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# **Emissions and behaviour of selected persistent organic pollutants (POPs) in the northern environment**

## **Part I: Development of the cycling model and emission and environmental data bases**

**J.M. Pacyna (coordinator),  
E. Brorström Lunden, J. Paasivirta,  
E. Runge and F. Wania**

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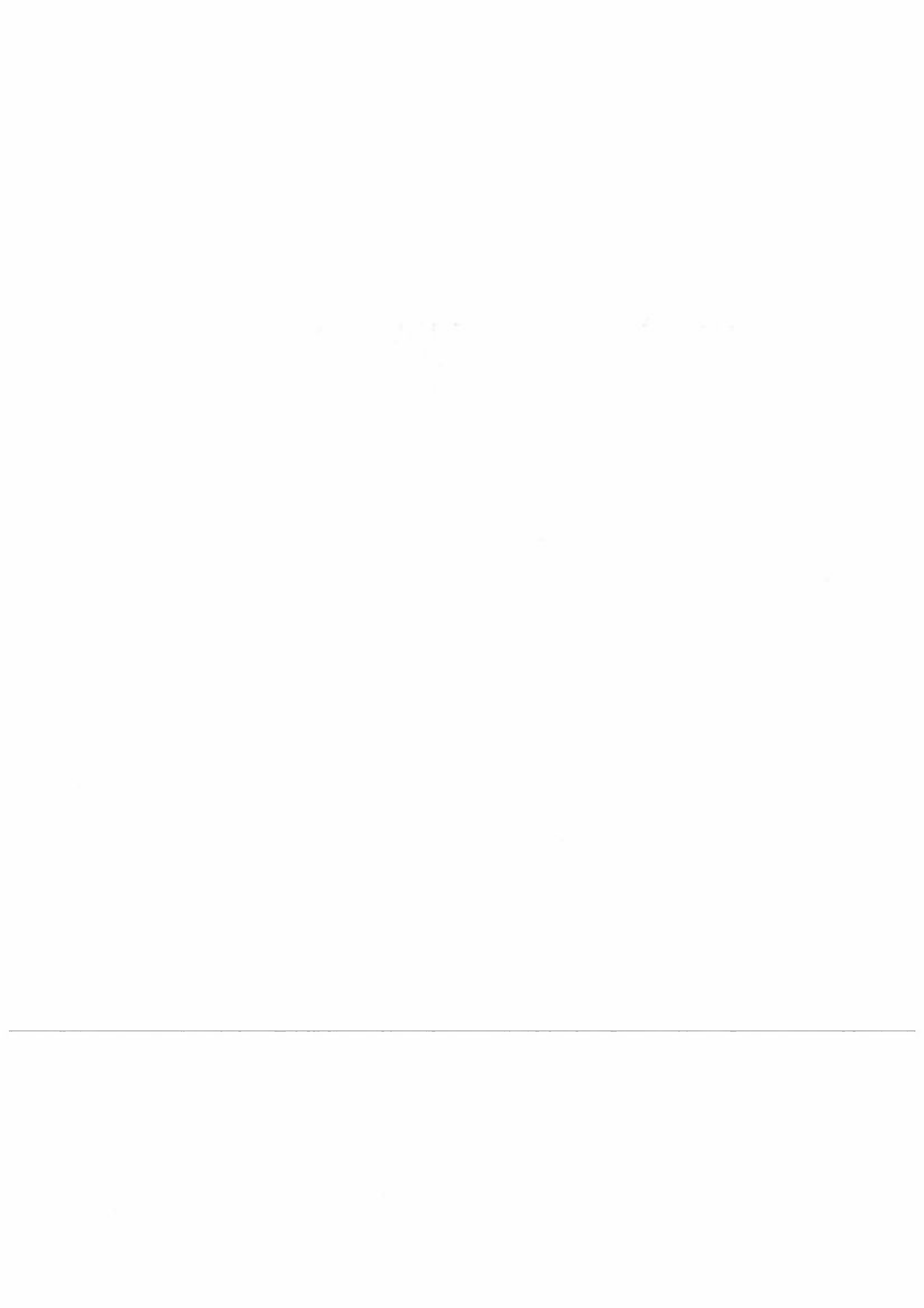
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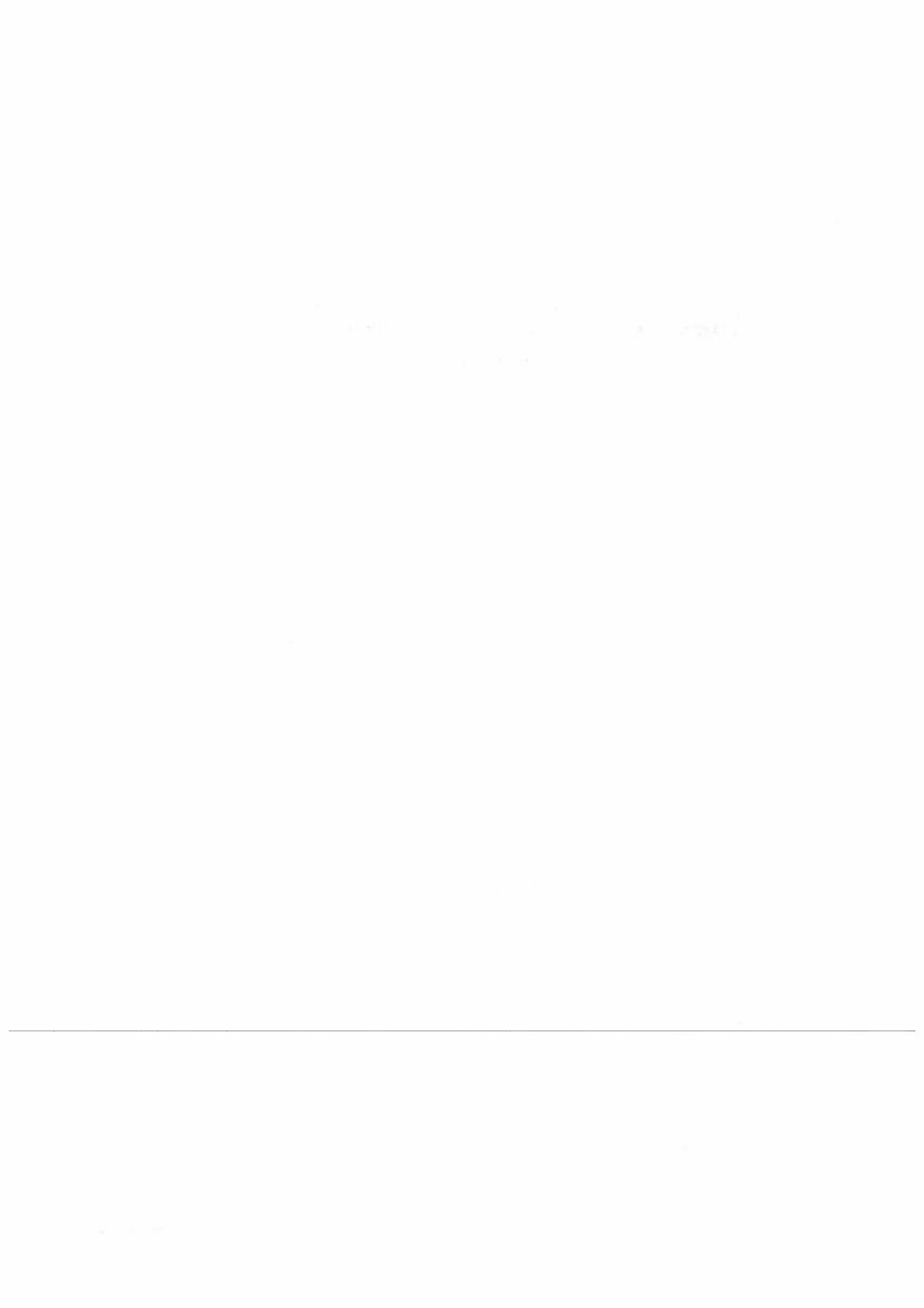
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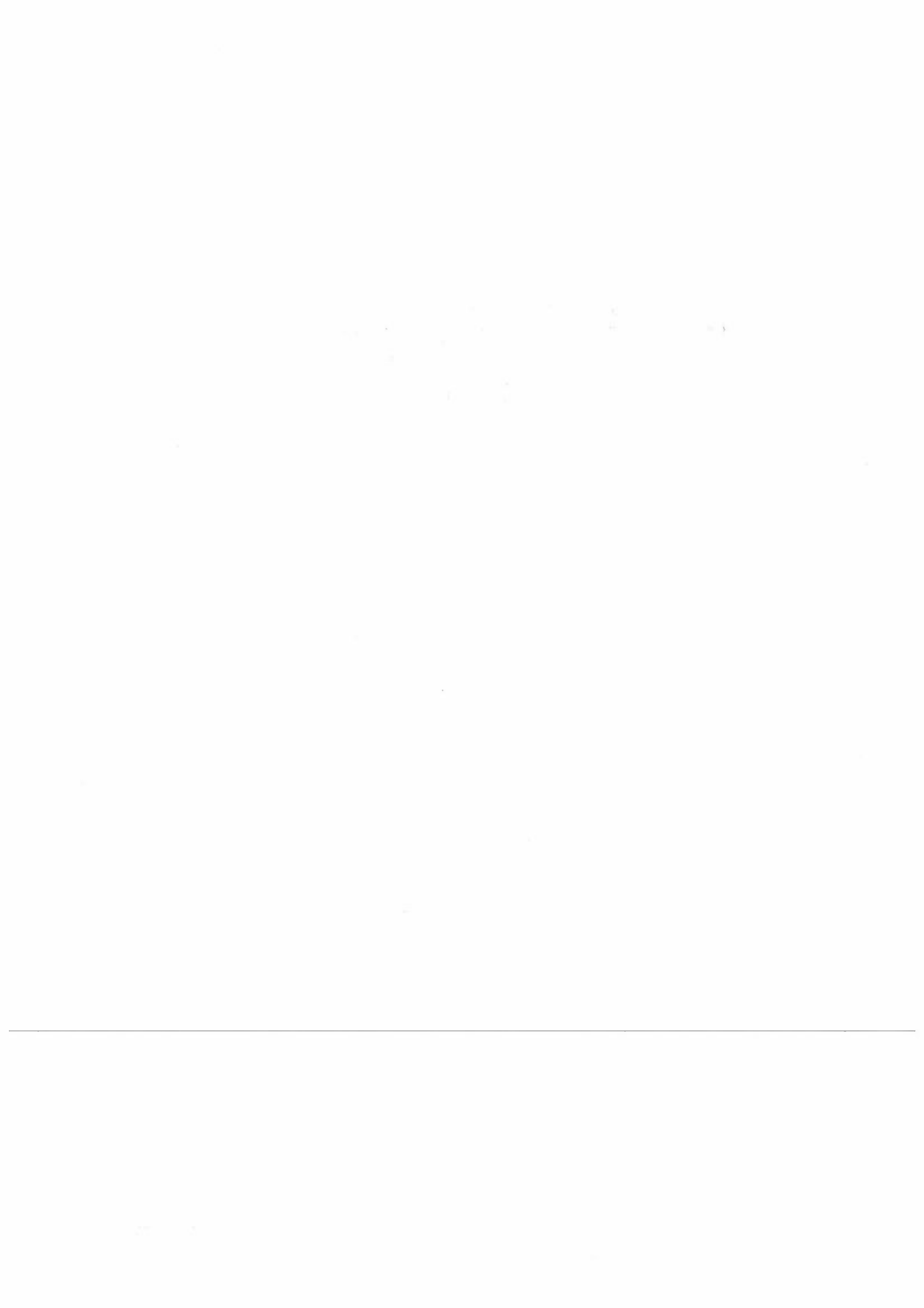
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## Summary

Recent NMR study concluded that although very incomplete, the information on emissions and concentration levels can be used in a simple multicompartiment mass balance model to quantitatively describe and possibly predict the regional fate of hexachlorobenzene, lindane, and fluoranthene in the Nordic countries. The overall goal of the reported project was to improve our knowledge on the inputs, transport, and migration of selected POPs in the Northern environments. A comprehensive steady-state mass balance model, so-called B-POP model, has been developed within the project. This model is based on the fugacity approach and comprises a number of compartments which are assumed to have homogeneous environmental conditions and chemical concentrations. Mass balance equations for all compartments were formulated and solved for the chemical fugacity in each compartment.

The B-POP model was then tested to describe the migration of PCBs. It was concluded that the model succeeds in outlining the general picture of PCB behaviour in the Baltic Sea at the end of the 1980s by identifying the major environmental pathways and reservoirs within the system. Considering the limitations inherent in the present version of the model the agreement between modelled and measured concentrations was surprisingly good. In most cases the calculated value was within the range and close to the average of the measured values.



# **Emissions and behaviour of selected persistent organic pollutants (POPs) in the northern environment**

## **Part I: Development of the cycling model and emission and environmental data bases**

### **1. Introduction**

Recently the NMR feasibility study has been carried out on the cycling of POPs in the regional environment (Pacyna et al., 1994). The main goal of this study was to review information on levels of POPs in various compartments of the environment, describe tools which can be used to simulate POP cycling on regional and global scale, and assess the environmental effects of POPs. Major conclusion of the project was that although very incomplete, the information on emissions and concentration levels can be used in a simple multicompartiment mass balance models to quantitatively describe and possibly predict the regional fate of hexachlorobenzene, lindane, and fluoranthene in the Nordic countries. The above conclusion can be regarded as a first step to better understanding of the POP cycling in the environment. It was also concluded that further improvement of our knowledge of the POP cycling is possible and a set of recommendations was defined in this connection. These recommendations called for obtaining more accurate and complete information on environmental inputs of POPs, broader and more complete basis of environmental data of the POP concentration levels in various compartments, and further improvement of models describing the migration of POPs through the environment.

### **2. The objectives of the project and its structure**

The overall goal of the reported project was to improve our knowledge on the inputs, transport, and migration of selected POPs in the Northern environments. The outcome of the recent feasibility study was a starting point in the project. In order to meet the above goal, the following tasks were carried out:

- further completion of the environmental data base established during the recent feasibility study with respect to the compounds and environmental compartments under study,
- collection of information and estimation of inputs of POPs to the individual compartments in the region of the study,
- ~~development of a non steady-state model which would simulate the environmental cycling of POPs, and~~
- elaboration of concentration trends for POPs in the studied region.

The following POPs have been selected:



- total PCBs, including the following congeners: CB-28, CB-52, CB-101, CB-105, CB-118, CB-138, CB-153, and CB-180,
- selected PAHs including anthracene, benzo(b)fluoranthene, benzo(a)pyrene, fluoranthene, phenanthrene, pyrene,
- total dioxins and furans including the following isomers: 2378-TCDD, 12378-PCDD, 123478-HxCDD, 123678-HxCDD, 123789-HxCDD, 1234678-HpCDD, 12346789-OCDD, 2378-TCDF, 12378-PCDF, 23478-PCDF, 123478-HxCDF, 123678-HxCDF, 234678-HxCDF, 123789-HxCDF, 1234789-HpCDF, 1234678-HpCDF, and 12346789-OCDF, and
- selected pesticides with focus on: HCHs, DDTs, DDE, chlordane, toxaphene, and HCB.

The study region covers the Nordic countries and surrounding marine environments, including the Baltic Sea, Kattegat, and Skagerrak. The Baltic Sea is a particularly interesting region for the following reasons:

- the Baltic Sea extends over a substantial latitudinal range and should, therefore, reveal differences of chemical behaviour between temperate and boreal environments,
- the region is heavily polluted by various POPs,
- the Baltic Sea is a relatively well studied marine region and some necessary input parameters for the model may be relatively easy to obtain, and
- there have been numerous measurements of the POP concentrations in various compartments of the Baltic Sea environment and these data may form a good data set for model validation.

The study has been divided into two parts. In the first part, reported here, the cycling model was developed and tested for PCBs.

In the second part the model will be applied for all studied components, and a set of recommendations concerning exposure to POPs in the Baltic Sea region will be presented.

Three working groups have been organized within the project concentrating on:

- modeling (Dr. Frank Wania),
- emissions (Dr. Erik Runge and Dr. Jozef M. Pacyna), and
- environmental data base (Dr. Eva Brorström-Lunden).

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The emission working group has collected information on releases of the studied compounds to various compartments of the environment.

It was intended to enlarge information on releases of POPs, collected during the previous NMR project (Pacyna et al., 1994). Major focus was on releases in the Baltic Sea region. Information on the Danish and Norwegian emissions is reported here.

The major task of the modeling group was to develop a steady-state model to study the cycling of the above mentioned POPs in the studied region. It was clear from the previous feasibility study that such model is necessary in order to describe the chemical behaviour of POPs, particularly in the region where emissions of POPs have been phased out and therefore concentrations in the environment are expected to decline. A first attempt to develop such model has been made very recently. Wania and Mackay used a two-region model to study the fate of a few POPs in Southern Norway and they found a better fit of the measured concentrations for the time-dependent than for the steady-state version of the model. An important message from this work was that the development and application of the non steady-state cycling model for POPs in the Northern environments is possible.

The third working group was aiming at the preparation of data base for validation of the model results. An extended review of information on the concentration levels of POPs measured in various environmental compartments in the Nordic countries has been prepared within the previous feasibility study. An update and completion of this information has been carried out within the reported project.

### 3. Development of the model

A comprehensive steady-state mass balance model, so-called B-POP model, has been developed within the project.

#### 3.1 Model structure

The concept and formulation of the B-POP model builds upon a previously described model of the long-term behaviour of PCBs in Lake Ontario (Mackay, 1989), which in itself is a modified version of the generic lake model QWASI (Mackay et al., 1983). The description of the model in this report will be kept brief and mostly restricted to those features which deviate from these previously described models. Some familiarity with the basics of fugacity modeling and the respective terminology (Mackay, 1991) may be required.

Very briefly, mass balance models based on the fugacity approach comprise a number of compartments which are assumed to have homogeneous environmental conditions and chemical concentrations. Expressions are defined for the capacity of each compartment to hold a chemical ( $Z$ -values in units of  $\text{mol}/(\text{m}^3 \cdot \text{Pa})$ ) and expressions describing transformation and intercompartmental transport processes ( $D$ -values in units of  $\text{mol}/(\text{h} \cdot \text{Pa})$ ). Mass balance equations for all compartments are formulated and solved for the chemical fugacity ( $f$  in units of  $\text{Pa}$ ) in each compartment. Concentrations (in  $\text{mol}/\text{m}^3$ ) are then calculated as products of  $Z$  and  $f$ , transport fluxes and transformation rates (in  $\text{mol}/\text{h}$ ) as products of  $D$  and  $f$ .

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Adopting a previously presented approach for modelling the fate of POPs in a large water body which accounts for the heterogeneity of the environment (Diamond et al., 1994), several sub-basins with distinct characteristics are identified within the Baltic Sea environment, described with a specific set of environmental parameters and interconnected through the formulation of water transport terms. These sub-basins are 1 Bothnian Bay, 2 Bothnian Sea, 3 Gulf of Finland,

4 Gulf of Riga, 5 Baltic Proper, 6 Danish Straits, and 7 Kattegat (Helsinki Commission, 1986). The Kattegat is in exchange with the North Sea, which is not a model compartment, but has been assigned the index 8. Due to its larger depth and pronounced vertical stratification the Baltic Proper sub-basin is additionally divided into a surface (index 5) and a bottom section (index 9). Each of these eight basins is represented in the B-POP model by a water and a surface sediment compartment, which are considered well-mixed and homogeneous in terms of environmental properties and chemical contamination. The water compartments of these sub-basins are connected through advective mixing processes as shown in Figure 1.

It is assumed that in both water and sediment the POPs are either present in truly dissolved form or attached to particulate organic carbon (POC). There are indications that a considerable fraction of POPs in the water column and in sediment pore water is associated with dissolved and colloidal organic carbon DOC. A DOC-bound PCB fraction is also occurring in the Baltic Sea. At the present stage it was considered premature to include partitioning to DOC in the model because crucial information is still missing. Partition coefficients  $K_{\text{DOC}}$  between water and DOC are very uncertain and seem to vary considerably depending on the nature of the organic material. Thus  $K_{\text{DOC}}$  values which are specific for the conditions in the Baltic Sea are required. There is currently also little information available on the concentrations and the dynamics of DOC in the Baltic Sea.

To describe an organic contaminant's behaviour in the aquatic system of the Baltic Sea with the present model it is therefore necessary to establish three mass balances for each sub-basin: I. for water, II. for POC, and III. for the contaminant. These mass balances are interdependent, i.e. the transport of POC will depend on the transport of water, and the contaminant's fate in turn depends on both POC and water movements. During steady state all the inputs of water, organic carbon and chemical to a sub-basin have to match all the outflows and losses.

### 3.2 Mass balance for water

A detailed study of the water balance of the Baltic Sea established for all seven sub-basins the mean water depth, water surface area, average precipitation input, evaporative loss and river water inflow as well as the freshwater exchange rates between them (Helsinki Commission, 1986). The total water flows between sub-basins can be derived from the fresh water flows using average salinity data in the various sub-basins or, if available, from measured values (e.g. the water exchange through the Danish Straits, Helsinki Commission 1986, Chapter 8). The depth of the vertical sub-division in the Baltic Proper is 36 m and the average vertical water exchange is estimated to be 1000 km<sup>3</sup>/a (Hinrichsen, Stockholm University, pers. comm.). Figure 1 summarises all steady-state water balance parameters including the average water residence times which can be estimated from these flow rates.

### 3.3 Mass balance for Particulate Organic Carbon (POC)

Previous fugacity lake models (Mackay et al., 1983; Mackay, 1989) generally included the sorption or partitioning of organic contaminants to solids which are

suspended in the water column, as well as contaminant transport in connection with the deposition and resuspension of such particles. For hydrophobic organic chemicals the association with the organic matter fraction of the suspended solids is dominant, while sorption to the mineral component tends to be negligible. To simplify model structure and to reduce the number of required input parameters, the B-POP model therefore describes particle-association and particle-mediated transport on the basis of particulate organic carbon (POC) instead of suspended solids.

The rates of mineralisation, sedimentation, resuspension and burial of POC are of eminent importance for the fate of POPs in the Baltic Sea. To assure the selection of a consistent set of parameters describing these processes, the B-POP model derives a complete POC mass balance for each sub-basin (Figure 2). For sub-basins without vertical sub-division the calculation of the POC balance is described in detail in Table 1, giving input parameters and equations. Because of the exchange of POC between surface and bottom water, the calculation for the Baltic Proper is more complex and summarised separately in Table 2. The selected input values are listed in Table 3.

Figure 3 shows the resulting steady-state mass balance for POC in the Baltic Sea as a whole. The POC balance reveals that autochthonous production by primary producers ( $110 \text{ g C}/(\text{m}^2\text{a})$ ) is by far the most important source of POC in the Baltic Sea, riverine inflow accounting only for a small fraction ( $2 \text{ g C}/(\text{m}^2\text{a})$ ). Most of the POC ( $100 \text{ g C}/(\text{m}^2\text{a})$ ) is mineralised before it settles to the surface sediment, where further decomposition takes place ( $8 \text{ g}/(\text{m}^2\text{a})$ ). Only a minor fraction ( $4 \text{ g}/(\text{m}^2\text{a})$ ) is buried in deeper sediment layers. There is a fairly intense recycling of POC occurring across the sediment-water interface. Figure 4 shows the respective mass balance for the Baltic Proper sub-basin. The shallow coastal areas experience strong resuspension and only minor burial, while the deep water bottoms have less intense resuspension and a fairly high burial rate. This general pattern of the dynamics of POC is in agreement with investigations in the Baltic Sea (Jonsson and Carman, 1994) and in other large aquatic systems such as Lake Superior (Baker et al., 1991).

### 3.4 Mass balance for POPs

#### 3.4.1 Phase Capacities Z

In the atmosphere the POPs occur in the vapour phase and attached to aerosol particles. In aqueous phases, i.e. in sea water, river water and surface sediment the POPs are present either in truly dissolved form or attached to POC. Between these sub-phases instant equilibrium partitioning is assumed to occur. Table 4 summarises the calculation of capacity values Z for each sub-compartment and bulk phase.

#### 3.4.2 Process Terms D

In the atmosphere the aerosol-attached fraction of the contaminant is subject to dry particle deposition and wet particle scavenging. POP vapours can diffuse into precipitation and directly into the sea water, whereby rain water is assumed to be in chemical equilibrium with the vapour phase.

In sea water the truly dissolved fraction can volatilise and diffuse into the surface sediment, while the POC-associated fraction is subject to particle settling and resuspension. Both fractions are subject to degradation processes and advective transport of the water phase. The truly dissolved POPs in sediment porewater can diffuse in the overlying water column, while the POC-attached fraction can be resuspended or buried to deeper sediment layers. Again, both fractions are subject to degradation. Table 5 summarises the derivation of rate parameters D for these processes.

### 3.4.3 Mass Balance Equations

Employing these Z and D-values two mass balance equations can be set up for each basin i (the index j refers to all basins adjacent to basin i), for the water and the surface sediment. In its current form the B-POP model is assuming a steady-state situation, i.e. the sum of all inputs of contaminant to a compartment matches the sum of the outputs.

The generic mass balances for the water and sediment compartment of a sub-basin i read

$$\begin{aligned} EW_i + DX_i \cdot FX_i + (DM_i + DQ_i + DV_i) \cdot FA_i + (DR_i + DT_i) \cdot FS_i + \sum_j (D_{ji} \cdot FW_j) = \\ = (DW_i + DV_i + DT_i + DD_i + \sum_j D_{ij}) \cdot FW_i \end{aligned}$$

$$FW_i \cdot (DT_i + DD_i) = FS_i \cdot (DR_i + DT_i + DS_i + DB_i)$$

The solution is greatly facilitated by deriving a D-value describing the net flow from the water column to the sediment. This makes it possible to eliminate the sediment fugacity FS from the water mass balance.

$$D_{sed_i} = \frac{(DT_i + DD_i) \cdot (DB_i + DS_i)}{DB_i + DS_i + DR_i + DT_i}$$

By summarising all the inputs with the exception of the inflow from neighbouring compartments in a parameter I

$$I_i = EW_i + DX_i \cdot FX_i + (DM_i + DQ_i + DV_i) \cdot FA_i \quad (\text{special case: } I_0 = 0)$$

and all D-values describing loss from the water column in a total D-value D<sub>tot</sub>

$$D_{tot_i} = D_{sed_i} + DW_i + DV_i + \sum_j D_{ij}$$

the generic mass balance for the water compartment of sub-basin  $i$  becomes

$$I_i + \sum_j (D_{ji} \cdot FW_j) = D_{toti} \cdot FW_i$$

Based on the connection of the sub-basins (Figure 1) the following set of mass balances for the water compartments can be formulated:

$$\begin{aligned} \text{Bay of Bothnia} \quad I_1 &= +D_{tot1} \cdot FW_1 - D_{21} \cdot FW_2 \\ \text{Bothnian Bay} \quad I_2 &= -D_{12} \cdot FW_1 + D_{tot2} \cdot FW_2 - D_{52} \cdot FW_5 \\ \text{Gulf of Finland} \quad I_3 &= +D_{tot3} \cdot FW_3 - D_{53} \cdot FW_5 \\ \text{Gulf of Riga} \quad I_4 &= +D_{tot4} \cdot FW_4 - D_{54} \cdot FW_5 \\ \text{Baltic Proper (S)} \quad I_5 &= -D_{25} \cdot FW_2 - D_{35} \cdot FW_3 - D_{45} \cdot FW_4 + D_{tot5} \cdot FW_5 - D_{65} \cdot FW_6 - D_{95} \cdot FW_9 \\ \text{Danish Straits} \quad I_6 &= -D_{56} \cdot FW_5 + D_{tot6} \cdot FW_6 - D_{76} \cdot FW_7 \\ \text{Kattegat} \quad I_7 &= -D_{67} \cdot FW_6 + D_{tot7} \cdot FW_7 - D_{87} \cdot FW_8 \\ \text{Baltic Proper (B)} \quad 0 &= -D_{59} \cdot FW_5 + D_{tot9} \cdot FW_9 \end{aligned}$$

If  $EW_i$ ,  $FX_i$ ,  $FA_i$ ,  $FW_8$  are input parameters, the  $I$  and  $D$  terms can be calculated and this system of eight linear equation can be solved numerically for the eight unknown water fugacities  $FW_i$ . It is then possible to calculate the sediment fugacities  $FS_i$

$$FS_i = FW_i \cdot \frac{DT_i + DD_i}{DB_i + DS_i + DR_i + DT_i}$$

Once the fugacities in water and sediment are known, concentrations  $C$  in  $\text{mol/m}^3$ , amounts  $M$  in  $\text{mol}$  and rates  $N$  of transport and transformation in  $\text{mol/h}$  are easily calculated employing the  $Z$  and  $D$ -values.

$$C = F \cdot Z \quad M = F \cdot Z \cdot V \quad N = F \cdot D$$

#### 4. Test of the model

The data on the occurrence and sources of POPs, which were generated during the Baltic Sea Project, are uniquely suitable input and evaluation parameters for the B-POP model. Because the current model is steady state, i.e. does not allow for changes to occur in time, a simulation has to focus on a specific time period where temporal changes in a contaminant's behaviour were minor. It was attempted to use the model to describe the fate of PCBs within the Baltic Sea environment during the late 1980s. Throughout the sixties and seventies the usage and thus the release of PCBs in Northern Europe underwent large changes, while in the eighties a slower development seemed to be established. Long term trend studies on PCB-levels in biota from the Baltic (Bignert et al., 1995) confirm relatively stable or only slightly decreasing concentrations of PCBs towards the late 1980s, when most of the measurements of the Baltic Sea Project were performed.

## 4.1 Selection of input parameters

### 4.1.1 Chemical properties

Although ideally the environmental behaviour of various PCB congeners should be modelled separately, this first and still preliminary effort uses concentrations for input and evaluation which refer to the sum of the six most commonly reported PCB isomers 52, 101, 118, 138, 153, 180, thus including one tetra-, two penta-, two hexa- and one heptachlorobiphenyl. The number of chlorine atoms in the PCB molecule strongly affects vapour pressures, aqueous solubilities and octanol-water partition coefficient. According to Falconer and Bidleman (1994) the vapour pressure of the sub-cooled liquid of PCB-52 is more than two orders of magnitude higher than of PCB-180. Similarly the  $K_{OW}$  of these compounds covers a range of one order of magnitude (Mackay et al., 1992). The use of a single set of physical-chemical properties is thus a major simplification. Table 6 lists the selected chemical property data, which reflect the typical properties of a hexachlorobiphenyl. Since the average temperatures in the Baltic Sea environment are considerably lower than room temperature, a temperature correction for vapour pressure and solubility is employed.

A tricky task is the selection of realistic degradation half-lives for POPs in the environment. Degradation rates established in the laboratory are not easily transferred to real environmental conditions. Two recent investigations on concentration profiles in sediment cores (Kjeller et al., 1995; Axelman et al., 1995) suggest that degradation of PCBs in Baltic sediments is very slow or not occurring to any significant extent at all. Therefore, an artificially long degradation half-life in surface sediment was used.

### 4.1.2 Source and emission information

The model requires three types of source and emission information: average bulk air concentrations  $CA_i$  (vapour and aerosol-bound phase), average bulk river water concentrations  $CX_i$  (truly dissolved and POC-bound phase) and an estimate of the direct discharge rate into water  $EW_i$ , which summarises input from industrial and urban sources and sewage treatment plants. The annually averaged air concentrations listed in Table 7 were estimated from recent measurements in numerous locations around the Baltic Sea (Brorström-Lundén et al., 1994). Measurements in various rivers draining into the Baltic Sea showed very variable PCB concentrations making a selection of an annually averaged value for all rivers draining into one sub-basin difficult. A typical value of 400 pg/L was used for the entire Baltic Sea with the exception of the two northernmost basins. Information on direct releases of PCBs into the Baltic Sea is, to our knowledge, non-existent. The values listed in Table 7 are very rough estimates based on comparable loading assessments performed in the Great Lakes region of North America (Thompson, 1992) and on basic information on population density and intensity of industrial development. Based on measurements by Schulz-Bull et al. (1995) the concentration of the water inflowing from the North Sea was assumed to be 60 pg/L.



### 4.1.3 Environmental parameters

Most environmental parameters which are required for the model calculations are related to the mass balances of water and POC and have been presented in Figure 1 and Table 3. Additional environmental input parameters – mostly mass transfer coefficients – are assumed to be the same throughout the Baltic Sea area and are listed in Table 8. These values were adopted from the Lake Ontario model (Mackay, 1989) assuming that the conditions in the Baltic Sea are similar to those from the Great Lakes region.

It should be mentioned that it is in many cases problematic and difficult to select a single input value to reflect annual and sub-basin-wide average conditions for a parameter which in reality may be vary variable both in time and space. Especially the parameters listed in Table 3 have to be considered as rather uncertain and preliminary estimates.

## 4.2 Presentation of test results

The results of the steady state calculations are shown in Figures 5 and 6. The mass balance of PCBs (sum of 6 isomers) in the entire Baltic Sea (Figure 5), which disregards regional differences by giving sums of the fluxes in the various sub-basins, suggests that there is a total annual input to the Baltic Sea of 880 kg PCB, of which 47 % is atmospheric deposition, 18 % riverine inflow, 21 % direct discharges, and 14 % inflow from the North Sea. It should be noted that part of the river-transported PCBs may have been originally atmospheric in origin, was deposited in the catchment area of the Baltic Sea and eventually leached to the marine environment. The annual input is balanced by an equally high total output of 880 kg. The contributions from the various loss processes are 34 % volatilisation, 2 % degradation in the water column, 46 % sediment burial, and 18 % outflow to the North Sea.

A closer inspection shows a net annual air-water exchange of 117 kg into the water, i.e. the Baltic Sea is a sink for atmospheric PCBs, which does not exclude the possibility that PCB volatilisation exceeds deposition during parts of the year. Major atmospheric deposition processes are wet and dry deposition of aerosol-associated PCBs. The net flux of 402 kg/a between water and surface sediment is also downward and major processes are the deposition and resuspension with POC. The overall pattern of PCB transport in the Baltic Sea environment is thus a downward flux from the atmosphere to the water to the surface sediment and finally to the deep sediment. This implies that the Baltic Sea acts as a sink for PCBs. This is in contrast with recent investigations in the Great Lakes of North America which suggest that volatilisation exceeds atmospheric deposition in these lakes and Lake Ontario (Mackay et al., 1994), Lake Michigan (Hornbuckle et al., 1995) and Lake Superior (Jeremiasen et al., 1995) now act as sources of PCBs to the regional atmosphere.

Figure 6 shows the calculated interbasin transport rates, input and loss terms and concentrations for each of the sub-basins. Not all sub-basins show a net atmospheric input of PCBs. The model calculations suggest that the Gulf of Finland acts as a source of PCBs to the atmosphere and that in the Bothnian Bay and the Gulf of Riga atmospheric deposition and volatilisation are almost equal.



Like the net water flux the net flux of PCBs between the sub-basins is almost always directed from the upstream basins towards the North Sea. The exception is that between Baltic Proper and Danish Straits virtually no net exchange of PCBs occurs. "Vertical" processes, i.e. those occurring within a basin tend to be more important than "horizontal", i.e. interbasin transport processes. In the Baltic Proper, for example, the calculated gross flux of PCBs to the bottom water is five times as high as the outflow to all other basins together – a result of the very efficient scavenging of PCBs by settling POC. The highly dynamic marine system in between Denmark and Southern Sweden (Basins 6 and 7) is an exception, in that horizontal exchange processes become dominant.

Riverine and atmospheric sources are of the same order of magnitude in most sub-basins, but a detailed analysis reveals that the sub-basins differ considerably in their source characteristics. Table 9 summarises the relative importance of various input and output processes in the sub-basins. In the large basin of the Bothnian Sea and Baltic Proper, PCB input is dominated by atmospheric deposition, while in the smaller and more densely populated Gulfs of Finland and Riga direct emissions and riverine inflow become far more important. Expectedly, the input of PCBs to the Danish Straits and the Kattegat is determined to a large extent by the inflow of water from the Baltic Proper and the North Sea. The Bothnian Bay finally has large contributions from both rivers and atmosphere. Differences are less pronounced with respect to PCB loss, as volatilisation and sediment burial are the major loss processes for all basins except the Danish Straits and the Kattegat, where interbasin flow is by far the most important loss pathway. In the deep Baltic Proper sediment burial become relatively more important.

Figure 6 also displays regional PCB concentration differences in water and sediment. The model suggests that the Gulf of Finland, due to high riverine inflow and relatively high population/industry density (St. Petersburg, Helsinki, Tallinn, Kohtla-Järve/Narva) should show elevated concentrations in both water and sediment. To a smaller degree this should also apply to the Gulf of Riga. The model also suggest fairly high concentrations in the bottom water and deep sediments of the Baltic Proper. For the remaining basins the model calculates rather uniform concentrations between 37 to 62 pg/L in sea water and 40 to 70 ng/g C in surface sediment.

#### **4.2.1 Response times**

Ratios of volume times bulk Z-value over sum of all D-values leaving a system are characteristic constants indicating the response time of this system to loading changes. A detailed discussion of derivation and interpretation of these time constants is given by Mackay (1989). Table 10 lists the time constants for the water column and the entire aquatic system for each of the sub-basins.

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The response time of the water column is approximately half a year indicating that a reduction of the PCB loadings, for example by reducing atmospheric or river concentrations, would be reflected by decreasing water column concentrations within a fairly short time period. This is because the residence time of PCBs in the water column is not determined by the hydraulic water residence time which is in the range of several years, see Figure 1), but by POC sedimentation.

(The smaller basins have smaller time constants because in these water advection is a more important loss process.) The response time of the entire aquatic system is much longer, in the range of 15 years. This indicates that volatilisation, sediment burial, degradation and outflow to the North Sea are too slow to effectively reduce PCB levels in the surface sediments of the Baltic Sea within a few years. This implies that loading reductions can only be expected to reduce water concentrations to a certain level which will be maintained for many years by resuspension from surface sediments. Not surprisingly, the deep sections of the Baltic Sea are particularly slow to respond. It can be hypothesised that the sediment concentrations in the Baltic Sea are likely to experience a long time lag of several years in responding to loading changes similar to what has been observed in Lake Ontario (Gobas et al., 1995). The sediment core from the Baltic Proper analysed by Kjeller and Rappe (1995) showed highest concentrations in the slice corresponding to the years 1974-1981, and thus well after the maximum PCB loadings in the Baltic had occurred.

### **4.3 Verification of test results**

One of the most interesting questions of course is, how the calculated concentrations and fluxes and their spatial patterns compare with measured concentration levels and transport rates?

#### **4.3.1 Water concentrations**

Two investigations of PCBs in the sea water of the Baltic Sea during the late 1980 (Schulz-Bull et al., 1994) revealed very heterogeneous concentrations in both space and time. Schulz-Bull et al. (1994) reported concentrations (six PCB isomers, dissolved phase and fraction adsorbed to suspended particles) between 8.5 pg/L and 1227 pg/L, with most concentration in the lower end of this range (10 to 100 pg/L). The calculated water concentrations (see Figure 6) are thus within the range of measured values. In neither study was it possible to discern clear geographical or seasonal patterns, which would allow the evaluation of the calculated regional differences.

#### **4.3.2 Sediment concentrations**

PCB concentrations in surface sediments of the Baltic Sea (Østfeldt et al., 1994) are less variable than the water concentrations. Østfeldt et al. (1994) reported values (six PCB isomers) between 67 and 238 ng/g LOI, while Broman et al. measured between 50 and 600 ng/g C. The calculated concentrations (Figure 6) are within this range of measured values, but may be a little bit too low. With respect to regional differences, the measured data indicate slightly decreasing concentrations towards the North, but especially within the Baltic Proper the measurements show large variations within rather small areas. Levels are less variable in the Northern basins and the measurements suggest higher concentrations in the Gulf of Finland compared to the Gulf of Bothnia, though not as clear a difference as predicted in the model.

#### **4.3.3 Concentrations in precipitation**

PCB concentrations in precipitation deduced from the model calculations are between 0.6 ng/l in the Bothnian Bay to 1.6 ng/l in the Danish Straits region.

Calculated PCB deposition fluxes (i.e. sum of wet deposition and dry particle deposition) are between 1.3 and 4.4 ng/(m<sup>2</sup>d). These values are lower than in measured precipitation samples by Järnmark et al. (personal communication), which for the entire Baltic Sea region show a median rain concentration of 0.46 ng/L and a median depositional fluxes of 0.54 ng/(m<sup>2</sup>d). Perhaps the measurements underestimated depositional fluxes by not wiping particles off the surface of the sampling area. Deposition measurements in the Kattegat by Brorström-Lundén (1995) which included particles sticking to the sampler surface yielded median values between 1.6 and 2.6 ng/(m<sup>2</sup>d), which compare better with a calculated depositional flux in this area of 3.1 ng/(m<sup>2</sup>d).

#### 4.4 Conclusions from the test results

It is believed that the B-POP model succeeds in outlining the general picture of PCB behaviour in the Baltic Sea at the end of the 1980s by identifying the major environmental pathways and reservoirs within the system. When judging the accuracy and precision of the simulation it is important to bear in mind the simplifications and assumptions made by the model. The most obvious are:

- the use of a single set of physical-chemical properties to describe a mixture of compounds with fairly variable volatility and hydrophobicity,
- the assumption of complete and instantaneous mixing of large and heterogeneous sub-basins,
- the assumption of steady state, i.e. stable conditions in time, including the omission of any seasonal variability of environmental parameters and concentrations,
- the uncertainty of many of the selected environmental input parameters, which in some cases are generic or estimated values.

Considering the limitations inherent in the present model the agreement between modelled and measured concentrations is surprisingly good. In most cases the calculated value is within the range and close to the average of the measured values. This agreement lends support to those model results not directly validated by comparison with measurements such as the response times.

## 5. Collection of emission data

Collection of emission data for the Baltic countries has been carried out within the project. This task has been completed for Denmark and Norway (SFT, 1993) and data for these countries are reported here.

### 5.1 PAHs

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The total PAH emissions in Norway in 1992 were as follows:

- Industrial sources: 58 t to the air and 20 t to the water
- Energy production: 63 t to the air, with 60 t from wood combustion and the rest from oil combustion
- Mobile sources: 12 t to the air
- Residential sources: 1 t to the air.

In Denmark, the emissions of fluoranthene in the 1990's were as follows:

- 26 t/y to the air
- 3.5 t/y to the water.

## 5.2 PCBs

The following statistics are available in Norway for 1992:

- Used in the country: 61 t
- Stored in the country: 1300 t
- Deposited as special wastes or exported: 384 t
- Unspecified use/storage: 265 t

## 5.3 Dioxins and furans

Major sources of dioxins and furans include:

- impurities in chemical compounds,
- accidental fires of certain compounds,
- some industrial processes,
- incineration of waste,
- incineration of (impregnated) wood and oil,
- combustion of gasoline with chlorinated additives.

In a Danish study, the measurements were carried out in 1989 at several different municipal and hospital incinerators. As a conclusion the average emission factors for the sum of dioxins and furans, were:

- 1.3 mg t<sup>-1</sup> waste burned in municipal incinerators, and
- 53 mg t<sup>-1</sup> medical waste burned in hospital incinerators (Manscher, 1990).

A reason for much higher emission factor for hospital incinerators is the use of smaller ovens, with smaller capacity, as well as the composition of the waste, containing more plastic, e.g. PVC products.

A factor of 1.5 % of the total sum of dioxins and furans was used to calculate the Nordic Toxicity equivalent (Manscher, 1990).

The total emission of dioxins and furans in Denmark in the 1980's was estimated 2.7 kg/year, an equivalent of 40.5 g/year · Nordic toxic units.

Estimates of dioxins in Norway in 1992 indicate the following emissions:

- 
- Industrial sources: 8 g/year · Nordic toxic units to the air and 5 g to the water
  - Commercial and residential sources: 15 g/year · Nordic toxic units to the air.

## 5.4 Pesticides

### 5.4.1 *Hexachlorocyclohexane (HCH) – Lindane*

Lindane was sold in Denmark in the following amounts (in kg/year):

1983	7300
1984	5800
1985	8400
1986	5151
1987	10385
1988	8145
1989	14609
1990	8356
1991	15317
1992	10058
1993	8640
Mean	9300

(Miljøstyrelsen, 1989; Miljøstyrelsen, 1992; Miljøstyrelsen, 1995a).

The main use of lindane in Denmark is as a fungicide on seeds in agriculture. Spreading of the treated seeds results in the deposition of 50 to 65 g lindane per ha. The minor use of lindane is also in products for pest treatment in house animals, e.g. dogs.

From 1 December 1994 the import and sale of lindane and lindane containing product is prohibited in Denmark and from 1 July 1995 the use of lindane and lindane containing products in Denmark is also prohibited (Miljøstyrelsen, 1995b).

The estimates of Danish emissions of lindane at the beginning of the 1990's are the following:

- 2.6 t/year to the air,
- 1.8 t/year to the water,
- 0.9 t/year to the soil, and
- about 40 kg/year to the sediments.

The use of lindane in Norway is prohibited.

### 5.4.2 *Hexachlorobenzene (HCB)*

The following emissions of hexachlorobenzene were reported in Denmark at the beginning of the 1990's:

- 1.3 t/year to the air, and
- 1.0 t/year to the water.

The emissions of HCB in Norway are lower, and in 1992 the following amounts were reported:

- 123 kg to the air, and
- 2 kg to the water.

No information has yet been available on the Danish and Norwegian emissions of other POPs studied in the reported work.

## **6. Collection of environmental data**

Collection of data on studied POPs has been carried out in order to summarize the available environmental data of these compounds in the Nordic countries. The previous data survey within the 1994 NMR project (Pacyna et al., 1994) has been completed with new data and arranged to better fit the structure of a new data base. The data are summarized in tables enclosed in Appendix A. The collection of data has mainly been based on scientific reports.

The organic compounds included are:

- PAHs,
- PCBs,
- dioxins,
- HCB, and
- selected pesticides: hexachlorocyclohexanes (HCHs), DDTs, and chlordane.

The data survey includes levels of POPs in air and deposition as well as in aquatic and terrestrial environments. The collection of data has mainly been concentrated on POP levels in compartments available for exchange processes. In the terrestrial environment concentrations of POPs in vegetation and soil are included.

In the marine environment, levels of POPs in sea water, sediment and biota have been collected. Up to now mussels have been used to represent biota in the marine water.

This summary shows that most data represent the marine environment while only a few investigations in the terrestrial environment are reported.

There is a lack of data from Finland and an effort will be made to collect more Finnish data.

The concentrations of different POPs given in the tables are mainly from rural and background areas. Data from urban and polluted areas will be included in the further update.

## 7. Acknowledgements

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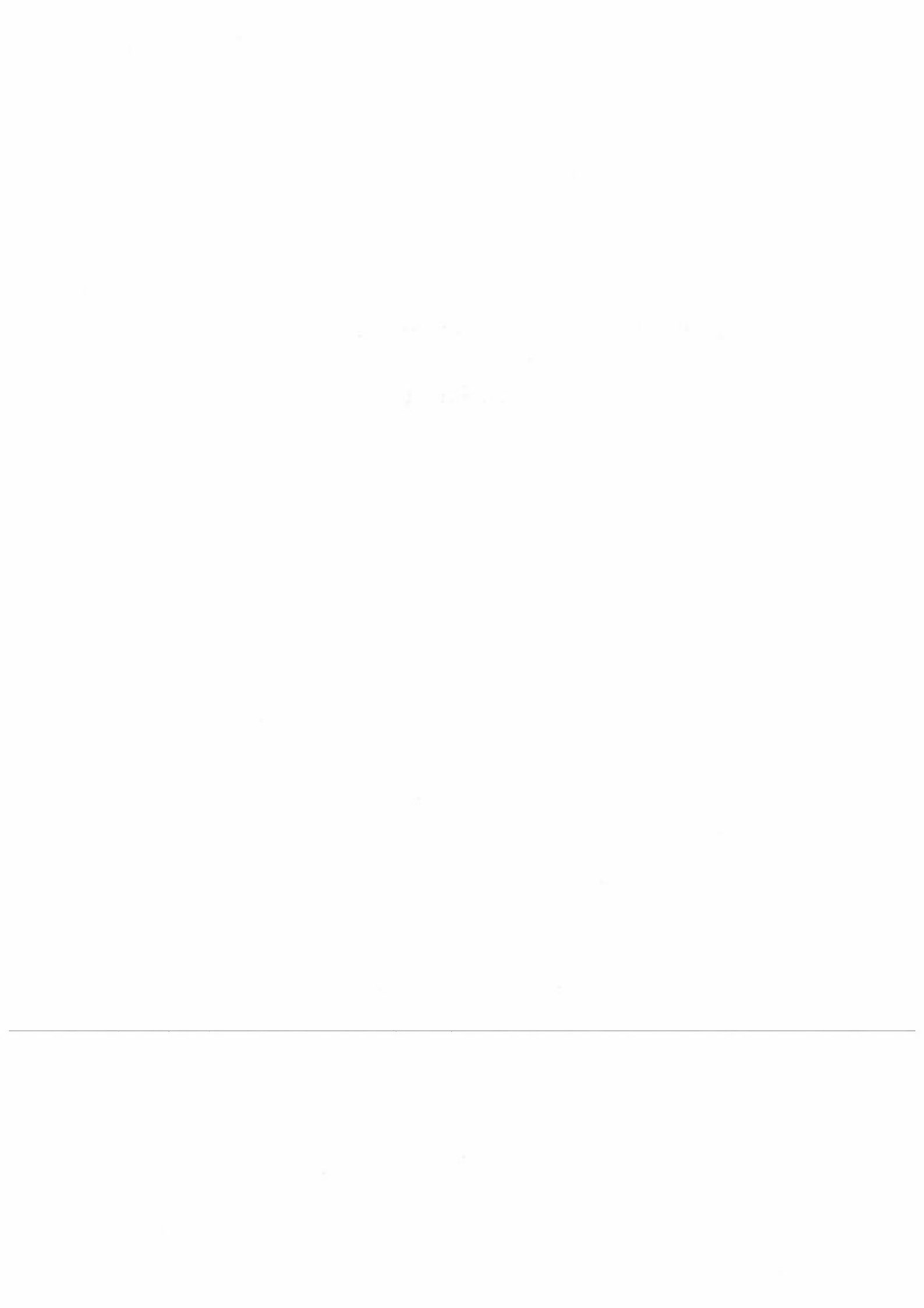
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## **Glossary**



## Glossary

(index  $i$  refers to sub-basin)

### Dimensions

$AW_i$	water surface area in $m^2$
$AS_i$	sediment surface area in $m^2$
$HS_i$	sediment depth in $m$
$VW_i$	volume of water in $m^3$
$VS_i$	volume of surface sediment in $m^3$

### Transport Rates of Water in $m^3/h$

$GX_i$	river water inflow
$GM_i$	precipitation
$G_{i,j}$	water flow from sub-basin $i$ to sub-basin $j$

### Transport and Transformation Rates of POC in $g C/h$

$OP_i$	autochthonous production
$OI_i$	inflow to sub-basin $i$ from neighbouring basins
$OO_i$	outflow from sub-basin $i$ to neighbouring basins
$OX_i$	riverine inflow
$OD_i$	deposition
$OE_i$	resuspension
$OB_i$	sediment burial
$OW_i$	mineralisation rate in the water column
$OS_i$	mineralisation rate in the sediment
$O_{5,9}$	transport rate from surface to bottom water in the Baltic Proper

### Environmental Parameters

$CW_{poc_i}$	POC concentration in sea water in $g/m^3$
$CX_{poc_i}$	POC concentration in inflowing river water in $g/m^3$
$ORG_s_i$	mass fraction organic carbon in surface sediment solids in $g/g$
$BP_i$	biological productivity in the water column in $g OC/(m^2 \cdot h)$
$facOW_i$	fraction of the POC input to the water column (i.e. river inflow, inflow from neighbouring basins and biological production), which is mineralised in the water column
$facOE_i$	fraction of POC gross-deposited to the sediment, which is resuspended
$facOS_i$	fraction of POC net-deposited to the sediment (i.e. deposition minus resuspension), which is mineralised in the surface sediment
$DENoc$	density of organic carbon in $g/m^3$
$DENM$	density of mineral matter in $g/m^3$
$DENS_i$	density of sediment particles in $g/m^3$
$SVG_i$	scavenging ratio

VAQ <sub>i</sub>	volume fraction aerosol in air in m <sup>3</sup> /m <sup>3</sup>
VSW <sub>i</sub>	volume fraction water in surface sediment in m <sup>3</sup> /m <sup>3</sup>
UQ <sub>i</sub>	dry deposition velocity in m/h
UA <sub>i</sub>	air-water mass transfer coefficient, air side in m/h
UW <sub>i</sub>	air-water mass transfer coefficient, water side in m/h
UT <sub>i</sub>	water-sediment mass transfer coefficient in m/h
TA <sub>i</sub>	air temperature in K
R	ideal gas constant in J/(K·mol)

### Chemical Properties

H <sub>i</sub>	Henry's law constant in Pa·m <sup>3</sup> /mol
P <sub>i</sub>	vapour pressure of the sub-cooled liquid in Pa
K <sub>ow</sub>	partition coefficient octanol to water
K <sub>poc</sub>	partition coefficient particulate organic carbon to water
HLW <sub>i</sub>	degradation half-life in water in h
HLS <sub>i</sub>	degradation half-life in surface sediment in h

### Model Parameters

all Z-values	explained in Table 4
EW <sub>i</sub>	direct emission rate into water in mol/h
I <sub>i</sub>	total input rate of chemical to water (direct, riverine and atmospheric) in mol/h
D <sub>sed</sub> <sub>i</sub>	D-value describing net transfer from water to sediment in mol/(h·Pa)
D <sub>tot</sub> <sub>i</sub>	sum of all D-values describing loss from the water compartment in mol/(h·Pa)
all other D -values	explained in Table 5
FA <sub>i</sub>	fugacity in air in Pa
FX <sub>i</sub>	fugacity in river water in Pa
FW <sub>i</sub>	fugacity in sea water in Pa
FS <sub>i</sub>	fugacity in surface sediment in Pa

## **Tables and figures**

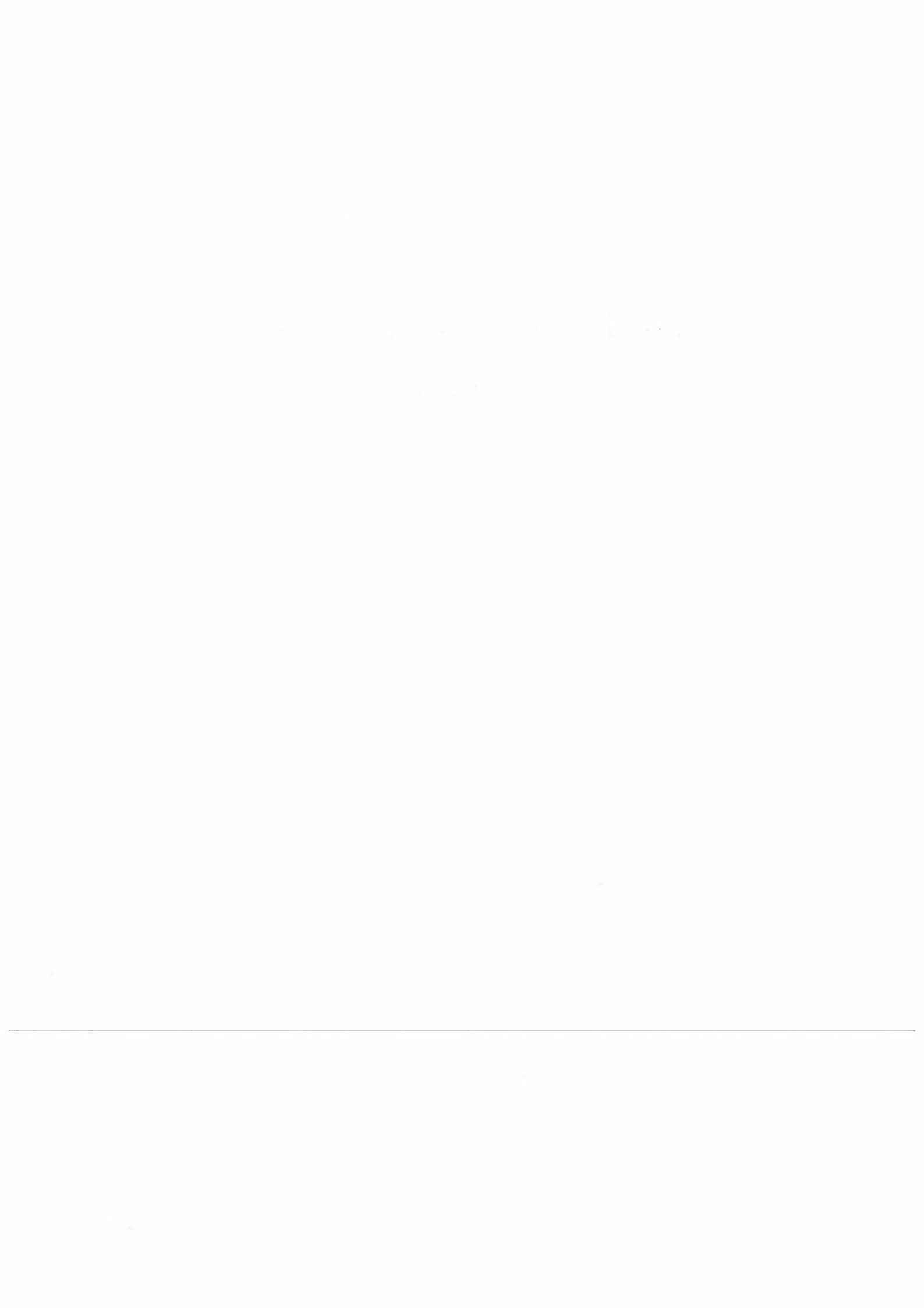


Table 1: Calculation of POC Mass Balance for Sub-Basins 1 to 4, 6 and 7 (for explanation of symbols see glossary).

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<b>Input Parameters</b> for each sub-basin i	
AW <sub>i</sub> , GX <sub>i</sub> , G <sub>i,i</sub> , G <sub>i,i</sub> (from water balance, Figure 1)	
CW <sub>poc,i</sub> , CX <sub>poc,i</sub> , BP <sub>i</sub> , facOW <sub>i</sub> , facOE <sub>i</sub> , facOS <sub>i</sub>	

---

<b>Calculation of POC Fluxes and Rates in g C/h</b> for each sub-basin i	
autochtoneous production	$OP_i = BP_i \cdot AW_i$
inflow from other basins j	$OI_i = \sum_j (G_{j,i} \cdot CW_{poc_j})$
outflow to other basins j	$OO_i = \sum_j (G_{i,j} \cdot CW_{poc_i})$
inflow with rivers	$OX_i = GX_i \cdot CX_{poc_i}$
mineralisation in water column	$OW_i = facOW_i \cdot (OI_i + OP_i + OX_i)$
resuspension from the sediment	$OE_i = \frac{(OI_i + OP_i + OX_i - OO_i - OW_i)}{(1 / facOE_i - 1)}$
deposition to the sediment	$OD_i = \frac{OE_i}{facOE_i}$
mineralisation in surface sediment	$OS_i = facOS_i \cdot (OD_i - OE_i)$
burial to deeper sediment layers	$OB_i = OI_i + OP_i + OX_i - OO_i - OW_i - OS_i$

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Table 2: Calculation of POC Mass Balance for the Baltic Proper (for explanation of symbols see glossary).

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**Input Parameters**

$AW_5, AW_9, GX_5, G_{5,j}, G_{j,5}$  (from water balance, Figure 1)

$CW_{poc5}, CW_{poc9}, CX_{poc5}, BP_5, facOW_5, facOW_9, facOE_5, facOE_9, facOS_5, facOS_9$

---

**Calculated as in Table 1**

$OP_5, OI_5, OO_5$  and  $OX_5$

---

**POC Transport from Bottom Water to Surface Water**

$$O_{9,5} = G_{9,5} \cdot CW_{poc_i}$$


---

**10 Unknown Parameters**

$OD_5, OD_9, OE_5, OE_9, OB_5, OB_9, O_{5,9}, OS_5, OS_9, OW_9$

---

**10 Equations**

The following set of equations is solved simultaneously using a numerical method

$$OI_5 + OP_5 + OX_5 + O_{9,5} + OE_5 = OO_5 + OW_5 + OD_5 + O_{5,9} \quad (1)$$

$$O_{5,9} + OE_5 = OD_5 + OW_5 + O_{9,5} \quad (1)$$

$$OD_5 = OE_5 + OB_5 + OS_5 \quad (2)$$

$$OD_9 = OE_9 + OB_9 + OS_9 \quad (2)$$

$$OE_5 = facOE_5 \cdot OD_5$$

$$OE_9 = facOE_9 \cdot OD_9$$

$$OS_5 = facOS_5 \cdot (OD_5 - OE_5)$$

$$OS_9 = facOS_9 \cdot (OD_9 - OE_9)$$

$$OW_9 = facOW_9 \cdot O_{5,9}$$

$$OD_5 \cdot AW_9 = (O_{5,9} - G_{5,9} \cdot CW_{poc_5}) \cdot (AW_5 - AW_9) \quad (3)$$


---

1) mass balance for water compartment,

2) mass balance for sediment compartment,

3) this equation is based on the assumption that on an area basis the POC settling rate to the shallow sediments and to the bottom water is the same.

Table 3: Selected input parameters describing concentrations and fate of particulate organic carbon in the Baltic Sea (for explanation of symbols see glossary).

	Bothnian Bay	Botnian Sea	Gulf of Finland	Gulf of Riga	Baltic Prp.(S)	Baltic Prp.(B)	Danish Straits	Kattegat
ORGS, m <sup>3</sup> /m <sup>3</sup>	0.05	0.05	0.05	0.05	0.05	0.08	0.05	0.05
CXpoc, µg/L	1000	1000	750	2500	3000	n.a.	1000	1000
CWpoc, µg/L	200	200	200	200	200	100	200	200
BP, gC/(m <sup>2</sup> ·a)	20	55	75	85	150	n.a.	125	125
facOW	0.65	0.70	0.80	0.75	0.85	0.65	0.70	0.70
facOS	0.60	0.65	0.70	0.75	0.75	0.50	0.75	0.75
facOE	0.75	0.70	0.70	0.80	0.85	0.60	0.75	0.75

Table 4: Calculation of Z-values in units of mol/(m<sup>3</sup>·Pa) (for explanation of symbols see glossary).

Input Parameters	
TA <sub>i</sub> , P <sub>i</sub> , H <sub>i</sub> , K <sub>OW</sub> , ORGS <sub>i</sub> , DENoc, DENM, CWpoc <sub>i</sub> , CXpoc <sub>i</sub> , VAQ <sub>i</sub> , VSW <sub>i</sub>	
Auxiliary Parameters	
$K_{poc} = 0.41 \cdot K_{OW}$	$DENS_i = \frac{I}{\frac{1-ORGS_i}{DENM} + \frac{ORGS_i}{DENoc}}$
Pure Phases	
water	$ZW_i = I / H_i$
POC	$Zpoc_i = ZW_i \cdot K_{poc}$
sediment particles	$ZS_i = ZW_i \cdot DENS_i \cdot ORGS_i \cdot K_{poc} / 1000000$
air	$ZA_i = I / R \cdot TA_i$
aerosols	$ZQ_i = ZA_i \cdot 6000000 / P_i$
Bulk Phases	
bulk water	$BZW_i = ZW_i + (CWpoc_i / DENoc) \cdot Zpoc_i$
bulk river water	$BZX_i = ZW_i + (CXpoc_i / DENoc) \cdot Zpoc_i$
bulk sediment	$BZS_i = VSW_i \cdot ZW_i + (1 - VSW_i) \cdot ZS_i$
bulk air	$BZA_i = VAQ_i \cdot ZQ_i + (1 - VAQ_i) \cdot ZA_i$

Table 5: Calculation of  $D$ -values in units of  $\text{mol}/(\text{h}\cdot\text{Pa})$  (for explanation of symbols see glossary).

<b>Input Parameters</b>	
$UA_i, UW_i, UQ_i, UT_i, SVG_i, HLW_i, HLS_i$ $AW_i, AS_i, VW_i, VS_i, GX_i, GM_i, G_{i,j}$ (from water balance, Figure 1) $OD_i, OE_i, OB_i, O_{5,9}$ (from POC balance, Tables 1 and 2) $ZW_i, Zpoc_i, ZA_i, ZQ_i, BZW_i, BZX_i, BZA_i, BZS_i$ (see Table 4)	
<b>Riverine Inflow</b>	
riverine inflow	$DX_i = GX_i \cdot BZX_i$
<b>Inter-Basin Transport</b>	
horizontal flow	$D_{i,j} = G_{i,j} \cdot BZW_i$
vertical upward flow	$D_{9,5} = G_{9,5} \cdot BZW_9$
vertical downward flow	$D_{5,9} = G_{5,9} \cdot ZW_9 + O_{5,9} \cdot Zpoc_5$
<b>Air-Water Exchange</b>	
wet deposition	$DM_i = GM_i \cdot (ZW_i + SVG_i \cdot VAQ_i \cdot ZQ_i)$
dry particle deposition	$DQ_i = AW_i \cdot UQ_i \cdot VAQ_i \cdot ZQ_i$
diffusive gas exchange	$DV_i = l \left/ \frac{1}{AW_i \cdot UA_i \cdot ZA_i} + \frac{1}{AW_i \cdot UW_i \cdot ZW_i} \right.$
<b>Water-Sediment Exchange</b>	
sedimentation	$DD_i = OD_i \cdot Zpoc_i / DENoc$
resuspension	$DR_i = OE_i \cdot Zpoc_i / DENoc$
sediment burial	$DB_i = OB_i \cdot Zpoc_i / DENoc$
water-sediment diffusion	$DT_i = AS_i \cdot UT_i \cdot ZW_i$
<b>Degradation</b>	
in the water column	$DW_i = VW_i \cdot BZW_i \cdot \ln 2 / HLW_i$
in the surface sediment	$DS_i = VS_i \cdot BZS_i \cdot \ln 2 / HLS_i$

Table 6: Physical-chemical properties and degradation half-lives of "PCBs" used in the model calculations.

molecular weight <sup>1</sup>	360.9 g/mol
melting point <sup>1</sup>	114 °C
entropy of fusion <sup>1</sup>	45.2 J/(Kmol)
vapour pressure <sup>2</sup>	$\log(p \text{ in Pa}) = 14.84 - 5399/(T \text{ in K})$
aqueous solubility <sup>3</sup>	$s \text{ in mol/m}^3 = -0,154 - 1497/(T \text{ in K})$
$\log K_{OW}$ <sup>1</sup>	7.0
degradation half life in water	50 years
degradation half-life in sediment	$\infty$

1) PCB-155, Mackay et al., 1992,

2) PCB-155, Wania et al., 1994,

3) PCB-155, W.Y. Shiu, University of Toronto, pers. comm.

Table 7: Assumed average concentrations of PCBs in the atmosphere above and in river water draining into seven sub-basins of the Baltic Sea. Also listed are the estimated direct discharge rates of PCBs and the concentration of aerosols in the atmosphere.

	Bothnian Bay	Botnian Sea	Gulf of Finland	Gulf of Riga	Baltic Proper	Danish Straits	Kattegat
CA in pg/m <sup>3</sup>	10	12	15	15	18	30	21
CX in pg/L	200	300	400	400	400	400	400
EW in kg/a	5	10	40	15	75	25	10
VAQ in m <sup>3</sup> /m <sup>3</sup>	1.0.10-11	1.5.10-11	2.5.10-11	2.5.10-11	2.0.10-11	2.0.10-11	2.0.10-11

Table 8: Selected environmental input parameters (for explanation of the symbols see glossary).

UA	1.00 m/h	UT	0.0004 m/h	DENoc	1,000,000 g/m <sup>3</sup>
UW	0.01 m/h	SVG	200000	DENM	2,600,000 g/m <sup>3</sup>
UQ	7.20 m/h	VSW	0.85	HS	0.01 m

Table 9: *Relative importance of various input and output pathways for PCBs in the sub-basins of the Baltic Sea (in percent).*

	Bothnian Bay	Bothnian Sea	Gulf of Finland	Gulf of Riga	Baltic Proper	Danish Straits	Kattegat
<b>output</b>							
direct emissions	11	8	32	31	17	12	4
river inflow	41	25	36	24	9	2	4
atmospheric deposition	39	49	29	42	49	17	9
interbasin flow	9	18	4	3	25	69	83
<b>output</b>							
volatilisation	44	35	37	43	28	10	8
degradation	2	2	2	1	3	0	0
sediment burial	30	43	35	44	54	7	4
interbasin flow	24	20	26	12	15	83	88

Table 10: *Calculated characteristic response times of the water column and the total aquatic system (i.e. water column and surface sediment).*

	response time of water in days	response time of aquatic system in years
Bothnian Bay	182	19.3
Bothnian Sea	160	14.2
Gulf of Finland	105	14.2
Gulf of Riga	32	14.3
Baltic Proper	195	16.6
Baltic Proper (Surface)	113	3.7
Baltic Proper (Bottom)	193	28.4
Danish Straits	16	2.5
Kattegat	25	1.9
Baltic Sea	151	18.3

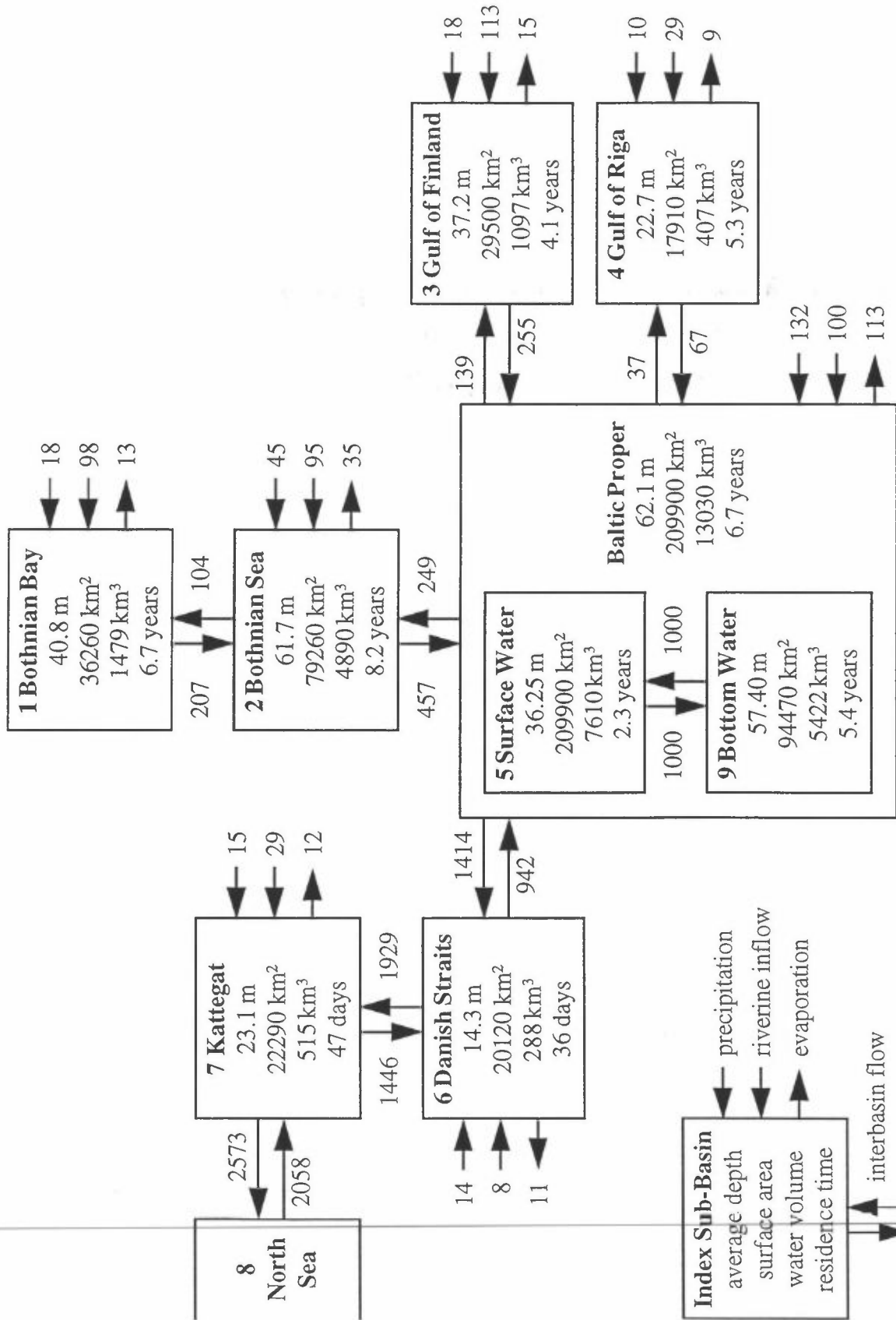


Figure 1 Water balance of the Baltic Sea (in km<sup>3</sup>/year)

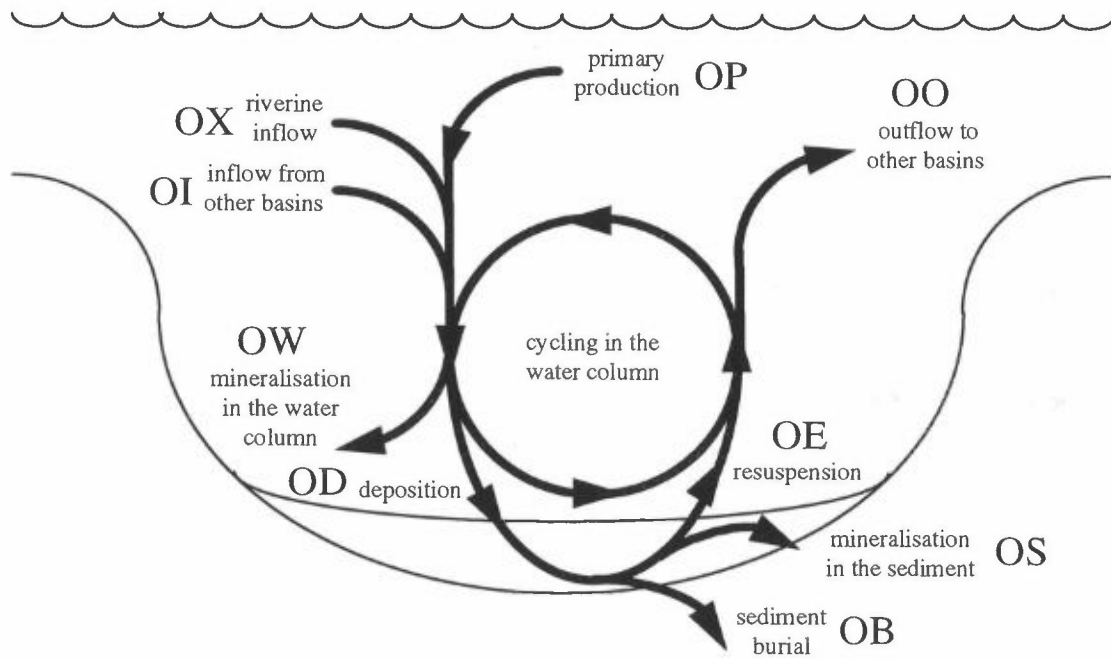


Figure 2 Particulate organic carbon transport fluxes and mineralisation rates are calculated for each sub-basin of the Baltic Sea.

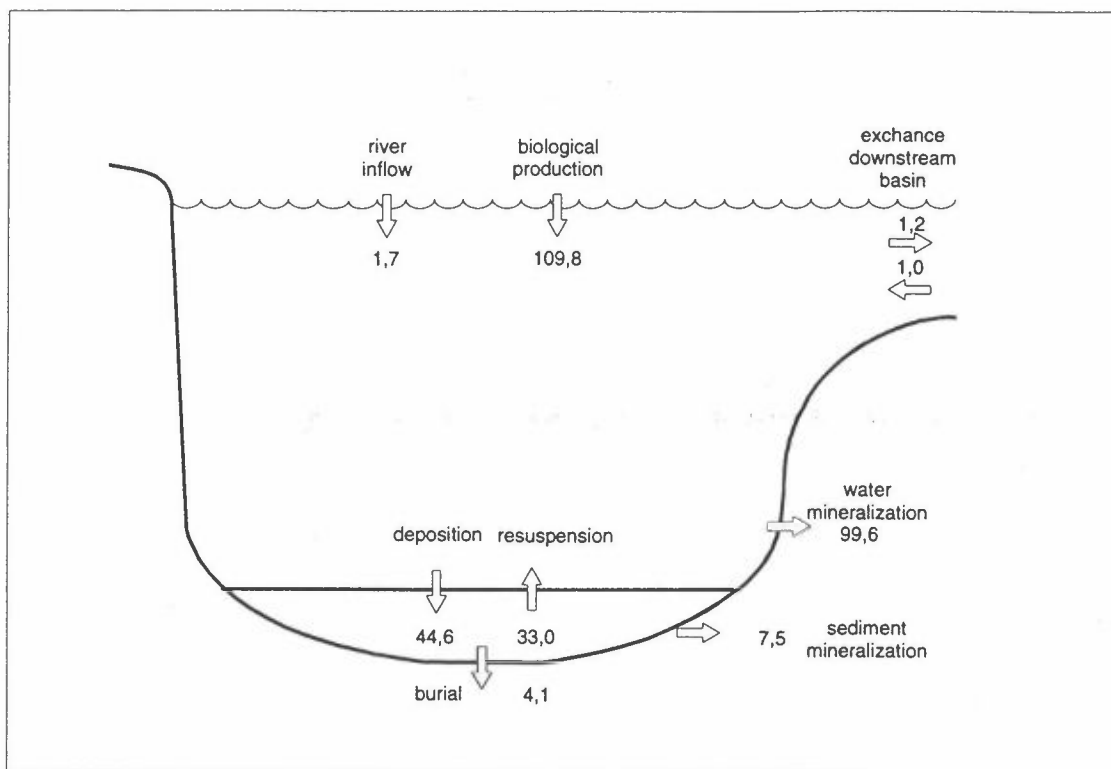


Figure 3 POC balance of the Baltic Sea (in  $\text{g}/(\text{m}^2 \cdot \text{year})$ )



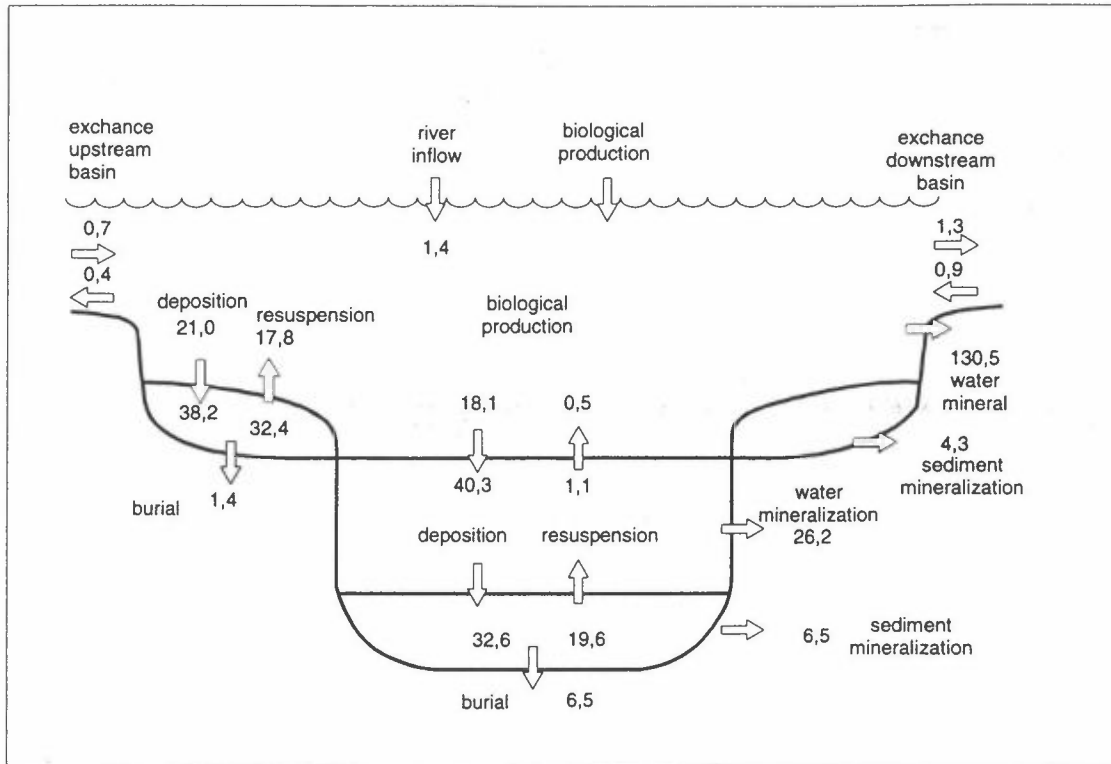


Figure 4 POC balance of the Baltic Proper (in g/(m<sup>2</sup>.year)).

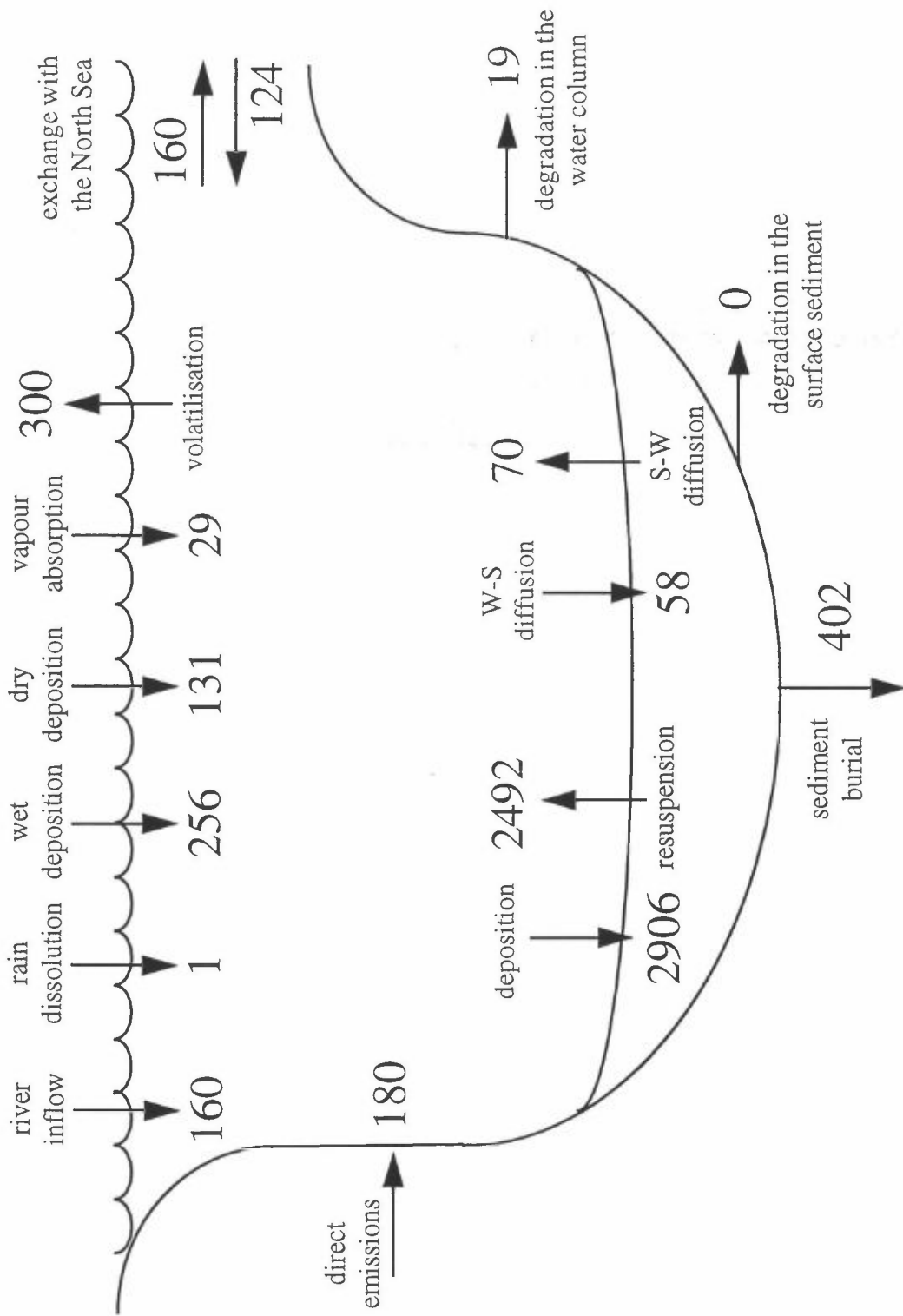


Figure 5 PCB mass balance of the Baltic Sea (in kg/year)

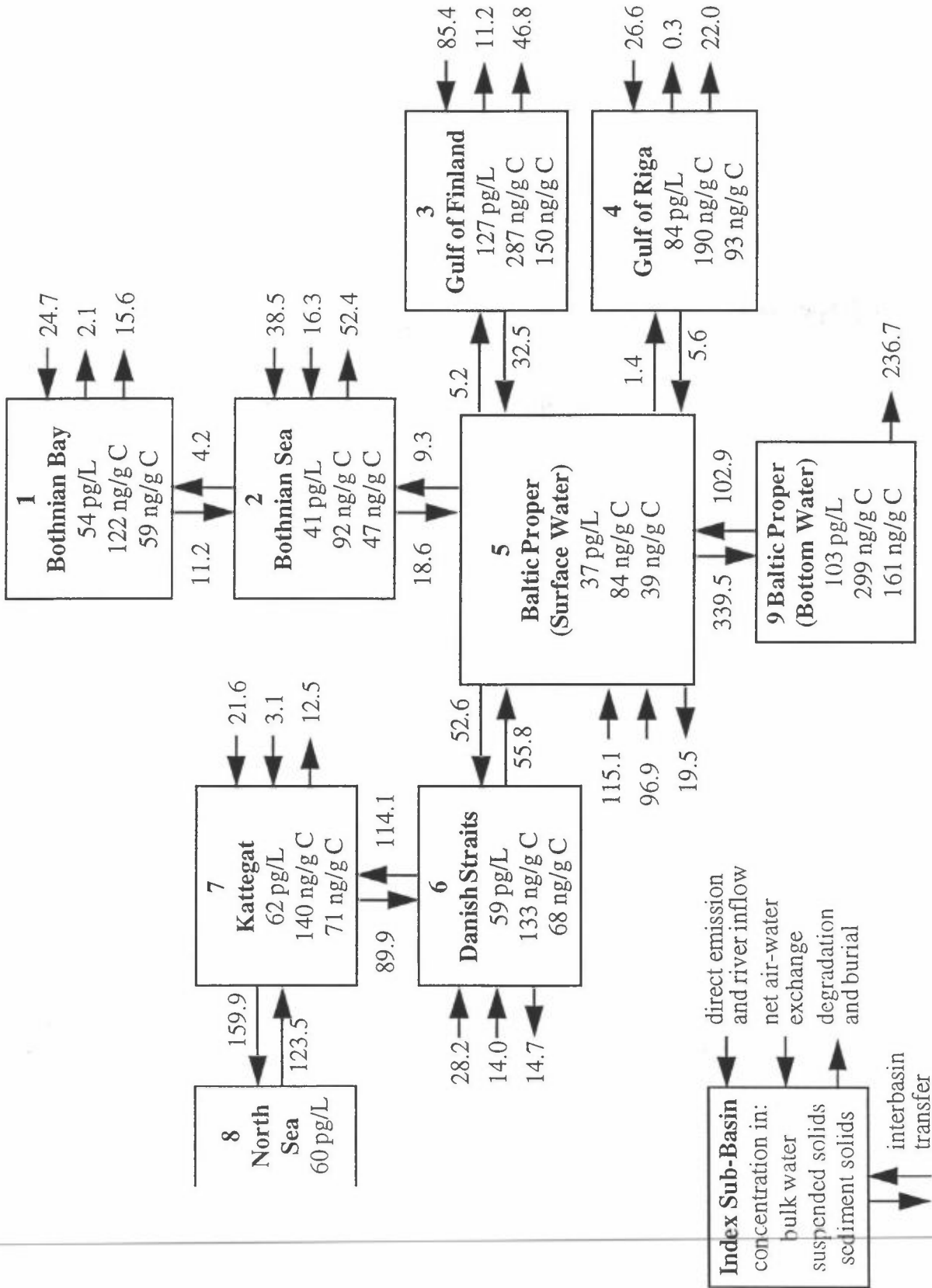
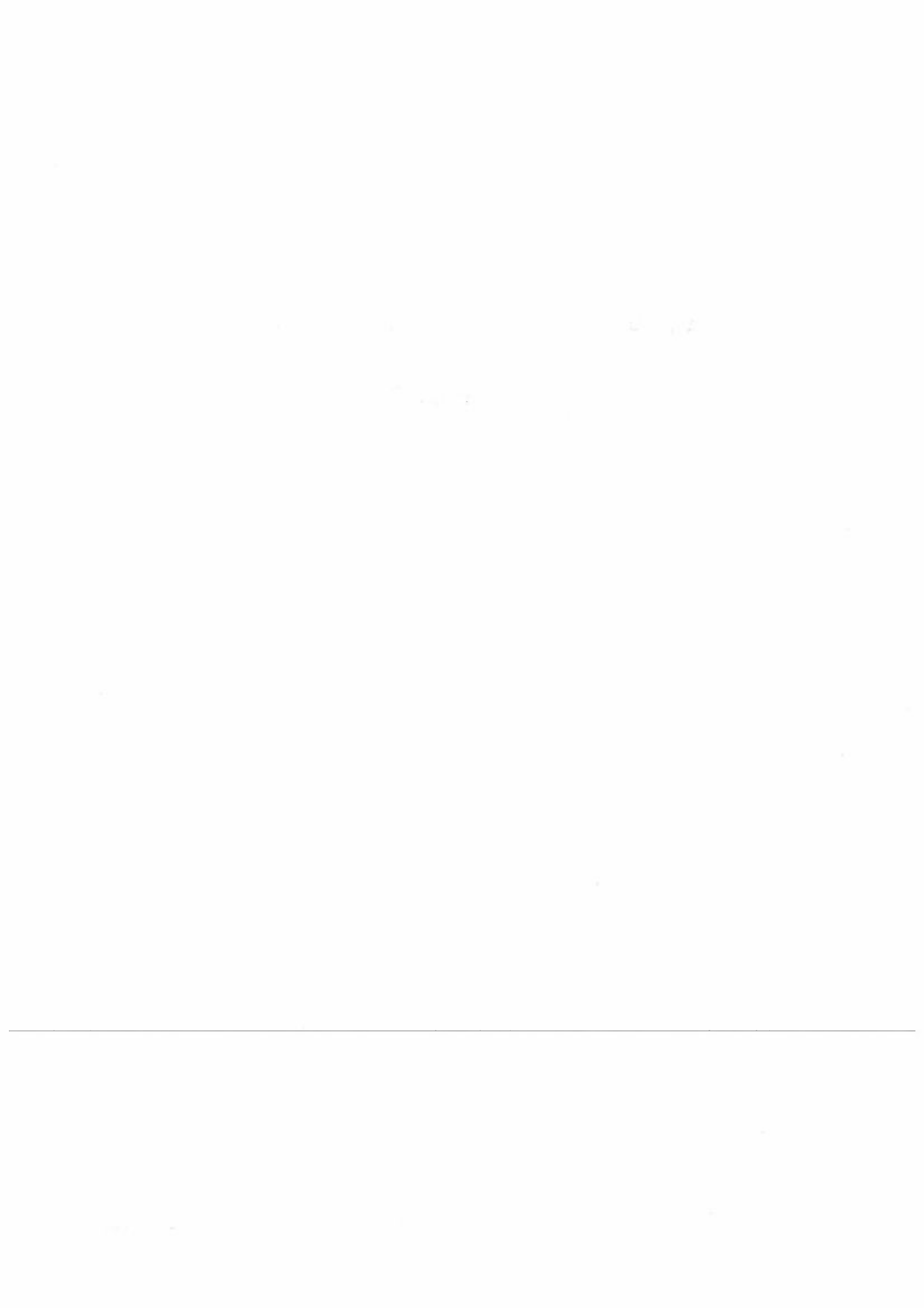


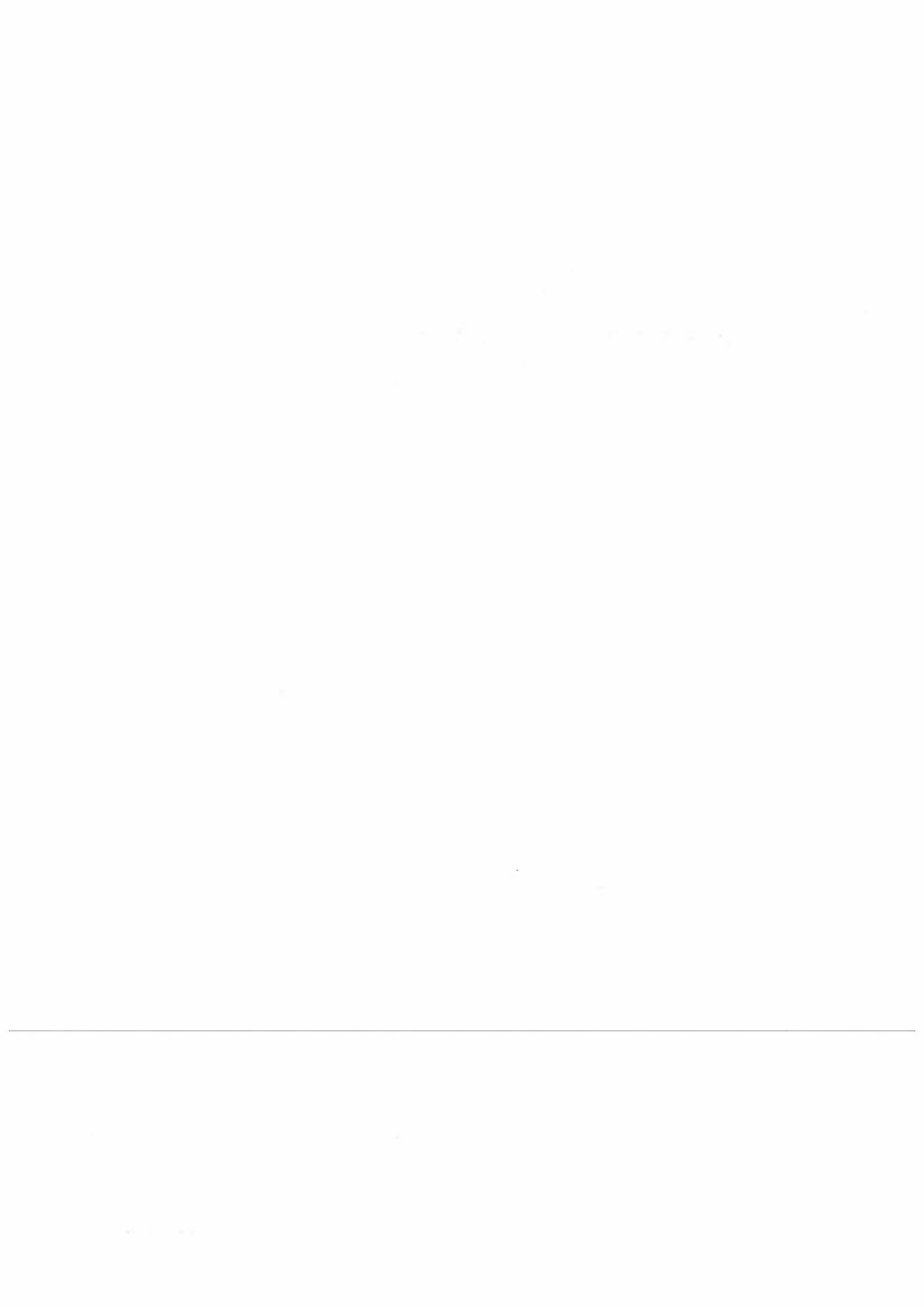
Figure 6 PCB mass balance of the Baltic Sea (in kg/year)

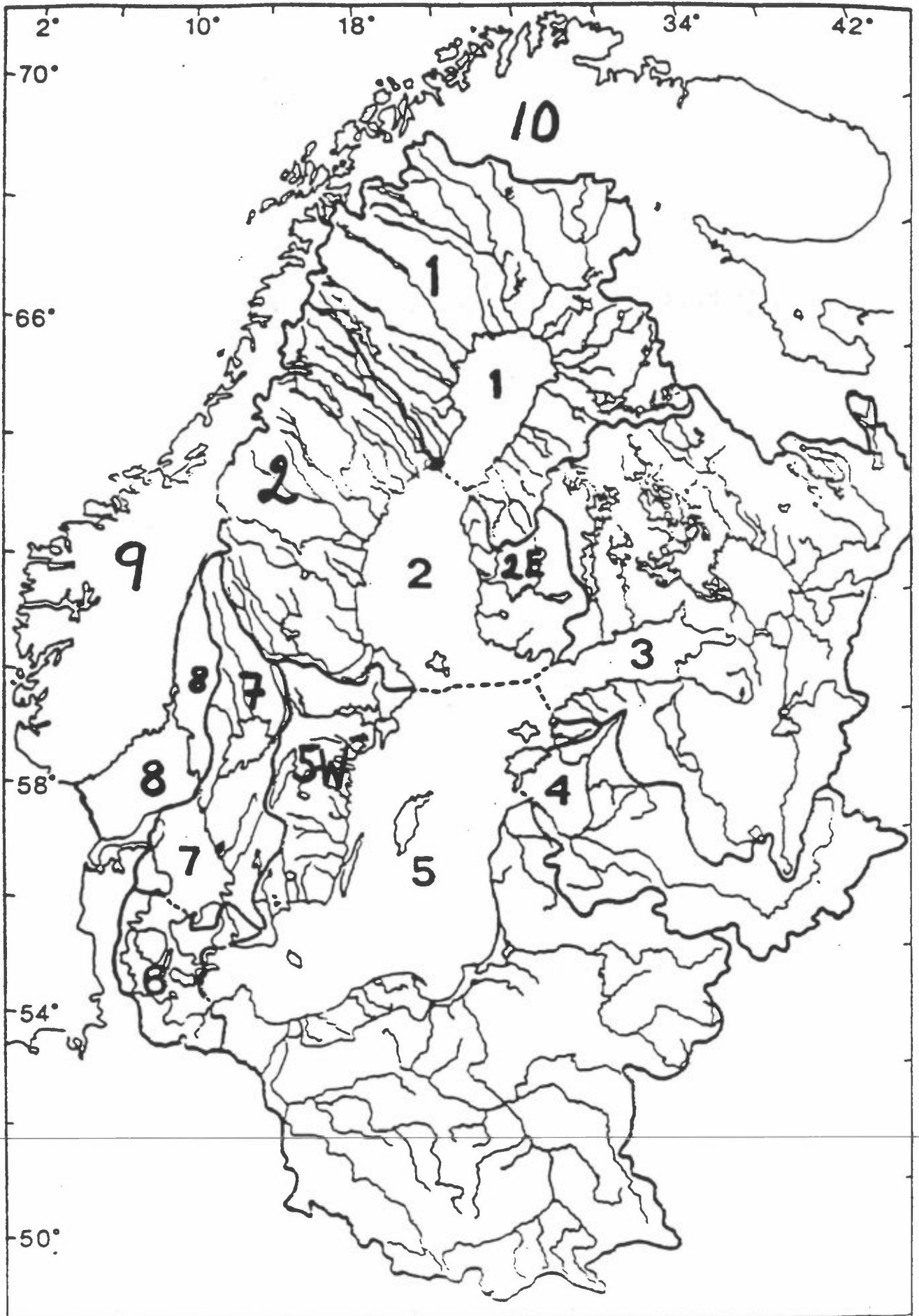
**Appendix A**  
**Environmental Data Base**



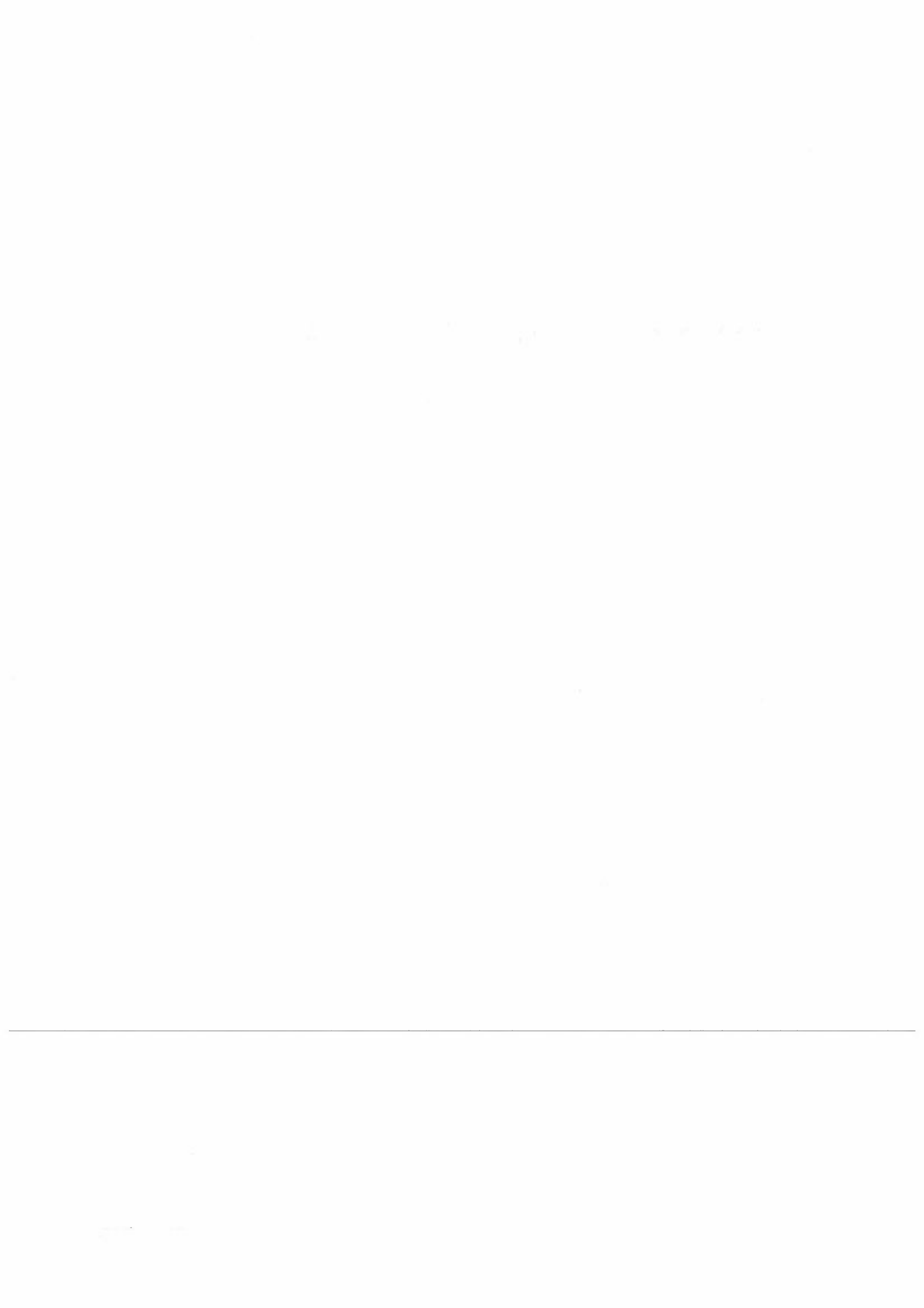
## Abbreviations and explanations to the tables

<b>Ref.</b>	See enclosed list of references
<b>Country</b>	The Nordic country where the investigation has been carried out
<b>Year</b>	Time for the measurements
<b>Seas.</b>	Seasons, the time of the year when the samples were collected
<b>Location</b>	The region where the measurements have been carried out
<b>Area code</b>	See enclosed map
<b>Site category</b>	The characteristics for the sampling site (if possible to define)
<b>Site numb.</b>	Number of sampling sites
<b>Obser.</b>	Number of observations per site









## Matrix Air

Ref	Country	Year	Seas.	Location	Area code	Site category	Site numb.	obser. num/site	PAH (number) ng/m <sup>3</sup>	PCB (number) pg/m <sup>3</sup>	HCH alpha pg/m <sup>3</sup>	HCH gamma pg/m <sup>3</sup>	HCB pg/m <sup>3</sup>	PCDF, PCDD (sum) pg/m <sup>3</sup>	DDE pg/m <sup>3</sup>	DDT pg/m <sup>3</sup>	Chlord. gamma pg/m <sup>3</sup>	Chlord. alpha pg/m <sup>3</sup>	Other data to be included	Comment
3	Nor	1981	Spring	No S	9	Inland	1	4-7			347-1230	154-410	55-230						Arctic	
4	Nor	1984	Surf	No S-N	9,10	coast/ Inland	3	12		PCB 5-8 Cl	289-1168	37-705	30-153				0.7-3.7	1.3-7.8	Arctic	
4	Nor	1984	Winter	No S-N	9,10	coast/ Inland	3	12		PCB 5-8 Cl	56-507	6-43	55-210				0.4-3.2	0.4-3.1	Arctic	
5	Nor	1992	Spring	No S-N	9,10	coast/ Inland	3	16		PCB 28-180	40-200	10-500	55-280				0.24-2.1	0.4-2.8	Arctic	
1	Swe	1984-85	Jan-Dec	Sw E	5W	coast	1	11-13		23-1200(tot)	190-3200	21->660	54->170				1.3-10	1.0-15	Urban	extreme values excluded
11	Swe	1984-85	Jan-Dec	Sw S-N	1,2,5w,7,8	coast/ Inland	14	4		<10->1000(tot)										
2	Swe	1986-87	Dep-May	Sw E	5W	coast	5	1	2.8-6.9 (15)											Urban
2	Swe	1989	Apr-Jun	Sw E	5W	coast	2	1						0.2-0.4						
6	Swe	1988-90	all	Swe. W	7	coast	1	14						0.27-5.2			0.4-1.7	1-1.9		
7	Swe	1988	wint	Sw W	7	coast	1	5	2.9-25 (11)											
8	Swe	1989-90	wint-spr.	Sw W	7	coast	1	12	1.5-30 (11)	6-120 (7)	14-550	9-1100								
9,10	Swe	1991-94	all	Sw W	7	coast	1	21	1.5-8.5 (11)	12-69(7)	10-150	10-520								
8,14	Swe	1991-94	all	Sw W	7,8	forest	1	12	1.1-5.9 (11)	11-18 (7)	24-110	44-270								
29	Swe	1994	all	Sw W	7	coast	1	9	0.7-16(11)	11-100(7)	18-63	12-200								
28	Swe		all	Sw E	5	sea	1	11		10-57(7)										

## Matrix Deposition

Ref	Country	Year	Seas.	Location	Area code	Site category	Site numb.	observ. numb.	PAH µg/m <sup>2</sup> day	PCB ng/m <sup>2</sup> day	HCH alpha ng/m <sup>2</sup> day	HCH gamma ng/m <sup>2</sup> day	HCB ng/m <sup>2</sup> day	PCDF, PCDD (sum) ng/m <sup>2</sup> day	DDE ng/m <sup>2</sup> day	DDT ng/m <sup>2</sup> day	Chlord. gamma ng/m <sup>2</sup> day	Chlord. alpha ng/m <sup>2</sup> day	Comment	
12	Den	1990	Jan-Dec	De	6	coast/ inland	2	12			2-25	5-190								
11	Swe	1984-85	Jan-Dec	Sw S-N	1,2,5w, 7,8	coast/ inland	14	4		<20->300 (tot)										Silicone oil net Extreme values excluded
7	Swe	1988	wint	Sw W	7	coast	1	5	1-7.1 (11)											Bulk sampler 1 m <sup>2</sup>
8	Swe	1989-90	wint-spri.	Sw W	7	coast	1	4-12	0.2-2 (11)	2.3-4.5 (7)	0.11-3.2	0.8-35								Bulk sampler 1 m <sup>2</sup>
9,10	Swe	1991-92	all	Sw W	7	sea	1	21	0.1-1.5 (11)	0.7-5.6 (7)	0.7-3.5	0.8-29								Bulk sampler 1 m <sup>2</sup>
9,10	Swe	1991-94	all	Sw W	8	sea	1	14	0.1-0.87 (11)	0.9-4.6 (7)	0.2-3.6	0.2-8.6								Bulk sampler 1 m <sup>2</sup>
8,14	Swe	1991-94	all	Sw W	8	forest	1	12	0.03-1.6 (11)	1.3-1.6 (7)	<0.1-7.2	<0.1-22	0.1-0.4							Bulk sampler 1 m <sup>2</sup>
29	Swe	1994	all	Sw W	7	coast	1	4-9	0.02-0.31 (11)	0.8-7.7 (7)	0.1-5.9	0.3-20	0.5-10	0.2-3	0.1-0.6					Bulk sampler 1 m <sup>2</sup>
31	Swe	1994	Apr-Oct	Sw E	5W	coast	1	7		0.2-5 (7)	0.05-3	0.1-10	<0.05-0.1	0.01-0.04	0.4-1.3					Bulk sampler 1 m <sup>2</sup>

Marine environment  
Matrix Sea water

Ref	Country	Year	Seas.	Location	Area code	Site category	Site numb.	obser. num/site	PAH (number)	PCB (number)	HCH alpha	HCH gamma	HCB	PCDF, PCDD (sum)	DDE	DDT	Chlord. gamma	Chlord. alpha	Comments
Surface	Ger	1983	May~Jun	Baltic	1,2,3,5	offshore	13	1			5-8	2-4							surface 5m
	Ger	1983	May~Jun	Kattegat	6,7		5	1			5-6	4-6							surface 5m
	Ger	1985	Aug	Skagerrak	8	offshore	1	1			3	3,2							surface 5m
	Ger	1985	Aug	No.W	9	offshore	4	1			1.3-2.7	0.5-2.1							surface 5m
	Swe	1988	May~Jun	Baltic	1,2,5	offshore	9	1			4-4.7	1.5-3.9							surface 5m
	Swe	1988	Jun~Jul	Baltic	1,2,5	coast	3	1	0.6-1.1 (11)					0.3-0.4					surface 9-12m
	Swe	1988	Jun~Jul	Baltic	1,2,5	offshore	6	1	0.5-0.8 (11)					0.2-0.5					

Marine environment  
Matrix Sediment

Ref	Country	Year	Seas.	Location	Area code	Site category	Site numb.	obser. num/site	PAH (number)	PCB (number)	HCH alpha	HCH gamma	HCB	PCDF, PCDD (sum)	DDE	DDT	Chlord. gamma	Chlord. alpha	Comments
									µg/g dw	ng/g dw	ng/g dw	ng/g dw	ng/g dw	pg/g dw	ng/g dw	ng/g dw	ng/g dw	ng/g dw	
22	Nor	1990		No. Atlantic	9	offshore	9	1	<0.04-0.3 (25)	0.07-0.64 (7)		<0.01-0.06	0.01-0.05		0.03-0.2 (sum DDT)		<0.01-0.06 (sum chlord.)		far offshore
21	Nor			North Sea/ Skagerrak			5	2											
23	Nor	1991		No W Olenstj	9	coast	8	1		0.8-3.6 (7)									
24	Swe	1990		Skagerrak	8	offshore/ coast	8	1	0.6-1 (11)	1.3-3.9 (7)	<0.04-0.14	<0.04-0.15	0.1-0.3		0.3-0.6	<0.2	<0.1		
24	Swe	1991		Skagerrak	8	offshore/ coast	5	1	0.8-1.5	1.9-2.8(7)	0.03-0.1	0.02-0.1	0.2-0.3		0.4-0.8	0.01-1	0.01-0.02	0.02-0.04	
24	Swe	1993		Skagerrak	8	offshore	9	1	0.5-2	1.7-4.6(7)	0.03-0.07	0.01-0.08	0.1-0.3		0.3-0.6	0.04-1.8	0.01-0.03	0.02-0.03	
32		1989-90		Baltic	1	offshore	2	1		8.6-8.9(7)			0.7-1		0.6-0.9	0.9-2.8			
32		1989-90		Baltic	2	offshore	2	1		3.2-7.3(7)			0.9-1.6		0.7-2	1-1.7			
32		1989-90		Baltic	5	offshore	8	1		16-34(7)			0.3-1.4		0.5-17	nd-1.3			
32		1989-90		Kattegat	6,7	offshore	6	1		1.5-11(7)			0.1-2.6		0.2-6	nd			
32		1989-90		Skagerrak	8	offshore	6	1		4.6-10(7)			0.1-1.2		0.1-1	nd			
33	Swe	1990		Skagerrak	8	costal	13	1	0.8-5.3	4-200(tot)			0.1-12						
34	Summary			Skagerrak	8	offshore			2-3(tot)	3-6(7)			<0.1-0.2						
34	Summary			Skagerrak	9	offshore			0.5-2(tot)	1-3(7)									

Marine environment  
Matrix Biota

Ref	Country	Year	Seas.	Location	Area code	Site category	Site numb.	obser. num/site	PAH (number) ng/g dw	PCB (number) ng/g dw	HCH alpha ng/g dw	HCH gamma ng/g dw	HCB ng/g dw	PCDF, PCDD (sum)	DDE ng/g dw	DDT ng/g dw	Chlord. gamma ng/g dw	Chlord. alpha	Comments
Mussels 26	Nor	1990-91		No. W	9	coast	5	1		2-5.5	0.2-0.5	0.2-0.5	0.04-0.07		0.34-26 (sum DDT)				50 mussels
23	Nor	1991		No W Olensfj	9	coast	8	1		4.3-10									
25	Den	1985-86		Kattegat, Belt Sea	6,7	coast/ offshore	25	1	22-670	12-140	2-12	4-18	0.4-2.6		15-81 (sum DDT)				contaminated sites included
27	Swe	1993		Sw W	8	coast				12-96			<2-3.5						det limit

Matrix Rivers

Ref	Country	Year	Seas.	Location	Area code	Site category	Site numb.	obser. num/site	PAH (number)	PCB (number)	HCH alpha	HCH gamma ng/l	HCB	PCDF, PCDD (sum)	DDE	DDT	Chlord. gamma	Chlord. alpha	Comments
35	Nor	1992		No S	8		10		nd-0.3 (7)			0.5-1.5							

Terrestrial environment  
Matrix Vegetation

Ref	Country	Year	Seas.	Location	Area code	Site category	Site numb.	obser. num/site	PAH (number)	PCB (number)	HCH alpha ng/g	HCH gamma ng/g	HCB ng/g	PCDF, PCDD (sum) ng/g	DDE ng/g	DDT ng/g	Chlord. gamma ng/g	Chlord. alpha ng/g	Comments
Needles	Swe	1984-86		Sw N	1		1	3		4-8 (tot)fw	2-6 (tot) fw				0.25-0.31 (tot) fw				Pine needle 84-86
15	Swe	1984-86		No,Sw	5w, 7,8,9		23	3		4-6 (tot)fw	1,4-2,9 (tot) fw				0.1-0.4 (tot) fw				Pine needle 84-86
16	Swe	1983-88		Den	6		1	5			0.5-3.8 fw	0.4-2.3 fw	0.08-0.6 fw		0.1-0.3 fw	0.24-0.55 fw			Pine needle 84-88
16	Swe	1983-88		Sw	5w		1	5			0.84-5.6 fw	0.29-2.0 fw	0.4-1.4 fw		0.03-0.21 fw	0.1-0.19 fw			Pine needle 84-88
8,14	Swe	1991-94	All	Sw W	8	forest	1	4-11	22-140 (11) dw	1,3-2,8 (7) dw	0.45-1,2 dw	0.67-1,5 fw	0.33-0.51dw						Spruce Needles first year
36	FI	1992	Dec	FIW	2E		8	1			1,1-1,3	1,1-3,3	0.16-0.37		0.11-1,2	0.04-0.49			
Mosses	Swe	1990-91	Sept, NOV	Sw S	5w		13	1-2	77-190(11) dw	2-4(7)dw			0.7-1,1dw						Mosses

Terrestrial environment  
Matrix Vegetation

	Ref	Country	Year	Seas.	Location	Area code	Site category	Site numb.	obser. num/site	PAH (number) ug/g dw	PCB (number)	HCH alpha	HCH gamma	HCB	PCDF, PCDD (sum)	DDE	DDT	Chlord. gamma	Chlord. alpha	Comments	
Humus	17	Swe	1990-91	Sept	Sw S	5w		5	1-4	0.1-1.5	2.6-28			0.5-3.2							
		Swe	1994	Mar	Sw W	8	forest	1	1	2,5	20										average of 30 samples



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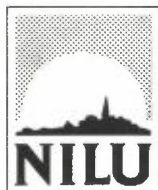
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ABSTRACT The overall goal of the reported project was to improve our knowledge on the inputs, transport, and migration of selected POPs in the Northern environments. A comprehensive steady-state mass balance model, so-called B-POP model, has been developed within the project. This model is based on the fugacity approach and comprises a number of compartments which are assumed to have homogeneous environmental conditions and chemical concentrations. Mass balance equations for all compartments were formulated and solved for the chemical fugacity in each compartment. The B-POP model was then tested to describe the migration of PCBs. It was concluded that the model succeeds in outlining the general picture of PCB behaviour in the Baltic Sea at the end of the 1980s by identifying the major environmental pathways and reservoirs within the system. Considering the limitations inherent in the present version of the model the agreement between modelled and measured concentrations was surprisingly good. In most cases the calculated value was within the range and close to the average of the measured values.			
NORWEGIAN TITLE			
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