

# Mercury Sequestration in Forests and Peatlands: A Review

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## ABSTRACT

Nearly all Hg in vegetation is derived directly from the atmosphere. Mass of Hg in forest vegetation (roughly  $0.1 \text{ mg m}^{-2}$ ) is about an order of magnitude smaller than that in the forest floor ( $1 \text{ mg m}^{-2}$ ) and two orders of magnitude smaller than that in the mineral soil ( $10 \text{ mg m}^{-2}$ ). Mass of Hg in peat ( $20 \text{ mg m}^{-2}$ ) is greater than the sum of that in mineral soil and the forest floor; wetlands usually sequester more Hg than associated uplands. The strong relationship of Hg to organic matter, associated with binding by reduced S groups, is fundamental to understanding Hg distribution and behavior in terrestrial systems. The stoichiometry of the Hg–C relationship varies; Hg–S relationships, though less variable, are not constant. Because of the Hg–organic matter link, landscape conditions that lead to differential soil organic matter accumulation are likely to lead to differential Hg accumulation. The ratio of methylmercury (MeHg) to total Hg is generally low in both vegetation (near 1.5%) and soil (<1%), but areas of poorly drained soils and wetlands are sites of MeHg production. The annual emission of anthropic Hg from the 48 contiguous states of the USA (144 Mg) is two orders of magnitude less than the pool of Hg in forests of those states (30 300 Mg). Peatlands, less than 2% of total land area, sequester more than 20 times annual emissions (2930 Mg). If global climate change affects C storage it will indirectly affect Hg storage, having a major effect on the balance between emissions and sequestration and on the global Hg cycle.

MERCURY (Hg) is considered a global pollutant (Fitzgerald et al., 1998; Jackson, 1997) because under ordinary conditions  $\text{Hg}^0$  [metallic;  $\text{Hg}(0)$ ] vaporizes readily and is widely distributed via atmospheric processes. In addition to natural sources of Hg, human activities since the beginning of the industrialized period have significantly increased the global reservoir of atmospheric Hg (Fitzgerald et al., 1998). Atmospheric Hg is deposited to terrestrial and aquatic systems, even in remote areas. There is some disagreement concerning the relative importance of atmospheric deposition (Fitzgerald et al., 1998) versus geologic sources (Rasmussen, 1994a) with respect to Hg in terrestrial and aquatic systems. Geographic trends in Hg that transcend geologic differences indicate that, at the minimum, atmospheric Hg contributes to a geologic background.

Most attention toward Hg pollution has focused on methylmercury (MeHg) because this potent neurotoxin can be concentrated more than a million-fold in the aquatic food chain (Rudd, 1995; Ullrich et al., 2001). Terrestrial systems are an important indirect source of atmospheric Hg to aquatic systems via runoff (Lee et al., 1994; Lorey and Driscoll, 1999; Swain et al., 1992); on an areal basis they receive more atmospheric Hg than do freshwater aquatic systems. They are also a significant reservoir of Hg, containing many times its an-

nual flux to and from the atmosphere (Mason et al., 1994). Fluxes in and out of the terrestrial reservoir affect the remainder of the global Hg cycle (Lindberg, 1996; Porcella, 1994). This review will focus on Hg pools in forests and peatlands in the temperate and boreal zones, and is a companion to a recent review of fluxes in and out of those systems (Grigal, 2002). The emphasis will be on representative studies of systems that are not near point sources of Hg. Most research has been carried out on forested systems, but reports from other land uses such as agriculture will also be included. Because many of the data are lognormally distributed, unless otherwise specified the geometric mean will be used to describe the central tendency of distributions.

## VEGETATION

### Foliage

The atmosphere is nearly the exclusive source of Hg in vegetation. Both greenhouse and laboratory studies have shown that Hg uptake from soil is limited, with roots acting as a significant adsorption site and a barrier for Hg transport to foliage (Grigal, 2002). Field observations are consistent with that premise, including positive correlations between foliar and atmospheric concentrations but none between foliar and root concentrations (Barghigiani et al., 1991), and positive correlations between foliar concentrations and growing season length (i.e., duration of physiologic activity) and inverse correlations with soil Hg (Fleck et al., 1999). Measurements of xylem sap in mature spruce and pine in Sweden indicated that only about 10% of Hg in litterfall could be accounted for by root uptake (Bishop et al., 1998). Some research has indicated the presence of a compensation point of atmospheric Hg, above which Hg is absorbed and below which it is emitted by foliage (Hanson et al., 1995; Lindberg, 1996). Nonetheless, foliage is a net sink for Hg with concentrations increasing with time. In conifers, concentrations increase with needle age (Fig. 1). Concentrations also increase over the growing season in both coniferous (Rasmussen, 1995) and deciduous trees (Lindberg, 1996). Some report that conifer needles tend to have higher Hg concentrations than deciduous leaves from the same location because collections from conifers usually include older foliage (Rasmussen et al., 1991), but such a tendency is not universal (Maňkovská, 1996). Plants of lower stature appear to have higher foliar concentrations than collocated trees (Rasmussen et al., 1991). This is consistent with uptake from a soil source of  $\text{Hg}(0)$  (Lindberg et al., 1992). Concentrations of Hg in nonvascular plants (mosses, fungi, and lichens) are nearly an order of magnitude higher than those in

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**Abbreviations:** MeHg, methylmercury; OM, organic matter; SOM, soil organic matter.

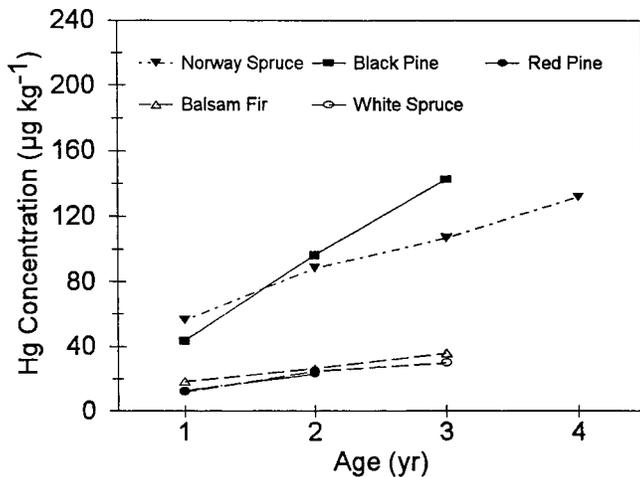


Fig. 1. Differences in Hg concentration of conifer needles with needle age. Data from Norway spruce [*Picea abies* (L.) H. Karst.] in Germany (Bombosch, 1983); Calabrian black pine (*Pinus nigra* J.F. Arnold subsp. *laricio* Maire) in Italy (Barghigiani et al., 1991); red pine (*Pinus resinosa* Aiton) in Minnesota (Fleck et al., 1999); and balsam fir [*Abies balsamea* (L.) Mill.] and white spruce [*Picea glauca* (Moench) Voss] in northern Ontario, Canada (Rasmussen, 1995).

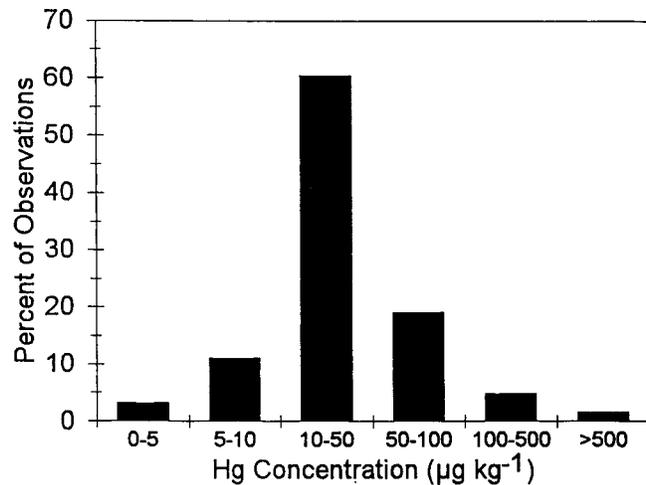


Fig. 2. Foliar concentrations of Hg reported in studies from Europe (Bombosch, 1983; Barghigiani et al., 1991; Maňkóvká, 1996; Munthe et al., 1998) and North America (Fleck et al., 1999; Grigal et al., 2000; Lindberg, 1996; Moore et al., 1995; Rasmussen et al., 1991; Rasmussen, 1994b, 1995; Zhang et al., 1995a) ( $n = 63$ ). This is only a snapshot of the extensive data that exist.

vascular plants (Moore et al., 1995) and may also reflect a soil source of Hg(0).

There is a wide range of reported foliar concentrations of Hg, depending on atmospheric concentrations and differences in uptake efficiency. For example, although there is a general increase in Hg concentration with needle age, concentrations in needles from western Europe are much higher than those from relatively remote North American sites (Fig. 1). In a Hg mining area in Italy, Austrian pine (*Pinus nigra* J.F. Arnold) needles had Hg concentrations up to 4000  $\mu\text{g kg}^{-1}$  associated with atmospheric concentrations of 600  $\text{ng m}^{-3}$  (Barghigiani et al., 1991). Most reported tree foliar concentrations are much lower, in the range 10 to 100  $\mu\text{g kg}^{-1}$  (Fig. 2) with a mean near the lower end of that range (26  $\mu\text{g kg}^{-1}$ ).

## Wood

The majority of the aboveground mass of forest vegetation is in bolewood (Birdsey and Heath, 1995). In a diverse group of deciduous hardwood and coniferous forests from the Northern Hemisphere, about 75% of the aboveground tree biomass (ranging from 60 to 90%) was woody bole (Johnson and Lindberg, 1992). Despite that predominance, there are relatively few reports of Hg concentration of woody material. Although some estimates are very high (e.g., 100–500  $\mu\text{g kg}^{-1}$ ; Nriagu and Pacyna, 1988, source not attributed), more recent studies using sophisticated analytical techniques have reported very low concentrations. For example, Grigal et al. (2000) reported a concentration of 5  $\mu\text{g kg}^{-1}$  in black spruce [*Picea mariana* (Mill.) Britton et al.] and 2  $\mu\text{g kg}^{-1}$  in aspen (*Populus tremuloides* Michx.). Moore et al. (1995) reported a 10  $\mu\text{g kg}^{-1}$  concentration in black spruce; Munthe et al. (1998) reported 13  $\mu\text{g kg}^{-1}$  in Norway spruce [*Picea abies* (L.) H. Karst.]; Fleck et

al. (1999) reported 4  $\mu\text{g kg}^{-1}$  in red pine (*Pinus resinosa* Aiton); and Lindberg (1996) reported 3  $\mu\text{g kg}^{-1}$  in hardwood species. A higher figure reported for black spruce in Ontario (average of 20  $\mu\text{g kg}^{-1}$ ; Zhang et al., 1995b) is suspect because the highest concentrations (37  $\mu\text{g kg}^{-1}$ ) were in annual tree rings formed between 1910 and 1930, before the peak of atmospheric Hg deposition in North America (Swain et al., 1992).

In the north-central USA, Hg in wood and associated bark ( $n = 95$ ) had a lognormal distribution with concentrations ranging from 0.6 to 12  $\mu\text{g kg}^{-1}$ , with more than 60% of observations in the range 1 to 4  $\mu\text{g kg}^{-1}$  and a mean of about 2.5  $\mu\text{g kg}^{-1}$  (E.A. Nater et al., personal communication, 1999) (Fig. 3). A wide variety of predominantly deciduous tree species, including both living and dead individuals, was sampled. There were few significant differences in concentration among species and none between living and dead trees. Concentrations of Hg were positively correlated in 15 pairs of bark and adjacent wood-only samples ( $r^2 = 0.48$ ), with wood concentrations significantly (more than one order of magnitude) lower than those in bark (mean = 0.9 vs. 12  $\mu\text{g kg}^{-1}$ ; E.A. Nater et al., personal communication, 1999). The source of the Hg in bark is probably long-term dry deposition, while that in wood is translocation from foliage (Barghigiani et al., 1991). The correlation between their concentrations is reasonable because they are both exposed, one directly and the other indirectly, to the same atmospheric pool of Hg. Similar differences and interpretations, albeit with higher concentrations (wood = 13  $\mu\text{g kg}^{-1}$ , bark = 200  $\mu\text{g kg}^{-1}$ ), were reported for Norway spruce (Munthe et al., 1998). Zhang et al. (1995a,b) reported smaller differences in concentration between black spruce wood (20  $\mu\text{g kg}^{-1}$ ) and bark (60  $\mu\text{g kg}^{-1}$ ). Higher concentrations in bark than wood suggest that branches, with proportionally more bark, would have higher Hg concentrations than the tree bole.

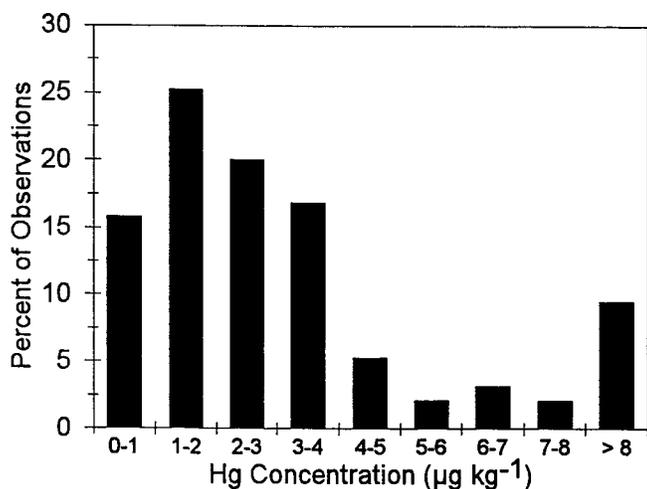


Fig. 3. Frequency distribution of Hg concentration in wood and associated bark from a variety of predominantly deciduous tree species ( $n = 95$ ) (E.A. Nater et al., personal communication, 1999).

The size of the Hg pool in forest vegetation depends on both concentrations and biomass. The aboveground carbon (C) content of forest vegetation in the contiguous 48 states of the USA is approximately  $48 \text{ Mg ha}^{-1}$  (Birdsey and Heath, 1995), and based on the mean Hg concentrations in vegetation the pool of Hg in that vegetation would be about  $66 \mu\text{g m}^{-2}$ . This is very near the estimate of  $80 \mu\text{g m}^{-2}$  for a deciduous hardwood forest in Tennessee, USA (Lindberg, 1996) and of  $60 \mu\text{g m}^{-2}$  for a coniferous spruce forest in a bog in Minnesota, USA (Grigal et al., 2000), but about three times that of a deciduous aspen forest in the upland surrounding the bog ( $20 \mu\text{g m}^{-2}$ ; Grigal et al., 2000). The latter estimate is similar to that for trees and shrubs in an ombrotrophic peatland in Ontario ( $15 \mu\text{g m}^{-2}$ ; Moore et al., 1995). These estimates are much lower than the estimate of  $680 \mu\text{g m}^{-2}$  for a Norway spruce forest in southern Sweden (Munthe et al., 1998). Very high concentrations in needles  $\geq 2$  yr old ( $530 \mu\text{g kg}^{-1}$ ) and in bark ( $200 \mu\text{g kg}^{-1}$ ) strongly influenced the latter estimate (Munthe et al., 1998). A high rate of dry deposition of Hg to the surfaces of that forest is a possible explanation for these high concentrations.

### Roots and Coarse Woody Debris

Data are rare for two other pools of Hg in forests. Mass of tree roots is about one-fifth that of aboveground material (Wharton and Griffith, 1993; Whittaker and Marks, 1975). Roots include both bark, likely to be high in Hg because of contact with mineral soil, and wood, likely to approximately equal to the concentration of aboveground wood. Concentrations in roots of Norway spruce in southern Sweden were similar to those for one-year-old needles (roots =  $49 \mu\text{g kg}^{-1}$ , needles =  $40 \mu\text{g kg}^{-1}$ ) (Munthe et al., 1998). In a contaminated area in Italy, root concentrations were about half those of needles up to about  $2000 \mu\text{g kg}^{-1}$  (Barghigiani et al., 1991) (compare needle concentrations with Fig. 2). Based on these limited data, general estimates of Hg concentration in roots are tenuous. If an estimate of Hg

in all living forest vegetation including roots is desired, an increase of about 20% in the aboveground estimate seems reasonable.

A second neglected pool of Hg in vegetation is coarse woody debris (CWD), which comprises both standing dead trees and dead wood on the forest floor. Increasing attention is being paid to CWD because of its importance in a wide variety of ecosystem processes (Harmon et al., 1986). The quantity and kind of CWD is determined by the balance between inputs and outputs, and varies widely with species, stage of stand development, climate, and forest disturbance regimes. Estimates of CWD range from less than  $10$  to  $40 \text{ Mg ha}^{-1}$  for forests in eastern and continental areas of North America (Duvall and Grigal, 1999; McCarthy and Bailey, 1994; Muller and Liu, 1991; Sturtevant et al., 1997; Tyrell and Crow, 1994) to a much higher mass, up to  $400 \text{ Mg ha}^{-1}$ , in moist old-growth conifer forests of the Pacific Northwest of North America (Harmon et al., 1987; Keenan et al., 1993; Spies et al., 1988). There are no data on Hg concentrations in CWD, nor are there data to indicate if it varies with stage of decay. Assuming that concentrations will be similar to those in live or freshly dead wood (from less than  $1$  to near  $10 \mu\text{g kg}^{-1}$ ; E.A. Nater et al., personal communication, 1999), Hg storage could range from  $1$  to  $40 \mu\text{g m}^{-2}$  in continental areas up to an unlikely  $400 \mu\text{g m}^{-2}$  in extreme situations. In most cases the Hg pool in CWD is therefore of the same order of magnitude but smaller than that in live vegetation.

The Hg in vegetation is an ephemeral pool, not a permanent sink for Hg. Vegetation modifies atmospheric Hg inputs and transfers them to soil reservoirs or back to the atmosphere. The potential exists for rapid release of Hg through natural phenomenon such as forest fire, for very slow release through vegetation death and microbial decomposition, or for intermediate rates of release following forest harvest.

### Methylmercury

Vegetation may also contain MeHg, from the atmosphere or produced in the plants (Rudd, 1995), or from soil via the transpiration stream (Bishop et al., 1998). The average ratio of MeHg to total Hg in tree litterfall, predominantly foliage, is nearly identical to that in open precipitation (1.5%) (Munthe et al., 1995, 1998; Schwesig and Matzner, 2000; Lee et al., 2000). In northwestern Ontario, Canada, tree and shrub leaves had a lower ratio of MeHg to total Hg (1.1%; Moore et al., 1995). About 1.5% of the total Hg mass in vegetation ( $30 \mu\text{g m}^{-2}$ ) was present as MeHg, with bryophytes contributing about 75% of the MeHg but only 50% of the total Hg (Moore et al., 1995).

### ANIMALS

In contrast to the emphasis on Hg accumulation in aquatic systems, and especially accumulation of MeHg, there has been little attention paid to Hg accumulation in terrestrial fauna. Although bioaccumulation is relatively well understood in the aquatic system (USEPA, 1997b, 2001), it does not appear to be predictable in

terrestrial systems; other factors influence accumulation (Nuorteva, 1990). A few general observations, however, can be made. First, analogous to aquatic organisms, MeHg is more toxic than Hg(II) to terrestrial organisms (Boening, 2000), and in feeding trials of terrestrial invertebrates it is retained to a greater degree than is Hg (II) (Boening, 2000). Second, because Hg concentrations in vegetation are relatively low, concentrations in herbivores such as deer are very low (Bombosch, 1983; Lodenius, 1994). Although the ratio of MeHg to total Hg approaches unity in the muscle tissue of carnivorous fish high in the food chain, ratios tend to be low in herbivorous terrestrial mammals (Bull et al., 1977; La-sorsa and Allen-Gil, 1995). Third, biomagnification is less pronounced in terrestrial than aquatic food chains (Lodenius, 1994). Because of low concentrations and limited biomagnification, the terrestrial pathway is not expected to be a significant source of Hg to animals higher on the food chain (USEPA, 1997b). Nonetheless, in general Hg concentrations are higher in terrestrial carnivores than herbivores (Boudou and Ribeyre, 1997).

As may be expected, piscivorous terrestrial predators have higher MeHg burdens than do strictly terrestrial predators (Boudou and Ribeyre, 1997). For example, body burdens of MeHg in otter (*Lutra lutra*) and mink (*Mustela lutreola*) may represent 30 to 50% of total Hg (Lodenius, 1994). The aquatic and terrestrial systems are often linked. There is at least one reported death of a terrestrial carnivore due to Hg toxicosis, that of a Florida panther (*Felis concolor coryi*) in Everglades National Park (Facemire et al., 1995). The major source of Hg to panthers in that area are raccoon (*Procyon lotor*), a piscivorous terrestrial predator (Facemire et al., 1995).

Earthworms are widely recognized as a bioindicator of heavy metal and organic pollution in soils. Because nearly all of the research regarding total Hg or MeHg effects on earthworms has been conducted at very high Hg concentrations (Rundgren et al., 1992), it is difficult to extrapolate to the low levels found in unpolluted environments. Cocking et al. (1994) found a direct relationship between Hg concentrations in earthworm tissue and in the soil from which they were collected ( $r^2 = 0.78$ ) over a range of soil concentrations from 100 to 50 000  $\mu\text{g kg}^{-1}$  and including uncontaminated and contaminated sites. At soil concentrations below about 400  $\mu\text{g kg}^{-1}$ , generally uncontaminated, concentrations in worms were greater than those in soil but seldom exceeded 1000  $\mu\text{g kg}^{-1}$  dry weight. Above that soil concentration, concentrations in worms were less than in soil but were all greater than 5000  $\mu\text{g kg}^{-1}$  dry weight. Few MeHg data are available, but earthworms from a contaminated site were reported to have 8 to 13% Hg as MeHg (Bull et al., 1977), in the same range as that reported in uncontaminated soils (see below). There is also considerable interspecific variation in Hg concentration in soil invertebrates (Lodenius, 1994). Accumulation of Hg in terrestrial fauna is an open topic for research.

## SOILS

### Introduction

Dominant forms of Hg in soil are  $\text{Hg}^{2+}$  [Hg(II)] and Hg(0), both in solution and adsorbed (Schuster, 1991; Yin et al., 1996). The strong relationship between Hg and organic matter (OM) (Meili, 1991), including soil organic matter (SOM), is fundamental to understanding its environmental behavior. Although Hg binds to ligands such as  $\text{OH}^-$  and  $\text{Cl}^-$  (Yin et al., 1996) and to clay minerals (Sarkar et al., 2000) in laboratory studies, in most terrestrial and aquatic systems the predominant form of Hg, Hg(II), is associated with OM (Schuster, 1991; Skyllberg et al., 2000). Laboratory work is usually done at Hg solution concentrations that may occur at contaminated sites but are unlikely away from such sites. The binding to OM in natural systems appears to be by reduced S groups that are at concentrations sufficiently high to bind all Hg (Skyllberg et al., 2000).

Some of the early literature reporting soil Hg may be biased upward; concentrations in soils are high enough so that ultraclean analytical techniques were not absolutely necessary but uncertainty would be greatest for soils with very low concentrations. Concentrations of Hg in soils tend to follow lognormal distributions, with some very high concentrations associated with industry or natural Hg mineralization. A compilation of data from soils from Europe, both arable (primarily agricultural soils,  $n = 48$ ) and natural (mainly forests with some pastures and peats,  $n = 50$ ), had nearly identical mean concentrations; about 140  $\mu\text{g kg}^{-1}$  (Fig. 4). Only 30% of the natural soils but 40% of the arable soils fell between 50 and 150  $\mu\text{g kg}^{-1}$  (Rundgren et al., 1992).

### Mercury-Soil Organic Matter Relationship

The close relationship between Hg and SOM can be illustrated by regional sampling of surface organic horizons in forests, referred to as mors from Sweden (Håkanson et al., 1990), as humus layers from Norway (Låg and Steinnes, 1978), and as the forest floor from the north-central USA (Nater and Grigal, 1992). All show similar changes in Hg concentration with changes in SOM (about 0.22  $\mu\text{g Hg}$  per g SOM), but the relationships are not identical, with Swedish data having the highest intercept and data from the USA having the greatest slope (Fig. 5). Arithmetic mean concentrations, from 150  $\mu\text{g kg}^{-1}$  in the USA to 190  $\mu\text{g kg}^{-1}$  in Norway to 250  $\mu\text{g kg}^{-1}$  in Sweden, may reflect a difference in historical loadings. Reports from surface organic horizons in central Europe are higher [300 to 400  $\mu\text{g kg}^{-1}$  in Schwesig et al. (1999) and 700 to 900  $\mu\text{g kg}^{-1}$  in Godbold (1994)].

The forest floor usually has a higher Hg concentration than does the underlying surface mineral soil. In the north-central USA, the mean Hg concentration in the forest floor was 140  $\mu\text{g kg}^{-1}$  while that of the underlying surface mineral soil (0- to 25-cm depth) was 20  $\mu\text{g kg}^{-1}$  (Fig. 6), associated with a higher SOM concentration in the forest floor (50 vs. 3% dry wt.). The ratio of Hg to SOM in the mineral soil, 2.5 times that in the forest

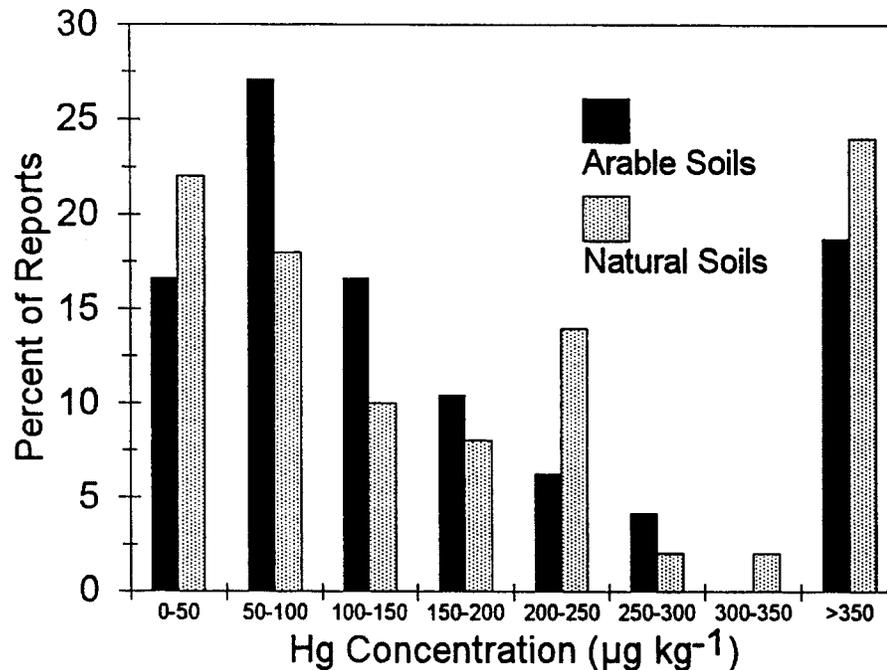


Fig. 4. Frequency distributions of Hg concentration in arable and natural soils from Europe, tabulated by Rundgren et al. (1992). Arable soils ( $n = 48$ ) are primarily agricultural soils, while natural soils ( $n = 50$ ) are mainly forests with some pastures and peats.

floor, was slightly greater than the nearly doubling of the ratio in 20 sites in Sweden (Lindqvist et al., 1991). The Hg-SOM relationship also continues into the mineral soil. Patterns of Hg concentration with depth mirror changes in SOM in both a chernozem, formed under grassland vegetation and characterized by high levels of SOM at the surface and a monotonic decline with depth (Fig. 7, top); and in a podzol, formed under forest vegetation with very high SOM at the surface and a subsurface accumulation (Fig. 7, bottom) (Anoshin et al., 1996). In both soils, the organic- and root-rich sod horizon (As) has high Hg concentrations compared with the lower horizons with less SOM. Although there are

pronounced differences in vertical patterns of Hg, these soils also represent different ecosystems whose vegetation and climate affect Hg input-output relationships.

### Stoichiometry

Although a general relationship exists between Hg and SOM, the stoichiometry varies widely. For example, the relationship in the forest floor across the north-central USA (Nater and Grigal, 1992) (slope equivalent to  $0.29 \mu\text{g Hg per g SOM}$ ,  $r^2 = 0.51$ ,  $n = 133$ ) significantly differs [ $F(1, 285) = 291$ ,  $p < 0.001$ ] from that in the 0- to 25-cm mineral soil horizon ( $0.44 \mu\text{g Hg per g SOM}$ ,  $r^2 = 0.53$ ,  $n = 155$ ). The higher Hg density of the SOM

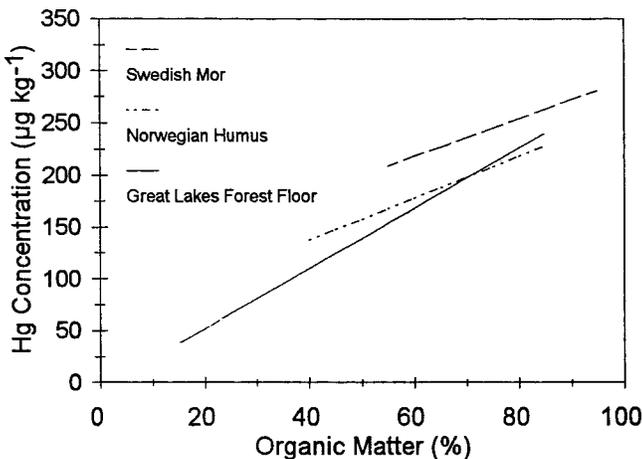


Fig. 5. Empirical relationships between Hg concentration and organic matter of surface organic horizons in forests: Swedish mor,  $n = 363$  (Håkanson et al., 1990); Norwegian humus,  $n = 700$  (Låg and Steinnes, 1978); Great Lakes forest floor from the north-central USA,  $n = 133$  (Nater and Grigal, 1992). Lines indicate approximate range of data from each study.

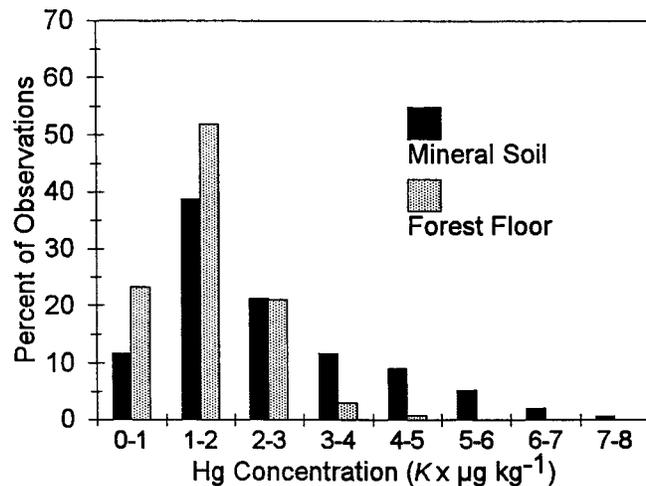


Fig. 6. Frequency distribution of Hg concentration in the forest floor and underlying surface mineral soil (0- to 25-cm depth) from sites located across the north-central USA ( $n = 133$ ) (data from Nater and Grigal 1992). The multiplier for the units,  $k$ , is 100 for forest floor and 10 for mineral soil.

in mineral soil implies that proportionally less Hg than C is lost during OM mineralization. Stoichiometry also varies with depth in mineral soil. In a mixed forest and field landscape in the north-central USA, the simple ratio between Hg and SOM in surface mineral soil (0–10 cm) was lower than that for the underlying 10 to 50 cm of soil (0.53 versus 0.78  $\mu\text{g Hg per g SOM}$ , respectively), with a greater rate of change in the surface (slope equivalent to 0.76  $\mu\text{g Hg per g SOM}$ ,  $r^2 = 0.84$ ,  $n = 53$ ) than in the underlying soil (0.61  $\mu\text{g Hg per g SOM}$ ,  $r^2 = 0.29$ ,  $n = 55$ ) [ $F(1, 105) = 397$ ,  $p < 0.001$ ] (data from Grigal et al., 1994). Higher Hg density of SOM or soil C with depth is commonly reported; average ratios for surface organic horizons (0.7  $\mu\text{g Hg per g C}$ ) increase in the A, B, and C horizons (1.5, 2.5, and 3.8  $\mu\text{g Hg per g C}$ , respectively) (Aastrup et al., 1991; Dudas and Pawluk, 1976; Gladkova and Malinina, 1999; Grigal et al., 1994; Schwesig et al., 1999). There do not appear to be appreciable differences in the proportion of Hg that is humic-bound among horizons, although slightly less Hg appears to be fulvic-bound in O horizons (Schwesig et al., 1999).

Soils in the same landscape also differ in Hg–SOM stoichiometry. A forested Antigo silt loam (coarse-loamy over sandy or sandy-skeletal, mixed, superactive, frigid Haplic Glossudalf) and a cultivated Sparta loamy sand (sandy, mixed, mesic Entic Hapludoll) occupying different parts of the same landscape were sampled over about 1000 ha (RUST Environment, 1994). Although there was a reasonable positive relationship between Hg and soil C for the mineral surface of the forested soil (slope 0.51  $\mu\text{g Hg per g C}$ ,  $r^2 = 0.40$ ,  $n = 54$ ), there was none for the cultivated soil ( $r^2 = 0.001$ ,  $n = 45$ , mean = 1.4  $\mu\text{g Hg per g C}$ ). Variation in the Hg–SOM stoichiometry may be related to historical loadings, degree of mineralization, or to the relative amount of reduced S groups.

Although data to examine the stoichiometry of the Hg–S relationship are limited, those from the north-central USA (Nater and Grigal, 1992) support a more constant relationship than between Hg and SOM. Changes in Hg concentration with S do not differ between the forest floor and the 0- to 25-cm mineral soil horizon [ $F(1, 285) = 0.12$ ,  $p = 0.73$ , pooled slope = 120  $\mu\text{g Hg per g S}$ ,  $r^2 = 0.86$ ,  $n = 288$ ]. This implies that differences in Hg concentration between the horizons simply reflect differences in S. Unfortunately, that uniform stoichiometry is not maintained in a limited sampling of soils in a small watershed in Minnesota (Skyllberg et al., 2000) (slope 40  $\mu\text{g Hg per g S}$ ,  $r^2 = 0.43$ ,  $n = 8$ ) nor in data from Russian soils (slope 95  $\mu\text{g Hg per g S}$ ,  $r^2 = 0.07$ ,  $n = 11$ ; Gladkova and Malinina, 1999). In both cases, the Hg–C relationship is stronger than Hg–S ( $r^2 = 0.85$  and 0.15, respectively). Although the Hg–S relationship holds promise for explaining Hg distribution, there are many other factors that influence the composition of SOM.

### Soil Physical Properties

Some early studies in the Canadian prairies reported lower Hg concentrations in organic-rich surface hori-

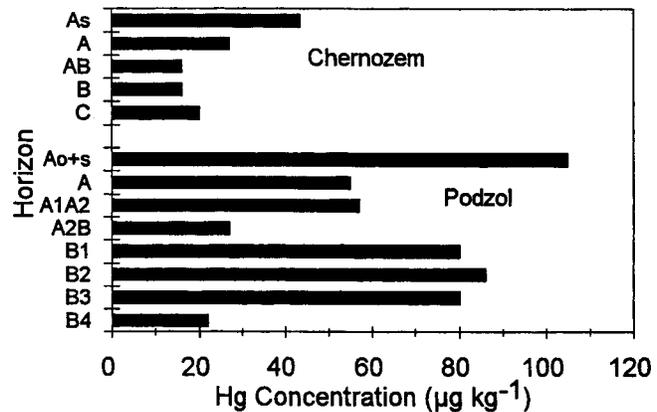


Fig. 7. Concentration of Hg with depth in an untilled chernozem (top) and podzol (bottom) from the Altai Territory, southwestern Siberia. Data to a 135-cm depth in the chernozem and a 150-cm depth in the podzol. Data from Anoshin et al. (1996); horizons with similar designations are combined.

zons than in subsurface horizons (Dudas and Pawluk, 1976 and references therein). This was ascribed to volatilization of Hg from the surfaces, and that hypothesis is a possibility. However, a more recent survey of Canadian prairie soils ( $n = 1076$ ) generally did not find that trend (R.G. Garrett and L.H. Thorleifsson, unpublished data, 1994). Higher Hg concentrations in subsurface than surface horizons were associated with clay-rich, shale-dominated glacial till. In conformity with those observations, most of the soils studied by Dudas and Pawluk (1976) had higher clay concentrations in subsurface than in surface horizons. They also reported that over all soils they studied, the clay fraction had significantly higher Hg concentrations (mean = 85  $\mu\text{g kg}^{-1}$ ) than did either the sand (12  $\mu\text{g kg}^{-1}$ ) or silt (24  $\mu\text{g kg}^{-1}$ ) fractions (Fig. 8). This is consistent with the concentration in silt plus clay of subsurface horizons of forested soils in the north-central USA (52  $\mu\text{g kg}^{-1}$ ) (Nater and Grigal, 1992).

Emphasis on concentrations can distort perceptions about Hg abundance. Although higher Hg concentrations are usually associated with higher SOM, soil bulk density and SOM are inversely related (Grigal et al., 1989). This leads to the paradox that horizons with high concentrations of Hg often have low mass per unit area. For example, although 20 Swedish sites had a mean Hg concentration of 250  $\mu\text{g kg}^{-1}$  in the surface organic horizon and only 30  $\mu\text{g kg}^{-1}$  in the B horizon, Hg mass was 1.6  $\text{mg m}^{-2}$  in the surface and 3.7  $\text{mg m}^{-2}$  in the B horizon (Lindqvist et al., 1991). Similarly, the average Hg mass in the forest floor from the north-central USA was 0.9  $\text{mg m}^{-2}$  while that in surface mineral soil was 5.7  $\text{mg m}^{-2}$ , despite the much higher Hg concentration in the forest floor (Fig. 6) (Nater and Grigal, 1992). Reported Hg mass in the forest floor ranges from about 0.5 to 4  $\text{mg m}^{-2}$  and for surface (to 25 to 40 cm) mineral soil ranges from 3 to 10  $\text{mg m}^{-2}$  (Aastrup et al., 1991; Fleck et al., 1999; Grigal et al., 1994, 2000; Lindqvist et al., 1991; Munthe et al., 1998; Nater and Grigal, 1992), with considerably higher mass reported in Tennessee, USA (Lindberg, 1996), and Germany (Schwesig et al., 1999; Schwesig and Matzner, 2000). The estimate of

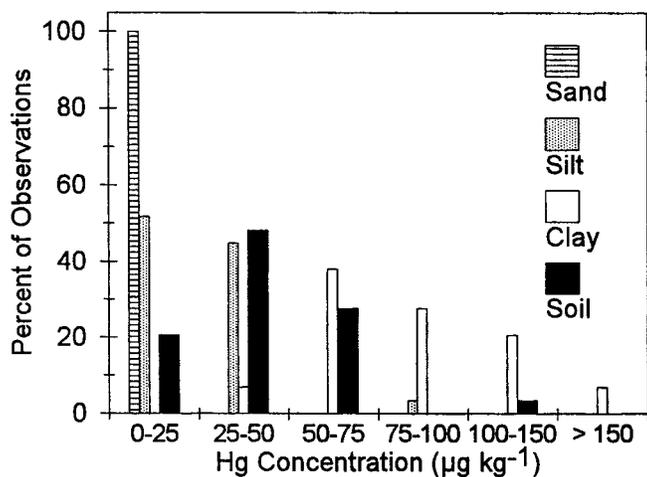


Fig. 8. Frequency distribution of Hg concentration in sand, silt, and clay fractions, and in total soil, of horizons of uncultivated soil from Alberta, Canada ( $n = 29$ ) (data from Dudas and Pawluk, 1976).

the Hg pool in mineral soil is strongly influenced by the depth that is considered. Even low concentrations of Hg can lead to substantial mass when accumulated over 200 to 300 cm, especially when associated with the higher bulk density usually occurring at depth. The Hg in deep soil horizons is a major source of uncertainty in pool estimation.

### Landscape

As a result of the Hg–SOM link, landscape conditions that lead to differential SOM accumulation are likely to lead to differential Hg accumulation. This provides an aid to understanding Hg distribution because fundamental pedological concepts related to SOM, such as differential horizon accumulation and the catena concept of increasing SOM downslope, can be used. Soil properties such as the redox status, cations, and especially the particle-size distribution (texture) all strongly affect SOM. High levels of cations, especially Ca, stabilize SOM and protect it from oxidation (Oades, 1988). The fine silt and clay soil fractions have the highest concentrations of SOM (Anderson et al., 1981; Zhang et al., 1988). Poorly drained soils with high water content and resulting low soil  $O_2$  have higher SOM than associated well-drained soils because of restricted microbial activity. Empirically, SOM in well-drained soils in both grasslands (Burke et al., 1989) and forests (Grigal and Ohmann, 1992; Homann et al., 1995) has been related to clay content, temperature, and precipitation.

These relationships affect Hg distribution. Nearly all the statistical variation in Hg concentration of the upper horizons of podzolic soils along a catena from the upper part of a slope to a river floodplain was explained by a combination of position along the catena (24%) and horizon differentiation (75%) (Gladkova and Malinina, 1999). Similarly, in transects from the summit to base of forested slopes, Hg content in surface soils increased from about 3.4 to 5.5 mg m<sup>-2</sup> (Grigal et al., 1994). In the same area, abandoned agricultural fields had very low SOM and less Hg (1.3 mg m<sup>-2</sup>) than immediately

adjacent forests (2.4 mg m<sup>-2</sup>) (Grigal et al., 1994). In cultivated soils in Alberta, poorly drained soils, higher in SOM than well-drained soils, tended to have higher Hg concentrations (Dudas and Pawluk, 1977). Within poorly drained soils, Hg and SOM concentrations were positively related (Dudas and Pawluk, 1977).

The role of vegetation in determining SOM in forests is the product of the complex interplay between production and decomposition. Over a range of temperate forest sites, productivity as measured by litterfall increased with mean annual temperature while SOM in both the forest floor and mineral soil concurrently decreased (Johnson, 1995). The decomposition rate, calculated by the steady state ratio,  $k$  (defined by Olson, 1963), increased with temperature but was lower for lignin-rich conifer than deciduous forests (Johnson, 1995). These contrasting trends are caused by opposing forces; more productive forests produce more organic inputs to the soil but they occur in climates and produce litter that favor rapid decomposition. Forests with greater rates of production and nutrient cycling can therefore be expected to have lower SOM and soil Hg concentrations, and resulting Hg content. In plantations of red pine, a species with little genetic variation, Hg concentrations in both plant tissue and soil were related to growing season length, but the relationship was positive for plant tissue and inverse for soil (Fleck et al., 1999). A longer growing season increased foliar uptake of Hg but also increased organic matter mineralization and Hg efflux from soil. This implies that the concern about Hg in aquatic systems at higher latitudes may be related to the short season for mineralization at those latitudes, leading to greater retention of SOM and Hg within the terrestrial system and greater potential for hydrologic transport to adjacent aquatic systems.

### Methylmercury

As stated earlier, the most toxic form of Hg is MeHg, with toxicity primarily manifested in aquatic systems. Depending on the circumstances, each of the three potential sources of MeHg to aquatic systems (internal production, flux from terrestrial watersheds, or atmospheric inputs) can be most important (Rudd, 1995). Atmospheric deposition of MeHg does not appear to be a dominant factor influencing the flux from terrestrial watersheds. Two Swedish watersheds, differing in wet MeHg inputs by a factor of three, had comparable MeHg flux (Lee et al., 1994). Only a small proportion of Hg in soils is present as MeHg, with the ratio of MeHg to total Hg ranging up to slightly more than 5%, but with more than 70% of the observations at <1% and a mean of 0.6% (Fig. 9). There is no consistent trend in the ratio, either with total Hg or with soil horizon. The proportion of MeHg in soils depends on factors other than those affecting total Hg.

Both the MeHg and total Hg concentrations in soil water are highly variable, as is the resulting ratio between them. From the limited available data, the ratio of MeHg to total Hg in soil water can be expected to range from about 0.15 to 15% (Grigal, 2002). Concentra-

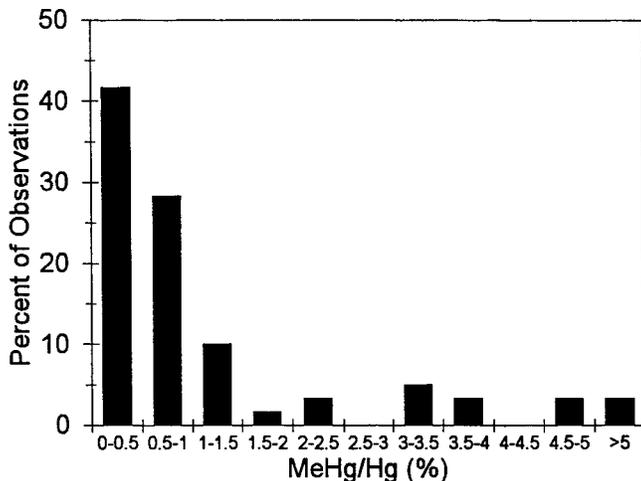


Fig. 9. Distribution of ratio of methylmercury (MeHg) to total Hg in horizons of both mineral and organic soils from Germany (Padberg and May, 1992; Schwesig et al., 1999; Schwesig and Matzner, 2000), Sweden (Hultberg et al., 1994; Munthe et al., 1998), and Ontario, Canada (Moore et al., 1995) ( $n = 60$ ).

tions of MeHg are higher in soil solution near the surface (e.g., Bishop et al., 1998; Branfireun et al., 1996), and near-surface concentrations were higher in peatland discharge zones and lower in recharge and lateral flow zones (Branfireun et al., 1996), all implying that a major source of MeHg is surficial or near-surface processes. Concentrations of MeHg in pore water also change seasonally, increasing from spring to summer (Branfireun et al., 1996). Flux of MeHg in streams also confirms this seasonality, with greatest flux in summer (Babiarz et al., 1998; Bishop et al., 1995; Rudd, 1995), albeit associated with lower flows than during spring snowmelt. The potential of wetlands and especially peatlands to act as sources of MeHg is reported so frequently that it is nearly axiomatic (Grigal, 2002). Over a diverse array of watersheds, there are positive relationships between proportion of wetland and MeHg concentration and flux, but no patterns with watershed size (Grigal, 2002). These empirical relationships do not directly address the mechanistic origin of the MeHg.

Sulfate-reducing bacteria are widely regarded as responsible for the bulk of Hg methylation (Morel et al., 1998; Ullrich et al., 2001). The observations of increased MeHg concentration near the soil surface and in summer and the association of MeHg flux with wetlands are consistent with bacterial sulfate reduction. Further evidence is the increased concentrations of MeHg that follow flooding of reservoirs, presumably stimulated by the decomposition of flooded organic matter (Bodaly et al., 1997). Decomposing plant tissue in wet and saturated sites in Ontario lost both mass and total Hg, but gained mass of MeHg (Heyes et al., 1998). Rates of MeHg production measured in a beaver pond were also higher than rates from sediments of lakes (Driscoll et al., 1998).

All these observations suggest that terrestrial sites of C accumulation and anoxia are likely to be sites of MeHg production. These same sites may also be sites of Hg(II) reduction and volatile loss. Wetlands and even season-

ally wet mineral soils are therefore likely sources of MeHg, which then has the potential to move to the aquatic system. This movement is even more likely because of the spatial distribution of such wet areas. They are often proximate to streams and act as variable source areas (Hewlett, 1982), parts of the watershed that contribute to stormflow and vary in size with the storm and with antecedent conditions. Additional work is needed to both assess MeHg production and total Hg loss in wet parts of terrestrial landscapes, and to place that production in a spatial context so that its contribution to aquatic systems can be ascertained.

## PEATLANDS—A UNIQUE CASE Accumulation

In peatlands, nearly continuous saturation leads to OM (peat) accumulation over hundreds of years (Gore, 1983). Because of the strong Hg-OM relationship, peatlands have the potential to be monitors of historic rates of deposition. In ombrotrophic bogs, which receive all their water and nutrients from the atmosphere, Hg accumulation rates are usually considered equivalent to deposition rates (Benoit et al., 1994). This assumption, however, deserves scrutiny. In Europe (Spain), in an area of Hg mining activity, deeper and older layers of an ombrotrophic bog, dated to about 4000 years before present (BP), have Hg concentrations near  $20 \mu\text{g kg}^{-1}$  corresponding to accumulation rates of about  $3.3 \mu\text{g m}^{-2} \text{yr}^{-1}$  (Martinez-Cortizas et al., 1999). A clear and continuous signal of anthropogenic Hg deposition occurred about 1500 BP. Near-surface concentrations are near  $450 \mu\text{g kg}^{-1}$ , with accumulation rates of about  $87 \mu\text{g m}^{-2} \text{yr}^{-1}$ . These recent rates are much higher than those from the north-central USA (Benoit et al., 1994) and Scandinavia (Jensen and Jensen, 1991). In the USA, the anthropogenic signal in an ombrotrophic bog occurred in about 1900, with prior accumulation rates of about  $7 \mu\text{g m}^{-2} \text{yr}^{-1}$ . Near-surface Hg concentrations, near  $60$  to  $70 \mu\text{g kg}^{-1}$ , yield accumulation rates of about  $38 \mu\text{g m}^{-2} \text{yr}^{-1}$  until about 1980 with a nonsignificant decrease to  $25 \mu\text{g m}^{-2} \text{yr}^{-1}$  in the following decade (Benoit et al., 1994). In seven ombrotrophic bogs in Scandinavia, accumulation rates between  $5$  and  $10 \mu\text{g m}^{-2} \text{yr}^{-1}$  in the early 1800s contrast with average near-surface concentrations of about  $75 \mu\text{g kg}^{-1}$  and rates of  $16 \mu\text{g m}^{-2} \text{yr}^{-1}$  (range from  $4$  to  $35 \mu\text{g m}^{-2} \text{yr}^{-1}$ ; Jensen and Jensen, 1991).

Differences in Hg accumulation among bogs may be due to differences in deposition, but other factors must be considered. Conversion of Hg concentrations to accumulation rates requires determination of concentration and bulk density with depth, and the relationship of depth to time. Even with reasonable quality assurance-quality control (QA-QC) procedures, multiple measurements (e.g., wet to dry weight conversion, bulk density determination, Hg determination, radionuclide analysis for dating) can lead to large uncertainties in the final estimate ( $\pm 73\%$  in older sediments; Gottgens et al., 1999). The assumption that ombrotrophic peatlands are

reliable geochemical archives is based on their spatial homogeneity and on absence of post-depositional Hg migration (Benoit et al., 1994). Post-depositional losses of Hg from bogs can occur through both temperature-related volatilization (Martinez-Cortizas et al., 1999) and fluxes of water. Continental bogs are subject to lowering of water tables of tens to hundreds of centimeters during periodic droughts (Verry, 1980), leading to increased oxygen at depths, increased mineralization, and potential loss of Hg. Alternatively, drier conditions may also decrease both methylation and Hg(II) reduction. Artificial drainage of peatlands increased both MeHg (Westling, 1991) and total Hg flux (Simola and Lodenius, 1982) in streamflow. Inter-bog or regional comparisons of rates of accumulation and inferred deposition to peatlands may also be influenced by differential deposition due to historical differences in surface vegetation.

### Mass

Mass of Hg in peatlands varies and usually differs from that in upland soil in the same region. For example, mass of nongeogenic Hg (derived from atmospheric deposition) to a 60-cm depth in upland soils in Bavaria, Germany, was about 120 mg m<sup>-2</sup>, while that to 70 cm in a nearby fen peatland was only 12 mg m<sup>-2</sup> and to 50 cm in a bog was 8 mg m<sup>-2</sup> (Schwesig et al., 1999). These peats are shallow. The Spanish bog was about 250 cm deep, and preindustrial and modern Hg deposition over about 4300 yr (Martinez-Cortizas et al., 1999) led to a total Hg accumulation of about 140 mg m<sup>-2</sup>, slightly higher than that in the German uplands. In the north-central USA, about 500 cm of peat has accumulated since 4500 BP (P.H. Glaser, University of Minnesota, personal communication, 2000) and preindustrial and modern Hg deposition (Benoit et al., 1994) has led to Hg accumulation of about 30 mg m<sup>-2</sup>. This is more than twice the 12 mg m<sup>-2</sup> accumulation in a shallower (170 cm) peatland in Ontario, Canada, about 400 km away (Moore et al., 1995), and about five times the mass of Hg in the forest floor and surface mineral soil in the north-central USA (about 6 mg m<sup>-2</sup>; Nater and Grigal, 1992).

## SUMMARY AND PERSPECTIVE

### Local Perspective

The relative size of terrestrial Hg pools addressed in this report can be summarized (Table 1). Mass of Hg in the surface mineral soil is more than an order of magnitude greater than that in vegetation, and four to five times greater than that in the forest floor. Mass of Hg in peat is generally greater than the sum of that in mineral soil and the forest floor; upland sequestration of Hg is subject to more pathways of loss than is wetland sequestration.

Assuming steady state, the turnover time of Hg in terrestrial systems can be computed (similar to Olson, 1963). A midrange estimate of current Hg deposition to forests is about 40 μg m<sup>-2</sup> yr<sup>-1</sup> (sum of throughfall plus litterfall) (Grigal, 2002). Turnover time of Hg in

**Table 1. Approximate size of terrestrial pools of Hg at specific sites as documented in this review.**

Pool	Midrange <sup>†</sup>	Maximum <sup>‡</sup>	Location
<b>Vegetation, μg m<sup>-2</sup></b>			
Foliage	20	370	southern Sweden <sup>§</sup>
Wood and bark	95	315	southern Sweden <sup>§</sup>
Roots	30	150	southern Sweden <sup>§</sup>
Coarse woody debris	30	400	northwestern USA <sup>¶</sup>
<b>Soil, mg m<sup>-2</sup></b>			
Forest floor	1.2	17	Germany <sup>#</sup>
Surface mineral soil	5.2	115	Germany <sup>††</sup>
Peat	15	145	Spain <sup>‡‡</sup>

<sup>†</sup> Approximate central tendency of published reports.

<sup>‡</sup> Maximum value reported.

<sup>§</sup> Norway spruce [*Picea abies* (L.) H. Karst.] forest, Gårdsjön, Sweden (Munthe et al., 1998).

<sup>¶</sup> Based on maximum reported mass of coarse woody debris in old-growth forests.

<sup>#</sup> Norway spruce forest, central Europe (Schwesig and Matzner, 2000).

<sup>††</sup> Norway spruce forest, central Europe (Schwesig et al., 1999).

<sup>‡‡</sup> Ombrotrophic bog in northwest Spain (Martinez-Cortizas et al., 1999).

living vegetation is therefore almost 4 yr, in coarse woody debris less than 1 yr, in the forest floor about 30 yr, in surface mineral soil more than 100 yr, and in peat nearly 400 yr. The annual litterfall of Hg-rich foliage from trees results in rapid turnover in the vegetation component. Even Hg present in the bolewood can be released relatively rapidly, by fire, for example. The slower turnover of Hg in the substrate, including the forest floor, mineral soil, and peat, is consistent with the dynamics of OM in those components. Relative to annual inputs, large stores of Hg are present in terrestrial systems.

Focusing on average quantities neglects the importance of spatial distribution. Conditions that lead to differential SOM accumulation, such as variation in vegetation, drainage, and landform position, are likely to lead to differential Hg accumulation. This is clearly demonstrated by Hg accumulation in peat (Table 1). The bulk density–OM relationship also influences Hg. High Hg concentrations in high-OM, minimally decomposed peat and in surface organic horizons in forests are associated with low bulk density, resulting in lower Hg mass than in mineral soil horizons of similar thickness. Higher Hg concentrations occur in the smaller size fractions of mineral soils; the fractions that are also the source of sediment to streams and rivers. Terrestrial flux of MeHg to aquatic systems is influenced by factors other than simply the low amounts of MeHg in vegetation or soil pools. In particular, specific sites in the landscape, especially poorly drained areas near streams, may be important sources of MeHg to aquatic systems.

### National (USA) Perspective

Although extrapolation is always problematic, the close association between Hg and OM allows data to be extrapolated to a national scale (Table 2). Some forests occur on peat so there may be some overlap in the categories of soil and peat in that table; the small area of peat (Table 2) helps mitigate that problem. Anthropogenic emissions of Hg from the contiguous 48 states for 1994–1995 were estimated to be 144 Mg (USEPA, 1997a) and total deposition was estimated to

**Table 2. Estimated Hg storage in forests and in peatlands in the contiguous 48 states of the USA. Estimates for forests based on ratios of Hg to C<sup>†</sup>, and for peatlands on Hg concentrations and peat mass.<sup>‡</sup>**

Component	Area		Hg		
	km <sup>2</sup>	% of USA	Mg	Mmol	mg m <sup>-2</sup>
Forest (sum)	2 452 000	32	30 300	151	12.3
Trees			200	1.00	0.082
Understory			8.7	0.044	0.004
Forest floor			1 350	6.72	0.55
Mineral soil§			28 700	143	11.7
Peat (sum)	105 000	1.4	2 930	14.6	27.9¶
Fibrists	1 970	0.03	43	0.21	21.6
Hemists	19 400	0.3	338	1.68	17.4
Saprists	83 600	1.1	2 550	12.7	30.4

<sup>†</sup> Area and carbon storage in forests from Birdsey and Heath (1995); Hg and C from this review and Nater and Grigal (1992).

<sup>‡</sup> Area and mass of peatlands from Malterer (1996); Hg concentrations with depth from Grigal and Nord (1983).

<sup>§</sup> To a 100-cm depth.

<sup>¶</sup> Weighted average.

be about 79 Mg (USEPA, 1997b). The Hg pool in forests, and primarily in their mineral soils, is more than two orders of magnitude greater than the annual emissions. Even peatlands, less than 2% of total land area, sequester more than 20 times the annual emissions. Because forests only occupy about one-third of the area of the USA, estimated annual deposition to them would be about 25 Mg. Even the relatively ephemeral vegetation component of forests contains almost an order of magnitude more Hg than annual deposition. If deposition is not spatially uniform, and that to forests is increased fourfold to account for throughfall and litterfall (Grigal, 2002), then forest vegetation contains about twice annual deposition. Based on their area, peatlands only receive about 1 Mg deposition per year; the mass of Hg within them is more than three orders of magnitude greater.

These numbers become more significant when evaluated in the context of potential climate change. Climate change can influence land use and forest structure, such as forest type and age class; both have the potential to profoundly influence C storage (Houghton et al., 1999; Gucinski et al., 1993) and by implication Hg storage. Similarly, the intricate feedbacks between climate change and peatland dynamics have the potential to significantly alter their C storage (Gorham, 1991; Hogg et al., 1992). If climate change leads to C loss, then the potential stores of Hg that are available for loss are large (Table 2). Because most loss is likely to be via volatilization to the atmosphere rather than via hydrologic pathways, this could have major effects on the global cycle of Hg.

### Research Needs

There are a many topics that require additional work before we can fully understand Hg in forests and peatlands. Some of the questions recognized through this review include:

- (1) Are there significant differences in Hg accumulation with tree species or forest cover types or geographic areas? Is variation related to soil dif-

ferences, or to location (related to differences in atmospheric concentration)?

- (2) Are significant quantities of Hg stored in coarse woody debris? How labile is that pool?
- (3) Are quantities or concentrations of Hg and MeHg accumulated in terrestrial fauna important for ecosystem health?
- (4) What is the spatial distribution of Hg in soils across the landscape; both its concentration and mass? How does Hg storage in soil vary with landscape drainage? How does the stoichiometry of the Hg-OM relationship vary both laterally and vertically? How much Hg is stored in deeper soil depths (beyond 100 cm), and how labile is it?
- (5) What is the role of variable source areas along stream channels as sources of Hg and MeHg from terrestrial systems?
- (6) Are ombrotrophic bogs reliable geochemical archives for Hg accumulation? What is the fate of Hg stored in peatlands under climate change? How will that Hg respond to increased or decreased precipitation? To higher temperature? Will more Hg be released to streams or will it be volatilized?
- (7) Are terrestrial systems simply conduits of MeHg from the atmosphere to aquatic systems, or are they sources of MeHg? What are the characteristics of terrestrial systems that increase or decrease production of MeHg?

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