substance	Application area	Year	Value	Uncertainty
EF(D,atm), EF(D,soil)	DecaBDE	construction	independent	9.0E–06	EF/10 – 10·EF
EF(D,atm), EF(D,soil)	HBCD	construction	1980–1997	2.5E–04	EF/10 – 10·EF
EF(D,atm), EF(D,soil)	HBCD	construction	2007	1.3E–04	EF/10 – 10·EF
EF(D,atm), EF(D,soil)	HBCD	construction	2020	7.8E–05	EF/10 – 10·EF
EF(D,atm), EF(D,soil)	BDE-47	construction	independent	9.0E–05	EF/10 – 10·EF

2.4.2.5.6 Recycling

Transport

As an exception, is has been chosen to discuss the application area *transport* first, because these data have also been used to estimate the EFs for *E&E*.

The atmospheric EFs from automobile recycling plants have been estimated based on data on the emission of dust provided by a Swiss recycling company (that did not want to be named in the report) and Metso Lindemann GmbH (Düsseldorf, Germany). The results of four studies carried out by the Swiss recycling company in 2002 and 2006 can be summarized as follows: scrap throughput 30–39 tons/h, dust emitted 184–600 g/h and dust concentrations in exhaust 3.7–8.9 mg/Nm^3. It has been assumed that most of the dust is generated by plastics and textile parts that will become ASR. The mass fraction of these materials that contain the majority of the BFRs is about 26% (see section 2.4.2.4.6). The best estimate atmospheric EF calculated from these data is 4.3E–05 for the past few years. According to data supplied by Metso Lindemann GmbH, the mean dust contents in exhaust were 100 mg/Nm^3 in the beginning of the eighties, 50 mg/Nm^3 in 1985, 20 mg/Nm^3 in 2005 and 10 mg/Nm^3 were estimated for the near future. The current dust contents are somewhat higher than in the Swiss study. However, the ratios between the past, present and future emissions have been taken into account.

E&E

As stated in section 2.4.2.4.4, there are two different recycling disposal routes: in specialised appliance recycling facilities and together with cars in automobile recycling plants. The atmospheric EF in an appliance recycling facility was recently determined as 5.0E–06 (Sakai et al., 2006c), which is one order of magnitude lower than the estimate for automobile recycling plants. For the temporal trend of the EF, the decrease in the PBDE air concentrations of one order of magnitude in an electronics recycling plant before and after a technical improvement have been taken into account (Takigami et al., 2006). The EFs of the two recycling disposal routes have been added according to the TCs given in figure 14 on page 40. As there were only automobile recycling plants before 1991, the resulting EFs are the same as for *transport* in the period 1980–1990.

Construction

The EFs for the recycling of construction materials have been assumed as equal to the EFs in the process *production* (see table 21).

Summary

A selection of the emission factors for the process *production* is listed in table 26. There is a steady but for *E&E* non-linear decrease for the intermediate year not included in the list.

Table 26 Emission factors applied in the dynamic SFA model for the process *recycling*

	Substance	Application area	Year	Value	Uncertainty
EF(R,atm)	all	E&E	1980	8.6E–04	EF/10 – 10·EF
EF(R,atm)	all	E&E	1990	3.2E–04	EF/10 – 10·EF
EF(R,atm)	DecaBDE	E&E	2007	1.5E–05	EF/10 – 10·EF
EF(R,atm)	DecaBDE	E&E	2020	9.5E–06	EF/10 – 10·EF
EF(R,atm)	HBCD	E&E	2007	1.7E–05	EF/10 – 10·EF
EF(R,atm)	HBCD	E&E	2020	1.2E–05	EF/10 – 10·EF
EF(R,atm)	BDE-47	E&E	2007	2.6E–05	EF/10 – 10·EF
EF(R,atm)	BDE-47	E&E	2020	2.1E–05	EF/10 – 10·EF
EF(R,atm)	all	transport	1980	8.6E–04	EF/10 – 10·EF
EF(R,atm)	all	transport	1985	4.3E–04	EF/10 – 10·EF
EF(R,atm)	all	transport	2002–2006	4.3E–05	EF/10 – 10·EF
EF(R,atm)	all	transport	2015	2.2E–05	EF/10 – 10·EF
EF(R,atm)	all	construction	dependent	see table 21	-

2.4.2.5.7 Incineration

Two emission pathways from MSWIPs have been considered: atmospheric emissions and emissions to surface water.

OECD (2004) estimated the emission factors of BFRs from incineration plants to air as 0 due to high decomposition rate, whereas Mark et al. (2006) estimated emissions of the PBDEs components PentaBDE to OctaBDE to be lower than 0.01% according to their measurements. The authors argued that specifying the emission factor more precisely would have led to a high uncertainty. The atmospheric emissions of PBDEs from an incineration plant were also measured by Tamade et al. (2002). From their data, an EF of 4.7E–07–9.4E–06 has been calculated for an incineration plant with an off-gas cleaning system and 3.3E–04–6.6E–03 for a plant without an off-gas cleaning system. The atmospheric EF of BFRs (not split up by individual substances and no reply on inquiries to the authors) that have been derived from results of a Norwegian study (Borgnes and Rikheim, 2004b), however, range between 3.0E–08 and 4.7E–08. These values are more than one order of magnitude lower than the calculated data from Tamade et al. Even though the comparability is not given, as not the same set of substances was measured, this difference is surprisingly big. According a personal communication by Y. Tamade, the reason of this difference could be due to the fact of the 60 times higher BFR input into the Norwegian plant. Up to him, the input and flue-gas concentrations do not linearly correlate. As concentrations applied in the Norwegian study were much higher than concentrations in municipal solid waste, these results might underestimate the EFs for real conditions. Based on three studies in Japan (including the study by Tamade et al.), Sakai et al. considered an atmospheric EF of 1.8E–06 as most appropriate (2006c). This number has also been selected for modelling for the current and the next decade. The temporal variation of the atmospheric EF has been estimated based on the change in dust concentrations in the flue-gas. Typical concentrations are 1 mg/Nm^3 to present, 5 mg/Nm^3 in 1991 and 150 mg/Nm^3 in 1985 before the Ordinance on Air Pollution Control that was enacted (LRV, 1985). The difference in the emissions between an incineration plant with and without an off-gas cleaning system determined by Tamade et al. (2002) is slightly larger but in the same order of magnitude.

There are only data on the emissions from the wet scrubber to hydrosphere in the Norwegian study (Borgnes and Rikheim, 2004b). From these data an EF to hydrosphere of 1.3E–09–2.3E–09 has been calculated, of which the mean has been applied in the model.

Table 27 Emission factors applied in the dynamic SFA model for the process *incineration*

	Substance	Application area	Year	Value	Uncertainty
EF(I,atm)	all	all	1980–1985	2.7E–04	EF/10 – 10·EF
EF(I,atm)	all	all	1991	9.0E–06	EF/10 – 10·EF
EF(I,atm)	all	all	2001–2020	1.8E–06	EF/10 – 10·EF
EF(I,hyd)	all	all	independent	1.8E–09	EF/10 – 10·EF

2.4.2.5.8 Sewerage

As shown in table 28, two emission factors have been considered in the process *sewerage*: emissions to hydrosphere caused by an overflow of the sewer system after strong precipitation events and emissions due to leaks in the sewer system to the underground (soil). The latter emission could also have accounted to the hydrosphere (groundwater) or to soil. As the substances might not reach surface water as a result of their limited mobility, soil has been selected as the receiving environmental compartment.

The fraction of the wastewater emitted to hydrosphere due to overflows has been estimated based on a study on the different input pathway of phosphorus to Lake Greifen. The contribution of the WWTP effluents and the overflows were estimated as 2.0 tons/year and 1.6 tons/year, respectively (Sennhauser, 2003). Taking into account the 96% elimination rate of phosphorus in WWTPs given in the report, an EF of 0.032 has been estimated. This number was considered as a realistic estimate for Switzerland, but a large domain of uncertainty was estimated (L. Rossi, Eawag, personal communication). The estimates on the percentage of the wastewater lost to groundwater range between different international studies between 1% and 11% for dry weather situations (Schluep et al., 2006). The authors conclude that the knowledge on these losses is insufficient. In the APUSS project description on the Eawag website the estimates on losses are summarized as 0–10%. The overall loss for Switzerland has been estimated as 1–2% (J. Rieckermann, Eawag, personal communication).

Table 28 Emission factors applied in the dynamic SFA model for the process *sewerage*

	Substance	Application area	Year	Value	Uncertainty
EF(Se,overflow)	all	all	independent	3.2E–02	±50%
EF(Se,leakage)	all	all	independent	1.5E–02	±50%

2.4.2.5.9 WWTP

The EFs from the process WWTP have been defined as the remainder of the TC of the BFRs into sewage sludge. These remainders are 0.037 for DecaBDE, 0.11 for both HBCD and BDE-47 (see section 1.1.1.1.1).

2.4.2.5.10 Landfill

Three different types of atmospheric emissions are expected to occur on landfills: emissions during unloading, emissions of dust caused by operations done on landfills and wind as well as gaseous emissions (see table 29).

No data on emissions during unloading on landfills were available. Therefore, an emission factor has been calculated from a study on emissions from opencast coal mining in India (Chaulya et al., 2002). A mean emission rate of overburden and coal unloading, given in grams per second, has been divided by the capacity of the unloader and the frequency of unloading. The emission factors determined are 11 g/ton for overburden unloading and 8 g/ton for coal unloading. The drop height on the coalmine for overburden and coal unloading was around 13 m and 3 m, respectively. The drop heights during unloading on landfills are probably also about 3 m. However, not all fractions dumped on landfills emit dust. APC residues are dumped in underground landfills; bottom ash and sludge from WWTP are not dusty. Most dust probably arises from demolition waste. On the other hand, the concentration of BFRs in small particles that are more likely to be emitted might be higher than average. Therefore, an EF of 1.0E–05 has been considered for the model. The annual emissions of dust from landfills in Switzerland due to operations and wind might be relatively low. As none of the landfill operators contacted was able to provide data or rough estimates, an assumption of 1 g annually released per ton in the landfill has been used for the model. The gaseous emissions have been estimated to be influenced by overlaying materials. On the other hand, the mean size of pieces might be smaller than in the use phase. The emissions have been estimated to be lower than from foams and textiles in the use phase. Therefore, the EFs from *E&E* in the process use might be most comparable and have been selected for the model.

Table 29 Emission factors applied in the dynamic SFA model for the process *landfill*

	Substance	Application area	Year	Value	Uncertainty
EF(L,unload)	all	all	independent	1.0E–05	EF/10 – 10·EF
EF(L,atm.d)	all	all	independent	1.0E–06/year	EF/10 – 10·EF
EF(L,atm.g)	DecaBDE	all	independent	6.1E–06/year	EF/10 – 10·EF
EF(L,atm.g)	HBCD	all	independent	6.1E–06/year	EF/10 – 10·EF
EF(L,atm.g)	BDE-47	all	independent	2.5E–05/year	EF/10 – 10·EF

2.5 Influence of future and past legislative and technical measures

2.5.1 Ban on the usage of DecaBDE

As stated in section 2.4.2.1.6, the consumption of DecaBDE is not expected to increase during the next decade. Therefore, an invariant consumption trend has been considered for the reference scenario. However, it is possible that the consumption will decrease markedly, either due to consumer pressure for green product design or a ban. Two different scenarios have been selected for the future consumption of DecaBDE in addition to the reference scenario (i.e. constant consumption until 2020):

- ban for all application areas except *E&E* in 2007
- ban for all application areas in 2007

2.5.2 Influence of an invariant waste management on emissions of DecaBDE

In order to demonstrate the influence of regulatory and technical measures taken in waste management on the emissions of BFRs, DecaBDE in E&E has been modelled omitting these changes in a special scenario “old fashion waste management”. In that scenario the disposal routes have been assumed to remain constant. This means that the TCs at end of life of products to landfill, incineration, recycling and export have been kept constant from the year 1980 to 2020. For the processes recycling and incineration the TCs to landfill, incineration, sewerage and export and for the final disposal of air pollution control residues, respectively, have been assumed to be invariant during the same period, too. Sewage sludge disposal routes, meaning the TCs to agriculture (i.e. soil), landfill and incineration as well as the TC from landfills to the sewerage system (leachate catchment factor) have also been kept constant. The same has been assumed for the atmospheric emission factors from recycling and incineration, meaning that air pollution control efficiency would have remained at the status of the year 1980.

The application area *E&E* has been selected, because it has been estimated to be most important for emissions to environment in Switzerland and in other countries. In Third World Countries probably only minor amounts of BFRs are used to flame-retard textiles and insulation materials. On the other hand, there is a considerable consumption of E&E as well as an import of WEEE to some of these countries.

2.5.3 Ban on the usage of HBCD in construction materials

For the reasons stated in section 2.4.2.1.6, a constant consumption of HBCD in the application areas E&E, transport and textile/furniture has been assumed. As for EPS and XPS, no market switch has been presumed and the growth rate of HBCD has been assumed to be equal to the increase in EPS and XPS consumption in the reference scenario. In an alternative scenario, a possible import stop in 2007 has been considered. The recycling of EPS and the subsequent reuse in the production of these insulation materials have been assumed to continue and to generate an unintended flow from *production* to *construction* and *use*. Theoretically, recycling can also lead to an uncontrolled contamination of polystyrene products with HBCD, which do not need to be flame-retarded. It is not clear whether flame-retarded polystyrene products are treated separately in the recycling process. Recycling of EPS could effect that concrete mixed with EPS in the future contains HBCD (Freilich, 2004 cited in ECB, 2006a).

Because the construction sector is by far the most important for HBCD, this scenario has been compared separately to the reference for this application area.

2.5.4 PentaBDE

No scenarios have been selected for PentaBDE in addition to the reference scenario due to the fact that this flame retardant had been banned in 2004 in Europe.

2.6 Environmental fate model

In order to estimate the fate of the BFRs under study in the environment and to calculate predicted environmental concentrations (PEC), a fugacity model amended with atmospheric deposition to both soil and hydrosphere has been used.

In a first step, degradation reactions have not been included except for the atmosphere, because of the influence of the degradation to the deposition. The substance flow originating from atmospheric emissions has been split up in the three compartments atmosphere, hydrosphere and soil.

2.6.1 Atmosphere

The atmospheric deposition has been calculated with the estimated deposition rate constant k_{dep} taking into account the atmospheric degradation: $J_{dep} = J_{in} \cdot (1 - 1/(k_{dep} + k_r)) \cdot k_{dep}/(k_{dep} + k_r)$. The atmospheric deposition substance flow has been split up to hydrosphere (5.6%) and soil (94.4%) according to their surface area fraction. These substance flows have been added to the emissions to hydrosphere and soil respectively. The mass remaining in the atmosphere has been estimated for each individual year according to the equitation $M_{atm} = J_{in} / (k_{dep} + k_r)$ assuming steady-state during one year.

The degradation rates have been derived from estimated half-lives. For DecaBDE, the geometric mean of 7620 hours (Palm et al., 2002a) and 37 hours (Söderström et al., 2004) – 531 hours – has been taken as a best guess estimate. For HBCD and BDE-47, the estimates 51.2 hours (Wania, 2003) and 256 hours (Palm et al., 2002a) have been taken into account. The deposition rates have been defined as the sum of dry and wet deposition. They have been estimated according to the equations that were adapted from Hirai and Sakai (2004):

The parameters used are the particle associated fraction of BFRs (Φ), the dry particle deposition velocity (v_{dep}), the height of the air compartment (h), the scavenging coefficient (SC), the precipitation (v_{rain}) and the air-water partition coefficient (K_{aw}).

The values of parameters applied and the way they have been estimated are given in the appendix in section 7.4.

Table 30 Fate of the atmospheric emissions of DecaBDE, HBCD and BDE-47

	DecaBDE	HBCD	BDE-47
Fraction of atm. emission degraded in air	2.3% (0.21–20.7%)	27.1% (19.3–38.9%)	22.0% (11.4–39.2%)
Fraction of atm. emission in deposition	97.5% (79.2–99.5%)	72.6% (60.8–80.6%)	77.1% (59.1–88.1%)
Fraction of atm. annual emissions that resides in air at steady-state	0.20% (0.13–0.26%)	0.23% (0.16–0.33%)	0.94% (0.48–1.68%)

In order to compare these estimates on the fate of BFRs in the atmosphere with field data, an attempt has been made to derive total elimination rates from empirical atmospheric half-life distances that were based on dated sediment cores from lakes along a latitudinal transect in North America (Breivik et al., 2006). The empirical half-distance for BDE-209 and BDE-47 were derived from surface flux data at different locations. The empirical half-life distances have been divided by an average wind speed of a range from 4–10 m/s that was read from the figures given in the paper to get half-lives. As no empirical half-life distances were available for HBCD, characteristic transport distances given in the literature (Palm et al., 2002b; Wania, 2003) have been converted into total elimination rates. The fraction of annual atmospheric emissions that has been estimated to reside in the air at steady-state has been derived from the reverse of the total elimination rates. The lower estimates of the fraction derived in this approach are about the same for DecaBDE and HBCD as in the approach above using the geometric mean of parameters pairs. For BDE-47, results are about the same in both approaches, but the uncertainty range is the largest for this substance.

Table 31 Fate of the atmospheric emissions of DecaBDE, HBCD and BDE-47 estimated with empirical half-life distances

	DecaBDE	HBCD *	BDE-47
Empirical half-life distance	566±101 km	527 / 543 / 1768 km	1168±942 km
Fraction of atm. annual emissions that resides in air at steady-state	0.21–0.75%	0.24–2.02%	0.10–2.41%

*: Calculated from characteristic transport distances (CTD) estimated by Wania (2003) with TaPL3 and ELPOS model and Palm et al. (2002b) with TaPL3 model.

However, as the second approach could not distinguish between deposition and degradation (table 31), the inputs to hydrosphere and soil compartments have been estimated with the data obtained in the first approach (table 30).

2.6.2 Hydrosphere and soil

For the hydrosphere, the chemicals partitioning at equilibrium into the different compartments water, sediment and aquatic organisms was modelled for each year. For the atmosphere and soil, only the resulting concentrations have been calculated. The calculations have been carried out with Safe-Pro, a fugacity model that had been developed by BMG Engineering AG.

The air has been assumed to have a height of 1000 m, which is about the mean height of the atmospheric boundary layer. The soil has been assumed as a mixed 1-box-model of a thickness of 0.1 m. In the first step neglecting any degradation, the emissions to soil (including atmospheric deposition on soil) of the individual years have been added. It has been assumed that the proportion of emissions released to hydrosphere (including the atmospheric deposition on surface water) resided in this compartment and only distributes between the different aquatic phases. The sediment has been assumed to cover the area of Swiss surface waters. The sedimentation rate in lakes is approximately 0.0035 m/year (e.g. Kohler, 2005). Therefore, it has been assumed that the amount released to hydrosphere would partition between the water, the biota and the yearly deposited sediment. In contrast to the soil, no mixing has been supposed. Therefore, a concentration time trend depending on the depth of the sediment layers has been derived. The full set of parameters of the compartments chosen is listed in the appendix (section 7.4).

In the second step, degradation rates in sediment and soil and dynamic trends have been included to the model. The atmospheric transport of the chemicals from and to Switzerland was not included. It has been assumed that the export is compensated by import from adjoining countries, because emissions in these countries have been supposed to be similar and the distance to the sea is only considerably shorter than the atmospheric half-life distances in one of the four main wind directions in Switzerland (from W, NNW, ENE and SSW; MeteoSchweiz, 2006). However, due to the long-range transport potential (Breivik et al., 2006), a high proportion of the atmospheric concentration and atmospheric deposition has been estimated to originate from foreign sources. As the atmospheric half-lives are much shorter than one year, no accumulation in the atmosphere over time has been expected. Therefore, the time trend has been derived by calculating the concentrations for each year individually. The export via the hydrosphere, i.e. with water and suspended sediments in rivers has been expected to be small, as the concentration in water is very low and only a small percentage of suspended sediments has been supposed to be exported from Switzerland. The import in food like fish or the export of dairy products might be of larger relevance.

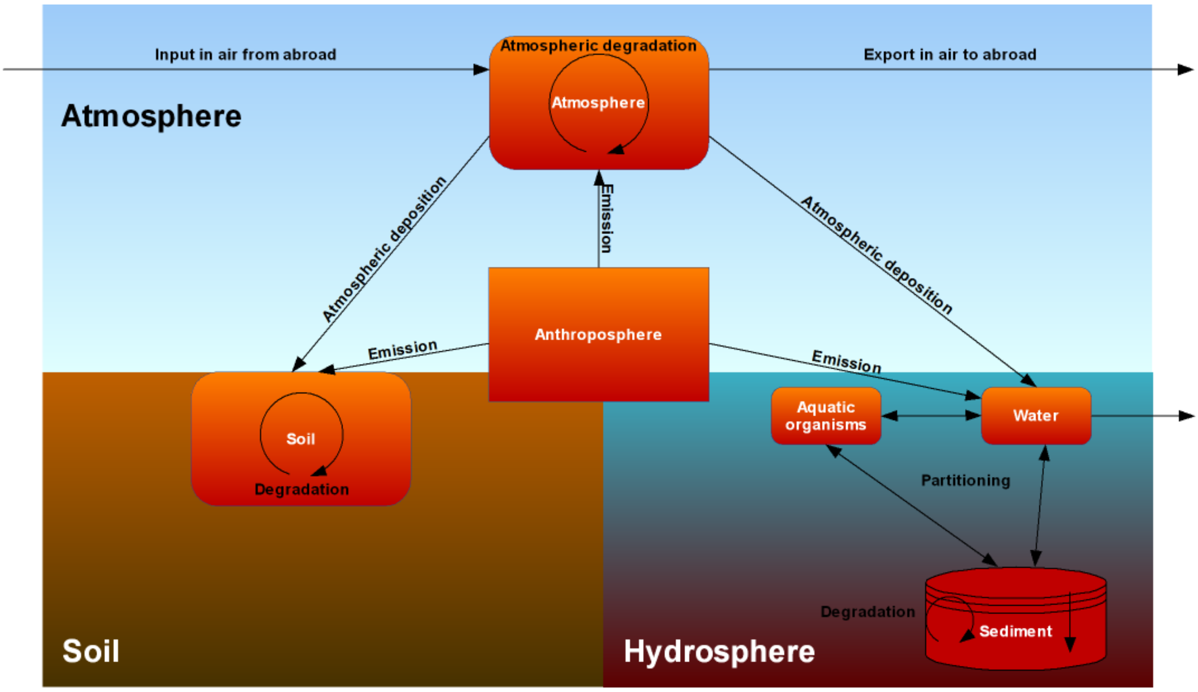


Figure 23 Scheme of the environmental fate model

The concentrations in the sediment and soil have been modelled with Safe-Pro for each individual year. They have subsequently been assembled taking into account the degradation rates.

DecaBDE

There were some data on degradation of DecaBDE (Parsons et al., 2004; Ahn et al., 2006; Gerecke et al., 2006; He et al., 2006), but either the type of degradation, the matrix or the temperature is not realistic for lake sediments or soils. The experiments by Gerecke et al. carried out with digested sewage sludge have been considered to be the most appropriate. They have found a half-life of approximately 2 years. Taking into account that the temperature in sediment is about 30 °C lower than in the experiment, the microbial community is not the same, the conditions are only partly anaerobic and the fact that the available energy is declining with the age of the sediment, the half-life was expected to be two orders of magnitude higher (A. Gerecke, personal communication). As there are aerobic conditions in soil, the degradation rate has been expected to be small. The photolytic degradation in soil, determined e.g. in Ahn et al. has been expected to be relevant for the very thin top layer only. The most appropriate method to estimate the biodegradation in soil has been assumed to be the attempt by Arnot et al. (2005) to develop a pragmatic method of selecting aerobic biodegradation environmental half-lives by the calibration of BIOWIN models 1, 3, 4, 5 (Urs Schenker, ETH Zurich, personal communication). For DecaBDE, a half-life of 3150 days has been estimated using this method.

HBCD

For the technical HBCD mixture, Gerecke et al. (2006) found a half-life of 0.66 days in digested sewage sludge under anaerobic conditions at 37 °C. In a similar study a half-life of approximately 5 days was found (Hunziker et al., 2004). Davis et al. (2006) found first-order reaction rates of 0.1278 d^{-1} (corresponding to a half-life of 5.4 days) in digested sludge, 0.0109 d^{-1} (63.4 days) in anaerobically and 0.0074 d^{-1} (93.7 days) in aerobically treated freshwater sediment at 20–22 °C. In a Swiss study, a sediment sample was taken in Lake Greifen at a depth of 31 m, sectioned and analysed (Kohler et al., 2006). The HBCD concentration was 1.27 ng/g d.w. in 1989, 1.76 ng/g d.w. in 1995 and 2.54 ng/g d.w. in 2001. Hence, the concentration of HBCD in the 12 years old sediment layer was 50% of what was found in the top layer. Even in 1974, the concentration was 0.51 ng/g d.w. Unpublished measurements in Lake Thun showed similar trends, but the concentrations were lower (Bogdal et al., 2006). Remberger et al. (2004) detected concentrations of HBCD in 30 (±7) and 40 (±7) years old sediment layers in two different sediment cores from the Stockholm archipelago. The concentrations were 25–33% of those found in the top layer. These environmental measurements indicate that the degradation rates obtained in simulations may not be applicable for the environment. It is not possible to determine a quantitative half-life of HBCD in sediment based on these studies. The temporal trend of the HBCD input to the sediment layers is the second unknown variable except the degradation rate. However, it is clear that the half-lives derived by Davis et al. are not relevant in these cases as HBCD was still present in sediment after a few decades. Even there was the risk of a circular reasoning and emissions to hydrosphere were not constant over the years, it could be concluded that a realistic half-life must be around 10 years or greater at environmentally relevant conditions. A best-guess half-life of 20 years has been assumed for the estimations.

Only half of the amount that gets into raw sewage sludge in WWTPs has been estimated to be in digester sludge due to degradation of which a fraction is used as a fertilizer (de Wit et al., 2007). For aerobic soils, a half-life of 63 days was found for HBCD (Davis et al., 2003) at 20 °C, which corresponds to a half-life of 119 days at 12 °C (ECB, 2007b). However, in another study carried out by the same authors, there were no indications of any transformation of HBCD during 112 days of incubation at 20±2 °C (Davis et al., 2004). The most appropriate biodegradation half-life has been assumed the 373 days derived by the method of Arnot et al. (2005). Because of the uncertainty in the degradation half-life, the concentration trend in soils has been estimated with and without any degradation.

BDE-47

For BDE-47, the situation is somewhat different: Besides the degradation of BDE-47 itself, also the degradation of higher brominated PBDEs has a (contrary) influence on the concentrations in the different environmental compartments. The extent of the formation of BDE-47 from other substances such as DecaBDE or different congeners of the technical OctaBDE mixture is highly uncertain and the concentrations of the latter have not been modelled quantitatively. Therefore, it has been chosen not to account for it in the estimations.

Gouin and Harner (2003) obtained data on half-life values for the PBDEs from the BIOWIN program adopting the approach recommended by Mackay for PCBs. The data suggested that PBDEs were likely to degrade slowly, having biodegradation half-lives longer than months. For BDE-47 a half-life of 10,000–30,000 hours (i.e. 1.1–3.4 years) in soil and >30,000 hours (i.e. >3.4 years) in sediments. Wania and Dugani (2003) made similar estimations with EPIWIN and estimated the half-lives as 3,600 hours (i.e. 150 days) in soil and 14,400 hours (i.e. 1.6 years) in sediment. In a recent study, a rapid loss of PentaBDE after exposure to soil microbes in a water environment was demonstrated. Enrichment cultures isolated from the experimentally contaminated soils showed the ability to swiftly utilize these congeners as a sole carbon source. Within 2 h in the first and even less in the second experiment, loss of all of the DE-71 added was observed (Vonderheide et al., 2006). If such a rapid degradation would be applicable for the environment, the BDE-47 concentrations in soil would be below the detection limit. As BDE-47 is found in various environmental matrices, it can be concluded that the half-lives determined in this experiment are not applicable for real conditions. Therefore, the half-life in soil estimated with the method of Arnot et al. (2005) being 384 days has been selected for the calculation in addition to the scenario without any degradation.

In a Taiwanese study, BDE-47 was totally degraded in the sediment from one river after 70 days, while in the sediment from another river no significant degradation could be detected within the same period (Yen et al., 2006). However, comparing half-lives estimated and experimentally determined with data from sediment cores, it appears that half-lives in the environment tend to be underestimated. BDE-47 concentrations in sediment sample from Lake Greifen were 0.53 ng/g d.w. in 1989, 0.64 ng/g d.w. in 1995 and 0.74 ng/g d.w. in 2001. Even in 1974, the concentration was 0.21 ng/g d.w. (Kohler, 2005). Also measurements in three sediment cores from Norway, the Netherlands and Germany showed that there was not a large difference in concentrations from recent sediment layers to 30 years older layers (Zegers et al., 2003). As stated above, it was logically not possible to determine a half-life from these data as the input trend is unknown, but it could be testified that the half-life must be in the order of magnitude of years or decades. As a best guess, the double of the minimum half-life estimated by Gouin and Harner (2003) being 8.6 years has been applied to the model.

Summary

The selected first-order degradation rates in sediment and soil are given in table 32. The concentration trend has also been estimated excluding any degradation for comparison purposes.

Table 32 First-order degradation rates in sediment and soil selected for the model [1/year]

	DecaBDE	HBCD	BDE-47
Degradation rate in sediment	3.47E−03	3.47E−02	1.01E−01
Degradation rate in soil	8.03E−02	6.78E−01	6.58E−01

3 Results and discussion

3.1 System improvement and update of data

In the period between the previous and the current study, several publications such as on consumption figures provided by the industry or peer-reviewed consumption trends (Prevedouros et al., 2004a) as well as on measurements in waste streams became available (Morf et al., 2005; Petreas and Oros, 2006; Sakai et al., 2006b). This fact allowed deriving more appropriate consumption estimates and reducing the uncertainty in the estimates.

DecaBDE

Results for DecaBDE at the end of the nineties determined in this study compared with data estimated in Morf et al., 2002 show that the total DecaBDE use determined in this study is about 170 tons/year which is roughly half of the amount estimated in Morf et al., 2002 (320 tons/year). The distribution pattern determined in the actual study is different, as the E&E seems to be more important according to the actual study (70% instead of 45%). On the other hand, transport accounts only for 10% instead for 30%. The difference for construction is small (20% instead of 25%). Textiles (not including interior textiles in vehicles) do not play a relevant role for the DecaBDE use in Switzerland.

As a result of the lower consumption and other factors (e.g. time trends, residence time) the stock in the use process has been modelled as 2400 tons compared to 5600 tons in Morf et al., 2002. Both studies showed a small stock decrease. The total flow from use and production to waste management accounts for roughly 150 tons/year compared to 370 tons/year and the stock in waste management (landfills) is about 350 tons compared to 620 tons for the same time estimated in Morf et al., 2002.

Total emissions to environment have been modelled as 50 kg/year compared to 2.2 tons/year in Morf et al. In the previous study, the vast majority was estimated to be emitted to the atmosphere (2.1 tons/year), while emissions to hydrosphere and to soil have been estimated as 10 kg/year and 45 kg/year, respectively. The emissions to hydrosphere and soil are similar in the present study. However, the atmospheric emissions have been estimated two orders of magnitude lower (15 kg/year) at the end of the nineties.

In the 2002 study diffuse emissions from the use phase were assumed to be the main emission source. This result was based on worst case estimates that had to be applied due to non available data at that time. In the present project, more field measurement data could be incorporated in the model. According to the calculations in the present study, the largest proportion of emissions is released from waste management, especially via the wastewater pathway. However, the atmospheric emissions seem to be underestimated in the present study based on chamber emission experiments and other measurements published (see section 4.2).

HBCD

For HBCD no data was available at all in Morf et al., 2002.

PentaBDE / BDE-47

Results for BDE-47 (or PentaBDE for the consumption trend) at the end of the nineties determined in this study have been compared with data estimated in Morf et al., 2002 for PentaBDE. The consumption of PentaBDE is about the double (around 4 tons/year) as estimated in the previous study for the end of the 1990s. As the consumption has been estimated to be declining at that time, the comparison is strongly dependent on the exact year looking at. While in the previous study the whole consumption was assigned to the application area *transport*, in the present study, also a reasonable share to E&E (32%) and textile/furniture

(12%) has been assumed. The reason of the high proportion in E&E is that due to import from Asia the consumption in this application area had not been declining as strongly as in other application areas. In a study in WEEE, the significant presence of PentaBDE could be shown (Morf et al., 2005). Construction materials, which have been estimated to be responsible for the largest proportion of the consumption in earlier years in the current study, have been estimated to consume approximately 18% of the PentaBDE by the end of the nineties. The largest share has been estimated to be consumed in the application area *transport* (38%).

The stock of BDE-47 in the use phase has been modelled as 55 tons corresponding to 140 tons of PentaBDE. A stock of 500 tons consisting of 91% PentaBDE in construction materials was estimated in the previous study. This difference mostly resulted from the higher estimate of consumption of PentaBDE in earlier years, especially in construction materials. Both studies show a clear stock decrease. The total flow of BDE-47 from use and production to waste management accounts for roughly 1.7 tons/year corresponding to 4.3 tons/year PentaBDE. For the same reasons as stated above, the flow estimated in the previous study was significantly higher (30 tons/year). Therefore and due to a longer residence time of construction materials in the use phase, the stock in waste management (landfills) has also been modelled much lower in this study (14 tons compared to 130 tons).

Total emissions of BDE-47 to environment have been modelled as 2.5 kg/year. This amount could not be directly converted to an emission flow of the sum of PentaBDE congeners due to their different physical properties (e.g. vapour pressure or the octanol air partition coefficient), but an emission figure of approximately 5 kg/year might be realistic. The emission of PentaBDE was estimated between two and three orders of magnitude higher (1.9 tons/year) in Morf et al., 2002. Like for DecaBDE, the largest part was estimated to be emitted to the atmosphere from diffuse sources in the previous study. In the previous study, emissions to hydrosphere and to soil have been estimated as 6 kg/year and 36 kg/year, respectively. While these estimates are one order of magnitude higher than in the current study, the atmospheric emission estimate is between two and three orders of magnitude higher. The reason for this difference is the same as stated above.

3.2 Temporal trend results

3.2.1 Temporal trends of the use figures

The estimated consumption trends for DecaBDE, HBCD and PentaBDE in Switzerland are based on the different approaches described in chapter 2.4.2.1. The bottom-up consumption estimate (including the estimate based on plastics consumption) as well as the available measurements in waste streams allowed splitting up the consumption in four application areas. The figures on industrial demand in Europe have been used to crosscheck the estimates. In the case of PentaBDE, the estimated use of PentaBDE in Europe (Prevedouros et al., 2004a) have been used to estimate the Swiss consumption in addition to the other approaches. The domain of uncertainty for the different substances and years could not be calculated. It has roughly been estimated to be between ±30% and ±50%. For the first half of the 1980s, the domain of uncertainty has been assumed as ±50% for DecaBDE and HBCD, dropping to ±30% by the end of the 1990s. The upper boundary of the uncertainty of the future consumption trend has been assumed to rise to 50% by the end of the period under study. The lower boundary is not given, as a future ban lowering the consumption drastically is possible. For PentaBDE, the uncertainty has been estimated as ±50% for the whole period, as the uncertainty in recent year has been assumed to be higher as for DecaBDE and HBCD.

DecaBDE

The consumption of DecaBDE in Switzerland has been estimated to increase during the 1980s and to reach a maximum of 220 tons/year in the beginning of the 1990s (figure 24). A small reduction to 175 tons/year by the end of the 1990s has been assumed to be followed by a nearly constant trend. The highest proportion of the amount consumed is in E&E, followed by the two application areas transport and construction. Textiles and furniture have been estimated to be of minor importance. The main reason of the decrease in the DecaBDE consumption has been estimated to be due to the reduction of the use in construction materials. DecaBDE was replaced by HBCD in XPS due to the implementation of a new process technique (see section below). As stated in the section 2.4.2.1.6, the consumption of DecaBDE has been assumed to stay constant during the next decade, if the substance would not be banned.

Compared with the per capita European industrial demand (converted on the base of the inhabitants of EU15 including Norway and Switzerland as well as the inhabitants of the whole Europe excluding Russia), the estimated Swiss consumption is higher. As imports of electronic equipment from Asia and America have been assumed to be of importance for DecaBDE, the higher estimate compared with the per capita industrial demand is reasonable. The consumption in Japan shows a different trend, but the per capita consumption is approximately in the same range. Due to an industry commitment, the DecaBDE consumption decreased since the beginning of the 1990s.

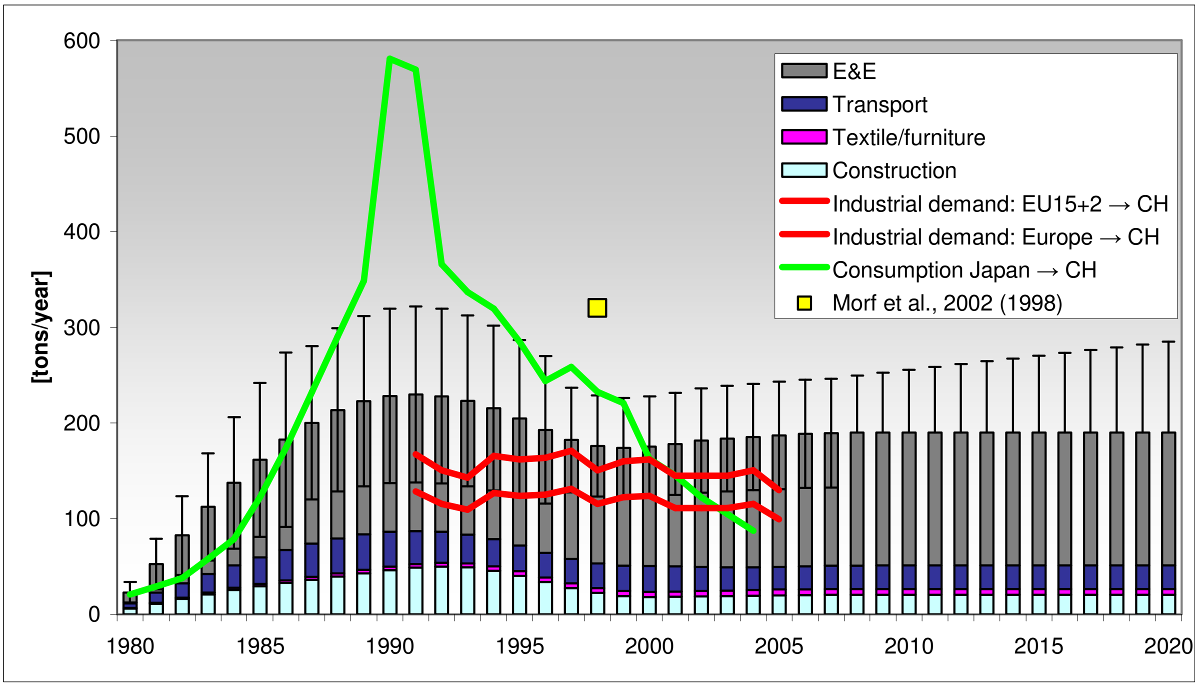


Figure 24 Estimated consumption trend of DecaBDE in Switzerland and comparison to industrial demand and Japanese data converted on a per capita base (source industrial demand: BSEF and CEFIC; source consumption Japan: Tokai et al., 2004)

HBCD

For HBCD, the consumption in Switzerland has been estimated to increase during the whole period from 1980 to 2020. The consumption has reached 100 tons/year in the mid-nineties and has almost doubled (181 tons/year) up to present. The construction materials EPS and XPS make up the by far most relevant share of the HBCD consumption. It amounts to 84% in the present year. The application areas transport (9%), textiles/furniture and E&E (3.5% each) are of smaller importance. A future increase of the use in the construction sector has been assumed. For the other application areas, the trend has been assumed to be constant over the next decade, if HBCD would not be banned (see section 2.4.2.1.6 and figure 25).

The per capita consumption in Switzerland is comparable to data on the European industrial provided by BSEF and CEFIC. As opposite to DecaBDE, imports of finished products have been estimated to be of minor importance, because European industry is the major HBCD consumer. 56% of the HBCD was consumed by the European industry in 1999 and 44% in 2003 (see table 6 and table 7 on page 23). Additionally, the trade between different continents is not as important for construction materials as for E&E. The step in the mid-nineties is caused by a flame retardant substitution. XPS foam that is exposed to a higher thermal impact during extrusion than EPS was flame-retarded with more stabile aromatic substances (DecaBDE) earlier. After the implementation of a process technique with reduced heat DecaBDE was substituted by the aliphatic substance HBCD (BASF, 2000 cited in Leisewitz and Schwarz, 2001).

As East European countries have stringent fire regulations (see section 2.4.2.1.3), the per capita conversion from the European industry demand has been assumed to be more appropriate to be based on the inhabitants of whole Europe (excluding Russia). As Switzerland has also stringent fire regulations, it is reasonable that the per capita use of HBCD is slightly higher than the European average (lower red line in figure 25). While the HBCD consumption in Asia is considerably lower than in Europe generally, Japan uses comparable amounts on a per capita base as Switzerland or Europe.

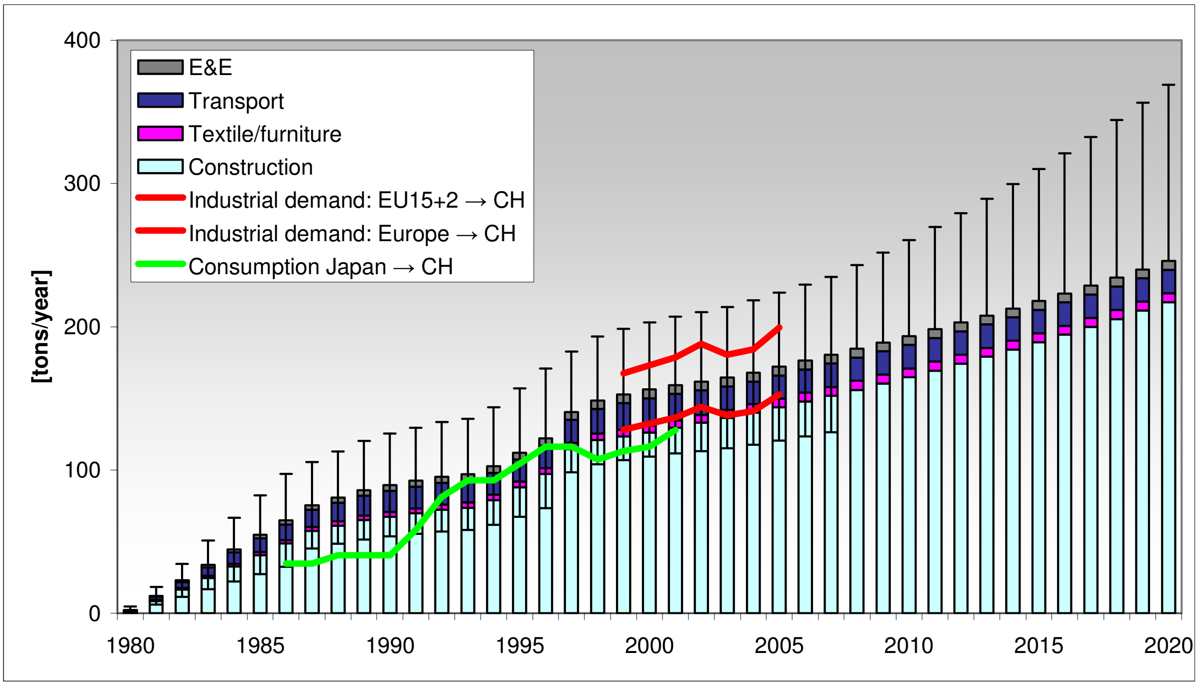


Figure 25 Estimated consumption trend of HBCD in Switzerland and comparison to industrial demand and Japanese data converted on a per capita base (source industrial demand: BSEF and CEFIC; source consumption Japan: Watanabe and Sakai, 2003)

PentaBDE

The consumption of PentaBDE in Switzerland has been estimated to increase during the 1980s and to reach a maximum of 14 tons/year in the beginning of the 1990s (see figure 26). A strong decrease has been estimated for the mid-nineties. In the year 2000, the consumption has been estimated as 2 tons/year. Virtually no consumption of PentaBDE has been assumed from the year 2006 onwards. Even though there is still a use of PentaBDE in some countries (e.g. China), the import in products to Switzerland has been estimated to be very small. In Asia, where PentaBDE had been used in printed circuit boards earlier, it has been estimated that it has been substituted with either TBBPA or non-halogenated flame retardants. Therefore, the amounts used in E&E might have decreased markedly in the past years. The largest share in the consumption has been estimated to be the use in rigid PUR foam in the construction sector. Flexible PUR foam in vehicles and furniture, PVC sheeting as well as printed circuit boards are other applications, where PentaBDE was used.

European industry demand data were only available for the years 1999 and 2001. The estimate for Switzerland is lower for this period compared to these numbers converted on a per capita base. As there are more stringent fire regulations for the United Kingdom for textiles and furniture, a large proportion of the European PentaBDE consumption has been assumed to be in flexible PUR foam in this area. Therefore, the estimated Swiss consumption has been smaller than estimated on the per capita base from the data supplied by BSEF and CEFIC. However, some of the difference has been assumed to be compensated by imports in products. The trend estimated by Prevedouros et al. (2004a) for Europe is also shown in figure 26. As it has been taken into account for estimating the consumption trend in Switzerland, it is not further discussed here. There were only data for the TetraBDE consumption trend available for Japan, but none for PentaBDE.

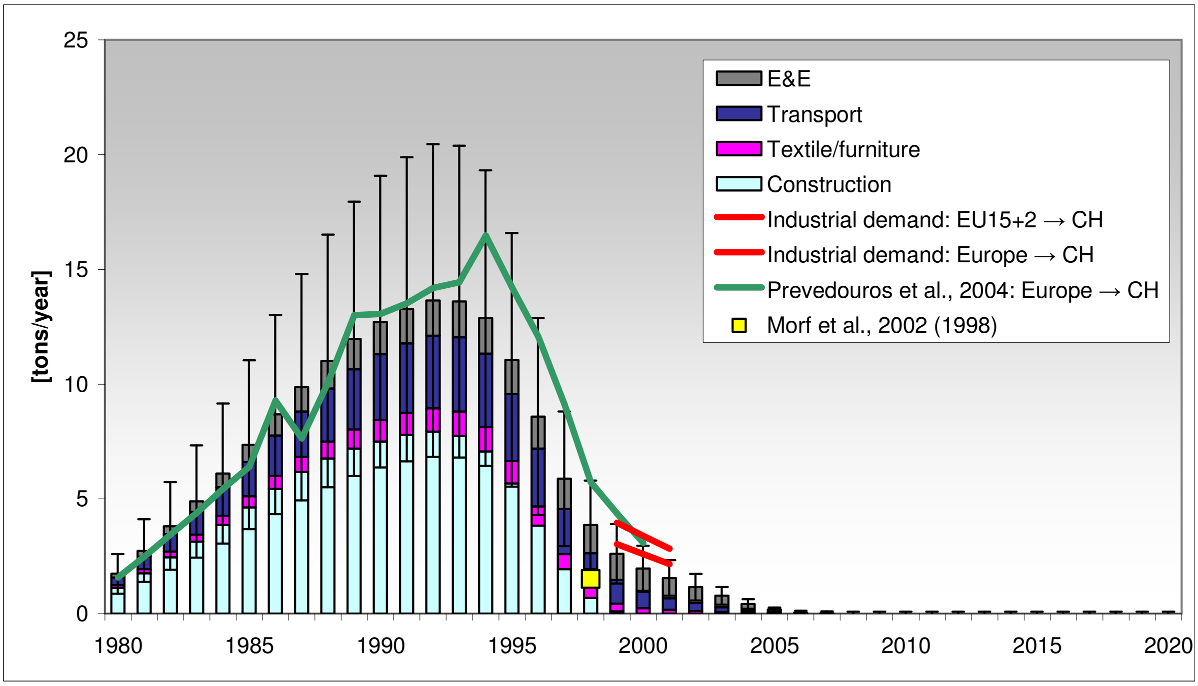


Figure 26 Estimated consumption trend of PentaBDE in Switzerland and comparison to industrial demand and estimates for Europe data converted on a per capita base (source industrial demand: BSEF and CEFIC)

3.2.2 Temporal trends of stocks and flows in the anthroposphere

DecaBDE

The stock of DecaBDE in the process *use* has been modelled to reach a maximum of 2,400 tons in 1999. A small stock decrease to 2,100 tons until 2020 is expected. Electrical and electronic equipment is the most relevant application area.

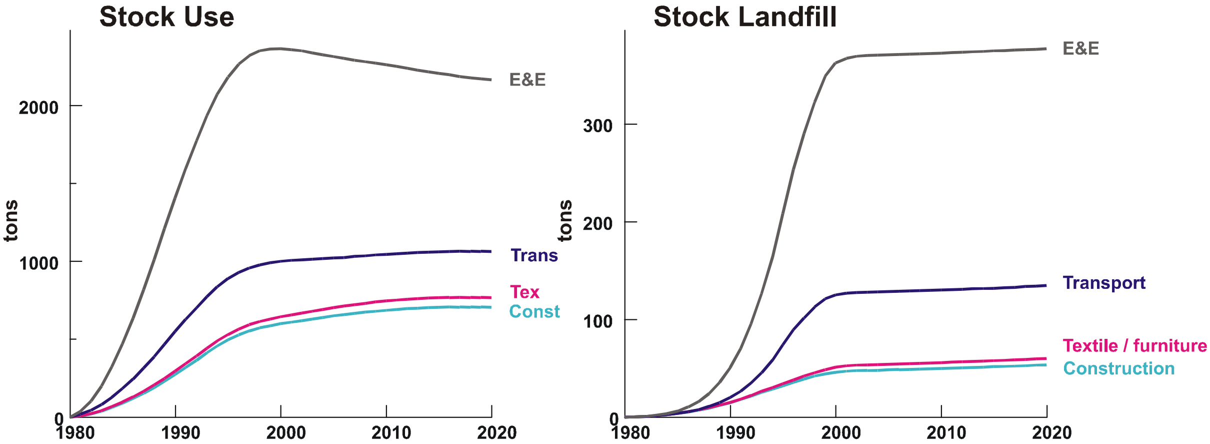


Figure 27 Trends of stocks of DecaBDE in the processes *use* and *landfill* (cumulative graphics)

The flow of DecaBDE to waste management increased markedly in past to about 190 tons/year at present (see section 7.6.1 in the appendix). Until the end of the period under investigation, it is expected to stay virtually constant. While a fraction of 32% to 45% has been modelled to be dumped on landfills (including dumping of recycled materials) between the 1980s and the mid-nineties, this fraction has significantly decreased to less than 1% at present. The fraction of the flow being incinerated in MSWIPs (including combustible residues from recycling) has been estimated to decline from 65% at the beginning of the 1980s to 50% in the mid-nineties, followed by an increase to 73% at present. On the other hand, the fraction in recycled materials being exported or reused in production increased from virtually zero to about 25% at present. The most important proportion of the latter flow is DecaBDE in metal fractions from WEEE and automobile recycling that have been assumed to be used in non-domestic industry. The amount of DecaBDE in exported APC residues is much smaller. The vehicles and E&E exported directly from the use phase to abroad are not included in the flow to waste management. At present, about 14 tons/year of DecaBDE are exported in old vehicles and about 2 tons/year in old electronic equipment.

As an example figure 28 shows the stocks and substance flows in the anthroposphere for the application area *E&E* for the year 2000 (figures of all application areas and substances are included in section 7.5 in the appendix). In that year the stock in the process *use* accounted for 1,360 tons (stock change −10 tons) and to 237 tons in the stock on landfills (stock change +4 tons). The largest substance flows are the imports to *production* and to *trade*, the export of DecaBDE in products to abroad as well as the internal flows from *trade* to *use* and from *use* to *recycling*. Smaller substance flows are between the processes *use* and *incineration* as well as between *recycling* and *incineration*. All the other substance flows have been modelled to be below 4 tons/year. Most of the DecaBDE in end-of-life products was recycled and incinerated subsequently; a smaller part was incinerated directly. About 100 tons of DecaBDE were destroyed in MSWIPs in the year 2000, while 30 kg were emitted to the environment.

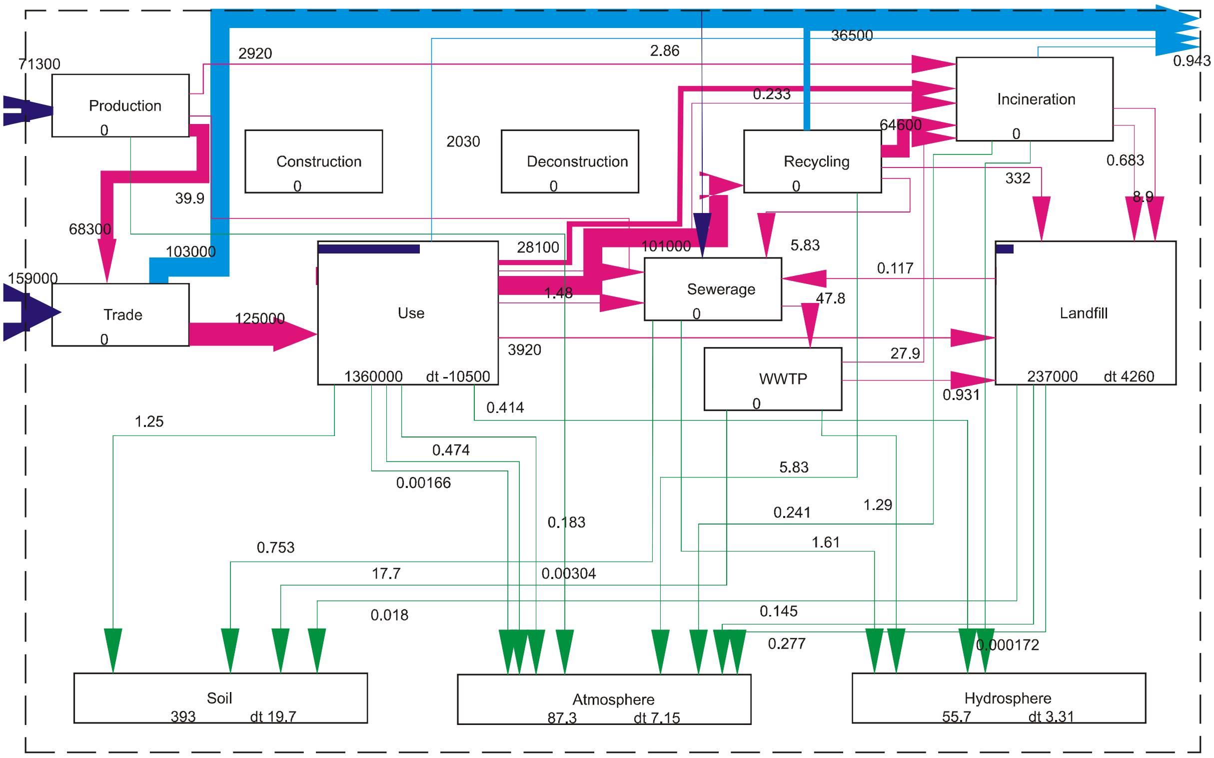


Figure 28 DecaBDE substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *E&E* for the year 2000

The amounts stored in waste management are almost one order of magnitude lower than in the use phase. Approximately 360 tons have been modelled to be on landfills in the year 2000, of which two third originate from the application area *E&E* (see figure 28). The highest growth rates of stocks on landfills were in the mid-nineties, when high amounts of products containing DecaBDE reached the end of service life and still a substantial part of the waste was dumped on landfills. In consequence of the technical ordinance on waste (Technische Verordnung über Abfälle, TVA) and the restrictions on the disposal of combustible waste on landfills, the accumulated BFR mass in landfills has increased much slower in recent years than before the mid-nineties. The contribution of the application areas E&E and transport are higher in landfills compared to the stock in the use phase. On the other hand, due to the longer residence time in the use phase, the contribution of construction materials is lower in landfills.

HBCD

As illustrated in figure 29, the stock of HBCD in the process *use* is about the same as for DecaBDE at present. In contrast du DecaBDE, the amount of HBCD in the use phase is not expected to decrease in the future, but to double until 2020. EPS and XPS insulation panels make up by far the largest proportion of the stock. The contribution of the other application areas to the stock is of minor importance due to the lower consumption and the shorter residence time in the use phase.

The flow to waste management amounts at present to approximately 30 tons/year, of which the vast majority originates from the use phase including construction and deconstruction processes (see section 7.6.1 in the appendix). Like for the stock in the use phase, also a doubling until 2020 is expected for this flow. About two third of the waste have been modelled to be dumped on landfills (including dumping of recycled materials) between 1980 and the beginning of the 1990s. This proportion has significantly decreased to less than 1% at present. On the other hand, the fraction of the flow finally being incinerated in MSWIPs has been modelled to increase from one third during the eighties to 70% at present. The reuse of recycled polystyrene from waste insulation panels in the industry has been expected to be negligible until the end of the nineties, but to increase subsequently to 20% of the flow to waste management in 2020. The proportion in recycled fractions and APC residues being exported has increased from virtually zero to about 20% at present. The most important proportion of the latter flow is HBCD in metal fractions from automobile and WEEE recycling that have been assumed to be used in non-domestic industry. The export of HBCD in APC residues is negligible. About 8 tons/year HBCD are estimated to be exported in old vehicles and 100 kg/year in E&E directly from the process *use*.

The stock in landfills is presently about 87 tons, which is by a factor four lower than for DecaBDE. The difference is mainly due to the fact that the residence time of insulation panels has been assumed to be 50 years, which causes a long lag between the consumption and the flow to waste management. Hence, only a small percentage of the amount of HBCD consumed in EPS and XPS got into the flow to waste management before the restrictions on the disposal on landfills came into effect. For the same reasons, the proportion of construction materials on landfills is not as large as in the stock in the use phase. The share of HBCD attributable to the other application areas amounts to about 40% to date.

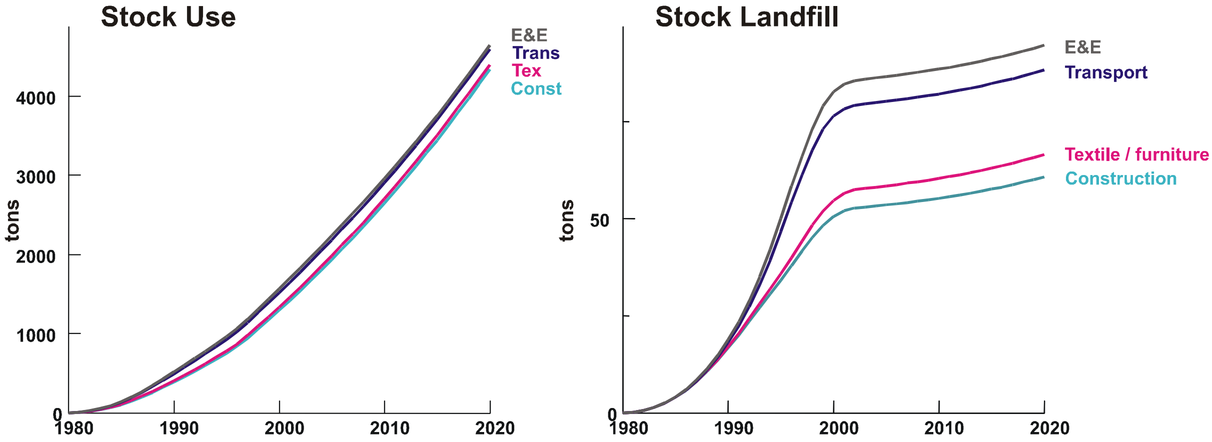


Figure 29 Trends of stocks of HBCD in the processes *use* and *landfill* (cumulative graphics)

BDE-47

The stock of BDE-47 in the process *use* has been modelled to reach a maximum of 56 tons in 1998 (see figure 30). A stock decrease to 18 tons until 2020 is expected. Construction materials have been estimated to be the most relevant application area. The proportion of BDE-47 in this application area has been modelled to be about 60% in 1998 and due to their longer residence time it is expected to converge at 100% in the next decade.

The flow to waste management has peaked in recent year at 2 tons/year. In contrast to DecaBDE and particularly to HBCD, a decrease of this flow by a factor of two until the year 2020 is expected (see section 7.6.1 in the appendix). While more than half of the waste was finally (i.e. including recycled fractions) dumped on landfills until the mid-nineties, this fraction has significantly decreased to about 1–2% in the period between 2002 and 2020. Approximately 40–50% of the flow to waste management was finally incinerated from 1980 until the mid-nineties. This fraction is expected to increase to 96% until 2020. The fraction of BDE-47 in recycled materials unintentionally reused in production has been estimated to be small over the whole period, reaching 1% in 2020. The export of recycled fractions increased from virtually zero to about 31% at the beginning of the present decade. About 350 kg/year and 150 kg/year in the application areas *E&E* and *transport* are estimated to be exported to non-domestic industry at present. As the proportion of these two application areas in the waste stream will become smaller over time, the proportion of the flow to be exported is expected to decrease to 1% by 2020. The export of BDE-47 in vehicles to be further used abroad is in the same order of magnitude as the export of recycled materials.

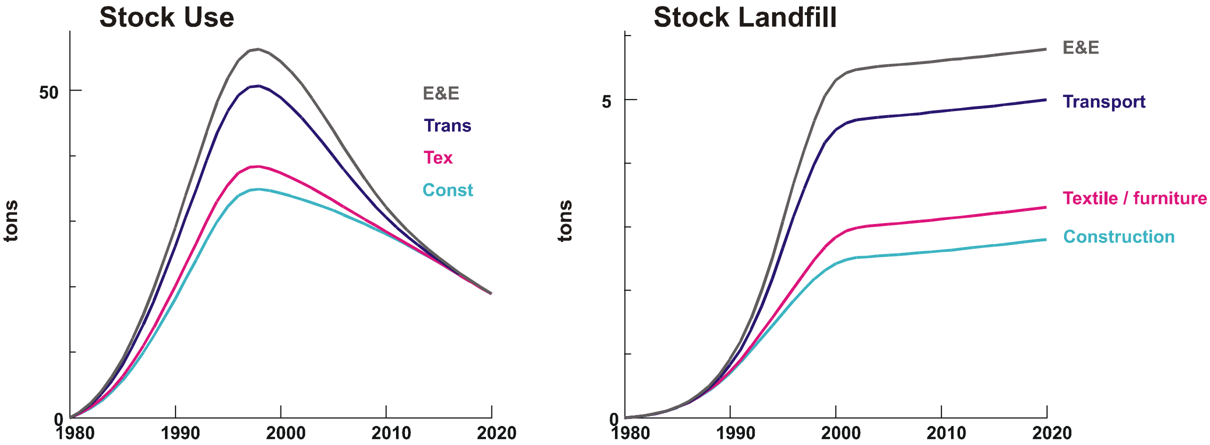


Figure 30 Trends of stocks of BDE-47 in the processes *use* and *landfill* (cumulative graphics)

As illustrated in figure 30, the stock on landfills is about a tenth of the stock in the use phase. The curve shape is comparable to DecaBDE and HBCD, as the most important factor was the change of policy in waste management. Apart from PUR foam used in the construction sector, dumped ASR have been modelled to make up a large proportion of the stock in landfills.

3.2.3 Temporal trends of flows and emissions to environment

The results from the dynamic SFA modelling are presented in the following sections. Please note that these results are based on the best estimate parameters depicted in the section 2.4.2. As the domain of uncertainty has been estimated to be large for the emissions to the environment and releases to wastewater, the emissions are also subject to large uncertainties. This aspect is discussed in section 3.5.

DecaBDE

The releases to sewerage from production processes have been most relevant, while smaller amounts originated from the use phase (6% in the year 2000), recycling (9%) and atmospheric deposition (7%). On the other hand, releases from landfills and from flame-retarded textiles to sewerage are negligible (see section 7.6.1 in the appendix). Wastewater has been modelled to be the most relevant emission pathway into soil and hydrosphere, especially in the past (note that even though emissions from sewerage and WWTP are accounted to the subsystem waste management, they are closely related with the processes *production* and *use*).

Kohler et al. (2003) measured the concentration of different PBDE congeners in sewage sludge from eight WWTP in the greater Zurich area taken in the years 1993 and 2002. The mean DecaBDE concentration of the eight samples was 220 ng/g d.w. (range 99–550 ng/g d.w.) in 1993 and 1100 ng/g d.w. (range 220–1700 ng/g d.w.) in 2002. The amount of annually produced sewage sludge (d.w.) is given as 209,000 tons/year in that publication. As the percentage of DecaBDE passing WWTP is minimal, the annual flows to WWTP have been derived as 46 kg/year (range 21–115 kg/year) in 1993 and 230 kg/year (range 46–355 kg/year) in 2002. Sewage sludge originating from 16 sites of a monitoring network was sampled and specific loads were calculated in a Swiss study (Kupper et al., unpublished work). The WWTP studied included (a) separate sewer systems and rural catchments without industrial activities apart from a few craft industries (b) combined sewer systems and rural catchments including some more craft industries and (c) combined sewer systems and urban catchments including industry and craft industry. However, the accuracy of results for DecaBDE is uncertain as no isotope labelled standard was used in the analytical method. The results are presented anyway: The mean specific load calculated from concentrations found in sewage was 6 mg/inhabitant/year (range 4–14 mg/inhabitant/year). The mean specific loads for the groups *a*, *b* and *c* were 4.5, 5.3 and 7.2 mg/inhabitant/year. The flow to WWTP in Switzerland that has been derived from the data is 45 kg/year (range 30–100 kg/year). In a recent Spanish study (Alonso et al., 2007), the concentration of PBDEs in sewage sludge from 25 different WWTPs was measured and releases to wastewater were estimated based on the results. The 95% confidence interval for DecaBDE is given as 4.4–8.4 mg/inhabitant/year. Converted to Switzerland, the annual releases to wastewater would be 33–62 kg/year. There are some other studies on concentrations in sewage sludge, but normally the number of people connected to the WWTP or the amount of sewage sludge produced is not given, which would result in a larger uncertainty when estimating releases from these data. The concentrations found in sludge range by up to two orders of magnitude.

In the present study, the substance flow into the sewer system has been modelled as 95 kg/year at the beginning of the 1990s and to drop to about 40 kg/year at present (see section 7.6.1 in the appendix). These numbers correspond quite well to the measurements in Switzerland and Spain. However, a decreasing trend during the past decade has been modelled, while the measurements in the greater Zurich area in 1993 and 2002 indicate a significant increase. It is uncertain, if the increase found at seven of eight sample locations can be extrapolated for Switzerland or if the result would have changed, when a larger number of WWTP in different areas of Switzerland would have been sampled. Releases from production or the use phase has not been estimated to increase, as the substance flows and stocks in these processes have approximately been constant over the past decade. Therefore, releases from waste management have been suspected to possibly be underestimated. With reservations of the accuracy of the measurements carried out by Kupper et al., the releases from the use phase have possibly been underestimated as the mean in the group *a*, where industrial are very limited and there are no inputs from storm water surface runoff, is not much lower than in group *c*.

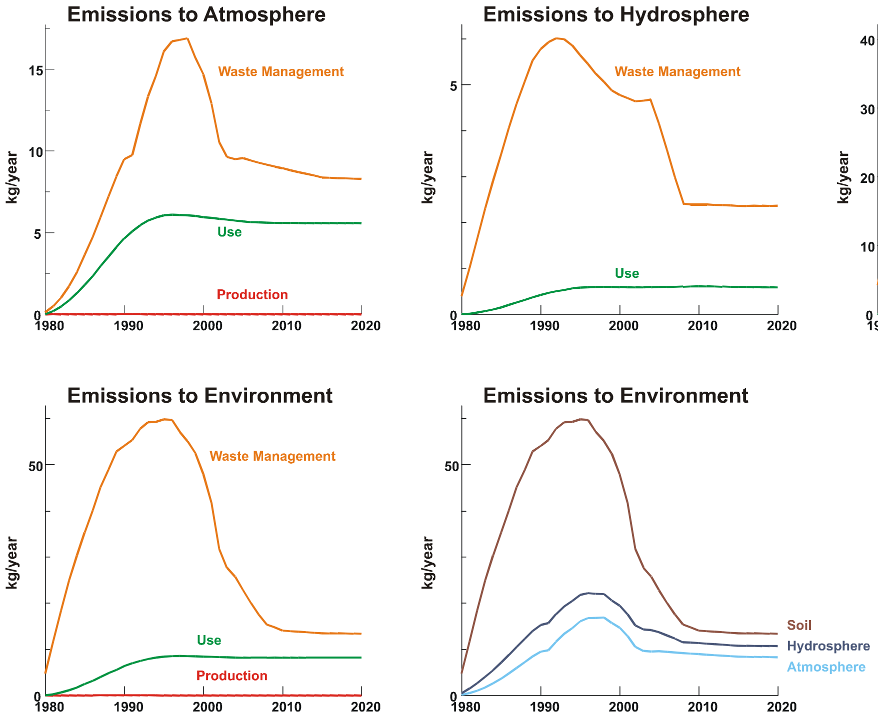


Figure 31 Emissions of DecaBDE to (*a*) atmosphere, (*b*) hydrosphere, (*c*) soil, environment split up by (*d*) source and (*e*) compartment (cumulative graphics)

Figure 31 shows that the total emissions of DecaBDE to the environment ranged between 10 and 60 kg/year during the period investigated. There was an increase until the middle of the 1990s, followed by a decrease until the late 2000s. The emissions have been expected to stay virtually constant within the next decade, when no ban has been assumed. Total emissions estimates at the end of the nineties are more than one order of magnitude lower than estimated in Morf et al. (2002), as worst case assumptions had to be applied in that study (see section 3.1).

Emissions to atmosphere made up to 17 kg/year, emissions to hydrosphere up to 6 kg/year and emissions to soil up to 40 kg/year at maximum.

The largest proportion of the emissions to environment has been in the flow from WWTP to soil (see figure 32). As the usage of sewage sludge as fertilizer in agriculture declined over time and was prohibited beginning October 1^{st} 2006, the emissions to soil decreased significantly. The emissions from WWTP effluents and sewer overflows to hydrosphere and due to leakages in the sewer system to soil also considerably contribute to the emissions. The most important sources for atmospheric emissions have been diffuse releases from products during the use phase (all application areas) and recycling processes (*E&E* and *transport*). Emissions of DecaBDE from production processes, building and vehicle fires, construction and deconstruction as well as from landfills have been estimated to be of minor importance. MSWIPs have been expected to contribute to a notable extent to atmospheric emissions during the eighties. As the emission factors were reduced due to technical measures taken in the off-gas treatment, the there are only minor emissions expected for the recent years.

From the figures in the appendix (section 7.6.3) for the individual application areas, it can be seen that *E&E* has been expected to be responsible for about half of the emission to the environment. Vehicles, construction materials as well as textiles and furniture together account for the other half.

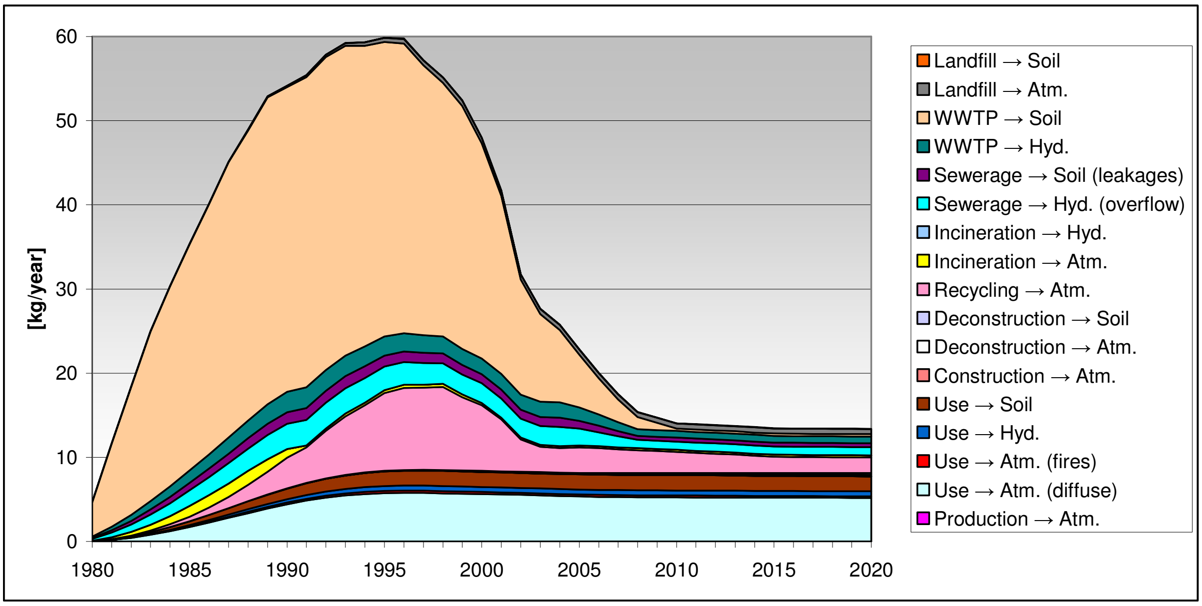


Figure 32 Emissions of DecaBDE to environment split up by the different emission sources

A study on the temporal trends of substance flows, stocks and emissions to the environment was recently carried out in Japan (Yamaguchi et al., 2006). It was written in Japanese and was made available by personal contacts to several research groups in Japan. Like in the present study, a lifecycle-based dynamic substance flow analysis was used to model the nation-wide DecaBDE emission quantity to atmosphere, hydrosphere and soil applying emission factors and different coefficients based on literature research. The emissions were modelled for the period 1978–2014. The emissions to atmosphere were modelled as 120 kg/year in 1991 as the maximum and as 50 kg/year in the present year. Maximum emissions to hydrosphere were modelled to be in the year 2000 (420 kg/year) and current emissions to be about 250 kg/year. A maximum was also modelled for the year 2000 for the emissions to soil (500 kg/year). The current emissions to soil were modelled as 150 kg/year. Converted on a per capita base to Switzerland, the current emissions are 3 kg/year to atmosphere, 15 kg/year to hydrosphere and 9 kg/year to soil (figures in the present study 8, 2 and 3 kg/year). The most relevant sources of the atmospheric emissions modelled in the Japanese study are the production of flame-retarded plastics, recycling processes and diffuse emissions from the use phase. For the emissions to hydrosphere, the production of DecaBDE (production of the substance), the production of flame-retarded textiles and the WWTP effluents are of most importance. Virtually all emissions of DecaBDE to soil were estimated to be due to the usage of sewage sludge in the agriculture. Compared to the Japanese study, a higher atmospheric emission from the use phase but lower emissions from production processes have been modelled. About 70% of the emissions to hydrosphere originate from the production of DecaBDE and flame-retarded textiles. As the former has never been taking place in Switzerland and the latter has been assumed to be small in this country, the difference in the emission estimates between Japan and Switzerland seems to be reasonable. The difference in the emissions to soil are most probably caused by higher releases from industrial processes to wastewater in Japan and the individual fraction of the sewage sludge that is used in the agriculture.

It can be concluded that the results from the two studies on the emissions including the temporal trends are pretty similar, taking into account the country-specific consumption and differences in waste management. However, this is not surprising as essentially the same literature data on emission factors were available for both studies.

HBCD

As opposite to DecaBDE, releases from production processes (11% in the year 2000) do not contribute as much to the wastewater pathway. Diffuse releases from the use phase (20%) and from recycling (4%) have been estimated to be of comparable importance, while atmospheric deposition has been expected to be the most important source (65%) (see section 7.6.1 in the appendix). Only two single measurements of atmospheric deposition in urban areas were carried out in Europe and measurements from USA are hardly comparable due to much lower HBCD consumption there. As the two European measurements carried out in Stockholm differed by almost a factor of seventy, the domain of uncertainty is high for this flow. Releases from landfills and from flame-retarded textiles to sewerage have been modelled to be negligible.

Based on concentrations found in sewage sludge originating from 16 sites of a monitoring network in Switzerland, the mean specific load was calculated (Kupper et al., unpublished work). The mean HBCD concentration in sludge was 149 ng/g d.w. (range 39–346 ng/g d.w.) corresponding to a specific load of 3.3 mg/inhabitant/year (range 0.4–6.4 mg/inhabitant/year). The mean specific loads for the groups *a*, *b* and *c* were 1.2, 2.5 and 4.8 mg/inhabitant/year. On a level for the whole Switzerland, the converted release to wastewater is 24 kg/year (3–48 kg/year). For HBCD, there was no uncertainty in the accuracy of results due to the analytical method used. HBCD was not investigated in the other Swiss (Kohler et al., 2003) or in the Spanish study mentioned (Alonso et al., 2007). Therefore, the concentrations in sewage sludge have been compared to concentrations from countries. In sludge from three WWTP in Stockholm, concentrations in the range of 21–54 d.w. was measured (Sellström et al., 1999). Mean HBCD concentrations in sewage sludge from the Netherlands, England and Ireland were measured as 175, 1401 and 3322 ng/g d.w. (Morris et al., 2004). The concentrations in the individual samples ranged from <0.6 to 9120 ng/g d.w. In a recent Swedish study sludge from fifty WWTP were measured and a mean concentration of 45 ng/g d.w. was determined (de Wit et al., 2007). There are large differences between among the different countries. The high levels in England and Ireland are expected to be a consequence of the usage of HBCD for textile back-coating, while the lower levels in Sweden might be due to a reduced HBCD consumption for building insulation. The concentrations in Switzerland are in-between and comparable to the mean concentration found in the Netherlands.

The release of HBCD to wastewater has been modelled as around 30 kg/year in recent years (see section 7.6.1 in the appendix). This substance flow is in good agreement with the release estimated based on the Swiss sewage sludge study. Compared to the mean specific loads from the different groups of WWTP catchments, the percentage of the input from atmospheric deposition might be somewhat too high and the release from the process *production* somewhat too low. On the other hand, the release from the use phase modelled is in good agreement with the field data. However, the range among the different groups was too high to draw a conclusion.

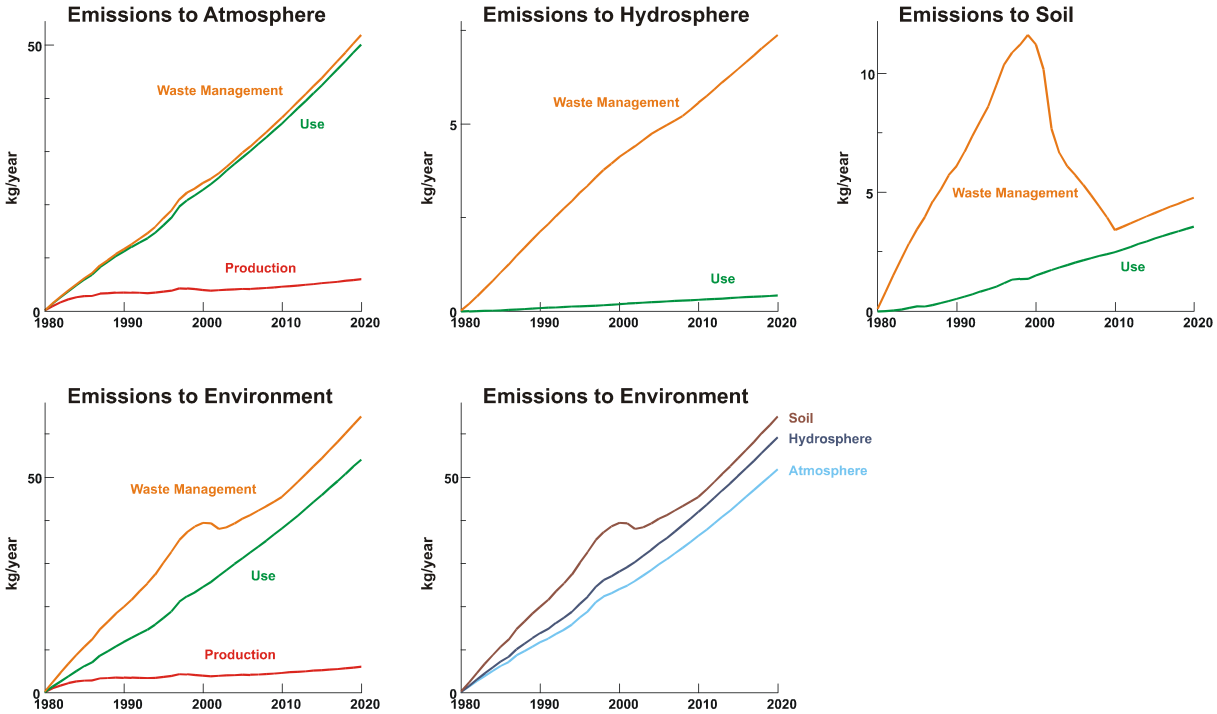


Figure 33 Emissions of HBCD to (*a*) atmosphere, (*b*) hydrosphere, (*c*) soil, environment split up by (*d*) source and (*e*) compartment (cumulative graphics)

Emissions of HBCD have been modelled in a similar range like DecaBDE, but in contrast to this substance, the emissions of HBCD have been increasing up to date and have been expected to continue increasing in future (see figure 33). The largest proportion of the emissions has been modelled to get to atmosphere. While these emissions have been expected to be as high as 50 kg/year in 2020, emissions to hydrosphere have been presumed as 7 kg/year for the same year. Emissions of HBCD to soil show a similar trend as for DecaBDE (figure 31), but the values are lower and peak significantly later.

The atmospheric emissions of HBCD from installed EPS and XPS insulation panels have been modelled to account for about half of the emissions to environment (figure 34). As the stock in the use phase has been growing over the whole period these diffuse emissions have also been modelled to increase to 30 kg/year in 2020. Atmospheric emissions from the manufacture of flame-retarded insulation panels have been modelled to amount to about 4 kg/year at present. Emissions from construction and deconstruction operations have also modelled to be of relevance, accounting for 8 kg/year at the moment. While emissions from deconstruction processes are expected to further increase for a long period due to growing substance flow from the use phase to the process *deconstruction*, emissions from process *construction* are highly dependant on the mass of HBCD consumed in insulation materials in future. Therefore, these emissions could be influenced rapidly by regulatory measures. HBCD is not retained in sewage sludge as effective as DecaBDE, due to its lower K_{OW}. Hence, a larger fraction of the substance entering into WWTPs is released to the effluent. The sewage sludge used in the agriculture led to emissions of 10 kg/year in 1999. As a result of fact that most of the sludge is incinerated today, this emission pathway could be reduced to about 2 kg/year at present. While emissions from the recycling of vehicles, insulation panels and E&E have been modelled to account for about 2% of the total, atmospheric emissions from the process *incineration* have only made up 0.1% starting from the nineties.

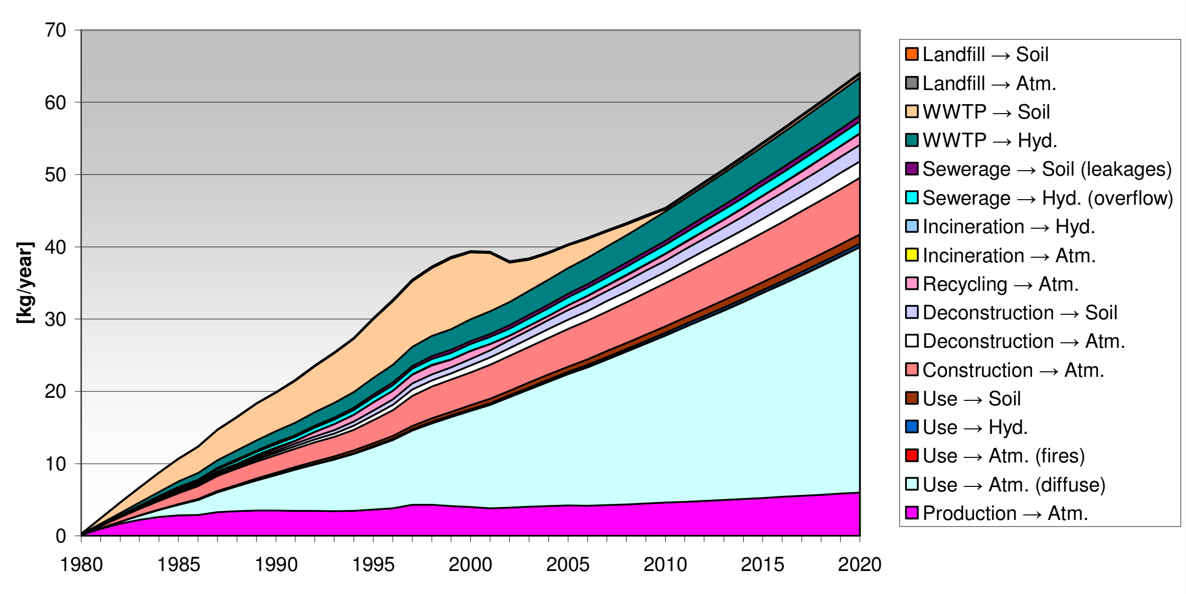


Figure 34 Emissions of HBCD to environment split up by the different emission sources

Construction materials are responsible for the majority of the emissions. The application areas *E&E*, *transport* and *textiles/furniture* have been modelled to account for 9 kg/year (23% of the total) at the end of the nineties and for 6 kg/year (15%) at present. The most important emission source within these three application areas are diffuse emissions during the use phase (3 kg/year between the mid-nineties and 2020), particularly from vehicles and textiles. The wastewater pathway including, especially emissions due to the usage of sewage sludge as a fertilizer and emissions from WWTP effluents to hydrosphere, produced emissions of 4 kg/year in the late nineties and produces 2 kg/year at present. Atmospheric emissions from automobile and electronic appliance recycling plants amounted to 1 kg/year at the end of the nineties. A 50% decrease has been expected for 2020. Figures of emissions from the individual application areas are included in the appendix (section 7.6.3).

BDE-47

The releases to sewerage from landfills (47% in the year 2000) and atmospheric deposition (39%) were most relevant, while smaller amounts originated from recycling (7%), the use phase (5%) and production processes (2%). As a result of the higher water solubility of BDE-47, flows from landfills to sewer have been expected to be of relevance for this substance (see section 7.6.1 in the appendix).

The study carried out in sewage sludge from eight WWTP in the greater Zurich area (Kohler et al., 2003) showed mean BDE-47 concentration of 58 ng/g d.w. (range 38–68 ng/g d.w.) in 1993 and 27 ng/g d.w. (range 12–38 ng/g d.w.) in 2002. The annual flows to WWTP have been derived as above as 12 kg/year (range 8–14 kg/year) in 1993 and 6 kg/year (range 2.5–8 kg/year) in 2002. The mean specific load calculated from concentrations found in sewage sludge originating from 16 sites of a monitoring network in Switzerland (Kupper et al., unpublished work) was 0.7 mg/inhabitant/year (range 0.3–2.9 mg/inhabitant/year). The mean specific loads for the groups *a*, *b* and *c* were 0.4, 0.7 and 1.3 mg/inhabitant/year. Converted for Switzerland, the number is 7 kg/year (2–21 kg/year).In contrast to DecaBDE, there was no problem in the accuracy of results for BDE-47 due to the analytical method used. In the Spanish study mentioned (Alonso et al., 2007), the 95% confidence interval for the specific BDE-47 load is given as 0.20–0.35 mg/inhabitant/year. Converted to Switzerland, the annual releases to wastewater would be 1.5–2.6 kg/year.

The release of BDE-47 to wastewater has been modelled to increase to 1.5 kg/year at the beginning of the present century and to drop to about 1 kg/year these days in the present study (see section 7.6.1 in the appendix). These numbers are clearly lower compared to the measurements in the greater Zurich area, especially for the first measurement. Also compared to the measurements carried out by Kupper et al., they correspond to the minimum estimate. On the other hand, the flows modelled in this study correspond quite well with the specific load estimated in the Spanish study. The modelled releases of BDE-47 to wastewater might be somewhat too low, but no significant difference between the confidence interval and the releases derived from the measurements in sewage sludge is expected. However, the releases to wastewater might have been underestimated, especially during the beginning of the period under investigation. Possible sources contributing to the releases in that period could be production processes, emissions from new products, atmospheric deposition and emissions from building fires. There are however not enough measurements in sewage sludge from that period.

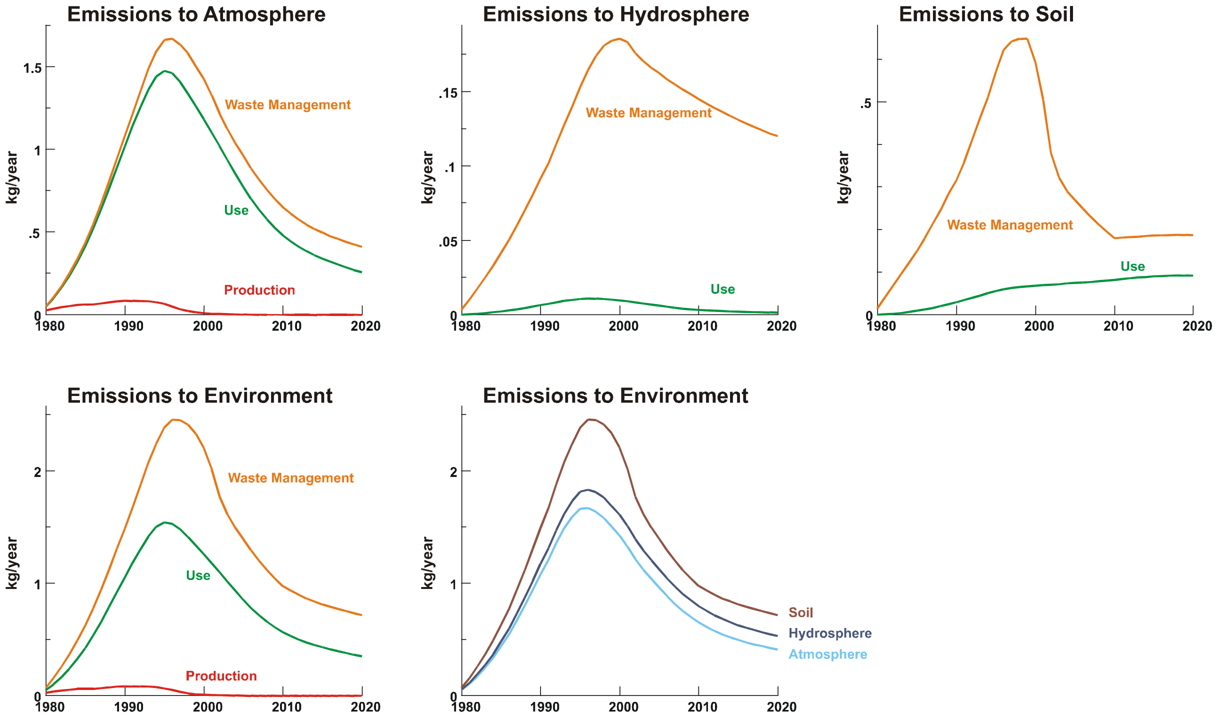


Figure 35 Emissions of BDE-47 to (*a*) atmosphere, (*b*) hydrosphere, (*c*) soil, environment split up by (*d*) source and (*e*) compartment (cumulative graphics)

Emissions of BDE-47 (figure 35) have been modelled as one order of magnitude lower than of DecaBDE and HBCD. There was an increase until the middle of the 1990s with a peak emission of 2.5 kg/year. Until the end of the investigated period, the emissions have been expected to drop below 1 kg/year. The highest proportion of the emissions has been modelled to be released to the atmosphere, while the wastewater pathway is of smaller relevance.

As illustrated in figure 36, diffuse emissions from products in the use phase have been expected to produce more than half of the total emission to the environment. Rigid and flexible PUR foams in constructions and in furniture as well as vehicles have been modelled to be the major sources for these emissions. Emissions from construction materials will continue for more than a decade, as the residence time in the use phase is longer compared to the other application areas. As there is no more consumption of PentaBDE in domestic production processes, emissions are expected to be virtually zero at present. Recycling of vehicles and

E&E has been modelled to contribute only during the nineties to a notable extent. The amount emitted from WWTP effluents has been modelled to stay approximately constant from the mid-nineties until the end of the period under investigation. Due to its higher volatility and mobility (water solubility), a notable proportion of BDE-47 is expected to be emitted from landfills to atmosphere and soil in 2020. Deconstruction of installed PUR foam has been modelled to produce increasing emissions to atmosphere and soil. However, they are still small compared to the emissions of HBCD caused by the same process. Figures of emissions from the individual application areas are included in the appendix (section 7.6.3).

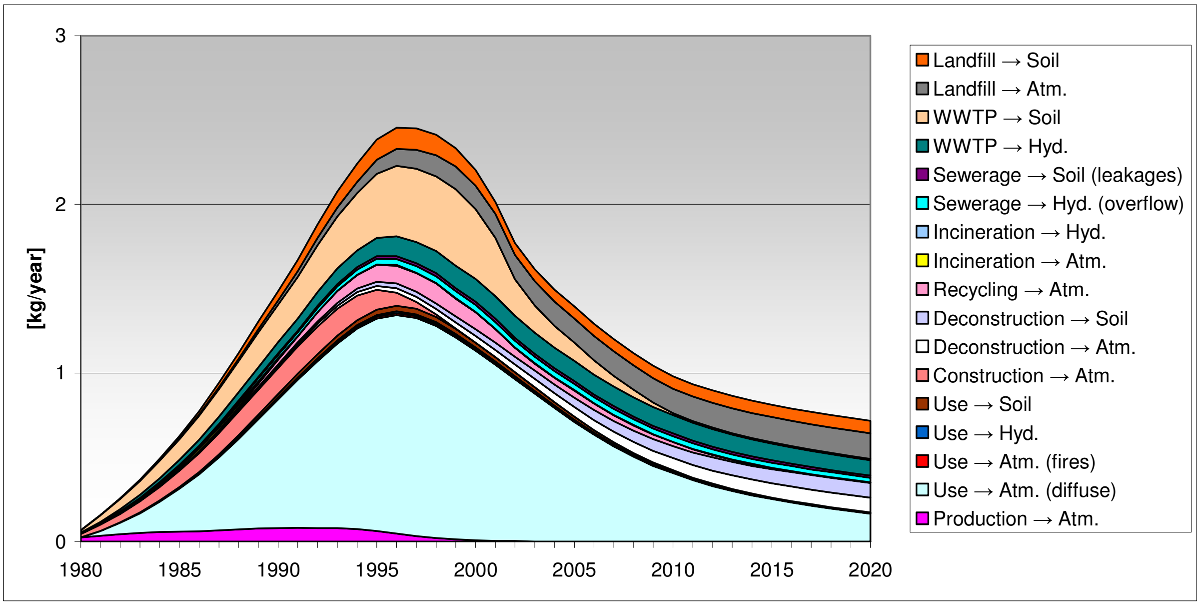


Figure 36 Emissions of BDE-47 to environment split up by the different emission sources

3.3 Comparison with EU risk assessment estimates

DecaBDE

The emission estimates in the EU risk assessment update of DecaBDE (ECB, 2004) are as follows (sum of regional and continental model): 35–145 kg/year to air, 12,530 kg/year to surface water, 159 kg/year to wastewater and 37,610 kg/year to industrial/urban soil for Europe (EU15). For some processes new information is available in the addendum of the EU risk assessment (ECB, 2005a), but no updated emission figures are given. Emission estimates from EU could not be compared directly with modelled Swiss data, because of different industry types, (fire) regulations and use patterns. Additionally, instead of emissions to the environmental compartments hydrosphere and soil, releases to wastewater were estimated only. In the EU risk assessment an 80% connection rate to the sewerage system was assumed in line with the recommendations in the latest Technical Guidance Document, which is much lower than for Switzerland.

Irrespective these facts and taking into account that Swiss population amounts to 1.88% of the population of EU15, the atmospheric emission of 9 kg/year modelled in the present study for 2004 is higher than the per capita emission in the EU. The releases to sewerage of 70 kg/year modelled for 2004 are also significantly higher than the per capita estimates made in the EU risk assessment for the EU. On the other hand, emissions to surface water (due to lower connection rate) and industrial/urban soil were estimated much higher in the EU risk assessment than in this study for Switzerland.

HBCD

In the February 2005 draft version of EU risk assessment of HBCD, total emissions of 4,537 kg/year to air, 11,228 kg/year to wastewater and 2,433 kg/year to surface water were estimated for Europe (EU15). In the October 2006 version, these estimates were revised down to 612 kg/year to air, 7,966 kg/year to wastewater and 2,248 kg/year to surface water, because of new data that got available.

The atmospheric emissions of 30 kg/year modelled in this project for 2005 are in-between the estimates made in the two draft risk assessment versions. On the other hand, the release to wastewater of 35 kg/year modelled for 2005 is significantly lower than the release estimates for the EU on a per capita base. This is mainly due to the fact that HBCD substance has been produced in the EU (the Netherlands) but not in Switzerland and the average industrial consumption of HBCD for textile back-coating has been estimated to be smaller than in the EU. Hence, this difference is reasonable taking into account the uncertainty.

BDE-47 / PentaBDE

The emission estimates in the EU risk assessment update of PentaBDE (ECB, 2000) are as follows: 43,200 kg/year to air, 5,260 kg/year to surface water, 180 kg/year to wastewater and 15,860 kg/year to industrial/urban soil for Europe (EU15). There is not much new information about emissions in the draft version of “Management Options for commercial PeBDE” (Norwegian Pollution Control Authority, June 2006). The atmospheric emissions of BDE-47 modelled for the year 2000 are about 1.5 kg/year. Taking into account the populations of EU15 and Switzerland and the fact that BDE-47 is only a part of the commercial PentaBDE product, there is a difference of two orders of magnitude between these estimates. It has to be emphasized that the release from PU foam, which was thought to make up the by far largest amount of the atmospheric emissions, was a worst-case estimate in the EU risk assessment. As argued in the section 2.4.2.5.4, the indoor air concentrations would need by far higher than all measurements conducted to result in such high atmospheric emissions. However, it is possible that these emissions have been underestimated in the present study. Especially for the atmospheric emissions from the use phase, there is a large range in estimated emission factors based on different methods.

The emissions of BDE-47 to hydrosphere and soil modelled for the year 2000 are about 0.18 kg/year and 0.7 kg/year, respectively. Converted on a per capita base to EU15, the emissions to hydrosphere would be 9.6 kg/year. For PentaBDE, that amount has been estimated as approximately 20 kg/year, which is two orders of magnitude lower than the sum of the emissions to surface and wastewater estimated in the EU RA (5,340 kg/year). However, the amount released to the sewer system would be reduced at a WWTP by a factor of about 10. The amount estimated to be emitted by “waste remaining in the environment” to surface water in the EU RA has been considered to be overestimated at that time. This is also the case for the emission to industrial soil, which was estimated to originate from “waste remaining in the environment” in the EU RA. The converted emission to soil modelled in this study (i.e. 37 kg/year for BDE-47 or approximately 70 kg/year for PentaBDE) is also two orders of magnitude lower than in the EU RA.

3.4 Influence of future and past legislative and technical measures

3.4.1 Ban on the usage of DecaBDE

For DecaBDE, two different scenarios have been compared to the reference scenario (see section 2.5).

A ban on the usage of DecaBDE in all application areas except *E&E* would lower the consumption by about 30%. The flow to waste management would be lowered from 190 tons/year by approximately 20 tons/year by 2020. The influence to the export flow has been modelled to be small, except for the export of goods from the process *trade*. The input to the sewer system has been modelled to decrease by about 7 kg/year to 23 kg/year by 2020. A reduction of the stock in the use phase from 2,300 tons in 2006 to 1,600 by 2020 has been modelled. On the other hand, there is virtually no influence on the stock in landfills, because the growth of that stock is in any case small these days and in future due to the prohibition of dumping combustible waste (see section 7.7.2 in the appendix).

If DecaBDE would be banned in 2007 for all application areas, the consumption would decrease to a small amount as only a minor proportion of waste materials containing DecaBDE has been expected to be reused in production. The flow to waste management has been modelled to drop to less than 50 tons/year in 2020. The releases to wastewater have been modelled to amount to about 8 kg/year in 2020. The influence on the stock in the use phase has been modelled to be much larger than if the usage of DecaBDE in E&E would be exempted from a ban. Instead of a decrease to 1,600 tons by 2020, the stock has been estimated as 550 tons for the same year (see section 7.7.2 in the appendix). This difference is of importance for diffuse emissions from the use phase.

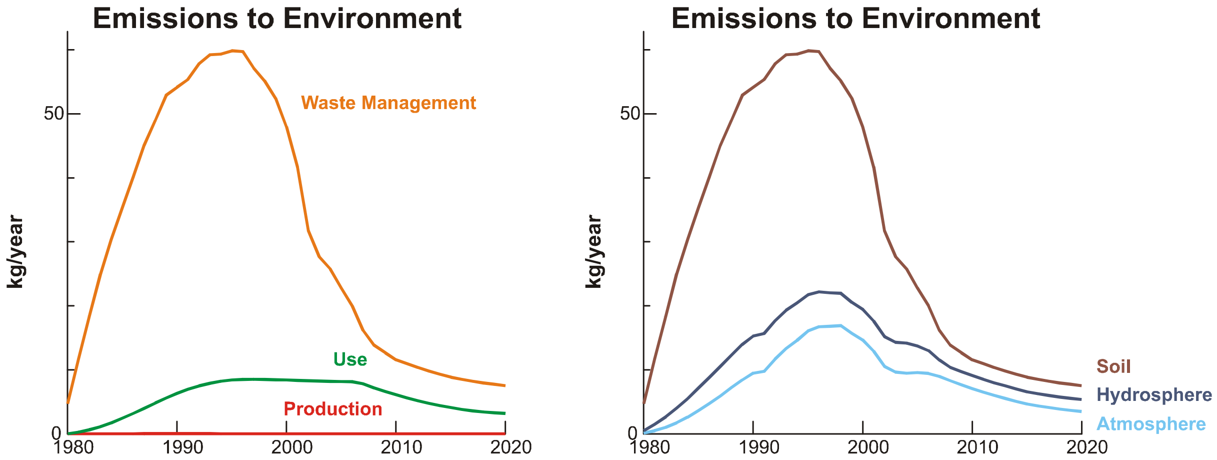


Figure 37 Scenario of a ban in 2007 except for E&E: emissions of DecaBDE to environment split up by source and compartment (cumulative graphics)

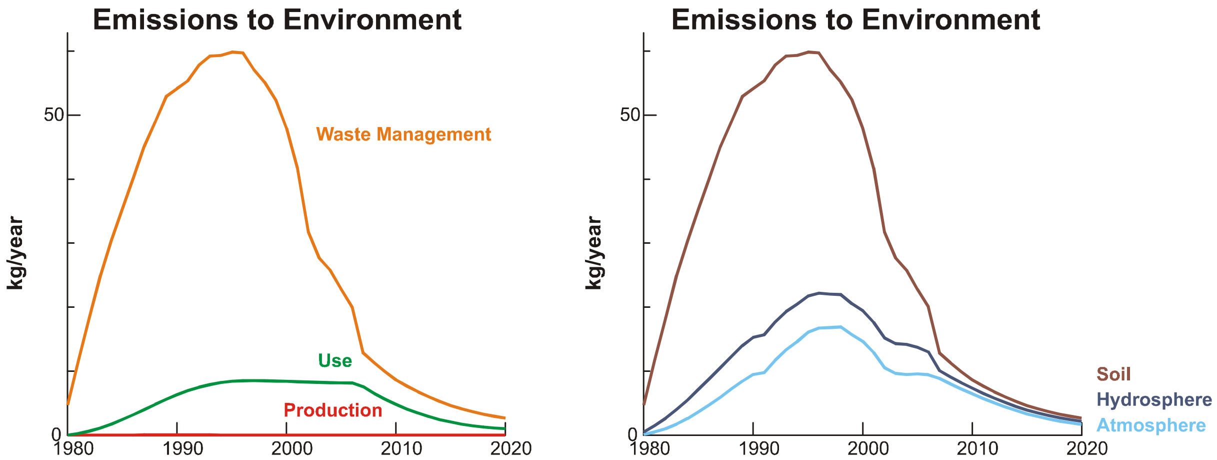


Figure 38 Scenario of a ban in 2007 for all application areas: emissions of DecaBDE to environment split up by source and compartment (cumulative graphics)

Emissions to the environment have been modelled to be 20 kg/year in the reference scenario for the present year and 13 kg/year for 2020 (see figure 31 on page 88). These emissions are divided in 8 kg/year to atmosphere, 2 kg/year to hydrosphere and 3 kg/year to soil. 8 kg/year originate from the use phase, while 5 kg are released from the waste management.

For the scenario with a ban in the application areas *transport*, *textile/furniture* and *construction*, the emissions of DecaBDE to the environment have been modelled as 9 kg/year for the year 2020 (see figure 37). Of these emissions, about 4 kg/year get to atmosphere and still about 2 kg/year to hydrosphere and 3 kg/year to soil. Split up by source, 4 kg/year are emitted from the use phase and 5 kg/year from the waste management. Thus, the atmospheric emissions from the use phase expected to be about 50% lower in 2020, if a ban would be enacted in 2007. The influence on the emissions from waste management and to the other environmental compartments is small. The reason why that the substance flow to waste management won't decrease much due to a ban on these applications within thirteen years and that atmospheric emissions have been estimated to be most dependent on the stock in the use phase.

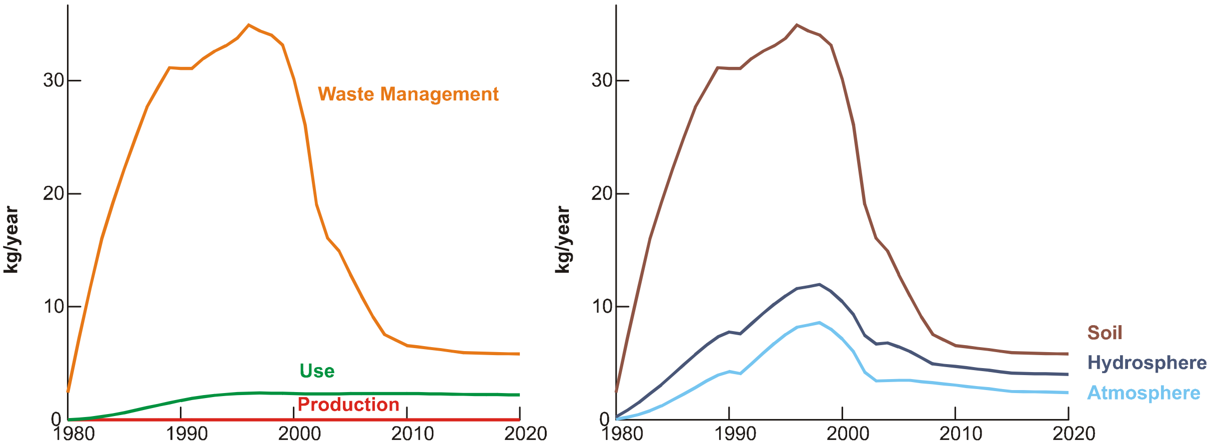
As illustrated in figure 38, in the scenario with a ban in all application areas it has been modelled that emissions to the environment in 2020 are about 3 kg/year, which is about a fifth of the emissions in the reference scenario. Thereof, 2 kg/year are emitted to atmosphere and 0.5 kg/year to both hydrosphere and soil. The emissions from the use phase have been modelled to account for 1 kg/year, which is eight times less than in the reference scenario. About 2 kg/year have been modelled to be emitted from the waste management, which is 60% less than in the reference scenario.

There are indications that the atmospheric emissions of DecaBDE and especially diffuse releases from the use phase have been underestimated based on data available at present (see section 4.2). Therefore, a ban would possibly reduce the emissions to the environment by more of a factor five by 2020.

3.4.2 Influence of an invariant waste management on emissions of DecaBDE

The model results of the "old fashion WM" scenario have been compared with the model results of the real situation in Switzerland (reference scenario) for the DecaBDE in the application area *E&E* only. The results demonstrate that in the "old fashion WM" scenario (without measures taken in the WM since 1980), the total emissions to the environment for the E&E application of DecaBDE would have been more than a factor 3 higher compared to the maximum emissions in reference scenario. As illustrated in figure 39, the maximum emissions in this case would have been at approximately 125 kg/year. The emissions in this scenario are even over one order of magnitude higher as in the realistic case for the present year and would decrease only slightly in the near future.

Emissions Reference Scenario



Emissions “Old Fashion Waste Management”

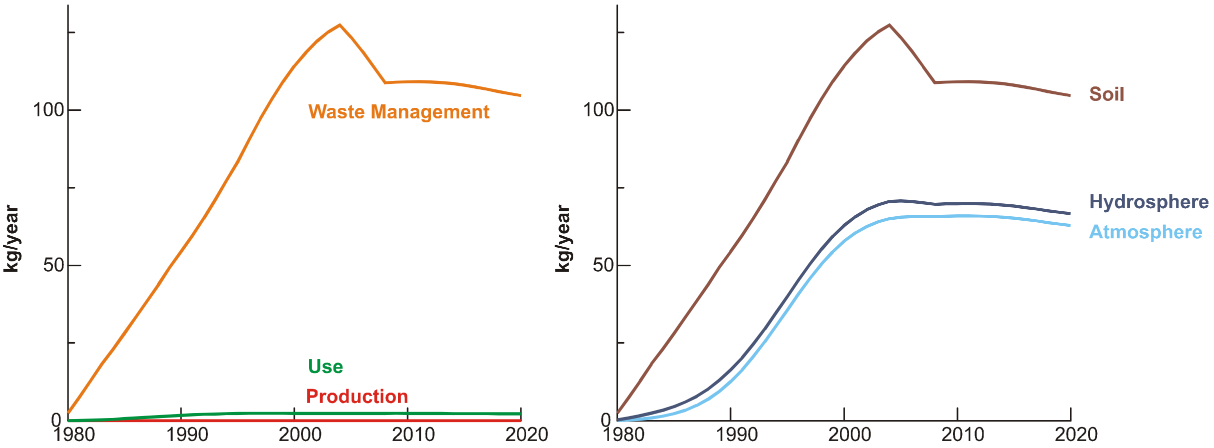


Figure 39 Emissions of DecaBDE in *E&E* to the environment: reference scenario (above) and “old-fashion waste management” scenario (below). Note the different scales.

The three main emissions sources in the “old-fashion waste management” scenario have been modelled as the atmospheric emissions from recycling and incineration and the usage of sewage sludge in the agriculture. The emissions through these pathways would be approximately 40 kg/year (2 kg/year in the reference scenario) from recycling to the atmosphere, 20 kg/year (0.2 kg/year) from incineration to the atmosphere and 40 kg/year (3 kg/year) from sludge to soil for the present year (see section 7.7.2 in the appendix). The emissions from sewerage, landfills and WWTP effluents would also increase, but none of them has been modelled to exceed 3 kg/year. While most of the emissions would be to soil in the beginning, the atmospheric emissions would make up 60% at present (36% to soil, 4% to hydrosphere).

The scenario results allow (a) demonstrating the effectiveness of real measures taken in the waste management of Switzerland regarding the reduction of emissions to the environment and (b) drawing conclusions regarding efficient measures for waste management in other regions of the world. They indicate significant differences for BFR emissions in countries that for example do not have state of the art incineration or recycling technology and have different sewage sludge and WEEE disposal/recycling schemes. Also the size of accumulated stocks as a future potential hazard will be rather different. Illegal combustion and landfill fires as well as production of dioxins and furans have not been taken into account in the model so far. Including these aspects that seem to be of high relevance especially in developing countries, the situation would be even more dramatic (Cuadra, 2005; Sakai et al., 2006a).

3.4.3 Ban on the usage of HBCD in construction materials

The influence of an import stop in 2007 is illustrated for the year 2020 (see figure 40 and figure 41). As the recycling of EPS and the subsequent reuse in the production of these insulation materials have been assumed to continue and to generate an unintended flow from *production* to *construction* and *use* (see section 2.5).

The stock accumulated in buildings and constructions would be about the double without an import stop compared to the situation with an import stop. This stock would further increase in the first case, while there would be a small decrease in the second case. It would last a few decades until this stock would be depleted. As there would be only a diminutive fraction dumped on landfills in the period 2007–2020, there would merely be a small difference in that stock. The atmospheric emissions mainly would be originating from diffuse emission from the stock in buildings and constructions and during production, construction and deconstruction. The emissions from the use phase would still be the largest in the import stop scenario, being about a third compared to the reference scenario. The emission from deconstruction would not be much smaller, while emissions from production and construction would one order of magnitude lower.

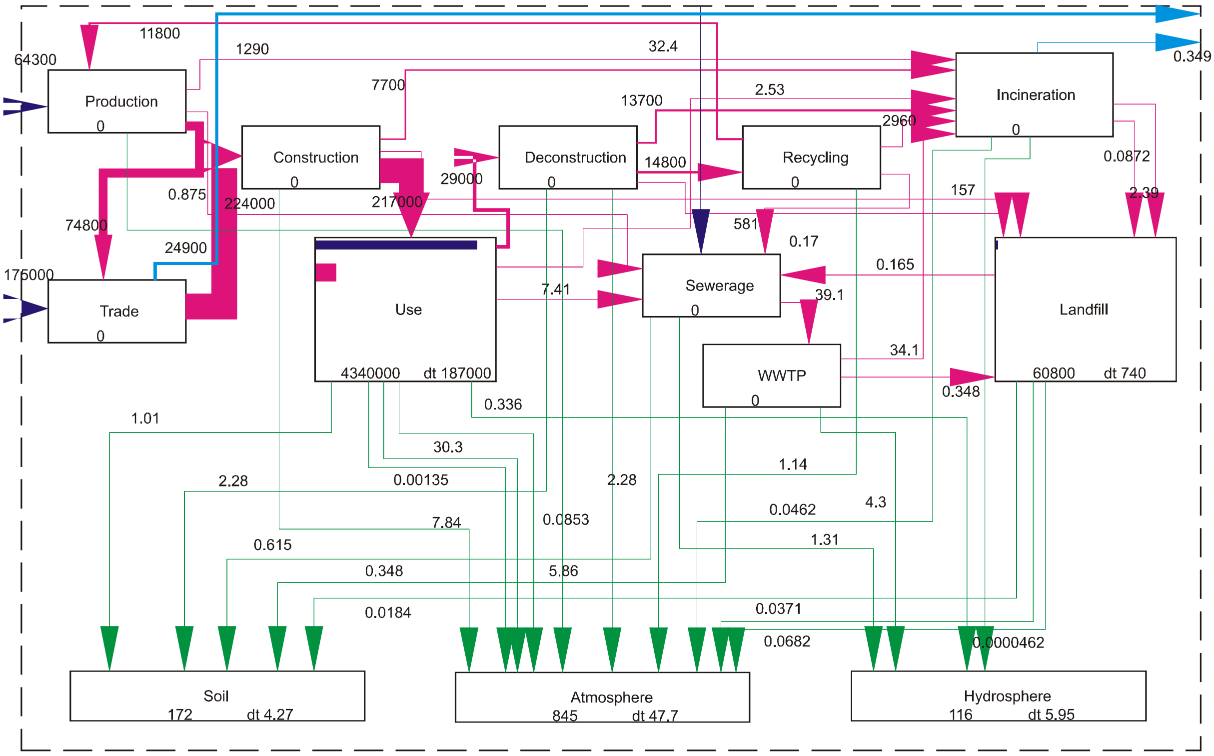


Figure 40 HBCD substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area construction: situation in 2020 with no measures taken. Note that the thickness of the arrows can not be compared among the different diagrams.

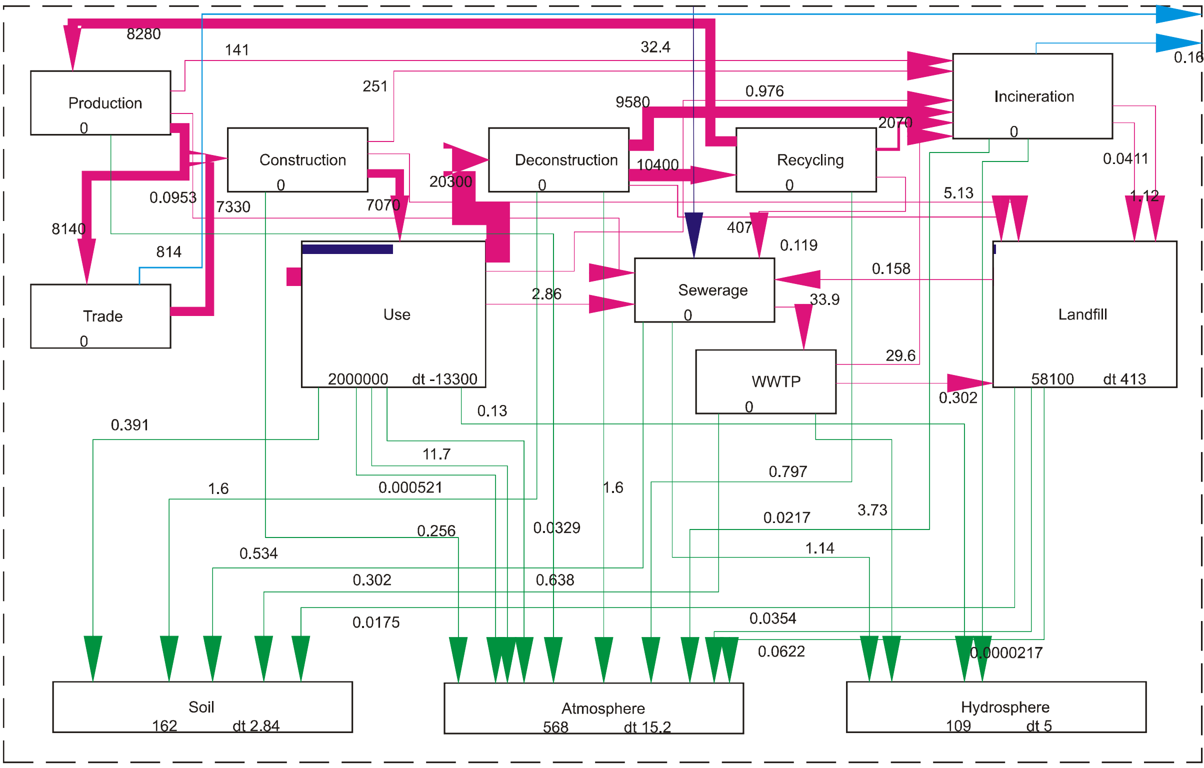


Figure 41 HBCD substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *construction*: situation in the same year with an import stop in 2007. Note that the thickness of the arrows can not be compared among the different diagrams.

As illustrated for the import stop scenario (figure 43), the emissions to the environment for the year 2020 would be about the same than for the late 1990s. Regulative measures are therefore considered to be able to avoid a further increase in emissions and to induce a reduction of one third compared to the situation in the reference scenario by 2020 (see figure 42). However, it would take more than half a century from now on until the emissions would be significantly lower.

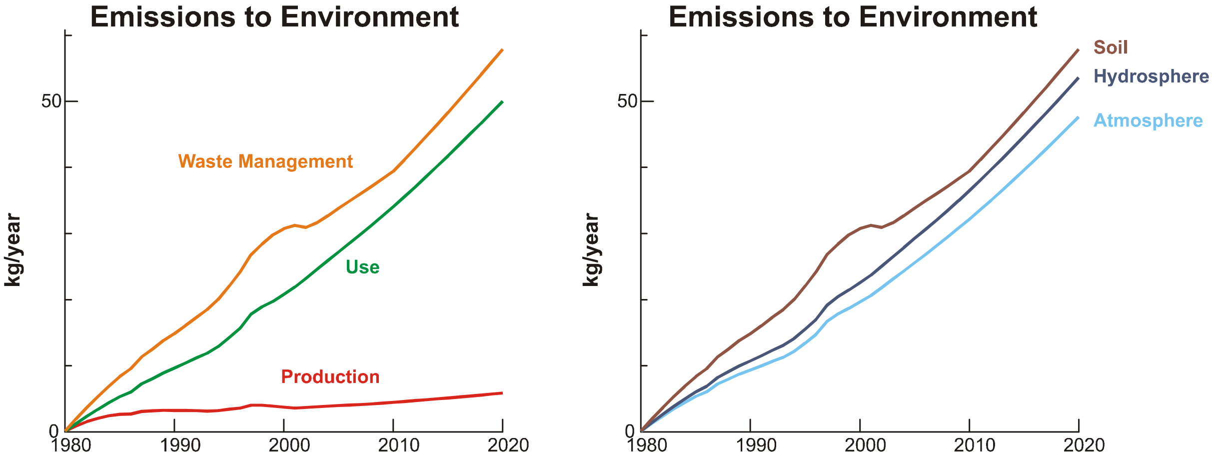


Figure 42 Emissions of HBCD from construction materials to environment split up by source and compartment: reference scenario. Note the different scale to the figure below.

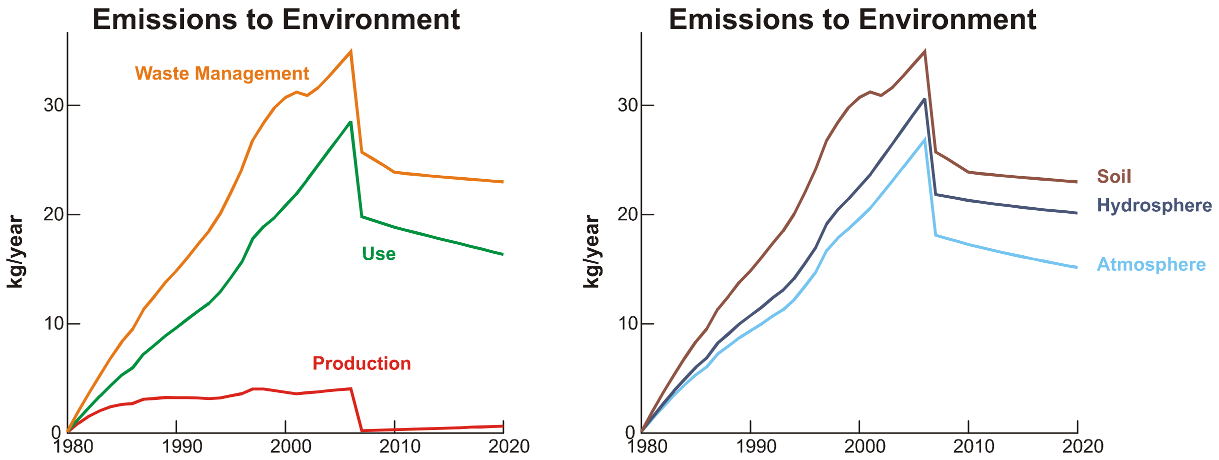


Figure 43 Emissions of HBCD from construction materials to environment split up by source and compartment: scenario import stop in 2007. Note the different scale to the figure above.

There would not be a considerable difference in the stock in buildings and constructions or in emissions to the environment in the period modelled for the import stop scenario, if no reuse of HBCD containing building materials would have been assumed. However, the time until the stock would be entirely depleted would be extended and diffuse emissions would continue longer. For figures of trend of the substance flow to waste management and of the emissions to the different environmental compartments see section 7.7.3 in the appendix.

3.5 Uncertainty of emissions to the environment

The uncertainty of the emissions to atmosphere, hydrosphere and soil has been estimated based on a Monte Carlo simulation for the year 2005. As the domain of uncertainty for the flows and stocks in the anthroposphere has been estimated to be much lower than for the releases to wastewater and the emissions to the environment, the stationary approach has been considered to be passable. The domain of uncertainty has been calculated with 10,000 iterations for the emissions from the different application areas to the three environmental compartments. The uncertainty has also been determined for the aggregated emissions from the different application areas, aggregated emissions to the environmental compartments and for the overall emissions to the environment. Here, the aggregated emissions from the four application areas are discussed for atmosphere, hydrosphere and soil separately.

Figure 44 shows the 90% confidence intervals for the emissions to atmosphere, hydrosphere and soil derived in the Monte Carlo simulation for 2005. The upper boundary of the confidence intervals is about one order of magnitude higher than the lower boundary for all substances and compartments. The largest factor (29) has been derived for the atmospheric emissions of HBCD, while all other factors are in a range of 8–13. The maximum values exceed the upper boundary by just over one order of magnitude for the emissions to hydrosphere and soil. The maximum value for atmospheric emissions of DecaBDE is more than two orders of magnitude higher than the upper boundary of the confidence interval.

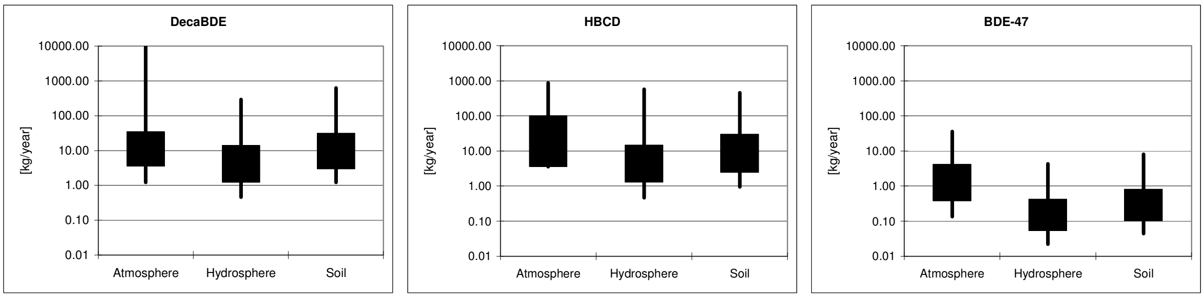


Figure 44 90% confidence intervals (box), minima and maxima emissions derived with the Monte Carlo simulation for the year 2005

The parameters with highest regression sensitivity for the atmospheric emissions of DecaBDE are the emission factors for diffuse releases from the use phase and from recycling processes. For emissions to hydrosphere and soil, the TC from production to sewerage is of highest relevance, especially in the application area *E&E*. Other important parameters for emissions to hydrosphere are diffuse emissions from the process *use* to hydrosphere, the emission factor of sewer overflows as well as the TC of DecaBDE to sewage sludge (negative regression sensitivity). For soil, also the parameters for diffuse emissions from the use phase to soil, the consumption of DecaBDE and the proportion of the sewage sludge used as a fertilizer are of high importance (see section 7.8 in the appendix).

For the atmospheric emissions of HBCD, the diffuse emissions from installed EPS and XPS insulation panels are by far of highest importance (regression sensitivity of 0.86). As this emission factor in the application area *construction* dominates the atmospheric emissions, the atmospheric emissions of HBCD have the largest confidence interval (see figure 44). Also the emission factors from *deconstruction*, *construction* and *production* as well as the consumption of construction materials have a high influence on the atmospheric emissions of HBCD. For application areas other than *construction*, the diffuse emission factors from *transport* and *textile/furniture* are of importance. For the emissions to hydrosphere, the flow from atmospheric deposition to sewerage and the releases from cleaning and mopping in the process *use* have the highest positive regression sensitivity, while it is negative for the TC of HBCD to sewage sludge in WWTP. The emissions from deconstruction processes and the atmospheric deposition that get into sewer are of highest relevance for the emissions of HBCD to soil.

For the atmospheric emissions of BDE-47, diffuse emissions from products in all application areas as well as emissions from the *deconstruction* have the highest regression sensitivity. Due to the lower K_{OW} of BDE-47, landfill leachates are of high relevance of emissions to hydrosphere. Also the input from atmospheric deposition to the sewer system and the TC of BDE-47 into sewage sludge (negative regression sensitivity) are of major importance. Landfill leachates as well as the input flow from atmospheric deposition to wastewater do have also a high regression sensitivity for emissions to soil, but the emissions from deconstruction operation are modelled to be of highest importance.

3.6 Environmental fate results

3.6.1 Atmosphere

Outdoor air concentrations

The outdoor air concentrations have been modelled by multiplying the best guess emissions for the year 2005 and the fractions of the annual emissions residing in the air at steady-state determined in table 30 on page 73. The upper boundary of the 90% confidence intervals as well as the maximum emissions have been multiplied by the high estimate of the fraction and vice versa. The temporal trends of the outdoor air concentration and the atmospheric deposition modelled would change over the years in analogy to the atmospheric emissions. Therefore, these figures are not given in this report.

The concentration calculated that way for DecaBDE is 0.5 pg/m^3 with a lower and upper boundary of 0.1 pg/m^3 and 2.3 pg/m^3 as illustrated in figure 45. The concentrations found in the literature ranged from 0.05 pg/m^3 (half of detection limit) to 74.5 pg/m^3 in rural and remote regions and from 1.5 to 11,500 pg/m^3 in urban areas (Strandberg et al., 2001; ter Schure et al., 2004b; Hoh and Hites, 2005; Chen et al., 2006). The highest concentration was found in the industrial zone of the southern Chinese city Guangzhou (not included in figure 45) and the highest concentration outside China was 878 pg/m^3 found in Chicago. Generally, the concentration ranged by one to three orders of magnitude within the same sampling site, showing the dynamics of emissions and atmospheric processes. The modelled Swiss DecaBDE concentrations are somewhat lower than levels in remote/rural areas and clearly lower than field data from urban environments.

The atmospheric concentration of HBCD derived from the model is 30 pg/m^3 (range 3.6–104 pg/m^3). Outdoor air concentrations found in Sweden were <1–280 pg/m^3 at background sites and 76 pg/m^3 and 610 pg/m^3 in Stockholm (Sternbeck et al., 2001; Bergander et al., 1995 cited in Remberger et al., 2004). A few measurements were also carried out in the vicinity of potential emission sources: on a landfill for construction and demolition waste (13 pg/m^3 and 180 pg/m^3), 350 m south of a textile industry area (19 pg/m^3 and 740 pg/m^3) and 10 m away from the exhaust of the ventilation system of a facility producing flame-retarded XPS (1070 ng/m^3) (Remberger et al., 2004). While the levels on a landfill and near textile industry were comparable to urban levels, the concentration next to the exhaust was three orders of magnitude higher. Concentrations found in remote, rural and urban in the USA were in a range of <0.07–11 ng/m^3. The somewhat lower concentrations compared to Sweden are not surprising, as the consumption of HBCD is lower in the States than in Europe. The concentrations modelled for Switzerland are somewhat lower than levels in Stockholm, on the landfills and in the vicinity of the textile industry area, but comparable or slightly higher than background levels in Sweden.

The outdoor air concentration calculated for BDE-47 is 0.22 pg/m^3 (range 0.04–1.7 pg/m^3) as shown in figure 45. Field data on remote concentrations range between 0.2 pg/m^3 and 5.6 pg/m^3 at sites in England, Ireland, Norway and Sweden (Jaward et al., 2004b; Lee et al., 2004; ter Schure et al., 2004b). Concentrations in rural and urban environments across Europe were found to range from 0.72 pg/m^3 to 80 pg/m^3 (Jaward et al., 2004a; Lee et al., 2004; Harrad and Hunter, 2006). It was found that United Kingdom had elevated levels, what is not surprising, because it has a history of PBDE production and has been a major user of PBDE-based flame retardants, owing to particularly stringent fire regulations. It was concluded that United Kingdom is clearly a regional source area for PBDEs to the European atmosphere (Jaward et al., 2004a). Levels in remote, rural and urban regions of the USA and Canada ranged between <detection limit and 48 pg/m^3 (Strandberg et al., 2001; Wilford et al., 2004;

Hoh and Hites, 2005; Harner et al., 2006). Air concentrations of <0.13–78 pg/m^3 were detected at sites in the Asian countries China, Japan, Singapore and South Korea (Jaward et al., 2005). Like for DecaBDE, also the highest concentration (2156 pg/m^3, mean of eight samples at one location) was found in the industrial zone of Guangzhou (Chen et al., 2006). This measurement is not included in figure 45, because the situation in a Chinese industrial zone is not though to be comparable to Switzerland and in contrast to DecaBDE and HBCD there are more field data from European or North American locations. The mean of the BDE-47 concentrations modelled for Switzerland are comparable to low concentrations found in remote/rural and urban areas. The upper boundary of the estimate seems to agree better with the field data.

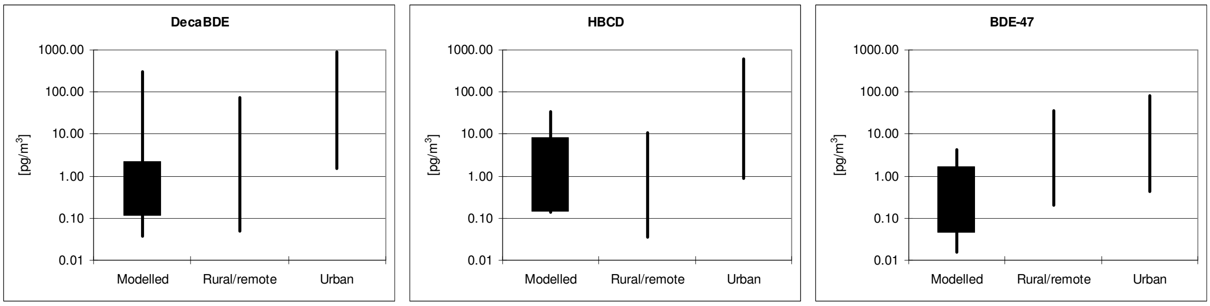


Figure 45 Modelled outdoor air concentrations of DecaBDE, HBCD and BDE-47 for the year 2005 and comparison to field data

Atmospheric deposition

As shown in figure 46, the atmospheric deposition for DecaBDE has been modelled as 0.23 µg/m^2/year (0.07–0.85 µg/m^2/year). The experimentally determined atmospheric deposition was 1.7 µg/m^2/year at the shore of Lake Thun, a pre-alpine lake in the center of Switzerland in summer 2006 (Bogdal et al., 2007). In Dübendorf, a Swiss municipality close to Zurich, the deposition was 3.6 µg/m^2/year in February 2006 (C. Bogdal, personal communication) measured with the same method. The mean deposition into the Baltic Sea was estimated based on measurements as 0.65 µg/m^2/year (ter Schure et al., 2004b). In Sweden bulk deposition rates were determined close to a MSWIP, in an urban and a rural reference site. The values found for DecaBDE were 23.3 µg/m^2/year, 5.4 µg/m^2/year and 1.6 µg/m^2/year (ter Schure et al., 2004a). The atmospheric deposition in the urban and the rural reference cite are comparable to the deposition rates found in Switzerland. Deposital fluxes of DecaBDE and BDE-47 to lakes were estimated based on concentrations in dated sediment cores taken from lakes along a latitudinal transect in North America. Surface fluxes of DecaBDE ranged from <0.003 to 0.094 µg/m^2/year in the three Arctic lakes, from 0.043 to 1.145 µg/m^2/year in the four mid-latitude lakes, and 67.8 µg/m^2/year in Lake Ontario (Breivik et al., 2006). In a similar study, surface fluxes of ~1–90 µg/m^2/year were found for the five Great Lakes (Li et al., 2006). While atmospheric deposition is the only relevant source for the levels found in the Artic lakes, input from WWTP and rivers has also been estimated to be of importance for the input to the Great Lakes. The modelled atmospheric deposition of DecaBDE is somewhat lower than the field data in Switzerland and compared to data from sites that might be comparable to Switzerland. The real emissions of DecaBDE tend to be at the upper boundary of the emission estimate, even though also foreign sources might be of importance. As DecaBDE has a long-range transport potential, even emissions from United Kingdom that has been shown to be a source area of lower brominated PBDE might contribute to the levels found in Switzerland. Less stringent regulatory measures or other recycling techniques as well as illegal burning of municipal and hazardous waste in neighbouring countries such as recently reported from Italy (Schweizer Fernsehen, 2007) could also have an effect on concentrations in the Swiss environment.

For HBCD, the atmospheric deposition modelled for the year 2005 is 0.7 µg/m^2/year (0.07–1.5 µg/m^2/year). As there are no field data available for Switzerland, literature data from other countries has been used for comparison purposes. Remberger et al. (2004) measured the bulk atmospheric deposition at three sites in Sweden and Finland (two measurements at each site). The deposition rates found were 0.007/0.58 µg/m^2/year at the Swedish west coast, 1.9/4.7 µg/m^2/year in Northern Finland and 2.0/134 µg/m^2/year in Stockholm. In a study carried out in the Great Lakes basin, wet-only deposition rates between 0.13 and 3.64 µg/m^2/year were found (Backus et al., 2005). The modelled value is comparable with the atmospheric deposition at Swedish and Finnish background locations and with the measurements carried out in the USA. As the HBCD consumption in North America is lower than in Europe, this comparison might not be valid. While to lower value determined in Stockholm is comparable with the modelled deposition rate, the higher is not. Based on these data there is an indication that the atmospheric emissions of HBCD might have been underestimated, but there are unquestionably not enough field data to draw a final conclusion.

The atmospheric deposition for BDE-47 has been modelled in the SFA as 0.02 µg/m^2/year (0.01–0.06 µg/m^2/year). The deposition rate determined at Dübendorf is 0.3 µg/m^2/year in February 2006 (C. Bogdal, personal communication). The bulk deposition rates determined in the vicinity of a MSWIP, in an urban and a rural reference site in Sweden are 3.0 µg/m^2/year, 1.1 µg/m^2/year and 0.1 µg/m^2/year (ter Schure et al., 2004a). For the Baltic Sea a deposition rate of about 0.07 µg/m^2/year (read from the figure) was determined (ter Schure et al., 2004b). Deposital fluxes of DecaBDE and BDE-47 to lakes were estimated based on concentrations in dated sediment cores taken from lakes along a latitudinal transect in North America. Surface fluxes of BDE-47 to North American lakes ranged from <0.01 to 0.15 µg/m^2/year in the three Arctic lakes and the four mid-latitude lakes and was 0.4 µg/m^2/year in Lake Ontario (Breivik et al., 2006). The modelled deposition rate seems to be low compared with the available field data in Switzerland and abroad, which is in line with the outdoor air concentrations. Like for DecaBDE, foreign sources might possibly contribute to the levels in Switzerland. The United Kingdom that has been shown to be a regional source area of lower brominated PBDEs (Jaward et al., 2004a) is within the characteristic travel distance estimated with TaPL3, ELPOS and CoZMo-POP model as well as empirical data (Breivik et al., 2006) or in the EMEP/MSC-E project (Vulykh et al., 2006). Based backwards trajectories for other substances emissions of PentaBDE from United Kingdom are likely to reach Switzerland during the appropriate meteorological conditions. Debromination of DecaBDE in the atmosphere might possibly contribute to the concentrations of BDE-47. Farrar et al. (2004) illustrate the potential for diffusive combustion processes to impact ambient levels of PBDEs: The air concentrations of BDE-47, BDE-99 and the sum of PBDEs (without DecaBDE) were elevated by a factor of 4, 7 and 25 during the Bonfire Event, a major anthropogenic combustion event in the United Kingdom. Atmospheric mission factors from open fires were also studied with a simulated landfill fire (Hirai et al., 2005). The emission factor of TetraBDE (expressed as g of TetraBDE emitted per g *DecaBDE* burnt) determined was 1.2E–03 (Sakai et al., 2006a). About 0.04% of the overall stock of DecaBDE in the use phase (i.e. 920 kg in Switzerland at present) has been estimated to be lost by fires (see section 2.4.2.5.4). Hence, the atmospheric emission of TetraBDE estimated from these numbers is about 1 kg/year. For OctaBDE, where an emission factor of 7.4E–02 was determined, the emissions from fires would amount to 66 kg/year.

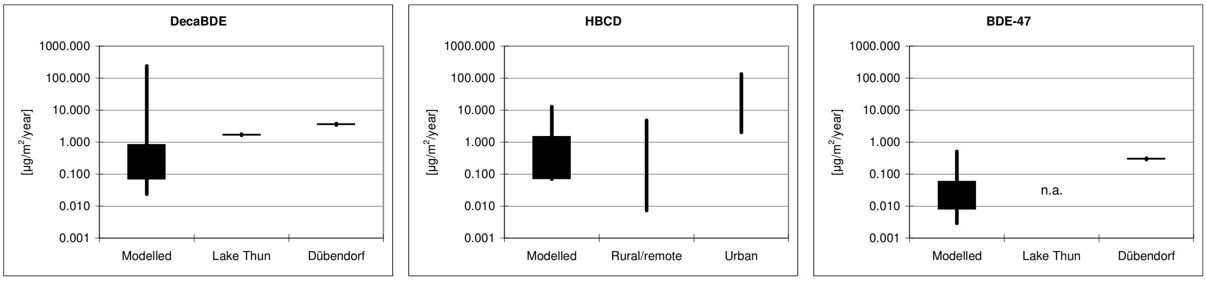


Figure 46 Modelled atmospheric deposition of DecaBDE, HBCD and BDE-47 for the year 2005 and comparison with field data

3.6.2 Hydrosphere

Sediment

The concentration of DecaBDE in freshwater sediments in Switzerland for the year 2005 has been modelled based on the confidence interval of the emissions (section 3.5) and the environmental fate model (section 2.6). The PEC derived is 0.32 ng/g d.w. with a lower and upper boundary of 0.09 ng/g d.w. and 1.1 ng/g d.w. as illustrated in figure 47. The PEC estimated in the EU risk assessment (continental scenario) is 20 ng/g w.w., which is signifycantly higher than the estimates for Switzerland (ECB, 2004). Levels found in the EU monitoring programme ranged between <0.25 ng/g d.w. and 13.7 ng/g d.w. at the sites in the United Kingdom, Ireland, Germany and France. At the western scheldt in Belgium/Netherland mean concentrations of 1300 ng/g d.w. were found (ECB, 2007a). There are two single measurements in two Swiss lakes available to date. The concentrations found in the dated layers of the sediment core was taken in Lake Greifen at a depth of 31 m are increasing from 1.1 ng/g d.w. by the year 1974 to 7.2 ng/g d.w. by 2001 (Kohler, 2005). The levels in a sediment core of Lake Thun increased from 0.08 ng/g d.w. by 1976 to 0.85 ng/g d.w 2001 (Bogdal et al., 2006). The difference in the concentration between these two lakes is believed to be due to their location and their catchment area. While Lake Thun is situated in a pre-alpine, low populated region, Lake Greifen is a small, shallow lake in the highly populated greater Zurich area. Nine WWTPs discharge into Lake Greifen. Therefore, concentrations of anthropogenic substances might possibly be lower than the Swiss average in Lake Thun and higher for the Lake Greifen. The concentration trend modelled is comparable to the field data from Lake Thun but significantly lower than the concentrations found in Lake Greifen. While increasing concentrations were found in the sediment of both lakes, a decrease has been modelled after the mid-nineties. However, when a half-life of 10 years instead of 200 years is assumed, the modelled concentration trend is increasing over the whole period. As there is a large uncertainty for the half-life of DecaBDE (see section 2.6.2), it is unclear whether the input steadily increased until today or the increase in the concentration of sediment cores is mainly caused by degradation. The best guess concentration modelled for sediments seems to be too low compared to the two measurements in Switzerland. As the atmospheric emissions and the subsequent atmospheric deposition might have been underestimated based on available literature data, this missing input might be responsible for the difference found in sediment cores. However, additional measurements to the two single sediment cores from Lake Thun and Lake Greifen would help to examine the evidence of this difference.

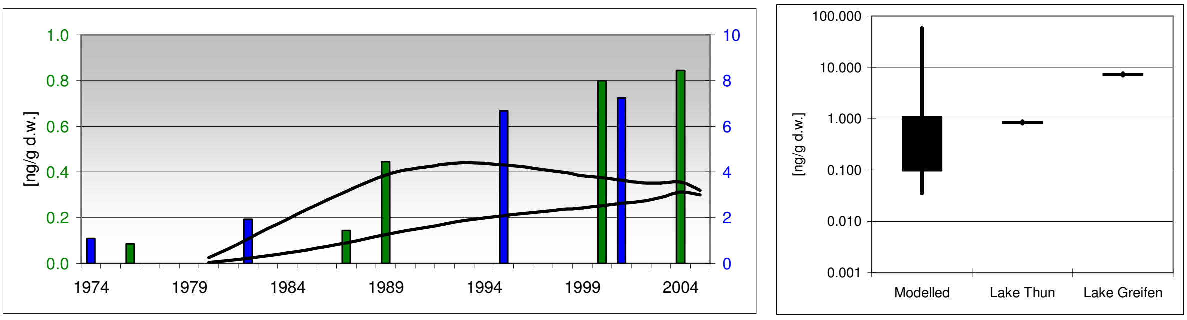


Figure 47 (a) Modelled DecaBDE concentration in sediment layers from different years: half-life of 200 years (upper line), half-life of 10 years (lower line). Concentrations in Lake Thun (green bars) and Lake Greifen (blue bars). Note the different scale on the right hand side for Lake Greifen. (b) Modelled concentration of DecaBDE for the year 2005 and comparison with field data.

The modelled concentration of HBCD in freshwater sediments in Switzerland for the year 2005 is 0.23 ng/g d.w. (boundaries 0.06 ng/g d.w. and 0.70 ng/g d.w.). The trend modelled is increasing over the whole period, irrespective of the choice of a half-life of 20 years or the assumption of no degradation. PEC calculated in the EU risk assessment is 1.4 ng/g d.w. in the continental scenario, which is the double of the upper bound modelled for Switzerland (ECB, 2007b). The HBCD concentration was 0.51 ng/g in 1974, 1.27 ng/g d.w. in 1989, 1.76 ng/g d.w. in 1995 and 2.54 ng/g d.w. in 2001 in a sediment sample taken in Lake Greifen (Kohler et al., 2006). Unpublished measurements in Lake Thun showed a similar trend (see figure 48), but the concentrations were lower by approximately a factor of five (Bogdal et al., 2006). The upper boundary of the estimate is comparable to the levels found in Lake Thun but somewhat lower than the concentrations in the sediment core from Lake Greifen.

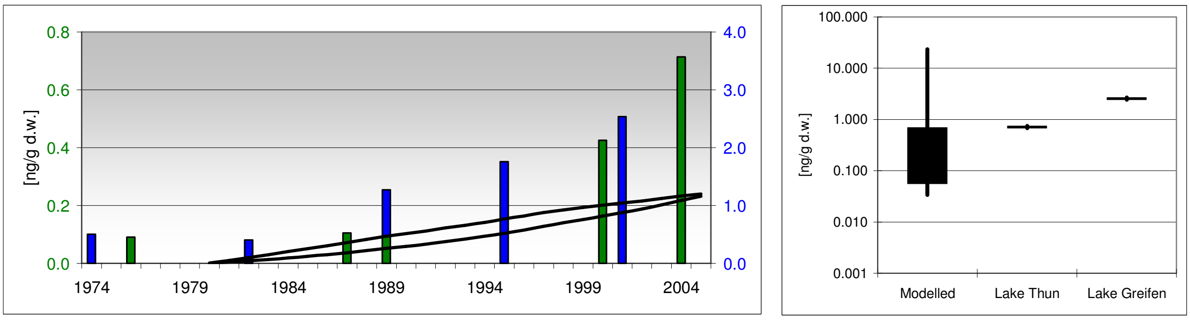


Figure 48 (a) Modelled HBCD concentration in sediment layers from different years: no degradation (upper line), half-life of 20 years (lower line). Concentrations in Lake Thun (green bars) and Lake Greifen (blue bars). Note the different scale on the right hand side for Lake Greifen. (b) Modelled concentration of HBCD for the year 2005 and comparison with field data.

The concentration of BDE-47 in freshwater sediments in Switzerland for the year 2005 has been modelled as 0.012 ng/g d.w. (boundaries 0.004 ng/g d.w. and 0.032 ng/g d.w). The PEC estimated in the EU risk assessment (continental scenario) is 32 ng/g w.w., which is significantly higher than the estimates for Switzerland (ECB, 2000). The BDE-47 concentration in sediment sample from Lake Greifen was 0.21 ng/g d.w. in 1974, 0.53 ng/g d.w. in 1989, 0.64 ng/g d.w. in 1995 and 0.74 ng/g d.w. in 2001 (Kohler, 2005). Unpublished measurements in a sediment core from Lake Thun demonstrated a similar trend (see figure 49), but the concentrations were lower by approximately a factor of five, which is the same difference as for HBCD (Bogdal et al., 2006). The modelled BDE-47 concentration is between one and two orders of magnitude lower than the measurements in the two top layers sediments from the two Swiss lakes. It has been concluded that either the emissions of BDE-47 has been underestimated or there is a significant contribution to the concentrations in sediment from debromination of higher brominated PBDEs in environment or due to fires or from foreign sources.

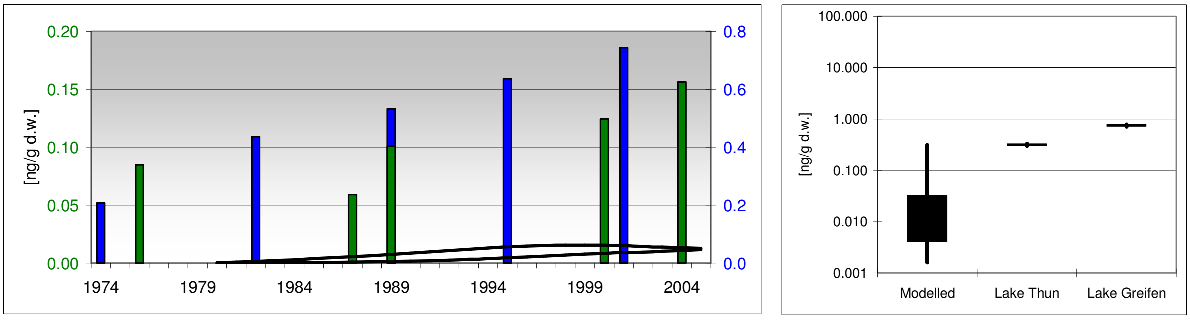


Figure 49 (a) Modelled BDE-47 concentration in sediment layers from different years: no degradation of BDE-47 (upper line), half-life of 8.6 years (lower line). Concentrations in Lake Thun (green bars) and Lake Greifen (blue bars). Note the different scale on the right hand side for Lake Greifen. (b) Modelled concentration of BDE-47 for the year 2005 and comparison with field data.

Water and aquatic organisms

The concentrations of dissolved DecaBDE, HBCD and BDE-47 in the water have been estimated with Safe-Pro for 2005 based on fugacity as 0.002 pg/L, 37 pg/L and 0.25 pg/L, respectively. As a result of the high octanol-water partition coefficient, the concentration of DecaBDE dissolved in surface water is extremely low. The concentrations of HBCD and BDE-47 have been estimated to be higher due to the lower K_{OW}. The PEC of HBCD calculated in the EU risk assessment (continental scenario) are 500 pg/L that is one order of magnitude higher than the estimate for Switzerland (ECB, 2007b). The difference between these modelled concentrations is mainly due to the fact, that a lower per capita industrial consumption for textile back-coating has been assumed for Switzerland (see section 3.3), which results in lower emissions to water and wastewater. Dissolved water concentrations of DecaBDE and tri–hepta-BDE Lake Thun were detected as <0.1 pg/L and 25 pg/L (Bogdal et al., 2007). BDE-47 made up a large fraction in tri–hepta-BDE. The dissolved phase water concentration of BDE-47 in Lake Michigan was 10±1.4 pg/L, while concentrations of 1.0±0.8 pg/L, 2.0±1.2 pg/L and 2.7±1.6 pg/L were estimated for the Lake Superior, Huron and Ontario (Streets et al., 2006). These estimates were obtained by setting congener-specific bioaccumulation factors for Lake Michigan equal to those for the other Great Lakes, and then rearranging to solve for the PBDE dissolved phase concentration of the other lakes. The concentration of BDE-47 in the dissolved phase modelled is up to two orders of magnitude lower than measured in Lake Thun and in the USA. The reasons for that difference might be the same as for the differences between the PEC and the concentrations found in sediments. DecaBDE and HBCD could not be compared to environmental levels, because the concentration of DecaBDE was below the detection limit in Lake Thun and no comparable measurements of HBCD were available.

The concentrations in aquatic organisms estimated with Safe-Pro for the year 2005 are 0.9 ng/g w.w. for DecaBDE, 0.7 ng/g w.w. for HBCD and 0.04 ng/g w.w. for BDE-47. These estimates are not valid for organisms at higher trophic levels such as fish, as biomagnification has not been considered. Some field data from Switzerland are presented anyhow. BDE-47 levels in whitefish samples taken from eight Swiss lakes ranged between 1.0 and 4.4 ng/g w.w. (Zennegg et al., 2003). Recently, Bogdal et al. (2007) measured the concentrations of

PBDEs in fat from muscle tissue of whitefish in Lake Thun. The levels of DecaBDE and tri–hepta-BDE found were 7.2 ng/g l.w. (range 3–10 ng/g l.w.) and 123 ng/g l.w. (range 110–140 ng/g l.w.), respectively.

3.6.3 Soil

The concentration of DecaBDE in soil in Switzerland for the year 2005 has been modelled based on the confidence interval of the emissions (section 3.5) and the environmental fate model (section 2.6). The PEC derived is 56 pg/g d.w. with a lower and upper boundary of 15 pg/g d.w. and 196 pg/g d.w. If no degradation is assumed, a concentration of 135 pg/g d.w. (range 37–470 pg/g d.w.) is modelled (see figure 50). Levels in agricultural soil, where no sewage sludge had been applied to, were found to range between 15 and 750 pg/g d.w. in Sweden. The concentrations in soils that had been fertilized with sewage sludge were 28–2,200,00 pg/g d.w. (Matscheko et al., 2002; Sellström et al., 2005). Humus layers in remote alpine forests have recently been investigated in the EU project MONARPOP. The DecaBDE concentration found in these soils were between 674 pg/g d.w. (5% quantile) and 7483 pg/g d.w. (90% quantile) (Knoth et al., 2007). PBDE concentrations in composts and digestates in Switzerland were determined (Brändli, 2006). The dominating compound was DecaBDE with a mean of 930 pg/g d.w. (range 600–31,000 pg/g d.w.). Based on the annual organic waste produced (770,000 tons/year in 2004), its organic and water content as well as the mass loss annual flows of 1.3 kg/year DecaBDE in Swiss composts and digestates have been estimated. The concentrations in composts and digestates can not be compared directly with soils. However, the concentrations are in a similar range as the levels in remote soils. The field data are one order of magnitude or more higher than the modelled concentrations, except for the soil in Sweden that was not fertilized with sewage sludge. The fact that only the top 5 cm (mean value) were investigated compared to the 10 cm in the model is not of relevance. Also the forest filter effect described by Breivik et al. (2006) and an elevated amount of precipitation in the alpine region can not explain the difference. Taking into account that the largest fraction of the emissions has been estimated to get to soil by the usage of sewage sludge as a fertilizer in the agriculture and the remote soil was never fertilized, the hypothesis of an underestimation of atmospheric emissions or a large input from foreign sources has been corroborated.

The HBCD concentration in soil has been modelled as 3.4 pg/g d.w. (boundaries 1.4 pg/g d.w. and 14 pg/g d.w.). When neglecting any degradation the concentration would be 48 pg/g d.w. (range 20–203 pg/g d.w.). The PEC calculated in the EU risk assessment (continental scenario) are 3000 pg/g d.w. for agricultural soil and 93 pg/g d.w. for natural and industrial soil (ECB, 2007b). As stated in section 3.3, the difference between the modelled concentrations for Switzerland and the EU is mainly due to the fact, that a lower per capita industrial consumption for textile back-coating has been assumed for Switzerland, which results in lower emissions to water and wastewater. There are only field data on soil concentration in sites that are in close vicinity of a potential emission source. In soil near XPS production plant, a formulator/compounder and a textile backcoater plant, concentrations in a range of 140,000–90,000,000 pg/g d.w. were found (Sternbeck et al., 2001; ECB, 2007b). Levels in soil 0.5 m under railway embankments ranged between 10,000 and 1,600,000 pg/g d.w. (Jansson, 2004 cited in ECB, 2007b). These data can obviously not be compared to the modelled concentrations. However, the concentration of 105,000 pg/g d.w. (range 17,000–372,000 pg/g d.w.) in composts and digestates (Brändli, 2006) as well as the somewhat higher field data for sediments compared to the model estimates give some evidence that emissions of HBCD might have been underestimated. More field data would be needed in order to examine the emission estimates.

The concentration of BDE-47 in soil has been modelled as 0.18 pg/g d.w. (boundaries 0.07 pg/g d.w. and 0.44 pg/g d.w.). If no degradation is assumed, a concentration of 3.9 pg/g d.w. (range 0.63–9.5 pg/g d.w.) is modelled. Concentrations in remote alpine soils were found to be between 97 pg/g d.w. (5% quantile) and 504 pg/g d.w. (90% quantile) (Knoth et al., 2007). In the United Kingdom, concentrations were found to be in a range of 7–1400 pg/g d.w. (Hassanin et al., 2004; Harrad and Hunter, 2006). In Spain, no detectable levels of PBDEs were found in soils from agricultural fields amended with sludge, while soil samples from an industrial area showed a mean BDE-47 level of 4600 pg/g d.w. (Sanchez-Brunete et al., 2006). In Sweden, concentrations in agricultural soil, where no sewage sludge had been applied to, were found to range between 6.8 and 31 pg/g d.w. The levels in soils that had been fertilized with sewage sludge were 52–450,000 pg/g d.w. (Matscheko et al., 2002; Sellström et al., 2005). The median concentration of BDE-47 was 800 pg/g d.w. (range 70–1500 pg/g d.w.) in Swiss composts and digestates (Brändli, 2006). Also in this environmental compartment the modelled BDE-47 concentration is lower than field data. Atmospheric deposition is probably the only relevance source for the concentrations found in remote soils. In contrast to DecaBDE, revolatilisation of the congeners with less than seven bromine from humus to air could be of importance (Knoth et al., 2007). Therefore, equilibrium between industrial or agricultural and remote soils is possible to a certain extent. However, underestimated atmospheric emissions are believed to be the main reason of the difference.

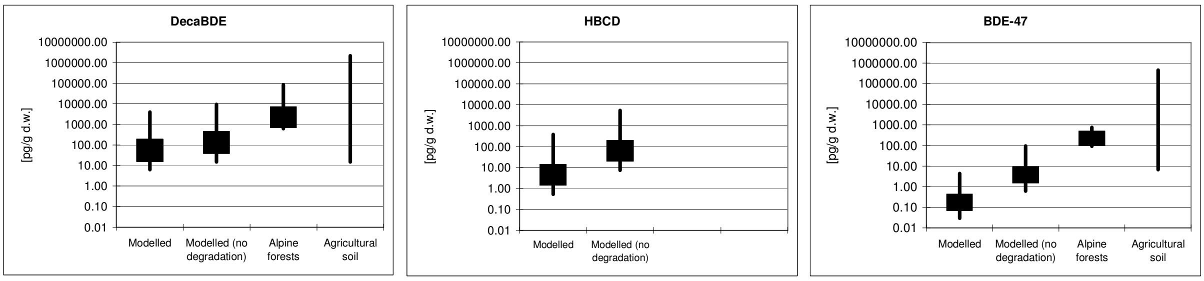


Figure 50 Modelled concentrations of DecaBDE, HBCD and BDE-47 in soil for the year 2005 and comparison with field data, where available

4 Summary and conclusions

4.1 Dynamic behaviour of the substance stocks, flows and emissions

4.1.1 Improvement and update of the SFA of the previous study

This project could improve the knowledge on consumption and emissions compared to the study by Morf et al., (2002). There are still relatively limited and sometimes highly variable field data available for emissions, which results in high domains of uncertainty for certain flows. Nevertheless, a more comprehensive time-dependent view of the anthropogenic metabolism could be elaborated and the most relevant emission sources could be identified. The fact that the process *use* has been modelled in more detail and the processes *construction* and *deconstruction* have been added facilitated to distinguish between different emission sources.

For DecaBDE, new investigations demonstrate that the consumption in the late nineties and as a consequence the stocks in the Swiss anthroposphere might have been estimated too high by Morf et al. (2002) according to data available at that time. Due to this fact as well as according to new data available on field measurements of emission factors during use and waste management, the total emissions to environment modelled in this study are about forty times lower than expected in Morf et al. Considering the uncertainty of the data available for the study in 2002, the differences to the new results are reasonable and acceptable. In opposite to the previous study, the diffuse emissions from the use phase have been estimated much smaller, because a few emission experiments have come available in the meantime. The measured values are significantly below worst case estimates used in Morf et al., 2002. Most relevant DecaBDE emission sources to soil and hydrosphere at the end of nineties are sewage sludge that is used as fertilizer in the agriculture, WWTP effluents as well as sewerage leakages and overflows. Most relevant atmospheric emissions originate from e-waste and automobile recycling and from goods during the use phase. In Morf et al., 2002 detailed information on the relevance was not yet available.

For HBCD, which was not included in Morf et al. (2002), the consumption and the resulting stocks in the anthroposphere, the substance flows and the emissions to the environment have been modelled. The EPS and XPS insulation panels have been identified as the most important application area and atmospheric emissions from these materials during the use phase (i.e. installed in buildings and constructions) have been estimated to be of largest relevance.

Based on new data, especially from the study carried out by Prevedouros et al. (2004a) and the data supplied by BSEF and CEFIC, the consumption estimate of PentaBDE in Switzerland has been lowered compared to the previous study. Particularly the consumption in the early 1990s might have been overestimated before. In consequence of this, also the modelled stocks and flows are lower compared to the previous study. The results from experiments on emissions ranged widely. Some of them showed an almost quantitative loss of BDE-47 from PUR foam. As such high emission factors would lead to much higher indoor air levels than detected, these experiments have been considered not to be applicable for real situations. However, the atmospheric emission factors chosen in the present study might be too low. Because of the lower consumption and stocks of PentaBDE modelled compared to be previous study and the lower emission factors, the atmospheric emissions have been estimated significantly lower than before.

The hypothesis made in the proposal *"Diffuse emissions from use and disposal activities are responsible for the occurrence of BFR in the environment"* could be corroborated with the model for HBCD. Diffuse emissions from insulation panels during the use phase including construction and deconstruction processes have been estimated to amount to the largest proportion of emissions. To a lower extent, the same is also true for PentaBDE, especially as these emissions might have been underestimated. The situation is somewhat less clear for DecaBDE, as large proportion of amount in the environment has been estimated to originate from sewage sludge. However, there is some evidence that diffuse emissions from the use phase have been underestimated.

4.1.2 Temporal trends of substance flows, stocks and emissions

High amounts of both DecaBDE and HBCD are stored in the use phase at present. The stock of PentaBDE is significantly lower. While the stock of DecaBDE is virtually stable and the stock of PentaBDE is decreasing, the amounts of HBCD stored are still increasing rapidly. Especially for HBCD, where the largest proportion is stored in construction materials that have a long residence time, big amounts would remain in the anthroposphere, even if a ban would be declared in the near future. As the use of DecaBDE in construction materials was reduced some years ago already and the largest part of the stock in the anthroposphere is made up by E&E, DecaBDE is not expected to remain as long in the system as HBCD. These rather huge and in the case of HBCD still rapidly increasing stocks are potential sources for actual and future emissions to the environment and have to be managed properly. The stock in the waste management (i.e. on landfills) is much lower and is not expected to generate high emissions or releases to wastewater for DecaBDE and HBCD. As long as no landfill fires will occur, the risk from landfills is expected to be rather small. For PentaBDE, the model demonstrates the potential of significance for releases from landfills to wastewater and for emissions.

The wastewater pathway is important for emissions to hydrosphere and soil, especially at the time, when large proportions of sewage sludge were used as fertilizer in agriculture. For HBCD, diffuse emissions from construction materials to atmosphere and emissions during construction and deconstruction processes are of high relevance. On the other hand, emissions from MSWIPs or landfills (except for PentaBDE, see above) are of minor relevance. Hence, in terms of DecaBDE and HBCD emissions from MSWIPs and landfills do not seem to be a major problem these days.

Emissions of DecaBDE and PentaBDE are expected to further decline or to be steady at maximum in future. On the other hand, HBCD emissions will continue to increase and are not expected to decline rapidly, if the substance would be banned (long residence time of insulation panels in the anthroposphere). After a potential ban of HBCD, if the on-going EU risk assessment would conclude that there is a risk, diffuse emissions from the use phase and deconstruction processes would continue over decades. For DecaBDE and PentaBDE, regulatory measures (e.g. incineration of sludge) and technical innovation (e.g. off-gas filter) led to a significant reduction of emissions during the past decade. Normally, a shift in emissions from production to use and finally to waste management over time is expected. As regulatory and technical measures (e.g. off-gas filters, incineration of sewage sludge), led to a relative reduction of emissions from waste management, this effect is not expected to happen in Switzerland for the BFRs under study. Especially for HBCD, emissions from the use phase are most important, which is not likely to change in future, even not in the first decades after a potential ban.

A large amount of BFRs leaves Switzerland via export to abroad. In 2007, about 200 tons/year of DecaBDE, 50 tons/year of HBCD and 2.5 tons/year of PentaBDE have been estimated. The largest part of that originates from trade (new goods) except for PentaBDE, but also the substance flows in the export of used cars to eastern European countries or to Africa and the export of e-waste are of relevance (8% of DecaBDE, 17% of HBCD, 50% of PentaBDE, sum of exports). A higher amount originates from waste management, where a large proportion of automobile shredder residues is incinerated abroad and metals from metal fractions have been assumed to be recovered in foreign steel works (23% of DecaBDE, 16% of HBCD, 50% of PentaBDE, sum of exports). These exported material flows need a close look at in order to prevent "exported emissions" to the environment due to non-adequate disposal abroad.

4.2 Environmental fate

The investigation on the environmental fate using a fugacity model shows that due to their physical properties, DecaBDE, HBCD and BDE-47 (main component of the PentaBDE technical mixture) are most likely to partition to organic phases. While biota might be an important intermediate stock, final sinks are soils and sediments. The best estimate outdoor air concentrations, atmospheric deposition rates as well as concentrations in sediment and soil are all predominantly lower compared to the available field data. It is therefore concluded that the atmospheric emissions might have been underestimated based on currently available data and knowledge on emission factors. The differences in sediment cores are also estimated to mainly result from the too low atmospheric depositions modelled. Also the high levels found in alpine soils support the hypothesis that atmospheric emissions might have been underestimated. The emissions to hydrosphere are easier to estimate based on measurements (WWTP effluents, sewage sludge), which diminishes the risk of a large inaccuracy. The substance flows to the sewer system modelled are in a quite good agreement with the loads derived from measurements (see section 3.2.3).

Especially the emissions from the use phase of products are supposed to be underestimated based on available literature data. However, the diffuse emissions from process *use* estimated have also been based on concentrations found in dust and the amount of dust in indoor environments (see section 2.4.2.5.4). As e.g. the particle bound fraction of DecaBDE has been estimated as >99%, dust is expected to be the major emission pathway for the diffuse emission from the use phase. Also the emissions from recycling, particularly where waste articles are shredded, could have been underestimated. There are no experimental data on BFR emissions from shredder plants. However, it has been shown that PBDE concentrations in ryegrass cultures placed in the vicinity of shredder plants exceeded background levels by one to three orders of magnitude (Wanner et al., 2007). For BDE-47, the degradation of higher brominated PBDEs in the environment or in fires could contribute to the levels found in the atmosphere, in sediments and in soils. As the BFRs under study have a long-range transport potential (Breivik et al., 2006), there might also be a net import with the air from abroad. Reasons for higher emissions in neighbouring countries could be a higher industrial and final consumption or a significant difference in waste management legislation and technology. Illegal burning of hazardous waste and frequent fires on illegal dumpsites may be of significance, too. However, these potential foreign sources are expected to be not the only explanations for the difference between modelled values and levels in the environment.

4.3 Data gaps

Even though there are a few emission experiments, the measurements are highly variable and not all application areas and relevant materials were investigated. For automobile recycling plants, especially for shredding operations, there is no study that determined the emission factors. A significant contribution of recycling processes to the atmospheric emissions have been modelled by Yamaguchi et al. (2006) as well as in the present study and also high levels of PBDEs were detected in the vicinity of shredding plants. An investigation quantifying the emissions from these recycling processes would thus be of high interest.

Long-term emission studies from products in the use phase under real conditions would be needed to testify, if the diffuse atmospheric emissions from the use phase have been underestimated based on currently available data. It is also not much known about the fate of these substances as soon as they have been emitted from the original products. Especially for DecaBDE, a large proportion is expected to adsorb quickly on surfaces and dust and is therefore not likely to be emitted to atmosphere. The amount of DecaBDE and HBCD accumulated in the annually produced dust in Swiss indoor environments has been estimated to be only in the low kg/year range. A study on the indoor emission pathways and distribution of BFRs or similar substances would also provide useful information on the human exposure to such chemicals. There are no measurements of releases from electronic appliance or automobile recycling facilities to wastewater. Even though there are some plants not producing any wastewater in their processes (like e.g. Immark AG), for others the releases including processes like washing hands or working clothes might be more relevant than estimated.

Major data gaps in the environment are missing field data on HBCD concentrations in soil and in the determination of realistic degradation half-lives. There are only measurements of the concentration of HBCD in soil in the vicinity of a potential source. Concentrations in agricultural, natural or even remote soils would provide important information for the risk assessment of HBCD. Many of the experiments on the degradation of BFRs in sediment and soil are not applicable to real conditions. As there is a large domain of uncertainty in the estimation of accurate half-lives based on these experiments, studies on the degradation in sewage sludge or the mathematical models, an attempt should be made to determine most realistic degradation rates.

4.4 Regulatory measures for risk reduction

Regulatory measures and technical innovation in Switzerland resulted in a positive impact on emissions. In order to take actions for risk reduction, human exposure in indoor and working environments should be analyzed and reduced if necessary. Levels found in house dust are not of big relevance for emissions to environment (atmospheric emission or via wastewater pathway), but they could have an impact on toddlers, if the endocrine disrupting effect of a specific BFR would be proved.

If the on-going EU risk assessment would find a risk for HBCD, risk reduction measures should be taken in the use phase. The replacement of intact EPS and XPS insulation panels might be a waste of resources and also be too expensive. Besides of using other insulation material or EPS and XPS with a substitute flame retardant (which has not been found yet), measures to reduce emission during renovation and deconstruction should be taken (e.g. not to break the panels). For DecaBDE, beside of a possible ban, the most important regulative measures seem to have already been taken in Switzerland (i.e. incineration of the sewage sludge, state-of-the-art technique in MSWIPs, prohibition of the dumping of combustible waste). The emissions from electronic appliance and automobile recycling plants have also been estimated to be lowered by technical measures. However, an investigation of these emissions and legislative measures if needed might be beneficial. For the further reduction of emissions from the wastewater pathway, measures to diminish sewer leakage and overflows in case of heavy rain, would lead to a reduction of the input of a whole range of pollutants to hydrosphere and soil.

5 References

Ahn, M. Y., T. R. Filley, C. T. Jafvert, L. Nies, I. Hua and J. Bezares-Cruz (2006). "Photodegradation of Decabromodiphenyl Ether Adsorbed onto Clay Minerals, Metal Oxides, and Sediment." Environ. Sci. Technol. **40**: 215–220.

Alaee, M., P. Arias, A. Sjodin and A. Bergman (2003). "An overview of commercially used brominated flame retardants, their applications, their use patterns in different countries/regions and possible modes of release." Environment International **29**(6): 683-689.

Albemarle Corporation. (2006). "SAYTEX Flame Retardants - Flame Retarding ABS." from http://www.specialchem4polymers.com/documents/indexables/contents/92/abs.pdf.

Alcock, R. E., A. J. Sweetman, K. Prevedouros and K. C. Jones (2003). "Understanding levels and trends of BDE-47 in the UK and North America: an assessment of principal reservoirs and source inputs." Environment International **29**(6): 691-698.

Alonso, E., A. de la Torre, M. A. Martínez and J. V. Tarazona (2007). "Polybrominated Diphenly Ethers (PBDEs) Emission from Spanish Urban Sewage Treatment Plants." Organohalogen Compounds.

Anderson, T. D. and J. D. MacRae (2006). "Polybrominated diphenyl ethers in fish and wastewater samples from an area of the Penobscot River in Central Maine." Chemosphere **62**(7): 1153-1160.

APME (1995). Plastics - A material of choice in building and construction, APME.

APME (2001). Plastics - A material of innovation for the electrical and electronic industry, APME.

Arnot, J., T. Gouin and D. Mackay (2005). Development and Application of Models of Chemical Fate in Canada: Practical Methods for Estimating Environmental Biodegradation Rates, Canadian Environmental Modelling Network.

Baccini, P. and H.-P. Bader (1996). Regionaler Stoffhaushalt. Heidelberg, Spektrum Akademischer Verlag.

Baccini, P. and P. H. Brunner (1991). Metabolism of the anthroposphere. Berlin, Springer-Verlag.

Backus, S., M. Archer, B. Harrison, D. Williams, D. C. G. Muir and M. Alaee (2005). Spatial and temporal distributions of brominated flame retardants (BFRs) in wet-only precipitation collected in the Great Lakes Basin. SETAC, Baltimore.

BAFU. (2007). "Evolution since 1988 – Particulate matter (TSP until 1996, PM10 since 1997): annual mean." from http://www.ubst.bafu.admin.ch/cgiluft/m_seit1988.pl?lang=en&kind=PM10_JM.txt.

Bogdal, C., M. Kohler, P. Schmid, M. Scheringer and K. Hungerbühler (2006). Endocrine disrupting chemicals in the ecosystem of Lake Thun. NRP50 Annual Meeting. Giessbach.

Bogdal, C., M. Kohler, P. Schmid, M. Scheringer and K. Hungerbühler (2007). "Partitioning of Polybrominated Diphenyl Ethers Between Air, Water, Sediment and Fish in Lake Thun (Switzerland)." Organohalogen Compounds.

Borgnes, D. and B. Rikheim (2004a). "Decomposition of BFRs and emission of dioxins from coincineration of MSW and electrical and electronic plastics waste." Organohalogen Compounds **66**.

Borgnes, D. and B. Rikheim (2004b). Målinger ved forbrenning av bromholdig avfall, Norsk renholdsverks-forening.

Brändli, R. C. (2006). Organic pollutants in Swiss compost and digestate, EPF Lausanne.

Breivik, K., A. Sweetman, J. M. Pacyna and K. C. Jones (2002). "Towards a global historical emission inventory for selected PCB congeners - a mass balance approach 2. Emissions." Science of the Total Environment **290**(1-3): 199-224.

Breivik, K., F. Wania, D. C. G. Muir, M. Alaee, S. Backus and G. Pacepavicius (2006). "Empirical and modeling evidence of the long-range atmospheric transport of decabromodiphenyl ether." Environmental Science & Technology **40**(15): 4612-4618.

Brunner, P. H., H. Daxbeck, G. Henseler, B. von Steiger, B. Beer and G. Piepke (1990). RESUB: Der regionale Stoffhaushalt im unteren Bünztal; die Entwicklung einer Methodik zur Erfassung des regionalen Stoffhaushaltes. Dübendorf, ETH Zürich, EAWAG, Abt. Abfallwirtschaft und Stoffhaushalt.

BSEF (2003). Major Brominated Flame Retardants Volume Estimates – Total Market Demand By Region in 2001.

BSEF (2006a). Building Insulation: EPS & XPS foams – Meet the EU's Energy Efficiency and Fire Safety Goals.

BSEF (2006b). VECAP – Annual Progress Report.

BSEF (2007). VECAP – Second Annual Progress Report.

Butt, C. M., M. L. Diamond, J. Truong, M. G. Ikonomou and A. F. H. ter Schure (2004). Spatial distribution of polybrominated diphenyl ethers in southern Ontario as measured in indoor and outdoor window organic films. Environmental Science & Technology. **38:** 724-731.

BUWAL (1992). Abfallkonzept für die Schweiz. Schriftenreihe Umwelt Nr. 173. Bern, Bundesamt für Umwelt, Wald und Landschaft.

BUWAL (1998). Abfallstatistik 1996. Umweltmaterialien Nr. 90. Bern, Bundesamt für Umwelt, Wald und Landschaft.

BUWAL (2003). Evolution of waste to be incinarated in MWIP and capacity of MWIP since 1996.

Cahill, R., L. Summerton and J. P. Clarke (2005). Green Chemistry and the Producer: Flame Retardants.

Carbotech AG (2000). Ökologische Beurteilung von EPS-Dämmstoffen, EPS Schweiz.

Carbotech AG (2004). Langzeitverhalten von EPS-Dämmstoffen, EPS Schweiz.

CEFIC (2006). Flame retardant fact sheet: Hexabromocyclododecane, CEFIC.

Chaulya, S. K., M. K. Chakraborty, M. Ahmad, R. S. Singh, C. Bondyopadhay, G. C. Mondal and D. Pal (2002). "Development of empirical formulae to determine emission rate from various opencast coal mining operations." Water Air and Soil Pollution **140**(1-4): 21-55.

Chen, L. G., B. X. Mai, X. H. Bi, S. J. Chen, X. M. Wang, Y. Ran, X. J. Luo, G. Y. Sheng, J. M. Fu and E. Y. Zeng (2006). "Concentration levels, compositional profiles, and gas-particle partitioning of polybrominated diphenyl ethers in the atmosphere of an urban city in South China." Environmental Science & Technology **40**(4): 1190-1196.

Chiu, C., G. Poole, B. Thibert, D. Cooper and L. Hawke (2006). "Measurement of PBDEs in Landfill Leachates." Organohalogen Compounds.

Cuadra, S. (2005). Child Labour and Health Hazards: Chemical Exposure and Occupational Injuries in Nicaraguan Children Working in a Waste Disposal Site. Department of Laboratory Medicine, Lund University.

Danish EPA (1999). Brominated Flame Retardants: Substance Flow Analysis and Assessment of Alternatives, Danish Environmental Protection Agency**:** 226.

Danon-Schaffer, M., J. Grace, R. Wenning, M. Ikonomou and W. Luksemburg (2006). "PBDEs in Landfill Leachate and Potential for Transfer from Electronic Waste." Organohalogen Compounds.

Davis, J., S. Gonsior and G. Marty (2003). Evaluation of Aerobic and Anaerobic Transformation of Hexabromocyclododecane in Soil. Study 021082. Midland, MI, Environmental Chemistry Research Laboratory. Toxicology & Environmental Research and Consulting. The Dow Chemical Company.

Davis, J. W., S. J. Gonsior, D. A. Markham, U. Friederich, R. W. Hunziker and J. M. Ariano (2006). "Biodegradation and product identification of [C-14]hexabromocyclododecane in wastewater sludge and freshwater aquatic sediment." Environmental Science & Technology **40**(17): 5395-5401.

Davis, J. W., S. J. Gonsior, D. A. Markham and G. T. Marty (2004). Investigation of the biodegradation of [14C]hexabromocyclododecane in sludge, sediment, and soil. Laboratory Project Study ID 031178. . Toxicology & Environmental Research and Consulting. The Dow Chemical Company, Midland, Ml.

de Boysere, J., E. Schmitt, M. Dietz, W. Krause and R. Walz (2007). Overview of Phosphorus Based Halogen Free Flame Retardants for Electric and Electronic Applications. 4th International Workshop on Brominated Flame Retardants, Amsterdam.

de Wit, C. A., K. Nylund, U. Eriksson, M. Haglund, A. Kierkegaard and L. Asplund (2007). Brominated Flame Retardants in Sludge from 50 Swedish Sewage Treatment Plants: Evidence of Anaerobic Degradation of HBCD and TBBPA. BFR2007, Amsterdam.

Diamond, M., C. M. Butt, B. Lam, J. Truong, N. Hernandez-Martinez, T. Labencki, R. Wu, A. Motelay-Massei and A. Simpson (2005). The Composition and Implications of Atmospherically-Derived Films on Impervious Surfaces. ISEEQS, Rehovot.

Dufton, P. (2003). Flame Retardants for Plastics. Market Report, rapra.

EBFRIP (2006). Update on TBBPA, EBFRIP.

ECB (2000). European Union Risk Assessment Report: pentabromo diphenyl ether, European Chemicals Bureau.

ECB (2004). European Union Risk Assessment Report: bis(pentabromophenyl) ether (Update), European Chemicals Bureau.

ECB (2005a). Addendum to the May 2004 Environmental Risk Assessment Report for Decabromophenyl ether, European Chemicals Bureau.

ECB (2005b). European Union Risk Assessment Report (draft): HBCD, European Chemicals Bureau.

ECB (2006a). European Union Risk Assessment Report (draft): HBCD, European Chemicals Bureau.

ECB (2006b). European Union Risk Assessment Report (draft): TBBPA, European Chemicals Bureau.

ECB (2007a). ADDENDUM to the May 2004 Environmental Risk Assessment Report for DECABROMODIPHENYL ETHER, European Chemicals Bureau.

ECB (2007b). European Union Risk Assessment Report (Draft): HBCD, European Chemicals Bureau.

Edwards, R. D., E. J. Yurkow and P. J. Lioy (1998). "Seasonal deposition of housedusts onto household surfaces." Science of the Total Environment **224**(1-3): 69-80.

Falconer, R. L. and T. Harner (2000). "Comparison of the octanol-air partition coefficient and liquid-phase vapor pressure as descriptors for particle/gas partitioning using laboratory and field data for PCBs and PCNs." Atmospheric Environment **34**(23): 4043-4046.

Farrar, N. J., K. E. C. Smith, R. G. M. Lee, G. O. Thomas, A. J. Sweetman and K. C. Jones (2004). "Atmospheric emissions of polybrominated diphenyl ethers and other persistent organic pollutants during a major anthropogenic combustion event." Environmental Science & Technology **38**(6): 1681-1685.

Fontana, M., J. P. Favre and C. Fetz (1999). "A survey of 40,000 building fires in Switzerland." Fire Safety Journal **32**(2): 137-158.

Frey, G. and P. Frischknecht (1989). Abfall und Recycling. Eine Dokumentation der Aktion Saubere Schweiz für Lehrer, Gemeinden und Umweltinteressierte.

Gächter, R. and H. Müller, Eds. (1987). Plastic Additives Handbook. Stabilizers, Processing Aids, Plasticizers, Fillers, Reinforcements, Colorants for Thermoplastics. Munich, Hanser.

Gearhart, J. and H. Posselt (2006). Toxic at any speed – Chemicals in cars and the need for safe alternatives, The Ecology Center.

Gerecke, A. C., W. Giger, P. C. Hartmann, N. V. Heeb, H.-P. E. Kohler, P. Schmid, M. Zennegg and M. Kohler (2006). "Anaerobic Degradation of Brominated Flame Retardants in Sewage Sludge." Chemosphere **64**(2): 311-317.

Gouin, T. and T. Harner (2003). "Modelling the environmental fate of the polybrominated diphenyl ethers." Environment International **29**(6): 717-724.

Greenpeace (2007). Guide to Greener Electronics: 3rd Edition.

Hamre, H. T. (2004). Bromerte flammehemmere i ee-avfall: Karakterisering av utlekkingsegenskaper. Oslo, SFT.

Hanari, N., K. Kannan, Miyake.Y, T. Okazawa, P. R. S. Kodavanti, K. M. Aldous and N. Yamashita (2006). "Occurrence of Polybrominated Biphenyls, Polybrominated Dibenzo-p-dioxins, and Polybrominated Dibenzofurans as Impurities in Commercial Polybrominated Diphenyl Ether Mixtures." Environ. Sci. Technol. **40**(14): 4400-4405.

Harner, T., M. Shoeib, M. Diamond, M. Ikonomou and G. Stern (2006). "Passive sampler derived air concentrations of PBDEs along an urban-rural transect: Spatial and temporal trends." Chemosphere **64**(2): 262-267.

Harrad, S., S. Hazrati and C. Ibarra (2006). "Concentrations of polychlorinated biphenyls in indoor air and polybrominated diphenyl ethers in indoor air and dust in Birmingham, United Kingdom: Implications for human exposure." Environmental Science & Technology **40**(15): 4633-4638.

Harrad, S. and S. Hunter (2006). "Concentrations of polybrominated diphenyl ethers in air and soil on a rural-urban transect across a major UK conurbation." Environmental Science & Technology **40**(15): 4548-4553.

Harrad, S., R. Wijesekera, S. Hunter, C. Halliwell and R. Baker (2004). "Preliminary assessment of U.K. human dietary and inhalation exposure to polybrominated diphenyl ethers." Environmental Science & Technology **38**(8): 2345-2350.

Hassanin, A., K. Breivik, S. N. Meijer, E. Steinnes, G. O. Thomas and K. C. Jones (2004). "PBDEs in European background soils: levels and factors controlling their distribution." Environmental Science & Technology **38**(3): 738-745.

Hayakawa, K., H. Takatsuki, I. Watanabe and S. I. Sakai (2004). "Polybrominated diphenyl ethers (PBDEs), polybrominated dibenzo-p-dioxins/dibenzofurans (PBDD/Fs) and monobromo-polychlorinated dibenzo-p-dioxins/dibenzofurans (MoBPXDD/Fs) in the atmosphere and bulk deposition in Kyoto, Japan." Chemosphere **57**(5): 343-356.

He, J. Z., K. R. Robrock and L. Alvarez-Cohen (2006). "Microbial reductive debromination of polybrominated diphenyl ethers (PBDEs)." Environmental Science & Technology **40**(14): 4429-4434.

Hirai, Y., A. Kida and S. I. Sakai (2005). Emission factors of PCDD/DF and PBDE by landfill fire simulation. Dioxin 2005, Toronto.

Hirai, Y., S. Sakai, K. Sato, K. Hayakawa and K. Shiozaki (2006). "Emissions of Brominated Flame Retardants from TV Sets." Organohalogen Compounds.

Hirai, Y. and S. I. Sakai (2004). Atmospheric Emission of BDE-209 in Japan. Dioxin 2004, Berlin.

Hoh, E. and R. A. Hites (2005). "Brominated flame retardants in the atmosphere of the east-central United States." Environmental Science & Technology **39**(20): 7794-7802.

Hüglin, C., R. Gehrig, U. Baltensperger, M. Gyselc, C. Monn and H. Vonmont (2005). "Chemical characterisation of PM2.5, PM10 and coarse particles at urban, near-city and rural sites in Switzerland." Atmospheric Environment **39**: 637–651.

Hunziker, R. W., S. Gonisor, J. A. MacGregor, D. Desjardins, J. Ariano and U. Friederich (2004). "Fate and Effect of Hexabromocyclododecane in the Environment." Organohalogen Compounds **66**: 2275-2280.

Jaward, F. M., N. J. Farrar, T. Harner, A. J. Sweetman and K. C. Jones (2004a). "Passive air sampling of PCBs, PBDEs, and organochlorine pesticides across Europe." Environmental Science & Technology **38**(1): 34-41.

Jaward, F. M., S. N. Meijer, E. Steinnes, G. O. Thomas and K. C. Jones (2004b). "Further studies on the latitudinal and temporal trends of persistent organic pollutants in Norwegian and UK background air." Environmental Science & Technology **38**(9): 2523-2530.

Jaward, T. M., G. Zhang, J. J. Nam, A. J. Sweetman, J. P. Obbard, Y. Kobara and K. C. Jones (2005). "Passive air sampling of polychlorinated biphenyls, organochlorine compounds, and polybrominated diphenyl ethers across Asia." Environmental Science & Technology **39**(22): 8638-8645.

Kemmlein, S., M. Bergmann and O. Jann (2006). "Emission Test Chamber Study: Specific Emission Rates of PBDE from Selected Materials under Various Conditions." Organohalogen Compounds.

Kemmlein, S., O. Hahn and O. Jann (2003a). Emissionen von Flammschutzmitteln aus Bauprodukten und Konsumgütern, BAM.

Kemmlein, S., O. Hahn and O. Jann (2003b). "Emissions of organophosphate and brominated flame retardants from selected consumer products and building materials." Atmospheric Environment **37**(39-40): 5485-5493.

Kim, Y. J., M. Osako and S. I. Sakai (2006). "Leaching characteristics of polybrominated diphenyl ethers (PBDEs) from flame-retardant plastics." Chemosphere **IN PRESS**.

Klatt, M. (2004). Emission of Hexabromocyclododecane from Polystyrene Foams into Gas Phase - Modeling versus Experiment. BFR 2004, Toronto.

Knoth, W., R. Bassan, C. Belis, G. Jakobi, M. Kirchner, N. Kräuchi, W. Mann, R. Meyer, W. Moche, J. Nebhuth, I. Offenthaler, I. Sedivy, P. Simoncic, M. Uhl, U. Vilhar and P. Weiss (2007). Polybrominated diphenyl ether (PBDE) in humus layers in remote forests. 4th International Workshop on Brominated Flame Retardants, Amsterdam.

Kohler, M., P. Schmid, P. C. Hartmann, M. Sturm, N. V. Heeb, M. Zennegg, A. C. Gerecke, E. Gujer, H. P. Kohler and W. Giger (2006). Occurrence and Temporal Trends of Hexabromocyclododecanes (HBCDs) in Swiss Lake Sediment. SETAC.

Kohler, M., M. Zennegg, A. C. Gerecke, P. Schmid and N. Heeb (2003). "Increasing concentrations of decabromodiphenyl ether (DecaBDE) in Swiss sewage sludge since 1993." Organohalogen Compounds **61**: 123-126.

Kohler, M., Zennegg, M., Hartmann, P.C., Sturm, M., Gujer, E., Schmid, P., Gerecke, A.C., Heeb, N.V., Kohler, H-P. E., Giger, W. (2005). "The historical record of brominated flame retardants and other persistent organic pollutants in a Swiss lake sediment core." 15th annual meeting of SETAC Europe, Lille, France.

Krause, C. (1991). "Umwelt-Survey: Messung und Analyse von Umweltbelastungsfaktoren in der Bundesrepublik Deutschland-Umwelt und Gesundheit." Band III c: Wohn-Innenraum: Raumluft. WaBoLu-Heft **4**.

Külling, D. (2002). Nährstoffe und Schwermetalle im Klärschlamm 1975–1999, FAL.

Kupper, T., L. F. de Alencastro, R. Gatsigazi, R. Furrer, D. Grandjean and J. Tarradellas (unpublished work). Concentrations and specific loads of brominated flame retardants in sewage sludge originating from a monitoring network in Switzerland.

La Guardia, M. J., R. C. Hale and E. Harvey (2006). "Detailed polybrominated diphenyl ether (PBDE) congener composition of the widely used penta-, octa-, and deca-PBDE technical flame-retardant mixtures." Environmental Science & Technology **40**(20): 6247-6254.

Lam, B., M. L. Diamond, A. J. Simpson, P. A. Makar, J. Truong and N. A. Hernandez-Martinez (2005). "Chemical composition of surface films on glass windows and implications for atmospheric chemistry." Atmospheric Environment **39**(35): 6578-6586.

Laube, A. and A. Vonplon (2004). Klärschlammentsorgung in der Schweiz – Mengen- und Kapazitätserhebung. Umwelt-Materialien Nr. 181. Bern, Bundesamt für Umwelt, Wald und Landschaft.

Lee, R. G. M., G. O. Thomas and K. C. Jones (2004). "PBDEs in the Atmosphere of Three Locations in Western Europe." Environmental Science & Technology **38**(3): 699-706.

Leidner, J. (1981). Plastics Waste: Recovery of economic value. Basel, Dekker.

Leisewitz, A. and W. Schwarz (2001). Flammhemmende Ausrüstung ausgewählter Produkte - anwendungsbezogene Betrachtung: Stand der Technik, Trend, Alternativen. Erarbeitung von Bewertungsgrundlagen zur Substitution umweltrelevanter Flammschutzmittel. Umweltbundesamt.

Li, A., K. J. Rockne, N. Sturchio, W. Song, J. C. Ford, D. R. Buckley and W. J. Mills (2006). "Polybrominated Diphenyl Ethers in the Sediments of the Great Lakes. 4. Influencing Factors, Trends, and Implications." Environmental Science & Technology **40**(24): 7528–7534.

Litten, S., D. J. McChesney, M. C. Hamilton and B. Fowler (2003). "Destruction of the World Trade Center and PCBs, PBDEs, PCDD/Fs, PBDD/Fs, and chlorinated biphenylenes in water, sediment, and sewage sludge." Environmental Science & Technology **37**(24): 5502-5510.

LRV (1985). Luftreinhalte-Verordnung vom 16. Dezember 1985, Schweizerischer Bundesrat.

MacGregor, J. A. and W. B. Nixon (1997). "Hexabromocyclododecane (HBCD): Determination of n-Octanol/Water Partition Coefficient." Wildlife International LTD 439C-104. Arlington, VA: Brominated Flame Retardant Industry Panel, Chemical Manufacturers Association.

Mark, F. E., H. Dresch, B. Bergfeldt, B. Dima, W. Grüttner, F. Kleppmann, K. Kramer, T. Lehner and J. Vehlow (2006). Mitverbrennung von Reststoffen aus der Verwertung von Elektro- und Elektronik-Altgeräten im MHKW Würzburg 2004. Müll und Abfall. **1**.

Matscheko, N., M. Tysklind, C. de Wit, S. Bergek, R. Andersson and U. Sellstrom (2002). "Application of sewage sludge to arable land-soil concentrations of polybrominated diphenyl ethers and polychorinated dibenzo-p-dioxins, dibenzofurans, and biphenyls, and their accumulation in earthworms." Environmental Toxicology and Chemistry **21**(12): 2515-2525.

MeteoSchweiz (2006). Typische Wetterlagen im Alpenraum, Bundesamt für Meteorologie und Klimatologie.

Moche, W., K. Stephan and G. Thanner (2004). Bromierte Flammschutzmittel in der aquatischen Umwelt. Wien, UBA.

Morf, L., R. Taverna, H. Daxbeck and R. Smutny (2002). Selected polybrominated flame retardants, PBDEs and TBBPA, Substance flow analysis, Buwal.

Morf, L. S., J. Tremp, R. Gloor, Y. Huber, M. Stengele and M. Zennegg (2005). "Brominated Flame Retardants in Waste Electrical and Electronic Equipment: Substance Flows in a Recycling Plant." Environ. Sci. Technol. **39**(22): 8691-8699.

Morose, G. (2006). An Overview of Alternatives to Tetrabromobisphenol A (TBBPA) and Hexabromocyclododecane (HBCD). U. o. M. Lowell, Jennifer Altman Foundation.

Morris, S., C. R. Allchin, B. N. Zegers, J. J. H. Haftka, J. P. Boon, C. Belpaire, P. E. G. Leonards, S. P. J. Van Leeuwen and J. De Boer (2004). "Distributon and fate of HBCD and TBBPA brominated flame retardants in north sea estuaries and aquatic food webs." Environmental Science & Technology **38**(21): 5497-5504.

OECD (2004). Emission scenario document on plastics additives, OECD.

Oliaei, F. (2005). Flame Retardants: Polybrominated Diphenyl Ethers (PBDEs) - Background Paper, Minnesota Pollution Control Agency.

Osako, M., Y. J. Kim and K. Kitamura (2005). Interaction between dissolved humic matter and micropollutants in landfill leachate. Tenth International Waste Management and Landfill Symposium, Sardinia.

Osako, M., Y. J. Kim and S. I. Sakai (2004). "Leaching of brominated flame retardants in leachate from landfills in Japan." Chemosphere **57**(10): 1571-1579.

Österreichische Länderversicherer. (2006). "Fast 2000 Fahrzeugbrände an 365 Tagen." Retrieved September 2006, from http://www.laenderversicherer.at/aktion/aktion.htm.

Palm, A., I. T. Cousins, D. Mackay, M. Tysklind, C. Metcalfe and M. Alaee (2002a). "Assessing the environmental fate of chemicals of emerging concern: a case study of the polybrominated diphenyl ethers." Environmental Pollution **117**(2): 195-213.

Palm, A., J. Sternbeck, L. Embertsén, A. Jonsson and U. Mohlander (2002b). Hexabromcyklodekan (HBCD) i Stockholm - modellering av diffusa emissioner. Stockholm, IVL Svenska Miljöinstitutet AB.

Parsons, J., B. Zegers, E. Skoczynska and P. De Voogt (2004). "Reductive debromination of decabromodiphenyl ether (BDE 209) by anaerobic sediment microorganisms." Organohalogen Compounds **66**.

Petreas, M. and D. Oros (2006). "PBDEs in California waste streams." Organohalogen Compounds.

PlasticsEurope (2004). Produktions- und Verbrauchsdaten für Kunststoffe in Deutschland unter Einbeziehung der Verwertung 2003, PlasticsEurope.

PlasticsEurope (2005). Plastics Business Data and Charts, KVE.

Posner, S. (2006). Survey and technical assessment of alternatives to TBBPA and HBCDD. Sundbyberg, Swedish Chemicals Inspectorate (KEMI).

Prevedouros, K., K. C. Jones and A. J. Sweetman (2004a). "Estimation of the production, consumption, and atmospheric emissions of pentabrominated diphenyl ether in Europe between 1970 and 2000." Environmental Science & Technology **38**(12): 3224-3231.

Prevedouros, K., K. C. Jones and A. J. Sweetman (2004b). "European-scale modeling of concentrations and distribution of polybrominated diphenyl ethers in the pentabromodiphenyl ether product." Environmental Science & Technology **38**(22): 5993-6001.

Rayne, S. and M. G. Ikonomou (2005). "Polybrominated diphenyl ethers in an advanced wastewater treatment plant. Part 1: Concentrations, patterns, and influence of treatment processes." Journal of Environmental Engineering and Science **4**(5): 353-367.

Reinhardt, T. and U. Richers (2004). Entsorgung von Schredderrückständen: Ein aktueller Überblick, Forschungszentrum Karlsruhe.

Remberger, M., J. Sternbeck, A. Palm, L. Kaj, K. Strömberg and E. Brorström-Lundén (2004). "The environmental occurrence of hexabromocyclododecane in Sweden." Chemosphere **54**(1): 9-21.

Sakai, S., Y. Hirai, T. Ninomiya, H. Aizawa, T. Nakano and Y. Muroishi (2006a). "Emissions inventory of polybrominated diphenyl ethers (PBDEs) on a homologue basis." Organohalogen Compounds.

Sakai, S., S. Takahashi, M. Osada and T. Miyazaki (2006b). "Dioxin-related compounds, brominated flame retardants and heavy metals in automobile shredder residue (ASR) and their behavior in high-temperature melting process." Organohalogen Compounds.

Sakai, S. I., Y. Hirai, H. Aizawa, S. Ota and Y. Muroishi (2006c). "Emission inventory of deca-brominated diphenyl ether (DBDE) in Japan." Journal of Material Cycles Waste Management: 56-62.

Sakai, S. I., Y. Hirai, S. Ota and K. Makiya (2005). Emission factors of PBDD/DFs and PBDEs from textile processing and BFR production, and the tentative PBDEs emission inventory. Dioxin 2005, Toronto.

Sanchez-Brunete, C., E. Miguel and J. L. Tadeo (2006). "Determination of polybrominated diphenyl ethers in soil by ultrasonic assisted extraction and gas chromatography mass spectrometry." Talanta **70**(5): 1051-1056.

Schädler, B. and R. Weingartner (2002). Komponenten des natürlichen Wasserhaushaltes 1961–1990. BWG: Hydrologischer Atlas der Schweiz. Bern.

Scheidl, K. (2006). Global Business Review: Polyethylene / Polypropylene. LME Plastics Seminar. London.

Schenker, U., M. MacLeod, M. Scheringer and K. Hungerbuhler (2005). "Improving data quality for environmental fate models: A least-squares adjustment procedure for harmonizing physicochemical properties of organic compounds." Environmental Science & Technology **39**(21): 8434-8441.

Schluep, M., M. Thomann, A. Häner, R. Gälli and G. Stucki (2006). Organische Mikroverunreinigungen und Nährstoffhaushalt. Eine Standortbestimmung für die Siedlungswasserwirtschaft. Umwelt-Wissen. BAFU.

Schneeberger, K. (1999). Kunststoffe in der Schweizer Bauindustrie, Dow Europe.

Schweizer Fernsehen. (2007). "Italien: Das «Drecksgeschäft» der Camorra." 10vor10, from http://www.sf.tv/sf1/10vor10/index.php?docid=20070413.

Sellström, U., C. A. de Wit, N. Lundgren and M. Tysklind (2005). "Effect of Sewage-Sludge Application on Concentrations of Higher-Brominated Diphenyl Ethers in Soils and Earthworms." Environ. Sci. Technol.

Sellström, U., A. Kierkegaard, T. Alsberg, P. Jonsson, C. Wahlberg and C. de Wit (1999). "Brominated flame retardants in sediments from european estuaries, the baltic sea and in sewage sludge " Organohalogen Compounds **40**: 383-386.

Sennhauser, W. R. A. (2003). Greifensee – Phosphoreintrag aus Trennsystem und Überläufen.

Simonson, M., P. Blomqvist, A. Boldizar, K. Möller, L. Rosell, C. Tullin, H. Stripple and J. Sundqvist (2000). Fire-LCA Model: TV Case Study. Fire Technology, Swedish National Testing and Research Institute.

Sjödin, A., H. Carlsson, K. Thuresson, S. Sjolin, A. Bergman and C. Ostman (2001). "Flame retardants in indoor air at an electronics recycling plant and at other work environments." Environmental Science & Technology **35**(3): 448-454.

Söderström, G., U. Sellström, C. A. De Wit and M. Tysklind (2004). "Photolytic debromination of decabromodiphenyl ether (BDE 209)." Environmental Science & Technology **38**(1): 127-132.

Song, M., S. Chu, R. Letcher and R. Seth (2006). "Fate, Partitioning, and Mass Loading of Polybrominated Diphenyl Ethers (PBDEs) during the Treatment Processing of Municipal Sewage." Environ. Sci. Technol. **In Press**.

Sternbeck, J., M. Remberger, L. Kaj, K. Strömberg, A. Palm and E. Brorström-Lundén (2001). HBCD i Svergie – screening ab ett bromerat flamskyddsmedel, IVL Swedish Environmental Research Institute.

Strandberg, B., N. G. Dodder, I. Basu and R. A. Hites (2001). "Concentrations and spatial variations of polybrominated diphenyl ethers and other organohalogen compounds in Great Lakes air." Environmental Science & Technology **35**(6): 1078-1083.

Streets, S. S., S. A. Henderson, A. D. Stoner, D. L. Carlson, M. F. Simcik and D. L. Swackhamer (2006). "Partitioning and bioaccumulation of PBDEs and PCBs in Lake Michigan." Environmental Science & Technology **40**(23): 7263-7269.

Suzuki, G., K. Nose, H. Takigami, S. Takahashi and S. Sakai (2006). "PBDEs and PBDD/Fs in House and Office Dust from Japan." Organohalogen Compounds.

Swiss Federal Statistical Office (2002). "Der spezielle Beitrag: Wandel der Bodennutzung in der Schweiz."

Takigami, H., Y. Hirai, Y. Matsuzawa and S. Sakai (2006). "Brominated flame retardants and brominated dioxins in the working environment and environmental emission – a case study at an electronics recycling plant." Organohalogen Compounds **68**.

Tamade, Y., S. Shibakawa, H. Osaki, S. Kashimoto, Y. Yagi, S. I. Sakai and T. Takasuga (2002). A study of brominated compound release from appliance-recycling facility. Dioxin 2002.

ter Schure, A. F. H., C. Agrell, A. Bokenstrand, J. Sveder, P. Larsson and B. N. Zegers (2004a). "Polybrominated diphenyl ethers at a solid waste incineration plant II: atmospheric deposition." Atmospheric Environment **38**(30): 5149-5155.

ter Schure, A. F. H. and P. Larsson (2002). "Polybrominated diphenyl ethers in precipitation in Southern Sweden (Skane, Lund)." Atmospheric Environment **36**(25): 4015-4022.

ter Schure, A. F. H., P. Larsson, C. Agrell and J. P. Boon (2004b). "Atmospheric transport of polybrominated diphenyl ethers and polychlorinated biphenyls to the Baltic Sea." Environmental Science & Technology **38**(5): 1282-1287.

Tokai, A., H. Yamaguchi, K. Tsunemi, Y. Yonezawa, M. Tominaga and J. Nakanishi (2004). Assessing risk of substitution with the example of deca brominated diphenyl ether in Japan. SRA.

VKE (2002a). Kunststoff im Automobil.

VKE (2002b). Verwertungspotenziale von Kunststoffabfällen (Nicht-Verpackungen) aus Gewerbe und Privathaushalten. UFOPLAN-Vorhaben, VKE, UBA.

VKE (2003). Kunststoffe in Elektro- und Elektronikgeräten, KVE.

von Arx, U. (1999). Bauprodukte und -Inhaltsstoffe, Buwal.

von Arx, U. (2006). Kupfer. Verbrauch, Umwelteinträge und -vorkommen. Umwelt-Wissen 06/01. Bern, Bundesamt für Umwelt (BAFU).

Vonderheide, A. P., S. R. Mueller-Spitz, J. Meija, G. L. Welsh, K. E. Mueller, B. K. Kinkle, J. R. Shann and J. A. Caruso (2006). "Rapid breakdown of brominated flame retardants by soil microorganisms." Journal of Analytical Atomic Spectrometry **21**(11): 1232-1239.

VREG (1998). Verordnung über die Rückgabe, die Rücknahme und die Entsorgung elektrischer und elektronischer Geräte, Schweizerischer Bundesrat.

Vulykh, N., S. Dutchak, E. Mantseva and V. Shatalov (2006). EMEP contrubution to the Preparatory Work for the Review of the CLRTAP Protocol on POPs. New Substances: Model Assessment of Potential for Long-range Transboundary Atmospheric Transport and Persistence of PentaBDE, Endosulfan, Dicifol, HCBD, PeCB, PCN. EMEP/MSC-E Technical Report 1/2006.

Waber, U. (2001). Elektronikschrott in der Schweiz. Bern, Bundesamt für Umwelt, Wald und Landschaft (BUWAL).

Wania, F. (2003). Assessing the Long-Range Transport Potential of Tetrabromobisphenol A and Hexabromocyclododecane Using Several Multimedia Transport Models, A report to the Bromine Science and Environment Forum.

Wania, F. and C. B. Dugani (2003). "Assessing the long-range transport potential of polybrominated diphenyl ethers: a comparison of four multimedia models." Environmental Toxicology And Chemistry / SETAC **22**(6): 1252-1261.

Wania, F. and D. Mackay (1995). "A global distribution model for persistent organic chemicals." Science of the Total Environment **160–161**: 211–232.

Wanner, A., L. Peichl, J. Köhler, S. Schädel, A. Rupprich and W. Körner (2007). Polybrominated diphenyl ether (PBDE) in Italian ryegrass exposed near to Bavarian shredder plants. BFR2007, Amsterdam.

Watanabe, I. and S. I. Sakai (2003). "Environmental release and behavior of brominated flame retardants." Environment International **29**(6): 665-682.

Weber, R. and B. Kuch (2003). "Relevance of BFRs and thermal conditions on the formation pathways of brominated and brominated-chlorinated dibenzodioxins and dibenzofurans." Environment International **29**(6): 699-710.

Wilford, B. H., T. Harner, J. P. Zhu, M. Shoeib and K. C. Jones (2004). "Passive sampling survey of polybrominated diphenyl ether flame retardants in indoor and outdoor air in Ottawa, Canada: Implications for sources and exposure." Environmental Science & Technology **38**(20): 5312-5318.

Wilford, B. H., G. O. Thomas, R. E. Alcock, K. C. Jones and D. R. Anderson (2003). Polyurethane foam as a source of PBDEs to the environment. Dioxin 2003, Boston.

Wragg, P. (2005). TFA DecaBDE Users Group Survey, Texconsul Ltd.

Yamaguchi, H., K. Tsunemi and A. Tokai (2006). "Estimation of environmental emission load of decabromodiphenyl ether in Japan applied dynamic substance flow analysis."

Yen, J.-H., W. Liao and Y.-S. Wang (2006). "Degradation of Polybrominated Diphenyl Ethers (PBDEs) in Sediments of Er-Jen River and Nan-Kan River Basin in Taiwan." Organohalogen Compounds **68**.

Zegers, B. N., W. E. Lewis, K. Booij, R. H. Smittenberg, W. Boer, J. de Boer and J. P. Boon (2003). "Levels of polybrominated diphenyl ether flame retardants in sediment cores from Western Europe." Environmental Science & Technology **37**(17): 3803-3807.

Zennegg, M., M. Kohler, A. C. Gerecke and P. Schmid (2003). "Polybrominated diphenyl ethers in whitefish from Swiss lakes and farmed rainbow trout." Chemosphere **51**(7): 545-553.

6 Glossary

ABS	Acrylonitrile butadiene styrene
APC residue	Air pollution control residue (in waste incineration plants)
APME	Association of Plastics Manufacturers in Europe (now PlasticsEurope)
ASR	Automobile shredder residue
BSEF	Bromine Science and Environmental Forum, Belgium
BFRs	Brominated flame retardants
CEFIC	European Chemical Industry Council, Belgium
d.w.	dry weight
DecaBDE	Decabromodiphenyl ether
E&E	Electrical and electronic equipment
EBFRIP	European Brominated Flame Retardant Industry Panel
EF	Emission factor
EPS	Expanded polystyrene
f_{oc}	Fraction of organic carbon
HBCD	Hexabromocyclododecane
HIPS	High-impact polystyrene
K_{AW}	Air-water partition coefficient
K_{FA}	Film-air partition coefficient
K_{OA}	Octanol-air partition coefficient
K_{OW}	Octanol-water partition coefficient
MFA	Material flow analysis
MMFA	Mathematical material flow analysis
MSWIP	Municipal solid waste incineration plant
Nm^3	Normal cubic meter
OctaBDE	Octabromodiphenyl ether
OECD	Organisation for Economic Co-operation and Development
PC	Proportion coefficient
PentaBDE	Pentabromodiphenyl ether
PE	Polyethylene
PEC	Predicted environmental concentration
PP	Polypropylene
PS	Polystyrene
PUR	Polyurethane

PVC	Polyvinyl chloride
RA	Risk assessment
RDF	Refuse derived fuel
Safe-Pro	Excel based fugacity model for Switzerland (developed by BMG Engineering AG)
SFA	Substance flow analysis
TBBPA	Tetrabromobisphenol A
TC	Transfer coefficient
VEPAC	Voluntary Emission Control Action Programme
VKE	Verband Kunststofferzeugende Industrie, Germany
VREG	Verordnung über die Rückgabe, die Rücknahme und die Entsorgung elektrischer und elektronischer Geräte
w/w	weight per weight
w.w.	wet weight
WEEE	Waste electrical and electronic equipment
WWTP	Wastewater treatment plant
XPS	Extruded polystyrene

7 Appendix

7.1 Balance equations used to generate the model

Inputs to system

i_1 *Drawn; value mainly affected by the amount consumed*
i_2 *Drawn; value mainly affected by the amount consumed*
i_8 *Externally calculated*

Production

f_{1-2} *Remainder of* $(i_1 + f_{6-1})$
$f_{1-7} = (i_1 + f_{6-1}) \cdot TC(P,inc)$
$f_{1-8} = (i_1 + f_{6-1}) \cdot TC(P,sew)$
$o_{1-11} = (i_1 + f_{6-1}) \cdot EF(P,atm)$

Trade

$f_{2-3} = (i_2 + f_{1-2}) \cdot (1 - TC(T,export)) \cdot TC(T,const)$
f_{2-4} *Remainder of* $(i_2 + f_{1-2})$
$o_{2-14} = (i_2 + f_{1-2}) \cdot TC(T,export)$

Construction

f_{3-4} *Remainder of* $(f_{2-3} + f_{6-3})$
$f_{3-7} = f_{2-3} \cdot TC(C,waste) \cdot (1 - PC(C,landfill))$
$f_{3-10} = f_{2-3} \cdot TC(C,waste) \cdot PC(C,landfill)$
$o_{3-11} = f_{2-3} \cdot EF(C,atm)$

Use

The equations in the process *use* are more complicated due to the utilization of residence times. Therefore, no balance equations are shown for this process.

Deconstruction

$f_{5-6} = (f_{4-5} - o_{5-11} - o_{5-13}) \cdot TC(D,recycling)$
f_{5-7} *Remainder of* $(f_{4-5} - o_{5-11} - o_{5-13})$
$f_{5-10} = (f_{4-5} - o_{5-11} - o_{5-13}) \cdot TC(D,landfill)$
$o_{5-11} = (f_{4-5}) \cdot EF(D,atm.d)$
$o_{5-13} = (f_{4-5}) \cdot EF(D,soil)$

Recycling

$f_{6-1} = (f_{4-6} + f_{5-6} - f_{6-8} - o_{6-11}) \cdot TC(R,reuse)$
f_{6-7} *Remainder of* $(f_{4-6} + f_{5-6} - f_{6-8} - o_{6-11}^{(1)} - o_{6-11}^{(2)})$
$f_{6-8} = (f_{4-6} + f_{5-6}) \cdot TC(R,sew)$
$f_{6-10} = (f_{4-6} + f_{5-6} - f_{6-8} - o_{6-11}) \cdot TC(R,landfill)$
$o_{6-11} = (f_{4-6} + f_{5-6}) \cdot EF(R,atm)$

$o_{6-14} = (f_{4-6} + f_{5-6} - f_{6-8} - o_{6-11}) \cdot \mathrm{TC(R,export)}$

Incineration

$f_{7-10}^{(1)} = (f_{1-7} + f_{3-7} + f_{4-7}^{(1)} + f_{4-7}^{(2)} + f_{5-7} + f_{6-7} + f_{9-7}) \cdot \mathrm{TC(I,APC\ res.)} \cdot \mathrm{PC(I,landfill)}$

$f_{7-10}^{(2)} = (f_{1-7} + f_{3-7} + f_{4-7}^{(1)} + f_{4-7}^{(2)} + f_{5-7} + f_{6-7} + f_{9-7}) \cdot \mathrm{TC(I,bot.ash)}$

$o_{7-11} = (f_{1-7} + f_{3-7} + f_{4-7}^{(1)} + f_{4-7}^{(2)} + f_{5-7} + f_{6-7} + f_{9-7}) \cdot \mathrm{EF(I,atm)}$

$o_{7-12} = (f_{1-7} + f_{3-7} + f_{4-7}^{(1)} + f_{4-7}^{(2)} + f_{5-7} + f_{6-7} + f_{9-7}) \cdot \mathrm{EF(I,hyd)}$

$o_{7-14} = (f_{1-7} + f_{3-7} + f_{4-7}^{(1)} + f_{4-7}^{(2)} + f_{5-7} + f_{6-7} + f_{9-7}) \cdot \mathrm{TC(I,APC\ res.)} \cdot (1 - \mathrm{PC(I,landfill)})$

Sewerage

f_{8-9} *Remainder of* $(i_8 + f_{1-8} + f_{4-8}^{(1)} + f_{4-8}^{(2)} + f_{6-8} + f_{10-8})$

$o_{8-12} = (i_8 + f_{1-8} + f_{4-8}^{(1)} + f_{4-8}^{(2)} + f_{6-8} + f_{10-8}) \cdot \mathrm{EF(Se,overflow)}$

$o_{8-13} = (i_8 + f_{1-8} + f_{4-8}^{(1)} + f_{4-8}^{(2)} + f_{6-8} + f_{10-8}) \cdot \mathrm{EF(Se,leakage)}$

WWTP

f_{9-7} *Remainder of* f_{8-9}

$f_{9-10} = f_{8-9} \cdot \mathrm{TC(W,sludge)} \cdot \mathrm{PC(W,landfill)}$

$o_{9-12} = f_{8-9} \cdot (1 - \mathrm{TC(W,sludge)})$

$o_{9-13} = f_{8-9} \cdot \mathrm{TC(W,sludge)} \cdot \mathrm{PC(W,agri)}$

Landfill

$f_{10-8} = m_{10} \cdot \mathrm{TC(L,leach)} \cdot \mathrm{PC(L,sew)}$

$o_{10-11}^{(1)} = m_{10} \cdot \mathrm{EF(L,atm.g)}$

$o_{10-11}^{(2)} = m_{10} \cdot \mathrm{EF(L,atm.d)} + (f_{3-10} + f_{4-10} + f_{5-10} + f_{7-10}^{(1)}) \cdot \mathrm{EF(L,unload)}$

$o_{10-13} = m_{10} \cdot \mathrm{TC(L,leach)} \cdot (1 - \mathrm{PC(L,sew)})$

7.2 Contacts established

7.2.1 Research groups and industrial research contacted

Information on consumption and trends

Prevedouros K, Sweetman AJ (Lancaster University)	Model of the production, consumption, and atmospheric emissions of pentabrominated diphenyl ether in Europe
Cederberg I (KemI, Sweden)	Consumption of HBCD and TBBPA
Kramer E (Fachhochschule NW)	BFR contents in plastics
Lyday PA (U.S. Geological Survey)	Bromine production trends

Emissions during use

Sakai SI, Hirai Y (Kyoto University, Japan)	Emission factors, emission inventory in Japan
Kemmlein S (BAM Germany)	Emission cell tests
Klatt M (BASF, Germany)*	Emission cell tests: HBCD from XPS boards
Herrmann T, Ball M (ERGO research Germany)*	Emission cell tests: TBBPA from computer monitors
Wilford B (Air Quality Research Branch Environment Canada)	Emissions of PBDEs from PUR foam
Gearhart G (Ecology Center, Michigan)	Emissions of PBDEs from cars
Harrad S (University of Birmingham)	Emissions of PBDEs from E&E and cars, pathways of house dust
Lambert J (Confederation of British Wool Textiles)	Emissions from textiles to wastewater, VECAP
Andersson L (KemI, Sweden)	Newest version of EU Risk Assessment draft on HBCD
Caley J (UK Environment Agency)	Newest version of EU Risk Assessment draft on TBBPA

*: no answer

Waste management

Sakai SI, Hirai Y (Kyoto University, Japan)	Emission factors, emission inventory in Japan
Tamade (Takuma, Japan)	Emissions from appliance recycling facility and from incinerator plant. Emission factors and transfer coefficients.
Borgnes D, Rikheim B (SFT, Norway)*	Emission factors and transfer coefficients from incineration plant
Wanner A (Bavarian Environment Agency)	Emissions from shredding plants
Rieckermann J (Eawag, Switzerland)	Leaching losses from sewerage system
Rossi L (Eawag, Switzerland)	Sewerage overflows
De Boer J (Netherlands Institute for Fisheries Research)	Emission factors and transfer coefficients from WWTP
North K (Environmental Compliance Division, Palo Alto)*	Emission factors and transfer coefficients from WWTP
Osako M (NIES, Japan)	Leaching from landfills

Townsend T (University of Florida)*	Leaching from landfills
Danon-Schaffer M (University of British Columbia)	Leaching from landfills
Petreas M (Department of Toxic Substances Control, California)	BFRs in WEEE and ASR

*: no answer

Information on concentration levels in environment

Kohler M, Gerecke A, Bogdal C, Schmid P, Zennegg M (EMPA, Switzerland)	Collaboration in the NFP50 project. Information on concentrations in sediments, sewage sludge, household dust and biosolids
Kupper T (Eawag, Switzerland)	Information on concentrations in sewage sludge and biosolids
Knoth W (Umweltbundesamt Germany and Monarpop)	Measurements of PBDE in humus layers in remote forests
Remberger M (Svenska Miljöinstitutet)*	HBCD measurements

*: no answer

Fires

Hirai Y (Kyoto University)	Emissions of BFRs into atmosphere
Simonson M, Lönnermark A, Blomqvist P (Swedish National Testing and Research Institute)	Emissions and life cycle assessment
Diamond M (University of Toronto)*	Emissions of BFRs into atmosphere
Litten ST (New York State Department of Environmental Conservation)	Emissions of BFRs into hydrosphere

Modelling

Kohler M, Gerecke A, Bogdal C, Schmid P, Zennegg M (Empa, Switzerland)	Environmental fate, degradation in sediments
Schenker U, Trudel D (ETH Zurich)	Degradation of BFRs in soil, pathways of BFRs in house dust
Tokai A (Center for Chemical Risk Management, AIST, Japan)	Model of the consumption, stock and waste of BFRs
Prevedouros K (Lancaster University)	Model on atmospheric emissions
Wania F (University of Toronto)	Environmental fate model
Braun C, Gälli R (BMG Engineering)	Environmental fate modelling
Reemtsma T (TU Berlin)	Modelling of indoor pathways, dust
Uhl M (Umweltbundesamt, Austria)	Modelling of indoor pathways, dust
Krieger U (ETH Zurich)	Atmospheric transport of particles, concentration, org. fraction
Hueglin C (Empa, Switzerland)	Atmospheric transport of particles, concentration, org. fraction
Baltensperger U (PSI, Switzerland)	Suspended particles, org. fraction in the Swiss atmosphere

7.2.2 Organisations, industry and national authorities contacted

Information on BFRs

The Bromine Science and Environmental Forum (BSEF)	Consumption trends of BFRs
European Brominated Flame Retardant Industry Panel (EBFRIP)	Consumption trends of BFRs
Verband der Europäischen chemischen Industrie (CEFIC)	Consumption trends of BFRs

Information on plastics

PlasticsEurope	Information on consumption of different plastic types and time trends
Kunststoff Verband Schweiz (KVS)	Information on consumption of different plastic types and time trends, Swiss specific data
EMS Grivory*	Information on BFR use in polyamides and other plastics
Huntsman Advanced Materials*	Information on BFR use in epoxy resins and other plastics

*: no answer

Information on consumer goods

Swiss customs authority	Import and export data of consumer goods

Information on electrical and electronic equipment

S.EN.S	Amounts of recycled household appliances
SWICO	Amounts of recycled office and consumer electronics
Swiss Consumer Electronics Association (SCEA)	Sale statistics of consumer electronics

Information on textiles and furniture

Textilverband Schweiz (TVS)	Information and production statistics of textiles
Textile Finishers Association (UK)	Information and results of VECAP
Verband der Schweizer Möbelindustrie (SEM)	Sale statistics of mattresses and upholstery furniture
interieursuisse	Information and sale statistics of textiles
Verband Schweizerischer Filialunternehmungen (VSF)	Information and sale statistics of textiles
Institut für Fußbodentechnik und Raumausstattung mbH (Cologne, Germany)	Information on flame retardants in carpets
Österreichisches Textil-Forschungsinstitut	Information on flame retardants in carpets
Swiss Retail Federation*	Sale statistics of textiles
Pfister	Sale statistics of textiles
Interio	Sale statistics of textiles
Ikea	Sale statistics of textiles

JAB	Sale statistics of textiles, BFRs used in textiles
Happy AG*	BFR use and production statistics of mattresses
bico	BFR use and production statistics of mattresses
roviva*	BFR use and production statistics of mattresses

*: no answer

Information on construction material

EPS Verband Schweiz*	HBCD use in EPS and XPS, recycling of EPS and XPS
Sarnafil	Imports and Swiss production, flame retardant use
Folag AG	Imports and Swiss production, flame retardant use
IMS Kunststoffe AG	Imports and Swiss production, flame retardant use
PVCH	Information on (historic) BFR use in PVC
Arbeitsgemeinschaft PVC und Umwelt e.V.*	Information on (historic) BFR use in PVC
Swiss Federal Office for the Environment (FOEN)	Consumption and domestic production of construction materials

*: no answer

Fires

Verband Kantonaler Feuerversicherungen (VKF)	Statistics on building and TV fires
Interkantonaler Rückversicherungsverband (IRV)	Statistics on building and TV fires
Bundesamt für Statistik	Statistics on building and TV fires
Bundesamt für Privatversicherungen	Statistics on building and TV fires
Gebäudeversicherung Zürich	Statistics on building and TV fires
Verband Schweizerischer Radio- und Televisions-Fachgeschäfte (VSRT)	Statistics on TV fires

Waste management

Swiss Federal Office for the Environment (FOEN)	Waste disposal statistics, sludge disposal statistics
Bachema AG	Analysis of bottom ash, APC residue and wastewater from incineration plant
Stiftung Auto Recycling Schweiz	End-of-life vehicles, transfer coefficients of ASR
Washington State Department of Ecology	PBDEs in cars and in ASR
Swiss recycling company (name undisclosed)	Emissions during recycling
Metso Lindemann GmbH, Düsseldorf	Emissions during recycling
Kehrichtverwertung Zürcher Oberland (KEZO)	Information on landfills
Deponie Lufingen	Volume of landfills leachates, mass on disposed materials, dust generated on landfills
Rhein-Main-Deponie GmbH / Hessisches Landesamt für Umwelt und Geologie	Dust generated on landfills

Flemish government Environment, Nature and Energy department	Releases of BFRs to wastewater

7.3 Plastic consumption in the domestic construction sector

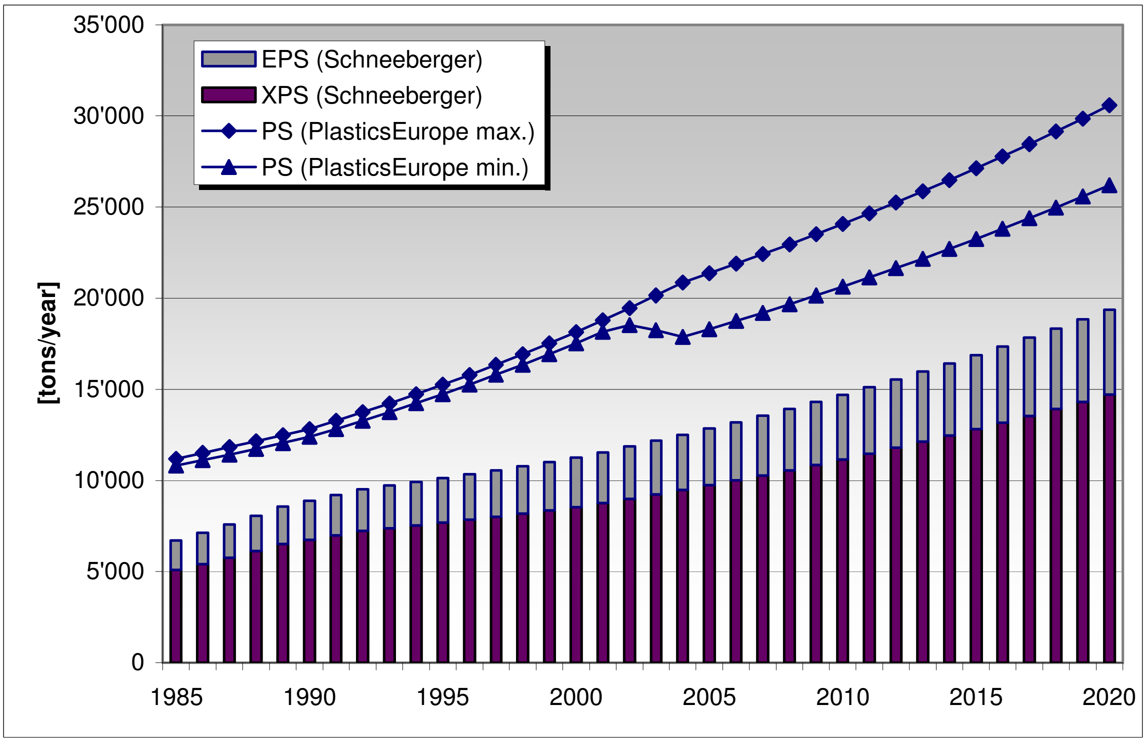


Figure 51 Consumption of expanded and extruded polystyrene in the Swiss construction sector (PS includes different polystyrene plastic types)

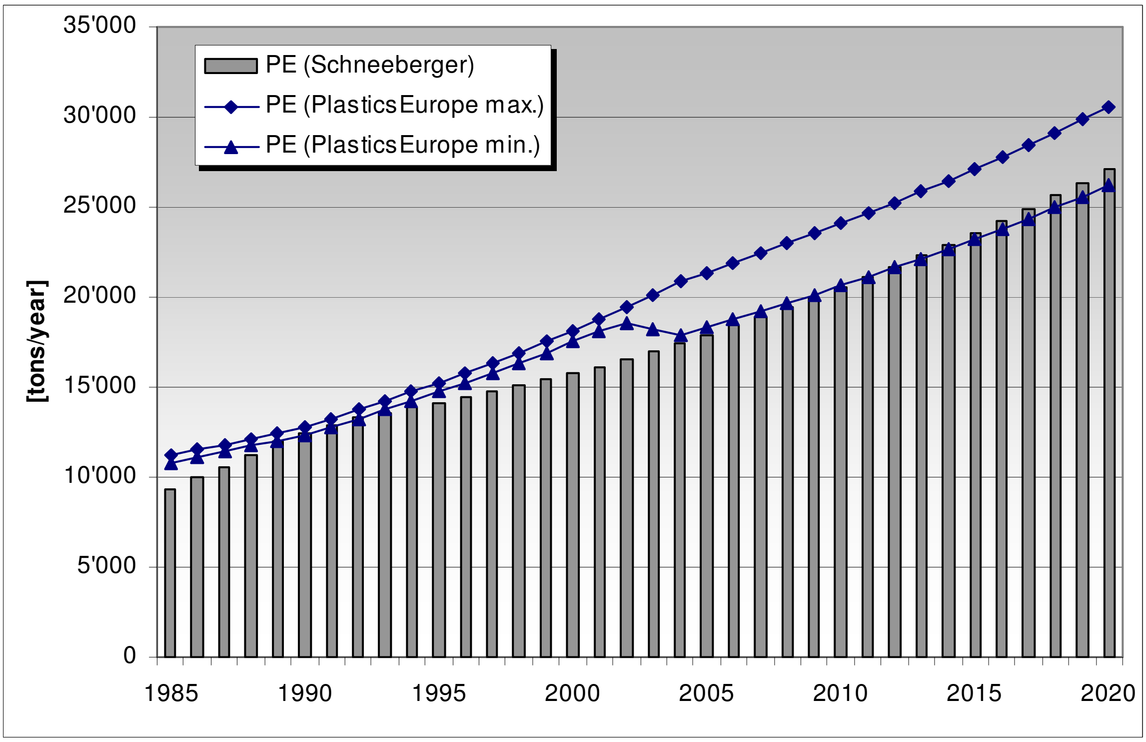


Figure 52 Consumption of polyethylene in the Swiss construction sector

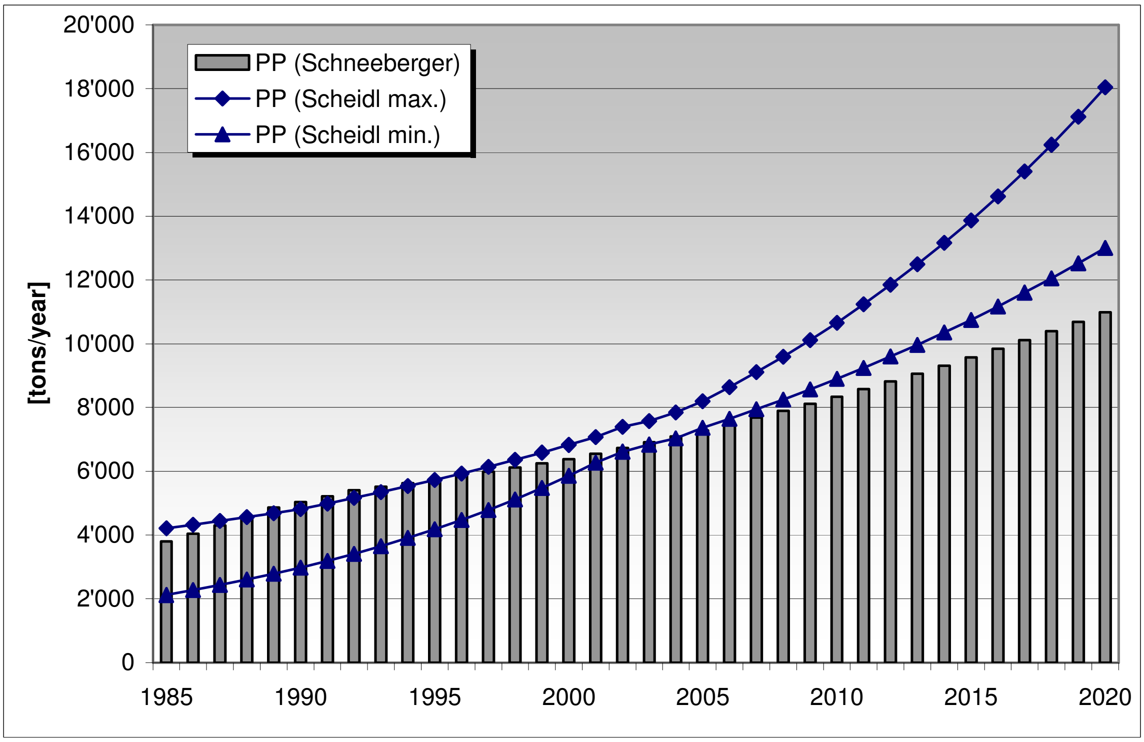


Figure 53 Consumption of polypropylene in the Swiss construction sector

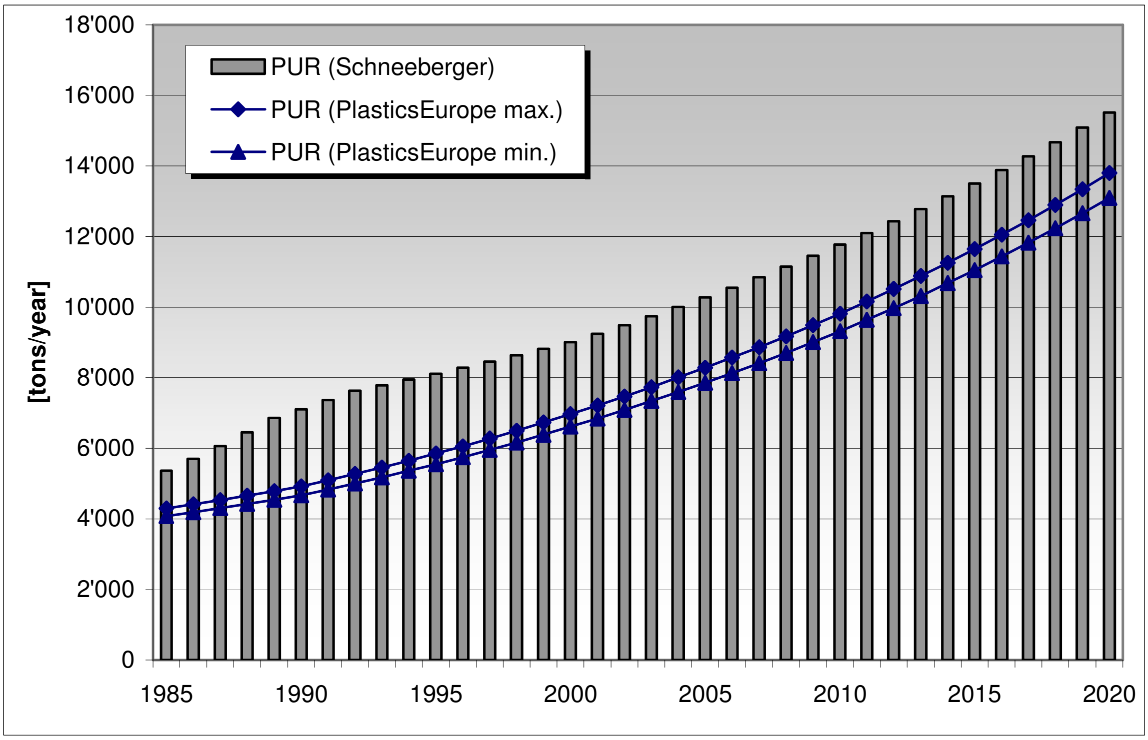


Figure 54 Consumption of polyurethane in the Swiss construction sector

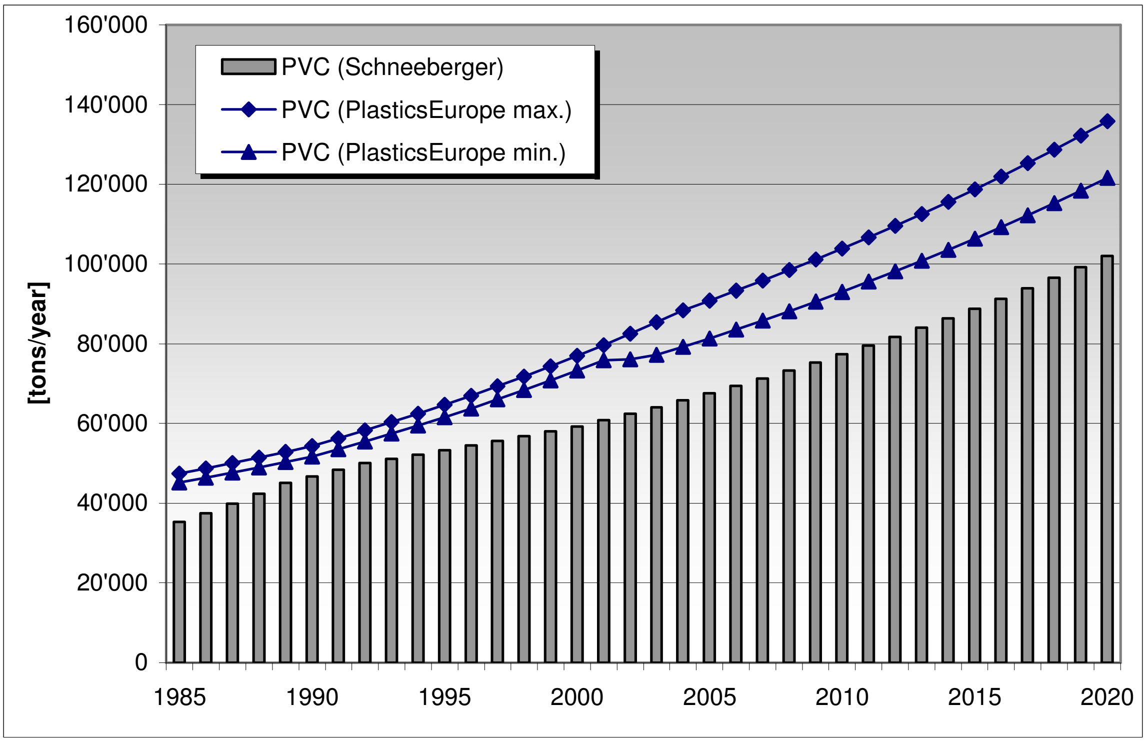


Figure 55 Consumption of polyvinylchloride in the Swiss construction sector

7.4 Environmental fate model

Equations for the estimation of the fate in the atmosphere

$\log K_p = \log K_{OA} + \log f_{OM} - 11.91$ (equation from Falconer and Harner, 2000)

$\Phi = 1 - 1 / (1 + K_p \cdot TSP)$

$k_{dep,dry} = \Phi \cdot v_{dep} / h$

$k_{dep,wet} = [\ \Phi \cdot SC \cdot v_{rain} + (1 - \Phi) \cdot v_{rain} / K_{AW}\ ] / h$

Table 33 Model parameters for estimating the fate of BFRs in the atmosphere

	Fast elimination	Slow elimination	Source
Scavenging coefficient (SC)	370,000	200,000	Wania and Mackay (1995), ter Schure et al. (2004b)
Total suspended particles (TSP)	50 µg/m^3	20 µg/m^3	BAFU (2007)
Organic fraction of aerosol particles (f_{OM})	0.246	0.155	Hüglin et al. (2005)
Height of atmospheric boundary layer (h)	1000 m		–
Dry particle deposition velocity (v_{dep})	10 m/h		Hirai and Sakai (2004)
Precipitation (v_{rain})	1.66E–04 m/h		Schädler and Weingartner (2002)

Table 34 Partitioning data estimated by least-square adjustment

	DecaBDE	HBCD *	BDE-47
$\log K_{OW}$	9.97	7.84	6.39
$\log K_{AW}$	–6.29	–3.56	–3.35
$\log K_{OA}$	18.25	11.40	10.44

*: Own calculation with *Least-Squares Adjustment Spreadsheet v1.1* (Schenker et al., 2005) using updated water solubility data.

7.5 Substance flows in the anthroposphere

In the following, the substance flows in the anthroposphere, including imports, exports and emissions to the environment are illustrated for the year 2000. The thickness of the arrows is relative to the substance flows. Note that it can not be compared among different diagrams.

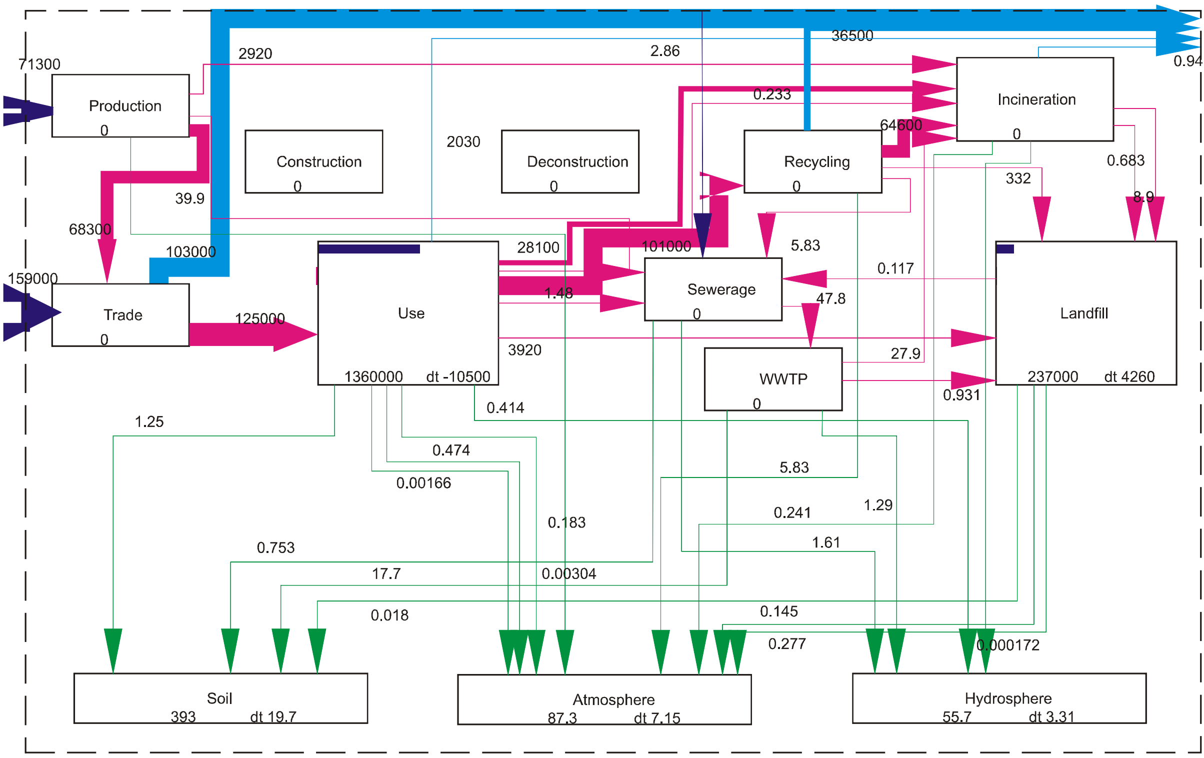


Figure 56 DecaBDE substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *E&E*.

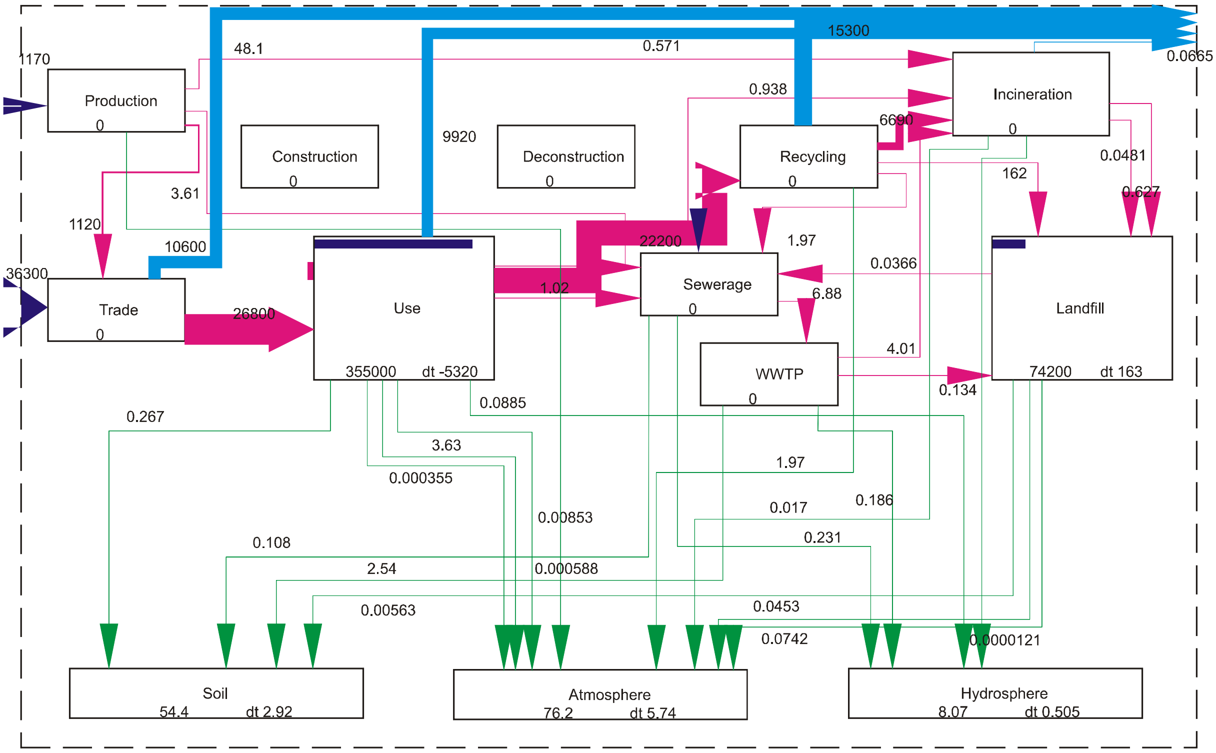


Figure 57 DecaBDE substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *transport*

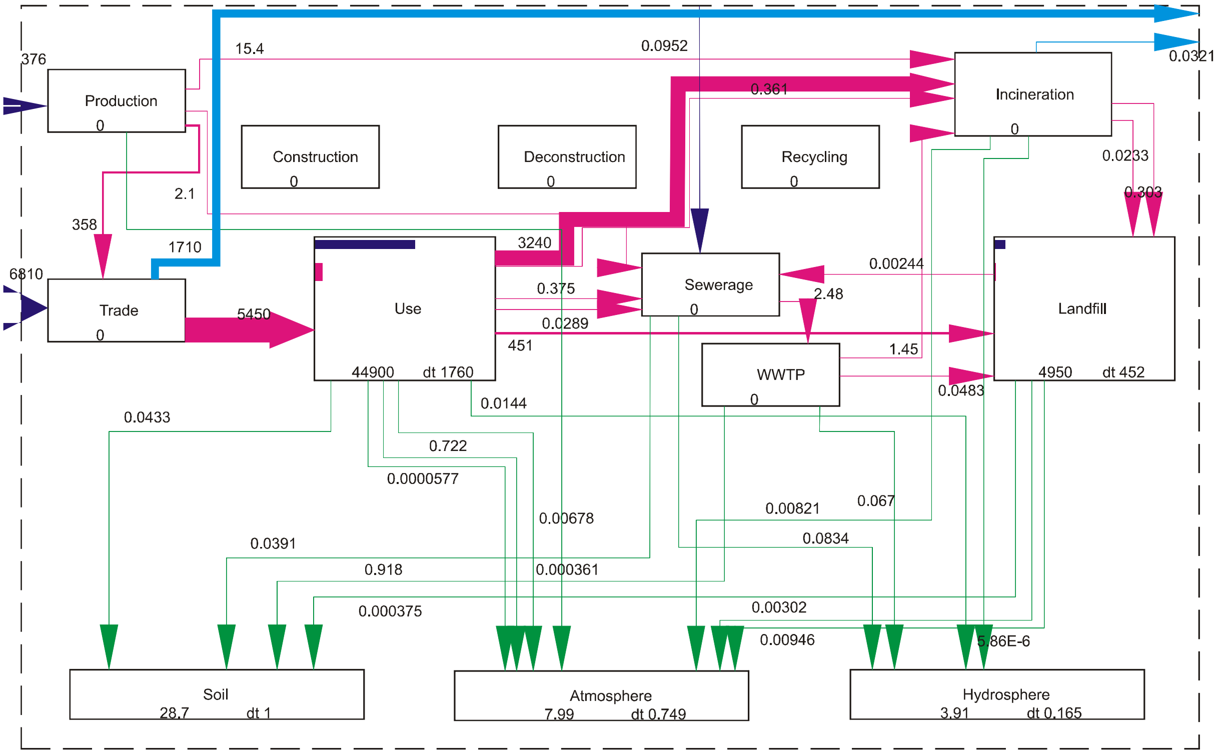


Figure 58 DecaBDE substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *textile/furniture*

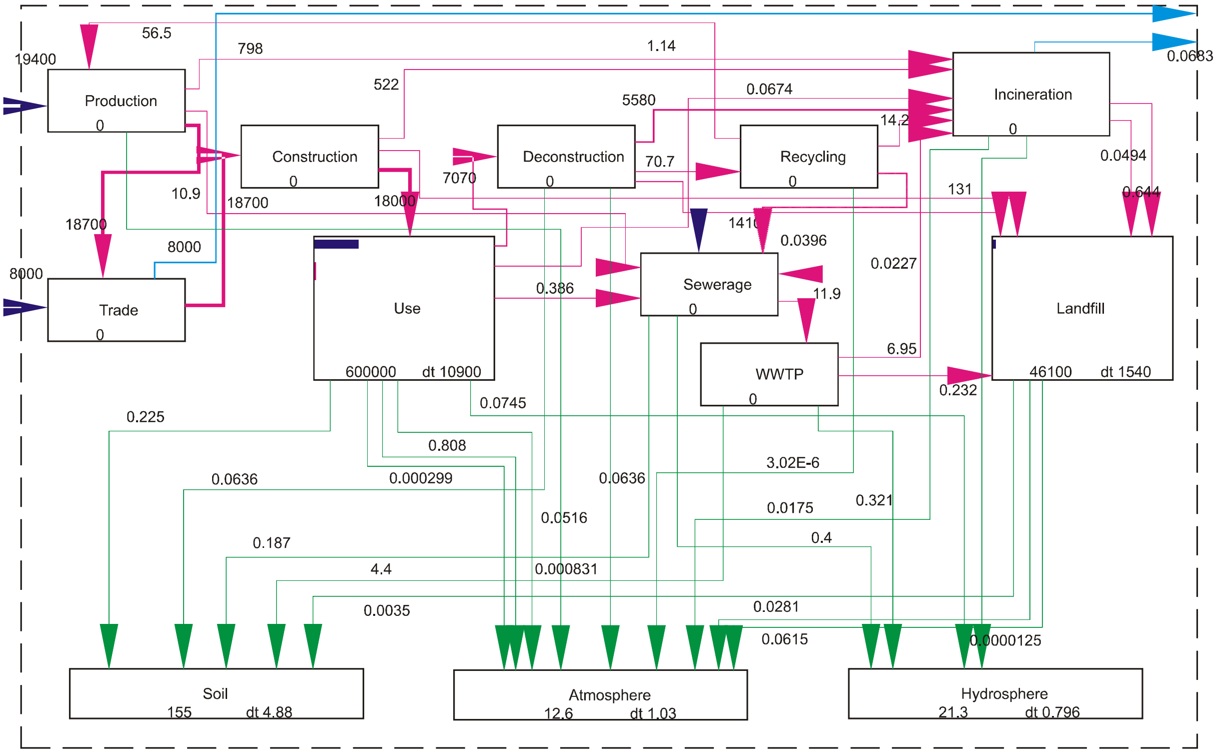


Figure 59 DecaBDE substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *construction*

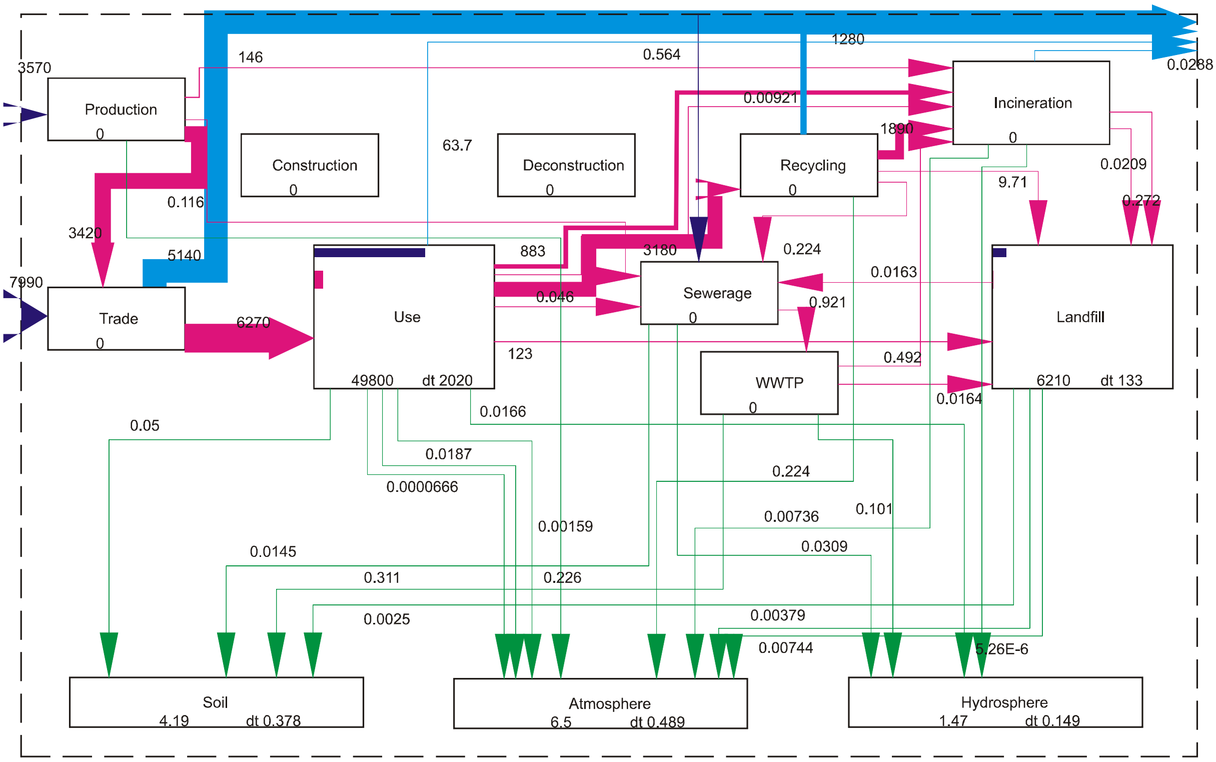


Figure 60 HBCD substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *E&E*

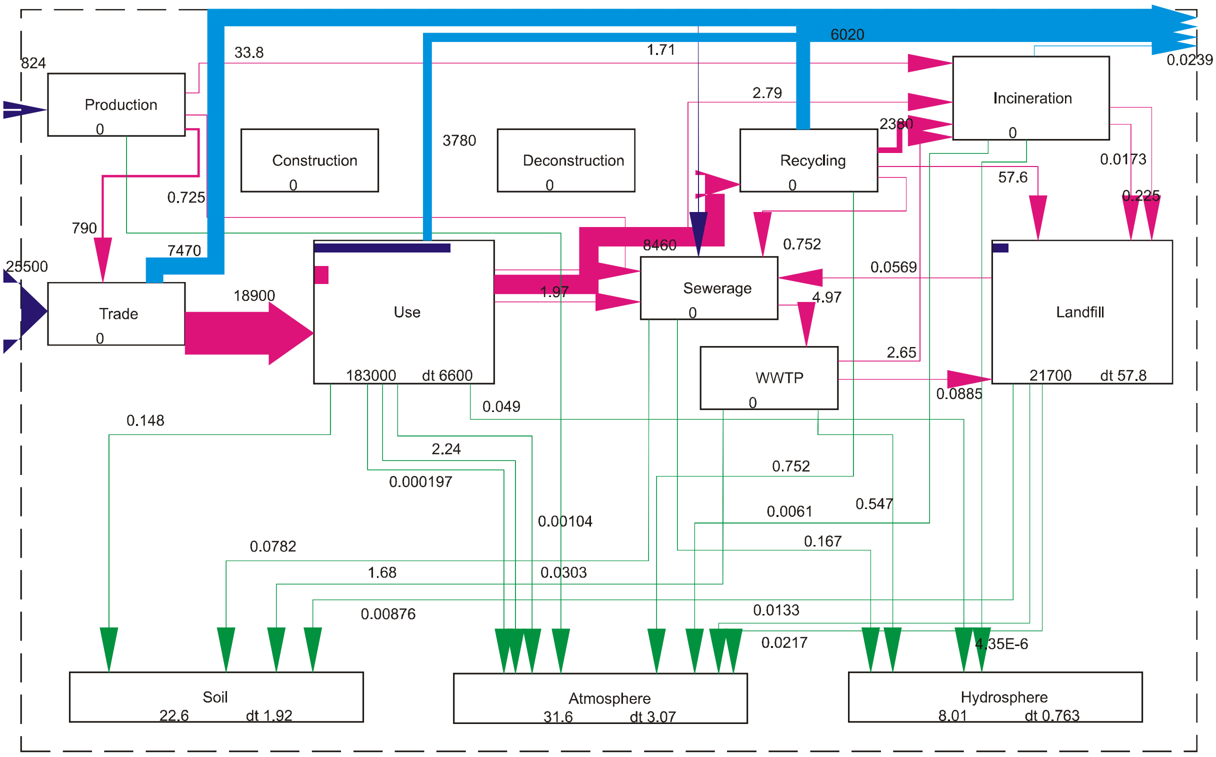


Figure 61 HBCD substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *transport*

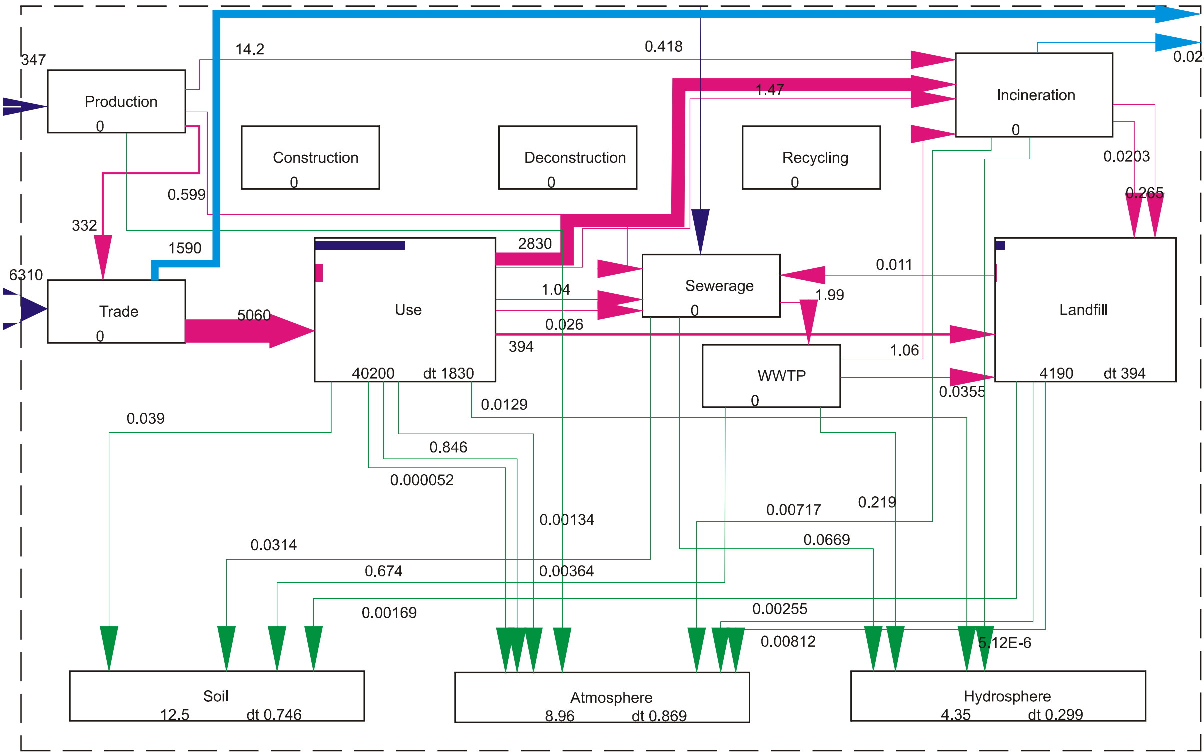


Figure 62 HBCD substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *textile/furniture*

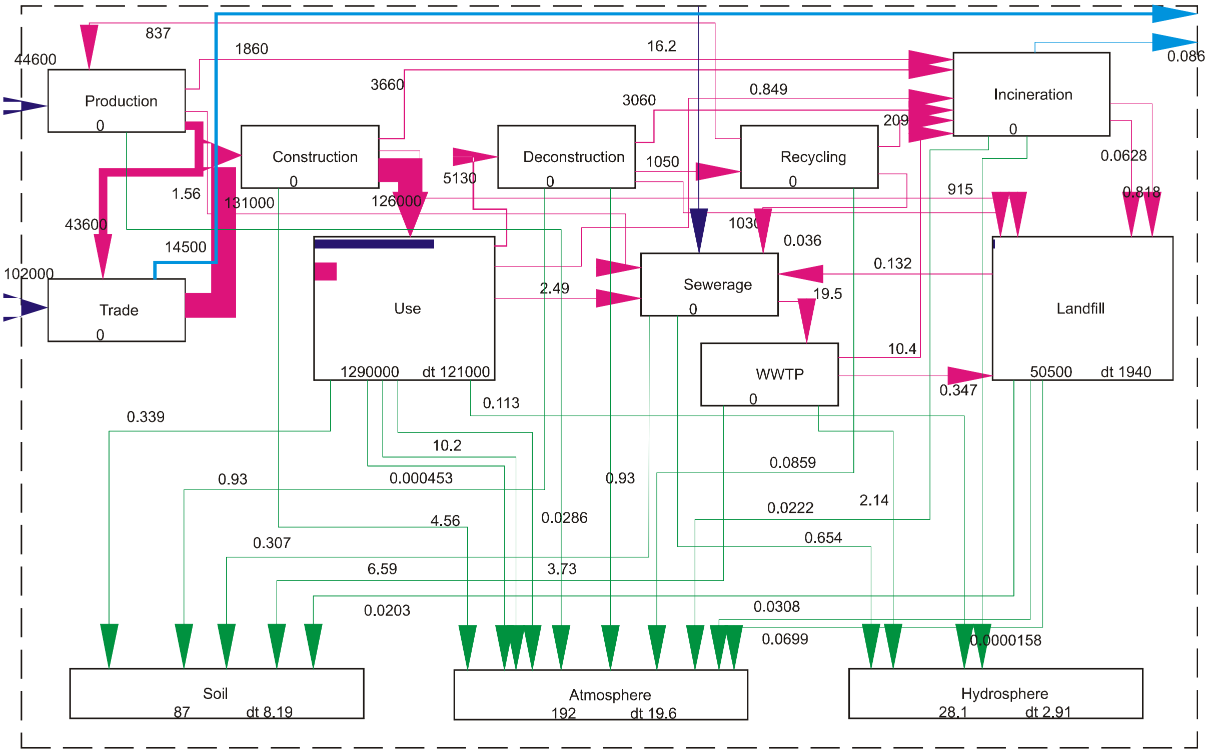


Figure 63 HBCD substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *construction*

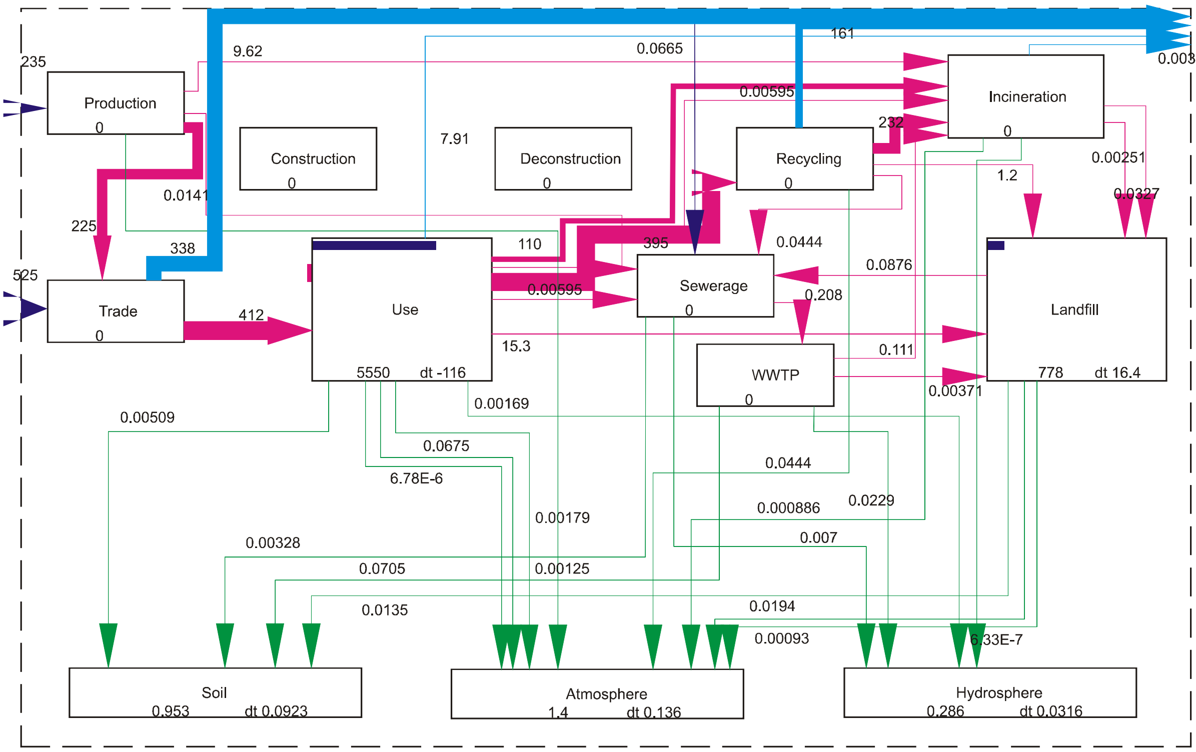


Figure 64 BDE-47 substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *E&E*

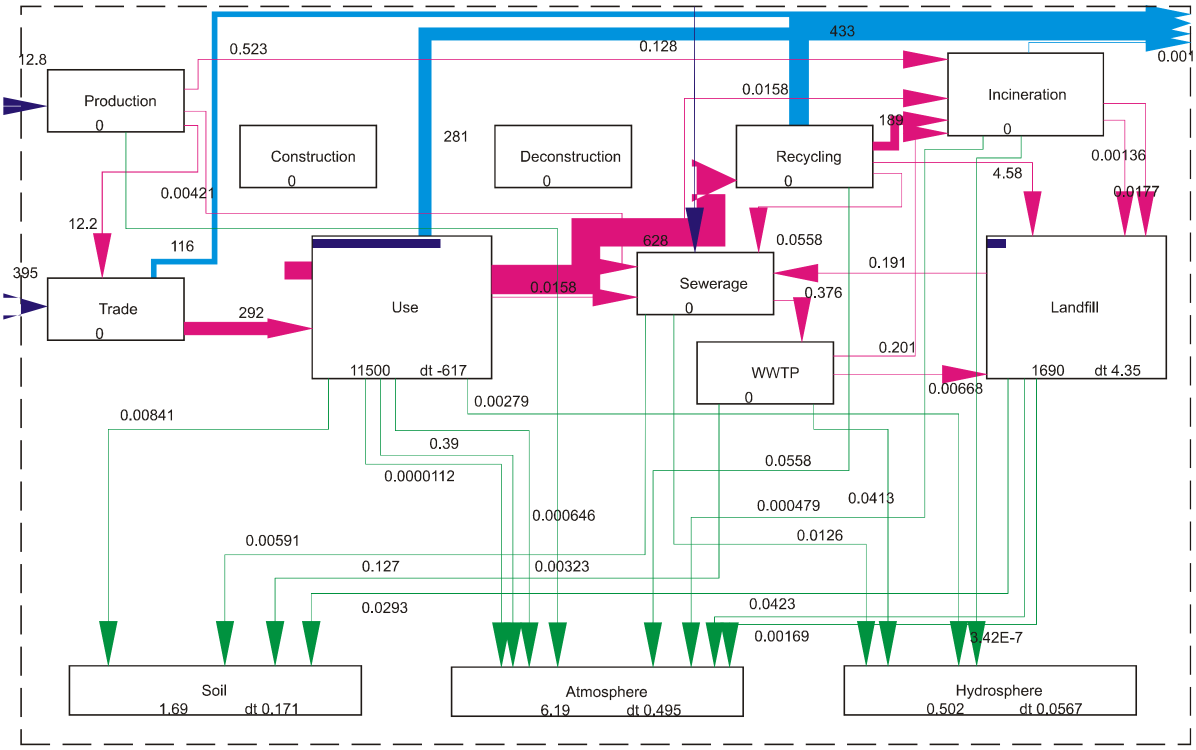


Figure 65 BDE-47 substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *transport*

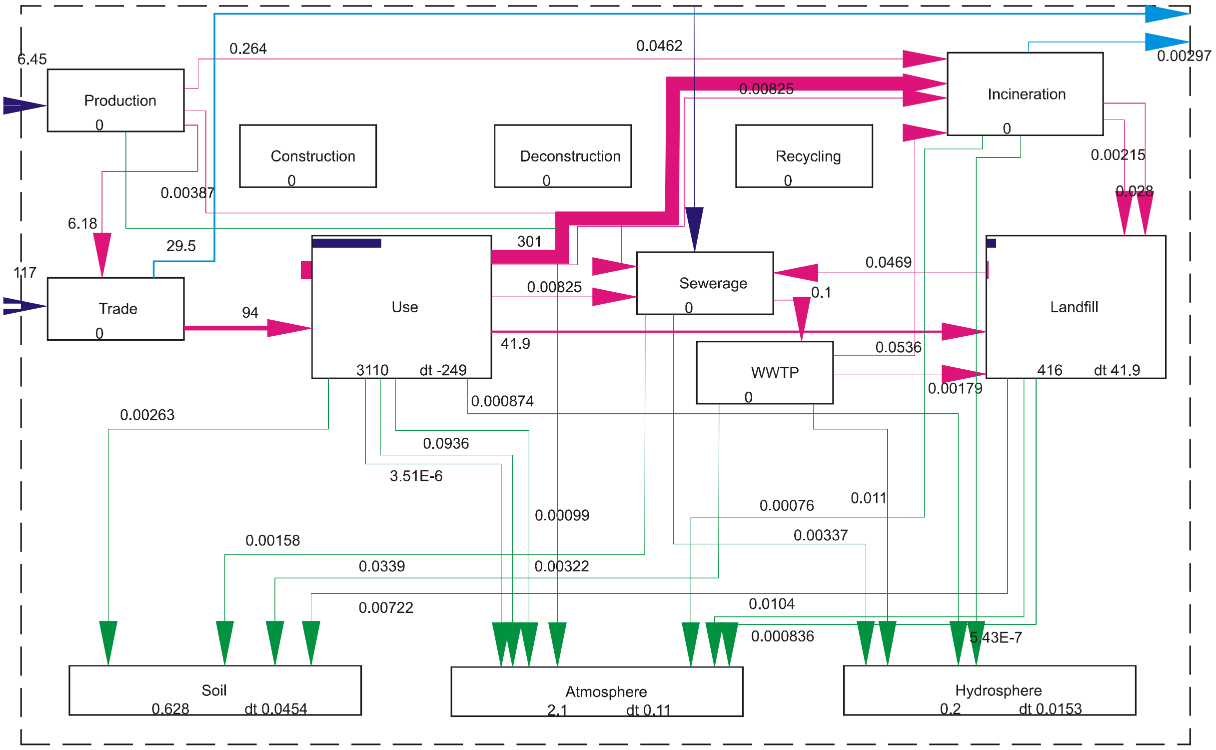


Figure 66 BDE-47 substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *textile/furniture*

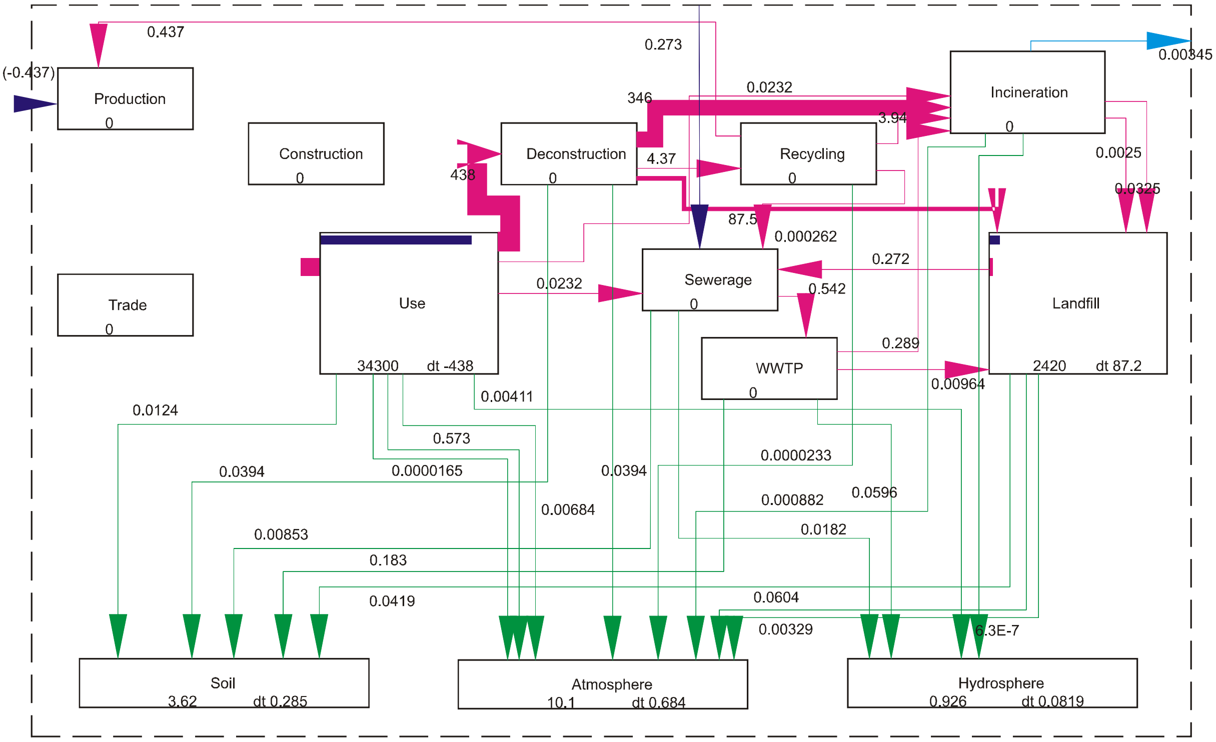


Figure 67 BDE-47 substance flows (kg/year) and stocks (kg) in the anthroposphere in the application area *construction*

7.6 Temporal trend results

7.6.1 Flows to use, waste management, export, sewerage and WWTP

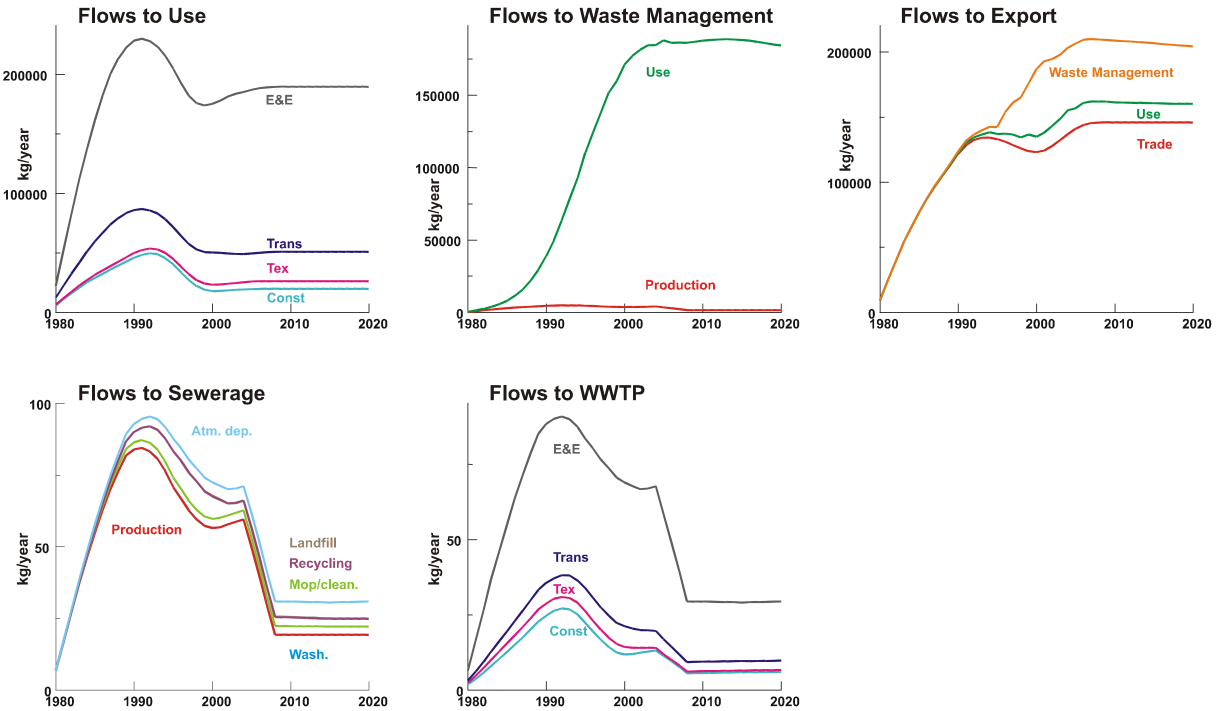


Figure 68 DecaBDE substance flows (sum of all application areas)

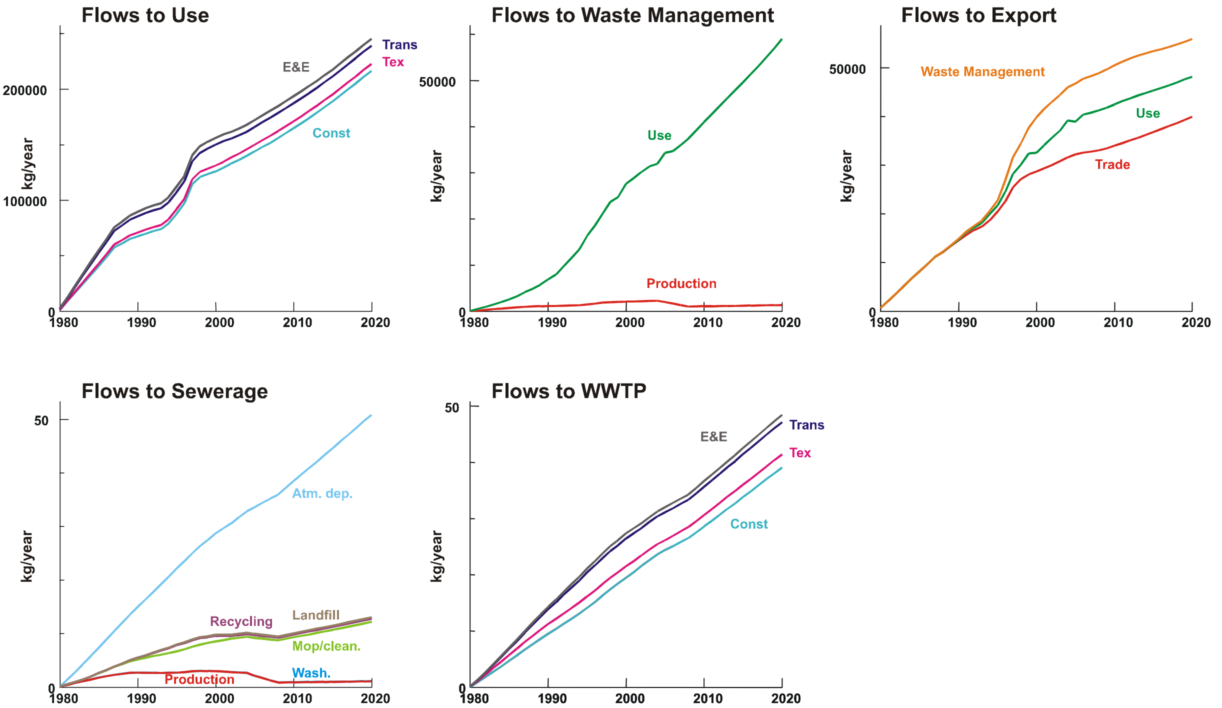


Figure 69 HBCD substance flows (sum of all application areas)

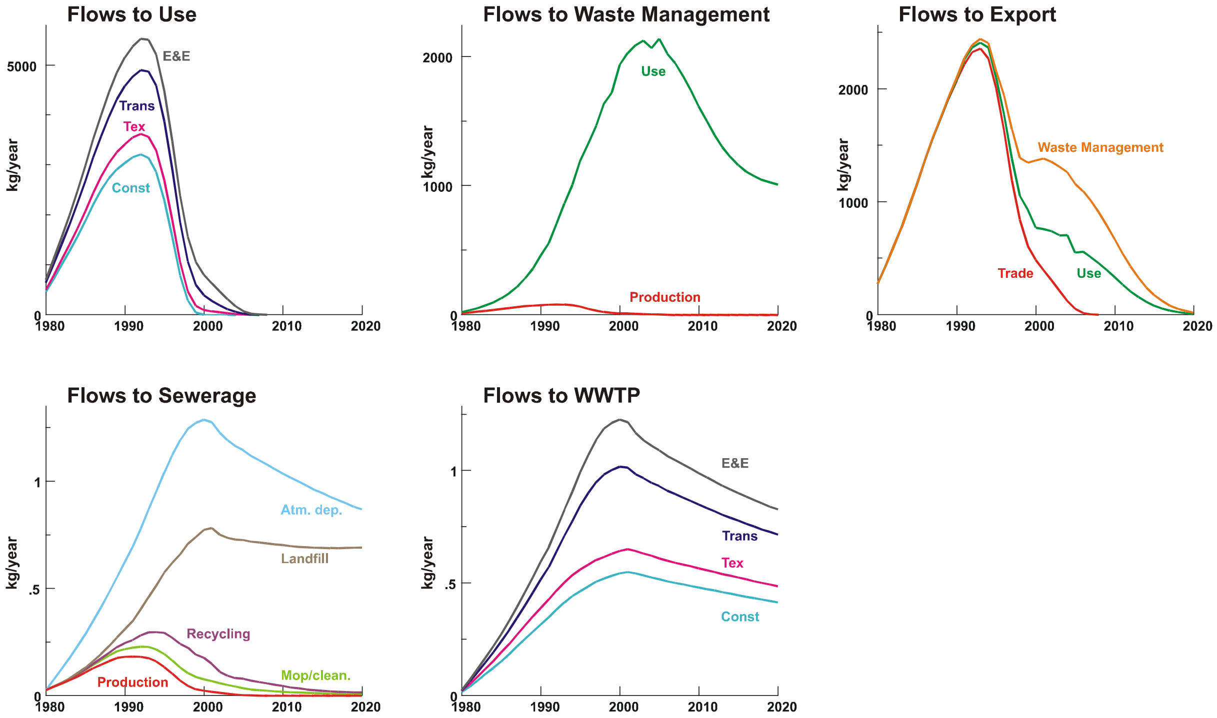


Figure 70 BDE-47 substance flows (sum of all application areas)

7.6.2 Flows to waste management split up by application areas

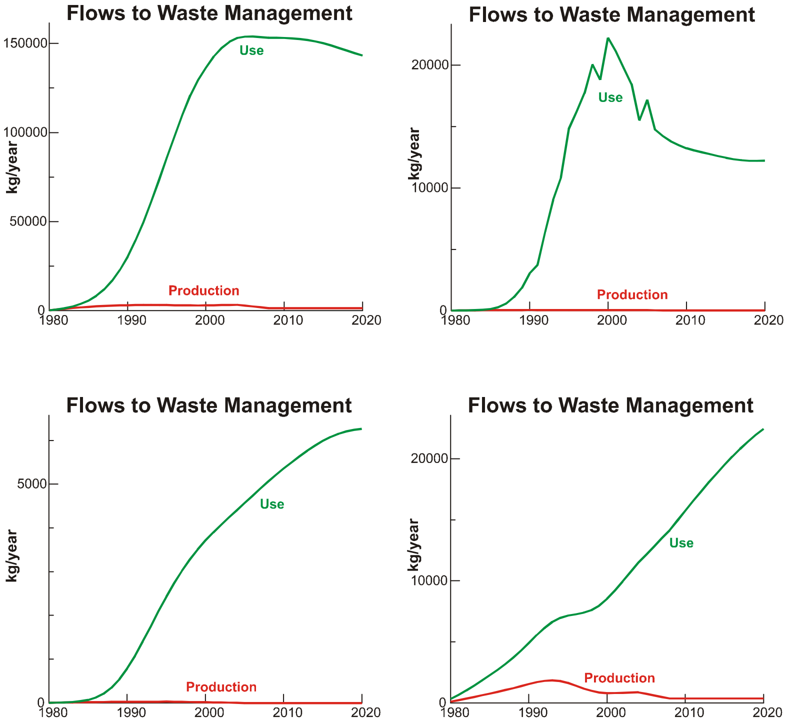


Figure 71 DecaBDE substance flows from production and use to waste management: (a) *E&E*, (b) *transport*, (c) *textile/furniture*, (d) *construction*

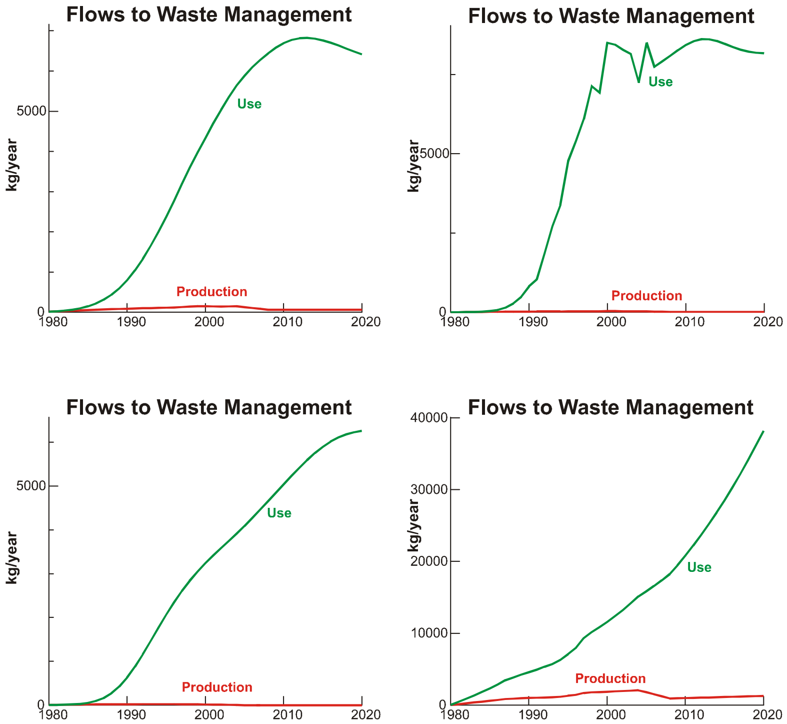


Figure 72 HBCD substance flows from production and use to waste management: (a) *E&E*, (b) *transport*, (c) *textile/furniture*, (d) *construction*

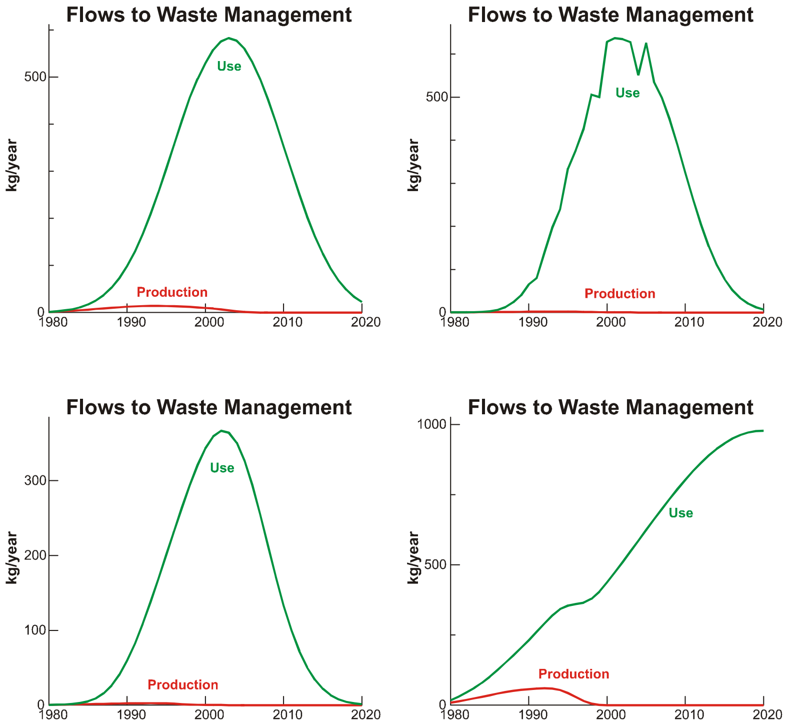


Figure 73 BDE-47 substance flows from production and use to waste management: (a) *E&E*, (b) *transport*, (c) *textile/furniture*, (d) *construction*

7.6.3 Emissions to environment split up by application areas

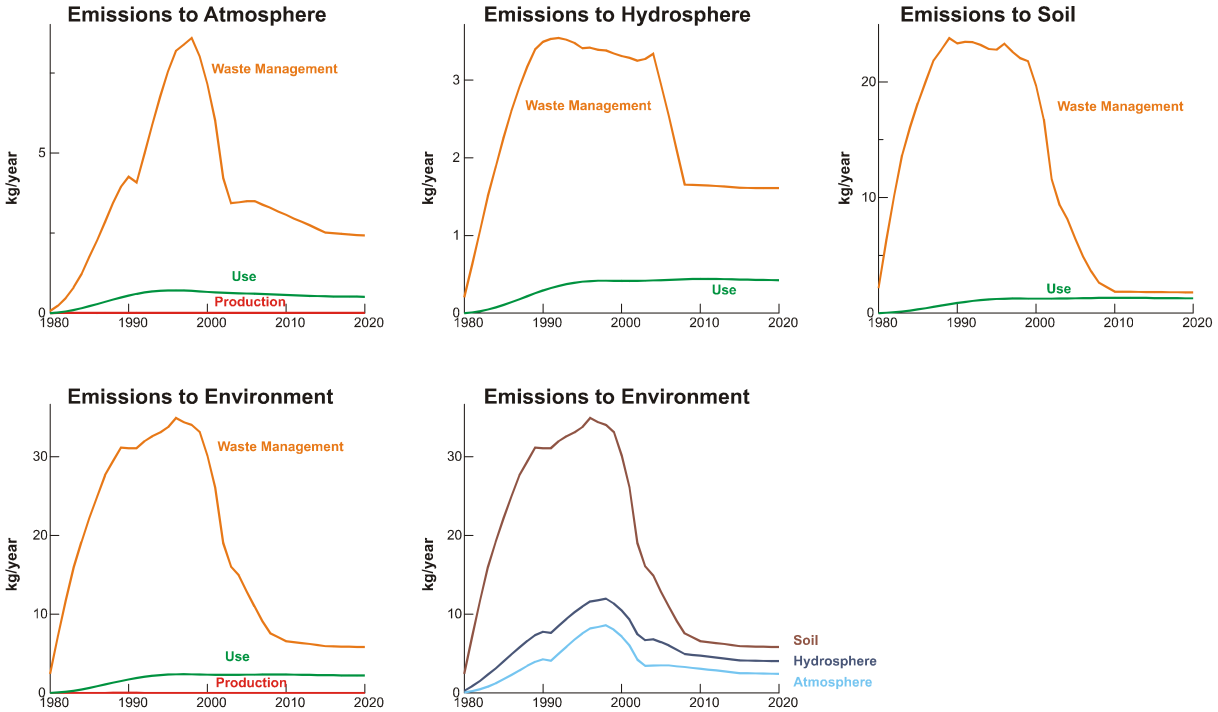


Figure 74 DecaBDE emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *E&E*

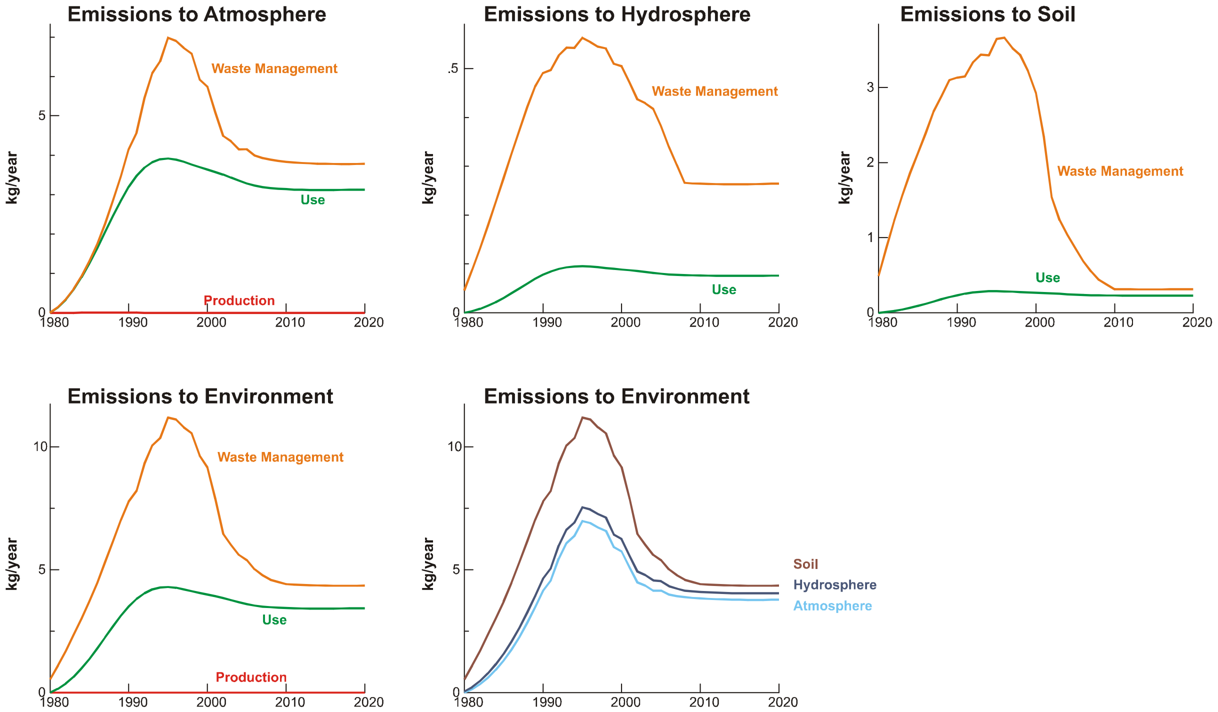


Figure 75 DecaBDE emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *transport*

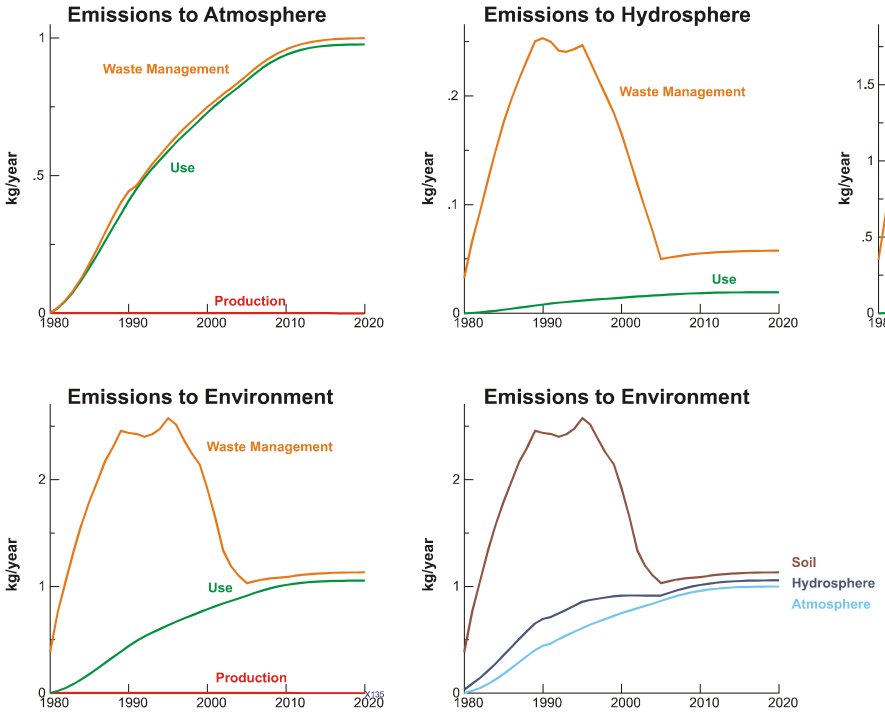


Figure 76 DecaBDE emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *textile/furniture*

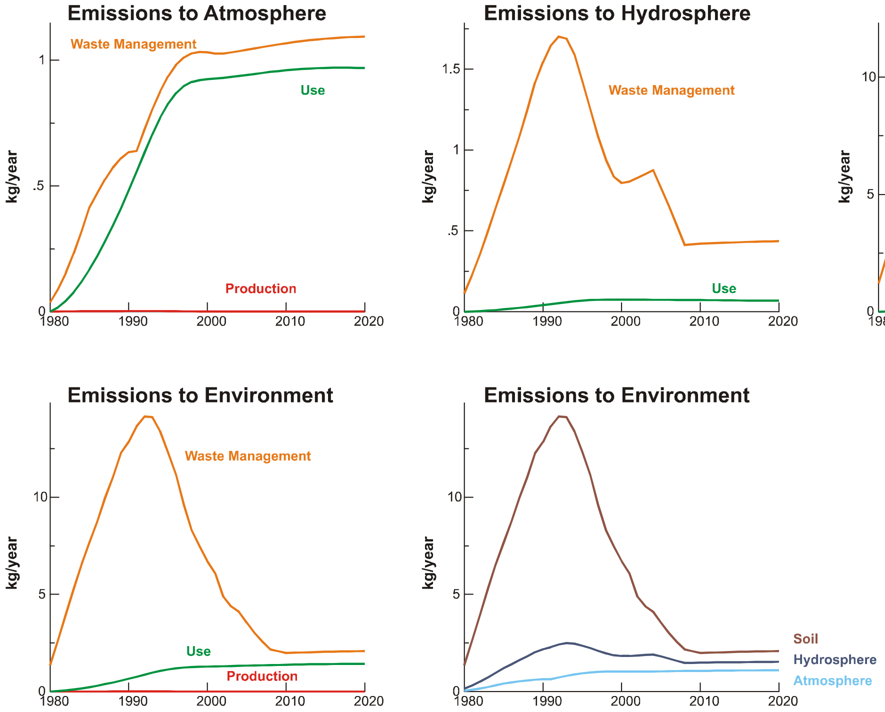


Figure 77 DecaBDE emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *construction*

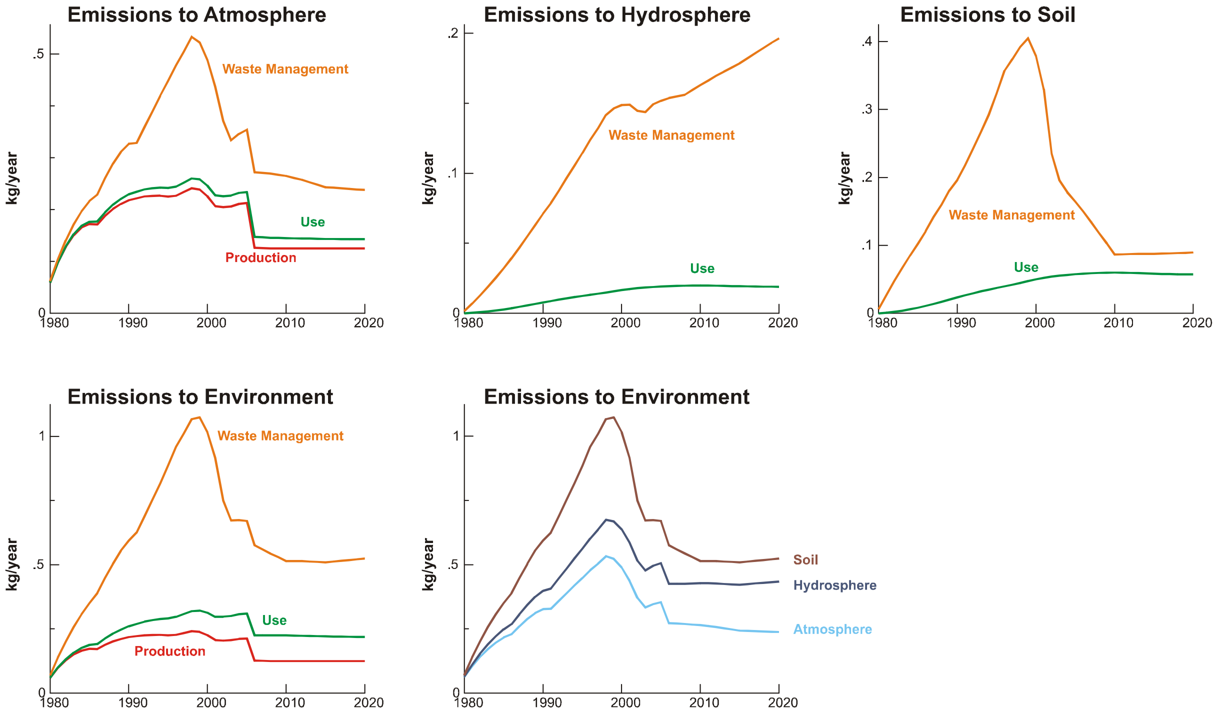


Figure 78 HBCD emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *E&E*

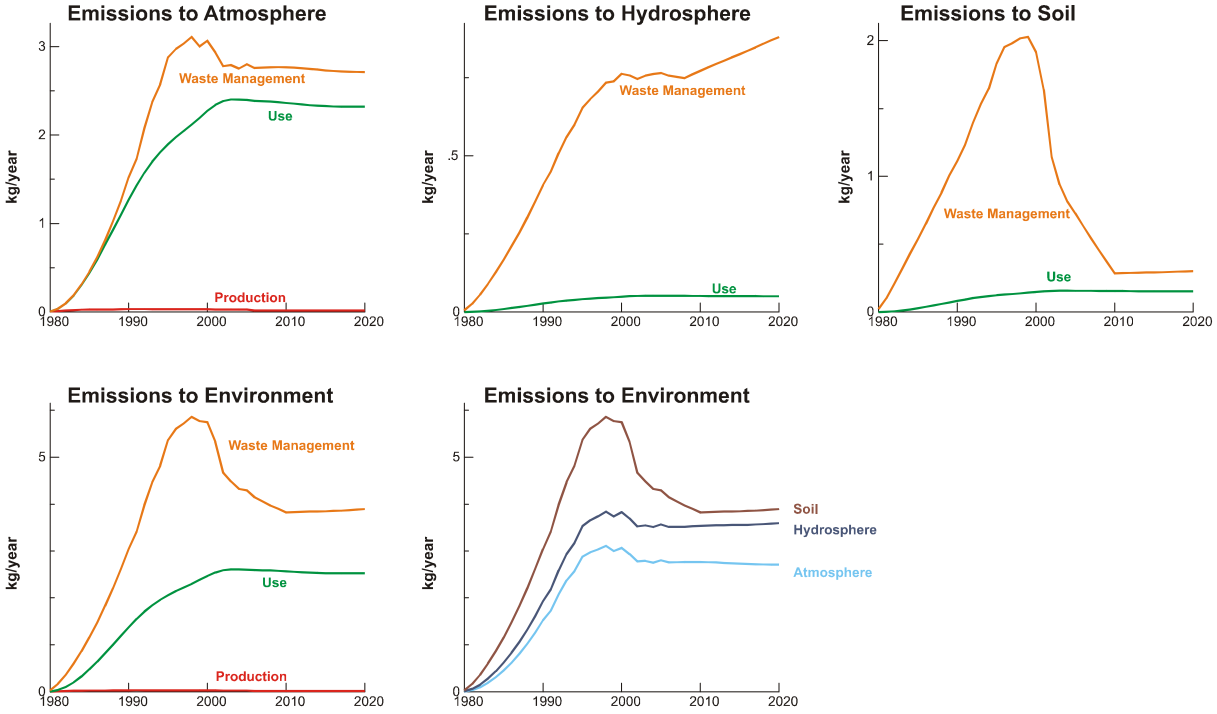


Figure 79 HBCD emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *transport*

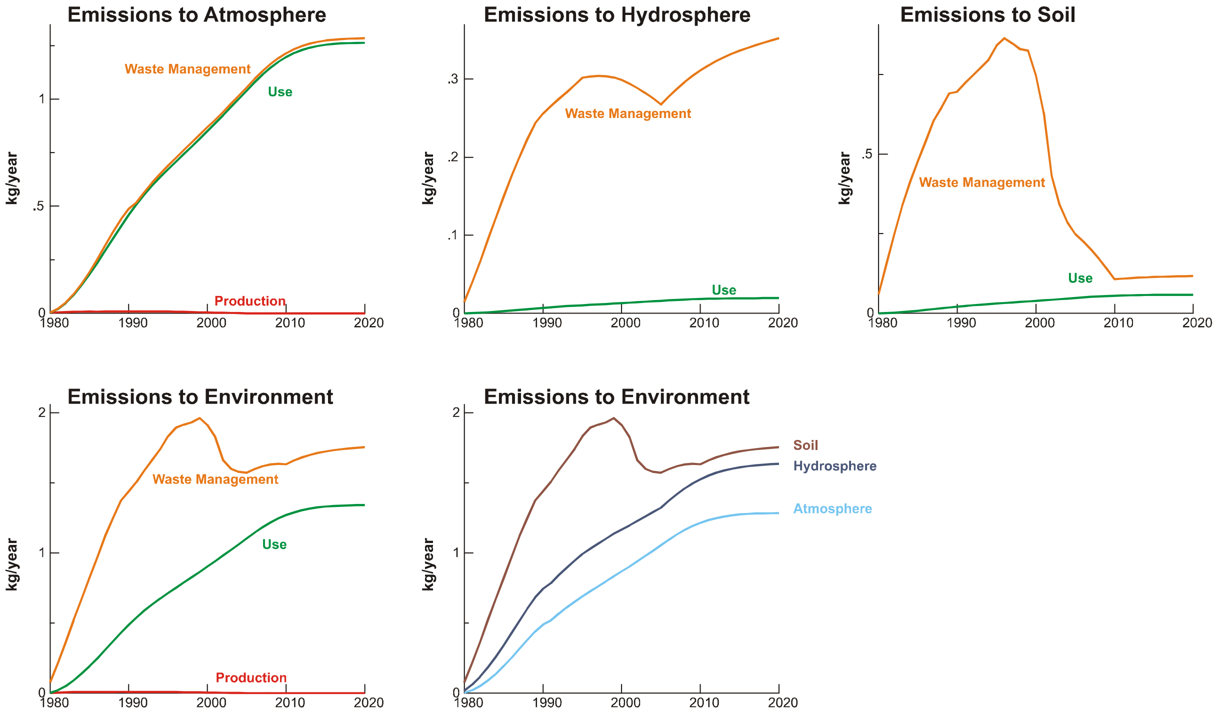


Figure 80 HBCD emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *textile/furniture*

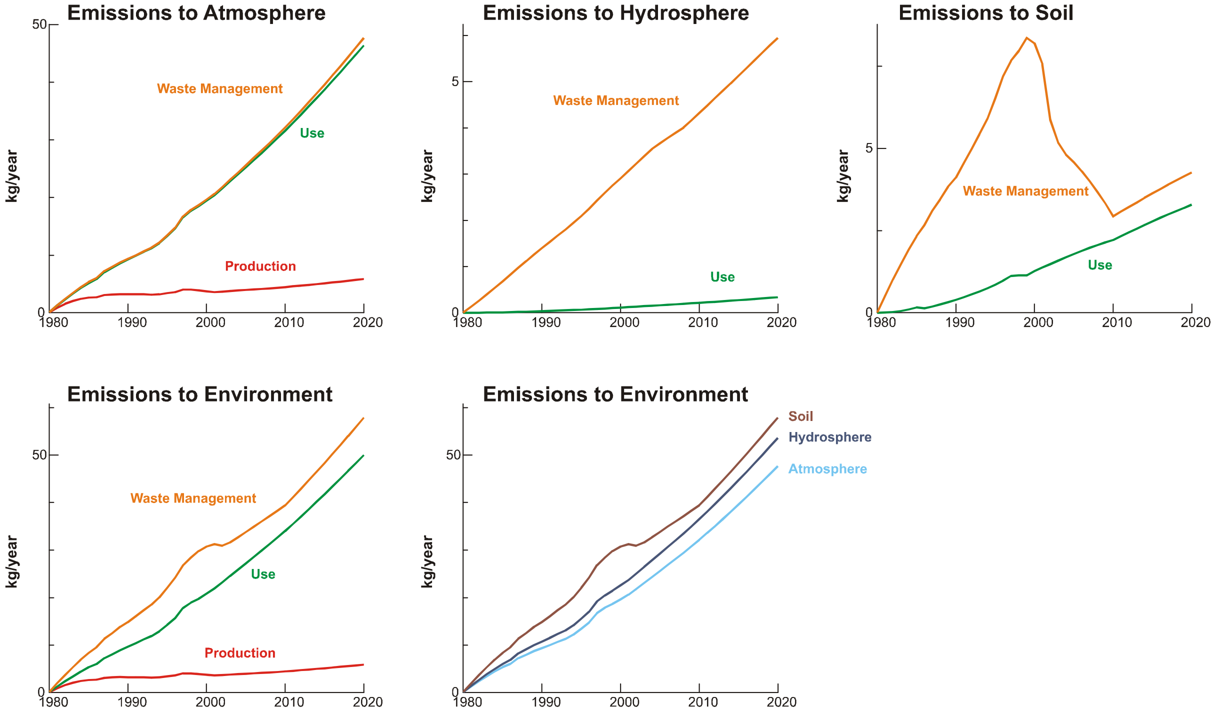


Figure 81 HBCD emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *construction*

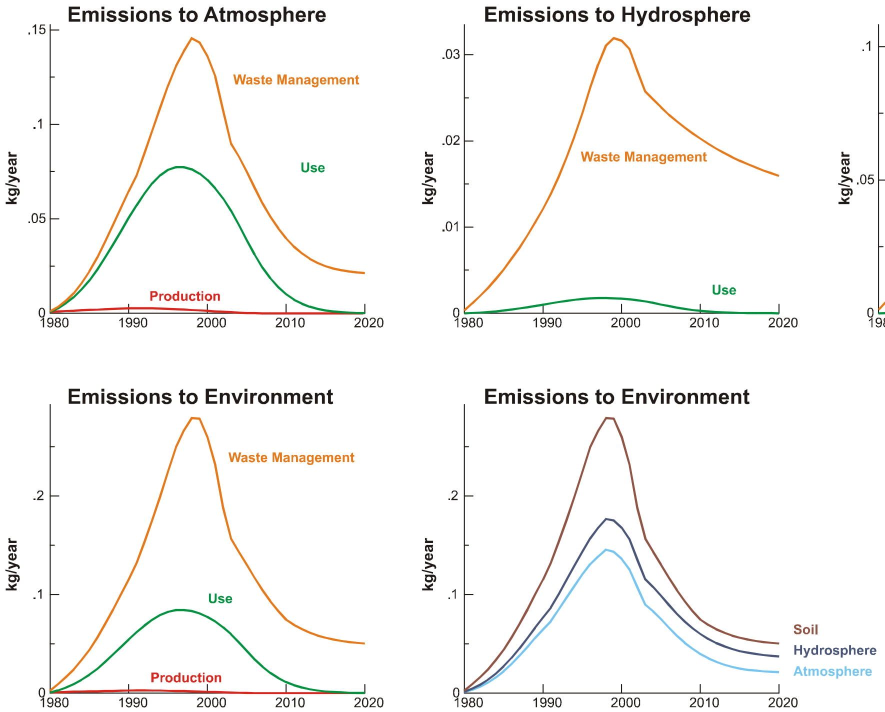


Figure 82 BDE-47 emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *E&E*

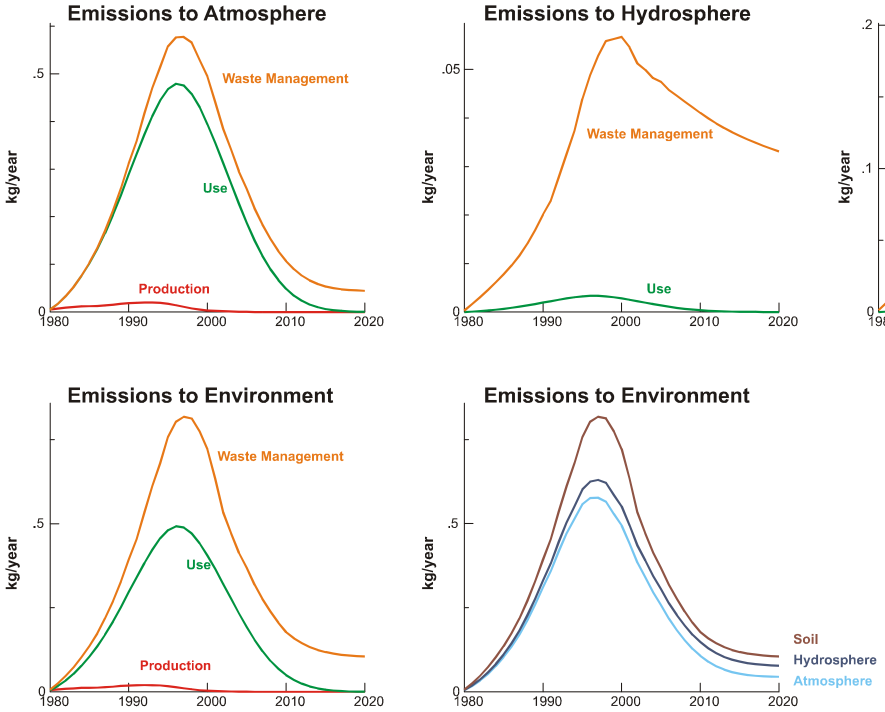


Figure 83 BDE-47 emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *transport*

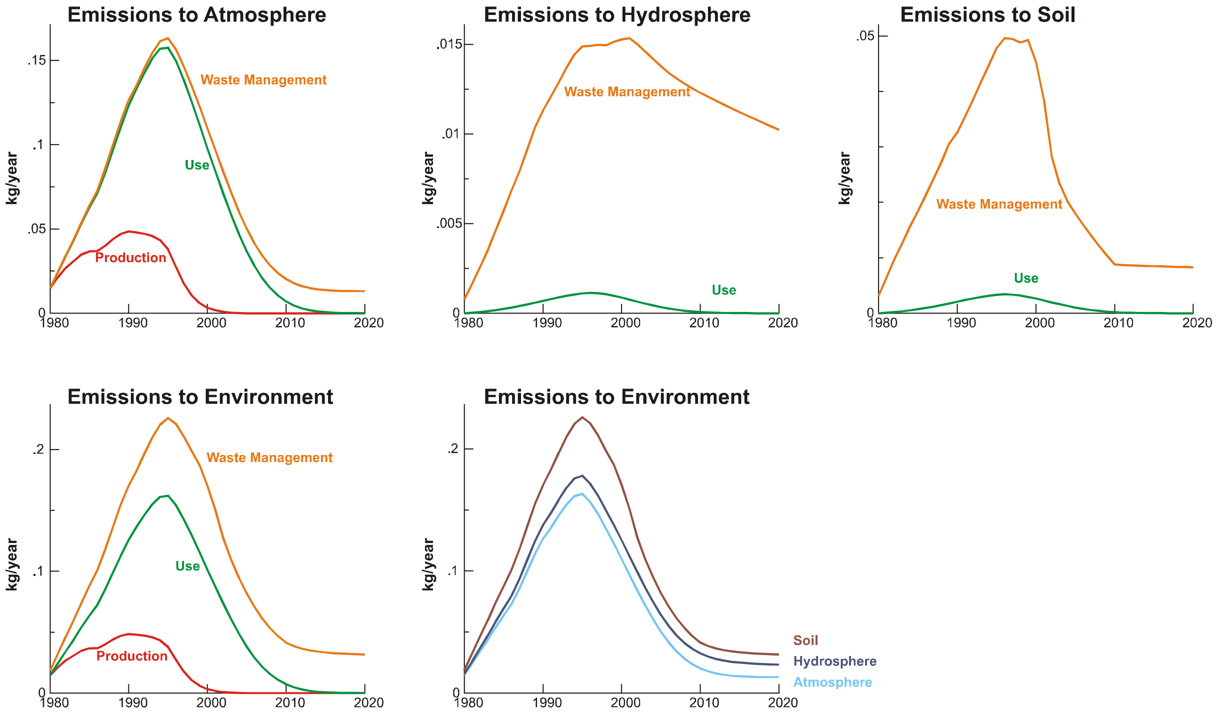


Figure 84 BDE-47 emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *textile/furniture*

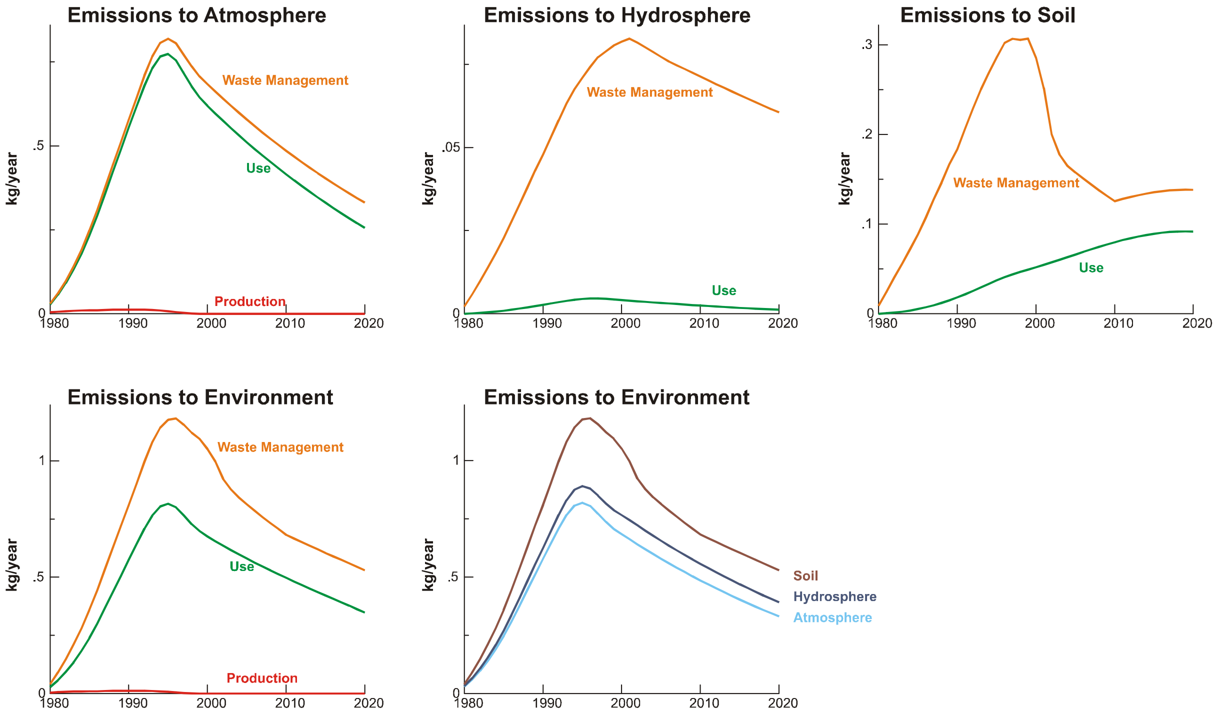


Figure 85 BDE-47 emissions to atmosphere, hydrosphere, soil and total emissions to the environment: *construction*

7.7 Temporal trend results – influence of future and past regulatory and technical measures

7.7.1 Ban on the usage of DecaBDE

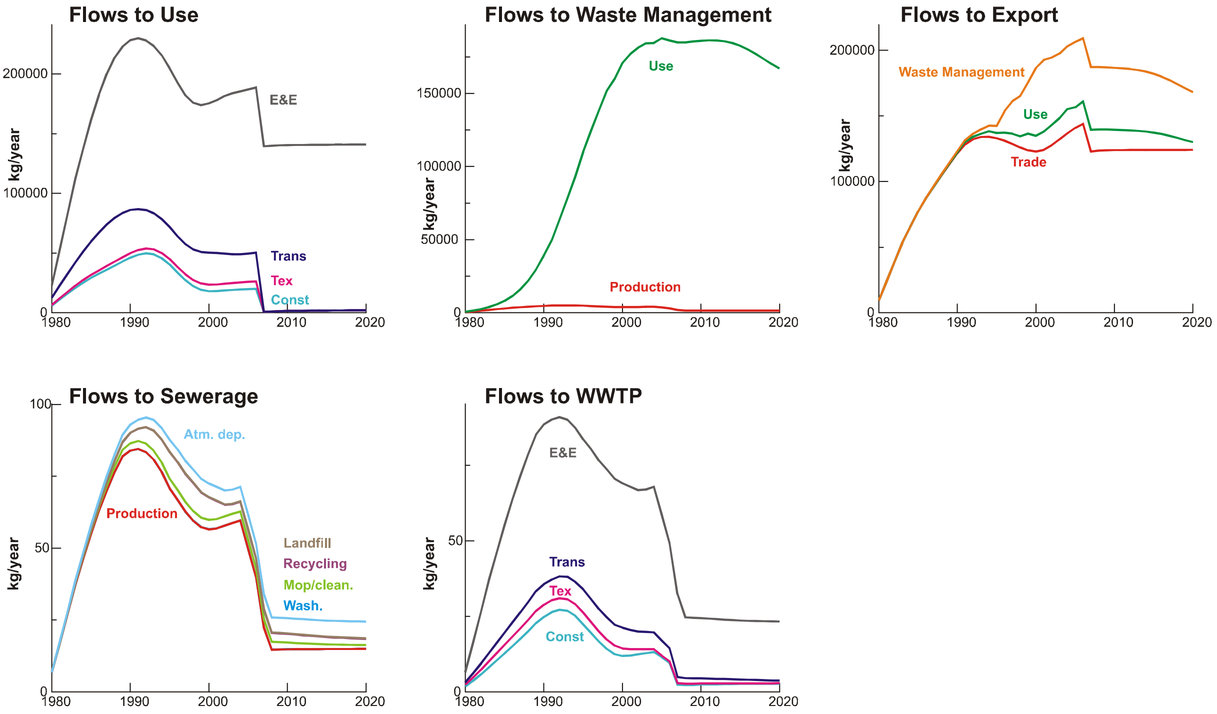


Figure 86 DecaBDE substance flows: scenario of a restriction by 2007 of the substance in all application areas except *E&E*

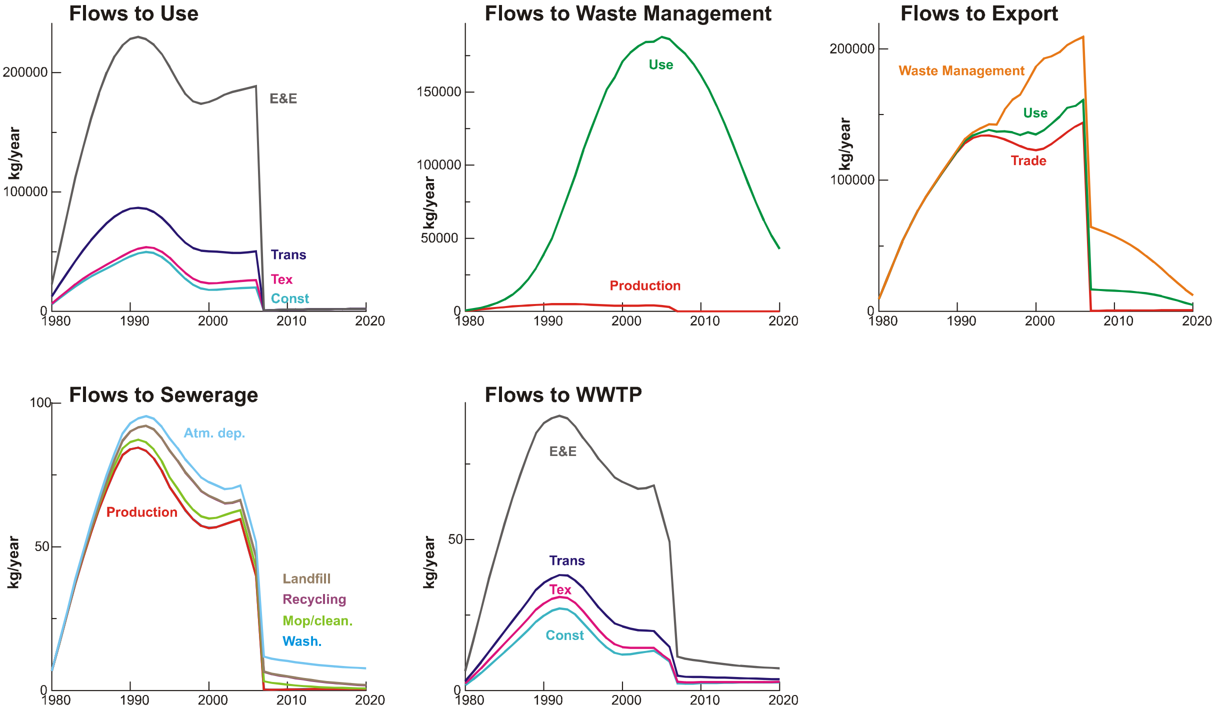


Figure 87 DecaBDE substance flows: scenario of a restriction by 2007 of the substance in all application areas

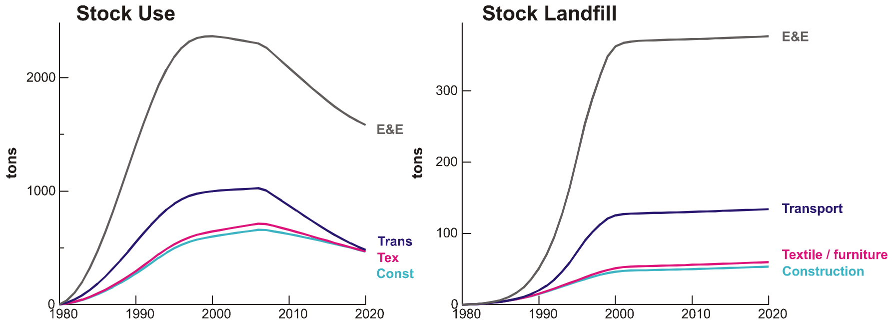


Figure 88 DecaBDE stocks in Use and Landfill: scenario of a restriction by 2007 of the substance in all application areas except *E&E*

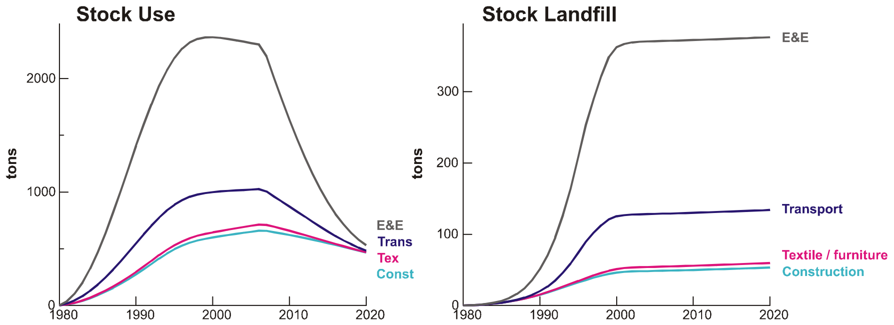


Figure 89 DecaBDE stocks in Use and Landfill: scenario of a restriction by 2007 of the substance in all application areas

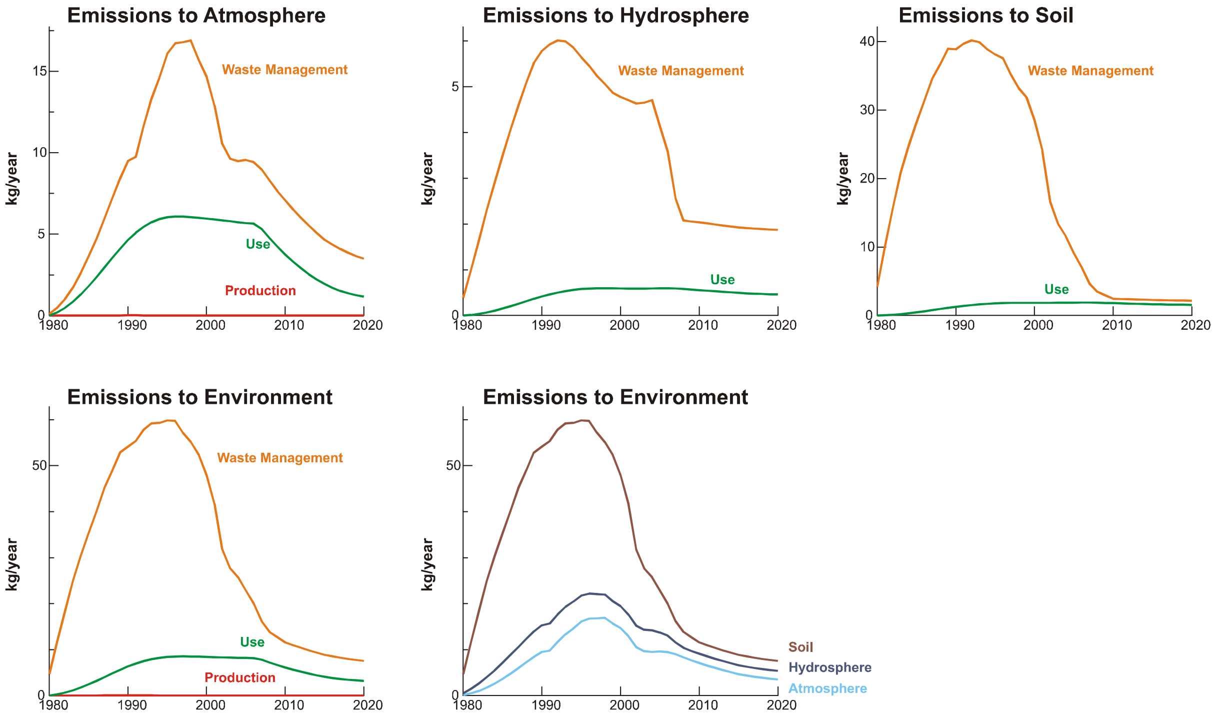


Figure 90 DecaBDE emissions to atmosphere, hydrosphere, soil and total emissions to the environment: scenario of a restriction by 2007 of the substance in all application areas except *E&E*

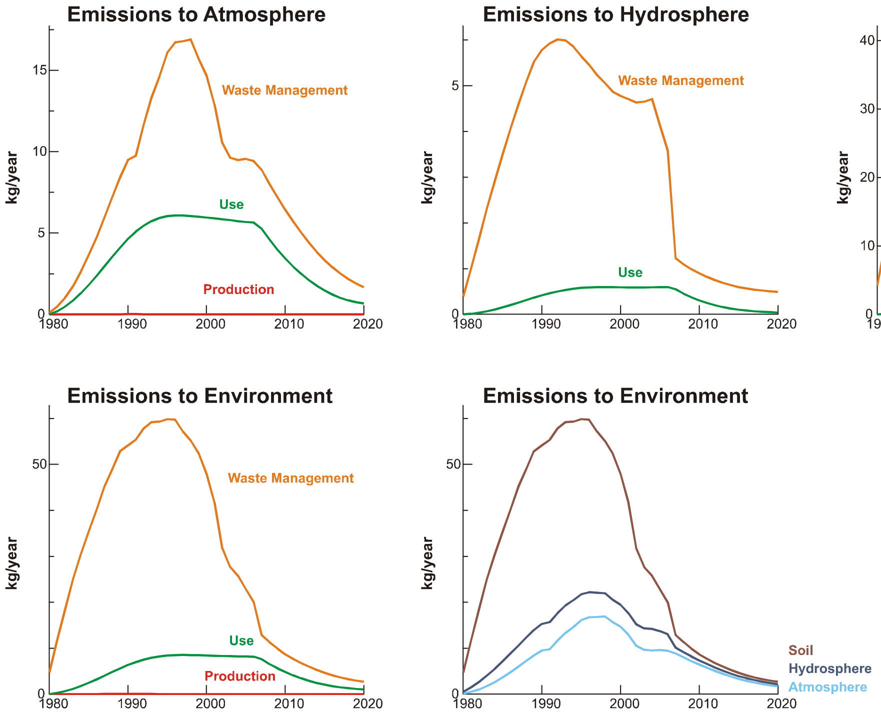


Figure 91 DecaBDE emissions to atmosphere, hydrosphere, soil and total emissions to the environment: scenario of a restriction by 2007 of the substance in all application areas

7.7.2 Influence of an invariant waste management on emissions of DecaBDE

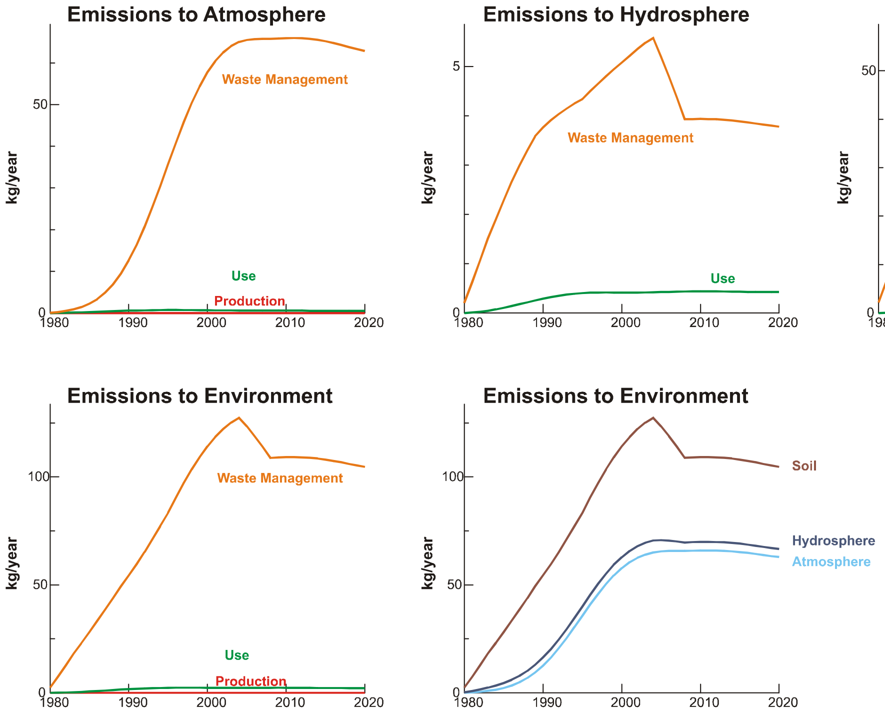


Figure 92 DecaBDE emissions from *E&E* to atmosphere, hydrosphere, soil and total emissions to the environment: scenario of an invariant waste management

7.7.3 Ban on the usage of HBCD in construction materials

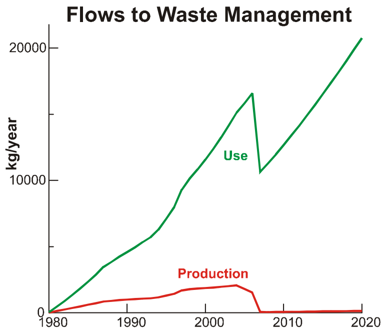


Figure 93 HBCD substance flows from production and use to waste management: scenario of a restriction by 2007 of the substance in construction materials (only application area *construction* shown)

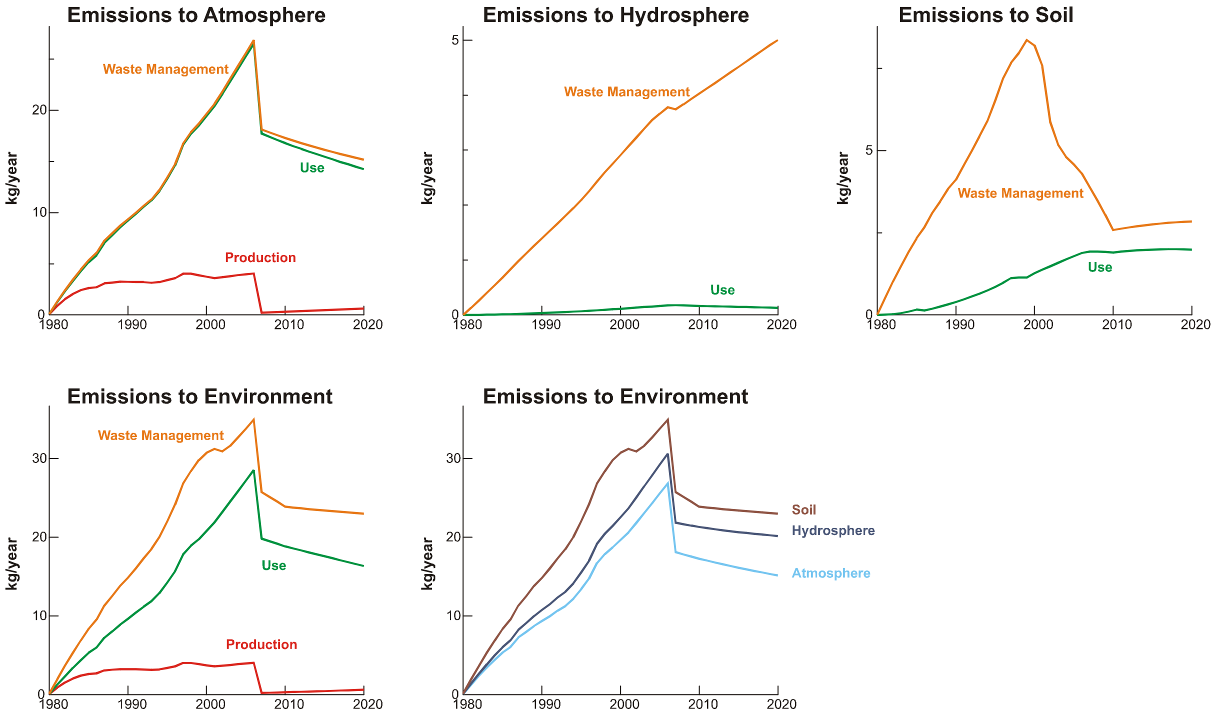
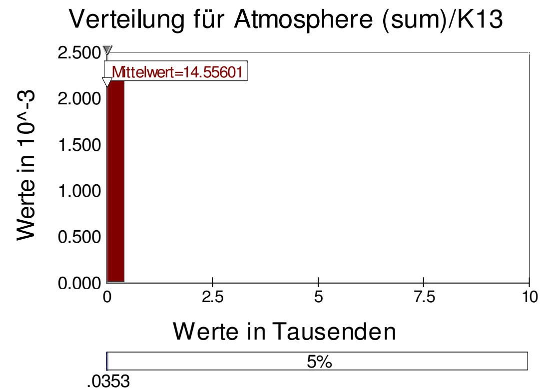
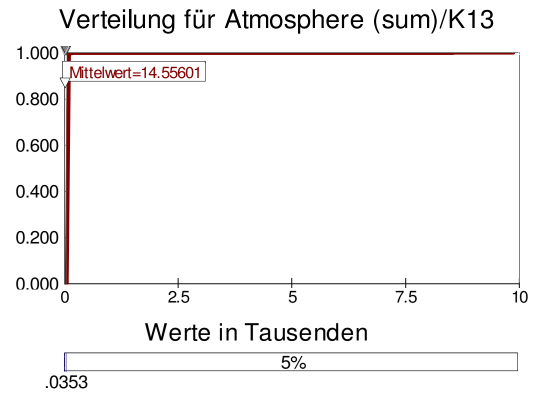


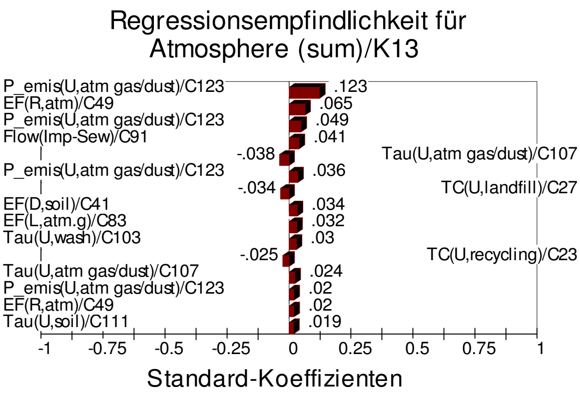
Figure 94 HBCD emissions to atmosphere, hydrosphere, soil and total emissions to the environment: scenario of in import stop by 2007 of the substance in construction materials (only application area *construction* shown)

7.8 Uncertainty of emissions to the environment

Monte Carlo simulation: emissions of DecaBDE to atmosphere





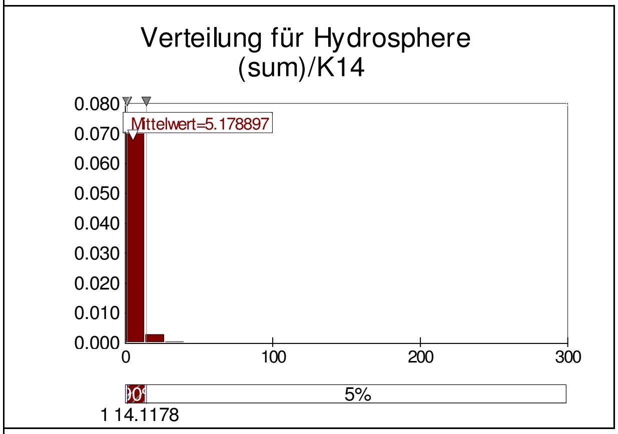


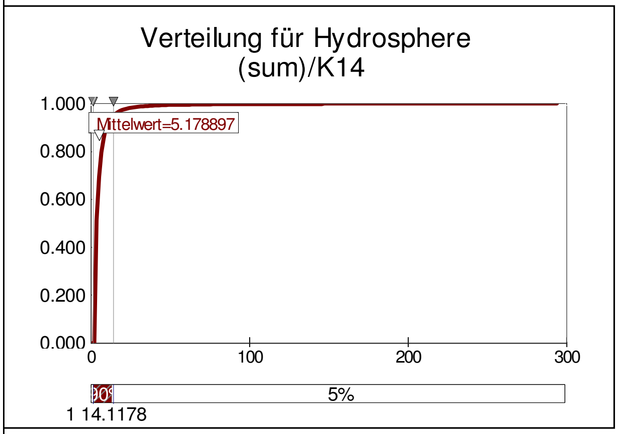
Übersichtsdaten	
Arbeitsmappenname	Input Deca 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	187
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 14:53
Simulationsende	04.04.2007 14:54
Simulationsdauer	00:00:57
Ausgangswert	1497890970

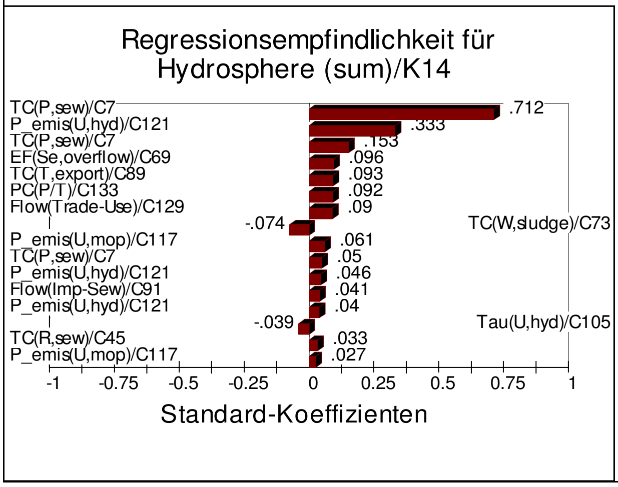
Übersichtsstatistiken			
Statistik	**Wert**	**Perz%**	**Wert**
Minimum	1.21	5%	3.57
Maximum	9864.13	10%	4.27
Mittelwert	14.56	15%	4.89
Standardabw.	100.02	20%	5.53
Varianz	10003.04	25%	6.07
Schiefe	95.60	30%	6.67
Wölbung	9412.86	35%	7.32
Medianwert	9.39	40%	7.95
Modus	5.59	45%	8.61
Linker X-Wert	3.57	50%	9.39
Linker P-Wert	5%	55%	10.19
Rechter X-Wert	35.34	60%	11.10
Rechter P-Wert	95%	65%	12.22
Diff. X	31.78	70%	13.63
Diff. P	90%	75%	15.29
Fehleranzahl	0	80%	17.39
Filter-Min.		85%	20.28
Filter-Max.		90%	25.18
Gefiltert	0	95%	35.34

Empfindlichkeit			
Rang	**Name**	**Regr.**	**Korr.**
#1	P_emis(U,atm gas/dust) / C123	0.123	0.612
#2	EF(R,atm) / C49	0.065	0.315
#3	P_emis(U,atm gas/dust) / C123	0.049	0.153
#4	Flow(Imp-Sew) / C91	0.041	-0.012
#5	Tau(U,atm gas/dust) / C107	-0.038	-0.035
#6	P_emis(U,atm gas/dust) / C123	0.036	0.230
#7	TC(U,landfill) / C27	-0.034	-0.017
#8	EF(D,soil) / C41	0.034	0.000
#9	EF(L,atm.g) / C83	0.032	0.012
#10	Tau(U,wash) / C103	0.030	0.002
#11	TC(U,recycling) / C23	-0.025	0.028
#12	Tau(U,atm gas/dust) / C107	0.024	-0.016
#13	P_emis(U,atm gas/dust) / C123	0.020	0.189
#14	EF(R,atm) / C49	0.020	0.115
#15	Tau(U,soil) / C111	0.019	-0.014
#16	TC(P,inc) / C5	0.000	-0.020

Monte Carlo simulation: emissions of DecaBDE to hydrosphere







Übersichtsdaten

Arbeitsmappenname	Input Deca 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	187
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 14:53
Simulationsende	04.04.2007 14:54
Simulationsdauer	00:00:57
Ausgangswert	1497890970

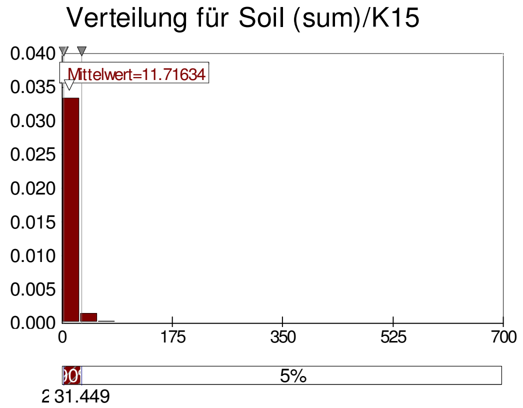
Übersichtsstatistiken

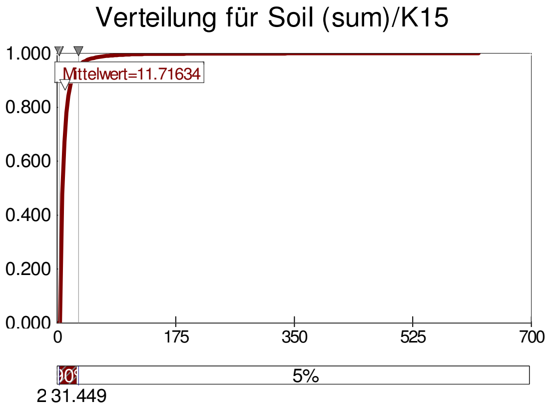
Statistik	Wert	Perz%	Wert
Minimum	0.46	5%	1.21
Maximum	293.78	10%	1.49
Mittelwert	5.18	15%	1.72
Standardabw.	7.96	20%	1.93
Varianz	63.32	25%	2.15
Schiefe	12.36	30%	2.35
Wölbung	286.92	35%	2.57
Medianwert	3.33	40%	2.80
Modus	1.83	45%	3.05
Linker X-Wert	1.21	50%	3.33
Linker P-Wert	5%	55%	3.63
Rechter X-Wert	14.12	60%	3.97
Rechter P-Wert	95%	65%	4.40
Diff. X	12.91	70%	4.93
Diff. P	90%	75%	5.54
Fehleranzahl	0	80%	6.42
Filter-Min.		85%	7.69
Filter-Max.		90%	9.58
Gefiltert	0	95%	14.12

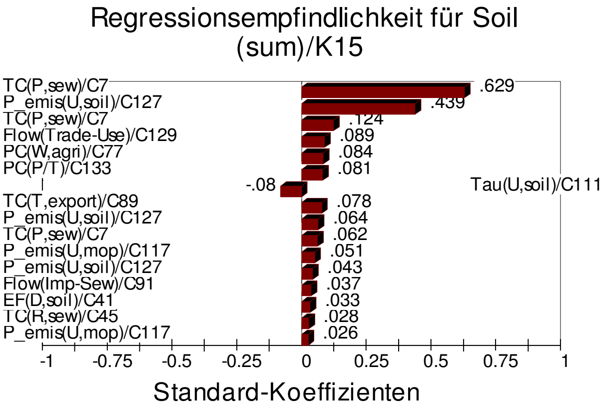
Empfindlichkeit

Rang	Name	Regr.	Korr.
#1	TC(P,sew) / C7	0.712	0.575
#2	P_emis(U,hyd) / C121	0.333	0.353
#3	TC(P,sew) / C7	0.153	0.202
#4	EF(Se,overflow) / C69	0.096	0.159
#5	TC(T,export) / C89	0.093	0.095
#6	PC(P/T) / C133	0.092	0.134
#7	Flow(Trade-Use) / C129	0.090	0.115
#8	TC(W,sludge) / C73	-0.074	-0.142
#9	P_emis(U,mop) / C117	0.061	0.081
#10	TC(P,sew) / C7	0.050	0.094
#11	P_emis(U,hyd) / C121	0.046	0.084
#12	Flow(Imp-Sew) / C91	0.041	0.108
#13	P_emis(U,hyd) / C121	0.040	0.075
#14	Tau(U,hyd) / C105	-0.039	-0.097
#15	TC(R,sew) / C45	0.033	0.076
#16	P_emis(U,mop) / C117	0.027	0.049

Monte Carlo simulation: emissions of DecaBDE to soil







Übersichtsdaten

Arbeitsmappenname	Input Deca 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	187
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 14:53
Simulationsende	04.04.2007 14:54
Simulationsdauer	00:00:57
Ausgangswert	1497890970

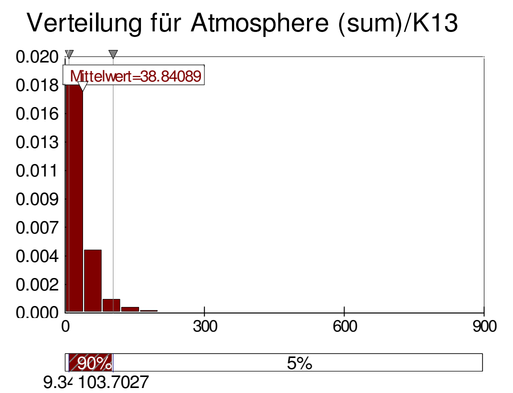
Übersichtsstatistiken

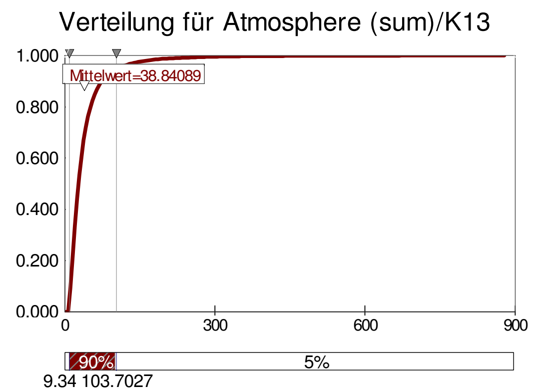
Statistik	Wert	Perz%	Wert
Minimum	1.22	5%	2.96
Maximum	622.24	10%	3.55
Mittelwert	11.72	15%	4.08
Standardabw.	17.27	20%	4.54
Varianz	298.16	25%	5.05
Schiefe	12.08	30%	5.53
Wölbung	268.88	35%	6.02
Medianwert	7.72	40%	6.51
Modus	4.22	45%	7.12
Linker X-Wert	2.96	50%	7.72
Linker P-Wert	5%	55%	8.36
Rechter X-Wert	31.45	60%	9.17
Rechter P-Wert	95%	65%	10.10
Diff. X	28.49	70%	11.23
Diff. P	90%	75%	12.68
Fehleranzahl	0	80%	14.59
Filter-Min.		85%	17.16
Filter-Max.		90%	21.59
Gefiltert	0	95%	31.45

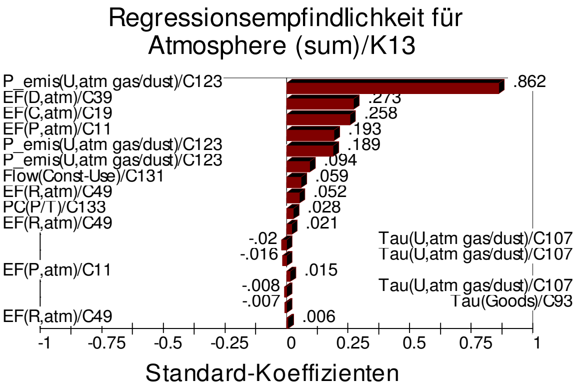
Empfindlichkeit

Rang	Name	Regr.	Korr.
#1	TC(P,sew) / C7	0.629	0.542
#2	P_emis(U,soil) / C127	0.439	0.426
#3	TC(P,sew) / C7	0.124	0.185
#4	Flow(Trade-Use) / C129	0.089	0.113
#5	PC(W,agri) / C77	0.084	0.131
#6	PC(P/T) / C133	0.081	0.130
#7	Tau(U,soil) / C111	-0.080	-0.101
#8	TC(T,export) / C89	0.078	0.108
#9	P_emis(U,soil) / C127	0.064	0.106
#10	TC(P,sew) / C7	0.062	0.080
#11	P_emis(U,mop) / C117	0.051	0.069
#12	P_emis(U,soil) / C127	0.043	0.076
#13	Flow(Imp-Sew) / C91	0.037	0.098
#14	EF(D,soil) / C41	0.033	0.041
#15	TC(R,sew) / C45	0.028	0.074
#16	P_emis(U,mop) / C117	0.026	0.029

Monte Carlo simulation: emissions of HBCD to atmosphere





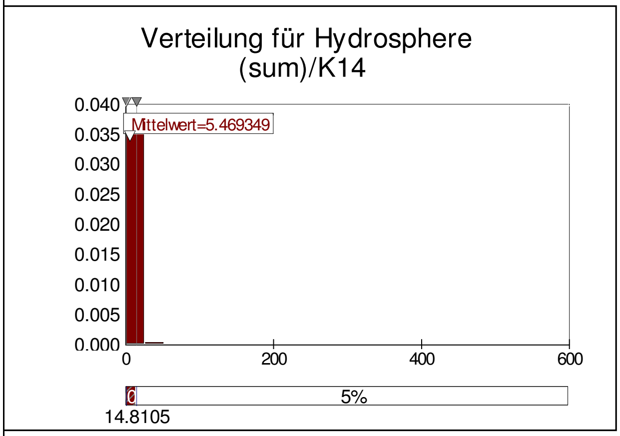


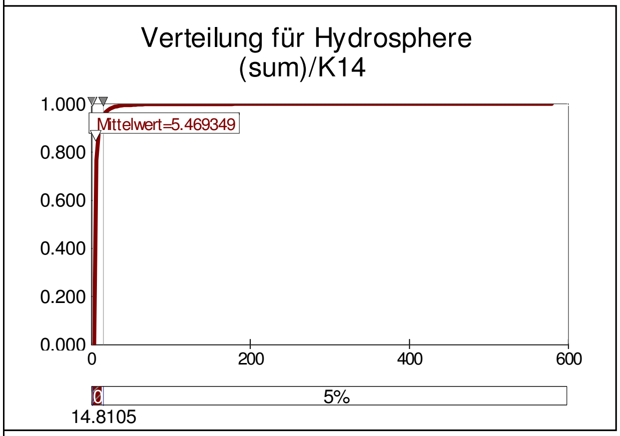
Übersichtsdaten	
Arbeitsmappenname	Input HBCD 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	188
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 17:01
Simulationsende	04.04.2007 17:02
Simulationsdauer	00:01:05
Ausgangswert	1087294550

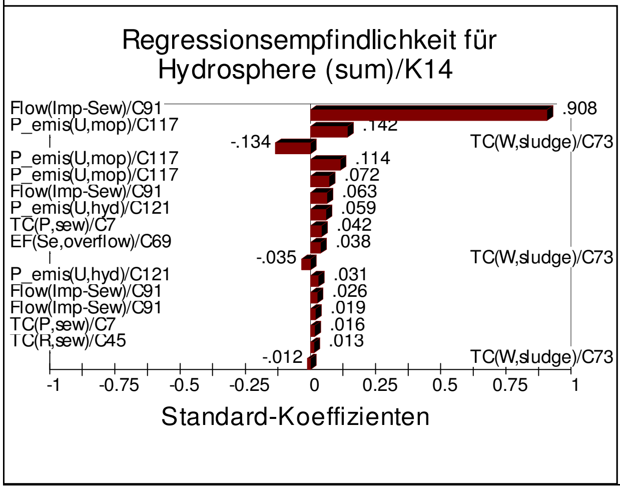
Übersichtsstatistiken			
Statistik	**Wert**	**Perz%**	**Wert**
Minimum	2.44	5%	9.35
Maximum	878.45	10%	11.45
Mittelwert	38.84	15%	13.45
Standardabw.	43.66	20%	15.15
Varianz	1906.29	25%	16.92
Schiefe	5.89	30%	18.67
Wölbung	64.24	35%	20.56
Medianwert	27.03	40%	22.55
Modus	15.02	45%	24.65
Linker X-Wert	9.35	50%	27.03
Linker P-Wert	5%	55%	29.71
Rechter X-Wert	103.70	60%	32.55
Rechter P-Wert	95%	65%	36.02
Diff. X	94.35	70%	40.03
Diff. P	90%	75%	44.72
Fehleranzahl	0	80%	51.53
Filter-Min.		85%	59.69
Filter-Max.		90%	73.02
Gefiltert	0	95%	103.70

Empfindlichkeit			
Rang	**Name**	**Regr.**	**Korr.**
#1	P_emis(U,atm gas/dust) / C123	0.862	0.637
#2	EF(D,atm) / C39	0.273	0.291
#3	EF(C,atm) / C19	0.258	0.260
#4	EF(P,atm) / C11	0.193	0.202
#5	P_emis(U,atm gas/dust) / C123	0.189	0.216
#6	P_emis(U,atm gas/dust) / C123	0.094	0.102
#7	Flow(Const-Use) / C131	0.059	0.102
#8	EF(R,atm) / C49	0.052	0.067
#9	PC(P/T) / C133	0.028	0.031
#10	EF(R,atm) / C49	0.021	0.028
#11	Tau(U,atm gas/dust) / C107	-0.020	-0.022
#12	Tau(U,atm gas/dust) / C107	-0.016	-0.005
#13	EF(P,atm) / C11	0.015	0.004
#14	Tau(U,atm gas/dust) / C107	-0.008	-0.016
#15	Tau(Goods) / C93	-0.007	-0.008
#16	EF(R,atm) / C49	0.006	0.018

Monte Carlo simulation: emissions of HBCD to hydrosphere


Verteilung für Hydrosphere (sum)/K14


Verteilung für Hydrosphere (sum)/K14


Regressionsempfindlichkeit für Hydrosphere (sum)/K14

Übersichtsdaten	
Arbeitsmappenname	Input HBCD 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	188
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 17:01
Simulationsende	04.04.2007 17:02
Simulationsdauer	00:01:05
Ausgangswert	1087294550

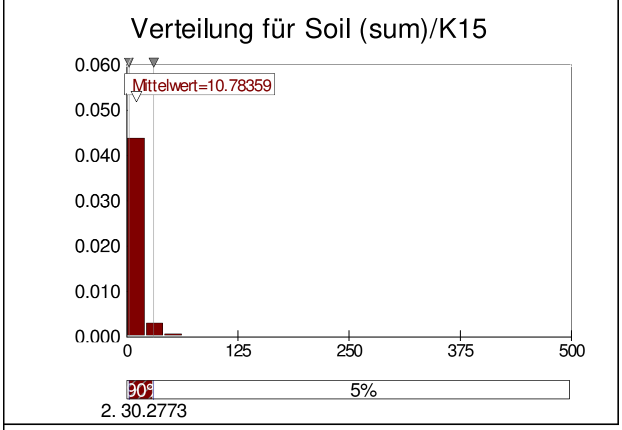
Übersichtsstatistiken

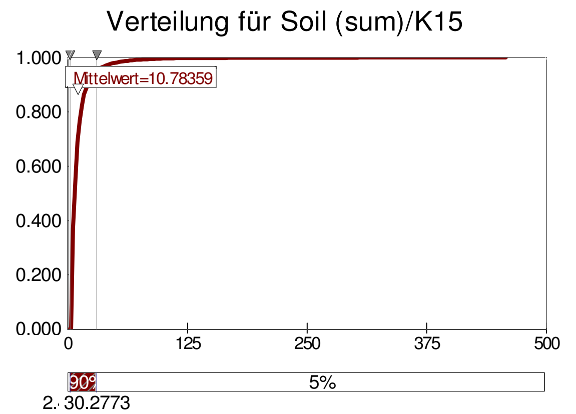
Statistik	Wert	Perz%	Wert
Minimum	0.46	5%	1.29
Maximum	579.28	10%	1.58
Mittelwert	5.47	15%	1.82
Standardabw.	9.39	20%	2.07
Varianz	88.08	25%	2.29
Schiefe	28.22	30%	2.53
Wölbung	1486.37	35%	2.76
Medianwert	3.58	40%	3.00
Modus	2.55	45%	3.28
Linker X-Wert	1.29	50%	3.58
Linker P-Wert	5%	55%	3.91
Rechter X-Wert	14.81	60%	4.29
Rechter P-Wert	95%	65%	4.76
Diff. X	13.52	70%	5.25
Diff. P	90%	75%	5.97
Fehleranzahl	0	80%	6.84
Filter-Min.		85%	8.14
Filter-Max.		90%	10.34
Gefiltert	0	95%	14.81

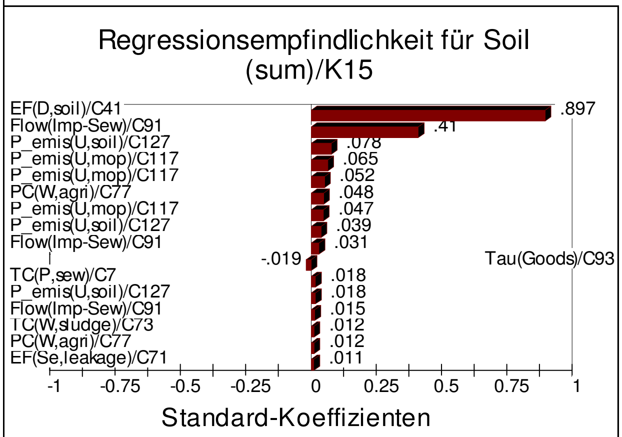
Empfindlichkeit

Rang	Name	Regr.	Korr.
#1	Flow(Imp-Sew) / C91	0.908	0.670
#2	P_emis(U,mop) / C117	0.142	0.231
#3	TC(W,sludge) / C73	-0.134	-0.285
#4	P_emis(U,mop) / C117	0.114	0.193
#5	P_emis(U,mop) / C117	0.072	0.134
#6	Flow(Imp-Sew) / C91	0.063	0.115
#7	P_emis(U,hyd) / C121	0.059	0.091
#8	TC(P,sew) / C7	0.042	0.077
#9	EF(Se,overflow) / C69	0.038	0.094
#10	TC(W,sludge) / C73	-0.035	-0.109
#11	P_emis(U,hyd) / C121	0.031	0.050
#12	Flow(Imp-Sew) / C91	0.026	0.043
#13	Flow(Imp-Sew) / C91	0.019	0.037
#14	TC(P,sew) / C7	0.016	0.054
#15	TC(R,sew) / C45	0.013	0.029
#16	TC(W,sludge) / C73	-0.012	-0.041

Monte Carlo simulation: emissions of HBCD to soil







Übersichtsdaten	
Arbeitsmappenname	Input HBCD 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	188
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 17:01
Simulationsende	04.04.2007 17:02
Simulationsdauer	00:01:05
Ausgangswert	1087294550

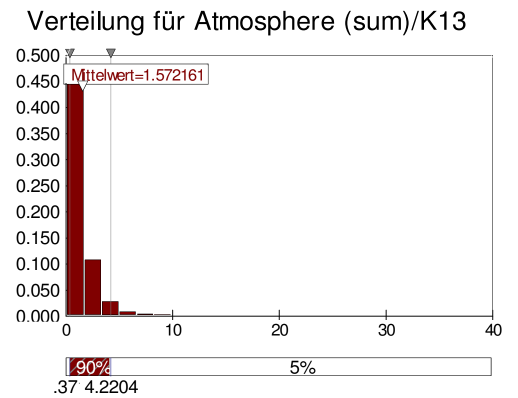
Übersichtsstatistiken

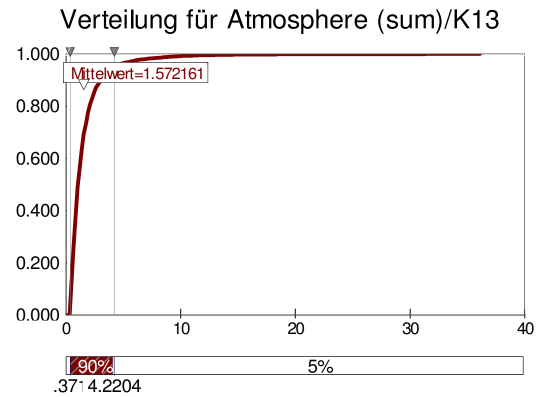
Statistik	Wert	Perz%	Wert
Minimum	0.95	5%	2.41
Maximum	457.52	10%	2.99
Mittelwert	10.78	15%	3.51
Standardabw.	15.51	20%	3.93
Varianz	240.71	25%	4.36
Schiefe	9.97	30%	4.84
Wölbung	177.41	35%	5.31
Medianwert	6.99	40%	5.79
Modus	3.72	45%	6.37
Linker X-Wert	2.41	50%	6.99
Linker P-Wert	5%	55%	7.65
Rechter X-Wert	30.28	60%	8.38
Rechter P-Wert	95%	65%	9.28
Diff. X	27.87	70%	10.34
Diff. P	90%	75%	11.77
Fehleranzahl	0	80%	13.55
Filter-Min.		85%	16.04
Filter-Max.		90%	20.44
Gefiltert	0	95%	30.28

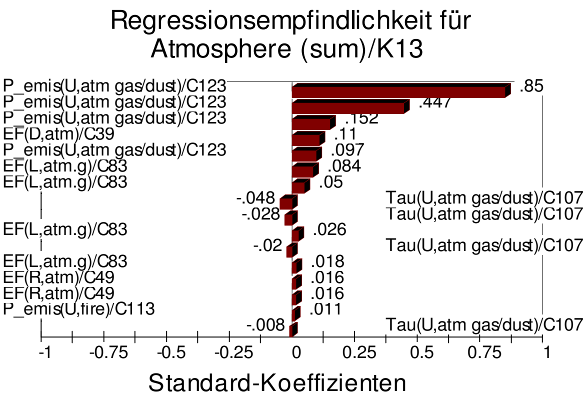
Empfindlichkeit

Rang	Name	Regr.	Korr.
#1	EF(D,soil) / C41	0.897	0.731
#2	Flow(Imp-Sew) / C91	0.410	0.364
#3	P_emis(U,soil) / C127	0.078	0.140
#4	P_emis(U,mop) / C117	0.065	0.102
#5	P_emis(U,mop) / C117	0.052	0.078
#6	PC(W,agri) / C77	0.048	0.111
#7	P_emis(U,mop) / C117	0.047	0.055
#8	P_emis(U,soil) / C127	0.039	0.074
#9	Flow(Imp-Sew) / C91	0.031	0.057
#10	Tau(Goods) / C93	-0.019	-0.013
#11	TC(P,sew) / C7	0.018	0.032
#12	P_emis(U,soil) / C127	0.018	0.015
#13	Flow(Imp-Sew) / C91	0.015	0.018
#14	TC(W,sludge) / C73	0.012	0.017
#15	PC(W,agri) / C77	0.012	0.035
#16	EF(Se,leakage) / C71	0.011	0.015

Monte Carlo simulation: emissions of BDE-47 to atmosphere





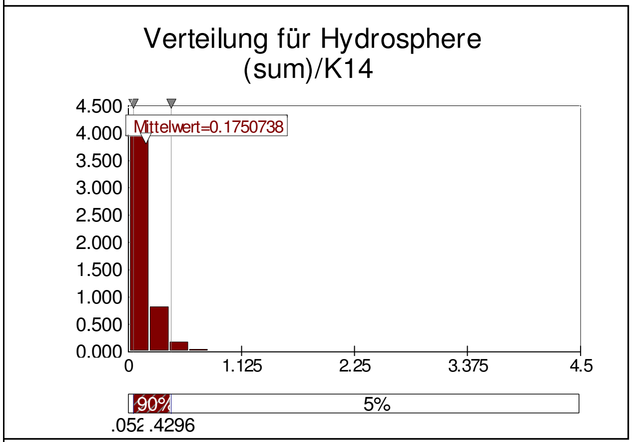


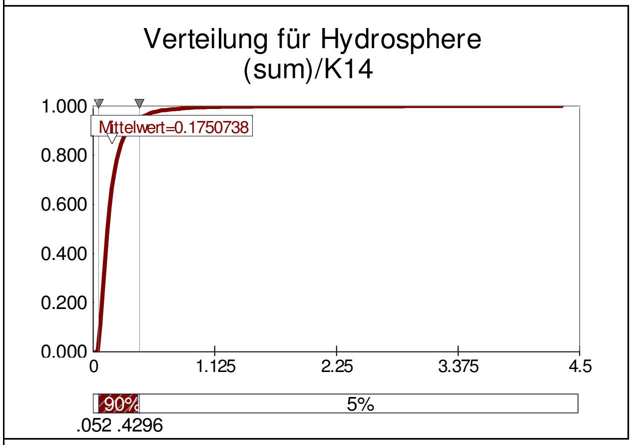
Übersichtsdaten	
Arbeitsmappenname	Input BDE-47 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	187
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 16:18
Simulationsende	04.04.2007 16:19
Simulationsdauer	00:01:02
Ausgangswert	456220090

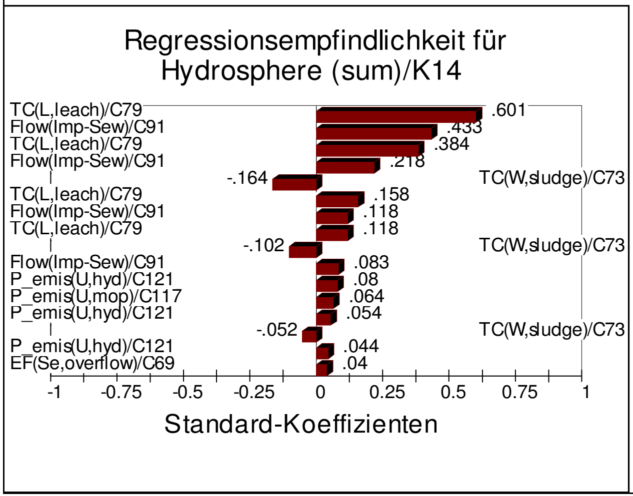
Übersichtsstatistiken			
Statistik	**Wert**	**Perz%**	**Wert**
Minimum	0.13	5%	0.37
Maximum	36.10	10%	0.45
Mittelwert	1.57	15%	0.52
Standardabw.	1.86	20%	0.59
Varianz	3.47	25%	0.67
Schiefe	6.23	30%	0.73
Wölbung	70.51	35%	0.80
Medianwert	1.05	40%	0.87
Modus	0.56	45%	0.96
Linker X-Wert	0.37	50%	1.05
Linker P-Wert	5%	55%	1.16
Rechter X-We	4.22	60%	1.29
Rechter P-We	95%	65%	1.43
Diff. X	3.85	70%	1.60
Diff. P	90%	75%	1.80
Fehleranzahl	0	80%	2.06
Filter-Min.		85%	2.43
Filter-Max.		90%	3.06
Gefiltert	0	95%	4.22

Empfindlichkeit			
Rang	**Name**	**Regr.**	**Korr.**
#1	P_emis(U,atm gas/dust) / C123	0.850	0.656
#2	P_emis(U,atm gas/dust) / C123	0.447	0.447
#3	P_emis(U,atm gas/dust) / C123	0.152	0.128
#4	EF(D,atm) / C39	0.110	0.146
#5	P_emis(U,atm gas/dust) / C123	0.097	0.118
#6	EF(L,atm.g) / C83	0.084	0.109
#7	EF(L,atm.g) / C83	0.050	0.065
#8	Tau(U,atm gas/dust) / C107	-0.048	-0.052
#9	Tau(U,atm gas/dust) / C107	-0.028	-0.046
#10	EF(L,atm.g) / C83	0.026	0.028
#11	Tau(U,atm gas/dust) / C107	-0.020	-0.040
#12	EF(L,atm.g) / C83	0.018	0.019
#13	EF(R,atm) / C49	0.016	0.016
#14	EF(R,atm) / C49	0.016	0.019
#15	P_emis(U,fire) / C113	0.011	0.021
#16	Tau(U,atm gas/dust) / C107	-0.008	-0.025

Monte Carlo simulation: emissions of BDE-47 to hydrosphere







Übersichtsdaten	
Arbeitsmappenname	Input BDE-47 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	187
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 16:18
Simulationsende	04.04.2007 16:19
Simulationsdauer	00:01:02
Ausgangswert	456220090

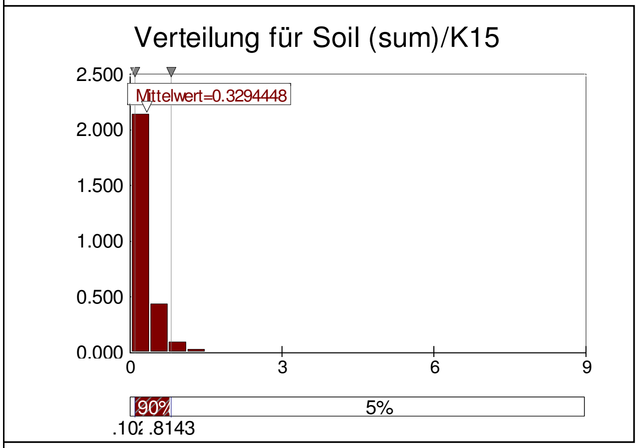
Übersichtsstatistiken

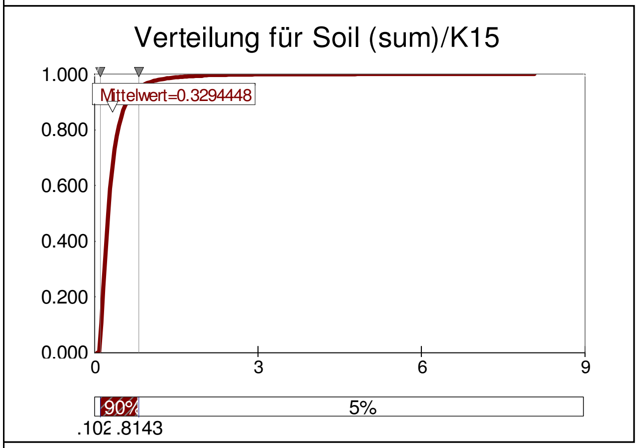
Statistik	Wert	Perz%	Wert
Minimum	0.02	5%	0.05
Maximum	4.33	10%	0.06
Mittelwert	0.18	15%	0.07
Standardabw.	0.17	20%	0.08
Varianz	0.03	25%	0.09
Schiefe	6.89	30%	0.10
Wölbung	99.13	35%	0.11
Medianwert	0.13	40%	0.11
Modus	0.09	45%	0.12
Linker X-Wert	0.05	50%	0.13
Linker P-Wert	5%	55%	0.14
Rechter X-We	0.43	60%	0.15
Rechter P-We	95%	65%	0.17
Diff. X	0.38	70%	0.18
Diff. P	90%	75%	0.20
Fehleranzahl	0	80%	0.23
Filter-Min.		85%	0.26
Filter-Max.		90%	0.32
Gefiltert	0	95%	0.43

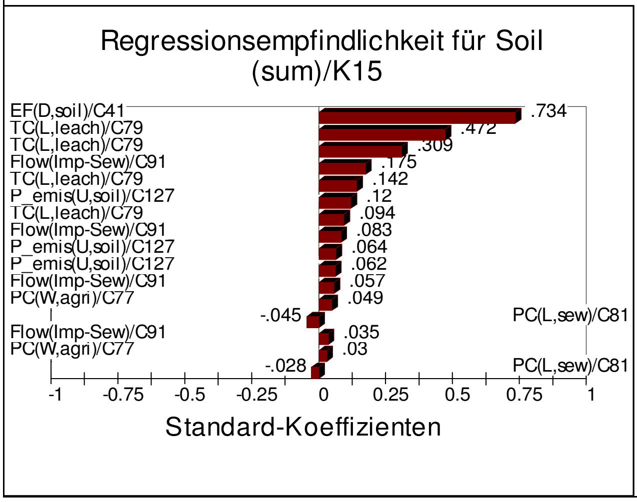
Empfindlichkeit

Rang	Name	Regr.	Korr.
#1	TC(L,leach) / C79	0.601	0.402
#2	Flow(Imp-Sew) / C91	0.433	0.339
#3	TC(L,leach) / C79	0.384	0.304
#4	Flow(Imp-Sew) / C91	0.218	0.190
#5	TC(W,sludge) / C73	-0.164	-0.222
#6	TC(L,leach) / C79	0.158	0.166
#7	Flow(Imp-Sew) / C91	0.118	0.097
#8	TC(L,leach) / C79	0.118	0.105
#9	TC(W,sludge) / C73	-0.102	-0.160
#10	Flow(Imp-Sew) / C91	0.083	0.077
#11	P_emis(U,hyd) / C121	0.080	0.067
#12	P_emis(U,mop) / C117	0.064	0.057
#13	P_emis(U,hyd) / C121	0.054	0.047
#14	TC(W,sludge) / C73	-0.052	-0.098
#15	P_emis(U,hyd) / C121	0.044	0.058
#16	EF(Se,overflow) / C69	0.040	0.065

Monte Carlo simulation: emissions of BDE-47 to soil


Verteilung für Soil (sum)/K15


Verteilung für Soil (sum)/K15


Regressionsempfindlichkeit für Soil (sum)/K15

Übersichtsdaten	
Arbeitsmappenname	Input BDE-47 2005.xls
Anzahl der Simulationen	1
Anzahl der Iterationen	10000
Anzahl der Eingaben	187
Anzahl der Ausgaben	20
Probenerhebungstyp	Monte Carlo
Simulationsbeginn	04.04.2007 16:18
Simulationsende	04.04.2007 16:19
Simulationsdauer	00:01:02
Ausgangswert	456220090

Übersichtsstatistiken

Statistik	Wert	Perz%	Wert
Minimum	0.04	5%	0.10
Maximum	8.07	10%	0.12
Mittelwert	0.33	15%	0.14
Standardabw.	0.31	20%	0.15
Varianz	0.10	25%	0.17
Schiefe	6.01	30%	0.18
Wölbung	79.86	35%	0.20
Medianwert	0.25	40%	0.21
Modus	0.18	45%	0.23
Linker X-Wert	0.10	50%	0.25
Linker P-Wert	5%	55%	0.27
Rechter X-Wer	0.81	60%	0.29
Rechter P-We	95%	65%	0.31
Diff. X	0.71	70%	0.34
Diff. P	90%	75%	0.38
Fehleranzahl	0	80%	0.43
Filter-Min.		85%	0.49
Filter-Max.		90%	0.59
Gefiltert	0	95%	0.81

Empfindlichkeit

Rang	Name	Regr.	Korr.
#1	EF(D,soil) / C41	0.734	0.491
#2	TC(L,leach) / C79	0.472	0.370
#3	TC(L,leach) / C79	0.309	0.270
#4	Flow(Imp-Sew) / C91	0.175	0.181
#5	TC(L,leach) / C79	0.142	0.144
#6	P_emis(U,soil) / C127	0.120	0.118
#7	TC(L,leach) / C79	0.094	0.089
#8	Flow(Imp-Sew) / C91	0.083	0.097
#9	P_emis(U,soil) / C127	0.064	0.083
#10	P_emis(U,soil) / C127	0.062	0.054
#11	Flow(Imp-Sew) / C91	0.057	0.043
#12	PC(W,agri) / C77	0.049	0.063
#13	PC(L,sew) / C81	-0.045	-0.062
#14	Flow(Imp-Sew) / C91	0.035	0.036
#15	PC(W,agri) / C77	0.030	0.046
#16	PC(L,sew) / C81	-0.028	-0.033