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Heavy metal and nitrogen concentrations in mosses are declining across Europe whilst some "hotspots" remain in 2010

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Abstract

In recent decades, naturally growing mosses have been used successfully as biomonitors of atmospheric deposition of heavy metals and nitrogen. Since 1990, the European moss survey has been repeated at five-yearly intervals. In 2010, the lowest concentrations of metals and nitrogen in mosses were generally found in northern Europe, whereas the highest concentrations were observed in (south-)eastern Europe for metals and the central belt for nitrogen. Averaged across Europe, since 1990, the median concentration in mosses has declined the most for lead (77%), followed by vanadium (55%), cadmium (51%), chromium (43%), zinc (34%), nickel (33%), iron (27%), arsenic (21%, since 1995), mercury (14%, since 1995) and copper (11%). Between 2005 and 2010, the decline ranged from 6% for copper to 36% for lead; for nitrogen the decline was 5%. Despite the Europe-wide decline, no changes or increases have been observed between 2005 and 2010 in some (regions of) countries.

Capsule: Heavy metal pollution remains high particularly in (south-)eastern Europe, whereas nitrogen pollution remains high in the central belt of Europe.

Keywords: biomonitoring; EMEP maps; heavy metals; nitrogen; moss survey

1. Introduction

Quantification of heavy metal concentrations in selected moss species provides a surrogate, time-integrated measure of the spatial patterns and temporal trends of heavy metal deposition from the atmosphere to terrestrial ecosystems (Harmens et al., 2007, 2008b, 2010). In addition, the nitrogen (N) concentration in mosses provides a good indication of areas exposed to high N deposition across Europe (Harmens et al., 2011, 2014). The analysis of elemental concentrations in mosses is easier and cheaper than conventional deposition analysis. Therefore, a much higher sampling density can be achieved than with deposition analysis (see Aas and Breivik, 2012). Despite potential confounding factors contributing to the variation in elemental concentrations in mosses (as discussed elsewhere in more detail e.g. Aboal et al., 2010; Harmens et al., 2008b, 2010, 2011, 2014 and references therein), the deposition of cadmium (Cd), lead (Pb) and N has been identified as the primary factor contributing to the spatial variation of concentrations in mosses at the European scale (Holy et al., 2009; Schröder et al., 2010a,b). For Cd and Pb, significant positive correlations were found between the concentration in mosses and the deposition modelled by the European Monitoring and Evaluation Programme (EMEP) for about two thirds or more of the countries participating in the European moss survey since 1990 (Harmens et al., 2012). However, correlations are weaker for mercury (Hg; Schröder et al., 2010b). For N, the relationship between concentrations in mosses and measured or modelled atmospheric deposition seems to by asymptotic, with saturation occurring in mosses in areas with high N deposition (Harmens et al., 2011, 2014). This suggests that the use of mosses as biomonitors of atmospheric deposition is primarily suitable for areas with low to medium N deposition. Nevertheless, the concentration in mosses is still a suitable indicator for identifying areas exposed to high atmospheric N deposition at the European scale.

The European moss survey has been repeated at five-yearly intervals since 1990 (Harmens et al., 2010, 2011) and the latest survey was conducted in 2010 with 25 countries reporting heavy metal concentrations and 15 countries reporting N concentrations in mosses (Harmens et al., 2013c). The European moss survey provides data on concentrations of ten heavy metals (arsenic (As), Cd, chromium (Cr), copper (Cu), iron (Fe), Hg, nickel (Ni), Pb, vanadium (V), zinc (Zn)) in naturally growing mosses, and since 2005 also for the metals aluminium (Al) and antimony (Sb) and for N. In 2010, a pilot study was conducted on the application of mosses as biomonitors of selected persistent organic pollutants (Harmens et al., 2013a, b). The geographical distribution of the moss concentrations of studied POPs reflected atmospheric deposition patterns and the level of urbanisation. Until 2000, the European moss survey has been coordinated by the Nordic Council of Ministers and since 2000 it has been coordinated by the ICP Vegetation (International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops) Coordination Centre at the Centre for Ecology and Hydrology, Bangor, UK. The ICP Vegetation was established in 1987 and is one of seven ICPs/Task Forces of the Working Group on Effects that reports to the United Nations Economic Commission for Europe (UNECE) Long-Range Transboundary Air Pollution (LRTAP) Convention on the effects of atmospheric pollutants on the environment and human health.

In the current paper, we report on the spatial patterns in 2010 and temporal trends of concentrations in mosses across Europe for heavy metals since 1990. Addition of the 2010 data has enabled the assessment of temporal trends in the last two decades. As the emissions and depositions of heavy metals (EEA, 2012; Travnikov et al., 2012) and to a lesser extent N (Fagerli et al., 2012) have declined across Europe in recent decades, we hypothesize that the concentrations of these metals in mosses have declined too. For the first time, we assessed similarities in the spatial variation of groups of metals by applying factor analysis at the European scale. Based on studies conducted at national level, we expect some distinct factors to be identified.

2. Materials and methods

Moss sampling

Moss samples of several carpet forming moss species were collected across Europe in 2010/2011 (Harmens et al., 2013c); throughout the paper we will refer to this as the survey from 2010. The moss sampling procedure and further preparation of the material for elemental analysis was done according to the guidelines described in the protocol for the 2010 survey (ICP Vegetation, 2010). Figure 1 shows the moss sampling sites across Europe in 2010: ca. 4,400 for heavy metals and ca. 2,400 for N. *Pleurozium schreberi* (Brid.) Mitt was the most frequently sampled species, accounting for ca. 42% of the samples for both heavy metals and N, followed by *Hylocomium splendens* (Hedw.) (23.5% and 15.3% for heavy metals and N, respectively) or *Hypnum cupressiforme* Hedw. (19.6% and 26.9%, respectively), and *Pseudoscleropodium purum* (Hedw.) (7.7% and 7.5%, respectively). Other moss species constituted 7.1% and 8.7% of the mosses sampled for heavy metals and N respectively. In some countries only selected areas were sampled, i.e. in Denmark mosses were collected from the Faroe Islands, in Italy from the Bolzano region, in the Russian Federation from Ivanovo, Kostromskyaya and Tikhvin-Lenigradskaya region, in Spain from Galicia, Navarra and Rioja region, and in Ukraine from Donetsk region.

Elemental analysis and quality assurance

The concentration of heavy metals and N were determined by a range of analytical techniques (Harmens et al., 2013c). All metal concentrations were expressed as mg kg^{-1} dry weight at 40 ^{o}C. As in previous surveys, a quality control exercise was conducted for assessing the analytical performance of the participating laboratories (Steinnes et al., 1997; Harmens et al., 2010). Moss reference material M2, containing elevated concentrations for most metals, and M3, containing background concentrations for most metals (Steinnes et al., 1997), were distributed amongst participating laboratories. Recommended values for the N concentration in M2 and M3 were established in the 2005 European moss survey (Harmens et al., 2010). In addition, some laboratories used other certified reference material for quality assurance. For determination of the elemental concentrations in the reference material, laboratories followed the same analytical procedure as used for the collected moss samples. Generally, data obtained indicated acceptable agreement between laboratories. However, outliers were identified for some laboratories for selected metals. This was considered the case when the values were outside the range of two standard deviations (as determined for the 2010 survey) from the mean recommended value for reference material M2 and/or M3 (Steinnes et al., 1997; Harmens et al., 2010). In 2010, the mean values ranged from 85% (for As; followed by 93% for V) to 105% (Sb) of the recommended values for M2 and from 92% (Cr) to 113% (As) for M3. For N the mean values of M2 and M3 were 101% and 102% of the recommended value respectively. Correction factors were applied when both M2 and M3 values were outliers for a specific metal, and sometimes corrections factors were also applied when only one reference value was identified as an outlier. Although applying correction factors enhanced compatibility of data between countries, it hardly affected the overall European mean and median values for the elements. As a consequence, it did not significantly affect the temporal trends reported for the whole of Europe (Harmens et al., unpublished).

Mapping

Maps were produced according to the method described by Harmens et al. (2008a); they show the mean concentration of each metal within individual EMEP grid squares (50 km x 50 km). Please note that the designations employed and the presentation of material in this paper do not imply the expression of any opinion whatsoever on the part of the United Nations concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries.

Statistical analysis

Factor analysis was used to identify how metals grouped together at sampling sites across Europe in 2010. In R (R Core Team, 2014; version 3.1.1) a correlation matrix was created from the logged metal concentrations in mosses. The ‘fa’ function of the R package ‘psych’ was used to perform a factor analysis using orthogonal (varimax) rotation and the maximum likelihood (ml) factoring method. The factor analysis was run using the correlation matrix, therefore variables were standardised (each has a variance of 1). The number of factors was set to three, based on examination of a scree plot. As data for Sb was only collected from 9 out of 25 countries, Sb was excluded from the factor analysis. As not all countries reported data for all metals, there were some missing values in the matrix, which were dealt with using pair-wise deletion, i.e. only missing values per site were removed from the analysis rather than all the measurements for a site.

Statistical analysis of temporal trends between 1990 and 2010 across Europe was performed according to the method described by Harmens et al. (2010). For each metal, data were only included for those countries that had determined the concentration for at least four out of the five survey years, although for Hg some countries were also included that had reported data for three out of the last four survey years. For Al, Sb and N, the temporal trend was determined between 2005 and 2010. For each metal, a general linear model including the geometric mean as the response and country and year as factors was then run using R. To account for differences in sample size between countries, the number of samples was included as a weights argument. Tukey tests were used to perform pairwise comparisons between years for each element.

3. Results

Spatial patterns of heavy metals in 2010

Spatial patterns of heavy metal concentrations in mosses in Europe in 2010 are shown in Figures 2 to 4 and median concentrations per country and metal are shown in Figure S1. Additional information per country is provided in Table S1. Please note that when we refer to countries, this does not necessarily mean the whole country but could mean a region(s) within a country if sampling was not across the whole country. The factor analysis identified three main factors explaining 68% of the total variance (Table 1). Factor 1 can possibly be explained by elements associated with mineral particles, mainly windblown dust, and contributes to 39% of the variance. Factor 1 is dominated by Al, Fe, V, Cr, but also includes As and Ni. Factor 2 is probably associated with long-range transport of air pollution and contributes to 18% of the variance. It is dominated by Cd and Pb, but also includes Zn. Factor 3 is dominated by Ni and Cu, and is most likely associated with local pollution sources; factor 3 contributes to 11% of the variance. Hg is not strongly associated with any of the factors, which most likely reflects its global nature and different chemistry compared to other metals, affecting its accumulation in mosses. The spatial variation in the strength of the association of the different elements contributing to each factor is shown in Figure S2.

The lowest concentrations of heavy metals in mosses were generally found in northern Europe and the highest concentrations in eastern and south-eastern Europe, resulting in a north-west to south-east gradient for many metals in 2010 (Figures 2 – 4).

This is particularly true for metals most likely associated with windblown dust. For Al, Fe, V and Cr, the highest median concentrations in mosses were generally found in Romania, Macedonia, Albania, Ukraine (Donetsk region) and Bulgaria (Figure S1 and Table S1). High Cr concentrations were found in Iceland. For the Faroe Islands (part of Denmark), relatively high concentrations of Al, Fe and V were also reported, but the concentration for Cr was rather low. Cr concentrations were also low in the Ukraine. Arsenic concentrations in mosses were particularly high in Macedonia, Romania, Bulgaria and Rioja (Spain), whereas the lowest concentrations were reported for the Faroe Islands.

For Cd, the highest median concentrations in mosses were observed in southern Poland, Slovakia, Croatia, Ukraine (Donetsk region), Belgium and Slovenia (Figure S1 and Table S1). However, in Belgium the median value has declined by 38% since 2005. Cd levels were lowest in north-west Scandinavia, Iceland, and western parts of France. Relatively low median values were also observed in Albania, Kosovo and regions in the Russian Federation. The highest Pb concentrations were found in southern Poland, Slovakia, Bulgaria, Kosovo, Ukraine (Donetsk region) and Slovenia, although the median concentration has declined between 31% and 50% in Slovakia, Bulgaria and Slovenia since 2005. Median Pb concentrations in mosses were lowest in northern European countries. In contrast to Cd and Pb, the Zn concentration in mosses has a rather homogenous distribution across Europe, with locally or regionally elevated concentrations being observed. The highest median values were found in Ukraine (Donetsk region), Poland, Belgium, Romania and Kosovo, whereas the lowest median values were reported for Albania, Faroe Islands, Macedonia, Iceland and Bulgaria.

Although the highest Cu concentrations in mosses were also found in parts of eastern Europe, i.e. Ukraine (Donetsk region), Slovakia, Russian Federation and

Bulgaria, low concentrations were reported in Albania, Kosovo and Macedonia (Figure S1 and Table 1). Ni concentrations were generally high in many parts of (south-)eastern European countries, however, high concentrations were also observed in Iceland and concentrations were low in Belarus. Whereas low concentrations of Cu and Ni were generally observed in northern Europe, locally high concentrations were detected at the Norwegian-Finnish-Russian border in the north due to the presence of strongly polluting Cu-Ni smelters in the Kola Peninsula at the Russian side of the border.

Hemispheric transport of Hg appears to result in a rather homogenous spatial pattern of Hg concentration in mosses across Europe. The highest levels of Hg were found in Albania and Macedonia, followed by Italy (Bolzano region), Poland and France. Relatively high levels of Hg were also reported for Norway and the Faroe Islands, and levels have increased since 2005 in some parts of Norway. High concentrations of Sb in mosses in Romania likely indicate a combination of high industrial and road traffic pollution in large areas of the country. High Sb concentrations were also reported for Slovenia and for highly-populated areas with high traffic density in other countries such as north-western France (including Paris), eastern Austria (Lower Inntal) and south-eastern Norway (around Oslo).

Temporal trends of heavy metals since 1990 (or later years)

Generally, heavy metal concentrations in mosses have continued to decline at the European scale between 2005 and 2010 (Figures 5 and 6, Tables 2 and 3). Between 1990 and 2010, the European average geometric mean concentration has declined significantly ($P < 0.001$) for all metals that were reported for that period. For As, the concentration had declined between 1995 and 2010, although not significantly at $P = 0.05$. For Hg, the concentration did not decline significantly between 1995 and 2010 (P

= 0.27; Table 2). Please note that the temporal trends between 2005 and 2010 based on countries that participated in both survey years (Table 3) can differ from the trends observed for the same period when comparing long-term trends between 1990 and 2010 (Table 2). The latter is based on countries that participated in four out of the five survey years. This generally includes some different and sometimes a lower number of countries in comparison to those participating in both the 2005 and 2010 survey, for example some countries have only participated in the most recent two or three surveys.

Between 1990 and 2010, the average median Pb and Cd concentration in mosses across Europe has declined by 77% and 51% respectively (Table 3). These declines are similar to those reported by EMEP for the modelled deposition across Europe, i.e. 74% and 51% for Pb and Cd respectively (Figure 5). The 14% decline in Hg between 1995 and 2010 was lower than the decline (27%) in EMEP modelled deposition across Europe. The average median As concentration in mosses has declined by 21% since 1995 and the average median concentration in mosses has declined between 11% (Cu) and 57% (V) for all the other metals between 1990 and 2010. For Al and Sb, the decline was 28% and 23% respectively since 2005 (Table 3).

Despite the further general decline in heavy metal concentrations in mosses at the European level between 2005 and 2010, no changes or increases have been observed at the (sub-)country level. For example, several countries reported an increase in Cd and Cu, an increase in Hg was found in Macedonia and Italy (Bolzano region), and an increase in Ni was found in Iceland and Croatia. Some clear country-specific results were also observed when comparing the results of the 2005 and 2010 European moss survey. In Belgium, the concentration in mosses has declined considerably for all metals since 2005. The same is true for Slovenia and Macedonia, although the decline has been lower compared with Belgium; for Macedonia, no change (As) or an increase (Hg) were also observed. In France, the concentration of metals associated with windblown dust has declined considerably between 2005 and 2010, particularly in eastern and southern parts of the country. For further details we refer to Harmens et al. (2013c).

Nitrogen

The spatial pattern of the N concentration in mosses was similar in 2005 and 2010, with lower values being observed for Finland than the rest of Europe (Figure 7). Generally, high concentrations were found in western, central and south-eastern Europe. The European average median or geometric mean value has not changed significantly since 2005 (a decline of 5%; Table 2). Whilst a considerable decline (30%) in the median N concentration in mosses was reported for Slovenia since 2005, an increase was reported for the Czech Republic (19% increase) and France (15%).

4. Discussion

Heavy metals – spatial patterns in 2010 and temporal trends since 1990

As in previous surveys (Harmens et al., 2010), the lowest concentrations of heavy metals in mosses were generally found in northern Europe and the highest concentrations in eastern and south-eastern Europe, resulting in an north-west to south-east gradient. For many metals (but not all) a north-south gradient is present in Scandinavia, reflecting both the higher population density in the south and the contribution of long-range transboundary air pollution from central Europe to the higher concentrations in the south (Steinnes et al., 2011). The three main factors identified in the factor analyis appear to be best explained by 1) windblown dust, 2) long-range transport of air pollution and 3) local pollution sources. While the factor analysis is useful for investigating broad trends in the associations of metals across Europe, the results do not provide any information on variation within individual countries (e.g. due to the presence of local point sources). Some countries have conducted factor analysis in more detail and have identified additional factors for 2010 by including more elements (e.g. Barandovski et al., 2013; Qarri et al., 2014; Špirić et al., 2013).

In recent decades, the general decline in emissions and subsequent deposition of heavy metals has resulted in a decrease in the heavy metal concentration in mosses at the European scale. Many emission sources have become cleaner, for example by using filters or other best available technologies, by changing from coal to gas as cleaner emission source or phasing out leaded petrol in many parts of Europe (Travnikov et al., 2012). In addition, some very polluting local emission sources have been shut down in recent decades. Emission abatement policies developed under the Convention on Long-range Transboundary Air Pollution (LTRAP) have targeted the metals Cd, Hg and Pb in the 1998 Aarhus Protocol on Heavy metals; the Protocol was amended in 2012. The implementation of emission abatement policies have contributed significantly to the decline in Cd and Pb pollution in Europe in recent decades. Since 1990, anthropogenic emissions in the EMEP region have declined by 90% for Pb and by approximately 65% for Cd and 60% for Hg. Nevertheless, long-range transboundary transport still contributes significantly to metal deposition in the majority of European countries (Travnikov et al., 2012). Because of the more hemispheric nature of transport of elemental Hg, European emission abatement policies are expected to have less impact on Hg than Cd and Pb deposition due to the contribution from other continents to Hg deposition in Europe. Nowadays, intercontinental transport is estimated to contribute more than 65% to total Hg deposition in the EMEP region (Travnikov et al., 2012). Therefore, both regional and global efforts are needed to reduce Hg pollution. Hence, in October 2013, the Minamata Convention on Mercury (a global Convention) was adopted by the Governing Council of the United Nations Environment Programme (UNEP). For Hg, large areas in Europe are still at risk of high critical loads exceedance for ecotoxicological effect. The risk of exceedance is lower for Pb, but exceedance still occurs in large areas of Europe, and the risk of exceedance is almost non-existent for Cd (Slootweg et al., 2010).

For Cd and Pb, the decline in the concentration in mosses since 1990 (77% and 51% respectively) was in good agreement with the decline in atmospheric deposition modelled by EMEP across Europe (74% and 51% respectively; Travnikov et al., 2012). For Hg, the decline in concentration in mosses (14%) was less than the decline in atmospheric deposition modelled by EMEP across Europe (27%). Similar results were found when the comparison of temporal trends for modelled deposition and concentrations in mosses was limited to the areas of Europe where countries reported concentrations in mosses in all survey years since 1990 (1995 for Hg; Ilyin et al., 2014). Previous analyses on the European scale have shown that EMEP modelled deposition is the main predictor for concentrations in mosses for Cd and Pb but not for Hg (Holy et al., 2009; Schröder et al., 2010b). It should be noted, however, that correlations between modelled deposition and concentrations in mosses for Cd and Pb are country-specific (Harmens et al., 2012; Ilyin et al., 2014). The lack of a strong correlation between modelled deposition and concentration in mosses for Hg may relate to the specific chemistry of Hg (Harmens et al., 2010, and references therein). Reduction of heavy metal pollution levels was accompanied by changes in the key source categories of both emissions and resulting deposition. For example, the prevailing contribution of road transport for Pb and metal production for Cd in 1990 were replaced by industrial and non-industrial combustion in 2010. Changes in sectoral composition of Hg emissions were less significant (Travnikov et al., 2012).

In agreement with the moss data, emissions for Cu have changed (0.5%) the least between 1990 and 2010 in Europe (EEA, 2012). For other metals, the decline in emissions between 1990 (1995 for As) and 2010 has been higher than the decline in concentrations in mosses in Europe, i.e. 30%, 73%, 59% and 43% for As, Cr, Ni and Zn respectively (EEA, 2012). The decline in deposition of heavy metals is generally lower than the decline in emissions possibly due to the contribution of wind-blown dust to the deposition of metals (Ilyin et al., 2007; Travnikov et al., 2012). Elevated fluxes of dust suspension are generally calculated for southern Europe and agricultural regions in southeastern and Eastern Europe (Ilyin et al., 2007). Whilst anthropogenic emissions of metals have generally declined in Europe, the relative contribution from wind re-suspension to metal deposition has increased between 1990 and 2010. Wind-blown dust consists of two components: the first component represents re-suspension of mineral dust with a natural content of metal that corresponds to the average metal concentration in the Earth' crust. This fraction tends to be high for metals associated with the Earth' crust, such as Al and Fe. The second component accounts for the legacy contribution of metals accumulated in soil and roadside dust due to previous (historic) atmospheric deposition. This fraction tends to be high for metals such as Cd and Pb (Ilyin et al., 2007). It should be noted that large uncertainties are associated with the legacy contribution of metals in soils and therefore the contribution of wind-blown dust to the calculated total deposition of metals. It appears that the currently high concentrations of metals in mosses in parts of eastern and south-eastern Europe are the result of a combination of the presence of still high local anthropogenic pollution sources, a high legacy component of heavy metals in windblown dust and the presence of mineral soils of some of these countries (Ilyin et al., 2007). For essential plant micronutrients such as Cu, Fe and Zn, background concentrations will be present in mosses due to internal cycling from old to new growing tissue, which also contributes to a lower decline in concentration in mosses compared to emissions.

Even in times of generally decreasing metal emission and deposition across Europe, temporal trends are different for different geographical scales, i.e. temporal trends are country or region-specific with no changes or even increases in metal concentrations in mosses being found (Harmens et al., 2010). Whereas many areas in a country have shown a decline, areas in the same or another country have shown no change or a considerable increase in metal concentrations in mosses since the previous survey in 2005 (Harmens et al., 2013c). Potentially confounding factors affecting temporal trends of metal concentrations in mosses at the country and European scale were discussed in more detail previously (Harmens et al., 2008b; 2010).

Nitrogen – spatial patterns in 2010 and temporal trends since 2005

For N, hardly any changes were observed in the N concentration in mosses since 2005. The non-significant decline (5%) in the European average median N concentration in mosses is in agreement with the 7% decline reported by EMEP for modelled total N deposition in the EU27 since 2005 (Fagerli et al., 2012). As in 2005, areas most exposed to high N deposition are located in western and central Europe. However, the magnitude of exposure in many northern, eastern and Mediterranean countries could not be assessed via the survey as those countries did not report on N concentrations in mosses. Whereas Germany participated in the 2005 survey, it did not participate in the 2010 survey, leaving a big gap in the data for central Europe. The relationship between site-specific N concentrations in mosses and modelled or measured deposition starts to show saturation at deposition rates of ca. 15 (Harmens et al., 2011) or 20 kg ha^{-1} y^{-1} (Harmens et al., 2014). Although this makes it difficult to assess the magnitude of exposure in areas with medium to high N deposition, the moss technique still allows the identification of the areas potentially most exposed. Some country-specific changes since 2005 are confounded by inclusion of data for more sites in 2010. For example, the relatively high decline in the median N concentration in mosses in Slovenia between 2005 and 2010 can be explained by including additional sampling sites (only about one-third of the sampling sites were the same in 2005 and 2010) and careful sampling to avoid the influence of canopy drip from trees as much as possible in the highly forested area of Slovenia. Generally, mosses affected by canopy drip have a higher N due to the higher N concentration in throughfall deposition (Kluge, 2013, Skudnik, 2014). The reported increase in France was confounded by the fact that the N concentration in mosses was determined at 88 sites in 2005, whereas it was determined at 442 sites in 2010.

Conclusions

The following conclusions can be drawn:

- Generally, areas in eastern and south-eastern European remain exposed to high levels of heavy metal pollution, whereas areas in the central belt of Europe remain exposed to high levels of N pollution. Participation of countries from these regions in future moss surveys is therefore highly recommended to monitor changes in the future.
- The implementation of air pollution abatement strategies in Europe in recent decades has contributed considerably to the general decline in heavy metal concentrations in Europe. The slower implementation of air pollution abatement policies in parts of eastern and south-eastern Europe has likely contributed to this area still having high levels of heavy metal pollution.

- Despite the general European decline in concentrations in mosses, country and region-specific temporal trends were observed, including no changes or increases in recent surveys.

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Figure legends

Figure 1. Sites where mosses were sampled for heavy metals (left) and nitrogen analysis (right).

Figure 2. Mean heavy metal concentration in mosses per EMEP grid square (50 km x 50 km) in Europe in 2010 for aluminium (Al), antimony (Sb), arsenic (As) and cadmium (Cd).

Figure 3. Mean heavy metal concentration in mosses per EMEP grid square (50 km x 50 km) in Europe in 2010 for chromium (Cr), copper (Cu), iron (Fe) and lead (Pb).

Figure 4. Mean heavy metal concentration in mosses per EMEP grid square (50 km x 50 km) in Europe in 2010 for mercury (Hg), nickel (Ni), vanadium (V) and zinc (Zn).

Figure 5. Average median metal concentration in mosses (± one SE) for countries that reported metal data for at least four survey years since 1990 (some countries reported three survey years since 1995 for Hg). The dots in the graphs show the decline in deposition across Europe as modelled by EMEP (Travnikov et al., 2012).

Figure 6. Average median metal concentration in mosses (± one SE) for countries that reported metal data for at least four survey years since 1990 (1995 for arsenic).

Figure 7. Mean nitrogen (N) concentration in mosses per EMEP grid square (50 km x 50 km) in Europe in 2010 (left) and medium value per country (right). Italy: Bolzano region, Spain: Navarra region.

Table 1. VARIMAX rotated factors of metal concentrations in mosses.

Loadings and explained variance of the first 3 factors are listed. Loadings higher than 0.5 are shown in bold.

Metal	Factor 1	Factor 2	Factor 3
Aluminium	**0.97**	0.20	0.12
Arsenic	**0.71**	0.24	0.24
Cadmium	0.23	**0.85**	0.03
Copper	0.29	0.41	**0.51**
Chromium	**0.79**	0.13	0.33
Iron	**0.89**	0.21	0.25
Lead	0.43	**0.77**	0.08
Mercury	0.33	0.15	0.19
Nickel	**0.52**	0.05	**0.74**
Vanadium	**0.85**	0.08	0.23
Zinc	-0.03	**0.52**	0.12
Variance	4.30	1.94	1.16
Variance (%)	39	18	11

Table 2. Average geometric mean values of heavy metal concentrations in mosses for countries that analysed these metals in at least four out of five survey years. The statistical significance (p-value) of survey year is also shown; for each metal, different letters indicate significant differences (at P = 0.05) between years.

Metal (no. of countries)	Average geometric mean (mg kg^{-1} or % for N)					P-value
	1990	1995	2000	2005	2010	Year
As (11)[1]	-	0.25[a]	0.21[a]	0.22[a]	0.20[a]	0.054
Cd (21)	0.37[a]	0.30[a,b]	0.23[b,c]	0.19[c]	0.19[c]	<0.001
Cr (20)	2.50[a]	2.18[a,b]	2.13[b,c]	1.91[a,b,c]	1.50[c]	<0.001
Cu (20)	7.62[a]	7.51[a,b]	6.90[b]	6.63[b]	6.99[b]	<0.001
Fe (20)	689[a]	589[a,b]	600[a,b]	506[b]	517[b]	<0.001
Hg (11)[1]	-	0.057[a]	0.054[a]	0.055[a]	0.050[a]	0.27
Ni (20)	2.63[a]	2.17[a,b]	2.19[a,b]	1.90[b]	1.82[b]	<0.001
Pb (19)	15.3[a]	9.23[b]	7.17[c]	4.95[c,d]	3.69[d]	<0.001
V (18)	3.92[a]	3.17[a,b]	2.88[b,c]	1.73[c,d]	1.82[d]	<0.001
Zn (22)	49.0[a]	37.7[a,b]	38.4[a,b]	33.1[b]	32.9[b]	<0.001
Al (13)[2]	-	-	-	1151[a]	812[a]	0.24
Sb (7)[2]	-	-	-	0.15[a]	0.11[a]	0.070
N (14)[2]	-	-	-	1.28[a]	1.21[a]	0.74

[1] For As and Hg sufficient data were only available for 1995 - 2010.

[2] For Al, Sb and N data were only available for 2005 and 2010.

Table 3. Decline in the average median heavy metal and N concentrations in mosses since the start of the European moss survey in 1990[1] and since the previous survey in 2005[2].

Element	Decline since 1990[3] (%)	Decline since 2005 (%)	Element	Decline since 1990 (%)	Decline since 2005 (%)
As	21	25	Pb	77	36
Cd	51	7	V	57	27
Cr	43	23	Zn	34	7
Cu	11	6			
Fe	27	15	Al	n.a.	28
Hg	14	20	N	n.a.	5
Ni	33	12	Sb	n.a.	23

[1] Based on data from countries that participated in at least four out of five survey years. For As countries were included that participated in four survey years since 1995, for Hg some countries were included that had data for three out of four survey years since 1995.

...[2] Based on data from countries that participated in both survey years.

[3] Decline since 1995 for As and Hg.

...n.a. = not available.

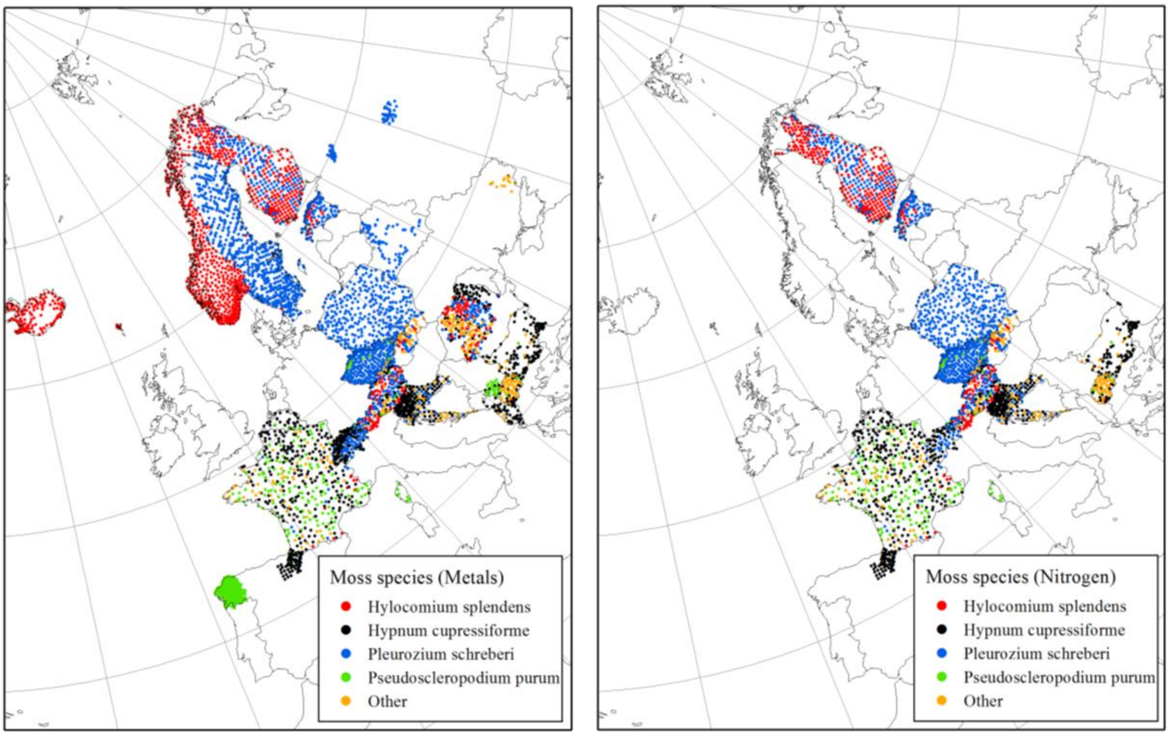


Figure 1. Sites where mosses were sampled for heavy metals (left) and nitrogen analysis in 2010 (right).

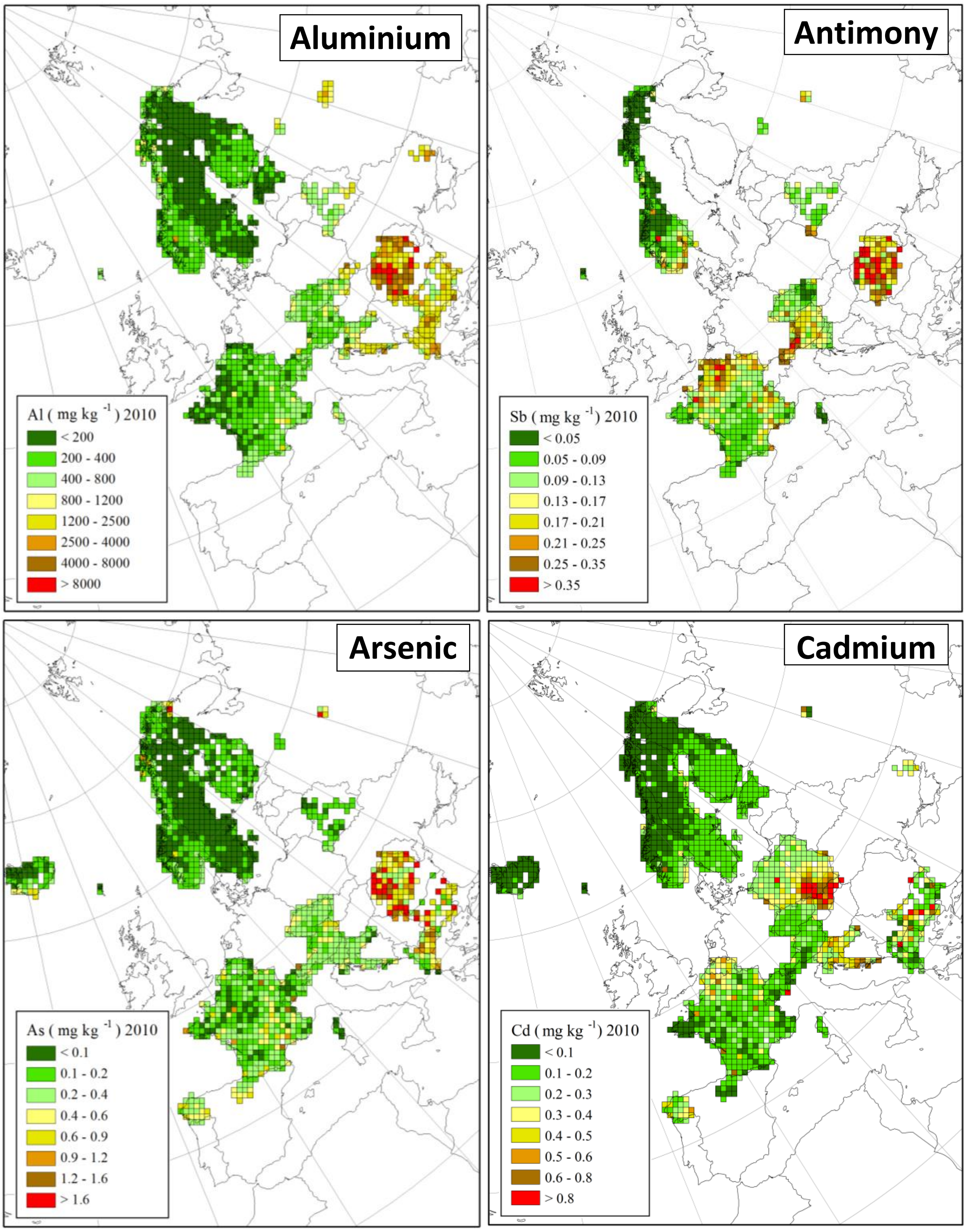
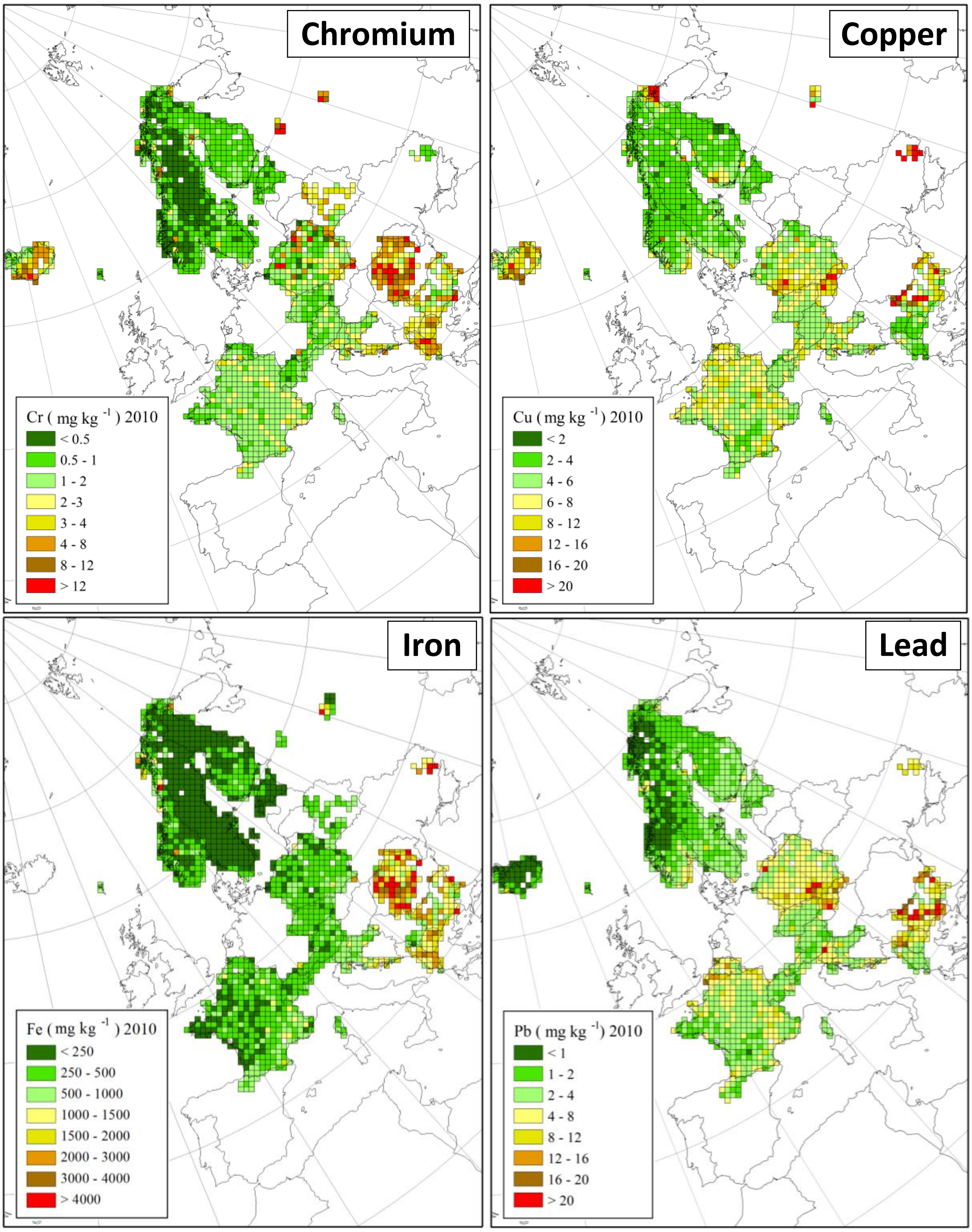


Figure 2. Mean heavy metal concentration in mosses per EMEP grid square (50 km x 50 km) in Europe in 2010 for aluminium (Al), antimony (Sb), arsenic (As) and cadmium (Cd).



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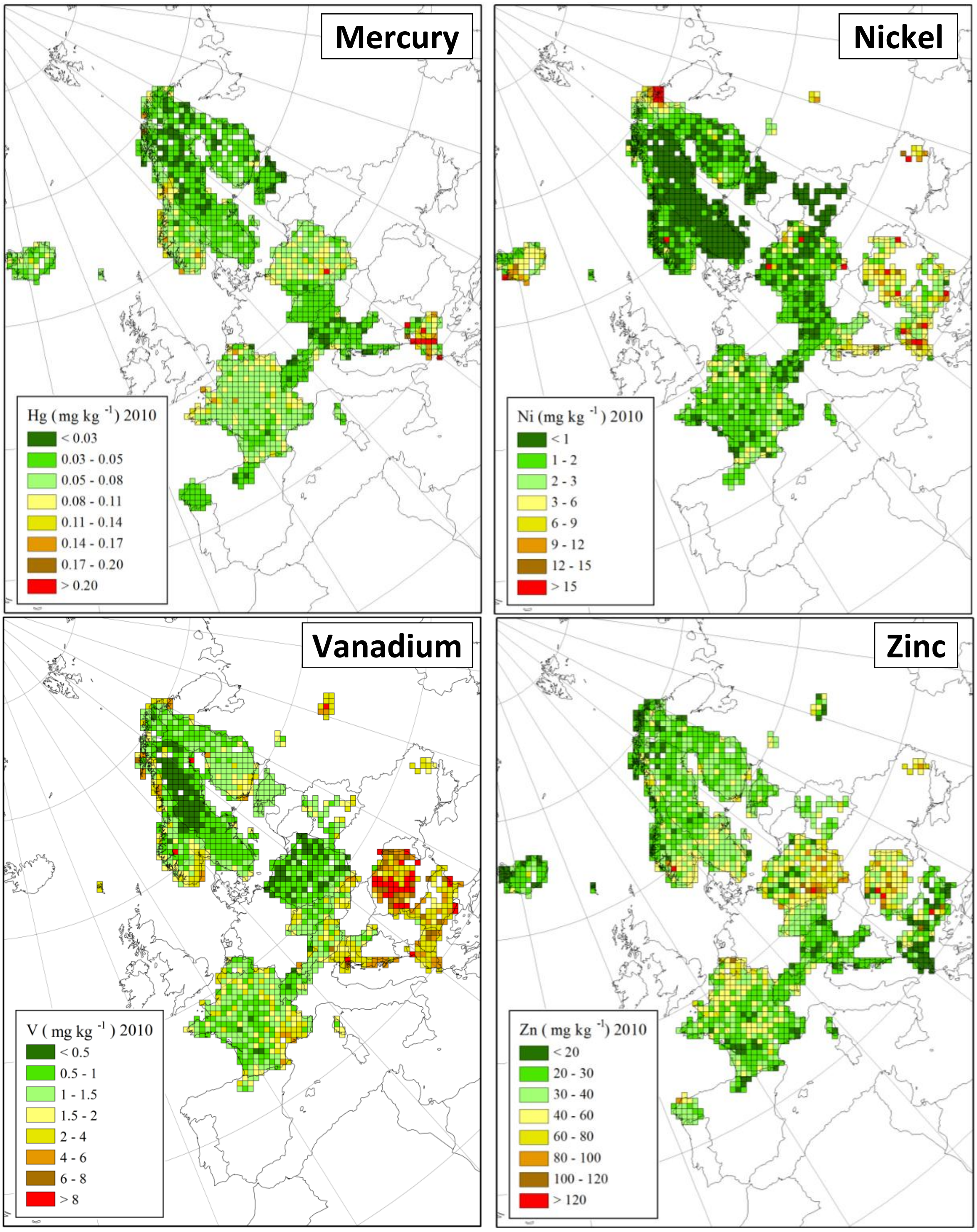


Figure 4. Mean heavy metal concentration in mosses per EMEP grid square (50 km x 50 km) in Europe in 2010 for mercury (Hg), nickel (Ni), vanadium (V) and zinc (Zn).

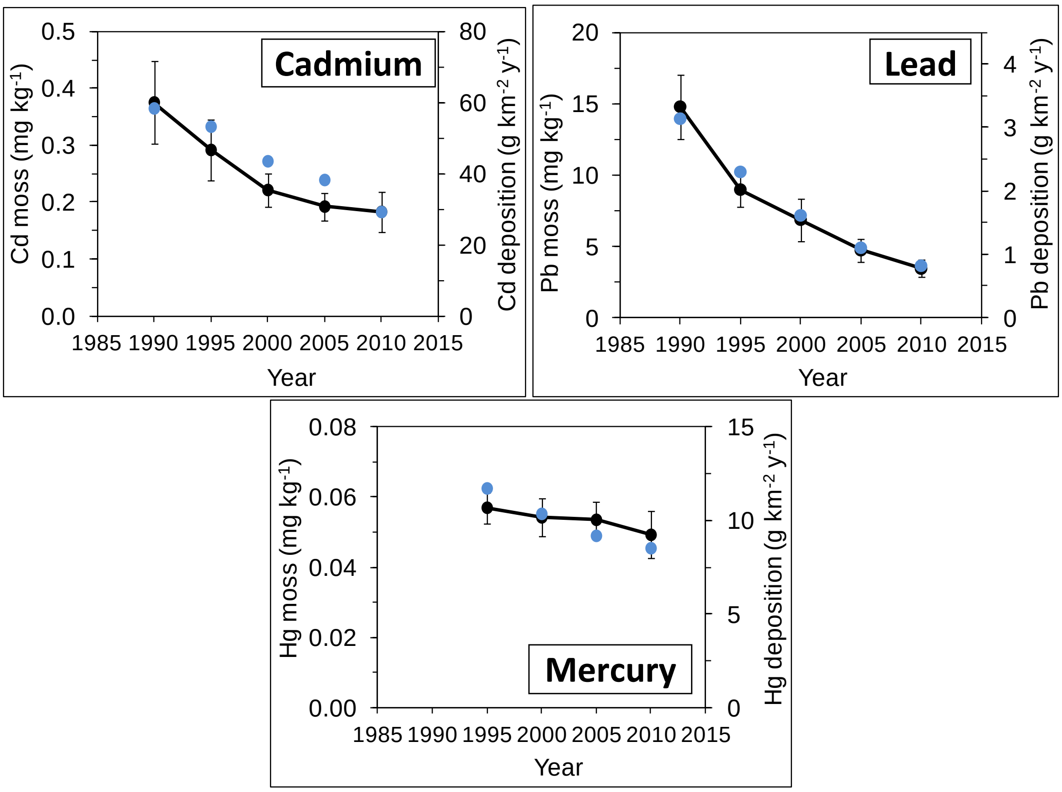


Figure 5. Average median metal concentration in mosses (± one SE) for countries that reported metal data for at least four survey years since 1990 (some countries reported three survey years since 1995 for mercury). The dots in the graphs show the decline in deposition across Europe as modelled by EMEP (Travnikov et al., 2012).

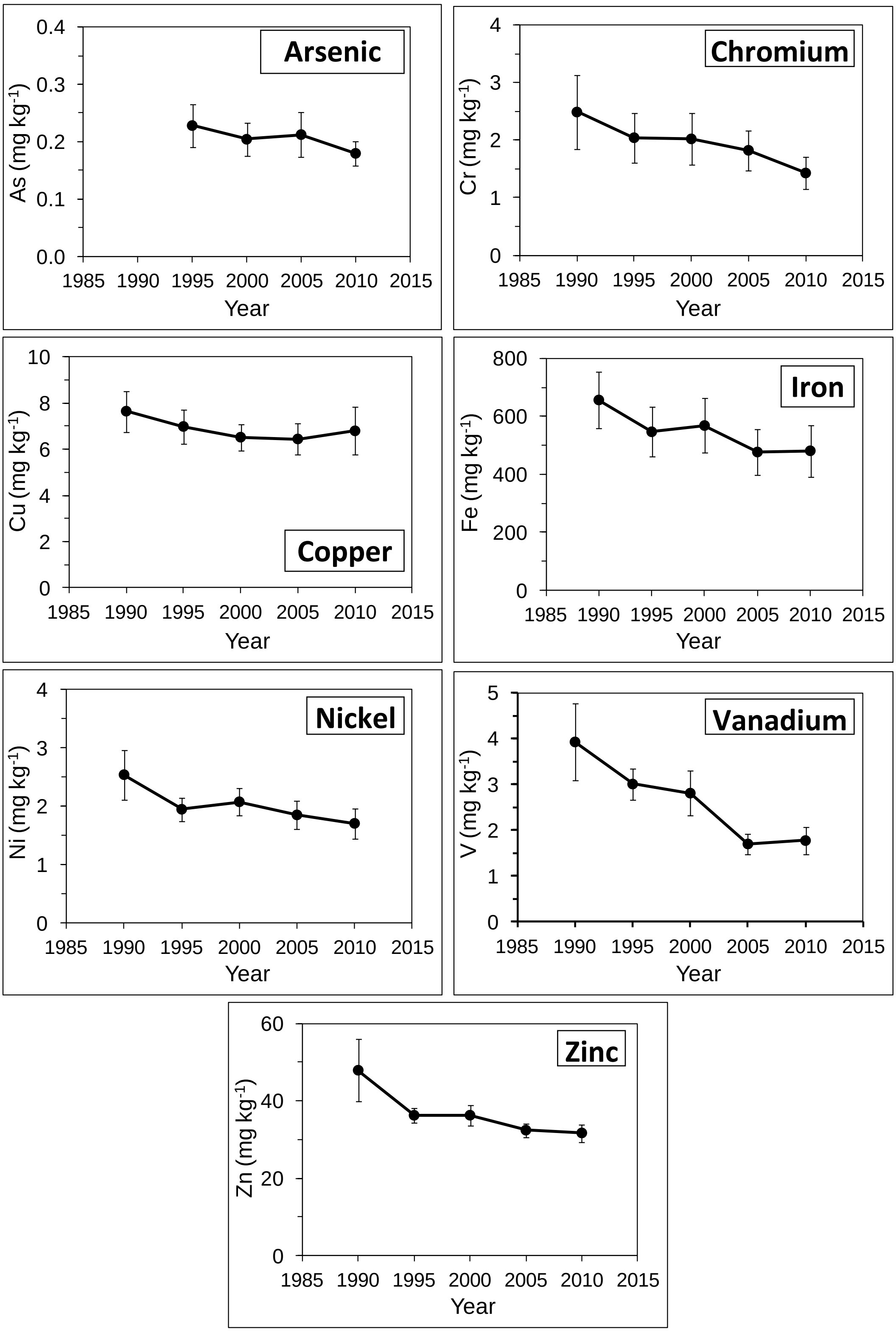


Figure 6. Average median metal concentration in mosses (± one SE) for countries that reported metal data for at least four survey years since 1990 (1995 for arsenic).

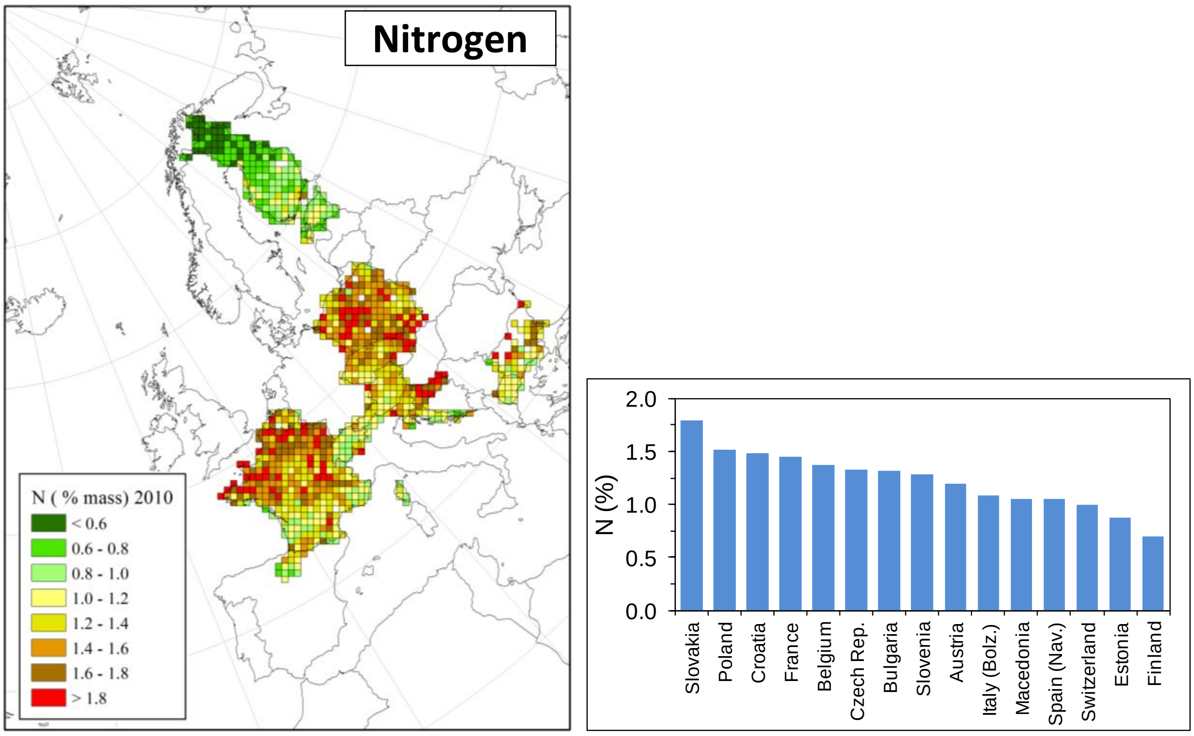
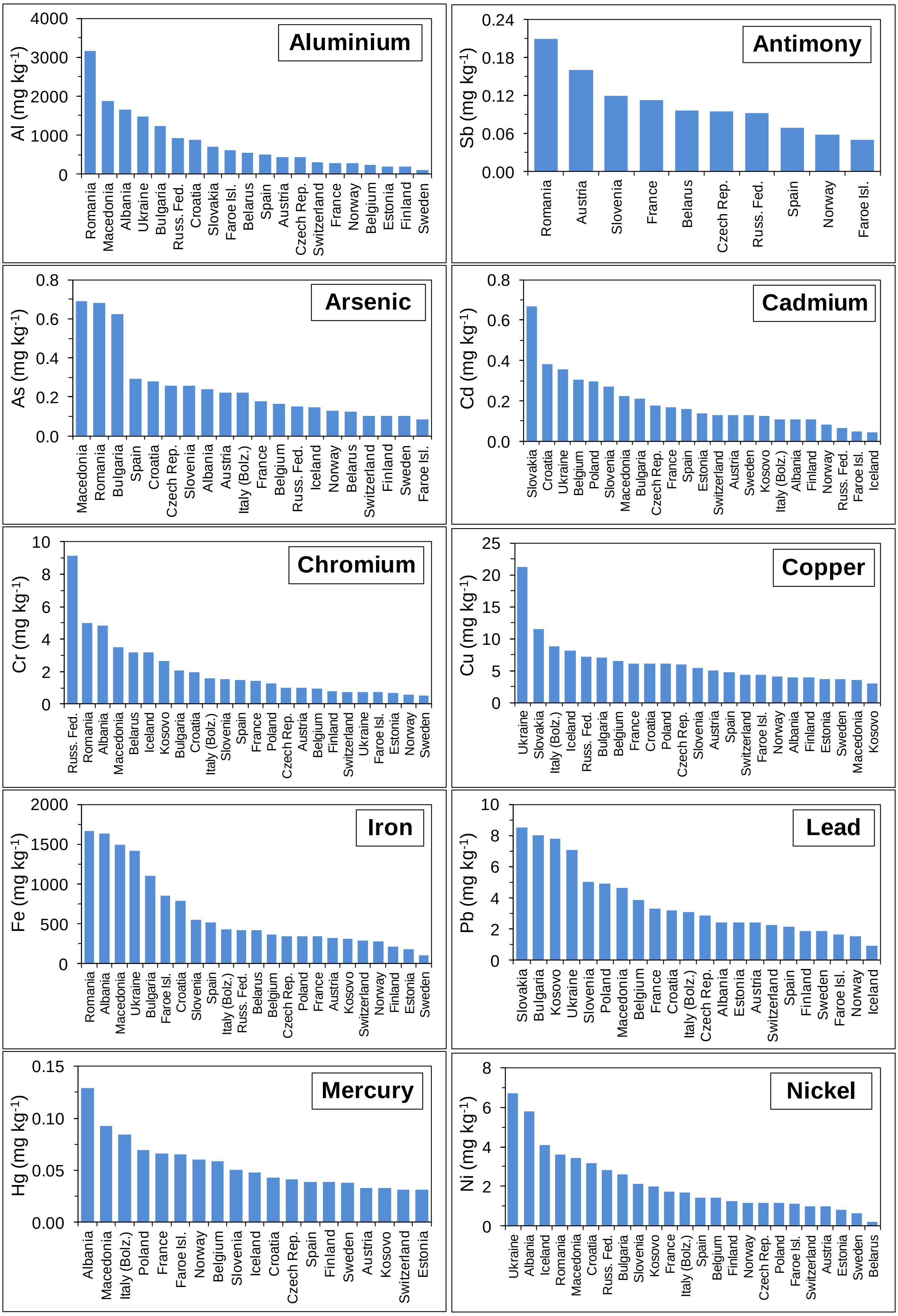


Figure 7. Mean nitrogen concentration in mosses per EMEP grid square (50 km x 50 km) in Europe in 2010 (left) and medium value per country (right). Italy: Bolzano region, Spain: Navarra region.



Aluminium
Al (mg kg-1)
4000
3000
2000
1000
0
Romania
Macedonia
Albania
Ukraine
Bulgaria
Russ. Fed.
Croatia
Slovakia
Faroe Isl.
Belarus
Spain
Austria
Czech Rep.
Switzerland
France
Norway
Belgium
Estonia
Finland
Sweden
Antimony
Sb (mg kg-1)
0.24
0.18
0.12
0.06
0.00
Romania
Austria
Slovenia
France
Belarus
Czech Rep.
Russ. Fed.
Spain
Norway
Faroe Isl.
Arsenic
As (mg kg-1)
0.8
0.6
0.4
0.2
0.0
Macedonia
Romania
Bulgaria
Spain
Croatia
Czech Rep.
Slovenia
Albania
Austria
Italy (Bolz.)
France
Belgium
Russ. Fed.
Iceland
Norway
Belarus
Switzerland
Finland
Sweden
Faroe Isl.
Cadmium
Cd (mg kg-1)
0.8
0.6
0.4
0.2
0.0
Slovakia
Croatia
Ukraine
Belgium
Poland
Slovenia
Macedonia
Bulgaria
Czech Rep.
France
Spain
Estonia
Switzerland
Austria
Sweden
Kosovo
Italy (Bolz.)
Albania
Finland
Norway
Russ. Fed.
Faroe Isl.
Iceland
Chromium
Cr (mg kg-1)
10
8
6
4
2
0
Russ. Fed.
Romania
Albania
Macedonia
Belarus
Iceland
Kosovo
Bulgaria
Croatia
Italy (Bolz.)
Slovenia
Spain
France
Poland
Czech Rep.
Austria
Belgium
Finland
Switzerland
Ukraine
Faroe Isl.
Estonia
Norway
Sweden
Copper
Cu (mg kg-1)
25
20
15
10
5
0
Ukraine
Slovakia
Italy (Bolz.)
Iceland
Russ. Fed.
Bulgaria
Belgium
France
Croatia
Poland
Czech Rep.
Slovenia
Austria
Spain
Switzerland
Faroe Isl.
Norway
Albania
Finland
Estonia
Sweden
Macedonia
Kosovo
Iron
Fe (mg kg-1)
2000
1500
1000
500
0
Romania
Albania
Macedonia
Ukraine
Bulgaria
Faroe Isl.
Croatia
Slovenia
Spain
Italy (Bolz.)
Russ. Fed.
Belarus
Belgium
Czech Rep.
Poland
France
Austria
Kosovo
Switzerland
Norway
Finland
Estonia
Sweden
Lead
Pb (mg kg-1)
10
8
6
4
2
0
Slovakia
Bulgaria
Kosovo
Ukraine
Slovenia
Poland
Macedonia
Belgium
France
Croatia
Italy (Bolz.)
Czech Rep.
Albania
Estonia
Austria
Switzerland
Spain
Finland
Sweden
Faroe Isl.
Norway
Iceland
Mercury
Hg (mg kg-1)
0.15
0.10
0.05
0.00
Albania
Macedonia
Italy (Bolz.)
Poland
France
Faroe Isl.
Norway
Belgium
Slovenia
Iceland
Croatia
Czech Rep.
Spain
Finland
Sweden
Austria
Kosovo
Switzerland
Estonia
Nickel
Ni (mg kg-1)
8
6
4
2
0
Ukraine
Albania
Iceland
Romania
Macedonia
Croatia
Russ. Fed.
Bulgaria
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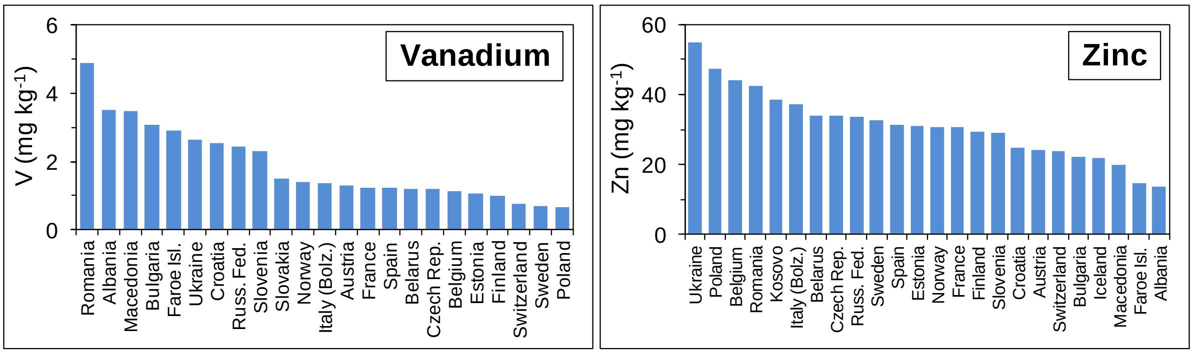

Figure S1. Medium heavy metal concentration in mosses per (region of) country in 2010.

Denmark: Faroe Islands; Italy: Bolzano region; Russian Federation: Ivanovo, Kostromskaya, Tikhvin-Leningradskaya region; Spain: Galicia, Navarra and/or Rioja region; Ukraine: Donetsk region.

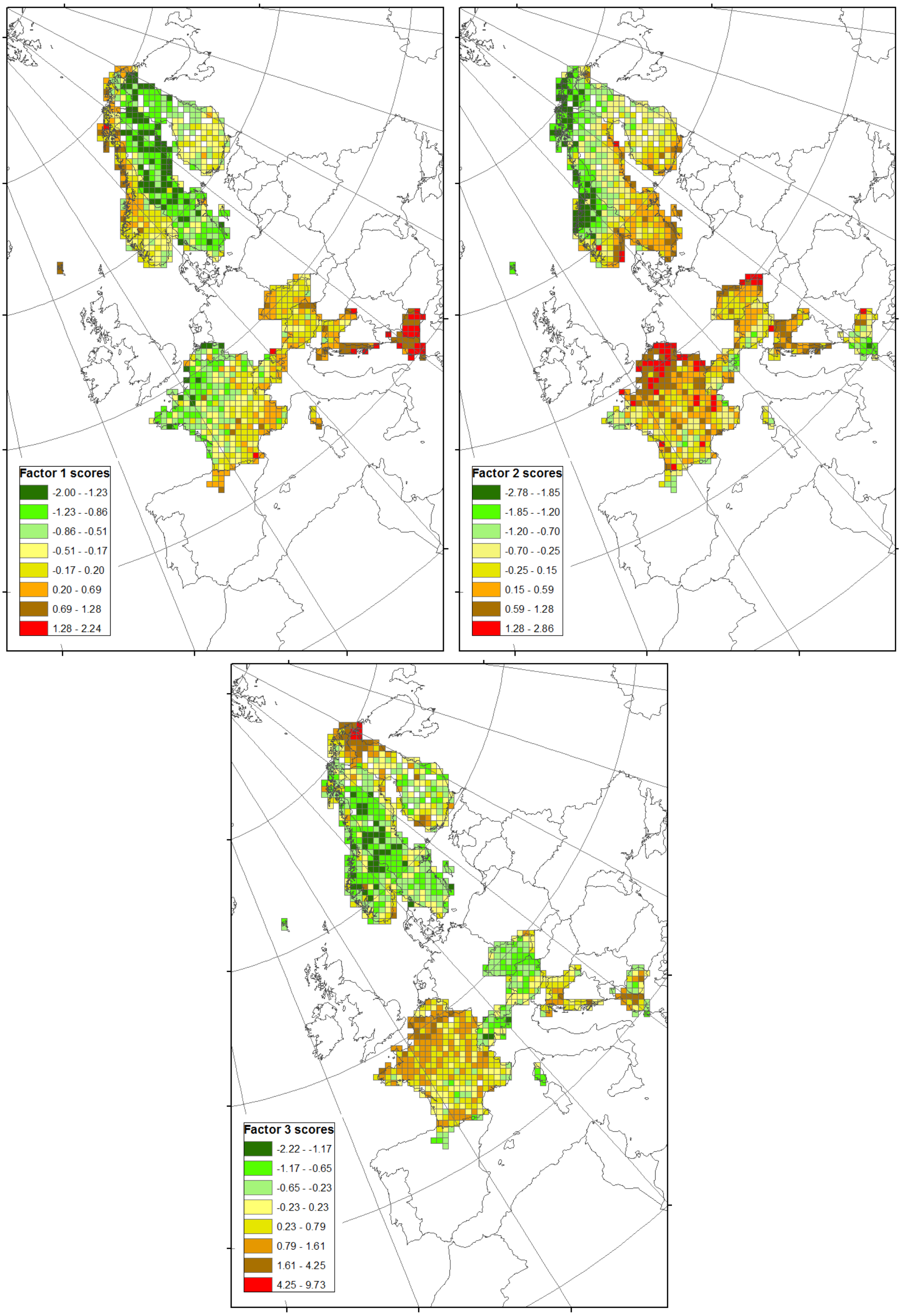

Figure S2. Spatial variation in the strength of association (factor scores) of the identified factors shown in Table 1. Factor 1: Al, As, Cr, Fe, Ni, V; factor 2: Cd, Pb, Zn, Factor 3: Cu, Ni. Note: Factors scores could only be determined for those sites that had data for all metals.

Table S1. Heavy metal (mg kg^{-1}) and nitrogen (mass %) concentrations in mosses in 2010.

	As	Cd	Cr	Cu	Fe	Hg	Ni	Pb	V	Zn	Al	Sb	N (%)
Albania													
Number	59	59	59	59	59	61	59	59	59	59	59		
Min	0.039	0.038	1.62	1.52	469	0.031	1.56	1.34	1.15	1.00	535		
Max	2.20	0.90	31.8	11.1	5488	2.23	131	19.7	16.9	68.1	6974		
Mean	0.42	0.17	6.35	4.31	1915	0.20	11.2	3.37	4.26	14.2	1975		
Median	0.24	0.11	4.83	3.96	1629	0.13	5.81	2.42	3.52	13.8	1650		
90th percentile	1.12	0.29	12.0	6.35	3287	0.31	21.8	4.83	6.63	22.8	3089		
Austria													
Number	221	221	221	221	221	221	221	221	221	221	221	221	221
Min	0.065	0.047	0.49	2.90	97.0	0.004	0.31	0.55	0.34	13.0	81.9	0.040	0.74
Max	1.78	1.10	6.50	29.0	2800	0.35	4.50	12.0	17.0	81.0	3024	1.60	2.20
Mean	0.32	0.16	1.28	5.56	411	0.037	1.18	2.64	1.55	25.6	444	0.19	1.26
Median	0.22	0.13	1.00	5.00	320	0.033	1.00	2.40	1.30	24.0	347	0.16	1.20
90th percentile	0.60	0.26	2.70	6.80	690	0.051	2.00	4.00	2.50	37.0	819	0.29	1.60
Belarus													
Number	76		76		76		76		76	76	76	76	
Min	0.006		0.66		194		0.086		0.59	21.5	267	0.026	
Max	0.34		5.23		1030		1.21		3.68	94.7	1650	0.34	
Mean	0.14		3.04		466		0.39		1.35	40.1	650	0.11	
Median	0.12		3.21		416		0.23		1.19	34.1	557	0.096	
90th percentile	0.20		4.64		788		0.85		2.20	66.7	1045	0.19	
Belgium													
Number	29	29	29	29	29	29	29	29	29	29	29		29
Min	0.052	0.092	0.54	3.27	171	0.020	0.72	2.12	0.41	16.6	114		0.79
Max	0.89	0.69	3.89	11.5	1109	0.32	22.7	12.5	2.76	132	696		2.30
Mean	0.19	0.33	1.08	6.80	377	0.068	2.14	5.14	1.19	52.4	275		1.38
Median	0.16	0.30	0.92	6.50	365	0.058	1.41	3.87	1.14	44.2	242		1.37
90th percentile	0.26	0.55	1.62	8.97	513	0.094	1.97	9.31	1.94	77.6	443		1.78
Bulgaria													
Number	60	129	129	129	129		129	129	129	129	129		99
Min	0.15	0.043	0.72	2.00	307		0.84	1.69	0.96	8.22	402		0.20
Max	10.8	7.75	38.1	270	8546		82.1	333	22.4	286	8886		2.94
Mean	1.08	0.39	3.46	12.2	1534		4.37	16.8	3.96	30.6	1493		1.38
Median	0.63	0.21	2.06	7.01	1101		2.61	8.00	3.07	22.2	1245		1.32
90th percentile	1.76	0.57	5.87	21.0	2824		6.44	21.9	7.52	45.4	2714		1.90
Croatia													
Number	121	121	121	121	121	121	121	121	121	121	121		119
Min	0.039	0.10	0.41	3.35	85	0.010	1.04	1.11	0.23	11.6	112		0.71
Max	0.77	1.42	8.55	16.1	4028	0.15	14.7	36.6	37.3	77.1	4493		2.93
Mean	0.30	0.43	2.25	6.55	881	0.043	3.70	3.79	3.50	27.1	1062		1.54
Median	0.28	0.38	1.94	6.06	789	0.043	3.16	3.21	2.55	24.8	878		1.49
90th percentile	0.54	0.74	3.91	9.32	1658	0.063	6.39	5.48	6.17	41.6	1995		2.35
Czech Republic													
Number	273	273	273	273	273	273	273	273	273	273	273	273	273
Min	0.068	0.092	0.46	3.26	150	0.019	0.37	1.17	0.44	20.1	184	0.001	0.70
Max	1.08	1.38	4.35	10.7	2072	0.11	4.47	42.1	6.10	105	3227	0.82	2.52
Mean	0.29	0.22	1.21	6.00	421	0.043	1.27	3.83	1.38	36.5	526	0.097	1.38
Median	0.26	0.18	1.01	5.92	348	0.041	1.15	2.85	1.18	33.9	435	0.096	1.33
90th percentile	0.46	0.34	2.10	7.84	692	0.058	2.01	5.77	2.23	47.3	797	0.15	1.86
Denmark (Faroe Islands)													
Number	7	7	7	7	7	7	7	7	7	7	7	7	
Min	0.071	0.034	0.56	3.48	511	0.054	0.90	1.30	2.23	12.7	461	0.039	
Max	0.12	0.080	0.84	4.63	1074	0.074	1.84	2.05	4.40	28.8	724	0.060	
Mean	0.086	0.057	0.72	4.09	842	0.065	1.23	1.72	3.01	17.2	617	0.048	
Median	0.084	0.049	0.71	4.27	853	0.064	1.12	1.66	2.91	14.8	612	0.050	
90th percentile	0.10	0.078	0.82	4.52	982	0.072	1.63	2.03	3.73	23.3	711	0.057	
Estonia													
Number		99	99	99	99	99	99	99	99	99	99		99
Min		0.080	0.36	0.92	93	0.022	0.43	1.29	1.05	19.3	79		0.65
Max		0.25	2.40	10.6	617	0.076	2.10	3.97	2.85	55.6	492		1.50
Mean		0.15	0.75	3.81	204	0.034	0.86	2.50	1.17	31.5	204		0.94
Median		0.14	0.68	3.67	180	0.031	0.82	2.41	1.07	30.9	188		0.88
90th percentile		0.21	1.04	4.81	317	0.047	1.15	3.33	1.48	39.7	294		1.28
Finland													
Number	201	426	426	426	426	202	426	426	426	426	426		426
Min	<0.10	<0.050	0.34	0.74	53	0.016	0.42	<0.75	<1.00	11.5	44		0.38
Max	0.38	0.44	14.0	55.1	2230	0.12	88.2	6.57	14.2	102	958		2.06
Mean	0.12	0.12	0.95	4.90	240	0.042	2.45	2.04	1.28	31.0	206		0.77
Median	0.10	0.11	0.80	3.91	209	0.039	1.24	1.87	1.00	29.5	187		0.70
90th percentile	0.17	0.18	1.38	7.45	411	0.067	3.43	3.17	1.79	43.2	318		1.11

Table S1 (continued).

	As	Cd	Cr	Cu	Fe	Hg	Ni	Pb	V	Zn	Al	Sb	N (%)
France													
Number	442	442	442	442	442	442	442	442	442	442	442	442	442
Min	<0.050	0.042	0.52	2.24	86	0.025	0.55	1.00	0.42	14.1	62	0.030	0.80
Max	2.46	1.21	5.91	16.2	3540	0.15	10.2	18.2	6.35	97.3	2020	0.58	2.71
Mean	0.26	0.20	1.55	6.36	429	0.069	2.00	4.03	1.45	34.3	357	0.14	1.47
Median	0.18	0.17	1.43	6.06	343	0.066	1.75	3.29	1.24	30.7	286	0.11	1.45
90th percentile	0.48	0.35	2.27	8.69	788	0.095	3.25	7.17	2.38	53.0	685	0.23	1.90
Iceland													
Number	144	144	144	144		144	144	144		144			
Min	0.045	0.012	0.68	3.06		0.025	0.83	0.27		8.4			
Max	1.38	0.29	18.1	47.4		0.16	40.7	72.1		194			
Mean	0.25	0.06	4.09	9.92		0.055	6.63	1.69		25.6			
Median	0.15	0.05	3.16	8.16		0.048	4.09	0.91		21.9			
90th percentile	0.50	0.12	8.36	16.7		0.088	14.4	2.03		36.2			
Italy (Bolzano region)													
Number	20	20	20	20	20	20	20	20	20	20			20
Min	0.14	0.060	0.78	5.72	232	0.054	0.89	1.76	0.91	17.7			0.68
Max	0.95	0.22	3.37	13.9	646	0.12	3.36	11.5	1.90	68.4			1.43
Mean	0.28	0.11	1.59	9.26	420	0.083	1.77	3.64	1.39	38.2			1.09
Median	0.22	0.11	1.59	8.88	431	0.084	1.69	3.11	1.37	37.1			1.09
90th percentile	0.38	0.14	2.17	12.0	546	0.11	2.27	4.57	1.63	54.7			1.34
Kosovo													
Number		25	25	25	24	25	24	25		25			
Min		0.028	1.63	2.46	124	0.009	1.22	2.62		14.3			
Max		3.05	4.55	3.93	3082	0.35	34.2	47.8		76.0			
Mean		0.37	2.72	3.12	582	0.055	6.08	12.13		37.8			
Median		0.13	2.63	3.04	312	0.033	2.00	7.78		38.5			
90th percentile		0.83	3.52	3.54	1085	0.091	24.4	20.7		55.2			
Macedonia													
Number	52	72	72	72	72	72	72	72	72	72	72		68
Min	0.077	0.068	1.03	1.97	513	0.010	1.25	1.87	1.00	1.00	537		0.68
Max	3.30	2.24	39.7	10.6	6348	0.60	51.7	22.0	17.4	365	8679		1.75
Mean	0.88	0.29	4.68	4.02	1732	0.11	6.43	5.40	3.95	29.7	2176		1.08
Median	0.69	0.22	3.48	3.54	1490	0.093	3.45	4.61	3.49	19.9	1878		1.06
90th percentile	2.01	0.44	7.38	6.21	2941	0.16	10.5	8.37	6.20	48.1	3373		1.29
Norway													
Number	463	463	463	463	463	463	463	463	463	463	463	463	
Min	0.020	0.009	0.16	1.38	27	<0.024	0.15	0.33	0.29	7.4	46	<0.001	
Max	4.84	1.87	47.9	443	24684	0.34	857	20.8	25.9	368	4581	1.17	
Mean	0.18	0.12	0.98	6.43	449	0.070	5.40	2.29	1.76	35.9	346	0.092	
Median	0.13	0.081	0.59	4.04	278	0.060	1.16	1.54	1.41	30.7	283	0.058	
90th percentile	0.26	0.23	1.55	7.29	685	0.11	2.79	4.85	3.03	57.5	565	0.20	
Poland													
Number		320	320	320	320	320	320	320	308	320			320
Min		0.003	0.20	1.46	110	0.029	0.14	1.54	0.11	7.46			0.78
Max		14.3	293	133	2618	0.76	108	141	4.69	211			2.86
Mean		0.45	3.58	6.94	405	0.072	2.20	6.73	0.77	51.8			1.56
Median		0.30	1.27	6.04	344	0.069	1.15	4.93	0.65	47.5			1.52
90th percentile		0.71	4.25	9.64	663	0.097	3.47	10.6	1.36	86.1			1.97
Romania													
Number	333		331		332		253		333	332	333	332	
Min	0.10		0.68		237		0.39		0.39	0.6	220	0.013	
Max	51.1		62.2		29500		35.9		58.3	1440	34400	16.5	
Mean	1.48		8.18		3000		4.99		7.56	56.0	4861	0.45	
Median	0.68		4.98		1670		3.60		4.89	42.3	3150	0.21	
90th percentile	2.76		18.9		6610		8.80		17.7	85.4	11620	0.63	
Russian Federation (Ivanovo, Kostromskaya, Tikhvin-Leningradskaya)													
Number	66	30	65	21	90		66		90	91	90	66	
Min	0.067	0.004	0.73	1.02	50		1.17		0.81	2.40	288	0.028	
Max	9.32	0.96	242	43.9	13600		11.3		23.40	172	13300	0.32	
Mean	0.46	0.17	22.4	10.2	1049		4.08		4.15	39.2	1826	0.12	
Median	0.15	0.068	9.16	7.22	419		2.82		2.45	33.6	922	0.092	
90th percentile	0.92	0.42	41.5	24.0	2470		9.48		11.1	59.5	3496	0.22	
Slovakia													
Number		67		67				67	67		67		67
Min		0.078		6.44				2.31	0.60		251		1.00
Max		3.39		90.4				58.4	10.2		5580		2.85
Mean		0.77		14.5				10.9	2.04		1043		1.84
Median		0.67		11.5				8.51	1.50		707		1.79
90th percentile		1.24		19.8				18.9	3.39		1926		2.39
Slovenia													
Number	102	102	102	102	102	63	102	102	102	102		102	102
Min	0.13	0.090	0.72	2.83	243	0.030	0.85	1.96	1.00	14.7		0.060	0.85
Max	0.83	1.05	13.7	11.4	1391	0.16	8.16	304	7.00	66.7		0.76	1.99
Mean	0.28	0.33	1.94	5.71	617	0.056	2.34	8.79	2.40	31.5		0.13	1.32
Median	0.26	0.27	1.56	5.42	548	0.050	2.12	5.01	2.30	29.0		0.12	1.29
90th percentile	0.41	0.53	2.69	7.29	934	0.070	3.34	8.63	3.57	48.7		0.18	1.70

Table S1 (continued).

	As	Cd	Cr	Cu	Fe	Hg	Ni	Pb	V	Zn	Al	Sb	N (%)
Spain (Galicia, Navarra, Rioja)[1]													
Number	211	211	39	39	39	211	64	64	39	186	39	39	64
Min	0.086	0.031	0.43	2.64	171	0.022	0.58	0.95	0.55	12.7	173	0.040	0.64
Max	2.69	1.57	4.77	9.81	1449	0.081	3.94	10.3	4.20	156	1459	0.15	1.80
Mean	0.39	0.22	1.83	4.83	610	0.040	1.60	2.75	1.36	32.9	597	0.072	1.09
Median	0.29	0.16	1.46	4.70	520	0.039	1.44	2.13	1.21	31.5	511	0.069	1.05
90th percentile	0.73	0.37	3.38	6.25	1089	0.053	2.88	4.86	2.08	44.7	1043	0.10	1.48
Sweden													
Number	602	602	602	602	602	602	602	602	602	602	602		
Min	0.080	0.023	0.11	1.38	28	0.016	0.22	0.43	0.16	13.4	25		
Max	0.45	0.39	10.8	23.5	2406	0.14	7.11	19.9	9.63	81.9	1303		
Mean	0.10	0.13	0.67	3.92	135	0.041	0.72	2.09	0.79	33.9	143		
Median	0.10	0.13	0.52	3.61	101	0.038	0.66	1.87	0.69	32.6	110		
90th percentile	0.10	0.21	1.14	5.57	218	0.062	1.02	3.38	1.26	46.8	254		
Switzerland													
Number	142	142	142	142	156	142	142	142	142	142	142		64
Min	0.027	0.034	0.21	2.61	101	0.018	0.17	0.71	0.22	11.1	81		0.64
Max	5.81	3.57	5.25	10.0	1732	0.076	5.93	12.7	4.27	170	2256		1.88
Mean	0.19	0.18	0.92	4.89	351	0.034	1.30	2.60	0.88	27.8	357		1.05
Median	0.10	0.13	0.75	4.37	286	0.031	1.00	2.24	0.74	23.7	295		1.00
90th percentile	0.27	0.26	1.55	6.68	601	0.047	2.50	4.15	1.44	40.7	626		1.38
Ukraine (Donetsk)													
Number		16	16	17	17		17	17	17	17	17		
Min		0.080	0.25	12.8	1003		4.70	5.56	0.98	37.6	821		
Max		0.52	2.40	62.1	7307		26.1	20.9	3.99	152	4664		
Mean		0.33	0.92	22.9	2437		8.94	8.74	2.51	63.7	1822		
Median		0.36	0.73	21.2	1414		6.70	7.07	2.63	54.9	1476		
90th percentile		0.40	1.59	33.1	6708		15.4	14.4	3.80	92.4	3177		

[1] As, Cd, Hg: all regions; Zn: Galicia and Navarra; Ni, Pb, N: Navarra and Rioja; Al, Cr, Cu, Fe, Sb, V: Navarra.
Number = number of sampling sites; Min = minimum; Max = maximum.