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Country-specific correlations across Europe between modelled atmospheric cadmium and lead deposition and concentrations in mosses

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Abstract

Previous analyses at the European scale have shown that cadmium and lead concentrations in mosses are primarily determined by the total deposition of these metals. Further analyses in the current study show that Spearman rank correlations between the concentration in mosses and the deposition modelled by the European Monitoring and Evaluation Programme (EMEP) are country and metal-specific. Significant positive correlations were found for about two thirds or more of the participating countries in 1990, 1995, 2000 and 2005 (except for Cd in 1990). Correlations were often not significant and sometimes negative in countries where mosses were only sampled in a relatively small number of EMEP grids. Correlations frequently improved when only data for EMEP grids with at least three moss sampling sites per grid were included. It was concluded that spatial patterns and temporal trends agree reasonably well between lead and cadmium concentrations in mosses and modelled atmospheric deposition.

Capsule: For the majority of European countries a significant positive correlation was found between modelled atmospheric cadmium and lead deposition and concentration in mosses.

Keywords: biomonitoring; EMEP; heavy metals; metal deposition; bryophytes

1. Introduction

Since 1979, the Convention on Long-range Transboundary Air Pollution has addressed major air pollution problems in the UNECE (United Nations Economic Commission for Europe) region through scientific collaboration and policy negotiation. The Convention has been extended by eight protocols that identify specific measures to be taken by countries to cut their emissions of air pollutants. The 1998 Aarhus Protocol on heavy metals targeted three harmful heavy metals (cadmium (Cd), lead (Pb) and mercury (Hg)) and entered into force in 2003. Within the Convention, the European Monitoring and Evaluation Programme (EMEP) i) collects emission data from Parties, ii) measures air and precipitation quality, and iii) models atmospheric transport and deposition of air pollutants. Deposition of the heavy metals Cd, Hg and Pb is modelled using the EMEP atmospheric transport model MSCE-HM (Travnikov and Ilyin, 2005) and is calculated from official emission data reported by the countries. The modelled data are verified against 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: in the period from 1990 to 2009 there were between 40 to 77 stations annually reporting measurement data on heavy metals to EMEP (http://www.nilu.no/projects/ccc/index.html). The EMEP monitoring network for Cd and Pb is scarce or absent in the southern and eastern parts of Europe, whereas Hg is primarily measured in northern Europe.

Under the Working Group on Effects of the Convention, the ICP Vegetation (International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops) has been coordinating the European moss survey since 2000. The survey has been repeated at five-yearly intervals since 1990 and the latest survey was conducted in 2005/6 with 28 countries participating and mosses being sampled at almost 6,000 sites across Europe. The European moss survey provides data on concentrations of twelve trace elements (Al, As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, V, Zn) in naturally growing mosses (Harmens et al., 2010). In 2005/06, the N concentration in mosses was also determined (Harmens et al., 2011b) and in the current ongoing survey in 2010/11, a pilot study was initiated in selected countries to determine the concentration of selected persistent organic pollutants (POPs), particularly polycyclic aromatic hydrocarbons (PAHs), in mosses (Harmens et al., 2011a).

In recent decades, mosses have been applied successfully as biomonitors of heavy metal deposition (Harmens et al., 2007, 2008b, 2010; Zechmeister et al., 2003) across Europe. Heavy metal concentrations in mosses provide a complementary, time-integrated measure of the spatial patterns and temporal trends of heavy metal deposition from the atmosphere to terrestrial systems, at least for the metals Cd and Pb (Aboal et al., 2010). It has been shown that at the European scale atmospheric deposition is the main factor determining the accumulation of Cd and Pb in mosses (Holy et al., 2010; Schröder et al., 2010). Compared to the EMEP monitoring network, the moss survey has the following main 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. Although the heavy metal concentration in mosses provides no direct quantitative measurement of deposition, this information has been derived in some countries by using regression or correlation approaches relating the results from moss surveys to deposition data (e.g. Berg and Steinnes, 1997; Berg et al., 2003; Schröder and Pesch, 2010; Thöni et al., 2011). Based on statistical relations between concentrations of Cd and Pb in modelled atmospheric deposition and mosses across Europe, deposition maps with a spatial resolution of 5 km by 5 km were calculated using a regression kriging approach for Germany (Schröder et al., 2011). However, based on a recent study, Bouquete et al. (2011) recommended that the results of moss biomonitoring studies should be regarded as qualitative or semi-qualitative, rather than attempting to provide absolute data, which may not be temporally representative, and may have a high degree of uncertainty associated with them, at least in Spain.

In the current study, we analysed in more detail the relationship between EMEP modelled atmospheric deposition of Cd and Pb and their concentration in mosses for individual European countries. Although previous studies have shown good correlations between both parameters at the European scale, other factors also contribute to the variation of Cd and Pb concentrations in mosses (Harmens et al., 2008b; Holy et al., 2010; Schröder et al., 2010). As these factors and their influence on the relationship is likely to be different for different countries and/or climatic regions (e.g. Thöni et al., 2011), we hypothesise that the correlations between both parameters will be country-specific, with good correlations expected in some but less good correlations expected in other countries.

2. Materials and methods

Determination of Cd and Pb concentrations in mosses

Moss samples were collected across Europe in 1990/1 (Rühling, 1994), 1995/6 (Rühling and Steinnes, 1998), 2000/1 (Buse et al., 2003) and 2005/6 (Harmens et al., 2008a, 2010). Throughout the paper we refer to the years of moss survey as 1990, 1995, 2000 and 2005 respectively. Because the mosses were collected in a range of habitats from the sub-arctic climate of northern Scandinavia to the hot and dry climate of southern Europe, it was not possible to sample just one carpet-forming moss species across Europe. *Pleurozium schreberi* was the most frequently sampled species in all surveys, accounting for 40.8 – 52.7% of the samples, followed by *Hylocomium splendens* (20.5 – 39.3%), *Hypnum cupressiforme* (7.4 – 22.0%) and *Pseudoscleropodium purum* (3.4 – 11.9%); other species constituted only 2.2 – 6.5% of the mosses sampled. The moss sampling procedure was according to the guidelines described in the protocol for the 2005 survey (ICP Vegetation, 2005). Only the last three years' growth of moss material was used for the analyses. The concentrations of Cd and Pb were determined by a range of analytical techniques; for further details we refer to the reports of the individual surveys (Buse et al., 2003; Harmens et al., 2008a, 2010; Rühling, 1994; Rühling and Steinnes, 1998). A comprehensive quality control exercise was conducted in 1995 (Steinnes et al., 1997) and 2005 (Harmens et al., 2010) with moss reference material being distributed amongst participating laboratories. In addition, some laboratories used other certified reference material for quality assurance. Recommended values were established in 1995 for moss reference material. For example, the recommended values for Cd and Pb for moss reference M2 were 0.454 ± 0.019 and 6.37 ± 0.43 mg kg^{-1} (mean ± standard deviation) respectively and 0.106 ± 0.005 and 3.33 ± 0.25 mg kg^{-1} respectively for moss reference M3 (Steinnes et al., 1997). No amendment of these recommended values was required in 2005 (Harmens et al., 2010). For further details we refer to Steinnes et al. (1997) and Harmens et al. (2010).

Modelling the deposition of Cd and Pb

Deposition of the heavy metals Cd and Pb was modelled using the EMEP atmospheric transport model MSCE-HM (Travnikov and Ilyin, 2005). MSCE-HM is a three-dimensional Eulerian-type chemical transport model driven by off-line meteorological data. The model takes into account heavy metal emissions from anthropogenic and natural sources, wind re-suspension of dust particles containing heavy metals, transport in the atmosphere, chemical transformations of mercury and ecosystem-dependent deposition to the surface. The model computation domain is defined on the polar stereographic projection. Its spatial resolution is 50 km × 50 km at 60°N. Modelled deposition of heavy metals was calculated from official emission data reported by the countries. The modelled data were verified against concentrations in air and precipitation measured at EMEP monitoring stations. The intrinsic uncertainty of the model (without uncertainty in reported emission data) is about 30 – 40% for concentrations in air, concentrations in precipitation and total deposition for Cd and Pb (Travnikov and Ilyin, 2005). The uncertainty of country-specific totals of heavy metal emission typically ranged between 30 – 60% and the overall uncertainty of measured wet deposition was around 20% for Cd and Pb. Modelling results agreed with measurement data with satisfactory accuracy, keeping in mind uncertainties of the emission and monitoring data. At most of the monitoring stations modelled and observed levels of Cd and Pb agreed within ±50% and the spatial correlation coefficient between modelled and observed values is between 0.6 – 0.9 (Ilyin et al., 2010).

Correlations between EMEP modelled deposition and concentrations in mosses

Country-specific Spearman rank correlations between various forms of EMEP modelled atmospheric deposition (dry, wet and total deposition) and concentrations in mosses for Cd and Pb were determined using SigmaPlot version 11. In this investigation, we computed the Spearman rank correlation coefficient r_s because the metal concentrations mostly proved not to be normally distributed. Although this non-parametric correlation method is less powerful than parametric methods if the assumptions underlying the latter are met, it is less likely to give distorted results when the assumptions fail. The coefficient r_s equals −1, if the two rankings are completely opposite to each other, r_s equals 0 if the rankings are completely independent and +1 if there is complete agreement between the two rankings. Within the interval [−1, +1] the strength of correlation can be classified as follows: 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 (Schröder et al., 2010).

As the last three years of moss growth was selected for heavy metal determination, representing the accumulation of Cd and Pb in mosses in the three years previous to sampling (ICP Vegetation, 2005), EMEP data were accumulated and averaged over the previous three years where possible. For 1990, the EMEP modelled data for 1990 were used as data for earlier years was not available. To assess the impact of using EMEP modelled data averaged over three years in comparison to modelled data for the year previous to moss sampling, correlations were also determined using only the EMEP modelled data for the year previous to moss sampling. Individual moss data were averaged per 50 km x 50 km EMEP grid before correlations were calculated. Moss data outside the mean ± 3 standard deviations were eliminated from the analysis leading to exclusion of 2 − 3% of the moss data.

In addition to calculating Spearman rank correlations, the moss concentration and modelled deposition data for individual countries were also normalized to their European mean values to assess the resemblance between spatial patterns for both data sets. For total deposition only EMEP grid cells were included where mosses were sampled, hence for calculation of the normalized values only data from the areas of the countries where mosses were sampled were used. For calculation of the European mean, the data per country were weighted by the area of EMEP grid cells in which mosses were sampled, i.e. more weight was given to countries where mosses were sampled in more grid cells. The normalized value of a country was then calculated as the mean concentration in mosses or mean total modelled deposition of that country divided by the European mean value for mosses or deposition respectively. Finally, temporal trends were compared per country for both datasets, including only data for EMEP grid cells where mosses were sampled in every survey year.

3. Results and discussion

Correlations between EMEP modelled deposition and concentrations in mosses

Previous analyses had indicated that total atmospheric deposition of Cd and Pb is the main factor explaining the variation in Cd and Pb concentrations in mosses across Europe (Holy et al., 2010; Schröder et al., 2010). However, other factors also contribute to spatial variation of heavy metal concentrations in mosses, including for example the variation in moss species sampled across Europe, land use in the area surrounding the moss sampling sites, altitude and competition for sea salt ions in coastal areas (Steinnes, 1995; Harmens et al., 2008b; Holy et al., 2010; Schröder et al., 2010). In addition, the temporal variability of metal concentrations can be high in some countries (Bouquete et al., 2011). In the current study, country- and metal-specific correlations were observed (Table 1 and 2, Figure 1 and 2) and correlations varied between years (Table 1). High correlations ($0.7 \leq r_s < 0.9$) were generally observed for the Czech Republic (except for 1990), Finland, Sweden, and for Pb also in Norway for the earlier years (Table 1). Moderate correlations ($0.5 \leq r_s < 0.7$) were generally found in France (for Pb in particular), Norway (for Cd) and Poland (with sometimes high correlations being observed). Other countries with moderate to high correlations for at least one of the metals for at least two survey years include Bulgaria, Iceland, Latvia, Switzerland, Ukraine and the United Kingdom. The generally lower correlations in Norway compared to Finland and Sweden might be related to the more complex topography of Norway with orographic deposition having a greater role. In addition, the lower correlations for Cd might be due to the competition with sea salt ions in the extensive coastal area of Norway (Steinnes, 1995). Significant positive correlations were found for about two thirds or more of the participating countries (except for Cd in 1990). As to be expected, non-significant or significant negative correlations were mainly found in smaller countries or in countries where mosses were sampled in a smaller number of grid squares (< 60), although this was not always the case (e.g. Iceland for Pb, Latvia, Ukraine, Switzerland). Negative correlations were significant only twice, i.e. in 1990 for Cd in Lithuania and Portugal.

As the heavy metal concentrations in mosses were determined over the last three years of growth before the date of sampling, it was assumed that the concentration in mosses represents the accumulation of Cd and Pb atmospheric deposition over the same period. Therefore, the metal concentration in mosses was compared with the average EMEP modelled annual deposition for the three years previous to moss sampling. However, this was not feasible for 1990 as only modelled annual deposition data was available for 1990 and not for the previous years. To investigate whether this would have any effect on the determined Spearman rank correlations, we also calculated the Spearman rank correlations based on the

EMEP modelled total deposition for the year previous to moss sampling for the years 1995 - 2005. As an example, the results for 2000 are shown in Table 2. The results for the year 2000 clearly indicate that the correlations per country are hardly affected by which EMEP modelled total deposition data were used. This might be explained by the fact that the relationship between the 1997 – 1999 (annual average) and 1999 EMEP modelled total deposition data was significantly linear with the 1999 values in general being slightly lower than the annual averages for 1997 – 1999 (data not shown). The previous European scale analyses had reached the same conclusion (Holy et al., 2010; Schröder et al., 2010). Therefore, the correlations determined for 1990 are not likely to be affected by the fact that only one year of EMEP modelled total deposition data was used.

An alternative explanation for the fact that correlations are hardly affected by the accumulation period for modelled deposition might be that the metal concentrations in mosses do not reflect the integration of air pollutants over a certain period as the moss might be in an unstable equilibrium with its environment, resulting in a high temporal variability of heavy metal concentrations in mosses (Boquete et al., 2011; Couto et al., 2004). However, whether this is true for other moss species than *Pseudoscleropodium purum* and for other climate conditions than the Mediterranean requires further investigation. For example, Berg and Steinnes (1997) and Thöni et al. (1996) found no seasonal variation in heavy metal concentrations for the moss species *Hylocomium splendens* and *Pleurozium schreberi* in Norway and Switzerland, respectively. The equilibrium between mosses and the environment is complex and depends on various factors. Inputs and outputs of elements in moss will depend on physicochemical (e.g. solubilization and leaching of elements, cation competition, anionic complexation) and biological processes (e.g. rate and type of growth, physiological activity, phenotypic adaptations). In addition, all of these variables will depend on environmental factors (pH, salinity, temperature), which may vary within short periods of time (Bouquette et al., 2011).

The impact of precipitation on the concentration of elements in mosses is unclear and there is no evidence that the intensity or frequency of precipitation affects heavy metal concentrations in mosses. On the one hand, rainwater may wash the moss, resulting in removal of the particulate material deposited on its surface, on the other hand, rainwater may dissolve elements adsorbed on moss tissue, thus facilitating their uptake. The most important way that precipitation may influence metals already present on the moss surface might be via exchange with other cations, including those from marine origin (Gjengedal and Steinnes, 1990; Steinnes et al., 1995). In the current study, Spearman rank correlations between Cd and Pb concentrations in mosses and EMEP modelled deposition (total, wet or dry) were not significantly affected by the fraction of EMEP modelled wet deposition (data not shown). The distribution of the annual deposition between the wet and dry part is controlled by a combination of two main factors: 1) the annual sum of precipitation and 2) the distribution of forests – dry deposition velocity of particles (and hence particulate species like Cd and Pb) to areas with tall vegetation is greater than that to areas with short vegetation. The combination of these two factors results in a relatively high fraction of dry deposition in central Europe (e.g. Czech Republic, Germany, Poland with moderate precipitation and high forested area) and in regions of southern Europe (Portugal, southern Spain, Italy). In the recent surveys mosses were sampled in northern Spain and Italy in mountainous regions with higher annual precipitation than other parts of these countries.

We also investigated whether Spearman rank correlations were affected by the form of atmospheric deposition, i.e. wet, dry and total deposition of Cd and Pb. The results show that indeed the correlations are affected by the form of atmospheric deposition, as shown in Figure 1 for 2000. As mosses accumulate both dry and wet deposition of heavy metals, one might expect the correlations to be highest for total atmospheric deposition, however this was not always the case. For Pb, 74% of the countries that had significant Spearman rank correlations showed the highest correlations with total deposition in 2000. However, for Cd this was the case for only 50% of the countries, with 22 and 28% of the countries showing the highest correlations with wet and dry deposition respectively in 2000. For other years of the survey, the number of countries showing the highest correlations with total deposition was also higher than the number of countries showing the highest correlations with either wet or dry deposition, except for Cd in 2005 when only 27% of the countries showed the highest correlation with total deposition. As to be expected, the highest variations in correlations for different atmospheric deposition forms (with often very low or even negative correlations) were generally observed in countries where mosses were sampled in a relatively small number of EMEP grid squares ($N \leq 30$), such as Belgium, Estonia, Hungary, Macedonia, Slovakia, Spain and Switzerland for 2000 (Figure 1).

In a previous study with nitrogen we found that the relationship between the total nitrogen concentration in mosses and EMEP modelled total atmospheric nitrogen deposition for Europe improved when the relationship was based on data for EMEP grid squares where at least five moss sampling sites were present (Harmens et al., 2011). This can be explained by the fact that atmospheric deposition of air pollutants is highly variable within each EMEP grid due to for example non-uniform distribution of emission sources within the grid cell, variation in roughness of vegetation including mosses, sub-grid variability of meteorological parameters, and orographic effects. Therefore, a single measurement of concentration in mosses can hardly characterize conditions for the model grid cell as a whole. Hence, for heavy metals we would also expect an improvement of the correlations between concentrations in mosses (site specific) and EMEP modelled total atmospheric deposition (per 50 km x 50 km grid) if EMEP grids with only one or two moss sampling sites were excluded from the analysis. Indeed, in the majority of countries (>70%) there was an improvement in the correlations, which appeared to be most pronounced for Pb (Figure 2). The improvement in correlations is observed despite the fact that the number of EMEP grids with the required data is lower than when all EMEP grids with moss data are included.

As for the nitrogen study (Harmens et al., 2011) we also compared the correlations including all EMEP grids with the correlations including EMEP grids with at least five moss sampling sites (data not shown). However, in contrast to the European-wide relationship established for nitrogen, in the current country-specific analysis with heavy metals the number of EMEP grids with at least five sampling sites was low in many countries, resulting in a decline in the number of significant correlations (i.e. correlations were significant in only eight of the 15 countries for which correlations could be determined) compared to including all EMEP grids or EMEP grids with at least three moss sampling sites (Figure 2). Only four (i.e. Czech Republic, Finland, Germany and Sweden) out of the 17 countries included in Figure 2 had more than 25 EMEP grids with at least five moss sampling sites, whereas in seven out of the 17 countries the number of EMEP grids with at least five moss sampling sites was less than 15.

Spatial patterns and temporal trends in moss concentrations and modelled deposition

To compare the spatial patterns of Cd and Pb concentrations in mosses and EMEP modelled total deposition, both datasets were normalized against the European mean (see Materials and Methods for details). Figure 3 and 4 show that the spatial patterns for 2005 are quite similar, i.e. regions in Europe with a deposition rate below (e.g. big parts of northern and western Europe) or above the European mean (e.g. Belgium, eastern part of Europe and parts of central Europe) also showed concentrations in mosses below or above the European mean respectively, particularly for Cd. Nevertheless some discrepancies can be observed: For Cd, modelled deposition is relatively high (i.e. the ratio of normalized deposition to moss concentration >1.5) in Macedonia, Spain and Lithuania compared to the concentration in mosses. The opposite is true for Belgium, Finland and the Russian Federation (i.e. the ratio of normalized moss concentration to deposition >1.5). For Pb, modelled deposition is relatively high in the Czech Republic, Germany and Iceland, whereas the opposite is true for Belgium, Bulgaria, Italy, Slovakia and Ukraine. The relatively high Pb concentrations in mosses in these countries result in relatively low normalized values in central Europe in comparison with the normalized deposition values (Figure 4).

Previously we reported on the similarity between temporal trends observed for Cd and Pb concentrations in mosses and EMEP modelled total deposition between 1990 and 2005 at the European scale (Harmens et al., 2011). In the current study, we compared the temporal trends in further detail for individual countries. Some examples are shown in Figure 5 and in general the temporal trends in concentrations in mosses agree reasonably well with the temporal trends in deposition at the national scale too. Nevertheless, in certain periods the decline of calculated deposition seemed to be underestimated in comparison to the decline of concentrations in mosses (e.g. for Cd in Lithuania and the Czech Republic between 1990 and 1995, for Pb in Estonia between 1990 and 1995) or vice versa (e.g. for Cd in Poland between 1995 and 2000 or for Pb in Slovakia between 1995 and 2000). The sometimes higher concentrations of Cd (Czech Republic and Lithuania) and Pb (Estonia) in mosses compared to modelled deposition in 1990 might reflect the presence of more local pollution sources that affect concentrations in mosses but are not included in modelled deposition. In many countries smaller local pollution sources closed down in the early 1990s. On the other hand, it might reflect more inadequate emission inventories in 1990 compared to later years.

Considering the uncertainties in the EMEP modelled deposition data (see Introduction) and the potential limitations and confounding factors in the use of mosses as monitors of atmospheric deposition (Aboal et al., 2010; Boquete et al., 2011; Harmens et al., 2008b; Steinnes, 1995), the spatial patterns and temporal trends of both data sets agree reasonably well for Cd and Pb. The results confirm once again that Cd and Pb concentrations in mosses can serve as a complementary method to determine spatial patterns and temporal trends of Cd and Pb deposition (Aboal et al., 2010; Harmens et al., 2010). Currently EMEP is conducting a case study to assess heavy metal pollution at country-scale levels, employing a spatial resolution finer (e.g. 5 km x 5 km) than that currently used (50 km x 50 km). The European moss survey will provide valuable field-based measurement data for the validation of the finer-resolution modelled atmospheric deposition (Ilyin et al., 2011).

4. Conclusions

The following main conclusions can be drawn:

- For Cd and Pb the correlations between concentrations in mosses and the EMEP modelled total atmospheric deposition are country- and metal-specific, with sometimes considerable variation being observed between years. However, significant positive correlations were found for about two thirds or more of the participating countries (except for Cd in 1990). Non-significant or significant negative correlations (only two) were mainly found in smaller countries or in countries where mosses were sampled in a relative small number of EMEP grid squares;
- Correlations were generally not affected by using EMEP modelled deposition data for the year previous to sampling or averaged over three years previous to sampling of the mosses. As expected, correlations mainly improved when the analysis was limited to using EMEP grids in which at least three moss sampling sites were present;
- For the majority of countries across Europe, the use of mosses as biomonitors of atmospheric deposition for Cd and Pb provides a valid, complementary method for

assessing the spatial patterns and temporal trends of atmospheric deposition for these metals.

The current study confirms that environmental monitoring programmes such as the moss survey are appropriate tools for national regulatory bodies in many European countries to assess the efficiency and effectiveness of national air pollution abatement strategies for the metals Cd and Pb. To further investigate the relationship between atmospheric deposition of Cd and Pb and their concentration in mosses and the robustness of this relationship, we recommend that countries sample mosses at EMEP monitoring stations and/or national deposition monitoring stations. The presence of a dense national heavy metal deposition monitoring network and measurement of concentrations in mosses at the same sites is likely to reduce the uncertainty in modelled deposition data and might provide further insight into why in one-third of the countries correlations were not significantly positive between the two data sets.

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Figure legends

Figure 1. Spearman rank correlation coefficients (r_s) between the EMEP modelled deposition (total, wet and dry) and concentrations in mosses for Cd and Pb for the moss survey year 2000. The modelled deposition data was based on the annual average of the three year sum for 1997 − 1999.

Figure 2. Spearman rank correlation coefficients (r_s) between the EMEP modelled total deposition and concentrations in mosses for Cd and Pb for the moss survey year 2005. Correlations are shown for all EMEP grids (black bar) where mosses were sampled or only for EMEP grids with at least three moss sampling sites (white bar) in at least 15 grids. The modelled deposition data was based on the annual average of the three year sum for 2003 − 2005.

Figure 3. Normalized values (relative to the overall European mean) of the average Cd and Pb concentration in mosses (2005/6) and EMEP modelled average total annual deposition (2003 – 2005) per country. For the calculation of the normalized values the areas of the countries where mosses were sampled were taken into account.

Figure 4. Maps of the normalized values per country (relative to the overall European mean) of the (top left) average Cd concentration in mosses (2005/6), (top right) EMEP modelled average annual total Cd deposition (2003 – 2005), (bottom left) average Pb concentration in mosses (2005/06) and (bottom right) EMEP modelled average annual total Pb deposition (2003 – 2005). For the calculation of the normalized values the areas of the countries where mosses were sampled were taken into account.

Figure 5. Examples of temporal trends of concentrations in mosses (bars) and EMEP modelled total atmospheric deposition (lines) for Cd (charts on the left) and Pb (charts on the right) between 1990 and 2005 for selected European countries.

Table 1. Spearman rank correlation coefficients (r_s) between the EMEP modelled total deposition data (annual average of three year sum) and concentrations in mosses for Cd and Pb for those countries that participated in at least three moss surveys during 1990 – 2005. N = number of EMEP grid cells (50 km x 50 km) for which moss data was available; n.d. = not determined; values in bold: $P \leq 0.05$.

	Cadmium (Cd)				Lead (Pb)				N			
Country	1990	1995	2000	2005	1990	1995	2000	2005	1990	1995	2000	2005
Austria	0.08	0.05	**0.34**	**0.30**	**0.52**	0.16	0.23	**0.39**	34	56	56	56
Bulgaria	n.d.	**0.45**	**0.57**	**0.56**	n.d.	**0.42**	**0.33**	**0.42**	n.d.	65-66	66-70	66-68
Czech Republic	0.09	**0.85**	**0.75**	**0.78**	0.19	**0.72**	**0.80**	**0.81**	18	44-45	48	47
Estonia	0.21	-0.20	-0.31	0.08	0.10	-0.05	-0.09	n.d.	28	30	30	31
Finland	**0.88**	**0.67**	**0.76**	**0.83**	**0.86**	**0.82**	**0.89**	**0.83**	158	159	156	151
France	n.d.	**0.52**	**0.42**	**0.47**	n.d.	**0.56**	**0.56**	**0.58**	n.d.	245-246	260	267
Germany	**0.49**	**0.52**	**0.43**	**0.39**	**0.33**	**0.45**	**0.44**	**0.43**	153-181	184-185	185	186
Iceland	0.13	0.05	0.21	**0.43**	**0.71**	**0.33**	**0.69**	**0.66**	43	45-46	44-45	45-46
Latvia	0.18	**0.65**	**0.70**	0.18	**0.53**	**0.37**	**0.50**	**0.39**	37	34	35-36	33-34
Lithuania	**-0.37**	-0.10	**0.40**	**0.52**	-0.10	**0.35**	0.30	0.26	37	38	37	37
Norway	**0.54**	**0.65**	**0.58**	**0.53**	**0.77**	**0.81**	**0.72**	**0.63**	179	176	172-173	176-177
Poland	**0.60**	**0.53**	**0.73**	**0.75**	**0.53**	**0.58**	**0.84**	**0.57**	112	145	35	35-36
Portugal	**-0.33**	0.09	0.14	n.d.	0.23	0.12	**0.39**	n.d.	55	53	53-54	n.d.
Slovakia	-0.45	0.42	0.00	0.05	**0.70**	0.16	0.07	0.09	18	21	24	21
Spain	n.d.	0.16	0.05	**0.40**	n.d.	0.01	0.21	**0.52**	n.d.	68-69	24	30
Sweden	**0.81**	**0.74**	**0.76**	**0.72**	**0.88**	**0.79**	**0.78**	**0.70**	199	200-202	173	183-184
Switzerland	**0.56**	**0.41**	**0.60**	**0.48**	**0.67**	0.32	0.29	**0.53**	28	29	29	29
Ukraine	n.d.	**0.50**	**0.53**	-0.07	n.d.	**0.39**	**0.58**	0.21	n.d.	38-39	32	23
United Kingdom	0.22	**0.46**	**0.71**	**0.47**	**0.54**	**0.48**	**0.70**	**0.60**	61-64	66	103	99

Table 2. Spearman rank correlation coefficients (r_s) between the EMEP modelled total deposition and concentrations in mosses for Cd and Pb for the moss survey year 2000. The modelled total deposition data was based on either the annual average of the three year sum (3 year) or the annual deposition in the year before moss sampling (1 year). N = number of EMEP grid cells (50 km x 50 km) for which moss data was available; values in bold: P ≤ 0.05.

	Cadmium (Cd)		Lead (Pb)		
Country	**3 year**	**1 year**	**3 year**	**1 year**	**N**
Austria	**0.34**	**0.29**	0.23	0.23	56
Belgium	0.14	0.24	-0.48	-0.49	15
Bulgaria	**0.57**	**0.62**	**0.33**	**0.32**	66-70
Czech Republic	**0.75**	**0.75**	**0.80**	**0.81**	48
Estonia	-0.31	-0.15	-0.09	-0.04	30
Finland	**0.76**	**0.77**	**0.89**	**0.89**	156
France	**0.42**	**0.42**	**0.56**	**0.58**	260
Germany	**0.43**	**0.42**	**0.44**	**0.44**	185
Hungary	-0.16	-0.20	0.35	0.25	27
Iceland	0.21	0.21	**0.69**	**0.68**	44-45
Italy	**0.34**	**0.31**	**0.32**	**0.32**	80-88
Latvia	**0.70**	**0.70**	**0.50**	**0.51**	35-37
Lithuania	**0.40**	**0.43**	0.30	**0.38**	37
Macedonia	0.02	0.07	-0.31	-0.44	17
Norway	**0.58**	**0.61**	**0.72**	**0.75**	172-173
Poland	**0.73**	**0.73**	**0.84**	**0.82**	34-35
Portugal	0.14	0.17	**0.39**	**0.38**	53-54
Russian Federation	-0.02	-0.01	**0.49**	**0.48**	74-82
Slovakia	0.00	-0.02	0.07	0.13	24
Spain	0.05	0.06	0.21	0.27	24
Sweden	**0.76**	**0.78**	**0.78**	**0.78**	173
Switzerland	**0.60**	**0.57**	0.29	0.24	29
Ukraine	**0.53**	**0.53**	**0.58**	**0.58**	32
United Kingdom	**0.71**	**0.67**	**0.70**	**0.67**	103

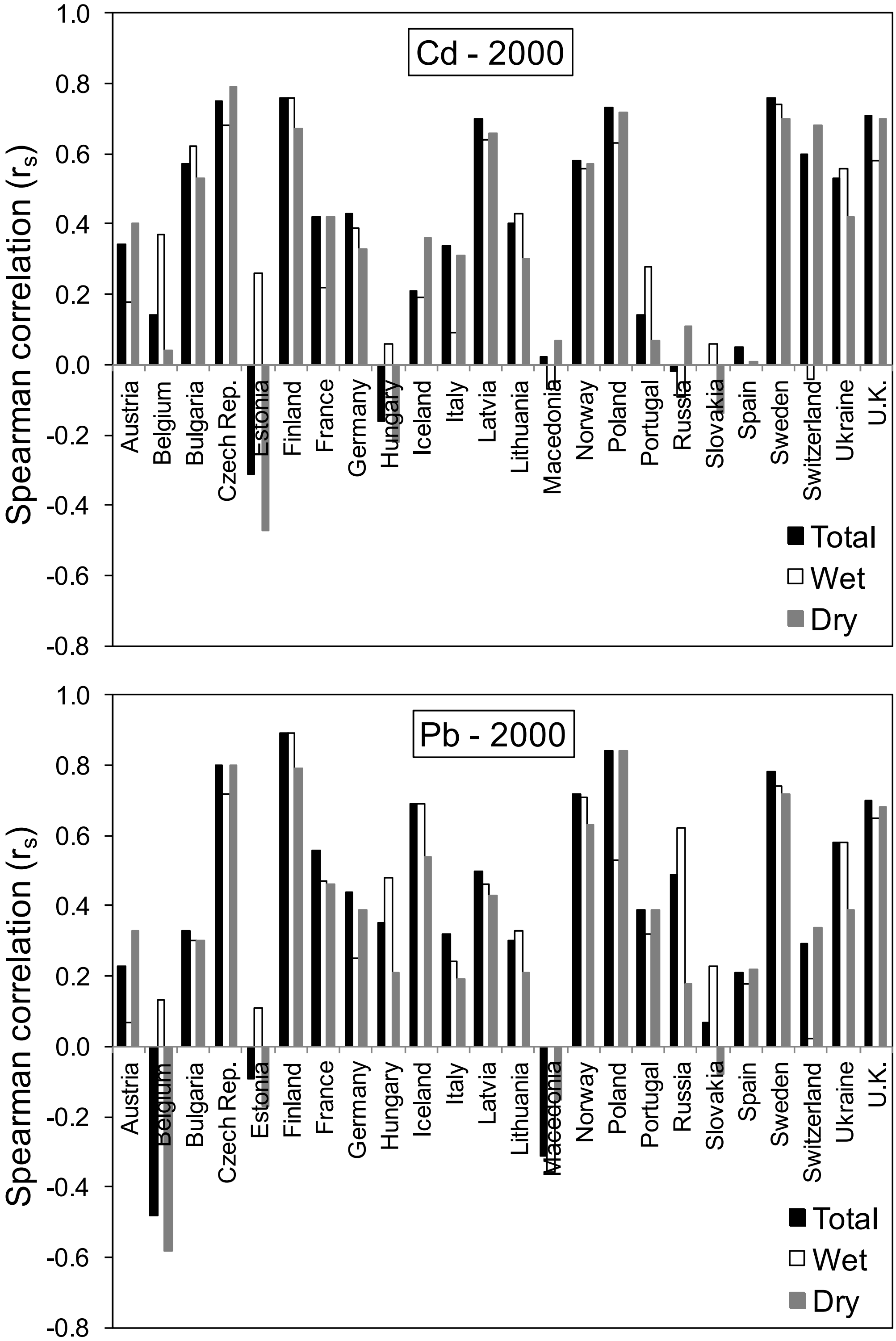
Cd - 2000
Pb - 2000
Spearman correlation (r_s)
1.0
0.8
0.6
0.4
0.2
0.0
-0.2
-0.4
-0.6
-0.8
Austria
Belgium
Bulgaria
Czech Rep.
Estonia
Finland
France
Germany
Hungary
Iceland
Italy
Latvia
Lithuania
Macedonia
Norway
Poland
Portugal
Russia
Slovakia
Spain
Sweden
Switzerland
Ukraine
U.K.
Total
Wet
Dry

Figure 1.

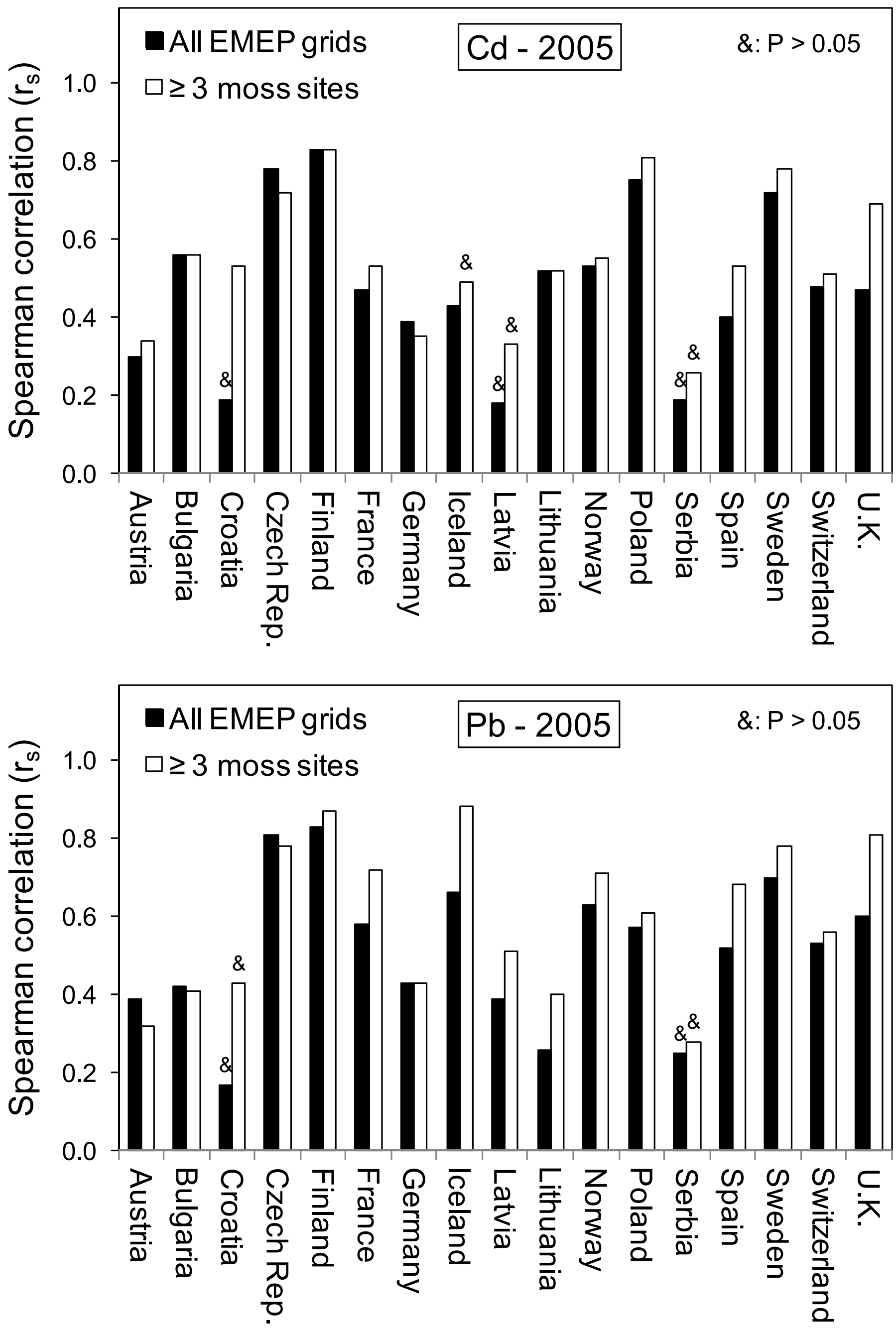
All EMEP grids
≥ 3 moss sites
Cd - 2005
&: P > 0.05
Spearman correlation (r_s)
1.0
0.8
0.6
0.4
0.2
0.0
Austria
Bulgaria
Croatia
Czech Rep.
Finland
France
Germany
Iceland
Latvia
Lithuania
Norway
Poland
Serbia
Spain
Sweden
Switzerland
U.K.
All EMEP grids
≥ 3 moss sites
Pb - 2005
&: P > 0.05
Spearman correlation (r_s)
1.0
0.8
0.6
0.4
0.2
0.0
Austria
Bulgaria
Croatia
Czech Rep.
Finland
France
Germany
Iceland
Latvia
Lithuania
Norway
Poland
Serbia
Spain
Sweden
Switzerland
U.K.

Figure 2.

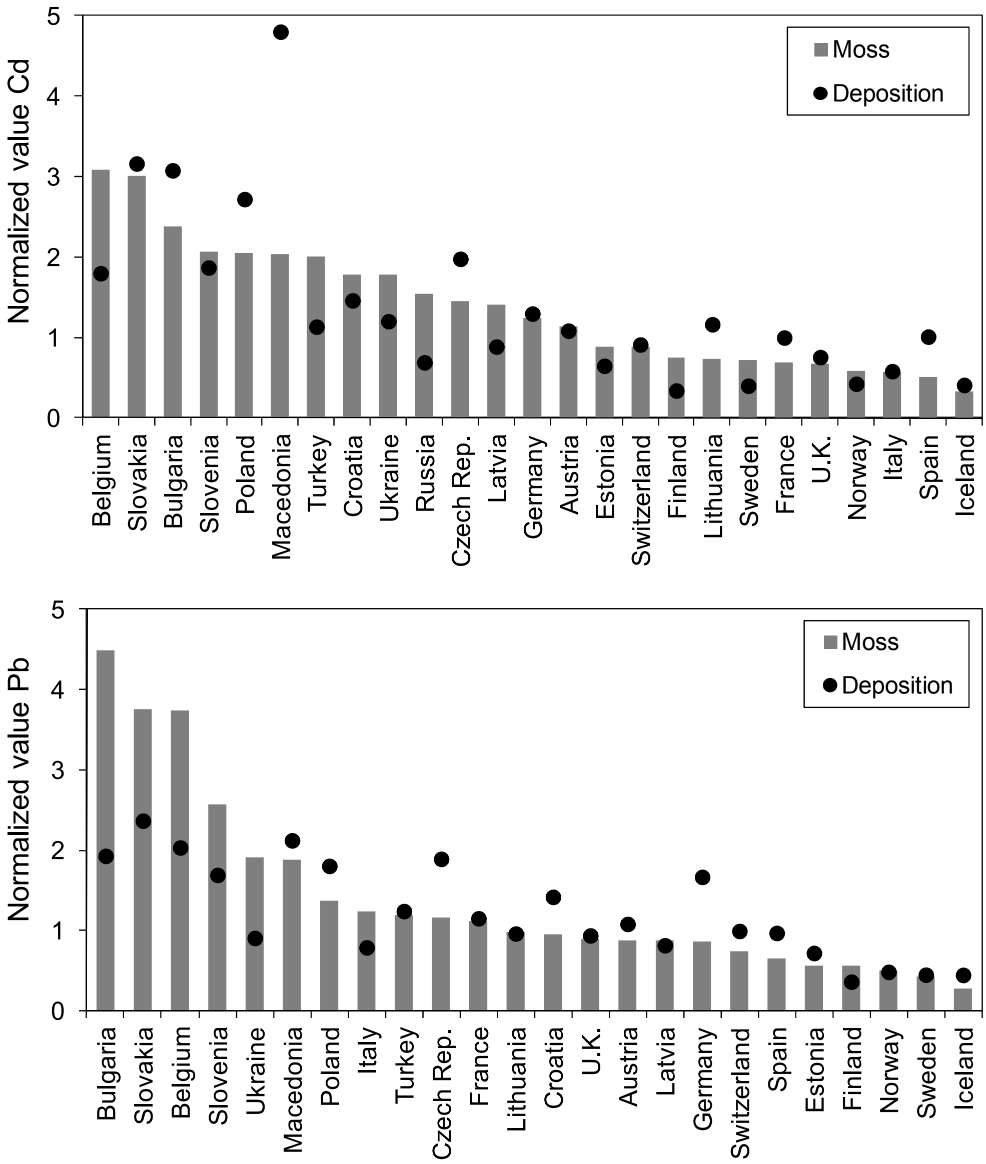
Normalized value Cd
Moss
Deposition
0
1
2
3
4
5
Belgium
Slovakia
Bulgaria
Slovenia
Poland
Macedonia
Turkey
Croatia
Ukraine
Russia
Czech Rep.
Latvia
Germany
Austria
Estonia
Switzerland
Finland
Lithuania
Sweden
France
U.K.
Norway
Italy
Spain
Iceland
Normalized value Pb
Moss
Deposition
0
1
2
3
4
5
Bulgaria
Slovakia
Belgium
Slovenia
Ukraine
Macedonia
Poland
Italy
Turkey
Czech Rep.
France
Lithuania
Croatia
U.K.
Austria
Latvia
Germany
Switzerland
Spain
Estonia
Finland
Norway
Sweden
Iceland

Figure 3.

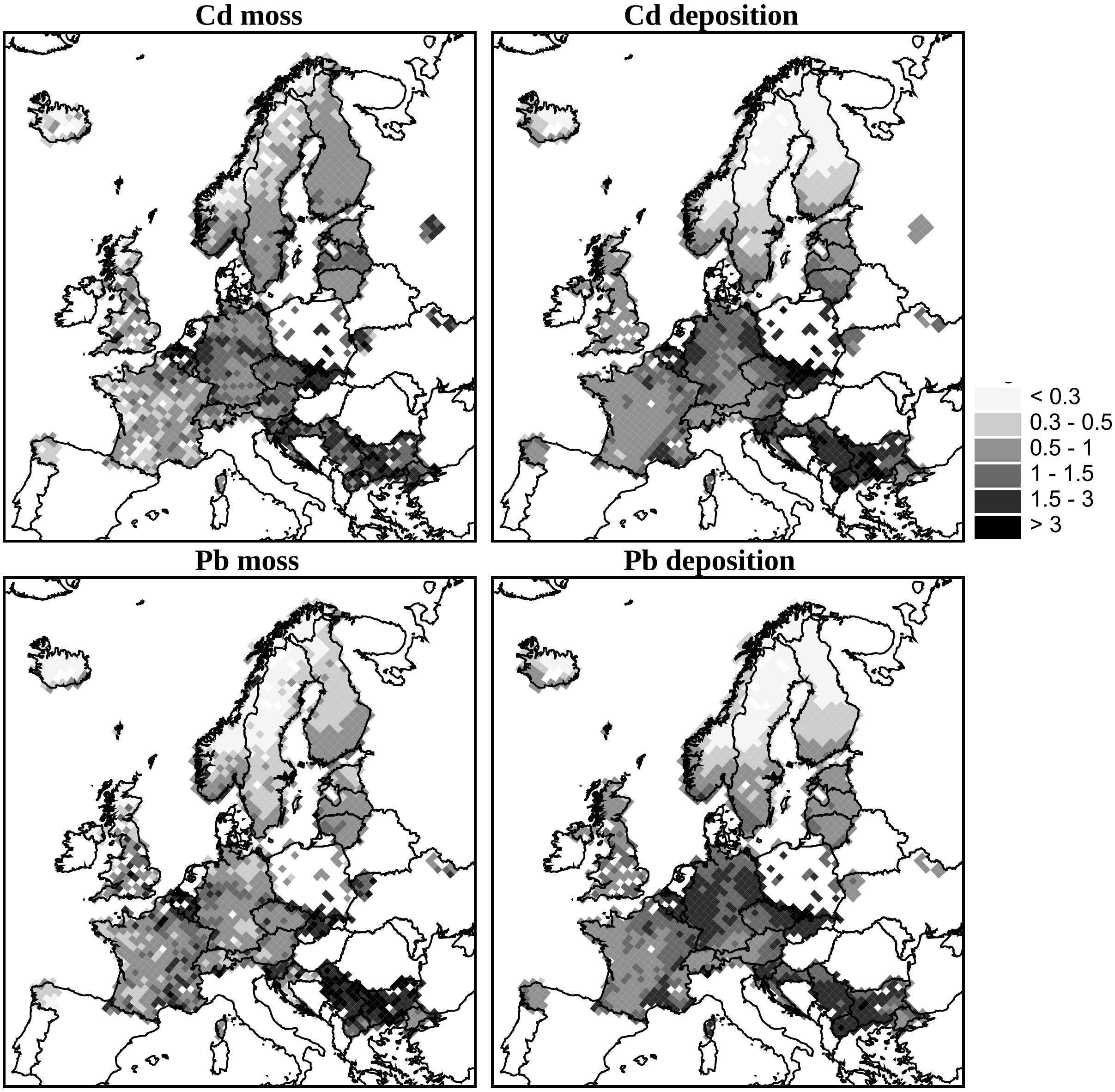
Cd moss
Cd deposition
Pb moss
Pb deposition
< 0.3
0.3 - 0.5
0.5 - 1
1 - 1.5
1.5 - 3
> 3

Figure 4.

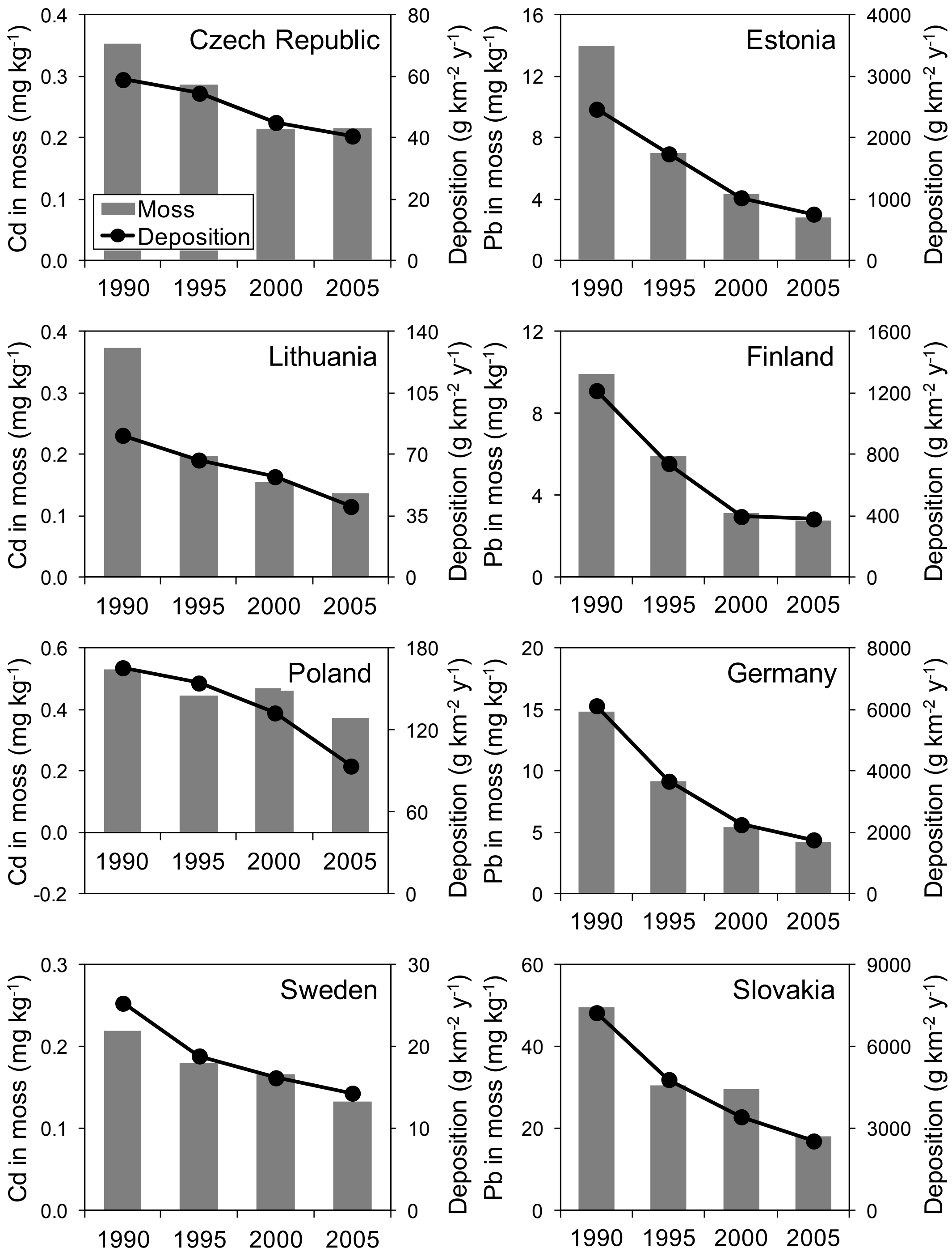
Czech Republic
Estonia
Lithuania
Finland
Poland
Germany
Sweden
Slovakia
Moss
Deposition
Cd in moss (mg kg-1)
Pb in moss (mg kg-1)
Deposition (g km-2 y-1)
1990 1995 2000 2005

Figure 5.