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METAL POLLUTION AROUND AN IRON SMELTER COMPLEX IN NORTHERN NORWAY AT DIFFERENT MODES OF OPERATION

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INTRODUCTION

Operations in and around metal smelters often cause substantial deposition of metal-containing dust, which may significantly affect human and environmental health in the surrounding areas. The classical approaches to study the extent of such pollution is either by measuring air concentrations based on aerosol collection on filters, or by analysis of surface soil samples. In recent years the use of biomonitoring techniques to record atmospheric deposition patterns around point sources of air pollution has proven to be a useful alternative to the conventional techniques. In particular the sampling of terrestrial mosses around pollution sources has appeared to be a powerful approach in studies related e.g. to thermal power plants (Folkeson, 1981) and Cu-Ni smelters (Äyräs et al., 1997). In the present work application of this technique to monitor the metal pollution from iron smelters is demonstrated.

Although the main problem of atmospheric metal deposition in Norway is associated with long-range transport from other European countries (Steinnes et al., 1989; Berg et al, 1995) there are a few industries that are sources of significant metal pollution on a local scale.

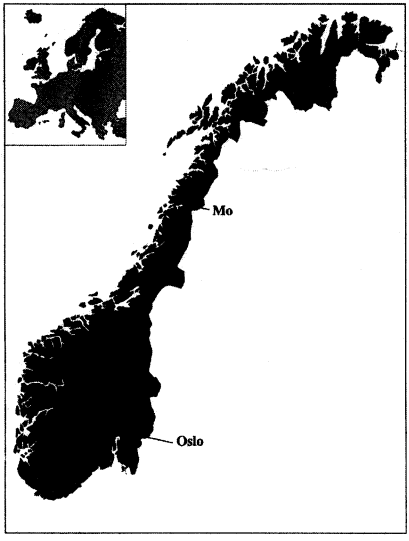

Fig. 1. Map of Norway showing the position of Mo i Rana

One of those is the iron industry at Mo i Rana, Nordland county (Fig. 1), established in 1946 and operated for four decades as a regular iron smelter. Financial and other considerations led to the decision to close this smelter in 1989 and use the available facilities for some alternative industries, including a ferrochrome smelter.

Prior to the re-orientation of the Mo industries the Norwegian State Pollution Control Authority required a survey to be carried out to assess how the change would affect the deposition of heavy metals from the industries. A monitoring network based on the collection of moss and surface soil samples was therefore established in 1989, and the samples were analysed for a given set of metals using standard atomic absorption (AAS) techniques (Steinnes et al., 1992). In 1993 the moss sampling was repeated and the moss samples were analysed for the same elements as before (Steinnes, 1995). The results showed that very extensive changes had occurred for some of the elements.

In the meantime epithermal neutron activation analysis (ENAA) had been demonstrated to be a powerful technique for multi-element analysis of mosses used for biomonitoring of trace element deposition (Frontasyeva et al., 1994). It was therefore decided to analyse the moss samples collected in the Mo area in 1989 and 1993 also by this technique. The ENAA appeared to provide additional information for almost 30 elements in these moss samples (Frontasyeva and Steinnes, 1995). Previously published work on deposition of trace elements in the vicinity of smelter industries appears to have been concentrated almost entirely on non-ferrous metal smelters. It was therefore thought that the results from the present work might be of more general interest than just as a separate case study. Consequently it was decided to combine all the above data and discuss them in a separate paper, in particular with respect to contributions from different sources to the observed heavy metals.

BIOMONITORING OF HEAVY METAL DEPOSITION USING MOSS ANALYSIS

Mosses have no root system and therefore normally receive their content of essential nutrients as well as other chemical substances from other sources than the soil. Atmospheric deposition is a major source of supply for a large number of elements to the moss. Moreover mosses have a high capacity to retain many elements. Mosses have been used for a long time to monitor the atmospheric deposition of heavy metals over large areas (Rühling and Tyler, 1973), and the moss technique is now employed every five years for monitoring of metal deposition on the European scale (Rühling, 1994; Rühling and Steinnes, 1998). Applications around point sources of metal emission have been limited in some cases because mosses are sensitive to high concentration of some heavy metals (Bengtsson et al., 1982; Tyler, 1990). In the case reported in this work however moss could be sampled as close as a few hundred metres from the emission sources.

Compared to analysis of surface soil the moss technique gives a relative measure of the deposition over a given period, normally the last 2-3 years. When employing *Hylocomium splendens*, such as in this work, the annual growth segments can be separated and the exposure period thus unambiguously defined. In the present case the last 3 years' growth was used. Sampling of surface soil on the other hand gives an integrated measure of components supplied from the atmosphere and firmly fixed in the upper few cm. Nothing can be said about the temporal trend of the deposition unless the soil sampling is carried out at time intervals and the incremental supply is calculated by difference.

SAMPLING AND ANALYSIS

The town of Mo i Rana is situated at 66° 20′ N, 14° 10′ E in the inner end of the eastbound Rana fjord, at about 60 km distance from the Norwegian Sea, and has around 20 000 inhabitants. The Rana fjord and the Rana valley extending eastwards from Mo are shielded by high mountains (700–1400 m altitude) in the north and south directions, as indicated by the topographic map in Fig. 2.

Fig. 2. Map showing the main topographic features of the area surrounding Mo i Rana

Correspondingly the prevailing local wind directions are either westerly or easterly. The metal industries are situated within the town area, about 1 km from the center, as indicated in Fig. 3.

Samples of the feather moss *Hylocomium splendens* and surface soil were collected in 1989 at 39 sites at distances of 1–42 km from the Mo industries. When planning the sampling network, particular attention was paid to the rather complex topography of the area surrounding Mo. The sampling network is shown in Fig. 4.

Soil samples were collected at 2–5 cm depth in the humic horizon. Five subsamples collected within an area of 10m x 10m were combined to form one sample. After drying and sieving (2 mm) 2-gram portions of the samples were decomposed in concentrated nitric acid (Steinnes et al., 1989). Concentrations of selected heavy metals were determined by flame atomic absorption spectrometry (AAS) using the flame (Cr, Fe, Cu, Zn, Pb), graphite furnace (Cd), or the hydride (As, Sb, Hg) technique.

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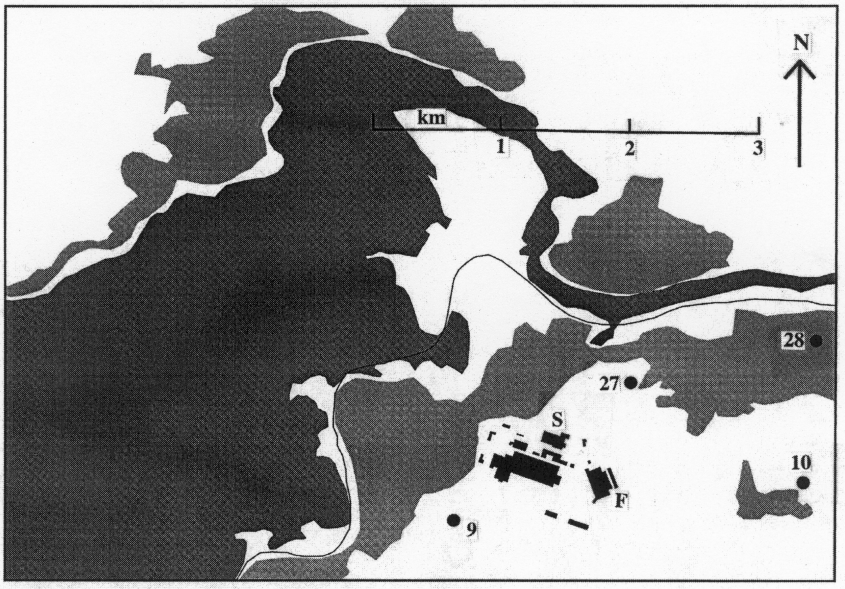

Fig. 3. Map of Mo i Rana showing the location of the industrial area and its main units after 1990. Grey colour: main urban area. Dark colour: Rana river and the innermost part of the Rana Fjord. S: Iron smelter. F: Ferrochrome smelter. 9, 10, 27, 28: sampling points, cfr. Fig. 4

The moss samples were collected and further prepared for analysis according to a standard procedure described elsewhere (Steinnes et al., 1992). Based on the analytical results for the surface soils, moss samples from 25 out of the 39 sites were selected for analysis. At these 25 sites a repeated sampling of moss was carried out in 1993.

Moss samples from the 1989 and 1993 collections were analysed for their concentrations of 35 elements using epithermal neutron activation analysis (ENAA) (Na, Mg, Al, Cl, K, Ca, Sc, V, Cr, Mn, Fe, Co, As, Se, Br, Rb, Sr, Zr, Mo, Ag, Sb, I, Cs, Ba, La, Sm, Eu, Tb, Yb, Lu, Hf, Ta, Au, Th, U) and 3 elements using AAS (Ni, Cu, and Zn). ENAA was carried out at the IBR-2 pulsed fast reactor in Dubna according to procedures described elsewhere (Frontasyeva et al., 1994; Frontasyeva and Steinnes, 1995). A summary of results for the moss samples is presented in Table 1. The accuracy of the analyses was checked by means of international standard reference materials run together with the present samples.

The underlying relations between the chemical parameters for the moss samples were investigated using factor analysis. The analyses were performed separately for the two data sets from 1989 and 1993 using principal component extraction, 1 as the eigenvalue factor selection criterion, and VARIMAX rotation of the extracted factors. Variables with factor loadings higher than 0.6 (cfr. Table 2) were assumed to contribute significantly to a given factor. Similarly factor analysis was applied to the soil data (cfr. Table 3).

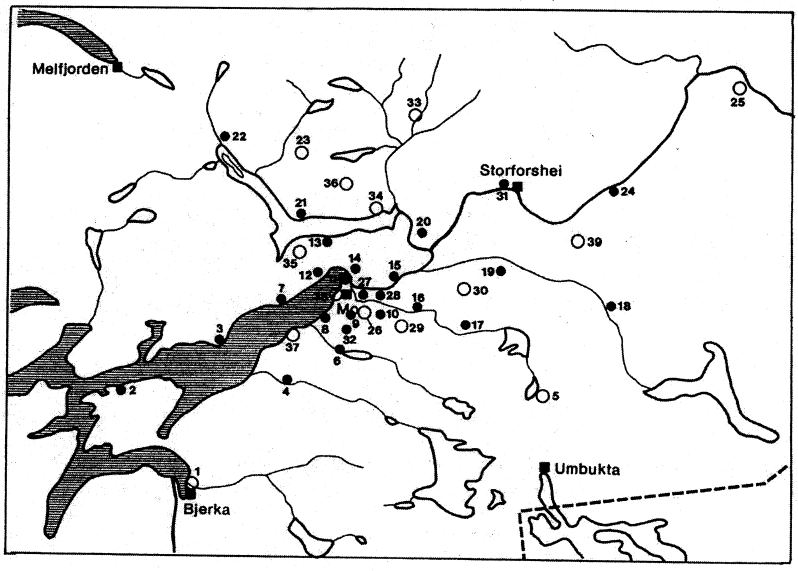

Fig. 4. Sampling network used in the present work. Closed circles: Soil samples (1989) and moss samples (1989 and 1993). Open circles: only soil samples

RESULTS AND DISCUSSION

Most of the elements are enriched in samples collected near Mo, indicating local sources. Of the elements studied in moss samples only Cl, K, Ca, Se, Rb, I, and Cs appear to be virtually independent of industrial and other activities within the urban area.

All the remaining elements seem to be influenced by these activities one way or the other by showing higher concentrations at sites within Mo than in the surrounding region. A closer examination of the data for sites close to the industrial area show the highest deposition occurring at sites 9 and 10 to the south for some elements whereas others exhibit higher concentrations in the moss at sites 27 and 28 in the north-eastern direction. However, this pattern changes for many elements between the two points of time, indicate that the character of the emission sources and distribution of emissions between them changed very much. Isopleths showing relative deposition patterns of 10 selected elements as depicted by the concentrations in moss in 1989 and 1993 are shown in Figs. 5 - 9. For all elements the geographical distribution is strongly affected by the topographical features of the region, allowing polluted air to be transported

Table 1. Summary of results obtained for 38 elements in 25 moss samples from the vicinity of the Mo i Rana iron smelter complex

Element	1993		1989	
	Mean (*ppm*)	Min- max (*ppm*)	Mean (*ppm*)	Min- max (*ppm*)
Na	310	127-732	294	93-635
Mg	2015	781-6520	1861	556-4230
Al	1186	293-4460	1244	243-3100
Cl	201	88-375	294	50-1110
K	3470	1640-4770	3845	1930-7160
Ca	1903	995-4830	2871	1450-6740
Sc	0.20	0.03-0.70	0.41	0.06-1.41
V	4.10	0.79-19.0	5.72	1.05-31.0
Cr	390	4.3-3900	11.7	0.5-50
Mn	325	61-917	384	89-1460
Fe	2440	300-8700	12280	700-72100
Co	0.68	0.07-3.0	0.61	0.06-2.2
Ni	2.84	<0.5-15.6	1.69	<0.5-6.96
Cu	8.23	3.5-20.3	12.3	2..9-29.0
Zn	88	16-542	99	31-397
As	0.37	0.06-1.56	0.62	0.06-2.20
Se	0.31	0.16-0.75	0.47	0.21-1.17
Br	6.36	3.5-11.4	6.94	3.6-12.2
Rb	12.5	3.3-26.4	17.2	6.7-46.2
Sr	8.9	<4-52	19.9	<4-50
Zr	3.0	<2-24	11.0	<2-46
Mo	0.16	<0.08-0.76	0.87	<0.08-3.85
Ag	0.057	<0.03-0.18	0.059	<0.03-0.16
Sb	0.166	<0.03-0.69	0.250	<0.05-0.76
I	1.82	<1.0-3.3	2.26	<1.0-4.3
Cs	0.28	0.05-0.89	0.37	<0.05-1.03
Ba	26.7	15-57	33.1	12-83
La	0.45	<0.10-1.38	0.69	<0.10-2.87
Sm	0.139	0.04-0.45	0.33	0.05-1.34
Eu	0.016	<0.01-0.13	0.031	<0.01-0.12
Tb	0.020	<0.005-0.096	0.019	<0.005-0.067
Yb	0.032	<0.010-0.120	0.069	<0.010-0.230
Hf	0.097	<0.04-0.36	0.179	<0.04-0.71
Ta	0.011	<0.003-0.038	0.043	<0.003-0.180
W	1.68	<0.6-9.4	1.71	<0.6-6.4
Au	0.0042	0.0006-0.037	0.0002	<0.0001-0.010
Th	0.114	0.02-0.47	0.267	0.04-1.10
U	0.035	0.03-0.22	0.143	<0.03-0.51

A brief discussion of each of these elements and other elements following the same trends is presented in the following:

Al: An obvious source of Al in air is mineral dust, which could originate from various processes within or outside the industrial area. The general concentration level in moss is similar in 1989 and 1993, but as shown in Fig. 5 the peak level is higher and the range of deposition is shorter in 1993, indicating a change to larger particles.

Sc: Like Al this element is a typical crustal component, and the geographical distribution patterns (Fig. 5) is similar, but the deposition is reduced by about a factor of 3 in 1993 relative to 1989. Similar temporal and spatial trends (not shown) are evident for other typical crustal elements such as lanthanoides, Ta, Th, and U.

V: The deposition patterns of V in 1989 and 1993 (Fig. 6) are very similar, but the level has declined by a factor of 2. Trends for Mo (not shown) are similar to those of V.

Cr: As is evident from Fig. 7 the chromium deposition shows an increase in 1993 by about a factor of 50, obviously associated with the new ferrochrome plant. A closer examination of the data indicates a slight shift of the main emission source to the south. A discussion of possible impacts on the local population or the surrounding environment of this substantial increase in Cr emission is beyond the scope of this paper.

Fe: The Fe deposition (Fig. 6) in the Mo area declined by a factor of 10 between 1989 and 1993, presumably mainly because the iron smelter was closed.

Ni: As indicated in Fig. 7 the deposition has increased by about a factor of 2, and the main source has moved southwards, in a similar way as for Cr. The deposition patterns for Co and Ag (not shown) look very similar to that of Ni.

Cu: The general deposition level (Fig. 8) has not changed much, but the patterns look different. In 1989 there seems to be some source northwest to the centre of the town in addition to the industrial area, whereas in 1993 emissions seem to be concentrated to the latter.

Zn: Relative small changes are evident, but the emissions appear to be more focused to the smelter area in 1993 (Fig. 9).

As: For this element (Fig. 8) a wider area of enhanced deposition presumably from more diffuse sources is apparent in 1989 than in 1993, where the emissions are more focused to the Mo industrial area.

Sb: Fig. 9 indicates that a source to the northeast of the industrial area seems to add to the emissions from the area itself. The deposition is more focused in 1993 than in 1989.

Results from factor analyses of the moss data from 1989 and 1993 respectively are shown in Table 2, and the distribution of the two most prominent factors in each case among the sampling sites are shown in Fig. 10. The 1989 moss factor analysis shows two distinct components. Factor 1 has high loadings for typical crustal elements (Al, Sc, Co, Sr, Sm, Yb, Hf, Ta, U) as well as for some elements often observed in pollution aerosols (Cr, Ni, Zn, Se, Sb) and shows the highest score at site 27 (cfr. Fig. 4) followed by sites 9 and 10. It may be considered as a general dust component related to activities mainly within the industrial area. Factor 2 on the other hand, with high loadings for Fe, V and As, is more concentrated to sites 9 and 10 and is probably specifically related to emissions from the iron smelter. Factors 3–5 appear to be more difficult to explain.

Table 2. Factor loadings from principal component analysis of moss data from 1993 and 1989. Only values >0.6 are listed

Элемент	Факторы 1989				Факторы 1993			
	1	2	3	4	1	2	3	4
Na				0.80		0.87		
Mg		0.63			0.78			
Al	0.81						0.60	
Cl			0.80					
K			0.82					0.67
Sc	0.89				0.66	0.67		
V		0.74			0.88			
Cr	0.88				0.95			
Mn	0.70						0.71	
Fe	0.62	0.71			0.72			
Co	0.93				0.88			
Ni	0.85				0.91			
Cu							0.79	
Zn	0.87				0.71			
As		0.75			0.80			
Se	0.85				0.61	0.63		
Br								
Rb								0.86
Sr	0.81							
Mo	0.74				0.77			
Sb	0.77					0.78		
I				0.86				
Cs								0.80
Sm	0.96					0.80		
Yb	0.90					0.85		
Hf	0.95					0.67		
Ta	0.90					0.80		
U	0.96				0.67	0.67		

In 1993, factor 1 with very high loadings for Cr, Ni, V, and Co is apparently associated with the ferrochrome smelter. The high scores at sites 8, 9, and 10 support this conclusion.

Factor 2 is more likely a dust factor, with highest scores on the sites north-east of the industrial area in 1993 similar to 1989.

A comparison of the levels in moss 1989 and 1993 (Table 1) shows that the deposition in Mo and the surrounding areas was distinctly higher in 1989 for Ca, Sc, Fe, Cu, As, Se, Sr, Zr, Mo, lanthanoides, Hf, Ta, Th, and U, which are mainly elements associated with operations related to the iron smelter and predominantly characteristic of the "dust" fraction. Elements showing distinctly higher deposition in 1993 are Cr, Ni, and Au. For the rest of the elements (Na, Mg, Cl, K, V, Mn, Co, Br, Rb, Ag, Sb, I, Cs, Ba, W) changes were small. This may mean that emissions from the original and the new industrial source were similar, or that the occurrence of these elements in the moss has no clear connection with activities in the Mo industrial area.

The concentrations of Cr, Fe, Cu, Zn, As, Cd, Sb, and Pb in natural surface soils (primary data listed in Steinnes et al., 1992) were generally higher in areas within or close to Mo than at locations farther away, indicating contamination from industry and other sources within the urban area.

The contamination was particularly pronounced for Fe, in which case the mean concentration near the industrial area (sites 9, 10, 26) was 51 times higher than the general background level in the region (sites 1, 5, 22) no doubt because of the long period of smelter operation. Similar ratios for other elements apparently associated with emissions from the iron smelter were as follows. As, 14; Cr, 5.0; Cu, 3.2; Zn, 3.2.

In the case of Sb, Cd, and Pb the data indicated a more general urban pollution pattern with no dominant contribution from any single point source, and these elements were therefore not included in the subsequent AAS analyses of moss samples.

Factor analysis of the soil data shows two distinct factors with high loadings for respectively Cr, Fe, Cu, Zn, As and Pb, Sb, Cd (Table 3).

Table 3. Factor loadings from principal component analysis of surface soil data

Элемент	Факторы	
	1	2
As	0.94	-0.03
Cd	0.02	0.54
Cu	0.88	0.38
Cr	0.90	0.14
Fe	0.96	0.06
Pb	0.46	0.80
Sb	0.06	0.83
Zn	0.78	0.50

In Fig. 11 concentrations of Cr, Fe, Cu, and Zn are plotted against corresponding values for moss from the same sites. Very strong correlations are evident, further supporting the assumption that the surface soil near Mo is contaminated with these elements from atmospheric deposition.

In the case of Hg, only 10 soil samples were analysed, showing concentrations of 0.07-0.12 ppm, which is within the normal range for the humic horizon in the region concerned (Steinnes and Andersson, 1991).

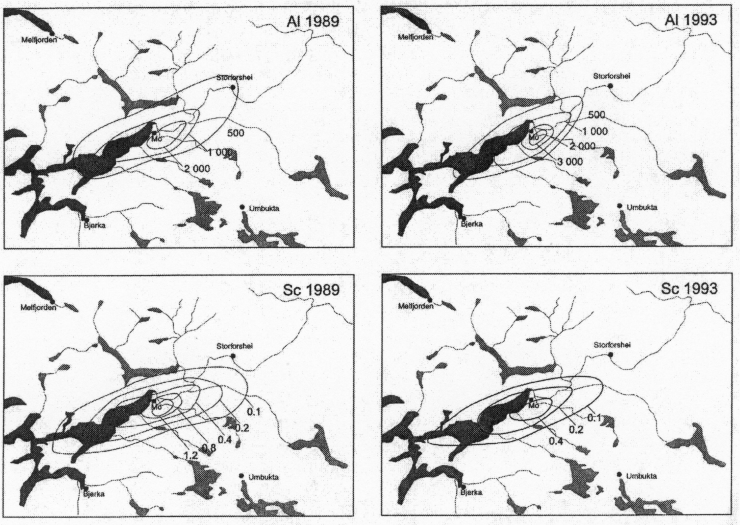

Fig. 5. Relative atmospheric deposition patterns of Al and Sc in he region surrounding the Mo industries as illustrated by analysis of moss samples collected in 1989 and 1993

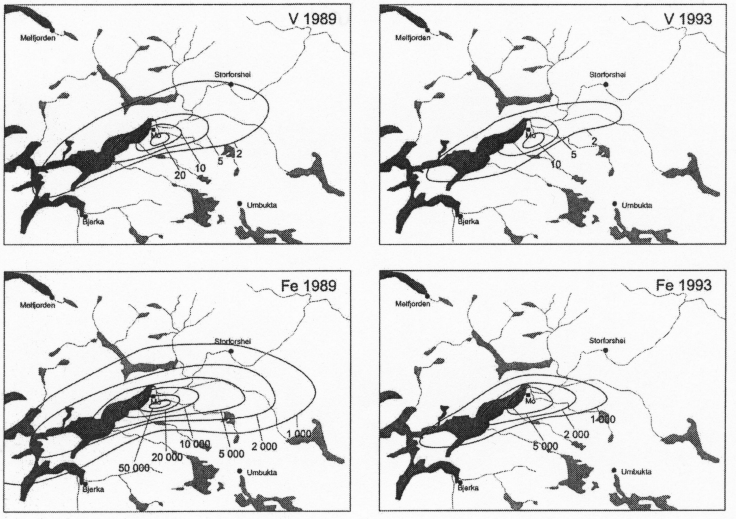

Fig. 6. Relative atmospheric deposition patterns of V and Fe in the region surrounding the Mo industries as illustrated by analysis of moss samples collected in 1989 and 1993

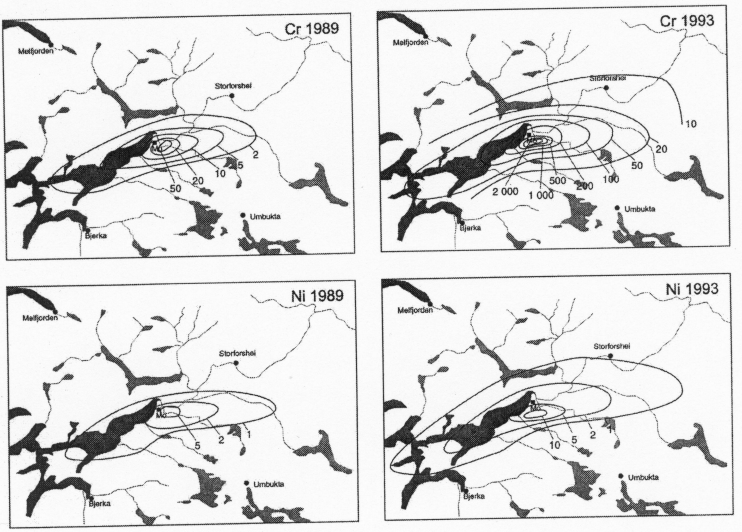

Fig. 7. Relative atmospheric deposition patterns of Cr and Ni in the region surrounding the Mo industries as illustrated by analysis of moss samples collected in 1989 and 1993.

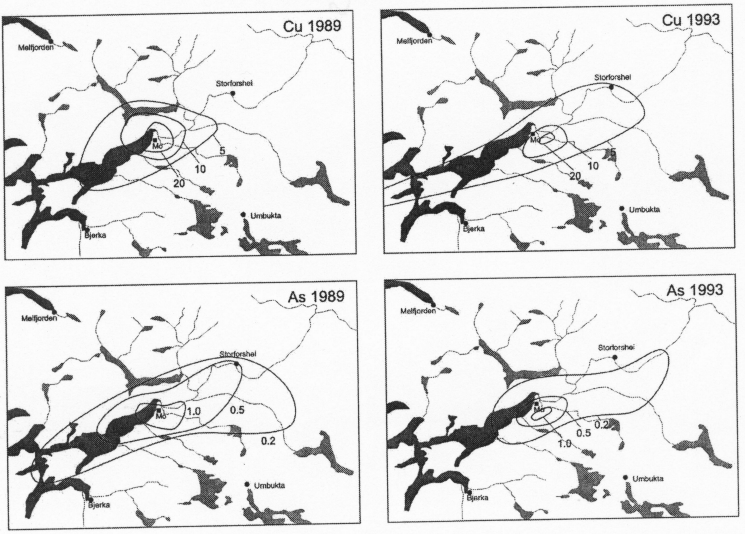

Fig. 8. Relative atmospheric deposition patterns of Cu and As in the region surrounding the Mo industries as illustrated by analysis of moss samples collected in 1989 and 1993

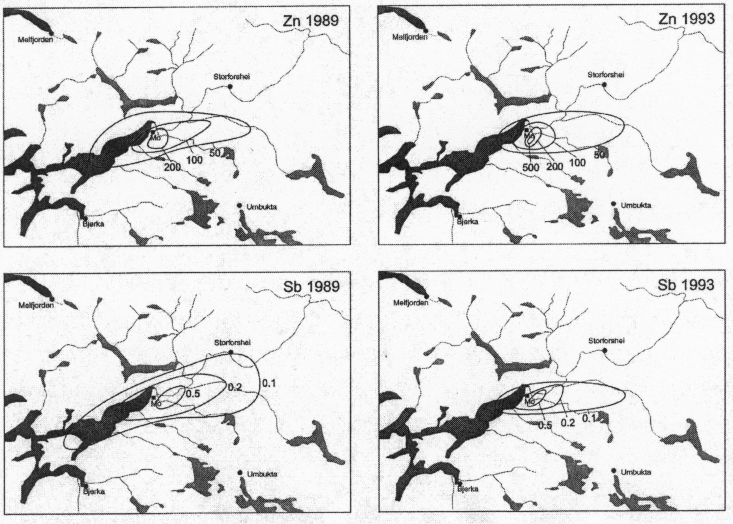

Fig. 9. Relative atmospheric deposition patterns of Zn and Sb in the region surrounding the Mo industries as illustrated by analysis of moss samples collected in 1989 and 1993

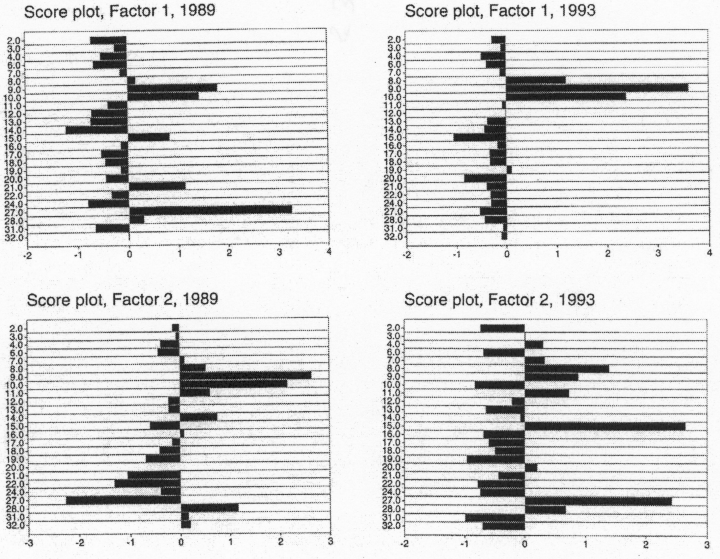

Fig. 10. Score plot for factors 1 and 2 from the factor analyses of 1989 and 1993 moss samples. Numbers of sampling sites are shown along the y-axes

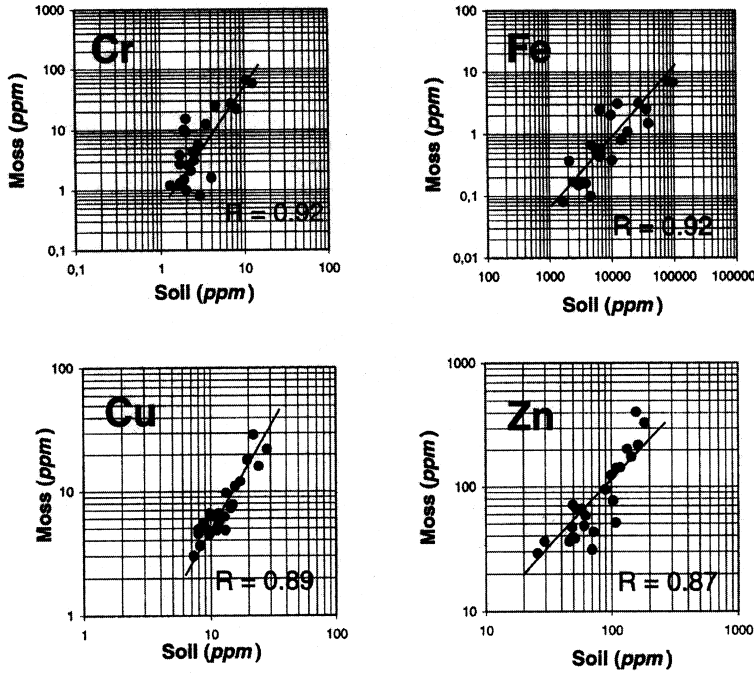

Fig. 11. Concentrations of Cr, Fe, Cu, and Zn in surface soil plotted against 1989 moss data from the same sites

CONCLUSIONS

The present work is a good example of the use of the moss biomonitoring technique to register the atmospheric deposition pattern as affected by local industry, and how it can be advantageously employed to register changes in pollution load following a re-structuring of the industrial activity. Whereas the deposition at Mo i Rana generally declined, the new processes introduced caused a substantial increase in Cr deposition and also some increase for Ni. Instrumental neutron activation analysis appeared to be a convenient analytical approach for this kind of study. By applying a dense sampling network and multivariate statistical treatment of the analytical data the main sources of air pollution were well characterized.

The experience from the present work indicates that the moss technique could be advantageously used to map the extent of heavy metal pollution also around many industrial complexes in Russia, in particular iron smelters. This was already indicated by some preliminary studies employing the moss technique and ENAA in the vicinity of the Magnitogorsk steel plant in South Ural (Frontasyeva et al., 2000).

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Изучение атмосферных выпадений тяжелых металлов в районе сталелитейного комплекса в северной Норвегии при различной ориентации производства

Для изучения атмосферных выпадений тяжелых металлов в окрестностях города Му-и-Рана в северной Норвегии до и после закрытия сталелитейного производства в 1989 г. и перехода на альтернативное ферросплавное производство применялся метод биомониторирования с помощью мхов-биомониторов. Образцы мха *Hylocomium splendens* собирались в одних и тех же точках пробоотбора в 1989 и 1993 г. Сочетание инструментального нейтронного активационного анализа (ИНАА) и метода атомной абсорбционной спектрометрии (ААС) позволило определить 38 элементов в собранных образцах мха. Для интерпретации аналитических результатов применялся факторный анализ. Выпадения Fe и других элементов, обычно ассоциируемых с земной корой, были выше в период работы завода в режиме сталелитейного производства. Существенно увеличилось выпадение Cr в связи с переориентацией завода на производство феррохрома. Также наблюдалось повышенное выпадение Ni и Au, в то время как выпадение Mn, Co, Ag, Sb и W осталось на прежнем уровне. Региональное распределение элементов-загрязнителей существенно зависело от топографии местности. Результаты анализа образцов природных поверхностных почв, собранных одновременно с первой серией мхов, явно свидетельствуют об их загрязнении многими металлами из атмосферных выпадений. ИНАА оказался мощным методом для подобного исследования. Описанный подход может с успехом применяться для изучения атмосферных выпадений тяжелых металлов вблизи сталелитейных заводов в России и других странах.

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Metal Pollution Around an Iron Smelter Complex in Northern Norway at Different Modes of Operation

The moss biomonitoring technique was employed to study the atmospheric deposition in and around the town of Mo i Rana, northern Norway, before and after closing an iron smelter and establishing alternative ferrous metal industries. Samples of *Hylocomium splendens* were collected from the same sites in 1989 and 1993. A combination of instrumental neutron activation analysis (INAA) and atomic absorption spectrometry was used to obtain data for 38 elements in these moss samples, and the analytical data were subjected to factor analysis. In general, the deposition was higher when the iron smelter was still in operation, in particular for Fe and for many elements normally associated with crustal matter. For Cr there was a substantially increased deposition due to the operation of a new ferrochrome smelter. Also for Ni and Au an increased deposition was observed, whereas for metals such as Mn, Co, Ag, Sb, and W there was no appreciable change. INAA proved to be a powerful tool for this kind of study. The regional distribution of pollutants was strongly dependent on the local topography. Samples of natural surface soils collected simultaneously with the first moss series showed clear signs of contamination with a number of metals from atmospheric deposition. The approach described in this work could be advantageously used to study atmospheric deposition of heavy metals around iron smelters in Russia and elsewhere.

The investigation has been performed at the Frank Laboratory of Neutron Physics, JINR.

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