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**Regional investigation of spatial-temporal variability of soil magnesium
- a case study from Switzerland**

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Abstract

Magnesium (Mg) is an essential element for plant growth and human health. Its availability and spatial distribution in soils depends on a wide variety of intrinsic and extrinsic factors. Understanding how Mg availability changes in space and time is crucial for preventing potential deficiencies. In 1987, a soil-monitoring network (FRIBO) was launched in the canton of Fribourg, Switzerland. It was based on 250 sites distributed evenly throughout the canton so as to include a large variety of soil types (Cambisols, Gleysols, Rendzinas, Regosols, Lithosols, Luvisols and Fluvisols) under three different land use types (croplands, permanent grasslands and mountain pastures). The aim of this research was to characterize the spatial and temporal variation of total and available forms of Mg in the agricultural soils of the canton of Fribourg and to discuss potential implications for Mg fertilization management. Total Mg concentration (Mg_T) averaged 5.5 g kg^{-1} , with small differences between land use types. Spatial distribution of Mg_T showed higher values on the southern part of the study area, mostly on Rendzinas and Cambisols. Average concentrations of available Mg forms were significantly different according to extraction methods, with water extraction (Mg_{H_2O}) having the lowest value (14.2 mg kg^{-1}) followed by calcium extraction (Mg_{CaCl_2} , 109.4 mg kg^{-1}) and ammonium acetate + EDTA extraction (Mg_{AAE} , 148.7 mg kg^{-1}). On average, permanent grasslands had significantly higher values for all Mg forms compared to croplands and mountain pastures, a result further corroborated by the analysis of spatial distribution. Intrinsic factors, such as soil type and terrain attributes, appeared to have a major influence on total Mg content, whereas available Mg forms depended mostly on extrinsic factors, such as land use type. Temporal analysis of soil available Mg forms revealed an overall increase between 1987 and 2016, especially after a land use change from croplands to permanent grasslands. In the light of the essential role of Mg for plant growth and development, as well as its critical role in animal health, the status of Mg should continue to be monitored in the FRIBO network and plant analysis should be implemented.

Keywords: soil magnesium, soil properties, terrain attributes, land use, spatial and temporal variability, Cambisols

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1. Introduction

Magnesium (Mg) is a vital element for plant growth being involved in various critical metabolic and physiological functions, such as chlorophyll and protein synthesis, enzyme activation, phosphorylation, photosynthesis and carbohydrate partitioning (Cakmak and Yazici, 2010; Farhat et al., 2016; Hawkesford et al., 2012; Marschner, 2012). Despite its importance, there has been little research concerning the role of Mg in agriculture (Cakmak and Yazici, 2010). Because latent Mg deficiency is difficult to detect, the importance of this element in agricultural research has been often under-evaluated (Gransee and Führs, 2013). Recently however, Mg has come into the spotlight as a limiting factor in crop productions subjected to continuous fertilization solely based on nitrogen (N), phosphorus (P) and potassium (K) (Cakmak and Yazici, 2010; Jayaganesh et al., 2011). This issue has become critical especially in more developed countries due to associated human health concerns (Guo et al., 2016; Long and Romani, 2014).

Magnesium is the 8th most abundant element on the Earth's crust and is present in a wide variety of minerals in the Mg^{2+} form (Maguire and Cowan, 2012; Mikkelsen, 2010). The majority of soil Mg (90-98%) is incorporated into soil minerals and is not immediately available for plant uptake (Sembayram et al., 2015). The availability of Mg for plants depends on the type of parent material as well as on weathering, fixation, erosion, leaching beyond the root zone, soil moisture and soil pH, high use of NPK fertilizers, and other anthropogenic activities (Guo et al., 2015, 2016; Gransee and Führs, 2013; Metson, 1974). Another factor that can potentially influence Mg availability is the antagonistic tendencies towards K and calcium (Ca) (Ding et al., 2006; Moore et al., 1961). Plants that suffer from Mg deficiency present a lower proportion of protein N compared to non-protein N. As a result, the rate of photosynthesis decreases in these plants and carbohydrates are accumulate in the leaves (Farhat et al., 2016; Hawkesford et al., 2012). If left unchecked, this condition can worsen,

causing an inhibition of plant growth, accelerated aging and reduced productivity (Guo et al., 2016, 2015; Verbruggen and Hermans, 2013). Sufficient availability of Mg can provide a resistance to aluminum (Al) toxicity and alleviate the toxicity effects of other heavy metals (Cakmak, 2013; Farhat et al., 2016; Rengel et al., 2016; Silva et al., 2001). Finally, Mg plays a critical role in protecting plants from biotic and abiotic stress, for example by improving the resistance to light and heat stress (Cakmak, 2008; Cakmak and Yazici, 2010; Gransee and Führs, 2013; Mengutay et al., 2013; Senbayram et al., 2015). The most common practices for increasing Mg availability are the application of farmyard manure and mineral fertilizer (Chowaniak and Gondek, 2009; Mazur and Mazur, 2015). In addition, no-till farming has been proven to increase Mg content in the soil surface layer (Chervet et al., 2016). Although Mg toxicity in plants can be equally problematic, it is quite uncommon and its effects are often negligible (Venkatesan and Jayaganesh, 2010).

In recent years, human Mg deficiency has also become an issue due to the lack of Mg content in plants coupled with the consumption of processed foods (Broadley and White, 2010; Maguire and Cowan, 2012; Guo et al., 2016; Long and Romani, 2014). In human metabolism this element is essential for the regulation of muscular contraction, blood pressure, insulin metabolism, cardiac excitability, vasomotor tone, nerve transmission and neuromuscular conduction (Gröber et al., 2015). Magnesium deficiency has also been found to be closely connected to age-related diseases as well as to Parkinson's disease (Gröber et al., 2015; Guo et al., 2016; Killilea and Maier, 2008; Long and Romani, 2014; Sun, 2018). For what concerns livestock-related diseases, Mg plays an important role in preventing grass tetany and milk fever (Hawkesford et al., 2012; Sun et al., 2013).

In the last decade, soil science has benefited from impressive advancements in the field of geographic information systems (GIS). This has led to the creation of a large multitude of geo-referenced soil databases from regional to global scales (McBratney et al.,

2003). Many recent soil fertility studies, however, have primarily focused on understanding the spatial variability of soil properties at field scale in relation to agricultural practices (Ferreira et al., 2015; Peukert et al., 2012; Usowicz and Lipiec, 2017; Vasu et al., 2017), so that less attention has been dedicated to the spatial variability of fertility at regional scale (Webster and Oliver, 2007). In recent years, soil scientists and agronomists have started understanding the effects that land use management, pedology and climate change have on soil fertility and nutrients at regional scales (Blanchet et al., 2017; Roger et al., 2014).

Our study focused on agricultural soils of the canton of Fribourg (western part of Switzerland). This region hosts a wide variety of agricultural practices which are adapted to different soil types, physiographic characteristics, soil properties and climate conditions. Agriculture is a very important activity in the canton; agricultural land covers more than 59% of the surface, and 70% of this is grassland mainly devoted to milk and cheese production. Currently, a growing number of farmers are converting their activities to the production of meat (Etat de Fribourg, 2018). This highlights the importance of Mg not only for maintaining sustainable yields, but also for ensuring high-quality crops for human consumption as well as for the health of livestock. To our knowledge, no research has been conducted on the status of Mg at a regional scale in Switzerland. The objectives of this study were to: (i) characterize the soil Mg status (total and available forms) in the agriculture soils of the canton of Fribourg (FRIBO network) according to four different extraction methods, (ii) analyze the spatial and temporal variation of soil Mg based on land use, soil type and physiographic characteristics, (iii) evaluate the spatial predictability of soil Mg forms at canton level, and (iv) discuss the implications for Mg fertilization management.

2. Material and methods

2.1 Study area

The canton of Fribourg (1,670 km², 4% of the area of the country) is located in the western part of Switzerland (46° 4' N; 7° 5' E). Due to its unique position as a transition area between the Swiss Midlands and the western Alps, this territory has a large variety of landscapes and ecosystems. The regional topography follows a northwest to southeast gradient and gentle slopes give way to steeper hills near the Alps. Other characteristics of the territory, such as geology and pedology, also follow the same topographical gradient. The climate of the study area is temperate-continental with generally cold winters (lowest mean monthly temperature of -0.1 °C in January) and mild summers (highest mean monthly temperature of 18.4 °C in July). The mean annual temperature during the 1981 - 2016 period was 8.9 °C. Mean annual precipitation is 1075 mm, with a minimal monthly value of 55 mm (February) and a maximal monthly value of 126 mm (May) (MétéoSuisse, 2016). Due to the influence of the Quaternary glaciation, the geology for this region is highly complex (Signer et al., 2000). This conformation resulted in molasses deposits subsequently covered by moraines in the north and a combination of alpine, flysch and calcareous formations in the south. These parent materials resulted in the distribution of Cambisols in the north and center of the canton and a combination of Regosols, Gleysols, Rendzinas and Lithosols in the south. Luvisols and Fluvisols occur throughout the whole canton in fewer areas (Fig. S1).

Our study is based on data from the FRIBO network, which was established in 1987 by the Agricultural Institute of the canton of Fribourg. The network is composed of 250 sampling sites distributed along a 2-by-2-km grid. Since the beginning of the monitoring, about 50 sites have been sampled annually (Blanchet et al., 2017; Roger et al., 2014). Each site has been sampled every 5 years. This study uses FRIBO data from the 6th cycle (2012-

2016). Eight of the 250 sites were excluded from our analyses due to very high soil organic matter (SOM) content causing some outliers in the different measured Mg forms.

The remaining 242 sampling sites can be classified in three distinct land use categories: 120 sites as croplands, 79 as permanent grasslands and 43 as mountain pastures (Fig. S1). In the Fribourg region, a meadow-maize-wheat-barley-rapeseed rotation is generally applied to croplands, but other crops (from tubers to vineyards) are also cultivated at some sites. The “permanent grasslands” category includes meadows that were sown at least 6 years before the soil sampling. Finally, the “mountain pastures” category includes all alpine grasslands used as pastures during the summer months. These types of land use reflect the spatial distribution of topography and soil characteristics (Fig. S1). The farmers of the FRIBO network apply fertilizers according to the Swiss guidelines for crops and grasslands (Sinaj and Richner, 2017). Because Mg currently receives little attention in Swiss agriculture, it is likely that Mg fertilization mainly occurred through the use of farm manure and NPK fertilizers with a low percentage of Mg.

2.2 Soil sampling and analysis

At each site, composite soil samples of the surface horizon (0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures) were obtained from 25 soil cores sampled over an area of 100 m². Plant residues were removed and samples were air-dried, sieved at 2 mm and further analyzed for different soil chemical properties (Table 1). The pH-H₂O, soil organic carbon (SOC), clay and sand content, and cation-exchange capacity (CEC) were measured according to the Swiss standard methods (Agroscope, 2011).

Soil total Mg (Mg_T), phosphorus (P_T) and potassium (K_T) concentrations were obtained by digestion of 0.25 g of soil previously treated in 5 ml of HF acid (40%) and 1.5 ml of HClO₄ (65%) according to the AFNOR standard X31-147 (1996) followed by determination with flame atomic absorption spectrometry. Soil available Mg forms were

measured by three methods used in routine analyses in Switzerland. The first method operated at an acidic pH (4.6) in the presence of ammonium acetate and EDTA as a complexing agent (Agroscope, 2011, Mg_{AAE}). The second method was based on a 2-hour-long analysis at a 1:10 soil to extract ($CaCl_2$ 0.0125 M) ratio (Agroscope, 2011, Mg_{CaCl_2}). The third method was based on a 1:10 soil to water ratio for a 16-hour period (Agroscope, 2011, Mg_{H_2O}). After extraction, Mg was determined by flame atomic absorption spectrometry.

2.3 Environmental variables for spatial predictions of Mg forms

The Digital Elevation Model (DEM) used for this study was projected according to the CH1903 Hotline Oblique Mercator Azimuth Center Geographic Coordinate System (Swisstopo, 2016). The original grid resolution of 5 meters was resampled through bilinear interpolation to 30 meters, as suggested by previous studies concerning the FRIBO network (Blanchet et al., 2017; Roger et al., 2014). This was done to reduce the noise of the elevation data. Various terrain attributes (slope, slope length, mid-slope position, curvature, planform curvature, profile curvature, standardized height, normalized height, SAGA Wetness Index, Vector Terrain Ruggedness, and Terrain Ruggedness) were obtained through the use of the System for Automated Geospatial Analysis (SAGA) (Conrad, 2006) and were used as environmental variables for the Mg spatial predictions. In addition, soil type and land use were included as environmental variables for all the 242 sites in order to investigate their spatial relationship with Mg.

2.4 Statistical analysis

The dataset was initially analyzed to determine the spatial variations of Mg forms in the agricultural soils of the canton (data from the 6th cycle, 2012-2016). This was done through the use of classical statistics and geostatistics. To assess the temporal variations of soil available Mg forms in the FRIBO network, current Mg_{AAE} and Mg_{CaCl_2} values were

coupled with values from the previous five sampling cycles. This new dataset was then analyzed using classical statistics.

2.4.1 Classical statistics

Initially, descriptive statistics (mean, median, standard deviation and coefficient of variation) were calculated for some general physico-chemical soil characteristics and different forms of soil Mg grouped according to the different land use classes. The next step was to conduct one-way analysis of variance (ANOVA) coupled with a Tukey-Kramer test to verify whether the physico-chemical soil properties and soil Mg forms were significantly different between the three categories of land use. In addition, the various Mg forms were plotted into boxplots according to land use categories and soil types to indicate their distribution, variation and significance. Finally, a principal component analysis (PCA) was performed on the multivariate dataset. Correlations among terrain attributes and soil variables by the projection of the 242 sites along the first and second PCA axis according to land use were investigated. All statistical analyses were performed on R Version 3.5.0 (R Core Team, 2014) using the packages “vegan” (Oksanen et al., 2018) and “agricolae” (Mendiburu, 2015).

2.4.2 Spatial analysis

2.4.2.1 Spatial dependence

We measured the spatial dependence of Mg_T , Mg_{AAE} , Mg_{CaCl_2} and Mg_{H_2O} using the geographical coordinates of the 242 sampling sites of the FRIBO database with the Geoda software (Anselin et al., 2006). To this aim, we calculated Local Indicators of Spatial Association (LISA) (Anselin, 1995). LISA indicators are statistics that measure spatial dependence and evaluate the existence of local clusters in the spatial arrangement of a given variable. They are based on the statistical index “I” developed by Moran (1950) to measure the global level of spatial autocorrelation of the variables in a study area. Moran’s I ranges from -1 (negative spatial autocorrelation) to 1 (complete spatial dependence), with 0

indicating the absence of spatial dependence. In this study, we used univariate LISA within a spatial lag of 10 kilometers (i.e. neighborhood considered for the analysis). This choice was made on the basis of the distance at which there was no neighborless sampling station (3.48 km). Next, we produced a correlogram for the four Mg variables. It showed that the value of the global Moran's I always remained between 0.1 (for spatial lags < 5 km) and -0.1 (for spatial lags > 60 km) which suggests no global spatial dependence. We decided to use the same spatial lag size as in our previous studies (Blanchet et al., 2017; Roger et al., 2014) to facilitate comparisons. Subsequently, for each sampling site the correlation between the measured variable and its mean within the 10 km neighborhood was calculated. The standardized scattergram for this relationship showed four distinct classes: a) high observed Mg values correlated with high Mg weighted values b) low observed values correlated with low weighted values, c) a low-high relationship and d) a high-low relationship. The attribution of sampling sites to these four classes depended on the results of a significance test. This test performed many random permutations among sites located in the spatial lag and compared the observed LISA values to the many LISA values corresponding to the random permutations (see details in Anselin, 1995). All sampling sites not significant were displayed in white. This allowed us to produce maps showing locations where there was significant spatial dependence of Mg variables and to identify unique sampling locations, i.e. a locally different behavior (outlier) from the other sampling sites of the same area.

2.4.2.2 Interpolation methods

Initially, a multivariate analysis was conducted to determine the multi-collinearity among environmental variables derived from DEM, soil type and land use for spatial predictions of soil Mg forms. The best predictors for the Mg forms were determined based on step-wise multiple linear regression (MLR) and were selected based on the minimum Akaike Information Criterion (AIC) (Akaike, 1976). The normality of the data distribution of soil Mg

forms was evaluated using the Shapiro-Wilk goodness-of-fit test. The test results suggested no need for transformation, and subsequent analyses were conducted on the untransformed data using JMP Version 11.3 (SAS Institute Inc., 2018) statistical package. The spatial structure of Mg forms was determined by means of a semi-variogram using the nugget/sill ratio as defined by Cambardella et al. (1994). The structure of the spatial dependence and residuals was modelled in VESPER based on automated variogram fitting (Whelan et al., 2002). No anisotropy was found among the selected variables; therefore, isotropy was assumed for all kriging calculations. All semi-variograms were fitted with the exponential model. Spatial interpolation was conducted based on ordinary kriging (OK), regression-kriging for the step-wise MLR model, as suggested by Hengl et al. (2004) and Odeh et al. (1995, 1994). The 95% confidence limits of the predictions were also calculated. Due to the limited number of points measured data relative to the study area size (1 per 6.8 km²), the entire dataset was used for modeling and a leave-one-out cross-validation was used for model validation.

2.4.3 Temporal Analysis

To understand the temporal variability of Mg in the FRIBO network, a two-step analysis was performed for Mg_{AAE} and Mg_{CaCl2}. *The first step* focused mainly on the evolution of both Mg forms for sites that remained under the same land use for 30 years, i.e. since the beginning of the FRIBO network. *The second step* considered the various changes in land use class that sites had undergone during 30 years. Initially, the dataset was sorted to identify the changes in land use between each cycle (for example, between cycle 1 and 2). This revealed that the sites underwent either a change from “cropland” to “permanent grassland”, or the reverse. No “mountain pasture” sites underwent a change. All sites that did not undergo a change in land use were placed in the “No Change” category for analysis. Once sites were placed in one of the three categories, an average value was calculated for each Mg form, land use change category and transition period. From these means, it was then possible to obtain a

slope of the evolution of the Mg form between cycles. These slopes were then used to calculate an overall average slope describing the evolution of Mg_{AAE} and Mg_{CaCl_2} according to the changes in various land use categories. An ANOVA was then performed, accompanied by a Tukey-Kramer test for pairwise comparisons for determining whether the slopes were significantly different.

3. Results and discussion

3.1 Soil properties

A detailed description of the status of the physical-chemical properties of the soils of the FRIBO network was already reported in past studies (Blanchet et al., 2017; Roger et al., 2014). Except for the base saturation cation-exchange capacity (Sat_{CEC}), all general physico-chemical properties were significantly different for the different categories of land use (Table 1). Most of the soil properties were highly variable, especially for croplands, most likely due to the different agricultural practices. Croplands were characterized by neutral soil reaction ($pH = 6.6$), sandy soils and low SOC (20.1 g kg^{-1}), whereas mountain pastures were characterized by moderately acidic ($pH = 5.8$), clay-rich soils and high SOC (50.6 g kg^{-1}). Differences between the land use categories have been found to heavily influence Mg availability (Mazur and Mazur, 2015; Senbayram et al., 2015). In addition, significantly higher amounts of K_T found in cropland soils could influence the availability of Mg and the ability of plants to absorb Mg due to antagonism effects (Moore et al., 1961).

3.2 Soil magnesium status

Total Mg averaged 5.5 g kg^{-1} and ranged from 2.8 to 13.0 g kg^{-1} across the study area (Table 1). The differences in Mg_T between land use types were not significant, varying from 5.5 g kg^{-1} (croplands) to 5.6 g kg^{-1} (permanent grasslands and mountain pastures) (Table 1). Similar trends were observed in relation to soil types, except for the significant difference between Cambisols and Luvisols (Fig. 1). Luvisols had significantly higher median Mg_T (7.8

g kg⁻¹) compared to Cambisols (5.1 g kg⁻¹). Interestingly, almost all the Luvisols occurred around or close to lakes, areas under intense and diverse management and cropping systems (Etat de Fribourg, 2014). This could explain the wider range and higher Mg_T values. Also, these areas have experienced depositions of finer materials, due to their position within floodplains, promoting the formation of clay-filled and water-rich soils with higher Mg_T content. Overall, the small differences in Mg between land use types and soil types suggest that soil physico-chemical properties may have a greater influence on Mg_T content (Fig. 2). For example, Mg_T was significantly correlated with total calcium ($r = 0.43$), base saturation cation-exchange capacity ($r = 0.46$), as well as soil silt content ($r = 0.39$) (Data not shown, Fig. 2).

The available forms of Mg showed a wide range of mean values, from 12.2 mg kg⁻¹ (croplands, Mg_{H₂O}) to 181.6 mg kg⁻¹ (permanent grasslands, Mg_{AAE}) (Table 1; Fig. 1). Permanent grasslands had the highest amount of available Mg for all forms compared to croplands and mountain pastures (Table 1). One possible explanation is that soils benefit from the fertilization with organic manure rich in Mg, which is common on permanent grassland and has been shown to increase the available Mg forms (Etat de Fribourg, 2014; Jeangros and Sinaj, 2018; Maltas et al., 2018). As expected, the available Mg forms (Mg_{AAE}, Mg_{CaCl₂} and Mg_{H₂O}) showed the highest variability (CV = 45%) across the canton compared to Mg_T (CV = 35%), with the widest range between 29.1 and 444.4 mg·kg⁻¹ (Mg_{AAE}). Similar trends were also observed for soil types (Fig. 1). Cambisols showed a wide range of data distribution for Mg_{AAE}, but had comparable ranges with other soil types for Mg_{CaCl₂} and Mg_{H₂O}. The lack of significant differences between soil types for the available Mg forms suggests that pedology plays a minor role in affecting Mg availability (Fig. 1). Because all available forms of Mg were strongly correlated to each other (Fig. 2), there were little differences between extraction methods, a result suggesting that availability of Mg can be determined using only one

extraction method. Similar to Mg_T , available Mg forms were also correlated with clay and silt content, base saturation cation-exchange capacity and Mg_T content (Fig. 2).

In summary, while Mg_T status seems to be less influenced by land use, available Mg forms appear to be partly sensitive to agricultural practices. Total Mg is primarily influenced by intrinsic factors (for example, soil type), while available Mg forms are influenced by a combination of intrinsic and extrinsic factors. According to our data, Mg_T content appears to be adequate for crop growth; however, given the soil properties of the study area, Mg availability to plants for optimal yields may need to be considered in the future. While the overall comparisons of all Mg forms between land use and soil types revealed few significant differences (Table 1, Fig. 1), spatial and temporal distribution is equally important for better understanding the overall regional trends for more site-specific Mg management.

3.3 Spatial autocorrelation of soil magnesium forms

Globally, the spatial distribution of the Mg forms (Mg_T , Mg_{AAE} , Mg_{CaCl_2} and Mg_{H_2O}) based on spatial autocorrelation followed a gradient that extended from the southwest to the northeast of the study area (Fig. 3a, b; Fig. S2a, b). Sites in high-high clusters (N=41) were located in the Veveyse, Glâne and Gruyère districts, while low-low clusters (N=44) were mostly found in the Singirè district. However, slight differences existed in the spatial distribution of the Mg forms.

The majority of measured Mg_T data did not show any spatial autocorrelation (Fig. 3a, N=146). Spatial autocorrelation occurred mostly in the central part of the study area, dominated by Cambisols, followed by the southern part, dominated by Redzinas and Lithosols (Fig. S1). High-high clusters of Mg_T were concentrated in the Veveyse district and in the area south of Glâne (Fig. 3a, N=29), both areas dominated by croplands and permanent grasslands. Low-low clusters occurred in the Broye district to the west of the study area, just east of Lake

Neuchâtel, and in the Singine district to the east of the study area (Fig. 3a, N=41). The sites were mostly under croplands and in Cambisols (Fig. S1).

The available Mg forms showed spatial autocorrelations similar to Mg_T (Fig. 3b; Fig. S2a&b). High-high clusters for all three available Mg forms (Mg_{AAE} , Mg_{CaCl_2} and Mg_{H_2O}) covered the whole southern part of the study area. They were predominantly in Cambisols under cropland and permeant grassland in Veveyse, Glâne and Gruyère districts. The spatial distribution of the low-low clusters was also very similar to the one observed for Mg_T , corresponding to stations located mainly in the Singine, Lac and Broye districts predominantly in croplands and Cambisols (Fig. S1).

Spatial autocorrelation analysis revealed mixed trends regarding relationships between sites. Most sites showed no spatial autocorrelation, followed by high-high and low-low sites, split approximately in half. While this analysis is very useful for site-to-site spatial variation, the between-sites spatial variation and distribution of Mg forms could also be useful, especially for regional planning.

3.4 Spatial prediction of soil magnesium forms

The prediction of spatial distribution of Mg forms between sites can be explained by the use of environmental factors (Hengl et al., 2004; McBratney et al., 2003; Odeh et al., 1995, 1994). In previous studies from the same area, Blanchet et al. (2017) and Roger et al. (2014) successfully used environmental variables to improve spatial predictions of P and K. After removing the auto-correlated environmental variables to avoid overfitting, we applied a step-wise multiple linear regression analysis (MLR) (Table 2). Only environmental variables significant at $p \leq 0.05$ and models with the lowest AIC were selected. The standard least square (SLS) values indicated that vector terrain ruggedness was statistically the most significant contributor to spatial predictions of Mg_T (Table 2). For Mg_{H_2O} , although all predictors were significant, their weighted contribution was very low. On the other hand, for

Mg_{AAE} and Mg_{CaCl_2} predictions, normalized height and land use had relatively higher weights. However, the modeling and validation results showed that, overall, the selected environmental covariates were able to explain only between 7% and 14% of the spatial variability of Mg forms. Because the use of regression kriging is not recommended when environmental variables are poor spatial predictors (Hengl et al., 2004), ordinary kriging was used for spatial predictions of Mg forms.

The spatial distribution of all Mg forms were similar, showing higher concentrations in the central part of the study area (Fig. 4; Fig. S3). The higher values for Mg_T in the southern part of the study area occurred mostly on Rendzinas, partially on Regosols, and mountain pastures (Fig. 4a). On the other hand, the higher concentrations of available Mg forms occurred on Cambisols and on a mixture of cropland and permanent grassland located around Lakes Morat and Neuchâtel (Fig. 4c; Fig. S3a, c). These results suggest that a complex interaction between soils and land use determines the spatial distribution of Mg forms, especially the available ones. The higher spatial variability of available forms in the croplands and permanent grasslands suggests that land use, in particular, has an influence on these Mg forms.

The nugget/sill ratio based on ordinary kriging was overall greater than 50% for the different Mg forms (Table 3). Based on the scale defined by Cambardella et al. (1994), a nugget/sill ratio greater than 50% is an indication of moderate to weak spatial dependency. The range values (Table 3) were high, varying from 15,000 to 50,000 m. This also suggests poor spatial dependency and indicates that accurate predictions between sites at finer scales are not to be expected.

The width of the 95% confidence interval (CI) varied spatially and between Mg forms. The highest CI for all Mg forms occurred on the edges of the study area, while the lowest occurred on the central part of the study area (Fig. 4b, d; Fig. S3b, d). The exponential

function used for ordinary kriging showed relatively high nuggets effect, which when above a certain fraction could require more observation points to lower kriging variance, especially around the edges (van Groenigen, 2000), like in our study area. The width of CI for all Mg forms was relatively small and uniform across the study area, implying consistent thresholds for fertilizer recommendations. However, the density of points used for spatial interpolation was only 1 per 6.8 km², which oversimplifies the spatial heterogeneity resulting from the average farm size, i.e. 0.22 km² according to the Etat de Fribourg (2018), and diverse management and fertilizer practices. This underscores the importance of an appropriate density of observation points in order to improve predictions especially at finer scales, like at farm and/or field level, and to potentially reduce the error (Roger et al., 2014; Blanchet et al., 2017).

3.5 Temporal trends

The collection of soil samples at intervals of 5 years since 1987 provided an opportunity to understand the evolution of available Mg forms through time. Overall, both available Mg forms (Mg_{AAE} and Mg_{CaCl_2}) increased between 1987 and 2016 (Fig. 5a, b), however not all changes were significant (Table 4). During the entire extent of the 6 cycles, 48 sites changed land use type. Thirty sites changed from croplands to permanent grasslands, while 18 changed in the opposite direction. Sites changing from permanent grasslands to croplands were characterized by a significant increase of both available Mg forms (Table 4). A transition from croplands to permanent grasslands significantly increased the concentration of Mg_{AAE} (Slope = 32.1), while the concentration barely increased in the opposite case (Slope = 1.4) (Table 4). The change from croplands to permanent grasslands caused an increase in Mg_{AAE} concentrations from 4.73 to 33.28%, while the opposite caused a strong decrease in the first two cycles (-14.12 and -7.92%, respectively) and a small increment (between 0.14 and 14.15%) in the remaining three cycles. The differences between land use classes can be

potentially explained by the different agricultural practices. Indeed, croplands are regularly fertilized (Sinaj and Richner, 2017), so it is reasonable to assume that additions of chemical Mg-fertilizers increased the availability of Mg. The high concentration and the increasing trend in permanent grasslands can be explained by the regular inputs of Mg through animal manure and by the reduced Mg leaching due to the dense and permanent soil cover provided by grassland species (Chowaniak and Gondek, 2009; Maltas et al., 2018).

3.6 Consequences of magnesium spatial-temporal distribution for magnesium fertilization

All Mg forms showed a wide range of variability. Gruyère, Veveyse and Glâne districts, characterized by permanent grasslands and mountain pastures on Regosols or Cambisols, had the highest Mg_T values. The highest levels of available Mg were mostly in regions dominated by permanent grasslands, however, the observed values were within a “low” to “moderate” range according to the current Swiss fertilization guidelines (Sinaj and Richner, 2017). The lowest levels of available and total Mg in regions dominated by croplands may potentially point to a negative relationship between agricultural practices and Mg availability. Although, on average, the current levels of Mg forms in soils do not indicate deficiencies for crops, the high levels of soil macronutrients, such as K, coupled with high NP fertilization in the study area could potentially lead to Mg depletion (Guo et al., 2016) and shortages for plant growth and optimum yields. Additionally, temporal trends for Mg_{AAE} and Mg_{CaCl_2} revealed significantly higher increase in soil, especially for sites that changed from croplands to permanent grasslands. This suggests a stronger depletion of Mg forms in croplands during the last 30 years and the relatively fast recovery of soil Mg in permanent grasslands due to addition of organic manure, which has been found to favor Mg availability (Eneji et al., 2003; Jeangros and Sinaj, 2018; Chowaniak and Gondek, 2009; Maltas et al.,

2018). The fact that croplands showed the lowest values for soil available Mg suggests that agricultural land is more vulnerable to alterations of available Mg. If this trend should continue, it could potentially become problematic for livestock that could be susceptible to dangerous diseases due to Mg shortage (Sun et al., 2013).

In the context of climate change and global warming (IPCC, 2013), plants may become more susceptible to variations of soil nutrients. A continuous monitoring of soil Mg levels may shed some light on the spatial and temporal trends of this element in the region. However, monitoring alone may be insufficient to estimate Mg impact on agriculture considering that the Mg content in the soil may not reflect the ability of plants to absorb it (Gransee and Führs, 2013).

Conclusion

We showed that Mg content presented a large variability throughout the FRIBO soil network. From a spatial perspective, total Mg was less variable than available forms. Total Mg was more abundant in the lower mountainous portion of the canton, while the available forms of Mg were more abundant in the central region dominated by permanent grasslands. Spatial distribution of total Mg was mainly related to intrinsic factors, such as soil type and properties, while the distribution of available Mg forms was more related to extrinsic factors, primarily land use practices. Temporal analysis of two available Mg forms (Mg_{AAE} and Mg_{CaCl_2}) showed an increase between 1987 and 2016, especially when land use changed from croplands to permanent grasslands. Although our data indicates that Mg is overall abundant in soils of the canton of Fribourg, the high content variability could create, at a local scale, limiting conditions for plant growth and, consequently, potential deficiencies for human and animal nutrition.

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Figure captions

Figure 1. Boxplots of the different Mg forms in the upper soil layer (0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures) in the FRIBO network according to land use and soil type.

Figure 2. Principal component analysis of Mg forms, environmental variables and soil characteristics determined at 0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures.

Fig 3. Cluster map of the Local Indices of Spatial Association (LISA) for (a) Mg_T and (b) Mg_{H_2O} with a spatial lag of 10 km. Results are significant at $p = 0.05$ (9999 permutations). Five distinct classes (colored circles) are represented. (i) red - high Mg values correlated with high weighted Mg values; (ii) blue - low Mg values correlated with low weighted Mg values; (iii) violet - a low-high relationship between Mg_T and weighted Mg, (iv) pink - a high-low relationship between Mg and weighted Mg, (v) white - no significant spatial dependence. The background shows the digital elevation model. The lower the altitude, the darker the color. (For interpretation of the references to color in this figure legend, the reader must refer to the web version of this article). Mg forms are determined at 0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures.

Figure 4. Spatial prediction maps of (a) Total magnesium (Mg_T), (b) 95% confidence interval (95% CI) based on ordinary kriging (OK), (c) Water extracted magnesium (Mg_{H_2O}), and (d) 95% confidence interval (95% CI) based on ordinary kriging (OK). Mg forms are determined at 0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures.

Figure 5. Temporal changes of (a) ammonium acetate extracted magnesium (Mg_{AAE}) and (b) calcium chlorite extracted magnesium (Mg_{CaCl_2}) over a 30-year period according to

different land use types. Mg forms are determined at 0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures.

Supplementary Figure 1. Sampling sites, soil types and land use types in the study area. Soil properties are determined at 0-20 cm for croplands and at 0-10 cm for grasslands and mountain pastures.

Supplementary Figure 2. Boxplots of the different Mg forms in the upper soil layer (0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures) in the FRIBO network according to soil type grouped by land use [Cropland, Permanent Grasslands (PG) and Mountain Pastures (MP)].

Supplementary Figure 3. Cluster map of the Local Indices of Spatial Association (LISA) for (a) Mg_{AAE} and (b) Mg_{CaCl_2} with a spatial lag of 10 km. Results are significant at $p = 0.05$ (9999 permutations). Five distinct classes (colored circles) are represented: (i) red - high Mg values correlated with high weighted Mg values; (ii) blue - low Mg values correlated with low weighted Mg values; (iii) white - a low-high relationship between Mg and weighted Mg, (iv) pink - a high-low relationship between Mg and weighted Mg, (v) white - no significant spatial dependence. The background shows the digital elevation model. The lower the altitude, the darker the color. (For interpretation of the references to color in this figure legend, the reader must refer to the web version of this article). Mg forms are determined at 0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures.

Supplementary Figure 4. Spatial prediction maps of (a) ammonium acetate extracted magnesium (Mg_{AAE}), (b) 95% confidence interval (95% CI) based on ordinary kriging (OK), (c) calcium chlorite extracted magnesium (Mg_{CaCl_2}), and (d) 95% confidence interval (95% CI) based on ordinary kriging (OK). Mg forms are determined at 0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures.

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Table 1.

Descriptive statistics [minimum and maximum value, mean, median, standard deviation (SD), and coefficient of variation (CV)] of the soil physico-chemical properties for all sites. Soil properties are determined at 0-20 cm for croplands and 0-10 cm for grasslands and mountain pastures. Different letters among land uses indicate a significant difference ($p < 0.05$) for a given variable (ANOVA followed by Tukey–Kramer pairwise comparisons test).

	Altitude	Amplitude	Humidity	Organic C	Total C	Total N	Total P	Total K	EC	CatCEC	Silt	Clay	Particle size
	m	°C	%	g kg ⁻¹	g kg ⁻¹	g kg ⁻¹	g kg ⁻¹	g kg ⁻¹	mol kg ⁻¹	cmol kg ⁻¹	%	µm	µm
<i>All sites (n = 242)</i>													
Minimum	M 29.5-1590	M 4 -75	M .6-8.09	M 4 5-121.58	M 8.4 7-752	M 8 1-772	M 2 1.005-52.1	M 6 7.65-100	M 1 .279-2.34	M 0 .67-22.62	M 1 .2		
Mean	M 81.15	M 7 0.38	M .32	M 94	M 30. 27.22	M 2 22.53	M 7 7.06	M 1 8.06	M 5 .96	M 0 4.12	M 1 .9		
Median	M 14.00	M 7 .00	M .20	M 97	M 27. 98.00	M 2 20.90	M 3 4.65	M 1 8.38	M 5 .93	M 0 3.81	M 1 .8		
SD	M 74.83	M 2 3.03	M .70	M 19	M 17. 09.80	M 1 5.43	M 7 .38	M 8 5.67	M 1 .29	M 0 .26	M 3 1.		
CV(%)	M 5.18	M 3 25.47	M 1.12	M 56	M 55. 32	M 4 3.39	M 2 9.10	M 4 6.99	M 2 0.75	M 3 3.13	M 2 42		
<i>Croplands (n = 120)</i>													
Minimum	M 29.50-995	M 4 -25	M .10-8.09	M 5 5-1590	M 8.4 7-454	M 8 79-772	M 1 .1-52	M 6 3.62-100	M 2 .44-1.64	M 0 .67-22.62	M 1 .7		
Mean	M 07.63	M 6 .63	M .56	M 10	M 20. 76.13	M 1 20.95	M 5 3.25	M 1 7.50	M 5 .87	M 0 5.51	M 1 .8		
Median	M 06.00	M 6 .00	M .50	M 06	M 17. 60.00	M 1 37.00	M 5 1.80	M 1 6.52	M 5 .84	M 0 5.13	M 1 .8		
SD	M 16.95	M 1 .14	M .68	M 89	M 10. 4.91	M 6 16.38	M 1 .02	M 6 7.65	M 1 .23	M 0 .01	M 3 3.		
CV(%)	M 9.25	M 1 6.09	M 0.31	M 18	M 54. 6.86	M 3 2.34	M 2 5.47	M 4 0.69	M 3 6.29	M 2 9.41	M 1 57		
Tukey Rank	c	a	a	c	c	c	a	c	a	b	a		
<i>Permanent Grasslands (n = 79)</i>													
Minimum	M 60-1015	M 4 -33	M .20-7.70	M 5 38-70.03	M 16. 13-752	M 1 1-694	M 2 .30-52.10	M 9 0.27-91.80	M 3 .60-2.34	M 0 .38-21.77	M 7 .8		
Mean	M 74.01	M 7 .64	M .23	M 51	M 36. 41.68	M 2 26.58	M 4 9.05	M 1 9.64	M 5 .11	M 1 3.15	M 1 .5		
Median	M 85.00	M 7 .00	M .20	M 89	M 33. 13.50	M 2 41.50	M 4 6.92	M 1 0.74	M 6 .09	M 1 2.82	M 1 .8		
SD	M 22.37	M 1 .74	M .57	M 82	M 10. 12.31	M 1 37.63	M 1 .69	M 7 2.21	M 1 .29	M 0 .78	M 2 .7		
CV(%)	M 5.81	M 1 01.38	M .17	M 65	M 29. 6.47	M 4 2.26	M 3 0.38	M 4 0.47	M 2 6.26	M 2 1.11	M 2 03		
Tukey Rank	b	b	b	b	b	b	b	b	a	a	b		

43) *Mountain pastures* (n =

in-Max	M	80-1590	8	-75	.60-7.44	4	09-121.58	22.	90-654	1	1-603	ε	1.59-49.67	1	7.65-87.24	1	.28-1.67	0	.27-19.25	7	.2
ean	M	274.63	1	8.65	.83	ε	59	50.	41.70	ε	98.14	2	3.93	2	6.65	5	.91	0	2.05	1	.2
edian	M	314.00	1	7.00	.70	ε	00	48.	46.00	ε	86.00	2	0.89	2	8.56	5	.83	0	2.00	1	.7
D	S	79.14	1	8.31	.72	ε	84	18.	11.36	1	54.07	1	.63	9	5.68	1	.35	0	.09	3	0.
V(%)	C	4.05	1	3.91	2.34	1	25	37.	2.59	ε	1.68	ε	0.22	4	7.67	2	8.19	3	5.66	2	24
Tukey Rank	T		a			c		a		a		c		a		a		b		b	

Table 2.

Summary results of the stepwise multiple linear regression of Mg forms.

Form ^a	Mg	Selected Predictors	RC (SLS*) ^b	p -value	^c A _{dj} R ²	K-fold R ²
T	Mg	Intercept	6.5	< 0.0001	0.07	0.04
		NH ^d	-1.2	0.0123		
		VTR ^d	62.6	0.0005		
		TRI ^d	-0.6	0.0023		
H ₂ O	Mg	Intercept	15.6	< 0.0001	0.07	0.06
		Slope	-6.2	0.0008		
		NH ^d	-3.8	0.0155		
		LULC ^d	1.6	0.0019		
AAE	Mg	Intercept	150.9	< 0.0001	0.12	0.08
		Slope	-3.2	0.0017		
		NH ^e	-75.2	< 0.0001		
		LULC ^d	34.9	< 0.0001		
CaCl ₂	Mg	Intercept	99.2	< 0.0001	0.14	0.12
		Slope	-2.4	0.0017		
		NH ^b	-28.8	0.0037		
		LULC ^d	24.6	< 0.0001		

^a Mg forms are determined at 0-20 cm for croplands and at 0-10 cm for grasslands and mountain pastures.

^b RC (SLS): regression coefficient of the respective predictors according to standard least square.

^c Adjusted R^2 refers to the coefficient of determination computed on the whole dataset, whereas K-fold R^2 was computed according to the leave one out Cross validation.

^d Abbreviations used for the predictors: VTR – Vector Terrain Ruggedness; TRI - Terrain Ruggedness Index; NH – normalized height; LULC – Land Use Land Cover

Table 3.

Semi-variogram parameters from ordinary kriging based on exponential function.

Forms ^b	Mg	Nugget Co	Partial sill C	Sill (Co + C)	Nugget/sill ratio	Range [m]	IC ^b	RMSE ^a
	Mg _T	4.1	3.3	7.4	0.56	5 0000	0	0.4
20	Mg _H	25.8	27.1	52.9	0.49	4 8962	62	2.2
AE	Mg _A	5337	4894	10231	0.52	4 0479	92	61
aCl2	Mg _C	1522	964	2485	0.61	1 4486	97	99

^a Mg forms are determined at 0-20 cm for croplands and at 0-10 cm for grasslands and mountain pastures.

^b AIC is Akaike Information Criterion; RMSE is root mean square error.

Table 4.

Land use change slopes and significance for Mg_{AAE} and Mg_{CaCl2}

Forms	Mg	Land Use Change (1210 sites, 6 cycles)	slope	Tukey Rank
AAE	Mg (30)	Cropland to Permanent Grassland	2.1	a
		Permanent Grassland to Cropland	4	b
		No Change (1162)	5	ab
CaCl2	Mg (30)	Cropland to Permanent Grassland	8.3	a
		Permanent Grassland to Cropland	.0	a
		No Change (1162)	.6	a

Highlights

- Spatial and temporal variability of soil Mg content in western Switzerland
- Total Mg content is influenced by soil forming processes
- Available Mg forms were primarily influenced by agricultural practices
- Temporal analysis of soil available Mg revealed an increase during the last 30 years

Journal Pre-proof

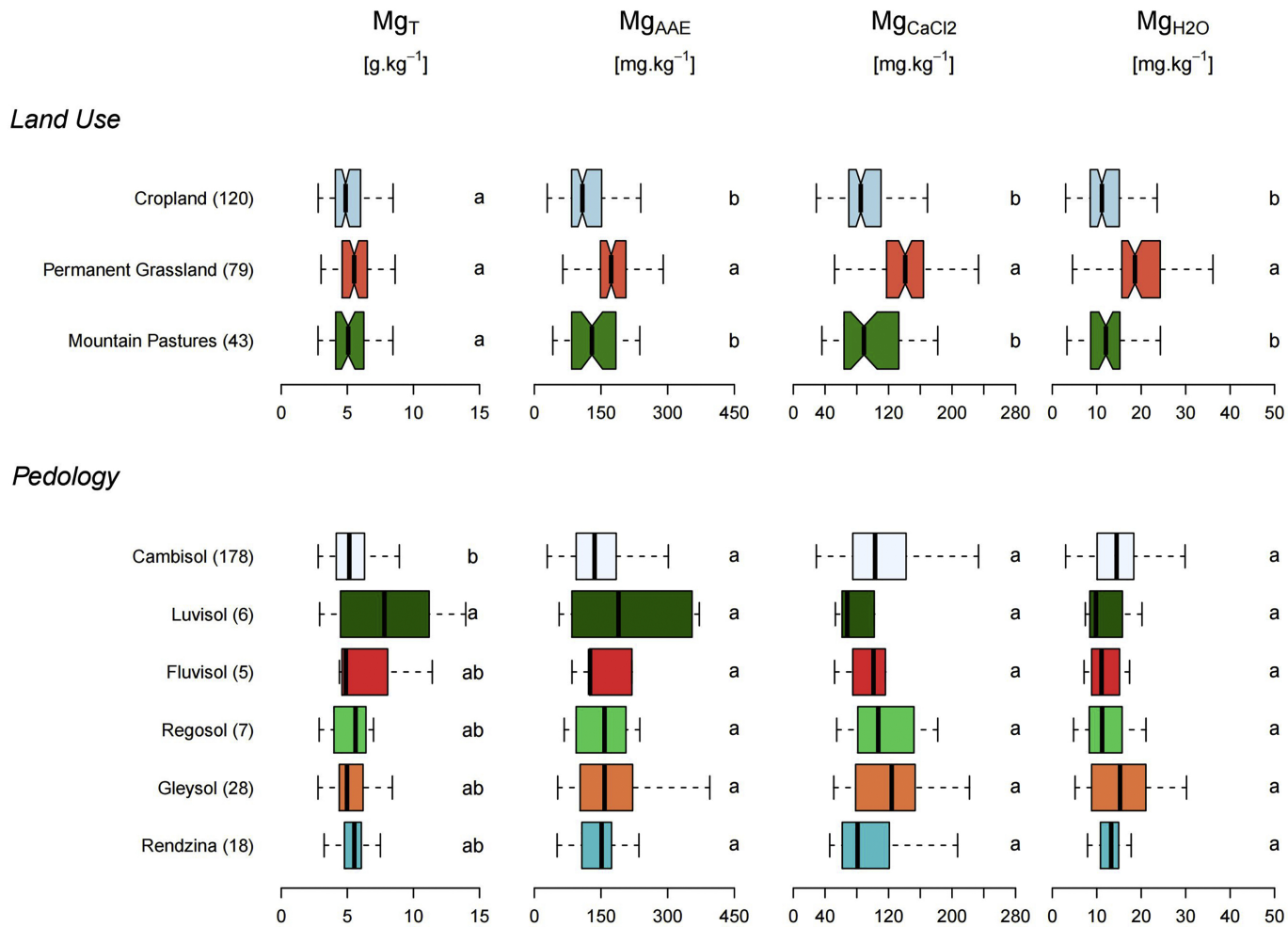


Figure 1

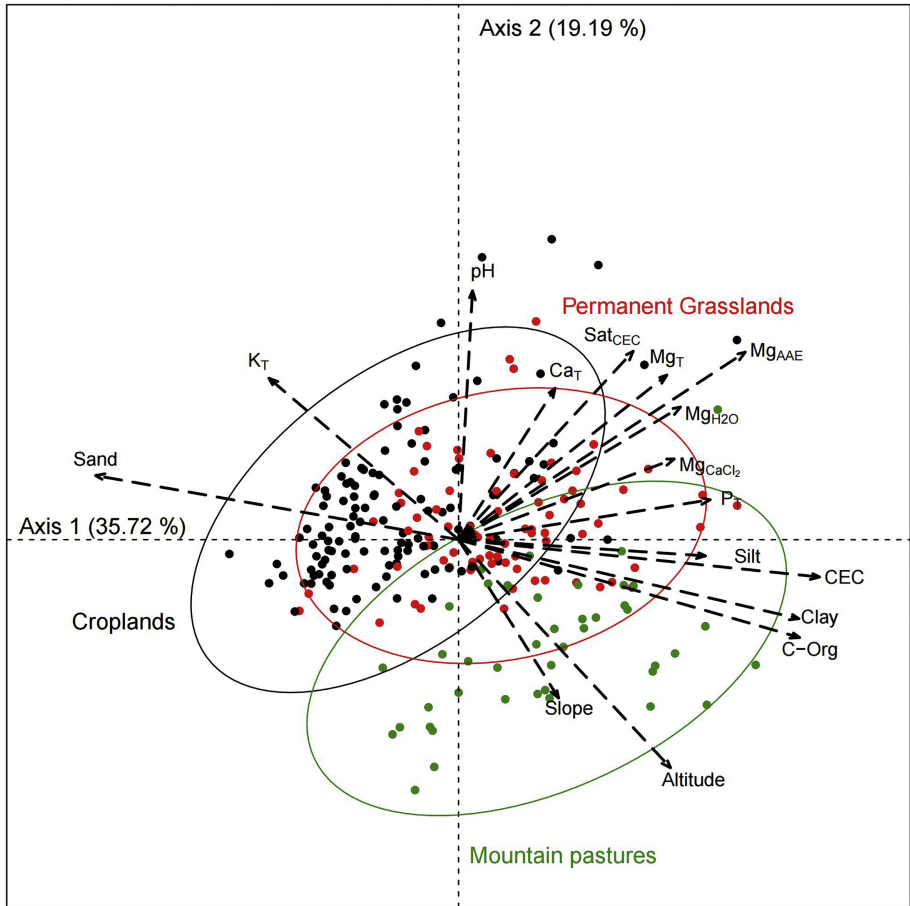


Figure 2

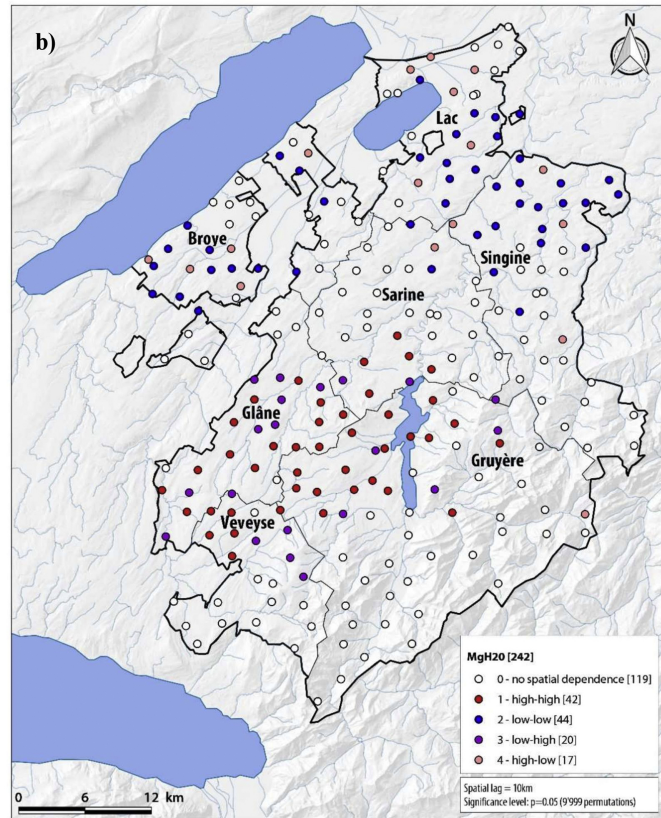
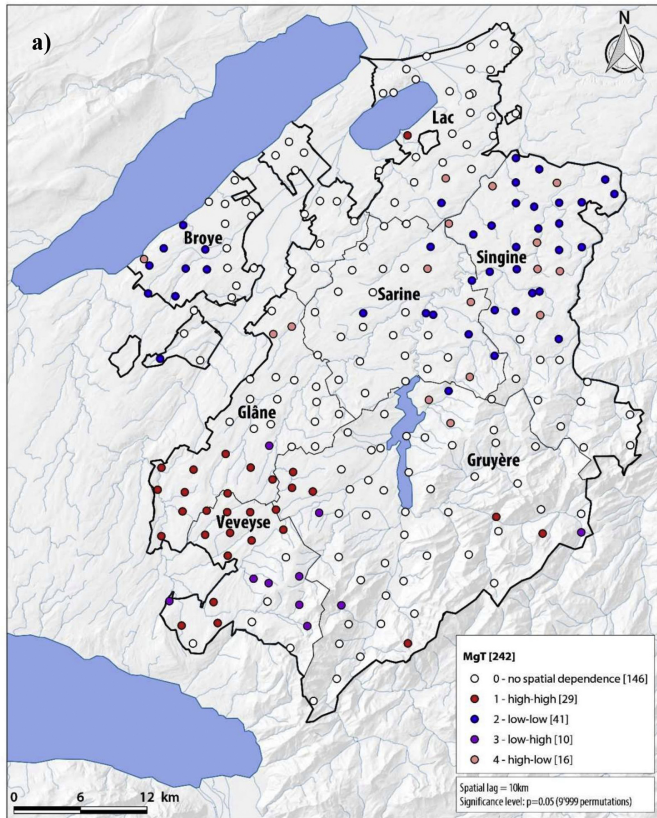


Figure 3

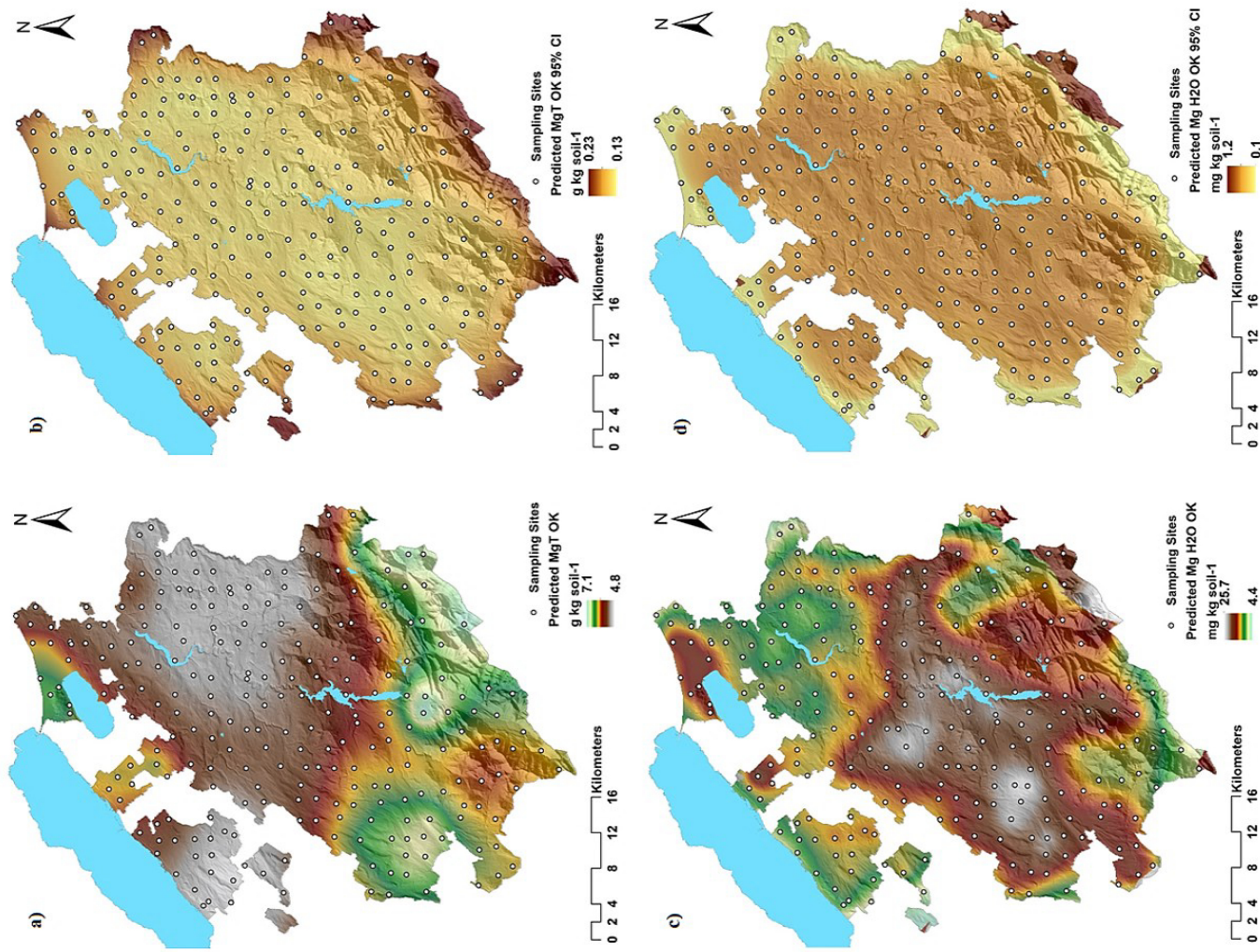


Figure 4

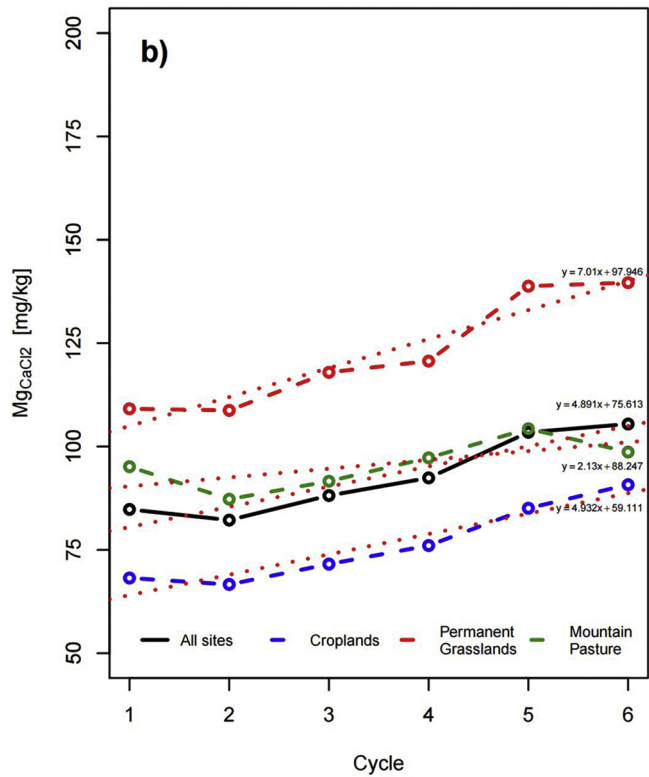
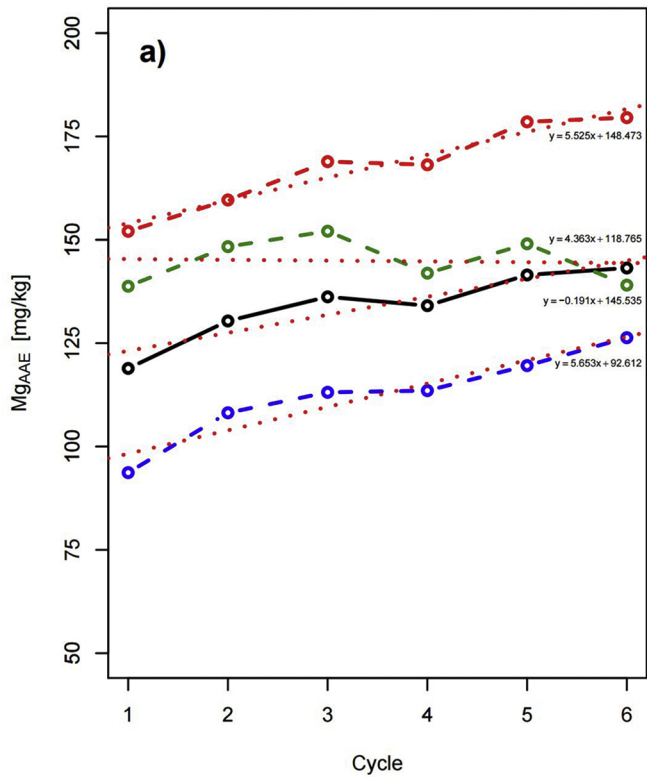


Figure 5