

# Epiphytic lichen distribution and plant leaf heavy metal concentrations in Russian–Norwegian boreal forests influenced by air pollution from nickel-copper smelters

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The Norwegian–Russian border area is exposed to air pollution from Russian nickel-copper smelters at Kola Peninsula. An attempt was made to relate distribution and abundance of epiphytic lichens to concentrations of sulphur, nickel and copper in birch and bilberry foliage and soil. Lichen cover showed significant correlation with Ni and Cu concentrations. However, since the deposition patterns of airborne heavy metals and sulphur dioxide (SO<sub>2</sub>) around the smelters differ, some plots experience low lichen cover, despite low heavy metal concentrations. These plots are affected by relatively high SO<sub>2</sub> emissions. Climatic variability within the study area may also play a role in explaining variation in lichen cover. For areas with uniform climate and physiognomy nearer than ca. 20 km from the smelters, Ni concentrations in birch leaves may prove useful for estimating the likelihood for recolonization to take place. The area closest to the smelters presently is much too polluted for lichen recolonization to occur.

## Introduction

Long-term emissions from the Russian copper-nickel smelters at Kola Peninsula close to the Norwegian and Finnish borders have led to extensive damage of surrounding vegetation (e.g. Aamlid 2002). The main components of air pollution from the smelters are sulphur diox-

ide-containing (SO<sub>2</sub>) gases and heavy metal particles, primarily of nickel (Ni) and copper (Cu). The northern boreal (subarctic) vegetation surrounding the smelters is dominated by birch (*Betula pubescens*) and dwarf shrub heaths, originally with thick mats of terricolous lichens (Tømmervik *et al.* 1998). The emissions were particularly high in the period from

1974 to 1984, and during and after this period, severe degradation of vegetation was evident (Tømmervik *et al.* 1995). Also epiphytic lichen vegetation was affected. *Hypogymnia physodes* and *Melanohalea olivacea* are the most common epiphytic lichens on birch stems in northern Fennoscandia, generally occurring from sea level to tree line in unpolluted sites. However, around the smelters there is a zone without any epiphytic lichens, the so-called epiphytic lichen desert zone (Hawksworth and Rose 1976), and the abundance of these two lichens outside the lichen desert zone is correlated, albeit not closely, with modelled patterns of air pollution (Aamlid and Skogheim 2001).

In air pollution studies, lichens are often used as biomonitors, since pollutants like heavy metals readily accumulate in the lichen thalli (Hawksworth and Rose 1976). However, too high concentrations eventually kill the lichens, as observed around the copper-nickel smelters, and in such heavily polluted areas native epiphytic lichens are not available for biomonitoring. In the area along the Russian–Norwegian border, epiphytic lichens are absent also from assumingly low-polluted sites (Aamlid and Skogheim 2001), where one could expect occurrences of epiphytic lichen populations. Thus, based on these previous investigations from the Norwegian–Russian border area, two questions arise: (1) What causes the low epiphytic lichen cover in apparently healthy, unpolluted sites? (2) Is it possible to relate the occurrence and absence of epiphytic lichens to concentrations of pollutants in other types of living biomass or in soil?

In order to elucidate in more detail the local variation in air pollution deposition and effects on vegetation, and to monitor future changes in vegetation and deposition rates, a large biomonitoring network was established, including plots both in Russia, Norway and Finland, and involving multidisciplinary personnel from the three countries. Data acquired through this network made it possible for us to answer the questions raised above. It was important to find a source of living tissue that could reflect the (potential) habitat of epiphytic lichens both in areas these lichens are absent and do occur. Living birch trees are found even close to the smelters despite the heavy pollution there (Tømmervik

*et al.* 1995, 2003, Kozlov 2005), and birch is also the primary host for the epiphytic lichens in the region. Thus, we considered birch leaves as a suitable plant tissue for use in correlative studies of plant chemistry and lichen distribution patterns. Bilberry (*Vaccinium myrtillus*) was also included in the study.

## Material and methods

### Study area

The study area is situated around the Russian nickel-copper refinery in Nikel and Zapolyarniy, two small towns at Kola Peninsula, close to the north-easternmost parts of Norway and Finland. It is hilly, located close to the arctic tree line and has a thin soil cover ( $30 \pm 5$  cm). Most of the area is covered by tills with coarse texture. Podzol is the most common soil type (Koptsik *et al.* 1999). The forests are mainly characterized by scarce tree stands of pine and birch and scarcely-developed ground vegetation, dominated by dwarf shrubs, mosses and lichens. However, denser and richer birch forests are found along the main rivers (Tømmervik *et al.* 2003). The smelter in Nikel was established in 1933. Emissions reached a level of 100 000 tonnes of SO<sub>2</sub> per year at the highest during the first 30 years of production. The emissions during this period affected only the vegetation cover in the near surroundings of the smelters (Kalabin 1991). From 1974 onwards, also Ni ore with a high content of S from the Norilsk ores in Siberia has been processed in the smelters. As a result, the emissions of SO<sub>2</sub> increased to 400 000 tonnes in 1979. Thereafter, the emission levels have slowly decreased, with ca. 160 000 tonnes being emitted in 2000 (Aamlid 2002). The high levels of SO<sub>2</sub> emissions resulted in a severe degradation of the natural environments in the area (Tømmervik *et al.* 1995). Emissions of heavy metals like Cu and Ni have also contributed to the damage on the vegetation in the area. After 1991, the pollution impact on the Norwegian side of the border was affected by the practice of reducing the output from the smelters when the dominant wind direction was towards Norway.

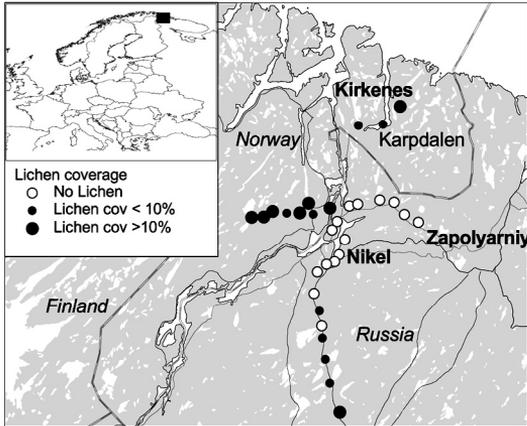


Fig. 1. Study area: the 31 study plots sampled in 2000.

### Estimation of lichen cover

Three transects extending north, south and west from the Cu-Ni refinery in Nikel were selected for the project (Fig. 1). Along these transects, a number of sample plots, used by all scientific disciplines involved (botany, vegetation ecology, soil science, geology and zoology) in the overall project, were located in order to fulfil all demands of each discipline. The sample plot design is schematically presented in Fig. 2. Each plot has a central point E, and four sub-plots A–D, in which biological, soil science and geological sampling and observations took place (Yoccoz *et al.* 2001). The nearest birch in the centre of each sub-plot was selected for measurement of occurrence and abundance of epiphytic lichens. In accordance with Jonsell (2000), sub-specific rank of *B. pubescens* was not used. In total, 31 plots were investigated in August 2000 (Fig. 1). The plots are located between 3 km and 38 km from Nikel. The lichen abundance was estimated at 150 cm above ground, chiefly in accordance with methods described by Aamlid *et al.* (2000), except that only one height level was used. A measuring tape with markers at each centimetre was placed around the stem, and the number of markers covering a single species was recorded for each aspect. All lichens, crustose as well as macrolichens, were included in the study. Crustose lichens were so scarce that they actually never were recorded, although they were occasionally observed on other parts of the stems. The lichen abundance is presented as rela-

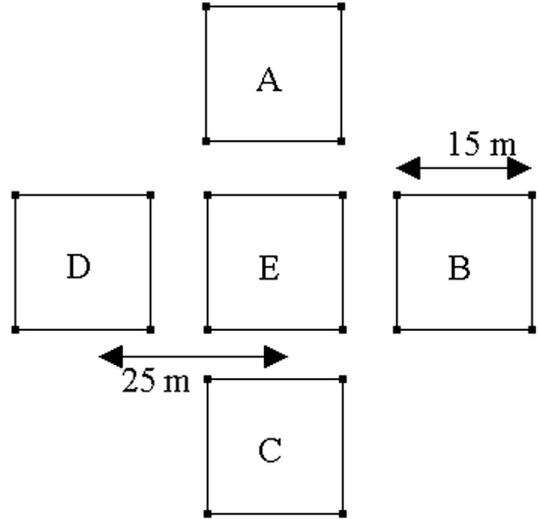


Fig. 2. Sketch of plot lay-out showing the position of the five sub-plots A–E. The birch trees closest to the centre of each of the sub-plots were selected ( $n = 5$ ).

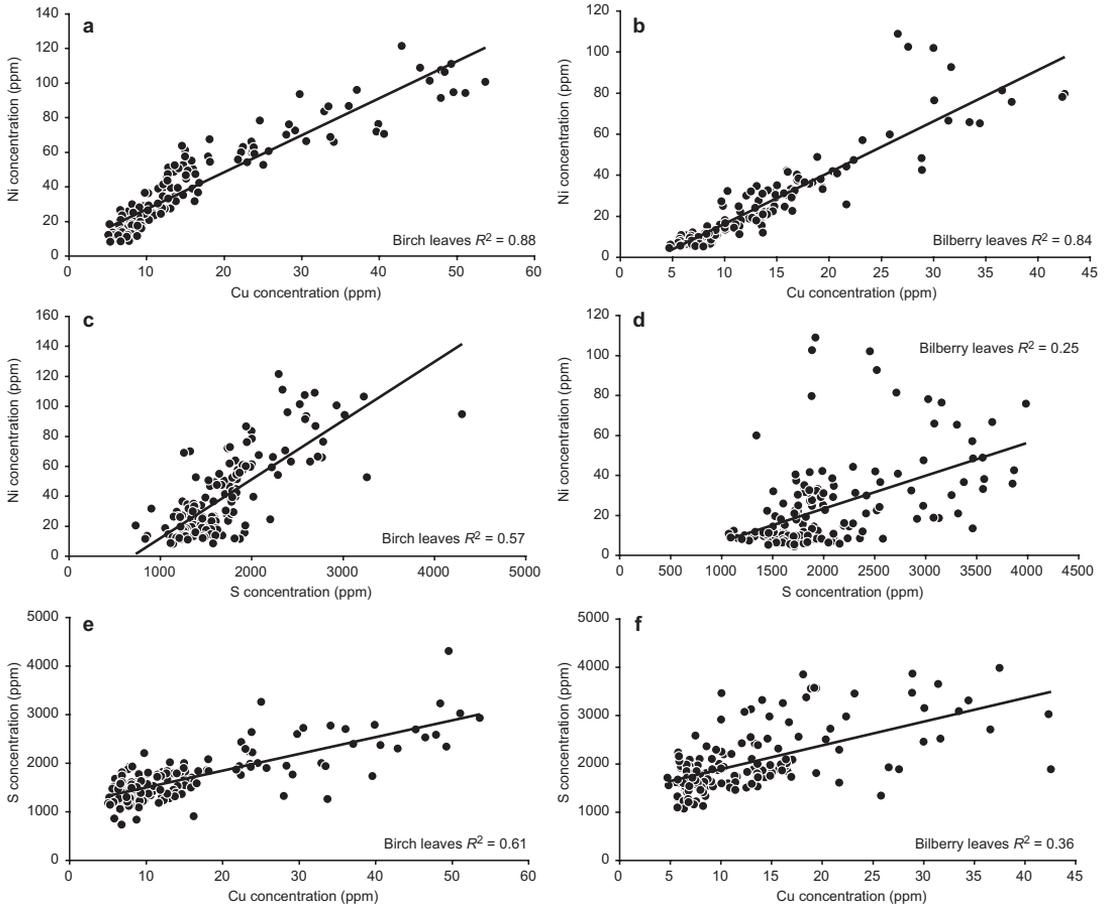
tive cover, *viz.* number of markers divided on the stem circumference (Aamlid *et al.* 2000). Thus, maximum score, *i.e.* cover, is 1 (or 100%).

### Chemical analyses of heavy metals and sulphur

From each birch checked for lichens, leaves were collected for analysis of chemical composition. In addition, leaves of bilberry from nearby plants, and soil samples, were taken and brought to the laboratory.

Plant samples were not washed prior to the analysis, due to potential leaching of elements from the interior of the plants. The element concentrations in the leaves were determined after nitric acid digestion, and atomic absorption spectrometry was used to determine the concentrations of Cu and Ni using procedures described previously (Lukina and Nikonov 2001). For preparation of AA standards and blanks commercial standards (J.T. Baker, The Netherlands and Merck, Germany) and Millipore purification system water were used. The S content was determined by colorimetry.

A- and C-horizon samples were air-dried and sieved through a 2-mm sieve. Sample fractions < 2 mm were digested with concentrated



**Fig. 3.** Correlation between copper, nickel and sulphur concentrations in birch and bilberry leaves in 155 samples from 31 plots in the Norwegian–Russian border area. (a) Ni versus Cu in birch, (b) Ni versus Cu in bilberry, (c) Ni versus S in birch, (d) Ni versus S in bilberry, (e) Cu versus S in birch, (f) Cu versus S in bilberry. Five samples were taken from five different plants at each plot.

nitric acid in autoclave in accordance with Norsk Standard NS 4770, *see* Reimann *et al.* (2001) for details. The element concentrations were determined using a Thermo Jarrell Ash ICP 61 instrument for ICP-AES analyses.

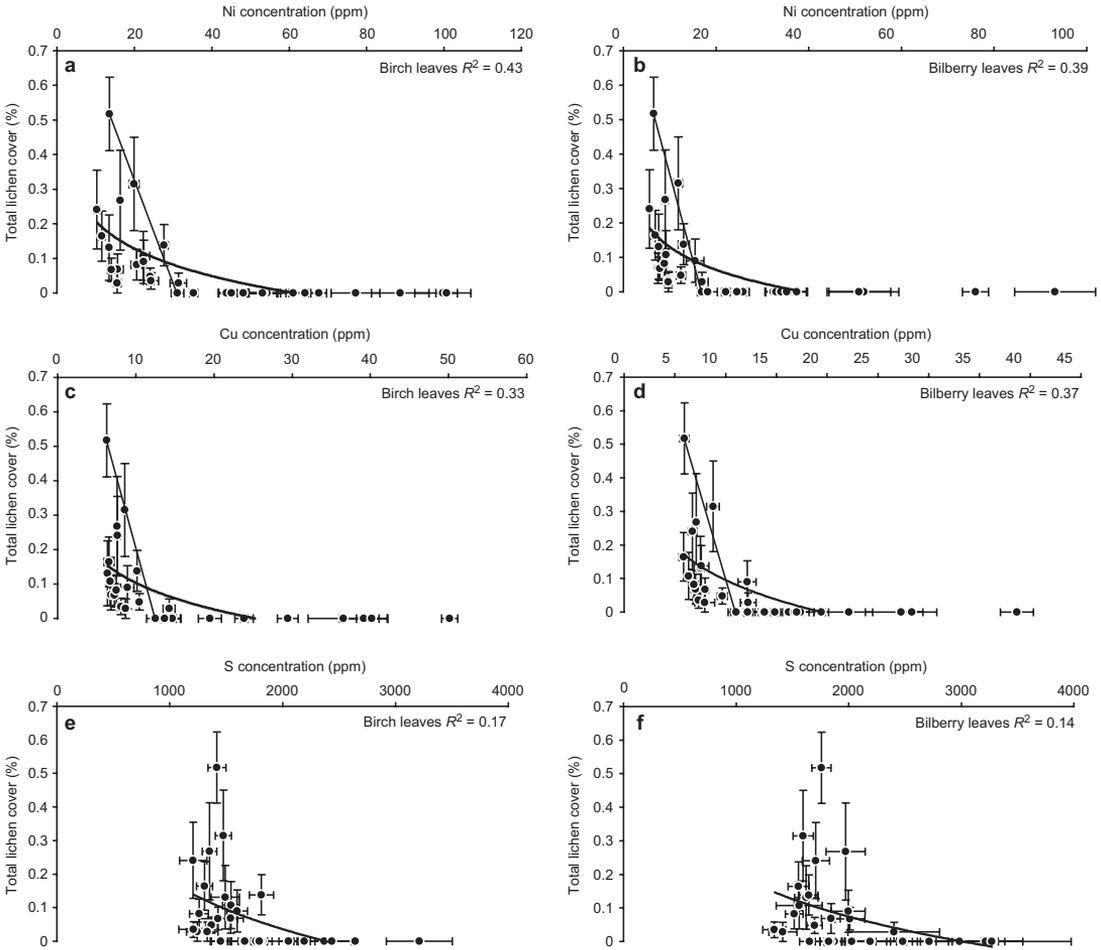
## Regression analysis

For the analyses involving lichen cover, correlation was estimated by means of logarithmic regression. Since the lichen data set contains several zero values, and since these values are proportions of maximum possible score, arcsine transformation of lichen data and logarithmic transformation of chemical data lead to slightly better regression lines. However, we choose to

present untransformed data for the sake of clarity. Linear regression generally shows a lower relationship between pollutant and biological response than does logarithmic regression (Richardson 1987). Thus, the latter is used here, except in the graphs in which element concentrations are compared.

## Results

The concentrations of Cu and Ni in both birch and bilberry leaves were strongly correlated (Fig. 3a and b). The maximum levels of Ni were slightly above 100 ppm in both species, whereas the maximum levels of Cu were slightly higher in birch (54 ppm) than in bilberry leaves (43 ppm).



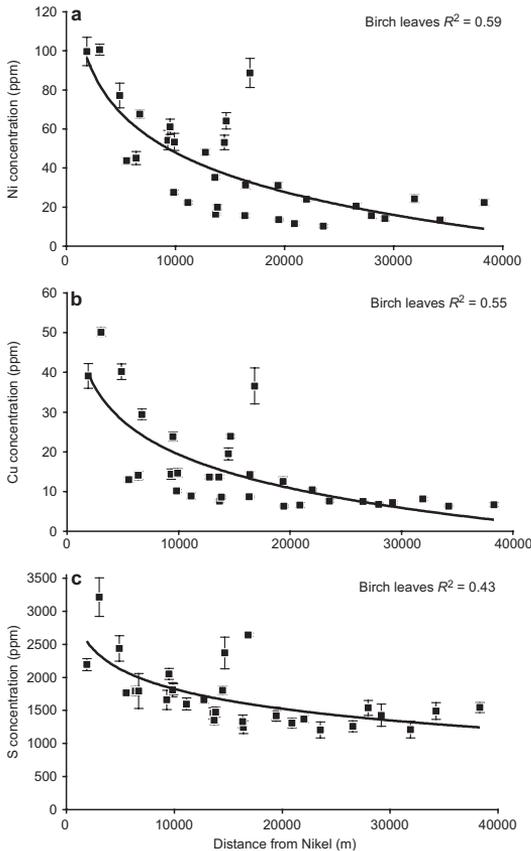
**Fig. 4.** Total relative lichen cover (as percentage of possible score) versus element concentrations in birch and bilberry leaves. (a) Ni concentrations in birch leaves, (b) Ni concentrations in bilberry leaves, (c) Cu concentrations in birch leaves, (d) Cu concentrations in bilberry leaves, (e) S concentrations in birch leaves, (f) S concentrations in bilberry leaves. Error bars, when larger than symbols, show  $\pm 1$  S.E.  $n = 5$ . The curved lines are the logarithmic regression lines (with the corresponding correlation coefficient), whereas the straight lines are the so-called potential maximum lichen cover lines, see text for further information.

S concentrations were more strongly correlated with Cu and Ni in birch leaves than in bilberry leaves (Fig. 3c–f), in both species reaching maximum mean levels slightly above 3200 ppm. Ni concentrations in birch leaves were closely correlated with Ni concentrations in bilberry leaves ( $R^2 = 0.92$ ) and in the A horizon of the soil ( $R^2 = 0.87$ ), but not with the values from the C horizon of the soil ( $R^2 = 0.19$ ), and similar relationships were found for Cu and S concentrations from the various tissues and soil horizons (not shown).

Of the 31 plots investigated, 15 were lacking epiphytic lichens (Fig. 1), and in the remain-

ing 16 plots, the total epiphytic lichen cover varied from 3% to 52% (Fig. 4). The most abundant lichen was *Melanohalea olivacea*. Other recorded species were, in decreasing order, *Hypogymnia physodes*, *Bryoria simplicior*, *Parmeliopsis hyperopta*, *Tuckermannopsis sepincola*, *P. ambigua*, *Parmelia sulcata* and *Cladonia coccifera*.

Total lichen cover showed a modest, but significant, correlation with heavy metal and S concentrations in birch leaves (Fig. 4). The chemical data reveal that all plots with high concentrations of heavy metals in leaves were lacking epiphytic



**Fig. 5.** Mean concentrations of Ni, Cu and S in birch leaves plotted against distance from Nickel. (a) Ni concentrations. Note that the plot with the third highest Ni concentration is situated between the smelters at Nickel and Zapolyarniy and is exposed to pollution from both the smelter in Nickel and the ore roasting facility in Zapolyarniy.  $R^2$  without this plot is 0.71. (b) Cu concentrations.  $R^2$  without the same plot as in a is 0.65. (c) S concentrations. Error bars, when larger than symbols, show  $\pm 1$  S.E.  $n = 5$ .

lichens (Fig. 4a–d). The lichen-containing plot with the highest concentrations of heavy metals had mean values of 31 ppm Ni in birch leaves (Fig. 4a), and 17 ppm Ni in bilberry leaves (Fig. 4b). The same values for Cu were 14 ppm (Fig. 4c) and 12 ppm (Fig. 4d), respectively. These values can be regarded as threshold levels for survival of epiphytic lichens in this area. Straight lines drawn from the plot point with maximum lichen cover to the first point with no lichen cover may be used to represent potential maximum lichen cover with varying concentrations of Ni and Cu (Fig. 4a–d). For S, it was not possible

to show such a potential maximum lichen cover line (Fig. 4e and f). Ni, Cu and S concentrations in the A horizon of the soil show similar relationships to epiphytic lichen distribution as do concentrations in bilberry leaves (not shown).

The results confirm that there still was quite an extensive zone around the smelter at Nickel without any epiphytic lichen cover. No plots closer than 9.8 km from Nickel had any lichen cover. All plots with high concentrations of heavy metals were situated close to Nickel or between Nickel and Zapolyarniy (Fig. 5) and were lacking epiphytic lichens. Ni concentrations in leaves were more closely correlated with distance from Nickel than Cu and S concentrations (Fig. 5a–c).

However, there were plots with low concentrations of heavy metals that also had a very low lichen cover (Fig. 4a–d). There were eight plots with Ni concentrations in birch leaves lower than 31 ppm (threshold value, *see* above) and with lichen cover < 10%. These plots were situated from 11.1 km to 31.9 km from Nickel, including four southern plots, two western plots, and two northern plots (Fig. 1). They also differed much in elevation (from 30 m to 268 m); hence, they were climatically diverse. Also among the plots with no lichen cover, there was much variation in distance from Nickel (1.9–19.4 km), and elevation (65–230 m). Thus, the linear relationships between lichen cover and distance from Nickel and elevation were low ( $R^2 = 0.07$  and 0.08, respectively, not shown). The plots with no lichen cover were situated either north or south of Nickel, whereas all plots west of Nickel had lichen cover (Fig. 1).

## Discussion

Concentrations of Ni, Cu and S in birch and bilberry leaves and in the A horizon of the soil from the epiphytic lichen desert zone around Nickel strongly indicate that air concentrations are much too high to sustain viable populations of epiphytic lichens. Based on the data from the plots with lichen cover, recolonization cannot be expected within the lichen desert zone until Ni and Cu concentrations drop below estimated threshold levels of birch leaves (*viz.* ca. 31 ppm Ni and ca. 14 ppm Cu). The epiphytic lichen

desert zone on the Norwegian side of the border seems to have been much larger at the beginning of the 1980s (Bruteig 1984), thus our results indicate that the outer parts of the early 1980s desert zone have experienced sufficient pollution reductions to allow recolonization of epiphytic lichens. However, our lichen cover studies were not undertaken on the same trees and plots as were used by Bruteig (1984), hence we cannot say with full certainty that the trees studied by us were lacking lichens in the 1980s, although emission loads during the 1970s and 1980s in combination with Bruteig's studies certainly indicate that this was the case.

As seen in Fig. 4a–d, the plots with low lichen cover and low heavy metal concentrations (points closest to origo) contributed considerably to reducing the relationships between these parameters. These plots with low lichen cover had differing climate and physiognomy. The two northern plots were situated at low elevation almost 30 km north of Nikel, but close to the sea. There are two factors that may explain the low lichen cover at these two plots. The predominant wind direction from Nikel is from south-southwest, leading to high SO<sub>2</sub> concentration in the air north and northeast of Nikel, also as far north as these two plots are situated (Bekkestad *et al.* 1995, Hagen *et al.* 2005). Measurements of precipitation chemistry from the adjacent localities Karpdalen and Karpbukta in the period 1991–2000 showed significantly higher concentrations of sulphate (SO<sub>4</sub>) than in background stations in northern Norway, which could be due to emissions from the smelters, but marine contributions of sulphate in this area may also be substantial (Hagen *et al.* 2005). Since Ni and Cu are present in aerosols, they are deposited close to the factory (cf. Bekkestad *et al.* 1995), and the concentrations of Ni and Cu in precipitation at Karpdalen and Karpbukta were much lower than at Svanvik (Hagen *et al.* 2005), which is located closer to the smelter in Nikel. Thus, the reduction of heavy metal concentrations with distance from pollution source is much more abrupt than the reduction of SO<sub>2</sub> concentrations. Increased exposure to SO<sub>2</sub> can cause chlorophyll degradation in lichens by exerting deleterious effects on the symbiosis between the fungal partner and its photobiont (LeBlanc and Rao 1973). The prox-

imity to the open sea and the north-facing terrain make these plots exposed to lower temperatures and stronger winds than further inland. These are factors that can restrain lichen establishment and growth in northern boreal forests (Bruteig 1998, Aamlid and Skogheim 2001). Thus, the low lichen cover at these two plots may result from a combination of SO<sub>2</sub> pollution and a harsh coastal climate.

The four plots south of Nikel with low lichen cover were all situated 16.4 km to 31.9 km from the factory, and they were all at 213 m elevation or higher. Thus, they were situated close to the tree line, which also experienced low temperatures and strong winds. Thus, the harsh climate certainly contributes to keeping the lichen cover low at these plots. An additional explanatory factor is related to air movement phenomena. Arctic cold air masses, especially during summer, can create inversions by overriding the warmer layers at ground level and forcing the warm, polluted air along the river drainage basins (Odasz-Albrigtsen *et al.* 2000). Prevailing high-elevation cold winds can force the lower-elevation, warm and polluted air from Nikel towards the 200 m to 400 m high hills and mountains surrounding Nikel (Odasz-Albrigtsen *et al.* 2000), where these plots were located. When this warm air rises, most of its heavy metals in aerosols will be dropped off and not transported further (Bekkestad *et al.* 1995). Thus, these episodes with high concentration of SO<sub>2</sub> can have deleterious effects on the epiphytic lichens at these southern plots, but without being reflected in the heavy metal concentrations of birch leaves. At similar high-elevation sites around Nikel, the vitality of birch and bilberry, expressed as photosynthetic efficiency, is lower than at distant, unpolluted reference sites (Odasz-Albrigtsen *et al.* 2000). This may indicate that these hillsides actually were exposed to air with high levels of SO<sub>2</sub> but low in heavy metals.

The two last plots with low lichen cover were situated 11.1 km and 16.3 km west of Nikel, thus quite close to the present lichen desert zone, but still they had relatively low heavy metal concentrations. They were situated within the 1980s lichen desert zone that experienced very high SO<sub>2</sub> concentrations in 1974–1984 (Bruteig 1984, Bekkestad *et al.* 1995). However, SO<sub>2</sub> con-

centrations in this area were later much reduced (Hagen *et al.* 2000), from ca.  $50 \mu\text{g m}^{-3}$  in summer 1975 to ca.  $15 \mu\text{g m}^{-3}$  in summer 1998 (*see* also maps reproduced in Tømmervik *et al.* 2003). The low recolonization of trunks at these plots probably spring from limited immigration of lichen diaspores after the long period of severe pollution. Since practically all epiphytic lichens died out during the severe pollution period (*cf.* Bruteig 1984, Aamlid 1992, Aamlid and Venn 1993), and the nearest surviving populations probably had reduced vitality and low production of sexual and asexual diaspores (*cf.* Mikhailova and Scheidegger 2001), the distance to the nearest unaffected propagating lichen populations was quite large, resulting in a very low number of immigrating diaspores. Dispersal of epiphytic lichens by means of diaspores can be quite slow and short-ranging (*e.g.* Armstrong 1990, Walser *et al.* 2004). Intermediate levels of pollutants still occurring at these plots also after the period of severe air pollution probably further reduced the success of recently immigrated diaspores (*cf.* Mikhailova 2002).

Despite the low correlations found between epiphytic lichen cover and heavy metal concentrations in plant leaves and soil, the use of Ni and Cu accumulation in birch leaves may be a useful technique for assessing the likelihood for survival of epiphytic lichens if transplanted to various areas in the lichen desert zone, and for evaluating required emission reductions for permitting recolonization by epiphytic lichens. This assumption is based on the fact that there is a good relationship between mean maximum lichen cover and heavy metal concentrations in birch leaves for plots at low elevations, as visualized by the “potential maximum lichen cover lines” in Fig. 4a–d. These straight lines are merely two-point lines, and should therefore not be given too much emphasis. Nevertheless, they can be used as rough estimates of how extensive lichen cover we can expect, under the regionally best climatic conditions, given that we know the concentrations of Ni and Cu in birch leaves. The low intra-plot variation in Ni and Cu values (*see* error bars in Figs. 3c and 4a) of birch leaves further support their applicability, both as surrogates for lichens in biomonitoring surveys, and as indicators of threshold values for heavy

metal tolerance in epiphytic lichens. However, as indicated above, at some distance from the factory, accumulation of Ni and Cu in plant tissue and soil do not correspond with annual mean  $\text{SO}_2$  concentrations in the air, and in such areas their applicability is apparently low. Moreover, as shown above, the regional variability in lichen cover induced by climatic variation must also be evaluated.

Not surprisingly, the highest concentrations of Ni, Cu and S were found closest to the Nikel smelter and the Zapolyarniy ore roasting facility, and reductions with distance show logarithmic correlations (Fig. 5). Our birch data revealed the same pattern as shown by Kozlov (2005) who also measured a decrease in concentration of Cu and Ni in birch leaves with increasing distance from smelters in another area at Kola Peninsula. Multiyear mean values peaked at 6.6 km south of the smelter, where they were 20–25 times higher than in the most distant study site 63 km from the smelter (Kozlov 2005). In a gradient study from Nikel, Steinnes *et al.* (2000) found maximum levels of 120 ppm Ni and 50 ppm Cu in birch leaves 7 km from the Nikel smelter, whereas the lowest concentrations of Ni (5 ppm) and Cu (5 ppm) were found 44 km from the smelter. Thus, maximum levels did not change considerably from the time Steinnes *et al.* (2000) had done their sampling (1991) until our sampling was undertaken (2000), despite observed reductions in pollution emissions in this period. Steinnes *et al.* (2000) and Aamlid *et al.* (2000) showed that birch leaves had higher Ni and Cu concentrations than bilberry leaves, which our measurements also showed (Fig. 4). Birch leaves are stickier than bilberry leaves, and hence air particulates more easily attach to birch leaves (Kozlov *et al.* 2000, Kozlov 2005). This may be the primary reason for the differences in heavy metal concentrations between birch and bilberry leaves.

To conclude, we found significant relationships between epiphytic lichen cover on birch stems and the heavy metal concentrations in birch and bilberry leaves. The deviant points can be explained by increased differences in accumulation of heavy metals and the dispersal of  $\text{SO}_2$  with increasing distance from pollution source, and by climatic variability within the

study area. Epiphytic lichens are shown to grow within an area that probably was a lichen desert zone during the 1980s (Bruteig 1984), but the concentrations of Ni, Cu and S in plant leaves and soil strongly indicate that the area closest to the smelters will not experience any recolonization of epiphytic lichens unless pollution emissions are strongly reduced, and maintained at a low level.

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