# 6 Modelling the environmental fate in the aquatic environment

In the following, environmental fate modelling approach for the aquatic environment is described. As for the terrestrial environment, the discription is generally distinguished into modelling of concentrations in water bodies and aquatic organisms (section 6.1 and 6.2, respectively).

## 6.1 Environmental fate modelling of water bodies

The environmental fate assessment methodology presented covers the environmental media 'soil' and 'water'. While the terrestrial environment is subject of Chapter 5, the medium 'water' will be addressed in the following.

When talking about 'water', in principle one may distinguish it according to its:

- composition: fresh and salty water,
- phases: liquid, solid and gaseous water, and
- macroscopic occurrence, i.e., water bodies at the surface or the subsurface (e.g., groundwater).

Gaseous water or water vapour is mostly part of the atmosphere. Solid water, i.e., ice and snow, is to some extent covered by a new terrestrial compartment (cf. section 5.1.11) noting that the influence of for example sea ice cover on the global environmental fate and snow on ecotoxicology may be important for some substances (e.g., Wania, 2003; Daly and Wania, 2004). As a result, only the distinction according to its composition and its occurrence may be relevant in the following discussion. However, at present only surface freshwater bodies are included in the assessment for reasons given in Chapter 7. In particular, disregarding subsurface water, i.e., ground water, is in line with most of the multimedia models in use today.

### 6.1.1 Compartments distinguished

When differentiating the freshwater environment into compartments, the most common division is into the freshwater body itself and a corresponding sediment (e.g., Mackay and Diamond, 1989; 1991; Mackay and Hickie, 2000; Mackay and Southwood, 1992; Mackay et al., 1992, 1996a; McKone, 1993b; Devillers et al., 1995; Wania and Mackay, 1995; Brandes et al., 1996; Rantio and Paasivirta, 1996; Severinsen et al., 1996; Wania, 1996; Scheringer et al., 2000a; Wania et al., 2000; MacLeod et al., 2001; Prevedouros et al., 2004). For the purpose of modelling non-volatile substances directly released into surface waters, Scheringer and co-workers developed a segmented model for the river Rhine following linear algebra formulations (Beck et al., 2000; Scheringer et al., 2000a). This model distinguishes two water compartments: one containing moving waters and another representing stagnant waters just above the sediment and in shallower regions of the river. Scheringer and co-workers, however, do not state to what extent the introduction of the stagnant water compartment influences the overall results and under which conditions its distinction is recommended. Therefore, the 'ordinary' distinction into water body and bottom sediment is made. However, it shall be noted already here that a distinction of stagnant water portions from flowing waters within the overall freshwater compartment will, nevertheless, be made in this study.

When performing spatially-resolved environmental fate assessments different zones are distinguished whose freshwater and sediment compartments may be allowed to vary in terms of dimensions and properties. There are many examples of such multi-zonal models published in the context of multimedia modelling (e.g., Mackay and Southwood, 1992; Devillers et al., 1995; Wania and Mackay, 1995; Wania, 1996; Rantio and Paasivirta, 1996; Mackay and Hickie, 2000; Wania et al., 2000; Prevedouros et al., 2004). Some of these distinguish between river stretches and lakes (e.g., Mackay and Southwood, 1992; Rantio and Paasivirta, 1996) or just between different parts of lakes (e.g., Wania, 1996; Mackay and Hickie, 2000). The spatial differentiation of the presented methodology allows to distinguish larger lakes from rivers in terms of dimensions and properties as well. This is also motivated by the fact that when computing at the high resolution as shown in Fig. 4-3 about 80 % of the zones contain freshwater compartments that only consist of streams. In the following, the respective dimensions and properties of the aquatic environment are defined.

### 6.1.2 Dimensions of the aquatic compartments

In order to define the dimension of the freshwater and sediment compartments, one needs to know their areas covered and their average depths. The area covered

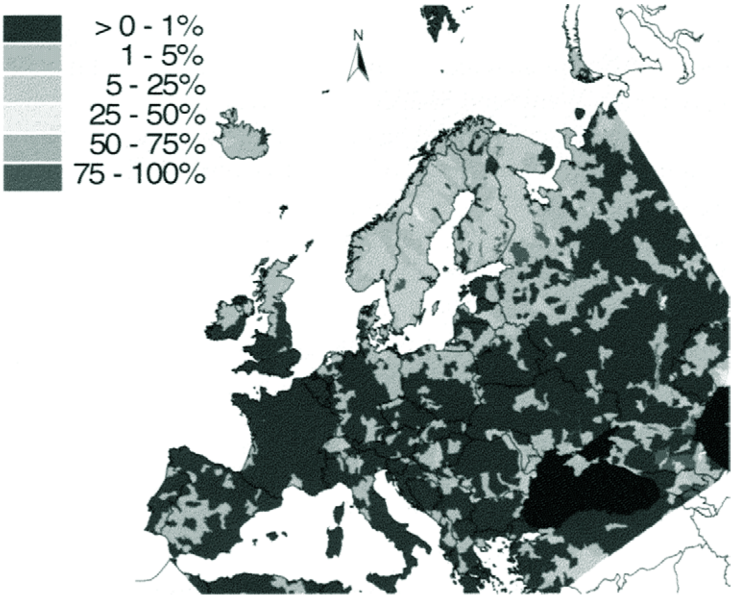

**Fig. 6-1:** Distribution of the predominance of freshwater bodies in the different zones distinguished by WATSON (note that the Black Sea and the Caspian Sea are presently not modelled)

by the freshwater compartment is determined by means of GIS land cover and hydrology-related datasets as presented in section B.4 and shown in Fig. 6-1. Usually, bottom sediments of freshwater bodies are perceived to have the same area as the water body as explicitly stated by Mackay et al. (1992) and Wania et al. (2000). As a result, the area fractions of freshwater bodies (cf. Fig. 6-1) also apply to their bottom sediments.

## Depth of freshwater bodies

The depth of the freshwater compartment is allowed to vary (a) by following the distinction between larger lakes and other freshwater bodies and (b) according to the general observation (in humid areas) that the channel of a river will increase with increasing catchment area (Finlayson and McMahon, 1995, see also sections B.2.1 and B.4). The latter downstream-directed volume increase only applies to those rivers whose drainage basins are further subdivided into several zones, referred to as 'large rivers'.

### Depth of the freshwater sediment

Most of the spatially differentiated multimedia models for which sediment depths are explicitly stated (e.g., Wania and Mackay, 1995; Wania et al., 2000) consider the depth of the active sediment layer to be invariant between zones. The depths of this layer for non-site-specific, generic or evaluative multimedia models range from 0.01 m (Mackay et al., 1992, 1996a) over 0.03 m (Mackay, 1991; Devillers et al., 1995; Brandes et al., 1996; Severinsen et al., 1996; European Commission, 2003b) to 0.05 m (Wania and Mackay, 1995; Scheringer et al., 2000a; Wania et al., 2000). It is noted that when a particular site is investigated zone-specific sediment depths may be available and, thus, used, as done in several studies (Mackay and Diamond, 1989; Mackay and Southwood, 1992; Rantio and Paasivirta, 1996; Wania, 1996; Mackay and Hickie, 2000) spanning a wider range (from 0.005 m for Lake Ontario, Mackay and Diamond, 1989, to 0.1 m for a river delta in a lake, Wania, 1996). For the purpose of the present study, a constant depth of 0.05 m is adopted for the sediment compartment.

## 6.1.3 Definition of the phases of aquatic compartments

Phases distinguished in freshwater bodies are usually water and abiotic suspended matter. Some environmental fate models also include fish (Mackay et al., 1992, 1996a; Mackay and Southwood, 1992; Devillers et al., 1995; Brandes et al., 1996; Severinsen et al., 1996). As noted by Mackay et al. (1992) and Brandes et al. (1996), fish usually play an insignificant role with regard to the overall fate of substances. Like suspended matter, however, these may contain appreciably high concentrations especially of less water-solvable substances. In line with many other multimedia models and chemical risk assessment frameworks (e.g., European Commission, 1996a), fish are not distinguished as phases or even as a compartment in the fate part but at least in the exposure assessment of the presented methodology. Due to the assumed equilibrium between phases within one compartment, a new process can optionally be included in the analysis which describes equilibrium partitioning of substances into freshwater fish that is caught and, thus, removed at a certain rate (cf. section 6.2). Together with other 'uptake-harvest' processes, the removal of caught freshwater fish upon exposure is part of a scenario analysis in section 9.3.3.

The composition of suspended matter as well as that of particles in the sediment varies considerably according to the information provided by several multimedia models (compiled in Table B-9 (Mackay et al., 1992, 1996a; Devillers et al., 1995; Rantio and Paasivirta, 1996) and in Table B-10 (Mackay, 1991; Mackay et al., 1992, 1996a; Devillers et al., 1995; Rantio and Paasivirta, 1996; Mackay and Hickie, 2000), respectively). Assuming that the organic carbon mass makes up 50 % of the organic matter mass (Schnitzer, 1978) with an organic matter density of 1400 kg/m$^3$ (Scheffer and Schachtschabel, 1989), the respective mineral matter mass fractions as well as their densities are obtained. As can be seen from both Tables, the resulting mineral matter densities tend to be lower than that of clay minerals (2200 - 2900 kg/m$^3$) and of quartz (2650 kg/m$^3$, Scheffer and Schachtschabel, 1989) ranging from 1500 to 2800 kg/m$^3$. While the organic carbon content of the sediment particles as given in Table B-10 spans the value range found in other publications (from 3 to 20 vol.-%, Mackay and Diamond, 1989; Mackay and Southwood, 1992; Brandes et al., 1996; Severinsen et al., 1996; Wania, 1996; Wania and Mackay, 1995), the values for the organic carbon content of suspended matter as shown in Table B-9 are located at the higher end of the range from 0.04 (Wania et al., 2000) to 0.4 in the deep water zone of a lake (Wania, 1996) or in the tropic zone (Wania and Mackay, 1995).

Like for other characteristics of the freshwater environment, a distinction of suspended solids in stream water and lakes is made. Generally, the organic matter content of suspended and deposited solids in streams is much smaller than that found in lakes. For this reason, the reported lower bound and upper bound values are used as an orientation for the characterisation of stream and lake solids, respectively (Table 6-1). The particle densities result according to the information on the respective organic carbon contents and the densities for organic and mineral matter. Two different mineral matter densities are used for the computation of suspended solid and sediment solid densities. For suspended matter, the mineral phase's density is set to that of clay minerals, i.e., 2550 kg/m$^3$ (Scheffer and Schachtschabel, 1989), whereas for sediment solids it is set to that of quartz, i.e., 2650 kg/m$^3$ (Scheffer and Schachtschabel, 1989).

Based on sediment yield data provided by Milliman and Syvitski (1992), the volume fraction of suspended solids in streams is set to $1 \cdot 10^{-3}$ vol.-% which appears to be applicable to non-Alpine mountainous, upland and lowland European rivers. As a result, the concentration of suspended matter in freshwater streams is set to 0.02469 kg/m$^3$. In the absence of more specific data, a value for the volume fraction of suspended matter of $1 \cdot 10^{-4}$ vol.-% is assumed for lakes which is of the same order of magnitude like the bulk of the reported values in the multimedia literature (Mackay et al., 1992, 1996a; Wania and Mackay, 1995; Rantio and Paasivirta, 1996; Mackay and Hickie, 2000).

The corresponding value for the volume fraction of solids in sediments also needs to be specified. Reported values range from 0.1 (Mackay and Hickie, 2000) to 0.4 (Mackay, 1991; Rantio and Paasivirta, 1996).[14] As the sediment compart-

**Table 6-1:** Characteristics of solids in the freshwater environment as used in the presented methodology

| Property | Particle type | Stream | Lake |
|---|---|---|---|
| Volume fraction of ... | suspended matter | $1 \cdot 10^{-5}$ [a] | $1 \cdot 10^{-6}$ [b] |
| | sediment solids | 0.2 | 0.2 |
| Mass fraction of organic carbon in ... | suspended matter | 0.02[c] | 0.4[d] |
| | sediment solids | 0.005[e] | 0.3[d] |
| Density of solids [kg per $m^3$][f] of ... | suspended matter | 2469 | 1539 |
| | sediment solids | 2627 | 1726 |

a.Selection of the value is guided by data in Milliman and Syvitski (1992).

b.Same order of magnitude like the bulk of the reported values in the multimedia literature (Mackay et al., 1992, 1996a; Wania and Mackay, 1995; Rantio and Paasivirta, 1996; Mackay and Hickie, 2000).

c.Half the value as used in Wania et al. (2000) assumed for 'pure' stream conditions.

d.In the deep water zone of a lake (Wania, 1996).

e.Although the smallest value found is 0.02 for the northern-boreal and the polar zones in Wania and Mackay (1995) an even smaller value is adopted for 'pure' stream sediments which usually have an organic matter content below 1 % (Scheffer and Schachtschabel, 1989) due to water erosion and oxic conditions.

f.Assumptions: half of the organic matter (OM) mass consists of organic carbon (Schnitzer, 1978) and taking the complement as mineral matter (MM); densities: 1400 $kg_{OM}/m^3$, 2550 $kg_{clay\ minerals}/m^3$ (for suspended solids) and 2650 $kg_{MM}/m^3$ (for sediment solids, Scheffer and Schachtschabel, 1989).

ments only comprise the active part of the overall sediments which is not as consolidated as the parts below, a value of 20 vol.-% is adopted here which corresponds to the median and the average of the volume fractions of sediments that consist of solids reported in the multimedia modelling literature.

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[14] Note that two values are used by Rantio and Paasivirta (1996): a value of 0.6 for sediment porosity corresponding to 40 vol.-% solids and a value of 0.05 for all segments concerning 'volume fraction of sediment solids'. It is unclear to the author how these match.

### 6.1.4 Processes considered for the aquatic environment

The set of processes included in the modelling exercise of multimedia models does not vary substantially between models for the aquatic environment (Table 6-2). In the presented methodology, the following processes that are not related to chemical transformation, speciation or radioactive decay are considered:

- water advection,
- sediment deposition,
- resuspension,
- sediment-water diffusive exchange, and
- sediment burial

noting that bioturbation beside resuspension may also be important for a remobilisation of heavy metals and other substances from sediments (Wania et al., 2000; Zoumis et al., 2001). Their formulation is given in Table 6-3 and further discussed in separate sub-sections to A.3 also stated in the Table. Note that degradation and radioactive decay may be defined for the aquatic environment analogously as for the terrestrial environment described in Table 5-5.

All of these processes are part of mass balances for suspended or deposited particulates or for water except for the diffusive exchange, a process which, however, may involve movement of colloidal matter as well (Mackay, 1991). The diffusive exchange between water bodies and sediments is included in the developed environmental fate model in an 'ordinary' way (cf. sections A.3.14 and A.3.15). The mass balance on water is described next while the components of the particle mass balance in the freshwater environment will be discussed in more detail in section 6.1.5.

#### Mass balance on water

For the mass balance on water, a harmonized set of Geographic Information System (GIS) data is used. Information on precipitation rates have been taken from New et al. (1999). Data on runoff and ground water recharge which are used to distinguish quickflow from baseflow waters have been provided by Döll et al. (2003). The quickflow waters drive the process of 'overland flow' (section A.3.4) whereas baseflow waters percolate through soil to the subsurface (section A.3.6) before exfiltrating into surface waters again. Note that except for few areas, for instance, in Spain and Africa, the general assumption of exfiltrating water flows holds.

According to the general assumptions as formulated in section 2.3, use shall be made of long-term average data. The data sources used provide average values for the period of 1961-1990 fulfilling this requirement. Due to the fact that

**Table 6-2:** Non-exhaustive overview about processes considered for the freshwater compartment by various multimedia models (note: chemical transformations are not listed)

| Model / reference | Air-water exchange | Water advection | Sediment deposition | Resuspension | Sediment-water diffusion | Sediment burial | Bioturbation |
|---|---|---|---|---|---|---|---|
| QWASI (Mackay and Diamond, 1989) | x | x | x | x | x | x | |
| EQC (Mackay et al., 1996a)[a] | x | x | x | x | x | x | |
| POPCYCLING-Baltic (Wania et al., 2000) | x | x | x | x | x | x | x |
| CHEMFRANCE (Devillers et al., 1995) | x | x | x | x | x | x | |
| Danish model (Severinsen et al., 1996) | x | x | x | x | x | | |
| River Rhine model (Scheringer et al., 2000a) | n/a[b] | x | x[c] | x | x[d] | | |
| SimpleBox 2.0 (Brandes et al., 1996) | x | x | x | x | x | x | |
| CalTOX (McKone, 1993b) | x | x | x | x | x | x | |

[a] A model with at least the same water-related processes, rates and compartment properties has previously been documented in Mackay et al. (1992).

[b] Not applicable for the substances investigated.

[c] Only affecting suspended particulates in the stagnant water compartment.

[d] There is a diffusive exchange between the moving and stagnant water compartment as well as between the stagnant water and sediment compartment.

environmental parameters are held constant during the present assessment ('quasi-dynamic' modelling), no storage change for example in soils is taken into account. The water balance is, thus, described at steady-state.

**Table 6-3:** Process formulations for the aquatic environment as used in the present assessment

| Name | Refer to section ... for more details | Formulation[a] | |
|---|---|---|---|
| Water body and sediment compartment | | | |
| Discharge | A.3.9 (p. 409) | $k_{w,\, z1 - z2,\, \text{discharge}}(z) = Q_{\text{discharge}}(z)$ | (6-1) |
| Water circulation in large lakes | A.3.10 (p. 410) | $k_{w,\, z_{\text{down}} - z_{\text{up,1}},\, \text{lake ciculation}}(z, w) = Q_{\text{discharge}}(z_{\text{down}}, w) \cdot fr\_Q_{\text{lake circulation}}(w)$ | (6-2) |
| Sedimentation in freshwater compartments | A.3.11 (p. 411) | $k_{w - ws,\, \text{sedimentation, pH}\vert\text{pH inv}\vert C_{\text{org}}}(p, w, z) = \dfrac{\text{A}(z) \cdot fr\_A(w, z)}{ED_{\text{bulk/aqueous, pH}\vert\text{pH inv}\vert C_{\text{org}}}(p, w, z)} \cdot v_{\text{sedimentation, derived}}$ | (6-3) |
| Resuspension of bottom sediment matter | A.3.12 (p. 412) | $k_{ws\text{-w, resuspension}}(ws, z) = \text{A}(z) \cdot fr\_A(w, z) \cdot v_{\text{resuspension, derived}}$ | (6-4) |
| Sediment burial | A.3.13 (p. 414) | $k_{ws,\, \text{burial, pH}\vert\text{pH}_{\text{inv}}}(p, ws, z) = \dfrac{\rho_{\text{solid phase}}(ws, z) \cdot Ksw(p, pH(ws, z))}{ED_{\text{bulk/aqueous, pH}\vert\text{pH}_{\text{inv}}}(p, ws, z)} \cdot \text{A}(z) \cdot fr\_A(w, z) \cdot v_{\text{burial, derived}}$ | (6-5) |
| Diffusion from sediment to water body | A.3.15 (p. 418) | $k_{ws - w,\, \text{diffusion, pH}\vert\text{pH inv}\vert C_{\text{org}}}(p, ws, z) = \text{A}(z) \cdot fr\_A(w, z) \cdot \dfrac{v_{\text{diffusion}}(ws)}{ED_{\text{bulk/aqueous, pH}\vert\text{pH inv}\vert C_{\text{org}}}(p, ws, z)}$ | (6-6) |

**Table 6-3:** Process formulations for the aquatic environment as used in the present assessment

| Name | Refer to section ... for more details | Formulation[a] | |
|---|---|---|---|
| Diffusion from water body to sediment | A.3.14 (p. 416) | $k_{w\text{-}ws,\ \mathrm{diffusion,\ pH\vert pH_{inv}}}(p, w, z) = \frac{\mathrm{A}(z)\cdot fr\_A(w, z)}{Ksw(p, pH(w, z)\vert pH(w))}\cdot\frac{1}{\rho_{\mathrm{suspended\ matter}}(w, z)}\cdot\frac{v_{\mathrm{diffusion}}(w)}{ED_{\mathrm{bulk/solid,\ pH\vert pH_{inv}}}(p, w, z)}$ | (6-7) |
| Freshwater fish | | | |
| Uptake by and catch of freshwater fish for non-volatile substances | A.3.8 (p. 404) | $k_{\mathrm{w,\ uptake+catch\ fish}} = \frac{BCF\_V/fw_{\mathrm{fish/water}}(p, r, e)\cdot P(r, n)}{Ksw\cdot ED_{\mathrm{bulk/solid}}\cdot\rho_{\mathrm{suspendend\ matter}}(w, z)}$ | (6-8) |

a. $A$: area of the zone [$m^2$]; $BCF\_V/fw$: bioconcentration factor [$m^3$/kg]; $d$: depth of a compartment [m]; $ED$: equilibrium distribution coefficient [-]; $fr\_A$: area fraction of a compartment within a zone [-]; $fr\_Q$: fraction of the discharge [-]; $fr\_v$: fraction of a process velocity [-]; $k$: process rate as used in the coefficient matrix [$m^3$/s]; $K$: solid-water partitioning coefficient [$m^3$/kg]; $Q$: discharge [$m^3$/s]; $r$: process rate [1/s]; $\rho$: density [kg/$m^3$]; $v$: process velocity [m/s] (for derived parameters: see text); symbols in parentheses denote a parameter's dependency on the exposure assessment framework ('$e$'), administrative unit ('$n$'), pollutant ('$p$'), receptor (or fish, '$r$'), compartment ('$w$' freshwater body, '$ws$': corresponding sediment) and/or the zone ('$z$')

The processing of the data related to the water balance is described in section B.5.2. The process 'discharge' or water advection is formulated based on runoff information (cf. Table 6-3 and section A.3.9).

### 6.1.5 Innovations as regards aquatic compartments

There are several innovations introduced as regards the modelling of the aquatic environment. These are

- the distinction of rivers from lakes in terms of the dynamics of the particles,
- the introduction of lake water circulation in those instances in which a distinction of large lakes is made; these consist of several zones that entirely consist of water.

These shall be presented in the following.

### Mass balance on particulates in the freshwater environment

Mass balances focusing on suspended organic particulates are explicitly addressed for example in Mackay et al. (1992), Brandes et al. (1996) and Wania et al. (2000) to varying degrees of sophistication. For the more sophisticated mass balances, data requirements are higher and include information for instance on the primary productivity of a water body, mineralisation rates, explicit information on organic matter entering the aquatic environment from soils, or inputs from sewers. Likewise a mass balance for suspended mineral matter may be formulated. This is addressed by all consulted multimedia model descriptions according to the organic carbon mass fractions reported especially for suspended matter. When assuming that 50 weight-% of organic matter consists of organic carbon (Schnitzer, 1978), the organic carbon mass fraction of suspended matter would need to amount to 50 weight-% for the suspended particles to entirely consist of organic material. All of the reported values found in the multimedia modelling literature are below this value implying that all models consider mineral particles to be present (cf. section B.5.4). SimpleBox 2.0 even explicitly considers the overall erosion rate from soil to water although suspended matter is understood to consist of non-living organic matter only (Brandes et al., 1996). For data availability reasons, the simpler approach for suspended particulates as presented for example by Mackay et al. (1992) is followed in the present study.

The rates of the processes 'sediment deposition', 'resuspension' and 'sediment burial' of the finer particles to which most of the substances will sorb strongly depend on the flow pattern or turbulence of the water body modelled, beside particle properties and fluid density (Shen and Julien, 1993). The 'fluid' or water density is assumed to be 1000 kg/m$^3$ in the freshwater environment. Although noting that the properties even of the 'finer particulates' active in contaminant transport will vary considerably, no differentiation for example into mineral and organic particles and/or colloids or 'flocs' (McCutcheon et al., 1993; Nicholas and Walling, 1996; Droppo et al., 1998) will be made here in terms of process velocities. Nevertheless, the distinction into larger lakes and other freshwater bodies as well as different zones within larger catchments (sections 4.3 and 6.1.2) allows a differentiated approach towards the determination of these process rates in the spatially-resolved impact assessment methodology presented.

In line with Scheringer et al. (2000a) and Beck et al. (2000), sedimentation is only allowed to take place in stagnant waters, however, without distinguishing these as separate compartments (cf. section B.5.4). In the absence of more specific information, it is assumed that the large lakes distinguished (cf. section B.2.1) entirely consist of stagnant waters. In contrast, only 5 % of the in-stream water volumes are assumed to allow for sedimentation due to their low amount of stagnant waters. This volume fraction is substantially lower than the 25 % that have been assumed by Scheringer et al. (2000a). However, these authors have not distinguished lakes explicitly. Also, their value appears to be rather high.

Another distinction is made in that sedimentation in freshwater bodies of zones that pour directly into the sea ('river mouth') is increased due to slower flow velocities, also called aggradation zone or "area of deposition" in an idealised fluvial system (Schumm, 1977). By default, the volume share of stagnant water is set to 21 % in these zones, provided they are part of a larger drainage basin. This is guided by the idea that a smaller percentage of the deposition rate is resuspended in these areas than in other 'pure' stream zones.

The process rates relevant for the particle mass balance in surface freshwater and their relationships are given in Table 6-4 based on a literature review (cf. section B.5.4), distinguished according to pure river and pure lake situations, respectively. Large lakes are considered not to contain (significantly) flowing waters which is why the values for all of the process velocities under pure lake conditions are directly given in Table 6-4. For mixtures of stagnant and flowing conditions, the overall applicable process rates are calculated according to the Eqs. (B-17), (B-22) and (B-27) of the general form:

$$\begin{aligned} v_{\text{process, total}} = {} & \mathit{Fraction}_{\text{stagnant water}} \cdot v_{\text{process, lake}} + \\ & (1 - \mathit{Fraction}_{\text{stagnant water}}) \cdot v_{\text{process, stream}} \end{aligned} \quad (6\text{-}9)$$

where

v : velocity of particles while undergoing either of the processes sedimentation, resuspension, or burial in the freshwater environment [m per s]

Fraction : volume or area fraction of stagnant waters [-].

**Table 6-4:** Particle mass balance for surface freshwater assumed in this study differentiated into a pure river and a pure lake situation

| Characteristics | Pure river | Pure lake |
|---|---|---|
| Sediment deposition rate [m/s] | $0.95 \cdot 0$ [a]$+ 0.05 \cdot 3.50 \cdot 10^{-6}$ [b]<br>$= 1.75 \cdot 10^{-7}$ | $3.50 \cdot 10^{-6}$ [b] |
| Resuspension rate [m/s] | $0.95 \cdot 1.143 \cdot 10^{-7}$ [c]$+ 0.05 \cdot 1.156 \cdot 10^{-6}$ [d]<br>$= 1.66 \cdot 10^{-7}$ | $1.156 \cdot 10^{-6}$ [d] |
| Burial rate [m/s] | $0.95 \cdot -1.143 \cdot 10^{-7}$ [e]$+ 0.05 \cdot 2.35 \cdot 10^{-6}$ [f]<br>$= 8.76 \cdot 10^{-9}$ | $2.35 \cdot 10^{-6}$ [f] |
| Ratio resuspension / sedimentation | 95.0 % [g] | 33.0 % |
| Ratio burial / sedimentation | 5.0 % | 67.0 % [h] |
| Ratio remainder / sedimentation | 0.0 % [i] | 0.0 % [j] |

a.As calculated for a pure in-stream sediment deposition rate by Eq. (B-20).

b.As calculated for a pure in-lake sediment deposition rate by Eq. (B-19).

c.As calculated for a pure in-stream resuspension rate by Eq. (B-26).

d.As calculated for a pure in-lake resuspension rate by Eq. (B-24).

e.As calculated for a pure in-stream burial rate by Eq. (B-31); see text for the explanation of the negative value.

f.As calculated for a pure in-lake burial rate by Eq. (B-29).

g.The highest reported value is 85 % for the shore line of a lake (Wania, 1996); due to the even higher flow rate in streams a higher value has been adopted; note that the 'effective' sedimentation is higher river mouth situations of larger rivers (Schumm, 1977) and in lakes.

h.Value selection guided by the burial at the deep water zone of a lake (Wania, 1996).

i.In case mineralization was to be considered, the value of 14 % as used in the POPCYCLING-Baltic model would be suggested to be used for organic matter in rivers (Wania et al., 2000).

j.In case mineralization was to be considered, the value of 7 % as used for the southern segment of Lac Saint Louis would be suggested to be used for organic matter in lakes (Mackay and Hickie, 2000), owing to the smaller biological activity and lower temperatures throughout the course of a year.

In section B.5.4, the mass balance for particles in the freshwater environment is discussed in more detail. In short, a value of $2.35 \cdot 10^{-6}$ metres per second for the net sedimentation (or burial) rate is identified to be appropriate for the full removal of all suspended particles from the water column of large lakes which are assumed to be 74 metres deep, disregarding the removal due to comparatively slow water advection. Following Stokes' law, this value falls in the range of velocities obtained for temperatures between 5 and 10°C for a spheric particle with a radius of 1 $\mu$m (i.e., of the clay fraction) and a density of 1539 kg/m$^3$ (assumed for suspended particles in lakes, cf. Table B-11). The settling velocities range from $2.21 \cdot 10^{-6}$ and $2.56 \cdot 10^{-6}$ metres per second for a temperature of 5 and 10°C and a corresponding (absolute) water viscosity of $1.518 \cdot 10^{-3}$ and $1.307 \cdot 10^{-3}$ kg/m/s (McCutcheon et al., 1993), respectively. Thus, the value appears to be reasonable.

For rivers, the same settling velocity is assumed under still-water conditions. However, it is allowed to be active only in 5 % of the water volume that are assumed to be stagnant as mentioned above. Furthermore, 95 % of the settled particles in streams are assumed to undergo resuspension so that the overall net sedimentation rate for rivers results to be $8.76 \cdot 10^{-9}$ metres per second.

Note that the value obtained for the pure in-stream sediment burial rate is negative. All of the velocities derived for streams (not only the velocity for burial, but also for sedimentation and resuspension (Eqs. (B-31), (B-20) and (B-26), respectively)) should, however, be regarded as hypothetical as these are not directly used in the process formulations (section A.3). These values are rather obtained in order to fulfil the following requirements:

- in order to provide a generally applicable computation of the overall process rates, the approach as given by Eq. (6-9) is followed which distinguishes between stagnant and non-stagnant domains within any freshwater compartment. Due to the heterogeneity of freshwater compartments ranging from pure streams over mixtures of streams and lakes to pure lakes, this methodological approach is adopted, and
- the overall in-stream net sedimentation rate, i.e., resulting sediment deposition rate minus resuspension rate as given in Table 6-4, shall amount to 5 % of the lake net sedimentation rate.

As a result, especially the negative value for the burial rate is hypothetical and is only used in order to yield a (non-negative) total process velocity. The hypothetical values for pure moving water conditions are, therefore, considered justified and scientifically defendable as long as they are consistently derived and used. As stated above, any water body is assumed to at least contain 5 vol.-% of still waters which assures that no negative end-values result.

As can be seen from Table 6-4, no mineralization is allowed to occur. In principle, one would need to distinguish between the mineral and organic phases of the respective sediment and suspended particles when taking account of mineralization. This is not done at present which is in line with Devillers et al. (1995), Severinsen et al. (1996) and Wania (1996).

## Water circulation in large lakes

In contrast to the process 'discharge' which flows from an upstream zone either into a further downstream one within a catchment or into coastal waters, i.e., out of the present model's scope, there is a new process introduced into the environmental fate assessment, termed 'water circulation in large lakes' (cf. Table 6-3). This has been deemed necessary due to the distinction of larger lakes from other water bodies for reasons detailed in the following.

Some of these larger lakes are fully contained within the zones identified by the HYDRO1k basin dataset (EROS Data Center, 1996). These are only considered in terms of a larger depth. However, many of these lakes extend over different zones. Their spatial differentiation according to the HYDRO1k basin dataset resembles a rather unnatural or non-intuitive herringbone that cuts the lakes into bands (Fig. 6-2). This is due to the derivation of sub-basins according to elevation data while lakes do not show marked slopes. All of these lake portions which are connected by the downstream flow of water constitute separate zones according to the presented methodology.

If only allowing downstream-directed discharge to occur between different lake portions, there would be no water exchange between a 'downstream' and an adjacent 'upstream' lake zone due to the cascading organisation of flows between zones according to the Pfafstetter code (cf. Fig. 4-4). Therefore, water advection is allowed to also move waters 'upstream' following this code.

The process 'water circulation in large lakes' is formulated as the reverse process of discharge so that one 'downstream' zone may pour a certain amount of water into maximally two 'upstream' lake zones. The amount of water involved is set to a share of the discharge flowing out of the 'downstream' lake zone as described in section A.3.10. At the same time the same amount of water flows back in order not to violate the water mass balance. The share of discharge undergoing this process is presently set to 100 %. The influence of this process on the exposure and impact results will be analysed in a sensitivity analysis in section 9.3.3.

## 6.2 Environmental fate modelling for aquatic organisms

WATSON at present does not include the marine environment which is why only freshwater organisms can be considered. Similar to vegetal produce (see section 5.2), freshwater fish are neither modelled as separate compartments nor constitute phases within compartments in the environmental fate model. Rather, they may be considered by means of a combined transfer-harvest process in which freshwater fish are assumed to be in equilibrium with the freshwater compartment and removed by means of catching. Considering harvest removal of substances in a multimedia modelling context has hardly been done (cf. section 5.2.3). These

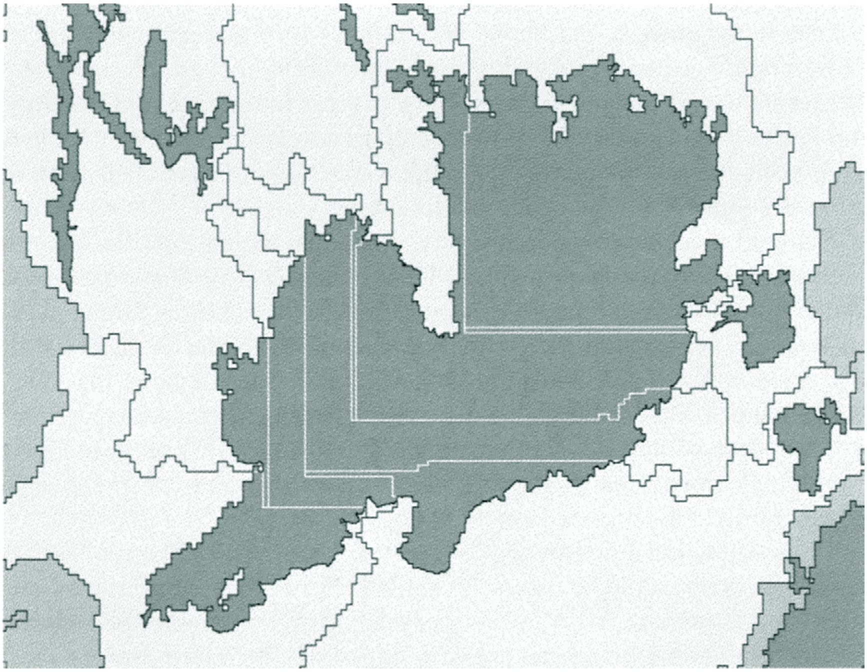

**Fig. 6-2:** Lake Vänern in southern Sweden as an example of a sub-division of larger lakes according to the spatial differentiation as provided by HYDRO1k basin dataset (EROS Data Center, 1996; dark grey: lakes; light grey: the Götälv catchment; water grossly flowing from north-east to south-west; lakes fully contained in one zone are also shown)

removal processes only addressed the harvest of terrestrial plants. The inclusion of a corresponding process for fish is, thus, novel in the realm of multimedia modelling. The respective equation is given in Table 6-3 (Eq. (6-8), p. 143).

In any case, human exposure towards freshwater fish consumption is part of the exposure assessment to be presented in the next Chapter (cf. section A.7.12).