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First Europe-wide correlation analysis identifying factors best explaining the total nitrogen concentration in mosses

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Abstract

In this study, correlation analyses were applied to quantitatively characterise the indicative value of mosses as biomonitors of atmospheric nitrogen (N) deposition and to identify factors which explain best the total N concentration in mosses. Correlations between the total N concentration in mosses and atmospheric N depositions and air concentrations were examined for the first time at a European scale. In addition, predictors such as urban and agricultural land uses, population and livestock density were integrated in the analyses to account for emission-related influences of land use. The analyses included data from mosses collected from 2781 sites across Europe within the framework of the European moss survey 2005/6, which was coordinated by the International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation). Modelled atmospheric N deposition and air concentration data were calculated using the Unified EMEP Model of the European Monitoring and Evaluation Programme (EMEP) of the Long-range Transboundary Air Pollution Convention. The modelled deposition and concentration data encompass various N compounds. In order to assess the correlations between moss tissue total N concentrations and the chosen predictors, Spearman rank correlation analysis and Classification and Regression Trees (CART) were applied. The Spearman rank correlation analysis showed that the total N concentration in mosses and modelled N depositions and air concentrations are significantly correlated ($0.55 \leq r_s \leq 0.68$, $p < 0.001$). Correlations with other predictors were lower than 0.55. The CART analysis indicated that the variation in the total N concentration in mosses was best explained by the variation in NH_4^+ concentrations in air, followed by NO_2 concentrations in air, sampled moss species and total dry N deposition. The total N concentration in mosses appears to mirror land use-related atmospheric concentrations and depositions of N across Europe to a high degree. The total N concentration in mosses is a valuable tool in identifying areas with high atmospheric N concentration and deposition at a high spatial resolution across Europe.

Keywords:
biomonitoring, correlation, deposition, moss, nitrogen

1 Background and goal

Preindustrial atmospheric levels of nitrous oxide (N_2O) measured in bubbles in glacier ice were around 285 ppm for many thousands of years. Human activity in the last 200 years has raised the N_2O level in the atmosphere which meanwhile approximates 310 ppm. The main anthropogenic sources for oxidised forms of N are transport, industry and energy production, estimated to contribute up to 70% of oxidised N (NO_x) emissions (Bragazza et al., 2005). Additional sources include soil emission, particularly under high N inputs for example in agricultural areas. Emission sources of reduced N (NH_y) compounds are primarily related to agricultural activities such as animal husbandry and the production and application of fertilizers. Forest fires are another important source of N emissions (Jovan and Carlberg, 2007). Deposition occurs when such emissions undergo complex chemical reactions in the atmosphere and get in contact with the earth´s surface (vegetation, soils) as wet deposition (rain, snow), occult deposition (cloud, fog) or dry deposition (dry particles, gas). There, they may accumulate in soils and plants such as mosses. Airborne chemicals can travel long distances from their sources and can therefore affect ecosystems over broad spatial scales and at locations far from the sources of emissions. Given European conditions, NH_y may be transported up to 500 km and NO_x effectively more than 1,000 to 1,500 km. Nitrate (NO_3^-) can have a transport range of up to 2,000 km (Slanina and Wayne, 2008).

Enhanced deposition of both NO_x and NH_y may be associated with acidification and eutrophication of ecosystems as well as changes in biodiversity (Erisman and de Vries, 2000; Galloway et al., 2008; Gundersen and Rasmussen, 1990; Pitcairn et al., 1998). Control of reactive N emissions to air is regulated under several European Union directives, such as the National Emission Ceilings Directive and Nitrates Directive and protocols of the Long-range Transboundary Air Pollution (LRTAP) Convention, such as the Gothenburg Protocol. The LRTAP Convention established a broad framework for co-operative action on reducing the impact of air pollution and set up a process for negotiating concrete measures to control emissions of air pollutants through legally binding protocols. In this process, the main objective of the European Monitoring and Evaluation Programme (EMEP) is to regularly provide governments and subsidiary bodies under the LRTAP Convention with qualified scientific information to support the development and further evaluation of the international protocols on emission reduction negotiated within the Convention. EMEP focuses on four main tasks: 1) collection of emission data, 2) measurements of concentrations in air and precipitation, 3) modelling of atmospheric transport and deposition of air pollutions and 4) integrated assessment. The storage and distribution of information on emissions and emission projections on N is the task of the Centre on Emission Inventories and Projections (CEIP). The Meteorological Synthesizing Centre West (MSC-W) is responsible for the modelling of sulphur (S), N, photo-oxidants and atmospheric particles.

Parties to the LRTAP Convention perform wet deposition monitoring at 70 regional monitoring sites across Europe within the framework of EMEP. The deposition measurement sites of the EMEP Chemical Coordinating Centre (EMEP-CCC) are spread over a large geographical area, so that, e.g. in the year 2000 Germany was represented by only eight measurement stations (Simpson et al., 2006b). In addition, deposition measurements sites are under-represented in southern and Eastern Europe. Depositions associated with acidification and eutrophication, as well as ozone are currently calculated by use of the EMEP MSC-W Unified Eulerian chemical transport model with a broad grid-size of 50 × 50 km^2 (Simpson et al., 2006a).

The EMEP depositions and air concentration models are each year validated against all the EMEP measurement data available throughout Europe. Furthermore, data outside the EMEP network are compared to the EMEP model in order to extend the data basis for the evaluation of the EMEP models. For instance, Simpson et al. (2006b) supplemented the EMEP deposition data by deposition measurements from the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests). The ICP-Forests Level II monitoring network started operations in 1994 and encompasses about 860 sites. However, applying quality assurance criteria Simpson et al. (2006b) had to exclude data from many sites and countries from the comparison with EMEP deposition data. Data from

160 ICP-Forests monitoring sites distributed over seven European countries and 140 EMEP grids fulfilled the quality assurance criteria. Regarding the 89 German ICP Level II-sites, 26 were in agreement with the respective quality criteria. The variability of the data quality from the ICP Forest sites proved to be considerable and the sites are not always representative for a larger area.

Even if Simpson et al. (2006b) concluded that the EMEP model performs rather well in reproducing patterns of N deposition in European forests, it is clear that an enhanced spatial resolution of the model results would be an advantage, e.g. when used for calculating exceedances of critical loads of acidification and eutrophication. At present, the EMEP model can be run on 10 km resolution, and emission data sets that can be used on this resolution are emerging. However, a dense network of observations is needed in order to validate whether the model results really improve on finer resolution. One step towards reaching this aim is to compare the calculated depositions and concentrations with observations from networks monitoring a phenomenon closely related to deposition and operating at a high spatial resolution.

Carpet-forming ectohydric mosses have successfully been used as biomonitors of atmospheric heavy metal deposition since the late 1960s (Rühling and Tyler, 1968, 1969, 1970). The moss technique is based on the fact that carpet-forming ectohydric mosses obtain most trace elements and nutrients directly from wet and dry deposition with little uptake from the substrate. Heavy metals accumulate in mosses and their concentration in moss tissue provides a surrogate, time-integrated measure of element deposition from the atmosphere to terrestrial systems (Harmens et al., 2008a). It is easier and cheaper than conventional deposition analysis as it avoids the need for deploying large numbers of deposition collectors with an associated long-term programme of routine sample collection and analysis. Hence, a much higher sampling density can be achieved than with conventional deposition analysis.

Carpet-forming ectohydric mosses might potentially be used as biomonitors of atmospheric N deposition as well (Pitcairn et al., 1995, 2006; Poikolainen et al. 2009; Salemaa et al., 2008; Solga et al., 2005; Zechmeister et al. 2008). A pilot-study in Scandinavian countries showed a strong linear relationship between the total N concentration in mosses and EMEP-modelled atmospheric N deposition rates

(Harmens et al., 2005). Therefore, for the first time in the European heavy metals in mosses survey the total N concentration was also determined in naturally-growing mosses in about 60% of the participating countries in 2005/6 (Harmens et al., 2008b). The European moss survey is currently coordinated by the International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation) and has been conducted every five years since 1990 (Harmens et al., 2008a). The total N concentration in mosses was measured at almost 3,000 sites in 16 European countries (Harmens et al., 2008b), providing a higher spatial resolution than the EMEP monitoring sites.

The aim of this study was to investigate whether ectohydric mosses can be used to monitor atmospheric air concentration and / or deposition of N at a high spatial resolution. Therefore, the correlations between N concentrations in mosses and modelled atmospheric concentrations and depositions of different N forms were examined. In addition, the influence of other predictors on the variation of the N concentration in mosses was investigated, including altitude, precipitation, distance to the sea, population and agricultural density, as well as urban, forested and agricultural land uses.

2 Materials and Methods

2.1 Total N concentration in mosses

Moss samples were collected during the European survey 2005/6 in Austria, Belgium, Bulgaria, Czech Republic, Estonia, Finland, France, Germany, Italy (Bolzano region), Latvia, Slovakia, Slovenia, Spain (Navarra), Switzerland, Turkey, and the United Kingdom at 2781 sites, which were representative in terms of a multivariate statistical ecoregionalisation of Europe (Hornsmann et al., 2008). As in previous European moss surveys, field sampling was conducted according to the guidelines set out in the monitoring manual for the 2005/6 survey (ICP Vegetation, 2005). These guidelines specify the moss species to collect as well as how and where to perform the sampling. In order to document whether these criteria could be reached the comprehensive WebGIS *MossMet* was used in the German moss survey 2005 (Pesch et al., 2007a). *MossMet* contains sampling site-specific information in terms of a check list concerning the mandatory specifications of the experimental protocol, detailed topographical and ecological site descriptions, and the measurement values of up to 40 metal elements recorded in 1990, 1995, 2000, and 2005. Furthermore, the surroundings of each sampling site are described by surface data on climate, altitude and land use (Pesch et al., 2007b). This approach is intended to be adopted by more participants of the European moss survey in 2010. In the current study, the N concentration in mosses were only related to a smaller set of site-specific and regional predictors such as EMEP modelled N deposition data and land use parameters (see Section 2.3).

Pleurozium schreberi was the most frequently sampled species (43.4%), followed by *Hylocomium splendens* (20.1%), *Hypnum cupressiforme* (18.9%), *Pseudoscleropodium purum* (11.8%) and other species (5.7%). For the determination of N, the moss samples were dried at 40° C and concentrations were determined according to either the Kjeldahl method (Kjeldahl, 1883) or via elemental analysis following the Dumas method (Dumas, 1831). N concentrations were expressed as percentage N based on dry weight. A quality control exercise was conducted to assess the analytical performance of the participating laboratories. The moss reference materials M2 and M3, first prepared for the 1995/6 European moss survey (Steinnes et al., 1997), were distributed amongst participating laboratories. In addition, some laboratories used certified reference material for quality assurance. For determination of the elemental concentrations in the reference materials, laboratories followed the same analytical procedure as used for the sampled mosses. The results indicated good agreement between laboratories and analytical techniques, and recommended values for the total N concentrations were established for the reference material (Harmens et al., 2008a). The accuracy of data submitted to the Programme Coordination Centre was assessed by inspecting them for extremes and by sending summarised data and draft maps to individual contributors for checking and approval before incorporating the final data into the database for further processing. To investigate whether mosses can be used as biomonitors of atmospheric N deposition and air concentration, the spatial variation in the N concentration in mosses was statistically compared (see Section 2.4) with EMEP modelled N deposition and air concentration data (see Section 2.2) and other potential predictors such as land use (see Section 2.3).

2.2 EMEP Unified model calculation of N depositions and air concentrations

The Unified EMEP model was developed at the Norwegian Meteorological Institute under the EMEP programme. The Unified EMEP model is a development of earlier EMEP models (Berge and Jakobsen, 1998; Jonson et al., 1998; fully documented in Fagerli et al., 2004; Simpson et al., 2003). The model has been extensively validated against measurements (Fagerli and Aas, 2008; Fagerli et al., 2003, 2007; Jonson et al., 2006; Simpson et al., 2006a,b, 2007; Tsyro et al., 2007). The EMEP Unified Eulerian Chemical Transport Model is a multi-layer atmospheric dispersion model designed for simulating the long-range transport of air pollution over several years. The EMEP model domain is centred over Europe and also includes most of the North Atlantic and the polar region (Fagerli et al., 2004; Simpson et al., 2003)[1]. The model comprises 20 vertical layers and is primarily intended for use with a horizontal resolution of ca. 50 × 50 km^2 (at 60° N) in the EMEP polar stereographic grid. The anthropogenic emission input data are generally based as far as possible upon emissions per sector and grid officially reported to the LRTAP Convention (Vestreng et al., 2004). The chemical scheme uses about 140 reactions between 70 species. The model allows for calculations of depositions to different types of land-cover within each grid cell.

2.3 Additional geodata

In addition to the modelled EMEP deposition and air concentration data, the N concentration in mosses was investigated for correlations with other geodata (Table 1). These other geodata include four site-specific characteristics and 19 regional characteristics which might influence the N concentration in mosses (Böhlmann et al., 2005; Bytnerowicz et al., 2002; Bytnerowicz and Fenn, 1996; Fenn et al., 2007; Fenn and Kiefer, 1999; Fernandez and Carballeira, 2002; Fowler et al. 1998; Frati et al., 2007; Jovan and Carlberg, 2007; Jovan and Mccune, 2006; Luo et al., 2003; Neirynck et al., 2007; Pesch et al., 2007b; Pitcairn et al., 2006). Together with the EMEP data the site-specific and regional characteristics are referred to as predictors in the CART-analysis (see Section 2.4). Raster information from surface maps were intersected with the monitoring sites and included in the correlation analysis. To account for the influence of the precipitation amount on the moss N loads, long-term

[1] http://www.emep.int/OpenSource/UniEMEPopenSource_documentation.html

monthly means (1961- 1990) were provided by the Global Climate Dataset (CL 2.0) in a resolution of 12.5 x 12.5 km². To supplement the site-specific data with regard to information on N emissions, proportions of land use were derived from the Corine Land Cover map 2000 (Keil et al., 2005). The area percentage of urban, forest and agricultural land use categories in a radius of 1, 5, 10 and 25 km (for forests), 1, 5, 10, 25 and 50 km (for agriculture) or 1, 5, 10, 25, 50, 75 and 100 km (for urban areas) around each raster cell was calculated and then projected onto either the 1 x 1 or 2 x 2 km² grid cells (Table 1). The sea spray-effect was assessed in terms of the distances of the monitoring sites to the coastlines of the Atlantic Ocean and the Baltic, Black and Mediterranean Sea. Further data used as predictors include the population density in a resolution of 100 x 100 m^2 as well as livestock density provided by EUROSTAT[2]. The latter was only available in terms of country averages.

Table 1: Predictors used for correlation analyses

2.4 Statistical analysis

Prior to the bi- and multivariate statistical analyses the moss and modelled deposition / air concentrations as well as additional land characteristics were intersected in a GIS environment. Regarding the EMEP modelling data the correlation analyses was performed with two sets of modelled N data: (1) one data set containing only those values representing the year of sampling and (2) one data set representing the whole three year accumulation period in terms of the mean deposition / air concentration. The three year accumulation period represents the moss tissue that was selected for total N analysis, i.e. the recent three years of moss growth.

Bivariate correlation coefficients were computed to indicate the strength and direction of the statistical relationship between the total N concentrations in mosses and EMEP modelled N depositions and air concentrations and additional influencing factors. There are several coefficients, measuring the degree of correlation, adapted to the levels of measurement according to Stevens (1946) or to the statistical distribution of the data. In this investigation, we decided to compute the Spearman rank correlation coefficient r_s because the total N concentration in mosses and most of the predicting variables proved to be not normally distributed. Such non-parametric

[2] http://epp.eurostat.ec.europa.eu/portal/page/portal/eurostat/home/ (June 2009)

correlation methods are less powerful than parametric methods if the assumptions underlying the latter are met, but are less likely to give distorted results when the assumptions fail. The coefficient r_s measures the correspondence between two rankings and assesses its statistical significance. An increasing rank correlation coefficient implies increasing agreement between rankings. The correlation coefficient r_s amounts for -1 if the two rankings are completely in opposite agreement, r_s equals 0 if the rankings are completely independent and +1 if the agreement between the two rankings is perfectly the same. Within this interval [-1, +1] the strength of correlation was classified according to Hagl (2008); r_s values < $|0.2|$ are very low, between $|0.2|$ and $|0.5|$ low, from $|0.5|$ to $|0.7|$ moderate, between $|0.7|$ and $|0.9|$ high and > $|0.9|$ very high. Statistical significance refers to the generality of the relationship and the likelihood the observed relationship occurred by chance.

In addition to non-parametric correlation analysis, classification and regression trees (CART) as introduced by Breimann et al. (1984) were applied to analyse the multivariate correlations between the tissue N concentration of mosses and characteristics of the surroundings of the 2781 sampling sites such as modelled air concentrations and depositions of N and land cover. CART does not make any assumptions regarding the distribution of the data and can use an explanatory variable more than once, so it is able to work with multiple-interrelated data. CART can reveal hierarchical and non-linear relationships among one dependent variable (tissue N concentration of mosses) and several descriptive variables (regional characteristics of the sampling sites such as N depositions and proportion of land cover) by sub-dividing a heterogeneous data set into more homogeneous sub-sets (classes, groups, nodes) by a series of nested binary 'if-then-else' splits. Each split maximises the homogeneity of the dependent variable. Each possible binary split for all variables is evaluated recursively for the best class separation until homogeneous end points (nodes) are reached. The predictor selected is the one for which the two new classes have the greatest within-group similarity for the response variable. The two new classes are then examined separately, with respect to each of the predictor variables, to see if they can be split again. The resulting dendrogram can have multiple branches, each of which represents a path to a particular combination of independent variables defining variable sub-spaces.

3 Results

The Spearman rank correlations coefficients r_s were computed to identify the relations between the different N depositions (dry and wet oxidised and reduced N deposition, total dry and total wet N deposition and total N deposition) and air concentrations (NO_2, HNO_3, NO_3^-, NH_3, NH_4^+) as well as site-specific and regional land characteristics on the one hand and the N concentration in mosses on the other hand. Figure 1 shows the rank correlation coefficients between the total N concentration in mosses and the modelled EMEP depositions and air concentrations and table 2 lists the rank correlation coefficients between the total N concentration in mosses and site-specific or regional characteristics. The correlation coefficients are only depicted with regard to the three years mean of all EMEP components since only minor differences exist to those correlation coefficients calculated for depositions / air concentration representing the year of sampling. The highest correlations (r_s = 0.64 – 0.68) exist between the N concentrations in mosses and NH_4^+ concentrations in air, wet and dry deposition of oxidised N, the total wet N deposition and the total N deposition. Regarding the regional land characteristics, the proportion of urban land uses in a radius of 75 - 100 km and agricultural land uses in a radius of 50 km around the monitoring sites showed the highest correlations with the total N concentration in mosses (r_s = 0.55 and r_s = 0.54, respectively). Low correlations (r_s < 0.5) were observed for altitude, precipitation, distance to sea, forested land use and population and livestock density.

Figure 1: Spearman rank correlation coefficients between N concentration in mosses and EMEP modelled air concentration and deposition of various N forms (three year mean)

Table 2: Spearman rank correlation coefficients between N concentration in mosses and site-specific and regional characteristics

Multivariate relations between the tissue N concentrations of mosses and modelled EMEP N depositions / air concentrations and potential site-specific and regional land characteristics (Table 1) were analysed using CART. The respective results are shown for the first three levels of the CART dendrogram in Figure 2. According to the root node of the dendrogram, the mean N concentration in mosses amounts to 1.22% N. As can be seen, the modelled NH_4^+ concentration in air is the most powerful predictor for the total N concentration in mosses: at sites with NH_4^+ concentrations below 0.63 mg m^{-3} the mean tissue N concentration of mosses equals 0.88% (node 1) whereas at sites with NH_4^+ concentrations above 0.63 mg m^{-3} the mean is 1.41% (node 2). The sampling sites classified in node 1 are further differentiated by the NO_2 concentrations in air: in mosses sampled at sites with NO_2 concentrations below 0.53 mg m^{-3} the N concentration in mosses is lower (0.65%) (node 3) than at locations where the concentrations exceed 0.53 mg m^{-3} (0.96%) (node 4). Node 2 is sub-divided by the sampled moss species: at sampling sites where *Abietinella abietina (Aa), Hylocomium splendens (Hs), Pleurozium schreberi (Ps), Rhytidiadelphus squarrosus (Rs*) or mosses of the genus *Scleropodium (Scl. sp.)* other than *Scleropodium purum* were sampled lower N concentrations in mosses were measured (1.29 %) (node 5) than at sites where *Brachythecium rutabulum (Br), Hylocomium splendens (Hs), Pseudoscleropodium purum* (Pp), Scleropodium Purum*[*] *(Sp), Thuidium abietinum (Ta), Thuidium tamariscinum (Tt)* or mosses of the genus *Brachythecium (Bra. sp.), Dicranum (Dic. sp.)* and *Homalothecium (Hom. sp.)* were sampled (1.52%) (node 6). Level 3 nodes 5 and 6 are furthermore split according to the total dry N deposition and the air concentration of ammonium. The CART dendrogram depicted in Figure 2 accounts for up to 44.8% of the total variance of the total N concentration in mosses measured at the 2781 sites.

Figure 2: CART-dendrogram of factors associated with N concentrations in mosses, explaining 44.8% of the total N concentration in mosses.

4 Discussion

This study reveals that mosses can potentially be used as biomonitors of total atmospheric deposition of N at a high spatial resolution. The highest, albeit moderate, bivariate correlations were found between EMEP modelled air concentrations and deposition of different N forms and the total N concentrations in mosses. Multivariate analysis identified modelled NH_4^+ concentrations in air as the most powerful predictor of the total N concentration in mosses. The total N concentration in mosses appears to mirror land use-related atmospheric

[*] Now collectively called *Pseudoscleropodium purum* (Hill et al. 2006).

concentrations and depositions of N across Europe as moderate bivariate correlations were also observed between the proportion of urban land uses (in a radius of 75 - 100 km) and the proportion of agricultural land uses (in a radius of 50 km). Correlations with other site-specific or regional characteristics were lower.

The applied statistical techniques are robust against skewed distribution of the data and the influence of potential outliers. Autocorrelations for the N concentrations in mosses exist but are rather weak (nugget / sill ratios of above 0.7 for four sub-regions in Europe). The assumption of independence for the target variable was therefore not violated. Furthermore, the existing co-linearities between the predicting variables are not a problem since each variable was investigated separately and not altogether as done in multivariate approaches like cluster or regression analyses. By applying CART, each predicting variable is examined separately with regard to the improvement of homogeneity they result in. Only the best predictor is chosen for each split.

This study may be regarded as a cross validation of European moss data and EMEP model data for N, but is complicated by both potential limitations in the of mosses as monitors of atmospheric N deposition (Harmens et al., 2008b; Zechmeister et al., 2008) and the uncertainties in the modelled nitrogen deposition, including uncertainties in emissions (Lieven et al., 2009). The moss technique provides a tool for validating the spatial pattern for modelled EMEP air concentrations and depositions of nitrogen compounds at a higher spatial resolution than can be achieved using the EMEP measurement stations throughout Europe. Atmospheric N input to terrestrial ecosystems is spatially variable due to e.g. distance to emission sources, variations in the aerodynamic roughness of vegetation, microclimate, canopy drip or orographic effects. As a result, site-specific inputs of N vary considerably from the mean annual deposition of a region. Indeed, when the total N concentration in mosses was plotted against site-specific bulk N deposition in Switzerland, a strong positive linear relationship (r = 0.95) was observed (Thöni et al., 2008). Nevertheless, previous studies in selected Scandinavian countries have also shown strong positive linear relationships between the total N concentration in mosses and EMEP modelled N deposition data (Harmens et al., 2005), which might be due to the absence of significant local pollutions sources in many of these parts of Scandinavia.

In Germany, the N concentrations in mosses (*Pleurozium schreberi*) and pine needles were compared with N deposition rates and N concentrations in precipitation (Mohr, 1999). The study covered 23 forest sites in northern Germany sampled between 1996 and 1998. The correlation (r) between the N concentration in mosses and pine needles was 0.87 ($p < 0.001$) and between the N concentration in mosses and N concentration in precipitation (measured in the year before the sampling) was 0.9 ($p < 0.001$). In 1998, the correlation between the N concentration in mosses and the annual N (NO_3^- + NH_4^+) deposition rate was 0.93 ($p < 0.01$). There is a need to measure atmospheric N deposition at selected moss sampling sites in other countries too in order to further investigate the robustness of the relationship with total N concentration in mosses.

In the current study, bivariate correlations between N concentrations in mosses and air concentration and / or deposition of different N species were quite similar for some N species, with multivariate analysis identifying NH_4^+ concentrations in air as the most powerful predictor of the total N concentration in mosses. However, Pitcairn et al. (2006) have shown that N concentration in mosses can respond differently to wet and dry deposited N. They concluded that N concentrations in mosses provide a good indication for N deposition at sites where deposition is dominated by NH_3, and is also valuable in identifying vegetation exposed to large concentrations of NH_4^+ and NO_3^-, in wet deposition dominated areas, such as hilltops and wind exposed woodland edges. In line with these findings, Pearce and Van der Wal (2008) suggested that tissue N concentration in mosses is more sensitive to N concentrations rather than deposition dose. It is therefore vital to gain a better understanding of the role concentration and dose and the various N forms play in threatening the survival of sensitive species and their habitats (Nordin et al., 2009). It should be noted that in general the EMEP model results agree better with measurements for the secondary components such as ammonium and nitrate in air. NH_3 concentrations differ greatly within small distances, which is impossible to reproduce with a model with a resolution of 50*50km. Moreover, whilst wet depositions are regularly validated against measurements, measurement data on dry deposition of nitrogen are scarce. Furthermore, the data that exists have been used to develop the dry deposition parametrisations that are used in the model – thus an independent validation is difficult.

As biomonitors of atmospheric N depositions, mosses can potentially be used for identifying areas at risk from high N deposition (Harmens et al., submitted). Increased deposition of atmospheric N is affecting biodiversity and the composition of natural and semi-natural vegetation in Europe (Bobbink et al., in press). The European Union Habitats Directive (92/43/EEC) promotes the maintenance of biodiversity and requires member states to take measures to maintain or restore natural habitats at a favourable conservation status. The Directive established the Natura 2000 network with the aim to assure the long-term survival of Europe's most valuable and threatened species and habitats. The provisions of the Directive require strict site protection measures, avoidance of deterioration and introduce a precautionary approach. Mosses and lichens are already routinely sampled in Denmark to assist with the assessment of Natura 2000 sites (Anderson et al., 2006). However, most likely a combination of the bioindicators / biomonitors will best describe the state on Natura 2000 sites (Nordin et al., 2009). It would be very useful if bioindicators / biomonitors could be applied throughout Europe to provide detailed information on the spatial patterns of N deposition and localise spatially varying exceedance of the critical N load values (Pitcairn et al., 2006). The conversion of the N concentrations in mosses to spatially highly resolved N deposition maps could be done by applying regression kriging as described by Hengl et al. (2007).

Recent pilot studies in Germany and France had already shown strong positive correlations between the N concentration in mosses and the proportion of agricultural land use around the moss sampling sites (Holy et al., 2008; Pesch et al., 2007b; Pesch et al., 2008). Although other factors such as urban land use, canopy drip and moss species were also identified as main factors determining the spatial variation in N concentration in mosses, modelled air concentrations and depositions were not yet included in those studies. The outcome of those studies is in agreement with other studies (Bytnerowicz et al., 2002; Frati et al., 2007; Jovan and Carlberg, 2007; Jovan and Mccune, 2006), which identified agriculture, especially the application of animal manure, animal feedlots and mast farms, as the major source of N in the environment. Further important sources are road traffic (Jovan and Mccune, 2006; Luo et al., 2003), urban emissions (Bytnerowicz and Fenn, 1996) and forest fires (Jovan and Carlberg, 2007).

5 Conclusions

The total N concentration in mosses mirrors land use-related atmospheric concentrations and depositions of N to a considerable degree and is therefore a valuable tool in identifying areas with high atmospheric N concentration and deposition at a high spatial resolution across Europe. The high number of measurement sites in the moss surveys can improve the evaluation of the EMEP N deposition and air concentration modelling. Studies are currently ongoing to cross-validate the N concentration in mosses with EMEP modelled air concentrations and depositions of N at a higher spatial resolution, i.e. 10 x 10 and 25 x 25 km^2 grids. Correlations between the two datasets can help to improve the spatial resolution of air concentration and deposition maps of N by means of a regression kriging approach based on surface maps of N concentrations in mosses (Hengl et al., 2007). The resulting high-resolution N deposition maps could potentially be used to assess atmospheric N inputs into protected areas like e.g. Natura 2000 areas. In order to enhance the robustness of the relationship between total N deposition and total N concentration in mosses, more measurements of site-specific air concentrations and depositions of N and other site-specific characteristics are required.

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Table 1: Predictors used for correlation analyses.

Predictor	Resolution	Data Source
Moss species	site-specific	
Altitude	site-specific	
Analytical method	site-specific	
Sea distance	site-specific	
Precipitation	12.5 km x 12.5 km	CRU
Population density	100mx100m	EEA[2]
Agricultural land uses (1, 5, 10, 25, 50 km radius)	1 km x 1 km	EEA[2]
Forested land uses (1, 5, 10, 25 km radius)	1 km x 1 km	EEA[2]
Urban land uses (1, 5, 10, 25, 50 km radius)	1 km x 1 km	EEA[2]
Urban land uses (75, 100 km radius)	2 km x 2 km	EEA[2]
Livestock density	Country-specific	EUROSTAT'
Wet deposition of oxidised nitroqen compounds	50 km x 50 km	MSC-West
Dry deposition of oxidised nitrogen compounds	50 km x 50 km	MSC-West[4]
Wet deposition of reduced nitrogen compounds	50 km x 50 km	MSC-West[4]
Dry deposition of reduced nitror: en compounds	50 km x 50 km	MSC-West[4]
Total wet deposition (oxidised +reduced)	50 km x 50 km	MSC-West[4]
Total dry deposition (oxidised +reduced)	50 km x 50 km	MSC-West[4]
Total N deposition (total dry+ total wet)	50 km x 50 km	MSC-West[4]
$N0_2$ concentration in air	50 km x 50 km	MSC-West
$HN0_3$ concentration in air	50 km x 50 km	MSC-West
NOç concentration in air	50 km x 50 km	MSC-West
NH_3 concentration in air	50 km x 50 km	MSC-West
NH_4 concentration in air	50 km x 50 km	Mo;c-vvest

Cl1mat1c Research Un1t, www.cru.uea.ac.uk

[2] European Environment Agency, http://www.eea.europa.eu/

[3] Statistical Office of the European Communities, http://epp.eurostat.ec.europa.eu

[4] Meteorological Synthesizing Centre-West of EMEP, http://met.no

Table 2: Spearman rank correlation coefficients between N concentration in mosses and site-specific and regional characteristics.

Predictor	r. ($p < 0.001$)
Altitude	-0.10
Population density	0.48
Precipitation	0.25
Livestock density (EUROSTAT)	0.42
Distance to the sea	0.25
Agricultural land use (1 km radius)	0.36
Agricultural land use (5 km radius)	0.49
Agricultural land use (10 km radius)	0.51
Agricultural land use (25 km radius)	0.52
Agricultural land use (50 km radius)	0.53
Forestal land use (1 km radius)	-0.11
Forestal land use (5 km radius)	-0.21
Forestal land use (10 km radius)	-0.23
Forestal land use (25 km radius)	-0.23
Urban land use (1 km radius)	0.15
Urban land use (5 km radius)	0.41
Urban land use (10 km radius)	0.49
Urban land use (25 km radius)	0.51
Urban land use (50 km radius)	0.54
Urban land use (75 km radius)	0.55
Urban land use (100 km radius)	0.55

Figure 1: Spearman rank correlation coefficients between N concentration in mosses and EMEP modelled air concentration and deposition of various N forms (three year mean).

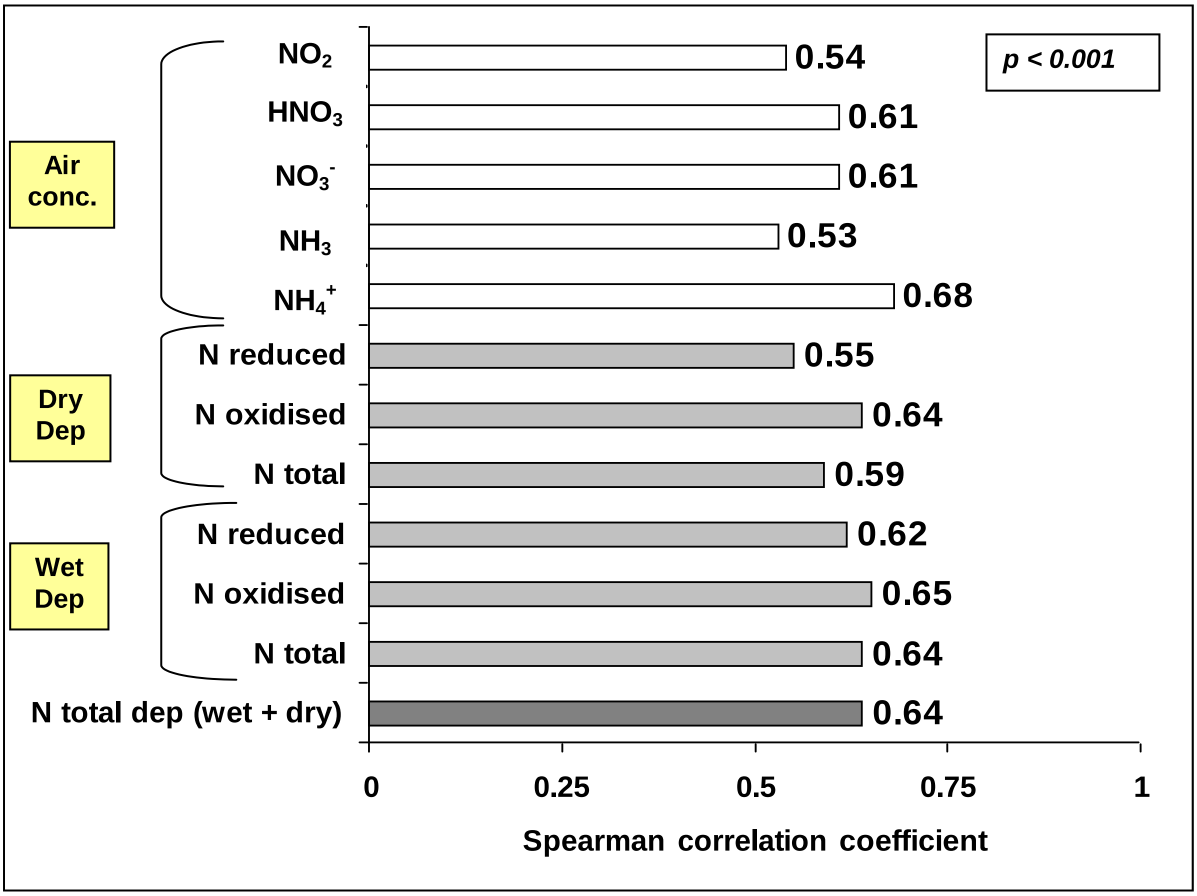
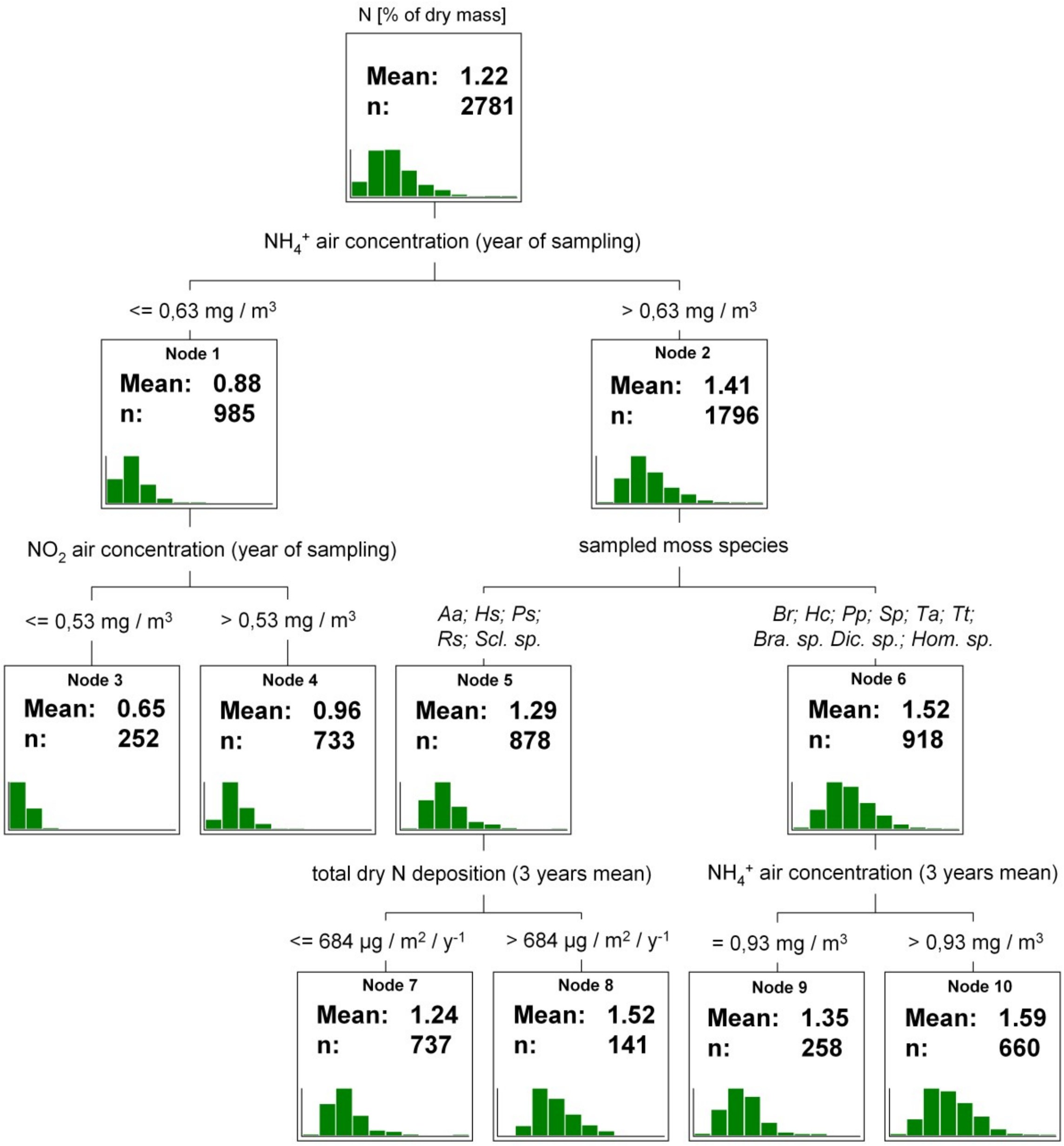


Figure 2: CART-dendrogram of factors associated with N concentrations in mosses, explaining 44.8% of the total N concentration in mosses.



Abietinella abietina (Aa), *Brachythecium* (Bra. sp.), *Brachythecium rutabulum* (Br), *Dicranum* (Dic. sp.), *Homalothecium* (Hom. sp.), *Hylocomium splendens* (Hs), *Pleurozium schreberi* (Ps), *Pseudoscleropodium purum** (Pp), *Rhytidiadelphus squarrosus* (Rs), *Scleropodium* (Scl. sp.), *Scleropodium purum** (Sp), *Thuidium abietinum* (Ta), *Thuidium tamariscinum* (Tt)

*Species classification is according to the nomenclature used during the 2005/6 European moss survey. *Scleropodium purum* and *Pseudoscleropodium purum* are now collectively called *Pseudoscleropodium purum* (Hill et al., 2006)