Restructuring the long-term moss biomonitoring of atmospheric deposition in Germany

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Abstract

The determination of atmospheric deposition in forests can be accomplished using technical sampling devices (bulk samplers, wet only samplers), biomonitors or modelling. In Europe, since 1990 moss sampled every five years at up to 7300 places in up to 36 countries was used as biomonitor. In the moss specimens, heavy metals (HM), nitrogen (N, since 2005) and persistent organic pollutants (POPs, since 2010) were determined. Germany participated in all surveys with the exception of that in 2010. For the moss survey 2015, the biomonitoring network applied in the 2005 campaign should be reorganized. To this end, a complex statistically based methodology including a decision support system was developed and implemented. Its application yielded a network with a reduction of sample points from 726 to 402. By use of the data collected in 2005 the performance of the reorganized network did not reveal significant loss of statistical validity.

Keywords: Decision support system; Minimum sample size; Spatial sampling design.

1. Introduction

Ectohydric mosses allow long-term monitoring the atmospheric deposition of several elements in the same matrix collected at many sampling sites covering large areas (Amodio *et al*. 2014). They accumulate dry, occult and wet deposition and by that enable the determination of elements far beyond their analytical detection and quantification limits. Therefore, since 1990 every 5 years the European Moss Survey (EMS) has been conducted covering up to 7300 sampling sites in up to 36 countries. In the moss specimens, heavy metals (HM), nitrogen (N, since 2005) and persistent organic pollutants (POPs, since 2010) were determined. As a long-term harmonized monitoring and mapping of background atmospheric deposition in areas remote from emission sources, the EMS enables the evaluation of the effectiveness of environmental policies such as mitigation of emissions (ICP Vegetation 2014). In Germany, the moss specimens were collected at 592 (1990), 1026 (1995), 1028 (2000), and 726 (2005) sites. Currently, Germany participates in the EMS 2015. The chemical analyses include Al, As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, V, Zn, N and POPs to be determined in moss specimens. One aim of the 2015 campaign was to evaluate whether the spatial density of the network applied for could be reduced compared to that in 2005. This article concentrates on this issue aiming at explaining a method enabling to reduce the number of sampling sites compared to previous surveys and the potentially associated loss of spatial validity of the monitoring results as far as possible. Thereby, those criteria should be regarded which were applied when the German moss biomonitoring network was reduced from 1028 in the year 2000 to 727 five years later (Pesch *et al*., 2008). The spatial representativity of the monitoring network should be preserved as well as requirements outlined in the following. Across Europe, the national networks contributing to the EMS should comply with international requirements defined by ICP Vegetation (2014). Some of them are given in the following: As in previous surveys in each country at least 1.5 moss samples / 1000 km^2. For the German territory this would result in 536 sites. If this is not feasible, a sampling density of at least two moss sample sites per EMEP grid (50 km by 50 km) is recommended. In Germany, this would correspond to 404 moss sampling sites which should be regarded as target number for the German moss survey 2015. A denser sampling network is recommended in areas where steep gradients in the deposition of heavy metals are to be expected based on previous surveys. To enable the analysis of temporal trends, it is recommended to collect specimens from the same sites as in the previous surveys. Regarding the determination of POPs, a lower sampling density may be performed due to potential financial limits. To assess a statistically valid number of sampling sites for a given ecoregion / landscape, country or whatever spatial unit, the respective minimum sample sizes should be calculated. Only pleurocarpous mosses should be sampled. The use of bryophytes other than *Hylocomium splendens* or *Pleurozium schreberi* must be preceded by a comparison and calibration of their uptake of heavy metals relative to the main preferred species. Sampling in the field should be conducted according to several criteria, some which being explained in the following: Each sampling point should be situated at least 3 m away from the nearest projected tree canopy, in gaps of forests (diameter > 10 m) or plantations (diameter > 5 m) primarily, without pronounced influence from canopy drip from trees, preferably on the ground or on the surface of decaying stumps. In nitrogen polluted regions with high density of livestock the horizontal distances to tree crowns should be more than 7 m. In habitats such as open heathland, grassland or peatland, sampling below a canopy of shrubs or large-leafed herbs should be avoided, as well as areas with surface run off on slopes. The sampling points should be located at sites representative of non-urban areas of the respective countries. In remote areas the sampling points should be at least 300 m from main roads (highways), villages and industries and at least 100 m away from smaller roads and houses. In order to enable comparison of the data from this survey with previous surveys, moss samples should be collected from the same or nearby, i.e. no more than 2 km away but with the same biotope conditions, sampling points as used in the most recent moss surveys. Sampling of mosses near monitoring stations of atmospheric HM, N or POPs is recommended in order to directly compare their concentration in moss with the accumulated atmospheric bulk deposition. Further, it is recommended to make one composite sample from each sampling point, consisting of five to ten (ten for POPs) subsamples, if possible, collected within an area of about 50 m by 50 m. In the composite sample only one moss species should be represented. Each locality must be given coordinates, preferably longitude and latitude (Greenwich coordinates, 360º system), suitable for common data processing. Based on this network design, the aims of the 2015 survey are to: Characterise the regional atmospheric deposition of HM, N and POPs in Europe; indicate the location of important HM, N and POPs emission sources and the extent of particularly polluted areas; produce maps of the deposition patterns of HM, N and POPs for Europe and analyse spatial trends; provide field-based evidence of the extent of long-range transboundary pollution in Europe; analyse temporal trends to establish the effectiveness of air pollution abatement policies within Europe; determine the effect of canopy drip on the concentration of HM and N in mosses by comparing moss samples from open fields and adjacent forest stands (Meyer *et al*. 2015 a, 2015 b; Skudnik *et al*., 2014). In addition to the above mentioned standard requirements of the EMS (ICP Vegetation, 2014), the Federal Environment Agency defined national requirements for the German moss survey network 2015. The network should be reduced to about 400 sites and, however, should enable the following statistical analyses without significant loss of validity: Multivariate analyses of measurement data and information on characteristic of the sampling sites and their surroundings which could influence the element concentrations in moss (Annex 5 of ICP Vegetation, 2014); correlation with technical deposition measurements (e.g. ICP Forests, ICP Integrated Monitoring, German deposition network); correlation with results from other biomonitoring networks, i.e. HM, N and POPs concentrations in leaves and needles collected in the German Environmental Specimens Bank (Nickel and Schröder, 2017 a); correlation with modelled atmospheric deposition (EMEP and LOTOS-EUROS, Nickel and Schröder, 2017 a); correlation with Critical Loads maps across Germany and for specific protected areas as for instance NATURA 2000.

2. Materials and methods

For operationalising the criteria *network efficiency* and *sufficiency* and those required for realising the objectives at the European spatial scale and at the German scale including administratively and ecologically defined sub-regions (**Section 1**), a multistep approach with an integrated decision support model was developed, implemented and applied for the reorganisation of the German moss survey network. By this, the MSS could be calculated for different spatial categories (territory of Germany as a whole, the German federal states, and ecological land classes covering Germany; **Section 1**) and the sampling density in specific areas (**Section 2.2**) could be determined. Furthermore, the sampling sites for the survey network 2015 could be selected (**Section 2.3**) and its performance tested based on the data collected in 2005 (Section 2.4).

2.1 Data

To cope with the European and German requirements for the moss survey monitoring network and the statistical analyses of the results, the following data sets were used: The potentially available sampling sites are those which were sampled in the campaign 2005/06. They were added by 27 sites located in the German federal state Saxony-Anhalt where moss specimens were sampled in 2010 / 2011 (Metzschker 2016) as well as by 28 sites in North-Western Germany investigated in 2012 / 2013 (Meyer *et al*. 2015 a, 2015 b). For the determination of sites which were sampled in previous surveys and of temporal trends, the comprehensive MossMet database with complete data sets for 1990, 1995, 2000 and 2005 was used. This database was added by geostatistically calculated surface estimations of As-, Cd-, Cr-, Cu-, Fe-, Hg-, Ni-, Pb-, Sb-, Ti-, V-, Zn- and N-concentrations in moss as well as by modelled deposition values of Cd and Pb on a 50 km by 50 km grid for Germany 2005 (EMEP Meteorological Synthesizing Centre – East, MSC-East), for Cd and Pb on a 7 km by 7 km grid for Germany 2007-2011 and for As, Cr, Cu, Ni, V and Zn on a 25 km by 25 km grid for Germany 2009-2011 calculated with LOTOS EUROS (Builtjes *et al*., 2017; Nickel and Schröder, 2017 a; Schaap *et al*., 2008; Schröder *et al*., 2016). This modelled deposition was used to identify regions with pronounced gradients and significant differences between monitoring and modelling data. Further, geographical coordinates and measurement data from several environmental monitoring networks, i.e. the German Environmental Specimens Bank, the German deposition network with is part of the EMEP deposition network, as well as those from ICP Integrated Monitoring and ICP Forests Level II were also included to enable to link and correlate the results. Data on forest ecosystem types in Germany (1:500,000; Schröder *et al*., 2015) as well as data on Ecological Land Classes of Europe covering Germany (ELCE40, 10 km by 10 km Europa, Hornsmann *et al*. 2008) were used for representativity analyses and the calculation of minimum sample sizes (MSS).

2.2 Minimum sample sizes for defined spatial categories

The calculation of the minimum sample sizes (MSS) should assure that the sample yield valid statistical result for different spatial levels of the moss survey in Germany. That means that the empirical mean value should be near the true mean value within a defined tolerance and at a specific probability. The MSS can be calculated with data from proceeding moss survey assuming that the causes for measurement variability also hold true for the survey 2015. The statistical estimation of MSS for Germany as a whole, its single federal states and ecological land classes was performed using an equation published by Hansen *et al*. (2013), ICP Vegetation (2014) and Schröder *et al*. (2016).

For the reorganisation of the monitoring network of the German moss survey 2015 the MSS were calculated for the whole territory of Germany, the federal states and the ecological land classes covering Germany using the data from the 2005 survey (n = 726) for As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, V, Zn and N. The statistical distribution was calculated by use of the test of Shapiro and Wilk (1965) with $\alpha = 0.05$.

2.3 Sample density in hot spot regions of element variability and of differences between HM concentrations in moss and modelled deposition

In addition to the three spatial scales mentioned in Section 2.1, regions with a high variability of HM and N deposition were identified. This was achieved by calculating the relative coefficient of variation (Vr). Vr was calculated for N, Cd, Cr, Hg, and Pb measured in moss 2005 / 06. Regions with Vr > 25 % were identified as hot spots of variation. Additionally, regions with remarkable differences between HM and N concentrations measured in biomonitors and modelled deposition values were identified based on investigations presented by Nickel and Schröder (2017 a). To this end, the HM deposition modelled with help of LOTOS-EUROS (Builtjes *et al*., 2017) and geostatistical surface estimations of HM concentration in moss were compared. Both spatial data were normalised and the deviation of the deposition and moss data for As, Cd, Cr, Cu, Ni, Pb, V, and Zn from the Germany wide median value was calculated. Regions with sites sampled in 2005 exceeding the mean difference by more than 25 % were stipulated for a higher sampling density in the 2015 survey. From modelled N deposition (Wichink-Kruit *et al*. 2014) a spatial consolidation was derived for southern Germany due to windward- / leeward-effects in mountainous regions.

2.4 Sample site selection for the moss survey network 2015

The sample site selection relies on the data introduced in **Section 2.1 and on a set of criteria used to confirm or remove former sampling** sites. The first step was to select out of the survey net 2005 sampling sites with a distance less than 5 km to sites of the German Environmental Specimens Bank, to those of the ICP Forests Level II (Seidling *et al*. 2014: Fischer *et al*. 2006), ICP Integrated Monitoring (ICP-IM, Kleemola and Forsius 2006) and the German deposition network. Furthermore, such moss survey sites were added lying in a distance of 5-10 km apart from the aforementioned environmental networks and being assigned to the same ecological land class (ELCE 40, Hornsmann *et al*. 2008) or the same ecosystem type (Schröder *et al*. 2015) were included into the moss survey network, too. The 28 sites sampled in 2011 in the federal state Saxony-Anhalt as well as seven sites already selected by the authorities of the federal state were also integrated into the moss survey network 2015. Finally, 25 sites in North-western Germany already sampled in 2012 and 2013 were selected for the moss monitoring net 2015 (Meyer *et al*. 2015 a, 2015 b). The latter sites include both open land sampling locations as well as such below canopies and intermediate ones. The second step aimed at thinning out the remaining sites of the survey network 2005 by about 50 %. To this end the spatial distances between the sites selected in the first step and the residual ones was calculated and a list of site pairs in the whole network derived. Based on this list, the following criteria were analysed: Number of preceding surveys at the same site in 1990, 1995, 2000 and 2005; former occurrence of *Pleurozium schreberi*; open land site character. These three criteria were aggregated in a decision matrix and applied in numerous combinations of weighting from 0 to 100 % to the pairs of sampling site. Thereby, the sampling pairs were structured according to increasing and decreasing spatial distance as well as by random. Accordingly, the thinning out of the remaining sites was conducted iteratively by a decision support model implemented in R (R Core Team, 2013), so that from each site pair the worse one in terms of multidimensional evaluation was eliminated: A) No site selected according to the first step criteria was dropped. B) For each EMEP grid at least one site should remain. C) To scale down the spatial pattern of sampling sites´ density of the 2015 survey network compared to that in 2005, a factor f applicable on the distances of the site pairs and below which no further thinning out should be done was introduced. The f values range between 1.5 and 2.5. D) For regions where a higher sampling density should be realised, i.e. regions with high spatial variability of deposition / accumulation and regions with remarkable differences (low or negative correlations) between moss concentrations and modelled deposition, maximum distances between sampling sites were decided for (15-35 km). E) MSS to be reached were defined according to the respective calculations explained in **Section 2.2**. For running the decision model, six versions of MSS were defined specifically for the federal states and the ecological land classes. MSS1-MSS4 were designed according to the results given in tables 3 and 4, calculating minima and maxima for different element groups and MSS5 was run without confinements. The federal state- and, respectively, ELCE-specific MSS1 are based on the means of the 12 element-specific MSS. The two MSS2 were derived from the MSS means for Cd, Hg, N, and Pb (priority elements). For Cd, Hg, and N, MSS maxima were taken to define MSS3. MSS4 relies on the maxima for Cd, Hg, N, Pb. In principle, the algorithm uses the MSS scenarios as constraints, i.e. does not remove any site below the threshold. MSS1-MSS5 were then applied and the results were evaluated according to **Section 2.4**. The best variant was used for refinements of the constraints defined in MSS6. For the sample site selection the manifold combination of the above mentioned model parameters enabled a broad range of variation. The weight factors for the number of preceding surveys, former occurrence of *Pleurozium schreberi* and open land site character were set to 0, 50 and 100 %. The f value was set to 1.5, 2.0 and 2.5 and the maximum distances to 15, 25 and 35 km. MSS1-5 and various MSS6 variants were applied and target sample size was set to n = 400. The results from more than 100 model runs were then analysed according to **Section 2.4** Finally, the model-based site selection with the highest degrees of fulfilment were refined through application of expert judgement based on criteria not included into the model (e. g. amendments of single sites in hot spot regions of pollution led to stronger geostatistical validity).

2.5 Evaluation of the survey networks 2005 and 2015

The evaluation of the reorganised moss survey network 2015 was performed by calculation of following statistical numbers: sample size, minimum of distances between the sampling sites [m]; percentage of open field sites [%]; mean number of sampling throughout the years 1990, 1995, 2000 and 2005; percentage of sites with occurrence of *Pleurozium schreberi* [%]; deficit for land class-specific MSS for As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, V, Zn, and N as defined by the proportion of elements with MSS reached [%]; federal states-specific of MSS for As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, V, Zn, and N as defined by the proportion of elements with MSS reached [%]; number of EMEP grids with one or zero moss sampling site; median values of concentrations As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, Ti, V, Zn, and N in Germany. The latter is of high importance in particular for temporal trend analysis. Medians of the sample 2005/06 should not be statistically different from the subsample used for the survey 2015/16. These statistical numbers were determined with continuously decreased sample size and documented as graphs to help evaluating the quality of the monitoring network. Additionally, the HM and N concentrations derived from the complete network 2005 with n = 726 sites were compared with the reduced network 2015 with n = 350, 400 and 450 sampling sites. To this end, for the German territory, federal states and ecological land classes minimum, maximum and percentiles (20, 50, 90) of HM and N concentrations in moss were compared for the 2005 vs. the 2015 network. The differences between the complete network (2005) and the reduced network (2015) were tested for statistical significance with $p > 0.05$ using the Mann-Whitney U-Test. Furthermore, the MSS was compared to the realised number of sampling sites for the complete network (2005) and the reduced one (2015) on three spatial levels: Germany as a whole, federal states, and ecological land classes. Then, the geostatistical validity of the complete and reduced survey samples (2005, 2015) were investigated in terms of spatial autocorrelation using variogram analysis (spatial range, nugget / sill ratio) and surface estimations via Kriging interpolation and mapping (Johnston *et al*. 2001). Finally, the results of geostatistical surface estimation were compared to the site-specific measurements with regard to the minimum, maximum, percentiles (20, 50, 90) and to statistical significance of differences (Mann-Whitney U-Test). These statistical analyses and Kriging interpolations were implemented in and run with R (R Core Team 2013).

3. Results

A total of 402 sampling sites were determined (figure 1). They include 152 sites according to the selection criteria and the thinning out explained in **Section 2.4** The network comprises sampling sites in most of the federal states of Germany: Baden-Württemberg (n = 30), Bavaria (n = 60), Brandenburg (n = 28), Hamburg (n = 3), Hesse (n = 28), Mecklenburg-West Pomerania (n = 24), Lower Saxony (n = 57), North Rhine- Westphalia (n = 48), Rhineland-Palatinate (n = 19), Saarland (n = 7), Saxony (n = 29), Saxony-Anhalt (n = 30), Schleswig-Holstein (n = 20), and Thuringia (n = 19). They include 78 ICP Forests Level II sites, 13 locations of the German Environmental Specimens Bank (two of them representing agrarian ecosystems, one forests, six ecosystems nearby urban agglomerations, and four near natural terrestrial ecosystems), five stations of the German deposition network (Schauinsland, Schmücke, Waldhof, Westerland, Zingst) as well as two sites of the ICP Integrated Monitoring (Forellenbach, Neuglobsow). This enables the correlation of moss data with technical deposition sampling and other biomonitoring networks. For the optimized model run to decrease the residual number of monitoring sites the site pairs were arranged according to increasing spatial distance. The criterion “occurrence of *Pleurozium schreberi* in preceding surveys” was weighted with 100, the “number of preceding moss samplings at that site” and “open land character” with 0 since this weighting yielded better results with regard to other more crucial criteria like measurement variance, MSS and geostatistical validity. The maximum distance between sampling sites was set to 20 km and the thinning out factor to f = 1.5. With regard to MSS and a target sample size of 400, the user defined MSS6 (table 1) as a refinement of MMS3 (ELCE 40) and MSS4 (federal states) was used, whereby the loss of subsamples with complied MSS was rather avoided for the ELCE 40 categories than for the federal states. Further refinement of modelling results was reached by enhancing the spatial density of the sampling sites in hot spots of deposition at the expense of other sites especially at the borders of Germany. By that, the geostatistical validity of the moss survey net could be improved. The 402 sites of the moss survey net 2005 contain 170 locations (= 42 %) with samplings in 1990, 1995, 2000, and 2005, 121 (= 30 %) with three preceding moss collections, 61 (= 15 %) with two and 35 (= 9 %) with one former sampling. In the survey 2005, *Pleurozium schreberi* was collected at 202 sites (= 50 %), *Hypnum cupressiforme* at 86 (= 21 %), and *Scleropodium purum* at 112 (= 28 %) locations. At two sites, other moss species were found. However, these two sites were included due to their small distance to stations of other relevant monitoring programs. Furthermore, 25 sites sampled in North-western Germany in 2012 and 2013 to explore the canopy drip effect were integrated (Meyer *et al*., 2015 a, 2015 b). At two additional sites, one of them with high and one with low deposition loads, each three single samples were collected and not pooled to investigate the variance due to the sampling procedure. Finally, eight sites were identified for specific POP-sampling. They were situated nearby (max. 2.4 km, mean 832 m) the sites of the German Environmental Specimens Bank where POPs are analysed and the ecosystem types there, and therefore enabling correlations POP concentrations in moss and other biomonitors. The German moss survey network 2015 encompasses 402 sampling sites, i.e. it was reduced by 45 % compared to the net applied in 2005. The mean value of preceding surveys was reduced by 5 % from 3.16 to 3.0. The percentage of *Pleurozium schreberi* was increase from 44 to 50 %. While in 2005 the MSS for ecological land classes was 54 %, it was reduced to 45 % in the network for 2015. The MSS were reached in 59 % of the federal states in 2015, whilst the respective number in 2005 was 72 %. 56 of the 202 EMEP-Grids in Germany encompassed one moss sampling site compared to 19 in 2005. Three EMEP grids do not contain any moss sampling sites however they do cover the German territory only partly. As proved by application of the Mann. Whitney U-Test, the median values of the concentrations of As, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Sb, V, Zn and N did not differ significantly between the 726 locations sampled in 2005 and the reduced network which should be sampled in 2015 applied to the measurements taken in 2005.

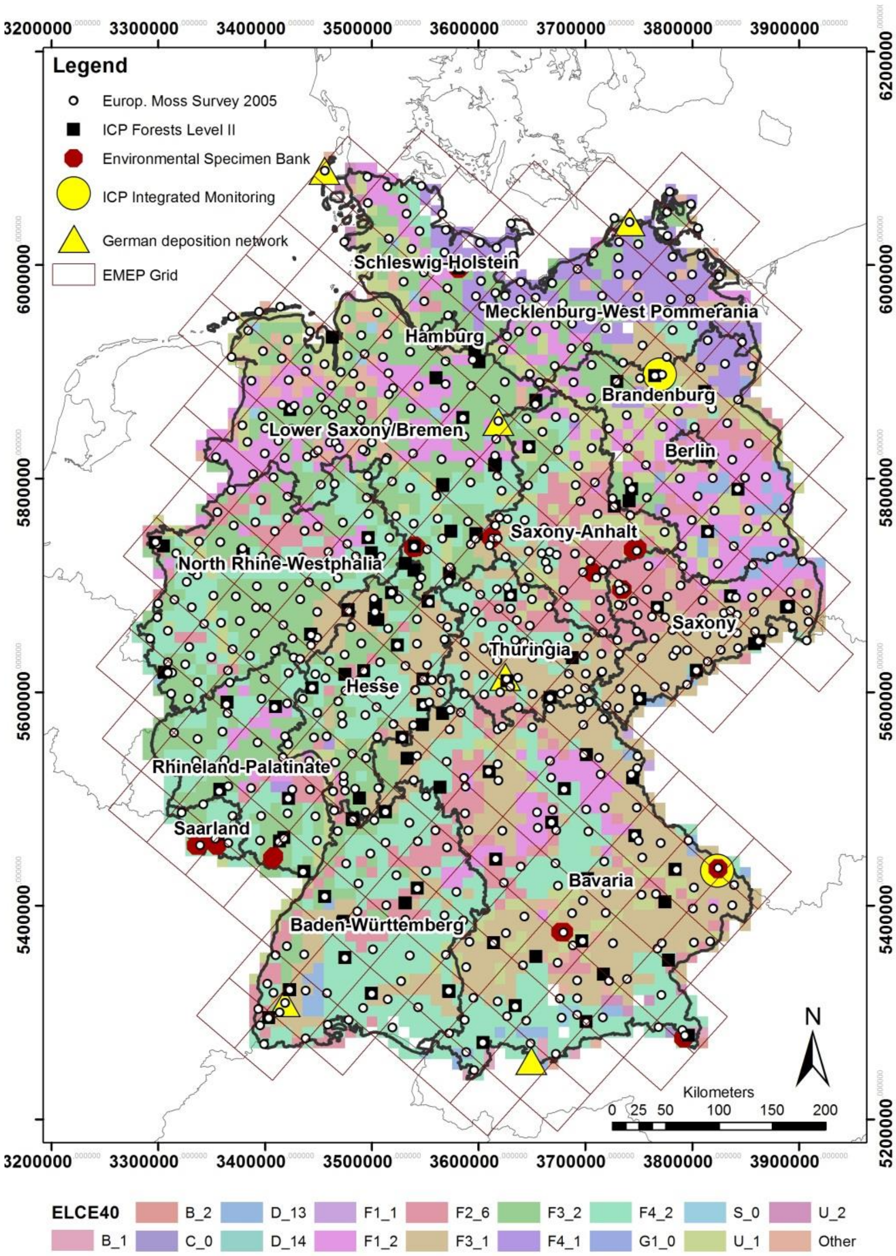

Figure 1. Spatial distribution of sampling sites from the European Moss Survey 2015

This could be proved for the territory of Germany (table 5) as well as for its federal states (supplementary table S1) and its ecological land classes (supplementary table S2). By that could be proved, that the reorganisation of the moss survey monitoring does not change significantly the elementary descriptive statistical characteristics of the German moss survey. The same conclusion can be drawn from the geostatistical analyses. Accordingly, the geostatistical validity of the monitoring network 2015 could be corroborated (supplementary figures S1-S4). Calculated from the thinned out network 2015, the strength of spatial autocorrelation in terms of the nugget / sill ratio is for nine out of twelve elements as high as or higher than derived by the full sample 2005. For six out of twelve elements, the percentile statistics derived from the geostatistical surface estimations are statistical significant. The MSS for the Partial sample 2005 and comparisons with the full sample 2005 reveal that a further reduction of the sample size would not yield any statistical improvement but a reduction of statistical validity. The mapping of ecological land classes and federal states, complying with or not achieving the MSS, identified deficits for Cd and Pb especially in Schleswig-Holstein, Thüringia, Baden-Württemberg, and partly in Saxony, Hesse and Rhineland-Palatinate.

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