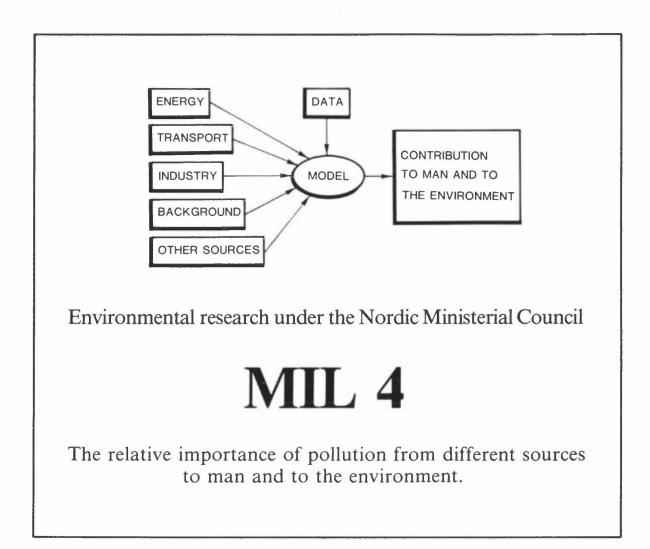
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# CONTRIBUTION FROM LONG RANGE ATMOSPHERIC TRANSPORT TO THE DEPOSITION OF TRACE METALS IN SOUTHERN SCANDINAVIA

Eiliv Steinnes



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Eiliv Steinnes\*

Affiliation: Department of Chemistry, University of Trondheim 7055 Dragvoll, Norway

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ABSTRACT

This report is an attempt to quantify the contribution of long-range atmospheric transport to the deposition of trace metals in Southern Scandinavia, as a basis for assessing the importance of the "background" component relative to other air pollution sources to the metal exposure of the population. The results, which reflect the situation about 1980, are presented as isopleths for each element and are based on wet deposition data from precipitation analyses and relative deposition data from moss analyses. The elements considered are V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb and Pb.

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Department of Chemistry, University of Trondheim, 7055 Dragvoll, Norway

## 1. INTRODUCTION

In air pollutant exposure studies aimed at identification of contributions from different source categories a "background" component due to long-range atmospheric transport from distant source regions must be taken into account. In some areas such as parts of Southern Scandinavia this long-range component may constitute a very significant part of the total exposure. The present report is an attempt to estimate the geographical variation in the "background" component of trace metal deposition in southern and central parts of Norway and Sweden during the time period around 1980, mainly on the basis of available data from moss surveys and precipitation analyses. The elements selected for study are V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, and Pb.

#### 2. AVAILABLE DATA

#### 2.1. Moss Surveys

Since the pioneer work of Rühling and Tyler (1), using the forest moss <u>Hylocomium splendens</u> to monitor relative deposition rates of trace elements from the atmosphere, several comprehensive moss surveys have been carried out in Scandinavia. In Sweden national surveys were performed in 1975 (2) and 1980. (3). Results from the 1980 study are presented jointly with those from a simultaneous Danish study in a recent report (4). In Norway, a national moss survey took place in 1977 (5). While the Swedish and Danish studies were based on atomic absorption spectroscopy (AAS) and comprised 10 elements, the Norwegian study included 27 elements owing to the use of instrumental neutron activation analysis (INAA) in combination with AAS (6).

The basis for using mosses as indicators of air pollution is that they absorb ions mainly from the atmosphere (7). Although this assumption is probably valid for many metal ions, it does not hold for all elements of interest in air pollution. The concentration of Mn in <u>Hylocomium</u> <u>splendens</u> e.g. is generally far too high to be explained from atmospheric uptake only, and its geographical distribution in Scandinavia (8,9), showing relatively low values along the coast rapidly increasing inland, may indicate that

Mn<sup>2+</sup> in the moss is being exchanged with sea-salt cations such as Mg<sup>2+</sup>. Also in the case of Zn some results from the Norwegian 1977 survey (5) indicate similar difficulties, but to a lesser extent. Moreover the sorption efficiency shows considerable variation among different trace elements (10). Finally the uptake of trace elements from dry deposition may be expected to depend on the particle size distribution of the aerosol and may in any case differ from that observed from wet deposition. In spite of these limitations however the moss technique may yield a quite reliable measure of relative deposition rates of most elements involved in the present work, at least in areas distant from the major sources.

#### 2.2. Precipitation data

Trace element deposition from the atmosphere in areas far from the source region may be assumed to depend largely on wet deposition (see section 2.4). Weighted mean values for trace elements in precipitation may thus allow a quite good estimate of the <u>total</u> deposition of the elements concerned during the collection period at a "background" sampling site. Up to a few years ago very few data existed in Scandinavia for trace metals in precipitation at "background" . locations. Recently however a fair number of data have become available from 4 locations in Norway (10-12) and 10 locations in Sweden (13-18). Observations from a

number of rural stations in Denmark (18,19) may also be of some help in estimating deposition levels in Southwest Sweden. The stations from where data sets are available are listed in Table 1 with information on the length of the sampling period and the elements studied. The locations of the various stations are shown on the map in Fig. 1.

#### 2.3. Air concentration data

Data for air concentrations of trace elements associated with particulates in "background" areas in Scandinavia are even more scarce than data on precipitation. Air concentration data cannot be used straight-forward to calculate deposition values, but they may still provide additional information when used in connection with other data.

One of the most comprehensive sets of data available to far for trace element concentration in air at "background" locations in Scandinavia seems to be that of Hanssen et al. (10) obtained during 11 months in 1978-79, where 48 hours' air filters and weekly precipitation samples were collected at Birkenes and several other Norwegian stations and subsequently analyzed for as many elements as possible by INAA and AAS. While most of the data for trace element contents in precipitation are somewhat questionable due to the very low concentration involved, the air concentration data from filter analyses are much more consistent. By selecting an element for which good data exist for both media, the ratio of element in air/element in precipitation can be established e.g. on the basis of weighted average values over the whole observation period. If the removal from the air by precipitation is equally efficient for other trace elements, one can use the same ratio to calculate what the weighted mean concentration of a given element should have been in precipitation, and consequently its deposition rate, from its corresponding value in air. This seems to be a reasonable assumption in the Birkenes case, where the data for most trace metals mainly associated with episodes of long-range transport are strongly intercorrelated in the air filter samples (10).

Air particulate data may also be useful to estimate relative contributions from different source regions to the trace metal deposition at a given site, either by air trajectory analysis on selected episodes (20,21) or by employing a sector sampler (22,23). A trajectory analysis of the above Birkenes data (21) indicates that for several trace elements more than 80 % of the amount deposited is likely to be due to long-distance atmospheric transport from areas out of Similar estimates were made in Sweden (22,23) Norway. based on sector samples from Sjöängen (Station 14 in Fig. 1) and calculation of the contributions from domestic sources based on known emission figures and meteorological data. In the samples from the south-east and south-west sector, only 20-30 % of the elements V, Ni and Pb could be accounted for by Swedish sources.

## 2.4. Dry deposition/wet deposition

Many precipitation samplers used in Scandinavia so far are of the open type, which measure some dry deposition in addition to the wet deposition. No attempts to <u>measure</u> the contribution of dry deposition to the total deposition seem to have been made in background locations of Sweden or Norway. Lannefors et al. (23) compared the measured wet deposition at Sjöängen with calculated dry deposition values to a forest surface, and arrived at figures of the order of 10-20 % dry deposition for elements associated with the small-particle fraction. In the present work we shall assume that the deposition values calculated from precipitation analyses closely represent the total deposition.

## 2.5. Temporal variations

Comparison between the results of the 1975 and 1980 moss surveys in Sweden (3) showed that the deposition of many heavy metals had been considerably reduced in different parts of the country. In most cases this is likely to be due to substantial reductions in the emission from Swedish industries. Some fetures however, such as the appreciable reduction in Pb (40 %) and Cd levels in Southwestern Sweden, would . indicate even a reduced supply of some elements by long-distance atmospheric transport from 1973-74 to 1978-79. Unfortunately none of the Swedish precipitation measurements mentioned above date further back than 1980, and all of them last for periods of 2 years or less. It is therefore difficult to know anything about the trend in recent years.

In Norway regular measurements have been carried out on samples from several precipitation stations with respect to Pb and Cd since 1978 (10-12). Weighted annual mean values are listed in Table 2. These results indicate that there has been no obvious change in the atmospheric supply of these elements during the period 1978-82.

In the present report all the available deposition values based on precipitation analyses are assumed to represent the 1980 level.

## 3. PRESENTATION AND DISCUSSION OF DATA

It was decided to present isopleth diagrams of trace element depositions based on moss analyses whereever such data existed and were considered reliable from a methodological point of view. In order to convert moss data to deposition figures, it is necessary somehow to estimate conversion factors. This was done on the basis of deposition data at Birkenes 1978-79 (10) and mean trace metal concentrations in the Birkenes area obtained from the 1977 moss survey (5). For most elements deposition rates were estimated from air concentrations (as outlined in Section 2.3), rather than calculated from precipitation data. In this way deposition figures were obtained for elements which were normally not detected in precipitation samples. A detailed account of this work will be published elsewhere (24). The conversion factors thus obtained were applied also to the Swedish 1980 moss data (3). Maxima on the moss diagrams likely to be caused by local sources were disregarded when drawing the isolines.

A possible decrease in trace metal depositions between 1975/76 and 1978/79 would lead to factors showing a too high deposition / moss content ratio. In that case the deposition levels as expressed by the isolines would . tend to be systematically low on the Swedish part of the map relative to the Norwegian. The extent of such errors is difficult to estimate, but they would hardly be expected to exceed 20-30 %. Deposition values based on precipitation analyses from individual stations are also plotted on the isopleth maps for comparison. Maps for each of the thirteen elements considered are shown in Figs. 2-14. In the following some comments on the various elements are presented.

## 3.1. Some general remarks

In Norway the group of elements most strongly associated with long-distance atmospheric transport (V, Zn, As, Se, Cd, Sb, Pb) shows a very rapid decrease in deposition inland from the southern coast. These elements also show a distinct, but less pronounced south-north gradient in Sweden (as far as available data can tell). The different trend in the two countries may be mainly associated with the distinct differences in topography yielding very different regional precipitation patterns. A higher relative contribution in Sweden to "background" pollutant levels from domestic sources may also be significant in this respect.

A second group of elements, including Cr, Mn, Fe, and Ni, shows a less distinct but still significant south-north gradient. For the remaining element Co and Cu the regional deposition differences appear to be smaller than for the other elements.

## 3.2. Vanadium

Vanadium is the element showing the least consistency between the data from the two countries. While the Norwegian moss data indicate a typical long-distance transport pattern, the Swedish data show no clear regional pattern at all, except for low levels in the north-west. Part of the difference could be associated with differences in analytical methodology (NAA in Norway, graphite furnace AA in Sweden).

## 3.3. Chromium

It is difficult to estimate "background" levels of Cr in Central Sweden due to very dominant contributions from local industries.

#### 3.4. Manganese

As indicated above (Section 2.1) the moss tecnique is not applicable for Mn, and the data from individual deposition measurements are too few to allow isolines to be drawn.

## 3.5. Iron

Fe data are available from the Norwegian moss survey, but the use of them is difficult because the Fe content is very sensitive to contribution from soil dust.

#### 3.6. Cobalt

The only thing that can be said so far about this element is that the deposition of Co in Southern Scandinavia is lower than for any other element discussed in this report.

## 3.7. Nickel

While the Ni deposition in Norway is affected by longdistance transport, domestic sources seem to be more important in determining "background" levels in Sweden.

## 3.8. Copper

Regional differences in copper deposition are apparently quite small over the whole area.

### 3.9. Zinc

It is possible that the moss does not give a completely adequate picture of the zinc deposition because it may absorb Zn from the substrate for physiological reasons and exchange it for seasalt cations (Section 2.1). Nevertheless the measured deposition values agree rather well with those estimated from moss analysis, except the values from ref. , (16) which appear systematically high for Zn.

## 3.10. Arsenic

The Swedish moss data for As are not consistent with those from the Norwegian survey, and it is therefore difficult to extend the isolines across the Swedish border. Also the absolute concentration levels in the moss are different in the two studies. Part of the discrepancy may be associated with differences in analytical methodology.

## 3.11. Selenium, antimony

No air quality data exist for Se and Sb in Swedish "background" areas so far.

## 3.12. Cadmium, lead

For these two elements there exist enough precipitation data to verify the general trend indicated from the moss surveys.

## 3.13. Uncertainty of data

The uncertainty of the diagrams presented in the figures depends on the uncertainty in the basic data used. It is believed that all data expressed in the form of isolines are accurate within a factor of two. In favourable cases the data may be accurate to  $\pm$  10-20 %.

#### 4. SUGGESTIONS FOR FUTURE WORK

<u>A.</u> Moss analysis is a useful means of obtaining relative deposition pattern for trace metals. Future national surveys in the Nordic countries should be coordinated such that the sampling takes place the same year, the same moss species is used and preferably the same elements are determined using the same analytical methods. If different methodologies are employed, samples should be exchanged for intercomparison.

<u>B.</u> More work should be done to establish conversion factors between trace element concentrations in moss and deposition values. In particular it is important to know if the same conversion factor can be used in different geographical locations.

<u>C.</u> Moss surveys cannot be performed very frequently. In order to follow the temporal trend in trace metal deposition more closely, it is important that precipitation samples from a fixed number of selected stations be analyzed regularly with respect to these elements. The above remarks on standardization of sampling and analytical techniques and exchange of intercomparison samples apply also in this case.

## 5. ACKNOWLEDGEMENTS

The author wants to thank all those investigators who submitted their unpublished data. In particular thanks are due to Dr. Åke Rühling who provided raw data from the 1980 moss survey in Sweden plotted on maps.

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Table 1. Available data on trace metal deposition in southern Scandinavia, based on precipitation analyses.

Loca	tion (see Fig. 1)		Sampling	
No.	Name	Investigator	period	Elements
1.	Birkenes	NILU (10-12)	78.8-82.12	Cu,Zn,As,Se, Cd,Sb,Pb
2.	Kise	" (12)	80.5-82.12	
3.	Narbuvoll	" (10,11)	78.8-82.12	_ " _
4.	Kårvatn	" (10-12)	78.8-82.12	_ " _
5.	Lammhult	Andersson and		
		Borg (13)	82.7-83.9	Cu,Zn,Cd,Pb
6.	Dalsbo	_ " _	83.1-83.9	- " -
7.	Lofsdalen	- " -	83.7-83.11	_ " _
8.	Uppsala	Andersson (14)	81.4-83.1	Mn,Cr,Fe,Ni,Cu
			×.	Zn,As,Cd,Pb
9.	Skånes Värsjö	Bergkvist (15)	80.6-82.8	V,Cr,Mn,Fe,Ni,
				Cu,Zn,Cd,Pb
10.	Svartberget	Grahn and Rosén (16)	80.10-81.9	V,Cr,Mn,Fe,Co,
				Ni,Cu,Zn,Cd,Pb
11.	Kullarna	- " -	80.10-81-9	<u> </u>
12.	Gårdsjön	_ " _	80.10-81.9	_ " _
13.	Nyköping,			
	2 stations	Grahn (17)	81.10-82.7	Mn,Fe,Cu,Zn,
				Cd, Pb
14.	Sjöängen	Hovmand (18)	82.1-82.12	Mn,Fe,Ni,Cu,
				Zn,Cd,Pb
15.	Jylland,			
	5 stations	Juhl Larsen (19)	80.6-81.10	Cu,Cd,Pb
16.	Denmark,			
	5-6 rural sites	Hovmand (18)	82.1-82.12	Mn,Fe,Ni,Cu,
				Zn,Cd,Pb

-		-			
		1978/79	1980	1981	1982
	Birkenes	11.4	8.4	9.1	12.9
	Kise	-	4.1	2.4	2.8
Pb .	Narbuvoll	1.9	1.6	1.3	1.1
	Kårvatn	2.0	1.8	1.4	1.7
	Jergul	1.2	0.6	0.7	1.1
	Birkenes	0.28	0.36	0.29	1.04 *)
	Kise	- 8	0.07	0.05	0.10
Cd	Narbuvoll	0.08	0.09	0.07	0.04
	Kårvatn	0.05	0.06	0.05	0.08
	Jergul	0.07	0.02	0.02	0.05

Table 2. Deposition of lead and cadmium in Norway (10-12) from precipitation data (mg  $m^{-2}y^{-1}$ ).

\*) Contamination problems suspected.

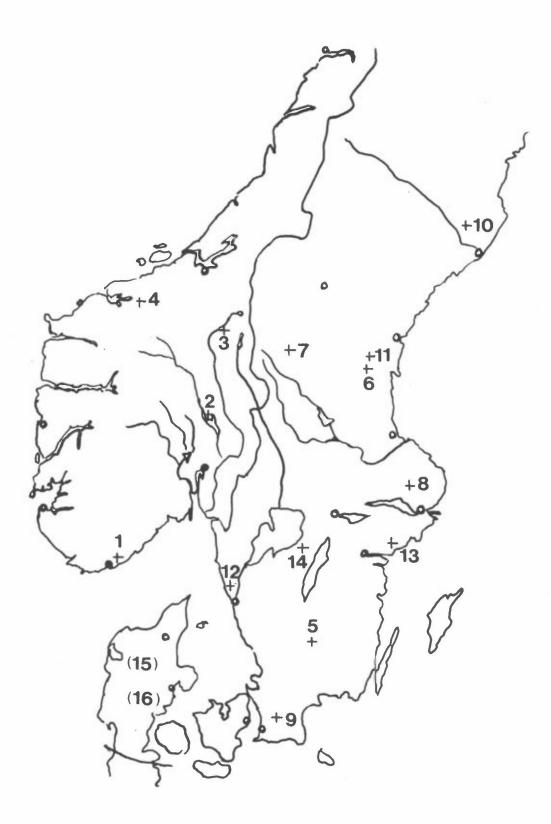


Fig. 1. Sampling sites for studies of trace metal deposition by precipitation analyses (cf. Table 1).

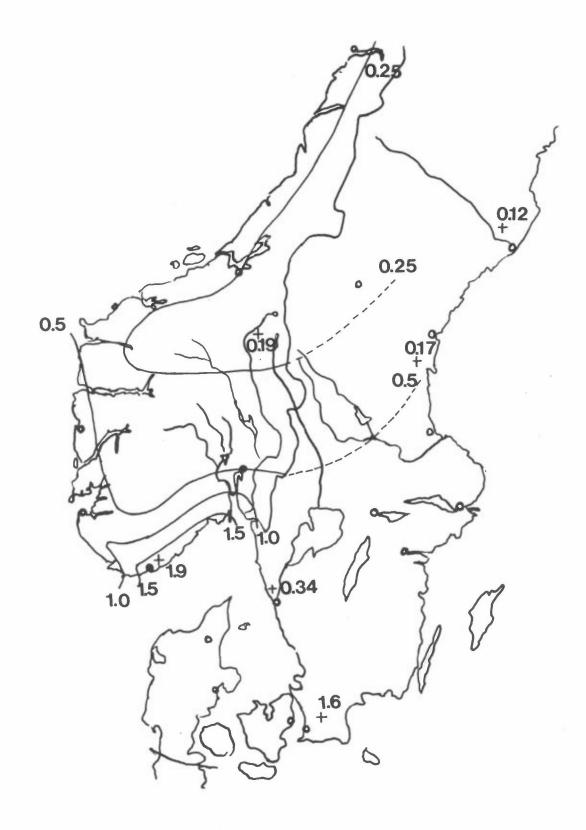


Fig. 2. Deposition of vanadium in Southern Scandinavia (mg  $m^{-2}y^{-1}$ )

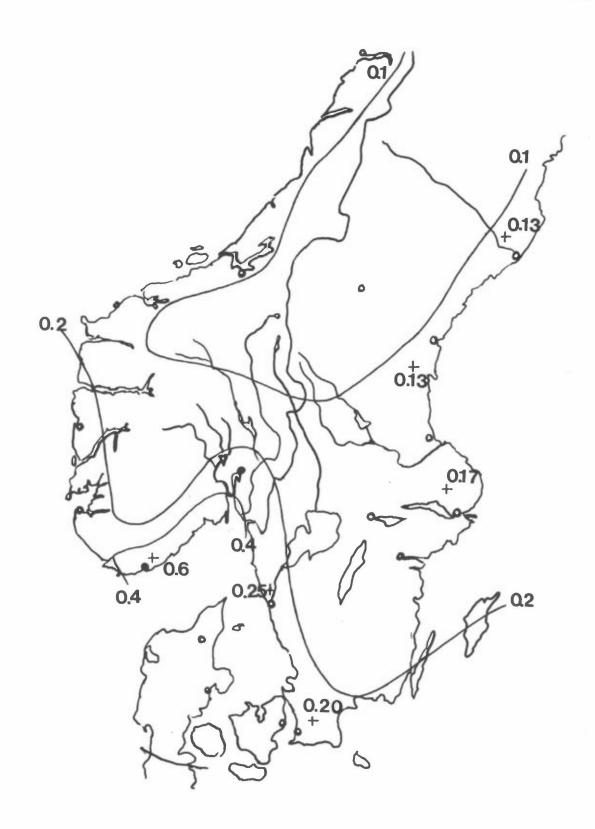
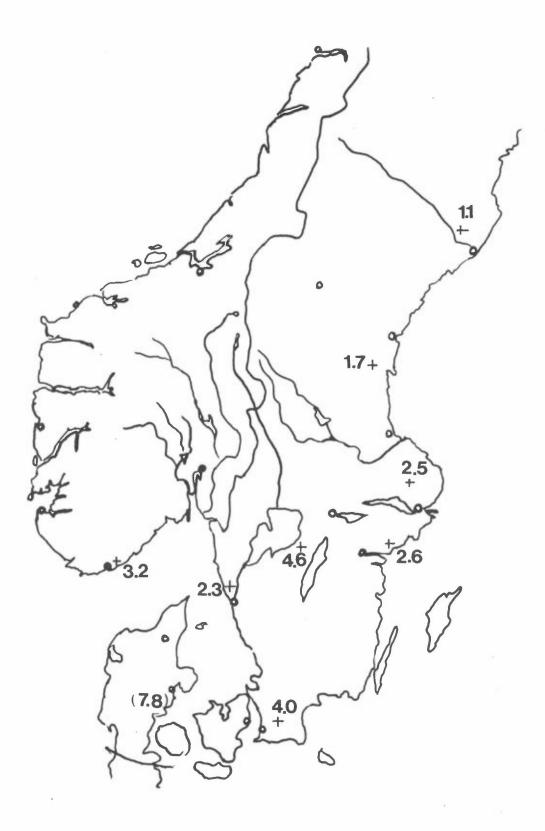


Fig. 3. Deposition of chromium in Southern Scandinavia (mg  $m^{-2}y^{-1}$ )



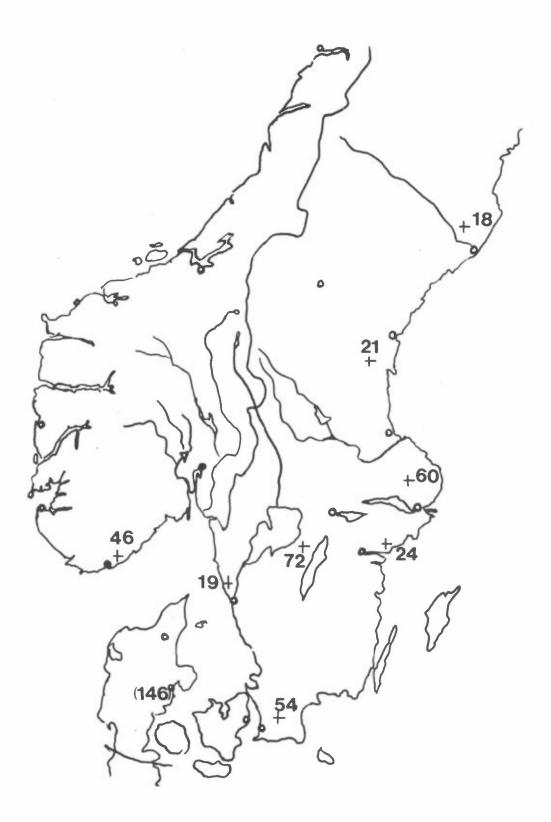


Fig. 5. Deposition of iron in Southern Scandinavia (mg  $m^{-2}y^{-1}$ )

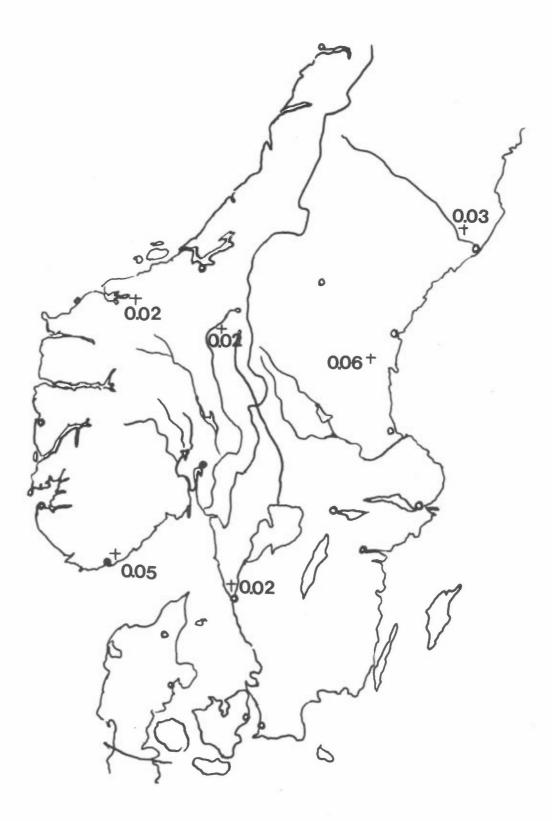


Fig. 6. Deposition of cobolt in Southern Scandinavia (mg  $m^{-2}y^{-1}$ )

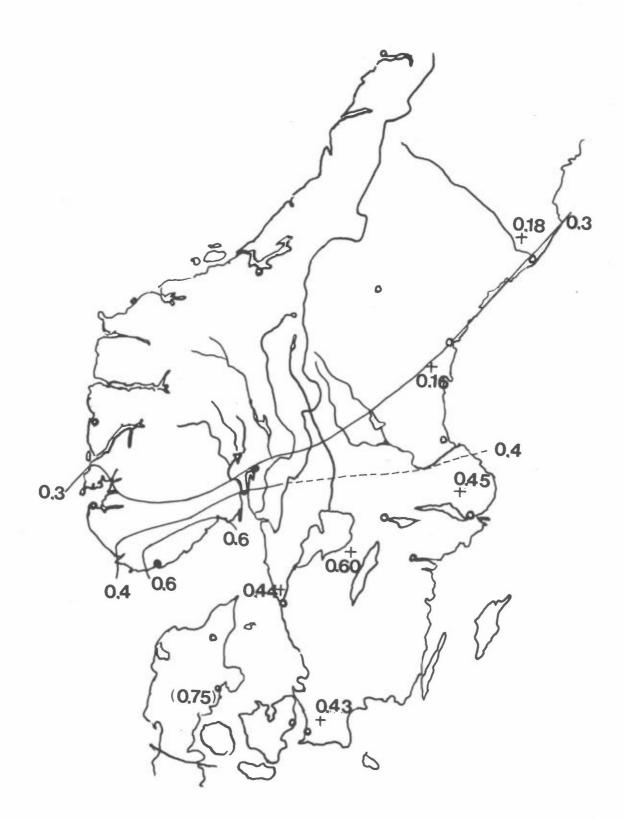


Fig. 7. Deposition of nickel in Southern Scandinavia ( $mg m^{-2}y^{-1}$ )

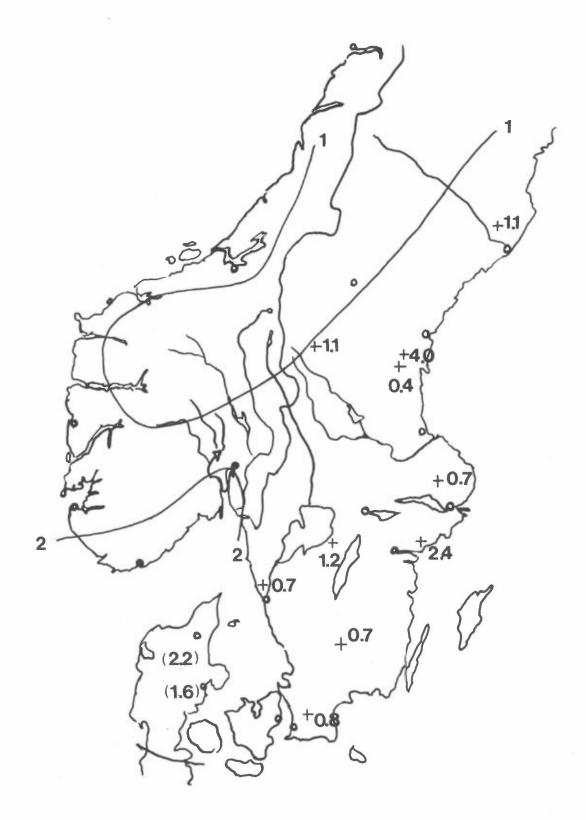
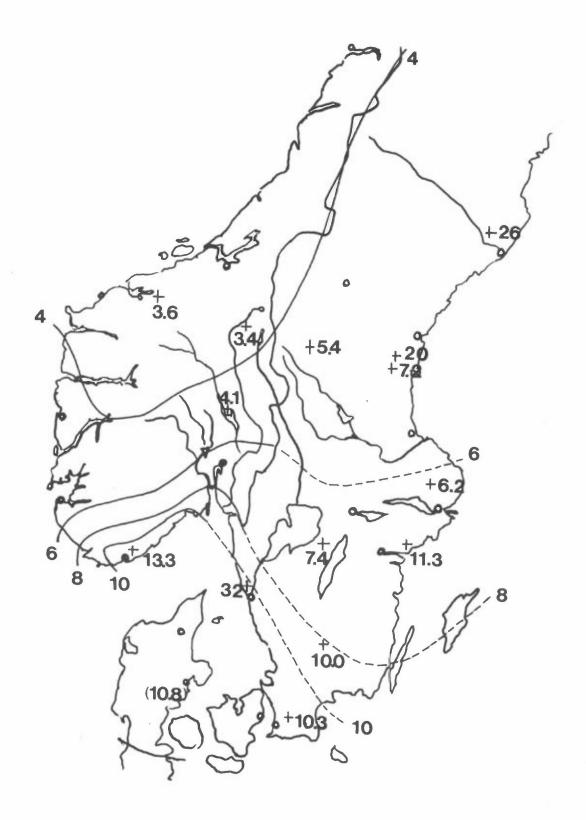
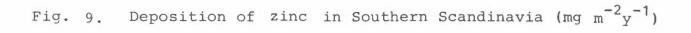


Fig. 8. Deposition of copper in Southern Scandinavia (mg m $^{-2}y^{-1}$ )





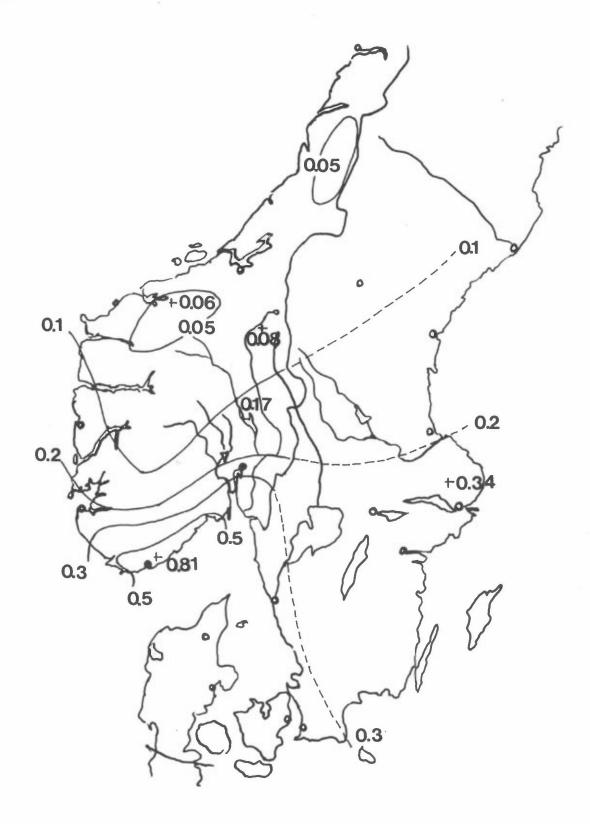


Fig. 10. Deposition of arsenic in Southern Scandinavia (mg  $m^{-2}y^{-1}$ )

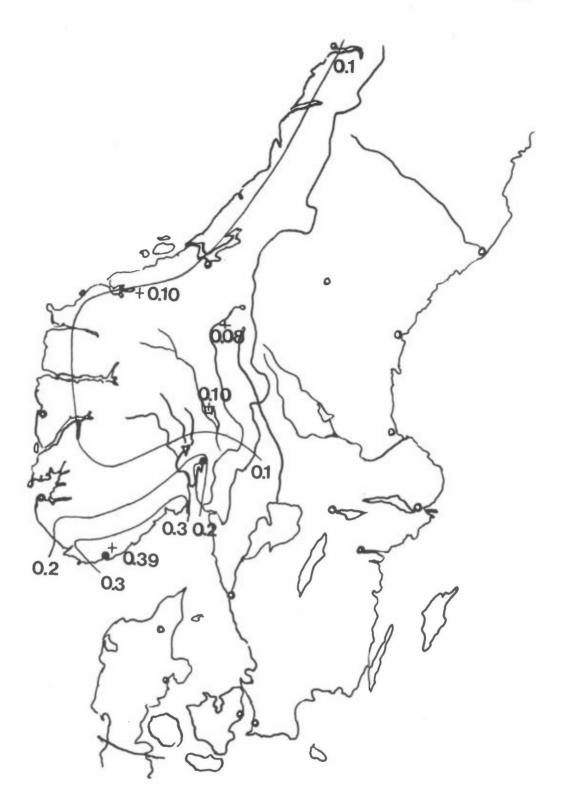


Fig. 11. Deposition of selenium in Southern Scandinavia (mg  $m^{-2}y^{-1}$ )

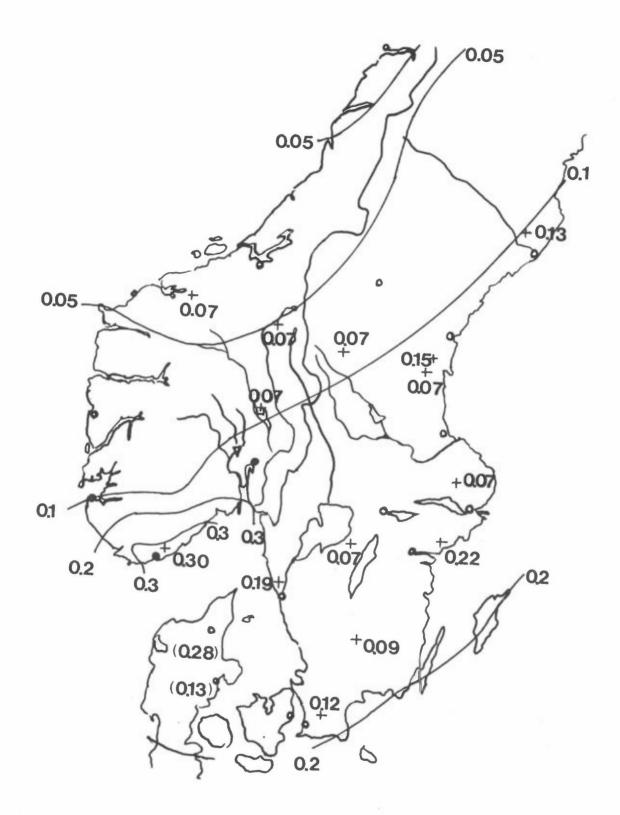
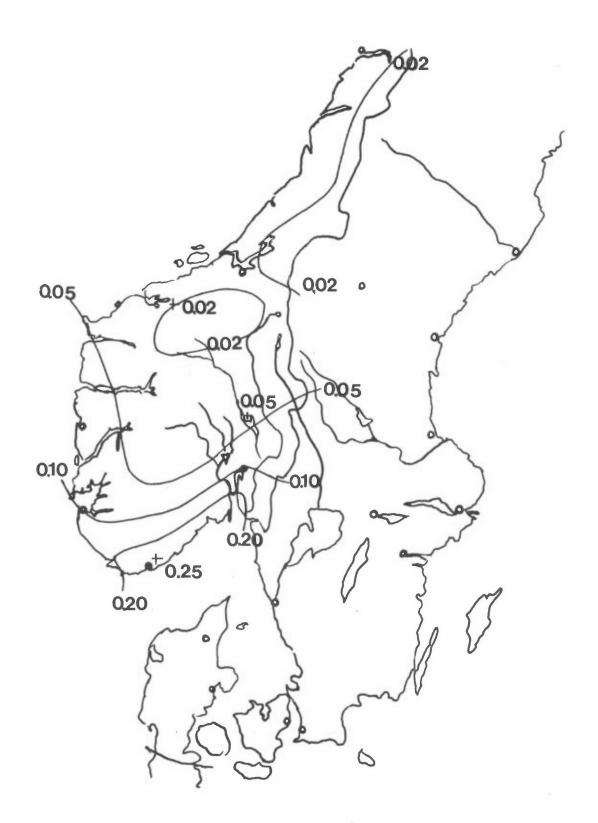
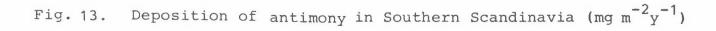
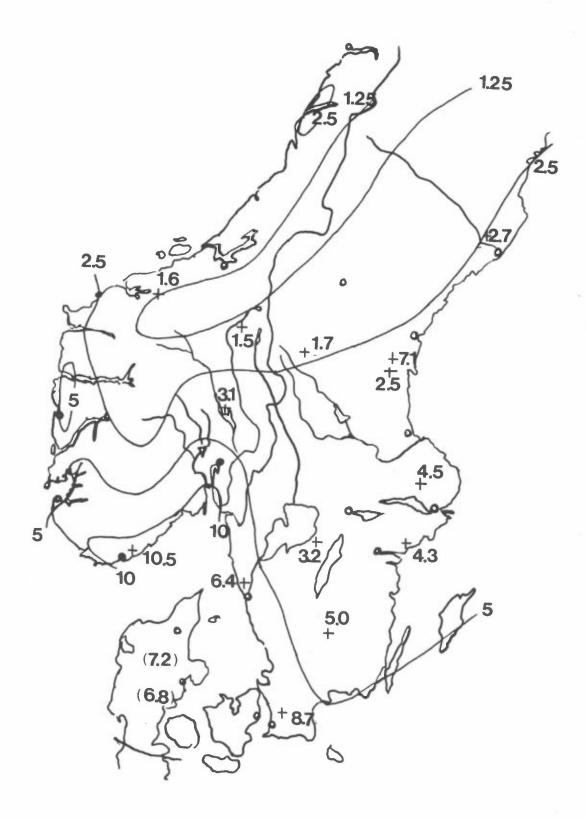


Fig. 12. Deposition of cadmium in Southern Scandinavia (mg  $m^{-2}y^{-1}$ )









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in Southern Scandinavia	NILU PROSJEKT NR. 0-8052				
FORFATTER(E)	TILGJENGELIGHET				
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TITLE

ABSTRACT (max. 300 characters, 7 lines) The contribution of long-range atmospheric transport to the deposition of trace metals in Southern Scandinavia was quantified to assess the "background" component relative to other air pollution sources. The results, which reflect the situation about 1980, are presented as isopleths for each element and are based on wet deposition data from precipitation analyses and relative deposition data from moss analyses. The elements considered are V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb and Pb.

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