



# MODELLING LONG-RANGE TRANSPORT AND EMISSIONS FROM LOCAL SOURCES OF NEW CONTAMINANTS IN THE ARCTIC

Scientific Report from DCE – Danish Centre for Environment and Energy

No. 499

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# Data sheet

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Abstract:	Contributions from local emissions and long-range transport of seven current-use flame retardants (FRs) to the environment in Nuuk, Greenland, were quantified and compared. The contribution to the local environment from local sources was significant compared to the contribution from long-range transport. Compound volatility was an important parameter. There was good agreement between modelled and measured air concentrations for five FRs, and good agreement of soil and seawater concentrations for four FRs. A human risk from indirect FR exposure via the environment, estimated from the derived environmental concentrations and standard daily intake values of air, fish and seal, showed no or low risk. Likewise, the environmental risk was low. This study showed that local sources were relevant for Arctic towns such as Nuuk, and the suggested method can also be used in other remote locations to assess emissions, source contributions and risks to humans and the environment
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# Contents

<b>Preface</b>	<b>5</b>
<b>Sammenfatning</b>	<b>6</b>
<b>Summary</b>	<b>8</b>
<b>1 Background and objectives</b>	<b>10</b>
<b>2 Method</b>	<b>12</b>
<b>3 Compound characteristics and environmental occurrence</b>	<b>14</b>
3.1 Physical/chemical parameters	14
3.2 Measurements in the Arctic	15
<b>4 Mass flows and emissions</b>	<b>17</b>
4.1 Mass flow and uses	17
4.2 Production	22
4.3 Emissions	24
4.4 Waste treatment and spatial distribution of emissions in the Northern hemisphere	31
<b>5 Long-range transport modelling with DEHM</b>	<b>37</b>
5.1 Model set-up and sensitivity analysis	37
5.2 Model evaluation	39
5.3 Assessment of source areas	46
<b>6 Local sources and environmental concentrations</b>	<b>50</b>
6.1 Local dispersion model	50
6.2 Additional potential sources and exposures	64
6.3 Beach litter	64
<b>7 Model evaluation and risk screening</b>	<b>66</b>
7.1 Model evaluation	66
7.2 Risk screening	71
<b>8 Conclusions</b>	<b>75</b>
<b>9 References</b>	<b>77</b>





## Preface

This project was funded by the programme *Miljøstøtte til Arktis* of the Danish Environmental Protection Agency (EPA).

The project was initiated in April 2020 and finalized in February 2022.

The project group consisted of:

Senior scientist Patrik Fauser, Aarhus University, Department of Environmental Science: Project leader, data acquisition, emission inventories, local model, exposure and risk screening, data analysis, dissemination.

Senior scientist Kaj Mantzius Hansen, Aarhus University, Department of Environmental Science: Global atmospheric transport modelling, data analysis, dissemination.

Senior scientist Jesper Heile Christensen, Aarhus University, Department of Environmental Science: Global atmospheric transport modelling, data analysis.

Senior scientist Peter Borgen Sørensen, Aarhus University, Department of Ecoscience: Local model, data analysis.

Professor Katrin Vorkamp, Aarhus University, Department of Environmental Science: Data acquisition, data analysis, implementation of knowledge and results in an AMAP context, dissemination.

Two meetings with the purpose to discuss project activities and coordinate them with other relevant activities were held in July 2020 and September 2021 in an advisory group consisting of: Mikala Klint (Ministry of Environment), Palle Smedegaard Nielsen (Government of Greenland), Rune Hjorth (Danish EPA), Maj Friis Munk/Niels Kurt Nielsen (Danish EPA), Patrik Fauser and Katrin Vorkamp (Aarhus University). However, Maj Friis Munk and Niels Kurt Nielsen were unable to attend the meetings.

Senior scientist Pia Lassen, Aarhus University, Department of Environmental Science, conducted peer assessment of the report and chief consultant Susanne Boutrup, Aarhus University, Danish Centre for Environment and Energy (DCE), conducted quality assurance.

## Sammenfatning

Dette studie omhandler syv flammehæmmere med nuværende anvendelse og som alle er fundet i miljøet i Arktis. Det drejer sig om tre organofosfat flammehæmmere: tri(2-chloroethyl) phosphate (TCEP), tri(chloroiso-propyl) phosphate (TCPP) og triphenyl phosphate (TPHP), tre bromerede flammehæmmere: 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), bis(2-ethylhexyl) tetrabromophthalate (BEH-TEBP) og hexabromobenzene (HBBz), og den chlorerede flammehæmmer dechlorane plus (DP).

Projektets formål var at kvantificere bidragene fra hhv. lokale emissioner og langtransport til miljøet i Nuuk i Grønland. Langtransport blev estimeret med kemi-transport modellen DEHM med anvendelse af emissioner fra produktion, plastik- og tekstil anvendelse og affaldsbehandling i ni globale regioner. Koncentrationer i miljøet blev desuden beregnet med en model for det lokale miljø indeholdende emissioner til luft, jord og vand fra lokal anvendelse af plastik og tekstiler samt fra affaldsforbrænding i Nuuk.

I forbindelse med modelberegningerne blev der brugt en række antagelser, tilnærmelser og justeringer af data. Disse omfattede: Emissionsfaktorer for de polybromerede diphenyl ether decaBDE og pentaBDE blev anvendt. Emissionsfaktorer til luft blev justeret mht. forskelle i molekylvægte, damptryk og temperatur (for udendørs anvendelse af produkter og affaldsbehandling). Der blev taget højde for usikkerheder i data ved at opstille et lavt og et højt emissionsscenarie.

En sammenligning af resultater fra modelsimulationer viste, at luftkoncentrationer fra de lokale kilder var to til tre størrelsesordener højere end luftkoncentrationer beregnet for langtransport, for TCEP, TCPP, TPHP (højt emissionsscenarie) og EH-TBB. Bidraget til luftkoncentrationer fra de lokale kilder var for TPHP (lavt emissionsscenarie), BEH-TEBP, HBBz og DP ca. én størrelsesorden højere sammenlignet med bidraget fra langtransport. De lokale kilder bidrog betydeligt mere end langtransport (fire til seks størrelsesordener) til forekomsten i jord, havvand og biota for alle flammehæmmere, på nær TCEP og HBBz. TCEP og HBBz har de højeste emissionsfaktorer til luft blandt hhv. organofosfat og bromerede flammehæmmere, hvilket havde en signifikant indflydelse på resultaterne. BEH-TEBP og DP har de laveste emissionsfaktorer til luft.

Til evaluering af modelresultaterne blev der anvendt målinger i luft, jord, havvand og biota. Der var relativt få måledata for de syv stoffer, hvoraf de fleste var fra Svalbard. Der var god overensstemmelse ( $\leq$  én størrelsesorden mellem modellerede og målte luftkoncentrationer for TCEP og TCPP og for et emissionsscenarie for EH-TBB, BEH-TEBP og HBBz, mens der var ringe overensstemmelse for TPHP ( $\geq$  to størrelsesordener) og DP ( $\geq$  tre størrelsesordener). I jord var de modellerede koncentrationer af alle flammehæmmere, på nær TCEP, sammenlignelige med eller op til to størrelsesordener lavere end målte jordkoncentrationer. Modellerede TCEP koncentrationer i jord var fire til fem størrelsesordener lavere end målte koncentrationer. Der var ingen BEH-TEBP målinger i jord. For havvand var modellerede koncentrationer af TCPP og TPHP (lavt scenarie), BEH-TEBP og HBBz (højt scenarie) sammenlignelige ( $\leq$  en størrelsesorden) med målte koncentrationer. For alle andre flammehæmmere var de modellerede koncentrationer i havvand mellem to



og fem størrelsesordener lavere end de målte koncentrationer. Der var ingen EH-TBB målinger i havvand. Der er kun få målinger i fisk (atlantisk torsk); for EH-TBB og HBBz var modellerede koncentrationer op til to størrelsesordener lavere sammenlignet med målinger. Modellerede koncentrationer i sælspek var sammenlignelige, dvs. en størrelsesorden lavere, for EH-TBB, og fire til fem størrelsesordener lavere for TPHP og DP, sammenlignet med målte værdier.

Simuleringer med den atmosfæriske kemi-transport model DEHM viste, at andelen af det langtransporterede bidrag til Arktis fra Europa var højere end regionens andel af de totale globale emissioner. Det modsatte var tilfældet for Kina, Japan og det øvrige Asien. Dette skyldes de globale transportmønstre, hvor transporten fra Europa og den asiatiske del af Rusland til Arktis, er den dominerende rute.

Indirekte eksponering af flammehæmmere via miljøet for mennesker, estimeret fra modellerede miljøkoncentrationer og standardværdier for dagligt indtag af luft, fisk og sæl, viste ingen eller lav risiko (risikokoefficient  $< 1E-04$ ). Den samme størrelsesorden i risikokoefficienten blev indikeret for torsk, her var toksicitetsdata dog mangelfulde.

## Summary

This study addresses seven current-use flame retardants (FRs), which have been detected in the Arctic environment. These include three organophosphorous FRs (OPFRs): tri(2-chloroethyl) phosphate (TCEP), tri(chloroiso-propyl) phosphate (TCPP) and triphenyl phosphate (TPHP), three brominated FRs (BFRs): 2-Ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), bis(2-ethylhexyl) tetrabromophthalate (BEH-TEBP) and hexabromobenzene (HBBz), and the chlorinated FR dechlorane plus (DP).

The purpose of the study was to quantify contributions from local emissions and long-range transport, respectively, in the Greenland environment in Nuuk. Long-range transport was studied with the atmospheric chemistry-transport model DEHM, based on emissions from production, plastic and textile use and waste treatment in nine global regions. Local concentrations were calculated using a local site model and emissions to air, soil and water from local use of plastic and textile products as well as waste incineration.

A number of assumptions, approximations and adjustments were made in the model calculations, including the following: Emission factors (EFs) were taken from the polybrominated diphenyl ethers (PBDEs) decaBDE and pentaBDE. EFs to air were adjusted for differences in molecular weights, vapour pressure (Vp) and temperature (in the case of outdoor product use and waste treatment). Uncertainties in EFs were taken into account by considering a low and a high emission scenario.

A comparison of model simulations showed that the air concentrations from local sources were two and three orders of magnitude (OM) higher than the air concentrations calculated for long-range transport for TCEP, TCPP, TPHP (high emission scenario) and EH-TBB. For TPHP (low scenario), BEH-TEBP, HBBz and DP the local sources contributed one OM or less to the air concentrations, compared to the contribution from long-range transport. For soil, marine water and biota, local sources were significantly more influential than long-range transport (four to six OM) for all FRs, except for TCEP and HBBz. TCEP and HBBz have the highest EFs (and Vp) to air among the OPFRs and the BFRs, respectively, which had a strong influence on this result. BEH-TEBP and DP have the lowest EFs to air.

The comparison with measured values was limited to relatively few studies reporting air, soil and water data for these seven compounds. The majority was from Svalbard. There was good agreement ( $\leq$  one OM) between modelled and measured air concentrations for TCEP and TCPP, and for one of the emission scenarios for EH-TBB, BEH-TEBP and HBBz. There was poor agreement for TPHP ( $\geq$  two OM) and even more so for DP. For the soil compartment, all FRs, except TCEP, had modelled concentrations that were comparable and up to two OM lower than measured soil concentrations. No soil measurements were found for BEH-TEBP. Modelled TCEP soil concentrations were between four and five OM lower than the measured concentrations. For seawater, modelled concentrations of TCPP and TPHP (low scenarios), and BEH-TEBP and HBBz (high scenarios) were comparable ( $\leq$  one OM) to measured concentrations. For all other FRs the modelled concentrations were between two and five OM lower than the measured concentrations. No measurements were found for EH-TBB. Measurements in fish (Atlantic cod) and seal are scarce,



but for EH-TBB and HBBz in cod the modelled concentrations were up to two OM lower compared to the measurements. Modelled values of EH-TBB in seal blubber are comparable (one OM lower) to measured values, and modelled values of TPHP and DP are four to five orders of magnitude lower than the measured values.

Simulations with the atmospheric chemistry-transport model DEHM showed that the contributed fraction to the Arctic from Europe was higher than the region's share of the total global emission, whereas the opposite was the case for China, Japan and the rest of Asia. This was due to the prevailing transport pattern, in which the transport from Europe and the Asian part of Russia into the Arctic was the largest transport route.

A human risk from indirect FR exposure via the environment, estimated from the derived environmental concentrations and standard daily intake values of air, fish and seal, showed no or low risk (Risk Characterization Ratio < 1E-04). The same OM for the environmental risk towards cod was found. However, toxicity data were missing or limited, which compromised the accuracy of the risk estimate.

# 1 Background and objectives

Since the inclusion of the polybrominated diphenyl ethers (PBDEs) in the Stockholm Convention on Persistent Organic Pollutants (POPs) in 2009 and 2017 for being persistent (P), bioaccumulative (B), toxic (T) and transported over long distances (LRT) the use of other brominated (BFRs) and organophosphorous flame retardants (OPFRs) has increased. Little information is available on identities and amounts of potential replacement substances. It has been reported that a mixture of bis(2-ethylhexyl) tetrabromophthalate (BEH-TEBP) and 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB) has replaced the commercial PentaBDE-mixture, which had primarily been used in the USA (Stapleton et al. 2008). In addition, hexabromobenzene (HBBz) has been found frequently in the Arctic (Vorkamp et al., 2019a). Specifically, these compounds have been among the non-PBDE BFRs with the highest air concentrations in the Canadian Arctic, Alaska, Svalbard and Iceland, and their concentration levels were often comparable to those of PBDEs. They were also found in Arctic animals, although usually in low concentrations (Sagerup et al., 2010; Harju et al., 2013; Vorkamp et al., 2019a). The OPFRs tri(2-chloro-ethyl) phosphate (TCEP), tri(chloroiso-propyl) phosphate (TCPP) and tri-phenyl phosphate (TPHP) have been found in Arctic air in concentrations that exceeded the concentrations of the banned PBDEs, as well as in other abiotic samples, while their detection in Arctic biota was limited (Vorkamp et al., 2019a). Both BFRs and OPFRs are used in applications in the polymer industries in plastics and electrical/electronic applications and in textile products. OPFRs are also used as softeners and anti-foaming agents, and they are used in hydraulic fluids and polishing products (Pantelaki et al., 2019).

Dechlorane plus (DP) is a chlorinated FR (CFR), which is currently under review in the Stockholm Convention on POPs. DP has been reported in several remote environments such as in the Arctic and Antarctic air and in seawater (Vorkamp et al., 2015; Möller et al., 2012b; Vorkamp et al., 2019b), in lichen in Antarctica (Kim et al., 2018) and the Tibetan Plateau (Yang et al., 2013) and in several species of Arctic wildlife (Schlabach et al., 2018; Simond et al., 2017; Vorkamp et al., 2015). Further, DP has also been found in global air (Rauert et al., 2018). DP has been manufactured since the 1950s, and it is currently registered as a high-production volume chemical in the United States (Ren et al., 2009). DP is used in polymers such as in electronic wiring and cables, the aviation- and automobile industry, as well as plastic casings for TVs and computers (Kurt-Karakus et al., 2019; Weil et al., 2015).

The LRT to the Arctic is important for an understanding of the global fate processes of these and other chemicals. LRT also is one of the criteria of the Stockholm Convention, in a classification of POPs and consequently, their global regulation. Arctic data from remote locations have been fundamental in the classification process, by providing evidence of LRT. However, recent findings suggest that for current-use chemicals, in particular those in consumer products, local sources in the Arctic may exist as well, as recently documented in a report of the Arctic Monitoring and Assessment Programme (AMAP) on Chemicals of Emerging Arctic Concern (CEACs) (AMAP, 2017). Given their use in many plastic materials, FRs could be among the group of chemicals emitted locally. The relative contribution of local sources compared to contributions from LRT are generally unknown. An important obstacle in the analysis of local versus distant sources is the confidential information of

the produced amounts and their development over time. Furthermore, emission inventories for the majority of environmental contaminants, including the investigated FRs, are absent. This was also highlighted in the AMAP report on CEACs: *“The lack of production, use and emission data hinders understanding of the environmental fate of many CECs<sup>1</sup>, and chemical producers are encouraged to be more forthcoming with such data. Where production and emission data do exist, environmental inventories should be compiled and validated with spatial and temporal trend data.”* (AMAP, 2017). The report further concluded: *“Such studies should be carried out for CECs in order to understand the most important sources and transport routes for their entry to the Arctic and to be able to prioritize actions to reduce discharges and emissions.”*.

Based on these challenges, the objectives of the project are to address and quantify local sources and LRT in the environment of Greenland, for the seven selected FRs (three BFRs, three OPFRs, DP), using Nuuk as an example of maximum human activity in Greenland. The study is based on model calculations and only uses measurements for comparisons with the modelled values. An important first step in the project is to compile information and data on production, use and disposal of FRs and FR-containing products and to establish emission estimates. Global distributions of emissions are set up and used to simulate LRT to the atmosphere in Nuuk with the Danish Eulerian Hemispheric Model (DEHM). A local-scale model is developed as a separate compartment model to simulate emissions and concentrations in air, soil and marine water in Nuuk from local sources. This gives a quantitative comparison of the two sources. Their sum are compared with environmental measurements for a critical evaluation of the models. Finally, a screening of the risk for humans and selected marine species is performed.

<sup>1</sup> Here the term „Chemicals of Emerging Concern“ (CECs) was used.

## 2 Method

The methodological procedure consists of four parts, as outlined in Figure 2.1. Parts 1 to 4 are performed for each of the seven compounds in Table 2.1 in order to investigate their global and local emissions, and exposure and risk to the local environment in Nuuk. In this study a mass flow consisting of production and manufacture of the compounds, indoor/outdoor use and waste treatment of the plastics and textile products in which they are used, is used.

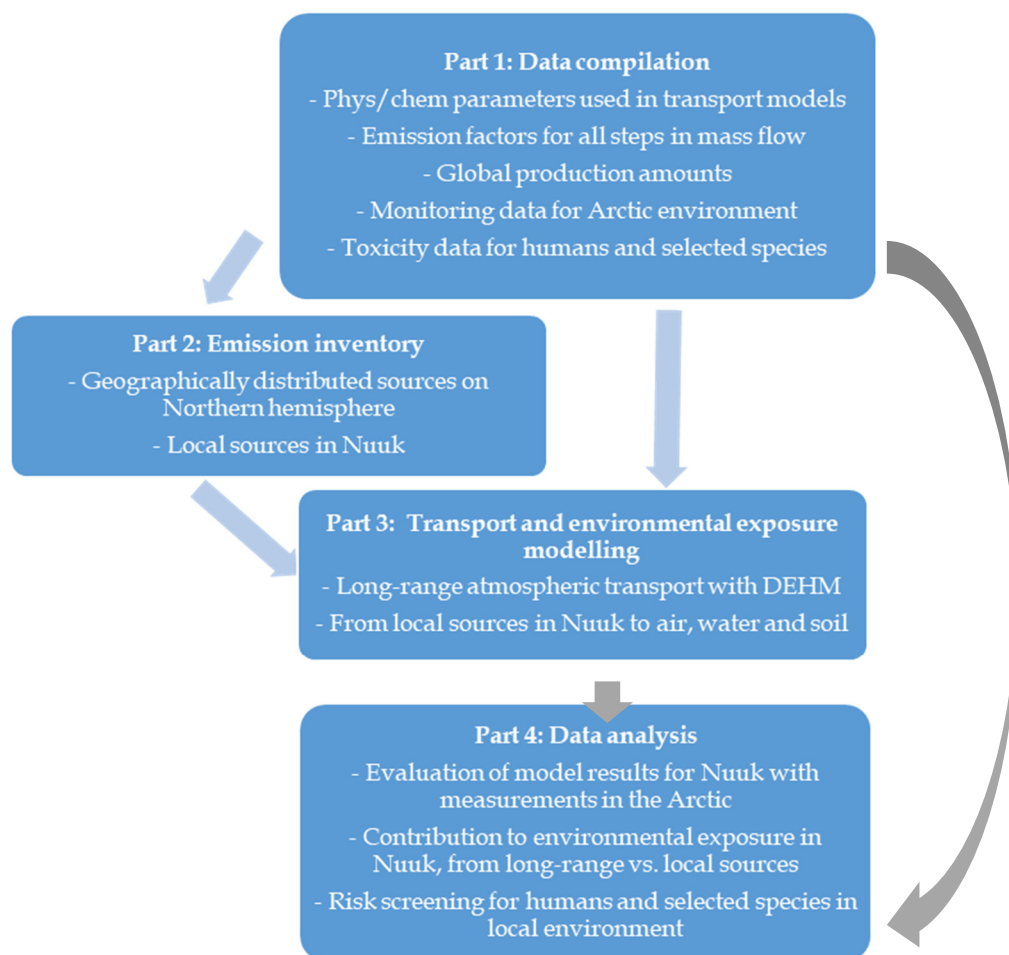


Figure 2.1 The four parts of the project.

Table 2.1 Flame retardants studied in this project.

Acronyme	Name	CAS number	Flame retardant group
TCEP	Tri(2-chloroethyl) phosphate	115-96-8	Organophosphorous
TCPP	Tri(chloroiso-propyl) phosphate	13674-84-5	Organophosphorous
TPHP	Triphenyl phosphate	115-86-6	Organophosphorous
EH-TBB	2-Ethylhexyl-2,3,4,5-tetrabromobenzoate	183658-27-7	Brominated
BEH-TEBP	Bis(2-ethylhexyl) tetrabromophthalate	26040-51-7	Brominated
HBBz	Hexabromobenzene	87-82-1	Brominated
DP	Dechlorane Plus	13560-89-9	Chlorinated

The data compilation (Chapter 3) comprises physical/chemical parameters used for calculating emissions (Chapter 4) from the Northern hemisphere, and atmospheric transport, deposition and exposure in the Arctic with DEHM



(Chapter 5). The data are also used for calculating emissions, transport and environmental exposures in the local environment, from sources in Nuuk (Chapter 6). Production amounts of the compounds and the geographical location of production sites on a global scale are compiled, considering data going back to 2012. If no such more recent data are available, older data are used (Chapters 3 and 4). Data on amounts going into manufacture of plastics and textiles, their indoor and outdoor use, and waste treatment are derived in a mass flow analysis. The Northern hemisphere is divided into nine global regions and the emissions from the different steps in the mass flow analysis are distributed in these using the light intensity as seen from space as a proxy of human activity and use of consumer products (Chapter 4).

Emission factors (EFs) to air, water and soil, for all steps in the mass flow, i.e. from compound production to waste treatment, are derived from the assessments of the brominated flame retardants pentabromodiphenyl ether (pentaBDE), and decabromodiphenyl ether (decaBDE), also representing proxies as no corresponding data are available for the compounds of this study (Chapter 4). EFs are corrected for differing vapour pressures and molecular weights, as well as for varying outdoor temperatures in the nine global regions.

Variations in the physical/chemical parameters, production amounts, and EFs are used in a sensitivity analysis of the emission estimates and the modelled concentrations in Nuuk (Chapters 5 and 6). The contributions to environmental exposure in Nuuk of the seven compounds from long-range transport vs. local sources are assessed based on model results. This also indicates the global regions with highest impact on the occurrence in the environment of Nuuk (Chapter 7).

Measurement data in air, soil, (sea)water and biota (fish: Atlantic cod (*Gadus morhua*), ringed seal (*Puca hispida*)) in the Arctic environment, i.e. Canada, Alaska, Finland, Greenland, Iceland, and Norway, are compiled (Chapter 3) for evaluating and verifying modelled concentrations (Chapters 5 and 6). The estimated environmental concentrations are evaluated in a risk screening, together with compiled toxicity data for human exposure and uptake of FR from air, fish and seal, and exposure to fish from FR uptake from water and biota (Chapter 7).

### 3 Compound characteristics and environmental occurrence

#### 3.1 Physical/chemical parameters

The environmental fate of organic contaminants is determined by their physical-chemical properties such as water solubility, vapour pressure, partition coefficients between different media, reaction rates and environmental half-lives in the different media.

For the modelling of LRT of the selected compounds, the partition coefficients are most important properties, which are applied to parameterize most environmental processes in the Danish Eulerian Hemispheric Model (DEHM). DEHM is described in detail in section 5.1. The air-water partition coefficient,  $K_{AW}$ , is, for example, used to parameterize the wet deposition and gas exchange between air and surface water, and the octanol-air partition coefficient,  $K_{OA}$ , is used to parameterize the distribution between the gas phase and particle phase in air and the gas exchange between air and soil. The partition coefficients are either determined in laboratory experiments or by an estimation method from the molecular structure of the compound.

We have made a literature study to identify possible input parameters to DEHM. There are at least three and for some components up to 20 different notions of values for the partition coefficients, although some are from the same estimation method. There are only few experimental values and the rest are calculated by estimation methods. We have chosen to apply the values by Zhang et al. (2016), which cover all substances of this study and which are estimated using the recognized EPI SUITE from US EPA (2021), see Table 3.1.1. The EPI SUITE estimates are in reasonable agreement with the few available experimental values and the values from other estimation methods. However, the values from a few other estimation methods are different from the values from EPI SUITE by several orders of magnitude (OMs), see section 5.1 for sensitivity study.

Table 3.1.1 Physical-chemical properties applied in DEHM.

	TCEP	TCPP	TPHP	EH-TBB	BEH-TEBP	HBBz
log $K_{AW}$	-5.98	-5.61	-5.79	-3.59	-4.91	-3.06
log $K_{OA}$	7.61	8.50	10.49	12.34	16.86	10.39
log $K_{OW}$	1.63	2.89	4.70	8.75	11.95	7.33
Vapour pressure (Vp) at 20°C (Pa)	3.76E+00	3.76E-03	7.51E-04	7.51E-05	1.89E-07	2.99E-03
OH reaction rate (cm <sup>3</sup> /mol-s)	2.20E-11	4.48E-11	1.08E-11	1.09E-11	2.18E-11	1.15E-14
Surface half-lives:						
$t_{1/2, \text{soil}}$ (hours)	2880	2880	1800	2880	2880	8640
$t_{1/2, \text{water}}$ (hours)	1440	1440	900	1440	1440	4320
$t_{1/2, \text{vegetation}}$ (hours)	1440	1440	900	1440	1440	4320
Snow parameters:						
$SbH$	0.98	1.04	0.98	0.74	1.29	0.26
$K_{hex}$	7.18	7.99	10.12	13.4	17.25	10.56
$\Delta H_{IA}$	-120.9	-130.5	-160.6	-152.5	-214.1	-102.8

Several physical-chemical characteristics, including the partition coefficients are temperature dependent, which has to be taken into account in the parameterizations in DEHM. The partition coefficients provided in the literature all refer to 25°C, unless no temperature was given. It has thus not been possible to determine a temperature dependence of the partition coefficients, and we have therefore not included the temperature dependence in this study.

Apart from the partition coefficients the most important physical-chemical input parameters in DEHM are the reaction rates for degradation in air and the environmental half-lives in the surface media soil, water and vegetation. Compared to the partition coefficients, fewer values are available for these parameters in the literature.

The literature search showed that all estimates for reaction rates with OH radicals in air, apart from one, where the source is not given, were based on estimates from EPI SUITE. All references apply a global average concentration of OH radicals to calculate a half-life in air from the reaction rates. We have applied the reaction rates from EPI SUITE directly because OH is one of the compounds included in DEHM through the O<sub>3</sub> chemical scheme.

Most literature estimates of half-lives in water and soil are also from EPI SUITE, while a few are based on other calculation methods. There is a slightly larger range for these parameters, but we have chosen to apply the values from EPI SUITE for these parameters as well. We assume that the half-life in vegetation is the same as the half-life in water (Hansen et al., 2008).

Several parameters are applied in the snowpack module to describe how much of a compound is depleted by snowfall and how much is retained in the snowpack. A range of descriptors calculated using the Liner Free Energy Relationship (LFER) is applied to parameterize these processes: the electron donor (H acceptor) of the compound (SbH), the hexadecane/air partition coefficient (K<sub>hex</sub>) and the enthalpy for temperature extrapolation ( $\Delta H_{IA}$ ). The three descriptors are calculated from the molecular structure of each compound using a group contribution method developed by Platts et al. (1999).

### 3.2 Measurements in the Arctic

Results from the modelling approaches were compared to measurements of the selected compounds in the Arctic. Ideally, comparisons would focus on Nuuk as the study region of this project, however, no environmental measurements are currently available from Nuuk for the compounds of this study. Some of the selected FRs have been analysed in air and biota samples from Greenland as part of the AMAP Core programme, the Danish-Greenlandic contaminant monitoring programme (Vorkamp et al., 2015). Furthermore, the following two projects have included the BFRs EH-TBB and BEH-TEBP as well as DP: "*New and updated time trends of persistent organic pollutants and their effects on peregrine falcon eggs from Greenland*" (Vorkamp et al., 2018) and "*POP/PBT characterisation of dechlorane plus and novel brominated flame retardants based on data from Greenland*" (Vorkamp et al., 2019b). As part of the ACCEPT cohort study, led by Prof. Eva Cecilie Bonefeld-Jørgensen, EH-TBB and DP have recently been determined in blood samples of the general population in Greenland, including inhabitants of Nuuk (Wielsøe et al., in press). All studies had in common that the levels of FRs were lower than typical POP concentrations, and often below detection limits for some of the samples. While the results for the peregrine falcon eggs (Vorkamp et al., 2018) and the

human blood samples (Wielsøe et al., in press) do not allow direct comparisons with the modelling results of this study, focussing on the atmospheric and the marine environment, data from the AMAP Core programme and the POP/PBT characterisation study are used for comparisons (Vorkamp et al., 2015; 2019b).

As mentioned in Chapter 1, all FRs selected for this study were assessed as CEACs by AMAP (AMAP, 2017). This circumpolar information was subsequently updated in a review article about the state of knowledge regarding the occurrence of current-use and non-regulated FRs in the Arctic, including the seven FRs of this study (Vorkamp et al., 2019a). This article reviews data for the atmospheric environment (including deposition and ice and snow measurements) as well as the terrestrial, freshwater and marine environment. It also presents updated air data for the BFRs and DP from the Canadian high Arctic monitoring station Alert. The majority of comparisons between measured and modelled data are based on the information compiled in this review.

As part of this project, a new literature search has been conducted to update the information reviewed in Vorkamp et al. (2019a). Web of Science was used to search for individual compound names, typical acronyms and CAS numbers as well as the terms “flame retardants”, in combination with “Arctic”. Focus was on the three BFRs and the three OPFRs. This literature search resulted in a list of 17 new publications addressing at least one of the selected compounds. The studies covered all environmental compartments (i.e. atmospheric, terrestrial, freshwater and marine environment) and various sample types, e.g. water, air, sediment, soil and biota. The majority of information was from Svalbard and the Canadian Arctic, with additional data from sea-based studies, Northern Norway and Alaska as well as one additional study from Greenland (Strobel et al., 2018). This study analysed OPFRs in ringed seals and polar bears from East Greenland, including *in vitro* experiments on their degradation. Although not representative of the same study region, data from this updated literature search were used in the comparisons of modelled results and measurements, as part of an evaluation of the model performance. These comparisons are described in more detail in Chapters 5 and 6.

## 4 Mass flows and emissions

### 4.1 Mass flow and uses

The mass flow of a FR follows the scheme in Figure 4.1.1.

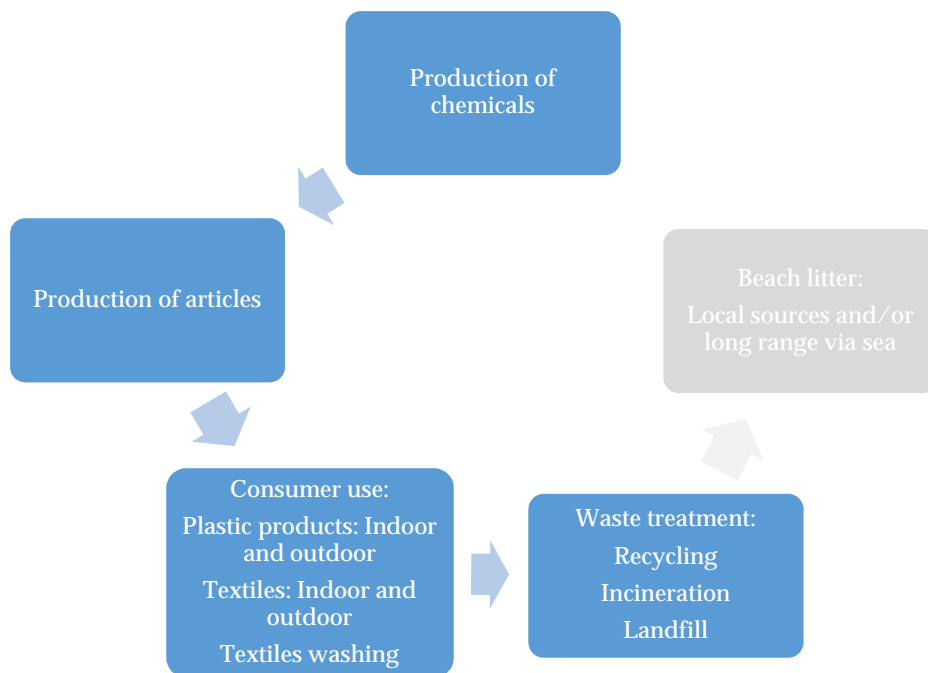


Figure 4.1.1 Mass flow of flame retardants with activities, from ECHA (2014), that potentially lead to FR emissions to the environment on a global and local scale. Beach litter is only included qualitatively (see Chapter 6).

The activities that lead to emissions are taken from the EU risk assessment report for decaBDE (ECHA, 2014) and represent the chain of potential activities that can lead to emissions of the FRs to air, water or soil, both on a global and a local scale. The use and emission characteristics of decaBDE, described in ECHA (2014), are applied for the BFRs, OPFRs and DP in this project. This is a valid assumption, as the more specific uses and emission pathways are similar for all FRs, see Table 4.1.1. All FRs have in common that they are mainly used in applications in the plastics, electrical/electronic applications and textile industries. They are additive FRs, i.e. they are physically combined with the material being treated rather than chemically bound (as in reactive FRs). This means that they have the potential to be released from the material to the environment through leaching and evaporation (Stapleton et al. 2008; Lai et al., 2015; Marklund et al., 2005; ECHA, 2014).

The occurrence of FRs at various concentrations in different environments is attributed to multiple factors such as compound and source characteristics, emission rates, chemical fate, and indoor/outdoor uses (Maddela et al. 2020; Saito 2007). Production of chemicals and articles, consumer use, both indoor and outdoor, are important activities that can lead to emissions to the environment where FRs emitted into the indoor environment may reach the outdoor environment through ventilation, cleaning and disposal of waste at dumpsites (ECHA, 2014; Wei et al. 2015; Hartmann et al., 2004). Emissions



from waste treatment can occur as small particles of a polymer product containing the FR (ECHA, 2014). These particles are primarily released to the industrial/urban soil compartment, but they may also end up in air or sediment. End-products with outdoor uses are most likely to be sources of this waste, where releases can occur over the lifetime of the product due to weathering and wear. This type of waste can also be generated during disposal of all types of plastic products. Surface water and groundwater are mainly influenced by treated and untreated wastewater discharges (Wei et al., 2015). Coarse particles with FRs can enter the soil or water systems and further leach into groundwater or react with biota, while fine particles and fumes can be transported over long distances (Li et al., 2019).

Monitoring studies of OPFRs in air and precipitations in remote areas and over the global oceans implied that certain OPFRs were subject to LRT and could reach the Arctic and Antarctica (Wei et al., 2015), despite their reported short atmospheric half-lives (Wei et al., 2015). Although OPFRs have higher emission potentials compared to BFRs and DP, due to their higher vapor pressures, the potential for atmospheric LRT is documented for the latter two and the transport pathways described above are assumed to be reasonable to use for all FRs in this project.

In order to calculate emissions for the specific activities in Figure 4.1.1 a distribution of FR masses into plastic and textiles and fractions of textiles that are washed, as well as the fractions of plastics and textiles that go into the different waste treatments, is needed. Fractions given for decaBDE (ECHA, 2014) are used for all FRs with adjustments for EH-TBB and BEH-TEBP where it is assumed that the fraction that is used for plastics is higher, and the fraction of plastics with outdoor applications is higher, compared to the other FRs, see Table 4.1.2.

Table 4.1.1 Uses, applications and release patterns of the OPFRs, BFRs and DP.

Acronym	Use and applications	Release patterns
TCEP	TCEP is widely used as a FR and additive plasticizer and viscosity regulator with FR properties in various domestic products, e.g. plastics (e.g. polyurethane, polyesters, polyvinyl chloride and other polymers), textiles, furniture, electronics, soft foams, PU mattresses, PUF fillers, etc. TCEP is also used in building materials, e.g. paints, wood preservation coatings, glass fibre wallpaper, acoustic ceiling coatings, etc., in baby products, e.g. nursing pillows, portable cribs, baby carriers, etc. It is used in other products such as flexible foams for automobiles and furniture, and in rigid foams used for building insulation in rigid polyurethane and polyisocyanurate foams, carpet backing, flame-laminated and rebonded flexible foam, flame-retardant coatings, most classes of thermosets, adhesives, cast acrylic sheet, and wood-resin composites such as particle board. In addition to the use in consumer articles, widespread uses are by professional workers in coating products and in offshore mining and building and construction work. It is the most common chlorinated OP present in the flexible and rigid polyurethane foam (PUF) (Maddela et al., 2020; Ingerowski et al., 2001; CEC, 2015; <a href="https://pubchem.ncbi.nlm.nih.gov/compound/8295#section=Use-and-Manufacturing">https://pubchem.ncbi.nlm.nih.gov/compound/8295#section=Use-and-Manufacturing</a> ; <a href="https://echa.europa.eu/da/substance-information/-/substanceinfo/100.003.744">https://echa.europa.eu/da/substance-information/-/substanceinfo/100.003.744</a> ). Concentrations of TCEP are e.g. 68.0 mg/g in acoustic ceiling coatings, 19.8 mg/g in soft PUFs, 32.0 mg/g in PUF fillers, 2.40 mg/g in glass fiber wall paper, 1.00 mg/g in wood preservation coatings, and 0.89 mg/g in polyurethane mattresses, respectively (Ingerowski et al., 2001).	TCEP release to the environment can occur from industrial use in the production of articles, consumer use of articles, and from outdoor use in long-life materials with low release rate (e.g. construction and building materials). Other release to the environment is likely to occur from outdoor use resulting in inclusion in materials (e.g. binding agent in paints and coatings or adhesives) ( <a href="https://echa.europa.eu/da/substance-information/-/substanceinfo/100.003.744">https://echa.europa.eu/da/substance-information/-/substanceinfo/100.003.744</a> ).
TCP	TCP is used as a FR, mainly in polymers such as polyurethane foams. It can be found in vehicles, mechanical appliances and electrical/electronic products (e.g. computers, cameras, lamps, refrigerators, washing machines), fabric, textile, and leather products; fillers; building construction materials. TCP can be found in plastic-based consumer articles, e.g. food packaging and storage, toys, mobile phones, rubber (e.g. tyres, shoes, toys), stone, plaster, cement, glass or ceramic (e.g. dishes, pots/pans, food storage containers, construction and isolation material), and apparel (e.g. clothing, mattress, curtains or carpets, textile toys), paper (e.g. tissues, feminine hygiene products, nappies, books, magazines, wallpaper) and wood (e.g. floors, furniture, toys), adhesives, sealants, binding agents; insulating materials; paints, lacquers and varnishes; intermediates; foam seating and bedding products; insulating foam. TCP is used by professional workers (widespread uses), in formulation or re-packing, at industrial sites laboratory chemicals and in manufacturing. ( <a href="https://echa.europa.eu/da/substance-information/-/substanceinfo/100.033.766">https://echa.europa.eu/da/substance-information/-/substanceinfo/100.033.766</a> ; <a href="https://pubchem.ncbi.nlm.nih.gov/compound/26176#section=Uses">https://pubchem.ncbi.nlm.nih.gov/compound/26176#section=Uses</a> ). Concentrations of TCP are e.g. 0.5-2.2 wt-% in different furnishings (e.g., couches, chairs, mattress pads, pillows and insulation foam from a laboratory-grade dust sieve) (Stapleton et al., 2009), 1-20 wt-% in wallpaper (Ni et al., 2007), 180 mg/g in PUF fillers, 1.10 mg/g in glass fiber wall paper, 0.15 mg/g in wood preservation coatings, 1.50 mg/g in polyurethane mattresses, 13.1 mg/g in polyurethane carpet backing and 0.22 mg/g in floor sealing material, respectively (Ingerowski et al., 2001). FRs are either incorporated in textile fibers, from manufacture, or subsequently added to the cover/upholstery/foam in	TCP release to the environment is likely to occur from indoor use in long-life materials with low release rates (e.g. flooring, furniture, electronic equipment, toys, construction materials, curtains, foot-wear, leather products, paper and cardboard products) and outdoor use in long-life materials with low release rate (e.g. metal, wooden and plastic construction and building materials). Releases can occur from outdoor use in long-life materials with high release rate (e.g. tyres, treated wooden products, treated textile and fabric, brake pads in trucks or cars, sanding of buildings (bridges, facades) or vehicles (ships)) and indoor use in long-life materials with high release rate (e.g. release from fabrics, textiles during washing, removal of indoor paints). Releases can also occur from industrial uses, such as manufacturing of the substance, formulation of mixtures, formulation in materials, in the production of articles, industrial abrasion processing with low release rate (e.g. cutting of textile, cutting, machining or grinding of metal) and industrial abrasion processing with high release rate (e.g. sanding operations or paint stripping by shot-blasting). Indoor use and outdoor use can result in TCP environmental emissions and inclusion into or onto materials (e.g. via binding agent in paints and coatings

	e.g. furniture, curtains, mattresses and carpets (DK-EPA, 2014). TCPP was found in 1 out of 3 office chairs (0.46 wt-%), in 2 out of 3 armchairs (0.44 wt-%), and no occurrence in 8 mattresses (DK-EPA, 2014).	or adhesives) ( <a href="https://echa.europa.eu/da/substance-information/-/substanceinfo/100.033.766">https://echa.europa.eu/da/substance-information/-/substanceinfo/100.033.766</a> ).
TPHP	<p>TPHP is used in polymers and for the manufacture of plastic products, textile, leather or fur, rubber products, electrical, electronic and optical equipment and machinery and vehicles. It can be found in consumer products such as electrical/electronic products (e.g. computers, cameras, lamps, refrigerators, washing machines), and in products with material based on fabrics, textiles and apparel (e.g. clothing, mattress, curtains or carpets, textile toys), paper (e.g. tissues, cosmetics and personal care products, nappies, books, magazines, wallpaper), rubber (e.g. tyres, shoes, toys) and plastic (e.g. food packaging and storage, toys, mobile phones), and in vehicles and machinery and mechanical appliances. TPHP is used in foam seating and bedding products; lubricants and greases; paints and coatings; photographic supplies, film, and photo chemicals; and in other plastic and rubber products. TPHP is used as FR agent, plasticizer for cellulose acetate and nitrocellulose; plasticizer in lacquers and varnishes; plasticizer in automobile upholstery, as a fireproofing agent, as a noncombustible substitute for camphor in celluloid, in adhesives and sealants and cosmetics and personal care products, and for impregnating roofing paper; found as one component of lubricating oil and hydraulic fluids. Industry uses are as FR in paint additives and coating additives and in adhesives and sealants; plasticizers; solvents (which become part of product formulation or mixture), and uses by professional workers (widespread uses), are in formulation or re-packing, at industrial sites and in manufacturing (<a href="https://echa.europa.eu/da/substance-information/-/substanceinfo/100.003.739">https://echa.europa.eu/da/substance-information/-/substanceinfo/100.003.739</a>; <a href="https://pubchem.ncbi.nlm.nih.gov/compound/8289#section=Use-and-Manufacturing">https://pubchem.ncbi.nlm.nih.gov/compound/8289#section=Use-and-Manufacturing</a>).</p> <p>TPHP was detected as the most prevalent OPFR in consumer products on the Japanese market in 2008, with concentrations ranging from 0.87 to 14,000, 0.56 to 2,600, 820 to 840, 0.63 to 12.0, 5.30 to 8.70, and 0.23 to 1.80 µg/g for LCD-TV, laptop computers, curtains, electrical outlets, insulation boards wallpaper and building materials, respectively (Kajiwara et al., 2011).</p> <p>TPHP varied from 0.3 to 10 wt-% for the plastic material used in computer video display units (Carlsson et al., 1997). In turbo oil, engine and accessory oil, and jet engine oil from an airport the levels of TPHP were reported as 6.10, 8.90 and 1.90 µg/g, respectively (Marklund et al., 2005). The occurrence of TPHP in the waste oil from cars, lorries and road-making machines varied from 0.80 to 1.90 µg/g (Marklund et al., 2005).</p>	<p>The release of TPHP to the environment is likely to occur from indoor use in long-life materials with low release rates (e.g. flooring, furniture, toys, construction materials, curtains, foot-wear, leather products, paper and cardboard products, electronic equipment), and outdoor use in long-life materials with low release rate (e.g. metal, wooden and plastic construction and building materials). Releases to the environment are also likely to occur from indoor use and outdoor use resulting in inclusion into or onto a materials (e.g. binding agent in paints and coatings or adhesives). Other releases to the environment of this substance is likely to occur from indoor use (e.g. machine wash liquids/detergents, automotive care products, paints and coating or adhesives, fragrances and air fresheners) and outdoor use resulting in inclusion in materials (e.g. binding agent in paints and coatings or adhesives). Release to the environment can also occur from manufacturing of the substance, production of articles and from formulation in materials and formulation of mixtures (ECHA: <a href="https://echa.europa.eu/da/substance-information/-/substanceinfo/100.003.739">https://echa.europa.eu/da/substance-information/-/substanceinfo/100.003.739</a>).</p>
EH-TBB	<p>Consumer and Industry uses of EH-TBB are as FR in foam seating and bedding products (<a href="https://pubchem.ncbi.nlm.nih.gov/compound/71316600#section=Use-and-Manufacturing">https://pubchem.ncbi.nlm.nih.gov/compound/71316600#section=Use-and-Manufacturing</a>).</p> <p>EH-TBB og BEH-TEBP are replacements for PentaBDE, which is the major constituent in PBDE formulations in the US (Stapleton et al., 2008). In the US PBDEs were widely used in textiles, plastics and polyurethane foam. PBDE treated articles were used in the home, in business settings, and in the transportation sector. Articles which were often treated with PBDEs include carpets, upholstery fabric, cushions,</p>	Probably similar to BEH-TEBP below.

	and plastics used as components in electrical appliances and equipment (US EPA, 2009). PentaBDE were added in concentrations between 5 and 30 wt-%. Major uses were in flexible polyurethane foam for furniture and upholstery, and other uses include additives in epoxy resins, phenolic resins, unsaturated polyesters and textiles, and in electrical and electronic equipment (EU, 2000)..	
BEH-TEBP	<p>BEH-TEBP is mainly used as FR plasticizer in polyvinylchloride, neoprene and electrical coatings. It is an ingredient in Firemaster 550 and BZ54 that are used in polyurethane foam products. It is used in plastic products and rubber consumer articles, such as electrical/electronic products (e.g. computers, cameras, lamps, refrigerators, washing machines); fabric, textile, and leather products; foam seating and bedding products; other plastic and rubber products, and in adhesives, sealants and polymers. It can be found in products with material based on rubber (e.g. tyres, shoes, toys) and plastic (e.g. food packaging and storage, toys, mobile phones). BEH-TEBP is used by professional workers (widespread uses), in formulation or re-packing and at industrial sites, in building &amp; construction work and scientific research in machinery, mechanical appliances and development (<a href="https://echa.europa.eu/da/substance-information/-/substanceinfo/100.043.099">https://echa.europa.eu/da/substance-information/-/substanceinfo/100.043.099</a>; <a href="https://pubchem.ncbi.nlm.nih.gov/compound/117291#section=Use-and-Manufacturing">https://pubchem.ncbi.nlm.nih.gov/compound/117291#section=Use-and-Manufacturing</a>).</p> <p>EH-TBB og BEH-TEBP are replacements for PentaBDE, see above for EH-TBB.</p>	The release of BEH-TEBP to the environment is likely to occur from indoor use in long-life materials with low release rate (e.g. flooring, furniture, toys, construction materials, curtains, foot-wear, leather products, paper and cardboard products, electronic equipment, and from outdoor use in long-life materials with low release rate (e.g. metal, wooden and plastic construction and building materials). Other release to the environment is likely to occur from indoor use (e.g. machine wash liquids/detergents, automotive care products, paints and coating or adhesives, fragrances and air fresheners) and outdoor use. Other release to the environment can occur from formulation of mixtures, formulation in materials, production of articles and in processing aids at industrial sites can occur from industrial use of articles where it is not intended to be released and where the conditions of use do not promote release ( <a href="https://echa.europa.eu/da/substance-information/-/substanceinfo/100.043.099">https://echa.europa.eu/da/substance-information/-/substanceinfo/100.043.099</a> ).
HBBz	HBBz is used as FR in polymers ( <a href="https://pubchem.ncbi.nlm.nih.gov/compound/6905#section=Uses">https://pubchem.ncbi.nlm.nih.gov/compound/6905#section=Uses</a> ).	Probably similar to BEH-TEBP.
DP	DP is applied as a non-reactive and non-plasticizing FR in mostly thermoplastic materials like polyethylene, polyvinyl acetate and polypropylene in for instance electronic applications and wire and cable applications. The DP contents in these products range from 5 % to 35 % (ECHA, 2007). It is also used in minor applications in polyester and epoxy resins, for example self-extinguishing phenolic resin laminated paper (ECHA, 2007). It is assumed that the main use of DP in plastic is similar to the previous use of DecaBDE (ECHA, 2014).	The mass flow and emissions of DP can be attributed to the activities in Figure 4.1.1, as shown in Hansen et al. (2020).

Table 4.1.2 Distribution in % of FRs fractions in plastics and textiles, and fractions that are subject to the different waste treatment procedures. From ECHA (2014) with adjustments for EH-TBB and BEH-TEBP.

Use distribution	DecaBDE (ECHA, 2014) used for TCEP, TCPP, TPHP, HBBz and DP		EH-TBB and BEH-TEBP	
	Plastics:	Textiles/furniture:	Plastics:	Textiles/furniture:
Fraction of chemical that is used in plastics or textiles:	52	48	90	10
Fraction of plastics/textiles with FR:	48	52	48	52
Fraction of plastics/textiles with outdoor applications:	0,1	5	10	5
Fraction of textiles subject to washing:	-	2	-	2
Fraction of recycled plastics/textiles:	19	0	19	0
Fraction of incinerated plastics/textiles:	37	50	37	50
Fraction of landfilled plastics/textiles:	44	50	44	50

## 4.2 Production

For the quantification of emissions related to the production of the selected FRs, it was relevant to collect information on production sites and quantities, preferably over time. This search was approached by compound and region, i.e. for North America (Canada, USA, Mexico), Europe (including country-specific information if available) and Asia, with some details for China and Japan. Production details were sought for the three BFRs and three OPFRs, while information on DP was used from Hansen et al. (2020).

Information on FR production in the USA was retrieved from the database Chemical Data Reporting (CDR)<sup>2</sup>, which generally was the most comprehensive source of information. However, the following four caveats have to be noted:

- i) The CDR database only included data through 2015, although an update through 2020 had been announced for the project phase. The data collection thus covered the period 2010-2015.
- ii) The production amounts frequently covered production and import. In some cases, it was specified whether the data related to manufacture or import, or both, but this was not always the case.
- iii) Some production volumes were confidential business information, for some years or the entire period, i.e. 2010-2015.
- iv) Some companies changed names over time or were part of mergers or acquisitions.

Besides company-specific data, the CDR database also reports National Aggregated Production Volumes. For TCEP, specific numbers were given, while orders of magnitude were provided for the other selected compounds except HBBz, for example approximately 23,000-45,000 tonnes of TCPP in 2015 (converted from US pounds). HBBz was the only FR with no information in the CDR database. According to Guerra et al. (2012), HBBz had been produced in the Great Lakes region, but production stopped in the 1980s. Additional information regarding US production and import was from US EPA (2015a; b), generally referring to the CDR database and thus mainly used for cross-checking. For TPHP, another US EPA report provided complementary data on National Aggregated production Volumes to those extracted from the CDR database (US EPA, 2019).

<sup>2</sup> <https://chemview.epa.gov/chemview>



Information on potential FR production in Canada and Mexico was sparse and mainly retrieved from only one report (CEC, 2015). With regard to the USA, this report referred to the CDR database and was thus also mainly used for cross-checking of information. Experts were not aware of any production of TCEP, TCPP, TPHP, EH-TBB or BEH-TEBP in Canada or Mexico, while HBBz was not included in the report (CEC, 2015). For Mexico, annual import quantities were stated of TCPP and TPHP, for five and eleven sites, respectively, with a maximum of 240 tonnes for TCPP in 2013 and generally increasing from 2009 to 2013 (CEC, 2015). For Canada, import volumes were given for EH-TBB and BEH-TEBP in 2011, of 10-100 tonnes each (Canada, 2019b).

The interpretation of production data from Europe was complicated by the fact that member states of the European Union (EU) had changed over time, meaning that information on FR import and production in the EU did not always refer to the geographical area. For example, production of TCEP in the EU was reported for 2004, but not for 2000/2001 (EU, 2009). However, in 2000/2001 import of TCEP into the EU originated from Russia and Poland. This may suggest that possibly, production of TCEP in 2004 was in Poland, having joined the EU in 2004.

For TCPP, four producers were mentioned in the EU as of 2000, three of these located in Germany and one in the UK, manufacturing approximately 38,000 tonnes of TCPP per year (EU, 2008). TPHP is registered with a production/import of 1000-10,000 tonnes in the EU, described as “manufacture” in the dossier<sup>3</sup>. Search of commercial vendors identified companies in Germany, the Netherlands and in the UK. However, these may be traders rather than producers. Recent and future updates on EU information may not include UK data. No registration in the EU could be found for EH-TBB or BEH-TEBP, while HBBz was pre-registered<sup>4</sup>. According to Verreault et al. (2007), HBBz was not a produced chemical in the EU. Both TPHP and BEH-TEBP are included in the Community Rolling Action Plan, which may generate more information on production and use. Regarding compound uses, the SPIN database<sup>5</sup> (Substances in Preparations in Nordic Countries) is an informative resource on use types and quantities in the Nordic countries. However, global approaches are needed in an Arctic context, which currently result in many data and information gaps.

Thus, it was not possible to retrieve detailed information on FR production in Asia although the literature suggests that substantial FR production takes place in Asia, especially in China: CEC (2015) mentioned China, Japan and South Korea as main users of flame retardants, but did not provide any quantitative data. The production of OPFRs was described as being on the rise in China. It was described as fragmented, with small to medium-size manufacturers (CEC, 2015). According to this report, Japan was assumed to produce chlorinated FRs for the domestic market, while South Korea was assumed to import chlorinated FRs. Internet searches revealed a multitude of manufacturers and/or suppliers in China for EH-TBB and BEH-TEBP. For TCCP and TPHP, a manufacture and/or supplier was found in the towns Zhengzhou and Zhenjiang, respectively. However, links on these web pages indicated

<sup>3</sup> <https://echa.europa.eu/information-on-chemicals/registered-substances/-/dis-reg/substance/100.003.739>

<https://echa.europa.eu/registration-dossier/-/registered-dossier/15972/3/1/2>

<sup>4</sup> <https://echa.europa.eu/substance-information/-/substanceinfo/100.001.613>

<sup>5</sup> <http://spin2000.net/>

that multiple other manufacturers and/or suppliers might exist in China. Covaci et al. (2011) reported the production of 600 tonnes/year of HBBz in Qindao (China) and its marketing by a company in Hangzhou (China). Better insights into Chinese manufacture and trade structures as well as knowledge of Chinese will be needed for an in depth analysis of former and present Chinese production of FRs.

Several sources have reported the production of HBBz in Japan (e.g. Covaci et al., 2011), with an annual production of 350 tonnes for the years 1994-2001 (Watanbe et al., 2003). A summary of the distribution of global production amounts, based on this overview containing several caveats and information gaps, is given in Figure 4.2.1.

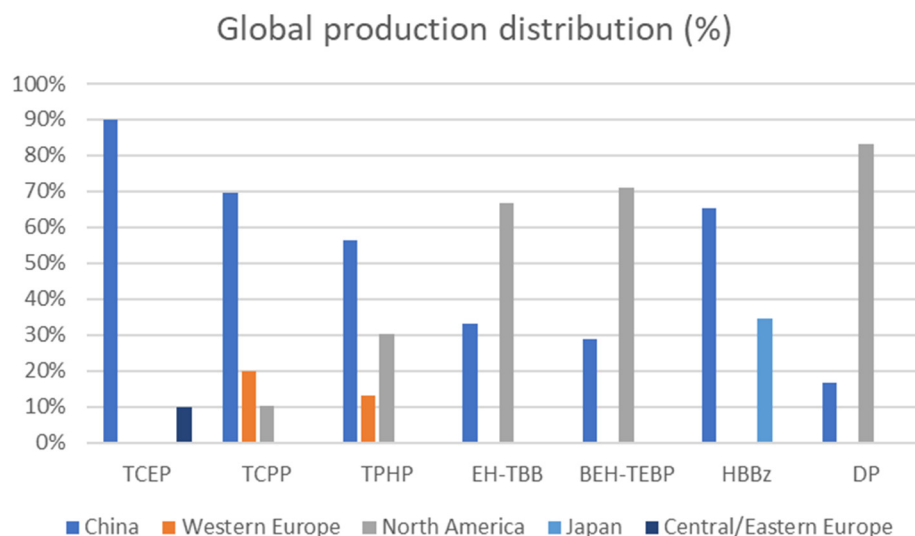


Figure 4.2.1 Global production distribution in %.

## 4.3 Emissions

### 4.3.1 Emission factors (EFs) from the literature

A top down approach based on the produced amount of each compounds is used to calculate the emitted amounts of compounds in every mass flow activity, see Figure 4.1.1. EFs in units “g compound emitted per g compound present in product or activity” ensures that the entire produced amounts are accounted for. The information needed for a bottom-up approach could be emission rates in units “g compound emitted per surface area or per mass of product or per product unit”, which requires the number and types of products and activities. Such detailed information is not available on a global scale.

A literature search revealed that there were no data available on EFs for the seven compounds that are applicable for the steps in the mass flow in Figure 4.1.1. The literature was then searched for flame retardants with similar molecular structures. For the BFRs, EFs from the ECHA assessment report on decabromodiphenyl ether (decaBDE) (ECHA, 2014) cover all activities in the mass flow, except for the production of chemicals. For production, the EF for pentabromodiphenyl ether (pentaBDE) in EC (2000) was used.

All compounds considered here are “additives which are notionally soluble in polymers and potentially volatile” (OECD, 2009). The three OPFRs and three BFRs are used in plastics, electronics and in textiles as well as in other

applications, which basically are similar to the uses for decaBDE and pentaBDE. Characteristically for FRs, they are added to polymers via physical mixing rather than chemical bonding; consequently, they can easily be released into the immediate surroundings by abrasion, diffusion, leaching, volatilization, etc., with possible long-range transport (Maddela et al., 2020; Wei et al., 2015). Similar uses in products and activities, both indoor and outdoor, similar waste management procedures and non-chemical bonding in the polymer matrix are expected. It is therefore assessed to be a reasonable assumption to perform an analogous emission analysis with similar release scenarios as done for decaBDE and pentaBDE in ECHA (2014) and EC (2000), respectively.

The EFs from ECHA (2014) and EC (2000) are compiled in Table 4.3.1.1 and are used as proxies for the compounds in this project. This approach is similar to the one used for modelling the long-range transport of dechlorane plus (DP) in Hansen et al. (2020). EFs are given for different environmental compartments for use in plastic products and textiles, and for each activity in the mass flow. In this project, it is assumed that EFs for electronics are similar to the EFs for plastic products in the use and waste treatment stages. Different EFs are reported and to cover this variation a low and a high emission scenario are defined from the available data.

Table 4.3.1.1 Emission factors (EFs) for decaBDE and pentaBDE (production only) from ECHA (2014) and EC (2000), respectively. A high and a low emission scenario are defined for the Northern hemisphere and the local scale (Nuuk). For further information on the local sources, see Chapter 6.

	Northern hemisphere		Local (Nuuk)		Environmental compartment
	Low emissions (g/t)	High emissions (g/t)	Low emissions (g/t)	High emissions (g/t)	
Production of chemical	0 <sup>1)</sup>	0.0005	-	-	air
	500	3000 <sup>1)</sup>	-	-	water
	-	-	-	-	soil
Production of articles (incl formulation and polymer processing)	8	24	-	-	air
	2	117	-	-	water
	5	0	-	-	soil
Consumer use of plastic products, indoor	24	500	24	500	air
	-	500	-	500	water
Consumer use of plastic products, outdoor	500	500	500	500	air
	16000	16000	16000	16000	water or soil
Consumer use of textiles, indoor	32	500	32	500	air
	-	500	-	500	water
Textiles washing	500	500	500	500	water
Consumer use of textiles, outdoor	500	500	500	500	air
	16000	16000	16000	16000	water or soil
Waste recycling	0.013	43	-	-	air
Waste incineration	0	1.80	.. <sup>2)</sup>	1.80 <sup>2)</sup>	air
Waste landfill:					
- Particle emissions from unloading	0	10	.. <sup>3)</sup>	.. <sup>3)</sup>	air
- Gas and particle during landfill operations	-	7	.. <sup>3)</sup>	.. <sup>3)</sup>	air
	0.68	0.68	.. <sup>3)</sup>	.. <sup>3)</sup>	water
Beach litter <sup>4)</sup> :					
- Local sources and/or long range via sea	(16000)	(16000)	(16000)	(16000)	water

- Not relevant.

<sup>1)</sup> TGD (2003), IC = 11, polymer industry.

<sup>2)</sup> In Nuuk, some plastics, e.g. fish nets/trauls, are packed and sent for recycling at Plastiks in Denmark. Other plastics are incinerated in the local incineration plant together with furniture, textiles, carpets etc. Electronics are not incinerated.

<sup>3)</sup> In Nuuk, insignificant amounts are deposited. Electronics are not deposited.

<sup>4)</sup> Beach litter is only considered qualitatively for Nuuk (Chapter 6).

OPFRs are rather different from decaBDE and pentaBDE in terms of molecular weights and physical-chemical properties, and potentially emission susceptibilities. Due to their moderate vapour pressure ( $V_p$ ) the release of OPFRs from consumer products and building materials are likely to occur (Wei et al., 2015). For example, Carlsson et al. (1997) showed that TPHP could be continuously emitted into indoor air during the normal operations of a computer. Maddela et al. (2020) found that there was a significant correlation among TCEP and other OPFRs, i.e. TnBP, TCPP and TDCPP, in indoor dust, implying that these OPFR congeners exist in the same predominant sources with similar emission properties and/or pathways. To investigate existing data for the three OPFRs, and to understand the variations in EFs or emission rates between different compounds, the literature search found the results shown in Table 4.3.1.2. The studies addressed consumer products, such as electronics, wall linings and furniture materials. Furthermore, EFs are shown from a study on thermal treatment on recycling in a closed chamber, i.e. board baking

or cooking and open burning (uncontrolled incineration) of OPFR-bearing typical e-wastes, such as plastic casings and printed circuit boards (Li et al., 2019). Table 4.3.1.2 shows that these EFs for OPFRs are in the same order of magnitude as EFs of the sum of 39 polybrominated diphenyl ethers ( $\Sigma 39\text{PBDE}$ ), found in a similar study with thermal treatment and open burning of plastics and printed circuit boards (Li et al., 2018). Apart from this, the data do not permit a comparison of EFs or emission rates between OPFRs individually or to BFRs that can be applied in this project to derive EFs.

Table 4.3.1.2 Emission rates and emission factors (EFs) found in the literature for the investigated OPFRs, as well as the OPFR bisphenol A bis(diphenyl phosphate) (BDP) and a study on the sum of PBDEs.

Chemical name	Material/process	Emission rate, emission factor, emission coefficient	Unit	Other	Reference
BDP	Personal computers	44	ng/unit/h	Desktop PCs in operation	OECD (2019)
TCEP	Monitors	<5 - 34	ng/unit/h	Unit specific emission rate	OECD (2019)
	TV set	1.4 (nd – 13.0)	µg/m²/h	Migration rates to solid extraction disc	Saito et al. (2007)
	Plastic casings (thermal treatment)	32	µg/g	Mass emitted from one unit mass of treated e-waste	Li et al. (2019)
	Printed circuit boards (thermal treatment)	14			
	Plastic casings (open burning)	2.2			
	Printed circuit boards (open burning)	1.4			
TCPP (TCIPP)	Personal computers	24	ng/unit/h	Desktop PCs in operation	OECD (2019)
	Monitors	<5 - 2465	Unit specific emission rate		
	Insulation board	0.35/0.21/0.7/0.6	µg/m²/h	Calculated from emission tests	
	Upholstery stool	28/36/77			
	Upholstery foam	77			
	Assembly foam (rough, new)	70			
	Assembly foam (smooth new)	50			
	Assembly foam (rough, stored)	130/140			
	Assembly foam (smooth, stored)	70/50			
	Mattress	0.012			
	PUR foam (porous surface, new)	70		Building materials	
	PUR foam (porous surface, old)	140			
	PUR foam (smooth surface, new)	50			
	PUR foam (smooth surface, old)	50			
	Wallpaper material	262.3 ~ 2166.8			
	TV set	0.42 (nd – 1.7)	µg/m²/h	Migration rates to solid extraction disc	Saito et al. (2007)
	Plastic casings (thermal treatment)	0.28	µg/g	Mass emitted from one unit mass of treated e-waste	Li et al. (2019)
	Printed circuit boards (thermal treatment)	132			
	Plastic casings (open burning)	3.3			
	Printed circuit boards (open burning)	2.2			
TPHP (TPP)	Personal computers	25	ng/unit/h	Desktop PCs in operation	OECD (2019)
	Printed circuit board	496		Unit specific emission rate	
	Desk top computer system	25 - 85			
	Monitors	23 ~ 133			
	Vinyl chloride products	5.0 (mean), 3.7 (min), 6.0 (max)	µg/m²/h	Micro chamber method testing	



	Electrical and electronic products at an in-service temperature of 60 °C	4.9			
	Electrical and electronic products at an in-service temperature of 60 °C	0.0004	-		
	Computer monitor	0.69 (nd – 20.7)	µg/m <sup>2</sup> /h	Migration rates to solid extraction disc	Saito et al. (2007)
	TV set	0.33 (nd – 6.7)			
	Plastic casings (thermal treatment)	20	µg/g	Mass emitted from one unit mass of treated e-waste	Li et al. (2019)
	Printed circuit boards *(thermal treatment)	52			
	Plastic casings (open burning)	3.9			
	Printed circuit boards (open burning)	31			
Σ39PBDE	Plastics and printed circuit boards (thermal treatment)	0.82–160	µg/g	Mass emitted from one unit mass of treated e-waste	Li et al. (2018)
	Plastics and printed circuit boards (open burning)	n.d.-91			

#### 4.3.2 Emission propensities and corrections of EFs

The emission rate of FRs in indoor and outdoor environments is attributed to several factors such as compound and source characteristics, binding of the compound in the material, time and indoor/outdoor conditions etc. (e.g. Maddela et al. 2020, Saito 2007, Ni et al., 2007).

One of the important factors for emissions of OPFRs and other compounds is the temperature. In one study an inverse linear relationship between temperature and emission rate of TCPP has been reported (Ni et al., 2007). However, other results disagree with this finding, as the emissions from a piece of foam increased with a factor of 12 when going from 23 °C to 55 °C (Liang et al., 2019). Similarly, the concentrations of different OPFRs, such as TCPP from PVC wallpapers significantly increased in the chamber air with an increase in temperature. Furthermore, the emission rates and the concentrations of OPFRs both indoors and outdoors increased during the summer and heating seasons (Maddela et al., 2020).

The relatively high EFs and use amounts for indoor use of consumer products entail that emissions from indoor use are potentially predominant. According to OECD (2019) indoor emissions from the surface of plastic products are probably similar throughout the world, and considering the relatively low variations in indoor temperatures it is assumed that indoor temperatures and service temperatures of consumer products indoor are constant, and identical EFs can be applied for indoor use in all parts of the world in relation to temperature.

Conversely, outdoor temperatures vary considerably during the year and across the world, as seen in Figure 4.3.2.1. EFs correlate significantly with vapour pressure (Vp) (Maddela et al., 2020). To account for the influence of temperature on the variation in EFs, the temperature dependence of Vps are considered. OECD (2019) evaluated different equations to estimate the temperature dependence of the Vp of FRs, by comparing measured and estimated Vps, and concluded that the Yaws estimation equation (Yaws, 1997) provided the most correct correlation within a range of normally occurring temperatures:

$$\log_{10}Vp = A + \frac{B}{T} + C \cdot \log_{10}T + D \cdot T + E \cdot T^2 \quad \text{Eq. 4.3.2.1}$$

where, Vp = vapour pressure (mm Hg), T = temperature (K), and A, B, C, D, and E are coefficients.

OECD (2019) has listed the coefficients for a number of plastic additives, including TPHP, i.e.  $A = 28.0972$ ,  $B = -5668.4$ ,  $C = -5.9768$ ,  $D = -3.1567 \cdot 10^{-9}$ ,  $E = 1.0751 \cdot 10^{-12}$ . Eq. 4.2.2.1 might be applicable to a wide spectrum of FRs, however, it still covers only some compounds and cannot be applicable to many plastic additives (OECD, 2019). From the comparison of measured and estimated Vps of tricresyl phosphate (TCP) and triphenyl phosphate (TPHP) in OECD (2019), it can be assumed that the temperature dependences of Vps are similar and parallel shifted. Accordingly, the TPHP coefficients in the Yaws equation can be used for other FRs, such as the OPFRs, BRs and DP. However, C, D and E have large variation between compounds. Using only  $A + B/T$  to describe the primary variation of Vp as a function of temperature the degrees of freedom are reduced notably. Moreover, the temperature interval applicable in this study is considerably lower than the one used in the Yaws equation in OECD (2019). It is now straightforward to derive A for the other FRs, by introducing known values for Vp at given temperatures in  $\log_{10} Vp = A + B/T$  with  $B = -5668$  for TPHP.

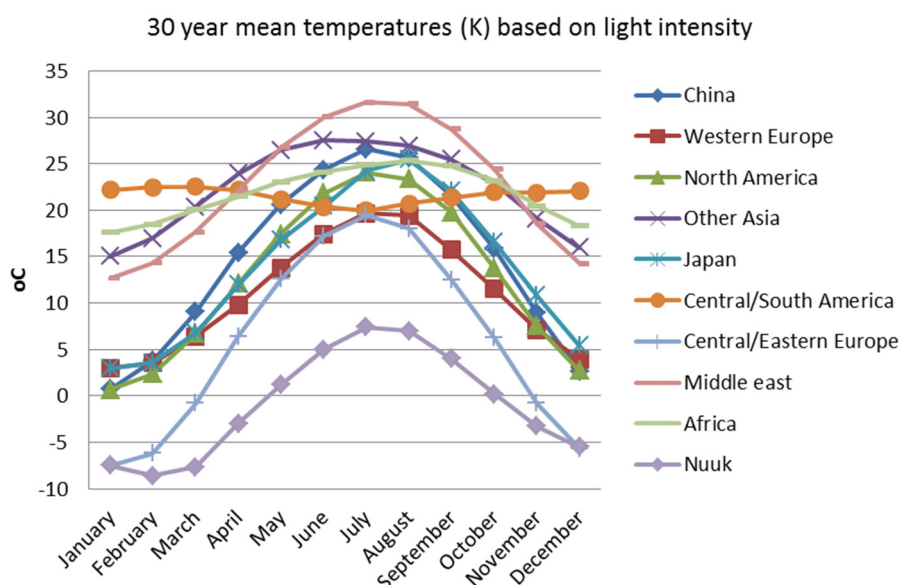


Figure 4.3.2.1 Monthly mean outdoor temperatures (°C) for the nine global regions and for Nuuk, where the temperature in each region is weighed according to the distribution of light intensity within the region.

Vps at 20 °C are shown in Table 3.1.1. Adjusting for the influence of temperature according to Equation 4.3.2.1 the Vps as a function of ambient temperature are shown in Figure 4.3.2.2.

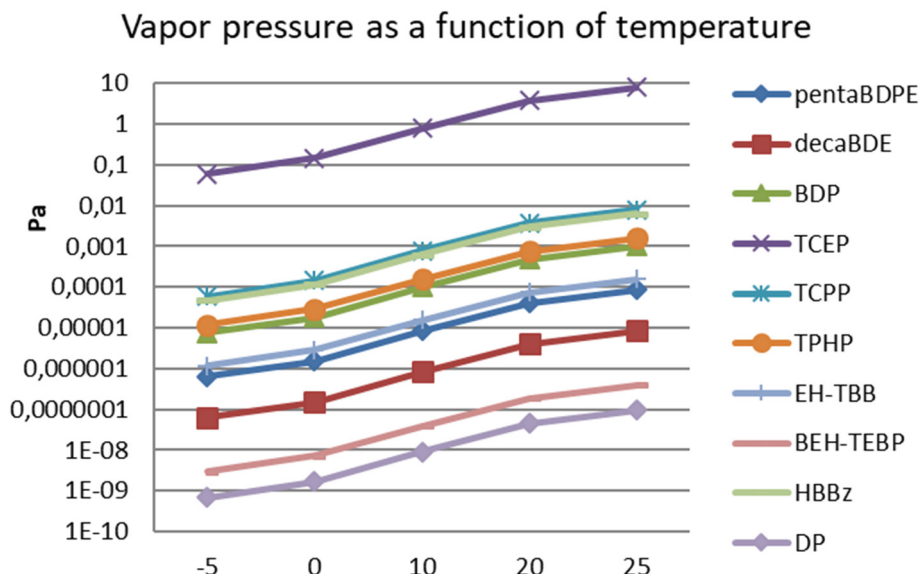


Figure 4.3.2.2 Vapour pressure as a function of temperature calculated from Yaws equation (Eq. 4.3.2.1). DecaBDE, pentaBDE and BDP are included as reference compounds.

The EFs to air for the two reference BFRs (decaBDE and pentaBDE) (Table 4.3.1.1), need to be adjusted in order to be used as EFs to air for BFRs, OPFRs and DP. One approach suggested by OECD (2019) assumes that the EF to air for most plastic additives, including FRs, is proportional to the Vp (25 °C) and inversely proportional to the molecular weight (M). Adjustment is done by using another compound with a known EF as a reference:

$$EF = EF(known) \cdot \frac{Vp/M}{Vp(known)/M(known)} \quad \text{Eq. 4.3.2.2}$$

OECD (2019) suggests to calculate EFs for BFRs from the decaBDE values, and EFs for OPFRs from values of the OPFR bisphenol A bis(diphenyl phosphate) (BDP) (CAS no 5945-33-5), using Eq. 4.3.2.2.

The OECD (2019) emission scenario document (ESD) provides the following measured emission rates during the use of electrical and electronic products at an in-service temperature of 60 °C:

DecaBDE: Emission rate = 0.03 µg/m<sup>2</sup>/h

BDP: Emission rate = <0.05 µg/m<sup>2</sup>/h

Even though “BDP was emitted from both product and test samples, and the results suggested that its actual emission rate is smaller than that of decaBDE” (OECD, 2019), it is assumed from the limited dataset that the measured EFs to air for BDP are approx. similar to the EFs for decaBDE. Accordingly, the decaBDE and pentaBDE EFs are used as proxies for OPFR EFs. Subsequently, the OPFR EFs to air are adjusted relative to BDP with Eq. 4.3.2.2.

EFs to air for the BFR and DP are adjusted relative to decaBDE and pentaBDE (only production) using Eq. 4.3.2.2.

Another approach from Lassen (1999) for BFRs suggests that the possible emission by volatilization (%) of PBDEs per year from products in service, is estimated from:

$$EF (\%) = 1.1 \cdot 10^6 \cdot Vp (mmHg \text{ at } 20^\circ C) \quad \text{Eq. 4.3.2.3}$$

Adjustment factors are calculated as EF to air (Eq. 4.3.2.3) for the OPFRs, BFRs and DP divided with EF to air (Eq. 4.3.2.3) for decaBDE and pentaBDE.

Both Eq. 4.3.2.2 and Eq. 4.3.2.3 are approximations. In Table 4.3.2.1, the mean correction factors for EFs to air are calculated for the seven compounds. These are valid for Vp at 20 °C. For outdoor use of plastics and textiles and for waste treatment Vps at the specific monthly mean outdoor temperatures are used to calculate the adjustment factors. The adjustment factors are then multiplied with the EFs to air in Table 4.3.1.1 for each of the seven compounds.

Table 4.3.2.1 Adjustment factors for EFs to air in Table 4.3.1.1, using Vp at 20 °C. For outdoor use of plastics and textiles and for waste treatment the factors must be based on Vps at the specific monthly mean outdoor temperatures.

Stages in mass flow	TCEP	TCPP	TPHP	EH-TBB	BEH-TEBP	HBBz	DP
All stages	13607	12.4	2.47				
Production				1.89	0.00425	75.0	0.0010
All stages except production				25.9	0.0565	1029	0.014

EFs to water and soil are assumed constant for varying temperatures and no compounds specific adjustment is made.

#### 4.4 Waste treatment and spatial distribution of emissions in the Northern hemisphere

The geographical distribution of emissions can be determined with high accuracy for the production of some FRs, as shown in Chapter 4.2. The FRs are applied in many products, materials and activities, and there are a large number of product manufacturing units all over the world, with unknown locations. This impedes a precise geographical distribution of emissions from manufacture, consumer use and waste treatment. However, due to the wide application in consumer products and large use we assume that the distribution correlates with population density, i.e. activity. A reliable proxy for this is the light intensity as seen from space (Hansen et al., 2020).

The global distribution of use and waste amounts was presented for DP in Hansen et al. (2020). The amounts were distributed in nine regions, i.e. Western Europe, Central/Eastern Europe, North America, Central/South America, China, Japan, Other Asia, Middle East, and Africa, corresponding to the presentation of the world consumption of FRs in 2019 in Figure 4.4.1. The approach in Hansen et al. (2020) is also used in this project where adjustments are done for reported use amounts in a region, which results in the distribution of use amounts in the different global regions as shown in Figure 4.4.2. For example, TCPP and TPHP are adjusted according to reported use amounts in the EU and US (van der Veen et al., 2012); EH-TBB is not registered in REACH and BEH-TEBP is a low production volume chemical in the EU, and uses of both are assumed skewed towards the US as they are replacements of pentaBDE, which was primarily used in the US; HBBz is mainly used in Japan (Verreault et al., 2007). Overall, the global distributions have only

smaller variations between the low and high emission scenarios. No similar data is available for textile use; therefore, the same global distribution as for plastics is used for textiles.

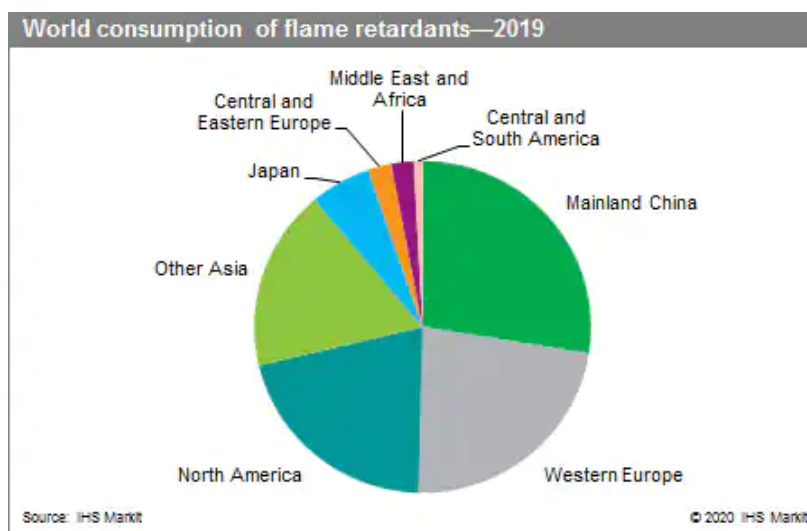


Figure 4.4.1 World consumption of flame retardants in 2019 (<https://www.ihs.com/products/chemical-flame-retardants-scup.htm>).

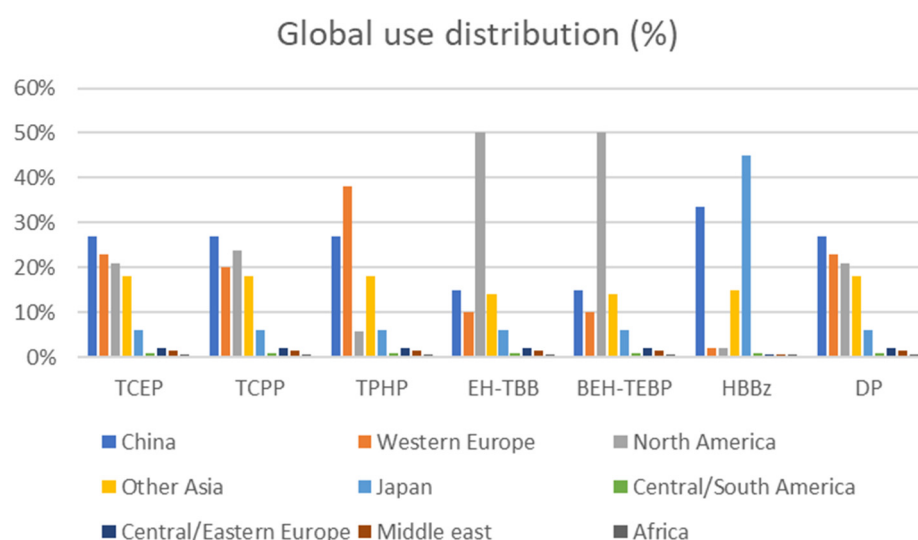


Figure 4.4.2 Global distribution of use amounts in %. The distributions have smaller variations between low and high emission scenarios.

Amounts of waste that go into waste treatment are assumed to consist of plastic waste from the regions' own use of plastics and electronics, c.f. Figure 4.4.2, and adjusted for import and export of plastic waste (Hansen et al., 2020). Only 5 % of the produced plastic enters the waste trade (Velis, 2014). Data on waste trade are obtained from UN Comtrade (Commodity code 3915) (UNComtrade UN Comtrade Database). Such data are not available for textile waste, and as before, the same assumptions and mass distributions as for plastic waste are used.

The monthly mean global emissions to air, water and soil were calculated for the low and high emission scenario, respectively, see Figure 4.4.3. This was done on the basis of the geographical distribution of production, use and waste amounts, the EFs in Table 4.3.1.1 with the correction factors in Table 4.3.2.1. Further it was also taking into account the specific monthly mean outdoor temperatures for outdoor use of plastics and textiles and for waste treatment, the distribution fractions into activities and products in Table 4.1.2.

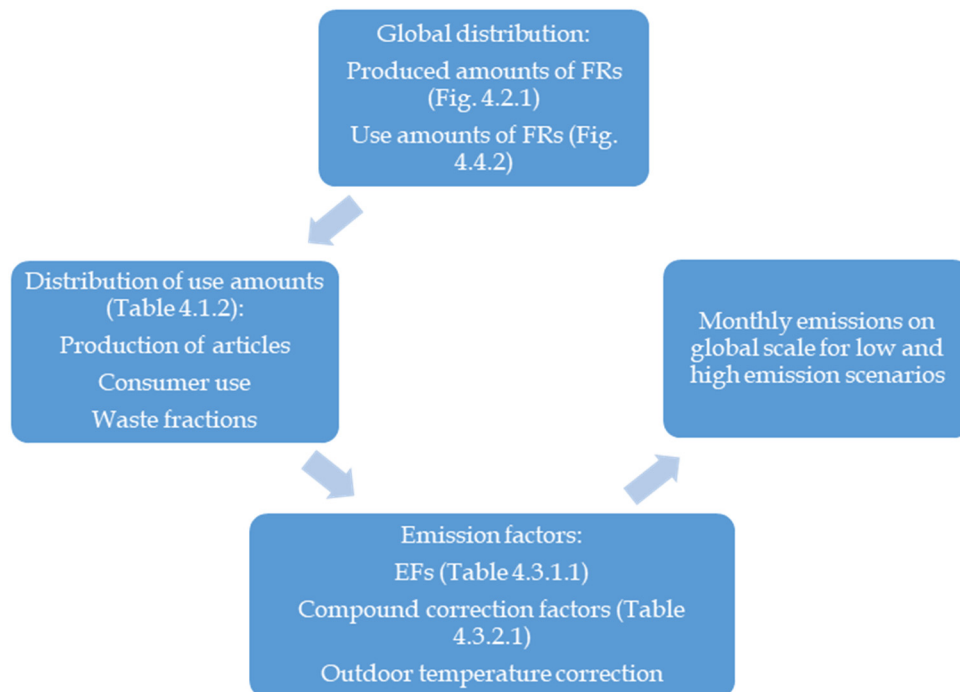


Figure 4.4.3 Procedure for calculating global monthly emissions.

The relative distribution of emissions between production of articles, consumer use and waste treatment is the same for the FRs. When a FR is produced within a region there is an additionally emission from the production of the compound. Table 4.4.1 shows the monthly mean emissions to air, water and soil for TCP as an example, for the low and high emission scenarios, respectively.

Table 4.4.1 Monthly mean emissions (January) for TCPP as an example. Low and high emission scenarios represent variations in EFs and produced amounts. The relative distribution of emissions between production of articles, consumer uses of plastics and textiles and waste treatment is the same for all FRs. Units are in t/month.

Activity	Compartment	Low emission scenario									High emission scenario								
		China	Western Europe	North America	Other Asia	Japan	Central/ South America	Central/ Eastern Europe	Middle East	Africa	China	Western Europe	North America	Other Asia	Japan	Central/ South America	Central/ Eastern Europe	Middle East	Africa
Production of chemical	air	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	2,8E-06	1,2E-06	4,2E-07	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00
	water	2,6E+00	9,6E-01	6,3E-01	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	3,2E+01	9,0E+00	4,7E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00
	soil	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00
Production of articles	air	9,6E-03	1,2E-02	7,7E-03	6,8E-02	3,1E-03	1,1E-02	1,6E-04	3,9E-03	2,8E-03	5,2E-02	5,8E-02	4,6E-02	3,7E-01	1,7E-02	6,2E-02	8,9E-04	2,1E-02	1,5E-02
	water	4,5E-03	3,7E-03	3,6E-03	3,0E-03	9,9E-04	1,7E-04	3,3E-04	2,5E-04	8,3E-05	4,8E-01	3,5E-01	4,2E-01	3,2E-01	1,1E-01	1,8E-02	3,5E-02	2,6E-02	8,8E-03
	soil	1,1E-02	9,2E-03	9,1E-03	7,4E-03	2,5E-03	4,1E-04	8,3E-04	6,2E-04	2,1E-04	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00
Consumer use of plastic products, indoor	air	3,4E-01	2,8E-01	2,8E-01	2,3E-01	7,7E-02	1,3E-02	2,6E-02	1,9E-02	6,4E-03	1,3E+01	9,7E+00	1,2E+01	8,7E+00	2,9E+00	4,8E-01	9,7E-01	7,3E-01	2,4E-01
	water	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	1,1E+00	7,9E-01	9,3E-01	7,0E-01	2,3E-01	3,9E-02	7,8E-02	5,9E-02	2,0E-02
Consumer use of plastic products, outdoor	air	3,1E-04	3,8E-04	2,5E-04	2,2E-03	1,0E-04	3,7E-04	5,3E-06	1,3E-04	9,2E-05	5,7E-04	6,3E-04	5,0E-04	4,0E-03	1,9E-04	6,7E-04	9,6E-06	2,3E-04	1,7E-04
	water/soil	1,9E-02	1,5E-02	1,5E-02	1,2E-02	4,1E-03	6,9E-04	1,4E-03	1,0E-03	3,4E-04	3,4E-02	2,5E-02	3,0E-02	2,3E-02	7,5E-03	1,3E-03	2,5E-03	1,9E-03	6,3E-04
Consumer use of textiles, indoor	air	4,0E-01	3,3E-01	3,3E-01	2,7E-01	9,0E-02	1,5E-02	3,0E-02	2,2E-02	7,5E-03	1,1E+01	8,5E+00	1,0E+01	7,6E+00	2,5E+00	4,2E-01	8,5E-01	6,4E-01	2,1E-01
	water	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	9,3E-01	6,9E-01	8,2E-01	6,2E-01	2,1E-01	3,4E-02	6,9E-02	5,1E-02	1,7E-02
Textiles washing	water	1,0E-02	8,4E-03	8,3E-03	6,8E-03	2,3E-03	3,8E-04	7,5E-04	5,7E-04	1,9E-04	1,8E-02	1,4E-02	1,6E-02	1,2E-02	4,1E-03	6,8E-04	1,4E-03	1,0E-03	3,4E-04
Consumer use of textiles, outdoor	air	1,4E-02	1,8E-02	1,2E-02	1,0E-01	4,7E-03	1,7E-02	2,4E-04	5,9E-03	4,2E-03	2,6E-02	2,9E-02	2,3E-02	1,9E-01	8,5E-03	3,1E-02	4,4E-04	1,1E-02	7,7E-03
	water/soil	8,6E-01	7,0E-01	7,0E-01	5,7E-01	1,9E-01	3,2E-02	6,4E-02	4,8E-02	1,6E-02	1,6E+00	1,2E+00	1,4E+00	1,0E+00	3,5E-01	5,8E-02	1,2E-01	8,7E-02	2,9E-02
Waste recycling	air	1,8E-06	1,7E-06	1,2E-06	1,1E-05	4,1E-07	7,1E-07	2,7E-08	6,9E-07	4,7E-07	1,1E-02	9,4E-03	7,7E-03	6,4E-02	2,5E-03	4,2E-03	1,6E-04	4,1E-03	2,8E-03
Waste incineration	air	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	2,0E-03	1,7E-03	1,4E-03	1,2E-02	4,5E-04	7,7E-04	3,0E-05	7,5E-04	5,1E-04
Waste landfill	air	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	1,2E-02	1,0E-02	8,5E-03	7,0E-02	2,7E-03	4,7E-03	1,8E-04	4,5E-03	3,1E-03
	air	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	8,5E-03	7,3E-03	6,0E-03	5,0E-02	1,9E-03	3,3E-03	1,3E-04	3,2E-03	2,2E-03
	water	8,4E-04	5,4E-04	5,5E-04	4,6E-04	1,3E-04	1,0E-05	5,5E-05	4,3E-05	1,4E-05	1,5E-03	8,8E-04	1,1E-03	8,3E-04	2,3E-04	1,8E-05	9,9E-05	7,8E-05	2,4E-05
Total monthly emissions	air	7,7E-01	6,4E-01	6,3E-01	6,7E-01	1,7E-01	5,6E-02	5,6E-02	5,1E-02	2,1E-02	2,5E+01	1,8E+01	2,2E+01	1,7E+01	5,5E+00	1,0E+00	1,8E+00	1,4E+00	4,9E-01
	water	3,4E+00	1,7E+00	1,4E+00	5,9E-01	2,0E-01	3,3E-02	6,6E-02	5,0E-02	1,7E-02	3,6E+01	1,2E+01	8,3E+00	2,7E+00	9,0E-01	1,5E-01	3,0E-01	2,3E-01	7,5E-02
	soil	1,1E-02	9,2E-03	9,1E-03	7,4E-03	2,5E-03	4,1E-04	8,3E-04	6,2E-04	2,1E-04	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00	0,0E+00

In Figure 4.4.4a and 4.4.4b, the total monthly mean emissions to air are shown for all FRs, illustrating the differences between FR emissions on account of corrections of EFs based on molecular weight (M) and vapour pressure (Vp), and the amount and location of chemical production facilities and reported regional use amounts. Figure 4.4.4a is for January with the low emissions scenario, representing a minimum in global emissions, and Figure 4.4.4b is for July with the high emission scenario, representing a maximum in global emissions.

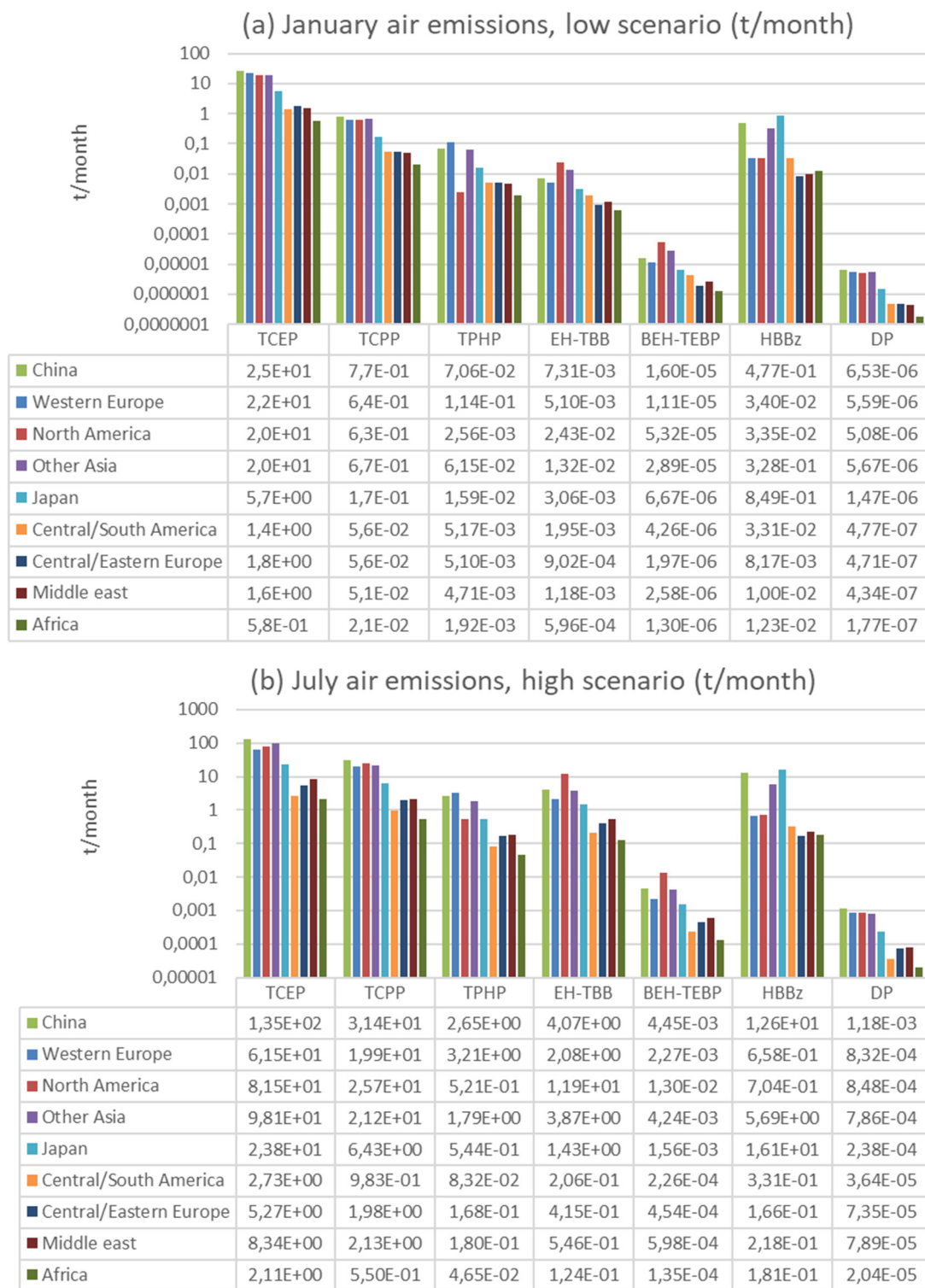


Figure 4.4.4a and b Global air emissions in (a) January with low emission scenario and (b) July with high emission scenario. Units are t/month.



In comparison, based on the EU risk assessment of TCPP, approximately 40 % of the TCPP in a finished product was available for release, whereas the total continental European and the total regional emission discharges of TCPP to air were estimated at 89.6 and 135 kg/d, respectively (Wei et al., 2015; EU, 2007). From Figures 4.4.4a and b the sum of Western and Central/Eastern European TCPP emissions to air vary from a minimum of 23.2 kg/d to a maximum of 729 kg/d. Air emissions representing the other months with low and high scenarios are in between these values. These estimates are comparable to the figures in Wei et al. (2015) and EU (2007).

## 5 Long-range transport modelling with DEHM

### 5.1 Model set-up and sensitivity analysis

The Danish Eulerian Hemispheric Model (DEHM) is a 3D atmospheric chemistry-transport model. DEHM was originally developed to study the atmospheric transport of sulphur and sulphate into the Arctic (Christensen, 1997). The model has since been further developed to include a comprehensive chemistry scheme with ozone and nitrogen oxides as well the possibility to run simulations with high temporal resolution in a limited area of the domain (Brandt et al., 2012). DEHM is defined using a polar-stereographic projection centered on the North Pole, a horizontal grid covering the majority of the Northern Hemisphere with a grid size of 150 km x 150 km, and with 29 vertical layers stretching into the stratosphere to a height of 100 hPa (approximately 16 km). DEHM has been used to study the atmospheric transport to the Arctic of several compounds, including organic contaminants such as  $\alpha$ -HCH (Hansen et al., 2004; 2008a;b) and decamethylcyclopentasiloxane (D5) (McLachlan et al., 2010, Krogseth et al., 2013), lead (Heidam et al. 2004) and mercury (Christensen et al., 2004; Skov et al., 2004).

DEHM is driven by meteorological data from the numerical weather prediction model WRF (Skamarock et al., 2008). The environmental distribution and fate of the organic compounds is determined by their physical-chemical properties such as the partition coefficients, reaction rates and half-lives in the surface media. We have applied the partition coefficients, reaction rates and surface media half-lives from section 3.1 as input to DEHM.

The physical-chemical parameters are uncertain and there is a range in values of the partition coefficients from the literature study, which is typically 2-3 OM, however, for one parameter and one compound (log  $K_{AW}$  of TPHP) there is 8 OM between the lowest and the highest reported value (See Table 5.1.1). A series of sensitivity simulations have been performed to investigate the influence of this uncertainty on the calculated concentrations. Calculations were made for each compound where each partition coefficient (Log  $K_{OA}$ , Log  $K_{AW}$  and Log  $K_{OW}$ ) is one OM lower and higher than the applied basic value, respectively.

Table 5.1.1 Basic values and highest and lowest literature value for the partition coefficients.

		TCEP	TCPP	TPHP	EH-TBB	BEH-TEBP	HBBz
log $K_{AW}$	Basic value	-5.98	-5.61	-5.79	-3.59	-4.91	-3.06
	Min.	-2.69	-1.53	-2.90	-1.05	-0.48	-2.20
	Max.	-6.94	-6.70	-10.70	-3.70	-6.30	-3.46
log $K_{OA}$	Basic value	7.61	8.50	10.49	12.34	16.86	10.39
	Min.	7.00	7.60	10.30	9.53	16.86	6.65
	Max.	8.90	10.00	14.00	12.40	18.00	10.40
log $K_{OW}$	Basic value	1.63	2.89	4.70	8.75	11.95	7.33
	Min.	1.44	2.59	3.30	7.73	9.34	3.68
	Max.	4.30	6.10	7.43	8.80	12.40	7.33

All sensitivity simulations are made for a four-year period (2009-2012) and changes are assessed by comparing the median atmospheric surface concentration for the whole period for each sensitivity analysis with the basic value for one station: Alert, Nunavut, Canada. The partition coefficients do not affect the simulated concentrations equally. Changes in Log  $K_{OW}$ , which is applied in the description of the fate of the compounds in the soil module, are barely affecting the atmospheric concentrations (Table 5.1.2). Largest differences were found for the compounds with lowest Log  $K_{OW}$  values, i.e. TCEP and TCPP, with a maximum of 2 % and 5 % difference in median concentrations, respectively.

Table 5.1.2 Changes in median atmospheric surface concentration for a four-year period used in the sensitivity simulations where the partition coefficients are one order of magnitude smaller (-) or larger (+) than the basic value, respectively.

	TCEP	TCPP	TPHP	EH-TBB	BEH-TEBP	HBBz
log $K_{OW}$ -	1.13	-4.80	-0.59	0.30	0.00	0.06
log $K_{OW}$ +	2.11	-1.89	0.47	-0.07	0.00	0.13
log $K_{OA}$ -	53.5	55.4	-1.99	-63.9	-1.37	145
log $K_{OA}$ +	-44.4	-33.5	30.5	942	0.10	-32.5
log $K_{AW}$ -	-8.16	-26.9	-40.6	-23.8	-10.3	-46.6
log $K_{AW}$ +	62.8	22.9	43.4	22.8	15.3	74.0

The difference is larger for changes in the Log  $K_{AW}$ , which is applied in the parameterization of the gas exchange between air and water and in the parameterization of wet deposition. When Log  $K_{AW}$  is one OM lower then the median concentration is lower for all compounds with up to minus 47 % for HBBz. When Log  $K_{AW}$  is one OM higher, the median concentration is up to 74 % higher for all compounds (Table 5.1.2).

Log  $K_{OA}$ , which is applied in the parameterization of the gas-particle distribution in air, has the largest influence on the median concentration, but the pattern is not the same for all compounds. In the sensitivity analysis where Log  $K_{OA}$  is one OM smaller than the basic value, the median concentration is up to -64 % lower for TPHP, EH-TBB and BEH-TEBP, which are the three compounds with highest Log  $K_{OA}$  values, while it is up to 145 % higher for the other three compounds. The median concentration is higher for the sensitivity analysis where Log  $K_{OA}$  is one OM larger for the three BFRs, while it is up to -44 % lower for the other three compounds. The largest difference is found for EH-TBB, where the difference in median concentrations is almost an OM for the simulation with one OM higher Log  $K_{OA}$  value than the basic simulation. The applied basic value for Log  $K_{OA}$  (12.34) is close to the maximum literature value (12.4). It is therefore less likely that there is a large difference to the basic value and therefore a large difference in the simulated atmospheric concentrations.

Disregarding the results for Log  $K_{OA}$  for EH-TBB, which is assumed not likely, there are not very large effects on the simulated concentration from the sensitivity analysis.

## 5.2 Model evaluation

The model simulations are evaluated by comparing the predicted concentrations to measurements of the studied compounds. Usually, the model performance of the model is evaluated by calculating statistical parameters of the data sets, such as correlation coefficients, bias and RMSE. However, for the compounds in this study, there are only measurements available from a few sites, and most of them are only available in summary form, i.e. given as a range of measured values, a mean and/or median concentration. It is therefore not possible to evaluate the model performance using regular statistics.

Furthermore, some of the available measurements are done using passive samplers for the analysis. These samples collect the pollutants from the air over long time periods, typically 3-12 months and an average air concentration is calculated using an assumed average air flow rate passing the sampler. The results have uncertainty and are difficult to compare to results from active sampling as well as the model results. It should also be noted that the air data available in the literature represent different years or time periods, possibly not reflecting the same emission situation as used in the model calculations.

We have extracted daily averaged concentrations from the lowermost atmospheric layer from DEHM corresponding to the coordinates of the site by interpolating the concentrations from the four nearest grid cells for the period 2009-2015. The median concentration of the measurements (if available) is compared to the predicted median concentration either for the whole period or for the specific period of measurements.

### 5.2.1 Longyearbyen, Svalbard

Salamova et al. (2014a) collected 34 samples of air particles in Longyearbyen, Svalbard over 48 hours using a high-volume sampler (HVS) between September 2012 and May 2013. The median concentration of the measurements is compared to the predicted median concentration for both the period 2009-2015 as well as the median concentration for the measurement period from September 2012 to May 2013 (Figure 5.2.1.1). The predicted median concentration and maximum concentration are similar for the two periods, while the minimum concentration is much smaller for the full period, since it contains concentrations from summer periods where there is a very little transport of air into the Arctic and hence very low concentrations. We have chosen to compare the measured concentrations to the sum of predicted gas and particle phase concentrations since the distribution between the two phases is uncertain for compounds with high Log  $K_{OA}$  values (Hansen et al., 2020).

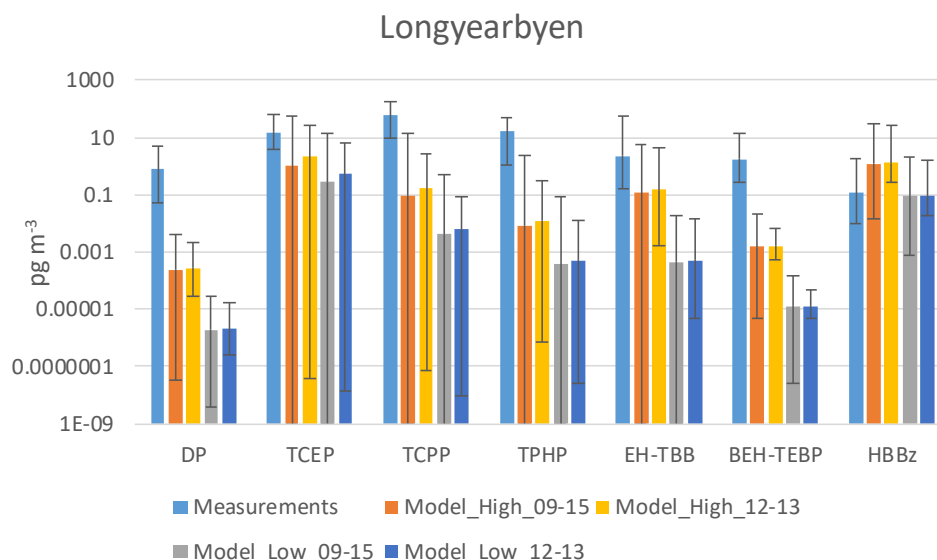


Figure 5.2.1.1 Measured and predicted median concentrations (bars) for the seven compounds. Upper and lower borders show maximum and minimum concentrations. \_09-15: predicted median concentrations for the years 2009-2015; \_12-13: predicted median concentrations for the years 2012-2013.

The predicted concentrations of DP, TCPP, TPHP and BEH-TEBP are about two OM lower than the measured concentrations for the high emission scenario and three for the low emission scenario, and the lowest measured values are higher than the highest simulated value. For TCEP and EH-TBB, the measured median concentrations are about one OM larger than the predicted median concentration for the high emission scenario, while there is an overlap between the range of measured and predicted concentrations. For EH-TBB, the median concentration for the low emission scenario is one OM lower than for the high emission scenario, while there is a smaller difference between the two scenarios for TCEP. For HBBz, the measured median concentration is between the predicted median concentrations of the high and the low emission scenario, but closest to the low.

There is no specific description of the location of the sampling site in the paper, but an illustration indicates that the measurements are made within the town. It is therefore possible that the measurements are affected by local sources, which could explain some of the differences.

Carlsson et al. (2018) placed three passive samplers of silicone membranes over three months in March-May 2015 and February-May 2016. There were measured high concentrations in the samplers of EH-TBB (mean value of 86 pg/m<sup>3</sup>), BEH-TEBP (mean value of 1867 pg/m<sup>3</sup>) and DP (mean value of 705 pg/m<sup>3</sup>), while the values of HBBz were under the detection limit. It can be difficult to directly compare these measurements from passive silicone sampler with the earlier results where there was used high volume samplers. The results from the passive silicone sampler showed considerably higher than the concentrations measured by Salamova et al. (2014a) who measured mean values of 7.0 pg/m<sup>3</sup> for EH-TBB, 2.7 pg/m<sup>3</sup> for BEH-TEBP and 1.2 pg/m<sup>3</sup> for DP. The measurements indicate that there are significant local sources, although the authors do not describe any possible sources to the high measured concentrations of EH-TBB and BEH-TEBP (Carlsson et al., 2018).

### 5.2.2 Ny Ålesund, Svalbard

Passive samples of air using XAD-PAS samplers were collected over one year from August 2014 to August 2015 at five sites in the area around Ny Ålesund, Svalbard and on an island in Kongsfjorden outside Ny Ålesund (Hao et al., 2020).

EH-TBB and BEH-TEBP were below detection limits. The concentration of HBBz was measured to be between 0.9 and 2.9 pg/m<sup>3</sup> with an average of 1.8 pg/m<sup>3</sup>. This is about an OM higher than concentrations measured in the nearby Longyearbyen (Salamova et al., 2014a).

There were no statistically significant spatial patterns in the five samples indicating minor influence of the research station in Ny Ålesund on the measured concentrations (Hao et al., 2020).

TCEP, TCPP and TPHP were analysed in the same five samples at average concentrations of 180 pg/m<sup>3</sup> (84-340 pg/m<sup>3</sup>), 73.9 pg/m<sup>3</sup> (54.5-91.8 pg/m<sup>3</sup>), and 85.1 pg m<sup>-3</sup> (31.3- 239 pg m<sup>-3</sup>), respectively (Han et al., 2020). The levels of TCPP are similar to the levels measured at Longyearbyen, while the concentrations of TPHP were about five times higher and the concentrations of TCEP about 10 times higher than the levels measured at Longyearbyen (Salamova et al., 2014a).

### 5.2.3 Alert, Nunavut, Canada

Four compounds: EH-TBB, BEH-TEBP, HBBz and DP were measured in combined gas and particle phase samples collected using a High Volume Sampler in the period 2008-2015 at Alert, Nunavut, Canada (Vorkamp et al., 2019b).

The compounds were not identified in all collected samples at the site, and the detection frequency varied from 3 to 26 % for EH-TBB, from 2 to 21 % for BEH-TEBP and from 1 to 12 % for HBBz. The median measured concentrations are compared to the modelled median concentrations over the full year.

The median measured concentrations for EH-TBB are about one OM larger than the median concentration predicted for the high emission scenario for five of the seven years and they are similar for the last two years (Figure 5.2.3.1). The predicted range for this scenario is within the measured range for all years. For the low emission scenario, the median concentration is two to three OM lower than the measured median and there is an overlap of the range for only three years.

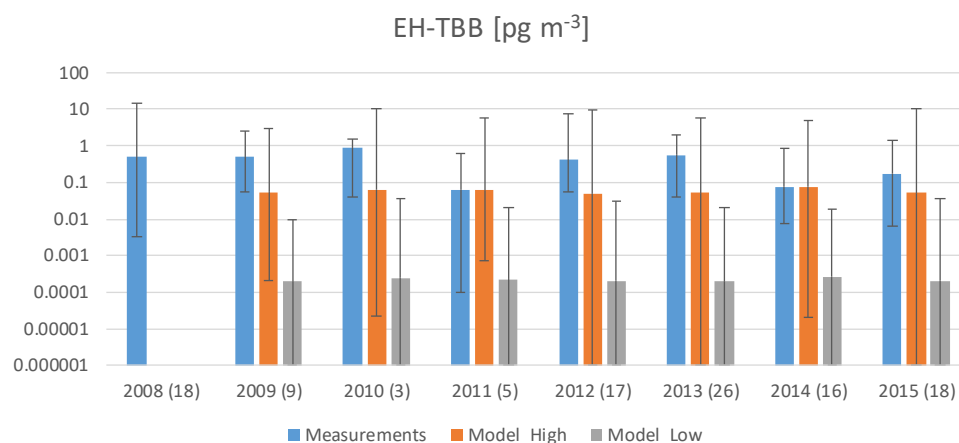


Figure 5.2.3.1 Median concentrations and range of measurements (blue bars) and predicted concentrations for the high (orange bars) and low (grey bars) emission scenarios for EH-TBB at Alert. For each year, the number of samples with detection of the compound is indicated in brackets.

There is a large year-to-year difference in measured median concentrations of more than two OM. The measured median concentrations for BEH-TEBP is two to four OM higher than the predicted median for the high emission scenario and four to six OM higher than the low emission scenario (Figure 5.2.3.2). There is only an overlap between measured and predicted concentrations for one year for the high emission scenario and none for the low.

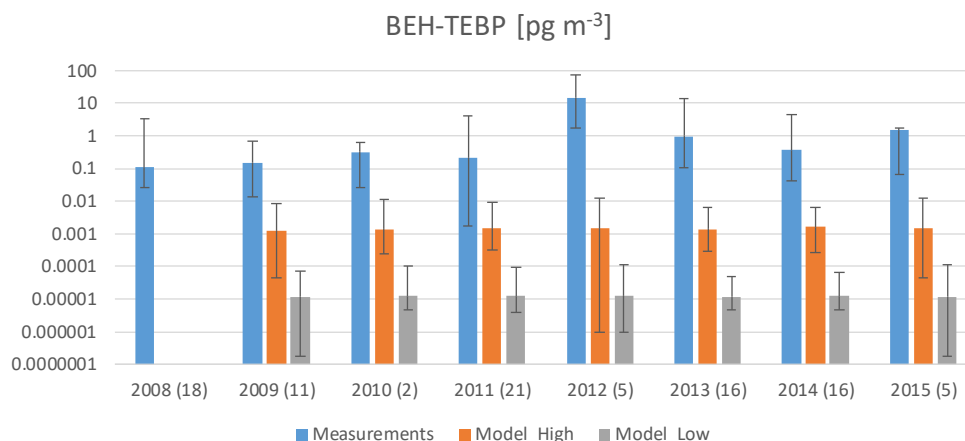


Figure 5.2.3.2 Median concentrations and range of measurements (blue bars) and predicted concentrations for the high (orange bars) and low (grey bars) emission scenarios for BEH-TEBP at Alert. For each year, the number of samples with detection of the compound is indicated in brackets.

The measured median concentrations for HBBz is similar to the median concentration for the low emission scenario the first four years and one OM lower for the last three years, while the predicted median concentrations for the high emission scenario is 1-2 OM larger (Figure 5.2.3.3). There is an overlap of the range for all years for the low emission scenario and for four of the seven years for the high emission scenario.

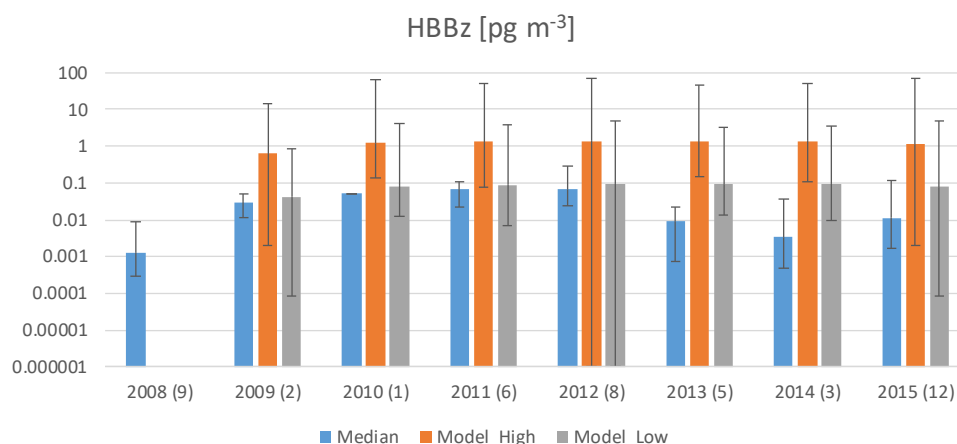


Figure 5.2.3.3 Median concentrations and range of measurements (blue bars) and predicted concentrations for the high (orange bars) and low (grey bars) emission scenarios for HBBz at Alert. For each year, the number of samples with detection of the compound is indicated in brackets.

The measured median concentration of the sum of DP isomers is more than two OM higher than the median concentration of the high emission scenario and more than four OM higher than the low emission scenario (Figure 5.2.3.4). There is no information available on the range of the sum of the two DP isomers from the measurements, but it is unlikely that there is an overlap.

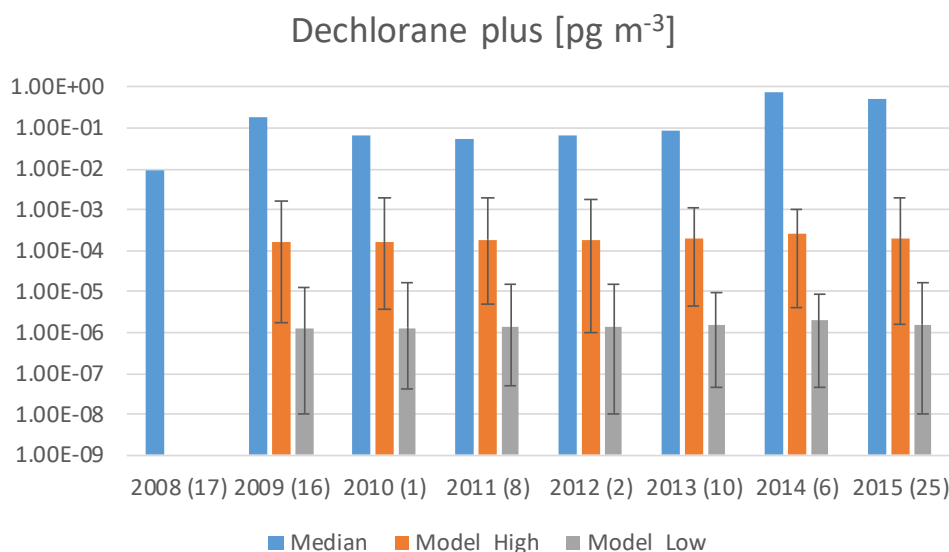


Figure 5.2.3.4 Median concentrations of measurements (blue bars) and predicted concentrations for the high (orange bars) and low (grey bars) emission scenarios for DP at Alert. For each year, the number of samples with detection of the compound is indicated in brackets. The range on the measurements was not available.

TCEP and TCPP were measured in seven and four samples, respectively, at Alert in 2009 using High Volume Sampler (Sühring et al., 2016). The measured median concentration is two and three OM higher than the median concentration predicted with the high and low emission scenario, respectively (Figure 5.2.3.5). In the same study, TPHP was analysed in 16 samples from 2012 and the measured median concentration is 4 and 5 OMs higher than the predicted median concentrations for the high and low emission scenario, respectively (Figure 5.2.3.5).



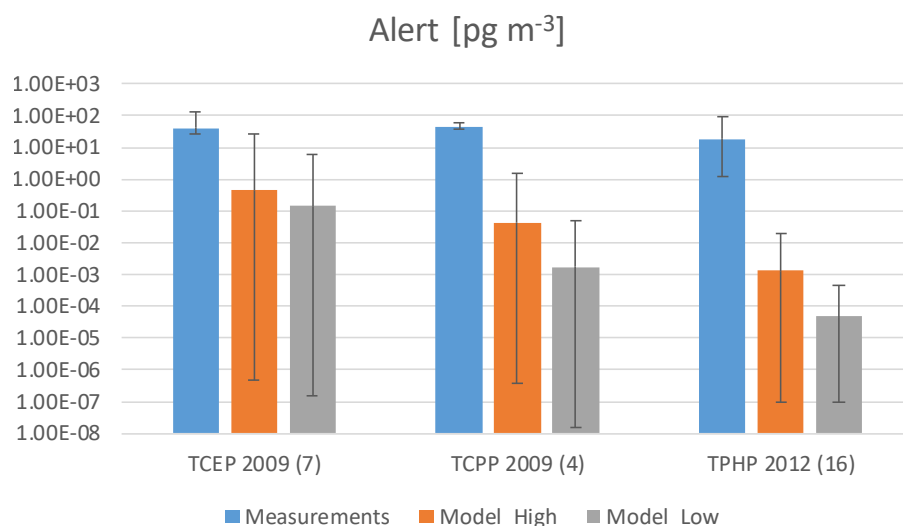


Figure 5.2.3.5 Measured and predicted median concentrations (bars) for the seven compounds. Upper and lower borders show maximum and minimum concentrations.

The predicted concentrations at Alert are lower but of similar OMs to the measurements from Longyearbyen for all compounds.

#### 5.2.4 Little Fox Lake, Yukon, Canada

The four components measured at Alert were also measured at Little Fox Lake using passive flow-through samplers deployed monthly between August 2011 and December 2014. The levels measured for the four compounds are similar to the measured levels at Alert and the pattern of the comparison with predicted concentrations are also the same.

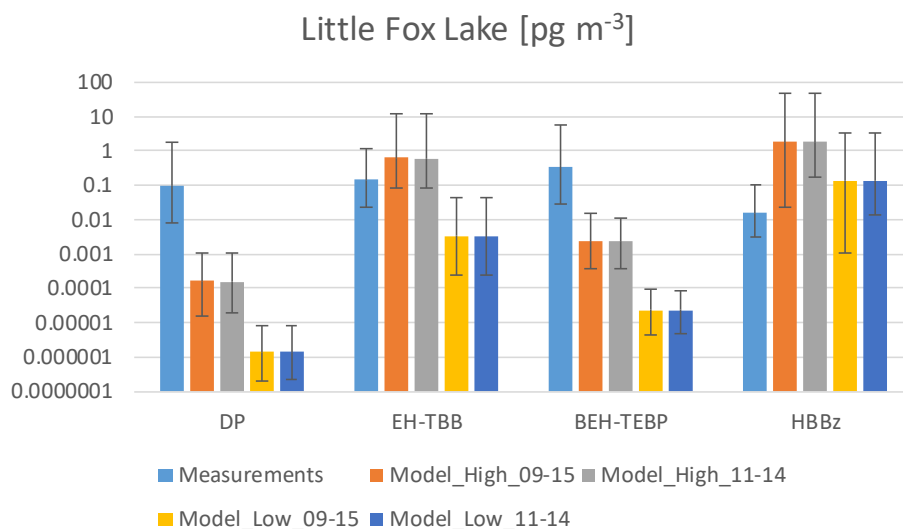


Figure 5.2.4.1 Median concentrations and range of measurements (light blue bars) and predicted concentrations for the high for the full period (orange bars) and for August 2011-December 2014 (grey bars) and for the low emission scenarios for the full period (yellow bars) and for August 2011-December 2014 (dark blue bars) for the four compounds.

### 5.2.5 Resolute Bay, Canada

Ten samples were collected using High Volume Sampler at Resolute Bay (Cornwallis Island, Nunavut, 74.70°N, 94.83°W) in 2012 and analysed for a range of compounds among others TCEP, TCPP and TPHP (Sühring et al., 2016). Measured median concentrations are also higher than the predicted median concentrations for both emission scenarios for these compounds, similarly to the results from Alert from the same study (Figure 5.2.5.1).

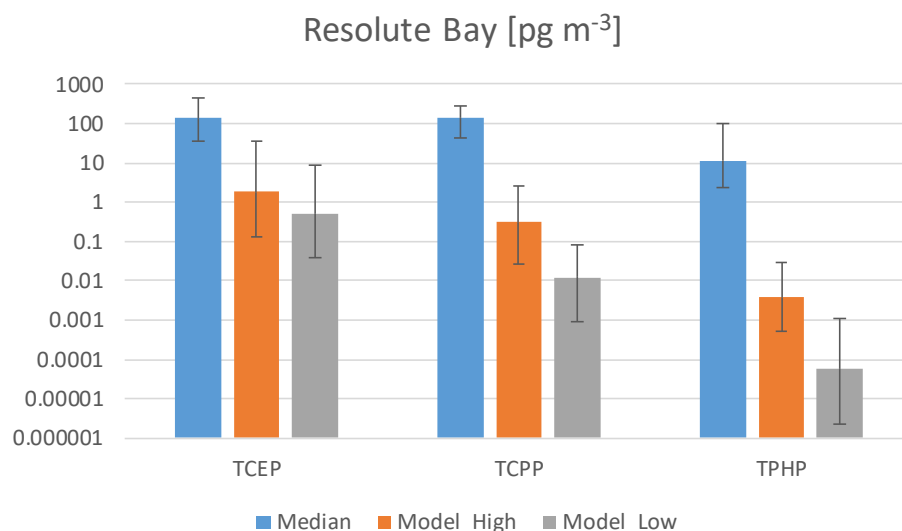


Figure 5.2.5.1 Median concentrations and range of measurements (light blue bars) and predicted concentrations for the high (orange bars) and for the low emission scenarios for the full period (grey bars) for the three compounds.

### 5.2.6 Ship cruises

Several measuring campaigns have been conducted on cruises that approach or enter the Arctic area. These measurements are difficult to use for model evaluation because sample collection were made during cruising of the ship and thus cover a large area which is not (typically) revisited.

Möller et al. (2011) measured several BFRs collected in samples during a cruise in the West Pacific Ocean from the East China Sea to the Arctic Ocean in the summer of 2010. In the Arctic part, the measured levels of HBBz were lower than  $0.5 \text{ pg/m}^3$  but no other of the compounds were detected in this study. These levels of HBBz are about an OM higher than the measurements at Alert and Little Fox Lake.

From the same cruise, the samples were analysed for TCEP, TCPP, and TPHP concentrations (Möller et al., 2012a), and the levels of these three compounds were within the same OM as the measurements at Resolute Bay and Alert.

Several cruises were made in the summer months of 2007-2013 in the Canadian waters, where TCEP, TCPP and TPHP were measured. These compounds showed similar median concentrations as the measurements from Alert and Resolute Bay although one maximum value of TPHP were 200 times higher than the maximum measured land concentration (Sühring et al., 2016), which could indicate that this sample is influenced by a large local source.

TCEP, TCPP, and TPHP were among a range of OPFRs measured in air samples in a cruise from the western Pacific Ocean into the Arctic Ocean between July and September 2018 (Na et al., 2020). The levels measured in the Arctic

were similar to the levels measured at Resolute Bay and the earlier cruise by Möller et al. (2012a).

### **5.2.7 Summary of comparison of predicted and measured air concentrations**

TCEP: The predicted concentrations of TCEP are about two OM lower than the measured concentrations at Alert and one lower at Longyearbyen for both scenarios.

TCPP: The DEHM model predicts three and four OM lower concentrations of TCPP for the high and low emission scenario, respectively than the measurements at Alert and Longyearbyen.

TPHP: The median TPHP concentration measured at Alert is about one OM lower than that measured at Longyearbyen, while the predicted median concentrations are about four and five OM lower at Alert and five and six OM lower at Longyearbyen, respectively.

EH-TBB: The measured median concentrations are within an OM of the high emission scenario and the ranges are overlapping at Longyearbyen, Alert and Little Fox Lake.

BEH-TEBP: Measured concentrations BEH-TEBP are two to four OM higher than the high emission scenario and a further two OM higher than the low emission scenario at Alert, Little Fox Lake and Longyearbyen.

HBBz: The measured concentrations of HBBz are of similar OM as the low emission scenario and about one OM lower than the high emission scenario, while the measured range overlaps the predicted ranges for both scenarios at Longyearbyen, Alert and Little Fox Lake.

## **5.3 Assessment of source areas**

We have calculated the contribution to Arctic atmospheric concentrations from the different source areas by running the model for a four-year period (2009-2012) with the emissions from each of the nine regions as a separate tracer in the model. The Arctic is defined as the area north of 67°N and the contribution is calculated as the fraction of the averaged air concentration from each region to the average of the sum of concentrations from the regions.

In general, the contributed fraction to the Arctic from Europe is higher than the region's share of the total global emission, whereas the opposite is the case for China, Japan and the rest of Asia. For North America, the fractions of the emissions and contributions are similar for most of the compounds, and both emissions and contributions are very low for South America, Middle East and Africa. This is due to the prevailing transport pattern, in which the transport from Europe and the Asian part of Russia into the Arctic is the dominant transport route.

TCEP: The largest contribution of TCEP is from Europe (64 %), despite a share of the emissions of 21 %, with a dominance of contributions from Western Europe (45 %). The Asian regions, which have the largest emissions (56 %) contribute with only 14 %. North America contribute with about 21 % of TCEP in the Arctic similar to the emission contribution (20 %).

### TCEP Emission and contribution to the Arctic [%]

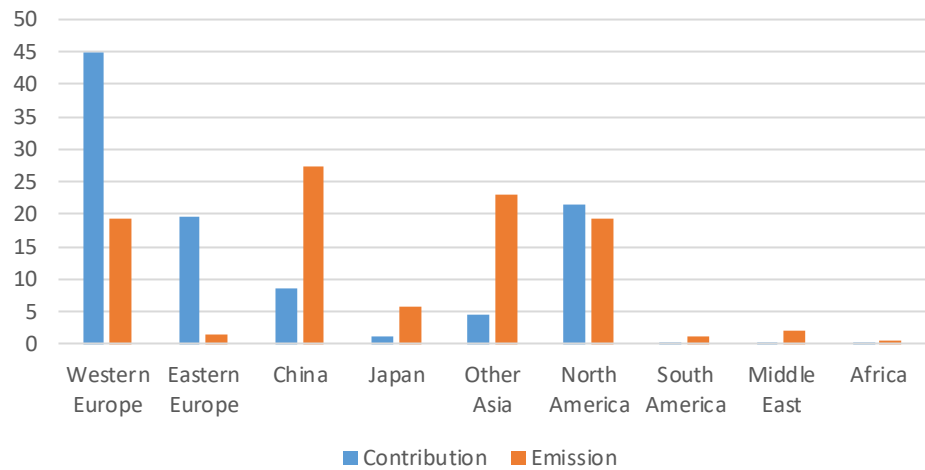


Figure 5.3.1 Share of global emission and contribution to the Arctic of TCEP from each of the nine emission regions.

TCPP: The tendency is the same for TCPP, although Western Europe is a more dominant source region contributing with 56% of the Arctic air concentrations.

### TCPP Emission and contribution to the Arctic [%]

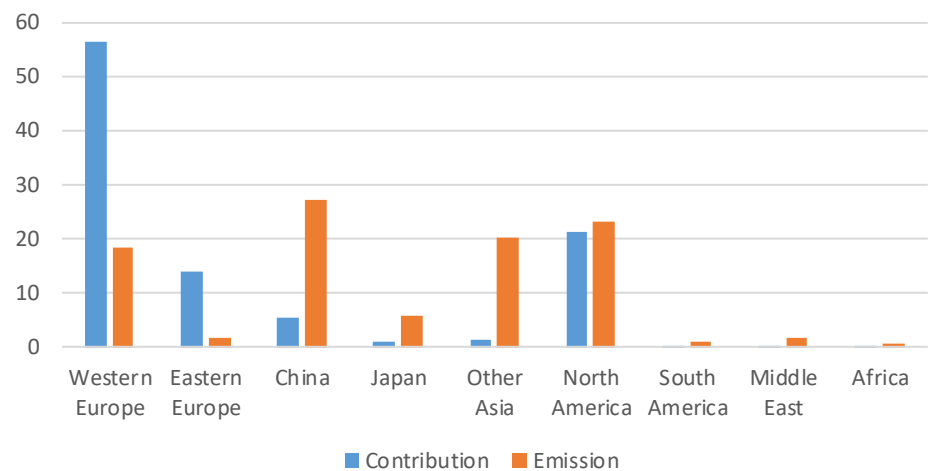


Figure 5.3.2 Share of global emission and contribution to the Arctic of TCPP from each of the nine emission regions.

**TPHP:** An even more dominant contribution from Western Europe is found for TPHP (71 %). The contribution from North America is lower than for TCEP and TCPP (3 %), due to the low emission share from this region (6 %).

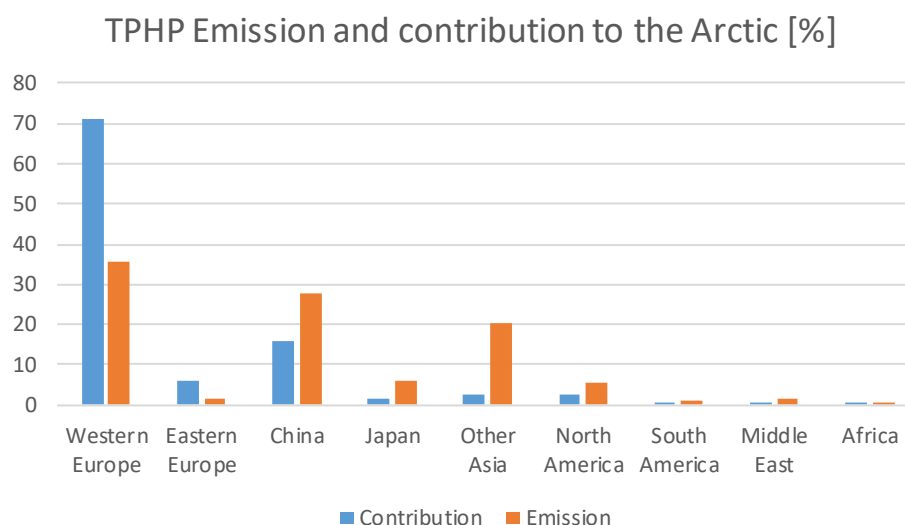


Figure 5.3.3 Share of global emission and contribution to the Arctic of TPHP from each of the nine emission regions.

**EH-TBB:** The largest fraction of the emissions of EH-TBB is from North America (48 %), which is also the largest contributor to the Arctic (32 %). A similar contribution comes from Western Europe despite emitting only 9 % of the global emissions.

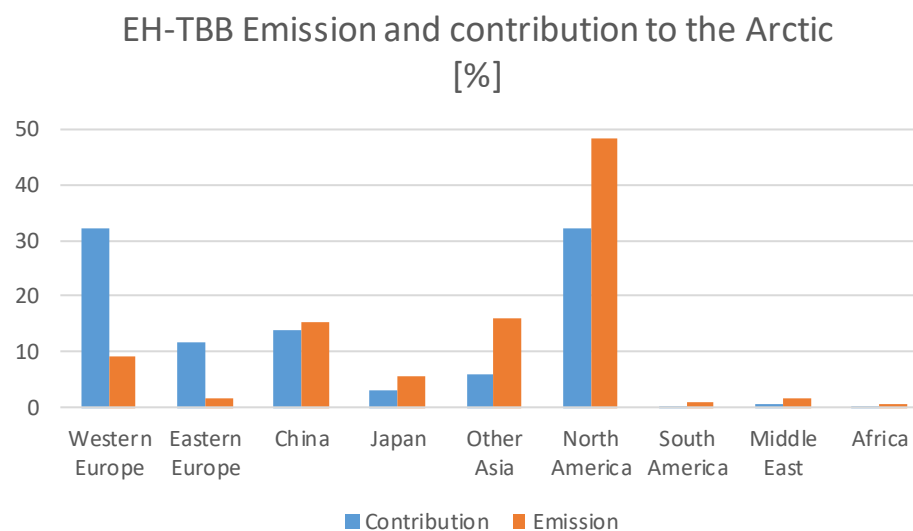


Figure 5.3.4 Share of global emission and contribution to the Arctic of EH-TBB from each of the nine emission regions.

**BEH-TEBP:** The emission pattern for BEH-TEBP is very similar to the pattern for EH-TBB, but the contribution to the Arctic from North America is much higher (51 %) than the contribution from Western Europe (22 %).

### BEH-TEBP Emission and contribution to the Arctic [%]

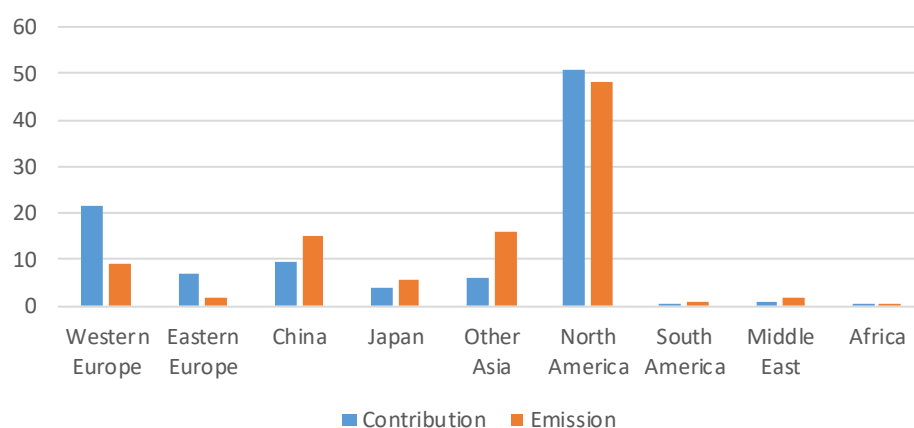


Figure 5.3.5 Share of global emission and contribution to the Arctic of BEH-TEBP from each of the nine emission regions.

**HBBz:** The largest source regions of HBBz is Japan (44 %), China (34 %) and Other Asia (16 %), with less than 2 % in North America and Western Europe. This is also reflected in the contribution, with the three main emitters also displaying the largest contributions.

### HBBz Emission and contribution to the Arctic [%]

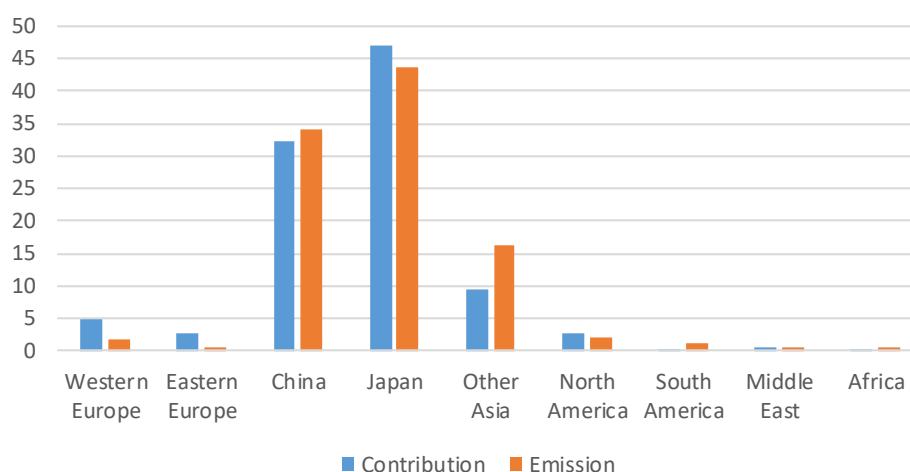


Figure 5.3.6. Share of global emission and contribution to the Arctic of HBBz from each of the nine emission regions.

## 6 Local sources and environmental concentrations

### 6.1 Local dispersion model

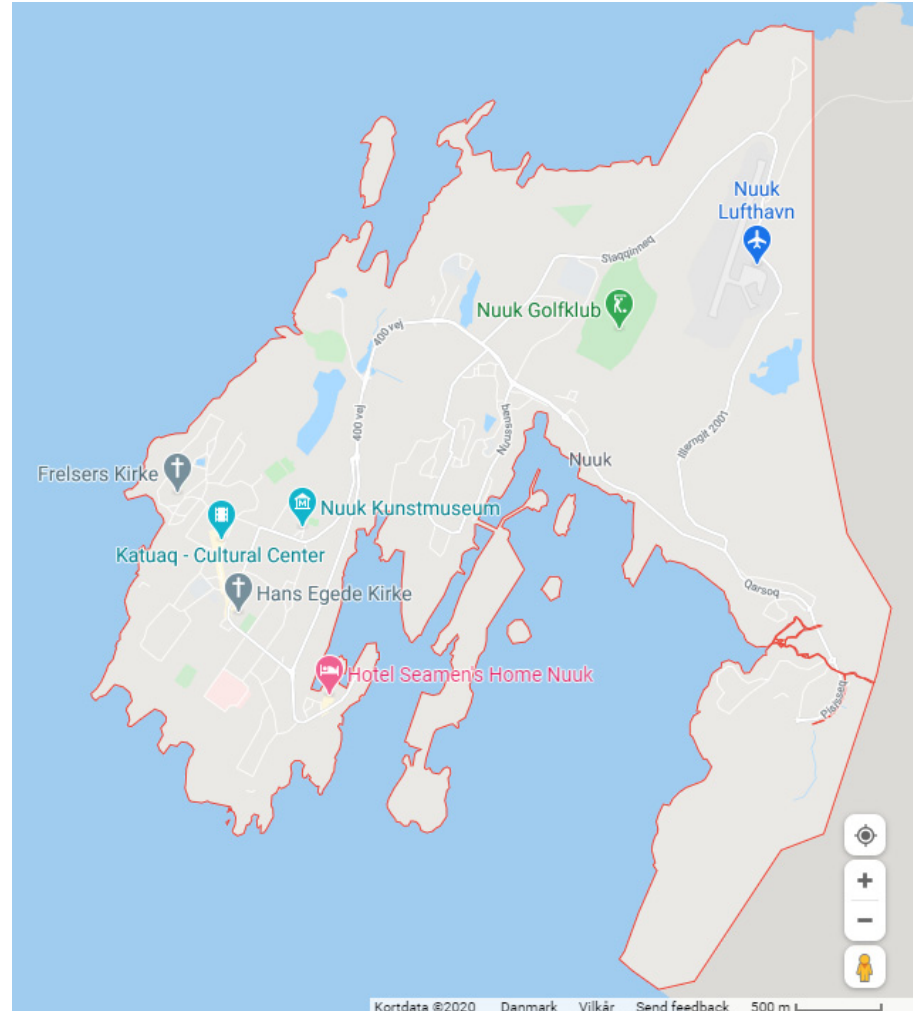


Figure 6.1.1 Outline of Nuuk city area

(<https://www.google.com/maps/place/Nuuk,+Gr%C3%B8nland/@64.1757251,-51.7164242,13z/data=!4m5!3m4!1s0x4ea20dce1ab32725:0x3de425fb4d692306!8m2!3d64.1814099!4d-51.6941381>).

The topography within 3.2 km of Nuuk contains significant variations in elevation, with a maximum elevation change of 297 m and a mean elevation above sea level of 16 m. Within 16 km, the elevation increases up to 1200 m. The area within 3.2 km of Nuuk is covered by sparse vegetation (68 %), water (17 %), and snow and glaciers (12 %), and within 16 km by sparse vegetation (48 %) and water (35 %). For 4.2 months a year, on average, the main wind direction is from south, and for 7.8 months, the main wind direction is from north (<https://weatherspark.com/y/29787/Average-Weather-in-Nuuk-Greenland-Year-Round>). The estimated area of Nuuk city with enclosed bay area (outlined in Figure 6.1.1) is approximately 16 km<sup>2</sup> (4 km x 4 km). The in-

dedicated boundary encloses the resident homes, electricity, heat and water supply plant Nukissiorfiit, waste incineration plant ESANI A/S (planned/under construction), and Dronning Ingrid's Hospital.

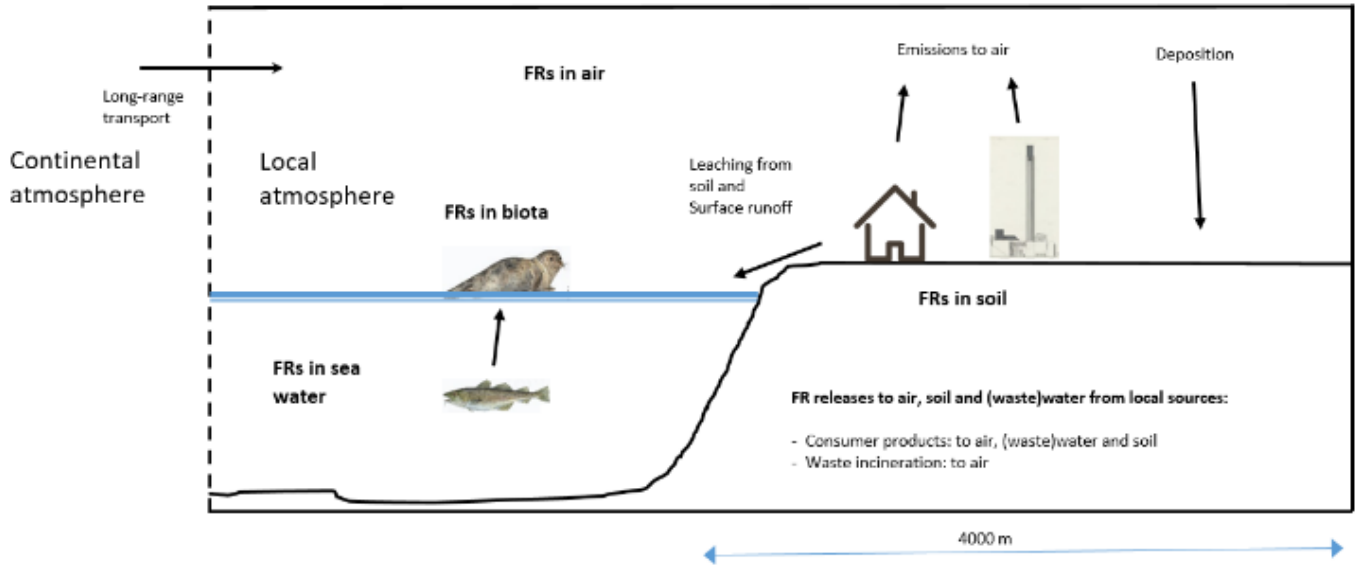


Figure 6.1.2 Local model with FR emissions from indoor and outdoor use of plastic and textile products, and waste incineration, with a simplified marine food-chain relevant for human consumption.

### 6.1.1 Local atmospheric compartment

Atmospheric dispersion modeling is performed for the model boundaries outlined in Figure 6.1.1 and the sources to FR emissions in Figure 6.1.2. A completely mixed box is assumed with in- and outflow of air from the surrounding ambient air:

$$\begin{aligned}
 & \frac{dC_{air,tot}(local)}{dt} \cdot V_{local} \\
 &= E_{air,consumer} + E_{air,waste} - \frac{F_{dep}}{h_{atm}/2} \cdot V_{local} - k_{1,air} \\
 & \cdot C_{air,gas}(local) \cdot V_{local} + w \cdot (C_{air,tot}(ambient) - C_{air,tot}) \cdot A_{wind}
 \end{aligned}
 \tag{Eq. 6.1.1.1}$$

Where  $C_{air,tot}(local)$  and  $C_{air,gas}(local)$  are monthly mean total (combined gas and particulate phases) and gas concentrations of the FRs ( $mg/m^3$ ) from local sources, respectively.  $dt$  is the time step.  $V_{local}$  is the air volume enclosing the city and the local sources, i.e. defined as the product of  $A_{local} = 4000 \text{ m} \times 4000 \text{ m}$ , and a vertical mixing height ( $h_{atm}$  in m), within which complete mixing of air is assumed with uniform concentrations of emitted FRs. Monthly mean mixing heights are based on the Weather Research and Forecasting Model (WRF) (Skamarock et al., 2008). The monthly mean wind speeds ( $w$  in m/s) in 100 m height are assessed to be representative of the transport of air over Nuuk, see Table 6.1.1.1.  $A_{wind} = 100 \text{ m} \times 4000 \text{ m}$  is the air area over Nuuk with height  $h_{atm}$  in the direction of the wind.

$E_{air}$  are monthly mean emissions in kg/month to the atmosphere from sources that emit FRs, i.e. indoor and outdoor consumer use of plastics and textiles,



and waste incineration, see Figure 6.1.2. Emissions are the product of the EF(local) (g/t) to air in Table 4.3.1.1 for the low and high emission scenarios, respectively, and activity data. Activity data are monthly mean used amounts of FR in products and waste (t/month). It is assumed that the main import of goods comes from Europe and products in the supermarket are the same as in Denmark. Greenland is part of Northern America in the light intensity calculations. In this study, it is assumed that residents in Nuuk have similar consumption and waste treatment patterns of plastic products and textiles as residents in Northern America. Consumed amounts of plastic and textile products are set to a fixed fraction of the amount used in Northern America, calculated as the population ratio, i.e.  $F = 5.058\text{E-}05$  (2021 numbers). Nuuk emissions are calculated as the use amount in Northern America multiplied with  $F$ , and subsequently multiplied with EF(local) that is adjusted for the monthly mean temperatures in Nuuk, see Chapter 4.3.2.

In Figure 6.1.1.1a and b, the total monthly mean emissions to air are shown for all FRs, which illustrate the differences between FR emissions on account of corrections of EFs based on  $M$  and  $V_p$ . There are no production facilities for FRs or for products containing FR in Nuuk. Figure 6.1.1.1a is for January with the low emissions scenario, representing a minimum in local emissions, and Figure 6.1.1.1b is for July with the high emission scenario, representing a maximum in local emissions.

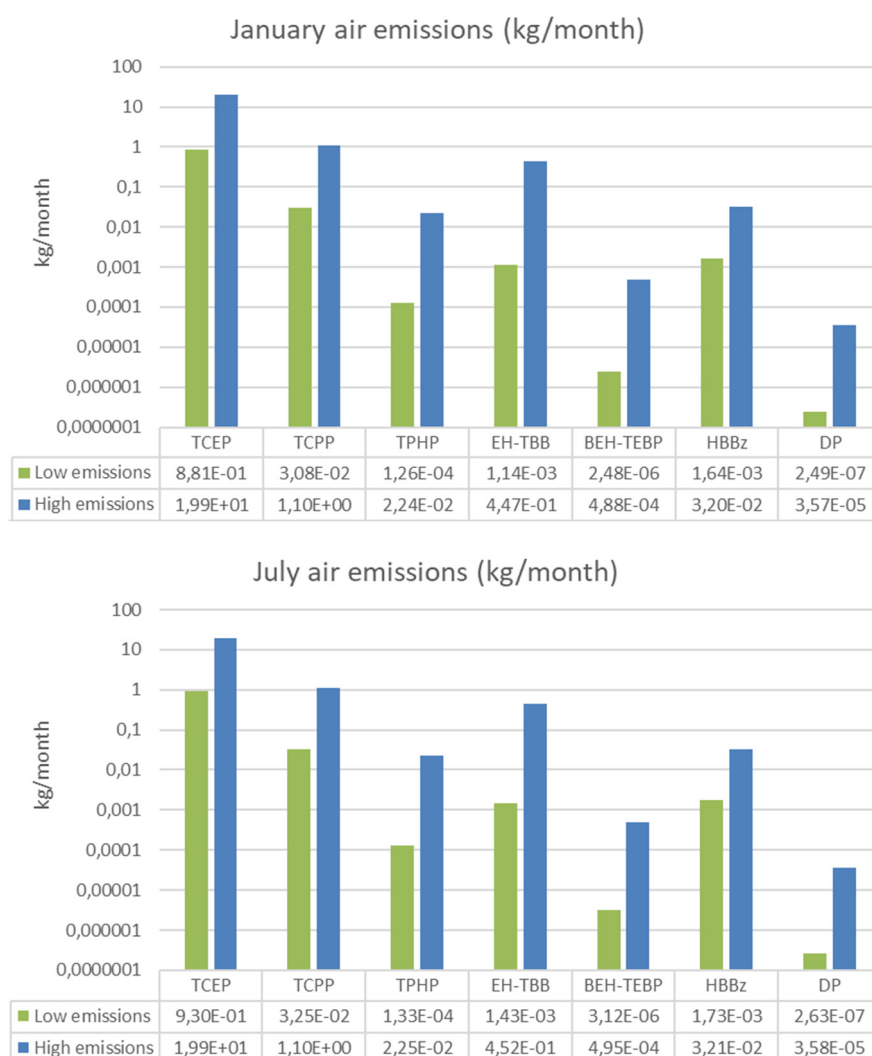


Figure 6.1.1.1a and b Local air emissions in (a) January with low emission scenario and (b) July with high emission scenario.

In Tables 6.1.1.1a and b, the emissions to air are broken down in consumer use of plastic and textiles indoor and outdoor, textile washing and waste incineration. The main contributors to air emissions are indoor use of plastic products and textiles where indoor emissions potentially will reach the outdoor environment via air exchange from ventilation etc. Indoor EFs to air are estimated for  $V_p$  (20 °C) and outdoor EFs to air are estimated for  $V_p$  corresponding to the monthly mean outdoor temperatures. This is reflected in the %-fractions originating from outdoor use that are approx. a factor 10 higher in July compared to January. January thus represents a minimum of emissions and July represents a maximum of emissions (worst-case). Only EH-TBB and BEH-TEBP have outdoor emissions from consumer use that exceed 20 % of the emitted amount, see Table 4.1.2 for the distribution percentages that are set for the different activities.

Table 6.1.1.1a and b FR air and water/soil emission distribution for the local sources in Nuuk. Low and high emission scenarios are shown for (a) January and (b) July emissions, which represent a minimum and a maximum (worst-case) in emissions, respectively.

(a) January		TCEP		TCPP		TPHP		EH-TBB		BEH-TEBP		HBBz		DP	
		Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions
		Percent													
Consumer use of plastic products. indoor	air	45.9	53.2	45.9	53.2	45.9	53.2	84.7	89.4	84.7	89.4	45.9	53.2	45.9	53.2
	water	0	28.9	0	29.4	0	29.4	0	20.5	0	20.5	0	29.4	0	29.4
Consumer use of plastic products. outdoor	air	0.009	0.001	0.009	0.001	0.009	0.001	1.9	0.10	1.9	0.10	0.009	0.001	0.009	0.001
	water or soil	2.1	1.0	2.1	0.9	2.1	0.9	94.7	73.0	94.7	73.0	2.1	0.9	2.1	0.9
Consumer use of textiles. indoor	air	53.7	46.7	53.7	46.7	53.7	46.7	13.2	10.5	13.2	10.5	53.7	46.7	53.7	46.7
	water	0	25.2	0	25.8	0	25.8	0	2.4	0	2.4	0	25.8	0	25.8
Textiles washing	water	0.7	0.0	1.1	0.5	1.1	0.5	0.1	0	0.1	0	1.1	0.3	1.1	0.5
Consumer use of textiles. outdoor	air	0.4	0.02	0.4	0.02	0.4	0.02	0.1	0.01	0.1	0.01	0.4	0.02	0.4	0.02
	water or soil	97.2	45.0	96.8	43.4	96.8	43.4	5.3	4.1	5.3	4.1	96.8	43.5	96.8	43.4
Waste incineration	air	0	0.0	0	0.002	0	0.002	0	0.001	0	0.002	0	0.001	0	0.002

Table 6.1.1.1a and b (continued)...

(b) July		TCEP		TCPP		TPHP		EH-TBB		BEH-TEBP		HBBz		DP	
		Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions
		Percent													
Consumer use of plastic products. indoor	air	43.4	53.1	43.4	53.1	43.4	53.1	67.3	88.2	67.3	88.2	43.4%	53.1%	43.4%	53.1%
	water	0	28.5	0	29	0	29	0	21	0	21	0	29	0	29
Consumer use of plastic products. outdoor	air	0.12	0.007	0.12	0.007	0.12	0.007	21.0	1.32	21.0	1.32	0.12	0.007	0.12	0.007
	water or soil	2.1	1.0	2.1	0.9	2.1	0.9	94.7	73.0	94.7	73.0	2.1	0.9	2.1	0.9
Consumer use of textiles. indoor	air	50.8	46.6	50.8	46.6	50.8	46.6	10.5	10.3	10.5	10.3	50.8	46.6	50.8	46.6
	water	0	25.5	0	26	0	26	0	2	0	2	0	26	0	26
Textiles washing	water	0.7	0.0	1.1	0.5	1.1	0.5	0.1	0.05	0.1	0.05	1.1	0.3	1.1	0.5
Consumer use of textiles. outdoor	air	5.6	0.3	5.6	0.3	5.6	0.3	1.2	0.1	1.2	0.1	5.6	0.3	5.6	0.3
	water or soil	97.2	45.0	96.8	43.4	96.8	43.4	5.3	4.1	5.3	4.1	96.8	43.5	96.8	43.4
Waste incineration	air	0	0.0	0	0.02	0	0.02	0	0.02	0	0.02	0	0.01	0	0.02

$F_{dep}$  is the monthly mean net atmospheric deposition (wet and dry) of compound in  $mg/m^2/month$ .  $F_{dep}(dry)$  is calculated as the settling velocity ( $v_{dep}(dry)$  in  $m/s$ ) multiplied with  $C_{air,tot}$ . Mean settling velocities for four different compounds, i.e. ozone,  $SO_2$ ,  $HNO_3$  and  $NH_3$ , and three different types of surfaces, i.e. tundra, representing the Nuuk area without snow or ice, water and ice/snow, are used to calculate mean settling velocities for land and water, respectively, see Table 6.1.1.2.  $F_{dep}(wet)$  cannot be extracted from the calculations, however, the settling velocities for dry deposition are for compounds that deposit rapidly compared to the FRs, which gives a conservative estimate of the dry deposition.

$k_{1,air}$  is the 1st order degradation rate of gas phase compound in the atmosphere in  $s^{-1}$ .

Assuming constant emissions and wind speeds, the steady-state air concentration is:

$$C_{air,tot} = \frac{\sum_i E_{air,i} - \frac{F_{dep}}{h_{atm/2}} V_{local} + w \cdot C_{air,tot}(ambient) \cdot A_{wind}}{\left( \frac{k_{1,air} \cdot V_{local}}{R} + w \cdot A \right)} \quad (Eq. 6.1.1.2)$$

Table 6.1.1.2 Parameters in the local dispersion model, compiled and estimated for 2019.

Month	w(100 m) (m/s)	$h_{atm}$ (m)	$v_{dep}(dry)$ (land) (m/s)	$v_{dep}(dry)$ (water) (m/s)
Jan-2019	8.1	330.9	0.0004	0.0074
Feb	6.7	227.0	0.0015	0.0101
Mar	7.7	426.8	0.0032	0.0053
Apr	7.2	485.5	0.0059	0.0045
Maj	5.6	497.7	0.0080	0.0046
Jun	5.9	494.8	0.0072	0.0045
Jul	4.2	557.8	0.0081	0.0042
Aug	4.1	472.0	0.0102	0.0051
Sep	5.6	399.1	0.0095	0.0052
Okt	6.0	278.4	0.0135	0.0076
Nov	7.1	279.6	0.0103	0.0083
Dec	7.9	281.7	0.0034	0.0081

Assuming wind speeds of 6 m/s it takes the air approx. 11 minutes to travel across Nuuk. With an approx. settling velocity of 0.003 m/s FRs in the air would have settled approx. 2 m during the 11 minutes. This means that only a very small fraction of FRs emitted within the Nuuk area have settled before the air reaches the outer boundaries of Nuuk. Similarly, the degradation is insignificant for local emissions of FRs.

These calculations suggest that local sources do not contribute significantly to the deposition of FRs in the local environment. The monthly mean FR concentration in air from local sources, Eq. 6.1.1.2 becomes:

$$C_{air,tot} = \frac{\sum_i E_{air,i}}{w \cdot A_{wind}} = \frac{\sum_i E_{air,i}}{w \cdot 4000 \cdot h_{atm}} \quad (Eq. 6.1.1.3)$$

In Table 6.1.2.2, monthly mean air concentrations for January and July, from local sources, are shown for Nuuk. Wind speeds and mixing heights are from Table 6.1.1.2, and FR emissions for a low and high emission scenario are from Figures 6.1.1.1a and b.

The contribution from specific local sources to the local air concentrations can be estimated from the %-distribution of emissions given in Tables 6.1.1.1a and b for January and July (worst-case emissions), respectively. It must be noted that the air concentration depends on the emissions as well as the wind speed and the mixing height, see Eq. 6.1.1.3. From Table 6.1.1.2, it is seen that the product of these two parameters is lower than average for July and higher than average for January, which confirms that the air concentration in July is higher than in January.

## 6.1.2 Soil compartment

FRs in air from local sources will not deposit locally in Nuuk. However, atmospheric deposition from long-range transport and emissions to soil and (sea) water from local sources will take place. Furthermore, the compounds can be transported via surface run-off and leaching from the soil to the sea where fish and seals can be exposed.

A homogeneously mixed 5 cm top soil layer with no macro pores and symmetry along the horizontal plane is considered. Assuming that the release of FRs from soils to the air is low because of their low Vps and high estimated soil sorption coefficients (Wei et al., 2015), and that the diffusive transport from the topsoil is negligible compared to deposition and vertical flow, the governing differential equation for the total ( $C_{soil,tot} = C_{soil,diss} \cdot R$ ) and dissolved compounds in the soil pore water ( $C_{soil,diss}$ ) (g/m<sup>3</sup>) in the top soil is:

$$\frac{dC_{soil,diss}}{dt} = \frac{F_{dep}}{h_{soil}} \Big|_{z=0} - q \cdot \frac{dC_{soil,diss}}{dz} - k_1 \cdot C_{soil,diss} + \frac{\sum_i E_{soil/water,i}}{4000^2 \cdot h_{soil}} \quad (\text{Eq. 6.1.2.1})$$

Where typical values for top soils;  $h_{soil}$  is the top soil layer thickness (approx. 0.05 m),  $\theta$  is the pore volume fraction in the soil (0.5),  $foc$  is the fraction of organic carbon in particulate matter (0.02 kg OC/kg dw),  $X_s$  is the density of soil (1.3 kg dw/L),  $q$  is the vertical flow of water from homogeneous top soil (0.2 m/year),  $R = (\theta + K_d \cdot X_s)$  is the retention factor,  $K_d = foc \cdot K_{oc}$  is the partitioning coefficient between dry matter and water in L/kg dw,  $K_{oc}$  is the partitioning coefficient (sorption coefficient) between organic carbon and water (L/kg OC),  $k_1$  is first order degradation rate of compound in soil in s<sup>-1</sup>, and  $dz$  is step in the vertical direction in m.

$E_{soil/water}$  (kg/month) is the local emission of total FR to soil and (waste)water, see Figure 6.1.2.1. As a worst-case assumption, all of  $E_{soil/water}$  is assumed to go to the soil compartment.

The deposition term ( $F_{dep}$  in g/m<sup>2</sup>/s) is the atmospheric deposition of total (particulate and dissolved) FR at the soil surface ( $z=0$ ). A mean settling velocity for January and July and from land and water ( $v_{dep}(\text{dry}) = 0.005$  m/s) is used to calculate deposition values from long-range transport, see last row in Table 6.1.2.2.

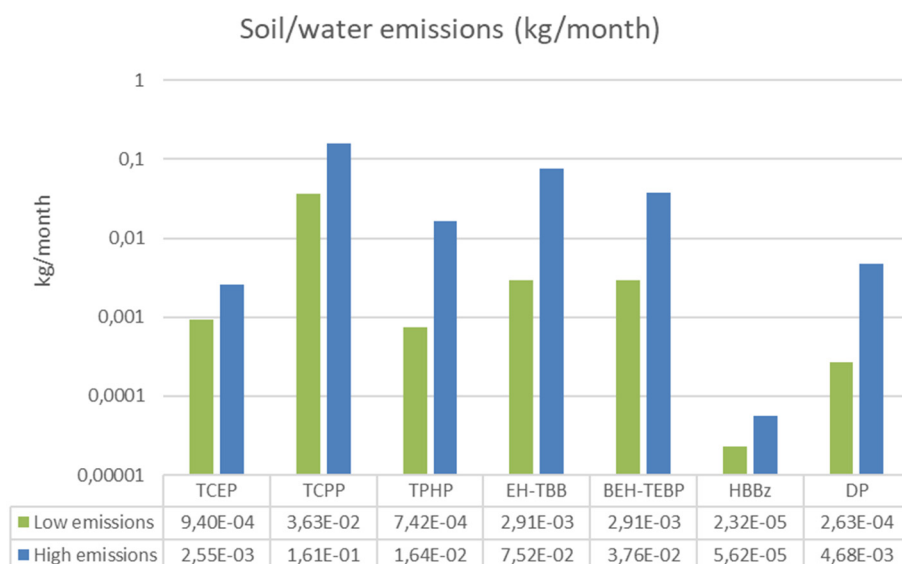


Figure 6.1.2.1 Local emissions to soil (sum of soil and (waste)water emissions) for low and high emission scenarios, respectively. The emissions are the same for all months as there are no corrections of EFs from temperature variations.

Steady-state conditions ( $\frac{dC_{soil,tot}}{dt} = 0$ ) can be assumed as we consider monthly mean concentrations; this yields the following steady-state concentration of dissolved compound in the top soil pore water:

$$C_{soil,diss} = \frac{F_{dep} + \frac{\sum_i E_{soil/water,i}}{4000^2}}{R \cdot (q + k_1 \cdot h_{soil})} \quad (\text{Eq. 6.1.2.2})$$

If the half-life ( $T_{1/2} = \frac{\ln 2}{k_1}$ ) of a compound is smaller than its Kd value, this indicates degradation of the compound in the top soil before it will be leached. Although a removal of particles from water leaching through the soil compartment is possible according to EU TGD (2003), as a worst-case it is assumed that particles are not withheld in the soil and the entire amount of FR in q goes to the marine compartment.

First order degradation rates in soil (derived from half-lives) are shown in Table 6.1.2.1. The linear relationship between Koc and Kow for most semi-volatile nonionizing organic compounds, suggested by Di Toro (1985), are assumed to be valid in this study and is used to calculate Koc values from literature values of Kow:

$$\log Koc = 0.00028 + (0.983 \cdot \log Kow) \quad (\text{Eq. 6.1.2.3})$$

Kow are from EPI Suite by Zhang et al. (2016).

Table 6.1.2.1 First order degradation rates in soil derived from half-lives in soil that are calculated with EPI Suite BIOWIN (Liagkouridis et al., 2015) and from Sverko et al. (2011) for DP.

	TCEP	TCPP	TPHP	EH-TBB	BEH-TEBP	HBBz	DP
$k_1$ (s <sup>-1</sup> )	1.6E-05	6.7E-08	1.1E-07	6.7E-08	6.7E-08	2.2E-08	2.2E-08
Koc (L/kg OC)	3.74E01	7.10E02	4.17E04	4.47E08	6.26E11	1.50E07	7.53E08
Kd (L/kg soil dw)	7.5E-01	1.42E01	8.34E02	8.95E06	1.25E10	3.00E05	1.51E07
R (retention factor)	1.47E00	1.89E01	1.09E03	1.16E07	1.63E10	3.90E05	1.96E07

In Table 6.1.2.3, the modelled monthly mean total and dissolved FR concentrations in soil are shown. Concentrations are from local source, from long-range transport (background) and total, which is the sum of the above. Concentrations from local sources are similar for all months. Measured soil concentrations are from Han et al. (2020), Hao et al. (2020), and Na et al., (2015). These studies report data from Svalbard, but are used for comparison and discussion in the absence of corresponding data from Nuuk.



Table 6.1.2.2 Modelled monthly mean air concentrations (pg/m<sup>3</sup>) in Nuuk. Concentrations are estimated from local sources in Nuuk and from long-range transport with DEHM. January and July represent minimum and maximum concentrations, respectively. The sum is calculated as the mean of modelled January and July air concentrations. Measured concentration data are the same as used in Chapter 5 for long-range evaluation, but only the lowest and highest measured concentrations are stated here for each FR. The deposition is based on long-range air concentrations calculated with DEHM. LB: Longyearbyen (Salamova et al., 2014), RB: Resolute Bay (Sühring et al., 2016), LFL: Little Fox Lake (Yu et al., 2015), Ship: Canadian Arctic (Sühring et al., 2016).

	TCEP		TCPP		TPHP		EH-TBB		BEH-TEBP		HBBz		DP	
	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions
Conc air (local) (pg/m <sup>3</sup> ) January	3.2E+01	7.1E+02	1.1E+00	3.9E+01	4.5E-03	8.1E-01	4.1E-02	1.6E+01	8.9E-05	1.8E-02	5.9E-02	1.1E+00	9.0E-06	1.3E-03
Conc air (local) (pg/m <sup>3</sup> ) July	3.8E+01	8.2E+02	1.3E+00	4.5E+01	5.4E-03	9.2E-01	5.9E-02	1.9E+01	1.3E-04	2.0E-02	7.1E-02	1.3E+00	1.1E-05	1.5E-03
Conc air (long-range) (pg/m <sup>3</sup> ) January	5.1E-01	2.1E+00	5.9E-03	1.9E-01	4.6E-04	1.3E-02	2.0E-04	7.0E-02	5.8E-06	1.0E-03	2.5E-03	4.7E-02	1.3E-06	1.8E-04
Conc air (long-range) (pg/m <sup>3</sup> ) July	1.5E-01	4.4E-01	5.5E-03	1.0E-01	9.3E-05	2.6E-03	8.0E-04	1.1E-01	4.0E-05	2.3E-03	1.1E-01	1.5E+00	4.6E-07	4.8E-05
Conc air (local+long-range) (pg/m <sup>3</sup> ) January (low) and July (high)	3.2E+01	8.2E+02	1.1E+00	4.5E+01	5.0E-03	9.3E-01	4.1E-02	1.9E+01	9.5E-05	2.3E-02	6.1E-02	2.8E+00	1.0E-05	1.5E-03
Measured air (pg/m <sup>3</sup> )	4.0E+00 (LB)	8.6E+02 (Ship)	1.0E+01 (LB)	2.8E+02 (RB)	1.1E+00 (LB)	1.9E+03 (Ship)	1.0E-04 (Alert)	5.8E+01 (LB)	1.7E-03 (Alert)	8.0E+01 (Alert)	3.0E-04 (Alert)	1.7E+00 (LB)	8.0E-03 (LFL)	5.0E+00 (LB)
Deposition (total) (mean Jan and July) (g/m <sup>2</sup> /s)	1.6E-15	6.5E-15	2.9E-17	7.2E-16	1.4E-18	3.8E-17	2.5E-18	4.6E-16	1.1E-19	8.3E-18	2.8E-16	3.8E-15	4.3E-21	5.6E-19

Table 6.1.2.3 Modelled monthly mean soil concentrations in Nuuk, and measured concentrations in soil.

	TCEP		TCPP		TPHP		EH-TBB		BEH-TEBP		HBBz		DP	
	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions
Conc soil,diss (local) (g/m <sup>3</sup> )	1.9E-08	5.0E-08	4.8E-06	2.1E-05	1.4E-09	3.1E-08	6.2E-13	1.6E-11	4.5E-16	5.8E-15	1.9E-13	4.7E-13	4.4E-14	7.7E-13
Conc soil,total (local) (pg/g dw)	2.1E-02	5.7E-02	7.0E+01	3.1E+02	1.2E+00	2.6E+01	5.6E+00	1.4E+02	5.6E+00	7.2E+01	5.8E-02	1.4E-01	6.6E-01	1.2E+01
Conc soil,diss (long-range) (g/m <sup>3</sup> )	1.3E-09	5.3E-09	1.6E-10	3.9E-09	1.1E-13	3.0E-12	2.2E-17	4.1E-15	7.3E-22	5.3E-20	9.6E-14	1.3E-12	3.0E-20	3.9E-18
Conc soil,total (long-range) (pg/g dw)	1.5E-03	6.0E-03	2.3E-03	5.7E-02	9.0E-05	2.5E-03	2.0E-04	3.6E-02	9.1E-06	6.6E-04	2.9E-02	3.9E-01	4.5E-07	5.8E-05
Conc soil,total (sum) (pg/g dw)	2.3E-02	6.3E-02	7.0E+01	3.1E+02	1.2E+00	2.6E+01	5.6E+00	1.4E+02	5.6E+00	7.2E+01	8.6E-02	5.3E-01	6.6E-01	1.2E+01
Measured soil (pg/g dw)	1.5E+02	2.9E+03	1.3E+02	6.6E+03	2.6E+02	3.8E+03	1.4E+01	6.4E+01	no data	no data	9.8E+00	1.5E+01	1.5E+06	2.0E+05

### 6.1.3 Marine compartment

Inflow of FRs to the marine environment in Nuuk constitutes emissions to water from local outdoor consumer use, atmospheric deposition to water areas from long-range transport, and leaching and surface run-off from soil. Removal processes are sedimentation of particles from bulk water, volatilization from water to air, degradation and bio-uptake of dissolved compounds. However, these removals are considered negligible compared to the removal of FR with the tidal water exchange. Due to the continuous filling up and emptying of the fjord system, there is a very strong tidal current along the north-west coast. Thus, it is likely that seawater emptied out during a tidal cycle from the considered volume will result in complete removed during one cycle. Assuming that the entire amount of FR in  $q$  enters the local marine water and the ambient inflowing tidal water has a FR concentration of zero (neglecting the contribution from long-range deposition on the deeper seawaters), the monthly mean concentration of total FR in seawater

( $C_{sea,tot}$  in  $\frac{mg}{m^3}$ ) can be found from:

$$\begin{aligned} \frac{dC_{sea,tot}(local)}{dt} \cdot V_{sea,local} &= \sum_i E_{water,i} + F_{dep,long-range} \cdot A_{sea,local} + q \cdot C_{soil,tot} \\ &\quad \cdot 4000^2 - 2 \cdot V_{tide}(t) \cdot C_{sea,tot}(local) \end{aligned} \quad (Eq. 6.1.3.1)$$

Which gives the following steady-state solution:

$$C_{sea,tot}(local) = \frac{\sum_i E_{water,i} + F_{dep,long-range} \cdot A_{sea,local} + q \cdot C_{soil,tot} \cdot 4000^2}{2 \cdot V_{tide}(t)} \quad (Eq. 6.1.3.2)$$

Assuming that the entire FR amount enters the local marine compartment 0 - 1 km from the shore  $A_{sea,local}$  is approx.  $17.3E06 \text{ m}^2$ . The lowest water level is 2.46 m under the mean water level in Nuuk ([https://www.dmi.dk/fileadmin/user\\_upload/Bruger\\_upload/Tidevand/2021/Nuuk.pdf](https://www.dmi.dk/fileadmin/user_upload/Bruger_upload/Tidevand/2021/Nuuk.pdf)), which gives a tidal local seawater volume exchange per day of:  $2 \cdot V_{tide} = 2 \cdot A_{sea,local} \cdot 2 \cdot 2.46 \text{ m} = 1.7E08 \text{ m}^3/\text{day}$ .

In Table 6.1.3.1, the FR concentrations in the local seawater, calculated from Eq. 6.1.3.2, are shown for the low and high emission scenarios, respectively. A separation of the sea concentration in a contribution from local sources and long-range transport is not straightforward, as  $q$  in Eq. 6.1.3.2 consists of contributions from both sources. However, local sources are the main contributor to FRs in  $q$ , as explained for the soil compartment, so FRs in  $q$  are considered to originate primarily from local sources. All the FR in  $q$  are assumed to be in the particulate phase.

Table 6.1.3.1 Modelled monthly mean concentrations in marine water, fish and seal in Nuuk. Concentrations are estimated from local emissions to water from outdoor product use and from long-range transport. Concentrations are mean values for January and July, representing minimum and maximum concentrations, respectively. Measured concentration data are selected as representative of lowest and highest particle or dissolved phase concentrations (> limit of quantification (LOQ)).

	TCEP		TCPP		TPHP		EH-TBB		BEH-TEBP		HBBz		DP	
	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions
Conc sea,total (local) (g/m <sup>3</sup> )	1.9E-10	5.1E-10	1.2E-08	5.2E-08	2.2E-10	5.0E-09	9.4E-10	2.4E-08	9.4E-10	1.2E-08	1.0E-11	4.6E-11	9.6E-11	1.7E-09
Conc sea,total (long-range) (g/m <sup>3</sup> )	1.4E-11	5.7E-11	2.5E-13	6.3E-12	1.2E-14	3.3E-13	2.2E-14	4.0E-12	1.0E-15	7.3E-14	2.5E-12	3.3E-11	3.8E-17	4.9E-15
Conc sea,total (sum) (g/m <sup>3</sup> )	2.0E-10	5.6E-10	1.2E-08	5.2E-08	2.2E-10	5.0E-09	9.4E-10	2.4E-08	9.4E-10	1.2E-08	1.3E-11	7.9E-11	9.6E-11	1.7E-09
Measured seawater (g/m <sup>3</sup> )	8.2E-07	8.6E-05	2.1E-07	6.5E-05	9.9E-10	6.3E-05	no data	no data	<LOQ	1.3E-09	<LOQ	2.0E-11	2.0E-12	1.2E-07
PECfish,meat (µg/kg)	2.6E-07	7.3E-07	9.4E-05	4.1E-04	8.6E-05	1.9E-03	9.9E-03	2.6E-01	1.2E-05	1.6E-04	1.3E-04	7.9E-04	9.6E-06	1.7E-04
PECseal,meat/blubber (µg/kg)	2.6E-08	7.3E-08	9.4E-06	4.2E-05	1.7E-05	3.8E-04	3.0E-03	7.7E-02	1.2E-06	1.6E-05	1.3E-04	7.9E-04	9.6E-07	1.7E-05
Measured fish (µg/kg ww)	no data	no data	no data	no data	no data	no data	<LOQ	2.8E-01	<LOQ	<LOQ	4.0E-03	7.2E-02	<LOQ	<LOQ
Measured seal (µg/kg ww)	no data	no data	no data	no data	<LOQ	7.2E+00	<LOQ	5.0E-01	<LOQ	<LOQ	<LOQ	<LOQ	6,7E-02 <sup>1)</sup>	1,1E-01 <sup>1)</sup>

<sup>1)</sup> Sum of syn-DP and anti-DP.

Fish caught in the local marine environment have been exposed to FRs in the water, and the Predicted Environmental Concentration (PEC) of FRs in fish meat is calculated according to EU TGD (2003):

$$PEC_{fish,meat} = PEC_{sea(local)} \cdot BCF_{fish} \cdot BMF_1 \quad (\text{Eq. 6.1.3.3})$$

Where;  $PEC_{sea(local)}$  is the predicted dissolved concentration in the local marine water. As a worst-case approach it is set to the total concentration ( $C_{sea,tot(local)}$ ).  $BCF_{fish}$  is the compound bioconcentration factor in fish and  $BMF_1$  is the biomagnification factor. These are found from the compound log Kow value according to EU TGD (2003), from the PubChem database (<https://pubchem.ncbi.nlm.nih.gov/>), and Vorkamp et al. (2019b), see Table 6.1.3.2.

According to EU TGD (2003) the direct uptake of compounds from the environment, i.e. from water and sediment, is only of minor relevance to top predators like seal. For a first tier (or trophic level) of predators a worst-case assumption is that they obtain their prey equally from the local and regional areas, respectively. For the second tier (the top predators) it can be assumed that they obtain their prey mainly (approximately 90 %) from the larger-scale regional marine environment (200 x 200 km<sup>2</sup>). The regional scale marine environment is assumed not to be influenced by the local activities in Nuuk due to dilution in the atmospheric and marine compartments. The concentration in top predator (seal) meat, is set equal to the concentration in blubber, and is found from (EU TGD, 2003) to be:

$$PEC_{toppred.meat} = 0.1 \cdot PEC_{sea(local)} \cdot BCF_{fish} \cdot BMF_1 \cdot BMF_2 \quad (\text{Eq. 6.1.3.4})$$

Where;  $PEC_{sea(local)}$  is set to 10 % of  $C_{sea,total(local)}$  and  $BMF_2$  is the biomagnification factor for top predators, which is based on log Kow and  $BCF_{fish}$ , see Table 6.1.3.2.

Table 6.1.3.2 Bioconcentration factors (BCF) for fish and biomagnification factors ( $BMF_1$  and  $BMF_2$ ) for fish and seal (top predator), respectively, from (EU TGD, 2003, <https://pubchem.ncbi.nlm.nih.gov/>, Vorkamp et al., 2019b).

	TCEP	TCPP	TPHP	EH-TBB	BEH-TEBP	HBBz	DP
$BCF_{fish}$ (L/kg)	1.29	7.94	190.6	3500	13	1000	100
$BMF_1$ (-)	1	1	2	3	1	10	1
$BMF_2$ (-)	1	1	2	3	1	10	1

In Table 6.1.3.1, measured concentrations in seawater are selected as representative of lowest and highest particle or dissolved phase concentrations (> limit of quantification (LOQ)). Measurement data are from Na et al. (2020), McDonough et al. (2018), Möller et al. (2010, 2011a, 2011b, 2011c), Sührling et al. (2021), and Gao et al. (2019, 2020). Measurements in fish (Atlantic cod) are from Schlabach et al. (2011), and Dreyer et al. (2019) and measurements in seal are from Sagerup et al. (2010) (reporting data for liver samples of ringed seals from Svalbard), Dam et al. (2011), and Strobel et al. (2018) (both reporting data for blubber of ringed seals from East Greenland), and Vorkamp et al. (2015) (reporting data for blubber of ringed seals from West Greenland). Only data > LOQ are considered.

## 6.2 Additional potential sources and exposures

Residents can be exposed in homes and other indoor environments via inhalation of gaseous and FRs bound to particles and dust, ingestion of dust, and dermal contact with consumer products and dust. Residents can be exposed to FRs in drinking water, e.g. groundwater or other drinking water reservoirs, and animals can be exposed via freshwater. Residents can be exposed via dermal contact to soil and children can be exposed via soil ingestion. Furthermore, residents and animals can be exposed from intake of contaminated vegetation, and humans from intake of livestock meat and milk products. These additional sources are not considered in the calculation of sources and exposure.

## 6.3 Beach litter

Beach litter on a local scale in Nuuk is only included on a qualitative basis. A more efficient waste collection and waste treatment strategy implemented in Nuuk in the last ten years has resulted in considerable less waste being discarded in the environment, including the sea, around Nuuk (personal communication with Frank Rasmussen, Head of environmental department in Sermersooq). A project (Strand in prep., 2022) was conducted from 2015 to 2019 to monitor the trend in occurrence of beach litter, including plastics and textiles, at three beaches on the outer islands in Godthåbsfjorden, se Figure 6.3.1, each with two surveys per year.

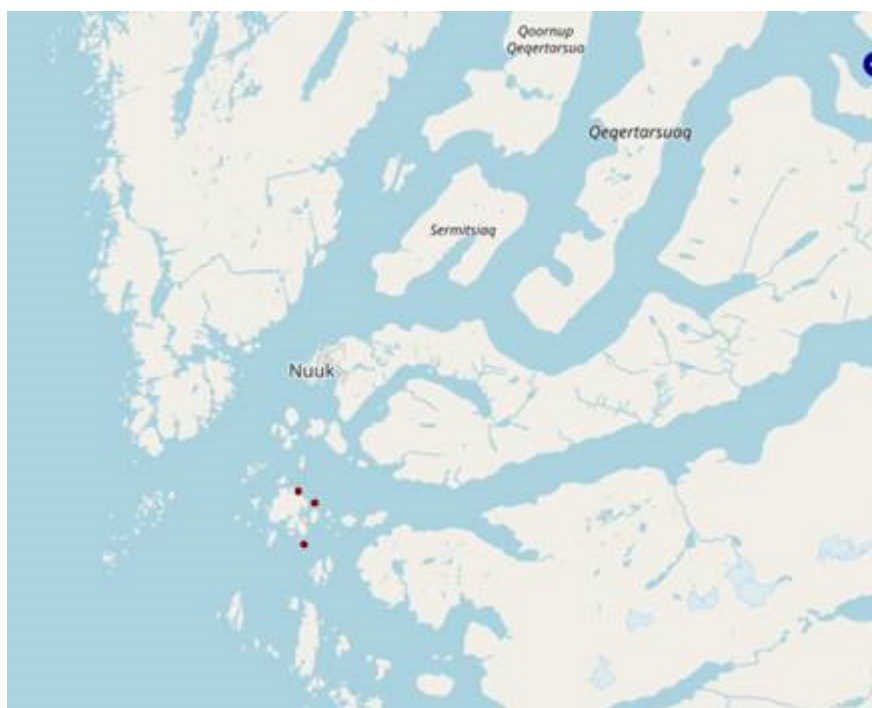


Figure 6.3.1 Sampling sites for beach litter on three beaches on the outer island in Godthåbsfjorden near Nuuk.

The findings show that plastic litter items predominantly have Danish or Greenlandic labels and are therefore assumed to be from local land- and sea-based sources, probably in Nuuk, and from smaller sources in the area and from ships. 300 m sections of the beaches were sampled, i.e. cleaned for litter, during each survey. Such random samples are not representative of the entire coastline and cannot be used for extrapolating the total amount of beach litter. In Figure 6.3.2 the litter categories relevant for FR use are shown, and it is seen

that these comprise approx. 11 % of the total number of litter items. PE, PP and PS make up the bulk of the items (approx. 89 %), but are not relevant to this study as they do not contain FR, c.f. Table 4.1.1.

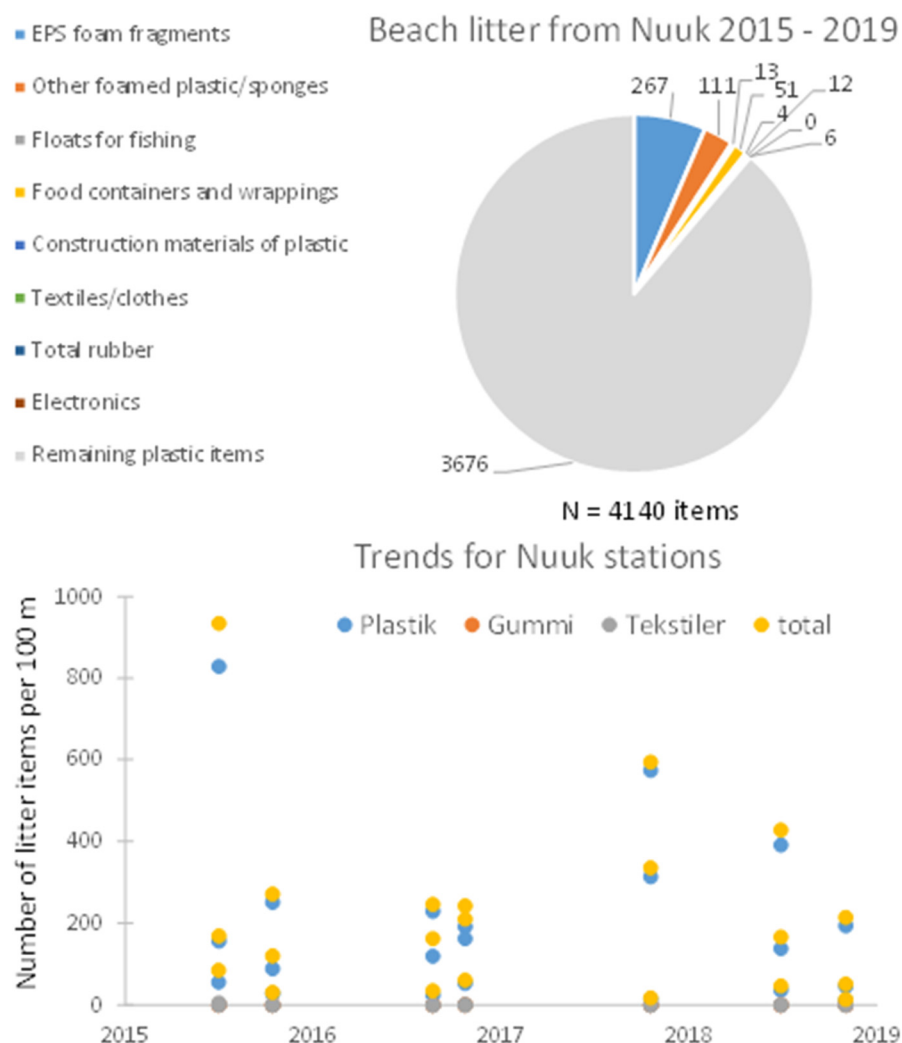


Figure 6.3.2 Litter categories relevant for use/content of FRs (upper figure), and trends for abundance and types of plastic, rubber, textile and total sampled beach litter (lower figure) on three beaches during three sampling surveys per year from 2015 to 2019 (Strand in prep., 2022).

Studies on migration, leaching and release of additives in plastic particles in the marine environment are scarce. However, a recent study specifically addressing leaching of  $\Sigma$ HBCDD from polystyrene foam macrodebris, larger than 2 cm with the outer 2-3 mm removed, that were collected on beaches, showed that leaching of  $\Sigma$ HBCDD occurred at rates of over 150 ng/g/d for relatively long durations (Aminot et al., 2020). Such numbers can be assessed in relation to FRs in plastics and textiles, and compared to the EF of 16000 g/t to water from outdoor use of plastic products and textiles in Table 4.3.1.1.

## 7 Model evaluation and risk screening

### 7.1 Model evaluation

#### 7.1.1 Local atmospheric compartment

In Table 6.1.2.2, monthly mean air concentrations (sum of particulate and gaseous phases) calculated from local sources and from long-range transport (background), respectively, are shown. January represents a minimum in concentrations, and July represents a maximum in concentrations. The sources' relative contributions to the air concentrations in Nuuk are calculated by dividing the local contributions with the long-range contributions, using the January and July concentrations in the low and high scenarios, respectively. Taking the logarithmic value of the ratio, gives a difference in orders of magnitude (OM), see Table 7.1.1. A comparison shows that the air concentrations from local sources are two and three OM higher than the air concentrations calculated for long-range transport for TCEP, TCPP, TPHP (high), and EH-TBB. For TPHP (low), BEH-TEBP, HBBz and DP the local sources contribute up to one OM more to the air concentrations than does the long-range transport.

As explained in Chapter 5 for long-range transport modeling with DEHM, air measurements are only available from a few Arctic sites for the FRs studied in this project. The same measurement data are used here for comparison with modelled total air concentrations in Nuuk, i.e. the sum of local and long-range contributions. The lowest and the highest measured concentrations are compared with the modelled total air concentrations for January and July, respectively (from Table 6.1.2.2). From Table 7.1.1 it is seen that the modelled air concentrations differ with one OM or less for TCEP, TCPP, EH-TBB (high), BEH-TEBP (low), and HBBz (high), with two and three OM for TPHP, EH-TBB (low), HBBz (low) and DP (low), and four OM for BEH-TEBP (high) and DP (high). The modelled concentrations exceed the measured concentrations, i.e. negative OM, for the low emission scenarios of TCEP, EH-TBB and HBBz. However, their modelled concentrations for the high emission scenarios are all below one OM for the highest measured concentrations. There are significant variations between measured lowest and highest concentrations, but for the three OPFRs and DP the differences compared to the modelled concentrations for the low and the high emission scenarios do not vary more than an OM of one.

The comparison of the modelled FR air concentrations from local sources vs. long-range transport indicates that local sources are the predominant contributors at Nuuk. This is more pronounced for the OPFRs and EH-TBB, whereas the air concentrations of two BFRs (BEH-TEBP and HBBz) and DP are more equally influenced by the two types of sources. BEH-TEBP and DP have the lowest Vps and therefore the lowest EFs to air. This is substantiated in the Arctic environment where the local outdoor emissions are significantly lowered due to the reduced EF to air. There is a good agreement between modelled and measured air concentrations for two of the OPFRs and for one of the emission scenarios for the three BFRs. There is a poor agreement between the measured and modelled concentrations for TPHP and even more so for DP. In Chapter 5, further details on a comparison between the measured vs. modelled background concentrations are given.

### 7.1.2 Soil compartment

The modelled total and dissolved FRs concentrations in the soil are given in Table 6.1.2.3. These are divided in contributions from local sources and from long-range transport. Local sources contribute with emissions to soil and (waste)water, and long-range transport contributes with deposition from the atmosphere. An assessment of their respective contributions to the soil concentration shows that local sources contribute with up to one OM more for TCEP and HBBz, four OM for TCPP, TPHP and EH-TBB, and five to six OM for BEH-TEBP and DP. BEH-TEBP and DP have the lowest EFs to air (see above), which will influence the long-range transport and thus the deposition to soil. EFs to soil and (waste)water from local sources are not affected by adjustments due to lower temperatures in Nuuk.

A comparison between modelled total soil concentrations, i.e. the sum of local and long-range contributions, and measured soil concentrations shows that modelled TCEP concentrations are an OM of four and five lower than the measured lowest and highest concentrations, respectively. All other FRs have modelled soil concentrations that are comparable to and up to two OM lower than measured soil concentrations. Possible reasons for the higher measured soil concentrations are simplifications in the local model and used parameters, e.g. for the leaching rates and soil characteristics. Further differences are that Svalbard, where the OPFR measurements are collected, has industrial activities from fossil fuel production, the airport and activities from tourism. No representative soil measurements are available for BEH-TEBP.

For the soil compartment, local sources are significantly more influential than long-range transport via atmospheric deposition for all FRs, except for TCEP and HBBz that have the highest EFs to air among the OPFRs and the BFRs, respectively. BEH-TEBP and DP have the lowest EFs to air and also the highest contribution to soil from local sources.

### 7.1.3 Marine compartment

The estimated FRs concentrations in the marine water are given in Table 6.1.3.1. These are divided into contributions from local emissions to water and leaching from soil, and from long-range atmospheric deposition to the sea. An assessment of their respective contributions to the seawater concentrations shows that local sources contribute up to one OM more for TCEP and HBBz and up to five and six OM for the other FRs. This corresponds with the soil results.

A comparison between modelled total seawater concentrations, i.e. the sum of local sources and long-range transport, and the lowest and the highest measured seawater concentrations shows that the modelled TCEP concentrations are four and five OM lower than the measured concentrations, for the low and high scenarios, respectively. Modelled TCPP and TPHP concentrations are comparable (up to one OM lower) for the low scenario, and three and four OM for the high scenario, respectively. BEH-TEBP and HBBz modelled values are comparable (minus one OM) for the high scenario. The DP modelled concentrations are two OM higher and two OM lower than the lowest and highest measured seawater concentrations, respectively. The long-range transport in the marine compartment is not included in the global model. This also includes deposition from the atmosphere to the marine compartment outside the area around Nuuk. Therefore, the ratio between local



and long-range related contamination for the marine compartment may over-predict the influence from local sources.

The ratio in contributions to FRs in seawater from local sources vs. long-range transport also applies to biota. Measurements in fish (Atlantic cod) and seal are scarce, but the values in Table 6.1.3.1 show that for EH-TBB and HBBz in cod the modelled concentrations are a maximum of two OM lower compared to the measurements. Measured values of EH-TBB in blubber from seal are comparable (one OM higher) to modelled values, and measured values of TPHP and DP are four to five orders of magnitude higher than the modelled values. These large differences could be caused by a higher BCF in blubber compared to the BCF for meat, which is used in the model.

#### 7.1.4 Comparison with other studies

Previous studies have shown a dominance of chlorinated OPFRs or an equivalence between the chlorinated and non-halogenated OPFRs in outdoor air (Wei et al., 2015), which is in agreement with this study where TCEP and TCPP have approx. three OM higher modelled air concentrations than TPHP. This reflects differences in EFs, their geographical distributions of e.g. production sites, long-range transport propensities among the OP analogues (Wei et al., 2015).

The proportion of non-halogenated OPFRs in air were the highest in urban regions (Takeshi et al., 2006; Green et al., 2008) due to the proximity to emission sources. In rural regions, the proportion decreased, possibly due to the decreased transport potential and/or the increased degradation of non-halogenated OPs during the transport processes. However, the dominance of non-halogenated OPFRs reported in some air samples from rural areas (Marklund et al., 2005; Salamova et al., 2014a, 2014b) suggests the possible influence of local sources in addition to remote contributions (Wei et al., 2015). Findings by Salamova et al. (2014b), Möller et al. (2012) and Cheng et al. (2013) indicate that atmospheric transport potentials of the non-halogenated OPFRs were similar to the halogenated OPFRs and that the non-halogenated OPFRs might also be transported to the remote areas via the atmosphere. This is also confirmed by the modelling results in this study, where the ratio between contributions to the air concentrations in Nuuk from local sources vs. long-range transport is similar for the chlorinated OPFRs (TCEP and TCPP) and the non-chlorinated OPFR (TPHP).

For the marine atmosphere, chlorinated OPFRs were also the prominent constituents, which was consistent with their increased persistence during long-range transport, compared to non-halogenated OPFRs (Wei et al., 2015). The predominance of TCPP in the European marine waters (Castro-Jimenez et al., 2014) and the dominance of TCEP in the Sea of Japan (Möller et al., 2012a), potentially demonstrates different use patterns of OPFRs between Europe and Asia (Wei et al., 2015). In the present study, it is found that the production sites, and thus emissions from production, are mainly in China for TCEP, whereas the produced fraction in Europe is larger for TCPP.

Compared with the BFRs, the sum of OPFR concentrations were one to two OM higher in the atmospheric particles in the European Arctic (Salamova et al., 2014a) and two to three OM higher for the atmosphere in the Great Lakes basin (Salamova et al., 2014b). In the present study, the modelled air concentrations of sum OPFRs are one and two OM higher than the sum of BFRs for

the low and high emission scenario, respectively. These findings confirm that OPFRs have larger emissions and/or long-range transport potentials compared to the BFRs, and that all studied FRs have potential for long-range transport to remote areas, including the Arctic (Möller et al., 2011b, 2012a; Cheng et al., 2013).

The FRs present in the atmosphere might be deposited in the aquatic and terrestrial environments via precipitation (Wei et al., 2015). Significantly higher sum OPFR concentrations were reported in urban compared with rural precipitation because of local emissions from urban settlements and road traffic. Studies (Takeshi et al., 2006; Bacaloni et al., 2008; Regnery et al., 2009, 2010a,b) state that long-range transport was the dominant input mode for the remote atmosphere. However, this is not confirmed in the present study.

Table 7.1.1 Model evaluation of contributions to concentration in Nuuk from local emissions vs. long-range transport, and of measured concentrations vs. modelled concentrations (sum of local emissions and long-range transport) in Nuuk. Modelled concentrations for the low emissions and high emissions are compared to lowest and highest measured values, respectively. The differences are expressed as orders of magnitude by taking the logarithmic value of the ratios. Negative values indicate higher modelled concentrations compared to measured concentrations.

	TCEP		TCPP		TPHP		EH-TBB		BEH-TEBP		HBBz		DP	
	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions
Air log(model: local sources/long-range) January (low) and July (high)	2	3	2	3	1	3	2	2	1	1	1	0	1	1
Air log(measured/model)	-1	0	1	1	2	3	-3	0	1	4	-2	0	3	4
Soil log(model: local sources/long-range)	1	1	4	4	4	4	4	4	6	5	0	0	6	5
Soil log(measured/model)	4	5	0	1	2	2	0	0			2	1	1	2
Marine water log(model: local sources/long-range)	1	1	5	4	4	4	5	4	6	5	1	0	6	6
Marine water log(measured/model)	4	5	1	3	1	4				-1		-1	-2	2
Fish log(measured/model)								0			1	2		
Seal log(measured/model)						4		1					5	4

Blanks: no measurements.

## 7.2 Risk screening

### 7.2.1 Human risk from indirect exposure via the environment

The daily dose (mg/kg bw) of FR is calculated for intake of air, fish and seal, from:

$$Dose(FR)_{air} = \frac{C(FR)_{air} \cdot IH(air)}{bw} \cdot \frac{BIO_{in}}{BIO_{oral}} \quad (\text{Eq. 7.2.1.1})$$

$$Dose(FR)_{fish\ or\ seal} = \frac{C(FR)_{fish\ or\ seal} \cdot IH(fish\ or\ seal)}{bw} \quad (\text{Eq. 7.2.1.2})$$

Standard daily intake values (IH) of air, fish and predator (seal), are found in EU TGD (2003). BIO<sub>in</sub> is the bioavailability for FR through inhalation and BIO<sub>oral</sub> is the bioavailability of FR through the oral route. C<sub>air</sub> is the calculated air concentration in Nuuk from local sources and long-range transport. C<sub>fish</sub> and C<sub>seal</sub> are PEC<sub>fish,meat</sub> and PEC<sub>toppred,meat</sub> from Eq. 6.1.3.3 and Eq. 6.1.3.4, respectively, and bw is the body weight of the considered human. Total dose of FR is the sum of doses for all media.

Table 7.2.1.1 Parameters used for calculation of human exposures via the environment (EU TGD, 2003).

Parameter	Explanation	Value	Units
IH(air)	Daily intake of air	20	m <sup>3</sup> /d
IH(fish)	Daily intake of fish	0.115	kg ww/d
IH(predator, seal)	Daily intake of seal	0.115	kg ww/d
BIO <sub>in</sub>	Bioavailability for FR through inhalation	0.75	-
BIO <sub>oral</sub>	Bioavailability for FR through oral route	1	-
bw	Body weight of human	70	kg bw

Calculation of human risk is based on the method outlined in the REACH Guidance documents, e.g. ECHA (2012; 2018). The Risk Characterization Ratio (RCR) is calculated for the sum of FR exposures from:

$$RCR(FR)_{total} = \sum_i \frac{Dose(FR)_i}{DNEL(FR)\ or\ RfD(FR)} \quad (\text{Eq. 7.2.1.3})$$

Where i is the medium (air, fish, seal) and DNEL is the Derived No Effect Level (mg/kg bw/d), and RfD is the Reference Dose (mg/kg bw/d). RCR > 1 represents a potential risk for the combined risk from all exposure pathways of FR. DNELs are derived from the critical doses, i.e. No Observed Adverse Effect Levels (NOAEL) or Lowest Observed Adverse Effect Levels (LOAEL), and from using relevant Assessment Factors (AF). AF are assigned based on the studies from which the NOAEL or LOAEL are derived. RfD is used for estimating the daily average exposure dose under which the risk of non-carcinogenic effects is expected to be reduced to a non-detected level if exposed to this dose level during the life time (Wang et al., 2021). Deriving AFs was beyond the scope of this study, therefore only available DNEL and RfD values are used. DNEL and RfD can be derived for different health endpoints, exposure pathways, and for adults or children. As a worst-case approach the lowest derived DNEL or RfD, typically for children, are used (Table 7.2.1.2). For further information, the references are given in the table.

The calculated sum(RCR) is 1E-05 and 1E-04, for the low and high emission scenarios, respectively. This indicates no or low risk for human intake of FRs

via inhalation of air, ingestion of fish and seal. Highest contributors to RCR are HBBz (low: 76 %, high: 45 %) and EH-TBB (low: 19 %, high: 42 %) from intake of fish and seal. The BFRs, DP and TPHP have a higher intake fraction of FRs from fish and seal, whereas TCEP and TCPP have higher and equal, respectively, intake fractions of FRs from air.

Inhalation is a common route of human exposure to OPFR (Maddela et al., 2020). In alignment with the results in this report, Luo et al. (2016) found that the calculated inhalation doses of chlorinated OPFRs around an e-waste recycling area and in an urban area in China were much lower than the RfD. This suggests that potential health risk due to inhalation exposure to particle-bound OPFRs in potentially more contaminated sites was low.

### 7.2.2 Environmental risk

The environmental risk screening is performed by calculating the RCR, which is the ratio between PEC and a Predicted No Effect Concentration (PNEC):

$$RCR = \frac{PEC}{PNEC} \quad (\text{Eq. 7.2.2.1})$$

As a worst-case assumption, PEC is the total amount of FR in water, i.e. the dissolved and particulate phases are 100 % bioavailable to the organisms. PNEC can be calculated as the quotient of a toxicological relevant concentration, such as L(E)C50, and a safety factor, which ranges from 1 to 1000. The accuracy of PNEC depends on the availability of dose–effect data for all environmental compartments and the sufficient number of species tested (e.g. Bruin et al., 2015). A safety factor of 1000 is typically used for the PNECs in Table 7.2.2.1.

A  $RCR < 0.1$  indicates no or low risk,  $0.1 \leq RCR < 1$  indicates moderate risk, and  $RCR \geq 1$  indicates high potential risk for adverse effects. RCR is determined for each FR and for the sum of all FRs, assuming additivity of risk. It is seen that the sum(RCR) for terrestrial organisms is  $4E-04$  with highest risk from TCPP and DP. The sum(RCR) for fish is  $2E-04$  with highest risks from EH-TBB and BEH-TEBP. These RCRs indicate low risk for terrestrial organisms and marine fish. However, it should be noted that toxicity data are missing or limited and the accuracy of PNECs are therefore debatable.

The ratio of contributions from local sources vs. long-range transport to the RCRs for soil organisms and fish, are similar to the ratios: “Soil log(model: local sources/long-range)” and “Marine water log(model: local sources/long-range)” in Table 7.1.1.

Garcia-Garin et al. (2020) identified TPHP and some other OPFRs in the tissues of fin whales and their main prey, krill (*Meganyctiphanes norvegica*), but mainly related the presence of OPFRs to the exposure to plastics. The uptake and potential accumulation of OPFRs is highly influenced by their physical-chemical properties, bioavailability and metabolism. In the present study the influence of log Kow values on bioconcentration and biomagnification is considered, c.f. Table 6.1.3.2, in accordance with the EU TGD (2003). Maddela et al. (2020) argue that the trophic biomagnification of OPFRs in the marine system has become the focus rather than in the terrestrial system. From the environmental risk screening (Table 7.2.2.1) the RCR for terrestrial organisms are an OM or more larger than RCR for fish, for TCEP and TCPP.

Table 7.2.1.2 Human risk screening. Modelled daily mean doses for human intake of air, incl. particles, fish (cod) and seal. Lowest DNEL or RfD are used in Risk Characterization Ratios (RCRs).

	TCEP		TCPP		TPHP		EH-TBB		BEH-TEBP		HBBz		DP	
	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions
Dose(air+part) (mg/kg/d)	6.9E-09	1.7E-07	2.4E-10	9.7E-09	1.1E-12	2.0E-10	8.8E-12	4.0E-09	2.0E-14	4.8E-12	1.3E-11	5.9E-10	2.2E-15	3.2E-13
Dose(fish) (mg/kg/d)	8.5E-13	2.4E-12	3.1E-10	1.4E-09	2.8E-10	6.2E-09	3.3E-08	8.4E-07	4.0E-11	5.2E-10	4.2E-10	2.6E-09	3.1E-11	5.6E-10
Dose(seal) (mg/kg/d)	8.5E-14	2.4E-13	3.1E-11	1.4E-10	5.6E-11	1.2E-09	9.8E-09	2.5E-07	4.0E-12	5.2E-11	4.2E-10	2.6E-09	3.1E-12	5.6E-11
Dose(total) (mg/kg/d)	6.9E-09	1.7E-07	5.8E-10	1.1E-08	3.4E-10	7.7E-09	4.2E-08	1.1E-06	4.4E-11	5.8E-10	8.5E-10	5.8E-09	3.4E-11	6.1E-10
Relative dose(FR/sum FRs) (%)	14%	13%	1.1%	0.9%	0.7%	0.6%	83%	85%	0.1%	0.04%	1.7%	0.4%	0.1%	0.05%
DNEL or RfD (mg/kg bw/d)	1.1E-02 <sup>1)</sup>	1.1E-02 <sup>1)</sup>	3.5E-02 <sup>1)</sup>	3.5E-02 <sup>1)</sup>	5.0E-01 <sup>2)</sup>	5.0E-01 <sup>2)</sup>	2.0E-02 <sup>3)</sup>	2.0E-02 <sup>3)</sup>	3.7E-01 <sup>4)</sup>	3.7E-01 <sup>4)</sup>	1.0E-04 <sup>3)</sup>	1.0E-04 <sup>3)</sup>	1.0E-02 <sup>5)</sup>	1.0E-02 <sup>5)</sup>
RCR = Dose(total)/DNEL	6.3E-07	1.6E-05	1.6E-08	3.2E-07	6.8E-10	1.5E-08	2.1E-06	5.5E-05	1.2E-10	1.6E-09	8.5E-06	5.8E-05	3.4E-09	6.1E-08
Relative RCR(FR/sum FRs) (%)	6%	12%	0.1%	0.2%	0.01%	0.01%	19%	42%	0.001%	0.001%	76%	45%	0.03%	0.05%

<sup>1)</sup> DNEL: ECHA (2018a).

<sup>2)</sup> DNEL: Larsson et al. (2018).

<sup>3)</sup> RfD: Wang et al. (2021).

<sup>4)</sup> DNEL: EPA (2016).

<sup>5)</sup> RfD: Wang et al. (2013).

Table 7.2.2.1 Environmental risk screening. Risk Characterization Ratios (RCRs) are calculated as the modelled concentrations in soil (Table 6.1.2.3) and marine water (Table 6.1.3.1) divided with lowest available PNECs for terrestrial organisms and fish, respectively.

	TCEP		TCPP		TPHP		EH-TBB		BEH-TEBP		HBBz		DP	
	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions	Low emissions	High emissions
Conc soil,total (sum) (pg/g dw)	2.3E-02	6.3E-02	7.0E+01	3.1E+02	1.2E+00	2.6E+01	5.6E+00	1.4E+02	5.6E+00	7.2E+01	8.6E-02	5.3E-01	6.6E-01	1.2E+01
PNEC(soil org) (pg/g ww (or dw))	3.4E05 <sup>1)</sup>	3.4E05 <sup>1)</sup>	1.5E06 <sup>2)</sup>	1.5E06 <sup>2)</sup>									7.5E04 <sup>5)</sup>	7.5E04 <sup>5)</sup>
RCR(soil org) = Conc soil,tot/PNEC(soil org)	6E-08	2E-07	5E-05	2E-04									9E-06	2E-04
Conc sea,total (sum) (g/m3)	4.0E-10	1.1E-09	2.4E-08	1.0E-07	4.5E-10	9.9E-09	1.9E-09	4.9E-08	1.9E-09	2.4E-08	2.6E-11	1.6E-10	1.9E-10	3.4E-09
PNEC(fish) (g/m3)	6.5E-02 <sup>1)</sup>	6.5E-02 <sup>1)</sup>	6.4E-02 <sup>2)</sup>	6.4E-02 <sup>2)</sup>	4.2E-04 <sup>3)</sup>	4.2E-04 <sup>3)</sup>	5.3E-04 <sup>6)</sup>	5.3E-04 <sup>6)</sup>	>2.0E-04 <sup>6)</sup>	>2.0E-04 <sup>6)</sup>	1.1E-02 <sup>4)</sup>	1.1E-02 <sup>4)</sup>		
RCR(fish) = Conc sea-water/PNEC(fish)	6E-09	2E-08	4E-07	2E-06	1E-06	2E-05	4E-06	1E-04	1E-05	1E-04	2E-09	1E-08		

<sup>1)</sup> EC (2008).

<sup>2)</sup> EU (2007).

<sup>3)</sup> Cristale et al. (2013).

<sup>4)</sup> Xiong et al. (2019).

<sup>5)</sup> Canada (2019a).

<sup>6)</sup> Schlabach et al. (2011).

Blanks: no data.

## 8 Conclusions

Modelled environmental concentrations in Nuuk were studied for three organophosphorous flame retardants (OPFR: tri(2-chloroethyl) phosphate (TCEP), tri(chloroiso-propyl) phosphate (TCPP) and triphenyl phosphate (TPHP)), three brominated flame retardants (BFR: 2-ethylhexyl-2,3,4,5-tetrabromobenzoate (EH-TBB), bis(2-ethylhexyl) tetrabromophthalate (BEH-TEBP) and hexabromobenzene (HBBz)) and one chlorinated flame retardant, dechlorane plus (DP). Concentrations from long-range transport following global emissions from production, plastic and textile product use and waste treatment, were calculated with the Danish Eulerian Hemispheric Model (DEHM) and compared to concentrations from estimated local emissions in Nuuk from plastic and textile product use and waste incineration, calculated with a local dispersion model.

Table 8.1 Summary of local vs long-range transport contributions to environmental concentrations, and model evaluation with measurements (good agreements in bold), with low and high emission scenarios.

	Local (L) sources vs. long-range transport (LRT)	Measured (ME) vs. Modelled (MO) concentrations
TCEP	L >> LRT (air)	ME = MO (air (high))
	L > LRT (soil, seawater, biota)	ME < MO (air (low))
		ME >> MO (soil, seawater)
TCPP	L >> LRT (air, soil, seawater, biota)	ME = MO (soil (low))
		ME > MO (air, soil (high), seawater (low))
		ME >> MO (seawater (high))
TPHP	L > LRT (air (low))	ME > MO (seawater (low))
	L >> LRT (air (high), soil, seawater, biota)	ME >> MO (air, soil, seawater (high), biota (high))
EH-TBB	L >> LRT (air, soil, seawater, biota)	ME = MO (air (high), soil, biota (high))
		ME << MO (air (low))
BEH-TEBP	L > LRT (air)	ME < MO (seawater (high))
	L >> LRT (soil, seawater, biota)	ME > MO (air (low))
		ME >> MO (air (high))
HBBz	L = LRT (air (high), soil, seawater (high), biota (high))	ME = MO (air (high))
	L > LRT (air (low), seawater (low), biota (low))	ME < MO (seawater (high))
		ME > MO (soil (high), biota (low))
		ME << MO (air (low))
		ME >> MO (soil (low), biota (high))
DP	L > LRT (air)	ME > MO (soil (low))
	L >> LRT (soil, seawater, biota)	ME << MO (seawater (low))
		ME >> MO (air, soil (high), seawater (high))

= is zero orders of magnitude difference.

< or > is one order of magnitude difference.

<< or >> is two or more orders of magnitude difference.

The contribution to the local environment from local sources was significant compared to the contribution from long-range transport. Compound volatility was estimated to be an important parameter; for the more volatile FRs the local sources had two to three orders of magnitude (OM) higher contributions to air concentrations. For the FRs with relatively lower volatility the local sources had four to six OM higher contributions to soil and seawater and biota.

There was good agreement ( $\leq$  one OM difference) between modelled and measured air concentrations for TCEP and TCPP, and for one of the emission



scenarios for EH-TBB, BEH-TEBP and HBBz. For the soil compartment TCPP and EH-TBB and one of the emission scenarios for HBBz and DP had modelled concentrations with good agreement ( $\leq$  one OM difference) compared to measured soil concentrations. For seawater, modelled concentrations of TCPP, TPHP, BEH-TEBP and HBBz were comparable ( $\leq$  one OM difference) to measured concentrations for one of the emission scenarios.

Disagreements between modelled and measured environmental concentrations are not straightforward to explain. There are several uncertainties in the approach used here. These are associated with the wide range of compound- and site-specific parameters that are primarily used in DEHM. Produced amounts and location of production sites were associated with uncertainties and especially the production sites in China were largely unknown. Chinese producers were predominant for the OPFRs and HBBz. Producers in North America (EH-TBB, BEH-TEBP and DP), and Western and Eastern Europe (OPFRs), and Japan (HBBz) were known, however the amounts were still uncertain. The geographical distribution of product manufacture and use, and waste treatment is on a proxy level. The local soil and seawater model comprises simplifications, e.g. regarding leaching rates, soil characteristics and water exchange for tidal water. Importantly, compound specific EFs to air, water and soil should be defined for production, product use and waste treatment, which are currently estimated from other FRs as proxies. Finally, measured values should be more representative of the specific site and season of sampling.

A human risk from indirect FR exposure via the environment, estimated from the derived environmental concentrations and standard daily intake values of air, fish and seal, showed no or low risk. The calculated Risk Characterization Ratios had highest contributions from HBBz, EH-TBB and TCEP. Likewise, the environmental risk was low, with main contributions from BEH-TEBP and EH-TBB. However, toxicity data were missing or limited, especially for the terrestrial compartment.

A quantitative comparison of contributions from local sources vs. long-range transport to the Arctic air, soil and marine environment, and a human and environmental risk screening, is novel for these FRs. Persistent Organic Pollutants (POPs) are defined, among others, as compounds that undergo long-range transport. For the compounds of current use, e.g. present in consumer products, local sources in the Arctic might exist as well. With a view to evaluate long-range transport, it is particularly important to better understand the potential presence and extent of local emission sources in the Arctic. This study showed that local sources were indeed relevant for Arctic towns such as Nuuk. Nuuk is not representative of a typical Arctic community as it is relatively large compared to other arctic communities. However, even smaller communities may have activities and use products that can lead to emissions of FRs and other chemicals to the local environment. The method suggested in this study can also be used in these cases to assess emissions, source contributions and risks to humans and the environment.

## 9 References

Abbasi, G., Li, L. & Breivik, K., 2019. Global historical stocks and emissions of PBDEs. *Environmental Science & Technology*, 53 (11): 6330–6340.

Additives for polymers, 2017. Global flame retardant market projected to reach US\$11.96 billion by 2025. *Additives for Polymers: Volume 2017, Issue 1*, January 2017, pp. 10-11. Available at: <https://www.sciencedirect.com/science/article/pii/S0306374717300143>

AMAP, 2017. AMAP Assessment 2016: Chemical of Emerging Concern. Arctic Monitoring and Assessment Programme (AMAP). Available at: <https://www.amap.no/documents/doc/AMAP-Assessment-2016-Chemicals-of-Emerging-Arctic-Concern/1624>

Bacaloni, A., Cucci, F., Guarino, C., Nazzari, M., Samperi, R. & Lagana, A., 2008. Occurrence of organophosphorus flame retardant and plasticizers in three volcanic lakes of Central Italy. *Environ. Sci. Technol.* 42: 1898-1903.

Brandt, J., Silver, J.D., Frohn, L.M., Geels, C., Gross, A., Hansen, A.B., Hansen, K.M., Hedegaard, G.B., Skjøth, C.A., Villadsen, H., Zare, A. & Christensen, J.H., 2012. An integrated model study for Europe and North America using the Danish Eulerian Hemispheric Model with focus on intercontinental transport. *Atmospheric Environment*, 53: 156-176.

Breivik, K., Armitage, J.M., Wania, F., Sweetman, A.J. & Jones, K.C., 2016. Tracking the global distribution of persistent organic pollutants accounting for E-waste exports to developing regions. *Environmental Science & Technology*, 50 (2): 798–805.

Bruin, Y.B., Peijnenburg, W., Vermeire, T., Posthuma, L., Hogendoorn, E. & Verhoeven, J.A., 2015. Tiered approach for environmental impact assessment of chemicals and their alternatives within the context of socio-economic analyses. *J. Clean. Prod.* 108: 955–964.

Canada, 2013. *Canada Gazette*, Part 1 147 (13), 2013, 605-614, 30 March 2013.

Canada, 2016. *Canada Gazette*, Part 1 150 (41), 2016, 2915-2945, 8 October 2016.

Canada, 2019a. Screening assessment certain organic flame retardants substance grouping 1,4:7,10-Dimethanodibenzo[a,e]cyclooctene, 1,2,3,4,7,8,9,10,13,13,14,14-dodecachloro-1,4,4a,5,6,6a,7,10,10a,11,12,12a-dodecahydro- Dechlorane Plus (DP). Chemical Abstracts Service Registry Number 13560-89-9. Environment and Climate Change Canada Health Canada, May 2019. Cat. No.: En14-369/2019E-PDF. ISBN 978-0-660-30091-7.

Canada, 2019b. State of the Science Report. Certain Organic Flame Retardants Substance Grouping; Ben-zoic acid, 2,3,4,5-tetrabromo-, 2-ethylhexyl ester (TBB) and 1,2-Benzenedicarboxylic acid, 3,4,5,6-tetrabromo-bis(2-ethylhexyl) ester (TBPH). Available at:

<https://www.canada.ca/en/environment-climate-change/services/evaluating-existing-substances/state-science-report-certain-organic-flame-retardants-substance-grouping-benzoic-acid-tetrabromo-ethylhexyl-ester-benzenedicarboxylic-acid-tetrabromo-bis-ethylhexyl-ester.html>

Carlsson, H., Nilsson, U., Becker, G. & Östman, C., 1997. Organophosphate ester flame retardants and plasticizers in the indoor environment: analytical methodology and occurrence. *Environ. Sci. Technol.*, 31: 2931-2936.

Carlsson, P., Vrana, B., Sobotka, J., Borgå, K., Bohlin Nizzetto, P. & Varpe, Ø., 2018. New brominated flame retardants and dechlorane plus in the Arctic: Local sources and bioaccumulation potential in marine benthos. *Chemosphere* 211: 1193-1202.

Castro-Jimenez, J., Berrojalbiz, N., Pizarro, M. & Dachs, J., 2014. Organophosphate ester (OPE) flame retardants and plasticizers in the Open Mediterranean and Black Seas atmosphere. *Environ. Sci. Technol.* 48: 3203-3209.

CEC, 2015. Enhancing Trilateral Understanding of Flame Retardants and Their Use in Manufactured Items: Supply Chain Analysis of Select Flame Retardants Contained in Manufactured Items that are used in Indoor Environments. Montreal, Canada: Commission for Environmental Cooperation. 33 pp.

Cheng, W.H., Xie, Z.Q., Blais, J.M., Zhang, P.F., Li, M., Yang, C.Y., Huang, W., Ding, R. & Sun, L.G., 2013. Organophosphorus esters in the oceans and possible relation with ocean gyres. *Environ. Pollut.* 180: 159-164.

Christensen, J.H., 1997. The Danish eulerian hemispheric model — a three-dimensional air pollution model used for the Arctic. *Atmospheric Environment*, 31(24): 4169-4191.

Christensen, J.H., Brandt, J., Frohn, L.M. & Skov, H., 2004. *Atmos. Chem. Phys.* 4: 2251-2257.

Covaci, A., Harrad, S., Abdallah, M.A.-E., Ali, N., Law, R.J., Herzke, D. & de Wit, C., 2011. Review - Novel brominated flame retardants: A review of their analysis, environmental fate and behavior. *Environment International* 37: 532-556.

Cristale, J., Katsoyiannis, A., Sweetman, A.J., Jones, K.C. & Lacorte, S., 2013. Occurrence and risk assessment of organophosphorus and brominated flame retardants in the River Aire (UK). *Environmental Pollution* 179: 194-200.

Dam, M.; van Bavel, B.; Rigét, F.; Rotander, A.; Polder, A.; Auðunsson, G.A. & Gabrielsen, G.W., 2011. "New" POPs in marine mammals in Nordic Arctic and NE Atlantic areas during three decades. *TemaNord* 2011:564.

Di Toro, D.M., 1985. A particle interaction model of reversible organic chemical sorption. *Chemosphere* 14: 1503-1538.

DK EPA, 2014. Kortlægning, sundheds- og miljøvurdering af flammehæmmere i tekstiler. Kortlægning af kemiske stoffer i forbrugerprodukter nr. 126, Miljøstyrelsen (in Danish).

Dreyer, A.; Neugebauer, F.; Lohmann, N.; Rüdél, H.; Teubner, D.; Grotti, M.; Rauert, C. & Koschorreck, J., 2019. Recent findings of halogenated flame retardants (HFR) in the German and Polar environment. *Environ. Pollut.* 253, 850-863.

Dulio, V., van Bavel, B., Brorström-Lundén, E., Harmsen, J., Hollender, J., Schlabach, M., Slobodnik, J., Thomas, K. & Koschorreck, J., 2018. Emerging pollutants in the EU: 10 years of NORMAN in support of environmental policies and regulations. *Environ Sci Eur* (30:5).

EC, 2008. Tris (2-chloroethyl) phosphate, TCEP, CAS No: 115-96-8, EINECS No: 204-118-5. Summary risk assessment report. Final report 26.05 2008, Germany. Final approved version.

EU, 2012. Directive 2012/19/EU of the European Parliament and of the Council of 4 July 2012 on waste electrical and electronic equipment (WEEE), 2012, pp. 38-71. Official Journal of the European Union L197.

EC, 2014. European Commission, 2014. Commission Directive 2014/79/EU. Off. J. Eur. Union L182/49.

ECHA, 2007. Hexachlorocyclopentadiene. European Chemicals Agency, Helsinki, Finland.

ECHA, 2012. Guidance on information requirements and chemical safety assessment, Chapter R.8: Characterisation of dose [concentration]-response for human health.

ECHA, 2014. Annex XV Restriction Report: Bis(Pentabromophenyl) Ether. European Chemicals Agency, Helsinki, Finland.

ECHA, 2018a. Screening report. An assessment of whether the use of TCEP, TCPP and TDCP in articles should be restricted. [https://echa.europa.eu/documents/10162/13641/screening\\_report\\_tcep\\_tcpp\\_td-cp\\_en.pdf/e0960aa7-f703-499c-24ff-fba627060698](https://echa.europa.eu/documents/10162/13641/screening_report_tcep_tcpp_td-cp_en.pdf/e0960aa7-f703-499c-24ff-fba627060698).

ECHA, 2018b. Inclusion of Substances of Very High Concern in the Candidate List for Eventual Inclusion in Annex XIV, Decision of the European Chemicals Agency, 2018. ED/02/2018, Helsinki, 3 January 2018.

EFSA, 2013. Scientific opinion on emerging and novel brominated flame retardants (BFRs) in food. Panel on contaminants in the food chain, European Food Safety Authority (EFSA), Parma, Italy, EFSA J. 10, 2013, 2908.

EPA, 2016. Pre-screening of REACH registration dossiers for 9 brominated flame retardants - A LOUS follow-up project. Environmental project No. 1821.

EU, 2007. EU Risk Assessment Report, Tris(2-chloro-1-methylethyl) Phosphate. TCPP. Draft, European Commission, European Union, Brussels.

EU, 2008. European Union Risk Assessment Report, Tris(2-chloro-1-methylethyl)phosphate (TCPP). CAS-No.: 13674-84-5; EINECS-No.: 237-158-7. Risk Assessment. EU RISK ASSESSMENT (europa.eu).

EU, 2009. European Union Risk Assessment Report, Tris(2-chloroethyl)phosphate, TCEP. CAS-No.: 115-96-8; EINECS-No.: 204-118-5. Risk Assessment, final approved version July 2009. RAR (europa.eu).

EU TGD, 2003. European Commission – Technical guidance document on risk assessment – Part I and II. Available at: Part I. [https://echa.europa.eu/documents/10162/16960216/tgdpart1\\_2ed\\_en.pdf](https://echa.europa.eu/documents/10162/16960216/tgdpart1_2ed_en.pdf) Part II: [https://echa.europa.eu/documents/10162/16960216/tgdpart2\\_2ed\\_en.pdf](https://echa.europa.eu/documents/10162/16960216/tgdpart2_2ed_en.pdf)

Fisk, P.R., Girling, A.E. & Wildey, R.J., 2003. Prioritisation of flame retardants for environmental risk assessment. Environment Agency.

Gao, X.; Huang, P.; Huang, Q.; Rao, K.; Lu, Z.; Xu, Y.; Gabrielsen, G.W.; Hallanger, I.; Ma, M. & Wang, Z., 2019. Organophosphorus flame retardants and persistent, bioaccumulative and toxic contaminants in Arctic seawaters: On-board passive sampling coupled with target and non-target analysis. Environ. Pollut. 253: 1-10.

Gao, X.; Xu, Y.; Ma, M.; Huang, Q.; Gabrielsen, G.W.; Hallanger, I.; Rao, K.; Lu, Z. & Wang, Z., 2020. Distribution, sources and transport of organophosphorous flame retardants in the water and sediment of Ny-Ålesund, Svalbard, the Arctic. Environ. Pollut. 264: 114792.

Garcia-Garin, O., Sala, B., Aguilar, A., Vighi, M., Vikingsson, G.A., Chosson, V., Eljarrat, E. & Borrell, A., 2020. Organophosphate contaminants in North Atlantic fin whales, Sci. Total Environ. 721: 137768.

Gewurtz, S.B.; Guerra, P.; Kim, M.G.; Jones, F.; Urbanic, J.C.; Teslic, S. & Smyth, S.A., 2020. Wastewater treatment lagoons: Local pathways of perfluoroalkyl acids and brominated flame retardants to the Arctic environment. Environ. Sci. Technol. 54: 6053-6062.

Green, N., Schlabach, M., Bakke, T., Brevik, E.M., Dye, C., Herzke, D., Huber, S., Plosz, B., Remberger, M., Schoyen, M., Uggerud, H.T. & Vogelsang, C., 2008. Screening of selected metals and new organic contaminants 2007. Norwegian Pollution Control Agency.

Guerra, P., Alae, M., Jiménez, B., Pacepavicius, G., Marvin, C., MacInnis, G., Eljarrat, E., Barceló, D., Champoux, L. & Fernie, K., 2012. Emerging and historical brominated flame retardants in peregrine falcon (*Falco peregrinus*) eggs from Canada and Spain. Environment International 40: 179-186.

Han, X., Hao, Y., Li, Y., Yang, R., Wang, P., Zhang, G., Zhang, Q. & Jiang, G., 2020. Occurrence and distribution of organophosphate esters in the air and soils of Ny-Ålesund and London Island, Svalbard, Arctic. Environ. Pollut. 263: 114495.

Hansen, K.M., Christensen, J.H., Brandt, J., Frohn L.M. & Geels, C., 2004. "Modelling atmospheric transport of  $\alpha$ -hexachlorocyclohexane in the Northern Hemisphere with a 3-D dynamic model: DEHM-POP". Atmospheric Chemistry and Physics 4: 1125-1137.

- Hansen, K.M., Christensen, J.H., Brandt, J., Frohn, L.M., Geels, C., Skjøth, C.A. & Li, Y.-F., 2008a. *J. Geophys. Res.* 113: D02310.
- Hansen, K.M., Halsall, C.J., Christensen, J.H., Brandt, J., Geels, C., Frohn, L.M. & Skjøth, C.A., 2008b. *Environ. Sci. Technol.* 42(8): 2943-2948.
- Hao, Y., Meng, W., Li, Y., Han, X., Lu, H., Wang, P., Yang, R., Zhang, Q. & Jiang, G., 2020. Concentrations and distribution of novel brominated flame retardants in the atmosphere and soil of Ny-Ålesund and London Island, Svalbard, Arctic. *J. Environ. Sci.* 97: 180-185.
- Harju, M., Herzke, D. & Kaasa, H., 2013. Perfluorinated Alkylated Substances (PFAS), Brominated Flame Retardants (BFR) and Chlorinated Paraffins (CP) in the Norwegian Environment e Screening 2013, Norwegian Institute for Air Research (NILU), 2013. Report no. 31/2013.
- Hartmann, P.C., Bürgi, D. & Giger, W., 2004. Organophosphate flame retardants and plasticizers in indoor air. *Chemosphere*, 57: 781-787.
- Heidam, N.Z., Christensen, J., Wahlin, P. & Skov, H., 2004. Arctic atmospheric contaminants in NE Greenland: levels, variations, origins, transport, transformations and trends 1990-2001. *Science of the Total Environment* 331, 5-28.
- Ingerowski, G., Friedle, A. & Thumulla, J., 2001. Chlorinated ethyl and isopropyl phosphoric acid triesters in the indoor environment - an inter-laboratory exposure study. *Indoor Air*, 11(3):145-9.
- Kajiwara, N., Noma, Y. & Takigami, H., 2011. Brominated and organophosphate flame retardants in selected consumer products on the Japanese market in 2008. *J. Hazard. Mater.* 192: 1250-1259.
- Kim, J.-T., Choi, Y.-J., Barghi, M., Yoon, Y.-J., Kim, J.-H., Kim, J. H. & Chang, Y.-S., 2018. Occurrence and distribution of old and new halogenated flame retardants in mosses and lichens from the South Shetland Islands, Antarctica. *Environ. Pollut.* 235: 302–311.
- Krogseth, I.S., Kierkegaard, A., Breivik, K., McLachlan, M.S., Hansen K.M. & Schlabach, M., 2013. *Environmental Science and Technology* 47(1). DOI: 10.1021/es3040208.
- Kurt-Karakus, P.B., Muir, D.C., de Jourdan, B., Teixeira, C., Martindale, J.E., Embers, H., Wang, X., Keir, M. & Backus, S., 2019. Bioaccumulation of selected halogenated organic flame retardants in Lake Ontario. *Environ. Toxicol. Chem.* 38(6): 1198–1210.
- McLachlan, M.S., Kierkegaard, A., Hansen, K.M., van Egmond, R., Christensen, J.H. & Skjøth, C.A., 2010. Concentrations and fate of decamethylcyclopentasiloxane (D5) in the atmosphere. *Environmental Science & Technology* 44(14): 5365-5370. DOI: 10.1021/es100411w.
- Lai, S.C., Xie, Z.Y., Song, T.L., Tang, J.H., Zhang, Y.Y., Mi, W.Y., Peng, J.H., Zhao, Y., Zou, S.C. & Ebinghaus, R., 2015. Occurrence and dry deposition of organophosphate esters in atmospheric particles over the northern South China Sea. *Chemosphere* 127: 195–200.

Larsson, K., de Wit, C.A., Sellström, U., Sahlström, L., Lindh, C.H. & Berglund, M., 2018. Brominated Flame Retardants and Organophosphate Esters in Preschool Dust and Children's Hand Wipes. *Environ. Sci. Technol.* 52: 4878–4888.

Liagkouridis, I., Cousins, A.P. & Cousins, I., 2015. Physical–chemical properties and evaluative fatemodelling of 'emerging' and 'novel' brominated and organophosphorus flame retardants in the indoor and outdoor environment. *Sci. Total Environ.* 524–525: 416–42.

Liang, Y., Liu, X. & Allen, M.R., 2019. The influence of temperature on the emissions of organophosphate ester flame retardants from polyisocyanurate foam: measurement and modeling. *Chemosphere*, 233: 347–354.

Luo, P., Bao, L.-J., Guo, Y., Li, S.-M. & Zeng, 2016. Size-dependent atmospheric deposition and inhalation exposure of particle bound organophosphate flame retardants. *Journal of Hazardous Materials* 301: 504–511.

Maddela, N.R., Venkateswarlu, K. & Megharaj, M., 2020. Tris(2-chloroethyl) phosphate, a pervasive flame retardant: critical perspective on its emissions into the environment and human toxicity. *Environ. Sci. Processes Impacts* 22: 1809.

Marklund, A., Andersson, B. & Haglund, P., 2005. Traffic as a source of organophosphorus flame retardants and plasticizers in snow. *Environ. Sci. Technol.*, 39: 3555-3562.

McDonough, C.A.; De Silva, A.O.; Sun, C.; Cabrerizo, A.; Adelman, D.; Soltwedel, T.; Bauerfeind, E.; Muir, D.C.G. & Lohmann, R., 2018. Dissolved organophosphate esters and polybrominated diphenyl ethers in remote marine environments: Arctic surface water distributions and net transport through Fram Strait. *Environ. Sci. Technol.* 52: 6208-6216.

Möller, A.; Xie, Z.; Sturm, R. & Ebinghaus, R., 2010. Large-scale distribution of dechlorane plus in air and seawater from the Arctic to Antarctica. *Environ. Sci. Technol.* 44: 8977-8982.

Möller, A., Xie, Z.Y., Cai, M.H., Zhong, G.C., Huang, P., Cai, M.G., Sturm, R., He, J.F. & Ebinghaus, R., 2011a. Polybrominated Diphenyl ethers vs alternate brominated flame retardants and Dechloranes from East Asia to the Arctic. *Environmental Science & Technology*, 45 (16): 6793–6799.

Möller, A., Xie, Z.Y., Caba, A., Sturm, R. & Ebinghaus, R., 2011b. Organophosphorus flame retardants and plasticizers in the atmosphere of the North Sea. *Environ. Pollut.* 159: 3660-3665.

Möller, A.; Xie, Z.; Sturm, R. & Ebinghaus, R., 2011c. Polybrominated diphenyl ethers (PBDEs) and alternative brominated flame retardants in air and seawater of the European Arctic. *Environ. Pollut.* 159: 1577-1593.

Möller, A., Sturm, R., Xie, Z.Y., Cai, M.H., He, J.F. & Ebinghaus, R., 2012a. Organophosphorus flame retardants and plasticizers in airborne particles over the Northern Pacific and Indian Ocean toward the polar regions: evidence for global occurrence. *Environ. Sci. Technol.* 46: 3127-3134.

- Möller, A., Xie, Z., Cai, M., Sturm, R. & Ebinghaus, R., 2012b. Brominated flame retardants and dechlorane plus in the marine atmosphere from South-east Asia toward Antarctica. *Environ. Sci. Technol.* 46(6): 3141–3148.
- Na, G., Hou, C., Li, R., Shi, Y., Gao, H., Jin, S., Gao, Y., Jiao, J. & Cai, Y., 2020. Occurrence, distribution, air-seawater exchange and atmospheric deposition of organophosphate esters (OPEs) from the Northwestern Pacific to the Arctic Ocean. *Mar. Pollut. Bull.* 157, 111243.
- Na, G.; Wei, W.; Zhou, S.; Gao, H.; Ma, X.; Qiu, L.; Ge, L.; Bao, C. & Yao, Z., 2015. Distribution characteristics and indicator significance of dechloranes in multi-matrices at Ny-Ålesund in the Arctic. *J. Environ. Sci.* 28: 8-13.
- Ni, Y., Kumagai, K. & Yanagisawa, Y., 2007. Measuring emissions of organophosphate flame retardants using a passive flux sampler, *Atmos. Environ.*, 41: 3235–3240.
- Pantelaki, I. & Voutsas, D., 2019. Review - Organophosphate flame retardants (OPFRs): A review on analytical methods and occurrence in wastewater and aquatic environment. *Science of the Total Environment* 649: 247–263.
- Platts, J.A., Butina, D., Abraham, M.H. & Hersey, A., 1999. Estimation of molecular linear free energy relation descriptors using a group contribution approach. *J. Chem. Inf. Comput. Sci.*, 39: 835-845.
- Rauert, C., Schuster, J.K., Eng, A. & Harner, T., 2018. Global Atmospheric Concentrations of Brominated and Chlorinated Flame Retardants and Organophosphate Esters. *Environ. Sci. Technol.* 52: 2777.
- Redfern, F.M., Lee, W.J., Yan, P., Mwangi, J.K., Wang, L.C. & Shih, C.H., 2017. Overview and perspectives on emissions of Polybrominated Diphenyl ethers on a global basis: evaporative and fugitive releases from commercial PBDE mixtures and emissions from combustion sources. *Aerosol Air Qual. Res.*, 17(5): 1117–1131.
- Regnery, J. & Püttmann, W., 2009. Organophosphorus flame retardants and plasticizers in rain and snow from Middle Germany. *Clean-Soil, Air, Water* 37: 334-342.
- Regnery, J. & Püttmann, W., 2010a. Occurrence and fate of organophosphorus flame retardants and plasticizers in urban and remote surface waters in Germany. *Water Res.* 44: 4097-4104.
- Regnery, J. & Püttmann, W., 2010b. Seasonal fluctuations of organophosphate concentrations in precipitation and stormwater runoff. *Chemosphere* 78: 958-964.
- Ren, G., Yu, Z., Ma, S., Li, H., Peng, P., Sheng, G. & Fu, J., 2009. Determination of Dechlorane Plus in serum from electronics dismantling workers in South China. *Environ. Sci. Technol.* 43(24): 9453–9457.
- Sagerup, K.; Herzke, D.; Harju, M.; Evenset, A.; Christensen, G.N.; Routti, H.; Fuglei, E.; Aars, J.; Strøm, H. & Gabrielsen, G.W., 2010. New brominated flame retardants in Arctic biota. *Klima- og Forurensningsdirektoratet, Rapportnr.* 1070/2010.



- Salamova, A., Hermanson, M.H. & Hites, R.A., 2014a. Organophosphate and halogenated flame retardants in atmospheric particles from a European Arctic site. *Environ. Sci. Technol.* 48: 6133-6140.
- Salamova, A., Ma, Y., Venier, M. & Hites, R.A., 2014b. High levels of organophosphate flame retardants in the Great Lakes atmosphere. *Environ. Sci. Technol. Lett.* 1: 8-14.
- Schlabach, M.; Remberger, M.; Brorström-Lunden, E. & Herzke, D., 2011. Brominated Flame Retardants (BFR) in the Nordic Environment. *TemaNord* 2011: 528.
- Schlabach, M., van Bavel, B., Baz-Lomba, J.A., Borgen, A., Gabrielsen, G.W., Götsch, A., Halse, A.K., Hanssen, L., Krogseth, I.S. & Nikiforov, V., 2018. Screening Programme 2017–AMAP Assessment Compounds.
- Schreder, E.D., Uding, N. & La Guardia, M.J., 2016. Inhalation a significant exposure route for chlorinated organophosphate flame retardants. *Chemosphere* 150: 499-504.
- Simond, A.E., Houde, M., Lesage, V. & Verreault, J., 2017. Temporal trends of PBDEs and emerging flame retardants in belugas from the St. Lawrence Estuary (Canada) and comparisons with minke whales and Canadian Arctic belugas. *Environ. Res.*: 156, 494–504.
- Skamarock, W.C., Klemp, J.B., Dudhia, J., Gill, D.O., Barker, D.M., Duda, M.G., Huang, X.-Y., Wang, W. & Powers, J.G., 2008. A description of the Advanced Research WRF version 3, National Center for Atmospheric Research Tech. Note, NCAR/TN-475+STR, 113 pp.
- Skov, H., Christensen, J.H., Goodsite, M.E., Heidam, N.Z., Jensen, B., Wählin, P. & Geernaert, G., 2004. Fate of elemental mercury in the arctic during atmospheric mercury depletion episodes and the load of atmospheric mercury to the arctic *Environ. Sci. Technol.* 38: 2373–2382.
- Stapleton, H.M., Allen, J.G., Kelly, S.M., Konstantinov, A., Klosterhaus, S., Watkins, D., Mcclean, M.D. & Webster, T.F., 2008. Alternate and new brominated flame retardants detected in U.S. house dust. *Environ. Sci. Technol.*, 42: 6910-6916.
- Stapleton, H.M., Klosterhaus, S., Eagle, S., Fuh, J., Meeker, J.D., Blum, A. & Webster, T.F., 2009. Detection of organophosphate flame retardants in furniture foam and US house dust. *Environ. Sci. Technol.*, 43: 7490-7495.
- Strand, J., *in prep.*, 2022. Amounts, composition and trends of marine litter on Greenlandic shorelines 2015-21. Data from the SUMAG-project Systematic sUrveys of Marine Litter in Greenland. Data are also available in the OSPAR beach litter database.
- Strobel, A.; Willmore, W.G.; Sonne, C.; Dietz, R. & Letcher, R.J., 2018. Organophosphate esters in East Greenland polar bears and ringed seals: Adipose tissue concentrations and in vitro depletion and metabolite formation. *Chemosphere* 196: 240-250.

Sun, Y.; De Silva, A.O.; St Pierre, K.A.; Muir, D.C.G.; Spencer, C.; Lehnher, I. & MacInnis, J.J., 2020. Glacial melt inputs of organophosphate ester flame retardants to the largest High Arctic lake. *Environ. Sci. Technol.* 54: 2734-2743.

Sverko, E., Tomy, G.T., Reiner, E.J., Li, Y.F., McCarry, B.E., Arnot, J.A., Law, R.J. & Hites, R.A., 2011. Dechlorane Plus and related compounds in the environment: a review. *Environmental Science & Technology* 45 (12): 5088–5098.

Sühring, R.; Diamond, M.L.; Bernstein, S.; Adams, J.K.; Schuster, J.K.; Fernie, K.; Elliott, K.; Stern, G. & Jantunen, L.M., 2021. Organophosphate esters in the Canadian Arctic Ocean. *Environ. Sci. Technol.* 55: 304-312.

Sühring, R., Diamond, M.L., Scheringer, M., Wong, F., Pucko, M., Stern, G., Burt, A., Hung, H., Fellin, P., Li, H. & Jantunen, L.M., 2016. Organophosphate esters in Canadian Arctic air: occurrence, levels and trends, *Environ. Sci. Technol.* 50: 7409-7415.

Takeshi, O., Takashi, A., Yoshinori, S. & Masahiro, F., 2006. Organic air pollutants inside and outside residences in Shimizu, Japan: levels, sources and risks. *Sci. Total Environ.* 366: 485-499.

US EPA, 2009. Polybrominated Diphenyl Ethers (PBDEs), Action Plan. U.S. Environmental Protection Agency 12/30/2009.

US EPA, 2015a. TSCA Work Plan Chemical Problem Formulation and Initial Assessment, Chlorinated Phosphate Ester Cluster, Flame Retardants; EPA Document #740-R1-5001. <https://www.epa.gov/assessing-and-managing-chemicals-under-tsca/tsca-work-plan-chemical-problem-formulation-and-0>

US EPA, 2015b. TSCA Work Plan Chemical Technical Supplement - Use and Exposure of the Brominated Phthalates Cluster, Flame Retardants; EPA Document #740-Q1-4002. [https://www.epa.gov/sites/production/files/2015-09/documents/bpc\\_data\\_needs\\_assessment\\_technical\\_supplement\\_use\\_and\\_exposure\\_assessment.pdf](https://www.epa.gov/sites/production/files/2015-09/documents/bpc_data_needs_assessment_technical_supplement_use_and_exposure_assessment.pdf)

US EPA, 2017. Assessing and managing chemicals under TSCA. Fact sheet: Assessing risks from flame retardants. <http://www2.epa.gov/assessing-and-managing-chemicals-under-tsca/fact-sheet-assessing-risks-flameretardants>, 2017.

US EPA, 2019. Proposed designation of triphenyl phosphate (CASRN 115-86-6) as a high-priority substance for risk evaluation. Available at: [https://www.epa.gov/sites/production/files/2019-08/documents/triphenylphosphate\\_115-86-6\\_high-priority\\_proposeddesignation\\_082319\\_0.pdf](https://www.epa.gov/sites/production/files/2019-08/documents/triphenylphosphate_115-86-6_high-priority_proposeddesignation_082319_0.pdf)

US EPA, 2021. Estimation Programs Interface Suite™ for Microsoft® Windows, v 4.1. United States Environmental Protection Agency, Washington, DC, USA.

van der Veen, I. & de Boer, J., 2012. Review - Phosphorus flame retardants: Properties, production, environmental occurrence, toxicity and analysis. *Chemosphere* 88: 1119–1153.

Verreault, J., Gebbink, W.A., Gauthier, L.T., Gabrielsen, G.W. & Letcher, R.J., 2007. Brominated Flame Retardants in Glaucous Gulls from the Norwegian Arctic: More Than Just an Issue of Polybrominated Diphenyl Ethers. *Environ. Sci. Technol.* 41: 4925-4931.

Vorkamp, K., Bossi, R., Rigét, F.F., Skov, H., Sonne, C. & Dietz, R., 2015. Novel brominated flame retardants and dechlorane plus in Greenland air and biota. *Environmental Pollution* (196): 284-291.

Vorkamp, K., Falk, K., Møller, S., Rigét, F. & Sørensen, P.B., 2018. Regulated and unregulated flame retardants in peregrine falcon eggs from Greenland. *Environmental Science & Technology* 52, 474-483. Available at: <https://doi.org/10.1021/acs.est.7b04866>

Vorkamp, K., Balmer, J., Hung, H., Letcher, R.J., Rigét, F.F. & de Wit, C.A., 2019a. Current-use halogenated and organophosphorous flame retardants: A review of their presence in Arctic ecosystems. *Emerging Contaminants* 5: 179-200.

Vorkamp, K., Rigét, F., Sanderson, H., Bossi, R., Hansen, K.M. & Skov, H., 2019b. POP/PBT characterisation of dechlorane plus and novel brominated flame retardants based on data from Greenland. Aarhus University, DCE – Danish Centre for Environment and Energy, 80 pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 339.

Wang, D.-G., Alaei, M., Byer, J.D., Brimble, S. & Pacepavicius, G., 2013. Human health risk assessment of occupational and residential exposures to dechlorane plus in the manufacturing facility area in China and comparison with e-waste recycling site. *Science of the Total Environment* 445-446: 329-336.

Wang, Y., Yang, X., Liu, Y., Zhang, Q., Xiao, H., Wang, Y., Yao, Y. & Sun, H., 2021. A low-volume air sampling method for legacy and novel brominated flame retardants in indoor environment using a newly developed sorbent mixture. *Ecotoxicology and Environmental Safety* 210: 111837.

Watanabe, I. & Sakai, S.-I., 2003. Environmental release and behavior of brominated flame retardants. *Environment International* 29: 665 – 682.

Wei, G.-L., Li, D.-Q., Zhuo, M.-N., Liao, Y.-S., Xie, Z.-Y., Guo, T.-L., Li, J.-J., Zhang, S.-Y. & Liang, Z.-Q., 2015. Review - Organophosphorus flame retardants and plasticizers: Sources, occurrence, toxicity and human exposure. *Environmental Pollution*, 196: 29-46.

Weil, E.D. & Levchik, S.V., 2015. Flame retardants for plastics and textiles: practical applications; Carl Hanser Verlag GmbH Co KG.

Wielsøe, M., Long, M., Bossi, R., Vorkamp, K. & Bonefeld-Jørgensen, E.C., 2022 (in press) Persistent Organic Pollutant exposures among Greenlandic adults in relation to lifestyle and diet: new data from the ACCEPT cohort. *Science of the Total Environment*.

Xiao, H., Shen, L., Su, Y., Barresi, E., DeJong, M., Hung, H., Lei, Y.-D., Wania, F., Reiner, E.J., Sverko, E. & Kang, S.-C., 2012. Atmospheric concentrations of

halogenated flame retardants at two remote locations: the Canadian High Arctic and the Tibetan Plateau, *Environ. Pollut.* 161: 154-161.

Xiong, P., Yan, X., Zhu, Q., Qu, G., Shi, J., Liao, C. & Jiang, G., 2019. A Review of Environmental Occurrence, Fate, and Toxicity of Novel Brominated Flame Retardants. *Environ. Sci. Technol.* 53: 13551–13569.

Yang, R., Zhang, S., Li, A., Jiang, G. & Jing, C., 2013. Altitudinal and spatial signature of POPs in soil, lichen, conifer needles, and bark of the southeast tibetan plateau. *Environ. Sci. Technol.* 47: 12736–12743.

Yu, Y., Hung, H., Alexandrou, N., Roach, P. & Nordin, K., 2015. Multiyear measurements of flame retardants and organochlorine pesticides in air in Canada's western sub-arctic, *Environ. Sci. Technol.* 49: 8623-8630.

Zhang, X., Sührling, R., Serodio, D., Bonnell, M., Sundin, N. & Diamond, M., 2016. Novel flame retardants: Estimating the physical-chemical properties and environmental fate of 94 halogenated and organophosphate PBDE replacements. *Chemosphere* 144: 2401-2408.

## MODELLING LONG-RANGE TRANSPORT AND EMISSIONS FROM LOCAL SOURCES OF NEW CONTAMINANTS IN THE ARCTIC

Contributions from local emissions and long-range transport of seven current-use flame retardants (FRs) to the environment in Nuuk, Greenland, were quantified and compared. The contribution to the local environment from local sources was significant compared to the contribution from long-range transport. Compound volatility was an important parameter. There was good agreement between modelled and measured air concentrations for five FRs, and good agreement of soil and seawater concentrations for four FRs. A human risk from indirect FR exposure via the environment, estimated from the derived environmental concentrations and standard daily intake values of air, fish and seal, showed no or low risk. Likewise, the environmental risk was low. This study showed that local sources were relevant for Arctic towns such as Nuuk, and the suggested method can also be used in other remote locations to assess emissions, source contributions and risks to humans and the environment.