

REPORT SNO 5222-2006

Trace metals in Norwegian surface waters, soils, and lake sediments – relation to atmospheric deposition



Pb in lake water, lake sediments, terrestrial moss and the humic surface layer of natural soils in 1995.

Norwegian Institute for Water Research

Main Office

P.O. Box 173, Kjelsås N-0411 Oslo Norway Phone (47) 22 18 51 00 Telefax (47) 22 18 52 00 Internet: www.niva.no

Televeien 3 N-4879 Grimstad Norway Phone (47) 37 29 50 55 Telefax (47) 37 04 45 13

Regional Office, Sørlandet Regional Office, Østlandet

Sandvikaveien 41 N-2312 Ottestad Norway Phone (47) 62 57 64 00 Telefax (47) 62 57 66 53

Nordnesboder 5 N-5008 Bergen Norway Phone (47) 55 30 22 50 Telefax (47) 55 30 22 51

Regional Office, Vestlandet Akvaplan-NIVA A/S N-9005 Tromsø

Norway Phone (47) 77 68 52 80

Telefax (47) 77 68 05 09

REPOR

Title Trace metals in Norwegian surface waters, soils, and lake sediments relation to atmospheric deposition	Serial No. 5222-2006	Date March 2006		
seaments – relation to atmospheric deposition	Report No. Sub-No.	Pages Price 70		
Author(s) Skjelkvåle, Brit Lisa, NIVA Steinnes, Eiliv, NTNU	Topic group	Distribution		
Rognerud, Sigurd, NIVA Fjeld, Eirik, NIVA	Geographical area Norway	Printed NIVA		
Berg, Torunn , NILU Røyset, Oddvar, NIVA				

Client(s)	Client ref.
Research Council of Norway (NFR) PROFO	134410/720
Norwegian Institute for Water Research (NIVA)	1.1.00-1.6.03
	/

Abstract

This report presents results from simultaneous trace metal surveys 1995 in Norway on atmospheric deposition based on moss analysis, the humic layer of soils, lake sediments, and surface waters. The samples of sediment and water were not taken at the same geographical locations as the moss and soil samples, but a direct comparison of the data sets is still feasible. Retrospective studies indicate that long-range transport has been a significant source of heavy metal contamination in southern Norway for the last couple of centuries, but has been considerably reduced over the last 20 years. Long-range transboundary air-pollution (LRTAP) is a very important source of of heavy metal contamination in the Norwegian environment, in particular for As, Cd, Sb, Hg, Pb, and Bi but also to some extent for V, Zn, Se, Mo, Sn, Te, and Tl. The corresponding geographic trend is reflected in samples of moss, surface soil, lake sediment, and lake water. A similar geographic trend is also indicated in the water samples for Be and Co, but not in samples of moss or soil humus. This is interpreted as a possible result of higher acidic deposition in the south wich leads to enhanced weathering of mineral matter and subsequent leaching to the lakes.

4 keywords, Norwegian		4 keywords, English				
1.	Tungmetaller	1.	Heavy metals			
2.	Atmosfærisk nedfall	2.	Deposition			
3.	Jord	3.	Soil			
4.	Innsjøer	4.	Lakes			

Brit Lisa Skjekvåle Research manager ISBN 82-577-4943-5

Buillisc Stiellwile Janh Nygard

Jarle Nygard Strategy director

Trace metals in Norwegian surface waters, soils, and lake sediments – relation to atmospheric deposition

Preface

This report presents a review of results from regional surveys carried out so far on trace metals in deposition, soil, sediment, and surface water in Norway. The report is based on recently and previously published projects from different monitoring surveys in Norway on moss, soil, and lake sediment. Only part of the lake water data has been published before.

The moss surveys in Norway were initiated and have been carried out by Eiliv Steinnes. Since 1990 Torunn Berg has been an important partner in this work. The soil surveys have also been initiated and conducted by Eiliv Steinnes, and several students of Steinnes at NTNU (O Njåstad and Tord Nygård) have been involved in writing up the results in previous reports. The sediment surveys have been performed and reported by Sigurd Rognerud and Eirik Fjeld.

In the present report Eiliv Steinnes has been responsible for writing the review on results from moss surveys and retrospective studies on peat cores, and Sigurd Rognerud has been responsible for writing the review on sediment surveys and retrospective studies based on sediment cores.

In several of the surveys discussed in this report, Oddvar Røyset has been in charge of the analytical work. Thanks to him we were able to obtain the results for trace metals from the lake survey in 1995. His great personal contribution made this possible in spite of the lack of funding for metal determinations.

Eirik Fjeld has been responsible for all the maps in the report

Brit Lisa Skjelkvåle has been responsible for reporting the results from the lake survey and for leading the work binding all the data from different surveys together.

Funding of this report has been provided by Research Council of Norway (NFR) under the programme PROFO Project number 134410/720 (1.1.00-1.6.03).

Funding of the surveys has been provided by Norwegian Pollution Control Authorities (SFT).

This is the first time we present result from all of these monitoring surveys in the same report. The amount of data collection and work prior to this report is enormous, therfore we are very pleased to present this report.

Oslo, May 2006

Brit Lisa Skjelkvåle

Contents

Summary	5
1. Introduction	6
2. Geographical patterns of trace metals in deposition, soils, lake	
sediments and surface waters in Norway	8
2.1 Trace metal in deposition as estimated from moss surveys	8
2.2 Trace metals in soils	9
2.3 Trace metals in lake sediments	10
2.4 Trace metals in lakes	11
3. Retrospective studies and trends in trace metals	13
3.1 Trends as estimated from peat cores	13
3.2 Temporal trends in metal accumulation estimated from lake	
sediments	14
3.3 Temporal trends in deposition estimated from repeated moss	
surveys	16
4. Comparison between elements in deposition (moss), soil, lake	
sediment and surface water	18
5. Summary	27
6. Reference List	29
Appendix A. Percentile tables	34
Appendix B. Maps	40

Summary

Long-range transboundary air pollution is the most important source of contaminants, including some heavy metals, to the natural environment in Norway. Since the mid 1970s several large regional surveys on heavy metals in moss, soil, sediments and surface water have been conducted to give an estimate of their "natural" levels and the relative contributions from natural and anthropogenic sources.

Retrospective studies based on lake sediments and ombrogenous peat cores indicate that long-range transport has been a significant source of heavy metal contamination in southern Norway for at least the last two centuries. The deposition of most heavy metals in Norway has been considerably reduced over the last 20 years, with the exception of local contamination from a few domestic metal smelters and contributions in the far northeast from Russian copper-nickel refineries.

This report presents results from the 1995 survey on atmospheric deposition based on moss analysis (Steinnes et al. 2001), the 1995 survey of trace metals in the humic layer of soils (Nygård 2000), the 1995-1996 survey of lake sediment (Rognerud and Fjeld 1999) and surface water in Norway (Skjelkvåle et al. 2001).

For all the surveys analytical results are available for 30-50 trace elements. The samples in the different surveys were not always taken at the same geographical locations, and a direct comparison of all data at the site level is therefore not possible. Some elements, however, show very similar geographical patterns in all compartments (moss, soils, sediments and surface waters), while others do not. This is because these media are affected by different physical and chemical processes influencing the mobilisation and thereby the concentration levels of the elements.

High concentrations of trace elements may be attributed to contribution from anthropogenic pollution input from point sources, such as in mining areas and near local industries, or from long-range transboundary air pollution (LRTAP). Appreciable input from LRTAP is mostly restricted to the southernmost part of Norway. Natural input is from geochemical anomalies. In addition, natural processes in the catchments can by different mechanisms contribute to the variability of the concentrations in surface waters and lake sediments, such as increased mobilisation and transport due to high levels of TOC or increased acidification, or hydrological conditions, such as long residence time.

Long-range transboundary air pollution is a very important source of heavy metal contamination in the Norwegian environment. LRTAP obviously provides an important contribution to the observed levels of As, Cd, Sb, Hg, Pb, and Bi, but also to some extent for V, Zn, Se, Mo, Sn, Te, and Tl. The corresponding geographic trend is reflected in samples of moss, surface soil, lake sediment, and lake water. A similar geographic trend is also indicated in the water samples for Be and Co but not in samples of moss or soil humus. This is interpreted as a possible result in the south of higher acidic deposition leading to enhanced weathering of mineral matter and subsequent leaching to the lakes. Observed concentrations of Cr, Ni, and Cu are also partly derived from air pollution, but in these cases point sources within Norway and in north-western Russia close to the Norwegian border are responsible.

1. Introduction

Exposure from long-range transboundary air pollution (LRTAP) has been evident since the early 1970s. This issue was first addressed during the interdisciplinary research program "Acid Precipitation - Effects on Forest and Fish" (Overrein et al. 1980) carried out during the period 1972-1980. This program focussed mainly on impacts from acidifying sulphur compounds. Research carried out during the same period, however, demonstrated that the problem was also evident for micro pollutants such as heavy metals (Rühling and Tyler 1973; Steinnes 1977) and PAH (Lunde and Bjørseth 1977). Over the last twenty years it has become evident from numerous scientific investigations (**Table 1**) that terrestrial as well as aquatic ecosystems in the southern part of Norway are to a great extent contaminated with a number of heavy metals derived from source regions in other parts of Europe.

Since the mid 1970s several large regional surveys on moss, soil, sediments and surface water have been conducted. An important purpose of all these surveys is to monitor metals in the natural environment in Norway and give an estimate of a "natural level" and the relative contribution of natural and anthropogenic sources. The different surveys give different type of information and have different advantages.

- The moss surveys are a surrogate of deposition input. The moss gives a time-integrated picture and the sample represent about a year of accumulation. The advantage with the method is that it can give a very detailed spatial picture of the metal deposition, which would have been very difficult and much more expensive by means of precipitation sampling.
- The soil surveys give an indication of the accumulation of metals in the soil over time. The present concentrations observed in the soil are to a large degree a result of many years of accumulation as most elements studied here are accumulated to some extent in the surface soil.
- The sediment surveys also give a time-integrated picture of the situation. The upper 0.5 cm of sediment, which is usually sampled for trace element determinations, represents the 2-3 last year of sediment accumulation. In addition the sediment holds information on variations in metals over time and can also give information about a reference condition, i.e. the concentration before the industrial revolution and what is the relative amount of pollutant added.
- Lake water gives an instant picture of the situation. The water chemistry varies through the year due to climatic variations (water renewal time) and biologically active periods. However, the method of taking an autumn sample is a surrogate of a weighted mean of the whole year. The concentration level in the water is important for aquatic biota. Lake water concentrations also give information on the general pollution level at present.

This report aims to:

- present a review of results from all regional surveys carried out on trace metals in deposition, soil, sediment and surface water in Norway;
- give a presentation of levels and geographical patterns of all the trace metal results from the 1995 lake survey;
- discuss the regional patterns and levels of trace metals in surface water in relation to other surface water properties and to results from the 1995 and 2000 surveys on atmospheric deposition based on moss analysis (Steinnes et al 2001), the 1995-1996 survey on sediment (Rognerud et al 1999), and the 1995 survey on the humic layer of natural soils (Nygård, 2000).

	Year	Number of sites	Elements	Reference
Moss/deposition	1977	514 sites nationwide	26 elements	Rambæk and Steinnes 1980,
				Steinnes et al. 1992
	1985	519 sites nationwide	16 elements	Steinnes et al. 1994
	1990	495 sites nationwide	33 elements	Berg et al. 1995
	1995	464 sites nationwide	51 elements	Berg and Steinnes 1997
	2000	repeat of 1995 survey	50 elements	Steinnes et al. 2001
Soils	1977	512 sites nationwide,	Cu, Zn, As,	Allen and Steinnes 1979, Allen
		humic surface layer	Se, Cd, Sb, Pb	and Steinnes 1980, Steinnes et
				al. 1997
	1981-1983	840 sites in southern Norway,	Cu, Zn, As,	Steinnes et al. 1989b
		humic surface layer	Cd, Pb	
	1985	400 sites nationwide in humic	29 elements	Bølviken and Steinnes 1987,
		(3-5 cm), illuvial (15-25 cm), and		Njåstad et al. 1994, Steinnes
		underground soil (60 cm)		and Njåstad 1995
	1995	464 sites nationwide in humic	33 elements	Nygård 2000
		layer (0-3 cm)		
Lake sediments	1987-1988	210 lakes nationwide	Pb, Hg, Cd, Ni	Rognerud and Fjeld 1993
	1995-1996	Same 210 lakes as in 87-88	34 elements	Rognerud and Fjeld 2001
Surface waters	1974-1975	165 lakes in southern Norway	Mn, Fe, Cu,	Henriksen and Wright 1978,
			Zn, Cd, Pb	Steinnes and Henriksen 1993
	1986	215 lakes in southern Norway	Cu, Zn, Cd, Pb	Henriksen et al. 1988, Steinnes
				et al. 1989a
	1995	995 lakes nationwide	53 elements	Skjelkvåle et al. 2001
				Skjelkvåle et al. 1996

Table 1. An overview of trace element surveys in mosses, soils, lake sediments and lakes in Norway

2. Geographical patterns of trace metals in deposition, soils, lake sediments and surface waters in Norway

2.1 Trace metal in deposition as estimated from moss surveys

Results from the first moss surveys as well as analytical data for precipitation samples from different parts of Norway (Hanssen et al. 1980) showed that the southernmost part of Norway was more exposed to deposition of several heavy metals than other parts of the country. On the basis of daily filter samples and air trajectory analysis at Birkenes, southern Norway during 1978-79 it was shown that air concentrations of heavy metals were highest when the air masses were supplied from south-easterly to south-westerly directions (Hanssen et al. 1980, Pacyna et al. 1984). A new sampling at Birkenes during 1985-86 was used to classify the samples with respect to emission areas (Amundsen et al. 1987, Amundsen et al. 1992). The results showed that metals deposited at Birkenes mainly originate from areas southwest, south, southeast and east of Birkenes. The origin of air from these directions is from industrialised regions in Europe. Very clean air was observed when the air came from sectors northwest through northeast, i.e. from the north Atlantic or from mainland Norway. Similar conclusions were reached independently for sulphur and nitrogen compounds (Overrein et al. 1980).

The first deposition surveys based on analysis of mosses from a small number of sites in Norway (Rühling and Tyler 1973; Steinnes 1977, Steinnes and Henriksen 1993) showed that the deposition of certain metals was several times higher in the south of the country than in the north. In the first nationwide moss survey in 1977 (Rambæk and Steinnes 1980; Steinnes et al. 1992), comprising 26 elements, it was demonstrated that several heavy metals, i.e. V, Zn, As, Cd, Sb, and Pb, showed a geographic distribution strongly indicating LRTAP. This is in agreement with the results from the sector analysis of aerosols at Birkenes mentioned above. In a factor analysis of the moss data (Schaug et al. 1990) the LRTAP elements V, Zn, As, Cd, Sb and Pb came out with high loadings in the first factor, explaining 20% of the variance in the data set. It was also clear that for other heavy metals such as Cr, Co, Ni, and Cu the deposition pattern was dominated by local point sources, within Norway or situated in north-western Russia close to the Norwegian border.

The moss surveys carried out in 1985 (Steinnes et al. 1994, Berg et al. 1995a), and 1995 (Berg and Steinnes 1997) verified the general patterns shown in the 1977 survey. More elements (Mo, Sn, Te, Tl, Bi) were added to the group dominated by LRTAP. The deposition of Pb in southernmost Norway was reduced by about a factor of 5 from 1977 to 1995, easily explained by facing out of leaded gasoline in many of the source countries. Analysis of stable lead isotope ratios (Rosman et al. 1998) shows that the contribution from different source regions to the Pb deposition differs considerably between different parts of the country, and also varies with time in the same region. Several other elements also exhibited a substantially reduced deposition in southern Norway during the same period, probably due to a combination of generally reduced emissions in the most important source regions and closing down of industries in some of the eastern European countries. For example the deposition of As and Cd in southern Norway as evident from the 1990 survey was only about 40% of the levels recorded in 1977 (Sjøbakk and Steinnes 1995). On the other hand, elements such as Cu and Ni supplied in large amounts to some of the areas in the north from smelters in Russia were still deposited at similar rates as 20 years ago.

The situation for mercury (Hg) as evident from moss analysis is different from that of the other heavy metals (Steinnes et al. 1991, Berg and Steinnes 1997). The geographical distribution was practically

unchanged from 1985 to 1995, and the difference between far south and far north was only about a factor of 2. This is in apparent contrast to wet deposition measurements of Hg (Iverfeldt 1991) which indicate a factor of 14 over the same south-north gradient. The difference may be explained by assuming that mosses (and other vegetation) absorb an appreciable amount of Hg⁰ in addition to Hg²⁺ which is the main form reflected in precipitation samples. Presumably the north-south gradient of atmospheric Hg⁰ in Norway is much less pronounced than for oxidised forms of Hg.

2.2 Trace metals in soils

Metals from atmospheric deposition will in most cases interact with the soil. Most of Norway consists of uncultivated land with podzols or similar soil types, where the surface horizon mainly consists of organic matter. These humus-rich topsoils will act as a temporary sink for many metals, and in the case of e.g. Pb, residence times of the order of centuries may be encountered (e. g. Tyler 1978). When it became evident that parts of Norway were receiving significant input of some metals from LRTAP, it was decided to investigate natural surface soils in the same systematic way as done for mosses. Samples from the humus layer were collected in 1977 at about 500 sites distributed all over the country, and analysed for Cu, Zn, Cd, and Pb (all samples) and As, Se and Sb (part of the material). The results (Allen and Steinnes 1979; Allen and Steinnes 1980; Steinnes et al. 1997) were very similar to those obtained for mosses collected simultaneously. Elements derived predominantly from LRTAP (Zn, As, Cd, Sb, Pb) were consistently much higher in the topsoil in southernmost Norway than in more northerly regions, whereas the concentration of elements such as Cu was apparently little affected by atmospheric deposition except in the vicinity of point sources.

High concentration of a metal in the topsoil does not necessarily imply anthropogenic input. The natural content of a metal in surface soils depends on its concentration in the underlying mineral soil, which again is in most cases related to the general geochemistry of the bedrock in the area. In order to find out to what extent the geographical differences could be associated with regional differences in bedrock composition, a second sampling campaign was organized in 1985, collecting soil samples from the illuvial horizon and the underground soil (about 60 cm depth) in addition to the humus layer. This survey verified the geographical patterns observed in the humus layer in the 1977 survey (Bølviken and Steinnes 1987; Njåstad et al. 1994; Steinnes and Njåstad 1995). The results obtained for the mineral soils, on the other hand, indicated no corresponding regional patterns for Pb, Zn, and Cd, which means that the regional patterns obtained for these elements are related to atmospheric deposition. The analytical technique employed (ICP-AES) did not allow determination of As and Sb.

In order to study the natural surface soil contamination in the most affected areas in more detail, an additional sampling was carried out during the period 1981-1983 including 840 sites in southernmost Norway covering an area of 35,000 km² (Steinnes et al. 1989b). Elements characteristic of LRTAP reached their maximum concentrations in the topsoil in a zone located about 20-50 km from the coastline, corresponding quite closely to the highest deposition of orographic precipitation. In this zone the following typical concentration levels (ppm) were observed in the raw humus (typical background values from central and northern Norway are shown in parantheses): Zn, 120 (40); As, 5 (1); Cd, 2.5 (<0.3); Pb, 180 (10). The concentrations observed for Pb were well above levels indicated in other studies to disturb microbial processes in forest soils (Bringmark et al. 1998). The surface soil concentrations observed for the same elements in Sweden (Johansson et al. 1995). The main reason for this may be the higher deposition in southern Norway due to orographic precipitation.

Heavy metals (Zn, As, Cd, and Pb) are removed rather slowly from the humus layer of natural soils (Berthelsen et al. 1995b, Samstad 1996) even in southernmost Norway where the input of acidic precipitation is considerable (Tørseth and Manø 1997) and soil acidification has proceeded much further than in areas further north (Steinnes et al. 1993). This may seem surprising since Zn and Cd in particular are known to be relatively weakly bound chemically in humic soils and therefore susceptible

to leaching (Tyler 1978, Tyler 1981). The removal of these metals from the humus layer, however, may in part be delayed by the action of mycorrhizal fungi, which tend to concentrate the available metals efficiently in the underground biomass, and thus probably play an active role in the turnover of the metals in the surface soil ecosystem (Berthelsen et al. 1995a).

The geographical distribution of Hg in surface soil in Norway (Låg and Steinnes 1978, Steinnes et al. 1991) was similar to that observed in moss, i.e. only slightly higher concentrations in the south.

2.3 Trace metals in lake sediments

Metals deposited on the lake surface, and supplied from the catchments, are likely to be scavenged by particles and deposited to the sediments. Lake sediments typically have concentrations of trace elements several orders of magnitude higher than those in overlying waters. For many metals the sediments appear to contain good temporal records. They are introduced to lakes through the weathering of rocks, as well as from atmospheric deposition. Analysis for metals in deeper sediment layers, deposited in pre-industrial times, will reveal the regional pattern of natural - or close to background concentrations. Lake sediments can therefore be regarded as an environmental archive, giving a unique possibility to assess the degree of impact of anthropogenic pollutants on freshwater systems.

A regional survey of Pb, Hg, Cd and Ni in sediments from 210 Norwegian lakes was carried out in 1987-1988 (Rognerud and Fjeld 1993). This study showed that a pronounced increase (3-6 fold) has occurred in concentrations of Hg, Pb, and Cd in surface sediments of lakes in southern Norway, as compared with sediments deposited in preindustrial times. In the case of Ni, an increase was found only in lakes in the vicinity of smelters, and in general only insignificant elevation of Ni concentrations was found. Generally, there was a clear south/north gradient and the most impacted sediments were found in the coastal and boreal areas of southern Norway. The pattern was very similar to that of wet deposition of sulphur and concentrations of the same metals in terrestrial mosses (Steinnes et al. 1992). By path analysis it was modelled how atmospheric deposition (concentrations of metals in moss as relative measure), water quality and lake specific data may influence the metal concentrations in surface sediments (Fjeld et al. 1994). The model indicated that both Hg and Pb were associated with sedimenting organic matter, and sediment concentrations were strongly influenced by atmospheric deposition and the amount of organic carrier particles in lake water. Cd behaved similarly, except for a weaker association with organic matter, and a negative effect of pH, indicating that Cd is mobilized during acidification. Geochemical contributions of Ni were much larger than atmospheric deposition. There was a negative effect of the ratio of catchment to lake area in the Pb model indicating an effective adsorption of Pb to humic substances in soils. Direct deposition on the surface is therefore an important pathway for Pb entering lakes. This is also true for Hg and probably also for other LRTAP- metals such as Cu, Sb, Bi, and As.

In 1996-1997 a second regional survey was carried out in the same 210 lakes as the first study (Rognerud and Fjeld 2001). It was a more extensive study including concentrations of 34 elements in surface and preindustrial sediments, grain size fraction, loss on ignition (LOI) and water chemistry. Surface sediments had considerably higher concentrations of Sb, Hg, Bi, Cd, As, and Pb than preindustrial sediments (2 to 7 times). Smaller, although statistically significant, increases in surface concentrations were also found for Cu, Zn and Ni (1.1 to 1.3 times). The spatial patterns of surface concentrations were quite similar for Hg, Sb, Bi and Pb. The concentrations were highest in the southern part of the country, with a pronounced decrease with latitude and altitude. The concentrations of Cd and Zn were generally highest in the south-eastern part of the country, whereas As showed highest concentrations of Cu and Ni, no clear regional gradients were detected, except for the area close to the Russian border. For elements such as Mo, V, Cr, and Co, without significantly elevated concentrations, no clear regional gradients were detected.

Detection of the effects of increased atmospheric deposition on concentrations in lake sediments is critically dependent on the size of the net fluxes to the sediments, and the natural background levels and their variability. Because high background levels also give increased variance, it is difficult to detect relatively small increase in the fluxes of elements with high background levels by statistical methods – even if the net sizes of the increased fluxes may be several orders of magnitude larger than for elements with low background levels. A small absolute increase in the concentrations in the surface sediments is easy to detect for LRTAP-elements with low background concentrations (Hg, Sb and Bi), whereas no significant increase is detected for other known LRTAP-elements (Mo and V; Berg and Steinnes 1997) with a small absolute increase and high background concentrations (*Figure I*). The absolute increase in concentrations of As, Pb and Zn was one to two orders of magnitude higher than for Hg, Sb and Bi. However, the relative increases were within a more narrow range (1.1-7.3) because of relatively high background concentrations for As, Pb, and especially Zn.



Figure 1. Principle to illustrate the difference between the possibility to detect anthropogenic contributions in cases of high and low background concentrations.

2.4 Trace metals in lakes

Studies related to connections between LRTAP and metal concentration in aquatic ecosystems have been less numerous than for terrestrial systems. The first large-scale study of lake waters in Norway was carried out in 1974-75 in 165 small, pristine lakes (Wright and Henriksen 1978). The survey focused on properties related to water acidification, but the analytical programme also included Mn, Fe, Cu, Zn, Cd and Pb. A spatial relation between metal concentration and polluted precipitation was evident for some metals. For Zn, and to a lesser extent for Pb, the data suggested that a substantial part was derived from atmospheric supply in lake waters of southern Norway (Wright and Henriksen 1978, Henriksen and Wright 1978). Later evaluation of these data (Steinnes and Henriksen 1993), suggested that Pb and Cd values from this survey were too high to be reliable, and that Mn and to some extent Fe were affected by acidic precipitation enhancing the leaching of these metals from mineral in soils and lake sediments. For Zn, it was concluded that airborne supply to the lakes and their catchments appeared to strongly affect the lake water concentration.

Data from the next nationwide survey of lakes in 1986 showed a clear geographical relation to LRTAP at least with respect to Zn and Pb (Henriksen et al. 1988, Steinnes et al. 1989b). A very distinct gradient for Zn inland from the southern coast was in accordance with similar gradients in deposition and in soil Zn in excess to the natural background (Steinnes et al. 1989b). The Zn observed in the lake water was most likely a combination of direct deposition on water surfaces and leaching from the contaminated topsoils in the catchment. Also for Pb there was a gradient inland from the coast, but the lake water concentrations were much lower than for Zn, because the release of Pb from the catchment is much slower and the residence time in the lake presumably much shorter.

In the autumn of 1995, coordinated national lake surveys were conducted in the Nordic countries, including Russian Kola (Skjelkvåle et al. 1999, Skjelkvåle et al. 2001). The 11 metals (Pb, Cd, As, Zn, Cu, Ni, Co, Fe, Mn, Cr, V) investigated in nearly 3000 lakes showed generally low values and distinct geographical patterns. It was concluded in this investigation that influence of LRTAP is the major important factor for distribution of Pb, Cd, Zn, and also to a certain degree Co. It was also concluded that total organic carbon (TOC) levels in lakes are important for Fe and Mn but also to a certain degree for As, Cr and V. Bedrock geology is the major controlling factor for Cu and Ni, with the exception of areas around the smelters in the Kola Peninsula, where the Cu and Ni concentrations in lakes are very high due to local airborne pollution. Bedrock and overburden geology is also an important factor for controlling the concentration levels of As, Co, Cr and V. Areas in the southern parts of Norway are affected by influence of LRTAP leading to Pb, Cd and Zn concentrations in lakes above limits set by environmental authorities.

The Norwegian part of the trace metals from the lake survey in 1995 has only been partly reported previously as a brief summary of the main results from about 500 of the lakes (Skjelkvåle et al. 1996).

3. Retrospective studies and trends in trace metals

Information on temporal trends in atmospheric deposition of metals comes from study of peat cores from ombrotrophic bogs and sediment cores from lakes. Documentation of trends in precipitation the last 20 years are documented by the Norwegian Monitoring Programme for Long-Range Transported air pollution (Johannessen 1995, Aas et al. 2005).

3.1 Trends as estimated from peat cores

In ombrotrophic bogs, the surface peat layer has no contact with ground water and hence is supplied with chemical substances only from the atmosphere. It is shown that deposition of elements such as Zn, As, Cd, Sb, Pb, and Bi in southern Norway during pre-industrial times probably was only about 1-2 % of that observed in recent decades (Hvatum et al. 1983; Hvatum et al. 1987; Steinnes and Jones 1995; Steinnes 1997). In the case of Pb, the concentrations of LRTAP-derived metals in near-surface peat show geographic patterns similar to those in moss and in natural surface soil (Steinnes 1997).

Particularly interesting information may be obtained from ²¹⁰Pb dated peat cores by studying stable Pb isotope signatures (Dunlap et al. 1999). In selected bogs from southern Norway it appears that only the uppermost few cm show isotope ratios characteristic of gasoline additive lead, whereas at somewhat greater depth where the Pb concentration is still high, the signature is distinctly different and remarkably similar to that of British coal (Eades et al. 2003). This shows that southern Norway was exposed to substantial deposition of Pb long before the advent of leaded gasoline, probably from non-ferrous metal smelting and coal burning in countries elsewhere in Europe.

Data for Hg in ombrogenous peat (Steinnes et al. 1991, Sjøbakk and Steinnes 1995) may provide some evidence to the ongoing discussion of the importance of anthropogenic versus natural sources to the current global Hg content in the atmosphere (Rasmussen 1994, Fitzgerald et al. 1998). Whereas the Hg concentration levels in six Norwegian bogs below 30 cm depth were 10-20 ng g⁻¹, presumably representing a pre-industrial atmospheric level, the concentrations in the surface peat representing recent conditions were 10-20 times higher. This indicates a substantial increase in atmospheric Hg, probably of anthropogenic origin, over the last few centuries.



Figure 2. Examples of trace element distributions in a peat core

3.2 Temporal trends in metal accumulation estimated from lake sediments

Temporal trends in accumulation of metals in lake sediments from southern Norway can be estimated based on the following surveys:

- The first national survey of heavy metals in lake sediments 1987-88. Concentrations of Hg, Pb, Cd and Ni were measured in the upper 3 sediment layers and enrichment factors were calculated (Rognerud and Fjeld 1993).
- The second national survey 1996-97. Changes in concentrations in surface sediments over the past 10 years were recorded (**Figure 3**, Rognerud and Fjeld 2001)
- Dated (²¹⁰Pb) sediment cores from 5 lakes in southern Norway (Kaste et al. 2001, Rognerud and Fjeld 2001).

A dated sediment core from Grindheimsvatn in southern Norway, located far from point sources, shows temporal trends in accumulation of LRTAP-metals comparable with what has been found in sediment cores from other parts of Scandinavia and peat cores from the Alps (Renberg et al. 1994, Weiss et al. 1999). There were two distinct periods of metal enrichment; betweeen 1870 and 1920, and between 1950 and 1980. From 1980 to present there has been a 2- to 5-fold decrease in accumulation rates of LRTAP-metals. The first period of enrichment was associated with the increasing coal burning in Europe, and the second enrichment period by increased industrialization and use of leaded gasoline after the Second World War. Since 1980, the introduction of unleaded gasoline and installation of efficient emission control equipment, as well as a general decrease in emissions from eastern Europe, has reduced the atmospheric deposition of metals (Pacyna 1998). Temporal accumulation rates of metals in lake sediments from 4 other lakes located along the coastline of southern Norway (Grenland, Grimstad, Arendal, Kristiansand and Sauda) showed the same trends up to 1920 as observed in Grindheimsvatn (Kaste et al. 2001, Rognerud and Fjeld 2001). However, during the period 1920 to 1985 there was significant atmospheric pollution from local smelters in the Arendal and Kristiansand area, which increased metal accumulation rates in sediments far above the LRTAP-metal signal. The importance of these smelters has been reduced during the last 10-15 years and the dominant source of the trace metals at present seems to be LRTAP.

The temporal trends observed in the dated cores, with a general increase in accumulation rates of metals in the period 1950 to 1980 followed by a decline from 1980 to 2000, are also indicated in the national sediment surveys. In the first survey 1987-88 there was a general increase in concentrations of Pb, Hg and Cd towards the sediment surface for the three uppermost centimeters of the sediment. Sedimentation rates in Nordic forest lakes are about 1.1 ± 0.5 mm yr⁻¹ (Kaste et al. 2001, Rognerud and Fjeld 2001). Thus, it can be assumed that the sediment layers (0-3cm) have been deposited during the last 30 years. If so, the increase in Pb and Cd sediment concentrations from the mid 1950s to mid 1980s is consistent with results from surveys of metal concentrations in mosses (Rühling and Tyler 1973, Steinnes et al. 1992) and Pb, Cd and Hg trends in dated sediment cores (Kaste et al. 2001). A decrease in concentrations of Pb in surface sediments was observed from the regional survey in 1987/88 to the regional survey in 1996/97 (Rognerud and Fjeld 1999). The decrease was most apparent in the most polluted areas in southern Norway.



Figure 3. Generalised map of Pb and Cd in pre-industrial sediments and present-day (1995) sediments (data from Rognerud and Fjeld 2001). Surface sediment is from 0-0.5 cm, whereas reference sediment (pre-industrial time) is from > 40 cm depth.



Figure 4. Yearly deposition of Hg, Pb and V to sediments in three lakes in southern Norway (Grindheimsvatn, Vatnebuvatn, Grovatn) from about 1840 to present time. In addition yearly deposition in a 10-year period around 1450 is shown to indicate deposition at a time before the atmosphere was polluted by metals (from Kaste et al. 2001).

3.3 Temporal trends in deposition estimated from repeated moss surveys

Generally the time trends in precipitation (**Figure 5**) are similar to those observed for the same elements in moss (**Figure 5**). In 1989-1990 a more extensive survey of bulk deposition was carried out at 6 stations in different parts of the country, using clean techniques for sampling and ICP-MS for the determination of 27 elements (Berg et al. 1994). Multivariate analysis of the data defined a component assigned to LRTAP explaining 42 % of the variation at Birkenes (Berg et al. 1994). The elements associated with this component (Pb, Zn, Cd, Sb, V, As) were essentially the same showing up in previous factor analyses of aerosol data at Birkenes (Amundsen et al. 1992) and moss data from different nationwide surveys (Schaug et al. 1990; Steinnes et al. 1994; Berg et al. 1995; Steinnes et al. 1997). Comparison of bulk deposition data from different stations with data for the same elements in moss collected near the precipitation station makes it possible to calibrate the moss data against bulk deposition of the same elements (Berg et al. 1995; Berg and Steinnes 1997).



Figure 5. Median concentrations of Pb, Cd and Zn in precipitation at Birkenes, Aust-Agder, 1976-2000 (Aas et al. 2005).

Pb in mosses (µg/g)



Figure 6. Atmospheric deposition of Pb and Cd in Norway in 1977, 1995, and 2000, illustrated by concentrations in moss ($\mu g/g$). Figures from Steinnes et al. 2001.

4. Comparison between elements in deposition (moss), soil, lake sediment and surface water

In this chapter the concentrations of all the investigated trace elements in the 1995 lake survey will be presented and discussed. The discussion is particularly focused in relation to published results from the surveys on moss, soil and lake sediment.

A common source of all trace metals in surface water is weathering of bedrock and surficial deposits in the catchment. Therefore the underlying process leading to the basic geographical pattern is the content of trace metals in the bedrock in combination with the weathering rate. Superimposed on this underlying pattern there are several other important factors leading to other geographical patterns. The major factors controlling trace element content in Norwegian surface waters can be listed as follows:

Sources:

- Geological input
 - trace metal content of bedrock and surficial deposits in combination with weathering rates.
- Atmospheric input
 - long-range transboundary air pollution (LRTAP) including several trace metals, from other parts of Europe
 - local point sources of air pollution within or near Norwegian borders
 - marine influence through atmospheric transport of marine salts and volatile biogenic emissions

Modifying factors:

- Chemical properties of catchment and lakes
 - organic matter is important for mobilizing metals from edaphic to aquatic systems. Organic matter forms complexes particularly with cations with high positive charge.
 - Redox potential is important for solubility of many metals. In areas with low redox potentials (reducing conditions) such as peat and bog areas mobilisation of particularly Fe and Mn may occur.
 - Fe and Mn hydroxides are very important carriers of many other elements
- Hydrological conditions
 - residence time of water in the catchment longer residence time allows longer contact time between percolating water and minerals, which then leads to higher concentrations.
 - amount of precipitation high water fluxes lead to short contact time.
 - in-lake processes, such as sedimentation rate.

Lake water chemistry in Norway is dominated by low ionic strength (dilute water), with low concentrations of nutrients (nitrogen and phosphorus compounds). This is mainly due to low weathering rates, low intensity of agriculture, and low population density. The bedrock in Norway and the surficial deposits derived from it are generally dominated by minerals resistant to chemical weathering. Consequently small amounts of ions are released to the water. The concentrations of base cations (Ca, Mg, Na, K) and alkalinity (HCO_3^{-}) are therefore generally low (Skjelkvåle et al. 1996, Henriksen et al. 1998). There are, however, general differences in the lake water chemistry between the different parts of the country, due to differences in hydrology, precipitation chemistry and amount, soil cover, and vegetation. From western to eastern Norway, there is a gradient from high to low precipitation (3500 to 600 mm·yr⁻¹) (illustrated as specific discharge in **Figure 7**) and from mountain areas with thin and patchy soils to forested areas with thick soils. This is reflected in lake water chemistry with low concentrations of base cations (Ca, Mg, Na, K), alkalinity (HCO_3^{-}) and total organic carbon (TOC) in the western areas relative to the eastern areas (**Figure 7**). Long-range transport of sulphur and nitrogen are deposited at the highest rates in southern and western Norway

and decrease northwards. The eastern part of Finmark in northern Norway is also affected by sulphur pollution from industry in the Kola Peninsula, northwest Russia. Southern and western parts of Norway are regions with lakes with low pH (pH < 5.5) (**Figure 7**) and negative values of ANC (Acid Neutralising Capacity). This general pattern in water chemistry is also affecting trace metals.

In the following each of the elements in surface waters is discussed with respect to geographical patterns, levels, and relations to possible sources and modifying factors and to results from surveys on deposition (moss), soil and lake sediment. Maps of each element for each matrix are presented in Appendix B. The data used are from the 1995 surveys of moss (Berg and Steinnes, 1997), humic layer of the soil (Nygård 2000), lake sediments (Rognerud and Fjeld 1999) and surface waters (Skjelkvåle et al. 1996). Many of the maps for moss, soil and lake sediment have been presented earlier although in slightly different formats. Most of the maps for surface water are presented here for the first time. It is also the first time that maps from all surveys are presented together.

An overview of the elements in question can be found in **Table 2**. Percentiles of all elements are presented in Appendix A. . The former table shows percentiles for the whole sampling set, while the latter shows percentiles divided into 10 subregions. The subdivision of these 10 regions is based on biogeographical criteria. However, since the biogeographic regions are a product of water chemistry and climatic factors, these regions are relatively well suited for picking up the differences in metal levels among the different regions of the country. This division is different from the one used in publications from Steinnes and coworkers, which is focussed on precipitation regimes.

Table 2. Periodic table illustrating elements included in the surveys. The trace elements presented in the present report are marked in yellow, pink, and green. Major ions associated with elements marked in light blue have also been determined but are not discussed in this report.

н		_															He
Li	Be											в	с	N	ο	F	Ne
Na	Mg											ΑΙ	Si	Р	Se	СІ	Ar
κ	Са	Sc	Ti	v	Cr	Mn	Fe	Со	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Мо	Тс	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Те	I	Xe
Cs	Ва	La	Hf	Та	w	Re	Os	Ir	Pt	Au	Hg	ті	Pb	Bi	Ро	At	Rn
Fr	Ra	Ac															
	•	•	Ce	Pt	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Но	Er	Tm	Yb	Lu	
			Th	Ра	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	

Major ions

Trace metals measured in moss, soil, sedimets and water

All observations (in surface waters) below detection limit of the method

Not for lake water (only moss, soil and sediments)

Not determined



Figure 7. Distribution of pH, TOC and non-marine Ca+Mg in Norwegian lakes in the autumn of 1995 and specific discharge (l/km²/sec) 1960-1990 (from Skjelkvåle et al. 1996).

Lithium - Li

The median value of Li in Norwegian lakes was 0.15 μ g/L, with concentrations ranging from <0.01 μ g/L to 5 /L. Previous surveys on wet deposition, moss, soil and sediment (Berg et al. 1994, Steinnes et al. 2001, Njåstad et al. 1994, Nygård 2000, Rognerud and Fjeld 1999) indicated only natural sources of Li in the natural environment in Norway. The geographical distribution of Li indicates that TOC is an important modifying factor for concentration levels in lake water.

Beryllium - Be

More than 65% of the lake population had Be-values below the detection limit of 0.01 μ g/L. Lakes with concentrations above detection limit were found in the eastern part of Norway. There was also a small area in the Telemark and Buskerud counties in Southern Norway where the Be concentrations in the lakes were above 0.2 μ g/L, with a maximum of 1.34 μ g/L. There were indications that Be may be more enriched in deposition than could be expected from only natural contributions in precipitation (Berg et al. 1994). Moreover the 1985 soil survey (Njåstad et al. 1994) indicated that the surface soil concentration levels cannot be explained only by mixing with underlying mineral soils. Be is an element that is mobilized by acidification but there was no indication that this had a major influence on the regional scale levels of Be in surface waters. The west-east gradient in surface water, parallel to the geographical pattern of TOC, indicated an influence of TOC on the Be content. The anomaly observed in Telemark was most probably due to bedrock geochemistry. The sediment survey indicated only geological sources.

Boron - B

The median value of B in Norwegian lakes was 1.3 μ g/L, with concentrations from <0.2 μ g/L to 40 μ g/L. One lake directly influenced by seawater had > 1000 μ g/L. The geographical pattern of B in lakes showed that this element was strongly influenced by seasalt from atmospheric deposition, and from old marine deposits. This is in agreement with conclusions from the moss survey 2000 (Steinnes et al. 2001) and the soil survey 1995 (Nygård 2000). The sediment survey showed very low concentrations of B.

Titanium – Ti

The median value of Ti in Norwegian lakes was 4.05 μ g/L, with concentrations from <0.4 μ g/L to well above 100 μ g/L. There was no clear geographical pattern for Ti in lakes. This indicated that the geochemistry of underlying bedrock and surficial deposits was the most important factor for Ti in lakes. This is also in line with previous studies on soils (Njåstad et al. 1994, Nygård 2000) which concluded that Ti in the humic layer was due to mixing from underlying mineral soils. This is also in line with the sediment survey, which concluded that Ti in sediment was dominated by geological sources (Rognerud et al. 1999). The moss survey concluded that Ti in moss was related to contribution from local soils (Steinnes et al. 2001). The pattern of Ti in lakes was different from that shown in soils and sediments, indicating that factors such as water quality (TOC) may be important for Ti concentrations in lakes in addition to the geochemistry.

Vanadium – V

More than 90% of the lake population showed V values below the detection limit of 0.3 μ g/L. The highest concentrations reached values up to 2 μ g/L. Lakes with concentrations above the detection limit were found scattered several places in Norway: in eastern Norway, southernmost part of Norway and in a small area in Sunnhordaland in western Norway. LRTAP has been identified as a source of V in moss surveys (Steinnes et al. 2001) and in studies of wet deposition (Berg et al. 1994). The soil surveys however did not reflect this pattern. The explanation for this is probably that the chemical form of V in the natural environment (oxy- and hydroxyions) does not favour adsorption to humic material (Varskog 1995). In the Nordic lake survey 1995 where the Norwegian data was evaluated together with data from Sweden, Finland, Denmark and Russian Kola (Skjelkvåle et al. 2001) it was concluded that V was slightly influence by the TOC content in the lakes. This is not evident when looking only at the Norwegian data. The sediments survey showed that V in the Norwegian

environment was controlled by two major processes: mobilisation along with Fe and influence from LRTAP (Rognerud and Fjeld 1999). Fe and TOC are also close interconnected and it can therefore sometimes be difficult to separate between the different processes leading to mobilisation of an element. The high V values in Sunnhordaland may be due to a geological anomaly.

Chromium – Cr

More than 65% of the lake population had Cr values below the detection limit of $0.1 \mu g/L$. The highest concentrations reached values slightly above $2 \mu g/L$. The geographical pattern of Cr resembles very much that of V. Lakes with concentrations above detection limit were found scattered several places in eastern Norway, southernmost part of Norway and in a small area of Sunnhordaland in western Norway. Results from moss surveys and studies of wet deposition did not show a clear relation to local sources and only a weak relation to LRTAP (Steinnes et al. 2001). The concentration levels of Cr in the humic layer of soils are interpreted as being influenced by mixing with underlying mineral soil layers (Nygård 2000). In the Nordic lake survey from 1995 (Skjelkvåle et al. 1999) it was concluded that Cr levels in lake water was not dominated by one single factor but both bedrock geology and TOC. Cr is also strongly connected with Fe and the mechanisms discussed for V above can also hold for Cr. The slightly elevated concentrations along the south-eastern coast may be attributed to influence from LRTAP. The high values in Sunnhordaland may be due to a geological anomaly.

Manganese – Mn

The median value of Mn in Norwegian lakes was 2.3 μ g/L, with concentrations from <0.2 to above 300 μ g/L. The geographical pattern of Mn strongly resembled that of TOC. Mn is released from Fe/Mn-hydroxides in bog areas were the redox-potential is sufficiently low that Mn⁴⁺ is reduced to Mn²⁺. Mn in sediments is associated with TOC. When the oxygen-level is decreasing, Mn goes into solution. The surface soil studies (Njåstad et al. 1994, Nygård 2000) indicated that Mn is of local geochemical origin, a view that is supported by precipitation studies (Berg et al. 1994). Mn (and Fe) is slightly higher in surface sediments than in preindustrial sediments probably due to effects of acidification and to redox-reactions in the water-sediment interface (Rognerud et al. 1999).

Iron – Fe

The median value of Fe in Norwegian lakes was 51 μ g/L, with concentrations from <15 μ g/L to above 300 μ g/L. The geographical pattern of Fe also strongly resembled that of TOC similarly to Mn. Fe are in the same way as Mn released from Fe/Mn-hydroxides in bog areas were the redox-potential is so low that Fe³⁺ is reduced to Fe²⁺. In contrast to Mn, Fe is strongly bound to dissolved organic matter in water, while Mn mostly occurs as free ions. Studies of atmospheric deposition (Berg et al. 1994; Steinnes et al. 2001) as well as the soil surveys (Njåstad et al. 1994, Nygård 2000) indicated negligible Fe contribution from air pollution. This conclusion is also supported from the sediment survey (see comments under Mn).

Cobalt - Co

The median value of Co in Norwegian lakes was $0.05 \ \mu g/L$, with concentrations from < $0.02 \ \mu g/L$ to above $0.2 \ \mu g/L$. The highest concentrations were found in eastern and south-eastern parts of Norway. Co is not a LRTAP element, but acidification may cause leaching of Co. Co is acossiated to Mn and both are mobilised during low red/ox potential (low oxygen). Results from moss surveys (Berg et al. 1994, Steinnes et al. 2001) showed that local point sources and soil dust were the most important sources to Co in terrestrial moss. Results from the 1985 (Njåstad et al. 1994, Varskog 1995) and the 1995 soil surveys (Nygård 2000) showed that the most important source to Co in the humic layer was the underlying mineral soil, and contribution from atmospheric deposition was evident only in the limited area affected by emissions from the Kola smelters. The sediment survey concluded that Co was accociated with Mn and Fe (Rognerud and Fjeld 1999).

Nickel - Ni

The median value for Ni in Norwegian lakes was 0.2 μ g/L, while lakes in eastern Finmark showed a median value of 0.55 μ g/L. Similar regional patterns were observed in terrestrial moss (Berg et al. 1994, Steinnes et al. 2001), lake sediments (Rognerud and Fjeld 2001) and soil humus layer (Nygård 2000). The high concentrations of Ni close to the Russian border were due to emissions from industry at the Kola Peninsula, and decreased rapidly westwards. More detailed investigations of Ni in lakes in eastern Finmark from other studies show that lakes at Jarfjordjellet close to the Russian border and the emission source, had Ni concentrations at 2.5 μ g/L (SFT 2005), while lakes more than 10 km away showed concentration < 1 μ g/L (Traaen and Rognerud 1996). Other lakes with Ni values above median value (up to about 5 μ g/L) were found along the west coast of Norway and scattered elsewhere. These values are most probably due to geochemical sources. A detailed investigation of lake sediments in southern Norway (Kaste et al. 2001) showed that there is an influence of LRTAP in this area of both Ni and Cu. This contribution from LRTAP however is too small to give a visible effect on the regional dataset.

Copper - Cu

About 40% of the lake population has Cu concentrations below the detection limit of 0.2 μ g/L. Two lakes are influenced by Cu mining activity in central Norway (e.g. Caledonian Cu-sulfide deposits in Central Norway) and therefore have very high Cu concentrations (26 and 37 μ g/L) There is a small anomaly of high concentrations in Sunnhordaland, western Norway, in the same area where a corresponding anomaly is shown for several other metals. Cu shows the same pattern as described above for Ni in terrestrial moss (Berg et al. 1994, Steinnes et al. 2001), lake sediments (Rognerud and Fjeld 2001) and soil humus lauer (Nygård 2000), with high concentrations near the Russian border in eastern Finmark. This pattern is due to emissions from industry at the Kola Peninsula, and is also observed for Cu in lake waters.

Zinc - Zn

The median value of Zn in Norwegian lakes was $3.6 \,\mu$ g/L, with concentrations from $<0.3 \,\mu$ g/L to 140 μ g/L. Two lakes in central Norway influenced by mining activity showed the highest values, while some other lakes with high concentrations were found scattered other places, probably due to geochemical anomalies. The lakes in southernmost Norway are most probably influenced by LRTAP (Skjelkvåle et al. 2001), both trough direct deposition but also mobilisation due to acidification. Zn is relatively mobile in acid soils, and in addition Zn is an element characteristic of LRTAP. Studies of transport in air (Amundsen et al. 1992) and terrestrial mosses showed that Zn is a dominant metallic component of LRTAP, although contribution from higher vegetation is a more important source than deposition for levels moss (Steinnes et al. 1994). The lake sediment survey concluded that geochemistry is the most important source for Zn. The LRTAP-signal is not easily visible in the lake sediments. Background levels of Zn are relatively high so that the additional contribution from LRTAP is difficult to detect (see illustration in illustrated in Figure 1). In addition, in acidic lakes more Zn is on soluble forms, and less is settled down to the sediment.

Germanium - Ge

The median value of Ge in Norwegian lakes was $0.15 \ \mu g/L$, with concentrations from < $0.05 \ \mu g/L$ to 4 $\mu g/L$. High values of Ge were found in the same two lakes that showed high Cu concentrations. There was also a small anomaly of high Ge concentrations in lakes in Sunnhordaland, western Norway, in the same area where anomalies of Cr and Cu were observed. There were also some lakes in southernmost Norway with high Ge concentrations, which may be interpreted as LRTAP although may seem unlikely as it is not confirmed either in the surveys on moss, soil, or sediment (Berg and Steinnes 1997, Nygård 2000, Rognerud and Fjeld 1999).

Arsenic - As

The median value of As in Norwegian lakes was 0.15 μ g/L, with concentrations from <0.1 μ g/L to 2 μ g/L. Two lakes had values up to 12 μ g/L. The pattern of As resembled very much that of Ge, with a

small anomaly of high As concentrations in lakes in Sunnhordaland, Western Norway, in the same area as for several other metals, and some lakes in southernmost Norway with high As concentrations due to LRTAP. The LRTAP origin of As has been shown in the surveys of moss, soil and sediments (Steinnes et al. 1997, Steinnes et al. 2001, Nygård 2000, Rognerud and Fjeld 1999).

Rubidium – Rb

The median value of Rb in Norwegian lakes was $0.4 \mu g/L$, with concentrations from $<0.02 \mu g/L$ to $>5 \mu g/L$. The main source of Rb is geological from weathering, but Rb may also be mobilized due to acidification. The moss surveys showed no effect of LRTAP. Higher concentrations of Rb in moss in southern Norway are attributed to enrichment from higher plants. Rb is mobilised due to acidification, which gives plants more access to Rb in acidified areas (Steinnes et al. 2001). Results from survey of the humic layer of the soil showed higher Rb concentrations in dry inland areas than in wet coastal areas, and this is attributed to loss of Rb due to exchange with marine cations in the areas strongly exposed to deposition of marine salts (Nygård 2000).

Strontium – Sr

The median value of Sr in Norwegian lakes was 4.7 μ g/L, with concentrations from <0.2 μ g/L to > 200 μ g/L. Strontium showed a relative good correlation with Ca at a rate Sr:Ca of 1:150 r²=0.45). The main source of Sr is geological from weathering, but the moss survey showed that there is also a marine component (Berg et al. 1994, Steinnes et al. 2001)

Yttrium – Y

The median value of Y in Norwegian lakes was 0.19 μ g/L, with concentrations from <0.003 μ g/L to > 2 μ g/L. The main source of Y is geological from weathering. Y is strongly bound to TOC, and concentrations are therefore higher in areas with high TOC in the lakes. Results from moss, soil, and sediment surveys indicated that the only significant source of Y is geological (Berg et al. 1994, Steinnes et al. 2001, Nygård 2000, Rognerud and Fjeld 1999).

Zirconium – Zr

The median value of Zr in Norwegian lakes was 0.016 μ g/L, with concentrations up to >0.2 μ g/L, while nearly 50% of the lakes had values below detection limit of 0.015 μ g/L. The main source of Zr is geological. Zr-minerals are very resistant to weathering and this explains the very low concentrations of Zr i surface waters. There was a tendency to higher Zr-values in the Oslo-fjord area, the reason for which is uncertain. The year 2000 moss survey showed that Zr in mosses were influenced by local soil dust (Steinnes et al. 2001) and in the 1985 soil survey Zr in the humic layer was attributed to contribution from the mineral soil (Njåstad et al. 1994)

Molybdenum – Mo

65% of the lakes had Mo concentrations <0.04 μ g/L, whereas maximum values exceeded 2 μ g/L. There was no large-scale geographical pattern for Mo. There was a small anomaly of higher concentrations in the Sunnhordland area. Mo is an LRTAP element as shown both from moss surveys, the sediment survey and wet deposition (Berg et al. 1994, Steinnes et al. 2001,Rognerud and Fjeld 1999) but a corresponding geographical pattern was not apparent for surface waters.

Cadmium – Cd

More than 60% of the lake population had Cd-values below the detection limit of 0.2 μ g/L. The highest concentrations reached values up to 1 μ g/L. Investigations of wet deposition, moss and sediment surveys have shown that Cd is strongly influenced by LRTAP (Amundsen et al. 1992, Berg et al. 1994, Steinnes et al. 2001, Rognerud and Fjeld 1999). Lakes with Cd concentrations above detection limit were mostly found in southernmost Norway. Cd is a relatively mobile ion that adsorbs weakly on organic material and minerals at pH < 6. The higher Cd concentrations in lakes in southernmost Norway are therefore most probably a combined result of LRTAP and mobilisation due to acidification.

Tin – Sn

The median value of Sn in Norwegian lakes was $0.5 \,\mu g/L$, with a few lakes > $1 \,\mu g/L$. Results from the 2000 moss survey (Steinnes et al. 2001) clearly showed that LRTAP was important for the occurrence of Sn in moss. There was no clear geographical pattern for Sn in lakes but in some areas, in particular Ryfylke in south-west Norway slightly higher values were observed, most probably due to geological sources.

Antimony – Sb

The median value of Sb in Norwegian lakes was $0.02 \ \mu g/L$, with concentrations up to $0.3 \ \mu g/L$. Results from the moss survey (Steinnes et al. 2001) clearly showed that LRTAP was important for the occurence of Sb in moss. The significance of LRTAP has been confirmed by investigations in air and wet deposition (Amundsen et al. 1992, Berg et al. 1994). For sediments this pattern was also very clear because the natural concentrations of Sb in sediments are relatively low compared to the anthropogenic contribution (Rognerud and Fjeld 1999). There was a clear geographical distribution of Sb in lakes showing high values in southern Norway, most probably due to LRTAP.

Barium – Ba

The median value of Ba in Norwegian lakes was 2.4 μ g/L, with concentrations up to 100 μ g/L. The geographical pattern of Ba in lakes showed a very distinct pattern with the highest values in eastern Norway. Soil and moss deposition surveys had previously showed that the highest Ba values were found inland in east Norway (Berg et al. 1994, Berg and Steinnes 1997, Steinnes et al. 2001, Nygård 2000). Comparison of humus layer with mineral soils in the 1985 soil survey (Njåstad et al. 1994) showed a distinct enrichment in the humus layer, which was explained by the "vascular pump" *i.e.* transport to the surface by uptake in plant roots and returned to the humus layer by leaching or litterfall (Steinnes and Njåstad 1995). The sediment survey showed a strong assosciation of Ba with Ti and Si (Rognerud and Fjeld 1999).

REE – Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu

All the REE showed much of the same geographical pattern although with different concentration levels. Median values ranged from <0.003 for Tm to 0.43 for Ce μ g/L. Results from previous surveys in wet deposition, moss and soils (Berg et al. 1994, Berg and Steinnes 1997, Njåstad et al. 1994) indicated only geological sources for the REE. The geographical patterns of REE in Norwegian lakes showed high values in eastern south Norway and along the coast in central Norway. This pattern may be related to influence from TOC and also to mobilisation due to acidification. The REE-pattern resembles that of Y, which may be explained by the similar chemical properties of these elements.

Thallium – Tl

More than 60% of the lake population had Tl-values below the detection limit of 0.006 μ g/L. The highest concentrations reached values up to 0.1 μ g/L. Results from previous surveys in wet deposition, moss and soils (Berg et al. 1994, Berg and Steinnes 1997, Nygård 2000) indicated that LRTAP is an important factor for the occurrence of Tl in the natural environment. The geographical distribution of Tl in lakes showed high values in southern Norway, probably due to influence from LRTAP.

Lead – Pb

The median value of Pb in Norwegian lakes was 0.17 μ g/L, with concentrations from <0.03 μ g/L to 15 μ g/L. The great importance of LRTAP for Pb is demonstrated in several of the surveys (Amundsen et al. 1992, Berg et al. 1994, Steinnes et al. 2001, Rognerud and Fjeld 1999, Skjelkvåle et al. 2001). The geographical pattern of Pb in lakes clearly confirmed the influence of LRTAP. In addition there was a small area in Sirdal/Kvinesdal in south-western Norway with very high Pb concentrations in water. This was not observed in any of the other surveys and most probably these lakes are also influenced by geological sources of Pb.

Bismuth - Bi

More than 75% of the Bi values in the lakes were below the detection limit of 0.02 μ g/L. The lakes with values above the detection limit were found scattered over the country. Results both from the soil, moss, and sediment surveys showed that Bi is related to LRTAP (Berg et al. 1994, Steinnes et al. 2001, Nygård 2000, Rognerud and Fjeld 1999). As for Sb, the signal in the sediments was very strong, due to low natural concentrations of Bi in the sediment compared to the anthropogenic contribution. The concentrations of Bi in water however were so low that it was not possible to conclude anything about sources.

Thorium – Th

More than 75% of the Th values in the lakes were below the detection limit of 0.015 μ g/L. The high Th values were mostly found in the eastern part of the country and may be connected to TOC. Th is easily mobilised with organic matter as it forms Th⁴⁺ ions.

Uranium – U

The median value of U in Norwegian lakes was $0.01 \,\mu g/l$, with values up to $1.5 \,\mu g/l$. The origin of U in lakes is most probably geochemical. U is similar to Th in that it forms U^{4+} which easly forms organic complexes. Therfore TOC is an important factor for mobilising U.

Nb, Ru, Rh, Pd, Ag, Hf, Ta, W, Os, Ir, Pt

All of these elements were found in very low concentration in surface waters. For all these elements > 90% of the values were below the detection limit of the method, which varied from 0.006 to 0.5 μ g/L.

5. Summary

Long-range transboundary air pollution (LRTAP) is the most important source of contaminants, including a number of heavy metals, to the natural environment in Norway. Since the mid 1970s several large-scale surveys on heavy metals in moss, soil, sediments and surface water have been conducted, in order to give an estimate of a "natural level" and the relative contribution of natural and anthropogenic sources.

This report presents results from the 1995 survey of surface water in Norway (Skjelkvåle et al. 2001) and compares the results with those from previus nationwide surveys of atmospheric deposition based on moss analysis (Steinnes et al. 2001), surface soils (Njåstad et al. 1994, Nygård 2000), and lake sediments (Rognerud and Fjeld 1999).

For all the surveys results are available for 30-50 trace elements. The samples in the different surveys were not always taken at the same geographical location so a direct comparison at each site is not possible. Some elements show similar geographical pattern in all compartments, while others do not. This is because each of the sampling media is influenced different physical and chemical processes that may alter the mobilisation of the elements and therby their concentration levels. This concerns factors such as e.g. redox reactions, complexation to organic compounds and mobilisation due to acidification.

Retrospective studies based on lake sediments and ombrogenous peat cores indicate that LRTAP has been a significant source of heavy metal contamination in southern Norway for the last couple of centuries. The deposition of most heavy metals in Norway has been considerably reduced over the last 20 years, with the exception of local contamination from a few domestic metal smelters and contributions in the north from Russian copper-nickel refineries.

High concentration of trace elements in mosses, soils, lake sediments and lake water may be attributed to contribution from:

- Point sources
 - Mining areas
 - Industrial point sources +
- LRTAP affecting the southernmost part of Norway in particular.

For the content of trace metals in soils, lake sediments and lake water some other sources and mechanisms may also be important for determining the concentration levels:

- Geochemical anomalies. A relatively large area in Sunnhordaland, western Norway show high concentration in lake water for a range of elements without any indication of anthropogenic sources. Local geochemical anomalies may also be source of high concentration levels at smaller spots (single lake or single soil sample).
- Mobilisation due to acidification. Some elements are very sensitive to changes in pH and are more easily mobilised at lower pH. In acidified areas of southern Norway increased mobilisation may partly explain higher concentrations e.g. of Cd.
- Organic matter is important for mobilizing metals from edaphic to aquatic systems. High content of TOC is positively correlated to high concentration of many trace elements in lakes.

LRTAP is a very important source of of heavy metal contamination in the Norwegian environment. LRTAP obviously provides an important contribution to the observed levels of As, Cd, Sb, Hg, Pb, and Bi, but also to some extent for V, Zn, Se, Mo, Sn, Te, and Tl. LRTAP in moss, soils, lake sediments, and lake water. Contributions from air pollution to observed levels of Cr, Co, Ni, and Cu are mainly derived from point sources within Norway and in northwestern Russia close to the Norwegian border. The remaining elements discussed in this report originate mainly from the local geochemical environment, but in some cases their concentrations surface water may be increased by enhanced leaching in catchments acidified by LRTAP.

6. Reference List

- Aas, W., Solberg, S., Berg, T., Manø, S., and Yttri, K. E. 2005. Overvåking av langtransportert forurenset luft og nedbør. Atmosfærisk tilførsel 2004. Statlig program for forurensningsovervåking Rapport 2102/2005, TA-926/2005, NILU-rapport OR-26/2005, Statens forurensningstilsyn, Oslo, Norge. 166 pp.
- Allen, R. O and Steinnes, E. 1979. Contribution from long-range transport to the heavy metal pollution of surface soil, p.271-274, <u>In</u>: *Proc. Int. Conf. Heavy Metals in the Environment, London 1979*, CEP Consultants, Edinburgh.
- Allen, R. O and Steinnes, E. 1980. Contribution from long-range transport to the heavy metal pollution of soils, p.102-103, In: Drabløs, D. and Tollan, A., *Ecological Impact of Acid precipitation*, Oslo-Ås.
- Amundsen, C. A, Hanssen, J. E., Rambæk, J. P., Semb, A., and Steinnes, E. 1987. Long-range transport of trace elements to southern Norway studied by INAA of air filters. *J.Radioanal.Nucl.Chem.* **114**: 5-12.
- Amundsen, C. A, Hanssen, J. E., Semb, A., and Steinnes, E. 1992. Long-range atmospheric transport of trace elements and main components in precipitation falling on Norway. *Atmos.Environ.* **26A** : 1309-1324.
- Berg, T., Røyset, O, and Steinnes, E. 1994. Trace elements in atmospheric precipitation at Norwegian background stations (1989-1990) measured by ICP-MS. *Atmos. Environ.* **28**: 3519-3536.
- Berg, T., Røyset, O, Steinnes, E., and Vadset, M. 1995. Atmospheric trace element deposition: Principal component analyses of ICP-MS data from moss samples. *Environ.Pollut.* **88**: 67-77.
- Berg, T., Royset, O., and Steinnes, E. 1995. Moss (Hylocomium splendens) used as a biomonitor of atmospheric trace element deposition: estimation of uptake efficiencies. *Atmos.Environ.* **29**: 353-360.
- Berg, T. and Steinnes, E. 1997. Recent trends in atmospheric deposition of trace elements in Norway as evident from the 1995 moss survey. *Sci.Tot.Environ.* **208**: 197-206.
- Berg, T. and Steinnes, E. 1997. Use of mosses (hylocomium slependens and pleurozium schreberi) as biomonitors of heavy metal deposition: from relative to absolute deposition values. *Environ.Pollut.* 98: 61-71.
- Berthelsen, B. O, Olsen, R. A, and Steinnes, E.,. 1995. Ectomycorrhizal heavy-metal accumulation as a contributing factor to heavy-metal levels in organic surface soils. *Science of the Total Environment* 170: 141-149.
- Berthelsen, B. O, Steinnes, E., Solberg, W., and Jingsen, L. 1995. Heavy-metal concentrations in plants in relation to atmospheric in precipitation falling on Norway. *Journal of Environmental Quality* **24**: 1018-1026.
- Bølviken, B and Steinnes, E. 1987. Heavy metal contamination of natural soils in Norway from long-range atmospheric transport: Further evidence from analyses of different soil horizons, p.291-293, In: *Heavy metals in the environment*, CEP Consultants, Edinburgh.
- Bringmark, L., Bringmark, E., and Samuelsson, B. 1998. Effects on mor layer respiration by small experimental additions of mercury and lead. *Sci.Tot.Environ.* **213**: 115-119.
- Dunlap, C. E, Steinnes, E., and Flegal, A. R. 1999. A synthesis of lead isotopes in two millennia of European air. *Earth Planet Sci.Letter* **167**: 81-88.

- Eades, L. J, Farmer, J. G., MacKenzie, A. B, and Bailey-Watts, A. E. 2003. Stable lead isotopic characterisation of the historical record of environmental contaminatiom in dated freshwater lake sediment cores from northern and central Scotland. *Sci.Tot.Environ.* **292**: 55-67.
- Fitzgerald, W. F, Engstrom, D. R, Mason, R. P, and Nater, E. A. 1998. The case for atmospheric mercury contamination in remote areas. *Environ.Sci.Technol.* **32**: 1-7.
- Fjeld, E., Rognerud, S., and Steinnes, E. 1994. Influence of environmental factors on heavy metal concentration in lake sediments in southern Norway indicated by path analysis. *Can.J.Fish.Aquat.Sci.* **51**: 1708-1720.
- Hanssen, J. E., Rambæk, J. P., Semb, A., and Steinnes, E. 1980. Atmospheric deposition of trace elements in Norway, p.116-117, <u>In</u>: Drabløs, D. and Tollan, A., *Ecological Impact of acid precipitation*, Proc. Int. Conf., Sandefjord, Norway.
- Henriksen, A., Lien, L., Traaen, T. S., Sevaldrud, I., and Brakke, D. F. 1988. Lake acidification in Norway present and predicted chemical status. *Ambio* 17: 259-266.
- Henriksen, A., Skjelkvåle, B. L., Mannio, J., Wilander, A., Harriman, R., Curtis, C., Jensen, J. P., Fjeld, E., and Moiseenko, T. 1998. Northern European Lake Survey - 1995. Finland, Norway, Sweden, Denmark, Russian Kola, Russian Karelia, Scotland and Wales. *Ambio* 27 : 80-91.
- Henriksen, A. and Wright, R. F. 1978. Concentration of heavy metals in small Norwegian lakes. *Water Research* **12**: 101-112.
- Hvatum, O. Ø, Bølviken, B, and Steinnes, E. 1987. Regional differences and temporal trends in heavy metal deposition from the atmosphere studied by analyses of ombrotrophic peat, p.2001-2003, In: Lindberg, S. E. and Hutchinson, T. C, *Heavy Metals in the Environment*, CEP Consultants, Edinburgh.
- Hvatum, O. Ø, Bølviken, B, and teinnes, E. 1983. Heavy metals in Norwegian ombrotrophic bogs, p.351-356, In: Hallberg, R, *Environmental Biogeochemistry. Ecol. Bull. (Stockholm)* 35,
- Iverfeldt, Å. 1991. Occurrence and turnover of atmospheric mercury over the Nordic countries. *Water Air Soil Pollut.* **56**: 251-265.
- Johannessen, T. 1995. Acidification in Norway. The Norwegian monitoring programme for long range transported air pollutants. *Water Air Soil Pollut.* **85**: 617-621.
- Johansson, K., Andersson, A., and Andersson, T. 1995. Regional accumulation pattern of heavy metals in lake sediments and forest soils in Sweden. *Sci. Tot. Environ.* **160/161**: 373-380.
- Kaste, Ø., Fjeld, E., and Rognerud, S. 2001. Environmental contaminants in lake sediments and fish in the Agder counties. NIVA-report 4334-2001, NIVA, 52pp pp.
- Låg, J. and Steinnes, E. 1978. Regional distribution of mercury in humus layers of Norwegian forest soils 28, 393-396 (1978). *Acta Agr.Scand.* 28: 393-396.
- Lunde, G and Bjørseth, A. 1977. Polycyclic aromatic-hydrocarbons in long-range transported aerosols. *Nature* **268**: 518-519.
- Njåstad, O., Steinnes, E., Bølviken, B, and Ødegård, M. 1994. Landsomfattende kartlegging av elementsammensetningen i naturlig jord: resultater fra prøver innsamlet i 1977 og 1985 oppnådd ved ICP emisjonsspektrometri. Rapport 94/027, Norwegian Geological Survey (NGU), Trondheim.
- Nygård, T. 2000. Sporelementer i humussjiktet i naturlig jordsmonn i Norge. Norwegian University of Sceince and Technology (NTNU), Cand.Scient.Thesis, 111 pp.

- Overrein, L., Seip, H. M., and Tollan, A. 1980. Acid precipitation Effects on forest and fish. Final report of the SNSF-project 1972-1980. Fagrapport FR 19-80, Oslo-Ås, Norway. 175 pp.
- Pacyna, J. M. 1998. Source inventories for atmospheric trace metals, p.Harrison, R. M. and Van Grieken, R., John Wiley & Sons, Inc , New York.
- Pacyna, J. M., Semb, A., and Hanssen, J. E. 1984. Emission and long-range transport of trace elements in Europe. *Tellus* **36B**: 163-178.
- Rühling, Å. and Tyler, G. 1973. Heavy metal deposition in Scandinavia. Water Air Soil Pollut. 2: 445-455.
- Rambæk, J. P. and Steinnes, E., 1980. Atmospheric Deposition of heavy metals studied by analysis of moss samples using neutron activation analysis and atomic adsorption spectrometry. Nuclear Methods in Environmental and Energy Research. 175-180. Conf-800433, Colombia, Missouri, US.
- Rasmussen, P. E. 1994. Current methods of estimating atmospheric fluxes of mercury in remote areas. *Environ.Sci.Technol.* **28**: 2233-2241.
- Renberg, I., Persson, M. W, and Emteryd, O. 1994. Pre-industrial atmospheric lead contamination detected in Swedish lake sediments. *Nature* **368**: 323-326.
- Rognerud, S. and Fjeld, E. 1993. Regional survey of heavy metals in lake sediments in Norway. *Ambio* 22: 206-212.
- Rognerud, S. and Fjeld, E. 1999. Landsomfattende undersøkelser av metaller i innsjøsedimenter. Statlig program for forurensningsovervåking Rapport 759/99, TA-1631/1999, Statens forurensningstilsyn, Oslo, Norway.
- Rognerud, S. and Fjeld, E. 2001. Trace element contamination of Norwegian lake sediments. Ambio 30: 11-19.
- Rosman, K. J. R., Chisholm, W., Boutron, C. F., Hong, S., Edwards, R., Morgan, V, and Sedwick, P. N. 1998. Lead isotopes and selected metals in ice from Law Dome, Antarctica. *Annals of Glaciology* 27: 349-354.
- Samstad, G.-R. 1996. Temporal changes in heavy metal supply to natural soils in Southern Norway (In Norwegian). Norwegian University of Science and Technology (NTNU), M.Sc.Thesis, 84 pp.
- Schaug, J., Rambæk, J. P., Steinnes, E., and Henry, R. C. 1990. Multivariate analysis of trace element data from moss samples used to monitor atmospheric deposition. *Atmos.Environ.* **24A**: 2625-2631.
- SFT. 2005. Norwegian monitoring programme for long-range transported air pollutants. Annual report Effects 2004. Report 941/2005, TA-2126/2005, Norwegian Pollution Control Authority (SFT), Oslo, Norway. 149 pp.
- Sjøbakk, T. E and Steinnes, E. 1995. Temporal trends in heavy metal deposition in Norway studied by moss analysis, p.88-91, <u>In</u>: Wilken, R. D, Förstner, U, and Knöchel, A, *Heavy metals in the environment*, CEP Consultants, Edinburgh.
- Skjelkvåle, B. L., Andersen, T., Fjeld, E., Mannio, J., Wilander, A., Johansson, K., Jensen, J. P., and Moiseenko, T. 2001. Heavy metal surveys in Nordic lakes: concentrations, geographic patterns and relation to critical limits. *Ambio* 30: 2-10.
- Skjelkvåle, B. L., Henriksen, A., Faafeng, B., Fjeld, E., Traaen, T. S., Lien, L., Lydersen, E., and Buan, A. K. 1996. Regional innsjøundersøkelse 1995. En vannkjemisk undersøkelse av 1500 norske innsjøer. Statlig program for forurensningsovervåking Rapport 677/96, Statens forurensningstilsyn, Oslo, Norway. 73 pp.

- Skjelkvåle, B. L., Henriksen, A., Vadset, M, and Røyset, O. 1996. Sporelementer i norske innsjøer. NIVA-report 3457-96, Norsk institutt for vannforskning, Oslo, Norway. 18 pp.
- Skjelkvåle, B. L., Mannio, J., Wilander, A., Johansson, K., Jensen, J. P., Moiseenko, T., Fjeld, E., Andersen, T., Vuorenmaa, J., and Røyset, O. 1999. Heavy metal surveys in Nordic lakes; harmonised data for regional assessement of critical limits. NIVA-report SNO 4039-99, Norwegian Institute for Water Research, Oslo. 71 pp.
- Steinnes, E. 1977. Atmospheric deposition of trace elements in Norway studied by means of moss analysis. Kjeller Report No. KR-154, Institutt for Atomenergi, Kjeller, Norway.
- Steinnes, E. 1997. Trace element profiles in ombrogenous peat profiles from Norway: Evidence of long range atmospheric transport. *Water Air Soil Pollut.* **100**: 405-413.
- Steinnes, E., Allen, R. O, Petersen, H. M., Rambæk, J. P., and Varskog, P. 1997. Evidence of large scale heavymetal contamination of natural surface soils in Norway from long-range atmospheric transport. *Sci.Tot.Environ.* 205: 255-266.
- Steinnes, E., Angell, V., and Andersson, E. M. 1991. Atmospheric deposition of mercury in Norway Temporal and spatial trends. *Water Air Soil Pollut.* **56**: 391-404.
- Steinnes, E., Berg, T., Sjøbakk, T. E, Uggerud, H. T., and Vadset, M. 2001. Atmosfærisk nedfall av tungmetaller i Norge. Landsomfattende undersøkelse i 2000. Statlig program for forurensningsovervåking SFTrapport 838/01, Statens forureningstilsyn, Oslo, Norway. 28 pp.
- Steinnes, E., Berg, T., Vadset, M, and Røyset, O. 1997. Atmosfærisk nedfall av tungmetaller i Norge. Landsomfattende undersøkelse i 1995. SFT-report 691/97, Norwegian Pollution Control Authority (SFT), Oslo.
- Steinnes, E., Hanssen, J. E., Rambæk, J. P., and Vogt, N. B. 1994. Atmospheric deposition of trace elements in Norway temporal and spatial trends studied by moss analysis. *Water Air Soil Pollut.* **74**: 121-140.
- Steinnes, E. and Henriksen, A. 1993. Metals in small Norwegian lakes relation to atmospheric deposition of pollutants. *Water Air Soil Pollut.* **7**: 167-174.
- Steinnes, E., Hovind, H., and Henriksen, A. 1989. Heavy metals in Norwegian surface waters, with emphasis on acidification and atmospheric deposition, p.36-39, <u>In</u>: Mouvet, C, Heavy Metals in the Environment, CEP Consultants, Edinburgh.
- Steinnes, E. and Jones, K. C. 1995. Temporal variations in atmospheric deposition of heavy metals studied by analysis of peat cores from ombrotrophic bogs, p.37-40, <u>In</u>: Wilken, R. D, Förstner, U, and Knöchel, A, *Heavy metals in the Environment, Vol. 1*, CEP Consultants, Edinburgh.
- Steinnes, E. and Njåstad, O. 1995. Enrichment of metals in the organic surface layer of natural soils: Identification of contributions from different sources. *Analyst* **120**: 1479-1483.
- Steinnes, E., Rambæk, J. P., and Hanssen, J. E. 1992. Large scale multi-element survey of atmospheric deposition using naturally growing moss as biomonitor. *Chemosphere* **35**: 735-752.
- Steinnes, E., Røyset, O, Vadset, M, and Johansen, O. 1993. Atmosfærisk nedfall av tungmetaller i Norge. Landsomfattende undersøkelse i 1990. Statlig program for forurensningsovervåking Rapport 523/93, Statens forurensningstilsyn, Oslo, Norway.
- Steinnes, E., Solberg, W., Petersen, H. M., and Wren, C. D. 1989. Heavy metal pollution by long range atmospheric transport in natural soils of Southern Norway. *Water Air Soil Pollut.* **45**: 207-218.

- Tørseth, K. and Manø, S. 1997. Overvåking av langtransportert forurenset luft og nedbør. Atmosfærisk tilførsel 1996. Statlig program for forurensningsovervåking Rapport 703/97, Statens forurensningstilsyn, Oslo, Norway. 203 pp.
- Traaen, T. S. and Rognerud, S. 1996. Forsuring og tungmetallforurensning i grenseområdene Norge/Russland. Årsrapport for 1995. SFT-Rapport 3458-96, SFT., Oslo, Norway. 21 pp.
- Tyler, G. 1978. Leaching rates of heavy metal ions in forest soil. *Water Air Soil Pollut.* 9: 137-148.
- Tyler, G. 1981. Leaching of metals from the A-horizon of a spruce forest soil. *Water Air Soil Pollut.* **15**: 353-369.
- Varskog, P. 1995. A study of chemical composition of Norwegian forest soils with relation to climatic, pedological and edpahic factors. Norwegian University of Sceince and Technology (NTNU), Dr.Scient. thesis pp.
- Weiss, D., Shotyk, W, Appleby, P. G, Kramers, J. D, and Cheburkin, A. K. 1999. Atmospheric Pb deposition since the industrial revolution recorded by five Swiss peat profiles: Enrichment factors, fluxes, isotop composition and sources. *Environ.Sci.Technol.* 33: 1340-1352.
- Wright, R. F. and Henriksen, A. 1978. Chemistry of small Norwegian lakes with special reference to acid precipitation. *Limnol.Oceanog.* 23: 487-498.

Appendix A. Percentile tables

Concentrations of trace metals (percentiles) in terrestrial moss in Norway (1995). Units: $\mu g g^{-1}$. Number of sites = 464. Data from Steinnes et al. 1997.

Element		Atomic No	10	25	50	75	90
Li	Lithium	3	0.065	0.11	0.18	0.26	0.36
Be	Beryllium	4	< 0.0004	0.0099	0.019	0.028	0.045
Mg	Magnesium	12	851	1175	1444	1758	2017
AI	Aluminium	13	166	229	288	380	550
Са	Calsium	20	2226	2747	3236	3873	4496
Ti	Titanium	22	21	30	43	64	90
V	Vanadium	23	0.73	1.4	2.3	3.7	5.5
Cr	Chromium	24	0.44	0.71	1.0	1.6	2.6
Mn	Manganese	25	99	165	247	397	581
Fe	Iron	26	208	348	475	736	1054
Со	Cobalt	27	0.12	0.18	0.24	0.38	0.58
Ni	Nickel	28	0.80	1.2	1.6	2.2	3.5
Cu	Copper	29	3.1	4.1	5.2	7.4	10
Zn	Zinc	30	16	28	38	52	67
Ga	Gallium	31	0.36	0.57	0.83	1.2	1.7
Ge	Germanium	32	0.45	0.71	0.94	1.2	1.6
As	Arsenic	33	<0.13	<0.13	0.21	0.36	0.66
Se	Selenium	34	<0.45	<0.45	<0.44	0.45	0.82
Rb	Rubidium	37	2.7	6.4	11	18	25
Sr	Strontium	38	5.0	8.7	13	19	26
Y	Yttrium	39	0.084	0.15	0.23	0.37	0.60
Zr	Zirconium	40	0.11	0.17	0.25	0.37	0.58
Nb	Niobium	41	0.022	0.050	0.078	0.13	0.20
Мо	Molybdenum	42	0.044	0.086	0.16	0.28	0.40
Cd	Cadmium	48	0.030	0.072	0.13	0.25	0.36
Sn	Tin	50	0.046	0.089	0.17	0.32	0.51
Sb	Antimony	51	0.025	0.046	0.092	0.20	0.31
Те	Tellurium	52	< 0.0023	< 0.0023	< 0.0023	0.0063	0.015
Cs	Cesium	55	0.042	0.098	0.18	0.32	0.54
Ва	Barium	56	10	17	24	35	51
La	Lanthanum	57	0.14	0.26	0.41	0.63	0.99
Ce	Cerium	58	0.26	0.51	0.81	1.2	2.0
Pr	Praseodymium	59	0.030	0.064	0.098	0.15	0.23
Nd	Neodymium	60	0.11	0.25	0.38	0.58	0.87
Sm	Samarium	62	0.016	0.042	0.071	0.11	0.19
Eu	Europium	63	0.0069	0.012	0.018	0.027	0.040
Gd	Gadolinium	64	0.019	0.045	0.069	0.12	0.19
Tb	Terbium	65	0.0024	0.0054	0.0095	0.015	0.023
Dy	Dysprosium	66	0.016	0.033	0.054	0.086	0.13
Но	Holmium	67	0.0027	0.0059	0.0096	0.015	0.024

Element		Atomic No	10	25	50	75	90
Er	Erbium	68	0.0083	0.017	0.028	0.045	0.067
Tm	Thulium	69	0.0002	0.0018	0.0036	0.0061	0.010
Yb	Ytterbium	70	0.0062	0.014	0.023	0.039	0.062
Lu	Lutetium	71	< 0.0001	0.0018	0.0032	0.0052	0.008
Hf	Hafnium	72	< 0.0026	< 0.0026	0.0036	0.010	0.019
W	Tungsten	74	0.012	0.031	0.061	0.11	0.21
Hg	Mercury	80	0.035	0.052	0.068	0.090	0.12
ТІ	Thallium	81	0.011	0.028	0.062	0.15	0.27
Pb	Lead	82	1.5	2.7	5.8	12	19
Bi	Bismuth	83	<0.024	0.085	0.19	0.38	0.76
Th	Thorium	90	< 0.0082	<0.0080	0.039	0.084	0.14
U	Uranium	92	0.010	0.025	0.039	0.065	0.090

Element		Atomic No	10	25	50	75	90
Li	Lithium	3	0.129	0.32	0.5	1.045	2.554
Be	Beryllium	4	0.01	0.04	0.07	0.15	0.262
В	Boron	5	2.4	3.34	4.22	5.405	6.864
Ті	Titanium	22	60	112.5	193	340.5	492.4
V	Vanadium	23	2.7	4.5	8	13.55	24.6
Cr	Chromium	24	1.4	2.6	3.9	7.35	15.26
Mn	Manganese	25	40.9	97	182	388.5	635.2
Со	Cobalt	27	0.359	0.62	1.02	2.11	3.87
Ni	Nickel	28	1.7	2.95	4.1	6.45	11.22
Cu	Copper	29	5.39	7.2	9	11.5	15.88
Zn	Zinc	30	23.29	38.4	51.7	72.3	93.64
Ge	Germanium	32	0.8	1.3	1.8	2.6	3.7
As	Arsenic	33	0.2	0.48	0.89	1.745	2.752
Rb	Rubidium	37	2.3	4.7	6.9	11.3	16.52
Sr	Strontium	38	9.79	17.4	25.6	37.25	55.74
Y	Yttrium	39	0.4	0.8	1.6	3.6	6.9
Zr	Zirconium	40	0.5	1.1	1.9	3.5	7.42
Мо	Molybdenum	42	0.09	0.23	0.39	0.675	1.024
Ag	Silver	47	0.07	0.19	0.34	0.535	0.858
Cd	Cadmium	48	0.12	0.27	0.45	0.69	1.05
Sn	Tin	50	<0.64	<0.64	<0.64	1.17	1.788
Sb	Antimony	51	0.0055	0.0905	0.212	0.452	0.798
Cs	Cesium	55	0.08	0.2	0.33	0.6	1.054
Ва	Barium	56	21	33.5	53	82	128
La	Lanthanum	57	0.6	1.2	2.1	4.8	11.42
Ce	Cerium	58	1.2	2.3	4.2	9.55	20.84
Pr	Praseodymium	59	0.1	0.3	0.5	1.1	2.5
Nd	Neodymium	60	0.6	1.05	2	4.3	9.86
Sm	Samarium	62	0.09	0.2	0.37	0.79	1.922
Gd	Gadolinium	64	0.09	0.2	0.365	0.77	1.869
Hf	Hafnium	72	0.01	0.03	0.06	0.12	0.23
TI	Thallium	81	0.05	0.09	0.15	0.24	0.34
Pb	Lead	82	10.9	18	35	63	95
Bi	Bismuth	83	0.04	0.08	0.12	0.23	0.353
Th	Thorium	90	0.1	0.21	0.37	0.78	1.494
U	Uranium	92	0.05	0.09	0.15	0.33	0.63

Concentrations of trace metals (percentiles) in humic layers of soils in Norway (1995). Units: $\mu g g^{-1}$ Number of sites = 469. Data from Nygård 2000.

Element		Atomic No	10	25	50	75	90
Li	Lithium	3	2.24	5.12	7.71	13.6	21
В	Boron	5	<1.63	<1.05	<1	1.63	4.75
S	Sulphur	16	970	2220	3330	4640	6560
Са	Calsium	20	881	2490	3670	5050	6808
V	Vanadium	23	26.3	43.3	58.6	75.4	87.9
Cr	Chromium	24	11.5	18.35	26.1	38.5	59.82
Mn	Manganese	25	119	420	1590	8180	40900
Fe	Iron	26	21100	46200	71000	120000	183000
Co	Cobalt	27	2.87	7.99	17.6	30	50.3
Ni	Nickel	28	7.42	12.5	19.4	30.9	58.4
Cu	Copper	29	14.3	26.5	41.7	60.6	90.8
Zn	Zinc	30	52	92.6	136	216	360
As	Arsenic	33	2.31	6.66	13.1	30.6	56.8
Se	Selenium	34	0.588	1.35	2.38	4.73	7.14
Br	Bromine	35	37.9	99.55	216	395	717.4
Sr	Strontium	38	8.02	17.4	24.9	33.2	45.84
Мо	Molybdenum	42	0.9427	2.61	4.98	10.8	22.9
Cd	Cadmium	48	0.215	0.462	0.865	1.44	2.62
Sb	Antimony	51	<0.0405	0.124	0.355	0.882	1.43
T	lodine	53	4.43	17.4	37.4	100	221
Cs	Cesium	55	0.461	1.16	1.78	2.81	4.184
Hg	Mercury	80	0.0677	0.147	0.271	0.423	0.573
Pb	Lead	82	22.4	58.8	108	198	286
Bi	Bismuth	83	0.18	0.3845	0.742	1.34	2.126

Concentrations of trace metals (percentiles) in lake sediments in Norway (1995-1996). Units: $\mu g g^{-1}$ Number of sites = 210. Data from Rognerud and Fjeld 1999.

Element		Atomic No	10	25	50	75	90
Li	Lithium	3	0.041	0.078	0.151	0.280	0.427
Be	Beryllium	4	<0.01	<0.01	<0.01	0.016	0.037
В	Boron	5	0.241	0.675	1.295	2.605	4.5614
Ti	Titanium	22	0.8	1.875	4.05	8.925	17.23
V	Vanadium	23	<0.3	<0.3	<0.3	<0.3	0.3
Cr	Chromium	24	<0.1	<0.1	<0.1	0.1	0.3
Mn	Manganese	25	0.5	1.3	3.2	9.025	24.12
Fe	Iron	26	<15	17.1	51.35	135.7	292.08
Co	Cobalt	27	<0.02	0.023	0.048	0.085	0.1936
Ni	Nickel	28	<0.1	0.1	0.2	0.4	0.7
Cu	Copper	29	<0.2	<0.2	0.3	0.6	1
Zn	Zinc	30	0.3	0.8	1.5	3.6	6.93
Ge	Germanium	32	<0.05	<0.05	0.068	0.153	0.3049
As	Arsenic	33	<0.1	<0.1	<0.1	0.14375	0.27
Rb	Rubidium	37	0.1	0.2	0.4	0.8	1.5
Sr	Strontium	38	1.2	2.2	4.7	10.1	20.22
Υ	Yttrium	39	0.02	0.03	0.08	0.19	0.41
Zr	Zirconium	40	<0.015	<0.015	<0.015	0.01625	0.038
Nb	Niobium	41	<0.04	<0.04	<0.04	<0.04	<0.04
Мо	Molybdenum	42	<0.04	<0.04	<0.04	0.1	0.22
Ru	Ruthenium	44	<0.02	<0.02	<0.02	<0.02	<0.02
Rh	Rhodium	45	<0.003	<0.003	<0.003	<0.003	<0.003
Pd	Palladium	46	<0.03	<0.03	<0.03	<0.03	<0.03
Ag	Silver	47	<0.01	<0.01	<0.01	<0.01	0.013
Cd	Cadmium	48	<0.02	<0.02	<0.02	0.03	0.06
Sn	Tin	50	<0.04	<0.04	0.05	0.08	0.14
Sb	Antimony	51	<0.01	0.012	0.02	0.04	0.0723
Ва	Barium	56	0.6	1.2	2.4	5.6	10.79
Ce	Cerium	58	0.028	0.067	0.1775	0.43775	0.9464
Pr	Praseodymium	59	0.005	0.012	0.0315	0.07	0.132
Nd	Neodymium	60	0.019	0.048	0.121	0.276	0.4964
Sm	Samarium	62	<0.015	<0.015	0.021	0.049	0.0973
Eu	Europium	63	<0.004	<0.004	<0.004	0.008	0.015
Gd	Gadolinium	64	<0.015	<0.015	0.021	0.04925	0.0946
Tb	Terbium	65	<0.002	<0.002	0.003	0.006	0.013
Dy	Dysprosium	66	<0.01	<0.01	0.015	0.04025	0.0893
Но	Holmium	67	<0.002	<0.002	0.003	0.006	0.014
Er	Erbium	68	<0.004	<0.004	0.008	0.02025	0.054
Tm	Thulium	69	<0.003	<0.003	<0.003	< 0.003	0.006
Yb	Ytterbium	70	<0.006	<0.006	0.007	0.017	0.039
Lu	Lutetium	71	<0.002	<0.002	<0.002	0.003	0.006
Hf	Hafnium	72	<0.03	< 0.03	<0.03	<0.03	<0.03
Та	Tantalum	73	<0.01	<0.01	<0.01	<0.01	<0.01
W	Tungsten	74	<0.02	<0.02	<0.02	<0.02	<0.02

Concentrations of trace metals (percentiles) in lakes in Norway (autumn 1995). Units: $\mu g L^{-1}$ Number of sites = 998. Data from this report.

Element		Atomic No	10	25	50	75	90
Os	Osmium	76	<0.5	<0.5	<0.5	<0.5	<0.5
Ir	Iridium	77	<0.006	<0.006	<0.006	<0.006	<0.006
Pt	Platinum	78	<0.02	<0.02	<0.02	<0.02	<0.02
ΤΙ	Thallium	81	<0.006	<0.006	<0.006	0.009	0.017
Pb	Lead	82	0.05	0.09	0.17	0.42	0.874
Bi	Bismuth	83	<0.02	<0.02	<0.02	<0.02	0.04
Th	Thorium	90	<0.015	<0.015	<0.015	<0.015	0.016
U	Uranium	92	<0.004	0.011075	0.0324	0.083875	0.18485

Appendix B. Maps

Maps of elements in lake water ($\mu g/L$), lake sediments ($\mu g/g$), terrestrial moss ($\mu g/g$) and the humic surface layer of natural soils ($\mu g/g$). Cerium (58) is used to illustrate alle the lanthanoides.

Element		Atomic no.	Lake water	Lake	Terrestrial	Natural
				sediments	moss	surface soil
Li	Lithium	3	x	x	x	x
Be	Beryllium	4	x	x	x	x
В	Boron	5	x	x		x
Ti	Titanium	22	x	x	x	x
V	Vanadium	23	x	x	х	x
Cr	Chromium	24	x	x	x	x
Mn	Manganese	25	x	x	x	x
Fe	Iron	26	x	x	x	
Со	Cobalt	27	x	x	x	x
Ni	Nickel	28	x	x	x	x
Cu	Copper	29	x	x	x	x
Zn	Zinc	30	x	x	x	x
Ge	Germanium	32	x		x	x
As	Arsenic	33	x	x	x	x
Rb	Rubidium	37	x		x	x
Sr	Strontium	38	x	x	x	x
Y	Yttrium	39	x	x	x	x
Zr	Zirconium	40	x	x	x	x
Мо	Molybdenum	42	x	x	x	x
Cd	Cadmium	48	x	x	x	х
Sn	Tin	50	x	x	x	x
Sb	Antimony	51	x	x	x	х
Ва	Barium	56	x	x	x	x
Ce	Cerium –	58	x		x	x
ТΙ	Thallium	81	x	x	x	x
Pb	Lead	82	x	x	x	х
Bi	Bismuth	83	x	x	x	x
Th	Thorium	90	x	x	x	х
U	Uranium	92	x	x	x	x

Lithium - Li



Beryllium Đ Be





Titanium Đ Ti

Vanadium Đ V

Chrome Đ Cr

Manganese Đ Mn Lake water Lake sediments Mn, μg/l Mn, % . ≥100 ≥10 10 1 ≤1.0 ≤0.01 **Terresterial mosses** Humic matter Mn, μg/g Mn, µg/g 5000 1000 1000 100 100 20 10

Rubidium Đ Rb

Strontium Đ Sr

Zirconium Đ Zr

Cadmium Đ Cd

Uranium Đ U

Antimony **Đ** Sb

Thallium Đ Tl

Bismuth Đ Bi

