

Mapping of ferrimagnetic susceptibility for screening of fly ash deposition

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Abstract

This article presents a case study in the industrial triangle Leipzig-Halle-Bitterfeld, the purpose of which was to assess the actual fly ash load in forest soils and to test if ferrimagnetic susceptibility can be used for a fast and cost efficient screening of deposited elements. Ferrimagnetic susceptibility was mapped in a raster of 1x1 km² and correlated with key nutrients, selected metals/heavy metals and Black Carbon. The predictive value of magnetic susceptibility was tested on the basis of linear regression models. Furthermore, multiple-regionalization techniques were used to model the spatial variation of fly ash. This includes an analysis of which environmental parameters are most important for the spatial model. The correlation between ferrimagnetic susceptibility, base saturation and the contents in Ca, Mg, Fe, Al and Cd (humus layers) was comparably high. The correlation with the content in Mn was weaker and the correlation with Black Carbon (humus layers) showed no clear trend. Linear regression based models with sufficient precision could be found for Ca, Mg and Mn, with lower precision for Cd and Black Carbon. No prediction was possible for Fe and Al. Multiple regression based modelling of the spatial variation of fly ash deposition was possible with a very high precision. A slightly differing set of model parameters was selected for different depth levels in the humus layer and mineral soil, comprising topographical and soil parameters and to a much lesser extent stand parameters. In conclusion, the usability of the proxy indicator ferrimagnetic susceptibility for screening of the deposited elements was proved.

Keywords: fly ash deposition, proxy indicator, ferrimagnetic susceptibility, linear regression models, spatial modelling, multiple regression based models.



1 Introduction

Forest ecosystems in Eastern Germany are still affected by long-term effects from former fly ash deposition caused by unfiltered lignite combustion in Czech, Polish, and German power plants until the early 1990s. Fly ash is defined as particle residue that enters the flue gas stream after lignite (brown coal) or hard coal (black coal) combustion. Observed deposition rates of the most affected regions in Eastern Germany and Eastern Europe range from 140 t / km² x a (industrial triangle Leipzig-Halle-Bitterfeld, Northeastern Germany) up to 457 t / km² x a (Upper Silesia, Poland) (Lux [19, 20], Lux and Stein [22], Strzyszcz et al. [29], Strzyszcz and Magiera [28], Klose and Makeschin [12]). In the industrial triangle Leipzig-Halle-Bitterfeld, an intensive industrialization has been taking place for almost 100 years and substantial deposition amounts have resulted, particularly from lignite combustion for energy production. The estimated deposition in the most important regional forest ecosystem, Dübener Heide, from 1910 to 2000 is 18 Mio t fly ash and 12 Mio t SO₂.

Based on the recognition of a dependence of fly ash deposition effects in Dübener Heide from the distance to the power plants in Leipzig-Halle-Bitterfeld, Lux [20] highlighted the necessity of defining deposition zones (planning units with homogeneous deposition impact) for an adapted forest ecosystem management. Enderlein and Stein [3], Lux and Pelz [21] and Lux [19] developed a sample plot based approach, where a classification of health state and growth potential in medium-aged Scots pine stands was used for a regionalization of deposition impact.

The degree of single tree growth reduction and needle losses were aggregated plot-wise to a factor, which indicated the intensity of damage. Based on the sample plot results, four deposition zones were defined, where deposition zone-specific silvicultural management measures were derived and delivered spatial information on additional costs and economic losses caused by fly ash deposition.

Impacts of fly ash deposition on chemical humus and soil properties are still evident (Fritz et al. [5], Fritz and Makeschin [6], Fürst et al. [7–9], Klose et al. [13], Klose and Makeschin [11], Koch et al. [15], Koch et al. [14]). However, the former forest growth and decline based definition of deposition zones cannot be used anymore due to considerable changes in regional forests health and species composition since the first inventories of Enderlein and Stein [3] and Lux [20]. In consequence, to derive management relevant information for today's forest management, an approach had to be developed to come to spatial information on the actual fly ash deposition loads in the regional forest soils. Therefore, the aim of this study was to assess the actual fly ash load in forest soils and to test if ferrimagnetic susceptibility can be used for a fast and cost efficient screening of formerly deposited elements.



2 Material and methods

2.1 Fly ash assessment approach

Magnetic susceptibility field assessment was carried out in Dübener Heide in a regular 1x1 km² grid (110 plots). The grid-wise measurements were done to map with high resolution the spatial variation of magnetic susceptibility and to test if the formerly observed distant dependent deposition gradient can be validated.

Magnetic susceptibility was measured with the MS2 meter susceptibility system of Bartington Instruments. The system is developed for detecting very low quantities of magnetic Fe-Oxides in compact (rocks) or loose substrates (mineral soil, humus layer). The susceptibility meter has a sensitivity of $0.1 - 1 \times 10^{-5}$ SI units and can be used in a single readout mode or transfers the measured values to a PC, where the data can be processed with the software Multisus (© Bartington). The Multisus program is using the Windows 3.1 or Windows 95/NT interface to record the magnetic susceptibility measurements of different field assessment sensors. The program allows for saving as file the results from a batch of individual samples or from a core. For single samples the results can be volume or mass specific and provision is made for automatic increments of depth for core measurements (Operation Manual Multisus 2.0, Bartington Inst. Ltd.).

The MS2 meter susceptibility system comprises a portable measuring instrument, the MS2 meter, and a variety of sensors. The meter displays the magnetic susceptibility value of the tested substrates when these are brought within the influence of one of the sensors, which are each designed for a specific application and sample type (Operation Manual MS2 system, Bartington Inst. Ltd.). At the field assessment, volume magnetic susceptibility was measured centimetre wise in 30 cm deep boreholes with the MS2H down-hole-probe sensor. The MS2H is a sub-surface probe for profiling the magnetic susceptibility of zones in 25 mm nominal diameter auger holes. Zones with a thickness down to 15 mm can be discriminated. The starting point 1 of the measurements was defined as first measurement after removing the litter (Oi). The Oi was removed, as a pre-study has shown that fly ash particles could never be detected in this layer (Fürst et al. [7]).

2.2 Correlation with fly ash components

When assessing ferrimagnetic susceptibility, a major question was, if the magnetic signal allows for concluding on the content of base cations, acid and heavy metal cations and Black Carbon as ecologically relevant components of fly ash deposition. Therefore, a test series on the correlation between magnetic susceptibility and the Ca, Mg, Fe, Mn, Al, Cd and Black Carbon content was carried out at 34 research plots, which are partially a subset of the above described 110 plots:

A. “ENFORCHANGE plots”: a subset of 12 plots origins from a regional research project called ENFORCHANGE (www.enforchange.de). Magnetic

susceptibility was assessed in situ (volume magnetic susceptibility) at the locations, where the chemical soil samples were taken in the profiles. A restriction for statistical analysis was the limited number of plots, which was predefined by the project frame. To widen the data basis for spatial trend analysis, plot collective B was included in the study.

B. “Monitoring plots”: 20 plots from Level-I monitoring and 2 other plots from a prevailing study were included. The precondition for their selection was the availability of soil chemical data, which were assessed according to the same standard as at the ENFORCHANGE plots (BMELV [1]). The Level-I plots belong to a European wide network of 6,000 soil monitoring plots for the assessment of long range transboundary air pollution with regular assessment of soil chemical values each five years. The two other monitoring plots were part of a habilitation thesis (Lorz [17]).

Additional laboratory magnetic susceptibility measurements were carried out for soil samples of plot collectives A and B. On this basis, field assessed volume magnetic susceptibility was converted into mass magnetic susceptibility by use of specific correction factors for humus layer and upper mineral soil. Hereby, it became possible to correlate the mass susceptibility ($\chi \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$) with contents of Ca, Mg, Fe, Mn, Al and Cd, which were assessed at the plot collectives a and B according to the Level-I standard procedure (BMELV [1], Fritz and Makeschin [6]) and with Black Carbon, which was assessed in the frame of a diploma thesis (Koschke et al. [16]). The correlation with Cd and Black Carbon was restricted to the humus horizons Oe and Oa. Furthermore, magnetic susceptibility was correlated with regionalized values of the base saturation from the ENFORCHANGE and Level-I plots. This was done to get further information on the spatial variance of magnetic susceptibility in dependence from the distance to the fly ash emitters.

2.3 Fly ash regionalization

The aim of the regionalization was among others to explain the spatial variation of the response variable magnetic susceptibility by auxiliary variables, which are characterized by a pertinent correlation with the response variable (Zirlewagen and von Wilpert [33]) and which are available in digital form. For the regionalization of magnetic susceptibility by use of a stepwise multiple regression approach in combination with geo-kriging, mean values of magnetic susceptibility measurements at three depth levels in the $1 \times 1 \text{ km}^2$ raster were used: Depth level 6–10 cm represents the zone in the humus layer, where in average, the highest magnetic susceptibility values were observed (Fürst et al. [7, 8]). For this zone, biased measurements can be excluded which can occur at the interface between airspace and humus layer (depth level 0–5 cm) due to technical particularities of the sensor. In addition, the likelihood of an impact of admixed particles from the mineral soil is low. Depth level 11–15 cm is situated in the transition zone between humus layer and upper mineral soil, which is characterized by great local variability of humus content in mineral soil and vice

versa due to bioturbation caused by wild boars. In most cases, a sharp border between humus layer and upper mineral soil does not exist. In this zone, an increased magnetic susceptibility is observed. Depth level 21–25 cm represents the local background value spectrum for the mineral soil as reference for the height of the magnetic susceptibility signal. A possible falsification of the measurements due to organic material, which can drop down into the bore hole when taking out the auger, can more or less be excluded in this zone.

3 Results

3.1 Magnetic susceptibility ranges

The observed magnetic susceptibility values from the field assessment (volume susceptibility) ranged in the humus layers from 0 up to 565 SI units $\times 10^{-5}$. Calculated as mass susceptibility, single outliers reached values of up to 800 $\chi \cdot 10^{-8} \text{ m}^3 \text{ kg}^{-1}$. In comparison, the regional background values, which were observed at the C(w) horizons vary between 0 (Podzol) and 10 – 20 SI units $\times 10^{-5}$ (Eutric Cambisols) or expressed as mass susceptibility, a natural background value range of 3–30 $\chi \cdot 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ was found. Considering the spatial variance of magnetic susceptibility, the highest single values were observed at the Western part of Dübener Heide, which was situated nearest and in the major regional wind direction to the former power plants. Here, also the broadest variability of the measured values was observed, which is supported by results of a pre-study (Fürst et al. [7]). The lowest values and the lowest variability were observed in the Northeastern part of Dübener Heide, which is situated farthest from the power plants. Within the bore holes, the highest mean values were achieved in the humus layers, in a depth of 8 and 9 cm. The highest variability of the measured values was observed in a depth from 10 to 12 cm. The lowest mean values and the lowest variability were found in the upper humus layer from 1 to 4 cm depth and in the mineral soil from 22 cm depth on.

3.2 Indicative value of ferrimagnetic susceptibility assessment for site potentials and risks

In trend, the correlation expressed by Pearson's correlation coefficient (r) between mass susceptibility and the selected cations and Black Carbon was higher in the humus layer compared to the mineral soil. Within the humus layer, correlation for Oa was higher than for Oe. The study revealed some problems using different plot types, where the assessment of magnetic susceptibility and the chemical characteristics were not in any case harmonized. The correlation between mass susceptibility and the Ca and Mg content was in trend higher at the ENFORCHANGE plots except for the Mg content at the Oe horizon. Taking the Fe, Al, Mn and Cd content, the results were vice versa. In addition, some trends were vice versa: the correlation with Fe and Al (with exception of Oa in case of Fe) was negative for all horizons at the ENFORCHANGE plots. At plot



collective B this came only true for Fe at the mineral soil horizon 3 (21–30 cm) and for Al at all three mineral horizons. The correlation with Black Carbon was negative for all plot types. When analyzing the distance dependence of the correlation, no spatial trend could be found for all elements, neither for Pearson's correlation coefficient, nor for the variability of the measurements expressed by the standard error. The accuracy of predicting soil chemical characteristics was tested by using linear regression equations. This step was restricted to humus horizons Oe and Oa as they were more intensively and directly impacted by fly ash deposition than the (upper) mineral soil. A linear regression model with comparably high coefficients of determination (R^2) could be derived for Ca (Fig. 1), Mg and Mn. The R^2 for Ca amounted to 0.51, for Mg to 0.52 and for Mn to 0.37. In all three cases, also small 95% confidence intervals were observed. This indicates a sufficient precision of the linear regression model. In contrast, the linear regression was not so clear for Cd and Black Carbon. In consequence, the R^2 values were lower and amounted in both cases to 0.09 and also the 95% confidence intervals were broader. Absolutely no linear regression could be calculated for Fe and Al. In both cases the R^2 values were approximately 0 and the 95% confidence intervals became very broad.

The model quality test by using the relation between measured and predicted values and the residuals revealed a visible coherence between measured and predicted values for Ca (Fig. 2), Mg and Mn with small 95% intervals. The model quality test for Cd and Black Carbon showed a slight coherence with broader 95% intervals. In contrast, and supporting the previous findings, no such coherence could be found for Fe and Al and the 95% intervals became very broad. Considering the residual histograms, these were slightly right skewed for

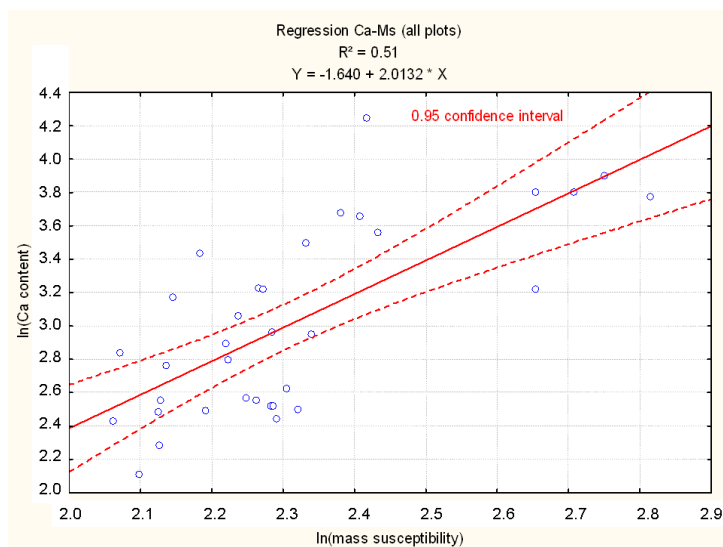


Figure 1: Ca content as linear regression function of magnetic susceptibility.

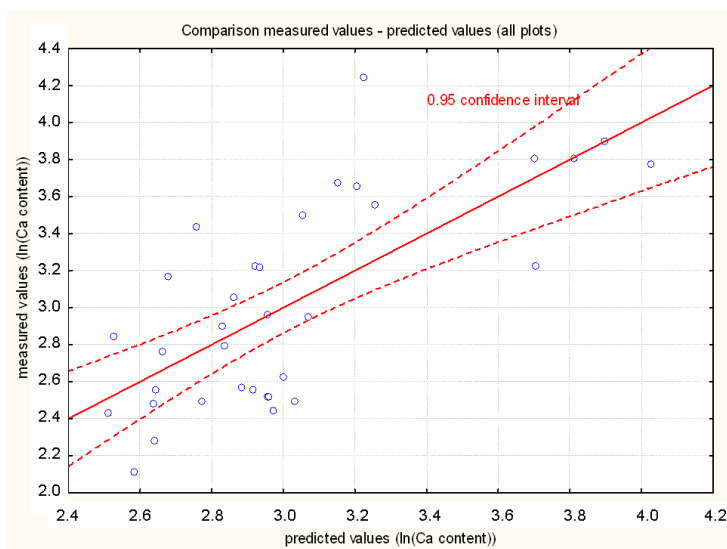


Figure 2: Comparison of measured and predicted values for Ca.

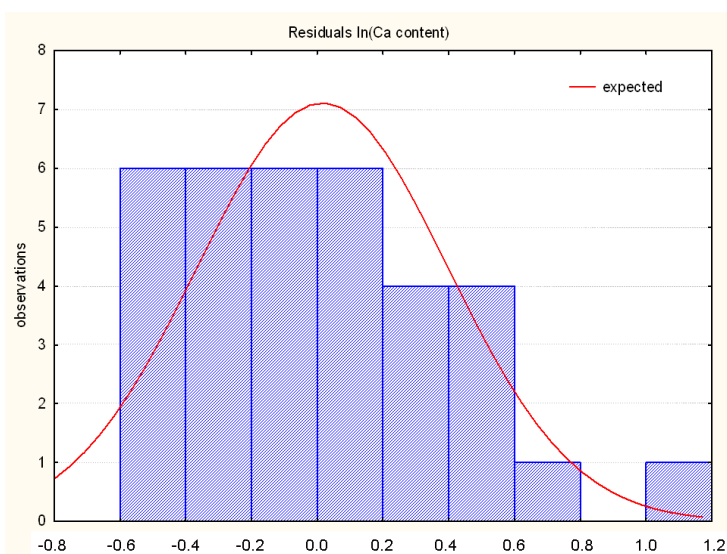


Figure 3: Histogram of residuals for Ca.

Ca (Fig. 3), Mg and Mn and a good coherence with the expected distribution of the observations was given. Also for Cd, the distribution corresponded very well to a standardized normal distribution. For Fe and Al the distribution of the residuals was left skewed and did not fit very well together with the expected distribution. The same applied for Black Carbon.

A stratification of the regression models into distance clusters of < 10 , $10-20$, $20-50$ and $50-75$ and > 75 km distance to the power plants resulted in an improvement of the R^2 of Fe and Al to 0.05 and 0.06, respectively. In addition, the coherence between measured and predicted values and the distribution of the residuals were improved. However, the sample sizes within each of the distance clusters are very small and in the consequence the 95% confidence intervals became very broad and let doubt about the quality of respective models. Contradictory results were obtained by a stratification of the regression models into plot collectives A and B. Taking Fe as an example, a clear linear regression model could be calculated for plot collective A with $R^2 = 0.43$. However, the 95% confidence interval became very broad due to the low number of plots. In contrast, the linear regression for plot collective B was much less clear and the R^2 amounted to 0.12. In this case, the 95% interval was smaller due to the higher number of plots. In both cases, the distribution of the residuals corresponded even less to a standardized normal distribution. The trends for Al were similar.

3.3 Regionalization of fly ash deposition

As a basis for the spatial model, a total of 21 auxiliary variables were identified in a stepwise selection process, which includes a global modelling approach for the whole area of Dübener Heide and a stratified modelling approach for the near distance zone up to 25 km. The spatial variation of magnetic susceptibility was predicted with high precision by a multiple linear regression model (Fürst et al. [8]). The use of a slightly differing set of model parameters for the different depth levels according to their explanatory value improved the prediction quality considerably and supported also the understanding of major drivers for magnetic particle deposition, storage, and vertical displacement in the forest soils.

For depth level 6-11 cm, the horizontal distance to the regionally most important industrial site Bitterfeld and the soil type (Podzol, semi-terrestrial sites) were the most important variables. They indicate slowed-down humus dynamics, which supports the accumulation of fly ash in the humus layer. For the depth level 11-15 cm, variables gain in importance, which describe the exposure (aspect) to major wind direction and thus indicate the probability of deposition. For depth level 21-25 cm, aspect and especially stand properties are most important. The latter give indication of the intensity of deposition driven by the variable combing-out effects. Consequently, the variables “coniferous” and “mixed” stands were highly relevant for the model.

As result of the regionalization, it was possible to identify strata with more or less comparable height of the magnetic signal. This is especially evident for depth level 6-10 cm (Fig. 4) and becomes less pronounced with increasing depth (Figs. 5 and 6). By using parameters for topography, soil type and forest stand type parameters, also the spatial variability within the strata can be modelled. This provides more detailed information for forest management planning than a simple zoning as proposed by Lux [20]. A further improvement in the representation of small scale variations of the magnetic signal was possible by using a stratified modelling approach (Fig. 7).

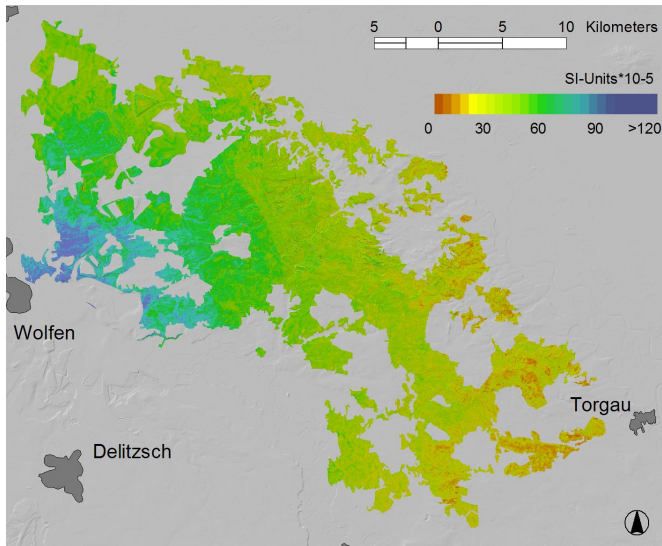


Figure 4: Spatial variability of magnetic susceptibility in the global model for depth level 6–10 cm.

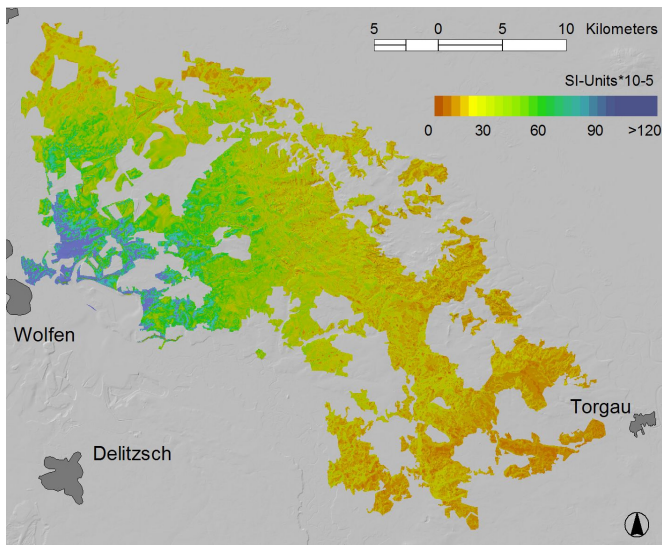


Figure 5: Spatial variability of magnetic susceptibility in the global model for depth level 11–15 cm.

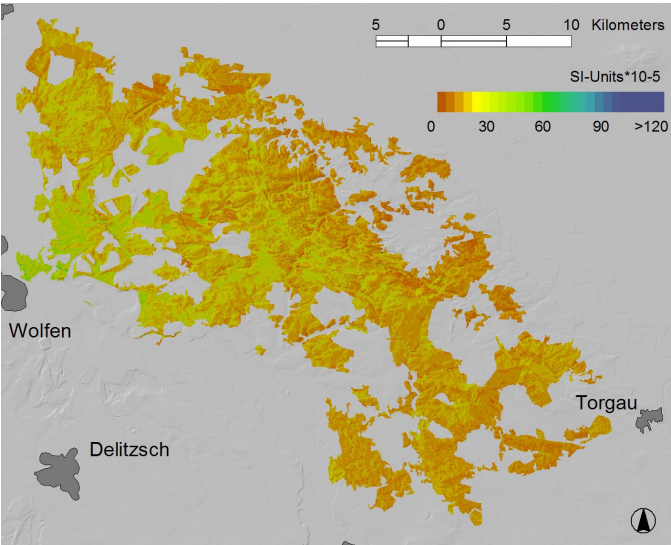


Figure 6: Spatial variability of magnetic susceptibility in the global model for depth level 21–25 cm.

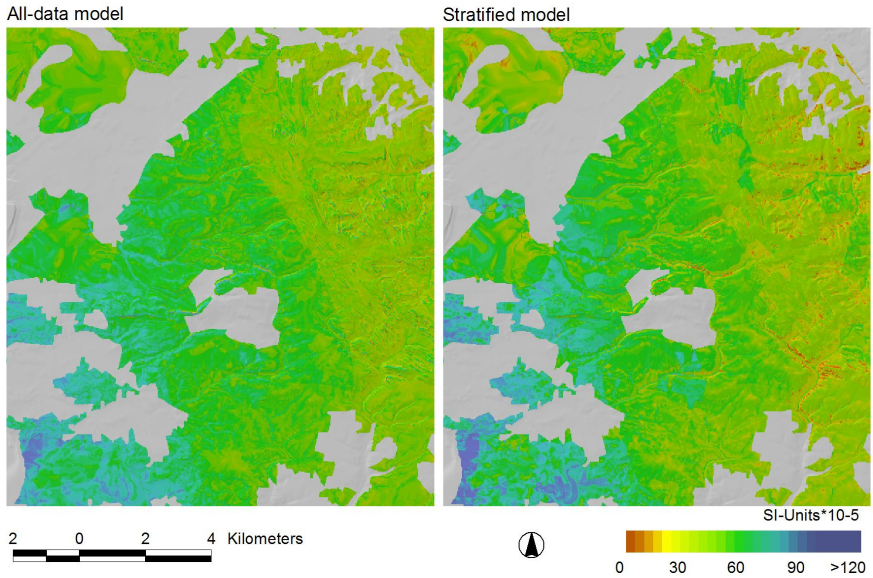


Figure 7: Zoom-in into differences between global (“all data”) and a stratified model considering high resolution information on small scale differences in magnetic susceptibility for the depth level 6–10 cm.

4 Discussion and conclusions

The use of ferrimagnetic field assessment to detect fly ash deposition was successfully tested and approved by numerous studies (e.g. Boyko et al. [2], Grimley et al. [10], Magiera et al. [24], Magiera and Strzyszc [25], Magiera and Zawadzki [23], Schibler et al. [27]). The major motivation of these studies was to get information on the spatial distribution of deposited fly ash. Magnetic susceptibility can also be easily correlated with a number of metals, especially Fe, Al, Mn and heavy metals (Lu and Bai [18], Magiera and Zawadzki [23], Wang and Qin [31], Zawadzki et al. [32]). The correlation varies in dependence from geographical origin, type of combustion material (lignite or hard coal) and land use type and cannot easily be transferred from one test region to another (Fialova et al. [4], Magiera and Zawadzki [23]).

Within the frame of the presented study, the correlation with Ca, Mg, Fe, Al, Mn, Cd and Black Carbon were tested. It was possible to develop a linear regression based model for predicting the content of Ca and Mg as important nutrients with ferrimagnetic susceptibility as model parameter. However, taking the relation between magnetic susceptibility and the Fe content as an example, some results were contradictory to the findings of other studies. A major impact factor might be the use of several plot types where the assessment of ferrimagnetic susceptibility and the chemical characteristics were not always well synchronized. Furthermore, mass susceptibility was not directly measured, but calculated by correction factors for humus layer and mineral soil. These correction factors can only describe an average correlation between field assessed volume and laboratory assessed mass susceptibility. In contrast to our results, Wang [30] found a very high and positive correlation between Black Carbon and mass susceptibility. Here, the applied analysis method and the hereby isolated part of the Black Carbon combustion continuum (Masiello [26]) might be the major impact factor. In addition, a multiple regression based approach might have been brought better results than the applied linear regression based approach.

Our study revealed that different sets of model parameters must be chosen to predict magnetic susceptibility at different depth levels. The different model parameters support the understanding of the deposition and accumulation process for different depth levels. Soil type related parameters such as "Podzol" and "Semi-terrestrial sites", which were very relevant for the depth level 6-10 cm (humus layer), indicate e.g. a slowed down humus dynamics, which supports a long-term accumulation of fly ash (Magiera and Zawadzki [23]). Topographical parameters such as "Streampower index" or "Slope length factor" were also relevant for the depth level 6-10 cm and can be explained with regard to their indication of humus accumulation or erosion. Stand properties play a minor role in the model for depth level 6-10 cm, but became more relevant for depth level 11-15 cm (transition zone between humus layer and mineral soil) and especially for depth level 21-25 cm (mineral soil). Probably, stand type impacts on the findings for the humus layer are widely superposed by soil type and orographic parameters, which decide upon humus dynamics. For depth level 11-15 cm,



variables describing orographic conditions were important. In addition, the divergence from western aspect was of high importance, which indicates the exposure against the major wind direction and thus the probability of deposition. In addition, stand properties (mixed forest) contribute to the model for this depth layer, though their explanatory value is lower compared to their importance for the model in depth level 21-25 cm. For depth level 21-25 cm (mineral soil), the importance of variables indicating soil and site properties is not existent anymore. Aspect and land surface characteristics, which indicate the deposition probability, play a major role together with stand properties. Here, the classes mixed forests and coniferous forests from Corine Landcover 1990 and 2000 were selected as model parameters. They can be considered as indicators for the probability that deposition was combed out by the crown layer. Coniferous and mixed stands have a higher surface roughness of the crown layer compared to deciduous stands and furthermore, the combing out effect of conifers in mixed or pure stands is extended to the whole year and not limited to the vegetation period compared to pure deciduous stands. The question is raised, why the stand type is not relevant for the model for the humus layer. For depth level 21-25 cm, the stand type might indicate a vertical displacement of magnetic iron complexes together with sesquioxides and humus complexes by initial podzolization processes. This is supported by the findings that (a) only coniferous or mixed types show an explanatory value and not deciduous types and that (b) the older Corine Landcover classification from 1990 contributes to the modelling in this depth layer and not the classification of 2000. In addition, the precipitation amount from 1971 to 2000 contributed to the model in the mineral soil. This might be in agreement with the hypothesis formulated before: locally higher precipitation amounts can support podzolization processes.

The grid-wise assessment and multiple-regression based modelling of magnetic susceptibility allowed for a complex spatial model for fly ash deposition under consideration of further environmental influence factors, which were in the past decisive for fly ash deposition. A high resolution spatial model was developed, which gives also information on micro-site differences of magnetic susceptibility as indicator for fly ash deposition and corresponds to the management need to develop strategies for a stand wise differentiated silvicultural treatment in dependence from growth relevant differences in the site potentials.

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