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Title: Metal accumulation in mosses across national boundaries: uncovering and ranking causes of spatial variation

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Abstract: This study aimed at cross-border mapping metal loads in mosses in eight European countries in 1990, 1995, and 2000 and at investigating confounding factors. Geostatistics was used for mapping, indicating high local variances but clear spatial autocorrelations. Inference statistics identified differences of metal concentrations in mosses on both sides of the national borders. However, geostatistical analyses did not ascertain discontinuities of metal concentrations in mosses at national borders due to sample analysis in different laboratories applying a range of analytical techniques. Applying Classification and Regression Trees (CART) to the German moss data as an example, the local variation in metal concentrations in mosses were proved to depend mostly on different moss species, potential local emission sources, canopy drip and precipitation.

Capsule: Factors affecting the spatial variation in metal accumulation in mosses were mapped by geostatistics and ranked by CART.

Keywords: CART; geostatistics; metal bioaccumulation; mosses; ordination.

1 Introduction

The European moss surveys of 1990, 1995 and 2000 enabled to map metal concentrations across the borderlines of the participating countries (Harmens et al., 2004; Schröder and Pesch, 2004). However, several studies revealed significant spatial variation and the metal concentration in mosses to be correlated with moss species, canopy drip, precipitation, altitude, distance to the sea and analytical techniques (Berg and Steinnes, 1997; Frahm, 1998; Herpin et al., 2004; Zechmeister et al., 2003). As contradicting results were reported (Čeburnis and Valiulis, 1999; Siewers and Herpin, 1998; Siewers et al., 2000), the interpretation of the moss data remained difficult (Herpin, 1997). Since environmental planning and policy need information which is valid at higher spatial levels than individual sampling sites, the integration of site-specific measurements and the connections between monitoring networks should be achieved (Ferretti, 2001; Parr et al., 2002). This implies the generalisation of measurement values from sampling sites to areas of greater extend (Miller et al., 2004). This study aimed at achieving this by cross-border geostatistical analysis of the moss survey data from Austria, Czech Republic, Germany, Italy, Slovakia, Slovenia, Poland, and Switzerland. A second aim was to rank factors that, in addition to metal deposition, affect the spatial patterns of metal bioaccumulation. Special attention was given to the question whether the effect of the metal analyses in participating laboratories cause significant differences of metal bioaccumulation data on both sides of the borders between the countries.

2 Materials and Methods

2.1 Data

The moss survey data of 1990, 1995 and 2000 (Table 1) covering the eight European countries were used to examine whether the measured values were spatially valid for both the sampling sites and adjacent areas. Detailed information on characteristics of the sampling sites (Table 2 rows 1 to 11) and their surroundings (Table 2 rows 12 to 30) was only available from the German Moss Monitoring Information System (GEMMIS). With the exception of the distance to the sea, all factors which could influence metal accumulation in mosses were restricted to the sampling sites or the area within a 5 km radius around them. This enabled to rank some causes of spatial variation underlying the nugget effects in the variogram analyses which often remained unexplained.

2.2 Statistical methods

The laboratory differences were tested by means of the Mann-Whitney U-Test. CART was used to ordinate the site-specific and regional factors that influence the metal bioaccumulation in addition to the atmospheric metal deposition. For all descriptive and inference statistical analyses SPSS 12.0 was used. The CART-analyses were performed using the SPSS module Answer Tree 3.1. For all geostatistical analyses the Geostatistical Analyst from the ESRI-product ArcGIS 9.1 was applied.

2.2.1 Classification Trees

CART enables processing large sets of mixed data, i.e. nominal, ordinal and metric scale data, without prior transformation. CART allows uncovering hierarchical and non-linear relationships among one dependent variable and several predictors. This is achieved by nested binary “if-then-else” splits, each maximising the homogeneity of the target variable. The Gini index is commonly used as impurity measure when the target variable is categorical. The predictor selected is the one for which the two new classes have both the greatest difference from each other and the greatest within-group similarity for the response variable. The two new classes are then examined separately with respect to each of the predictor variables to see if they can be split again. CART does not make any assumptions regarding the distribution of the data and can use predictors more than once, thus multiple interrelations can be detected (Breiman et al. 1984).

2.2.2 Geostatistics

Since monitoring data should also be valid for areas beyond the sampling sites, surface maps should be produced from measurement data if they are spatially auto-correlated. Spatial auto-correlation can be examined by variogram analysis. The mean squared differences of all pairs of measurement values (semi-variances) were calculated for so called bins of a variogram map to derive an experimental semi-variogram. This procedure may result in more than one semi-variogram value per lag. In order to perform kriging it is necessary to adapt a defined variogram model to the experimental variogram. This can be achieved by means of mathematical models fitted to the experimental variogram in terms of a least-squares regression line. A 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. Spatial variability within the first lag can be caused by measurement errors and other confounding factors resulting in high semi-variances. Such nugget effects remain unexplained in most investigations.

Based on the variogram model, several kriging methods can be used for spatial predictions. They all minimise the estimation variance and rely on weighted averaging of the measured values within a chosen kriging window. For the interpretation of the kriging estimations one needs to know how far the predictions deviate from the quality-controlled measurement values. To achieve this, iteratively each measurement point is extracted from the sample and, based on the respective variogram model, estimated again using the surrounding auto-correlated measurement values. This cross-validation quantifies how well the model estimates values at locations without measurements. Differences between measured and estimated values can be described by the mean error (ME), the root mean square standardised error (RMSSE), the median percentile error (MPE), the MPE adjusted to the ratio of the empirical value range and the absolute cross-validation error range (MPEc), and the coefficient of correlation by Pearson (C).

The ME equals the average value of the differences between measured and estimated values and is a measure of the degree of bias. If the ME equals 0, the model can be assumed to be unbiased. By setting the respective measured value to 100%, the difference between the measured and predicted values may be calculated in percent. The MPE then is the median value of the percentile deviations, and enables a comparison of the quality of the estimation of several measured variables, e.g. metal concentrations. The MPEc adjusts the MPE with regard to the different ranges of the measurement values and the cross-validation errors. The RMSSE is a measure of the mean squared deviation between measured and estimated values standardised by the kriging variance. Ideally, the RMSSE should equal 1. C quantifies the correlation between the measurements and estimations (Olea, 1999).

3 Results

3.1 Spatial patterns of bioaccumulation

In order to map the spatial patterns of the metal bioaccumulation across Austria, the Czech Republic, Germany, Italy, Slovakia, Slovenia, Poland, and Switzerland, data from these countries were analysed geostatistically with regard to the concentration of As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, V, and Zn in mosses. The Gauß-Krüger coordinate system (date: Potsdam; ellipsoid: Bessel; 15th meridian) was used as a geographical reference system. To be able to detect temporal trends, only data sets were selected from the national databases where identical moss species were collected since 1990 or 1995. As a result, in 1990 94 (Hg) to 477 (Cr, Cu, Fe, Ni) measurement values were analysed (Table 3). For the 1995 and 2000 surveys, 1061 (Hg) to 1257 (Cr) and 1072 (Hg) to 1266 (Cd, Fe) values were used for the computations, respectively. Figure 1 illustrates the geographical locations of the sampling sites and the moss species collected. The mapping of the metal bioaccumulation was restricted conservatively to 60 km around the monitoring sites, which clearly is far beneath the maximum auto-correlation range (Figure 2).

The descriptive statistical analyses reveal high variability with regard to all elements and years (Table 3). The coefficients of variation exceed 100% in 15 of 30 cases. Highest variability was found for Cu in 1995 (218%) and Hg in 1995 (317%) and 2000 (281%). In addition, all elements show highly left skewed distributions, expressed by a skewness of above 1 in all cases. The highest skewness was found for Cd in 1995 (21.7) and 2000 (10.8) and for Hg in 1995 (30.6) and 2000 (11.9). The large differences between the 98th percentile and the maximum values furthermore indicate the existence of outliers.

Regarding the calculation of experimental semi-variograms, the average distance of each measurement site to its nearest neighbour was set as a starting point for the lag size (Webster and Oliver, 2001). This is considered a “safe practice” when encountering irregular sampling configurations because it allows to give consideration to the spatial distribution of sampling sites (Olea, 1999). The number of lags assigned to each lag size should enable an optimal detection of the autocorrelation structure. In most cases hardly any spatial autocorrelation was found using the raw data. Since all elements showed highly left skewed distributions, the metal concentrations in mosses were transformed lognormally and variogram analyses and kriging was carried out with the transformed values. Such transformations were done for all elements and years except for V in 1990 and in 1995 (Table 4). In seven cases even the lognormally transformed data showed no spatial autocorrelation. Therefore, sites showing the highest ratios of squared cross-validation errors and kriging variances were excluded iteratively until spatial autocorrelation could be detected. If the sill cannot be visualised in the variogram window even when applying high lag sizes and numbers of lags, this is indicative of deterministic trends in the data. Therefore, second order polynomial functions were calculated and then subtracted from the measurement values. The residuals were then used for variogram analysis and for the kriging procedures. Except for V in 1990 such a universal kriging approach was performed in all cases (Table 4). For Cd, Cr, Ni and Pb in 1990 no autocorrelation could be detected due to pure nugget effects.

In all cases spherical variogram models fitted the experimental semi-variogram values best. All models showed high nugget-sill ratios (Table 4), indicating high local variability of the measurement data; apart from two exceptions these ratios were above 0.5. The highest nugget effect was detected for Cu in 1990, accounting for 84% of the sill. The auto-correlation ranges were between 91 and 333 km. Except for V in 1990 and 1995, a lognormal universal kriging approach was used to estimate values for a 16 km by 16 km grid. The grid resolution was set according to the average mean nearest neighbour distance of all measurement sites. A four-sector neighbourhood was defined to avoid directional bias and a maximum of five points was accounted for in each sector to estimate a certain point. The results of the kriging procedures for Zn concentrations in mosses in 1990, 1995 and 2000 are shown as an example (Figure 2). The maps illustrate that, except for Germany, a continuous decrease of Zn bioaccumulation occurred with time. The highest Zn values were estimated for Slovakia (in 1990) and for Katowice in Poland (all three campaigns).

To describe the quality of the surface estimations, the ME-, RMSSE-, MPE-, MPEc-, and the C-values were calculated from the results of cross-validation (Table 5). Whereas the ME indicated neither underestimation nor overestimation in almost all cases, this was not true for the RMSSE. With the exception of Hg in 1990 and 1995, the variances calculated from the cross-validation errors by average were higher than the theoretical kriging variances. A maximum of this ratio was observed for Pb in 1995. Due to local variability and the variances referred to in Table 3, high MPE-values were observed in almost all cases. The MPEc allowed comparing the different elements more reliably, ranging from 11.7% for Hg to 35.1% for As in 1995. C was lowest for As in 1990 and 1995 and reached values above 0.5 in all but six cases.

3.2 Variability due to laboratories, altitude and sea spray

The U-tests proved significant differences on both sides each of the frontiers between the eight European countries (Table 6). Concerning the effects of altitude and sea spray on the metal accumulation in Germany no clear trends could be identified (Table 7).

3.3 Ranking factors of spatial variability

CART allowed ranking the factors causing the spatial variability of the metal concentrations which were geostatistically quantified for arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), nickel (Ni), lead (Pb), antimony (Sb), titanium (Ti), vanadium (V) and zinc (Zn) in terms of the nugget effect. Using Ni as an example, it is demonstrated how to read a CART-Dendrogram (Figure 3).

The statistical distribution of Ni concentrations is depicted in the box on top of the two dendrograms. The mean Ni concentration in mosses was 2.63, 1.86 and 1.33 µg g^{-1} in 1990, 1995 and 2000, respectively. Although for the Ni accumulation in 1990 no statistically relevant factor could be detected, in 1995 as well as in 2000, the moss species was the most powerful predictor. For 1995 the following two subgroups resulted from the first split: node 1 encompasses 778 samplings sites where the mean Ni concentration in *Pleurozium schreberi* and *Scleropodium purum* was 1.66 µg g^{-1}; 201 sites were clustered into node 2 with a mean Ni concentration in predominantly *Hypnum cupressiforme* of 2.65 µg g^{-1}. Node 1 was subdivided by the spatial density of traffic routes within 5 km around the sampling sites. If the density of traffic routes was less than 6.45%, then the mean Ni concentration in mosses was 1.62 µg g^{-1} (node 3). In cases where the traffic density exceeded this value, the mean Ni concentration was 2.73 µg g^{-1} (node 4). Node 2 was split by the percentage of urban areas within 5 km of the monitoring sites. If this percentage was below 8.24%, the mean Ni concentration in mosses was 2.49 µg g^{-1} and if it exceeded 8.24% the mean Ni concentration was 3.57 µg g^{-1}. The CART-tree for Ni in 2000 shows similar properties as the one for 1995.

In further CART-models the other twelve metals were defined as the target variable. The results are summarised here with regard to the hierarchy and the frequency of the predictor variables within the decision trees. The moss species most frequently subdivided the root node into two sub-nodes (11 times) and occurred most frequently throughout all levels of the decision trees (14 times). In most cases *H. cupressiforme* was separated from the *P. schreberi* and *S. purum*. In general, the subgroups in which *H. cupressiforme* was present showed higher metal bioaccumulation than the subgroups with *P. schreberi* and *S. purum*. The latter two species formed sub-nodes where the metal concentrations showed distributions similar to those in the root node. Subordinated to the moss species, predictors related to potential emission sources around the monitoring sites, canopy drip and precipitation proved to be the most dominant predictors in the calculated decision trees, i.e. 11, 10 and 8 times respectively. Altitude and distance of the sampling sites to the sea occurred five and two times, respectively.

4 Discussion

This investigation is the first geostatistical mapping of the metal accumulation in mosses across the national borders of Austria, the Czech Republic, Germany, Italy, Slovakia, Slovenia, Poland, and Switzerland. The variogram analyses depicted spatial autocorrelation although high nugget effects gave hints on the existence of small scale variability of the metal loads in the mosses. Furthermore, this study includes the first CART-analysis of the factors associated with the spatial variability of the metal bioaccumulation. Mainly the moss species, potential emission sources around the monitoring sites, canopy drip and precipitation were proved to cause the spatial variability of the metal accumulation. Furthermore, different techniques for digestion and analysis seem to influence the measurements. However, geostatistical analyses did not ascertain discontinuities of metal bioaccumulation at the national borders due to different analytical techniques, as other factors proved to be more important in explaining the variation in metal concentrations in mosses.

Siewers and Herpin (1998) investigated the correlation between the metal concentrations in *P. schreberi* and *S. purum* and between *P. schreberi* and *H. cupressiforme.* No distinct tendency could be derived from the results of regression analysis due to the low amount of samples and high dispersion around the regression function. Köhler and Peichl (1993) found that *H. cupressiforme* accumulates at least twice as much metal as *P. schreberi*. However, Rühling and Tyler (1968) found that the metal accumulation was only slightly higher in *H. cupressiforme* than in *Hylocomium splendens* and *P. schreberi.* In contrast, Folkeson (1979) detected much higher Pb concentrations in *H. cupressiforme* compared to *H. splendens* and *P. schreberi.* Throughout Germany, Schröder et al. (2002) measured significantly higher concentrations of As, Cd, Hg, Ni and Pb in *H. cupressiforme* compared with the other mosses which were collected from different sites. This observation may be explained by the surface morphology of *H. cupressiforme*, which is likely to enhance the capture of metals from the atmosphere.

Fernández et al. (2000, 2002) stated that an interspecies calibration for *H. cupressiforme* and *S. purum* is urgently needed. Interspecies calibration tests of metal concentrations in *P. schreberi, S. purum* and *H. cupressiforme* in the Czech Republic revealed insignificant ($p > 0.05$) differences in aluminium (Al), As, Cd, cobalt (Co), Cr, Cu, Fe, molybdenum (Mo), Ni, Pb, sulphur (S), V and Zn concentrations in *P. schreberi* and *S. purum,* whilst in *H. cupressiforme* these elements accumulated up to 112-225% of the concentration found in *P. schreberi* and *S. purum* (Suchara and Sucharová, 1998). *H. cupressiforme* had significantly higher concentrations of Al, Co, Mo, Ni, Pb and Zn than *P. schreberi* and *S. purum*. It was hypothesized that the higher metal concentrations in *H. cupressiforme* were due to contamination rather than to a higher efficiency of element capture. Thöni et al. (1996) found higher concentrations of Ni, Pb and Zn in *H. cupressiforme* than in *P. schreberi* at three sampling sites, but no species-specific variation for Cu. However, these differences were small compared with the variability of several other characteristics at each of the sites. These results were corrobated by Zechmeister (1994) who found slightly higher concentrations in *H. cupressiforme* than in *P. schreberi* and *H. splendens* but significant higher amounts only for Ni and Cd. The slopes of regression relationships between *P. schreberi* and the moss species *H. splendens, H. cupressiforme* and *Rhytidiadelphus squarrosus* sampled at the same sites in the UK were in most cases and for most metals close to unity, suggesting no species-specific efficiencies in the capture of metals from the atmosphere (Ashmore et al., 2000). Possibly the differences in metal accumulation between moss species are metal-specific and vary with climate conditions and other site characteristics.

The precipitation is an important factor controlling wet atmospheric deposition of elements, washing out dry deposition, leaching elements from plants and affecting moss production. Main variables co-operating with the precipitation are altitude, geomorphology and wind conditions. The amount of precipitation could be correlated with the metal bioaccumulation due to atmospheric rain out and wash out-effect (Čeburnis and Valiulis, 1997; Frahm, 1998; Zechmeister, 1995). But heavy rain could also remove dry deposition from mosses before incorporation or contaminate the mosses by splash erosion (Herpin, 1997). There is a strong correlation between altitude and precipitation, and therefore correlation between metal concentration in mosses and altitude have been proven empirically (Fowler et al., 1988; Soltes, 1992, Zechmeister, 1995). However, partial regression analyses showed significant ($p < 0.05$) positive and negative correlations between the precipitation without the effect of altitude and concentrations of Ag, Ba, Bi, Cd, Cr, Cs, Cu, Fe, In, Mo, Ni, Pb, Rb, S, Se, Sn, Tl, V, Zn (r_p = 0.18-0.55) and Mn (r_b = -0.27) in moss samples in the Czech Republic. Although strong positive correlations between precipitation and metal deposition were proven, no correlations were found between precipitation and the leaching of elements (except Mn) from mosses or the initiation of moss production (Sucharová and Suchara, 2004b).

The predictors which represent the canopy drip effect were of third importance according to the CART-analyses. This is in agreement with Økland et al. (1999) who proved correlations between metal concentrations in *H. splendens* and the density of trees and crowns, respectively. However, this is contrary to findings reported by Fernández and Carballeira (2002) and Čeburnis and Steinnes (2000). In the Czech Republic, moss samples were collected in large open spaces in forests and were therefore little affected by throughfall and canopy drip from trees. The concentration of the mainly geogenous elements Al, As, Co, Cr, Fe, Hg, S and V in the mosses decreased significantly ($p < 0.01$) with the forest cover percentage in a 5-km radius around the sampling plots (Sucharaová and Suchara, 2004a), indicating an effective filtering of dust particles by forests.

Altitude is easy to determine but a rather complex explanatory variable. Many more or less strong covariables occur along altitudinal gradients: precipitation, vegetation, wind speed, air density, spatial density of settlements and anthropogenic activities, which can influence the emission and deposition of air pollutants and moss production. Consequently, the results on the correlation between altitude and the accumulation of metals in mosses are contradictory. Gerdol et al. (2002) found decreasing concentrations of Al, Ca, Co, Cr, Fe, Ni, Mo, Ni, Pb in *H. splendens* with increasing altitude. They hypothesised that this was due to increasing biomass production and decreasing metal binding capacity with increasing altitude. The opposite was observed for Cd, Cu, Mg, Na, Zn. Zechmeister (1995) found positive correlations between the concentrations of Pb, Cd, Zn and S in mosses and altitude and precipitation. However, he stated that this could also be related to the atmospheric deposition by increasing amounts of rain and reduced vegetation cover at higher altitudes. This was corroborated by Sucharova and Suchara (2004b).

The concentrations of Al, As, Co, Cr, Cu, Fe, Hg, Ni, S, and V in mosses throughout the Czech Republic decreased significantly ($p < 0.05$) with altitude. Even after filtering out the effect of precipitation as a tight covariable, the concentrations of all these elements and the concentrations of Cd, Mo, Pb and Zn in mosses were negatively correlated with increasing altitude (Sucharová and Suchara, 2004a). Without the precipitation effect, significant negative relationships between altitude and Ag, Be, Bi, Ce, Ga, In, La, Li, Pr, Se, Th, U and Y and significant positive relationships for Cs and Rb were also proven (Sucharová and Suchara, 2004b). Even after elimination of the precipitation effect in the variable altitude, the variable still remained too complex to reveal exact causality. Most of investigated elements may be bound on particulate carriers, which concentration decreases with altitude. On the other hand, increased concentrations of Cs and Rb in mosses might be explained by increased concentration of these elements in the local mountain rocks and release of Cs and Rb to the environment by erosion.

5 Conclusion

The main factors that influence the bioaccumulation of metals in mosses could be ranked as follows: moss species, potential emission sources around the monitoring site, canopy drip and precipitation. They should be identified before producing transboundary maps on metal bioaccumulation. This will only be feasible if all participating countries provide additional information about site characteristics as currently is done in e.g. the German moss surveys. Furthermore, a harmonized procedure for digestion and analysis should be aimed for.

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Figure 1: Locations of the moss monitoring sites from eight European countries.

The figure depicts the locations of the sampling sites that were used for the geostatistical analysis of metal concentrations in mosses in eight central European countries. In the map to the left sites are depicted that were sampled in 1990 as well as in 1995 and 2000. The map to the right illustrates monitoring sites that were sampled both in 1995 and in 2000. The maps also show the moss species that were collected in these surveys (1990-1995-2000 and 1995-2000, respectively).

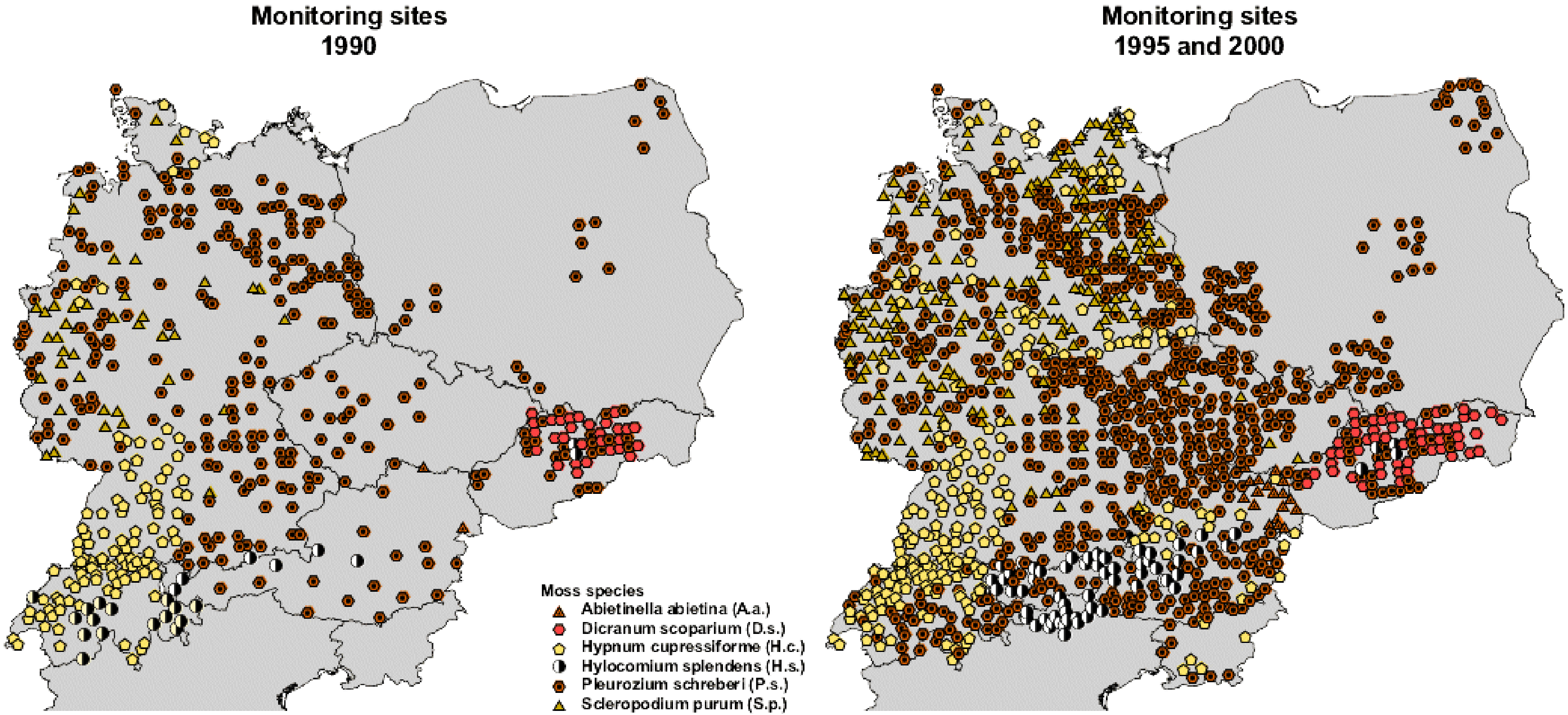

Figure 2: Spatial patterns of the bioaccumulation of Zn in 1990, 1995 and 2000 in Central Europe.

Figure 2 depicts the spatial patterns of the bioaccumulation of zinc in mosses in 1990, 1995 and 2000 in eight central European countries. Variogram analysis and lognormal universal kriging was applied to estimate values for 16" 16 km² grid squares.

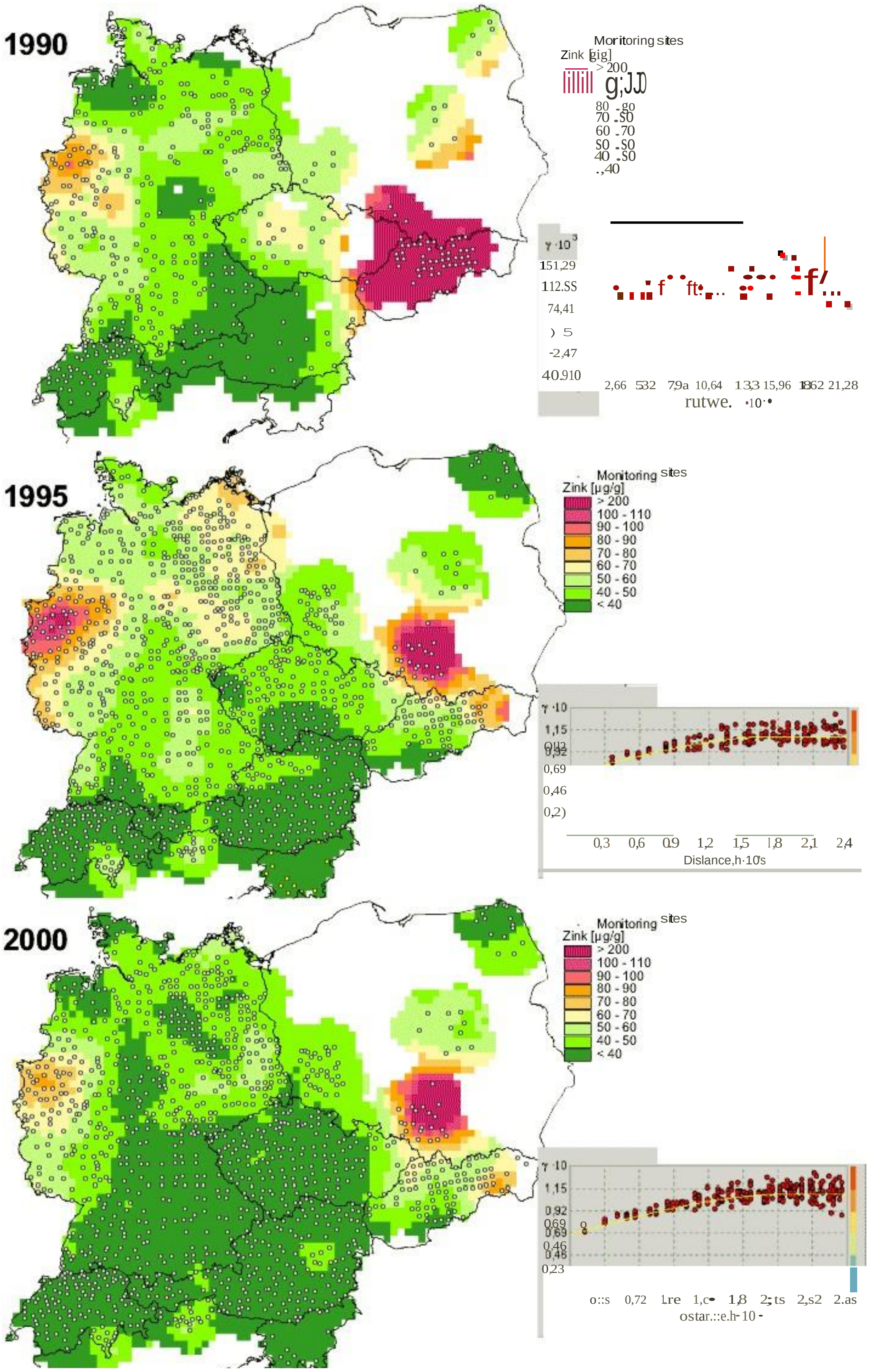

Figure 3: CART-analyses for Ni concentrations in mosses in Germany for 1995 and 2000.

In both years moss species were the most important predictor, splitting each root node into one node containing sites with mainly *Pleurozium schreberi* (*P.s.*) and *Scleropodium purum* (*S.p.*) (with relatively low Ni bioaccumulation) and one node containing mainly *Hypnum cupressiforme* (*H.c.*) with relatively high Ni bioaccumulation. Further splitting occurred due to the percentage of areas related to traffic emissions (P_Verkehr) and urban areas (P_Urb).

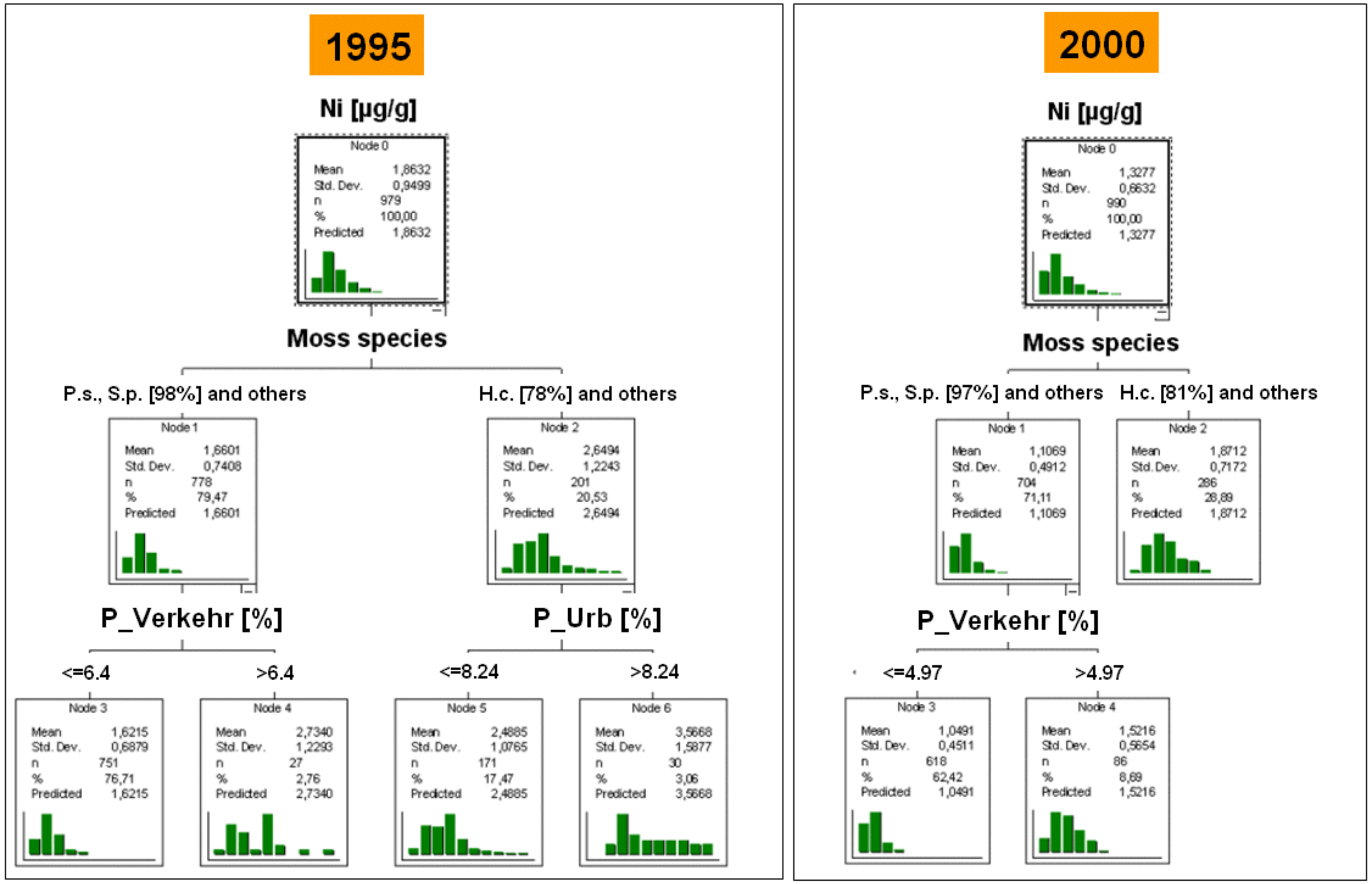

Table 1: Data used for geostatistical estimation and testing of laboratory bias.

	Country	As			Cd			Cr			Cu			Fe			Hg			Ni			Pb			V			Zn		
		1990	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**
Analytical device*	Austria	3	8	8	2	7	7	2	2	2	2	2	2	2	2	2	3	3	3	2	2	2	2	2	2	2					
	Czech Rep.	7	2	1	7	2	1	7	2	1	7	2	1	3	2	1		3*	3*	7	2	1	7	2	1	7	2	1	3	2	1
	Germany	4	1	1	7	1	1	2	1	1	2, 3	1	2	2, 3	1	2		1	6	2	1	1	7	1	1	2	1	1	2, 3	1	2
	Italy		1	1		1	1		1	1		1	1		2	2		3*	3*		1	1		1	1		1	1		1	1
	Poland				3	3	3,4	3	3	3	3	3	3	3	3	3				3	3	3	3	3	3,4	3		3	3	3	3
	Slovakia		7	9	3	7	7	3	7	9	3	2	3	3	2	9		6	3*	3	7	9	3	7	7		2	9	3	2	9
	Slovenia		9	9		9	9		9	9					9	9		9	9											9	9
	Switzerland	1	1	1	1	1	1	1	1	1	2	1	2	2	1	2	6	-	6	1	1	1	2	1	1	1	1	1	2	1	2
Number of sampling sites**		**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**	**1990**	**1995**	**2000**
	Austria	26	183	182	26	181	184	26	183	180	26	183	184	26	183	184	26	182	184	26	183	184	25	183	184	26	183	184	26	183	184
	Czech Rep.	17	147	147	17	147	147	17	147	147	17	147	147	17	147	147		145	145	17	147	147	17	147	147	17	147	147	17	147	147
	Germany	234	630	629	234	630	629	293	630	629	293	630	630	293	630	629		630	630	293	630	630	293	630	629	293	630	629	293	630	628
	Italy		20	20		20	20		20	20		20	20		20	20		20	20		20	20		20	20		20	20		20	20
	Poland				18	78	78	18	78	78	18	78	78	18	76	78				18	77	78	18	78	78	18		78	18	78	78
	Slovakia			82	55	73	82	55	73	82	55	73	82	55	73	82		73	82	55	73	82	54	73	82		73	82	52	66	82
	Slovenia		11	11		11	11		11	11					11	11		11	11											11	11
	Switzerland	68	115	115	68	115	115	68	115	115	68	115	115	68	115	115	68			68	115	115	68	115	115	68	115	115	68	115	115

* Analytical techniques are decoded as follows:

1 ICP-MS
2 ICP-OES
3 AAS
3* AAS amalgam.preconcentration - AMA 256 Altec
4 AAS hydrid
6 AAS cold-vapour
7 AAS graphite furnace
8 AAS flow injectiuon
9 INAA

** Only those sites are listed that were used for the geostatistical analyses presented in this paper

Table 2: Predictors for metal accumulation in mosses in Germany used in Classification Trees (*numbers relate to Corine landcover-categories level 3).

<table>
<tr><th colspan="2">Predictor</th><th>Spatial reference</th><th colspan="2">Source</th></tr>
<tr><td colspan="2">Inclination</td><td rowspan="11">sampling site</td><td colspan="2" rowspan="11">GEMMIS
(Schröder and Pesch 2004)</td></tr>
<tr><td colspan="2">Direction</td></tr>
<tr><td colspan="2">Land use category</td></tr>
<tr><td colspan="2">Moss species</td></tr>
<tr><td rowspan="7">Distance to the next ...</td><td>... road</td></tr>
<tr><td>... motorway</td></tr>
<tr><td>... housing estate</td></tr>
<tr><td>... industrail area</td></tr>
<tr><td>... tree</td></tr>
<tr><td>... shrub</td></tr>
<tr><td>... vegetation</td></tr>
<tr><td colspan="2">Agricultural land
(including crop land)</td><td rowspan="16">area within a radius of 5 km of each sampling site</td><td>211, 243</td><td rowspan="15">Corine Landcover*</td></tr>
<tr><td colspan="2">Agricultural crop land</td><td>211</td></tr>
<tr><td colspan="2">Agricultural land and natural vegetation (without crop land)</td><td>243</td></tr>
<tr><td colspan="2">Meadows and pastures</td><td>231</td></tr>
<tr><td colspan="2">Forets, woodland and shrubs, fruit growing area</td><td>311, 324, 313, 312, 222</td></tr>
<tr><td colspan="2">Mixed forests</td><td>313</td></tr>
<tr><td colspan="2">Coniferous forests</td><td>312</td></tr>
<tr><td colspan="2">Deciduous forest</td><td>311</td></tr>
<tr><td colspan="2">Woodland and shrubs</td><td>324</td></tr>
<tr><td colspan="2">Fruit-growing area</td><td>222</td></tr>
<tr><td colspan="2">Urban area</td><td>111, 112</td></tr>
<tr><td colspan="2">Industrials area</td><td>121</td></tr>
<tr><td colspan="2">Urban and industrial areas</td><td>111,112, 121</td></tr>
<tr><td colspan="2">Landfill and minig dump</td><td>132</td></tr>
<tr><td colspan="2">Tidelands, saltmeadows, sands, dunes</td><td>423, 421, 331</td></tr>
<tr><td colspan="2">Traffic routes</td><td>Encoded by 190</td><td></td></tr>
<tr><td colspan="2">Distance to the sea</td><td rowspan="3">local (sampling site)</td><td colspan="2">Distance grid</td></tr>
<tr><td colspan="2">Altitude</td><td colspan="2">UNEP Grid</td></tr>
<tr><td colspan="2">Precipitation
(Sum of the means 1961 to 1990)</td><td colspan="2">German Weather Service (DWD)</td></tr>
</table>

Table 3: Descriptive statistical variables of the metal concentration in mosses in eight central European countries for 1990, 1995 and 2000.

	n	mean [µg/g]	min [µg/g]	max [µg/g]	var.c. [%]	skew.	50perc. [µg/g]	98perc. [µg/g]
1990								
As	345	0.55	0.10	13.70	173	9.6	0.36	2.42
Cd	418	0.49	0.13	5.70	96	4.5	0.34	1.90
Cr	477	2.8	0.3	30.6	87	5.6	2.1	8.1
Cu	477	10.1	1.8	84.2	65	4.3	8.8	27.1
Fe	477	963	51	6257	100	2.8	650	4498
Hg	94	0.06	0.01	0.27	59	2.9	0.05	0.15
Ni	477	2.7	0.1	11.6	54	1.7	2.4	6.2
Pb	475	21.8	4.2	359.0	125	7.2	15.3	98.2
V	422	3.5	0.6	19.0	63	3.0	3.0	11.3
Zn	474	62.1	13.2	353.0	82	3.0	44.9	245.0
1995								
	n	**mean [µg/g]**	**min [µg/g]**	**max [µg/g]**	**var.k. [%]**	**skew.**	**50perc. [µg/g]**	**98perc. [µg/g]**
As	1106	0.29	0.001	2.69	98	3.5	0.22	1.12
Cd	1255	0.40	0.03	6.29	107	7.2	0.29	1.58
Cr	1257	2.4	0.05	61.4	192	6.8	1.3	17.2
Cu	1246	10.5	2.1	650.3	218	21.7	8.2	27.0
Fe	1255	564	68	10560	113	6.8	408	2356
Hg	1061	0.06	0.001	6.44	317	30.6	0.05	0.18
Ni	1245	2.0	0.1	33.4	84	7.6	1.6	6.7
Pb	1246	12.7	1.7	499.0	186	14.6	8.7	50.8
V	1168	1.8	0.1	17.9	79	4.9	1.5	5.2
Zn	1250	51.3	14.2	821.0	65	11.4	45.2	113.5
2000								
	n	**mean [µg/g]**	**min [µg/g]**	**max [µg/g]**	**var.k. [%]**	**skew.**	**50perc. [µg/g]**	**98perc. [µg/g]**
As	1186	0.24	0.02	2.21	97	3.2	0.16	0.97
Cd	1266	0.29	0.05	7.17	128	10.8	0.21	1.10
Cr	1262	1.6	0.3	26.2	149	6.1	1.0	8.6
Cu	1256	7.4	2.7	41.5	48	4.2	6.8	15.6
Fe	1266	526	5	9981	122	6.8	365	2258
Hg	1072	0.08	0.02	3.44	281	11.9	0.05	0.26
Ni	1256	1.6	0.4	12.6	74	4.0	1.3	5.1
Pb	1255	7.8	0.7	104.2	121	4.7	5.1	36.8
V	1255	2.0	0.2	30.3	128	4.9	1.2	10.1
Zn	1265	42.8	14.4	589.9	63	9.2	37.1	106.4

var.k.[%]	coefficient of variation
skew.	skewness
50perc.[µg/g]	50th percentile = median
98perc.[µg/g]	98th percentile

Table 4: Variogram and kriging variables of the metal concentration in mosses in eight central European countries for 1990, 1995 and 2000.

1990					
Dataset	**Trend**	**Logn**	**Outl.**	**Range [km]**	**N/S-Ratio**
As	2nd	yes	1	197	0.62
Cu	2nd	yes	0	98	0.84
Fe	2nd	yes	0	162	0.62
Hg	2nd	yes	0	91	0.63
V	none	no	0	182	0.26
Zn	2nd	yes	0	150	0.65
1995					
Dataset	**Trend**	**Logn**	**Outl.**	**Range [km]**	**N/S-Ratio**
As	2nd	yes	0	146	0.62
Cd	2nd	yes	3	230	0.66
Cr	2nd	yes	0	154	0.64
Cu	2nd	yes	3	333	0.51
Fe	1st	yes	0	137	0.64
Hg	2nd	yes	1	201	0.60
Ni	2nd	yes	0	166	0.71
Pb	2nd	yes	2	201	0.68
V	2nd	no	4	159	0.63
Zn	2nd	yes	0	179	0.64
2000					
Dataset	**Trend**	**Logn**	**Outl.**	**Range [km]**	**N/S-Ratio**
As	2nd	yes	0	178	0.54
Cd	2nd	yes	0	146	0.59
Cr	2nd	yes	0	234	0.51
Cu	2nd	yes	0	312	0.58
Fe	2nd	yes	0	142	0.67
Hg	2nd	yes	4	126	0.68
Ni	2nd	yes	0	163	0.72
Pb	2nd	yes	0	122	0.47
V	2nd	yes	0	168	0.53
Zn	2nd	yes	0	230	0.61

Trend	order of polynomial function substracted from the measurement values
LogNormal	Lognormal kriging performed
Outl.	Number of outliers excluded from the geostatistical analysis
Range [km]	largest distance of observed and modelled autocorrelation
N/S-Ratio	nugget to sill ratio

Table 5: Cross validation results for kriging of the metal concentration in mosses in eight central European countries for 1990, 1995 and 2000.

1990					
Dataset	**ME [µg/g]**	**RMSSE**	**MPE [%]**	**MPEc [%]**	**C**
As	-0.01	1.42	31.9	29.6	0.24
Cu	0.08	1.01	20.7	17.1	0.67
Fe	-1.04	1.16	30.5	19.3	0.79
Hg	0.00	0.99	32.9	25.3	0.30
V	-0.01	1.19	21.5	13.3	0.68
Zn	-0.48	1.24	17.8	14.2	0.81
1995					
Dataset	**ME [µg/g]**	**RMSSE**	**MPE [%]**	**MPEc [%]**	**C**
As	0.00	1.14	40.3	35.14	0.29
Cd	0.00	1.10	19.0	12.96	0.84
Cr	-0.06	1.09	34.2	26.70	0.72
Cu	-0.18	1.20	18.5	14.82	0.62
Fe	-21.13	1.26	31.4	24.35	0.62
Hg	0.00	0.84	23.7	11.70	0.74
Ni	-0.03	1.19	25.9	20.94	0.56
Pb	-1.01	1.93	28.4	26.65	0.44
V	0.00	1.04	27.3	24.12	0.47
Zn	-0.56	1.29	16.9	14.38	0.62
2000					
Dataset	**ME [µg/g]**	**RMSSE**	**MPE [%]**	**MPEc [%]**	**C**
As	-0.01	1.43	28.5	24.7	0.40
Cd	-0.01	1.32	20.9	16.1	0.71
Cr	-0.08	1.17	25.4	20.5	0.72
Cu	-0.03	1.09	16.8	14.8	0.68
Fe	-17.83	1.35	28.0	25.2	0.62
Hg	0.00	1.77	20.4	17.9	0.64
Ni	-0.03	1.19	25.9	20.9	0.56
Pb	-0.11	1.36	27.6	16.6	0.79
V	-0.07	1.45	26.4	22.7	0.71
Zn	-0.66	1.44	17.5	14.3	0.59

ME Mean error
RMSE Root mean standardised error
MAE Mean absolute error
MPE median percental error
MPEc Median percental error corrected
C Correlation coefficient after Pearson

Table 6: Significant differences across national boundaries related to the analytical methods used in the 1995 and 2000 heavy metals in mosses surveys in Austria (AU), the Czech Republic (CZ), Germany (DE), Italy (IT), Slovakia (SL), Poland (PL), and Switzerland (CH).

		AU/CH	AU/DE	AU/IT	AU/SL	CH/DE	CZ/DE	CZ/PL	PL/DE	CZ/SL	PL/SL*	%
As	1995	■	■	■	▨	■	■	▨	▨	▨	▨	100
	2000	■		■	■	■	■	▨	▨	■	▨	86
Cd	1995				■			■	■	■		40
	2000		■	■	■	■		■	■	■	■	80
Cr	1995		■	■	■	■		■		■	■	70
	2000	■		■	■	■	■			■	■	70
Cu	1995		■	■	■	■	■	■	■	■	■	90
	2000	■		■	■	■		■	■	■	■	80
Fe	1995	■		■		■			■	■	■	60
	2000	■	■		■	■		■		■	■	70
Hg	1995	■	■	■		▨	■	▨	▨	■	▨	83
	2000	■	■	■	■	▨		▨	▨	■	▨	83
Ni	1995			■	■					■	■	40
	2000		■	■	■	■	■		■	■		70
Pb	1995	■	■	■		■	■	■	■	■	■	90
	2000	■	■	■	■	■		■	■	■		80
V	1995	■		■		■	■	▨	▨		▨	57
	2000		■	■	■	■	■	■	■	■	■	90
Zn	1995		■	■	■	■	■	■	■		■	80
	2000	■	■	■	■			■		■	■	70
%		60	65	90	79	83	50	73	71	90	80	

■ Difference significant ($p<0.05$)
(blank) number of values too small
% percentage of detected difference with $p<0.05$
▨ no comparison possible

Table 7: Differences in accumulation of metals in mosses in Germany due to altitude and sea spray.

		as			cd			cr			cu			fe			hg			ni			pb			sb			ti			v			zn		
		90	95	00	90	95	00	90	95	00	90	95	00	90	95	00	90	95	00	90	95	00	90	95	00	90	95	00	90	95	00	90	95	00	90	95	00
Altitude [m]	200	■						■	■		■	■		■	■			■			■	■	■	■	■			■	■			■	■		■	■	■
	500		■	■				■		■		■	■	■	■	■			■					■			■		■	■	■	■			■	■	■
	700							■	■			■	■	■	■	■				■				■					■	■	■	■				■	■
	800							■	■			■	■	■	■	■													■	■	■	■					
	1000							■	■			■	■	■	■						■								■	■	■						
Distance to ocean [km]	10	■		■					■	■		■				■					■	■		■	■		■	■			■						
	30	■						■	■	■				■	■						■	■	■	■	■		■	■	■					■			
	50	■						■	■	■				■	■						■	■	■	■	■			■	■					■			
	80	■	■	■				■	■	■	■			■	■	■					■	■	■	■	■			■	■		■			■			
	100	■	■	■				■	■	■	■	■		■	■	■				■	■	■	■	■	■			■	■	■	■		■	■			
	120	■	■	■				■	■	■	■			■	■	■			■	■	■	■	■	■	■			■	■	■	■		■	■			
	150	■	■	■				■	■	■				■		■			■	■	■	■	■	■	■			■	■	■	■		■	■			

■ Difference in metal loads significant (p<0.01)