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Review of the European moss survey (1990 – 2005): deliverables and use of data

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Summary

Carpet-forming, ectohydric mosses obtain most trace elements and nutrients directly from precipitation and dry deposition with little uptake from the substrate and are therefore suitable biomonitors for atmospheric heavy metal and potentially also nitrogen deposition. A European moss biomonitoring network for heavy metals was established in 1990 and moss surveys have since then been repeated at five-yearly intervals, including in the UK. Analysis of nitrogen was included for the first time in the 2005/6 survey. As sampling of mosses is much easier and cheaper than the collection of deposition samples, a much higher spatial density is achieved in moss than deposition monitoring networks. As the density of monitoring stations of atmospheric heavy metal and nitrogen deposition is low across the UK and Europe, the moss data provide a valuable tool for cross-validation of atmospheric deposition fields obtained by atmospheric transport and deposition models applied across Europe (by EMEP) and the UK (e.g. in the UK rural heavy metal monitoring network). This review provides an overview of the deliverables of the European moss survey and how the moss data are being used within the LRTAP Convention and within the UK.

Detailed statistical correlation analyses between the moss data and atmospheric concentrations and depositions of the heavy metals cadmium, lead and nitrogen and other regional (e.g. land use, precipitation, population and agricultural density) or site-specific (e.g. altitude, distance to the sea) characteristics has shown that the heavy metal and nitrogen concentration in mosses is primarily determined by the atmospheric concentrations and depositions of these elements. Across Europe and the UK, the spatial variation in the concentrations of cadmium and lead in mosses corresponds well with the spatial variation in modelled atmospheric depositions. The same is true for temporal trends across Europe between 1990 and 2005. At the European level, correlations are lower for mercury (mercury was not included in the UK moss survey), which is most likely due to technical difficulties of the analysis of mercury and the greater uncertainties associated with mercury in atmospheric transport models. In comparison with modelled deposition of mercury, the concentration in mosses tends to underestimate the decline in mercury deposition between 1995 and 2005. Although the relationship between modelled atmospheric deposition of nitrogen and its concentration in mosses shows at lot of scatter and correlations are moderate at the UK and European scale, good linear relationships have been shown for selected countries measuring bulk deposition at the moss sampling sites. Good linear relationships between EMEP modelled atmospheric nitrogen deposition and the concentrations in mosses were also observed in selected Scandinavian countries.

It was concluded that he moss data are a useful tool for cross-validation of atmospheric transport and deposition models at a high spatial resolution. The models are generally only validated with low density measurement network of element concentrations in air and precipitation. In Europe, these low density monitoring networks are scarcely or not at all present in southern and eastern Europe. Any discrepancies in spatial or temporal trends highlight areas of research that require further detailed analysis in the future, for example regarding factors potentially contributing to lower correlations between modelled depositions and moss concentrations. The nitrogen in moss data are currently used for cross-validation of EMEP modelled deposition at a higher resolution than 50 x 50 km^2 grid, i.e. 10 x 10 and 25 x 25 km^2 grid. Some countries in Europe (e.g. Denmark) already measure the nitrogen concentration in mosses at regular intervals as part of an integrated assessment of the state of Natura 2000 sites. Based on the deliverables so far and the future usefulness of the data, we recommend the UK to take part in the next European moss survey planned for 2010/11. A

further recommendation is to reduce the temporal frequency of sampling to every 10 years if the 2010/11 survey indicates that the reduction in heavy metal and nitrogen concentration of mosses has not declined substantially since the 2005/6 survey.

Acknowledgements.

We wish to thank Defra for providing financial support for the ICP Vegetation, including this study. We would like to thank the participants of the European moss survey 1990 – 2005 for providing data to the ICP Vegetation Programme Coordination Centre and Winfried Schröder, Roland Pesch and Marcel Holy (University of Vechta, Germany) in particular for conducting detailed statistical analyses. We thank Ilia Ilyin (EMEP/MSC-East) and Hilde Fagerli (EMEP/MSC-West) for providing EMEP data and conducting data analyses.

1. Background

The European moss biomonitoring network was originally established in 1990 to estimate atmospheric heavy metal deposition at the European scale and has since then been repeated at five-yearly intervals (Harmens *et al.*, 2008a). The moss technique is based on the fact that carpet-forming, ectohydric mosses obtain most trace elements and nutrients directly from precipitation and dry deposition with little uptake from the substrate. The technique provides a surrogate, time-integrated measure of metal and potentially nitrogen deposition from the atmosphere to terrestrial ecosystems. It is easier and cheaper than conventional precipitation analysis as it avoids the need for deploying large numbers of precipitation collectors with an associated long-term programme of routine sample collection and analysis. Therefore, a much higher sampling density can be achieved than with conventional precipitation analysis. Mosses have been sampled from up to 7,000 sites from up to 28 countries in each survey. In 2001, the ICP Vegetation Programme Coordination Centre took over the coordination of the European moss survey from the Nordic Council of Ministers as requested by the Long-range Transboundary Air Pollution (LRTAP) Convention. The most recent survey was conducted in 2005/6 and for the first time the nitrogen concentration in mosses was determined in the majority of participating countries (ca. 3,000 sites in 16 countries) to establish whether mosses can also be used as biomonitors of atmospheric nitrogen deposition at the European scale (Harmens *et al.*, 2008b). The next European moss survey is scheduled for 2010/11.

2. Deliverables

The following data and maps have been delivered since 1990:

- Heavy metal concentration in mosses for the following ten metals: arsenic, cadmium, chromium, copper, iron, mercury, lead, nickel, vanadium, zinc. For 2005/6, data are also available for aluminium and antimony.
- Total nitrogen concentration in mosses for 2005/6.
- Maps of heavy metal and nitrogen concentration in mosses showing the mean concentration per 50 x 50 km^2 EMEP grid.
- Identification of areas exposed to high atmospheric deposition of heavy metals and nitrogen at a high spatial resolution.
- Temporal trends in heavy metal concentration in mosses as an estimation for trends in atmospheric heavy metal deposition.
- Identification of areas affected by long-range transport and local pollution.

3. Use of moss data within the LRTAP Convention

3.1 Heavy metals

3.1.1 Protocol on Heavy Metals

In 2006, the data on cadmium, lead and mercury concentrations in mosses was included in a review conducted by the Task Force on Heavy Metals on the sufficiency and effectiveness of the 1998 Protocol on Heavy Metals. This review contained the best available scientific information on the effects of deposition of heavy metals from long-range atmospheric transport (Task Force on Heavy metals, 2006).

3.1.2 Cross-validation with the EMEP atmospheric transport model

Within the LRTAP Convention, deposition of heavy metals is modelled using the EMEP atmospheric transport model MSCE-HM (Travnikov and Ilyin, 2005). The modelled data are verified against heavy metal concentrations in air and precipitation measured at EMEP monitoring stations. However, the number of EMEP monitoring stations and their spatial distribution across Europe is limited. For example, in 2006 and 2007, there were 66 measurement sites, of which only 29 measured heavy metals in both air and precipitation (Figure 1; Aas and Breivik, 2009). In 2007, there were 22 sites measuring at least one form of mercury, which is six more sites than in 2006. Some countries have measurement campaigns but no long-term commitment to monitoring heavy metals. The EMEP monitoring network for cadmium and lead is scarce or absent in southern and eastern Europe, whereas mercury is primarily measured in northern Europe. Compared to the data from EMEP monitoring network, the moss survey has two advantages: i) the density of the moss monitoring network is much higher and ii) their spatial distribution is wider, including parts of southern and eastern Europe (see Figure 1). Although deposition fluxes of heavy metals and their concentrations in mosses cannot be compared directly, it is possible to compare the spatial distribution of deposition and concentrations in mosses. In addition, temporal trends since 1990 can be compared for both data sets.

Spatial trends

In a pilot study in 2005, EMEP/MSC-East used the measurement data of the European moss survey for lead as a cross-validation for the performance of the EMEP atmospheric transport model MSCE-HM (Travnikov and Ilyin, 2005). The mean lead concentration of lead in mosses per EMEP grid for 2000/1 (representing the accumulated lead concentration over the last three years of growth) was compared with the average atmospheric deposition of lead simulated by the EMEP model for the years 1997 – 1999 using correlation analysis (Ilyin and Travnikov, 2005).

A significant positive correlation coefficient ($R = 0.56$) indicated that the EMEP model managed to mimic the spatial pattern of lead pollution levels for the whole of Europe (Figure 2a). The correlation coefficient was not as high as normally obtained when the model is verified with concentrations in precipitation measured at the EMEP network. However, it should be noted that the lead concentrations in mosses were not only determined in areas with background levels of lead pollution, but also in relatively polluted areas. In addition, the concentration of metals in mosses can be affected by factors such as proximity to the sea and contamination by windblown soil dust, in particular in mining areas and dry regions of Europe. Therefore, the correlation between modelled lead deposition and its concentration in mosses can vary from one part of Europe to another. As a result, country-specific correlation coefficients were observed.

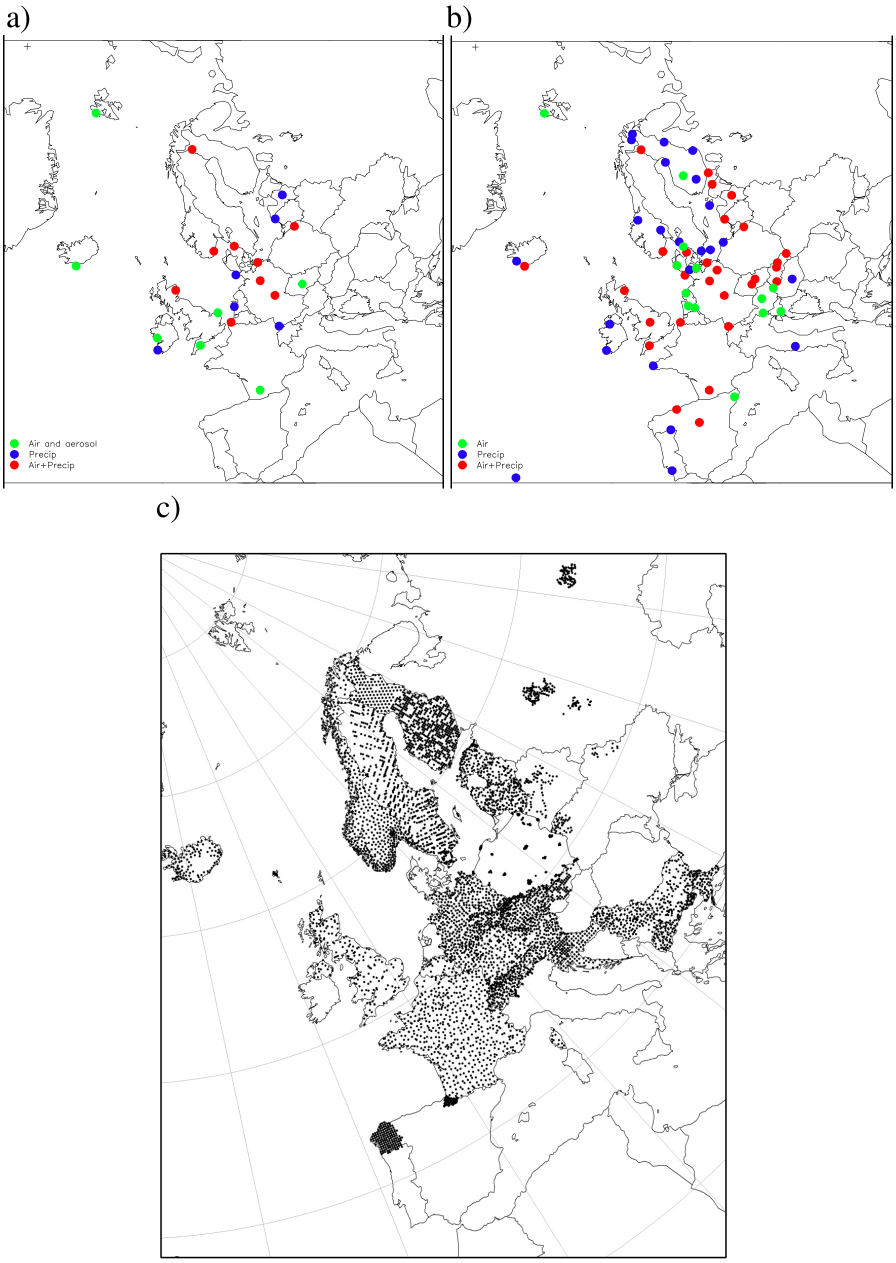

Figure 1. EMEP measurement network of a) heavy metals (+ Cyprus outside the map area) and b) mercury in 2007 (Aas and Breivik, 2009) and c) the moss sampling sites in the European survey of 2005/6.

When a comparison was performed between lead concentrations in mosses and modelled total lead deposition for selected grid cells in Scandinavia where EMEP monitoring stations are situated, i.e. a comparison was performed at locations representative for the EMEP task (modelling long-range transboundary air pollution), a very high correlation of 0.91 was found (Figure 2b). Scandinavian emissions are relatively low and lead pollution levels are mainly caused by long-range transport (and possibly by natural emissions and re-emissions). The high correlation indicates that the EMEP model simulates atmospheric transport well and the lead concentration in mosses can be used as an estimate of atmospheric heavy metal deposition at a high spatial resolution in background areas.

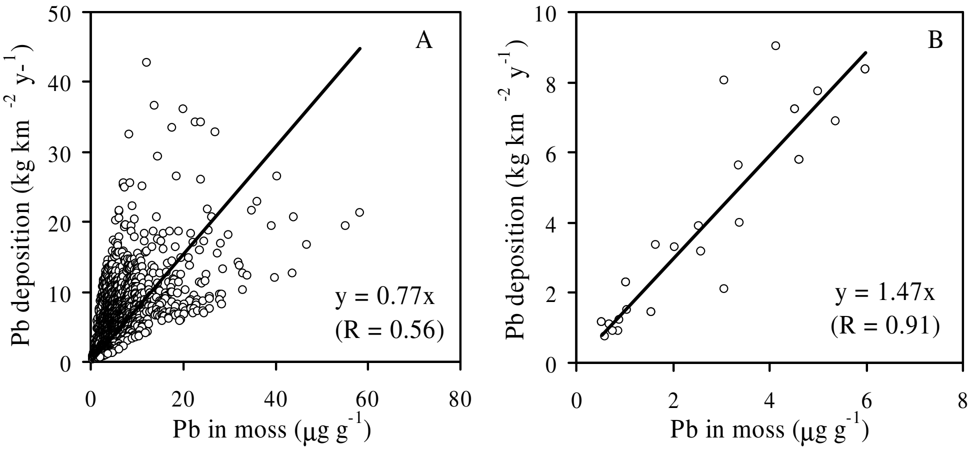

Figure 2. a) Modelled total depositions of lead versus measured lead concentrations in mosses accumulated over 1997 – 1999 across Europe and (b) at sites with background levels of lead pollution in Scandinavia (Norway, Sweden, Finland). Modified after Ilyin and Travnikov (2005).

In 2009, the cross-validation study with the EMEP atmospheric transport model was extended to include all the priority metals of the LRTAP Convention, i.e. cadmium, lead and mercury. Spatial correlation analyses and comparisons of temporal trends were conducted for all survey years. Preliminary data analysis showed that the spatial pattern of cadmium and lead concentrations in mosses and modelled deposition agree reasonably well, i.e. regions with higher deposition had generally higher concentrations in mosses and vice versa in 2005/6 (Figure 3). For mercury, the spatial pattern showed less similarity. For lead, the concentration in mosses appears to be relatively higher than the modelled deposition in Bulgaria, Slovakia, Belgium, Slovenia and the Ukraine, whereas the opposite appears to be the case for the Czech Republic, Croatia and Germany. For cadmium, the lower concentration in mosses compared to modelled deposition in Macedonia is most striking. Cadmium deposition may seem underestimated in Belgium, Russia, Finland, Latvia, and overestimated in the FYR Macedonia, north-western Spain and Lithuania. This comparison does not provide strict evidence that calculated deposition is underestimated or overestimated in certain areas, but it can indicate regions which deserve more thorough attention regarding validation of modeled pollution levels. However, considering the intrinsic uncertainty of the EMEP model (30 – 40% for total deposition; Travnikov and Ilyin, 2005), high uncertainties in heavy metal emission data and potential limitations in the use of moss data as monitors of atmospheric deposition (see Harmens *et al.*, 2008a), the spatial patterns of both data sets agree reasonably well, at least for cadmium and lead.

Figure 4 summarizes correlation coefficients for lead and cadmium obtained for all countries which participated in the 2005/6 moss survey. In a number of countries, the correlation coefficient exceeds 0.5, and in more than half of the countries it exceeds 0.4. However, there are countries where correlation is low or even negative. As a rule, the coefficient for lead is higher than that for cadmium. It is also important to note that area covered by measurements of mosses and to which the coefficient relates varies greatly among countries (Figure 4).

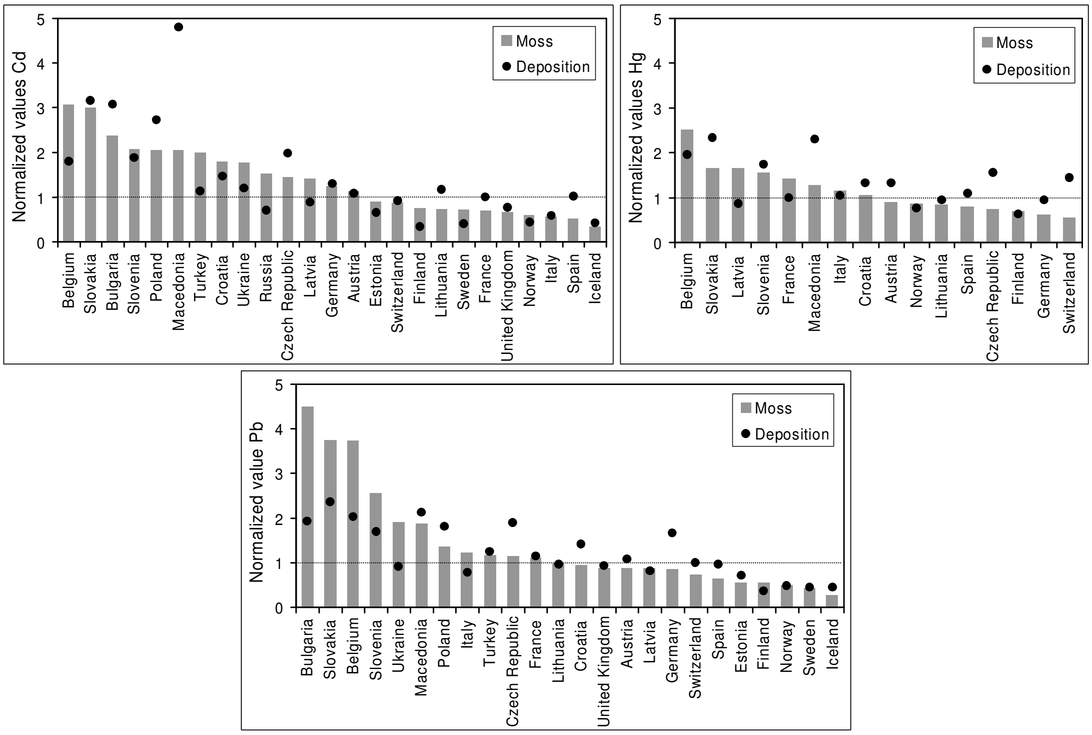

Figure 3. Normalized values (relative to the overall mean) of the average cadmium (top, left), mercury (top, right) and lead (bottom) concentration in mosses (2005/6) and EMEP modelled average annual deposition (2003 – 2005) per country.

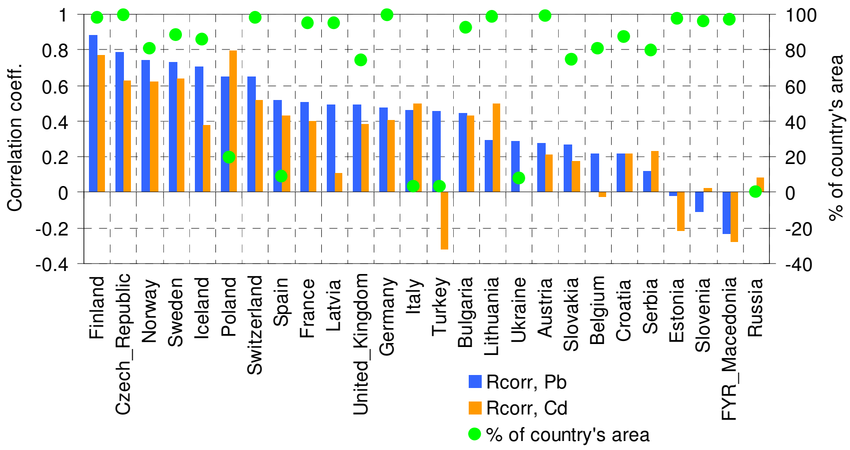

Figure 4. Correlation coefficients between modelled total depositions of lead and cadmium and concentrations in mosses surveyed in 2005/6 in European countries, and area of countries covered by moss measurements.

Measurements of mercury concentrations in air, precipitation and mosses are technically more difficult and likely to be more uncertain compared to those of cadmium and lead. Therefore, the interpretation of results for mercury is more complicated. From a viewpoint of modelling of atmospheric transport, uncertainties for mercury can also be high. That is why the correlation between depositions and concentrations of mercury in mosses is lower than that for lead or cadmium. For Switzerland, the correlation coefficient is 0.5 for 2005/6, for other countries it is lower or even negative.

Temporal trends
Initial data analysis indicated that the temporal trends in metal concentration in mosses agree reasonably well with the trends in metal deposition modelled by EMEP. Between 1990 (1995 for mercury) and 2005, the metal concentration in mosses had declined by 73, 46 and 20% across Europe, whereas the modelled deposition had declined by 70, 41 and 30% for lead, cadmium and mercury respectively (Figure 5).

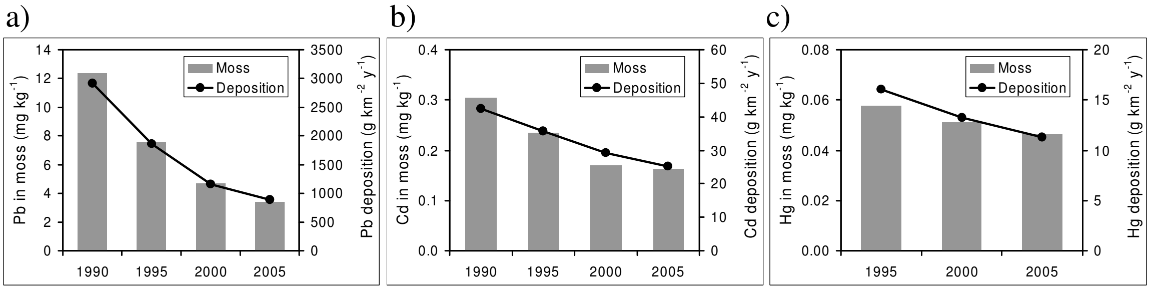

Figure 5. Temporal trends in the heavy metal concentration in mosses and EMEP modelled atmospheric deposition for (a) lead, (b) cadmium and (c) mercury. Source of deposition data: EMEP/MSC-East.

In summary, an initial analysis shows that for Europe as a whole, temporal trends in cadmium, lead and mercury concentrations in mosses agree well with the trends in depositions modelled by the EMEP atmospheric transport model. Further data analysis will be conducted in the future, in particular regarding country-specific temporal trends and factors that might contribute to discrepancies in temporal trends between heavy metal concentration in mosses and modelled atmospheric deposition.

3.1.3 Linking to other European databases: identifying factors contributing to the spatial variation of heavy metal concentrations in mosses

As a contribution in kind, Prof. Winfried Schröder and colleagues at the University of Vechta, Germany conducted a more detailed statistical analysis on factors influencing the spatial variation of heavy metal concentrations in mosses (Harmens *et al.*, 2009). Bivariate correlation coefficients were computed to indicate the strength and direction of the statistical relationship between the heavy metal concentrations in mosses and EMEP modelled depositions and additional factors that might influence the heavy metal concentration in mosses (see Table 1). These additional factors include both site-specific and regional characteristics. The moss data were linked to the following regional characteristics available from European databases:

- Precipitation, long-term monthly means (1961 - 1990) as provided by the Global Climate Dataset (CL 2.0) at a resolution of 12.5 x 12.5 km².
- Proportions of land use 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, 25 km (for forests and agriculture) or 1, 5, 10, 25, 50 and 100 km (for urban areas) projected onto 1 x 1 or 2 x 2 km² grid cells.
- 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.
- Population density in a resolution of 100 x 100 m^2 provided by the European Environment Agency.

Table 1. Spearman rank correlation coefficients (r_s) between heavy metal concentrations in mosses and i) EMEP modelled total depositions and emissions and ii) other site-specific or regional characteristics. All values are significant at P = 0.001, except * (P = 0.05); n.s. = not significant.

Independent variable	Cd	Hg	Pb
Cd concentration moss		0.06	0.65
Hg concentration moss			0.44
EMEP Cd depositions	0.63		
EMEP Hg depositions		0.20	
EMEP Pb depositions			0.73
EMEP Cd emissions	0.49		
EMEP Hg emissions		0.14	
EMEP Pb emissions			0.65
Altitude	0.14	-0.07	0.18
Precipitation	-0.05	0.17	0.11
Population Density	0.19	n.s.	0.16
Sea Distance	0.33	-0.18	0.31
Proportion of Urban Land Uses (1 km radius)	0.10	n.s.	0.09
Proportion of Urban Land Uses (100 km radius)	0.40	-0.17	0.41
Proportion of Agricultural Land Uses (1 km radius)	0.09	n.s.	0.18
Proportion of Agricultural Land Uses (25 km radius)	0.20	n.s.	0.31
Proportion of Forested Land Uses (1 km radius)	n.s.	n.s.	-0.06
Proportion of Forested Land Uses (25 km radius)	-0.03*	-0.04*	-0.14

Bivariate analysis of the data (Table 1) showed the highest correlations between the cadmium and lead concentration in mosses and i) modelled EMEP depositions ii) EMEP total emissions and iii) the proportion of urban land use in a 100 km radius. Correlations between the mercury concentration in mosses and modelled EMEP depositions or anthropogenic emissions were low.

Multivariate analyses showed that for cadmium and lead the modelled EMEP deposition was the main factor determining the variation of their concentration in mosses, whereas for mercury the variation of the concentration in mosses was primarily determined by the moss species sampled (Harmens *et al*., 2009).

3.2 Nitrogen

Within the LRTAP Convention, deposition of nitrogen is modelled using the Unified EMEP model (Simpson *et al*., 2003; Fagerli *et al*., 2004). The Unified EMEP model is designed to calculate air concentrations and deposition fields for major acidifying and eutrophying pollutants, photo-oxidants and particulate matter. The modelled data are verified against nitrogen concentrations in air and precipitation measured at EMEP monitoring stations. As with heavy metals, the number of EMEP monitoring stations and their spatial distribution across Europe is limited, with southern and eastern Europe being under-represented compared to northern and central Europe. In addition, there are only up to 24 EMEP stations which have had a long-term commitment to measuring nitrate and ammonium in precipitation since 1980 and measurements of concentrations of various nitrogen forms in air since the late 1980's (Fagerli and Aas, 2008). Compared to the data from the EMEP monitoring network, the moss survey has two advantages: i) the density of the moss monitoring network is much higher and ii) their spatial distribution is wider, including southern and eastern Europe. Although deposition fluxes of nitrogen and their concentrations in mosses cannot be compared directly, it is possible to compare the spatial distribution of deposition and concentrations in mosses.

3.2.1 Cross-validation with the EMEP atmospheric transport model

The spatial distribution of the nitrogen concentration in mosses was similar to that of the total nitrogen deposition modelled by EMEP for 2004, except that the modelled nitrogen deposition tended to be relatively lower in eastern Europe (Figure 6). However, the relationship between total nitrogen concentration in mosses and modelled total nitrogen deposition, based on averaging all sampling site values within any one EMEP grid square, showed considerable scatter (Figure 7a). Some of the scatter can be explained by relating site-specific nitrogen concentrations in mosses with total modelled nitrogen depositions averaged over a bigger area (50 x 50 km^2). Actual deposition values vary considerably within each EMEP grid cell due to for example topography, vegetation, local pollution sources and climate.

The apparent asymptotic relationship shows saturation of the total nitrogen in mosses above a nitrogen deposition rate of approximately 10 kg ha^{-1} y^{-1}. It is not clear, however, whether this is due to an overestimation of modelled deposition at these sites, or that it indicates a non-linear relation between nitrogen deposition and total nitrogen concentration in mosses. This exercise may be regarded as a cross validation of moss data and EMEP model data for nitrogen, but is complicated by both the limitations in the use of mosses as monitors of atmospheric nitrogen deposition and the uncertainties in the modelled nitrogen deposition, including uncertainties in emissions. When the total nitrogen concentration in mosses was plotted against site-specific nitrogen deposition values in for example Switzerland, a strong positive linear relationship was observed (Figure 6b). In addition, a good correlation was found between the nitrogen concentration in mosses and the EMEP modelled deposition fluxes in some of the Scandinavian countries (Harmens *et al.*, 2005). There is a need to measure atmospheric nitrogen deposition at selected moss sampling sites in other countries too in the future in order to further investigate the robustness of the relationship with total nitrogen concentration in mosses.

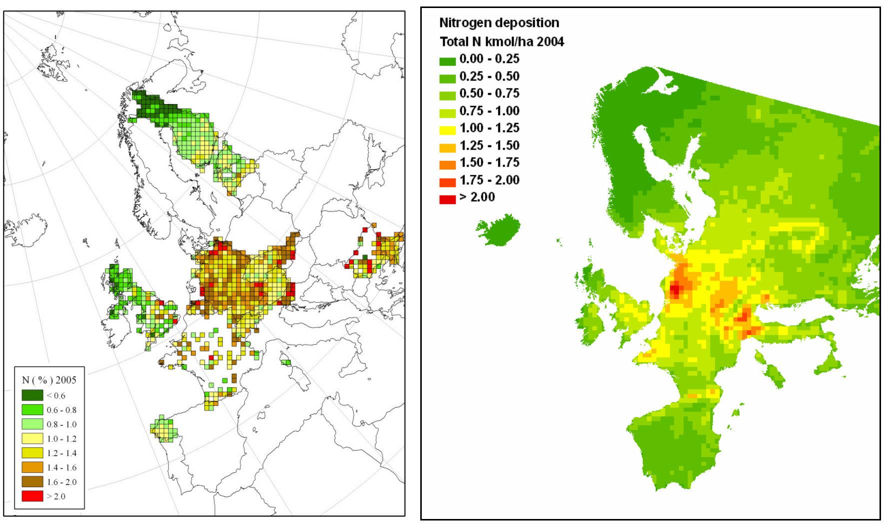

Figure 6. (a) Mean concentration of nitrogen in mosses per EMEP grid square in 2005/6 and (b) modelled nitrogen deposition per EMEP grid square in 2004. Source of deposition data: EMEP.

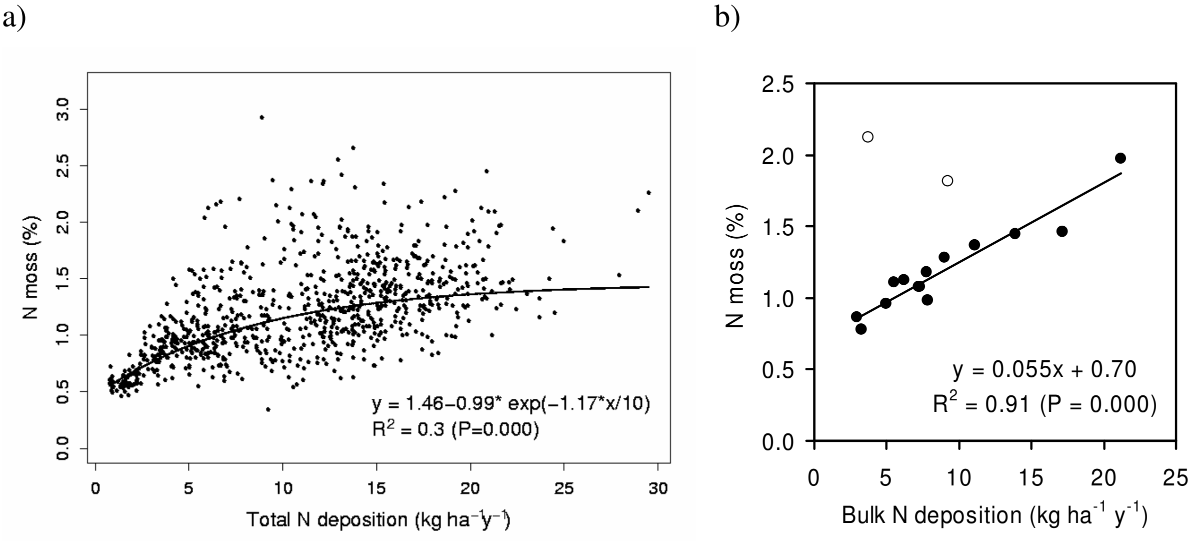

Figure 7. (a) Relationship between EMEP modelled total nitrogen deposition (2004) and averaged nitrogen concentration in mosses (2005/6) per EMEP grid square across Europe and (b) relationship between measured bulk nitrogen deposition rate and nitrogen concentration in mosses in Switzerland; the open symbols were excluded from the regression.

3.2.2 Linking to other European databases: identifying factors contributing to the spatial variation of nitrogen concentrations in mosses

As a contribution in kind, Prof. Winfried Schröder and colleagues at the University of Vechta, Germany, conducted a more detailed statistical analysis on factors influencing the spatial variation of nitrogen concentrations in mosses. Data on modelled nitrogen depositions and air concentrations were provided by EMEP/MSC-West. In addition to the predictors described above for heavy metals (see section 3.1.3), livestock density was included as a predictor, using data provided by EUROSTAT. Bivariate analysis of the data showed the highest, albeit moderate Spearman rank correlations between the total nitrogen concentration in mosses and modelled EMEP atmospheric deposition (r_s = 0.55 – 0.65) or air concentrations (r_s = 0.54 – 0.63) of various nitrogen forms (Table 2). However, these correlation coefficients were of a similar order of magnitude as the spatial correlation coefficients reported between EMEP modelled and measured concentrations of nitrogen concentration in air and precipitation. The EMEP model underestimates the nitrate and ammonium concentrations in precipitation by 10-30% and overestimates oxidised nitrogen concentration in air by ca. 30%; modelled reduced nitrogen concentrations in air are close to the measurement values (Fagerli and Aas, 2008). Moderate correlations were observed for the proportion of urban and agricultural land use (with correlations increasing with increasing radii), followed by population and livestock density. Low to very low correlations were found for the other tested predictors. In general, the total nitrogen concentration in mosses appears to mirror land use-related atmospheric nitrogen depositions.

Multivariate relations between the nitrogen concentration in mosses and modelled EMEP nitrogen depositions/air concentrations and other site-specific and regional land characteristics showed that the ammonium concentration in air was the most powerful predictor of the total nitrogen concentration in mosses, followed by nitrogen dioxide concentrations in air (at sites with ammonium concentrations below 0.63 mg m^{-3}) and moss species (at sites with ammonium concentrations above 0.63 mg m^{-3}; Harmens *et al.*, 2009).

Table 2. Spearman rank correlation coefficients (r_s) between total nitrogen concentrations in mosses and i) EMEP modelled depositions and air concentrations of different nitrogen forms and ii) other site-specific or regional characteristics. Correlation coefficients with EMEP depositions and air concentrations were based on median nitrogen concentrations in mosses per EMEP 50 x 50 km^2 grid. All correlations with $p < 0.001$ except for livestock density ($p < 0.01$).

EMEP modelled	N form	r_s	Other predictors	r_s
Air concentration	NO_2	0.54	Proportion urban land use (1 km radius)	0.15
	$NH_3 + NH_4^-$	0.61	Proportion urban land use (100 km radius)	0.55
	$HNO_3 + NO_3^-$	0.63	Proportion agricultural land use (1 km radius)	0.36
	Sum all N air	0.59	Proportion agricultural land use (50 km radius)	0.53
Deposition	Wet oxidised	0.65	Proportion forested land (1 km radius)	-0.11
	Total (wet + dry)	0.64	Proportion forested land (25 km radius)	-0.23
	Total wet	0.64	Population density	0.48
	Dry oxidised	0.64	Livestock density	0.42
	Wet reduced	0.62	Precipitation	0.25
	Total dry	0.59	Distance to sea	0.25
	Dry reduced	0.55	Altitude	-0.10

Further statistical analysis of the relationship between total nitrogen concentrations in mosses and EMEP modelled nitrogen depositions and air concentrations will be conducted in the future, testing EMEP data at a lower resolution (10 x 10 km^2 and 25 x 25 km^2) and averaging deposition data over longer time periods. This will allow further cross-validation of the modelled EMEP and moss data for nitrogen at a higher spatial resolution.

4. Use of moss data within the UK

4.1. Heavy metals

In the UK, the heavy metal concentrations in mosses for the most recent two surveys have been used to map deposition fields of heavy metal according to the procedure described by Ashmore *et al*. (2002). In 2005, the moss calibration (i.e. relationship between bulk deposition and concentration in moss) was applied as derived by co-located sampling of mosses at the monitoring sites of the rural Heavy Metal Deposition Network (HMDN) in 2004/5 (Fowler *et al*., 2006). The deposition fields and calculated total deposition of As, Cd, Ni, Pb, V and Zn derived from the 2005 moss survey (based on 168 sampling sites) were compared with the deposition maps and calculated total deposition from the UK HMDN for 2004 (based on 14 monitoring sites; Fowler *et al*., 2006). The monitoring sites in the rural deposition network and the sampling sites for mosses in the 2005 survey are shown in Figure 8. It should be noted that there is not always perfect agreement between the average of the grid cell and the moss samples for two reasons: firstly, the local map values are also affected by the surrounding measurement points, due to the interpolation routine used. Secondly, the grid cell reflects the weighted average to the different land-cover forms, while deposition to moss represents deposition to short vegetation only.

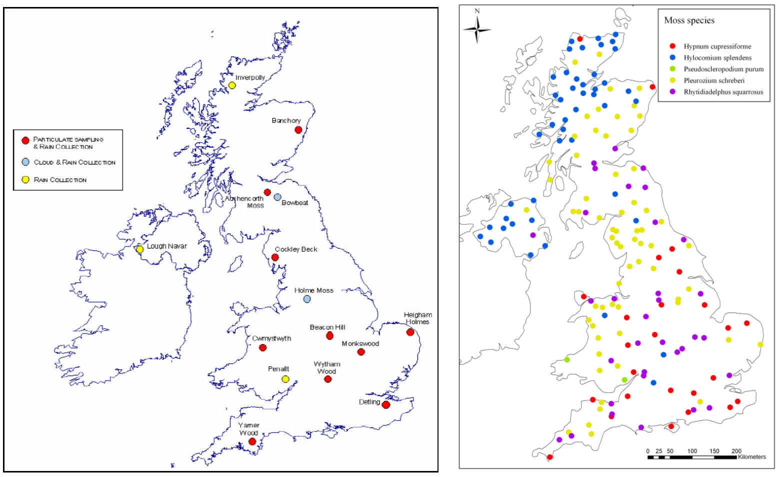

Figure 8. a) The UK rural heavy metals monitoring network (Fowler et al., 2006) and b) the sampling sites in the UK 2005 moss survey.

a) b) c) d)

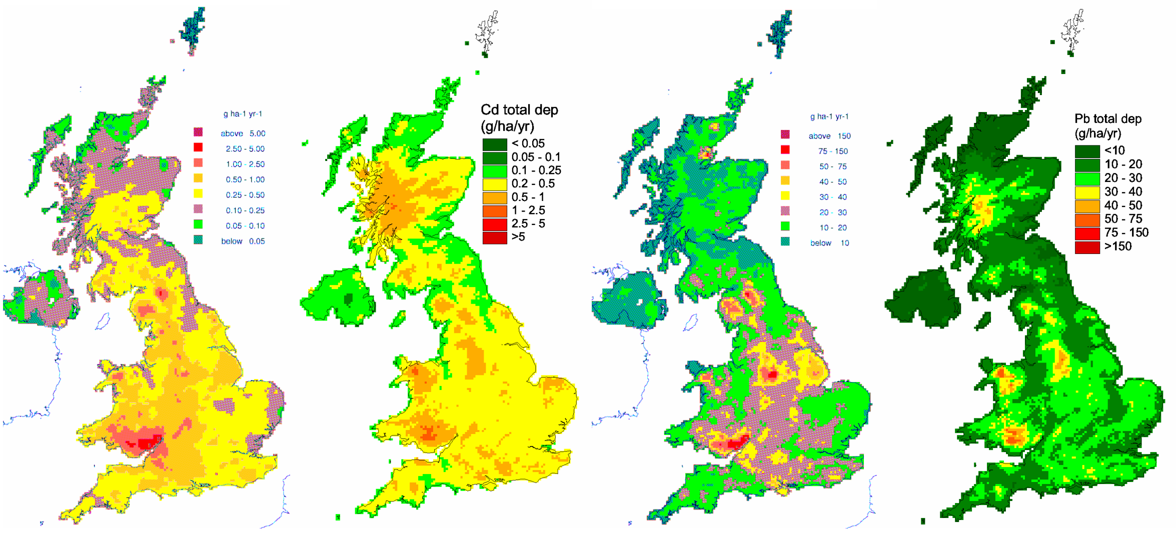

Figure 9. Comparison of cadmium (a, b) and lead (c, d) deposition maps, based on the 2005 moss survey (a, c) and the HMDN (b, d).

Both the total deposition values and spatial patterns were similar between the different deposition estimates of cadmium, lead (Figure 9) and vanadium, but different for arsenic, nickel and zinc. The total deposition estimates for arsenic and nickel based on the moss data were higher than those based on the HMDN, whereas the opposite was found for zinc. The higher values based on the moss data might be due to moss concentrations being affected by windblown dust representing historical deposition of these metals. The spatial patterns differed for arsenic, nickel and zinc for both methods. HMDN derives higher deposition estimates for the West Highlands of Scotland for cadmium and zinc. The HMDN approach has the advantage of being a more direct measurement of the deposition, but it is more likely to miss out localised hotspots, due to the smaller number of sites. In addition, elevated measurement values at a HMDN measurement station affects a large area as seen by the high estimates of deposition predicted for the West Highlands of Scotland.

4.2. Nitrogen

The concentration of tissue nitrogen was determined for 170 moss samples collected in 2005 from 170 sites distributed across the UK. These nitrogen concentrations have been compared with estimates of nitrogen deposition based on the Unified EMEP model for 2004 (see section 3.2.1) and the Concentration Based Estimated Deposition (CBED; Smith *et al.*, 2000) values for nitrogen averaged for 2003-2005. The CBED data consists of three sets of values:

- Average – for all vegetation types;
- Moorland – assuming all land cover is low growing vegetation;
- Woodland – assuming all land cover is woodland.

The Unified EMEP model (50 x 50 km^2 grid) tends to underestimate the total annual nitrogen deposition when compared with the CBED data (5 x 5 km^2 grid), but shows a higher correlation ($r = 0.46$) with the moss data than CBED ($r = 0.26$; Figure 10a). However, the correlation between CBED modelled nitrogen deposition and the concentration in mosses improves to the same level as that for EMEP data when the modelled habitat-specific rather than averaged deposition values are compared with the total nitrogen concentration in mosses (Figure 10b).

a) b)

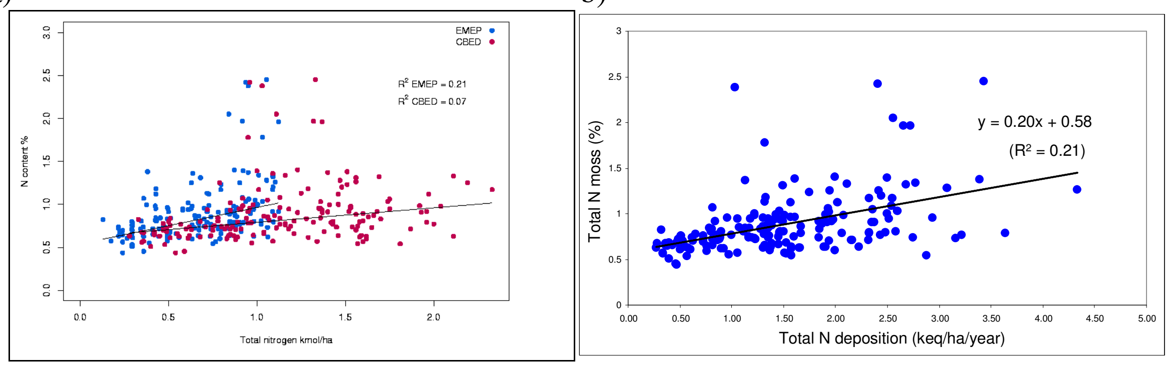

Figure 10. (a) Relationship between the N concentration in mosses (2005) and average CBED (2003 – 2005 annual average) or EMEP (2004) total nitrogen deposition estimates and (b) percent nitrogen in mosses versus habitat-specific total nitrogen deposition at UK sampling sites.

5. Discussion, conclusions and recommendations

Detailed statistical analysis has shown that variation in heavy metal and nitrogen concentration in mosses across Europe is best explained by variation in their atmospheric concentration and deposition. Deposition maps of heavy metals in the UK based on the moss data indicate that they are spatially most similar to measurement-based modelled deposition maps for those metals, such as cadmium and lead, which are not essential micronutrients for plants and therefore not metabolised in biological tissues. Lower correlations at the European level for mercury compared to cadmium and lead most likely reflect i) difficulties related with the analysis of mercury in atmospheric and plant material (and therefore higher uncertainties in the measurement data) and ii) higher uncertainties for modelling atmospheric transport of mercury.

Cross-validation of moss data and EMEP model data for heavy metals and nitrogen is complicated by both the limitations in the use of mosses as monitors of atmospheric deposition (Harmens *et al.*, 2008a,b) and the uncertainties in the modelled deposition, including uncertainties in emissions data. Although mosses seem to be more suitable as biomonitors of heavy metals than nitrogen, national studies in which bulk nitrogen depositions are actually measured at the moss sampling sites (Figure 6b) suggest that mosses can be suitable biomonitors of atmospheric nitrogen deposition at the local scale. More measurements of site-specific nitrogen deposition and other site-specific characteristics are required to establish the robustness of the relationship between total nitrogen deposition and total nitrogen concentration in mosses.

Recently it has been suggested that the total nitrogen concentration in mosses can potentially be used as a high spatial resolution tool in identifying Natura 2000 sites at risk from atmospheric nitrogen deposition (Harmens *et al.*, submitted). However, it is unclear yet whether the total nitrogen concentration in mosses can be used as early warning for nitrogen impacts on Natura 2000 habitats or as part of an integrated assessment of the state of habitats. Most likely, a combination of the bioindicators/biomonitors will best describe the state on Natura 2000 sites. Some countries (e.g. Denmark) already include measurements of the nitrogen concentration in mosses in an integrated assessment of the state of Natura 2000 sites (Andersen *et al.*, 2006).

Conclusions

1. Mosses are a cheap and useful tool for estimating spatial and temporal trends in atmospheric deposition of heavy metals and potentially nitrogen.
2. The European (including the UK) moss survey provides a measurement network with a higher spatial resolution than can ever be achieved with atmospheric deposition monitoring networks.
3. The moss network provides a useful tool for validating the performance of atmospheric deposition models at a high spatial resolution, in particular in areas in which EMEP monitoring stations are scarce or absent, i.e. in southern and eastern Europe.
4. For nitrogen, the moss data can be used as an additional parameter in an integrated assessment of the state of ecosystems and the identification of areas at risk from high atmospheric nitrogen pollution.

Recommendations

1. At its 21st meeting in 2008, the ICP Vegetation Task Force recommended to conduct the next European moss survey in 2010/11.
2. Based on this review and the decision of the ICP Vegetation Task Force, we recommend to continue the UK moss monitoring network and take part in the next European moss survey in 2010/11.
3. Considering the decline in atmospheric heavy metal deposition in recent decades, a reduction in the frequency of the European moss survey may be considered for later years.
4. We recommend to continue cross-validation between the moss survey data and atmospheric depositions modelled by EMEP, aiming at further identification of

factors potentially contributing to disparity between the data sets, in particular at country-level.

5. For nitrogen, cross-validation with EMEP modelled depositions at a higher spatial resolution (10 x 10 km^2 and 25 x 25 km^2) and longer-term annual average should be investigated.
6. We recommend that more European countries participate in the nitrogen survey in the future and conduct site-specific measurements of atmospheric nitrogen deposition fluxes in order to further investigate the robustness of the relationship with the total nitrogen concentration in mosses.

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Annex 1. Publications

Scientific papers

Published

Harmens, H., Norris, D.A., Koerber, G.R., Buse, A., Steinnes, E., Rühling, Å. (2008). Temporal trends (1990 – 2000) in the concentration of cadmium, lead and mercury in mosses across Europe. Environmental Pollution 151: 368-376.

Schröder, W., Pesch, R., Englert, C., Harmens, H., Suchara, I., Zechmeister, H.G., Thöni, L., Maňkovská, B., Jeran, Z., Grodzinska, K., Alber, R. (2008). Metal accumulation in mosses across national boundaries: uncovering and ranking causes of spatial variation. Environmental Pollution 151: 377-388.

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Harmens, H., Norris, D. and the participants of the European moss survey. Spatial and temporal trends in heavy metal accumulation in mosses in Europe (1990-2005). Environmental Pollution.

Harmens, H., Norris, D., Cooper, D., Hall, J. and the participants of the European moss survey. Spatial trends in nitrogen concentrations in mosses across Europe in 2005/2006. Environmental Pollution.

Schröder, W., Holy, M., Pesch, R., Harmens, H., Ilyin, I. and the participants of the European moss survey. Are cadmium, lead and mercury concentrations in mosses across Europe primarily determined by the atmospheric deposition of these metals? Journal of Soils and Sediments.

Schröder, W., Holy, M., Pesch, R., Harmens, H., Fagerli, H. and the participants of the European moss survey. First Europe-wide correlation analysis identifying factors best explaining the total nitrogen concentration in mosses. Atmospheric Environment.

Reports for the LRTAP Convention

Note: every year progress with the European moss survey is reported in the annual ICP Vegetation report and the technical documents of the Working Group on Effects.

In addition, data from the European moss survey have been used in the following reports for the Convention:

Harmens, H., Cooper, D., Norris, D., Schröder, W., Pesch, R., Holy, M., Fagerli, H. (2009). Comparison of modelled nitrogen deposition and nitrogen concentrations in mosses. In: Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe in 2007. EMEP Status Report 1/2009.

Ilyin, I., Rozovskaya, O, Sokovyh, V., Travnikov, O., Aas, W. (2009). Heavy metals: transboundary pollution of the environment. EMEP Status Report 2/2009.

Gusev, A., Iliyn, I., Rozovskaya, O., Shatalov, V., Sokovych, V., Travnikov, O. (2009). Modelling of heavy metals and persistent organic pollutants: new developments. EMEP/MSC-E Technical Report 1/2009.

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Moss survey reports and other reports containing moss survey data

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