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Long distance nitrogen air pollution effects on lichens in Europe

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Abstract: The epiphytic lichen flora of 25 European ICP-IM monitoring sites, all situated in areas remote from air pollution sources, was statistically related to measured levels of SO_2 in air, NH_4^+, NO_3^- and SO_4^{2-} in precipitation, annual bulk precipitation, and annual average temperature. Significant regression models were calculated for eleven acidophytic species. Several species show a strong negative correlation with nitrogen compounds. At concentrations as low as 0·3 mg N l^{-1} in precipitation, a decrease of the probability of occurrence is observed for *Bryoria capillaris*, *B. fuscescens*, *Cetraria pinastri*, *Imshaugia aleurites* and *Usnea hirta*. The observed pattern of correlations strongly suggests a key role of NH_4^+ in determining the species occurrence, but an additional role of NO_3^- cannot be ruled out. Some species show a distinct response to current levels of SO_2 as well. It may be concluded that long distance nitrogen air pollution has strong influence on the occurrence of acidophytic lichen species.

Key words: acidophytic lichens, air pollution, ammonia, nitrogen, precipitation chemistry.

Introduction

For many years sulphur dioxide (SO_2) air pollution was regarded as the most important cause of decline of epiphytic lichens in large parts of Europe (Barkman 1958; Hawksworth & Rose 1970). During the last few decades the levels of SO_2 have dropped considerably, and, as a consequence, numerous species have become common again in the populated areas of western Europe (Hawksworth & McManus 1989; van Dobben 1993; van Herk *et al.* 2002). At the same time many species preferring an acid bark have become diminished (van Herk & Aptroot 1998); some species such as *Hypogymnia physodes* even showed a considerable country-wide decrease in abundance over the same period in the Netherlands (van Herk 2001), this being most pronounced in areas with a high cattle density. High levels of nitrogen compounds in air, viz. ammonia (NH_3), appeared to lead to a complete disappearance of acidophytic species, resulting in communities dominated by nitrophytic species (van Herk 1999). Such a shift in species composition was shown to be governed by a high bark pH due to the alkaline properties of NH_3 rather than to an increased availability of nitrogen compounds (de Bakker & van Dobben 1988; van Herk 2001). The pH of *Quercus* bark is positively correlated with the NH_3 concentration in air, resulting in an upward shift of approximately two pH units at high NH_3 levels (van Herk 2001).

Virtually nothing is known so far about long distance effects of the emission of nitrogen compounds on lichens. Although a relatively large proportion of emitted NH_3 is known to be deposited as NH_3 close to the source (*c.* 10% within a 100m downwind distance), it is a common misconception that NH_3 emissions do not disperse very

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widely. Calculations have shown that approximately 25% of emitted NH_3 reaches a downwind distance of 100–1000 km as ammonium (NH_4^+), and is deposited mainly in wet precipitation (Asman & van Jaarsveld 1990). Deposition of unaltered NH_3 reaches only 50 km downwind due to an atmospheric chemical reaction of NH_3 into $(NH_4)_2SO_4$ (ammonium sulphate) or NH_4NO_3 (ammonium nitrate). At greater distances from the source virtually no NH_3 will be left. Deposition of NH_4^+ does not increase the pH of bark, in contrast to the deposition of NH_3. However, when NH_4^+ is nitrified into NO_3^-, for example on mossy trunks, it might add considerably to acidification.

Several publications have appeared discussing concern about the lichen flora and vegetation of native woodlands in areas remote from pollution sources. Acid rain is often mentioned as a cause of the decline in (acid sensitive) *Lobarion* communities (Gauslaa 1985; Gilbert 1986). A reduced bark pH, attributable to wet acidic deposition, has been found in several *Lobarion* studies. There is increasing evidence that some of the floristic changes attributed to wet acidic deposition may be caused by nitrogen (Farmer *et al.* 1992). Farmer (1997) more particularly indicates ammonia as a possible cause of decline of *Lobaria* at certain sites in Britain.

Acidophytic lichen species and communities in the boreal coniferous zone have been subject to many air pollution studies, usually focusing on SO_2. In some studies, nitrogen was considered as well. Holopainen (1983) found ultrastructural changes in *Bryoria capillaris* and *Hypogymnia physodes* near a fertilizer plant and a pulp mill in Finland, mainly attributable to SO_2 and nitrogen compounds. Bruteig (1993), using *H. physodes* as a monitor for nitrogen and sulphur deposition throughout Norway, observed effects of nitrogen only relatively close to the sources of emission. Bråkenhielm & Qinghong (1995) investigated 15 reference areas in Sweden (some of these are also considered in this study) and found a mean sensitivity index of lichens increasing from south to north and from west to east, concomitant with decreasing deposition levels of nitrogen and sulphur. Due to highly correlated environmental variables, it was not possible to conclude what factor was responsible for the observed distribution pattern of lichens. None of these studies describe a clear long distance effect of nitrogen compounds.

No long distance effects of nitrogen compounds have been reported from the coniferous montane belt in Central Europe. Ruoss (1999) found the occurrence of nitrophytic lichens to be correlated with agricultural land use in Switzerland, but effects on acidophytic lichens in more distant areas were not mentioned.

In the Netherlands, *Bryoria fuscescens* can serve as a good example of a species that has retreated gradually to areas remote from ammonia emitting sources. This species is nearly extinct (Aptroot *et al.* 1998). Barkman (1958) found it to be a fairly common epiphyte, but practically all epiphytic occurrences disappeared during the 1960s and 70s. The few erratic occurrences on *Pinus* stumps in inland dune areas disappeared in the 1990s, and terrestrial occurrences in coastal dunes (Ketner-Oostra 1972) have nearly vanished even more recently. Changes in these lichen-rich dune grasslands were attributed to nitrogen deposition (Ketner-Oostra & van der Loo 1998). This is remarkable because these sites are at least 70 km (not downwind) from large-scale nitrogen (NH_3 and NO_x) sources.

The quantities of nitrogen oxides (NO_x), mainly emitted by road traffic, have increased considerably in many European countries. Nash (1976) considered NO_2 unlikely to be an important lichen phytotoxin since fumigation experiments showed damage only at very high levels. However, in a few recent urban studies (Loppi *et al.* 1996; Davies *et al.* 2002) effects from NO_x are reported.

The aim of the present study is to find out whether lichen species are affected by nitrogen or other air pollutants in areas only subject to long range air pollution. This has been done by statistically comparing the composition of the lichen flora of such sites distributed over large parts of Europe, to *in situ* measured levels of air pollution (SO_2) and concentrations in precipitation (NH_4^+, NO_3^-, SO_4^{2-}). Differences in the amount of annual bulk precipitation and annual average temperature are also considered as factors of importance in governing lichen species composition.

This study was performed within the framework of the *International Cooperative Programme on Integrated Monitoring of Air Pollution Effects* (ICP-IM). Since 1989, ICP-IM has been part of the *Effects Monitoring Strategy* under the UN ECE Convention on *Long Range Transboundary Air Pollution*. Twenty-three countries were involved in this programme up to 2000, with 70 monitoring stations in Europe and Canada. One of the biological monitoring programmes is the subprogramme *Trunk Epiphytes*, which comprises 37 monitoring stations in Europe. The availability of a wide range of chemical, physical and biological data in ICP-IM monitoring stations provided the opportunity to study the effects of long distance effects on epiphytic lichens.

Material and Methods

The observations of lichens were made on trunks of living trees between 50 and 200 cm above ground level, either by the line cover, point frequency or the species list method. These methods, as well as the monitoring and analytical methods of the chemical variables, are described in the Manual for Integrated Monitoring (UN ECE, 1998) and are available on the internet.

Data gathered in ICP-IM were received from the ICP-IM Programme Centre of the Finnish Environment Institute in Helsinki. Observations on *Trunk Epiphytes* were made at 37 monitoring stations. Participating countries were approached for more detailed information on the epiphytic data. Besides the data of the ICP-IM database, data of monitoring stations of the European Monitoring and Evaluation Programme (EMEP) were used to fill some gaps in the physico-chemical data. It is important to stress that the ICP-IM sites are all specifically selected to occur in natural areas, remote from local sources of air pollutant emissions. Variables that were studied in relation to the occurrence of epiphytic lichens are: the annual average temperature, the annual amount of precipitation, the concentrations of SO_4^{2-}, NH_4^+ and NO_3^- in precipitation, and the concentrations of SO_2 in air. Twenty-five stations (see Fig. 1, Table 1) have appropriate data to perform the statistical analysis explaining the occurrence of lichen species. For some of these stations, time series were available spanning up to four sampling events in a period of 20 years. Adjacent years of observation were excluded from the analysis to prevent pseudo-replication in the lichen data.

To be able to relate the occurrence of species to the environmental factors in a statistically valid way, the physico-chemical variables should not demonstrate too much correlation. The correlation matrix of the scalar variables is given in Fig. 2. One correlation was found to be quite high (0·81 for NH_4^+/NO_3^-). It was decided that for this data analysis no predictor series should be dropped from the dataset. Some implications will be discussed below.

After preparing the data and making the above data summaries, the occurrences (absence/presence) of 82 epiphytic lichen species were subjected to stepwise logistic General Linear Modelling (GLM) in the statistical programme package S-Plus with the 6 predictors (pred.) presented in Table 1 and Fig. 2. The GLM has the general formula in which p is the probability of occurrence of a particular species:

$$\log\left(\frac{p}{1-p}\right)=a+c_1\times pred._1+\cdots+c_x\times pred._x+d_1\times(pred._1)^2+\cdots+d_x\times(pred._x)^2$$

The quality of a GLM-regression is given as the difference between the deviance (scaled Error Sum of Squares) of the null model ($\log\left(\frac{p}{1-p}\right)=a$) and the deviance of the calculated model with predictors. This so-called explained deviance is Chi-square distributed with the number of predictor variables as the degrees of freedom (df). The same holds for adding single terms to the model. The added explained deviance is of course equal to the difference in the explained deviance of the model before and after addition of the term with the degrees of freedom being one. The probability of the Chi-square for term additions as well as for the overall model is equal to the probability that the explained deviance is caused by random variation. The GLM is formulated to add automatically and iteratively significant predictor terms ($p_{\text{explained deviance}}<0{\cdot}05$) to the model.

It was not possible to calculate a valid model for a certain species in the following cases: 1, the species records are scarce (less than 8 out of 49 data series); 2, the species is generally present (in more than 41 out of 49 data series); 3, none of the predictors adequately explains the occurrence of the species. Only overall models with at least 15% explained deviance were considered as valid.

Results

Physico-chemical data

Measured concentrations of ammonium N in precipitation (see Table 1) were

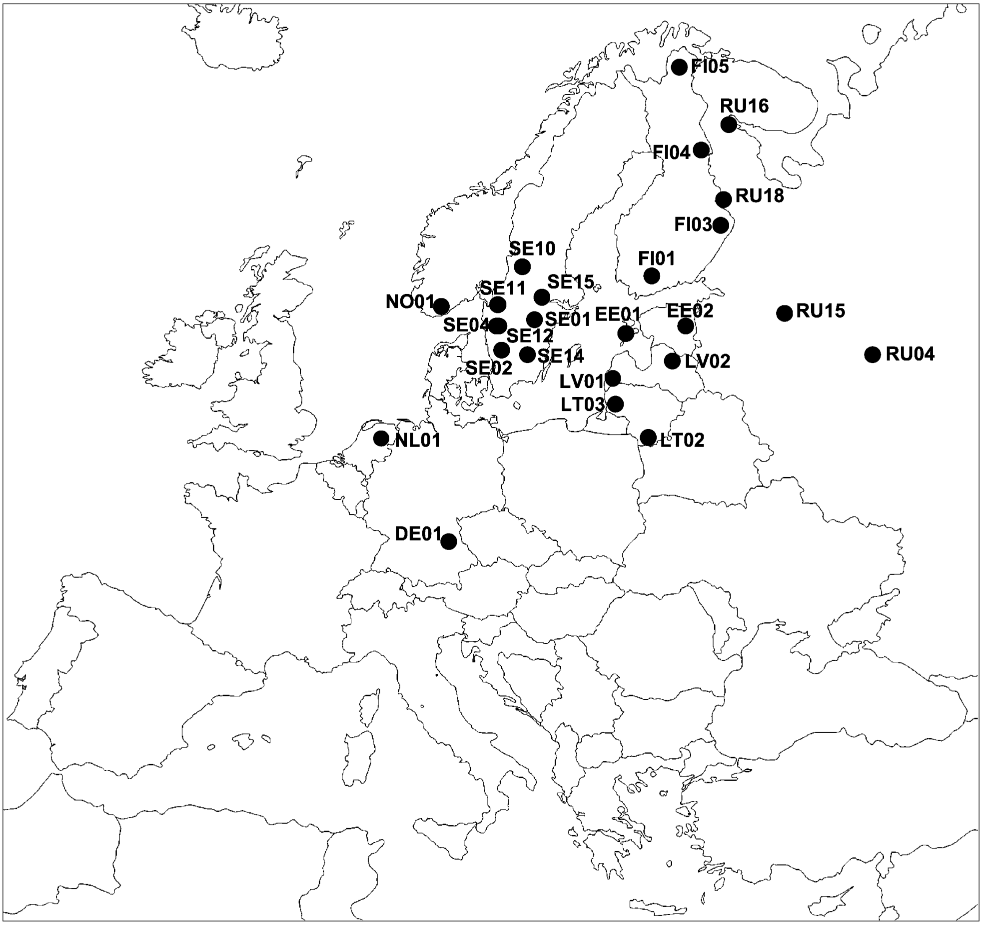


FIG. 1. Locations of ICP-IM monitoring stations in Europe where the observations were taken. Codes are explained in Table 1.

lowest at Vuoskojärvi (0·02–0·04 mg l^{-1}) and Pesosjärvi (0·06–0·09 mg l^{-1}), both in Northern Finland, while the highest ammonium levels occurred in Zemaitija, Lithuania (1·1–1·3 mg l^{-1}) and Lheebroekerzand, Netherlands (0·9–2·3 mg l^{-1}). The spatial pattern of nitrate N in precipitation is very similar to that of ammonium (Fig. 2, $r = 0{\cdot}81$). Stations with lowest and highest values are only slightly different being Kamalahti in Russia (0·10 mg l^{-1}) and again Vuoskojärvi (0·08–0·15 mg l^{-1}) with the lowest values, and again Zemaitija (0·63–0·87 mg l^{-1}) and Lheebroekerzand (0·53–0·93 mg l^{-1}) with the highest values. There are rather strong correlations between sulphate and ammonium ($r = 0{\cdot}74$) and sulphate and nitrate ($r = 0{\cdot}67$). Lowest sulphate S values occurred at Pesosjärvi (0·23–0·47 mg l^{-1}) and Vuoskojärvi (0·28–0·32 mg l^{-1}), while again Zemaitija (1·3–2·1 mg l^{-1}) and Lheebroekerzand (0·8–2·5 mg l^{-1}) showed the highest values in precipitation.

The spatial pattern of sulphur dioxide in air appears quite different from most other predictors, a relatively high correlation was only found between SO_2 and SO_4^{2-} in precipitation ($r = 0{\cdot}61$). Lowest concentrations of sulphur dioxide S were measured at Kindlahöjden, Sweden (0·19 µg m^{-3}) and Forellenbach, Germany (0·23–0·36 µg m^{-3}) and highest levels at Zemaitija (1·5–3·5 µg m^{-3}) and Lheebroekerzand (1·1–5·8 µg m^{-3}).

The annual average temperature appeared to be correlated positively with all predictors for air pollution, most obviously with ammonium ($r = 0{\cdot}68$), nitrate ($r = 0{\cdot}65$) and sulphate ($r = 0{\cdot}57$). Strong correlations between the annual amount of precipitation and predictors for air pollution were not found. The amounts of precipitation ranged between *c.* 270 mm yr^{-1} at Kamalahti, Russia, and *c.* 1600 mm yr^{-1} at Birkenes, Norway or *c.* 1700 mm yr^{-1} at Forellenbach.

Lichens

Using GLM it was possible to determine significant models for fifteen lichen species out of the original 82 species. Four out of the fifteen species have significant terms for temperature and/or precipitation only; these species will not be discussed. The remaining eleven species have significant terms for either NO_3^- or NH_4^+ in precipitation, SO_2 in air, or a combination of these terms with or without temperature. Table 2 gives the frequency of occurrence of the eleven species in the data set, the number of significant predictor terms, the explained deviance and the overall significance, respectively.

All eleven species for which a valid model was calculated have a wide natural distribution throughout Europe. The species considered are therefore not limited by a restricted range, except for *Phlyctis argena* which reaches north only into southern Scandinavia, probably due to its preference for warmer conditions. All species are known to show a preference for acid bark (Wirth 1991) and can therefore be considered as 'acidophytic'.

Among the eleven species with a valid model, five species show significant correlations with NH_4^+ in precipitation (Fig. 3C), three species with NO_3^- in precipitation (Fig. 3B) and five species with SO_2 in air (Fig. 3D). A valid model in which SO_4^{2-} is present could not be calculated for any of the species. Eight species show a positive response to the annual mean temperature (Fig. 3A); none of the eleven species shows a response to the annual bulk precipitation. All except two species appeared to show a response to more than one predictor series (Table 2), often the annual average temperature and one predictor for air pollution.

The majority of the eleven species considered show a strong negative correlation with nitrogen compounds. At very low concentrations (*c.* 0·3 mg N l^{-1} of either NH_4^+ or NO_3^- in precipitation), a decrease of the probability of occurrence can be observed for *Bryoria capillaris*, *B. fuscescens*, *Cetraria pinastri*, *Chaenotheca ferruginea*, *Imshaugia aleurites* and *Usnea hirta*.

Four species, viz. *Bryoria capillaris*, *B. fuscescens*, *Imshaugia aleurites* and *Chaenotheca ferruginea* appear to be correlated negatively with NH_4^+ in precipitation. They all show a gradual decrease of their probability of occurrence from normally present at near-zero levels of NH_4^+ to absence at *c.* 1·0 mg N l^{-1} or more (Fig. 3C). *Lecanora pulicaris* shows a different behavour in its positive correlation with NH_4^+; from levels of *c.* 1·0 mg N l^{-1} its probability of occurrence increases to being quite common at *c.* 2·3 mg N l^{-1}.

Two species, viz. *Cetraria pinastri* and *Usnea hirta*, correlate negatively with NO_3^- in precipitation. The first is present only at levels <0·2 mg N l^{-1}; the second shows a gradual decrease within the whole measured range of NO_3^- (Fig. 3B). Surprisingly, *Bryoria fuscescens* shows a positive correlation with NO_3^-; it gradually increases within the whole measured range. *Chaenotheca chrysocephala*, *Cladonia digitata*, *Evernia prunastri*, *Lecanora pulicaris* and *Phlyctis argena* appear to correlate negatively with SO_2 in air (Fig. 3D). All species except *Bryoria fuscescens*, *Cetraria pinastri* and *Imshaugia aleurites* show a positive correlation with the annual average temperature (Fig. 3A).

TABLE 1. *Annual average temperature, annual bulk precipitation, concentrations of* SO_4^{2-}, NH_4^+ *and* NO_3^- *in precipitation, and concentrations of* SO_2 *in air for 25 ICP-IM monitoring stations in Europe where physico-chemical data were collected. For most of the stations several observation series of non-adjacent years are included*

Station code	Station name	Country	Year	Annual mean temperature °C	Precipitation mm yr^{-1}	SO_4^{2-} in precipitation mg S l^{-1}	NO_3^- in precipitation mg N l^{-1}	NH_4^+ in precipitation mg N l^{-1}	SO_2 in air μg S m^{-3}
DE01	Forellenbach	Germany	1992	6·5	1136	0·71	0·53	0·59	0·36
DE01	Forellenbach	Germany	1995	5·9	1696	0·56	0·43	0·38	0·23
EE01	Vilsandi	Estonia	1994	8·7	741	0·96	0·29	0·29	0·25
EE01	Vilsandi	Estonia	1996	8·7	325	1·15	0·58	0·70	0·79
EE01	Vilsandi	Estonia	1998	8·7	507	0·68	0·37	0·31	0·33
EE02	Saarejärve	Estonia	1995	5·8	336	1·76	0·26	0·21	0·24
EE02	Saarejärve	Estonia	1998	5·8	640	0·76	0·28	0·34	0·54
FI01	Valkeakotinen	Finland	1990	4·8	667	0·52	0·26	0·22	1·46
FI01	Valkeakotinen	Finland	1994	3·4	563	0·55	0·28	0·11	1·07
FI01	Valkeakotinen	Finland	1997	4·1	547	0·36	0·25	0·10	0·57
FI03	Hietajärvi	Finland	1990	2·9	469	0·59	0·31	0·16	1·02
FI03	Hietajärvi	Finland	1994	1·8	632	0·43	0·23	0·09	0·50
FI03	Hietajärvi	Finland	1997	2·0	574	0·30	0·18	0·07	0·86
FI04	Pesosjärvi	Finland	1991	−0·5	571	0·47	0·17	0·09	0·61
FI04	Pesosjärvi	Finland	1994	−0·2	486	0·30	0·17	0·06	0·49
FI04	Pesosjärvi	Finland	1998	−2·1	649	0·23	0·14	0·06	0·65
FI05	Vuoskojärvi	Finland	1991	−0·8	378	0·32	0·08	0·02	0·98
FI05	Vuoskojärvi	Finland	1994	−1·1	314	0·28	0·15	0·04	0·49
FI05	Vuoskojärvi	Finland	1997	−1·1	321	0·31	0·10	0·04	0·56
LT02	Dzukija	Lithuania	1993	7·7	817	1·01	0·39	0·95	1·92
LT02	Dzukija	Lithuania	1996	7·7	377	0·89	0·50	0·85	3·17
LT03	Zemaitija	Lithuania	1994	8·4	469	2·09	0·87	1·11	3·48
LT03	Zemaitija	Lithuania	1996	8·4	550	1·25	0·63	1·30	1·47
LV01	Rucava	Latvia	1994	6·9	791	1·06	0·44	0·71	0·86
LV01	Rucava	Latvia	1998	6·6	906	0·66	0·64	0·76	0·40
LV02	Zoseni	Latvia	1994	5·1	811	1·53	0·38	0·93	0·91

Table 1 *Continued.*

Station code	Station name	Country	Year	Annual mean temperature °C	Precipitation mm yr^{-1}	SO$_4^{2-}$ in precipitation mg S l^{-1}	NO$_3^-$ in precipitation mg N l^{-1}	NH$_4^+$ in precipitation mg N l^{-1}	SO$_2$ in air µg S m^{-3}
LV02	Zoseni	Latvia	1998	5·0	865	0·54	0·40	0·41	0·60
NL01	Lheebroekerzand	Netherlands	1978	8·3*	575	1·91	0·84	1·16	5·76
NL01	Lheebroekerzand	Netherlands	1989	8·3*	685	2·47	0·93	2·31	3·06
NL01	Lheebroekerzand	Netherlands	1996	7·6	516	1·15	0·77	1·50	1·94
NL01	Lheebroekerzand	Netherlands	1998	7·1	1320	0·79	0·53	0·85	1·14
N001	Birkenes	Norway	1986	7	1613	1·23	0·75	0·77	0·75
RU04	Oka Terrace	Russia	1993	4	1403	0·43	0·10	0·61	0·66
RU15	Tayozhny Log	Russia	1994	5·3	707	0·91	0·18	0·27	1·57
RU16	Velikiy	Russia	1994	−0·1‡	407	0·54	0·14	0·13	1·40
RU18	Kamalahti	Russia	1993	2·2‡	269	1·65	0·10	0·22	1·65
SE01	Tiveden	Sweden	1987	4·1‡	734	1·42	0·67	0·69	2·91
SE01	Tiveden	Sweden	1992	4·1‡	779	1·07	0·68	0·58	1·19
SE02	Berg	Sweden	1988	5·1‡	1154	0·97	0·58	0·53	2·92
SE04	Gårdsjön	Sweden	1996	3·9	850	0·69	0·54	0·48	0·61
SE10	Tandövala	Sweden	1987	2‡	734	1·42	0·67	0·69	2·91
SE10	Tandövala	Sweden	1992	2‡	754	0·47	0·29	0·21	1·19
SE11	Tresticklan	Sweden	1987	3·9‡	891	0·77	0·39	0·33	2·91
SE11	Tresticklan	Sweden	1992	3·9‡	650	0·85	0·58	0·60	1·19
SE12	Svarted	Sweden	1987	3·9‡	999	1·02	0·51	0·48	2·91
SE12	Svarted	Sweden	1992	3·9‡	908	0·93	0·64	0·61	1·19
SE14	Aneboda	Sweden	1995	5·1	615	1·04	0·64	0·61	1·08
SE14	Aneboda	Sweden	1997	5·1	782	0·59	0·49	0·47	0·33
SE15	Kindlahöjden	Sweden	1998	4·1	897	0·45	0·34	0·28	0·19

*Long year average.

‡Grid estimate.

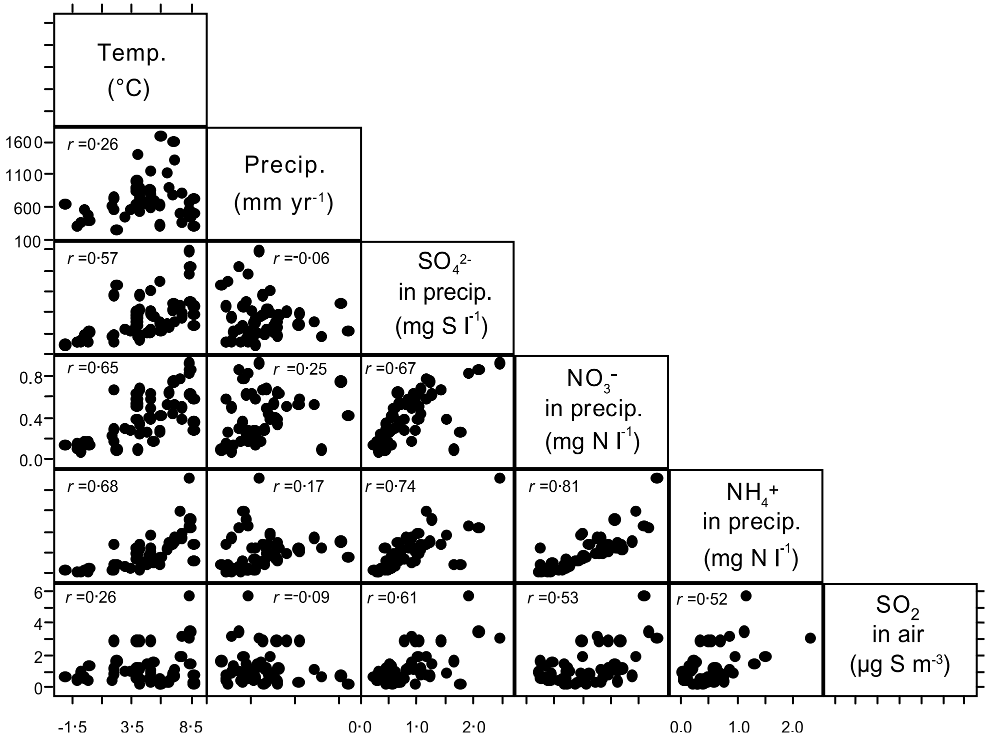


FIG. 2. Scatter plots for combinations of predictor variables, viz. annual average temperature, annual bulk precipitation, concentrations of SO_4^{2-}, NO_3^- and NH_4^+ in precipitation, and concentrations of SO_2 in air (expressed as °C, mm yr^{-1}, mg S l^{-1}, mg N l^{-1}, mg N l^{-1} and µg S m^{-3} respectively). Correlation coefficients (*r*) are shown.

TABLE 2. *Lichen species for which a valid regression model with either SO_4^{2-}, NO_3^- or NH_4^+ in precipitation, or SO_2 in air could be calculated, with their number of occurrences, the predictor terms, the explained deviance and the significance of the model. Nomenclature follows Purvis* et al. *(1992)*

Species	Number of occurrences	Predictors	Explained deviance %	Significance
Bryoria capillaris	14	Temp. & NH_4^+	25·41	$5{\cdot}83 \times 10^{-4}$
B. fuscescens	23	NH_4^+ & NO_3^-	37·12	$3{\cdot}46 \times 10^{-6}$
Cetraria pinastri	8	NO_3^-	27·71	$5{\cdot}09 \times 10^{-4}$
Chaenotheca chrysocephala	8	Temp. & SO_2	58·62	$2{\cdot}81 \times 10^{-6}$
C. ferruginea	10	Temp. & NH_4^+	37·77	$8{\cdot}56 \times 10^{-5}$
Cladonia digitata	8	Temp. & SO_2	45·42	$4{\cdot}99 \times 10^{-5}$
Evernia prunastri	9	Temp. & SO_2	41·68	$5{\cdot}88 \times 10^{-5}$
Imshaugia aleurites	21	NH_4^+	45·77	$3{\cdot}12 \times 10^{-8}$
Lecanora pulicaris	8	Temp. & SO_2 and NH_4^+	55·67	$2{\cdot}18 \times 10^{-5}$
Phlyctis argena	8	Temp. & SO_2	39·90	$1{\cdot}66 \times 10^{-4}$
Usnea hirta	12	Temp. & NO_3^-	30·50	$2{\cdot}44 \times 10^{-4}$

Discussion

A significant model with one or more terms for air pollution could be calculated for only eleven species out of the original 82 species. This small number is partly due to the fact that, for the calculations, only records of absence or presence of species could be used. The abundance data collected, either as line cover or point frequency, appeared to be too heterogeneous for a proper comparison between the stations. Data based on different tree species needed to be aggregated as well since separate datasets based on only one tree species appeared too small for statistical treatment. Between the stations, however, the data are comparable because in all cases complete species lists are available.

In all, 49 data series from 25 stations have been used, i.e. often several data series (several non-consecutive years) per station. In all cases, these data series are grossly independent; thus the species and abiotic data concerned refer only to records made during that particular year.

Approximately half of the eleven species considered show a distinct negative correlation with NH_4^+ or NO_3^- in precipitation. However, because of the very high correlations between these two predictors (Fig. 2) strong effects cannot be assigned unequivocally to one single predictor variable. Care must be taken when the occurrence of a particular species is explained, for example the negative correlation between the abundance of *Cetraria pinastri* and the concentration of NO_3^- may eventually be reduced to NH_4^+ or the reverse may be applicable to, for example, *Imshaugia aleurites*. A response to both NO_3^- and NH_4^+ may be the case as well. The observed positive correlation of *Bryoria fuscescens* with NO_3^- is most probably an artefact due to its strong negative correlation with NH_4^+ combined with the strong correlation between NO_3^- and NH_4^+.

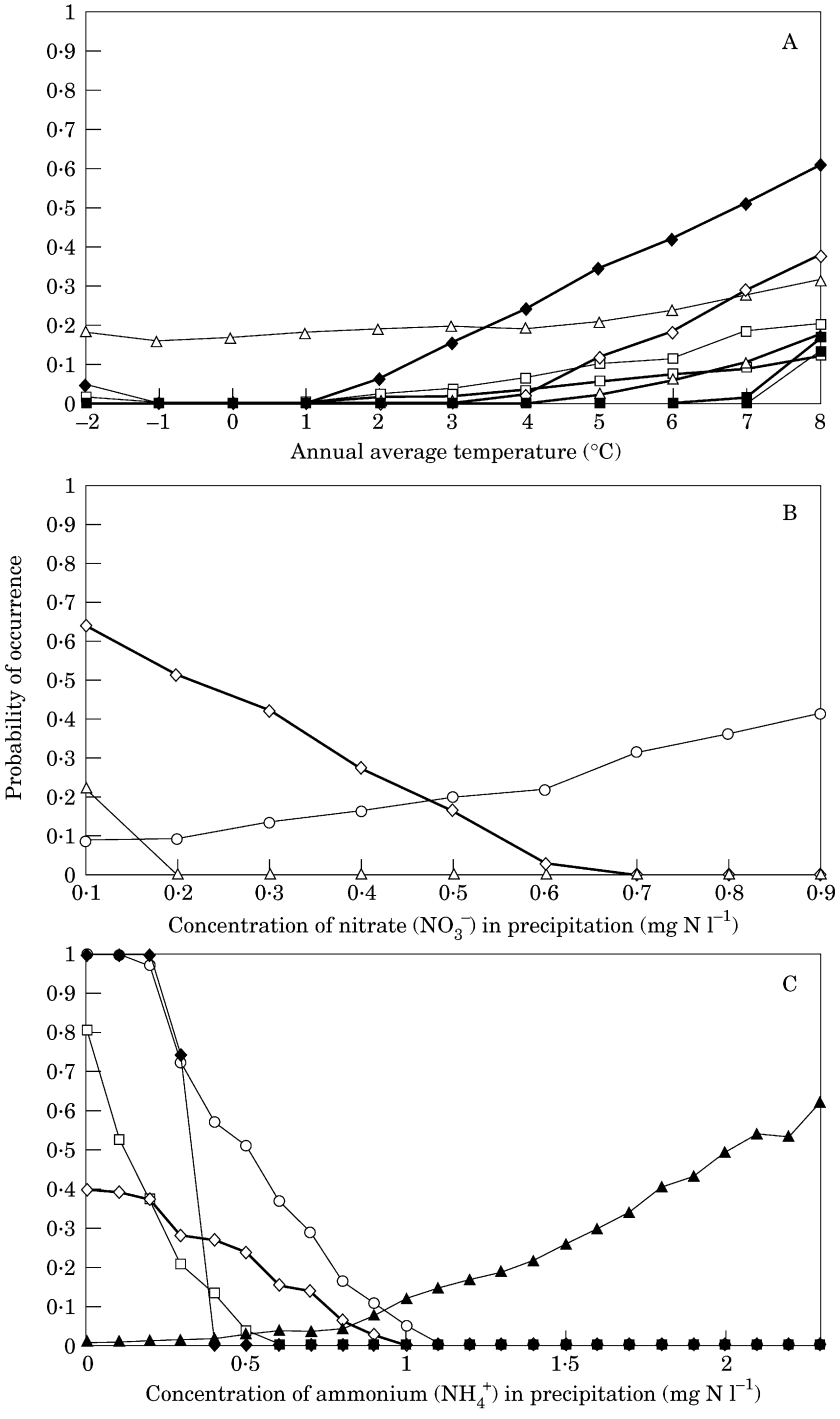
We considered dropping one predictor variable (e.g. NO_3^-) from the dataset, but this would not have solved this problem, as significant fits would then have been assigned to a 'second best' predictor variable (i.e. NH_4^+). Nevertheless, the observed pattern of correlations strongly suggests a key role of NH_4^+ and a possible additional role of NO_3^- in determining the distribution of a range acidophytic species.

Surprisingly, only five species considered show a distinct response to SO_2. Present day species distribution patterns are not always correlated with the current concentration of SO_2 in the atmosphere. However, historic levels of SO_2 (not necessarily in the same spatial patterns as at present) may still be important for the present species distribution as the European lichen vegetation might not have recovered fully from the effects of SO_2 during the past 30 years.

The annual amount of precipitation correlates with none of the eleven species, although it was expected that at least some of them (e.g. *Bryoria capillaris*, *B. fuscescens* and *Cetraria pinastri*, cf. Wirth 1991) would have shown a preference for high levels of precipitation.

It may be concluded that long distance transport of nitrogenous air pollution is important in determining the occurrence of acidophytic lichen species and constitutes a threat to natural populations that has so far been strongly underestimated. There are three principal possible explanations for the observed sensitivity to nitrogen compounds of acidophytic species: 1, sensitive species react to changes of bark pH; 2, species react readily to increased nitrogen (NH_4^+ and/or NO_3^-) content of precipitation or bark; 3, species react to increased growth of other epiphytes, like algae, mosses, or other lichens.

It seems unlikely that an increased bark pH due to adsorption of NH_3 explains all or most of the observed changes. The majority of the monitoring stations are probably outside the reach of substantial amounts of NH_3. Locally, however, an increase of the pH may be significant, for example at Lheebroekerzand in the Netherlands, which is one of the few stations where nitrophytic species such as *Xanthoria polycarpa* and *Physcia tenella* became common during the last decade (especially on twigs and branches). Kermit & Gauslaa (2001) reported an unusually high pH of canopy twigs in an area in central Norway. It is known that NH_3 may cause features as described by Kermit and Gauslaa (e.g. dominance of *Parmelia*


A
1
0·9
0·8
0·7
0·6
0·5
0·4
0·3
0·2
0·1
0
–2
–1
0
1
2
3
4
5
6
7
8
Annual average temperature (°C)
B
Probability of occurrence
0·1
0·2
0·3
0·4
0·5
0·6
0·7
0·8
0·9
Concentration of nitrate (NO_3^-) in precipitation (mg N l^{-1})
C
0
0·5
1
1·5
2
Concentration of ammonium (NH_4^+) in precipitation (mg N l^{-1})

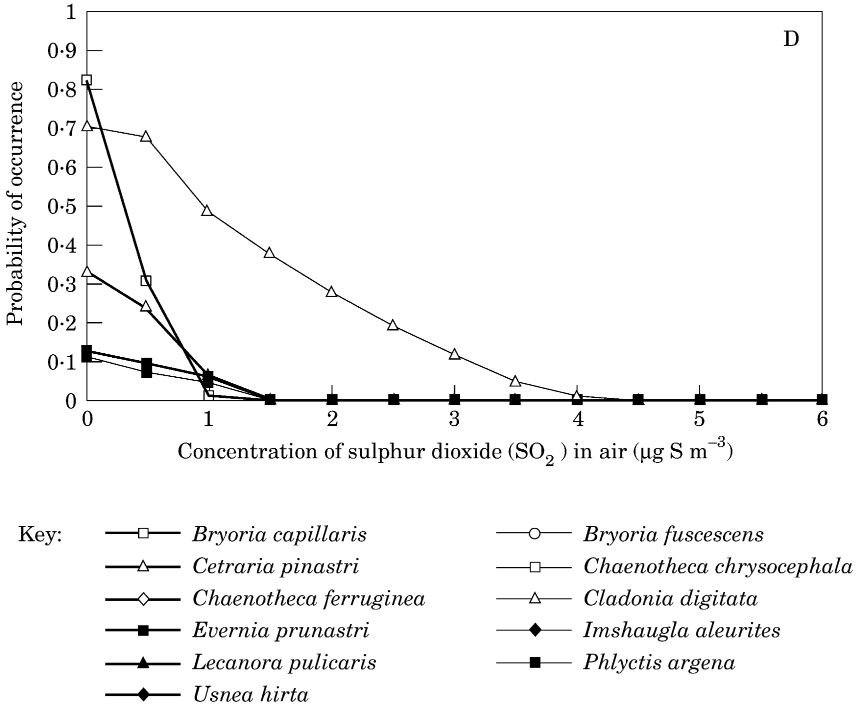


FIG. 3. Probability of occurrence of the eleven lichen species investigated as a function of four predictor variables. A, annual average temperature (°C); B, concentration of nitrate (NO_3^-) in precipitation; C, concentration ammonium (NH_4^+) in precipitation; D, concentration of sulphur dioxide (SO_2) in air. Only probabilities of species that react to variables are shown.

exasperatula), however, nitrogen concentrations in rainfall in this area (0·1 mg N l^{-1}) seem too low to make NH_3 a likely cause.

Acidification of bark due to nitrification of ammonium needs to be considered as well, especially in relation to *Lobarion* communities. At present, there is not enough information available to assess the extent of this process in for example mossy bark. As *Lobarion* species are not treated in the present paper, more research is necessary to assess the role of nitrogen in this context.

A direct effect of an increased nitrogen content of precipitation or bark seems most likely. Barkman (1958) considered the *Hypogymnietalia physodo-tubulosae* as a both ‘strongly acidiphilous’ and ‘strictly nitrophobous’ order. This may explain why some acidophytic lichens seem to be affected by nitrogen at large distances from the source and at low levels, when nitrophytic lichens are not yet found. Our results show that at least some very sensitive acidophytic species seem to react readily to a slightly increased nitrogen content due to NH_4^+, while nitrophytic species were shown earlier to react primarily to an increased pH due to NH_3 (de Bakker & van Dobben 1988; van Herk 2001). As NH_4^+ is transported further than NH_3, it can be concluded that it may affect acidophytic lichens at great distance from the source. Similar conclusions follow from multivariate analysis (van Herk 2001): 44·7% of the variance of acidophytic species (cumulative occurrence) could be explained by bark pH, but an additional 22·5% by atmospheric and bark NH_4^+, whereas nitrophytic species (cumulative) appeared to react only to bark pH (53·6% variance).

During the LICONS (Lichen conservation) conference in Switzerland 1999, it was suggested that effects of nitrogen on sensitive species may be caused indirectly through an increased growth of algae. Algae on the surface of lichens are reported to inhibit their development. It has been observed by Benfield (1994) that old parts of thalli are often attacked first, fall off, and subsequently the open space is filled by algae. Bråkenhielm & Qinghong (1975) found in their study at reference areas in Sweden that the spatial pattern for algae was opposite to the pattern observed for sensitive lichens. In general, algae have often increased where sensitive acidophytes have disappeared. However, both changes may be caused by the same process, i.e. increased nitrogen availability. A strong negative correlation does not necessarily indicate a causal relationship. So far there is no indication that increased growth of mosses or other lichen species plays a significant role.

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