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Heavy metals and nitrogen in mosses: spatial patterns in 2010/11 and long-term temporal trends (1990-2010) in Europe

Report by the International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops

Summary

The present report, prepared by the International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops, was drafted in response to the request of the Executive Body for the Convention on Long-range Transboundary Air Pollution, as set out in the 2012–2013 workplan for the Implementation of the Convention (ECE/EB.AIR/109/Add.2, item 3.5, ongoing activities, para. (c)).

It provides an analysis of heavy metal and nitrogen concentrations in mosses in 2010/11 and long-term temporal trends for heavy metals since 1990 and for nitrogen since 2005 in Europe. The moss data provide field-based evidence of the deposition of heavy metal and nitrogen pollution to vegetation.

Contents

		Paragraphs	Page
I.	Introduction	1–2	3
II	Spatial patterns of cadmium, lead and mercury concentrations in mosses in 2010 and changes since 2005	3–6	4
III.	Spatial patterns of other heavy metal concentrations in mosses in 2010 and changes since 2005	7–12	7
IV.	Temporal trends (1990–2010) of heavy metal concentrations in mosses agree well with trends in atmospheric deposition modelled by the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe	13–15	11
V.	Spatial patterns and temporal trends (2005–2010) of nitrogen concentrations in mosses	16–17	14
VI.	Conclusions	18	15
VII.	Recommendations	19–20	16

Figures

		Page
1.	Mean cadmium, lead and mercury concentration in mosses per EMEP[1] grid cell in 2010	6
2.	Mean arsenic, chromium and copper concentration in mosses per EMEP grid cell in 2010	8
3.	Mean nickel, vanadium and zinc concentration in mosses per EMEP grid cell in 2010	10
4.	Temporal trend of cadmium, lead and mercury concentration in mosses compared with the trend of EMEP-modelled deposition for these heavy metals	12
5.	Temporal trend of arsenic, chromium, copper, nickel, vanadium and zinc concentration in mosses	13
6.	Mean total nitrogen concentration in mosses per EMEP grid cell in 2005 and 2010	14
7.	Relationship between nitrogen concentration in mosses and EMEP-modelled total nitrogen deposition in 2005 for EMEP grid cells where at least five moss sampling sites were present and measured bulk nitrogen deposition rates in Switzerland in 2005 and 2010	15

[1] Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe.

I. Introduction

1. Carpet-forming mosses do not have a root system and therefore obtain most trace elements (e.g., heavy metals) and nutrients (e.g., nitrogen) directly from atmospheric (wet and dry) deposition. The analysis of elemental concentrations in mosses provides a time-integrated measure of atmospheric deposition to terrestrial systems. It is easier and cheaper than conventional precipitation analysis as it avoids the need for deploying large numbers of deposition collectors with an associated long-term programme of routine sample collection and analysis. Hence, the moss survey provides a complementary method to assess spatial patterns and temporal trends of atmospheric deposition of heavy metals and nitrogen deposition to vegetation and to identify areas at risk from air pollution.

2. The first European-wide moss survey was conducted in 1990 and since then the survey has been repeated every five years. The most recent European survey was conducted in 2010/11(described as 2010 from here onwards), when mosses were collected at over 4,500 sites in 25 countries for heavy metal analysis and at some 2,400 sites in 15 countries for nitrogen analysis (table 1). In addition, six countries determined the concentration of selected persistent organic pollutants, particularly polycyclic aromatic hydrocarbons, at a selected number of sites (see ECE/EB.AIR/WG.1/2013/8).

Table 1
List of countries that submitted data for the 2010 European moss survey[2]

Albania	**Finland**	**Slovakia**
Austria	**France**	**Slovenia**
Belarus	Iceland	**Spain**[d]
Belgium	**Italy**[b]	Sweden
Bulgaria	**Macedonia**	**Switzerland**
Croatia	Norway	Ukraine[e]
Czech Republic	**Poland**	
Denmark[a]	Romania	
Estonia	Russian Federation[c]	

Note: Countries in bold submitted data on heavy metals and nitrogen, the other countries submitted only data on metals. Countries submitting data for only particular regions or areas are indicated in the table notes.

[a] Faroe Islands.
[b] Bolzano region.
[c] Ivanova, Kostromskaya and Tikhvin-Leningradskaya regions.
[d] Galicia, Navarra and Rioja regions.
[e] Donestsk region.

[2] Although not a Party to the Convention on Long-range Transboundary Air Pollution, Kosovo (United Nations administered region, Security Council resolution 1244 (1999)) submitted its heavy metal data for the 2010 European moss survey.

II. Spatial patterns of cadmium, lead and mercury concentrations in mosses in 2010 and changes since 2005

3. As in previous surveys, the lowest concentrations of heavy metals in mosses were generally found in Northern Europe, although higher concentrations were reported for some metals near local sources. Low to intermediate heavy metal concentrations in mosses were generally observed in Western and Central Europe. The highest concentrations were often found in South-Eastern Europe, with localized lower concentrations being observed. The spatial pattern of mercury concentrations in mosses was more homogeneous across Europe due to its greater hemispheric long-range transport.

4. *Cadmium*: Cadmium concentrations in mosses were generally low in Northern Europe (figure 1). The cadmium levels were lowest in north-west Scandinavia, Iceland and the western parts of France. However, in France the median value has increased since 2005. This is likely due to the higher uncertainty and potential underestimation of the cadmium concentrations in mosses in 2005, particularly in areas with low cadmium concentrations in mosses in 2005. Relatively low median values were also observed in Albania, Kosovo[3] and the Russian Federation. Very high levels of cadmium were observed in Romania, followed by Slovakia, Croatia, Ukraine and Belgium. However, in Belgium the median value has declined by 38 per cent since 2005. Whereas a decline has also been found in other countries, several countries reported an increase of the median value since 2005. The average median cadmium concentration in mosses has declined by only 7 per cent — from 0.21 milligrams (mg) per kilogram (kg^{-1}) in 2005 to 0.20 mg kg^{-1} in 2010.

5. *Lead*: Very high lead concentrations in mosses were still reported in Romania (figure 1), where the use of leaded petrol has been banned completely only since January 2012. In addition, the presence of large industrial areas (including metallurgical works or melting plants) located in Baia Mare, Magoaja, Letca, Cergau, Zagra and Copsa Mica, contribute to the high concentration of lead in mosses. Relatively high lead concentrations were also found in Bulgaria, Kosovo,[3] Slovakia, Slovenia and Ukraine, although the median lead concentration in mosses has declined between 31 per cent and 50 per cent in Bulgaria, Slovakia and Slovenia since 2005. Relatively high lead concentrations were also found in parts of southern Poland. The huge decline of lead in mosses in Belgium since 2005 (decrease of the median value by 74 per cent) suggests that the implementation of clean air policies for heavy metals are finally starting to pay off in Belgium in line with neighbouring European countries. A slight increase in the median lead concentration in mosses since 2005 was only reported for Croatia, which might be due to the relatively low median value reported for 2005 (compared to neighbouring countries). The average median lead concentration in mosses had decreased by 36 per cent, from 5.62 mg kg^{-1} in 2005 to 3.57 mg kg^{-1} in 2010.

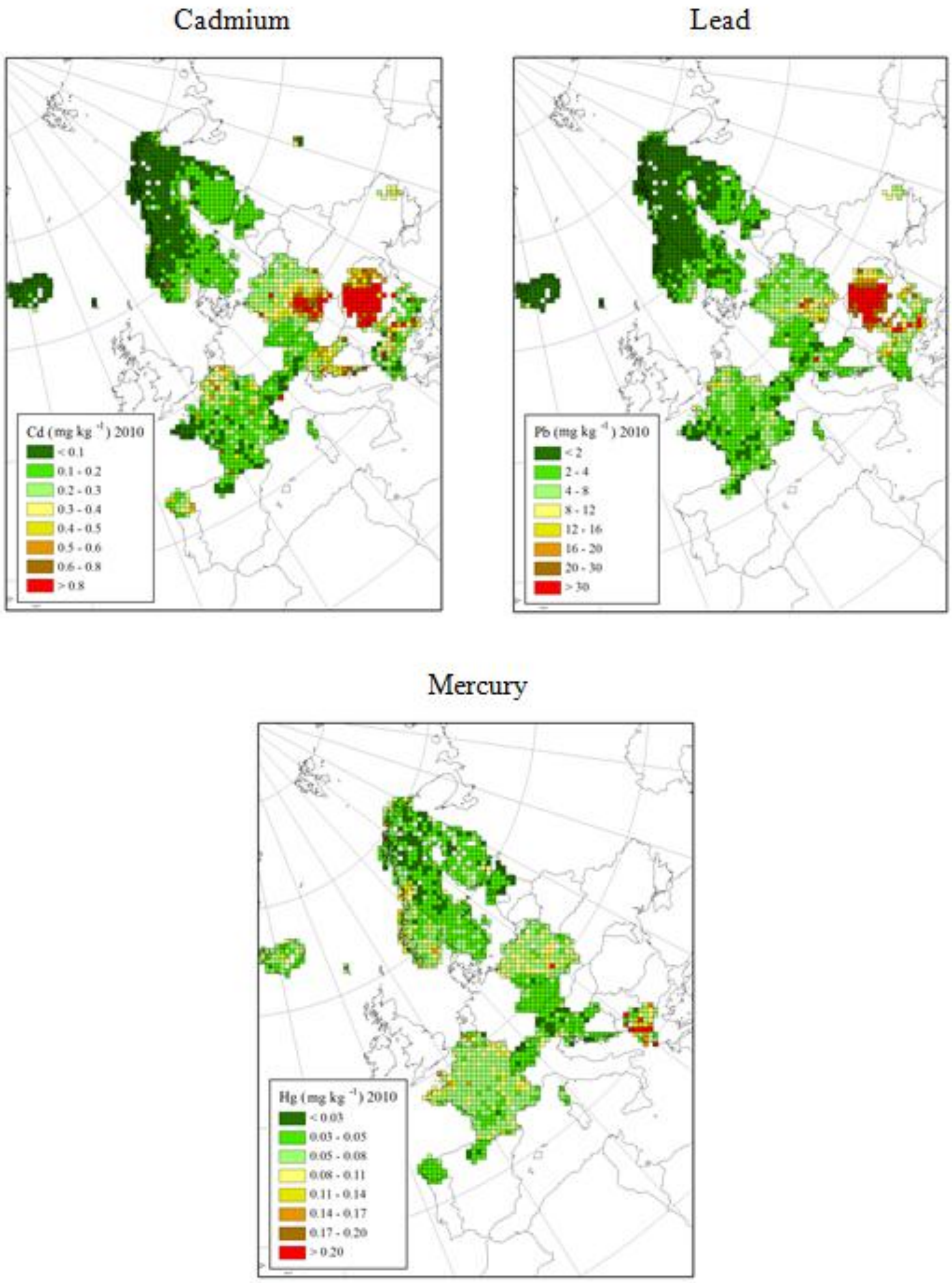
6. *Mercury*: In contrast to other metals, mercury is a global pollutant and can be transported in the atmosphere around the globe. Therefore, emission sources located on other continents have a significant impact on mercury pollution in Europe. Due to the long residence time of gaseous mercury in the atmosphere, most of it will be transported outside Europe. The global nature of mercury pollution appears to result in a more homogenous spatial pattern of mercury concentration in mosses in Europe compared to many other metals. The highest levels of mercury in mosses were found in Albania and the former Yugoslav Republic of Macedonia, followed by Italy, Poland and France (figure 1). In

[3] Kosovo (United Nations administered region, Security Council resolution 1244 (1999)) is neither a Party to the Convention on Long-range Transboundary Air Pollution nor a member State of ECE.

France, particularly areas with relatively high mercury concentrations in 2005 showed a considerable decline in the concentrations in mosses in 2010. Relatively high levels of mercury were also reported for Norway, and the levels have increased since 2005 in many parts of Norway. Arctic mercury depletion events (episodes in polar areas where gaseous elemental mercury is transformed to oxidized species) might be contributing to the elevated mercury concentrations in mosses in northern Norway. Whereas in many areas in southern Finland the mercury concentration in mosses has increased since 2005, the opposite was true for many areas in northern Finland. As with many other metals, since 2005 a considerable decline in mercury concentrations in mosses was reported for Belgium (decrease of the median value by 59 per cent). The average median mercury concentration in mosses has declined by 20 per cent — from 0.066 mg kg^{-1} in 2005 to 0.053 mg kg^{-1} in 2010.

Figure 1

Mean cadmium, lead and mercury concentration in mosses per EMEP[4] grid cell (50 km x 50 km) in 2010



[4] Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe.

III. Spatial patterns of other heavy metal concentrations in mosses in 2010 and changes since 2005

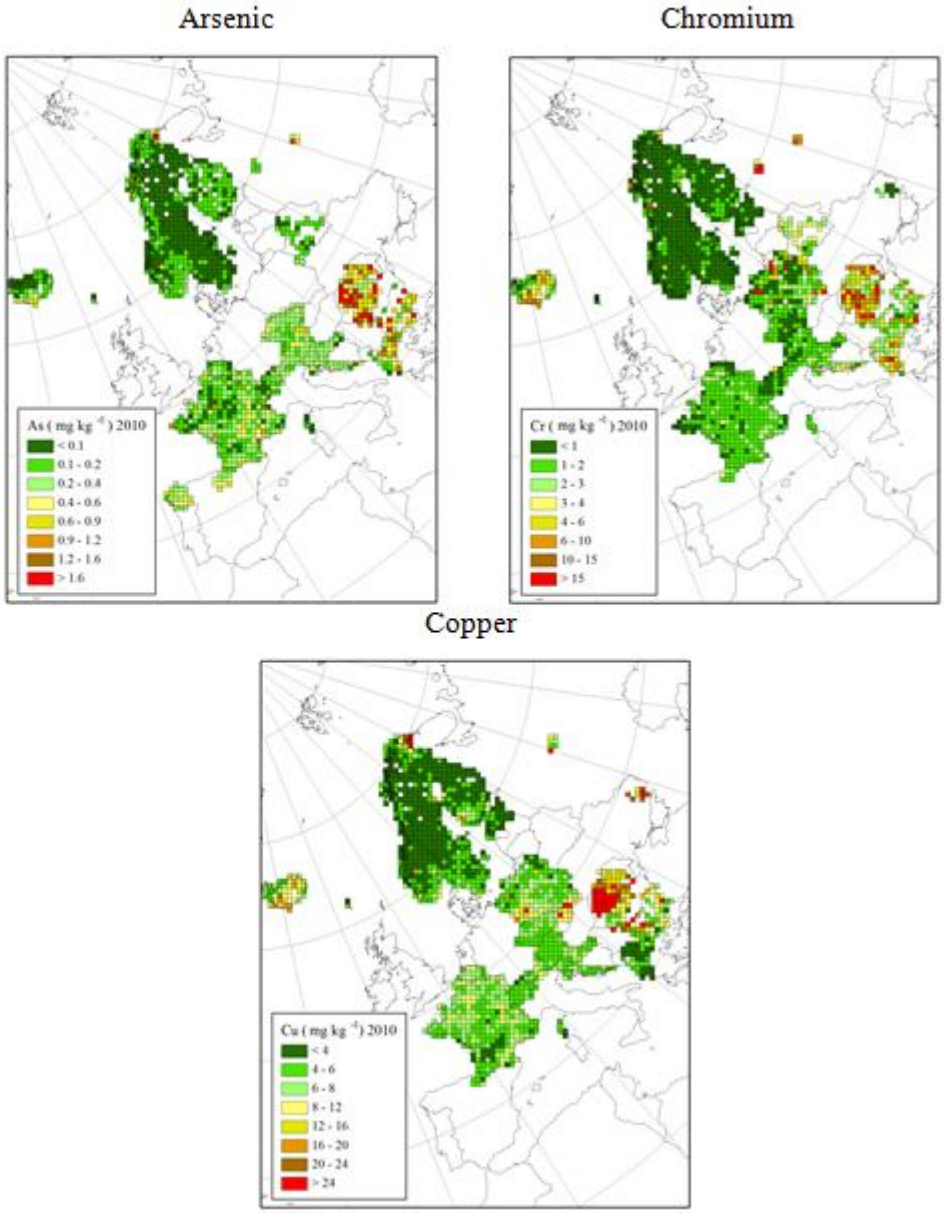
7. *Arsenic*: Arsenic concentrations in mosses were generally low in Northern Europe (figure 2), but have changed in these areas since 2005, with both increases and decreases being observed. High levels of arsenic were observed in South-Eastern Europe (Bulgaria, Romania and the former Yugoslav Republic of Macedonia). Since 2005, considerable reductions in the median arsenic concentration were observed in Belgium (76 per cent), Italy (Bolzano region, 52 per cent), France (51 per cent) and Slovenia (41 per cent). The median arsenic concentration has increased in Spain since 2005, primarily due to an increase of the concentration in mosses sampled in Galicia (as a decline was observed in Navarra). In Spain, the highest arsenic values were found in mosses in Rioja. The average median arsenic concentration in mosses has declined by 25 per cent — from 0.27 mg kg^{-1} in 2005 to 0.21 mg kg^{-1} in 2010.

8. *Chromium*: Chromium concentrations in mosses show a clear east-west gradient, with the highest values being observed in Eastern Europe, e.g., Albania, Belarus, Romania, the Russian Federation and the former Yugoslav Republic of Macedonia (figure 2). The high values in Belarus, Romania and the Russian Federation might be explained partly by using neutron activation analysis, a technique that generally results in higher chromium concentrations as other techniques require acid digestion of the samples first, and this digestion is generally not complete for chromium. However, this cannot explain the increase in the median values of chromium in Belarus and the Russian Federation since 2005. In general, the determination of chromium concentrations in mosses is associated with considerable uncertainty. A considerable decline of the median value of chromium between 2005 and 2010 was reported for Belgium (79 per cent), Spain (77 per cent), Ukraine (61 per cent), Italy (Bolzano region, 53 per cent) and the former Yugoslav Republic of Macedonia (49 per cent). Changes in the Russian Federation since 2005 are confounded by the fact that mosses were sampled from different regions in 2005 and 2010. The decline in the former Yugoslav Republic of Macedonia might be partly due to using neutron activation analysis in 2005 but not in 2010 as the analytical technique. The average median chromium concentration in mosses has declined by 23 per cent — from 2.37 mg kg^{-1} in 2005 to 1.82 mg kg^{-1} in 2010.

9. *Copper*: The highest copper concentrations in mosses were found in parts of Eastern Europe, i.e., Bulgaria, Slovakia, Romania and Ukraine (figure 2). However, low copper concentrations were found in Albania, Kosovo and the former Yugoslav Republic of Macedonia. As in previous years, the lowest copper concentrations were found in Northern Europe, although locally high concentrations were detected at the Norwegian-Finnish-Russian border in the north due to the presence of very polluting copper-nickel smelters in the Kola Peninsula on the Russian side of the border. In several countries the copper concentration in mosses has declined between 2005 and 2010, particularly in Belgium, Bulgaria and the former Yugoslav Republic of Macedonia (decline by more than one third). The high increase in the Ukraine since 2005 can to some extent be explained by sampling being limited to the Donetsk region in 2010, which is quite an industrialized region. The average median copper concentration in mosses has declined by only 6 per cent — from 6.96 mg kg^{-1} in 2005 to 6.53 mg kg^{-1} in 2010.

Figure 2

Mean arsenic, chromium and copper concentration in mosses per EMEP[5] grid cell (50 km x 50 km) in 2010



10. *Nickel*: In general, the highest nickel concentration were found in South-Eastern European countries (Albania, Bulgaria, Croatia, Romania, Russian Federation, the former Yugoslav Republic of Macedonia and Ukraine), although high nickel concentrations were also observed in Iceland (figure 3). As for copper, locally high concentrations of nickel were detected at the Norwegian-Finnish-Russian border in the north due to the presence of very polluting copper-nickel smelters in the Kola Peninsula on the Russian side of the

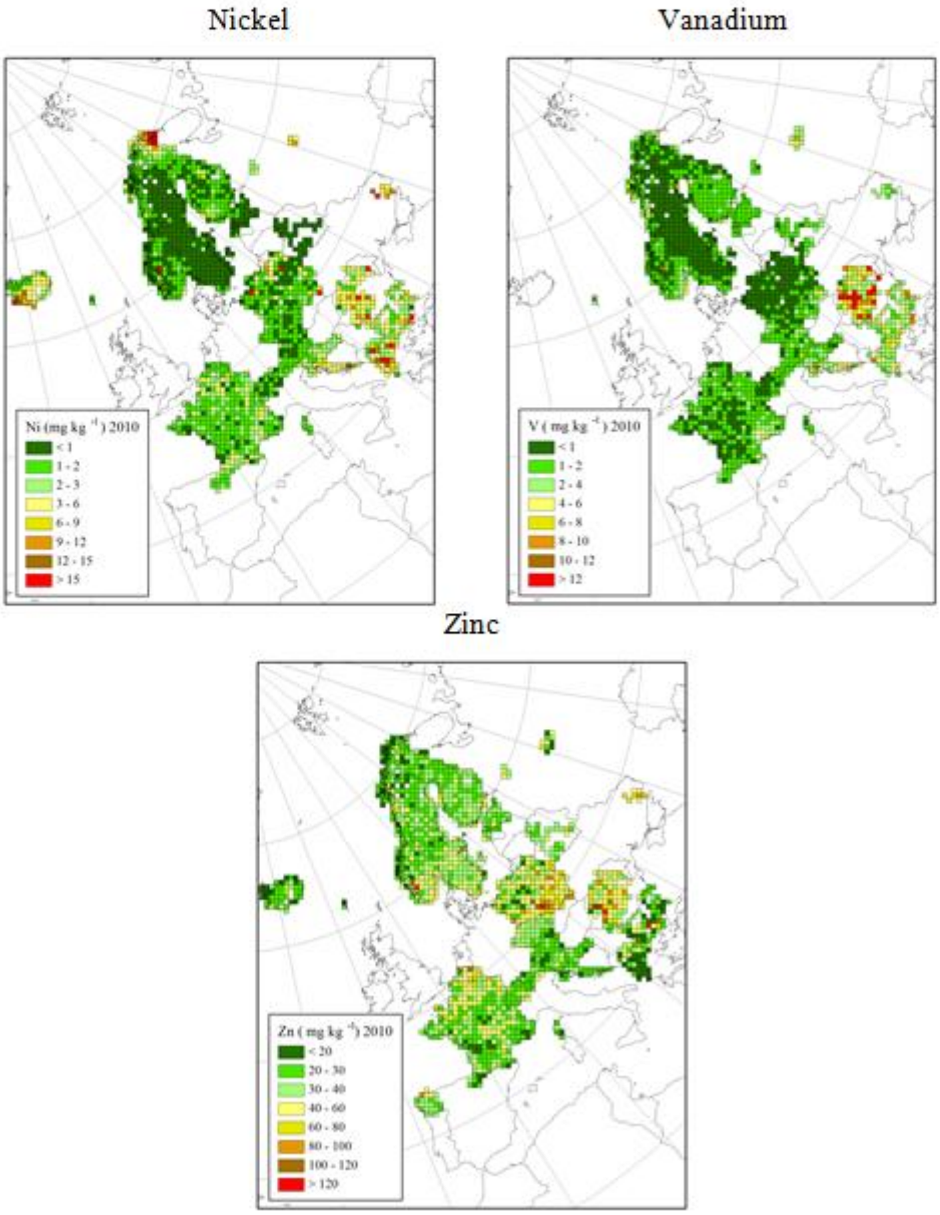
[5] Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe.

border. The spatial pattern for nickel is similar to that reported in 2005, although nickel concentrations in mosses have declined considerably in Belgium (decline of median value by 64 per cent), northern Spain (61 per cent) and eastern parts of France. Remarkable also is the homogeneously distributed low concentrations of nickel in Belarus in comparison with 2005. In northern Italy (Bolzano region) and the former Yugoslav Republic of Macedonia the median value for nickel has declined by 41–42 per cent since 2005. For Ukraine, the increase in the median value of nickel concentrations in mosses since 2005 is most likely due to sampling mosses in a more polluted region (Donetsk) in 2010. The average median nickel concentration in mosses has declined by 12 per cent — from 2.21 mg kg^{-1} in 2005 to 1.94 mg kg^{-1} in 2010.

11. *Vanadium*: In 2010, the highest vanadium concentrations were reported for mosses in South-Eastern Europe, i.e., Albania, Bulgaria, Romania and the former Yugoslav Republic of Macedonia (figure 3). Relatively high median concentrations were also observed on the Faroe Islands, similarly to 2005. Low vanadium concentrations were generally found in the rest of Europe. As for many other metals, the decline in the median concentration of vanadium in mosses between 2005 and 2010 is remarkable for Belgium (75 per cent). Since 2005, a decline in the median value of more than 45 per cent was observed in France, Italy (Bolzano region), Slovakia and the former Yugoslav Republic of Macedonia. Increases in the mean values per Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) grid since 2005 were observed in parts of northern Finland, the north-west coast of Norway and in many areas of Austria. The average median vanadium concentration in mosses has declined by 27 per cent — from 2.36 mg kg^{-1} in 2005 to 1.72 mg kg^{-1} in 2010.

12. *Zinc*: Of all metals, the zinc concentration in mosses has the most homogenous distribution across Europe, with locally or regionally elevated concentrations being observed (figure 3). In 2010, the highest median values were found in Belgium, Kosovo, Ukraine, Poland and Romania, whereas the lowest median values were reported for Albania, Bulgaria, the Faroe Islands, Iceland and the former Yugoslav Republic of Macedonia. As for many other metals, the decline in the median concentration of zinc in mosses between 2005 and 2010 is remarkable for Belgium (43 per cent) and the former Yugoslav Republic of Macedonia (44 per cent). The average median zinc concentration in mosses has declined by only 7 per cent — from 33.4 mg kg^{-1} in 2005 to 31.0 mg kg^{-1} in 2010.

Figure 3
Mean nickel, vanadium and zinc concentration in mosses per EMEP grid cell (50 km x 50 km) in 2010



IV. Temporal trends (1990–2010) of heavy metal concentrations in mosses agree well with trends in atmospheric deposition modelled by the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe

13. The decline in emission and subsequent deposition of heavy metals across Europe in recent decades has resulted in a decrease in the heavy metal concentration in mosses since 1990, with the decline continuing since the previous moss survey in 2005 (table 2). In general, the decline in metal concentrations in mosses was higher between 1990 and 1995 (or 2000) than in later years. The metal concentration in mosses has declined the most for lead, due to the abolishment of leaded petrol, and the least for copper.

Table 2

Decline in the average median heavy metal and nitrogen concentrations in mosses since the start of the European moss survey in 1990[6] and since the previous survey in 2005[7]

Element	*Decline since 1990 (%)*	*Decline since 2005 (%)*
Antimony	n.a.	23
Arsenic	26[a]	25
Cadmium	51	7
Chromium	43	23
Copper	11	6
Iron	52	15
Lead	77	36
Mercury	23[a]	20
Nickel	33	12
Nitrogen	n.a.	5
Vanadium	57	27
Zinc	34	7

Abbreviation: n.a. = not available.

[a] Decline since 1995 for arsenic and mercury as only a few countries have reported concentrations in mosses for these metals in 1990.

14. For cadmium, lead and mercury, the temporal trends in concentrations in mosses are in good agreement with trends reported for atmospheric deposition modelled by EMEP. Between 1990 and 2010, the average cadmium and lead concentration in mosses has declined by 51 per cent and 77 per cent respectively, whereas the average modelled cadmium and lead deposition in the EMEP domain has declined by 51 per cent and 74 per cent respectively (figure 4). Between 1995 and 2010, the average mercury concentration in

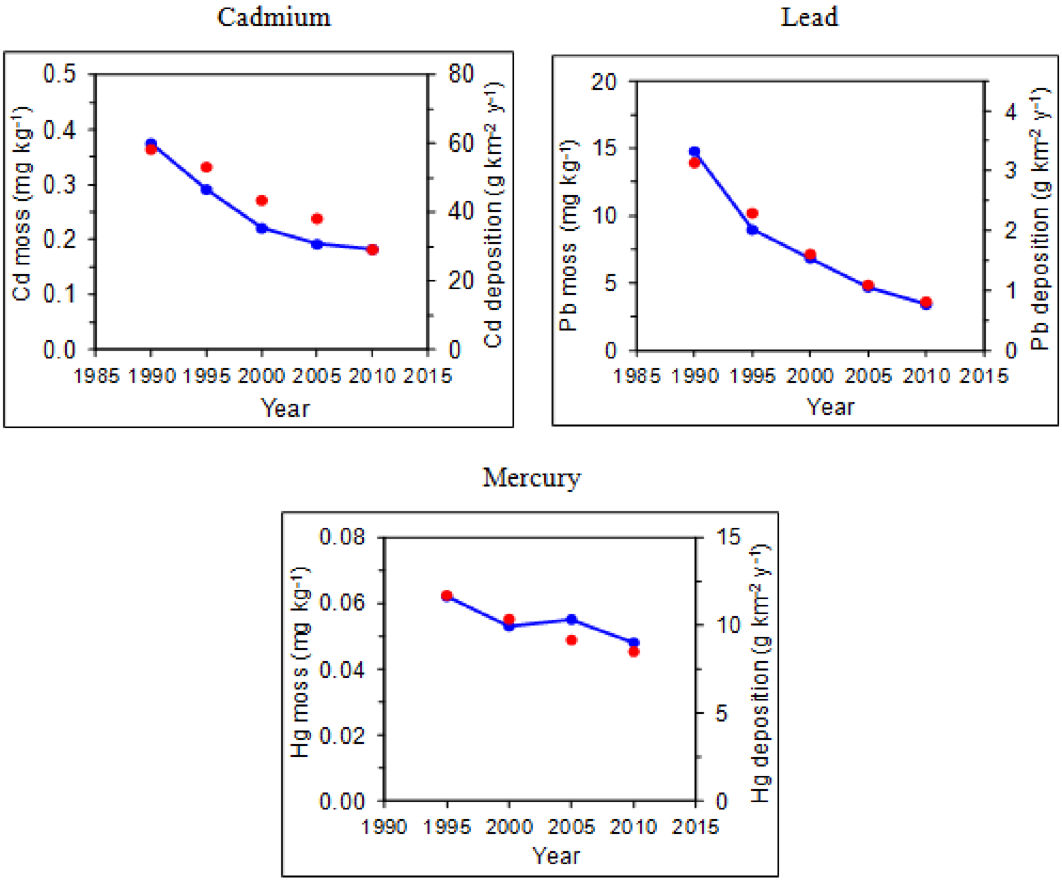
[6] Based on data for countries that reported data for at least four out of the five survey years.

[7] Based on data for countries that reported data for both 2005 and 2010.

mosses has declined by 23 per cent, whereas the average modelled mercury deposition in the EMEP domain has declined by 27 per cent.

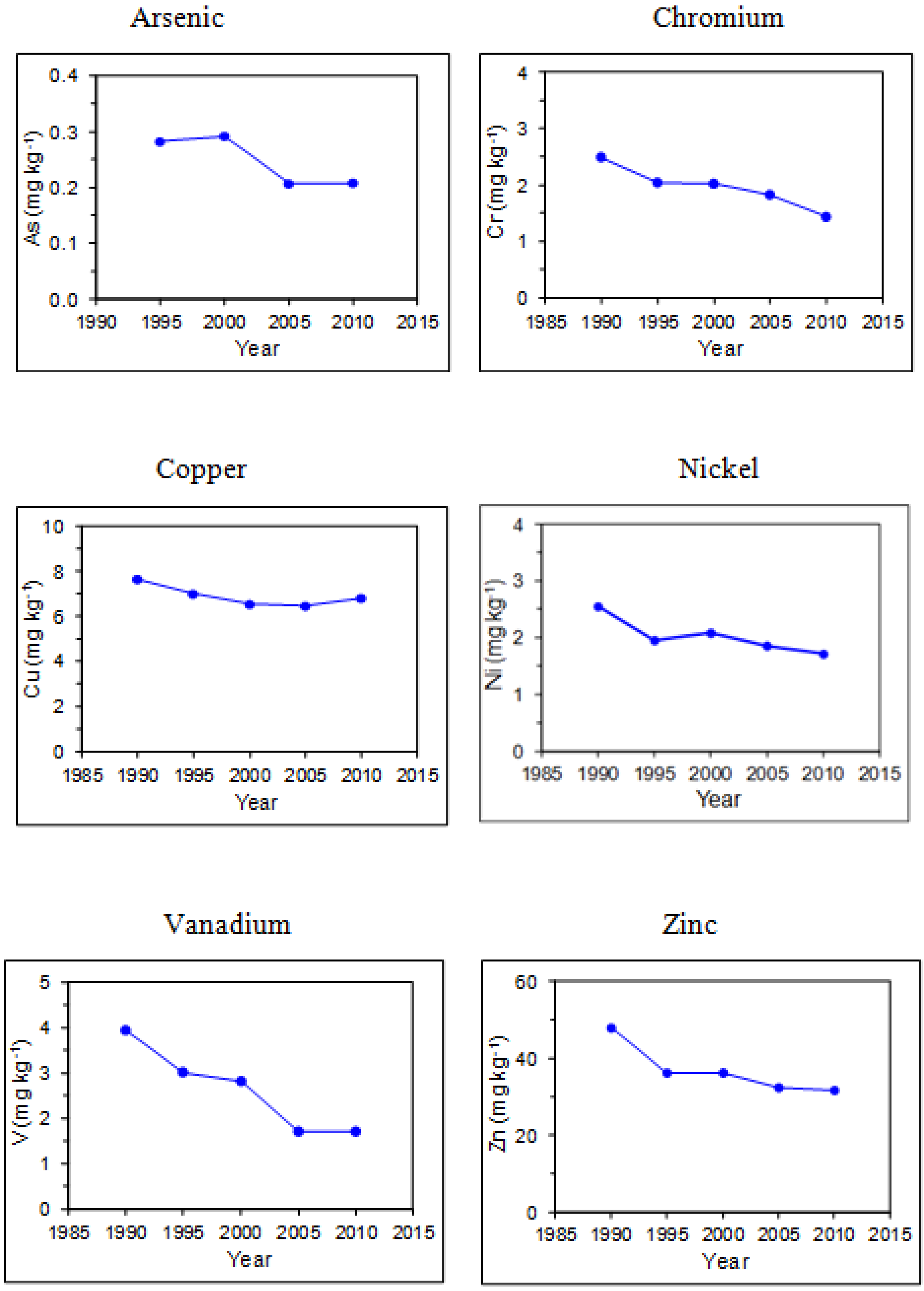
Figure 4

Temporal trend of cadmium, lead and mercury concentration in mosses (blue dots and line) compared with the trend of EMEP-modelled deposition for these heavy metals (red dots)



15. For other metals, the decline in concentrations in mosses also follows the decline in reported emissions since 1990, with the lowest decline being reported for copper for both variables (figure 5). However, on a national or regional scale within countries deviations from the general European trend were sometimes found, i.e., temporal trends were country- or region-specific, with no changes or even increases being observed between survey years. Therefore, even in times of generally decreasing metal deposition across Europe, temporal trends can be different for different geographical scales.

Figure 5
Temporal trend of arsenic, chromium, copper, nickel, vanadium and zinc concentration in mosses

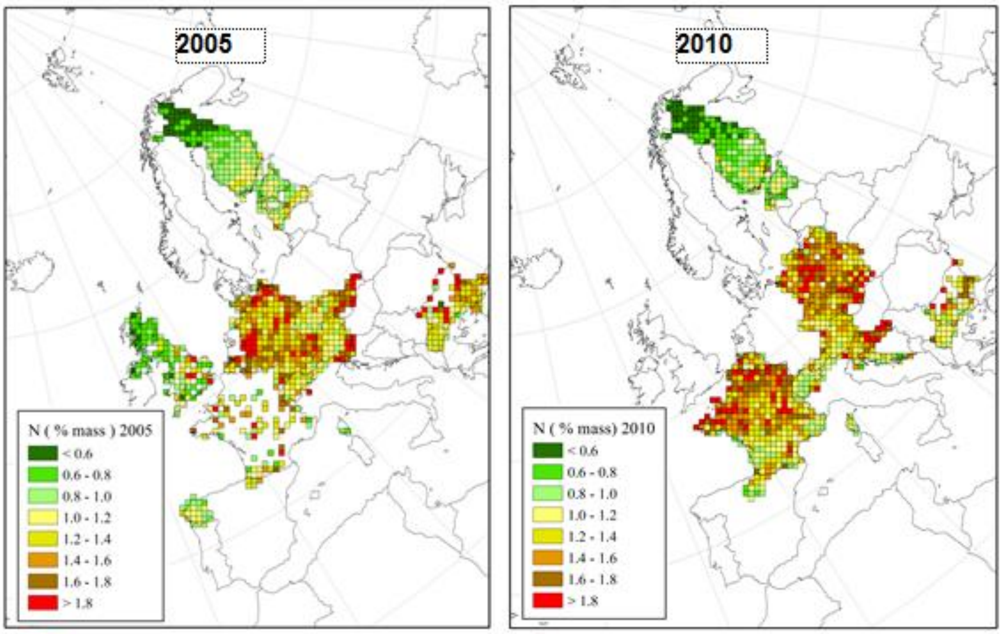


V. Spatial patterns and temporal trends (2005–2010) of nitrogen concentrations in mosses

16. The spatial pattern of the nitrogen concentration in mosses was similar in 2005 and 2010, with lower values being observed for Finland than in the rest of Europe (figure 6). Generally, high concentrations of nitrogen were found in Western, Central and South-Eastern Europe. In addition, the average median value for nitrogen has hardly changed since 2005: from 1.26 per cent in 2005 to 1.19 per cent in 2010, a decline of 5 per cent for the 13 countries reporting nitrogen concentrations in mosses in both 2005 and 2010. This decline is in agreement with the almost 7 per cent decline reported by EMEP for modelled total nitrogen deposition in the 27 member States of the European Union between 2005 and 2010. The considerable decline (30 per cent) reported for Slovenia is most likely due to either a more careful sampling campaign, trying to avoid the sampling of mosses affected by canopy drip from trees, or the fact that the majority of sampling sites were different for 2005 and 2010. Increases in the median values were found in the Czech Republic (19 per cent increase) and France (15 per cent). For France it should be noted that the total nitrogen concentration in mosses was determined at 88 sites in 2005, whereas it was determined at 442 sites in 2010.

Figure 6

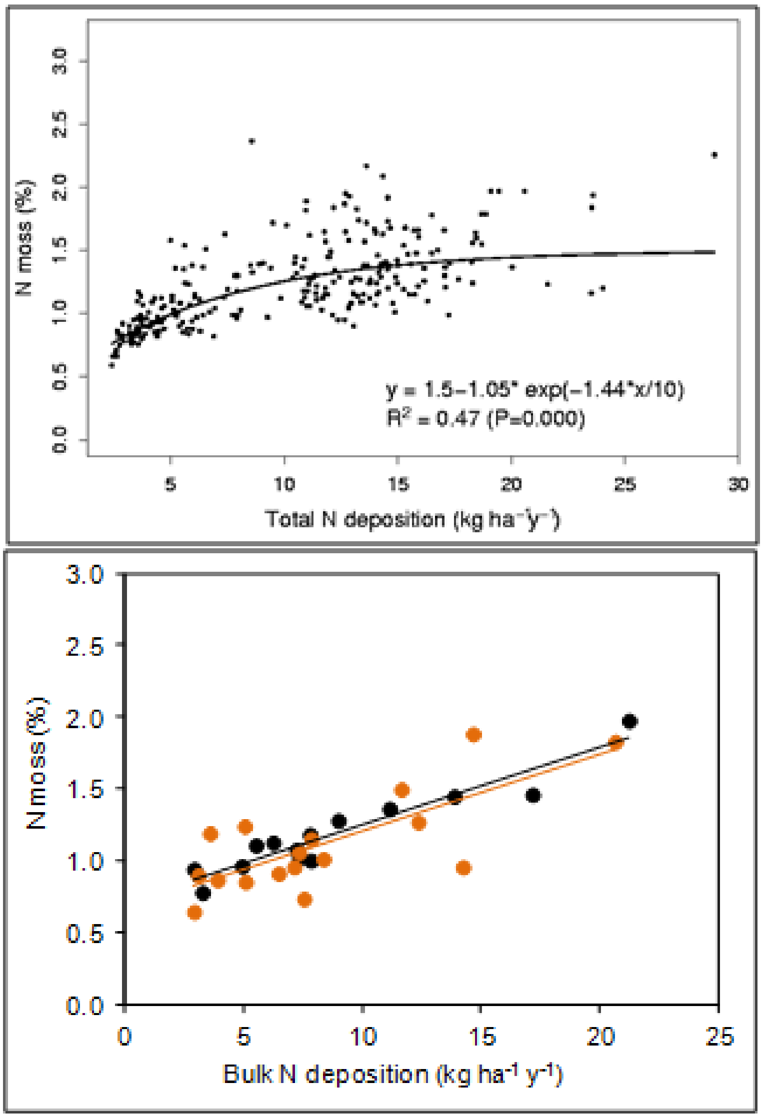
Mean total nitrogen concentration in mosses per EMEP grid cell (50 km x 50 km) in 2005 and 2010



17. The relationship between the total nitrogen concentration in mosses and EMEP-modelled deposition has not been analysed yet for 2010; however, it is expected that the relationship will be similar to the one reported for 2005 (figure 7). In 2005, this relationship showed saturation occurring at a modelled deposition rate of around 15 kg of nitrogen (N) per hectare (ha^{-1}) per year (y^{-1}). In Switzerland, a linear relationship was found between the nitrogen concentration in mosses and measured bulk nitrogen deposition at the site level in 2005, and an almost identical relationship was found in 2010 (figure 5).

Figure 7

Relationship between nitrogen concentration in mosses and (left) EMEP-modelled total nitrogen deposition in 2005 for EMEP grid cells (50 km x 50 km) where at least five moss sampling sites were present and (right) measured bulk nitrogen deposition rates in Switzerland in 2005 (black dots and line) and 2010 (orange dots and line)



VI. Conclusions

18. Based on the current survey, the following conclusions were drawn:

(a) Moss biomonitoring provides a cheap, complementary method to deposition analysis for the identification of areas at risk from high atmospheric deposition fluxes of heavy metals and nitrogen and for monitoring changes over time;

(b) For the priority metals cadmium, lead and mercury and for nitrogen the decline in average median concentrations in mosses across Europe is in agreement with that reported for modelled atmospheric deposition;

(c) Despite the general European decline in concentrations in mosses between 2005 and 2010 (and also since 1990), country and region-specific temporal trends were observed;

(d) Despite the apparent success of the implementation of air pollution abatement techniques in large areas of Europe, further measures are required in South-Eastern Europe to reduce the relative high emissions of heavy metals. For nitrogen, more stringent air pollution abatement strategies are required across Europe to reduce the areas at risk from adverse effects of elevated atmospheric nitrogen deposition.

VII. Recommendations

19. Based on the current survey, the following recommendations were made:

(a) As ecosystems and human health are still predicted to be at risk from adverse effects of heavy metals and nitrogen in the future, the moss survey should be continued to monitor any future trends in heavy metal and nitrogen deposition in Europe, with the next survey recommended for 2015–2016;

(b) Further stimulation of the participation in South-Eastern European countries for both heavy metals and nitrogen is encouraged;

(c) In addition, more countries are encouraged to report on the nitrogen concentration in mosses in the future;

(d) It is recommended to use the newly available data for 2010 to further assess the performance of the EMEP models, particularly the model that estimates the atmospheric deposition of the priority heavy metals cadmium, lead and mercury;

(e) For nitrogen, it is recommend to investigate in further detail the relationship between measured total nitrogen deposition and the total nitrogen concentration in mosses at the site level.

20. Further details of the outcome of the 2010/11 European moss survey and temporal trends since 1990, can be found in the full report.[8] The full report also contains country-specific contributions from 13 countries: Albania, Austria, Bulgaria, Croatia, Czech Republic, Finland, Norway, Poland, Slovakia, Spain, Sweden, Switzerland and the former Yugoslav Republic of Macedonia.

[8] H. Harmens, D. Norris and G. Mills, *Heavy metals and nitrogen in mosses: spatial patterns in 2010/2011 and long-term temporal trends in Europe* (Bangor, United Kingdom: ICP Vegetation Programme Coordination Centre, Centre for Ecology and Hydrology, 2013). Available from http://icpvegetation.ceh.ac.uk/publications/documents/Finalmossreport2010-11forweb.pdf.