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

H. Harmens[a]*, I. Ilyin[b], G. Mills[a], J.R. Aboal[c], R. Alber[d], O. Blum[e], M. Coşkun[f], L. De Temmerman[g], J.Á. Fernández [c], R. Figueira[h], M. Frontasyeva[i], B. Godzik[j], N. Goltsova[k], Z. Jeran[l], S. Korzekwa[m], E. Kubin[n], K. Kvietkus[o], S. Leblond[p], S. Liiv[q], S.H. Magnússon[r], B. Maňkovská[s], O. Nikodemus[t], R. Pesch[u], J. Poikolainen[n], D. Radnović[v], Å. Rühling[w], J.M. Santamaria[x], W. Schröder[u], Z. Spiric[y], T. Stafilov[z], E. Steinnes[aa], I. Suchara[ab], G. Tabors[t], L. Thöni[ac], G. Turcsányi[ad], L. Yurukova[ae], H.G. Zechmeister[af]

[a] Centre for Ecology and Hydrology, Environment Centre Wales, Deiniol Road, Bangor, Gwynedd LL57 2UW, UK. hh@ceh.ac.uk; gmi@ceh.ac.uk

[b] Meteorological Synthesizing Centre East of EMEP, Krasina pereulok, 16/1, 123056 Moscow, Russian Federation. ilia.ilyin@msceast.org

[c] University of Santiago de Compostela, Faculty of Biology, Department of Ecology 15782 Santiago de Compostela, Spain. jesusramon.aboal@usc.es; jangel.fernandez@usc.es

[d] Environmental Agency of Bolzano, 39055 Laives, Italy. Renate.Alber@provinz.bz.it

[e] National Botanical Garden, Academy of Science of Ukraine, Timiryazevs'ka St. 1, 01014 Kyiv, Ukraine. blum@nbg.kiev.ua

[f] Canakkale Onsekiz Mart University, 17100 Çanakkale, Turkey. coskunafm@yahoo.com

[g] Veterinary and Agrochemical Research Centre, Leuvensesteenweg 17, 3080 Tervuren, Belgium. Ludwig.DeTemmerman@coda-cerva.be

[h] Jardim Botânico da Universidada de Lisboa, Lisbon, Portugal. pcrfigueira@alfa.ist.utl.pt

[i] Joint Institute for Nuclear Research, Str. Joliot-Curie 6, 141980 Dubna, Russian Federation. marina@nf.jinr.ru

[j] Institute of Botany, Polish Academy of Sciences, Lubicz 46, 31512 Krakow, Poland. b.godzik@botany.pl

[k] St Petersburg State University, St Petersburg, Russian Federation. pinexpert@mail.ru

[l] Jožef Stefan Institute, Department of Environmental Sciences, Jamova 39, 1000 Ljubljana, Slovenia. zvonka.jeran@ijs.si

[m] University of Opole, Poland. korzekwas@wp.pl

[n] Finnish Forest Research Institute, Kirkkosaarentie 7, 91500 Muhos, Finland. Eero.Kubin@metla.fi; Jarmo.Poikolainen@metla.fi

[o] Institute of Physics, Savanoriu Ave 231, 02300 Vilnius, Lithuania. kvietkus@ktl.mii.lt

[p] Muséum National d'Histoire Naturelle, 57 rue Cuvier, Case 39, 75005 Paris, France. sleblond@mnhn.fr

[q] Tallinn Botanic Garden, Kloostrimetsa tee 52, 11913 Tallinn, Estonia. siiri.liiv@botaanikaaed.ee

[r] Icelandic Institute of Natural History, Hlemmur 3, 125 Reykjavík, Iceland. sigurdur@ni.is

[s] Institute of Landscape Ecology, Slovak Academy of Science, Štefánikova Str. 3, 814 99 Bratislava, Slovakia. bmankov@stonline.sk

[t] University of Latvia, Riga, Latvia. nikodemu@lanet.lv; guntis@lanet.lv

[u] Chair of Landscape Ecology, University of Vechta, PO Box 1553, D-49356 Vechta, Germany. rpesch@iuw.uni-vechta.de; wschroeder@iuw.uni-vechta.de

[v] Faculty of Science, University of Novi Sad, Trg D. Obradovica 4, 21000 Novi Sad, Serbia. dragan.radnovic@dbe.uns.ac.rs

[w] Humlekärrshultsvägen 10, 572 41 Oskarshamn, Sweden. ake.ruhling@telia.com

[x] University of Navarra, Irunlarrea No 1, 31008 Pamplona, Spain. chusmi@unav.es

[y] Oikon Ltd., Institute for Applied Ecology, Avenija V. Holjevca 20, 10020 Zagreb, Croatia. zspiric@oikon.hr

[z] Saints Cyril and Methodius University, PO Box 162, 1000 Skopje, FYR Macedonia. trajcest@iunona.pmf.ukim.edu.mk

[aa] Department of Chemistry, Norwegian University of Science and Technology, 7491 Trondheim, Norway. Eiliv.Steinnes@chem.ntnu.no

[ab] Silva Tarouca Research Institute for Landscape and Ornamental Gardening, Kvetnove namesti 391, 252 43 Pruhonice, Czech Republic. suchara@vukoz.cz

[ac] FUB-Research Group for Environmental Monitoring, Alte Jonastrasse 83, 8640 Rapperswil, Switzerland. lotti.thoeni@fub-ag.ch

[ad] Szent István University, Gödöllő, Hungary. turcsanyi.gabor@kti.szie.hu

[ae] Institute of Botany, Bulgarian Academy of Sciences, Acad. G.Bonchev Str., Block 23, 1113 Sofia, Bulgaria. yur7lild@bio.bas.bg

[af] University of Vienna, Althanstraße 14, 1090 Vienna, Austria. Harald.Zechmeister@univie.ac.at

* Corresponding Author: Harry Harmens

E-mail address: hh@ceh.ac.uk

Tel.: +44-1248-374512

Fax: +44-1248-362133

**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

97 their emissions of air pollutants. The 1998 Aarhus Protocol on heavy metals targeted three
98 harmful heavy metals (cadmium (Cd), lead (Pb) and mercury (Hg)) and entered into force in
99 2003. Within the Convention, the European Monitoring and Evaluation Programme (EMEP)
100 i) collects emission data from Parties, ii) measures air and precipitation quality, and iii)
101 models atmospheric transport and deposition of air pollutants. Deposition of the heavy metals
102 Cd, Hg and Pb is modelled using the EMEP atmospheric transport model MSCE-HM
103 (Travnikov and Ilyin, 2005) and is calculated from official emission data reported by the
104 countries. The modelled data are verified against concentrations in air and precipitation
105 measured at EMEP monitoring stations. However, the number of EMEP monitoring stations
106 and their spatial distribution across Europe is limited: in the period from 1990 to 2009 there
107 were between 40 to 77 stations annually reporting measurement data on heavy metals to
108 EMEP (http://www.nilu.no/projects/ccc/index.html). The EMEP monitoring network for Cd
109 and Pb is scarce or absent in the southern and eastern parts of Europe, whereas Hg is
110 primarily measured in northern Europe.
111 Under the Working Group on Effects of the Convention, the ICP Vegetation
112 (International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and
113 Crops) has been coordinating the European moss survey since 2000. The survey has been
114 repeated at five-yearly intervals since 1990 and the latest survey was conducted in 2005/6
115 with 28 countries participating and mosses being sampled at almost 6,000 sites across
116 Europe. The European moss survey provides data on concentrations of twelve trace elements
117 (Al, As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, V, Zn) in naturally growing mosses (Harmens et al.,
118 2010). In 2005/06, the N concentration in mosses was also determined (Harmens et al.,
119 2011b) and in the current ongoing survey in 2010/11, a pilot study was initiated in selected
120 countries to determine the concentration of selected persistent organic pollutants (POPs),
121 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

147 of Cd and Pb concentrations in mosses (Harmens et al., 2008b; Holy et al., 2010; Schröder et
148 al., 2010). As these factors and their influence on the relationship is likely to be different for
149 different countries and/or climatic regions (e.g. Thöni et al., 2011), we hypothesise that the
150 correlations between both parameters will be country-specific, with good correlations
151 expected in some but less good correlations expected in other countries.
152
153 **2. Materials and methods**
154 *Determination of Cd and Pb concentrations in mosses*
155 Moss samples were collected across Europe in 1990/1 (Rühling, 1994), 1995/6 (Rühling and
156 Steinnes, 1998), 2000/1 (Buse et al., 2003) and 2005/6 (Harmens et al., 2008a, 2010).
157 Throughout the paper we refer to the years of moss survey as 1990, 1995, 2000 and 2005
158 respectively. Because the mosses were collected in a range of habitats from the sub-arctic
159 climate of northern Scandinavia to the hot and dry climate of southern Europe, it was not
160 possible to sample just one carpet-forming moss species across Europe. *Pleurozium schreberi*
161 was the most frequently sampled species in all surveys, accounting for 40.8 – 52.7% of the
162 samples, followed by *Hylocomium splendens* (20.5 – 39.3%), *Hypnum cupressiforme* (7.4 –
163 22.0%) and *Pseudoscleropodium purum* (3.4 – 11.9%); other species constituted only 2.2 –
164 6.5% of the mosses sampled. The moss sampling procedure was according to the guidelines
165 described in the protocol for the 2005 survey (ICP Vegetation, 2005). Only the last three
166 years' growth of moss material was used for the analyses. The concentrations of Cd and Pb
167 were determined by a range of analytical techniques; for further details we refer to the reports
168 of the individual surveys (Buse et al., 2003; Harmens et al., 2008a, 2010; Rühling, 1994;
169 Rühling and Steinnes, 1998). A comprehensive quality control exercise was conducted in
170 1995 (Steinnes et al., 1997) and 2005 (Harmens et al., 2010) with moss reference material
171 being distributed amongst participating laboratories. In addition, some laboratories used other

172 certified reference material for quality assurance. Recommended values were established in
173 1995 for moss reference material. For example, the recommended values for Cd and Pb for
174 moss reference M2 were 0.454 ± 0.019 and 6.37 ± 0.43 mg $kg^{-1}$ (mean ± standard deviation)
175 respectively and 0.106 ± 0.005 and 3.33 ± 0.25 mg $kg^{-1}$ respectively for moss reference M3
176 (Steinnes et al., 1997). No amendment of these recommended values was required in 2005
177 (Harmens et al., 2010). For further details we refer to Steinnes et al. (1997) and Harmens et
178 al. (2010).
179
180 *Modelling the deposition of Cd and Pb*
181 Deposition of the heavy metals Cd and Pb was modelled using the EMEP atmospheric
182 transport model MSCE-HM (Travnikov and Ilyin, 2005). MSCE-HM is a three-dimensional
183 Eulerian-type chemical transport model driven by off-line meteorological data. The model
184 takes into account heavy metal emissions from anthropogenic and natural sources, wind re-
185 suspension of dust particles containing heavy metals, transport in the atmosphere, chemical
186 transformations of mercury and ecosystem-dependent deposition to the surface. The model
187 computation domain is defined on the polar stereographic projection. Its spatial resolution is
188 50 km × 50 km at 60°N. Modelled deposition of heavy metals was calculated from official
189 emission data reported by the countries. The modelled data were verified against
190 concentrations in air and precipitation measured at EMEP monitoring stations. The intrinsic
191 uncertainty of the model (without uncertainty in reported emission data) is about 30 – 40%
192 for concentrations in air, concentrations in precipitation and total deposition for Cd and Pb
193 (Travnikov and Ilyin, 2005). The uncertainty of country-specific totals of heavy metal
194 emission typically ranged between 30 – 60% and the overall uncertainty of measured wet
195 deposition was around 20% for Cd and Pb. Modelling results agreed with measurement data
196 with satisfactory accuracy, keeping in mind uncertainties of the emission and monitoring

197 data. At most of the monitoring stations modelled and observed levels of Cd and Pb agreed
198 within ±50% and the spatial correlation coefficient between modelled and observed values is
199 between 0.6 – 0.9 (Ilyin et al., 2010).
200
201 *Correlations between EMEP modelled deposition and concentrations in mosses*
202 Country-specific Spearman rank correlations between various forms of EMEP modelled
203 atmospheric deposition (dry, wet and total deposition) and concentrations in mosses for Cd
204 and Pb were determined using SigmaPlot version 11. In this investigation, we computed the
205 Spearman rank correlation coefficient $r_s$ because the metal concentrations mostly proved not
206 to be normally distributed. Although this non-parametric correlation method is less powerful
207 than parametric methods if the assumptions underlying the latter are met, it is less likely to
208 give distorted results when the assumptions fail. The coefficient $r_s$ equals −1, if the two
209 rankings are completely opposite to each other, $r_s$ equals 0 if the rankings are completely
210 independent and +1 if there is complete agreement between the two rankings. Within the
211 interval [−1, +1] the strength of correlation can be classified as follows: $r_s$ values <|0.2| are
212 very low, between |0.2| and |0.5| low, from |0.5| to |0.7| moderate, between |0.7| and |0.9| high
213 and > |0.9| very high (Schröder et al., 2010).
214 As the last three years of moss growth was selected for heavy metal determination,
215 representing the accumulation of Cd and Pb in mosses in the three years previous to sampling
216 (ICP Vegetation, 2005), EMEP data were accumulated and averaged over the previous three
217 years where possible. For 1990, the EMEP modelled data for 1990 were used as data for
218 earlier years was not available. To assess the impact of using EMEP modelled data averaged
219 over three years in comparison to modelled data for the year previous to moss sampling,
220 correlations were also determined using only the EMEP modelled data for the year previous
221 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

247 metal-specific correlations were observed (Table 1 and 2, Figure 1 and 2) and correlations
248 varied between years (Table 1). High correlations ($0.7 \leq r_s < 0.9$) were generally observed for
249 the Czech Republic (except for 1990), Finland, Sweden, and for Pb also in Norway for the
250 earlier years (Table 1). Moderate correlations ($0.5 \leq r_s < 0.7$) were generally found in France
251 (for Pb in particular), Norway (for Cd) and Poland (with sometimes high correlations being
252 observed). Other countries with moderate to high correlations for at least one of the metals
253 for at least two survey years include Bulgaria, Iceland, Latvia, Switzerland, Ukraine and the
254 United Kingdom. The generally lower correlations in Norway compared to Finland and
255 Sweden might be related to the more complex topography of Norway with orographic
256 deposition having a greater role. In addition, the lower correlations for Cd might be due to the
257 competition with sea salt ions in the extensive coastal area of Norway (Steinnes, 1995).
258 Significant positive correlations were found for about two thirds or more of the participating
259 countries (except for Cd in 1990). As to be expected, non-significant or significant negative
260 correlations were mainly found in smaller countries or in countries where mosses were
261 sampled in a smaller number of grid squares (< 60), although this was not always the case
262 (e.g. Iceland for Pb, Latvia, Ukraine, Switzerland). Negative correlations were significant
263 only twice, i.e. in 1990 for Cd in Lithuania and Portugal.
264 As the heavy metal concentrations in mosses were determined over the last three
265 years of growth before the date of sampling, it was assumed that the concentration in mosses
266 represents the accumulation of Cd and Pb atmospheric deposition over the same period.
267 Therefore, the metal concentration in mosses was compared with the average EMEP
268 modelled annual deposition for the three years previous to moss sampling. However, this was
269 not feasible for 1990 as only modelled annual deposition data was available for 1990 and not
270 for the previous years. To investigate whether this would have any effect on the determined
271 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

296 environmental factors (pH, salinity, temperature), which may vary within short periods of
297 time (Bouquette et al., 2011).
298 The impact of precipitation on the concentration of elements in mosses is unclear and
299 there is no evidence that the intensity or frequency of precipitation affects heavy metal
300 concentrations in mosses. On the one hand, rainwater may wash the moss, resulting in
301 removal of the particulate material deposited on its surface, on the other hand, rainwater may
302 dissolve elements adsorbed on moss tissue, thus facilitating their uptake. The most important
303 way that precipitation may influence metals already present on the moss surface might be via
304 exchange with other cations, including those from marine origin (Gjengedal and Steinnes,
305 1990; Steinnes et al., 1995). In the current study, Spearman rank correlations between Cd and
306 Pb concentrations in mosses and EMEP modelled deposition (total, wet or dry) were not
307 significantly affected by the fraction of EMEP modelled wet deposition (data not shown).
308 The distribution of the annual deposition between the wet and dry part is controlled by a
309 combination of two main factors: 1) the annual sum of precipitation and 2) the distribution of
310 forests – dry deposition velocity of particles (and hence particulate species like Cd and Pb) to
311 areas with tall vegetation is greater than that to areas with short vegetation. The combination
312 of these two factors results in a relatively high fraction of dry deposition in central Europe
313 (e.g. Czech Republic, Germany, Poland with moderate precipitation and high forested area)
314 and in regions of southern Europe (Portugal, southern Spain, Italy). In the recent surveys
315 mosses were sampled in northern Spain and Italy in mountainous regions with higher annual
316 precipitation than other parts of these countries.
317 We also investigated whether Spearman rank correlations were affected by the form
318 of atmospheric deposition, i.e. wet, dry and total deposition of Cd and Pb. The results show
319 that indeed the correlations are affected by the form of atmospheric deposition, as shown in
320 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

346 excluded from the analysis. Indeed, in the majority of countries (>70%) there was an
347 improvement in the correlations, which appeared to be most pronounced for Pb (Figure 2).
348 The improvement in correlations is observed despite the fact that the number of EMEP grids
349 with the required data is lower than when all EMEP grids with moss data are included.
350 As for the nitrogen study (Harmens et al., 2011) we also compared the correlations
351 including all EMEP grids with the correlations including EMEP grids with at least five moss
352 sampling sites (data not shown). However, in contrast to the European-wide relationship
353 established for nitrogen, in the current country-specific analysis with heavy metals the
354 number of EMEP grids with at least five sampling sites was low in many countries, resulting
355 in a decline in the number of significant correlations (i.e. correlations were significant in only
356 eight of the 15 countries for which correlations could be determined) compared to including
357 all EMEP grids or EMEP grids with at least three moss sampling sites (Figure 2). Only four
358 (i.e. Czech Republic, Finland, Germany and Sweden) out of the 17 countries included in
359 Figure 2 had more than 25 EMEP grids with at least five moss sampling sites, whereas in
360 seven out of the 17 countries the number of EMEP grids with at least five moss sampling
361 sites was less than 15.
362
363 *Spatial patterns and temporal trends in moss concentrations and modelled deposition*
364 To compare the spatial patterns of Cd and Pb concentrations in mosses and EMEP modelled
365 total deposition, both datasets were normalized against the European mean (see Materials and
366 Methods for details). Figure 3 and 4 show that the spatial patterns for 2005 are quite similar,
367 i.e. regions in Europe with a deposition rate below (e.g. big parts of northern and western
368 Europe) or above the European mean (e.g. Belgium, eastern part of Europe and parts of
369 central Europe) also showed concentrations in mosses below or above the European mean
370 respectively, particularly for Cd. Nevertheless some discrepancies can be observed: For Cd,

371 modelled deposition is relatively high (i.e. the ratio of normalized deposition to moss
372 concentration >1.5) in Macedonia, Spain and Lithuania compared to the concentration in
373 mosses. The opposite is true for Belgium, Finland and the Russian Federation (i.e. the ratio of
374 normalized moss concentration to deposition >1.5). For Pb, modelled deposition is relatively
375 high in the Czech Republic, Germany and Iceland, whereas the opposite is true for Belgium,
376 Bulgaria, Italy, Slovakia and Ukraine. The relatively high Pb concentrations in mosses in
377 these countries result in relatively low normalized values in central Europe in comparison
378 with the normalized deposition values (Figure 4).

379 Previously we reported on the similarity between temporal trends observed for Cd and
380 Pb concentrations in mosses and EMEP modelled total deposition between 1990 and 2005 at
381 the European scale (Harmens et al., 2011). In the current study, we compared the temporal
382 trends in further detail for individual countries. Some examples are shown in Figure 5 and in
383 general the temporal trends in concentrations in mosses agree reasonably well with the
384 temporal trends in deposition at the national scale too. Nevertheless, in certain periods the
385 decline of calculated deposition seemed to be underestimated in comparison to the decline of
386 concentrations in mosses (e.g. for Cd in Lithuania and the Czech Republic between 1990 and
387 1995, for Pb in Estonia between 1990 and 1995) or vice versa (e.g. for Cd in Poland between
388 1995 and 2000 or for Pb in Slovakia between 1995 and 2000). The sometimes higher
389 concentrations of Cd (Czech Republic and Lithuania) and Pb (Estonia) in mosses compared
390 to modelled deposition in 1990 might reflect the presence of more local pollution sources that
391 affect concentrations in mosses but are not included in modelled deposition. In many
392 countries smaller local pollution sources closed down in the early 1990s. On the other hand, it
393 might reflect more inadequate emission inventories in 1990 compared to later years.

394 Considering the uncertainties in the EMEP modelled deposition data (see
395 Introduction) and the potential limitations and confounding factors in the use of mosses as

396 monitors of atmospheric deposition (Aboal et al., 2010; Boquete et al., 2011; Harmens et al.,
397 2008b; Steinnes, 1995), the spatial patterns and temporal trends of both data sets agree
398 reasonably well for Cd and Pb. The results confirm once again that Cd and Pb concentrations
399 in mosses can serve as a complementary method to determine spatial patterns and temporal
400 trends of Cd and Pb deposition (Aboal et al., 2010; Harmens et al., 2010). Currently EMEP is
401 conducting a case study to assess heavy metal pollution at country-scale levels, employing a
402 spatial resolution finer (e.g. 5 km x 5 km) than that currently used (50 km x 50 km). The
403 European moss survey will provide valuable field-based measurement data for the validation
404 of the finer-resolution modelled atmospheric deposition (Ilyin et al., 2011).
405
406 **4. Conclusions**
407 The following main conclusions can be drawn:
408 • For Cd and Pb the correlations between concentrations in mosses and the EMEP
409 modelled total atmospheric deposition are country- and metal-specific, with sometimes
410 considerable variation being observed between years. However, significant positive
411 correlations were found for about two thirds or more of the participating countries (except
412 for Cd in 1990). Non-significant or significant negative correlations (only two) were
413 mainly found in smaller countries or in countries where mosses were sampled in a relative
414 small number of EMEP grid squares;
415 • Correlations were generally not affected by using EMEP modelled deposition data for the
416 year previous to sampling or averaged over three years previous to sampling of the
417 mosses. As expected, correlations mainly improved when the analysis was limited to
418 using EMEP grids in which at least three moss sampling sites were present;
419 • For the majority of countries across Europe, the use of mosses as biomonitors of
420 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.

## Acknowledgements

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## References

445 Aboal, J.R., Fernández, J.A., Boquete, T., Carballeira, A., 2010. Is it possible to estimate
446 atmospheric deposition of heavy metals by analysis of terrestrial mosses? Science of
447 the Total Environment 408, 6291-6297.
448 Berg, T., Hjellbrekke, A., Rühling, Å., Steinnes, E., Kubin, E., Larsen, M.M., Piispanen, J.,
449 2003. Absolute deposition maps of heavy metals for the Nordic countries based on the
450 moss survey. TemaNord 2003:505, Nordic Council of Ministers, Copenhagen,
451 Denmark.
452 Berg, T., Steinnes, E., 1997. Use of mosses (*Hylocomium splendens* and *Pleurozium*
453 *schreberi*) as biomonitors of heavy metal deposition: from relative to absolute values.
454 Environmental Pollution 98, 61-71.
455 Boquete, M.T., Fernandez, J.A., Aboal, J.R., Carballeira, A., 2011. Analysis of temporal
456 variability in the concentrations of some elements in the terrestrial moss
457 *Pseudoscleropodium purum*. Environmental and Experimental Botany 72, 210–216.
458 Buse, A., Norris, D., Harmens, H., Büker, P., Ashenden, T., Mills, G., 2003. Heavy metals in
459 European mosses: 2000/2001 survey. ICP Vegetation Programme Coordination
460 Centre, Centre for Ecology and Hydrology, Bangor, UK.
461 http://icpvegetation.ceh.ac.uk
462 Couto, J.A., Fernandez, J.A., Aboal, J.R., Carballeira, C., 2004. Active biomonitoring of
463 element uptake with terrestrial mosses: a comparison of bulk and dry deposition.
464 Science of the Total Environment 324, 211–222.
465 Gjengedal, E., Steinnes, E., 1990. Uptake of metal ions in moss from artificial precipitation.
466 Environmental Monitoring and Assessment 14, 77-87.
467 Harmens, H., Mills, G., Hayes, F., Norris, D. and the participants of the ICP Vegetation,
468 2011a. Air pollution and vegetation. ICP Vegetation annual report 2010/2011.
469 http://icpvegetation.ceh.ac.uk

Harmens, H., Norris, D. and the participants of the moss survey, 2008a. Spatial and temporal trends in heavy metal accumulation in mosses in Europe (1990-2005). Programme Coordination Centre for the ICP Vegetation, Centre for Ecology and Hydrology, Bangor, UK. http://icpvegetation.ceh.ac.uk

Harmens, H., Norris, D. A., Cooper, D.M., Mills, G., Steinnes E., Kubin, E., Thöni, L., Aboal, J.R., Alber, R., Carballeira, A., Coşkun, M., De Temmerman, L., Frolova, M., Gonzáles-Miqueo, L., Jeran, Z., Leblond S., Liiv, S., Maňkovská, B., Pesch, R., Poikolainen, J., Rühling, Å., Santamaria, J. M., Simonėiė, P., Schröder, W., Suchara, I., Yurukova, L., Zechmeister, H. G., 2011b. Nitrogen concentrations in mosses indicate the spatial distribution of atmospheric nitrogen deposition in Europe. Environmental Pollution 159, 2852-2860.

Harmens, H., Norris, D.A., Koerber, G.R., Buse, A., Steinnes, E., Rühling, Å., 2007. Temporal trends in the concentration of arsenic, chromium, copper, iron, nickel, vanadium and zinc in mosses across Europe between 1990 and 2000. Atmospheric Environment 41, 6673-6687.

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

Harmens, H., Norris, D.A., Steinnes, E., Kubin, E., Piispanen, J., Alber, R., Aleksiayenak, Y., Blum, O., Coşkun, M., Dam, M., De Temmerman, L., Fernández, J.A., Frolova, M., Frontasyeva, M., González-Miqueo, L., Grodzińska, K., Jeran, Z., Korzekwa, S., Krmar, M., Kvietkus, K., Leblond, S., Liiv, S., Magnússon, S.H., Maňkovská, B., Pesch, R., Rühling, Å., Santamaria, J.M., Schröder, W., Spiric, Z., Suchara, I., Thöni, L., Urumov, V., Yurukova, L., Zechmeister, H.G., 2010. Mosses as biomonitors of

494 atmospheric heavy metal deposition: spatial and temporal trends in Europe.
495 Environmental Pollution 158, 3144-3156.
496 Holy, M., Pesch, R., Schröder, W., Harmens, H., Ilyin, I., Alber, R., Aleksiayenak, Y., Blum,
497 O., Coşkun, M., Dam, M., De Temmerman, L., Fedorets, N., Figueira, R., Frolova,
498 M., Frontasyeva, M., Goltsova, N., González Miqueo, L., Grodzińska, K., Jeran, Z.,
499 Korzekwa, S., Krmar, M., Kubin, E., Kvietkus, K., Larsen, M., Leblond, S., Liiv, S.,
500 Magnússon,S., Maňkovská, B., Mocanu, R., Piispanen, J., Rühling, Å., Santamaria, J.,
501 Steinnes, E., Suchara, I., Thöni, L., Turcsányi, G., Urumov, V., Wolterbeek, H.T.,
502 Yurukova, L., Zechmeister, H.G., 2010. First thorough identification of factors
503 associated with Cd, Hg and Pb concentrations in mosses sampled in the European
504 Surveys 1990, 1995, 2000 and 2005. Journal of Atmospheric Chemistry 63, 109-124.
505 ICP Vegetation, 2005. Heavy metals in European mosses: 2005/2006 survey. Monitoring
506 manual. ICP Vegetation Programme Coordination Centre, CEH Bangor, UK.
507 http://icpvegetation.ceh.ac.uk
508 Ilyin I., Rozovskaya, O., Sokovyh, V., Travnikov, O., Varygina, M., Aas, W. Uggerud, H.T.,
509 2010. EMEP Status Report 2/2010. Heavy Metals: Transboundary Pollution of the
510 Environment. http://www.msceast.org
511 Ilyin I., Rozovskaya, O., Travnikov, O., Varygina, M., Aas, W. Uggerud, H.T., 2011. EMEP
512 Status Report 2/2011. Heavy Metals: Transboundary Pollution of the Environment.
513 http://www.msceast.org
514 Rühling, Å., 1994. Atmospheric heavy metal deposition in Europe – estimation based on
515 moss analysis. NORD 1994:9. Nordic Council of Ministers, Copenhagen, Denmark.
516 Rühling, Å., Steinnes, E., 1998. Atmospheric heavy metal deposition in Europe 1995-1996.
517 NORD 1998:15, Nordic Council of Ministers, Copenhagen, Denmark.

518 Schröder, W., Holy, M., Pesch, R., Harmens, H., Ilyin, I., Steinnes, E., Alber, R.,
519 Aleksiayenak, Y., Blum, O., Coşkun, M., Dam, M., De Temmerman, L., Frolova, M.,
520 Frontasyeva, M., González Miqueo, L., Grodzińska, K., Jeran, Z., Korzekwa, S.,
521 Krmar, M., Kubin, E., Kvietkus, K., Leblond, S., Liiv, S., Magnússon,S., Maňkovská,
522 B., Piispanen, J., Rühling, Å., Santamaria, J., Spiric, Z., Suchara, I., Thöni, L.,
523 Urumov, V., Yurukova, L., Zechmeister, H.G., 2010. Are cadmium, lead and mercury
524 concentrations in mosses across Europe primarily determined by atmospheric
525 deposition of these metals? Journal of Soil and Sediments 10, 1572-1584.
526 Schröder, W., Holy, M., Pesch, R., Zechmeister, H.G., Harmens, H., Ilyin, I. 2011. Mapping
527 atmospheric depositions of cadmium and lead in Germany based on EMEP deposition
528 data and the European Moss Survey 2005. Environmental Sciences Europe 23:19, 1-
529 14.
530 Schröder, W., Pesch, R., 2010. Long-term monitoring of the metal accumulation in forests
531 measured by use of the moss technique. European Journal of Forest Research 129,
532 475-488.
533 Steinnes, E., 1995. A critical evaluation of the use of naturally growing moss to monitor the
534 deposition of atmospheric metals. The Science of the Total Environment 160/161,
535 243-249.
536 Steinnes, E., Rühling, Å., Lippo, H., Mäkinen, A., 1997. Reference material for large-scale
537 metal deposition surveys. Accreditation and Quality Assurance 2, 243-249.
538 Thöni, L., Schnyder, N., Krieg, F., 1996: Comparison of metal concentrations in three species
539 of mosses and metal freights in bulk precipitations. Fresenius' Journal of Analytical
540 Chemistry 354, 703-708.

Thöni, L., Yurukova, L., Bergamini, A. Ilyin, I., Matthaei, D., 2011. Temporal trends and spatial patterns of heavy metal concentrations in mosses in Bulgaria and Switzerland: 1990-2005. Atmospheric Environment 45, 1899-1912.

Travnikov, O., Ilyin, I., 2005. Regional model MSCE-HM of heavy metal transboundary air pollution in Europe. EMEP/MSC-E Technical report 6/2005. Meteorological Synthesizing Centre - East, Moscow, Russian Federation. http://www.msceast.org

Zechmeister, H.G., Grodzińska, K., Szarek-Łukaszewska, G., 2003. Bryophytes. In: Markert, B.A., Breure, A.M., Zechmeister, H.G. (Eds.). Bioindicators and biomonitors. Elsevier Science Ltd., Amsterdam, pp. 329-375.

551 **Figure legends**

552

553 **Figure 1.** Spearman rank correlation coefficients ($r_s$) between the EMEP modelled deposition
554 (total, wet and dry) and concentrations in mosses for Cd and Pb for the moss survey year
555 2000. The modelled deposition data was based on the annual average of the three year sum
556 for 1997 − 1999.

557

558 **Figure 2.** Spearman rank correlation coefficients ($r_s$) between the EMEP modelled total
559 deposition and concentrations in mosses for Cd and Pb for the moss survey year 2005.
560 Correlations are shown for all EMEP grids (black bar) where mosses were sampled or only
561 for EMEP grids with at least three moss sampling sites (white bar) in at least 15 grids. The
562 modelled deposition data was based on the annual average of the three year sum for 2003 −
563 2005.

564

565 **Figure 3**. Normalized values (relative to the overall European mean) of the average Cd and
566 Pb concentration in mosses (2005/6) and EMEP modelled average total annual deposition
567 (2003 – 2005) per country. For the calculation of the normalized values the areas of the
568 countries where mosses were sampled were taken into account.

569

570 **Figure 4**. Maps of the normalized values per country (relative to the overall European mean)
571 of the (top left) average Cd concentration in mosses (2005/6), (top right) EMEP modelled
572 average annual total Cd deposition (2003 – 2005), (bottom left) average Pb concentration in
573 mosses (2005/06) and (bottom right) EMEP modelled average annual total Pb deposition
574 (2003 – 2005). For the calculation of the normalized values the areas of the countries where
575 mosses were sampled were taken into account.

576

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

580

581 **Table 1**. Spearman rank correlation coefficients ($r_s$) between the EMEP modelled total
582 deposition data (annual average of three year sum) and concentrations in mosses for Cd and
583 Pb for those countries that participated in at least three moss surveys during 1990 – 2005. N =
584 number of EMEP grid cells (50 km x 50 km) for which moss data was available; n.d. = not
585 determined; values in bold: $P \leq 0.05$.
586

| | 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 |

587
588

589 **Table 2**. Spearman rank correlation coefficients ($r_s$) between the EMEP modelled total
590 deposition and concentrations in mosses for Cd and Pb for the moss survey year 2000. The
591 modelled total deposition data was based on either the annual average of the three year sum
592 (3 year) or the annual deposition in the year before moss sampling (1 year). N = number of
593 EMEP grid cells (50 km x 50 km) for which moss data was available; values in bold: P ≤
594 0.05.
595

| | 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 |

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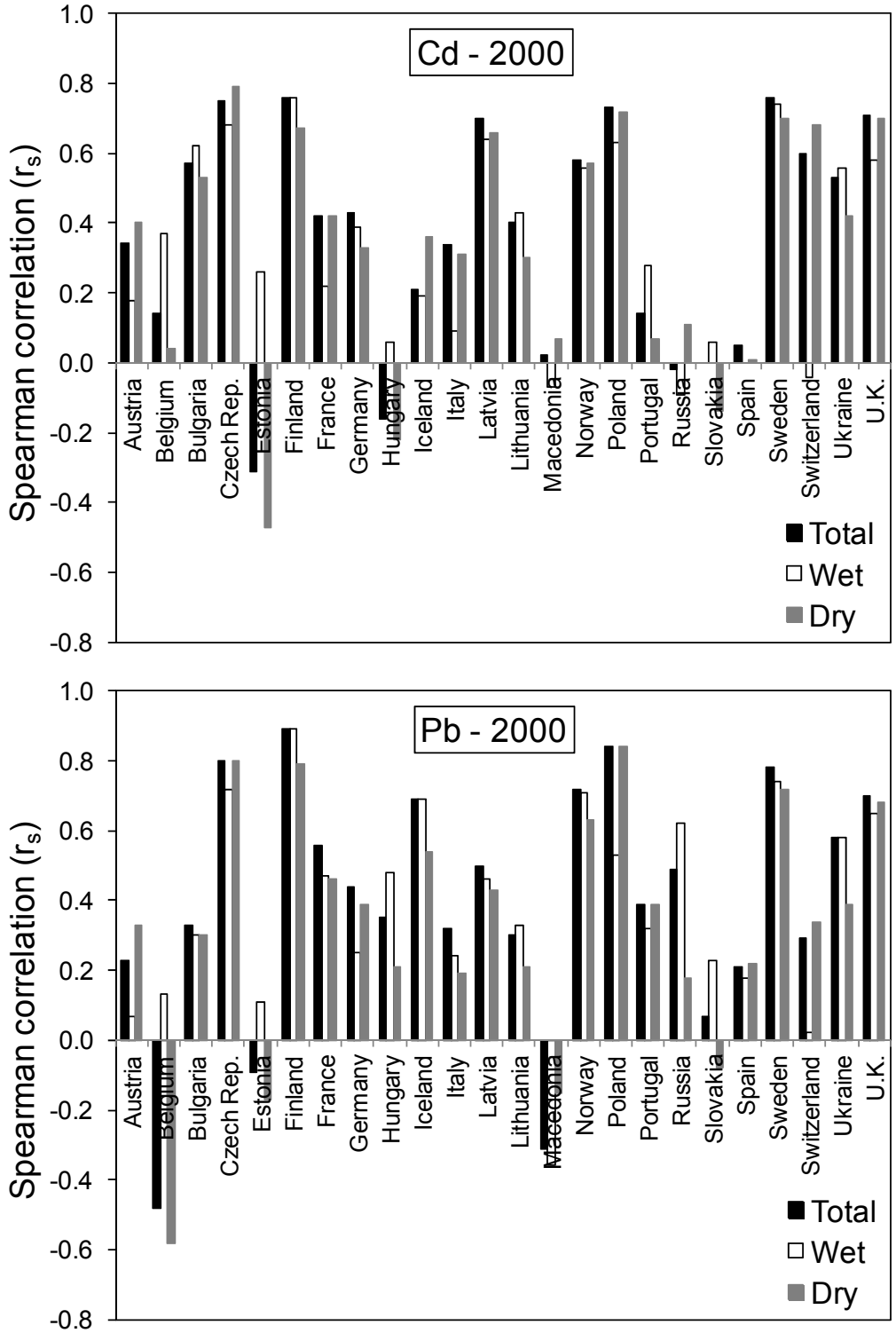
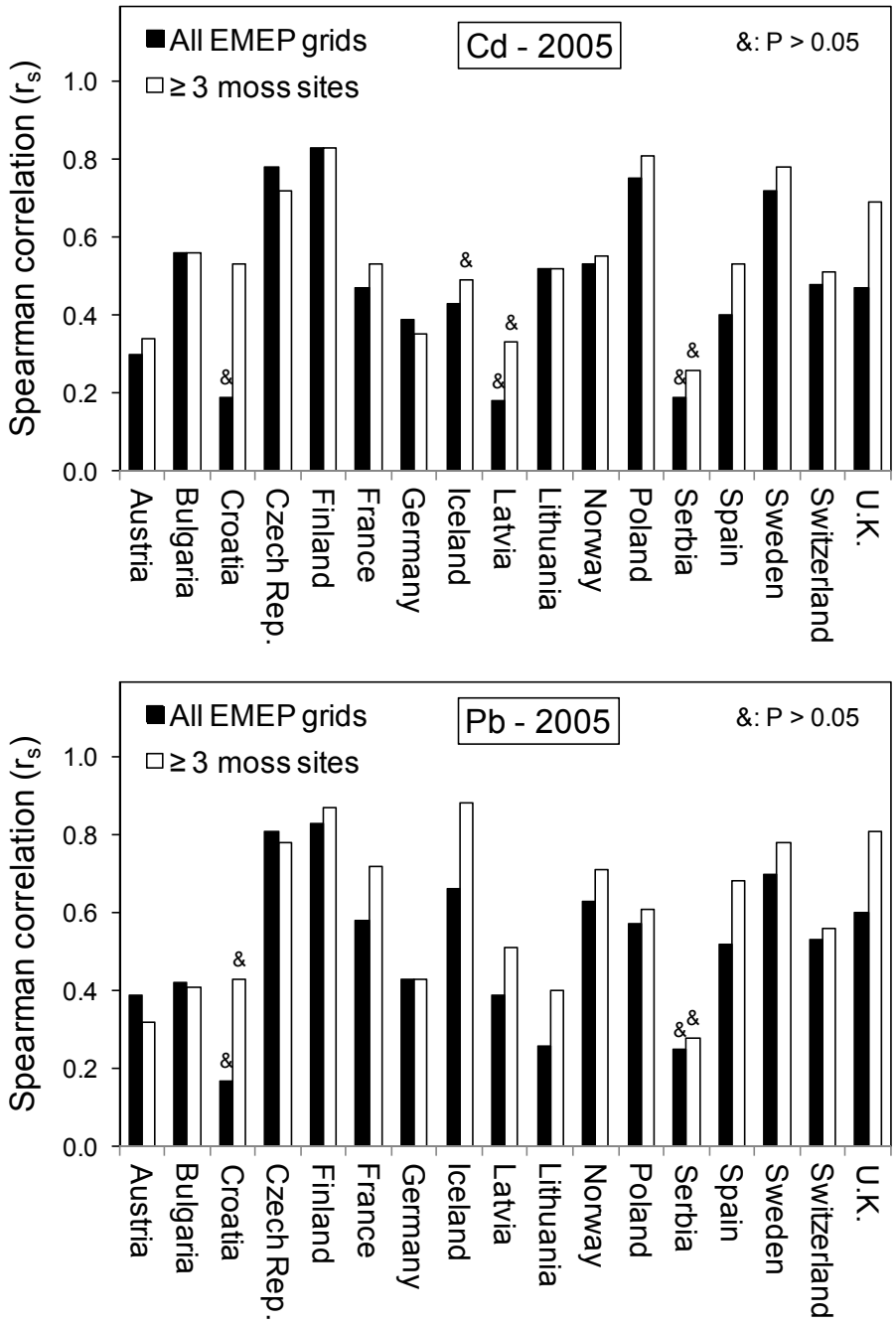
597




598

599 **Figure 1.**

600




601

602 **Figure 2.**

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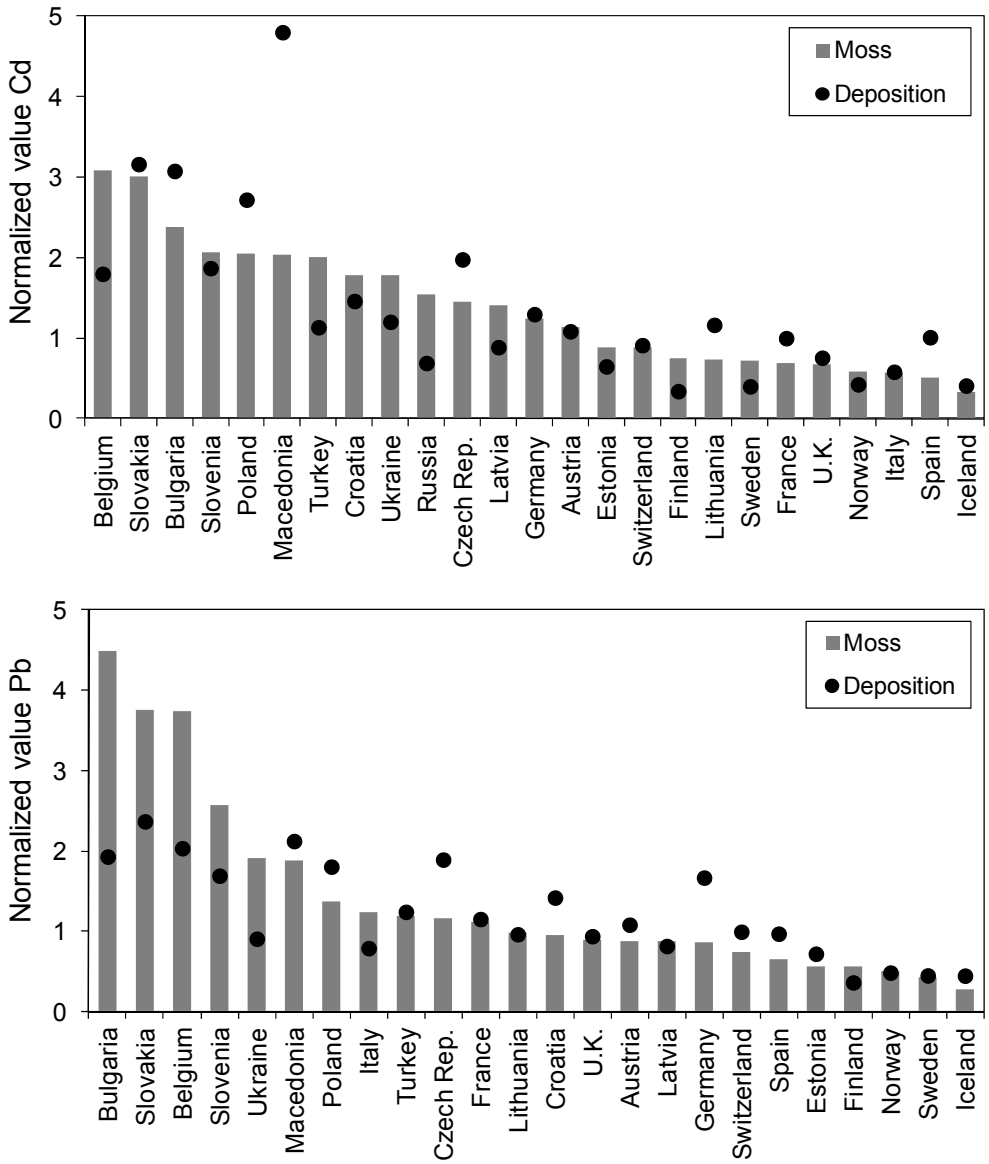


604

605 **Figure 3.**

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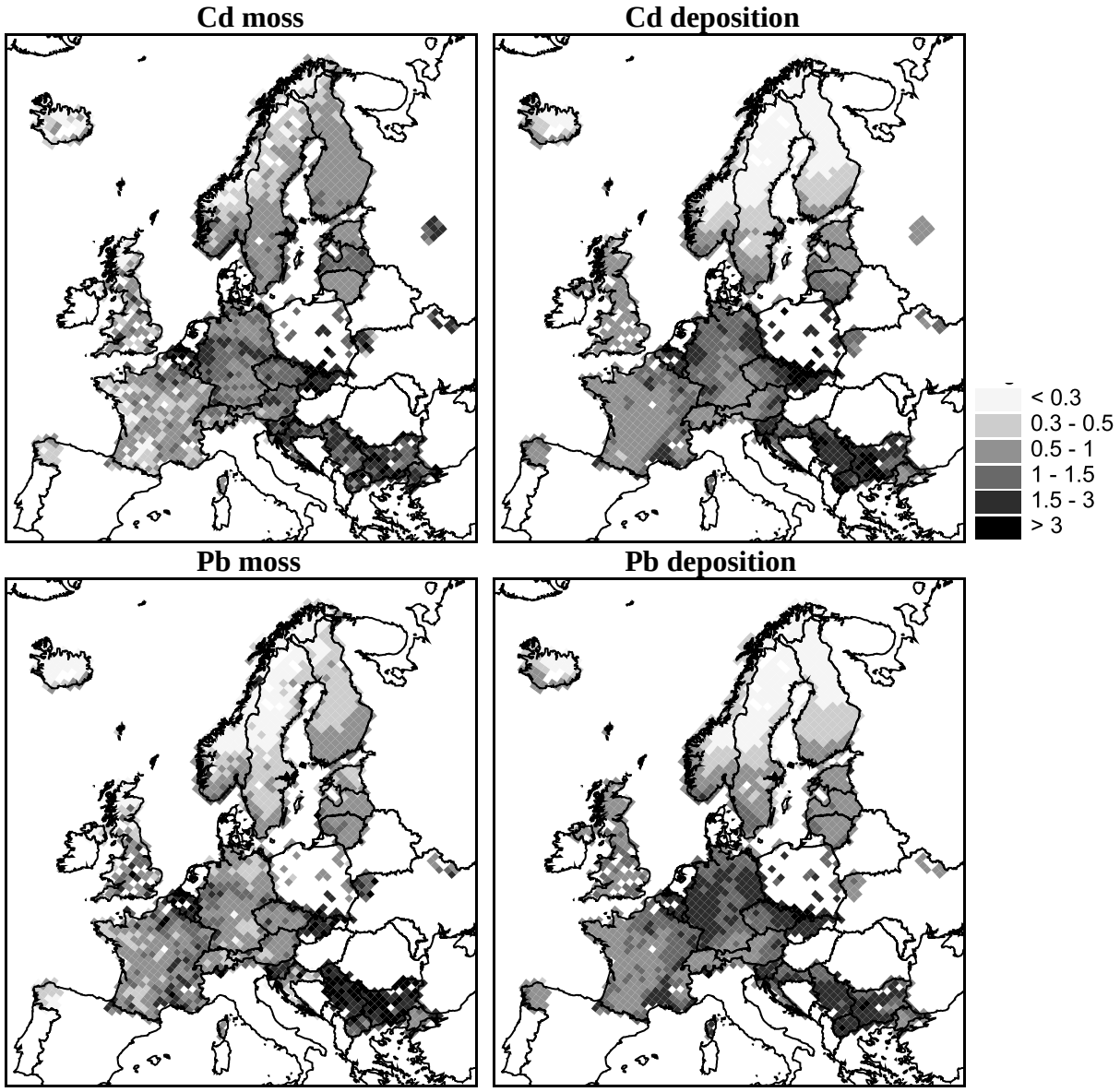


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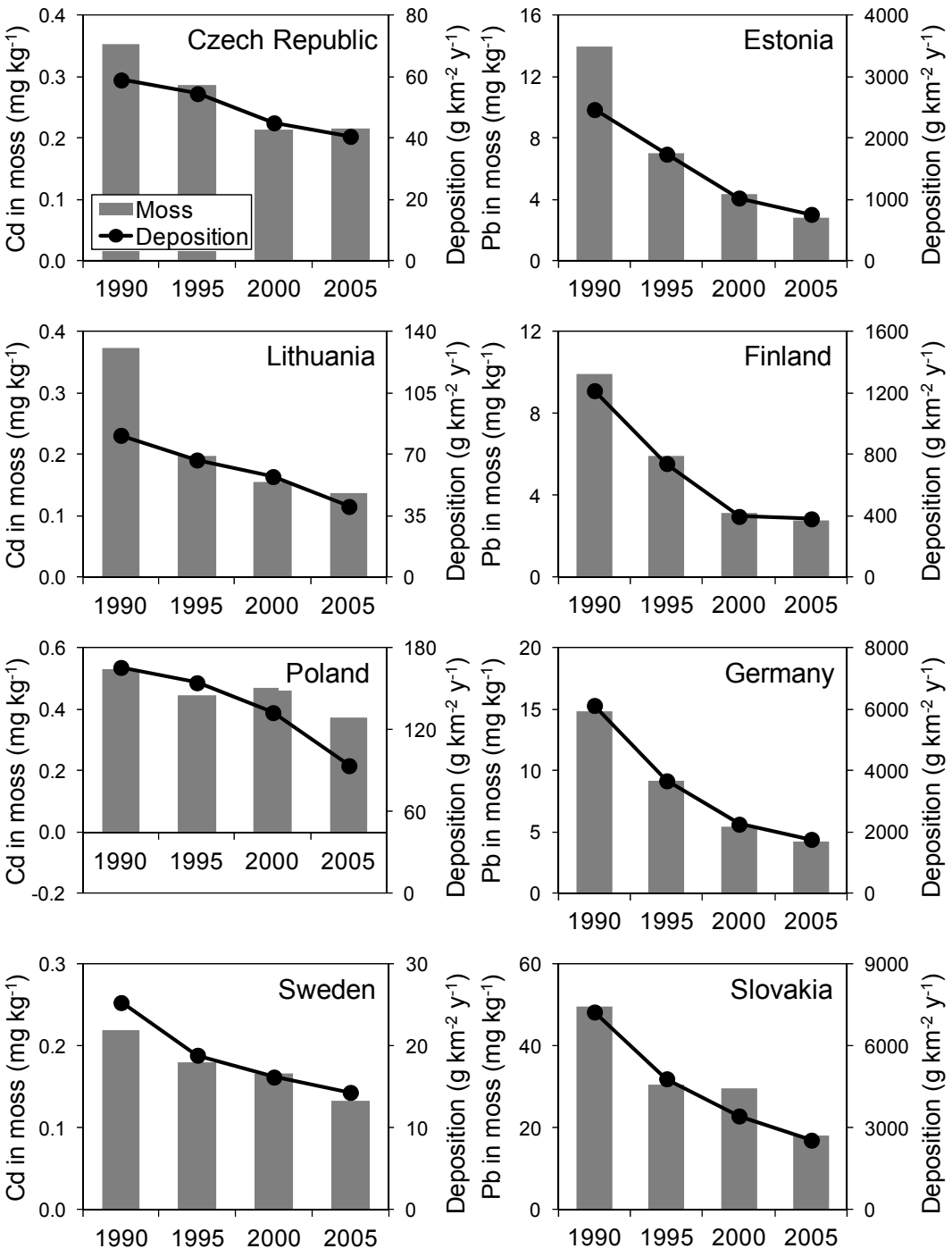
611 **Figure 4.**

612




613

614 **Figure 5.**

615