Schröder et al. Environmental Sciences Europe 2012, **24**:20
http://www.enveurope.com/content/24/1/20

Environmental Sciences Europe
a SpringerOpen Journal

RESEARCH

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Does spatial auto-correlation call for a revision of latest heavy metal and nitrogen deposition maps?

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Abstract

Background: Within the framework of the Convention on Long-range Transboundary Air Pollution atmospheric depositions of heavy metals and nitrogen as well as critical loads/levels exceedances are mapped yearly with a spatial resolution of 50 km by 50 km. The maps rely on emission data and are calculated by use of atmospheric modelling techniques. For validation, EMEP monitoring data collected at up to 70 sites across Europe are used. This spatially sparse coverage gave reason to test if the chemical and physical relations between atmospheric depositions and their accumulation in mosses collected at up to 7000 sites throughout Europe can be quantified in terms of statistical correlations which, if proven, could be used to calculate deposition maps with a higher spatial resolution. Indeed, combining EMEP maps on atmospheric depositions of cadmium, lead and nitrogen and the related maps of their concentrations in mosses by use of a *Regression Kriging* approach yielded deposition maps with a spatial resolution of 5 km by 5 km. Since spatial auto-correlation can make testing of statistical inference too liberal, the investigation at hand was to validate the 5 km by 5 km deposition maps by analysing if spatial auto-correlation of both EMEP deposition data and moss data impacted on the significance of their statistical correlation and, thus, the validity of the deposition maps. To this end, two hypotheses were tested: 1. The data on deposition and concentrations in mosses of heavy metals and nitrogen are not spatially auto-correlated significantly. 2. The correlations between the deposition and moss data lack statistical significance due to spatial autocorrelation.

Results: As already published, the regression models corroborated significant correlations between the concentrations of heavy metals and nitrogen in atmospheric depositions on the one hand and respective concentrations in mosses on the other hand. This investigation proved that atmospheric deposition and bioaccumulation data are spatially auto-correlated significantly in terms of Moran's I values and, thus, hypothesis 1 could be rejected. Accordingly, the degrees of freedom were reduced. Nevertheless, the results of the calculations regarding the reduced degrees of freedom indicate that the statistical relations between atmospheric depositions and bioaccumulations remained statistically significant so that hypothesis 2 could be rejected, too.

Conclusions: The positive auto-correlation in data on atmospheric deposition and bioaccumulation does not call for a revision of the 5 km by 5 km deposition maps published in recent papers. Therefore we can conclude that the European moss monitoring yields data that support the validation of modelling and mapping of atmospheric depositions of heavy metals and nitrogen at a high spatial resolution compared to the 50 km x 50 km EMEP maps.

Keywords: Biomonitoring, Concentrations of Cd, Pb and N in mosses, Atmospheric depositions of Cd, Pb and N, EMEP deposition network and modelling, ICP Vegetation

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Background

Measurements of atmospheric depositions are needed as a basis to evaluate environmental quality. To this end, deposition data are, amongst others, used to calculate exceedance maps for critical loads. Critical loads are defined as quantitative estimates of an exposure to one or more pollutants below which significant harmful effects on specified ecosystem functions are not expected to occur according to present knowledge [1]. In Europe, the control of heavy metals and reactive nitrogen emissions to air is regulated under several directives of the European Union and protocols of the Long-range Transboundary Air Pollution (LRTAP) Convention. Under the LRTAP Convention, the European Monitoring and Evaluation Programme (EMEP) collects emission data from European countries in order to model atmospheric transport and depositions of air pollutants. Amongst others, depositions of cadmium (Cd), lead (Pb) and nitrogen (N) are calculated using chemical transport models yielding deposition maps with a grid size of 50 km by 50 km. The modelling results are validated by use of deposition data collected at EMEP monitoring sites. However, the number of EMEP measurement stations is rather limited across Europe and EMEP stations are generally under-represented in Southern and Eastern Europe. In 2005, 53 EMEP stations measured the concentration of nitrogen compounds in precipitation and wet deposition, whereas up to 41 stations reported air concentrations of nitrogen compounds [2]. In case of heavy metals, the number of EMEP measurement stations accounts for up to 70 throughout Europe [3].

For ecosystem-specific evaluations of exposure in terms of atmospheric depositions or critical loads information with high spatial resolution is crucial [4-10]. To enhance the spatial resolution of the deposition maps data on phenomena that are physically and statistically related with depositions and collected at higher spatial density could

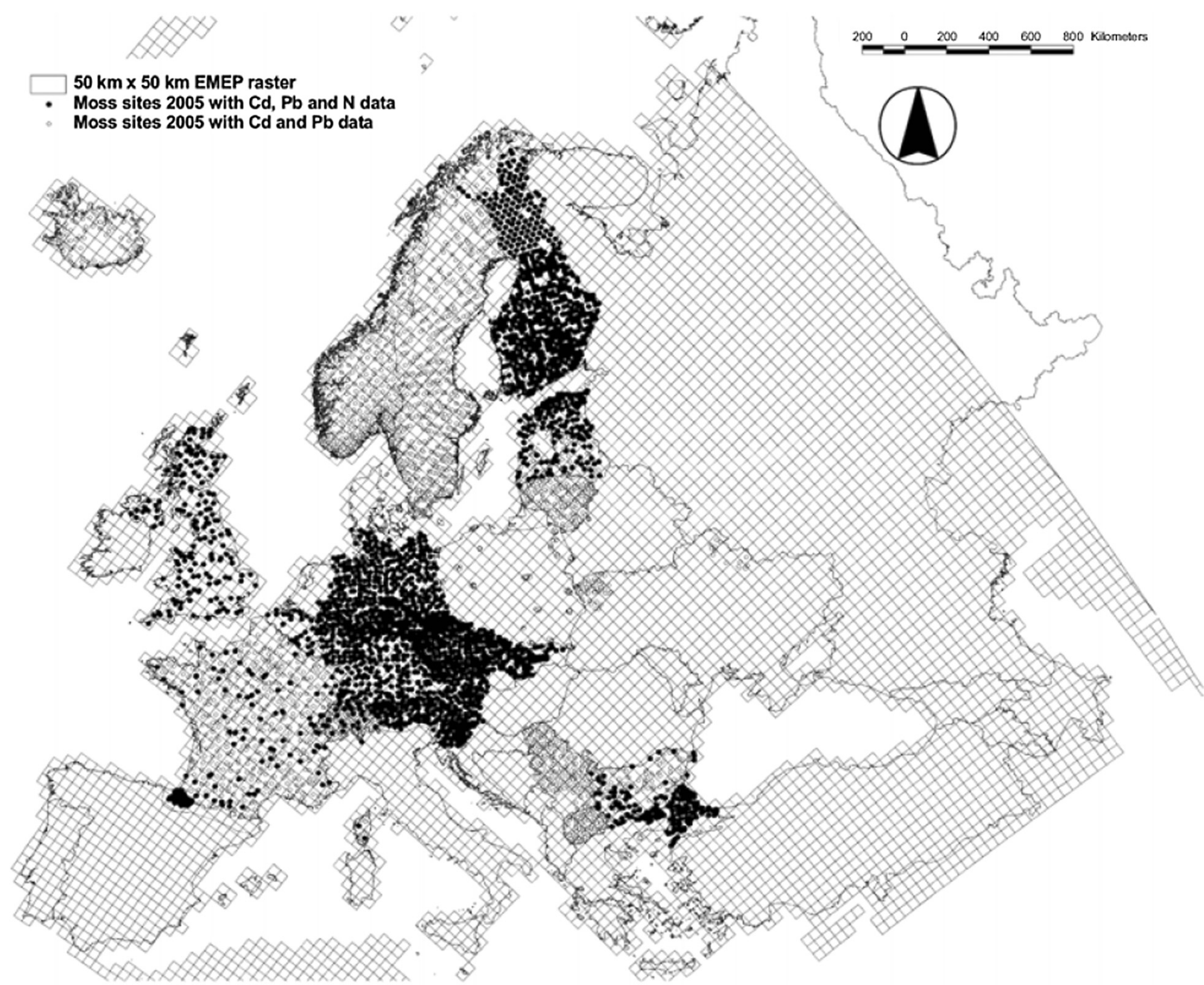

Figure 1 Moss sampling sites 2005 and EMEP raster across Europe.

be utilised. Once substances emitted to air have been deposited, they can accumulate in plant biomass, as for instance in mosses. The European moss biomonitoring network encompassing up to 7000 sites was established in 1990 and has been repeated every five years since then [3]. Carpet-forming, ectohydric mosses obtain most trace elements and nutrients directly from precipitation, occult deposition and dry deposition. Therefore, the moss technique has been shown to provide a complementary, time-integrated measure of element deposition from the atmosphere to terrestrial systems quantifying the potential availability of potentially harmful substances such as heavy metals [3] or nutrients such as nitrogen [2]. With the moss technique a much higher sampling density can be achieved than with conventional deposition analysis. The national moss surveys across Europe are coordinated by the ICP Vegetation and follow recommendations regarding sampling, preparation and chemical analyses of the mosses put down in an experimental protocol. Figure 1 shows the distribution of moss species sampled in the 2005 survey together with the EMEP 50 km x 50 km raster.

The European moss monitoring produces datasets at high spatial resolution which was used to evaluate the performance of the EMEP model [11] and to calculate deposition maps with a spatial resolution of 5 km by 5 km through modelling the statistical relations between atmospheric deposition and bioaccumulation of of Cd, Pb and N by use of Regression Kriging [12,13]. The corresponding methodology and results can be summarised as follows: The EMEP deposition maps were intersected within a GIS with Kriging maps on N, Cd and Pb accumulations in mosses. The maps were calculated by Ordinary Kriging on basis of the variograms presented in the 'Results' section of this paper. Next medians were calculated for all moss estimations within each EMEP grid cell. Both moss data and corresponding modelled deposition values were ln-transformed and their relationship investigated and modelled by linear regression analysis. The regression models corroborate that the Cd concentration in mosses is correlated with the EMEP modelled total Cd deposition across Europe (regression coefficient according to Pearson, $r_p = 0.67$; regression coefficient according to Spearman, $r_s = 0.69$). The coefficient of determination is $R^2 = 0.44$. The same is true for Pb with $r_p = 0.76$ and $r_s = 0.77$ and $R^2 = 0.58$ [13]. The regression analysis of the estimated N concentrations in mosses and the modelled EMEP depositions, too, resulted in clear linear regression patterns with coefficients of determination of $R^2 = 0.62$ and Pearson correlations of $r_p = 0.79$ and Spearman correlations of $r_s = 0.70$, respectively [12]. The regression equations were applied on the moss kriging estimates of the element concentration in mosses. The respective residuals were projected onto the centres of the EMEP grid cells and were mapped using variogram analysis and ordinary kriging. Finally, the residual and the regression map were summed up to the map of total N, Cd, and Pb deposition in terrestrial ecosystems throughout Europe. This was done for a 5 km by 5 km raster which was chose due to the results of nearest neighbourhood statistics: All nearest neighbour distances of all moss sites were calculated in ArcGIS 10.0 and summarised in terms of quantile statistics. The 10^{th} quantile was chosen in order to adjust the interpolation raster to the high density of the moss monitoring net approximating ca. 5000 m (exact value: 5468.5 m).

By application of this environmental mapping methodology the EMEP maps could be improved in both spatial resolution and, by adding more empirical data, in terms of validation aspects. Due to the use of moss data the maps furthermore depict direct impacts of atmospheric pollution to terrestrial ecosystem functions since the uptake of pollutants by plants can be seen as the first step towards an effect.

Auto-correlation is a widespread phenomenon in environmental systems [14,15]. In statistics, the auto-correlation of a random process is defined as the similarity of, or correlation between, values of a process at neighbouring points in time or space. Auto-correlation describes the similarity between observations as a function of the separation of time and space intervals between them. Positive auto-correlation means that the individual observations contain information which is part of other, timely or spatial neighbouring, observations. Subsequently, the effective sample size will be lower than the number of realized observations. Negative auto-correlation can have the opposite effect, thus, making the effective sample larger than the realized sample [16]. Therefore, autocorrelation can have several implications for calculating statistics of measurement data in terms of statistical inference testing [17,18]. Initially, investigations of statistical implications of auto-correlation concentrated mainly on time series analysis and were followed by investigations of the impacts of spatial auto-correlation on inference testing methods. For instance, it could be shown, that positive spatial auto-correlation enhances type I errors, so that parametric statistics such as Pearson correlation coefficients, are declared significant when they should not be [19]. These findings gave reason for the investigation at hand aiming at validating recently published deposition maps which were derived by a Regression Kriging approach [12,13].

Results

The results are presented in Figures 2, 3, 4 and Tables 1 and 2. Variogram analyses (Figures 2, 3, 4) reveal that the concentrations of Cd, Pb and N in mosses measured at 5731 (Cd, Pb) and 2781(N) sites, respectively, exhibit positive spatial auto-correlation. The measurement values were transformed log normally due to the highly skewed

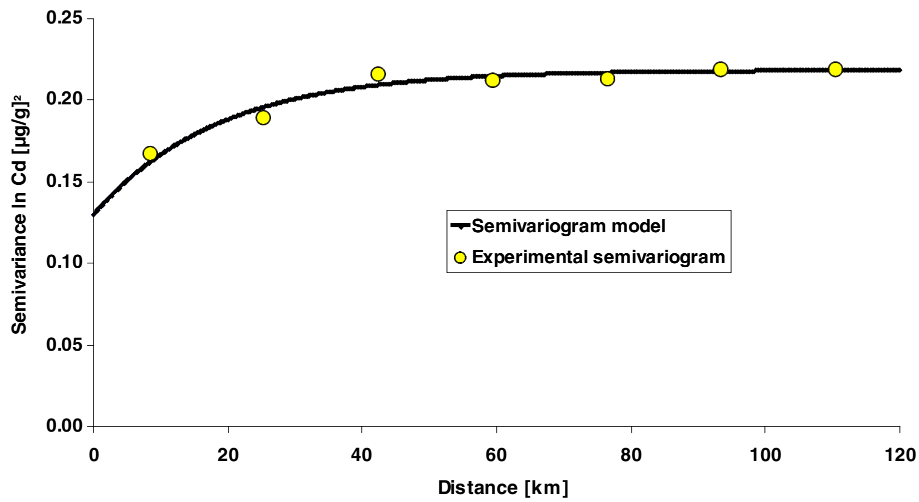

Figure 2 Semi-variogram of Cd concentrations in mosses (ln-transformed) [Exponential semi-variogram model; nugget effect: 0.13; sill: 0.09; nugget/sill [%]: 59; range [km]: 59.3].

data distributions of the elements investigated (Skewness Cd = 8.1; skewness Pb = 11; skewness N=1). With variogram analysis experimental semi-variances are calculated in terms of half of the average squared differences of all pairs of measurement values within each distance interval. The mean nearest neighbour distances were chosen as a starting point for the distance intervals resulting in 15.6 km for Cd, 15.8 km for Pb and 16.5 km for N. The width of the variogram window was set so that both the increase and the flattening of the semi-variance values with the separation distance could be clearly observed. Then, semi-variogram models were fitted to the experimental semi-variograms by a least squared regression line. The variogram model can be described by three parameters: range, sill and nugget-effect. The range equals the maximum separation distance for which a distinct increase of semi-variogram values, and therefore spatial autocorrelation, can be observed. The sill corresponds to the semi-variance assigned to the range. High spatial variability within the first distance interval can be caused by measurement errors and other confounding factors resulting in nugget-effects. Accordingly, the variogram model will tend to cut the ordinate of the variogram plot above the origin. Even though such a high nugget effect can be observed for Cd, Pb and N a distinct increase of experimental semi-variances with separation distance proves that spatial autocorrelation exists in all three cases.

Table 1 corroborates by means of calculated Moran's I values for the same distance intervals that this positive spatial auto-correlation is also statistically significant.

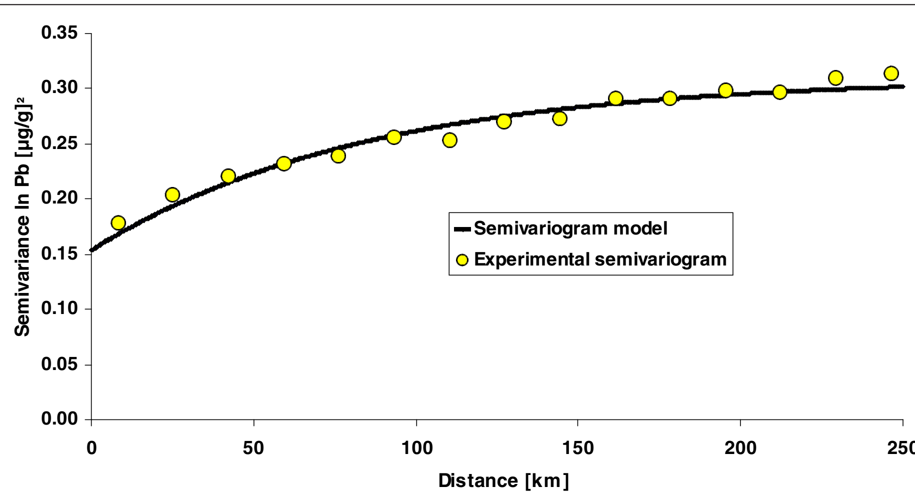

Figure 3 Semi-variogram of Pb concentrations in mosses (ln-transformed) [Spherical semi-variogram model; nugget effect: 0.19; sill: 0.12; nugget/sill [%]: 61.2; range [km]: 255].

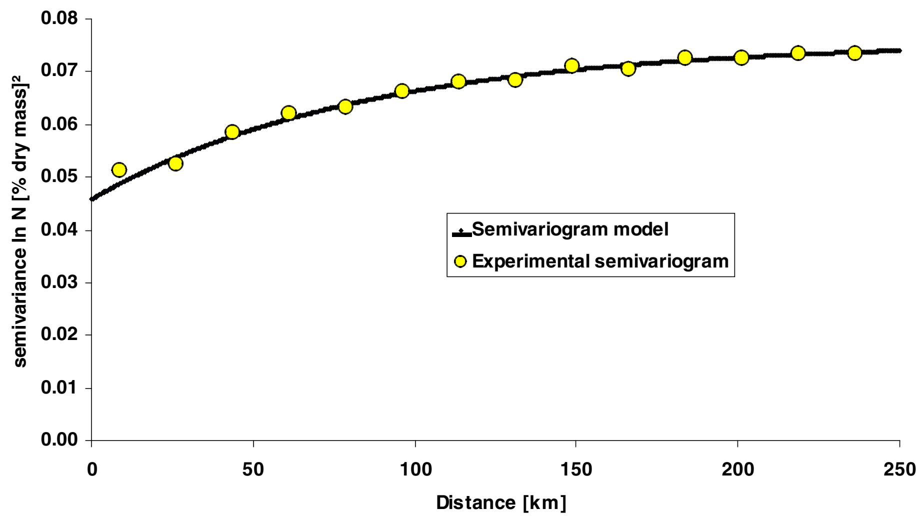

Figure 4 Semi-variogram of N concentrations in mosses (ln-transformed) [Spherical semi-variogram model; nugget effect: 0.046; sill: 0,03; nugget/sill [%]: 60.5; range [km]: 209].

Moran's I values range from approximately +1 to −1, where positive Moran'I values represent positive and negative Moran's I values represent negative spatial autocorrelation [20]. P-values may be calculated for each of the derived Moran's I values and therefore the statistical significance of spatial autocorrelation can be assessed.

Table 1 Moran's I for Cd, Pb and N concentrations in mosses regarding the first 20 distance intervals according to mean nearest neighbour distance (Cd: 15.6 km; Pb: 15.8 km; N 16.5 km)

Distance interval	Cd	Pb	N
1	0.73	0.53	0.47
2	0.57	0.62	0.49
3	0.52	0.58	0.41
4	0.45	0.55	0.41
5	0.45	0.53	0.38
6	0.41	0.48	0.34
7	0.39	0.47	0.34
8	0.38	0.45	0.31
9	0.35	0.44	0.30
10	0.35	0.41	0.28
11	0.34	0.39	0.27
12	0.32	0.38	0.27
13	0.30	0.35	0.25
14	0.28	0.34	0.26
15	0.29	0.33	0.25
16	0.27	0.31	0.26
17	0.25	0.30	0.27
18	0.24	0.29	0.28
19	0.23	0.28	0.28
20	0.24	0.27	0.27

All coefficients are statistically significant ($p < 0.001$).

Consequently, this positive spatial auto-correlation was accounted for in the calculation of statistical correlation between Cd, Pb and N (for N: dry, wet and total) medians for EMEP cells and the corresponding EMEP according to [21]. Table 2 contains some descriptive statistical measures for all variables investigated. The results of the correlation analysis show that the auto-correlation considerably reduces the degrees of freedom. Despite this, the correlations remained statistically significant ($p < 0.01$ for Cd and Pb; $p < 0.05$ for N) (Table 3). As a result, both hypotheses which have been tested were to be falsified. Thus, the 5 km by 5 km deposition maps which have been calculated based on the correlations between atmospheric depositions and bioaccumulations, and by means of Regression Kriging [12,13] are statistically valid and could be used for ecosystem-specific exposure evaluations or calculations of critical loads.

Discussion

Neighbouring measurement values along time series or across geographic space that are more similar or less similar than expected for randomly associated pairs of measurements are positively auto-correlated or negatively auto-correlated, respectively. Temporal and spatial autocorrelation is a widespread property of environmental variables and as such the result of abiotic and biotic processes and their interrelations. Thus, spatial patterns existing across the whole spectrum of spatial scales are functional in ecosystems and not the result of pure random effects. This fact conflicts with the assumptions of statistics such as, e.g., the independence of observations.

Table 2 Descriptive statistics for the medians of Cd, Pb and N estimations within EMEP grid cells and corresponding EMEP modelling results

	n	Min	Max	Mean	Stabw	1st quartil	Median	3rd quartil
N estimations in moss [% in dry mass]	769	0.3	2.9	1.2	0.4	0.9	1.2	1.4
N total despostion [kg/ha*a] year of sampling	769	97.0	2901.2	1023.6	558.9	541.3	1068.5	1403.8
N total despostion [kg/ha*a] sum	769	256.2	8919.4	3069.2	1650.3	1634.5	3223.4	4212.7
Cd estimations in moss [µg/g]	1534	0.020	3.520	0.184	0.163	0.096	0.150	0.225
Cd total despostion [g/ha*a] year of sampling	1534	2.7	722.7	34.6	37.9	14.9	27.6	42.6
Cd total despostion [g/ha*a] sum	1534	10.3	2105.6	106.7	117.2	45.8	82.3	126.8
Pb estimations in moss [µg/g]	1523	0.45	137.85	5.25	6.31	2.20	3.50	5.70
Pb total despostion [g/ha*a] year of sampling	1523	83.5	5274.1	1068.6	723.1	500.5	941.5	1460.3
Pb total despostion [g/ha*a] sum	1523	323.7	16650.6	3237.0	2157.9	1555.9	2855.6	4346.7

The problem with auto-correlated data is that an observation at a certain point in time or space does not bring 100 % additional information and, hence, cannot be accounted for one full degree of freedom due to its similarity with neighbouring measurements [22,23]. Taken the computation of a Pearson or Spearman correlation coefficient as an example, positive spatial auto-correlation of the two variables, e.g. atmospheric deposition and concentrations in mosses, provoke that the coefficient is declared too often significant. The fact that ecological reality in terms of auto-correlation often violates the assumption of inference statistical methods is of crucial importance for ecological sampling design, analysis and evaluation of field experiments and surveys [22,24,25]. The same holds true for spatial analysis of landscapes [26], including for instance testing the significance of the relation between spatially auto-correlated data at the landscape level [27]. The latter case was examined in this investigation by example of data on atmospheric deposition and physically related concentrations of heavy metals and nitrogen in mosses. Even when accounting for spatial auto-correlation and applying the method proposed by [21] the relation between deposition and bioaccumulation remained statistically significant.

Conclusion

The positive auto-correlation in data on atmospheric deposition and concentrations in mosses does not call for revision of the 5 km by 5 km deposition maps published recently [12,13]. Therefore, the European moss monitoring yields data that support the validation of modelling and mapping of atmospheric depositions of heavy metals and nitrogen at a high spatial resolution. The validation of the 5 km by 5 km deposition maps in terms of the auto-correlation tests presented in this investigation allows for the maps to be used to calculate critical loads exceedances complementing the ecotoxicological endpoint 'accumulation'. Thus, the complementary use of data derived from two internationally harmonized monitoring networks, the

Table 3 Spearman correlation coefficients corrected for the existence of spatial autocorrelation for Cd, Pb and N concentrations in mosses and the corresponding deposition rates for Cd, Pb as well as N (for N: dry, wet and total)

				Degrees of freedom		p-values	
		n	Spearman	Original	Corrected	Original	Corrected
Cd	Year of sampling	1534	0.66	1532	59	<0.001	<0.001
	sum	1534	0.64	1532	58	<0.001	<0.001
Pb	Year of sampling	1523	0.73	1521	23	<0.001	0.008
	sum	1523	0.73	1521	23	<0.001	0.01
N total wet	Year of sampling	769	0.63	767	13	<0.001	0.029
	sum	769	0.64	767	13	<0.001	0.029
N total dry	Year of sampling	769	0.59	767	16	<0.001	0.028
	sum	769	0.59	767	16	<0.001	0.027
N total	Year of sampling	769	0.63	767	13	<0.001	0.026
	sum	769	0.64	767	13	<0.001	0.026

In each case the correlations were both calculated for the concentration of the elements in the year the sampling was performed as well as for the sum according to the accumulation period of three years.

EMEP deposition measurement and the ICP Vegetation moss monitoring, allows for synergies enhancing the spatial validity of deposition maps and subsequent products.

Methods

The EMEP deposition data for the year 2005 and the moss concentration data collected within the International Co-operative Programme on Effects of Air Pollution on Natural Vegetation and Crops (ICP Vegetation, http://icpvegetation.ceh.ac.uk) were analysed in a two step procedure: Firstly, the deposition and moss data were mapped by use of Regression Kriging (see 'Introduction') [12,13]. Secondly, in this investigation we analysed how spatial auto-correlation in the modelled deposition data and the moss data influences the testing of statistical inference. To this end, two hypotheses were tested: 1. The data on deposition and concentrations in mosses of Cd, Pb and N are not spatially auto-correlated significantly. 2. The correlations between the deposition and moss data lack statistical significance due to spatial auto-correlation. Both hypotheses were tested through calculation of:

- Experimental and modelled semi-variograms of ln transformed moss data for Cd, Pb and N;
- Amount and significance of spatial auto-correlation for the first ten distance classes of the semi-variograms by use of Moran's I [20];
- Significance of correlations between data on atmospheric deposition and concentrations in mosses with regard to the potential reduction of degrees of freedom due to positive spatial auto-correlation according to [21].

The extension Geostatistical analyst from ESRI ArcGIS 10.0 was used for calculation of semi-variograms. The software SAM v4.0 (Spatial Analysis in Macroecology) was applied in order to calculate Moran's I values and to account for spatial auto-correlation when testing the correlation between EMEP values and moss data for statistical significance [22].

Competing interests
No competing interests do exist.

Authors' contributions
WS wrote the text. RP conducted the computations. HH, HF and II supported the work by dealing with the validity of experimental and modelling data. All authors read and approved the final manuscript.

Acknowledgement
We thank the United Kingdom Department for Environment, Food and Rural Affairs (Defra; contract AQ0810 and AQ0816), the UNECE (Trust Fund) and the Natural Environment Research Council (NERC) for funding the ICP Vegetation Programme Coordination Centre at CEH Bangor, UK. The contributions of many more scientists in 2005/6 and all the funding bodies in each country are gratefully acknowledged (see [2,3], for details).

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Received: 8 November 2011 Accepted: 29 April 2012
Published: 9 June 2012

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doi:10.1186/2190-4715-24-20
Cite this article as: Schröder *et al.*: Does spatial auto-correlation call for a revision of latest heavy metal and nitrogen deposition maps?. *Environmental Sciences Europe* 2012 **24**:20.