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K. Pilegaard

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See page 3 of cover.

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Contents

Deposition of airborne metals around the lead-zinc mine in Maarmorilik monitored by lichens and mosses

K. PILEGAARD

Pilegaard, K. 1994. Deposition of airborne metals around the lead-zinc mine in Maarmorilik monitored by lichens and mosses. – Meddr Grønland, Biosci. 43, 20 pp. Copenhagen 1994-11-30.

The deposition of heavy metals around the Pb-Zn mine in Maarmorilik (Greenland) was monitored during the years 1979–1990 by analyses of concentrations in *in situ* lichens (*Cetraria nivalis* and *Umbilicaria lyngei*) and higher plants (*Rhododendron lapponicum*).

Concentrations of the metals Ag, As, Cd, Cu, Hg, Pb, Sb and Zn decreased with increasing distance from the mining and milling complex according to the model: $y = ax^b + c$ where y = concentration, x = distance, c = background concentration, a and b = constants.

The spread was most pronounced west of Maarmorilik. Exposed Pb-Zn mineralizations in the area were found not to influence the overall deposition pattern. The transport of pollutants to higher altitudes was little.

Airborne pollution with Cd, Pb and Zn was monitored with suspended *Sphagnum*-bags during a period with ship-loading of concentrates and compared to a period without this activity. There was a strongly increased deposition of airborne Cd, Pb and Zn during periods of ship-loading. The primary sources of pollution were the concentrate conveyor and the ship-loader. *Sphagnum*-bags were also used to monitor the effects of remedial actions carried out in the mining town.

Analyses of the concentrations of Pb in *Cetraria* during the years 1979 to 1990 showed that pollution during the last years was only about half as large as during the early years.

This decrease is attributed to the pollution abatement carried out at the mining complex.

Key words:
Heavy metal deposition, Greenland, mining, pollution.

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Introduction

The Black Angel mine in Maarmorilik is situated in West Greenland at 71°8'N, 51°15'W in an area without any other pollution sources. The mine was in operation from 1974 to 1990 and had a yearly production of approximately 40,000 t Pb-concentrate and 150,000 t Zn-concentrate (Table 1). The concentrates were shipped from Maarmorilik from June to December when the fjords were free from ice. The concentrates were placed in an enclosed storage until shipping, and ships were loaded with a 400 m long conveyor, which from 1982 was covered by a roof but not totally enclosed. The tailings from the production plant (crushing mill and flotation plant) were discharged directly to the sea (Asmund *et al.* 1991). Significant amounts of Cd, Cu, Pb and Zn have been released to the marine environment from the mining operation. This has resulted in increased metal concentrations in organisms such as seaweed, mussels, prawns and fish (Johansen *et al.* 1991). The lichen flora around Maarmorilik is rich in species restricted to calcareous rocks and alkaline substrates (Hansen 1991).

The present paper presents a series of investigations of airborne spread of metals from the mining and milling complex using biological monitors: *in situ* vegetation (shrubs, mosses and lichens) and transplants (moss bags). The first investigation was carried out in 1979. This was followed by more detailed investigations covering a larger area in 1980 and 1982. Moss bag surveys were performed in 1980 and 1982 in order to identify pollution sources and evaluate pollution abatement. Finally an evaluation of changes in atmospheric deposition up to the closure of the mine in 1990 is given based on results from

Table 1. Composition of ore, concentrates and tailings of the Black Angel Mine 1982–83 (Greenex A/S, pers. comm. 1984).

Element	Ore	Zn-conc.	Pb-conc.	Tailings	Unit
Amount	675,000	130,596	34,188	510,208	ton
Zn	11.8	57.5	6.2	0.47	%
Pb	4.0	1.6	70.6	0.22	%
Fe	13.3	5.4	2.1	16.0	%
Cd	623	3043	340	32.4	$\mu g\ g^{-1}$
Cu	293	889	2243	126	$\mu g\ g^{-1}$
As	82.4	25.2	416	58.7	$\mu g\ g^{-1}$
Ag	30.9	25.1	439	4.8	$\mu g\ g^{-1}$
Hg	15.2	69.1	9.1	0.7	$\mu g\ g^{-1}$

a continuous monitoring programme of atmospheric deposition started in 1983 (Asmund *et al.* 1988).

Materials and methods

Details of sampling and materials are given in the sections covering the individual investigations. The following preparation and chemical analysis procedures were the same for all samples.

The samples were carefully cleaned for extraneous material and dried for 3 days at 60°C. The elements Ag, Cd, Cu, Fe, Pb and Zn were determined by atomic absorption spectroscopy (AAS). One gram of dry material was digested to total dissolution in a mixture of 10 ml 14 N HNO_3 (Merck, *pro analysi*) and 10 ml de-mineralized H_2O and evaporated to dryness. The residue was dissolved in 25 ml 1 N HNO_3. The solutions were filtered and stored in polyethylene bottles in a refrigerator. The concentrations of the metals were determined with a Perkin-Elmer model 5000 spectrophotometer with graphite furnace HGA-76, D_2-arc background compensation and automatic sampler AS-1.

The following elements were determined by instrumental neutron activation analysis (INAA): Na, K, Sc, Cr, Fe, Co, Zn, As, Se, Br, Rb, Sr, Ag, Sb, Ba, Cs, La, Ce, Sm, Eu, Yb, Hf, Au, Hg, Th and U. Dried samples (0.2–0.4 g) were placed in a small plastic vessel. The samples were irradiated in the 10 MW heavy-metal moderated reactor DR3 at Risø National Laboratory. The irradiation times were 1–12 h, the decay times 3–7 d and 25–30 d with counting times 1–4 h. Two separate measurements were made in order to analyze as many elements as possible. The countings were made with a Ge(Li) detector. Detailed description of the applied INAA-method is given by Christensen & Damsgaard (1985).

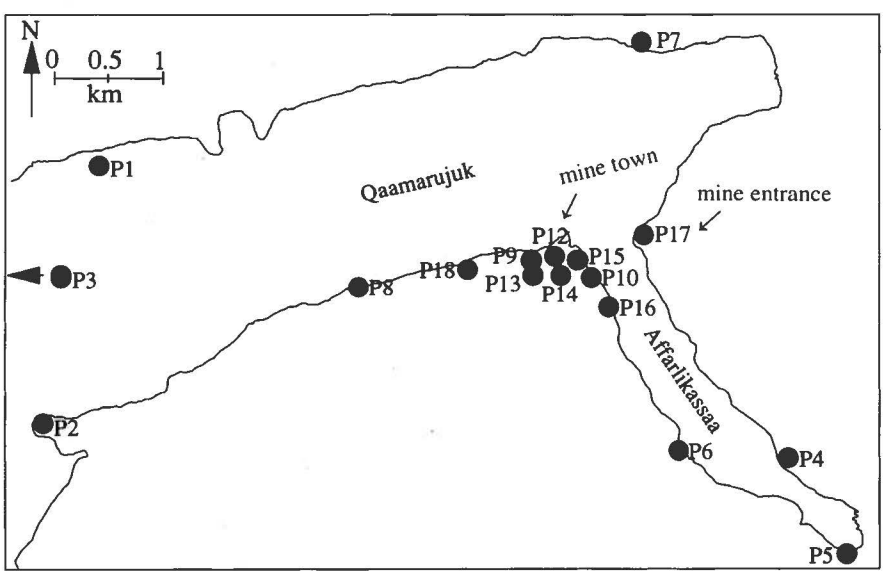

Fig. 1. Localizations of sampling sites in the area around Maarmorilik in the pilot investigation 1979.

Table 2. Concentrations of elements (μg g^{-1}) in *Cetraria nivalis* sampled in 1979.

Station	Direction*	Distance (m)*	Ag	Cd	Cu	Fe	Pb	Zn
P10	NW	53	1.09	12.7	13.7	420	5650	1580
P15	NNW	87	1.05	13.9	25.2	416	4470	1630
P14	WNW	280	0.70	7.5	13.5	244	1890	853
P13	WNW	510	0.37	3.7	7.9	317	786	474
P17	NE	630	0.187	2.9	3.2	182	511	261
P18	W	1350	0.22	4.0	6.9	183	637	385
P 6	SSE	2050	0.134	1.37	2.1	179	245	166
P 8	W	2120	0.141	1.33	2.9	103	228	228
P 7	N	2500	0.29	0.93	2.1	175	130	132
P 4	SE	2700	0.062	0.90	1.6	141	89	114
P 5	SE	3760	0.100	1.13	1.8	106	135	124
P 1	W	5120	0.107	1.31	1.0	247	144	233
P 2	WSW	5200	0.079	1.09	2.1	215	119	119
P 3	WSW	10240	0.053	0.79	1.8	148	74	106
Qeqertanguit	W	11250	0.043	0.43	–	64	40	68
Uummanaq	SW	60000	0.013	0.11	5.5	242	26	42

* From mining town (arbitrary pollution centre).

Pilot investigation

Samples

Samples of plant species were collected in March 1979. The species were chosen according to their abundance throughout the area and their expected suitability as monitor organisms of airborne metal pollution. The species were the dwarf shrub *Rhododendron lapponicum* L. and the lichens *Cetraria nivalis* (L.) Ach. and *Umbilicaria lyngei* Schol.

Rhododendron lapponicum is a 10–20 cm high dwarf shrub with elliptical and hairy leaves. Top shoots were collected, and in the laboratory leaves were separated from the stems. Only green leaves were used for the analyses. Leaves from one year remain green through the next summer.

Cetraria nivalis is an epigeic fruticose lichen that grows in loose tufts. The thallus is erect, sparingly branched, 2–6 cm high, straw yellow with a strong yellow or yellowish-brownish coloured band towards the base of the living part. There is almost always a thick mat of dead material below the living parts. At each site 5–10 tufts were taken to form a composite sample. The material was, whenever possible, tufts growing on a mat of moss or at least on a mat of dead material of the lichen itself. This sampling procedure was followed in order to avoid contamination due to direct contact with the soil. In the laboratory, samples were carefully cleaned to remove all foreign material.

Umbilicaria lyngei is a foliose epilithic lichen with a smooth dark grey to black thallus with a diameter of 1–5 cm. The species was sampled from granitic blocks scattered all over the area. The lichens were separated from the substratum by cutting the umbilicus with a knife (stainless steel). The area around the umbilicus was removed before chemical analyses in order to avoid attached mineral particles.

Table 3. Concentrations of elements (μg g^{-1}) in *Umbilicaria lyngei* sampled in 1979.

Station	Direction	Distance (m)	Ag	Cd	Cu	Fe	Pb	Zn
P10	NW	53	0.90	8.0	21.9	473	2320	1310
P15	NNW	87	1.41	6.9	34.5	692	2470	1500
P14	WNW	280	0.88	8.8	36.0	640	1790	1174
P12	WNW	380	0.45	6.2	22.1	541	953	780
P13	WNW	510	0.28	1.95	21.3	590	746	533
P18	W	1350	0.25	4.3	15.3	362	606	563
P 6	SSE	2050	0.146	0.69	7.9	292	348	192
P 8	W	2120	0.158	1.09	8.5	297	370	279
P 7	N	2500	0.102	0.74	2.8	282	169	138
P 5	SE	3760	0.087	0.46	4.9	238	200	161
P 1	W	5120	0.056	0.77	3.6	268	148	133
P 2	WSW	5200	0.093	0.55	4.8	294	219	181
P 3	WSW	10240	0.052	0.36	3.4	331	106	106
Uummanaq	SW	60000	0.044	0.12	6.6	452	11	47

Table 4. Concentrations of elements (μg g^{-1}) in *Rhododendron lapponicum* sampled in 1979.

Station	Direction	Distance (m)	Ag	Cd	Cu	Fe	Pb	Zn
P10	NW	53	1.02	6.2	19.0	397	2080	1550
P15	NNW	87	0.41	5.0	12.9	253	764	804
P14	WNW	280	0.20	5.2	12.0	229	374	585
P13	WNW	510	0.15	1.57	12.1	288	249	316
P 9	WNW	530	0.23	3.1	11.0	209	355	496
P17	NE	630	0.069	0.74	8.5	194	101	201
P18	W	1350	0.086	0.86	9.1	91	151	216
P 6	SSE	2050	1.11	0.36	8.6	107	31	78
P 8	W	2120	0.026	0.32	7.9	71	42	87
P 7	N	2500	0.018	0.26	5.8	89	18	73
P 4	SE	2700	0.026	0.36	8.3	127	10	73
P 5	SE	3760	0.40	0.18	8.7	79	5	81
P 1	W	5120	0.032	0.34	7.1	79	24	81
P 2	WSW	5200	0.052	0.29	9.4	94	16	68
P 3	WSW	10240	0.565	0.18	7.3	86	11	50
Uummanaq	SW	60000	–	0.03	–	101	0.3	20

Sampling sites

Samples were collected from 17 sites within 12 km from Maarmorilik and at a reference site (Ummanaq) 60 km away (Fig. 1). The sampling sites were all close to sea level. This was due to the inaccessibility of the mountain area in the winter time. The way of transportation was by car on the frozen fjords.

Table 5. Regression equations ($y=ax^b$) of element concentrations (y) vs. distance (x) (1979).
(* p<0.05, ** p<0.01, *** p<0.001, n.s. = non significant).

Regression equation			n	% var	S_b	signif. of slope ≠ 0
[Ag]	Cetraria	$= 14.94x^{-0.61}$	15	89.0	0.06	***
[Ag]	Umbilicaria	$= 19.86x^{-0.65}$	13	93.2	0.05	***
[Ag]	Rhododendron	$= 1.72x^{-0.36}$	15	16.1	0.23	n.s.
[Cd]	Cetraria	$= 190x^{-0.67}$	15	91.4	0.05	***
[Cd]	Umbilicaria	$= 183x^{-0.67}$	13	82.2	0.09	***
[Cd]	Rhododendron	$= 198x^{-0.79}$	15	87.2	0.08	***
[Cu]	Cetraria	$= 215x^{-0.57}$	14	82.3	0.08	***
[Cu]	Umbilicaria	$= 326x^{-0.50}$	13	78.0	0.08	***
[Cu]	Rhododendron	$= 30x^{-0.16}$	15	72.7	0.03	***
[Fe]	Cetraria	$= 887x^{-0.21}$	15	55.9	0.05	**
[Fe]	Umbilicaria	$= 1423x^{-0.19}$	13	68.6	0.04	***
[Fe]	Rhododendron	$= 1420x^{-0.33}$	15	80.7	0.05	***
[Pb]	Cetraria	$= 209149x^{-0.89}$	15	94.4	0.06	***
[Pb]	Umbilicaria	$= 419x^{-0.64}$	13	93.8	0.05	***
[Pb]	Rhododendron	$= 165479x^{-1.10}$	15	87.4	0.12	***
[Zn]	Cetraria	$= 17492x^{-0.58}$	15	90.0	0.05	***
[Zn]	Umbilicaria	$= 17848x^{-0.56}$	13	90.1	0.06	***
[Zn]	Rhododendron	$= 21661x^{-0.68}$	15	93.0	0.05	***

Results and discussion

The samples were analyzed for the elements Ag, Cd, Cu, Fe, Pb and Zn (Tables 2–4). The distance from Maarmorilik was calculated from an arbitrary pollution centre, because of the complexity of the distribution of pollution sources. The centre was chosen as the middle point between stations 10 and 15 which showed the highest concentrations of Pb. There was a steep decrease in concentrations in all monitor organisms with increasing distance from this point. Even at the most distant of the sampling stations (10,240 m) around Maarmorilik the concentrations of several elements were still far above the reference station at Ummanaq 60 km away.

Regression lines of the concentration on distance were calculated (Table 5). Logarithmic transformations of both distance and concentration gave the highest correlation coefficients.

The pilot investigation showed that the concentrations of Ag, Cd, Cu, Fe, Pb and Zn decreased with increasing distance from the mining town. The highest concentrations were found near the conveyor belt from the concentrate storage to the ship-loader, which indicated that upwhirling of dust by transport and loading of the concentrates was the major source of airborne metal pollution in the area. The pilot investigation also showed that the greatest spread was found in the direction westward of Maarmorilik.

Extensive investigations

Aims

Based on the findings in the pilot investigation the following questions arose:

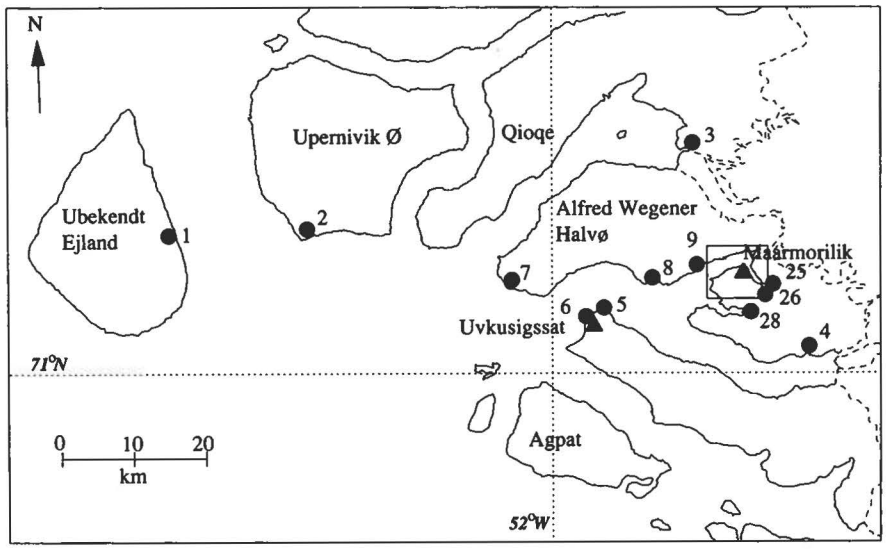

Fig. 2. Localizations of sampling sites in the area around Maarmorilik. The outlined area is shown in Fig. 3.

1) How far from Maarmorilik can the elevated deposition of metals be traced?
2) What is the infiuence of exposed mineralizations on the general level of metals of the lichens in the area?
3) How is the distribution of pollutants affected by the topography of the area?

In order to answer question 1 material of *Cetraria nivalis* was sampled along a transect from Maarmorilik and westwards to Ubekendt Ejland, 80 km from Maarmorilik.

To illustrate the influence of an exposed mineralization on the metal content of lichens (question 2) samples of *Cetraria nivalis* and *Umbilicaria lyngei* were taken close to a pyrite vein with a Pb-Zn mineralization, 13 km ESE of Maarmorilik.

The influence of the topography (question 3) was illustrated by a set of samples of *Cetraria* from sea level to

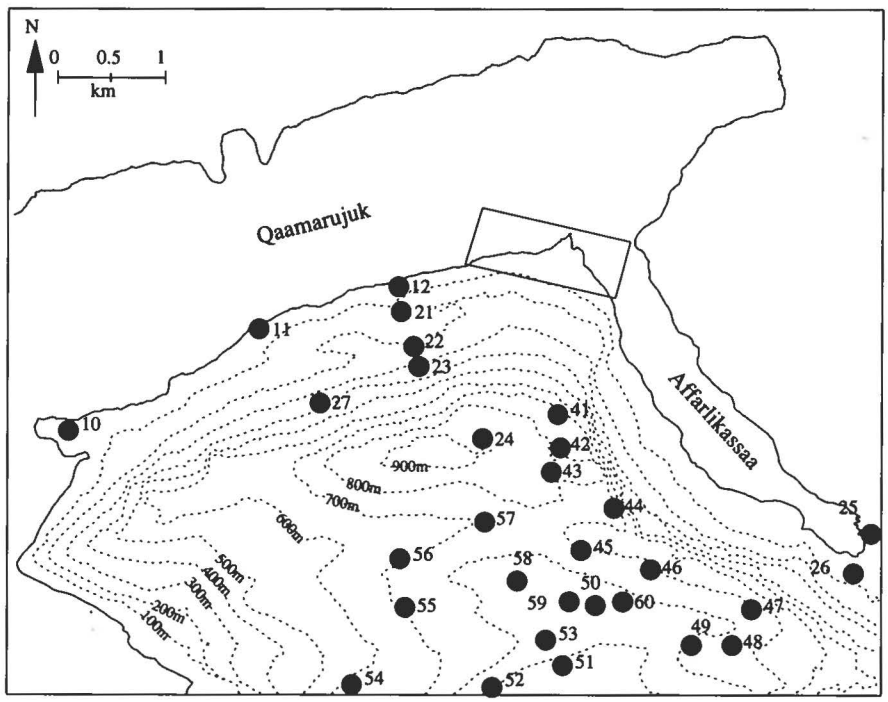

Fig. 3. Localizations of sampling sites in the area around Maarmorilik. The outlined area is shown in Fig. 4.

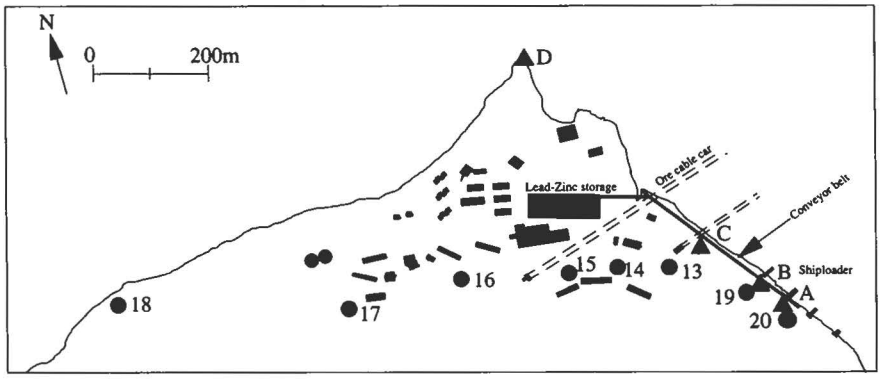

Fig. 4. Localizations of sampling sites in the mining town.

900 m a.s.l. at a distance of 1.8 km westward of Maarmorilik.

In addition to the above mentioned sample series material was collected from a number of other localities in 1980. In 1982 samples were collected on the Maarmorilik plateau.

Samples

Transect westwards

Cetraria nivalis was collected from 15 stations (nos. 1, 2, 5, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, and 18 (Figs. 2–4)) from Maarmorilik and westwards: the stations were chosen in such a way that the natural logarithm to the distance from the concentrate conveyor in the mining town increased equidistantly, according to the findings in the pilot-investigation. The station closest to the conveyor was at a distance of 85 m and the most distant station at 80,000 m. At each station several tufts of the lichen were collected and mixed to a composite sample.

Transect uphill

Cetraria nivalis was collected at a distance of 1,800 m from Maarmorilik at different altitudes: 100 m a.s.l., 200 m, 300 m, and 900 m (stations nos. 21, 22, 23, and 24 (Fig. 3). At each station several tufts were sampled and mixed to a composite sample. The absence of samples between 300 m and 900 m a.s.l. is due to the inaccessibility of the mountain at these altitudes.

Around a Pb-Zn mineralization

Cetraria nivalis and *Umbilicaria lyngei* were sampled along a transect crossing a pyrite vein with Pb-Zn mineralization. The thickness of this vein is approximately 1 m. It is exposed over a length of 250 m in an area with dolomite marble as bed-rock 13 km ESE of Maarmorilik.

Samples of the two lichen species were taken at 9 stations (nos. 29–37) at distances up to 400 m perpendicular to the vein in the directions north and south. The samples of *Cetraria* consisted of tufts growing on moss

Table 6. Concentratons of metals (μg g^{-1}) in *Cetraria nivalis* sampled along a transect westward of Maarmorilik (1980).

Station	Distance m	Ag *n* = 5	As *n* = 2	Cd *n* = 5	Cu *n* = 5	Hg *n* = 2	Pb *n* = 5	Sb *n* = 2	Zn *n* = 5
13	85	1.11	1.02	12.79	10.6	0.26	4280	0.99	1810
14	190	0.76	0.91	8.52	10.1	0.15	3110	0.78	1150
15	265	0.55	0.85	7.13	6.6	0.13	2370	0.45	700
16	460	0.38	0.80	4.38	5.5	0.12	1400	0.56	470
17	660	0.38	0.49	3.08	4.4	0.07	1070	0.28	310
18	1,045	0.33	0.54	3.51	4.3	0.09	990	0.36	350
12	1,800	0.26	0.25	1.75	2.3	0.08	410	0.23	240
11	2,960	0.15	0.24	1.22	1.7	0.07	170	0.22	150
10	5,120	0.11	0.23	0.80	1.4	0.07	101		95
9	8,250	0.21	0.21	0.72	2.3	0.08	89	0.11	83
8	13,750	0.12	0.18	0.70	1.9		81		80
5	21,000	0.07	0.17	0.19	1.6		9		25
7	33,000	0.06	0.25	0.24	1.5		15		35
2	60,500	0.10	0.20	0.21	2.2		3		17
1	80,000	0.07	0.07	0.08	1.5		5		12

Table 7. Regression equations ($y=ax^b+c$) of element concentration (μg g^{-1}) (y) in *Cetraria nivalis* as a function of distance (x) from Maarmorilik and background concentration (c). (* $p<0.05$, ** $p<0.01$, *** $p<0.001$, n.s. = non significant).

Regression equation		n	% var	S_b	signif. of slope ≠ 0
[Ag] =	$14.74x^{-0.600} + 0.06$	60	94.4	0.04	***
[As] =	$23.25x^{-0.632} + 0.16$	25	79.3	0.09	***
[Cd] =	$550.9x^{-0.808} + 0.09$	71	95.7	0.05	***
[Cu] =	$196.7x^{-0.678} + 1.5$	65	81.7	0.09	***
[Hg] =	$1.91x^{-0.599} + 0.06$	16	78.4	0.11	***
[Pb] =	$1943280x^{-1.182} + 2.0$	75	94.1	0.08	***
[Sb] =	$23.20x^{-0.687} + 0.08$	20	86.5	0.09	***
[Zn] =	$85263x^{-0.831} + 14.4$	69	94.0	0.06	***

or humus on the bed-rock, whereas the samples of *Umbilicaria* consisted of thalli growing horizontally on loose blocks (1–2 m in diameter) of granodiorite, which were scattered over the area.

Supplementary samples

In 1980 *Cetraria* was collected at 9 additional stations in the area (nos. 3, 4, 6, 19, 20, 25, 26, 27 and 28 (Figs. 2–4). In 1982 20 samples of *Cetraria* were collected on the Maarmorilik plateau (nos. 41–60) (Fig. 3).

Analyses

All samples were analyzed for Cd, Cu, Pb and Zn by atomic absorption spectrophotometry, some samples were also analyzed for Ag and Fe. Samples from the westward transect were also analyzed for the elements Na, K, Sc, Cr, Fe, Co, Cu, Zn, As, Se, Br, Rb, Ag, Cd, Sb, Cs, Ba, La, Ce, Sm, Eu, Yb, Hf, Au, Hg, Pb and Th by instrumental neutron activation analysis. All elements were analyzed in duplicates. Some elements (Ag, Cd, Cu, Fe, Pb and Zn) in samples from the westward transect were analyzed in 5 replicates.

Table 8. Average concentration (μg g^{-1}) of elements in *Cetraria nivalis* showing no relation to distance from Maarmorilik.

Element	mean	s.e.	n
Sc	0.106	0.002	30
Cr	4.17	0.08	30
Fe	211	3.5	30
Rb	3.95	0.05	20
Sr	14.3	1.7	4
Cs	0.081	0.001	27
Ba	15.3	0.4	12
La	1.17	0.025	30
Ce	2.51	0.05	30
Sm	0.153	0.002	30
Eu	0.0227	0.0003	30
Tb	0.0170	0.0012	3
Yb	0.030	0.001	12
Lu	0.0061	0.0004	5
Hf	0.041	0.002	18
Au	0.0021	0.001	24
Th	0.173	0.004	30
U	1.11	0.14	7

Results

Transect westwards

The mean concentrations of elements in *Cetrania* sampled at 15 stations with increasing distance in the westward direction from Maarmorilik are shown in Table 6. The standard devlation was proportional to the concentration, whereas the coefficient of variation was rather constant This indicated that a logarithmic transformation would stabilie the variance, which was confirmed by Bartletts test. This test showed that, after transformation of the data, variance homogeneity was found for most elements.

For some elements heterogeneity still existed This was, however, due to high coefficients of variation at very low concentration levels close to the limts of detection by the analytical method applied. Therefore, the tranormation of data to natura logarithm of the original values has been applied in the followlng statistical analyses.

In order to describe the distance relationship further an analysis of variance and a regression analysis of element concentration versus distance was performed according to the model:

$$y = ax^b + c$$

or

$$\log(y - c) = \log(a) + b \log(x)$$

where y = concentration in lichen, c = background concentration in lichen, and x = distance from Maarmorilik.

Negative values after subtraction of background values were dismissed from the data sets before calculation of the regressions.

In the analysis of variance the mean square of deviations from regression was used as the denominator mean square against which the significance of linear regression was tested. This was done because most of the residual mean square was due to the analytical variance and not the biological variance since samples were composite. Linear regressions were found highly significant for Ag, As, Cd, Cu, Hg, Pb, Sb and Zn (Table 7, Fig. 5). Average concentrations for the other elements are given in Table 8.

The amount of the total variance accounted for by regression on distance was very high (78%–96%). The slopes of the lines were different for the different elements with Pb showing the steepest slope followed by Zn, Cd, Sb, Cu, As, Ag, Hg. A statistical analysis showed that the difference between the slopes of Cd and Zn were not significant, whereas the slopes of both Ag and Pb differed significantly from the Cd-Zn value.

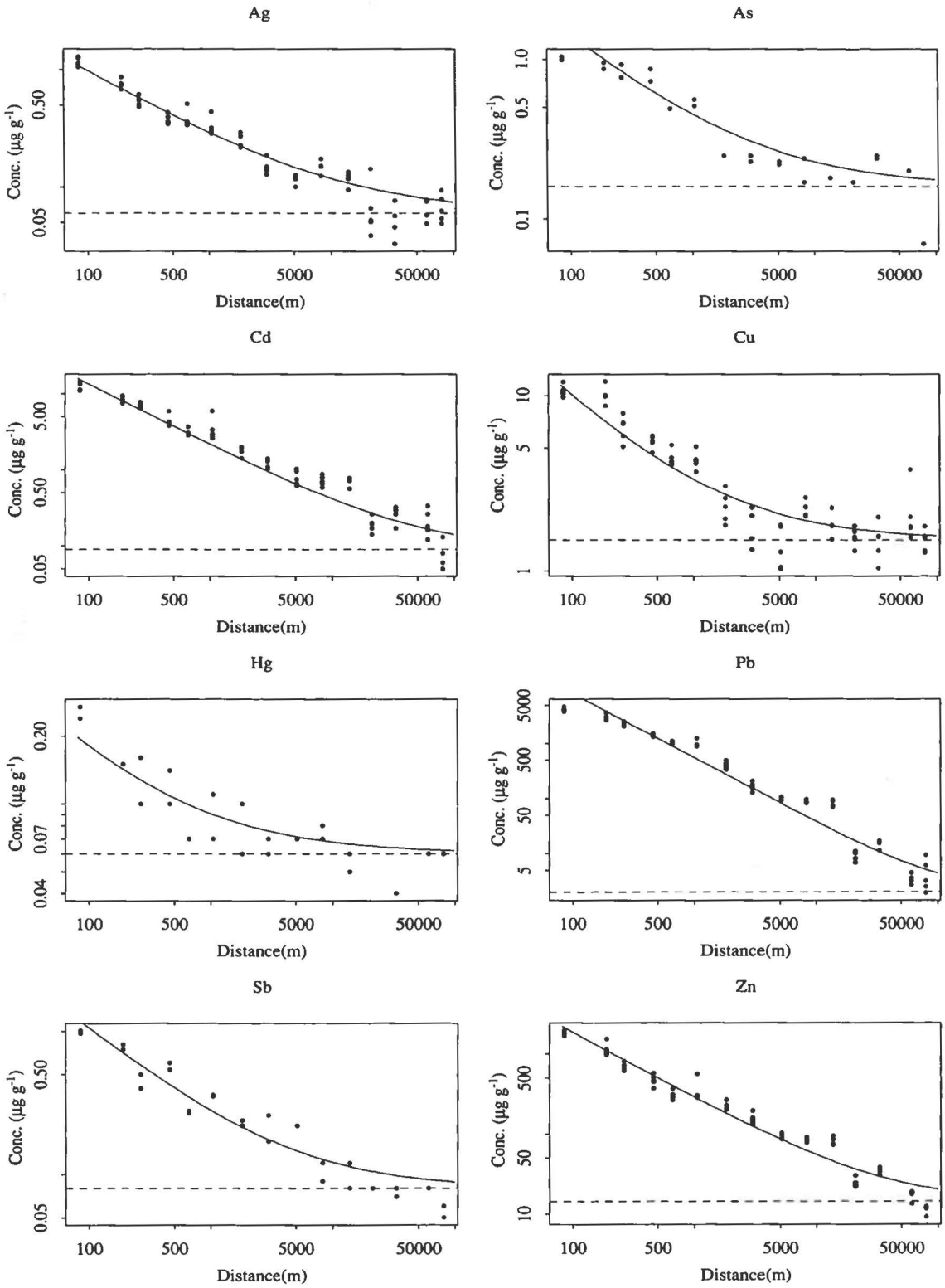

Fig. 5. Regression lines of metal concentrations in *Cetraria nivalis* versus distance in the westerly direction from Maarmorilik. Dashed lines represent background concentrations.

Table 9. Varimax rotation of 4-factor solution for *Cetraria nivalis*.

	Commu-nality	Factor 1	Factor 2	Factor 3	Factor 4	s.d.
Na	0.9421	–0.850	0.211		0.408	0.073
Cr	0.7468	–0.656	0.491			0.152
Cu	0.9177	0.942				0.087
Zn	0.9785	0.933	–0.216	0.247		0.044
As	0.9166	0.899		0.325		0.087
Se	0.8090	0.831				0.132
Ag	0.6619	0.641				0.175
Cd	0.9888	0.926	–0.214	0.283		0.032
Sb	0.9714	0.916	–0.281	0.167	0.163	0.051
Ba	0.7215	0.777				0.159
Hg	0.9498	0.961				0.068
Pb	0.9811	0.902	–0.260	0.281	–0.143	0.041
K	0.7707		0.719			0.144
Sc	0.9141		0.862	0.282		0.088
Fe	0.8200		0.794			0.128
Co	0.8376	–0.550	0.700			0.121
La	0.8863		0.758	0.544		0.102
Ce	0.9422		0.777	0.580		0.073
Sm	0.9563		0.928	0.291		0.063
Eu	0.9038		0.932			0.094
Yb	0.7051		0.754			0.164
Hf	0.9124	–0.461	0.693	0.359	0.301	0.089
Br	0.7090				0.644	0.163
Au	0.2065					0.269
Rb	0.9225			0.922		0.084
Cs	0.7412			0.780		0.153
Th	0.9731		0.493	0.729		0.049
27	22.7859	10.132	7.319	3.593		Variances
100	84.39	37.5	27.1	13.3		Percentages

The method of factor analysis was applied in order to reduce the amount of data variables. The factor analysis method applied in this study is described by Heidam (1981 and 1982). By means of a correlation matrix a large set of intercorrelated variables are reduced to a smaller set of uncorrelated variables, the factors. The factors can be interpreted as groups of element with different origin, and the loading of each factor on each station (the factor score) can be calculated. The factor analysis relies on the existence of a correlation matrix of the variables. In case of missing values the program is able to generate new values by an iterative process using the factor scores. Only elements determined in more than 70% of the samples were included. The number of factors to be retained in the model is determined by the eigenvalues; *i.e.* factors that contribute a variance < 1 are excluded from the model. After construction the principal factor model is rotated to fulfill the varimax criterion of either maximum or minimum loadings of the factor.

The applied program calculates factor loadings, communalities, absolute and relative factor variance contributions, standard deviations of factor loadings and factor scores. The factor loadings are simply the correlation coefficients between the original variables and the factors. The communality is the fraction of variance explained by the model. Twenty seven elements were determined in more than 70% of the samples: Na, K, Sc, Cr, Fe, Co, Cu, Zn, As, Se, Br, Rb, Ag, Cd, Sb, Cs, Ba, La, Ce, Sm, Eu, Yb, Hf, Au, Hg, Pb and Th. Correlation matrices of the logarithmically transformed concentrations were calculated. The matrices were diagonalized and analyses of eigenvalues for significance performed by means of a test statistic which measures individual eigenvalues in terms of the residual variance. The first 10 eigenvalues showed that the number of factors to be retained was 4.

The rotated 4-factor solution is given in Table 9. Factor loadings less than three standard deviations have been deleted for the sake of clearness. The 4-factor solution explained 84% of the total variation. The first factor (accounting for 38% of the total variation) included elements originating from the mining activity. Prominent members (loadings > 0.7) of this factor were: Cu, Zn, As, Se, Cd, Sb, Ba, Hg and Pb. Na and Cr were strongly negatively correlated with this factor. Factor 2 (accounting for 27% of the total variation) had the prominent members: K, Sc, Fe, Co, rare earth elements, Yb and Hf. These elements are probably of crustal origin. Factor 3 (accounting for 13% of the total variation) had the prominent members Rb, Cs and Th. Factor 4 (accounting for 7% of the total variation) had Br as the dominating element. No clear sources can be identified for the last two factors. Factor 1 (the mining factor) shows a clear decrease with distance from Maarmorilik, while the other factors show no relation to distance (Fig. 6).

Transect uphill

The metal concentrations in *Cetraria* at increasing altitudes at a distance 1800 m from Maarmorilik are shown in Table 10. All the metals (except Fe) were found in significantly lower concentrations at an altitude of 900 m a.s.l. than close to the sea. The steepest gradient was found for Pb followed by Cd, Zn, Ag and Cu, which is in accordance with the order of the slopes of the regression lines in the westward transect.

The concentrations found at 900 m a.s.l. correspond to the concentrations found in the westward transect at the following distances: Ag 59,000 m, Cd 26,000 m, Cu 6,000 m, Pb 22,000 m and Zn 20,000 m.

Around a Pb-Zn mineralization

The results of the analyses of metal concentrations in *Cetraria* and *Umbilicaria* sampled along a transect perpendicular to a Pb-Zn bearing ore are shown in Table 11. The results are means of double analyses, and the coefficient of variation is shown in the table. The coefficient of variation was calculated as:

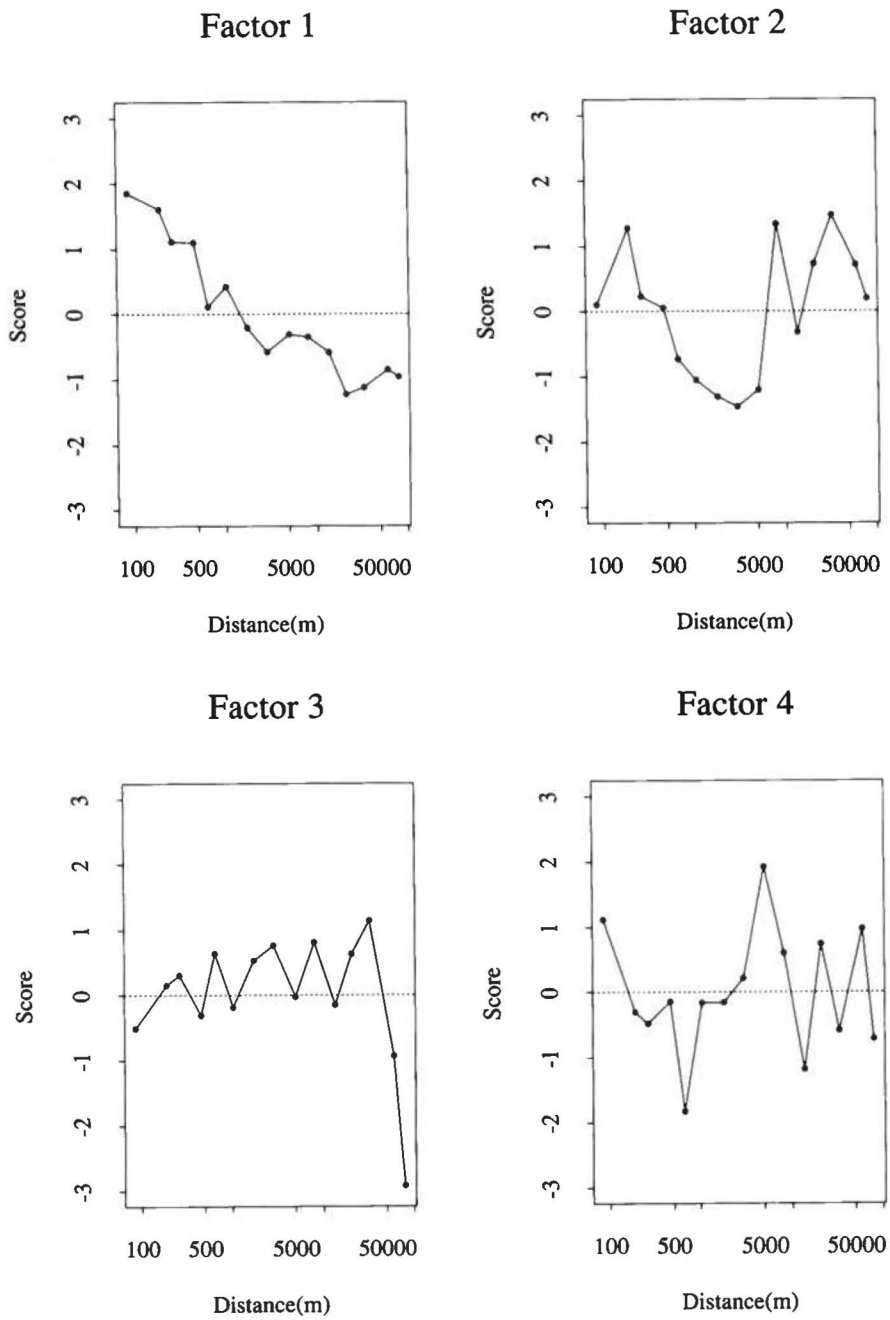

Fig. 6. Factor scores as a function of distance in the westerly direction from Maarmorilik.

Table 10. Concentrations of metals (μg g^{-1}) in *Cetraria nivalis* at increasing altitudes at a distance of 1,800 m from Maarmorilik.

Station	Altitude m a.s.l.	n	Ag	Cd	Cu	Fe	Pb	Zn
12	10	5	0.26	1.75	2.3	134	404	233
21	100	2	0.173	1.64	1.5	145	241	188
22	200	2	0.137	0.88	1.1	99	143	96
23	300	2	0.121	0.82	1.2	149	105	111
24	900	2	0.048	0.26	1.3	203	19	39

Table 11. Concentrations of metals (μg g^{-1}) in *Cetraria nivalis* and *Umbilicaria lyngei* sampled along a transect crossing a Pb-Zn mineralization.

Station	Distance	Ag		Cd		Cu		Fe		Pb		Zn	
		Cet	Umb	Cet	Umb	Cet	Umb	Cet	Umb	Cet	Umb	Cet	Umb
29	380 m N	0.053	0.074	0.14	0.22	1.17	2.03	189	519	7.4	9.7	18	29
30	250 m N	0.038	0.083	0.17	0.24	1.23	2.50	178	556	5.1	10.0	17	34
31	150 m N	0.029	0.065	0.12	0.20	0.88	2.30	185	658	5.4	10.3	17	37
32	50 m N	0.050	0.123	0.17	0.23	1.28	2.50	183	538	8.6	9.4	24	52
33	25 m N	0.060	0.077	0.67	0.37	1.41	3.65	346	545	32.0	17.3	124	140
34	2 m S	0.062	0.161	0.15	0.15	1.41	3.19	237	568	37.2	67.7	27	36
35	100 m S	0.044	0.116	0.09	0.08	1.42	2.30	251	475	5.4	8.6	17	35
36	200 m S	0.037	0.093	0.08	0.15	0.90	3.47	119	405	4.7	9.1	16	24
37	400 m S	0.045	0.091	0.10	0.09	1.01	2.84	156	427	5.6	8.6	18	28
COV(%)		9.4	12.4	15.9	15.5	13.1	15.8	22.7	11.9	19.1	7.2	5.6	14.5

$$COV = 100 \times \sqrt{\frac{1}{2N} \sum (\log(x_1) - \log(x_2))^2},$$

where N = number of sample sets.

High concentrations of Ag, Cd, Cu, Pb, and Zn were found close to the mineralization. The high values were, however, only found within very short distances from the vein: 2 m S, 25 m N and 50 m N. The terrain slopes to the north with station 37 (400 m S) at an altitude of 800 m a.s.l. and station 29 (380 m N) at 700 m a.s.l.

To test the significance of the elevated concentrations a two-way analysis of variance was carried out on the data of both species (Table 12). Significant differences between stations were found for Cd, Pb, and Zn, between species for Ag, Cu, Fe, and Zn; where *Umbilicaria* had the highest concentrations. The metal concentrations of the two species were closely correlated. The correlation coefficients (Table 12) were all positive, although only significant for Cd, Pb, and Zn.

The concentrations found at distances of 100 m or more from the mineralization can be regarded as the general concentration level in the area. These are for *Cetraria:* Ag 0.040 μg g^{-1}, Cd 0.11 μg g^{-1}, Cu 1.1 μg g^{-1}, Pb 5.5 μg g^{-1} and Zn 17 μg g^{-1}. These concentrations are slightly higher than the background concentrations for Greenland.

Supplementary samples

A number of supplementary samples of *Cetraria* were taken throughout the area. The results of the metal analyses are shown in Table 13. Station 19 (Fig. 4) was taken very close (12 m) to the conveyor belt and showed accordingly high concentrations, but unfortunately very little material was available at this station. Stations 25 and 26 (Fig. 3) were placed 3,800 m SE of Maarmorilik for comparison with the pilot investigation. Station 28 (Fig. 2) was placed 6,000 m S of Maarmorilik but separated from the mining town by a 1,000 m high mountain massif. The concentrations found at this station were very low, close to background concentrations. Concentrations of the same level were also found at station 4 (Fig. 2) which was placed 15,000 m SE of Maarmorilik. The concentrations found at station 6 (Fig. 2) at Uvkusigssat were generally lower than those found at the nearby station 5. Station 6 was, however, sheltered from Maarmorilik by high mountains. Station 3 (Fig. 2), 20 km NW of Maarmorilik, showed generally low concentrations except for Cd and Zn which were found in higher concentrations than expected according to the distance from Maarmorilik.

Results of the analyses of the 20 samples (nos. 41 to 60, Fig. 3) of *Cetraria* from Maarmorilik Plateau sampled in 1982 are given in Table 13. Generally there is only little variation between the concentrations throughout the area. The mean concentrations ± standard deviations are: Cd 0.30 ± 0.09, Pb 9.8 ± 5.1 and Zn 27 ± 7. The

Table 12. Statistical treatment (two-way analysis of variance and correlation coefficient) of the metal concentrations in *Cetraria nivalis* and *Umbilicaria lyngei* sampled along a transect crossing a Pb-Zn mineralization (*$p < 0.05$, **$p < 0.01$, *** $p < 0.001$).

	Ag	Cd	Cu	Fe	Pb	Zn
F between stations	3.023	3.741*	1.130	2.225	11.546**	29.973***
F between species	68.624***	0.179	86.914***	119.015***	0.032	46.717***
Correlation coefficient of species	0.508	0.754*	0.061	0.499	0.855**	0.963***

Table 13. Concentrations of metals (μg/g) in *Cetraria nivalis* at various localities in the area around Maarmorilik.

Station	n	Ag	Cd	Cu	Fe	Pb	Zn
3	2	0.187	1.14	1.82	224	7.3	31
4	2	0.053	0.06	1.38	126	1.8	11
6	2	0.277	0.11	2.02	320	4.8	16
19	2	1.292	14.78	10.47	331	6820	2010
20	2	0.781	9.31	7.93	325	3740	1250
25	2	0.106	0.91	1.15	93	99	93
26	2	0.119	1.11	1.40	155	123	116
27	2	0.093	0.43	1.26	104	65	48
28	2	0.027	0.07	0.99	119	3.8	24
41	2		0.28	1.2		19.6	35
42	2		0.29			9.1	26
43	2		0.23	1.8		7.4	25
44	2		0.30			10.0	30
45	2		0.37			10.7	36
46	2		0.37			14.9	35
47	2		0.36			19.0	42
48	2		0.26			14.7	25
49	2		0.38			9.0	32
50	2		0.58			4.0	31
51	2		0.21			3.2	27
52	2		0.21			9.5	16
53	2		0.24			5.3	15
54	2		0.24			3.6	23
55	2		0.30			5.7	19
56	2		0.21			9.3	22
57	2		0.35			19.3	27
58	2		0.36			6.6	33
59	2		0.25			7.1	17
60	2		0.28			8.9	22

highest Pb and Zn concentrations were found along the edge of the plateau towards the fjords Qaamarujuk and Affarlikassaa. The concentrations at these stations are 2–3 times higher than in the south-western part of the plateau.

Discussion

Concentrations in relation to distance from Maarmorilik

The main objective of this study was to describe the spread of metals in the direction west of Maarmorilik, in which direction the greatest spread was found in the pilot investigation. The results of this part of the investigation were summarized in Table 7 and Fig. 5. Significant regressions of concentrations vs. distance were found for Ag, As, Cd, Cu, Hg, Pb, Sb and Zn. The amounts of the total variations accounted for by the regressions were very high (78–96%). A factor analysis resulted in a model with four factors accounting for 84% of the total variation. The first factor included the elements with significant regressions and the elements Se and Ba. The elements Na and Cr were negatively correlated to this factor. The factor scores of factor 1 decreased with increasing distance from Maarmorilik (Fig. 6). The distribution of the elements with significant regressions and Se and Ba can clearly be ascribed to pollution from Maarmorilik.

Sources and spread of metals

The main sources of airborne metal pollution in the mining complex were waste dumps, the crusher in the mine, the cable car, which transported ore to the mill, the concentrate storage and the conveyor belt and ship-loader. The concentrations found in lichens point to the conveyor belt and ship-loader as the most important sources. Ship-loading is going on for about a week up to 8 times a year in the ice-free period (June to December). The meteorological conditions during the periods with ship-loading are therefore crucial for the spread of metals. The contribution of other sources of pollution will be of greatest importance in the periods intervening ship-loading.

The concentrates consist mainly of PbS and ZnS, respectively. Small amounts of more soluble Pb– and Zn–compounds may be present in the concentrates together with other metals present in the ore, especially Cd in the Zn-concentrate, Ag in the Pb-concentrate, and Cu in both concentrates (Table 1). The Cu in the concentrates has its origin in both the ore and in $CuSO_4$ added in the flotation process.

The formation and transportation of dust from the concentrates were mainly caused by the wind. The pick-up velocities (*i.e.* the air speed required to lift and carry particles) have been determined for various types of coarse dusts (granite, silica, coal) by Djamgouz and Ghonein (1974). They found pick-up velocities for granite for the following size classes: 10–35 μm: 4 m sec^{-1}, 35–75 μm: 6 m sec^{-1}, and 75–105 μm: 7 m sec^{-1}. Dust may be raised by other means than wind, *e.g.* by vehicle passage and cleaning up after ship-loading. Other important factors in the spread of particles are wind direction and humidity. Wet dusts require higher wind-speeds to be lifted than dry dusts, and rainfall washes out suspended particles from the atmosphere.

Winds with speeds required to lift and carry concentrates particles are frequent in Maarmorilik, blowing mainly towards the west. The horizontal dispersion of dusts from Maarmorilik perpendicular to the westward transect is limited by high mountains (1,000 to 1,500 m) along the Qaamarujuk fjord and further out in the Ummanaq fjord complex, and the vertical dispersion is believed to be small due to meteorological conditions and gravity. The analyses of *Cetraria* from different altitudes (Table 10) thus showed very little spread in the vertical direction as compared to spread along the westward transect.

The extent of pollution

Background values of elements in *Cetraria* have been determined in a large number of samples (Pilegaard 1987). Background levels for Greenland and for the areas closest to Maarmorilik (the Sarqaq Valley, 130 km S and

Table 14. Concentrations (μg g^{-1}) of metals in *Cetraria nivalis* from background localities in Greenland (Pilegaard 1987) and from the Northwest Territories, Canada (Puckett & Finegan 1980).

Locality	n		Ag	Cd	Cu	Fe	Pb	Zn
Sarqaq	4	$\bar{x}$	0.02	0.06	1.5	210	1.7	20
		s.d.	0.02	0.02		105	1.1	10
Thule and	5	$\bar{x}$		0.11	1.5	94	1.1	11
Melville Bay		s.d.		0.03		40	0.6	6
Northwest	15	$\bar{x}$			6.2	260	5.6	25
Territories		s.d.			1.5	300	2.3	15

Melville Bay and Thule, 600–1000 km N) are given in Table 14 together with values from the Northwest Territories, Canada (Puckett and Finegan 1980). The regression line of Pb (Fig. 5) showed a very steep gradient and background levels were not reached within the investigation area; *i.e.* the concentrations at 80,000 m were still significantly higher than the background levels. Cd and Zn reached background levels at 80,000 m, Ag, As and Sb at around 10,000 m, and Hg and Cu already at around 1000 m.

Heterogeneity around the regression lines

Some heterogeneity exists around the regression lines (Fig. 5). This heterogeneity was due mainly to other variations than analytical variation. Since the biological variation was minimized by pooling of samples, the remaining part of the variation was due to localization of stations. The actual sampling sites might have differed with respect to the immediately surrounding topography, and therefore the exposure to the pollution. Another explanation of the heterogeneity is that the spread of pollutants did not necessarily follow a fixed model all the way out of the fjord system. Extraordinary spread may have arisen at broads, *e.g.* between stations 8 and 5 and beyond station 7. Conversely extraordinary high deposition of particles may be found where the wind meets obstacles, *e.g.* rock walls. The heterogeneity was, however, rather small and had no great impact on the general pattern of spread of pollutants from Maarmorilik.

Spread of metals from an exposed Pb-Zn mineralization

To find out to which extent exposed mineralizations might influence the metal concentrations of lichens, samples of *Cetraria* and *Umbilicaria* were taken in the vicinity of a Pb-Zn mineralization. The metals Cd, Pb and Zn were found in elevated concentrations (up to 10 times higher than the regional background) very close (25 m) to the exposed mineralization. The spread of metals was found to be greatest in the direction north of the mineralization, which was down-slope. The two species showed the same variations in concentrations although the growth places and substratum were different. *Cetraria* was growing on the ground and *Umbilicaria* on lose blocks of granodiorite and thus elevated (about 1 m) from the ground. The close similarity in the concentration pattern of the two species supports the assumption that metals are spread as wind-blown particles from the mineralization.

The concentrations in lichens found at distances of 100–400 m from the mineralization can be regarded as a regional background level. Other mineralizations are found in the area, but the distance were at least 800 m and influence from these can therefore be neglected. The regional concentration found in this area is generally low and close to the background level for Greenland; but pollution from Maarmorilik might be responsible for the slightly elevated concentrations of Pb.

The influence of exposed mineralizations could be traced at a maximum distance of 50 m. Therefore the general pattern in the lichens around Maarmorilik is not believed to be influenced by exposed mineralizations, which are mainly found E, SE and S of Maarmorilik and near Uvkusigssat in a height of 1200 m a.s.l.

Sphagnum bag investigations

In order to investigate the conveyor belt and the shiploader as pollution sources, the "moss-bag"-technique (Goodman & Roberts 1971) was applied. This technique was developed in Great Britain and has been used in the monitoring of airborne metal pollution around industrial complexes (Goodman & Roberts 1971; Little & Martin 1974; Ratcliffe 1975; Cameron & Nickless 1977) and mineralizations (Pilegaard 1993). The main advantage of the method is that position as well as time and duration of the exposure can be adapted to the actual pollution situation.

Sphagnum bags were exposed during a period when a ship was loaded with concentrates in 1980 and compared with an equally long period without this activity immediately following the other. As a result of the 1980-investigation the conveyor belt was covered with a roof and a similar investigation was performed in 1982 to examine the effects of this remedial action.

Materials and methods

Sphagnum girgensohnii Russ. was sampled in an area without local pollution sources in Southern Sweden. The moss was rinsed for impurities, washed in demineralised H_2O, and dried at room temperature. Two grams of moss were placed in nylon bun nets, which were immersed in distilled H_2O and then formed to balls with a diameter of approximately 5 cm. The moss bags were kept in a deep-freezer until exposure.

Sphagnum bags were exposed during loading of the ship in the period 19 August to 23 August 1980, where a total of 6,600 t Pb and 17,200 t Zn was loaded. Four bags were placed suspended 1.5 m above the ground on wooden gallows-like stands at stations A, B, C, and D (Fig. 4). At the end of the ship-loading the bags were replaced by 4 new ones, which were exposed during a period of the same length. In 1982 a set of moss bags were exposed during ship-loading from 1–5 August and after loading from 5–11 August. The total amounts loaded in 1982 were 2, 000 t Pb and 17,200 t Zn. The bags were placed at the same stations as in 1980 and at stations A and D four replicates were exposed.

Results

The results of the analyses of metal concentrations are shown in Fig. 7. The concentrations in the reference moss bag were Cd: 0.46 μg g^{-1}, Pb: 16 μg g^{-1}, and Zn: 78 μg g^{-1}, shown as horizontal lines. There was a marked increase in the concentration of all the investigated metals in the moss bags exposed during loading both in 1980 and in 1982. The concentrations of Cd, Pb and Zn increased significantly also in the period after loading. The highest accumulations of Cd, Pb and Zn during the loading were found at station B.

The results show a strongly increased metal pollution during the loading of the concentrates. It can be implied from the distribution patterns that the pollution was caused by loss of concentrate from the conveyor or the ship-loader or whirling up of dust when the concentrate fell down into the hold of the ship. The concentrations found at station A (which was placed immediately above the ship-loader) were lower than those found at station B and C.

The absolute concentration increase, *i.e.* the difference between the concentration after exposure and the initial concentration, was highest for Pb followed by Zn and Cd in this order. The order of amounts of metals produced from the mine is Zn > Pb > Cd.

Increases in the concentrations of Cd, Pb and Zn were also found during the second exposure period (after loading). During this period, however, the highest concentrations were obtained at station A.

During loading in 1982 the resulting concentrations of Cd, Pb and Zn in the moss bags were 50% lower than in 1980. Otherwise, the same pattern was found in the two years. The amount of Zn loaded during the two years was the same, whereas the amount of Pb loaded in 1982 was only 1/3 of that in 1980.

Discussion

The pattern of concentrations found in the moss bags during loading can be explained by: 1) loss from the conveyor at station B, or 2) whirling up of dust from the loading of the ship, blown in the direction of station B. The source of pollution in the period after loading was probably loss of concentrate at the ship-loader. By examination of the increases in element concentrations during the second exposure, it can be deduced that the loss was primarily of Zn-concentrate.

Cd is mainly found in the Zn-concentrate, the Zn/Cd ratio being approximately 190. The ratio calculated on the basis of the absolute concentration increase in the mosses exposed during loading in 1980 was Zn/Cd: 200, which is in good agreement with the concentrates. The Pb/Zn ratio during loading in 1980 was 0.38. The ratio for the mining period up to 1980 was 0.35. The Pb/Zn ratio calculated on the basis of the absolute concentration increase in the mosses exposed during loading was 1.4 at station B falling to 0.25 at station A. The ratio in *Cetraria nivalis* from stations near the conveyor belt was 3. The mosses were wet at the beginning of the exposure, and it is known from other investigations (Clough 1975) that the uptake of metals is more efficient when the mosses are wet. In both years Pb-concentrate was loaded first. At sampling after the exposure period the mosses were dry, but the possibility of the mosses being moistened during the exposure by rain or dew cannot be excluded. However, as mentioned above the *in situ* vegetation also showed a higher Pb/Zn ratio than expected. The explanation of the discrepancy in the ratios might be a difference in spread of particles due to a difference in particle size distribution in the two concentrates. Another contributing factor rnight be the different retention of the metals in the moss tissue. Rühling & Tyler (1970) have shown that the retention of Pb is stronger than that of Zn and Cd. The Pb/Zn ratio varies between the stations, which indicates that the winds might have changed during the exposure period.

The results from the two years are somewhat contradictory because an improvement is found for the Zn-concentrate (Zn and Cd), but not for the Pb-concentrate. If the 50% reduction for Zn and Cd should hold for Pb also, the concentrations in the moss bags should only have been 1/6 of the concentrations in 1980. The explanation for this discrepancy might be that the linearity of the relationship between air concentration and deposition does not extend to situations with very high air concentrations of particulate matter (Vestergaard *et al.* 1986). The results nevertheless indicate that the remedial actions

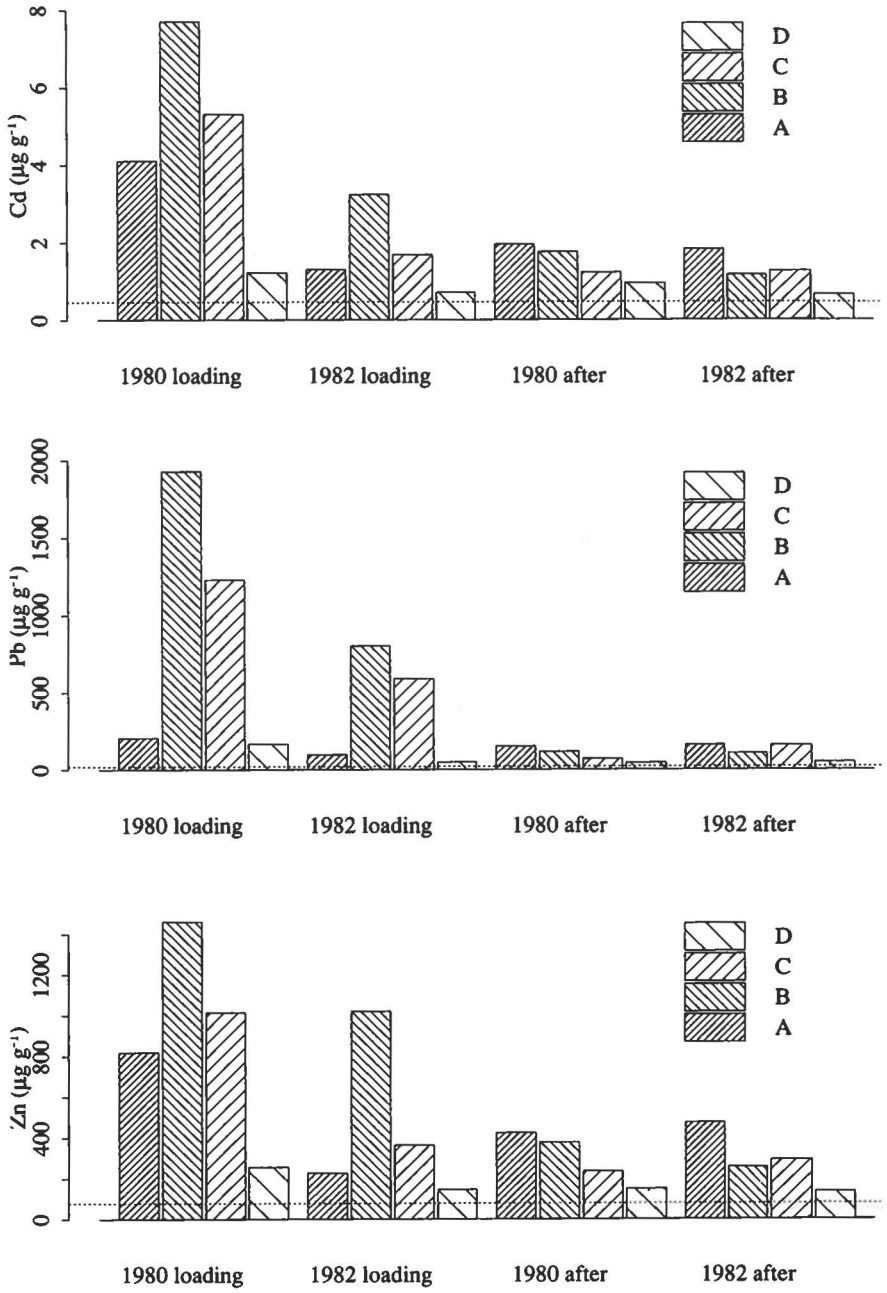

Fig. 7. Concentrations of Cd, Pb and Zn (µg g^{-1}) in bags of *Sphagnum girgensohnii* during and after loading of concentrates in 1980 and 1982. A, B, C and D are the four different stations. Horizontal lines represent background concentrations.

performed between the two periods (covering of the conveyor belt with a roof) have improved the situation.

Other investigators (*e.g.* Cameron & Nickless 1977) have presented the problem that some moss material from the bags might have been lost. In this investigation, however, no loss was observed as the exposed bags had the same dry weight as the unexposed reference bag. The mosses have reacted very quickly to the ambient metal concentrations in the air, probably because of the extremely high amounts of Pb accumulated during the first 10 hours after exposure. The total exposure time of 100 hours seems to be sufficient, at least during loading. It can generally be recommended to keep the exposure time as short as possible to prevent loss from the bags and reduce the influence of climatic changes.

The survey has shown that the moss bag method is effective in the monitoring of the relative amounts, duration, time and locality of airborne metal deposition. The results show that there was a strongly increased metal air pollution in Maarmorilik during the period when concentrates were loaded in the ship. The primary sources of this pollution were the conveyor and ship-loader. These re-

Table 15. Regression coefficients ± standard errors (s.e.) for individual years.

Year	log (a)	b	n
1979	12.16±0.29	−0.85±0.04	9
1980	14.46±0.69	−1.18±0.08	15
1982	11.81±2.10	−0.72±0.35	5
1983	13.07±1.36	−1.00±0.16	11
1984	15.73±1.36	−1.37±0.16	10
1985	11.80±1.18	−0.93±0.14	15
1986	11.42±1.08	−0.81±0.13	14
1987	13.40±1.21	−1.15±0.15	17
1988	15.67±1.18	−1.44±0.15	16
1989	15.28±1.17	−1.38±0.15	15
1990	14.80±1.45	−1.30±0.19	11

sults are in agreement with those obtained by the investigation of metals in the *in situ* vegetation.

Development in deposition with time

Sampling of *Cetraria nivalis* became a part of the monitoring programme for the mining activity from 1983. The sampling and analysis of lichens from stations along the fjords were carried out by the Geological Survey of Greenland and the Greenland Environmental Research Institute (Asmund *et al.* 1988; Asmund 1991). This material enables an evaluation of the changes in atmospheric deposition during the life-time of the mine.

Comparison of concentrations in the years 1979–1990

The change in atmospheric deposition over time is illustrated by comparing the regression lines of Pb for the individual years at stations in the westward direction (Table 15). The regression equations were compared by means of analysis of covariance (Freund *et al.* 1986). A test for heterogeneity of slopes showed that the slopes of the individual years were not significantly different. Therefore an analysis of covariance with a common slope was carried out and the intercepts of the individual years calculated (Table 16). Based on the comparison of the adjusted treatment means (Table 17) it is found that the intercepts generally can be divided into three different groups. The first group comprises the years 1979, 1980, 1982 and 1983; the second group the years 1984, 1985 and 1986; the third group the years 1987, 1988, 1989, 1990. The intercepts are significantly lower in the later years than in the earlier. The years 1984–1986 seem to be a transition period.

Table 16. Regression coefficient ± standard errors (s.e.) calculated with common slope.

Year	log (a)	b	n
1979	14.00±0.38	−1.12±0.04	9
1980	13.99±0.40	do.	15
1982	14.21±0.40	do.	5
1983	14.08±0.43	do.	11
1984	13.61±0.43	do.	10
1985	13.37±0.40	do.	15
1986	13.88±0.39	do.	14
1987	13.18±0.39	do.	17
1988	13.13±0.40	do.	16
1989	13.26±0.39	do.	15
1990	13.46±0.39	do.	11

Based on the adjusted treatment means it can be calculated that if the emission level in 1979–1983 is set to 100% the level in 1987–1990 is 44%; provided that the uptake and retention in the lichens are directly proportional to the deposition.

Another approach to the question of development with time is simply to look at concentrations in samples from the same stations over the years. Sampling was performed in 4 or more years at 5 stations at different distances in the direction west of Maarmorilik (Fig. 8). If the level of 1979–1983 is taken as the reference pollution level, the average levels of these stations were in 1984: 43%, 1985: 46%, 1986: 41%, 1987: 31%, 1988: 30%, 1989: 35% and 1990: 34%. This is a little lower than the values derived from the regression equations. However, the tendency is the same.

Discussion

Campbell (1976) and Nieboer & Richardson (1981) have discussed the theory of the distance relationship:

$$y = ax^b + c, \text{ or } \log(y - c) = \log(a) + b \log(x),$$

where y = concentration in lichen, c = background concentration in lichen, and x = distance from Maarmorilik. The exponent (b) is specific for the pollution source. Changes in source strength only lead to changes of the constant (a), while (b) remains constant. Changes in emission characteristics (*i.e.* particle size distribution) or changes in the relative importance of different sources affect (b). The constant (a) is affected if the total emission is changed and/or if the relative contribution from sources geographically apart are changed.

The presented results show no significant changes in the exponent (b), whereas there is a significant change in the constant (a). It can thus be concluded that the emission from the mine has decreased, but the emission characteristics have remained constant. It should be noted that a change in emission only can be measured in the lichens after some time due to the long mean residence time of lead in lichens (Persson *et al.* 1974). The findings

Table 17. Comparison of adjusted treatment means of analysis of covariance. Significance of difference: *** = p<0.001, ** = p<0.01, * = p<0.5, n.s. = non significant.

Year	1979	1980	1982	1983	1984	1985	1986	1987	1988	1989	1990
1979	–	n.s.	n.s.	n.s.	n.s.	*	n.s.	**	**	*	n.s.
1980	n.s.	–	n.s.	n.s.	n.s.	*	n.s.	***	***	**	*
1982	n.s.	n.s.	–	n.s.	n.s.	*	n.s.	**	**	**	*
1983	n.s.	n.s.	n.s.	–	n.s.	**	n.s.	***	***	**	*
1984	n.s.	n.s.	n.s.	n.s.	–	n.s.	n.s.	n.s.	*	n.s.	n.s.
1985	*	*	*	**	n.s.	–	*	n.s.	n.s.	n.s.	n.s.
1986	n.s.	n.s.	n.s.	n.s.	n.s.	*	–	**	**	*	n.s.
1987	**	***	**	***	n.s.	n.s.	**	–	n.s.	n.s.	n.s.
1988	**	***	**	***	n.s.	n.s.	**	n.s.	–	n.s.	n.s.
1989	*	**	**	**	n.s.	n.s.	*	n.s.	n.s.	–	n.s.
1990	n.s.	*	*	*	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	–

are in agreement with the pollution abatement carried out at the mine.

Conclusions

The spread of metals from the Maarmorilik mining complex was found to be most pronounced in the direction west of the mining town. Analyses of the lichen *Cetraria nivalis* showed that significant regressions vs. distance were found for the elements Ag, As, Cd, Cu, Hg, Pb, Sb and Zn. The concentrations of these elements decreased with increasing distance from the mining town. A factor analysis with a 4-factor model resulted in a factor including the above mentioned elements and Se and Ba. The scores of this factor clearly decreased with increased distance from Maarmorilik. The highest concentrations of metals related to the mining activity were found close to the conveyor belt and ship-loader. Concentrations found in this area were up to 4000 times higher (Pb) than the background level. The background level of Pb was not reached within an 80 km distance westward of Maarmorilik.

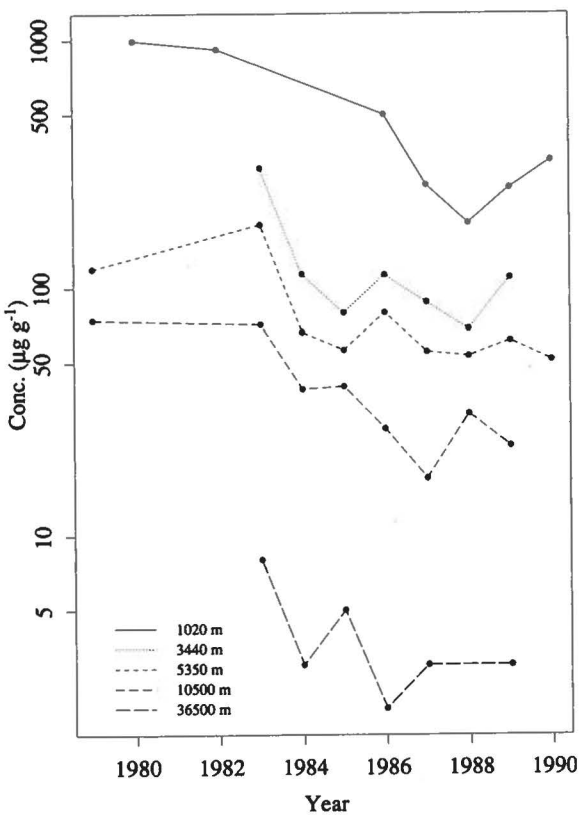

Fig. 8. Development in Pb-concentration (µg g^{-1}) in *Cetraria nivalis* at 5 discrete distances during the years 1979–1990.

Background levels for Cd and Zn were reached at 80 km, for Ag, As and Sb at 10 km, and for Hg and Cu at 1 km. The pollution was generally confined to low altitudes.

Exposed mineralizations were found not to contribute to the overall concentration pattern, since locally elevated concentrations around these could be traced only within a very short distance (50 m).

The results of the *Sphagnum* bag investigations showed that there was a strongly increased metal air pollution in Maarmorilik during the period where concentrates were loaded in the ship. The primary sources of this pollution were the conveyor and ship-loader. These results are in agreement with those obtained by the investigation of metals in the *in situ* vegetation.

An analysis of deposition of Pb during the years 1979–1990 showed that pollution in the years 1987–1990 was only about half of that in the years 1979–1983. Since the production of the mine was almost stable during the years, the remedial actions carried out must be the cause for the lowering of the pollution.

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