

# Does forestry contribute to mercury in Swedish fish?



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# Does forestry contribute to mercury in Swedish fish?

Report from a workshop  
at the Royal Swedish Academy of Agriculture and Forestry  
21–22 September 2006



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## Foreword

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*There are three kinds* of environmental pollution. In my point of view they are addressed publicly in a reversed order of importance, which is unfortunate: 1) Plastic, glass, paper and other garbage spread in the terrain – this is mainly an aesthetic problem. 2) Too much nitrogen, phosphorus, potassium and micro nutrients in places where they fertilize the wrong species – mainly a waste of resources. 3) Release of organic and inorganic substances, often man-made (like drugs and pesticides), including poisonous elements and chemicals, that affect biological systems. This often ends up hitting humans negatively and sometimes even dangerously. Increased mercury content in fresh water fish related to forestry activities is an alarm of the third category above, and thus especially important to investigate further.

In September 2006 a workshop was jointly organized by SLU's Department of Aquatic Sciences and Assessment, the Swedish Environmental Research Institute (IVL) and the Royal Swedish Academy of Agriculture and Forestry (KSLA) in the Academy's premises in Stockholm. The workshop was also sponsored by the IUFRO Forest Operations Ecology Research Group on Analyses and Management of Watershed Impacts.

The organizers found that there is an urgent need to:

- Assemble the information that is available about forestry's influence on the mercury problem and thus define the "state of the art".
- Determine what can be done to better answer the questions about what, if any, measures should be taken to limit the eventual effect of forestry on the mercury problem.

The workshop participants shared views and experiences before departing. The outcome was found to be "state of the art" and is here assembled in the Royal Academy's official journal. Sixteen of the contributors have summarized the workshop (pp. 9–23) and eight separate articles discuss the topics from different angles.

Much of the atmospheric deposition of anthropogenic mercury has accumulated in forest soils. This is now being released into streams and lakes, sometimes in association with forestry activities, thus poisoning fish and other creatures. Especially after final felling, but also after thinning operations, the water table is rising and more mercury may be released. Logging in non-frozen wet areas

also has a greater potential to release mercury. Still we have to harvest forest trees, and there are many reasons to try to avoid damage to the soil structure that heavy machinery like forwarders can inflict. Lowering the water table by ditching likely reduces the risk of mercury release, but biodiversity reasons argue against ditching. Few activities in forestry are either totally positive or negative, and we need the products and services from forestry. So we have to look for a good balance.

The obvious result of the workshop is establishing the need for more and better research in order to provide a better basis for making the necessary trade-off decisions on the activities concerning mercury and forestry. But development of less-damaging harvesters and forwarders, along with other technologies and techniques is also needed to mitigate the mercury problem.

Åke Barklund  
Academy secretary and Director

# Introduction

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## Does forestry contribute to mercury in Swedish fish?

Mercury in fish remains both a major environmental problem and a scientific puzzle. Despite most Swedish forest lakes being remote from sources of Hg emissions to the atmosphere, tens of thousands of lakes still have fish with unacceptably high levels of mercury. Although there exists some evidence of a link between atmospheric loading of mercury and methylmercury in fish, other factors such as land use and lake trophic status also influence mercury levels in fish. A major part of the mercury deposited from the atmosphere to the earth's surface is accumulated in forest soils. Consequently, factors which lead to an increased mobilization of this soil mercury to aquatic ecosystems may influence levels of mercury in fish.

Recent research from Sweden, Canada and Finland all indicate a connection between forestry operations and the input of mercury and/or the much more toxic methylmercury species to aquatic ecosystems. While these different lines of evidence are sufficient to raise the question of whether forestry is a significant factor in the problem of mercury in freshwater fish, there is not enough information available in Sweden today to provide a satisfactory answer as to how important forestry is for the mercury contamination in freshwater fish, and what management options might exist to mitigate an eventual contribution to the mercury problem by silviculture.

Due to the sensitivity and importance of this issue, there is an urgent need to:

1. Assemble the information that is available about forestry's influence on the mercury problem and thus define the "state of the art".
2. Determine what can be done to better answer the questions about what, if any, measures should be taken to limit the eventual effect of forestry on the mercury problem.

The Royal Swedish Academy of Agriculture and Forestry (KSLA) hosted a two day workshop addressing these issues that was jointly organized by the Department of Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences (SLU) and the Swedish Environmental Research Institute (IVL). This special issue of KSLA's *Tidskrift* presents the consensus document that resulted from that meeting, as well as summaries of some of the presentations. While many issues remain, we hope that these results of the conference will be a step on the way to a more satisfactory understanding of forestry's role in the unacceptable levels of mercury so commonly found in the fish of boreal lakes, and how to mitigate that problem.

Kevin Bishop  
Swedish University of Agricultural Sciences  
Dept. of Aquatic Sciences and Assessment

John Munthe  
Swedish Environmental Research Institute  
IVL, Gothenburg

## Authors

Kevin Bishop, Swedish University of Agricultural Sciences SLU, Dept. of Environmental Assessment

Craig J. Allan, University of North Carolina at Charlotte, Dept. of Geography and Earth Sciences

Lage Bringmark, Swedish University of Agricultural Sciences SLU, Dept. of Environmental Assessment

Edenise Garcia, University of Montreal, Sustainable Forest Management Network

Sofie Hellsten, Swedish Environmental Research Institute IVL

Andrew Heyes, University of Maryland, Chesapeake Biological Laboratory

Lars Högbom, The Forestry Research Institute of Sweden

Kjell Johansson, Swedish Environmental Protection Agency; SLU, Dept. of Aquatic Sciences and Assessment

Anja Lomander, Swedish Forestry Board

Robert J. Mackereth, Ontario Ministry of Natural Resources, Centre for Northern Forest Ecosystem Research

Markus Meili, Stockholm University, Dept. of Applied Environmental Science

John Munthe, Swedish Environmental Research Institute IVL

Mats Nilsson, Swedish University of Agricultural Sciences SLU, Dept. of Forest Ecology and Management

Petri Porvari, Finnish Environment Institute SYKE

Ulf Skjällberg, Swedish University of Agricultural Sciences SLU, Dept. of Forest Ecology and Management

Rasmus Sorensen, Swedish University of Agricultural Sciences SLU, Dept. of Environmental Assessment

Matti Verta, Finnish Environment Institute SYKE

Therese Zetterberg, Swedish Environmental Research Institute IVL, Gothenburg

Staffan Åkerblom, Swedish University of Agricultural Sciences SLU, Dept. of Environmental Assessment

# Forestry's contribution to Hg bioaccumulation in freshwaters: assessment of the available evidence

KEVIN BISHOP, CRAIG J. ALLAN, LAGE BRINGMARK, EDENISE GARCIA, SOFIE HELLSTEN, LARS HÖGBOM, KJELL JOHANSSON, ANJA LOMANDER, MARKUS MEILI, JOHN MUNTHE, MATS NILSSON, PETRI PORVARI, ULF SKYLLBERG, RASMUS SORENSSEN, THERESE ZETTERBERG, STAFFAN ÅKERBLOM

## Summary

Mercury (Hg) levels are alarmingly high in fish from lakes across Fenno-Scandia and northern North America, with distinct regional hotspots. Long-range transport by air pollution is the ultimate origin of most of the Hg in fish. Most of the Hg that arrives in the rain remains in the soil. This soil retention has protected aquatic ecosystems from the full effect of the Hg in atmospheric deposition. The resulting Hg build up in the soils may be a threat to the soil biota. But it is definitely a threat to fish, since anything that increases rates of transfer from soils to water will make the problem even worse in fish and other aquatic biota. Of the different forms of Hg, methylmercury (MeHg) is by far the most susceptible to biomagnification in the aquatic food chain, and the factors that promote methylation (such as more reducing conditions due to elevated water tables) are of particular concern.

Recent studies have shown that forest harvest operations are associated with increased levels of total Hg ( $Hg_{tot}$ ) and MeHg in run-off and the biota. While only a few relevant studies have been conducted, they are consistent in pointing to forestry operations increasing the total loading of mercury to aquatic ecosystems. In a set of Canadian studies on uptake by the biota, increases in fish and periphyton concen-

trations between two- and fourfold were seen. When it comes to Hg in run-off, the smallest effect of forest harvest on Hg outputs was a doubling of  $Hg_{tot}$  and MeHg outputs. Several other boreal studies found concentration increases in  $Hg_{tot}$  by a factor of two to ten. Increases in MeHg concentrations were generally larger, between four- and twentyfold in a given year. In one case, such increases were associated not with harvest, but only with creation of a temporary skidding road. The duration of increased MeHg concentrations appears to persist for at least five years.

From these studies we estimate that 10–25 percent of the mercury now in the fish of forested, high-latitude landscapes can be attributed to forest harvest. The assumptions underlying this estimate are that forest harvest increases the uptake of Hg in fish by a factor of two to four for a period of ten years, and that one percent of the landscape is harvested each year. If thinning operations also have a similar affect on bioaccumulation, and there is one thinning before final harvest, the figure for forestry's "share" rises to 15–35 percent.

This result should be evaluated in the light of two important considerations. One is that forest operations raise Hg/MeHg outputs to levels that are similar to those seen from wet-

lands. The second is that forest harvest is not what created the elevated Hg levels in the soils. And even if harvest increases the output from soils, we do not know the degree to which other forestry practices might be increasing Hg retention while the forest is growing – for instance due to increased mor layer thickness, or lowered water tables.

When it comes to what can be done to mitigate forest harvest impacts, most of the current recommendations in Sweden appear helpful based on current knowledge, such as leaving riparian buffer zones, reducing stream disturbance with bridges, and avoiding operations in moist areas. Two types of recommendation warrant further consideration. One is the creation of humus traps, since they might promote methylation. Restoration of wetlands and reduction of the degree of drainage will also increase Hg/MeHg loadings to aquatic ecosystems, but the magnitude of the effect will depend on the type of wetland and the local geochemistry.

However accurately these conclusions reflect the current state of knowledge, it must be emphasized that there are very few studies specifically focused on this issue. Therefore there is an urgent need to advance our understanding of how forestry influences the cycling of Hg, its bioaccumulation in aquatic ecosystems, and Hg affects on the soil biota.

## Introduction

Alarming levels of mercury in the fish of many remote forest lakes have long been a concern in Fenno-Scandia and northern North America (Johnels *et al.*, 1967; Björklund *et al.*, 1984; Håkansson *et al.*, 1990; Lucotte *et al.*, 1995). The initial focus in the 1970's and 1980's was on the role of mercury (Hg) deposition and acidification (Johansson *et al.*, 1991). This has given way during the last decade to

an awareness of the role that catchment processes play in methylating Hg and making a part of the atmospherically deposited Hg available for bioaccumulation in aquatic ecosystems (Rudd, 1995; St. Louis *et al.*, 1994; Meili, 1997). This pointed to the potential role that forest management has in the Hg problem:

*The great challenge for science and policy is to define and evaluate the prospective management alternatives, such as riparian management in forestry and reservoir regulation that might mitigate the enduring threat posed by Hg output from catchment soils to surface waters.* (Bishop and Lee, 1997.)

Several studies have since indicated that harvesting and other management/manipulation of forests in the boreal zone increases the loading of methylmercury exported from catchments (e.g. Porvari *et al.*, 2003) or the amount of mercury found in downstream fish (e.g. Garcia and Carignan, 2000). This raises serious questions about the potential role that forestry has played in the disastrous situation we now have, where mercury levels in pike and perch exceed health recommendations for human consumption in more than half of the Swedish lakes (Figure 1), and large parts of North America and Finland.

The issue of forestry's contribution to this problem has attracted the attention of both the international scientific community and governments (Erickson, 2003; Fjeld, 2003; and Finland's Environmental Admin., 2003) as well as the media and NGOs. That concern has been amplified by the forest salvage operations that followed Hurricane Gudrun in Scandinavia (Munthe *et al.*, 2007).

While inorganic Hg ( $\text{Hg}_{\text{tot}}$ ) is the main mercury form in the atmospheric input, the dominant form in fish is MeHg. The transformation

of  $Hg_{tot}$  to MeHg occurs naturally in semi anoxic environments, i.e. water-saturated nutrient-rich zones in peatlands, riparian zones, and sediments (e.g. Meili, 1997). Under humid hydrological regimes, such zones are abundant naturally (e.g. extended wetlands in boreal and tropical regions), or can be created/induced by land use changes such as forest management or damming for hydro-power.

Watershed manipulations can result in an increase of fish Hg levels downstream by an order of magnitude, without any additional input of Hg (e.g. Munthe and Hultberg, 2004). This can be attributed to increased mobilization of existing MeHg from catchments and/or an increased net methylation of natural and anthropogenic Hg stored in soils. (In Sweden the Hg concentrations in the organic layer of forest soils has increased by circa threefold due to anthropogenic deposition [Johansson *et al.*, 1995]). Removal of trees will increase the water table and result in warmer soils during summer/autumn (Rosén *et al.*, 1996). The disturbance associated with logging also increases the hydrological connectivity along superficial flow pathways as well as the output of DOC which is associated with  $Hg_{tot}$ /MeHg.

Thus there is reason to believe that logging could be associated with both increased Hg outputs and increased Hg net methylation. Indeed, several recent studies have shown this. In Canada elevated concentrations of MeHg levels in zooplankton (Garcia and Carignan, 1999)

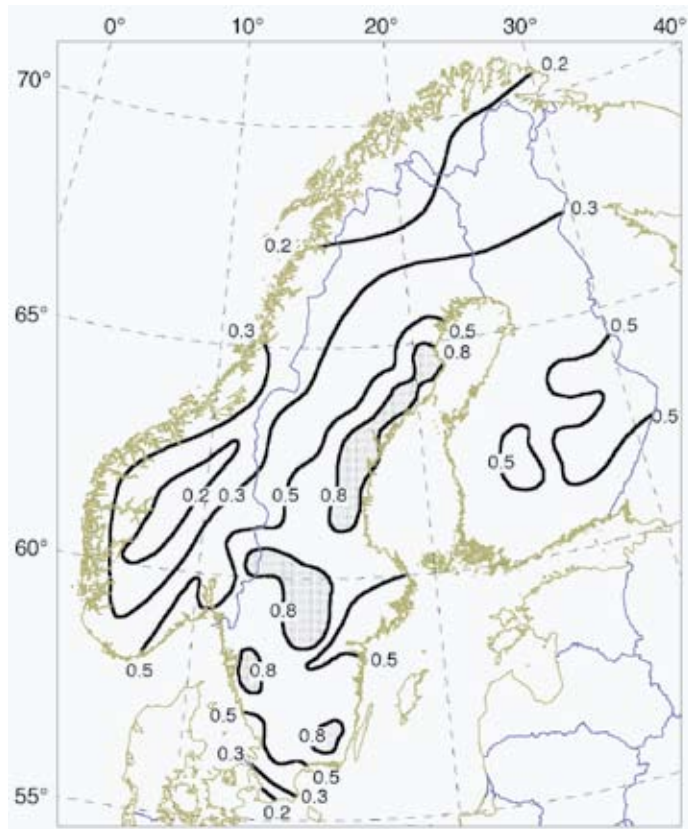


Figure 1. Mercury mg/kg in 1-kg pike (Munthe *et al.*, 2004). The concentration of mercury exceeds the FAO guideline of 0.5 mg/kg in circa half of Sweden's 100,000 lakes.

and  $Hg_{tot}$  levels in pike (Garcia and Carignan, 2000) were observed in lakes with catchments that had been clearcut. In Finland levels of both  $Hg_{tot}$  and MeHg were found to increase markedly in stream run-off after clearcutting (Porvari *et al.*, 2003). In Sweden, the marked increase in run-off MeHg concentrations observed when a temporary skidding road crossed a long term monitoring catchment (Munthe and Hultberg, 2004) attracted a great deal of attention. This also raised the level of public concern about modern forestry, as witnessed by a number of articles in the media (e.g. WWF

letter to Västerbotten Kuriren Feb. 18, 2003, and subsequent responses.)

In the Swedish and Finnish studies, the increased output of MeHg was not limited to the year of the clearcutting but continued for at least five years. There is clearly a justified concern that forestry can contribute to the Hg problem in fish. Furthermore, it could be hypothesized that by minimizing the disturbances in riparian zones where much of the methylation potential lies, the impacts of forestry on methylation and export of Hg can be reduced substantially. With the information available today though, it is difficult to assess the magnitude of that eventual forestry contribution, or the possibilities for mitigating that contribution through altered forest practices.

#### KSLA workshop:

##### Does forestry contribute to mercury in fish?

While different lines of evidence are sufficient to raise the question of whether forestry is a significant factor in the problem of mercury in freshwater fish, there is not enough information available today to provide a satisfactory answer as to whether this is indeed the case. And if so, what management options might exist to mitigate an eventual contribution to the mercury problem by silviculture.

Given the sensitivity and importance of this issue, SLU and IVL organized an international symposium to:

1. Assemble the information that is available about forestry's influence on the mercury problem and thus define the "state of the art" to answer the question 'Does forestry contribute to mercury in freshwater biota?'
  2. Define what can be done to better answer the questions about what, if any, measures should be taken to limit the eventual effect of forestry on the mercury problem.
- Since the focus of the workshop is on

Swedish forestry, KSLA agreed to serve as the official host for the workshop, which brought together researchers, forest owners and managers, government authorities and NGOs. The workshop was held in September 2006. This paper presents the consensus achieved at that workshop. Summaries of individual presentations accompanying this document can be found in this special issue of the KSLA Tidskrift (<http://www.ksla.se>).

#### How much of the mercury in fish can be attributed to forestry?

Given that there are elevated levels of mercury in forest soils, does forestry harvest increase the loading of methylmercury to aquatic ecosystems and thus increase Hg bioaccumulation?

Elevated levels of mercury in fish and aquatic invertebrates after clearcutting were first reported from Canada by Garcia and Carignan (1999, 2000 and 2005), who compared 20 lakes which had unharvested catchments with nine lakes that had been clearcut (Table 1). The Hg in fish higher in the food chain continued to increase several years after the harvest. Later studies in the same area (Desrosiers *et al.*, 2006) also found increases in the Hg concentrations of periphyton. In five of the eight lakes with clear-cut catchments, the increases were statistically significant, with the increase ranging from zero to a factor of eight (Table 2).

These pioneering studies from Canada remain the only ones that have studied the effect of harvesting on mercury in the aquatic biota. The other studies of forestry affects have considered the concentrations and fluxes of  $Hg_{tot}$  and/or MeHg in run-off. The first study that showed increased mercury in run-off was that by Porvari and colleagues in Finland (Table 3). Harvesting of a seven hectare stand increased  $Hg_{tot}$  fluxes by a factor of four and MeHg by a

Table 1. Concentration of total mercury ( $Hg_{tot}$ ) in fish after clearcutting (1996–1998). N.B. concentrations vary with trophic level and size. From García and Carignan (2005).

Common name	Latin name	Trophic level	Standardized length (mm)	$Hg_{tot}$ (mg kg <sup>-1</sup> )	
				Reference lakes (n=20)	Lakes influenced by clearcutting (n=9)
White sucker	<i>Catostomus commersoni</i>	Bentivore	325	0.55–1.23	1.08–2.41
Yellow perch	<i>Pesca flavescens</i>	Omnivore	135	0.68–1.08	0.99–1.51
Walleye	<i>Stizostedion vitreum</i>	Top predator	318	0.98–1.00	1.87–2.78
Northern pike	<i>Esox lucius</i>	Top predator	560	1.00–2.59	2.39–3.58

Table 2. Concentration of methylmercury (MeHg) in periphyton (ng g<sup>-1</sup>). Periphyton refers to algae, bacteria and fungi that are attached to rocks or to the leaves and stems of submerged vegetation. Values in bold indicate significant changes. From Desrosiers et al. (2006).

Lake	Before clearcutting	After clearcutting (year 1)	After clearcutting (year 2)
DF2	1.7 ± 1.3	<b>8.5 ± 3.4</b>	–
DF5	5.2 ± 3.1	<b>10.7 ± 4.9</b>	–
DF7	5.4 ± 4.4	<b>44.9 ± 16.1</b>	–
DF9	7.6 ± 4.3	7.3 ± 2.3	–
K3	4.3 ± 2.4	<b>6.4 ± 2.8</b>	<b>41.2 ± 13.0</b>
K4	7.7 ± 1.6	<b>18.2 ± 3.5</b>	<b>28.4 ± 8.9</b>
K8	6.4 ± 3.2	4.6 ± 2.6	9.2 ± 4.4
DA9	4.2 ± 1.5	15.0 ± 15.3	11.7 ± 8.7

factor of six. The increased fluxes resulted from both a doubling in run-off and concentration increases. It was difficult to distinguish the effect of the harvest itself from the subsequent site preparation. Recent, unpublished data shows that the harvest effect on this site has persisted for over five years (Porvari, pers. comm.).

In Sweden, the effect that simply driving logging machinery through the forest could have was elegantly demonstrated at Gårdsjön where a temporary logging track inadvertently placed across a small, long-term reference catchment

increased outputs of MeHg by a factor of over five (Munthe and Hultberg, 2004) for more than half a decade (Table 3). There has also been an increase in  $Hg_{tot}$ , but the largest effects on  $Hg_{tot}$  were observed during the first year, as was also the case for dissolved organic carbon (DOC).

The large harvesting effect is clear in these published studies. Preliminary results from two newer, as yet unpublished harvest studies were also presented at the workshop. On one, the Balsjö catchment in northern Sweden, harvest

Table 3. Losses of MeHg and Hg<sub>tot</sub> from managed forests. Concentrations are medians if not otherwise specified.

Location	Period	Stream		Stream		Comments
		Concentration (ng l <sup>-1</sup> )		Transport (g km <sup>-2</sup> yr <sup>-1</sup> )		
		Hg <sub>tot</sub>	MeHg	Hg <sub>tot</sub>	MeHg	
S. Sweden						
Gårdsjön <sup>1</sup> , F1 before track	1994–1998	3.56	0.05	2.31	0.03	Forest reference area (3.7 ha) before and after soil disturbance (logging tracks) in the autumn of 1999.
Gårdsjön <sup>1</sup> , F1 after track	1999–2001	4.30	0.22	3.03	0.15	
Gårdsjön <sup>2</sup> , F2	1999–2006	8.1	0.53	–	–	Forest stand (3.3 ha) which was harvested in 1999.
Götaland <sup>2</sup>						
Coniferous forest	1987–2004	4.1	0.12	–	–	Leaching coefficients (median) from coniferous forests and clearcuts.
Clearcut	1999–2006	7.5	0.51	–	–	
N. Sweden						
Svartberget <sup>3</sup>	1993–1998	4.6	0.20	1.6	0.07	Mature coniferous forest (42 ha) and wetland (8 ha).
Balsjö <sup>4</sup>						
Before harvest	2005–2006	5.7	0.3	0.29	0.013	Forest stand (37 ha) harvest in April 2006 and a reference area (20 ha).
Reference	2005–2006	6.4	0.4	0.40	0.025	
S. Finland <sup>5</sup>						
Before harvest	1994–1997	8.13	0.15	0.81	0.02	Forest stand (7.1 ha) harvest in 1997/98. Soil scarification has been carried out in the clearcut.
After harvest	1998–2000	12.02	0.35	3.44	0.13	
Reference	1994–1997	4.90	0.33	0.64	0.03	
Reference	1998–2000	4.74	0.33	0.74	0.04	
Canada <sup>6</sup>						
Ontario						Forest stand clearcut in July 2004. Careful soil scarification on the southern slopes. Average values are shown.
Before harvest	2003–2004	7.19	0.16	7.3	0.016	
After harvest	2004–2006	6.16	0.37	–	–	*) N. B. Values on the load for the year 2004 and 2006 are only based on six months observations.
2004				1.3*	0.083*	
2005				6.2	0.141	
2006				0.6*	0.013*	

1) Munthe and Hultberg, 2004; 2) Munthe *et al.*, 2007; 3) Lee *et al.*, 2000; 4) Sörensen, unpublished data; 5) Porvari *et al.*, 2007; 6) Allan, unpublished data.

had occurred just a few months before the conference, and it is too early to draw any conclusions. Another study in Ontario, Canada, has been underway for several years. In that study, the effects on concentrations of Hg<sub>tot</sub> and MeHg were limited. The increased run-off that resulted from forest harvest, however, doubled the output of Hg from the catchment (Table 3).

The range of responses to harvesting should

not be a surprise given the large variability in the outputs from different catchments (Table 3). But there is a clear indication that forest harvest leads to increases in Hg<sub>tot</sub> and MeHg loading, and increased bioaccumulation in the aquatic biota.

On a broad scale, factors such as wetland area in forested catchments and the DOC concentrations have previously been correlated with

Table 4. Specific output of MeHg from upland areas versus wetland/riparian/discharge areas, in comparison to fluxes from the Gårdsjön F1 catchment.

Region	Site	Period	Specific MeHg output	
			Recharge area g/km <sup>2</sup> /yr	Discharge area g/km <sup>2</sup> /yr
N.W. Ontario <sup>1</sup>	ELA			
	Upland/Wetland 1	1991	0.01	0.18
	Upland/Wetland 2	1991	0.01	0.28
N. Sweden <sup>3</sup>	Svartberget-Kallkällbäcken	1993	0.05	0.89
	Västrabäcken	1993	0.05	0.21
Wisconsin <sup>4</sup>	Allequash Creek	1992–1994	...	0.21–0.55
S.W. Sweden <sup>2</sup>	Gårdsjön			
	F1 catchment	Autumn 1991	0.09	0.24
	F2 catchment	Winter 1991	0.01	0.2

1) St. Louis *et al.*, 1994; 2) Lee *et al.*, 2000; 3) Bishop *et al.*, 1997; 4) Krabbenhoft *et al.*, 1995.

higher levels of Hg in fish (Meili, 1997). The indication that wetlands are effective hotspots of MeHg output from the terrestrial ecosystem has been borne out by studies that separate the output from different parts of the landscapes. Wetlands and especially the organic rich riparian zones commonly found bordering boreal watercourses have much higher outputs of

MeHg, per unit area, than well drained forest soils (Table 4). The difference between outputs of MeHg from organic soils and mineral soils are comparable to those between harvested and unharvested catchments (Figure 2).

### Upscaling from catchments to landscapes

When comparing the range of MeHg outputs from uplands and wetlands, the span is of

the same order of magnitude as the change in output before and after harvest/tracking. But the area of wetlands is relatively constant for a catchment, whereas new areas are impacted by forestry every year.

When thinking of forestry’s impact on Hg in a landscape perspective, we need to scale up from the individual catchment studies. This involves

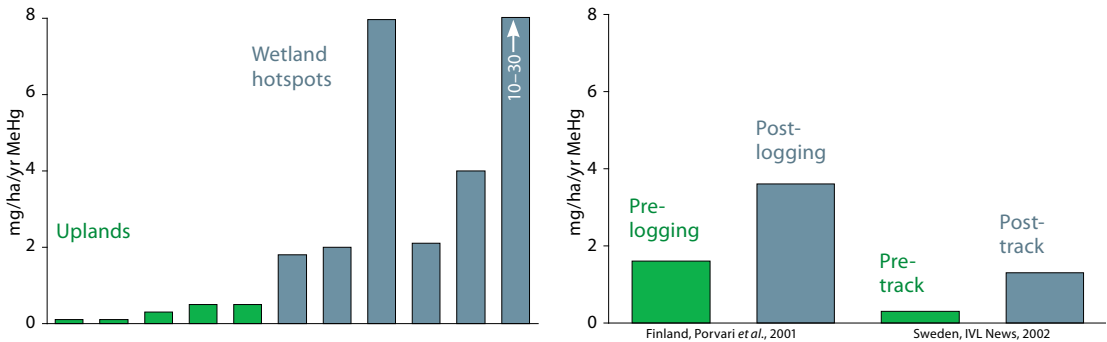


Figure 2. Comparison of the differences in MeHg outputs between wetland/riparian hotspots and upland forest soils on the left (from Table 4), and between logged and unlogged catchments on the right (from Table 3). Note that the scales of the Y-axes are the same.

estimating both the duration of a forestry impact and the areal extent. Assuming a 100 year rotation period, one percent of the productive forest area is harvested each year. The magnitude and duration of that impact are variable, judging from the existing studies. A two- to fourfold increase in loading and bioaccumulation lies within the range of what has been observed. The effect has also been observed to persist up to at least five years, with no examples so far of when the logging effect has disappeared. Thus we will assume a ten year duration of impact. In this situation, Hg loading to surface waters and bioaccumulation will increase to a point where 10–25 percent of the Hg in fish is associated with the increased Hg outputs from final felling. If there is a thinning during the rotation that has a similar impact, then the surface water loading and bioaccumulation will increase by 15–45 percent.

In another landscape scale assessment, Munthe *et al.* (2007) calculated the potential increase in mercury losses to run-off from forest soils across southern Sweden as a consequence of the increased felling of trees resulting from Hurricane Gudrun. The loss coefficients from growing forests and storm-felled sites were calibrated with the help of measured data from earlier studies on conventionally managed forests and harvested forests that were not affected by the storm. The  $Hg_{tot}$  and MeHg from growing forest were estimated to be 4.1 and 0.12 ng l<sup>-1</sup>, respectively. From harvested sites, the loss was 7.5 ng l<sup>-1</sup> for  $Hg_{tot}$  and 0.51 ng l<sup>-1</sup> for MeHg (see Table 1). By comparing the area of felled forest before and after the storm, the estimated increase in total mercury losses was three percent, and 11 percent for methylmercury in Götaland County (Munthe *et al.*, 2007).

#### Assumptions and calculations

The entire catchment is forested and Hg bioaccumulation is directly proportional to output.  
One percent of the landscape is impacted by harvest each year, and will remain impacted for ten years.  
That means ten percent of the landscape is impacted at any given point in time.  
The range of output/bioaccumulation from impacted areas is estimated to be between two to four times that of growing forest.  
The output and bioaccumulation from growing forest is normalized to a value of 1.

Unimpacted output/bioaccumulation from 90 percent of catchment:  $0.9 * 1 = 0.9$

Impacted output/bioaccumulation from 10 percent of catchment:

Low estimate  $0.1 * 2 = 0.2$

High estimate  $0.1 * 4 = 0.4$

Total output<sub>(low estimate)</sub> is  $0.9 + 0.2 = 1.1$ , relative to the output from the unimpacted landscape of 1. The total output<sub>(high estimate)</sub> is  $0.9 + 0.4 = 1.3$ . This means there will be a 10–30 percent increase in output/bioaccumulation relative to a forest landscape with only growing forest and no harvest. Of the Hg in fish, 0.1–0.3 of 1.1–1.3, or 9–23 percent can thus be attributed to forestry.

It must, however, be emphasized that there are many uncertainties in these estimates. They are based on very few studies, and the results of these studies span a large range of impacts. The sites studied may also not be a representative sample of the landscape. For instance the data is largely from sites without wetlands. And even if the data was a fair representation of the landscape, there will be local variations in the forest area impacted, and the sensitivity of the areas. There are also likely to be regional differences, both in operational methodologies, and the susceptibility of the landscape to forest impact – for instance between shallow soils with large rainfall in the southwest of Sweden, and regions with deeper soils and less rainfall in Finland or central Canada. This variable sensitivity is exemplified by the difference between the Ontario study (Table 4) where a clear felling gave little change in concentrations, to the Gårdsjön situation where a single logging road led to a manifold increase in run-off concentrations of MeHg.

There are two more factors to be born in mind as well when evaluating the forestry impact. The first is that it was not forestry that created the excess of mercury that has accumulated in superficial soils as a result of long range transport of mercury. The second factor is that we do not know the extent to which growing, managed forests may (or may not) release less MeHg to aquatic ecosystems than unmanaged forests. We simply have no information on what the mercury concentration would be in an unmanaged forest landscape without harvest.

If the growing, managed forest sequesters more of the atmospherically deposited mercury in the soil, or transforms a smaller fraction of the mercury output to the methylmercury fraction that bioaccumulates, then this could offset the increased loading during the harvest phase. Factors in the managed forest that might reduce

MeHg loading are thicker forest mor layer that “dilutes” the Hg accumulated in that horizon, or the greater depth of oxic conditions in the soil associated with lower water tables in drained forest that is growing quickly.

### Strategies for mitigating the mercury impacts of forest harvest

Given that harvest increases the mercury loading to aquatic environments, it is appropriate to consider the strategies for mitigating that effect. These strategies need to be based on an understanding of the processes that increase methylmercury output in association with forest harvest.

The two most central processes that need to be distinguished when effects of forestry activities are evaluated are: 1) Mobilization and transport of Hg and MeHg, and 2) Net formation of MeHg. There are no detailed studies on these processes in forest soils. Based on findings from sediments and wetland soils, a theoretical discussion about possible effects by forest activities may be useful at this point. These theoretical speculations should be verified or refuted by future studies.

### Mobilization and transport of Hg and MeHg

Both Hg and MeHg are strongly associated to three types of ligands in soil solution, ground and surface waters: 1) halides (Cl, Br), 2) inorganic sulfides ( $\text{HS}^-$  and  $\text{S}^{2-}$ ), and 3) organic thiols ( $\text{RS}^-$ ). In oxidized freshwater systems and soils having measurable quantities of dissolved organic matter (DOM), dissolved Hg and MeHg are almost 100 percent associated to thiols (Skjellberg *et al.*, 2003). Under oxygen deficient conditions, both thiols (associated to DOM) and inorganic sulfides and polysulfides may contribute to the solubility of Hg and

MeHg (Skylberg, 2008). This means that any process that increases the solubility and concentrations of DOM and inorganic sulfides will increase the solubility and subsequent transport of Hg and MeHg.

Possible effects of forestry activities on the mobilization of Hg and MeHg

#### Clearcutting and thinning operations

Both activities reduce the number of trees and thus the evapotranspiration. It is known that clearcutting results in approximately a doubling of the input of water to soil. This means that the water table will move closer to the soil surface. This in turn means that organic rich surface horizons (such as O/A, E, Bh horizons of Spodosols and Inceptisols) will be periodically water saturated. This will inevitably result in a mobilization of DOM and, under certain conditions, a lowering of the redox-potential when oxygen is depleted due to microbial degradation of soil organic matter under water saturated conditions. Inorganic sulfides may form if the redox-potential is low enough. Furthermore, the hydraulic conductivity is higher in the organic rich surface horizons, enhancing lateral transport of soil water. Thus, clearcutting and thinning generally promote  $Hg_{tot}$  and MeHg mobilization and transport from soils to surface waters, in the form of thiol- and inorganic sulfide complexes. Once inorganic Hg/MeHg-sulfide complexes reach surface waters and streams they may be oxidized. Once oxidized Hg and MeHg will be 100 percent complexed and transported by thiols associated with DOM.

#### Ditching and other forestry activities

Because ditching will lower the water table, a rise in redox-potential is likely in surface horizons of soils. This means that inorganic sulfides are not present and all Hg and MeHg will be complexed by thiols in DOM. Shortly after

ditching, concentrations of DOM in run-off are commonly very high. This will lead to an initial, large output of Hg and MeHg associated with DOM to surface waters. In the longer term, however, the solubility of DOM is likely to decrease since the water table is lowered to soil horizons dominated by mineral matter. In these soil horizons adsorption of DOM to mineral soil surfaces control and decrease the solubility and transport of Hg and MeHg to streams and surface waters. Thus, in the long-term, ditching down to mineral soil may lower the mobilization and transport of Hg and MeHg from soils to surface waters. The point in time of break-even between the initial enhanced export of Hg/MeHg and less mobilization in the long run, as compared to a non-ditched control, needs to be determined experimentally.

Liming may affect the solubility of Hg and MeHg indirectly via dissolution of DOM. In highly decomposed organic soils stabilized by aluminum, liming may significantly increase the solubility of DOM (Skylberg and Magnusson, 1995), whereas the solubility of less decomposed organic matter may show little effect after liming.

#### Net formation of MeHg

Transformation of Hg to MeHg is an important process in wetland soils (St Louis *et al.*, 1994) and in freshwater or marine sediments (Benoit *et al.*, 1999). The net production is the result of accidental methylation processes associated mainly to the activity of sulfate reducing bacteria (SRB, Gilmour *et al.*, 1992) and demethylation processes. The methylation process is governed by four major factors: 1) easily available organic compounds like fatty acids and low molecular weight organic acids acting as electron donors for SRB, 2) sulfate acting as the electron acceptor for SRB, 3) high temperature, and 4) dissolved, neutral Hg-sulfide complexes.

Fatty acids are produced after degradation of natural organic matter produced by plants and microorganisms in soils. Sulfate is often available in forest soils but may be limiting the process at highly oxygen deficient conditions. The neutral Hg-sulfides are passively transported over the plasma membrane into the cells of SRB, where the methylation process takes place. The great importance of dissolved neutral Hg-sulfide complexes for net methylmercury production was recently shown for highly contaminated pulp fiber rich sediments (Drott *et al.*, 2007).

Demethylation processes may be both biotic and abiotic. They occur mainly under oxidized conditions and results so far indicate that demethylation rates are less variable and smaller than methylation rates under most conditions in sediments (Lambertsson and Nilsson, 2006). Thus, forestry activities are less likely to effect demethylation rates.

Possible effects of forestry activities on the net formation of MeHg

#### Clearcutting and thinning operations

After both clearcutting and thinning, easily degradable NOM from twigs, leaves, needles, fine roots, etc., boost the activity of various microorganisms. Soil temperatures are enhanced when the solar energy reaches the ground and soil moisture increases. When the ground water table rises and oxygen is depleted, dissolved neutral Hg-sulfides may increase in concentration. If sulfate is not limiting, methylation rates will likely increase. In certain low-lying positions in the landscape with wetland soils, net methylation “hot-spots” may be formed. These hot-spots may even receive increased fluxes of Hg, associated with DOM or inorganic sulfides carried by greater lateral flow rates after clearcut of shallow groundwater.

#### Ditching and other activities

Because ground water levels are lowered by ditching, surface soils are oxidized. This should hamper methylation reactions by lowering concentrations of dissolved neutral Hg-sulfides in soil solution. The effect of liming and soil scarification is essentially unknown. Soil compaction is likely one major effect caused by several forestry activities involving heavy machinery. Soil compaction will decrease the pore volume, enhancing oxygen deficiency and reduced conditions. Thus, soil compaction will likely promote net methylation.

To summarize the key processes, clearcutting and thinning operations may enhance mobilization of Hg/MeHg and production of MeHg by:

- 1) Raising ground water table to surface horizons with a high potential for MeHg production;
- 2) Increasing input of easily degradable plant debris;
- 3) Increasing DOM concentrations in soils and run-off;
- 4) Increasing inorganic sulfide concentrations in soils.

Ditching may enhance demethylation and reduce methylation rates by:

- 1) Lowering ground water tables and oxidizing surface horizons of soils with a high potential for MeHg production;
- 2) In the long run decreasing the mobilization of DOM and thus the transport of Hg and MeHg with DOM.

However, these theoretical discussions need to be verified by experiments in the field and in the laboratory. When it comes to the formation of dissolved, neutral Hg-sulfides available for methylating bacteria, the strong link between iron (Fe) and sulfur biogeochemistry is likely to have a great influence. Terrestrial systems are

most often rich in iron, especially compared to marine systems where iron can be limiting biogeochemical processes. In mineral forest soils, as well as in sediments, high concentrations of Fe may control and lower the availability of neutral Hg-sulfides to a minimum (by formation of FeS) and thus inhibit the transformation of Hg to MeHg. This has been verified in experimental systems of wetlands with added Fe (Mehrota and Sedlak, 2005). In fact, high methylation rates in organic soils may be explained by lack of iron. To complicate the picture even more, iron-reducing bacteria (FeRB) recently have been proven to significantly contribute to MeHg production in sediments (Fleming *et al.*, 2006). It may very well be that iron-reducing bacteria contribute significantly to methylation rates in reduced, minerogenic soil.

Tree species effects on mobilization and net formation of MeHg are unknown. Wetland restoration may lead to increased MeHg production caused by flooding and depletion of oxygen. However, until the effects of iron chemistry on methylmercury production are determined and interpreted on a process level, we can only speculate on the range of response that will be found in different types of wetlands.

### Practical recommendations

The understanding of processes controlling the output of Hg from catchments to surface waters, and subsequent bioaccumulation is not sufficiently developed to confidently recommend ameliorative measures for forestry. While waiting for a better understanding of processes, there is a set of recommendations from the Swedish forestry board (found at [www.svo.se](http://www.svo.se)) to reduce the mercury impact of forest harvest. Several of those measures clearly seem appropriate:

- Avoid soil scarification and transportation of timber in moist and wet areas. Be careful in the riparian zones adjacent to watercourses. Use temporary bridges when watercourses or wet areas have to be passed. Where clear-felled areas are both large and frequent, for example in the area hit by the storm Gudrun, it is extra important to avoid machine tracks. Place the slash so that the water upstream will not be dammed up.
- Avoid creation of large clear-felled areas in connection to moist areas. The larger the clear-felled area, the higher ground water level. Fluctuations of the ground water level in humus rich horizons increase the risk of methylation and leaching of mercury. Wait about ten years before harvest of a nearby site.
- Avoid clear-felling of moist areas. In moist areas, the ground water table is already near the soil surface. Therefore, if possible, leave a shelterwood stand which reduces the increase in ground water table. Avoid soil damage by harvesting when the soil is frozen.
- Collaborate with other land owners within the same catchment area. This may imply finding proper routes on dry land and construction of bridges, which increase the possibility to protect streams, watercourses and moist areas from damage caused by transport. When and where a clear-felling will take place could also be a subject for discussion and coordination, with the goal to keep the proportion of clear-felled area low within any given catchment.

One recommendation that appears sensible, but deserves to be tested is to:

- Clear ditches in such manner that the transport of humus to watercourses is not increased.
- Create humus traps for both new and old ditches. Block old ditches which haven't increased forest production, when such an increase was the reason for digging the ditches in the first place.

While minimizing the concentration of humus could reduce mercury loading to surface waters, the creation of small dams to trap humus may create sites for methylation. Flooding forest for hydropower reservoirs is known to increase mercury methylation and bioaccumulation dramatically (Tremblay *et al.*, 1998). Retaining humus at low flows may also create situations where high flows can liberate large quantities of accumulated humus.

The general recommendation for forest management to reduce the degree of drainage to restore the area of wetlands should also be reconsidered. Wetlands, as noted earlier, generally have higher outputs of MeHg than better drained soils. While the value of more wetlands may outweigh the mercury risks, the presence of a potential goal conflict between the national strategy for “thriving wetlands” and the desire to reduce MeHg in fish should be recognized. Better knowledge of the processes that mobilize and generate MeHg should improve the ability to identify the risk associated with specific restoration projects since the output of MeHg from wetlands will vary with the type of wetland and the local geochemistry.

## Conclusion

Loss of mercury from forested catchments is a significant source of mercury loading to surface waters. The degree of loading varies strongly depending on the characteristics of the catchment. Forest harvest can be a major contributor to this mercury loading. This additional loading can persist for at least five years. The decisive factor for the importance of this additional mercury burden from forestry is how it affects the mercury levels in the downstream biota.

More research and monitoring is needed for a better assessment of the effects of forestry and other alterations of the landscape, including drainage, forest thinning, clearcutting, site preparation, drainage improvement, and wetland restoration. A better understanding of processes is also needed to distinguish the role of increased net methylation from increased mobilization of MeHg from soils. This knowledge is most urgently needed in areas where a large portion of the catchment will be affected, and where the downstream aquatic ecosystem is particularly sensitive to increased loadings.

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# Consequences of the storm Gudrun in Sweden – increased mercury loads to aquatic systems

SOFIE HELLSTEN, JOHN MUNTHE, THERESE ZETTERBERG

Mercury leaching from forest soils to aquatic systems is expected to increase in the south of Sweden following the storm event “Gudrun”, which occurred in January 2005. Methylmercury (MeHg) is the most toxic form of mercury (Hg) in the environment. In most cases, methylmercury constitutes only a small proportion of the total mercury concentration in soil and water. However, methylmercury bioaccumulates in aquatic food chains and therefore nearly 100 percent of the mercury in fish can be in this toxic form. Methylmercury can damage the central nervous system and is particularly harmful to unborn foetuses. The Swedish National Food Administration (Livsmedelsverket) recommends pregnant and breastfeeding women not to consume freshwater fish like perch, pike and eel. Many lakes in Sweden have fish with elevated concentrations of mercury; therefore, preventing mercury from entering the aquatic system is of great importance.

Although the pollution load of mercury to the environment has decreased, mercury has accumulated in soils over time. Forest soils can act as a buffer for mercury, preventing it from entering aquatic systems. However, studies in Sweden (Munthe and Hultberg, 2004) and Finland (Porvari *et al.*, 2003) have shown that forestry operations can increase the leaching of methylmercury from forests to aquatic systems,

as more favourable soil conditions for methylation of mercury to methylmercury are created, and changes in hydrology enhances leaching of methylmercury already present in the soil.

The storm Gudrun caused significant damage to the forestry industry in the south of Sweden. 270,000 hectares of forest was damaged and about 110,000 to 130,000 hectares had to be logged as a consequence of the massive wind-throw. The significant increase in clearcuts and the extensive forestry operations following the storm is expected to increase the leaching of methylmercury from forest soil to streams and lakes. In order to evaluate the effect on the mercury leaching, IVL Swedish Environmental Research Institute carried out measurements in a number of streams in the storm affected area. In addition to the measurements, a theoretical calculation of the leaching of mercury was carried out to evaluate the potential increase of mercury leaching at a regional level, and to identify areas where the effects can be severe. In this calculation it was assumed that the leaching of mercury from the storm af-

Table 1. Leaching coefficients applied in this study for total mercury ( $Hg_{tot}$ ) and methylmercury (MeHg) from forest soil (growing forest and clearcut). Source: Hellsten *et al.*, 2006.

	Growing forest			Clearcut		
	Median	Min	Max	Median	Min	Max
$Hg_{tot}$ (ng l <sup>-1</sup> )	4.1	0.59	36.3	7.5	2.8	19
MeHg (ng l <sup>-1</sup> )	0.12	0.002	6.15	0.51	0.03	11

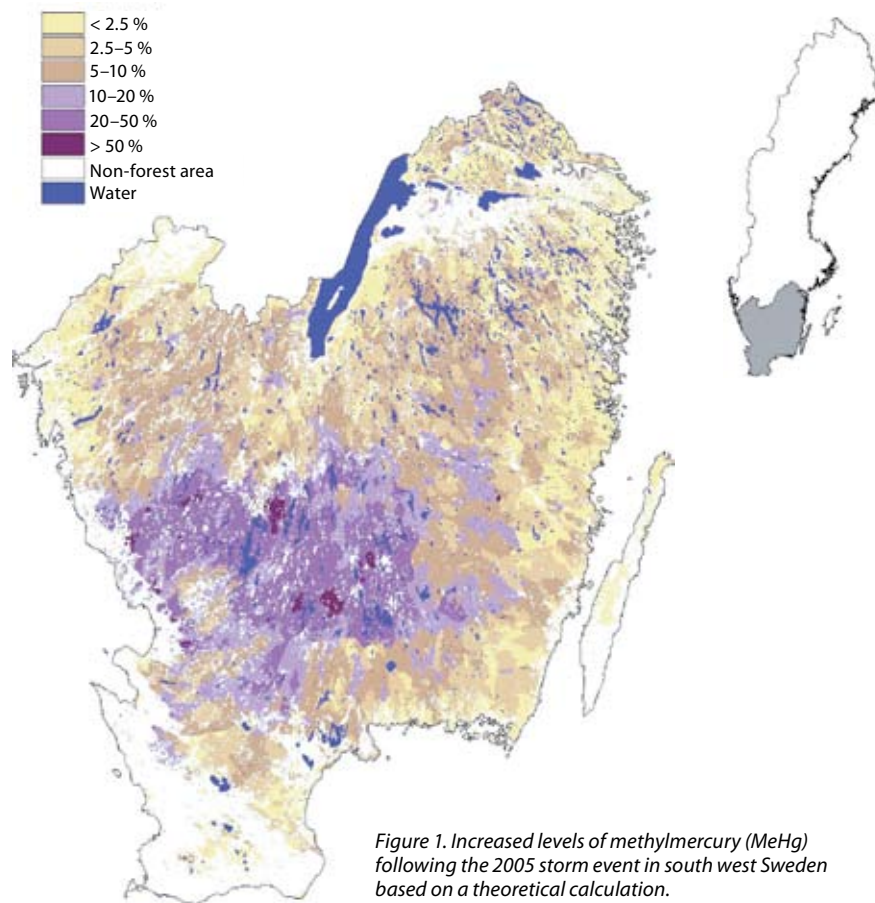


Figure 1. Increased levels of methylmercury (MeHg) following the 2005 storm event in south west Sweden based on a theoretical calculation.

affected areas were of the same magnitude as from conventional clearcuts. Information on storm felled areas, hydrology and typical leaching coefficients (Table 1) were used to estimate the potential increase of methylmercury leaching in the south of Sweden. The leaching coefficients are based on measurements from conventional clearcuts, independent of the storm. However, due to the extensive clearing operations following the storm, where normal precautions could not be carried out, the soil damage, and hence the mercury leaching may have been even

greater. An extensive description of the input data used and the method applied can be found in Hellsten *et al.* (2006).

The results of the theoretical calculation (Figure 1) indicate that the leaching of methylmercury from forest soil can increase by 11 percent in the south of Sweden as an effect of the storm. In some of the worst affected areas, the increase may be more than 50 percent. This effect is expected to be long-lasting, as previous studies have shown that forestry operations can affect leaching of methylmercury for more than

Table 2. Measurements of total mercury and methylmercury in seven storm affected catchments in south west Sweden. Source: Munthe et al. (2007).

Catchment	Hg <sub>tot</sub> (ng l <sup>-1</sup> )	MeHg (ng l <sup>-1</sup> )	Degree of damage
Nissan, Ni12	7.9	1	Small
Nissan, Ni2	9.1	1.9	Moderate
Nissan, Ni4	7.5	1.1	Moderate
Nissan, Ni5	5	0.51	Moderate
Nissan, Ni14	14	3.6	Moderate but large damage in stream
Nissan, Ni10	15	3.4	Severe
Nissan, Ni13	16	3.3	Severe

five years (Munthe and Hultberg, 2004). IVL's measurements in small streams in the storm affected area (Table 2) supported the results of the calculation, as the level of methylmercury in these streams increased in relation to degree of damage (Munthe *et al.*, 2007). In January 2007, another storm "Per", which was almost as severe as Gudrun, caused further damage to the forestry industry in areas that were already badly damaged by Gudrun. It is therefore likely that the mercury loads from forests to surrounding streams and lakes will increase even more.

This study has, in agreement with many other studies, shown that factors disturbing or changing the forest soil and the hydrology can promote methylation and/or increase the transportation of mercury from forest soils to surrounding streams and lakes. Seen from a wider perspective, and considering any potential future climate change effects, such as increased precipitation or more frequent and severe storm events, this study points to the need to further evaluate the effect of these occurrences on the mercury load to aquatic systems.

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# Forest harvesting and Hg accumulation in aquatic organisms from Canadian boreal shield lakes

EDENISE GARCIA

## Introduction

Forest harvesting and wildfires are important disturbances in Canadian forests, affecting each year up to one percent of the boreal forest. Current management forest practices propose that logging and fire have similar impacts on ecosystems and that forestry operations can emulate the effect of natural fires. As part of the Canadian Network of Centres of Excellence on Sustainable Forest Management, a multi-disciplinary study was conducted in 38 boreal shield lakes to compare the effects of both disturbances on water quality and on the ecology of aquatic organisms. The study of the effects of forest harvesting and wildfires on mercury bioaccumulation and biomagnification was part of this network and is presented here.

In forested areas, mercury tends to accumulate in the upper organic layer of the soil. Therefore, major watershed disturbances such as forest harvesting and wildfires resulting in the loss of forest cover and consequently in soil exposure may alter Hg export rates to lakes and influence in-lake reactions that determine Hg fate. For example, clearcut logging increases run-off and the transport of dissolved organic carbon (DOC) and Hg, and can stimulate the bacterial production of methylmercury (MeHg), the most toxic form of Hg. The activity of heavy machinery during logging can also create depressions in the catchment where prevalent anoxic conditions favor the bacterial methylation of Hg. Wildfire may produce a

nutrient pulse in lakes, increasing phytoplankton biomass and influencing the availability of MeHg to the biota. Forest fires may also reduce the pool of Hg in catchment soils through elemental Hg volatilization to the atmosphere. The objective of this study was to determine and compare the effects of forest clearance by wildfire and logging on MeHg contamination of zooplankton and fish.

## Study lakes

This study was conducted in 38 lakes located on the Canadian Shield in central Quebec. Nine lakes had 9–72 percent of their catchment clearcut in 1995, whereas another nine lakes were impacted by high intensity fire in this same year. Seven of these lakes had around 100 percent of their watershed burnt and in two lakes 25 percent and 50 percent of the catchment area were not affected by fire. The catchments of 20 reference lakes have remained undisturbed for at least 70 years. The 38 drainage lakes were selected on the basis of comparable size, depth, and catchment morphometry. Wetlands, which are known as important sites of MeHg production, were present in the catchments of 23 lakes, and occupied  $1.8 \pm 0.4$  percent of the catchment area on average.

## Zooplankton

Zooplankton were chosen because they repre-

sent an important source of food for small, juvenile or pelagic fish. Additionally, these short-lived invertebrates respond very quickly to alterations in the water quality. Zooplankton samples were collected near the deepest point in each lake, from 1996 to 1998, in spring (within two weeks of ice-out in late May to early June), in mid-summer (July), and in late summer (late August to mid-September). Samples were divided into two subsamples: one for MeHg analyses and other for taxonomic identification. MeHg is known to magnify through food webs, therefore the presence of predatory species would be expected to increase MeHg levels in a bulk sample of zooplankton. Inversely, immature individuals would be expected to accumulate less MeHg than adults, thereby influencing negatively MeHg levels in the whole sample. Taxonomic analyses indicated that the samples collected in the three groups of lakes were quite comparable in terms of zooplankton composition and proportion of different taxonomic groups.

MeHg concentrations in bulk zooplankton in reference lakes ranged from 22 to 202 ng·g<sup>-1</sup> dry mass. In forest harvested and fire-impacted lakes, MeHg concentrations varied from 35 to 240 ng·g<sup>-1</sup> dry mass and from 17 to 377 ng·g<sup>-1</sup> dry mass, respectively. Mean MeHg concentrations in zooplankton from forest harvested lakes were significantly higher than in reference and in fire-impacted lakes during mid-summer, in the three study years, as well as in spring and late summer 1998 (Figure 1). Differences

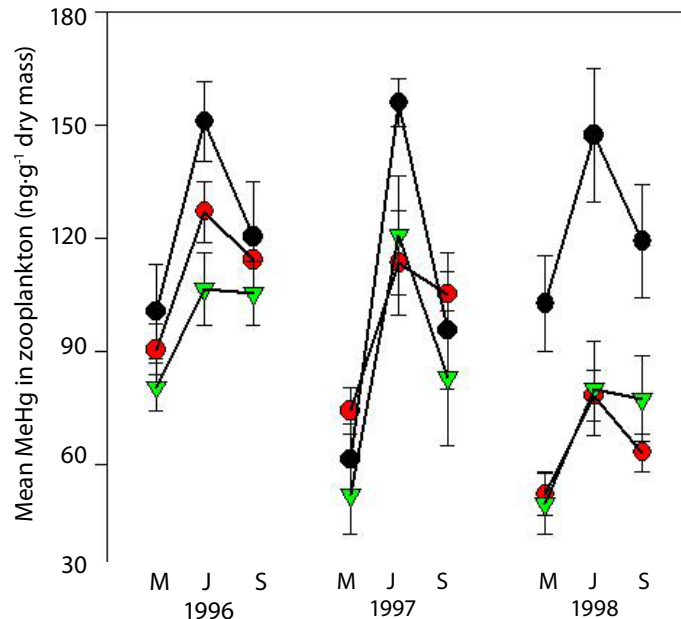


Figure 1. Average methylmercury concentrations in zooplankton from reference (green triangles,  $n = 20$ ), forest harvested (black circles,  $n = 9$ ) and fire impacted lakes (red circles,  $n = 9$ ), in May (M), July (J) and September (S) 1996, 1997 and 1998. Error bars correspond to standard error (Garcia and Carignan, 1999).

in MeHg levels between fire-impacted and reference lakes were not significant at any study period.

Although the observed among-group differences in MeHg rely on a comparison of post treatment data rather than on a comparison of pre and post treatment, there are clear indications that forest harvesting influences MeHg concentrations in zooplankton. Indeed, a significant correlation between MeHg in zooplankton and deforested catchment area (DEF) normalized to lake volume (VOL) was observed in forest harvested lakes ( $r = 0.60$ ,  $p < 0.01$ ). The DEF: VOL ratio has been shown in turn to explain 86 percent of the variability of DOC in the forest harvested lakes (Carignan *et al.*, 2000). These findings are supported by those

observed in a before and after-logging study also conducted in boreal Canadian Shield lakes (Desrosiers *et al.*, 2006), which showed an increase in MeHg concentrations in periphyton mats one or two years following clearcut relative to pre-logging values.

Because Hg tends to associate with humic substances in the upper layer of the soil, the impact of logging on the bioavailability of Hg in aquatic systems may be initially controlled by an increased co-transport of Hg-bound to organic matter, as suggested by the higher DOC concentrations in forest harvested lakes relative to reference lakes, in all periods. In addition, the positive relationship between MeHg levels in zooplankton and DOC loading to lake water or DOC concentration in the three groups of lakes ( $r$  between 0.30 and 0.76,  $p < 0.05$ ) supports this mechanism. Besides affecting Hg speciation by stimulating the bacterial methylation of inorganic Hg, inputs of organic matter in lake water can also decrease MeHg photodegradation rates (Sellers *et al.*, 1996). Additionally, decreased photoproduction of volatile dissolved gaseous mercury, and thus reduction of mercury evasion has been observed in forest harvested lakes (O'Driscoll *et al.*, 2004). Other factors affecting methylation rates and MeHg availability, such as sulfate concentrations and pH, could also explain part of the variability in MeHg in zooplankton observed among the three groups of lakes

Even if fire affected a larger proportion of the watershed area of most lakes (> 90 percent) than did logging, significant differences in MeHg concentration in zooplankton were not observed between fire-impacted and reference lakes. High intensity fires may have reduced the Hg pool by volatilization (Sigler *et al.*, 2003). Alternatively, high phytoplankton biomass in burnt lakes (Planas *et al.*, 2000) may have caused a biological dilution of the available Hg, as sug-

gested by the significant negative correlation between chlorophyll *a* and MeHg observed during the first year following fire. Likewise, observed increases in biovolume of crustacean organisms in the same group of burnt lakes (Patoine *et al.*, 2002) may have resulted in a growth dilution effect.

## Fish

Between six and 23 specimens of up to the three most abundant species in each lake were selected to maximize the range in size of the different species. A total of 642 individuals distributed in 63 fish populations were analyzed. A boneless, skinless filet of dorsal muscle was removed from each fish and used for total Hg determinations. The most abundant species collected were northern pike (*Esox lucius*), wall-eye (*Stizostedion vitreum*), yellow perch (*Perca flavescens*), white sucker (*Catostomus commersoni*), whitefish (*Coregonus clupeaformis*), lake trout (*Salvelinus namaycush*), brook trout (*Salvelinus fontinalis*), and burbot (*Lota lota*). Because Hg increases with fish size, only Hg concentrations in fish of a standardized length were used for inter-lake comparisons. We regressed Hg concentrations for each species in each lake against fish total length (average  $r^2 = 0.85$ ), and estimated Hg concentrations in fish of a standardized length, which corresponded to the mean total length for all individuals from a given species. The adopted standardized lengths were: 560 mm for Northern pike; 318 mm for walleye; 135 mm for yellow perch; 325 mm for white sucker; 310 mm for lake whitefish; 365 mm for lake trout; 240 mm for brook trout; and 590 mm for burbot.

Mercury concentrations ranged from 0.2 to 20.2 mg/kg dry mass all fish sizes and species together. For all species, average Hg concentrations in fish from harvested impacted lakes were

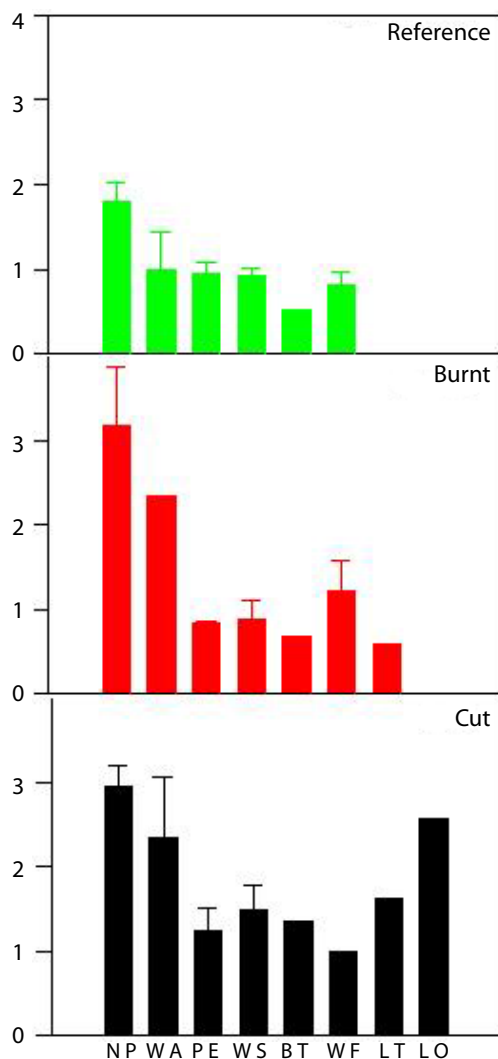


Figure 2. Average mercury (Hg) concentrations in different species of fish of a standardized length from lakes with logged, burnt, or undisturbed catchments. Error bars correspond to standard error. BT = brook trout (*Salvelinus fontinalis*); LO = burbot (*Lota lota*); LT = lake trout (*Salvelinus namaycush*); NP = northern pike (*Esox lucius*); PE = yellow perch (*Perca flavescens*); WA = walleye (*Stizostedion vitreum*); WF = lake whitefish (*Coregonus clupeaformis*); WS = white sucker (*Catostomus commersoni*). Note that Hg concentrations are reported on a dry-mass basis, and should be multiplied by 0.2 in order to obtain concentrations on a wet-mass basis (Garcia and Carignan, 2005).

higher than in fish from reference ones (Figure 2), with the differences ranging from 25 percent (whitefish) to 140 percent (walleye). Mercury levels in fish from forest harvested lakes also tended to be higher than in fire impacted lakes for all species excepting top predators northern pike and walleye (Figure 2). These two species were the only ones from fire impacted lakes that showed significantly higher concentrations than fish from reference lakes.

Despite the higher Hg concentrations in fish from forest harvested lakes, among-group variations in fish trophic position rather than watershed disturbances could be responsible for the observed differences. As mentioned before, MeHg concentrations tend to increase with fish trophic position, which in turn varies with food chain length. Therefore, the same species of fish may occupy different trophic positions in different lakes, depending on the number of species present in the lakes. In this study, stable isotopes of nitrogen corrected for baseline variations were used to determine the trophic position of fish. This approach is based on the enrichment in  $^{15}\text{N}$  from prey to predator ( $\delta^{15}\text{N}$ ) because of the preferential excretion of  $^{14}\text{N}$ . In summary, the higher the  $\delta^{15}\text{N}$ , the higher the trophic position occupied by a given species. We then compared the relationships between Hg levels and fish trophic position in both disturbed and undisturbed lakes (Figure 3).

Mercury concentrations in fish from the three groups of lakes increased with  $\delta^{15}\text{N}$ . The highest Hg levels were observed in predatory species, namely northern pike, walleye and burbot, whereas fish occupying the lowest trophic levels, such as the benthivorous white sucker and the planktivorous whitefish tended to show relatively low Hg concentrations. The slopes of the regressions did not vary significantly (ANCOVA,  $F = 0.06$ ,  $p = 0.81$ ), which indicates that the biomagnification of Hg along the

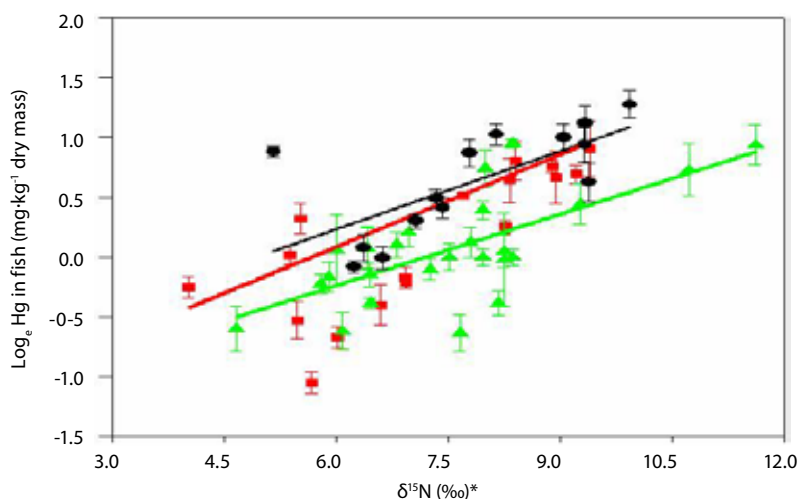


Figure 3. Relationships between mercury concentrations and fish  $\delta^{15}\text{N}$  for reference lakes (green triangles and line), and for lakes with cut (black circles and line) and burnt catchments (red squares and line). The  $\delta^{15}\text{N}$  values are corrected for baseline variations. Error bars correspond to standard deviation (Garcia and Carignan, 2005).

food chain was similar in the three groups of lakes. However, variations in the intercept were significant ( $p < 0.0001$ ), with the highest values being observed in forest harvest lakes. These results suggest that Hg availability to organisms at the base of the food chain in forest harvested lakes was higher than in reference and fire impacted lakes. From the back transformation of logarithmic values showed in Figure 3, it was estimated that, in average, Hg concentrations in fish from clearcut lakes were 43 percent and 48 percent higher than in fish from reference and fire impacted lakes, respectively, occupying an equivalent trophic position. Similarly to zooplankton, mercury in fish normalized for  $\delta^{15}\text{N}$  from cut lakes showed a significant correlation with the ratio of clearcut area to lake volume ( $r = 0.82$ ,  $p < 0.001$ ) and the ratio of clearcut area to lake area ( $r = 0.74$ ,  $p < 0.01$ ). In addition, Hg in fish in the pooled lakes showed a significant positive correlation with DOC

( $r = 0.41$ ;  $p < 0.001$ ) and was negatively related to pH ( $r = 0.38$ ,  $p < 0.01$ ).

Among the fire impacted lakes, Hg levels tended to be significantly higher in fish from the two lakes with partially burnt watershed, comparatively to those that had their watershed completely burnt. In fact, Hg concentrations in fish from these two lakes were the highest among all study lakes. It seems that partial loss of forest cover through fires may result in elevated Hg levels in the biota.

Overall the results presented here indicate that forest harvesting activities may increase Hg levels in the aquatic biota. The disruption in Hg cycle following clearcutting may provoke an almost immediate and relatively long lasting effect in terms of MeHg accumulation at organisms occupying lower trophic levels, such as zooplankton. Given that MeHg tends to be biomagnified along the food chain, high and long-lasting concentrations of MeHg in or-

ganisms at the base of the food chain have an important effect on the accumulation of MeHg in fish, particularly in top predatory species. Indeed, a significant relationship was observed between MeHg levels in zooplankton and total Hg concentrations in northern pike (Figure 4). Therefore, the presence of water bodies with intensive fishing activities should be taken into account in forestry management plans.

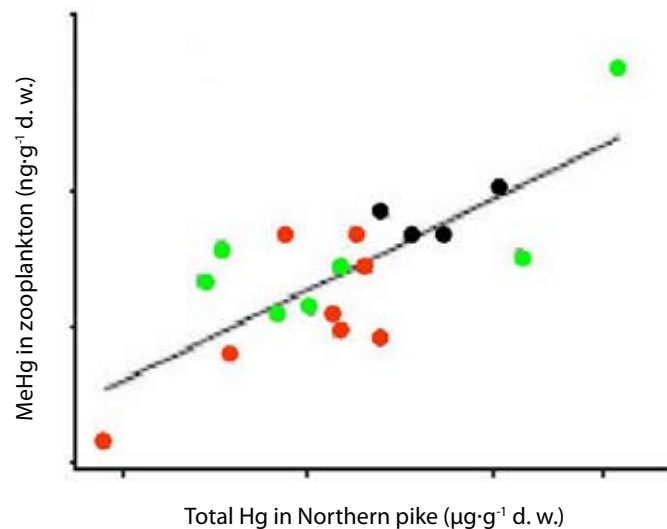


Figure 4. Relationship between Hg concentrations in 560-mm northern pike and MeHg concentrations in zooplankton, expressed on a dry weight basis ( $r = 0.79$ ,  $p = 0.0000$ ,  $n = 19$  lakes). Black circles = harvested lakes ( $n = 4$ ); red circles = fire impacted lakes ( $n = 7$ ); green circles = reference lakes ( $n = 8$ ) (Garcia and Carignan, 2000).

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# Forestry practices cause extreme mercury and methylmercury output from boreal forest catchments

PETRI PORVARI, MATTI VERTA, JARMO LINJAMA, JOHN MUNTHE

## Synopsis

We observed significant increases in the run-off export of total mercury ( $Hg_{tot}$ ) and methylmercury (MeHg) from a spruce forest catchment after clearcutting. Here we show that clearcutting, combined with site preparation prior to planting, may act as an important additional source of  $Hg_{tot}$  and MeHg for forest lakes.

## Boreal forests as sources of $Hg_{tot}$ and MeHg

Hg accumulates efficiently in boreal forest soils. Increased concentrations in the mor layer are a result of atmospheric deposition. Although many investigations have identified the accumulation in forest soils as a potential risk for increased loading to surface waters, very little is known about the factors controlling the mobility of  $Hg_{tot}$  and MeHg in soils.

In boreal forests, timber harvesting is usually carried out as regeneration felling or thinning. Regeneration felling in boreal spruce stands normally takes the form of clearcutting. The surplus of  $Hg_{tot}$  and MeHg output loads from logged boreal forests might therefore result in elevated MeHg concentrations in fish. At least a significant increase in the  $Hg_{tot}$  and MeHg load from catchments to lakes for a number of years is to be expected. Human exposure to highly toxic MeHg through the consumption of fish is the principal public health concern with Hg in the environment.

## Study design

The treatment site was a small (0.071 km<sup>2</sup>) Norway spruce forested catchment situated in Janakkala, southern Finland (61°01'N, 24°45'E). The landscape of the site has low elevation with maximum of circa ten metres' difference of elevation inside the catchment. The soil of the catchment is dominated by coarse sand moraines with a thin humus layer (circa five centimetres) on the top and only circa five percent peatland. The stand mainly (98 percent of growing stock volume) consisted of mature Norway spruce and the rest of deciduous tree species (two percent). The mean age of trees was 76 years in 1995. The upper part of the catchment (eight percent) had a seedling stand of Norway spruce planted in 1990.

The site was clearcut in autumn 1997. A buffer zone of 20 metres' breadth and 50 metres' length was left on both sides of the outflow stream. The outflow stream is a man-made dike of circa 100 metres' length. One year after clearcutting, the soil was treated to promote the growth of newly planted trees by a process called mounding, in which the soil is turned over with a large plow into raised mounds so that it is drier and warmer.

The effects of the forestry practices were studied by comparing  $Hg_{tot}$  and MeHg concentrations in water samples of outflow stream water and  $Hg_{tot}$  and MeHg output fluxes before (1994–1996) and after the treatments (1998–2005).

## Concentrations

Before the treatments  $\text{Hg}_{\text{tot}}$  ( $8.0 \pm 4.0 \text{ ng l}^{-1}$ ) and  $\text{MeHg}$  ( $0.22 \pm 0.37 \text{ ng l}^{-1}$ ) concentrations and the output fluxes were of the same level as in studies carried out in forest catchments with upland and wetland sites in boreal zone.

After the treatments the annual flow-weighted  $\text{Hg}_{\text{tot}}$  concentrations ( $10.5\text{--}17.2 \text{ ng l}^{-1}$ ) were 1.1–2.1 times higher and  $\text{MeHg}$  ( $0.41\text{--}$

$3.18 \text{ ng l}^{-1}$ ) 2.3–18 times higher than before the treatments. Total organic carbon (TOC) concentrations of run-off water peaked during the first year after clearcut, and then returned to the same level as during the calibration period.

## Mercury loads and water discharge

The monthly  $\text{Hg}_{\text{tot}}$  and  $\text{MeHg}$  output loads, as well as water discharge were all significantly higher after clearcutting. The mean annual loads and discharge were circa 1.5 times higher for  $\text{Hg}_{\text{tot}}$ , seven times for  $\text{MeHg}$  and two times for water discharge after the treatments.

## Annual loads

After the treatments the annual  $\text{Hg}_{\text{tot}}$  output loads were 2–7 times greater and  $\text{MeHg}$  output loads 5–52 times greater than before the treatments.

## Causes for leaching

The changed hydrological conditions, i.e. saturated soils and increased water discharge, had a major impact on the  $\text{Hg}_{\text{tot}}$  and  $\text{MeHg}$  output. Clearcutting decreases the amount of evapotranspiration from the forest and therefore increases the amount of water in the watershed. The soil becomes saturated, and soil that was formally aerobic be-

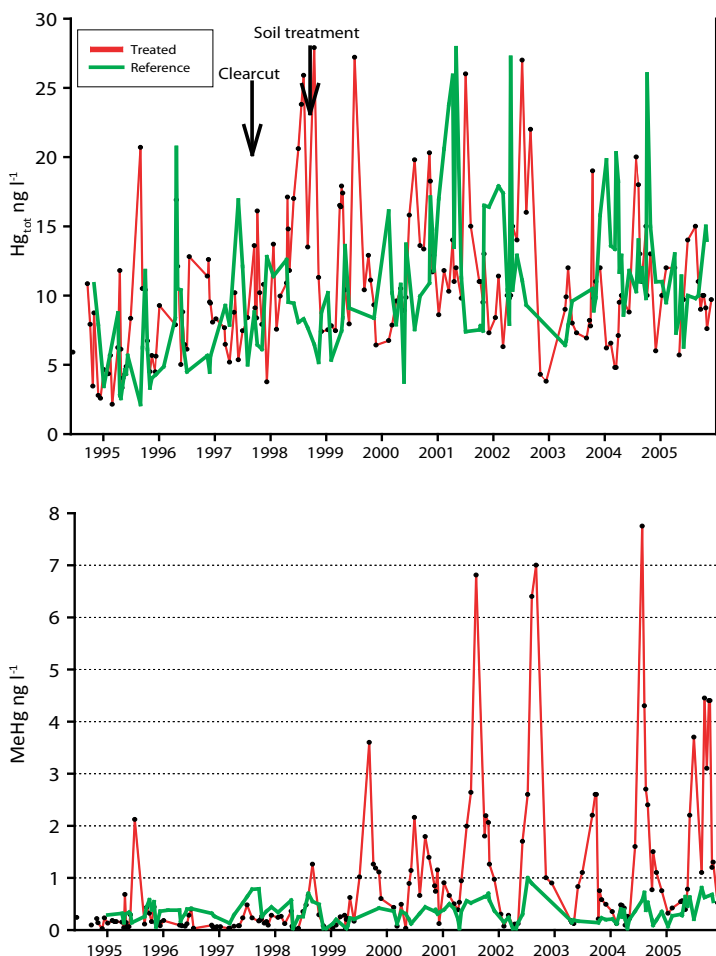


Figure 1.  $\text{Hg}_{\text{tot}}$  and  $\text{MeHg}$  concentrations in outflow stream of treated and reference catchments.

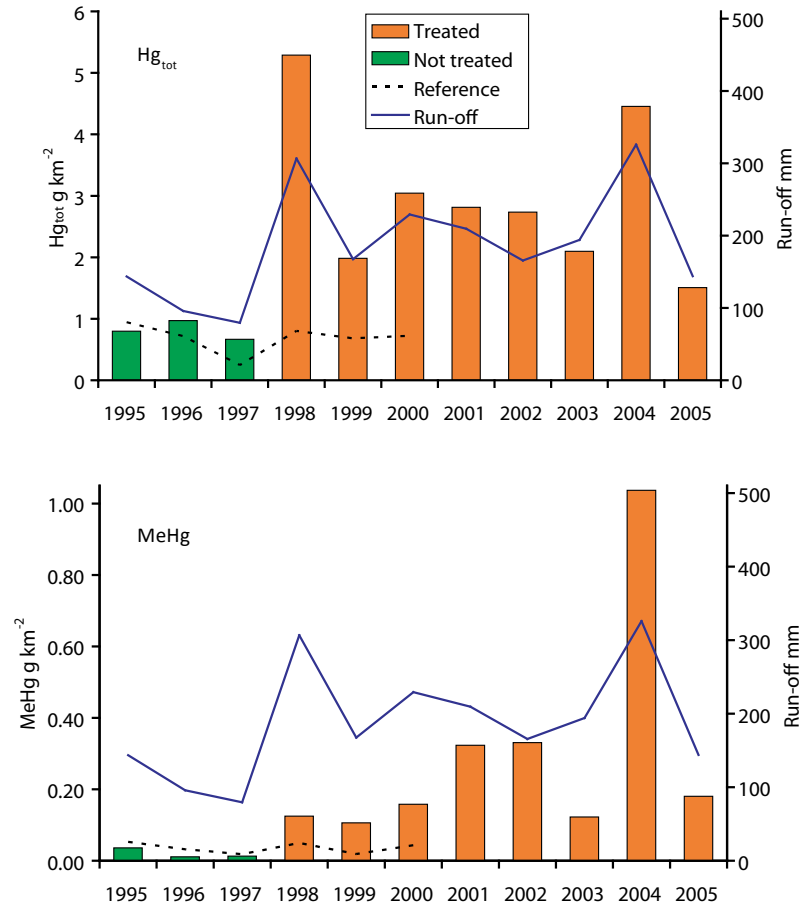


Figure 2. Annual run-off and output loads of  $Hg_{tot}$  and MeHg from the treated and reference catchments.

comes anaerobic. Those are just the conditions needed for the bacteria that methylate mercury to thrive.

The changed hydrological pathways in the soil led to increased leaching of TOC and dissolved organic carbon (DOC). This change seems to be temporary and only lasts for one–two years. It is not possible to distinguish between permanent changes in hydrological path-

ways (causing long-term increases in MeHg leaching) and methylation of soil bound Hg. In any case, since the MeHg effect is more long-lasting, the MeHg apparently has a higher mobility in the soil water.

The discrete effects of the different forest treatments (clearcutting, mounding, afforestation) could not clearly be distinguished. However, already in the next summer and early au-

tumn after the clearcutting the water discharge peaked in summer and early autumn, which increased  $Hg_{tot}$  and MeHg loads. Still, the average increase in water discharge only doubled after the treatments compared to the extreme peaking of MeHg concentrations. Even if the monitoring period after clearcutting was too short to conclude the effect of harvest alone, these results suggest that the soil preparation had a major effect on Hg methylation and MeHg load.

## Conclusions

In conclusion, clearcutting with soil treatment significantly increased  $Hg_{tot}$  and MeHg concentrations as well as output loads in run-off water from a spruce forest catchment. The increase in TOC after harvest was temporary, but elevated concentrations and output loads of  $Hg_{tot}$  and MeHg seem to be more long lasting. The  $Hg_{tot}$  and especially the enormous MeHg surplus load might therefore result in elevated MeHg concentrations in fish. The determining factors and time scale of increased Hg exports from logged forest catchments as well as the significance of Hg leaching from catchments to lakes should be studied in more detail in boreal forest regions and especially in areas with intensive forestry.

## The concerns of the Swedish Forest Agency

ANJA LOMANDER

Research has localized and identified processes in the forest floor and in wetland areas that increase the leaching of bioavailable mercury (Hg) to aquatic ecosystems. It has also shown the role of water table fluctuations in stimulating sulphur reducing bacteria which are effective in the methylation process of Hg. The results indicate the potential effect forest management has on leakage of Hg and methylmercury (MeHg) to aquatic systems and therefore the Swedish Forest Agency emphasizes the importance of further research within this area.

At present, not enough knowledge is available to evaluate the actual contribution of total Hg and MeHg to aquatic ecosystems from various forestry activities, and as such the possibility of mitigating these effects by changing practice. However, in case a coming evaluation shows that the contribution from certain forest practices is not negligible, it ought to be followed by an evaluation of management alternatives. Therefore, the Swedish Forest Agency considers studies on the effects of different harvest strategies (for example whole tree harvest, stump extraction) and soil scarification, as well as the effects of machine tracks in relation to soil type and soil moisture content being of high priority. Furthermore, studies of the effects of riparian buffer zones, ditching and sedimentation basins/humus traps on the Hg biogeochemistry would be required as well as the effect of liming/wood ash recirculation. The effects of climate change on the methylation process are also in great demand. In this context, studies of the effects of a warmer/wetter climate as well as

studies on the effects of a change in tree species are important. The latter example refers to the discussion about increasing the amount of deciduous tree species in order to make forest stands more resistant to storm felling. However this change would probably increase the amount of easily available carbon and could therefore have an effect on the methylation potential of the soil. An increased understanding of the natural factors controlling the methylation process in forest soil, and how anthropogenic influence affects it, is also of importance. From this knowledge, it would be possible to identify "hot spots" for the methylation of mercury and adapt forestry practices to them.

Despite the uncertainties mentioned above, the Swedish Forest Agency considers the indications of a connection between forest machine activities and increased concentrations of MeHg in run-off water presented hitherto as being serious. Therefore preliminary advice to forest owners and operators has been expressed as follows:

- Avoid soil scarification and transportation of timber in moist and wet areas. Be careful in the riparian zones against small rivers and watercourses. Use temporary bridges when watercourses or wet areas have to be passed. Where clear-felled areas are both large and frequent, for example in the area hit by the storm Gudrun, it is extra important to avoid machine tracks. Place the slash so that the water upstream will not be dammed up.

- Clear ditches in such manner that the transport of humus to watercourses is not increased. Create humus traps for both new and old ditches. Block old ditches which haven't caused the intended increase in production.
- Avoid creation of large clear-felled areas in connection to moist areas. The larger clear-felled area, the higher ground water level. Fluctuations of the ground water level in humus rich horizons increase the risk of methylation and leaching of mercury. Wait about ten years before harvest of a nearby site.
- Avoid clear-felling of moist areas. In moist areas, the ground water table is already near the soil surface. Therefore, if possible, leave shelterwood stand which reduces the increase in ground water table. Avoid soil damages and harvest when the soil is frozen.
- Collaborate with other land owners within the same catchment area. This may imply finding proper routes on dry land and construction of bridges, which increase the possibility to protect streams, watercourses and moist areas from damage caused by transportation. When and where a clear-felling will take place could also be subject for collaboration, in order to keep the proportion of clear-felled area low, within a catchment area.

These advices are of a general character and are therefore also suitable for counteraction of nutrient leaching from clear-felled areas as well as transport of sediment and organic material which may disturb aquatic ecosystems.

# Sulphur chemistry controls both transport of mercury forms and methylation reactions in sediments, wetland and forest soils

ULF SKYLLBERG

## Mobilization and production of methylmercury in soils

In order to understand effects of forestry practices, or other human activities, on the biogeochemistry of mercury (Hg) and methylmercury (MeHg) in soils and surface waters we have to separate two major processes:

1. Changes in mobilization and subsequent transport of Hg/MeHg.
2. Net production of MeHg.

An increased mobilization means that equilibria or kinetics of Hg/MeHg associated with the solid and aqueous phases are shifted in favour of the latter. Thus, the total concentration of Hg/MeHg in the soil-soil solution system may be unchanged but changes in environmental factors like pH, redox-potential and/or concentrations of Hg/MeHg complexing ligands cause a shift in favour of soluble species. Most important for an enhanced mobilization is increased concentrations of dissolved inorganic sulphides (predominantly  $\text{HS}^-$ ) and dissolved organic matter (DOM).

A net production of MeHg, on the other hand, means that the total concentration of MeHg is increased in the system (soil-soil solution) as a whole, independent on thermodynamic shifts among different phases. Even if this distinction may appear obvious, several studies making conclusions about MeHg production have in fact ignored mobilization ef-

fects. This holds for studies on the effect of sulfate additions to wetland soil-solution systems (Jeremiason *et al.*, 2006; Branfireun *et al.*, 1999; 2001). In these three studies, enhanced concentrations of MeHg in soil solution after amendment of sulfate were interpreted to be caused by increased MeHg production. This may very well be true, but in order to justify such a conclusion measurements of total concentrations of MeHg (in solution and associated to the solid phase) are needed. An addition of sulfate to wetland soils in most cases would result in enhanced concentrations of inorganic sulfides as a consequence of sulfate reduction, which in turn will increase the dissolved concentration of MeHg, as well as Hg. Thus, increased concentrations of MeHg in soil solution after amendment of sulfate may simply be caused by sulphide formation and a shift in the equilibrium in favour of dissolved MeHg-sulphides. Because MeHg production mediated by sulfate reducing bacteria (SRB) has been shown to be promoted by sulphate addition in laboratory experiments (Gilmour *et al.*, 1992), in reality it is likely that both processes (1 and 2 above) occur simultaneously as a consequence of sulfate addition. In line with this, Harmon *et al.* (2004) found that MeHg increased several times, and significantly, in pore water, whereas the increase

of total concentrations in the sediment were much smaller and insignificant as a consequence of sulphate addition to wetland mesocosms.

### Sampling and sample treatment

Another important aspect associated with the issue of mobilization or net production of MeHg is that soil and water samples are taken and treated under oxygen free conditions. Drott and co-workers (2007a) showed that the concentration of MeHg in a sediment pore water was underestimated up to 100 times if precautions were not undertaken to protect the samples from contact with air during pore water extraction and filtration (Figure 1). This effect was caused by oxidation of Fe and Mn oxy/hydroxides in reduced sediment samples and subsequent flocculation/sorption of MeHg-species prior to filtration. In less reduced (oxygen deficient) sediment samples the effect was smaller. Thus, in studies of effects caused by forestry, soil and water samples need to be protected. Especially soil and groundwater samples which have a low redox-potential (are oxygen deficient).

### Controls of dissolved Hg and MeHg forms in forest and wetland soils

Both Hg and MeHg are strongly complexed by reduced organic sulphur groups, so called thiols (Skylberg *et al.*, 2006; Karlsson and Skylberg,

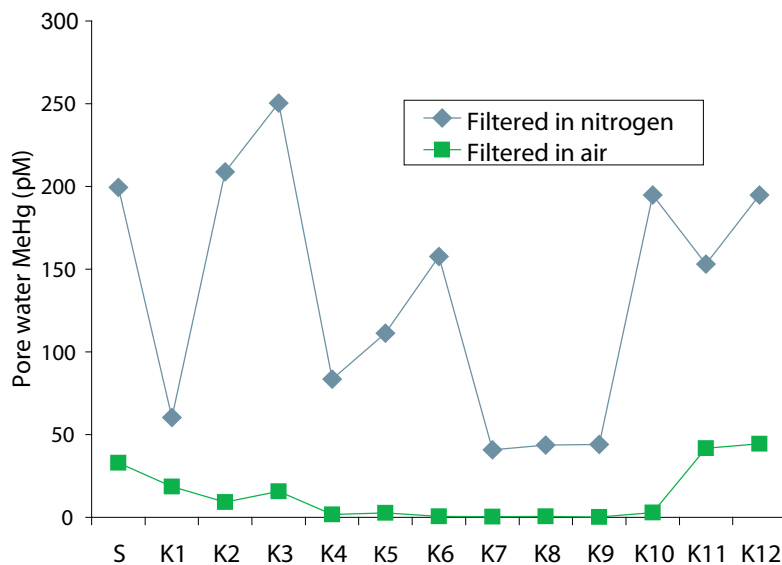


Figure 1. Concentrations of methylmercury in pore water from 13 sediment samples (Drott *et al.*, 2007a).

2003). This means that under oxidized conditions in soils and waters more than 99.9 percent of Hg and MeHg in the aqueous phase are in the form of metal-thiol complexes (Skylberg *et al.*, 2003). Whether the thiols are mainly of low molecular weight (e.g. cysteine) or thiol groups associated to large macromolecules in DOM is currently unknown. Concentrations of chloro- and hydroxy-complexes, which in some literature are mentioned as quantitatively important in soils and waters, contribute with less than 0.1 percent.

The chemistry gets more complicated when the redox-potential decreases and inorganic sulphides get into play. As illustrated in Figure 2, both Hg and MeHg shift from mainly organic complexes (Hg-DOC and MeHg-DOC) to inorganic sulphide complexes ( $[\text{Hg}(\text{HS})_2] + [\text{HgSHS}^-] + [\text{HgS}_2^{2-}] + [\text{HgS}^0]$  and  $[\text{MeHgSH}] + [\text{MeHgS}^-]$ ) when the concentration of inorganic sulphides ( $[\text{HS}^-] + [\text{H}_2\text{S}(\text{aq})]$ ) approaches the

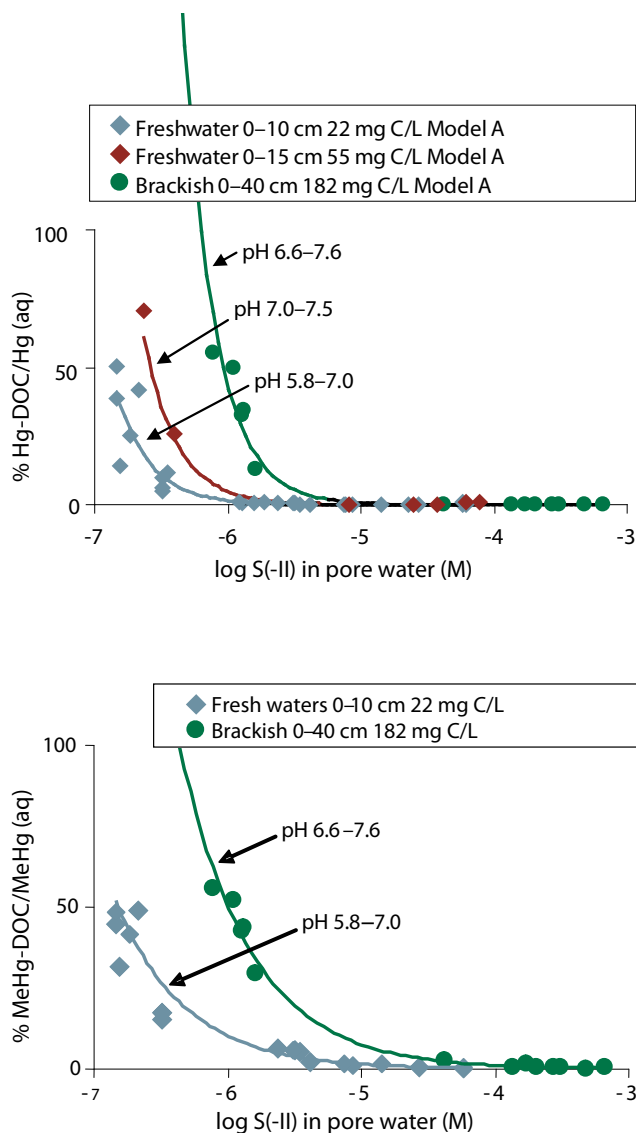


Figure 2. Shift from organic (Hg-DOC and MeHg-DOC) to inorganic sulphide complexes of Hg and MeHg in pore waters for sediments reported by Drott et al. (2007b). In the upper figure the sum of Hg-DOC and Hg-sulphides = 100 percent Hg in pore water and in the lower figure the sum of MeHg-DOC and MeHg-sulphides = 100 percent MeHg in pore water. Model A refers to the chemical model described in Drott et al. (2007b), including the complexes  $Hg(RS)_2$ ,  $Hg(HS)_2$ ,  $HgSHS^-$  and  $HgS_2^{2-}$ , but excluding  $HgS^0$  and the formation of  $HgS(s)$ . The model for MeHg includes only  $MeHgHS^0$  and  $MeHgSR$  (Skylberg et al., unpublished).

micro-molar level. Note that the slope of the relationship between  $S(-II)$  and MeHg-DOC (%) is flatter than for Hg. This is because two  $HS^-$  groups are involved for each Hg molecule in the formation of most Hg-sulphides, in opposition to one  $HS^-$  group for each MeHg molecule in the only MeHg-sulphide ( $MeHgSH^0$ ). As a consequence, the shift to a predominance of inorganic sulphide complexes is more abrupt for Hg (Figure 2) and in the concentration range  $10^{-6}$ – $10^{-5}$  M of sulphides, Hg is more dominated by inorganic sulphide complexes than MeHg. On the other hand, the inorganic sulphides may get quantitatively important for the complexation of MeHg at slightly lower concentrations than for Hg. It should be noted that these small differences are within the error of the stability constants used to calculate these distributions (Drott *et al.*, 2007b). In conclusion, both Hg and MeHg shift from organic to inorganic complexes within the sulphide concentration range  $10^{-7}$ – $10^{-6}$  M, and at  $10^{-5}$  M of inorganic sulphides both Hg and MeHg are highly dominated by inorganic sulphide complexes even if DOM concentrations are exceptionally high.

To this “picture” we need to add the absolute concentrations of Hg and MeHg. A common situation in many soils and sediments contaminated by diffusive sources of Hg is that the total concentration of Hg is 0.1–1.0 ppm ( $mg\ kg^{-1}$ ) and the total concentration of MeHg is 1–10 ppb ( $\mu g\ kg^{-1}$ ). Thus, the concentration of Hg commonly is 10–1,000 times

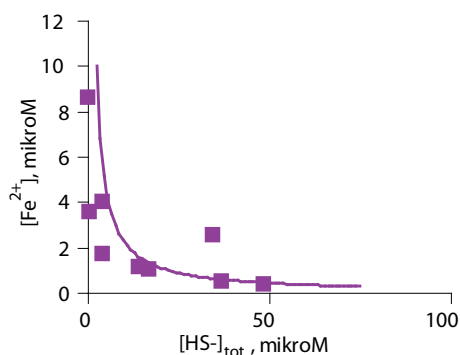


Figure 3. Relationship between dissolved Fe and HS<sup>-</sup> in a lake sediment. The solid line describes the theoretical model for the reaction  $\text{FeS(s)} + \text{H}^+ = \text{HS}^- + \text{Fe}^{2+}$ ;  $K = 10^{-3.6}$ . This is the solubility product for amorphous FeS(s), Macinawite (Skylberg et al., unpublished).

higher than the concentration of MeHg. Because Hg may form a solid phase with inorganic sulphides, as HgS(s) or mixed Fe/HgS(s)-phases, its relative solubility, in relation to total amounts in the sediment or soil, is lower than for MeHg. This relative solubility is expressed as the partitioning coefficient  $K_d$  ( $\text{L kg}^{-1}$ ). In sediments, soils and wetlands,  $\log K_d$  varies between 4.5 and 6.5 for Hg and between 2.0 and 4.5 for MeHg. Thus, the relative solubility is in general approximately 100

times (2.0 log-units) higher for MeHg than for Hg. This higher solubility of MeHg therefore “compensates” for its lower total concentrations, resulting in approximately one order of magnitude lower concentrations of MeHg and Hg in soil solution and sediment pore waters.

### Effects of iron geochemistry on Hg and MeHg solubility

To take the discussion one step further, indirect effects on the solubility of inorganic sulphides also needs to be considered. Of major concern is the formation of FeS(s), which in turn will regulate concentrations of e.g. HS<sup>-</sup>, and indirectly concentrations of Hg- and MeHg-sulphides. In Figure 3 the relationship between dissolved Fe (dominated by the Fe<sup>2+</sup>-ion) and HS<sup>-</sup> is illustrated for a lake sediment. Low sulphide concentrations represent the partly oxidized 0–5 centimetres surface layer. Note the inverse relationship and the good fit of the theoretical model.

Because enhanced concentrations of HS<sup>-</sup> means enhanced solubility of Hg and MeHg, an increase in the concentration of Fe<sup>2+</sup> results

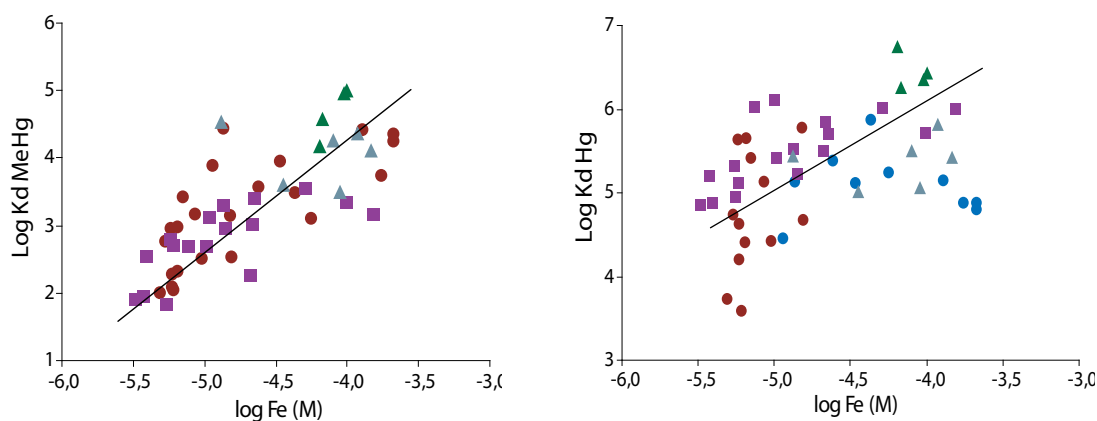


Figure 4. Relationships between log concentrations of total dissolved Fe and partitioning coefficients ( $\log K_d$ ) for MeHg and Hg in different types of sediments (Skylberg et al., unpublished).

in a decreased solubility of Hg and MeHg. This relationship is illustrated for some sediments in Figure 4. These two plots show that an increased concentration of iron in pore water is correlated to a decreased solubility of both MeHg and Hg. The reason for this is that an increase in Fe(II)-ions results in a decrease in  $\text{HS}^-$ , because the solubility product  $[\text{Fe}^{2+}] \times [\text{HS}^-] = 10^{-10.6}$  (at pH 7.0) illustrated in Figure 3 needs to be fulfilled in presence of FeS(s). As a secondary effect, also concentrations of dissolved Hg and MeHg complexed by  $\text{HS}^-$  ( $\text{Hg}(\text{HS})_2$ ,  $\text{HgSHS}^-$ ,  $\text{HgS}_2^{2-}$  and  $\text{MeHgHS}^0$ ) will decrease when Fe(II) increases.

### The role of neutral Hg-sulphides for methylmercury production

In a recent study (Drott *et al.*, 2007b) it was shown, both in short-term laboratory experiments as well as under long-term in sediments, that the concentration of neutral Hg-sulphides to a high degree control the methylmercury production. However, in the surface of the most firm sediment types (without substantial bio-

turbation and mixing) the variation in input of energy-rich organic matter overruled the effect of neutral sulphide concentrations. In Figure 5 the relationship between concentrations of neutral Hg-sulphides is plotted against the methylation rate constant ( $K_m$ ) determined after 48 hours of incubation under aerobic conditions at 23°C. Note that total concentrations of Hg in pore water had no effects on the methylation rate constant.

Figure 5 give additional support for a passive uptake of neutral inorganic Hg-sulphides by methylating bacteria, and that these bacteria methylate Hg and excrete MeHg back into the soil or sediment aqueous phase.

### Implications for forestry effects on mobilization and production of MeHg

Any forestry activity resulting in a production of soluble inorganic sulphides ( $\text{H}_2\text{S}$ ,  $\text{HS}^-$ ) or dissolved organic matter will enhance the solubility of both Hg and MeHg. If the solubility products of HgS(s) or of mixed Fe/HgS(s) phases are exceeded, the enhanced solubility

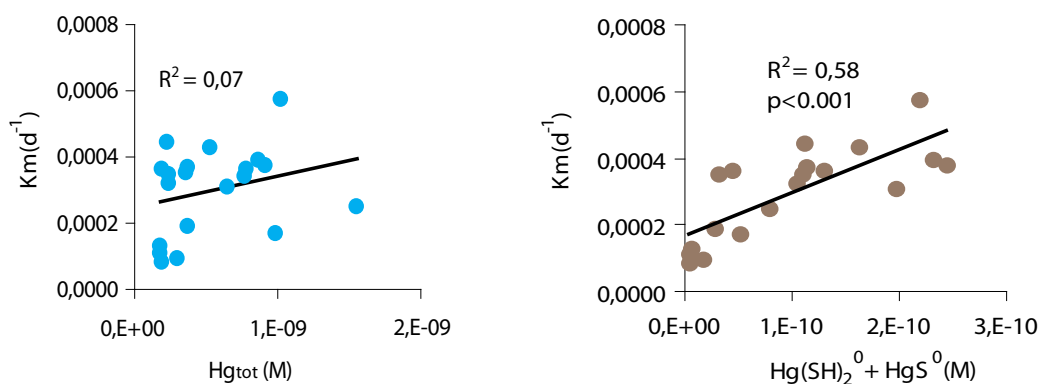


Figure 5. The relationship between pore water concentrations of total Hg (to the left) and neutral Hg-sulphides (to the right) and the methylation rate constant ( $K_m$  day<sup>-1</sup>). Data from Drott *et al.* (2007b).

of Hg will be less than for MeHg. If sources of energy-rich organic matter are available for methylating organisms like SRB, any enhancement of dissolved sulphides also will result in an enhanced production of MeHg, owing to the formation and biouptake of neutral, dissolved Hg-sulphides. It is therefore absolutely essential to differentiate between MeHg mobilization effects, caused by a shift of equilibria from solid to aqueous phases, and net MeHg

production caused by methylation processes. Furthermore, iron and sulphur are strongly associated through the formation of FeS(s). This means that iron chemistry will affect both the solubility and the methylation reaction through its control of inorganic sulphide concentrations in solution. Thus, in order to have full control on Hg/MeHg mobilization and MeHg production reactions, the biogeochemistry of C, Fe and S needs to be understood and followed in detail.

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# International efforts to control mercury emissions

KJELL JOHANSSON

Due to emissions of mercury to air the concentration of mercury in fish has increased in many parts of the world. Mercury is nowadays present all over the globe in fish at concentrations that adversely affect human beings and wildlife (UNEP 2002). In addition microbiological activity in soil appears to be very sensitive to mercury, and significant impacts may already be taking place in forest soils over large parts of Europe. Since emitted mercury is transported over wide areas and national borders before deposition, the only way to solve these environmental problems is by international co-operation. During recent years some important international organizations have placed regional mercury pollution on their agenda.

## UN ECE

The UN/ECE Convention on Long-range Transboundary Air Pollution (CLRTAP) has provided a framework for controlling and reducing the damage to human health and the environment caused by transboundary air pollution. In the years from its entering into force (1983) until today, the main focus of the effect-oriented work under the convention has been the problems of acidification and eutrophication caused by anthropogenic emissions of sulphur and nitrogen compounds. These problems include damage to human health, surface waters, vegetation, materials and cultural heritage by air pollutants.

In the 1990s, problems related to heavy metals, their accumulation in ecosystems and their impact on the environment and human

health became increasingly important. Specific attention was devoted to deriving effect-based abatement strategies. A UN-ECE protocol on Heavy Metals was signed by 36 parties in Århus (Denmark) in 1998. It addresses three particularly harmful metals: cadmium, lead and mercury. According to one of the basic obligations, the parties will have to reduce their emissions of these three metals below the levels in 1990 (or an alternative year between 1985 and 1995). The Protocol aims at cutting emissions from industrial processes, fuel combustion and waste incineration. It lays down stringent limit values for emissions from stationary sources and suggests best available techniques (BAT) for these sources. The Protocol requires the parties to phase out leaded petrol and introduces measures to lower heavy metal emissions from other products. In article 6 of the protocol the parties were encouraged to support the development of an effects-based approach to optimize future control strategies.

Since 1997 five workshops have been organized to develop effect-based approaches and critical limits for heavy metals. A manual for calculating critical loads has been finalized. The aim of the manual is to provide guidance in defining critical loads for heavy metals (lead, cadmium and mercury) for both terrestrial and aquatic ecosystems, the approaches can be summarized as:

- General methodological aspects of calculation and mapping critical loads of heavy metals.
- Mass balance models and input data to calculate critical loads.

- Critical limits that are essential to carry out the critical load calculations.
- Transfer functions, describing i.e. the relation between mercury concentrations in precipitation and biota, while accounting for the impact of soil and water chemical properties.

Regarding mercury, two endpoints have been chosen for calculating critical loads: mercury concentrations in fish and in the organic top layer of forest soils. These two receptors are highly sensitive to increased mercury deposition.

To calculate critical loads for mercury in soils a simple mass balance is used. The method is based on the balance of all relevant fluxes into or out of the organic topsoil layer (humus layer) in forest soil. Assuming steady state for this soil layer implies that the internal metal cycling within the ecosystem can be ignored. In consequence the critical loads of mercury can be calculated from the sum of tolerable outputs from the humus layer by harvest and leaching minus the natural inputs by weathering release. Weathering can however be ignored outside volcanic and ore rich areas.

For the mercury endpoint in fish, the critical level of atmospheric deposition is calculated by linking the concentrations in fish directly to the main immissions through two different transfer functions describing the relationship of their mercury concentrations at steady state. With this method there is no need for mass balance consideration or detailed understanding of ecosystem processes. One of the transfer functions ( $TF_{HgSite}$ ) relates mercury concentrations in biota to the mercury concentration in precipitation at watershed steady state. Once the mercury concentration in the standard fish is established for an ecosystem, the mercury concentration in other fish types can be addressed by means of the transfer function  $TF_{HgBio}$ , describing the

deviation from the standard fish.

Detailed descriptions of the methods to calculate critical loads and levels of mercury can be found in the Mapping Manual on the website <http://www.unece.org/env/>.

The manual has been used for a first mapping of critical loads of mercury, lead and cadmium in the ECE region. Ten countries have reported on mercury. The critical mercury concentrations in precipitation were exceeded in almost all grid cells for which they have been computed.

A Task Force on Heavy metals in the UNECE organization has now presented basic information for a discussion on the revision of the protocol on heavy metals. This discussion is now going on. An important issue is if there should be a revised protocol in 2009 or if this process should be postponed.

## EU

In 2002 the EU Commission presented a report to the Council concerning mercury from the chlor-alkali industry. This considered the fate of 12,000–15,000 tonnes of surplus mercury resulting from the sector's conversion away from the mercury cell process. The council reacted by inviting the commission to present "a coherent strategy... with measures to protect human health and the environment from the release of mercury based on a life-cycle approach, taking into account production, use, waste treatment and emissions". The strategy should also provide a basis for the Community's input to international debate on mercury at the UNEP Governing Council in February 2005. In 2005, the European Commission presented its Community Mercury Strategy (<http://ec.europa.eu/comm/environment/chemicals/mercury/index.htm>).

A key aim of the strategy is to reduce mercury levels in the environment and human ex-

posure, especially from methylmercury in fish. However, eliminating the problem of methylmercury in fish will probably take decades, as present levels are due to past emissions, and would take long time to decline even without further releases. The community has already taken much action to reduce mercury emission and uses. This highlights the importance of full implementation of existing measures by Member States and of making progress at the global level.

The strategy therefore has the following objectives:

- Reducing mercury emissions.
- Reducing the entry into circulation of mercury in society by cutting supply and demand.
- Resolving the long-term fate of mercury surpluses and societal reservoirs (in products still in use or in storage).
- Protecting against mercury exposure.
- Improving understanding of the mercury problem and its solutions.
- Supporting and promoting international action on mercury.

In order to reduce emissions of mercury the following actions has been initiated – the commission will:

- Assess the effects of applying IPPC (directive on large coal burning plants) on mercury emissions and consider if further action is needed.
- Encourage Member States and industry to provide more information on mercury releases and prevention and control techniques, so conclusions can be drawn in BREFs (on best available technique) helping to reduce emissions further.
- Undertake a study of options to abate mercury emissions from small scale coal combustion.

- Review Member States' implementation of community requirements on the treatment of dental amalgam waste.

## UNEP

On a global scale United Nation Environment Programme (UNEP) is an important organization. On the basis of a Global Mercury Assessment report the UNEP Governing Council concluded in 2003 that there is sufficient evidence of significant global adverse impacts from mercury to warrant further international action to reduce the risks to humans and wildlife from the release of mercury to the environment. The Governing Council decided that national, regional and global actions should be initiated as soon as possible and urged all countries to adopt goals and take actions, as appropriate, to identify populations at risk and to reduce human-generated releases (<http://www.chem.unep.ch/mercury/default.htm>).

The Governing Council has requested UNEP, in co-operation and consultation with other appropriate organisations, to facilitate and conduct technical assistance and capacity building activities to support the efforts of countries to take action on mercury pollution. In response to this request, UNEP has established a mercury programme. The objective of the programme is to support the efforts of countries to take actions regarding mercury pollution and identify exposed populations and ecosystems.

The programme is focusing on the following activities:

- Facilitate and conduct technical assistance and capacity-building activities (develop risk communication material and guidance material, organizing workshops and training sessions, develop a clearinghouse for mercury information).

- Develop a report summarizing supply, trade and demand information for mercury.
- Facilitate the development of partnerships between Governments, IGOs, NGOs and the private sectors.
- Promote mobilization of technical and financial resources from Governments, relevant international organisations and other partners in order to support national, regional and global efforts and capacity-building.

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# Changes to groundwater and surface water Hg transport following clearcut logging: a Canadian case study

CRAIG J. ALLAN, ANDREW HEYES, ROBERT J. MACKERETH

Researchers from Canada and the USA have been monitoring the water and chemical fluxes in two small watersheds in Northwestern Ontario, Canada since January 2003. The Escape Lake Shoreline Harvesting Project is designed to test the efficacy of current forest harvest management guidelines in protecting water quality and fish habitat in the near stream (riparian) zone of boreal forest streams. The study site is located approximately 75 kilometres

northeast of Thunder Bay, Ontario at 48° 22' N and 89° 19' W (Figure 1). The study site comprises two small watersheds. Escape 0 (E0) is a small 0.60 hectare catchment with no discernible surface drainage and Escape 1 (E1) is a larger 26.31 hectare catchment drained by an intermittent surface stream that exits through a road culvert (Label C in Figure 1). E0 serves as our long-term reference catchment and remains unlogged. The E1 catchment was clearcut

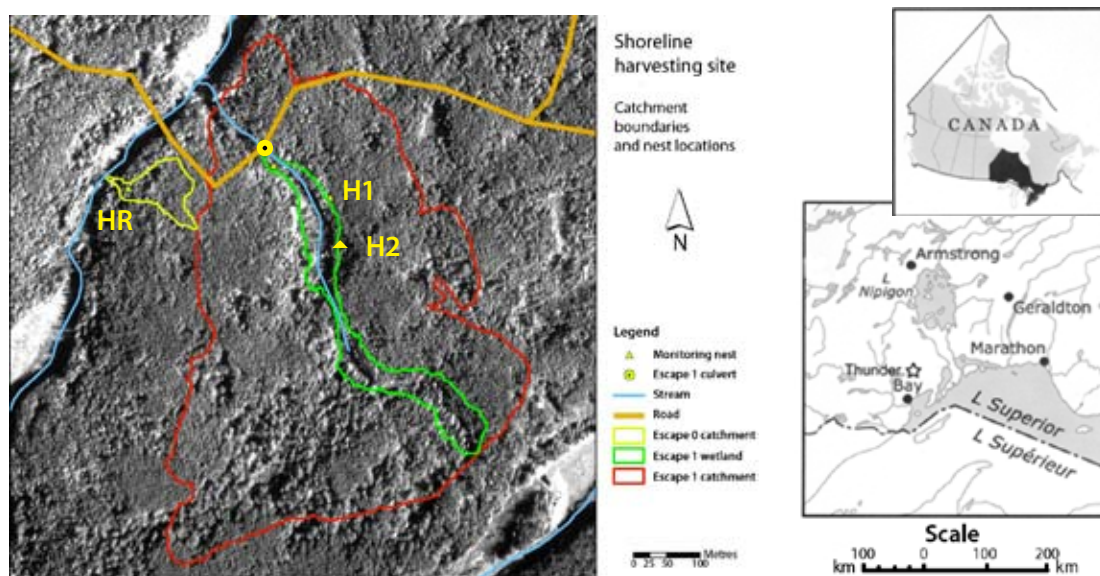


Figure 1. Location and site map including monitoring locations of Escape Lake forest harvest study.



Figure 2. Before and after photographs of the E1 experimental watershed. The 0.2 hectare valley bottom wetland that was not clearcut is outlined in orange. Rutted and water-filled skidder trails are evident in the after clearcutting photograph.

logged from July through August 2004 with only the valley bottom wetland (approximately ten percent of the watershed) not clearcut (Figure 2).

One of the unique aspects of the Escape Lake study is that it was specifically designed to assess the impacts of clearcut logging on the groundwater/surface water interface in the near stream zone. The study site as for most of the boreal shield is characterized by a veneer of boulder-rich glacial till of varying thickness overlying granitic bedrock. In order to monitor

the groundwater movement in this challenging environment a series of groundwater wells were installed with a mobile drilling rig in January 2003 (Figure 3).

After installation of the groundwater wells in the reference and study catchment groundwater levels were monitored continuously and groundwater and culvert outlet water samples were collected on a weekly schedule during snowmelt and the growing season (April–September) and monthly thereafter. Monitoring sites are presented in Figure 1. Water quality constituents

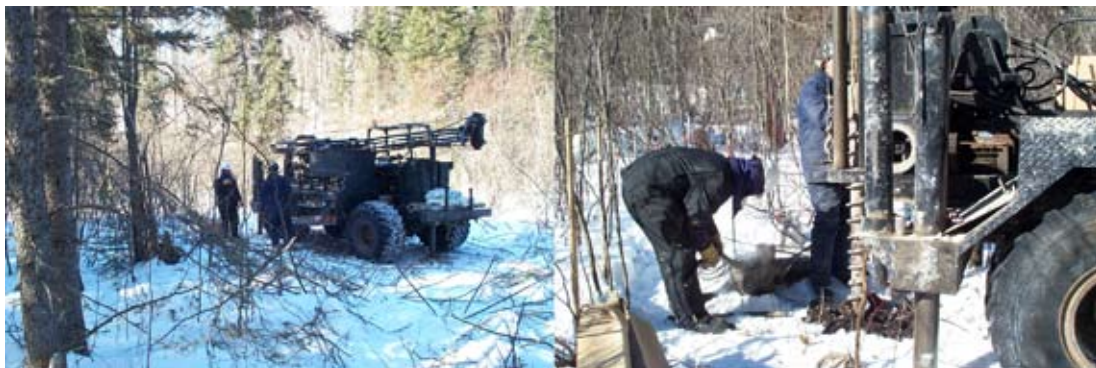


Figure 3. Installation of groundwater wells in Escape 1 watershed January, 2003.



Figure 4. Escape 1 groundwater wells before and after logging.

analyzed included total suspended solids (TSS), turbidity, conductivity, pH, calcium (Ca), magnesium (Mg), sodium (Na), potassium (K), sulfate ( $\text{SO}_4$ ), chloride (Cl), nitrate ( $\text{NO}_3$ ), ammonium ( $\text{NH}_4$ ), total nitrogen (TN), ortho phosphorus (ortho-P), total phosphorus (TP), aluminum (Al), copper (Cu), nickel (Ni), cadmium (Cd), lead (Pb), zinc (Zn), methylmercury (MeHg) and total mercury ( $\text{Hg}_{\text{tot}}$ ). Soil moisture and soil temperature measurements were also measured on a continuous basis at various locations in the watersheds. Pictures of the groundwater wells before and after logging are presented in Figure 4.

The median and range of MeHg and  $\text{Hg}_{\text{tot}}$  concentrations for the various groundwater and surface water monitoring sites before and after logging are presented in Figures 5 and 6. H1 and H2 are groundwater monitoring sites on the logged south facing hillslope in E1. W is the groundwater monitoring site in the unlogged wetland of E1. C is the culvert surface water sampling station draining the logged E1 catchment. HR is the groundwater sampling station for the unlogged E0 hillslope reference catchment. The P after each station name represents concentration of MeHg and  $\text{Hg}_{\text{tot}}$  at that station after logging. Statistically significant

changes in post logging  $\text{Hg}_{\text{tot}}$  concentrations were only observed for hillslope groundwaters (H1 and H2) with concentrations increasing (Mann-Whitney,  $P = 0.025$ ). Statistically significant changes in post logging MeHg concentrations were only evident for the wetland sampling location (W) with concentrations declining (Mann-Whitney,  $P = 0.025$ ).

No statistically significant changes in post logging Hg concentration were observed at the E1 culvert outlet (Figure 5 and 6).

The lack of a significant increase in MeHg or  $\text{Hg}_{\text{tot}}$  in run-off waters draining the logged E1 catchment is a significantly different post logging Hg response than that observed for a logged Finnish catchment (Povari *et al.*, 2003) and the forwarder disturbed Lake Gårdsjön catchment in southern Sweden (Munthe and Hultberg, 2003). At present there is no physical explanation as to why the concentration responses differ between this study and the European sites. However, it is likely that the lack of a run-off concentration response observed in this study is related to lower levels of Hg accumulation in catchment soils that is the result of historically lower atmospheric deposition at the Canadian site as compared to the European sites. Soil samples from the Escape

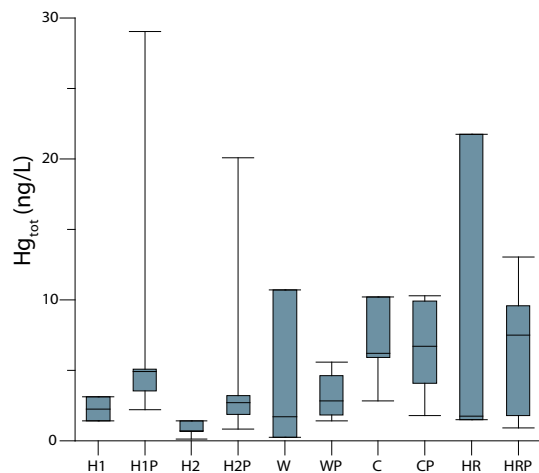


Figure 5.  $Hg_{tot}$  concentrations in surface and groundwaters, pre and post logging.

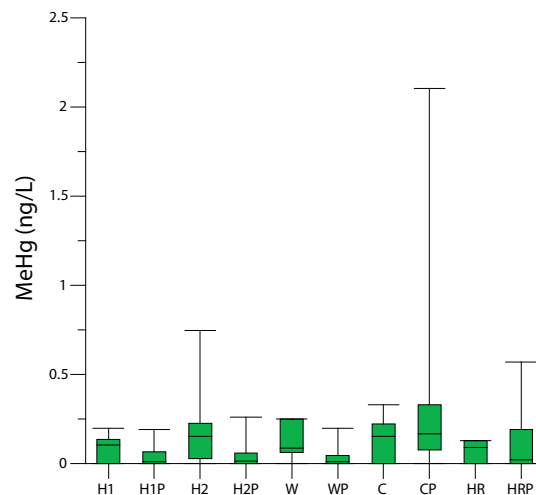


Figure 6. MeHg concentrations in surface and groundwaters, pre and post logging.

Lake watersheds are currently undergoing analysis to determine their Hg content. Despite the lack of an increase in run-off Hg concentration, the transport of methyl and total Hg from the logged E1 watershed has increased significantly in the first two years after logging. The driver for the increase in Hg transport from the logged

E1 catchment is the increase in water flux as a result of reduced transpiration and interception loss (Hornbeck et al., 1993). The increase in monthly post logging hillslope run-off as compared to the pre logging run-off relationship between the reference (E0) and experimental (E1) catchment is presented in Figure 7.

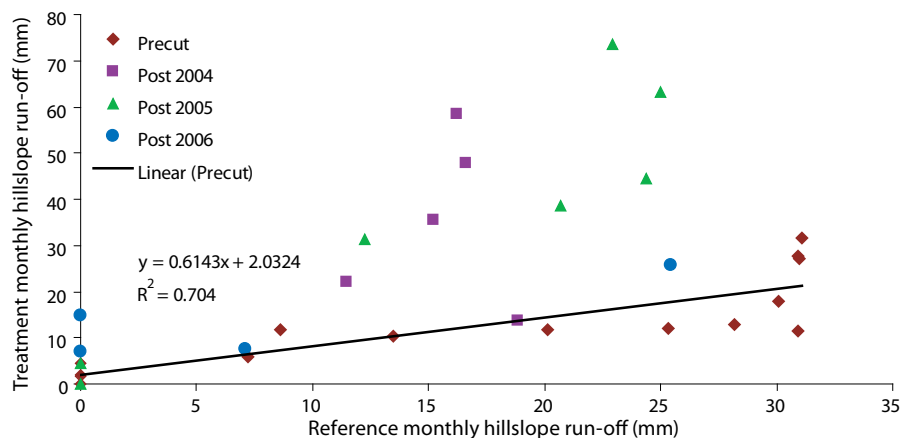


Figure 7. Pre and post logging hillslope run-off.

By multiplying the measured post logging Hg concentrations by the water flux predicted from the pre logging run-off relationship we estimate an 81 percent MeHg, and a 116 percent  $Hg_{tot}$  increase in transport from the logged E1 catchment in the first 22 months after logging. In a related study we have been measuring the hydrologic response of two logged watersheds since 1995 at a site immediately west of the Escape Lake study site. The observed increase in annual run-off at the Coldwater Lakes study site which is in a similar climate, vegetative and hydrologic setting has averaged 51 percent for the two catchments and is still observable eight years after logging (Figure 8). From these results we expect the increased Hg transport to persist at the Escape Lake site as a result of the increased run-off yield for at least a decade with the percent increase in Hg transport proportional to the increased water yield. The increased water flux and associated increase in Hg transport from

clearcut catchments provides a mechanism to explain the elevated MeHg concentrations observed in the biota of lakes impacted by watershed clearcutting in the eastern Precambrian Shield region of Canada (Derosiers et al., 2006, Garcia and Carignan 1999, Garcia and Carignan 2000).

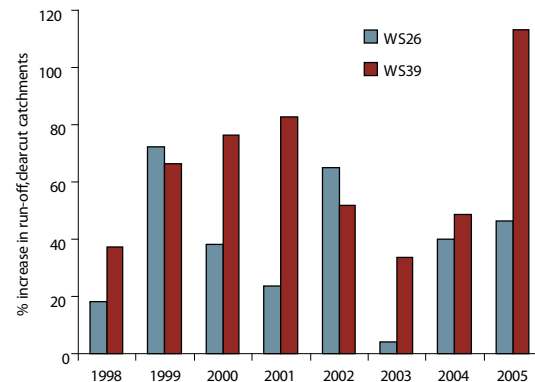


Figure 8. Increased water yield from Coldwater Lakes clearcut catchments.

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# Is mercury a threat to more than waters: soil respiration effects

LAGE BRINGMARK

## Large-scale accumulation in humus layers

Although European mercury emissions were reduced in recent decades, this happened to a lesser degree for Hg than for other pollutants. As a consequence Hg levels in forest soils continue to increase. Atmospheric Hg transported over very long, even global, distances is effectively intercepted in tree canopies and subsequently deposited to forest floors. Deposition of Hg in forests is substantial even in remote areas and to a large extent retained in the organic humus layers. In combination with other co-varying pollution factors, there is a risk of detrimental effects on soil organisms and on recycling of soil carbon and other biological processes. This risk is of chronic nature, slowly developing, and thus less dramatic than the mobilisation of methyl-Hg to aquatic systems due to disturbances associated with forest management. There is close coupling between atmospheric deposition and a steady increase of Hg in upper soil layers. The necessary remedy is lowering of soil stores viewed in a very long-term perspective.

The accumulation of Hg in forest humus layers has led to large-scale gradients. In Sweden, there is about  $0.35 \mu\text{g}\cdot\text{g}^{-1}$  in the southern parts descending to  $< 0.25 \mu\text{g}\cdot\text{g}^{-1}$  in the far north. Levels in central Europe are higher, with local concentrations exceeding  $2 \mu\text{g}\cdot\text{g}^{-1}$  in some places. With original unpolluted levels estimated to  $0.07 \mu\text{g}\cdot\text{g}^{-1}$ , Swedish concentrations in the upper organic soil have increased three to five times.

In the international work to reduce anthropogenic emissions under the Convention on Long-term Atmospheric Pollution, CLRTAP, there is an intent to base regulations on the concept of *critical loads* (see presentation by Kjell Johansson, this volume). This is the load of a pollutant to ecosystems below which long-term harmful effects do not occur. In the calculation of critical loads, the starting point is a critical limit for sensitive parts of the ecosystems. For Hg, the biological critical limit and the method for calculation of the critical load were introduced by Swedish experts. Further work is much desired to consolidate the scientific acceptance internationally. The present state of our assessments is presented here.

## Hg mass balance for the humus layer

At Aneboda in South Sweden about  $17 \mu\text{g}\cdot\text{m}^{-2}$  of Hg reaches the forest floor annually by way of throughfall, i.e. wet deposition under the canopy. An additional quantity of  $23 \mu\text{g}\cdot\text{m}^{-2}$  is coming via plant litterfall. These estimates were based on measurements in the Integrated Monitoring Programme of Swedish EPA in 1997–2000. For Hg, internal circulation in the tree-soil system is small, so almost all Hg reaching the forest floor is regarded as coming from external sources. Thus, Hg in throughfall plus litterfall is a convenient estimate of the total deposition to the forest. Transports by leaching in the soil were quantified to  $3 \mu\text{g}\cdot\text{m}^{-2}$ ,

while stream transports were about  $2 \mu\text{g m}^{-2}$ . These numbers clearly demonstrate that the major part of the deposited Hg is retained in upper soil layers.

A recent doctoral thesis by Staffan Åkerblom at the Department of Environmental Assessment, SLU, showed that Hg leaching from the humus layer was closely associated with dissolved organic material. There is a relation between Hg/carbon in the solid material and Hg/carbon in the dissolved material. The relation seems to have a general validity for different sites. This means that if Hg content of the solid material is known, monitoring of dissolved organic carbon, DOC, can be used to calculate the leaching of Hg. More research on formation of DOC and on validity of the Hg ratios is desirable.

### Field assessments of soil microbial effects

We look for co-variation between microbial activity and Hg as well as other metals in homogeneous 50x50 soil plots (Figure 1 and 2).

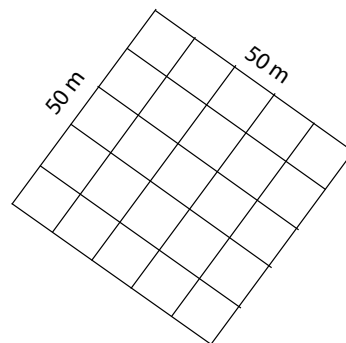
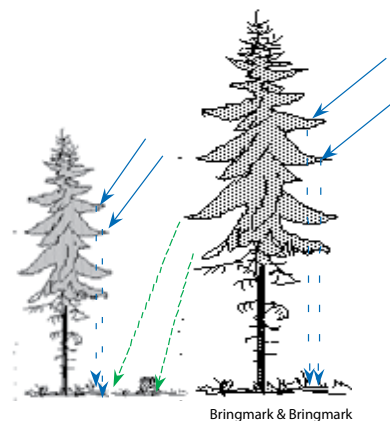


Figure 1. Forest soil sampling plot subject to heterogeneous atmospheric deposition.

Negative correlation is taken as an indication of adverse effects. The selected plots have homogeneous forest stands and soils but are heterogeneous in terms of metal loads. Variable metal interception in the canopies leads to spatially uneven deposition to the forest floor and hence variation in the humus layer concentrations. Potential microbial activity in standardised laboratory climate is measured in samples from the upper part of humus layers. Correlations with metal concentrations are calculated. Samples are collected with ten metre spacing to ensure absence of spatial autocorrelation and statistical independence.

The advantage of the method is that correlations can be searched within sites. You need not evaluate samples that are less comparable in the large-scale regional gradients, where conditions of soils and climates are more variable than within sites. The drawback is co-variation of many factors making it unclear what actually determines the microbial activity. We have looked for alternative explanations to microbial effects, e.g. organic quality, but could

not find any. There was a pronounced south-west to north-east gradient for twelve soil plots in southern Sweden with successively weaker correlations between Hg concentrations and microbial activity towards the north-east (Figure 3). This is taken as circumstantial evidence that we actually register a pollution effect in south-west Sweden, where pollution loads are higher. However, Pb co-varies rather closely with Hg both in large-scale

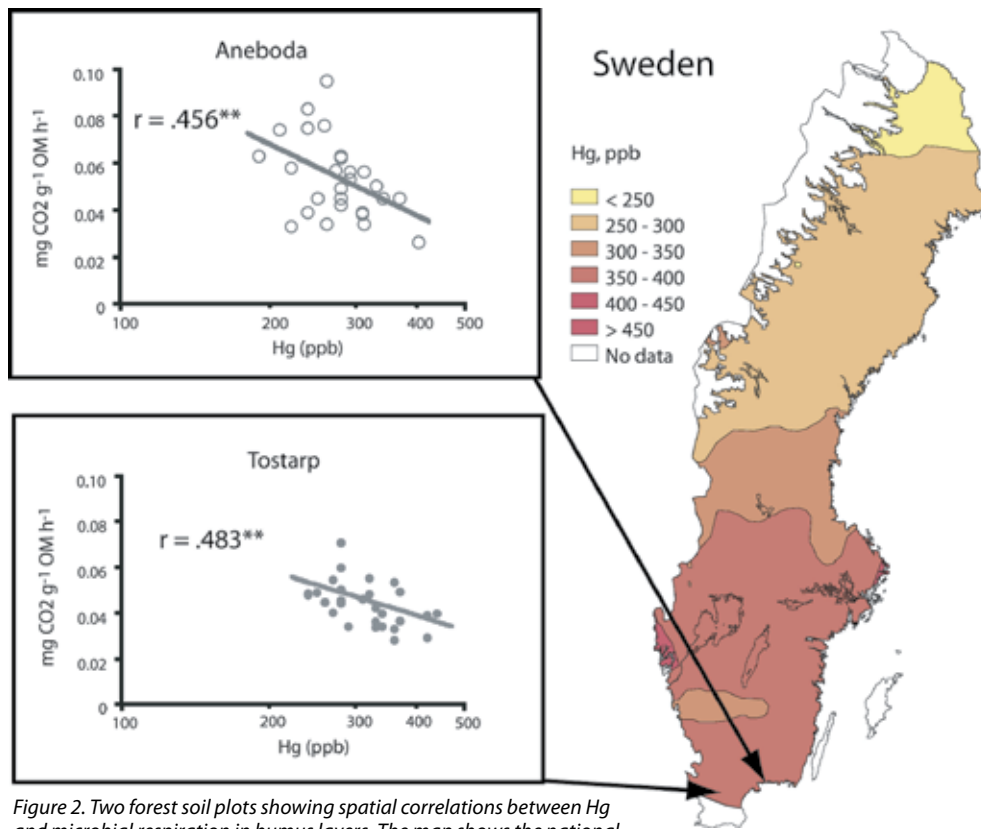
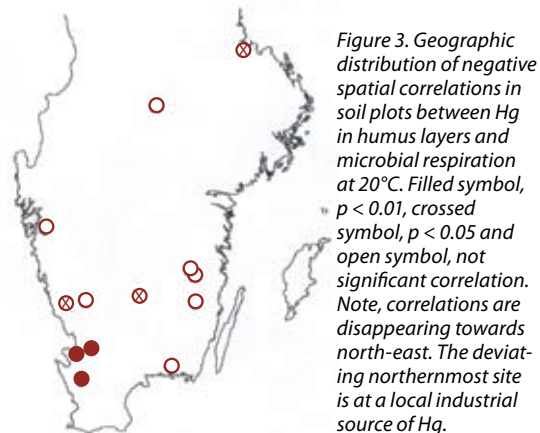


Figure 2. Two forest soil plots showing spatial correlations between Hg and microbial respiration in humus layers. The map shows the national gradient of mercury levels in humus layers.

gradients and within soil plots so effects of the two metals cannot be separated in the field investigations.

### Experimental effect levels

In order to demonstrate effects that can be attributed solely to single metals such as Hg, experiments with added metal salts to humus layer samples were set up. To create realistic conditions as far as possible, the experiments were of long duration (> six months) with low doses applied at start. New organic material was added during the experiment to serve as



microbial fuel. Thus there was time for metal immobilisation and adjustment of the microbial community. Microbial respiration was monitored throughout the experiments as the effect variable.

From the cumulated respiration values, effects were calculated. Five percent reduction of microbial activity (EC5) occurred at  $2 \mu\text{g}\cdot\text{g}^{-1}$  in a south Swedish material, while a north Swedish material had EC5 as low as  $0.9 \mu\text{g}\cdot\text{g}^{-1}$ . The latter EC5 value is not very much higher than levels actually occurring in the field sites and even lower than some central European levels. Original Hg levels in the two materials were not differing very much. The explanation for different EC5 values should probably be sought in the availability of carbon. Due to the preservation of the organic material during long, cold winters of the north, fresh substrate for microorganisms always remains in the north leading to a vital but also more sensitive microbial community. A conclusion is that different soil organic materials are not strictly comparable with regard to EC5 values.

The information in the literature on soil Hg effects is rather sparse, especially when confined to forest humus layers. We need to broaden the set of examples on microbial effects. One interesting possibility is the study of microbial communities by their phospholipid patterns. Observations of increased microbial Hg tolerance would also be useful if recorded, the implication being adaptation to toxic exposure.

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## Risk assessment summarized

The risk assessment is based on the following system of conclusions:

1. Humus layers are identified as vulnerable compartments of ecosystems due to their great capacity to retain Hg and other metals and the large presence of biological life in the layers.
2. Past and present Hg accumulation is substantial in humus layers with present Swedish levels three–five times elevated compared to pre-industrial conditions.
3. Small scale and regional field patterns of Hg and microbial respiration indicate possible effects in South Sweden, but co-variation with Pb is confounding.
4. Effect levels for Hg determined in laboratory experiments are not safely higher than highest observed field concentrations.

**Conclusion:** From this work we conclude that the critical limit for Hg in humus layers should be set to the levels currently seen in South Sweden, i.e. at  $0.5 \mu\text{g Hg}\cdot\text{g}^{-1} \text{ org.}$

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Tens of thousands of Swedish lakes have fish with unacceptably high levels of mercury. Recent research from Sweden, Canada and Finland indicate a connection between forestry operations and the input of mercury and/or the much more toxic methylmercury species to aquatic ecosystems. While these studies raise the question of whether forestry is a significant contributor to the problem of mercury in freshwater fish, there is not enough information to provide a satisfactory answer as to how important forestry actually is for the mercury contamination in freshwater fish, or what management options might exist to mitigate an eventual contribution to the mercury problem by silviculture.

The Royal Swedish Academy of Agriculture and Forestry (KSLA) hosted a two day workshop addressing these issues. This special issue of KSLA's Tidskrift presents the consensus document that resulted from that meeting, as well as summaries of some of the presentations. While many issues remain, we hope that these results of the conference will be a step on the way to a more satisfactory understanding of forestry's role in the unacceptable levels of mercury so commonly found in the fish of boreal lakes, and how to mitigate that problem.



**Royal Swedish Academy of Agriculture and Forestry**

Visiting address Drottninggatan 95 B, Stockholm  
P. O. Box 6806, S-113 86 Stockholm, Sweden  
tel +46 (0)8-54 54 77 00, fax +46 (0)8-54 54 77 10  
[www.ksla.se](http://www.ksla.se), [akademien@ksla.se](mailto:akademien@ksla.se)

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