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Xylem sap as a pathway for total mercury and methylmercury transport from soils to tree canopy in the boreal forest

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Abstract. Conifer needles are an important link in the cycling of Total Mercury (THg) and Methylmercury (MeHg) in the boreal ecosystem due to the high THg and MeHg concentrations in litterfall. Translocation within the tree of Hg from soils to the crown canopy has been assumed to be a minor source of the Hg in litterfall. This paper, however, is the first to present direct observations of THg/MeHg transport from the soil via xylem sap. Xylem sap concentrations of THg and MeHg were measured in sap drained from different levels along the boles of freshly cut 100 year old Norway spruce (*Picea abies*) and Scots pine (*Pinus sylvestris*). The trees came from a mixed stand growing on podzolized till soils at the Svartberget Forest Research Station in N. Sweden. Soil solution concentrations of THg and MeHg at different levels in the soil profile were measured for comparison.

Concentrations of THg in xylem sap ranged from 10–15 ng L^{-1} in both the Scots pine and Norway spruce. Concentrations of MeHg varied from 0.03 ng $L^{-1}$to 0.16 ng L^{-1}, with higher values in Scots pine than Norway spruce. If these concentrations are representative of the transport from soils to needles in xylem sap at this site, then only 3% of the MeHg in litterfall (0.12 mg ha^{-1} yr^{-1}) and 11% of the THg (26 mg ha^{-1} yr^{-1}) can originate via this pathway. The upward transport via xylem sap is larger relative to the open field inputs (84% of THg and 17% of MeHg). Comparison of soil solution and xylem sap THg/MeHg suggested some degree of THg exclusion during water uptake in Scots pine and Norway spruce, but MeHg exclusion only in Norway spruce.

Introduction

In the boreal forest, most of the total mercury (THg) and methylmercury (MeHg) input to soils occurs in the form of litterfall. Recent budget studies of THg and MeHg inputs to forest ecosystems have shown that litterfall contributes ca half of the THg input to the soil and up to over 80% of the MeHg input (Hultberg et al. 1994; Iverfeldt 1991; Lee et al., in press; Munthe et al. 1995). Litterfall can be enriched by dry deposition of different atmospheric pollutants including Hg. The “dry deposition” of atmospheric

Hg in the forest canopy occurs through different mechanisms, such as surface adsorption of water soluble Hg forms and particulate Hg, Hg^0 uptake through the stomata or surface oxidation of Hg^0 to water soluble forms (Iverfeldt 1991; Munthe et al. 1995). An alternative source of some Hg in litterfall, however, is "recycling" of the soil Hg store.

The high degree to which past deposition of THg and MeHg has been retained in soils, possibly combined with methylating processes, has created a large pool of THg and MeHg in boreal forest soils (Lee et al. 1994; Munthe et al. 1996). Although the mobility and reactivity of the soil Hg and MeHg bound to organic material may be limited, the presence of this store, combined with tree uptake of water via roots, and reduction processes which can generate volatile elemental Hg (Hg^0) create the possibility of Hg mobilization from soils to the tree canopy. Thus when attempting to quantify the input of recent anthropogenic Hg emissions to the boreal ecosystem via litterfall, it is important to assess the degree to which mobilization of the accumulated Hg in the soil from past emissions is contributing to the Hg in contemporary litterfall. (Johnson & Lindberg 1995).

There are several forms in which Hg can be transferred from the soil to the crown canopy. One form is gaseous Hg^0 that evaporates from the soil surface or moves up through the tree bole dissolved in xylem sap. Mercury from the soil reaching the crown canopy as Hg^0, and its reaction products, could be deposited on needles, or assimilated into them, in the same way as recent atmospheric emissions. Measurement of Hg emissions from forest soils in SW Sweden using chamber techniques suggest values of up to 1 ng m^{-2} hr^{-1}, which translates to annual emissions of up to 90 mg ha^{-1} yr^{-1} (Schroeder 1989; Xiao et al. 1991). This value is similar to recent micrometeorological measurements of Hg emission from forest soils at a more temperate latitude (Carpi and Lindberg submitted). These emissions are significant compared to annual THg litterfall inputs in Sweden of ca 200 mg ha^{-1} yr^{-1}. Measurements of Hg^0 emissions in transpiration are more provisional, but suggest that this too could be an important mechanism by which the soil store of Hg re-enters the atmosphere (Hanson et al. 1995).

Mercury can also be transferred from soils to the crown canopy in association with the transpiration process as THg or MeHg moving up through the bole in xylem sap. Most, but not all studies of translocation of Hg within plants suggest that there is little movement of Hg from roots to the above ground portions of plants (Godbold 1994; Lodenius 1994). Some of these studies have found that soils contaminated with Hg did not significantly effect THg in leaves or needles (Cocking et al. 1995; Fukuzaki et al. 1986; Lindberg et al. 1979). Laboratory studies have also found that 95–99% of Hg taken up by the roots stay in the roots (Beauford et al. 1977; Godbold & Hüttermann 1988).

The importance of ambient air concentrations for needle concentrations have, on the other hand, been demonstrated along the deposition gradient around a point source of atmospheric Hg emissions (Tamura et al. 1985).

While these studies give good reason to accept the general picture that translocation of THg and MeHg from the roots in xylem sap is not a significant source of Hg in needles, there is still reason to quantify this process. To begin with, there are several studies by Siegel & Siegel and coworkers (1987) which suggest a relation between vegetation uptake and soil contamination. Field studies have also tended to compare foliar THg to the amount in soils, rather than the concentration in soil solution which is more immediately available for transfer into the root, and may not be directly related to the solid soil store of Hg. Furthermore, few of the field studies have looked at MeHg in either vegetation, solid soil or soil solution despite indications that organic forms of Hg are more susceptible to translocation (Dolar et al. 1971). Finally, the laboratory studies (Beauford et al. 1977; Godbold & Hüttermann 1988) have tended to look at seedlings rather than mature trees, and worked with concentrations much higher than those found in boreal forests.

This study investigated THg and MeHg (though not Hg^0) transport from soils to needles via the xylem sap in several Scots Pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*). The study also compared the xylem sap concentration to the concentrations found in the soil solution of the rooting zone to assess the degree to which the roots can filter out THg and MeHg when taking up water.

Methods

Xylem sap is composed of water taken in by the roots which then moves up the tree bole to the needles where most of the water is transpired. With measurements of THg and MeHg in the xylem sap and an estimate of evapotranspiration, the annual transport of THg and MeHg from soils to the tree canopy can be estimated. Samples of the xylem sap for determination of MeHg and THg were extracted from the tree bole using the displacement method described by Glavac et al. (1990).

In the application of that method reported here, one meter logs were cut from the tree bole with a hand saw. The logs were then taken to the laboratory where a 10 cm wide band of bark and the underlying phloem were removed from both ends of the log to preclude the possibility of contaminating xylem sap with fluid from the phloem. A hand saw was then used to remove a few cm from the bottom of the log to expose fresh wood protected from contamination during transport from the field site.

The prepared logs were clamped into a vertical position, and a rubber sleeve cut from an inner tube was attached to the upper end of the log. Water was poured into this sleeve, creating a pool in the sleeve. A pool just several cm deep creates sufficient pressure on the log's pore structure to displace xylem sap from many conifers, including Scots pine and Norway spruce.

Three trees each of mature Norway spruce and Scots pine, all with their crowns in the dominant canopy layer, were felled on July 31, 1995. Xylem sap was extracted from 1 m long logs in each tree taken from the bole 0.5–1.5 m, and 4.5–5.5 m above ground. The first 100 ml of xylem sap that drained from each log was discarded, and then four 100 ml samples were drained sequentially into acid-washed Teflon bottles. The total amount drained from each log, 500 ml, is ca 5% of the xylem sap in each log. As a check that none of the water added at the top of the log contaminated the xylem sap draining from the bottom of the log, a strong dye was mixed into the water poured into the sleeve at the top of the log, and checked for visually in the drained sap.

The first and third drainage samples were collected for MeHg, while the second and fourth were collected for THg. If contamination was present on the face of the log, a dilution as more xylem sap drains would be expected. The mean difference between the first and second samples was less than 5% for both THg and MeHg (Table 1). This was taken as an indication that the samples were not contaminated at the freshly cut face of the log.

The soil solution from a profile in the stand where the study trees grew was extracted from bulk soil samples of specific soil horizons by centrifuging 100 g portions of soil at 14,000 r.p.m. for 20 minutes. This corresponds to a soil suction between pF 3 and 4 in different parts of the sample. The techniques for this extraction were developed and reported more fully in earlier studies (Bishop et al. 1995).

All Hg samples were collected in Teflon or Pyrex bottles that were acid-washed (24 hours in 20% HNO_3 and 1% HCl). The samples were stored in the dark at 4 °C before analysis. The THg samples were preserved with 1 ml of Suprapure HCl (Merck) per 100 ml of sample. All the THg and MeHg analyses were conducted at the Swedish Environmental Research Institute. Samples for THg analysis were oxidized with BrCl prior to reduction with $SnCl_2$ (Bloom & Crecelius 1983). The Hg was then preconcentrated on a gold trap and analyzed using a double amalgamation helium dc-plasma atomic emission method (Iverfeldt & Lindqvist 1982). Methylmercury was analyzed using GC-CVAFS after aqueous phase ethylation using a distillation preparation step (Bloom 1989; Horvat et al. 1993). The detection limit for THg was 0.05 ng L^{-1}, and 0.03 ng L^{-1} for MeHg. The uncertainty in each individual analysis was 5%, but greater (ca 25%) below three times the detection limit.

Samples with MeHg concentrations below the detection limit were assigned a concentration of 0.01 ng L^{-1}, and an uncertainty of 200%.

Study site

The experiment was conducted at the Svartberget Research Forest (64°14′ N, 19°46′ E) in northern Sweden, 60 km NW of Umeå in a mixed stand of hundred year old Scots Pine (*Pinus sylvestris* L.) and Norway spruce (*Picea abies* (L.) Karst) growing at 260 m above sea level on the lower part of the East-facing slope of a blocky moraine deposit. The soil profile was a typical humoferric podzol. The mor layer was ca. 7 cm thick, and the bleached E horizon was also ca. 7 cm thick. Beneath these layers was the strongly colored Bhs horizon that extended from 7 to 17 cm depth in the mineral soil, albeit with large local variation. The next layer down in the soil profile was the 15 cm Bs horizon. The silt and clay content was ca. 20% in the unsorted till soil. During the experiment, the water table was ca. 70 cm below the soil surface.

An earlier study of tree water uptake used hydrological isotopes to trace the depth distribution of water uptake in trees from this stand during July 1991 (Bishop & Dambrine 1995). The average depth of water uptake by Norway spruce was at 2 cm depth in the soil profile (i.e. 2 cm into the mineral soil of the E horizon), while Scots pine had an average water uptake depth of 12 cm (i.e. in the Bhs horizon).

The mercury deposition at Svartberget in open field precipitation between 1993 and 1995 was 31 mg ha^{-1} yr^{-1} of THg and 0.7 mg ha^{-1} yr^{-1} of MeHg. Litterfall deposition was 239 mg ha^{-1} yr^{-1} of THg and 3.7 mg ha^{-1} yr^{-1} of MeHg (Bishop & Lee 1997). Throughfall data is also being collected, but will be reported at a later date. If input to the soil is considered to be litterfall plus open field deposition, then litterfall comprised 84% of the annual MeHg input, and 88% of THg input to the soil.

Results

Due to cost limitations, only a selection of the xylem sap drained from the tree boles was analyzed initially. A number of consistent patterns emerged from this first selection of samples, but where necessary to clarify uncertainties, further samples were analyzed (Table 1).

The xylem sap concentrations of THg from both tree species (Figure 1) were grouped within a narrow range of concentrations (10 ng L^{-1} to 16 ng L^{-1}), with a mean THg concentration in Norway spruce xylem sap of 12.3 ng L^{-1} and 13.5 ng L^{-1} in Scots pine.

Table 1. Total Mercury and Methylmercury in xylem sap.

Species	Tree #	Tree height (m)	Diameter breast height (cm)	Tree age (yrs)	Sample	First log 0.5–1.5 m height		Second log 4.5–5.5 m height	
						THg ng L^{-1}	MeHg ng L^{-1}	THg ng L^{-1}	MeHg ng L^{-1}
Scots pine	1	14.5	18	108	First	14.7	0.08	–	0.08
					Second	14.5	0.12	–	–
Scots pine	2	14.7	19	107	First	15.5	0.16	–	0.03
					Second	–	–	–	0.01*
Scots pine	3	16.8	23	106	First	9.9	0.13	–	0.10
					Second	10.9	0.12	–	0.07
Norway spruce	4	16.4	21	103	First	12.9	0.05	13.4	0.01*
					Second	–	0.04	14.3	0.03
Norway spruce	5	14.4	20	114	First	10.7	0.01	15.7	0.07
					Second	–	–	–	–
Norway spruce	6	15.5	18	97	First	13.3	–	12.8	0.01*
					Second	–	–	–	0.05
Scots pine	Average					13.5	0.13	–	0.06
Norway spruce	Average					12.3	0.03	14.1	0.04

[*] Methylmercury concentrations below the detection limit of 0.03 ng L^{-1} have been assigned an estimated value of 0.01 ng L^{-1} with an analytical uncertainty of ±200% in the error bars on Figure 1.

The MeHg concentrations were 100 to 1000 times lower, and had a larger relative variation (from below the detection limit of 0.03 ng L^{-1} to 0.16 ng L^{-1}). Although the analytical uncertainty is relatively great near the detection limit, the average MeHg concentration of Scots Pine xylem sap was significantly greater (95% confidence level) than that of Norway spruce xylem sap at the 0.5–1.5 m level in the bole (0.13 ng L^{-1} vs. 0.03 ng L^{-1}), as calculated using Student's *t*-test, and the assumption of a standard deviation in individual measurements of 0.04 ng L^{-1}. At the 4.5–5.5 m level in the bole, the difference between Scots pine (0.06 ng L^{-1}) and Norway spruce (0.03 ng L^{-1}) was not significant.

Soil samples were collected two weeks prior to the felling of the trees to allow soil solution at the time of sampling to be taken up into the xylem sap in the bole. Subsequent analysis of the estimated transpiration rate during those two weeks indicate that it took roughly one week for soil water taken up by the tree roots to reach the level of sampling in the bole. The method for this calculation is presented in Bishop & Dambrine (1995).

During the period between soil sampling and tree sampling, the volumetric moisture content in the mor layer was ca. 10%, and 20–35% in the mineral soil. Samples of soil were bulked from the 7 cm thick mor horizon, 0–7 cm depth (A horizon), 7–17 cm depth (Bhs horizon), 17–35 cm (Bs horizon) and 35–55 cm (B horizon). Soil solution centrifuged from these samples had THg and MeHg concentrations that were highest in the mor layer, and declined rapidly down through the soil profile (Figure 1).

If the concentrations measured in Scots pine and Norway spruce xylem sap on this occasion in July are assumed to be representative of the average concentration of MeHg and THg transported annually from roots to the tree canopy, the upward transport of MeHg and THg via this pathway can be calculated from the annual xylem sap flux. The 15 year average difference between runoff and open field precipitation was 325 mm (365 mm during the 1995 hydrological year from Oct. 1994 to Oct. 1995). This difference is the sum of interception, evaporation and transpiration. The transpiration component was estimated to be 200 mm yr^{-1}. With an average xylem sap concentration from Scots pine and Norway spruce of 13 ng L^{-1} THg and 0.06 ng L^{-1} MeHg, the annual upward transport is 26 mg ha^{-1} of THg and 0.12 mg ha^{-1} of MeHg.

The decline in xylem sap MeHg concentrations with height in the Scots pine boles gave concentrations similar to those of Norway spruce at 5 m height in the bole. Thus the estimated MeHg transport from soil to needles was similar for Scots pine and Norway spruce.

The soil solution concentrations of THg and MeHg were similar to those seen at Svartberget during the summer in previous studies (Bishop et al.

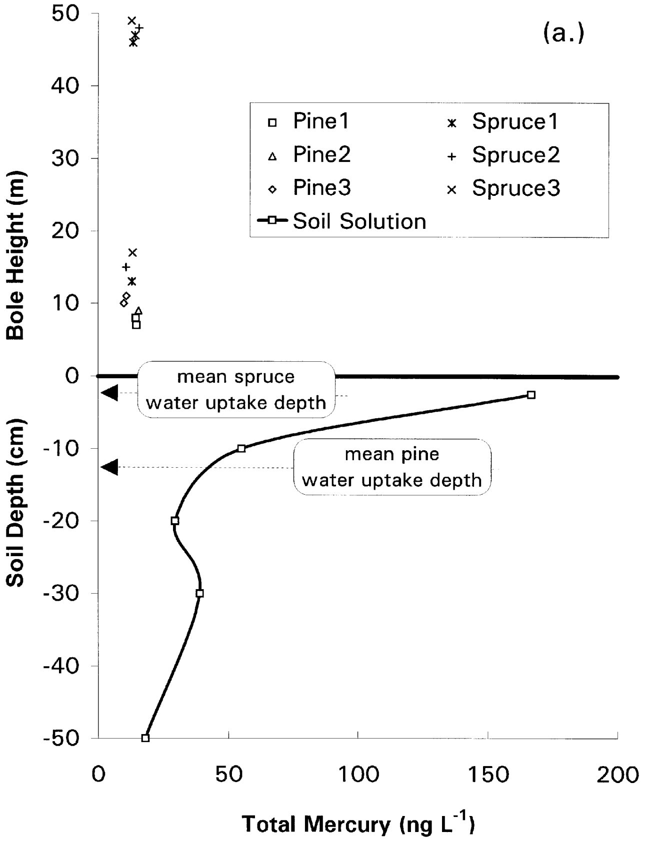

Figure 1. Profile of (a.) THg and (b.) MeHg from soil solution up through xylem sap in the tree bole. Each data point represents a single sample, with the analytical uncertainty in the MeHg measurements demarcated by error bars. (See Methods section for a description of the uncertainty estimates). Please note the different y-axis elevation scales below the soil surface (cm) and above the soil surface (m). The average xylem sap MeHg concentrations at the 1 m and 5 m level in the bole are connected by a line (dashed for Scots pine and solid for Norway spruce). The mean level of water uptake for Norway spruce and Scots pine as determined by hydrological isotopes (Bishop and Dambrine 1995) are indicated.

1995), and the values in water extracted from organic soils are comparable to some values in peat pore water from another region (Branfireun et al. 1996). The rapid THg/MeHg concentration decrease with depth coincides with the decrease in soil solution DOC down through the soil profile. The depth related THg/MeHg concentration decline means that the soil solution concentrations where water is taken up by Norway spruce (2 cm average depth in the mineral soil) are higher than those surrounding Scots pine roots at 13 cm where the mean depth of water uptake by Scots pine was found in the earlier study by Bishop & Dambrine (1995).

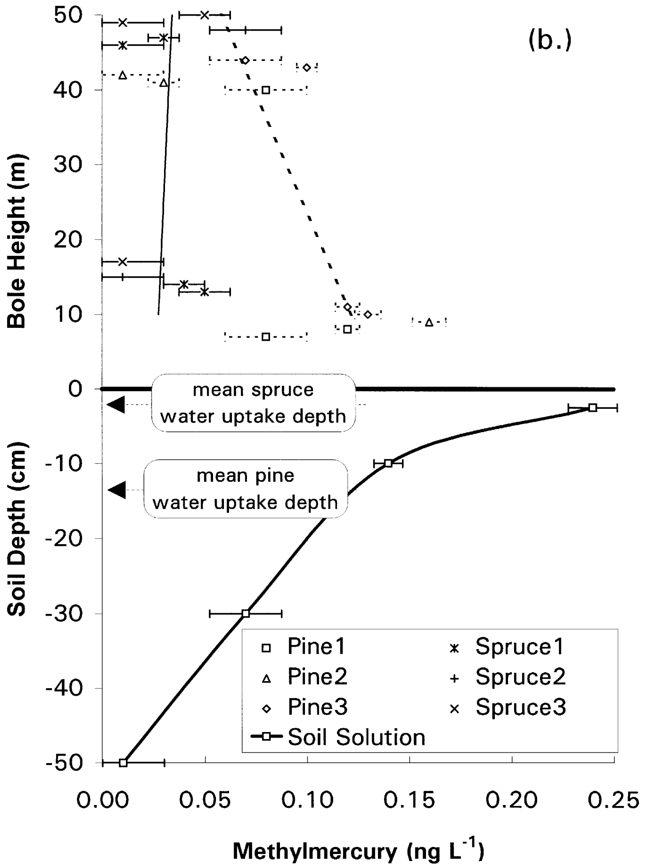

Figure 1. Continued.

Discussion

The major assumption in this experiment is that the MeHg/THg concentration samples on this one occasion are representative of the mean concentration moving upwards in xylem sap through the year, or more precisely during the growing season from May through September. Previous sampling has shown an increase in soil solution MeHg concentrations between July and October by as much as 100% (Bishop et al. 1995). There is also a trend in runoff concentration of MeHg from lowest values during spring flood in April, increasing through the summer and autumn, with flow-related variation (Lee et al. 1995). Total mercury, however, does not have a similar seasonal trend in soil water or runoff.

Thus, in so far as soil water concentrations are proportional to xylem sap concentrations, the values in the July sampling are not expected to be

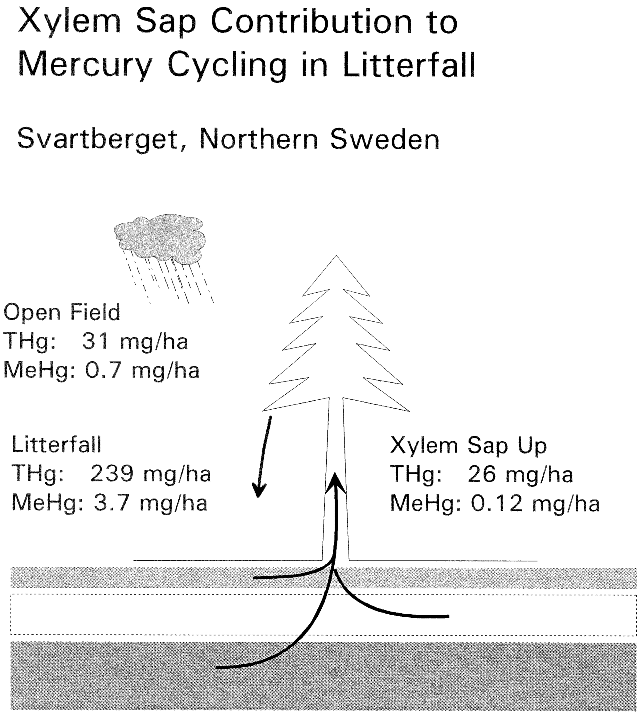

Figure 2. Annual flux of THg and MeHg from the soil to the crown canopy in xylem sap, together with the observed deposition in open field precipitation and litterfall between 1993 and 1995, from Bishop and Lee (1997).

unrepresentative of those during the growing season. July is also the period of maximum evapotranspiration (Eriksson 1986). Finally, even if the volume weighted mean xylem sap concentrations differed by a factor of two or three from those found in July, that would not change the overall conclusion that transport of THg and MeHg in xylem sap is a small component of the THg and MeHg in litterfall.

Despite the exclusion of much of the THg/MeHg in soil solution, however, xylem sap does transmit both MeHg and THg to the tree crown in the boreal forest. The calculated amount of THg going upwards in xylem sap is similar to that coming down in open field precipitation (Figure 2). The amount of MeHg transported by xylem sap is ca 17% of that in open field precipitation.

Comparison of the soil solution to xylem sap concentrations suggest that THg uptake in both species, and MeHg uptake in Norway spruce, is limited by the roots. If THg had been taken up passively so that xylem sap concentrations reflected those in soil solution at the level of water uptake, the xylem sap concentrations would have been considerably higher, especially in Norway spruce which takes up more water closer to the soil surface where soil solution THg concentrations were highest. The presence of an exclusion/regulation mechanism is consistent with the findings of studies on other elements, though the degree of exclusion was not as complete as seen in laboratory studies on mercury uptake (Beauford et al. 1977; Godbold & Hüttermann 1988). The very tight grouping of xylem sap THg concentrations suggests a strong control within the tree on these xylem sap concentrations.

The degree of uniformity in xylem sap MeHg concentration and root exclusion was less than for THg. This suggests a less effective root exclusion or internal control system for MeHg. In the case of Scots pine, the levels in xylem sap just above the ground are similar enough to those in soil solution at the mean depth of water uptake that there may be little exclusion at all, though the concentration of MeHg decreased further up in the tree bole. By contrast, Norway spruce, which takes up more water closer to the soil surface than Scots pine (and thereby from soil solution with higher MeHg concentrations) had generally lower xylem sap MeHg concentrations than Scots pine. The greater degree of root exclusion by Norway spruce roots may be related to a higher degree of mycorhizzal association in the more superficial soil layers.

The low xylem sap MeHg concentrations relative to the analytical uncertainty make it inappropriate to speculate too much on eventual differences in the xylem sap concentration of Norway spruce and Scots pine relative to soil solution. The large difference between litterfall MeHg/THg input and the calculated xylem sap transport, however, contributes to the strength of the central conclusion that xylem sap is a relatively small source of the MeHg and THg found in needles falling to the forest floor.

Determining whether the major source of the large THg/MeHg amount in litterfall is long range atmospheric transport or mobilization of the soil store of Hg via mechanisms other than xylem sap transport of THg and MeHg remains one of the central challenges in quantifying the global cycling of mercury.

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