FLUXES, POOLS, AND TURNOVER OF MERCURY IN SWEDISH FOREST LAKES

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ABSTRACT. In boreal forest lakes, high Hg concentrations in fish are common, even in remote areas. Data from recent Swedish surveys were synthesized to assess sources, sinks, fluxes and turnover rates of Hg in headwater lakes of different type, to determine and quantify controlling factors and their relative influence, and to discuss implications for the contamination of fish and potential remedial actions. In humic lakes, annual input of Hg is dominated by the transport from forest soils, whereas in clearwater lakes, direct deposition on the lake surface is often more important. Direct deposition is more likely to dominate during the growing season when the water discharge in forest streams is low. Sedimentation of Hg is largely controlled by the water residence time and is related to water quality. In clearwater lakes, most of the Hg load is deposited in the sediment, which is in contrast to humic lakes, and which partly explains the susceptibility of headwater lakes to atmospheric Hg pollution. The fish community contains about half or more of the methylmercury pool in the water column, and internal recycling in the food web may be important. However, only a minor fraction of the total annual Hg load to lakes is channeled through the fish community. As this allows large variations in the ecological fractionation and bioavailability of Hg, differences in Hg fluxes can only partly explain the wide range of Hg concentrations found in fish from different lakes. The annual Hg load to headwater lakes exceeds the total Hg pool in the water column by a factor of up to 20. Single remedial measures aiming at the removal of Hg from the water column are therefore likely to have little success. Local actions should aim primarily at reducing the bioavailability of the Hg entering the lakes and need to be repeated continuously.

1. Introduction

In thousands of Swedish lakes with no obvious source of contamination, Hg concentrations in fish exceed health advisory limits (0.5 to 1.0 mg kg^{-1}, wet weight), and the situation is not improving despite a dramatic reduction of atmospheric Hg emissions (Håkanson *et al.*, 1988; Lindqvist *et al.*, 1991). Hg concentrations in the water column are very low, typically on the order of a few µg m^{-3} (Meili *et al.*, 1991). It has been proposed that the high Hg concentrations in fish may occur even at low Hg loading to lakes as a result of an efficient recycling of methylmercury in lacustrine food webs (Verta, 1984). Consequently, the turnover rate of Hg in lakes may be of crucial importance both for the Hg concentrations in biota and for the selection of appropriate measures to counteract high Hg levels in natural fish populations.

Hg concentrations in fish are typically highest in lakes with low ionic strength, low pH, low productivity and high water color (cf. Lindqvist *et al.*, 1991, and Verta, 1990, for recent reviews). Many

of these lakes are situated in boreal areas where shallow podzolic soils on igneous bedrock prevail, and where the vegetation is dominated by coniferous forest and mires. The lakes which are most susceptible to atmospheric pollution with acidifying substances, heavy metals and radionuclides are headwater lakes, where the water supplied has a short average contact time with soils before entering the lake.

Although many processes in the biogeochemical behavior of Hg are not understood in detail, data from recent surveys (summarized in Lindqvist *et al.*, 1991) allow the estimation of environmental Hg fluxes with sufficient accuracy to assess the importance of major Hg pathways in lakes. Syntheses of this type are useful to assess sources, fluxes and turnover rates of Hg in different lake types, to determine and quantify controlling factors and their relative influence, as well as to discuss implications for the contamination of fish and potential preventive or remedial actions. Hg contamination of lacustrine fish in Sweden is most severe in southern and central regions of the country (Håkanson *et al.*, 1988; Chapter 12 in Lindqvist *et al.*, 1991). The calculations in this paper are focused on these areas, but many conclusions may be valid for other boreal regions of similar character.

2. Sources and sinks of mercury in remote lakes

In lakes without any known direct discharge of Hg to the lake or its tributaries, Hg can be supplied either by direct deposition on the lake surface, via surface runoff from the catchment, or via groundwater inflow. As Hg concentrations in clear groundwaters are extremely low (Aastrup *et al.*, 1991; Chapter 8 in Lindqvist *et al.*, 1991), and as the inflow of clear groundwater is of minor importance in most Swedish forest lakes as a result of shallow soils, this pathway is not treated separately here.

Direct deposition can occur through precipitation or through dry deposition of gaseous or particulate Hg. Few measurements of dry deposition of Hg on water surfaces are available, but they indicate that the fluxes are lower than wet deposition where dense vegetation such as forest is absent (Chapters 6 and 7 in Lindqvist *et al.*, 1991; cf. Meili, 1991). The Hg input to forest lakes from their catchment is of significant magnitude due to the strong association of Hg with humic matter (Iverfeldt and Johansson, 1988; Johansson and Iverfeldt, 1990; Mierle, 1990; Johansson *et al.* 1991). Most of this loading is a result of atmospheric deposition of Hg onto the soils of the catchment and subsequent transport to forest streams, whereas the contribution from bedrock erosion generally is very small (Chapter 8 in Lindqvist *et al.*, 1991).

Potential sinks of Hg in drainage lakes are the outflow, the sediment and the atmosphere. Little is known about the quantitative impact of Hg volatilization from water bodies; it is here, similar to dry deposition, assumed to be of minor importance on an annual basis in the type of drainage lake lakes considered. Outflow and sedimentation of Hg on the other hand are both of potential importance.

3. Fluxes of mercury in forest lakes

The amount and the relative importance of direct (wet) deposition on lake surfaces and transport to lakes from their tributaries varies both regionally and seasonally and is dependent on climate and lake hydrography (Meili, 1991; Meili *et al.*, 1991).

Lakes in boreal areas are often small and shallow drainage lakes with a comparatively large catchment area. As a result, water residence times are typically on the order of a few months to a few years (data base from Håkanson *et al.*, 1988). Many headwater lakes are stained by humic substances, which is related partly to the surficial and rapid flow of water in the shallow organic soils surrounding these lakes, partly to the short water residence time in the lake basin, counteracting sedimentation of humic matter (Rasmussen *et al.*, 1989).

Hg input to forest lakes from the catchment is strongly dependent on the transport of humic matter (Iverfeldt and Johansson, 1988; Johansson and Iverfeldt, 1990). In surface runoff from forested watersheds, the concentrations of both Hg and organic matter vary spatially and temporally, but the ratio of

Hg/C in remote areas appears to be similar in many areas, with annual mean values around 0.25 μg g^{-1}, at concentrations of organic C on the order of 20 and 15 g m^{-3} in southern and central Sweden, respectively (Meili, 1991). The annual surface runoff is about 0.4 m^3 m^{-2} yr^{-1} both in the south and in the north, although varying along an east-west gradient (Ahlmann, 1976, Falkenmark, 1979). The annual runoff export of Hg from typical forest areas can be calculated as a product of the values above.

Direct deposition of Hg on lake surfaces can be estimated from the Hg concentration in precipitation and the annual amount of precipitation (Iverfeldt, 1991; Chapter 6 in Lindqvist *et al.*, 1991). The strong correlation of the annual mean concentration of Hg and H$^+$ in precipitation also allows to estimate Hg deposition in remote areas from the deposition of acid (Meili, 1991). Although temporal and spatial variations probably are considerable, typical Hg deposition values can be assigned to remote areas in different regions, about 16 μg m^{-2} yr^{-1} in southern Sweden and 9 μg m^{-2} yr^{-1} in central Sweden.

The outflow of Hg from lakes is a product of the Hg concentrations in lake waters and the water flux. Horizontal Hg fluxes through drainage lakes are dominated by comparatively short periods of high water flow usually occurring in spring (snow melt) and autumn (Meili *et al.*, 1991; cf. Iverfeldt and Johansson, 1988; Mierle, 1990). During these periods, Hg concentrations in lakes are elevated above the annual average, which has to be accounted for when calculating annual fluxes, especially in lakes with a short water residence time. Simulations of different conditions within potential ranges resulted in values corresponding to 20% and 80% of the total load in clearwater and brownwater lakes, respectively.

Hg concentrations in forest lake waters are highly correlated to the concentration of humic substances (Meili *et al.*, 1991), which is a function of a lake's hydrology and thus dependent on its mean depth, the size of its catchment area and the annual precipitation and evaporation (Rasmussen *et al.*, 1989). In Swedish headwater forest lakes, the water color typically ranges from 20 to 200 g Pt m^{-3}, the mean depth from 1 to 6 m, and the ratio of catchment and lake area from 4 to 40 (SNV, 1986; data base from Håkanson *et al.*, 1988). Hg concentrations are about 2-3 μg m^{-3} in clearwater lakes and about 4 to 6 μg m^{-3} in brownwater lakes of southern and central Sweden (Meili *et al.*, 1991).

Sedimentation rates of Hg were calculated as the difference between volume-weighted outflow and total load. Simulations of different conditions typically resulted in values corresponding to 80% and 20% of the total load in clearwater and brownwater lakes, respectively.

4. Turnover of mercury in forest lakes — a mass balance study

In order to assess the Hg turnover in the most susceptible lakes in the areas where Hg contamination in remote lakes is most severe, calculations were focused on a set of model lakes representing the range of typical headwater lakes in the boreal forest regions of southern and central Sweden during a hydrologically representative year. Calculations were performed separately for a clearwater lake (characterized by a long water residence time, a deep water column and a little catchment area) and a brownwater lake (characterized by a short water residence time, a shallow water column and a large catchment area). The model is further based on the following conditions, part of which have been discussed above:

- the catchments are dominated by igneous bedrock and covered with coniferous forest on podzolic soils,
- no lakes or mires are situated upstream of the model lakes,
- the human impact on the lakes and their catchment is minimal,
- no major atmospheric point sources of Hg occur(red) within 100 km,
- wet deposition of Hg is similar over forests and lake surfaces,
- dry deposition on and reemission from lake surfaces is insignificant,
- the Hg/C ratio in runoff water is constant in the whole region.

This simplified approach was deliberately chosen to assess the relative importance of Hg transport across a typical land-water interface in different types of lakes. The model headwater lakes may thus represent only part of the lakes in the boreal forest region. For example, the runoff load of Hg is overestimated if large

lakes are located in the drainage area, but underestimated in areas where the soils are contaminated by historical point sources, which appears to be the case for large areas of Sweden (Håkanson *et al.*, 1990; Johansson *et al.*, 1991; Chapter 4 and 8.1 in Lindqvist *et al.*, 1991). Hydrological and catchment parameters were assigned regionally representative values. All natural variability of e.g. water fluxes, water chemistry, lake productivity, geology, soil and vegetation characteristics is therefore not accounted for in the calculations, but potential ranges of some values are given. Fluxes are volume-weighted, and pools are averaged over time.

The total Hg load to Swedish forest lakes is about 10 to 40 μg m^{-2} yr^{-1} in clearwater lakes and 50 to 120 μg m^{-2} yr^{-1} in brownwater lakes (Table I; Fig. 1). The large difference in loading between these lake types may be one of the main causes for the elevated Hg levels in fish typically found in humic lakes (McMurtry *et al.*, 1989; Verta, 1990; Chapter 12 in Lindqvist *et al.*, 1991).
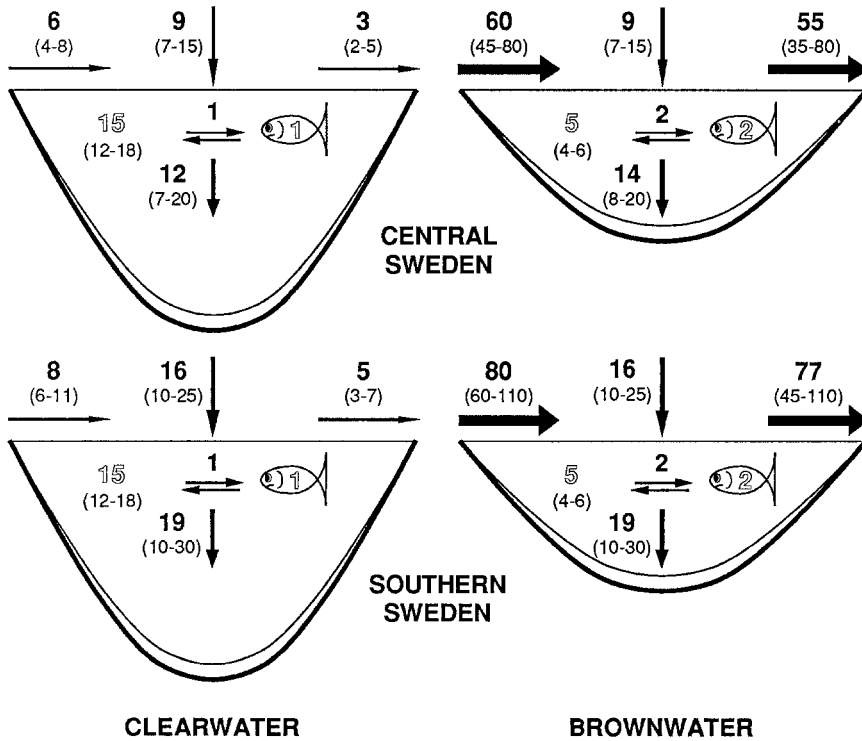

Figure 1. Mass balance study for a set of model lakes covering the typical range of remote headwater lakes in the boreal forest region of southern and central Sweden (see Table I for lake characteristics). Hg fluxes (μg m^{-2} yr^{-1}, bold numbers) and pools (μg m^{-2}, outlined numbers) are given per unit lake surface for a hydrologically representative year. Model values are shown together with typical ranges based on the spatial variability of lake hydrology and Hg concentrations.

Table I. Hg fluxes and pools in a set of model lakes covering the range of typical headwater lakes in the study regions of central and south Sweden during a hydrologically representative year. Background data are given as typical values, Hg data as typical ranges of annual means based on the spatial variability of lake hydrology and Hg concentrations.

REGIONAL FACTORS	Central Sweden		Southern Sweden	
Annual precipitation (m yr^{-1})	0.7		0.8	
Hg concentration in precipitation (μg m^{-3})	13		20	
Annual area-specific runoff water discharge (m yr^{-1})	0.4		0.4	
Organic carbon in forest runoff (g m^{-3})	15		20	
Hg/C ratio in runoff water (μg g^{-1})	0.25		0.25	
MODEL LAKE CHARACTERISTICS	**Clearwater lake**		**Brownwater lake**	
Lake water color (g Pt m^{-3})	20		200	
Mean depth (m)	6		1	
Ratio of catchment area / lake surface area	4		40	
Mean water residence time (mo)	36		1	
Sedimentation (% of Hg load)	80		20	
Hg CONCENTRATION AND POOL	**Clearwater lake**		**Brownwater lake**	
Mean Hg concentration in lake water (μg m^{-3})	2-3		4-6	
Hg pool in lake water (μg m^{-2})	12-18		4-6	
Hg FLUXES in HEADWATER LAKES	**Central**	**South**	**Central**	**South**
Atm. wet deposition of Hg (μg m^{-2} yr^{-1})	7-15	10-25	7-15	10-25
Runoff load of Hg (μg m^{-2} yr^{-1})	4-8	6-11	45-80	60-110
Total load of Hg (μg m^{-2} yr^{-1})	11-23	16-36	55-90	70-120
Outflow of Hg (μg m^{-2} yr^{-1})	2-5	3-7	35-80	45-110
Sedimentation of Hg (μg m^{-2} yr^{-1})	7-20	10-30	8-20	10-30

Table II. Pools, biotic uptake and turnover of total and methylated Hg in the epilimnion (water and pelagic biota) of small headwater lakes (cf. Table I).

	Clearwater lake	Brownwater lake
Surface area (km^2)	1	0.1
Mean depth of epilimnion (m)	$\leq$5	$\leq$2
Total Hg concentration in epilimnion water (μg m^{-3})	2	5
MeHg concentration in epilimnion water (μg m^{-3})	$\leq$0.2	$\leq$0.5
Fish density (g m^{-2})	5	5
Fish production (g m^{-2} yr^{-1})	5	5
MeHg concentration in fish (μg g^{-1})	0.2	0.4
Total Hg pool in epilimnion water (μg m^{-2})	$\leq$10	$\leq$10
MeHg pool in epilimnion water (μg m^{-2})	$\leq$1	$\leq$1
MeHg pool in fish (μg m^{-2})	1	2
Total load of Hg (μg m^{-2} yr^{-1})	~20	~80
Hg flux through fish community (μg m^{-2} yr^{-1})	~1	~2

Both fluxes of Hg entering lakes are of significant magnitude. Runoff loading dominates in a majority of lakes (cf. Verta, 1990). The relative importance of the two input fluxes, however, varies dramatically with lake type. The ratio of runoff load and direct deposition varies from around 0.6 in clearwater lakes to around 6 in brownwater lakes, where it may be even higher close to historical point sources (Håkanson *et al.*, 1990; Meili, 1991). Evidently, this ratio will change during the year together with the hydrological conditions (Meili *et al.*, 1991). Direct deposition is more likely to dominate in summer, when water discharge in forest streams is low. As the biological activity in lakes is highest in summer as well, the importance of Hg deposition on lake surfaces for the Hg concentrations in biota may be underestimated by mass balance calculations on an annual basis. On the other hand, the bioavailability of deposited Hg may be comparatively low (Meili, 1991).

A finding of great importance is that the annual input of Hg to most lakes is considerably higher than the Hg pool in the water column (1 to 20 times, depending on lake type). This implies that Hg is supplied at a rate at which the Hg pool is usually replaced several times a year (cf. Fitzgerald and Watras, 1989). Single remedial measures within lakes of this type aiming at the removal of Hg from the water column are therefore likely to have little success.

The Hg pool in the water column of the model lakes is about 4 to 18 μg m^{-2}. It may have been slightly underestimated, since the Hg concentrations in the hypolimnion can exceed the surface concentrations during periods of stratification (Bloom and Watras, 1989). The hypolimnion volume, however, is often smaller than the epilimnion volume, especially in small and shallow lakes. The lake water pool of Hg is not varying very much regionally despite differences in Hg loading patterns and pathways. This may be due to regional differences in sedimentation patterns as a result of different summer length, lake productivity or humus characteristics, or due to selective rapid sedimentation of directly deposited Hg which accounts for a significant part of the regional differences in load in clearwater lakes.

The estimated sedimentation rates were 7 to 30 μg m^{-2} yr^{-1}, which coincides very well with data from sediment traps (Jönsson *et al.*, 1988; Grahn *et al.*, 1988; Johansson *et al.*, 1988; Bengtsson *et al.*, 1988) and with calculations based on dated sediment cores in similar lakes (e.g. El-Daoushi and Johansson, 1983). Regional variations in sedimentation behavior may affect both actual Hg concentrations and historical changes found in sediment Hg profiles (Johansson, 1985; Meili, 1991). Despite different amounts and pathways of Hg entering lakes, Hg sedimentation rates are similar in different lake types, whereas the concentrations in fish are higher in brownwater lakes. This indicates that different processes control sedimentation and biotic uptake, which may contribute to the weak or lacking correlation between Hg concentrations in sediments and fish (Chapter 11 in Lindqvist *et al.*, 1991).

Lake sediments appear to act as an efficient trap for Hg entering lakes. Consequently, the Hg load per unit lake surface from the tributaries will decrease significantly (up to 70%) with increasing number, size and proximity of upstream lakes in the catchment. This is supported by observations on the Hg export from small catchments (Chapter 8.3 in Lindqvist *et al.*, 1991). At the same time, the relative importance of direct deposition increases. The susceptibility of headwater lakes to Hg pollution can partly be explained by a higher Hg load per unit lake surface.

Most environmental studies of Hg pollution are reported from heavily contaminated systems, where sediments become important as an internal source of Hg after the reduction of emissions (e.g. Parks, 1988; Parks and Hamilton, 1987). As a result, external diffuse sources have often been overlooked, and the importance of sediments as an internal source of Hg overestimated, especially in "unpolluted" lakes. Sediments and hypolimnion waters, however, may still influence the bioavailability of Hg by acting as a methylation site or a temporary storage pool for methylated Hg (Bloom and Watras, 1989), which may be transferred to fish via zoobenthos, zooplankton or intense resuspension.

In general, it can be assumed that in summer, vertical Hg fluxes prevail in forest lakes, whereas in spring and autumn, horizontal fluxes dominate. It may therefore be of importance both for mass balance calculations and for the bioaccumulation of Hg, that the length of the growing season is decreasing rapidly from the south to the north of the study area.
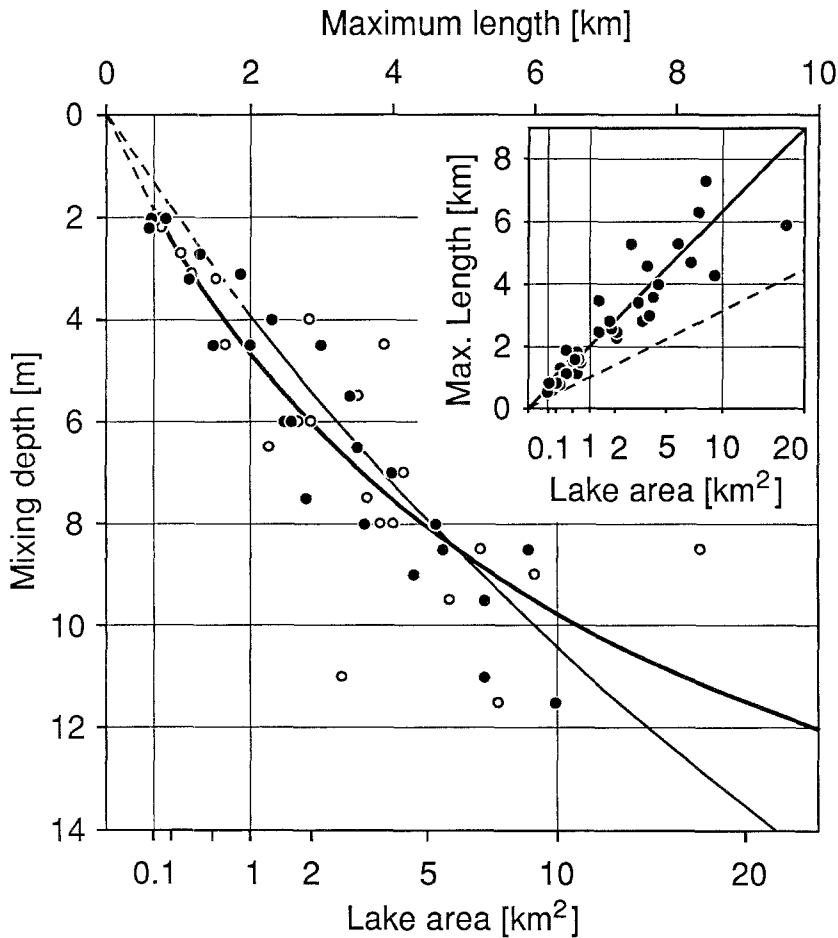
Figure 2. Depth of the mixed layer of surface water in 24 Swedish lakes of different size during the season of maximum stratification (July-August). The lower boundary of the mixed layer is defined as the depth at which the temperature gradient exceeds >0.5 °C m^{-1}. The relationship between mixing depth and the maximum lake length, defined as the largest straight line across continuous open water, is shown with black symbols. The bold line shows a model for the relationship between mixing depth [m] and maximum length [km], which takes into account that in very large lakes, mixing depth rarely exceeds 20 m ($D_{mix} = 20 \cdot L_{max} / (L_{max} + 6.6)$). Water mixing depth is in fair agreement with the depth below which sedimentation is not resuspended by water turbulences ($D_{T-A} = 45.7 \cdot L_{max} / (L_{max} + 21.4)$; thin line; for lakes >1 km^2; Håkanson and Jansson, 1983).

Maximum length can be estimated from the surface area (inset). The dashed line shows the theoretical maximum length in circular lakes (L_{max} = diameter = $2 \cdot \sqrt{(A / \Pi)} = 1.13 \cdot \sqrt{A}$). The solid line shows the geometric mean ratio of $L_{max} / \sqrt{A}$ in 38 Swedish lakes. In lakes for which the shape is not known, maximum length can be estimated from the lake area ($L_{max} = 2.0 \cdot \sqrt{A}$). The resulting relationship of mixing depth with estimated L_{max} is shown with white symbols.

5. Implications for biota

Another mass balance study is applied to assess the pool of Hg in the lake water and its bioavailability in the epilimnion (Table II, Figure 1). For this purpose, the model lakes above are stocked with a fish community of 5 g m^{-2}, with an annual production per biomass of 1 g g^{-1} (including youngs of the year). These values are typical for oligotrophic forest lakes (Chapman, 1978; Nyberg and Håkanson, unpublished data). Hg concentrations in fish are typically around 0.2 µg g^{-1} (wet weight) in clearwater lakes and 0.4 µg g^{-1} in polyhumic lakes. These values are given as an average for the whole community including top predators and fry, and are typical for forest lakes (cf. Chapters 11 and 12 in Lindqvist et al., 1991). Hg in fish is assumed to consist exclusively of MeHg (e.g. Westöö, 1973; Grieb et al., 1990). The Hg pool in fish is fairly representative for the total biotic pool in the open water, as it is larger than the pool of both total and methylated Hg in all other organisms including zooplankton, phytoplankton and bacterioplankton (Meili, unpublished data). MeHg in surface water is set as 10% of total Hg, which is in the upper part of the range found in lake surface waters (Parks et al., 1989; Bloom and Watras, 1989; Lee and Hultberg, 1990; Lee and Iverfeldt, 1991).

The depth of the mixed layer of surface water depends on the lake area as a result of wind action (Fig. 2). Brownwater lakes are usually smaller than clearwater lakes. The size of the model lakes was set to 0.1 km^2 (brownwater) and 1 km^2 (clearwater), resulting in mixing depths of 2 m and 5 m, respectively. The mean depth of the epilimnion in the whole lake is smaller, because mixing depth exceeds water depth in shallow areas, and because the mixed layer is more shallow earlier in the season.

The results in Table II and Figure 1 show that only a minor part (ca. 10 to 20%) of the total Hg pool in the epilimnion is stored in fish, but at least half of the MeHg pool (cf. Verta, 1984; Fitzgerald and Watras, 1989). Comparing the flux of Hg to fish with the total load on lakes, it is evident that usually less than 5% of the annual Hg input to lakes is channeled through the fish community. Consequently, both direct deposition and runoff input supply enough Hg to dominate the biotic uptake in lakes by themselves. This allows for large variations in bioavailability and concentrations of Hg in aquatic organisms at a given load on a lake, and implies that the bioavailability of the loaded Hg rather than the load itself is of crucial importance for the resulting Hg concentration in fish. Local remedial measures should therefore aim primarily at reducing the bioavailability of the Hg entering the lakes and need to be repeated continuously. If the turnover time of Hg in whole fish communities is similar to the turnover time of biomass, i.e. about one year (see above), the response time to changes in the Hg bioavailability will be on the order of a few years.

Acknowledgments

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