Secondary enrichment of soil by alkaline emissions: the specific form of anthropogenic soil degradation near magnesite processing factories and possibilities of land management

Nora Pollakova[1], Ján Hamar[1], Vladimir Simansky[1], Agata Bartkowiak[2], and Joanna Lemanowicz[2]

[1]Slovak University of Agriculture in Nitra
[2]University of Science and Technology Bydgoszcz

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Abstract

Over the past 90 years, anthropogenic degradation of soil caused by alkaline, magnesium-rich dust deposit has presented a serious problem near magnesite processing factories in Jelšava and in Lubeník (Slovakia). The objective of this study was to investigate the chemical and biological soil properties in 14 sampling sites at different distances from factories, and based on the results, to propose further use of affected land. Results revealed that the available Mg 3–68 fold exceeded very high content for texturally medium soils at all grassland sampling sites, and areas close factory contained up to 14.4–17.4 g kg-1. Higher excess of available Mg caused significant increase of soil pH (up to 9.39) and worsened the conditions for the growth of vegetation. As a result, lower stock of newly formed organic matter (0.50–0.96 g kg-1 of labile carbon) with consequently weaker enzymatic activity occurred. Therefore, enrichment by organic matter provides a measure to support the biological activity of soil. The content of monitored heavy metals (Zn, Cu, Pb and Ni) was not related to Mg and did not influence the enzymatic activity of soil. Because alkaline emissions have decreased by 99.8% since 1970, the application of classical measures (mechanical removal of the Mg-rich crust, incorporation of gypsum and manure to the soil), or newer methods (growing of Mg hyper-accumulating plants) can offer more lasting positive effects than those of 50 years ago. This study concluded that Mg-rich, alkaline dust deposition causes long-lasting anthropogenic soil degradation.

1 Introduction

Slovakia belongs to one of the countries with economical-valuable occurrence of natural- crystalline magnesite, and behind China, Russia, North Korea and Turkey belongs to the largest producer of magnesite in the world. Production is localised at two sites: Jelšava (Slovak magnesite processing plant) and Lubeník (Slovmag). Mining of magnesite and its processing to refractory materials is a very dusty operation that negatively influences whole ecosystems including biota. Landscapes have been changed, and craters, waste dumps and heaps have been formed (Huttmanová et al., 2015). Unfortunately, after the installation of new technologies and effective filters, considerably less attention was paid to the research of the locality than in the past. Therefore, some scientific literature used in this research is older and mostly written in Slovak.

Contrariwise with situation in Slovakia, Yang et al. (2012) had claimed that magnesite processing has expanded around the world mainly during the past 30 years, mainly in China, which nowadays accounts 44 % the production in the world. Despite relatively short period of production, the environment is already severely damaged in the surrounding of magnesite mining and calcination plants (Fu et al., 2011). In Slovakia, magnesite has been mined and processed for more than 90 years therefore there are many long-term experiences in the degradation of environment, but also in the reclamation of affected soil. Therefore, some methods successfully applied in land reclamation may be useful also on localities recently contaminated by magnesite dust. Soil contamination by Mg-rich, alkaline emissions is a serious, long-lasting problem.

The magnesite processing plant in Jelšava (founded in 1923) and in Lubeník (founded in 1934) did not produce large amounts of clinker and the common technology of shaft furnaces caused moderate dustiness. However, since 1958, a change in the processing technology (from shaft to rotation furnaces), increased production in new enterprise and insufficient filtration, caused significant dustiness in the surroundings of the factories, mainly along the direction of predominant winds from the north-west to the south-east. From a mineralogical perspective, Mg-rich, alkaline emissions contained 35%–50% amorphous MgO and 10%–20% of other minerals (such as periclas, dolomite, and calcite) (Šály & Minďáš, 1995). Near magnesite processing factories, the maximum permitted concentration of 150 t $km^{-2}year^{-1}$ (i.e. 12.5 g m^{-2} per 30 days) of alkaline dust deposition has been exceeded during 50 years. However, after installing new technologies and improved dust filters (Amertherm in 1984), the situation has improved (Bobro & Hančulák, 1997).

Alkaline dust emitted during magnesite processing has caused the anthropogenic alkalinisation of more than 12,700 ha of agricultural land and 6,600 ha of forests, as well as the contamination of water and damage of soil biota. In particular, the finest dust fractions (0.04–0.063 mm) contain the highest proportion of free MgO particles emitted furthest from the source. Because MgO particles and amorphous MgO have a large surface area, they are highly active, have high absorption ability for gases and liquids, and react chemically with substances in the soil and plant tissues. In the affected area, mainly highly active amorphous MgO has caused an increase in the original soil pH from slightly acidic (5.5–6.5) to alkaline (8.5–9.5) (Hronec et al., 1992).

Alkaline emissions reduce the production of plant biomass through direct devastation of vegetation by dustiness, as well as by damage to assimilatory organs. Alkaline solutions formed by the reaction of alkaline emissions with air humidity also directly harm the leaves and bark of plants (Machín & Navas 2000). Indirect damage to vegetation includes anthropogenic alkalinisation of soil with alkaline dust fallout, leading to the unavailability of macro- and micro-nutrients (Yang et al., 2012; Wang et al., 2015a) and thus decreased biological activity and diversity (Katuz et al., 2001; Mihál et al., 2015). In closer surroundings of magnesite processing plants, considerable deposition of Mg-rich, alkaline dust has caused vast damage of local plant communities. Currently, only several resistant species are present (Fazekaš et al., 2018). In localities most affected by alkaline dust deposition, the vegetation completely disappeared and a tight, solid Mg-rich, 5–8 mm thick crust was formed on the soil surface, inhibiting the proper soil functioning (Hronec et al., 1992; Fazekaš et al., 2019).

Previous information has demonstrated that all components of the environment near magnesite processing factories are considerably deteriorated, and despite the adoption of many measures, this unfavourable state continues. The amount of Mg-rich, alkaline fallout depends on both the distance from the source and the direction of predominant winds carrying and spreading alkaline emissions. Hence, in this work we investigated and evaluated the anthropogenic degradation of selected chemical and biological soil properties caused by alkaline emissions coming from magnesite processing factories in Lubeník and Jelšava. Soil properties were assessed along the direction of the prevailing winds (carrying and spreading alkaline emissions), at different distances from factories. Based on obtained results, an approach to land use in the studied localities was proposed.

2 Materials and methods

2.1 Study area

The territory affected by alkaline dust deposition extends in the valley along the River Muráň (48º60'–48º66' N, and 20º15'–20º27' E, about 270 m a.s.l.). It is located in the Revúcka Highland, a geomorphological complex of the Slovak Ore Mountains, and a sub-province of the Inner Western Carpathians. Long-term annual average rainfall at the area is 728 mm, and the average annual temperature is 8.2 °C. Prevailing winds in the territory are in the direction from north-west to south-east (Climate-Data.Org, 2019). The geology of the loaded area is very complex. The older Paleozoic is represented by granite, and the younger by magnesite, on which were developed Cambisols. Rendzic Leptosols were formed on Mesozoic limestones, dolomites, and shales. Cambisols were formed on Neogene gravel, Luvisols on Pleistocene loam, and Haplic Fluvisols were formed on Holocene alluvial deposits (Geological map, 2019).

2.2 Soil sampling and samples preparation

Soil sampling sites (Figure 1) were designed based on the quantity of fallen alkaline dust (g m^{-2} 30 $days^{-1}$) for the period 1980–1990. Values were published by Turčan Consulting (1992), and results were map processed. The studied area started in the north-west, approximately 1 km before the first magnesite processing factory in Lubeník, and continued down the alluvium of the River Muráň alongside the factory in Jelšava and finished in the south-east, behind the village of Gemerské Teplice. Following the direction of prevailing winds (north-west to south-east), 14 sampling sites covered by natural vegetation were selected. At each site, soil-sampling plots (30 m × 30 m) were designated, and from these, networks of 10 m × 10 m were assigned to collect nine soil subsamples per each plot (resulting in 126 subsamples). To represent the average conditions of the plot, the 9 soil subsamples were pooled and formed composite sample for each of the 14 sampling sites. Because in the past, most of the studied area was used as arable land, soil samples were collected from the topsoil (0–30 cm).

In the laboratory, soil samples were air dried at laboratory temperature (20 °C), ground and sieved via a mesh diameter of 2 mm, and for determination of organic carbon with a mesh size of 0.25 mm.

2.3 Soil analysis

Soil pH was measured potentiometrically in a 1:2.5 suspension of dry soil to distilled water; carbonates (CO_3^{2-}) volumetrically using 10% HCl; the content of available magnesium and calcium were analysed by method of Mehlich III (Mehlich, 1984); total soil organic carbon (C_T) was determined using the method of Tyurin (Kononova, 1966) by soil sample oxidation in the mixture of $K_2Cr_2O_7$ and H_2SO_4; and labile carbon (C_L) i.e. oxidisable by 5 mmol dm^{-3} $KMnO_4$ in an acidic medium of 2.5 mmol dm^{-3} H_2SO_4 according to Loginow et al. (1987).

Total content of heavy metals, magnesium and calcium were assayed after samples mineralization in the mixture of HF and $HClO_4$(Crock & Severson, 1980); and the available forms of heavy metals were extracted with 1 mol dm^{-3} HCl according to Rinkis method (Novozamsky et al., 1993). Subsequently, the elements were determined by atomic absorption spectroscopy using PU 9100X spectrometer (Philips).

For the determination of soil enzymatic activity were used standard methods, namely dehydrogenase activity (DEH) [E.C.1.1.1] was determined using method of Thalmann (1968), and catalase activity (CAT) [E.C.1.11.1.6] by method of Johnson & Temple (1964). Alkaline phosphatase (AlP) [E.C.3.1.3.1] as well as acid phosphatase (AcP) [E.C.3.1.3.2] by method of Tabatabai & Bremner (1969).

2.4 Statistical analysis

Each analysis was completed in three replications. Results shown in Figures 2, 4, and 5 represent the average values (mean ± SD). One-way analysis of variance (ANOVA) and the least significant difference (LSD) were used to compare investigated parameter means for the different sampling sites at P <0.05. A correlation matrix was used to assess the relationship between pH, content of carbonates, total and labile organic carbon, and the contents of total and available magnesium and calcium, enzymatic activity, and total and available forms of Cu, Ni, Pb, and Zn. For the expression of total and available Mg content dynamics following the direction of prevailing winds, quadratic polynomial regression models were used. All statistical analyses were performed using Statgraphics Centurion XV.I software (Statpoint Technologies, Inc., USA).

3 Results

3.1 Influence of alkaline dust fallout on basic chemical characteristics and quantity of heavy metals in soil

Alkaline dust generated during magnesite processing contains beside magnesium also the calcium, therefore Figure 2a, b and c presents the total and available contents of Mg and Ca determined at all 14 investigated sites. Results showed that sample sites close to factories 3–4 folds exceeded the natural regional background content of total Mg in topsoils (9.1–15.2 g kg^{-1}).

Consequently, the available Mg 3–68 fold exceeded very high content for texturally medium soils (>0.255 g kg^{-1}) at all grassland sampling sites, even at distance of 10 km from factories. The dynamics of changes in total as well as available Mg concentration in soil (depending on the direction of prevailing winds and therefore alkaline emissions spreading) are clearly documented in the quadratic polynomial trend (Figure 3a and b). While both forms of Ca showed no trends (Figure 3c and d), the highest contents of total Mg were found in the sampling sites near both magnesite processing plants, particularly in Jelšava. The contents of available Mg gradually increased in the direction of predominant winds, beginning near the factory in Lubeník, and the highest concentration was determined near the factory in Jelšava. The same sampling sites also had the highest pH values and carbonate content (Figure 3e and f).

Sites situated in close proximity to both sources of pollution recorded the highest values of soil pH_{H2O} 7.60–9.39. With increased distance from the factories, pH values, together with Mg content, demonstrated a decreasing tendency (pH_{H2O}7.04–7.98). Soil pH was highly significantly affected by the content of total and available magnesium (r = 0.788; r = 0.894; $P<0.001$) respectively, while calcium did not result in significant increase of soil pH. Accordingly, no significant relationship was found between total Mg and Ca and also between available Mg and Ca (Table 1). Increased carbonate concentration (CO_3^{2-}) more or less followed the localities heavily loaded by Mg-rich dust fallout and correlated with the available and total Mg content in the soil (Table 1, Figure 2). Conversely, there was no linkage between carbonate content and total or available calcium.

Figure 4 displays the concentrations of total and available zinc, copper, lead, and nickel on studied soils. The contents of analysed metals varied by sampling sites. This was also confirmed by analysis of variance. Increase of available Ni content was associated with an increase in the total Ca concentration (r = 0.663; $P < 0.01$) not Mg (r = 0.335; $P > 0.05$). In addition, the concentration of available Ni (r = 0.811; $P < 0.001$) and Cu (r = 0.566; $P < 0.05$) significantly correlated with the content of available Ca. We found no statistically significant relationship between total or available Mg and total or available forms of monitored heavy metals (Zn, Cu, Pb, and Ni).

3.2 Influence of alkaline dust deposition on the storage of soil organic matter and enzymatic activity

Soil degraded by high amount of Mg-rich, alkaline dust fallout, especially in localities where a solid Mg-rich crust has been formed on the surface, is characterised by low stock and altered quality of soil organic matter. Total organic carbon (C_T) was in range 5.4–24.3 g kg^{-1}, i.e. low content predominated (Figure 5a).

The dynamics of changes in total as well as labile carbon (C_L) content depending on the direction of alkaline emissions spreading is documented in the quadratic polynomial trend (Figure 6). The lowest contents of C_T and especially C_L were found in sampling sites 6–9 close to the factory in Jelšava. The same sampling sites contained the highest quantity of total but mainly available Mg (Figure 2a and b). Although the C_T content was lower in the localities most polluted with alkaline dust fallout (Figure 5a), there was no significant relationship between C_T and total and available Mg (Table 1). Conversely, there was a significant negative correlation between the labile fraction of organic matter (C_L) and the available Mg (r = -0.617; $P < 0.05$), suggesting that in the most affected areas, limited formation prevails and therefore a low quantity of new, labile organic matter. Formation of new, labile organic compounds was significantly impeded also by high pH_{H2O} values (r = -0.602; $P < 0.05$), as shown in Table 1.

ANOVA test disclosed significant differences in enzymatic activities on the sampling sites. The highest activity of dehydrogenases, catalase, alkaline as well as acid phosphatases were reported in the soil collected from sites 11 and 13, which also contained the highest quantity of C_L (Figure 5).

We determined a significant decrease in soil enzymatic activity because of increased Mg content (Table 1). In particular, depending on the available Mg content, the alkaline phosphatase, acid phosphatase, dehydrogenase and catalase activities significantly decreased (r = -0.613; -0.640; -0.574; -0.610; $P < 0.05$).

Moreover, the activity of acid phosphatase was negatively influenced by increased pH_{H2O} (r = -0.608; $P < 0.05$). Conversely, alkaline phosphatase activity increased in accordance with the content of available Ca (r = 0.538; $P < 0.05$). Thus, in the affected area, the excess of available Mg, as well as increased pH values, and decreased content of labile soil organic matter were associated with Mg-rich, alkaline dust deposition and caused a significant decrease in soil enzymatic activity.

4 Discussion

4.1 Influence of Mg-rich alkaline dust deposition on chemical properties of affected soil

Calcium and magnesium are important macronutrients necessary for all living organisms. However, problems might arise due to not only their shortage, but also their excess. Excess of both macronutrients has a negative effect on plants through increased pH, reduced availability of many micronutrients, and also heavy metals (Balakrishnan et al., 2000; Guo et al., 2016).

The natural regional background content of total Mg in topsoils unaffected by alkaline deposition occurs within a range of 9.1–15.2 g kg^{-1} (Čurlík & Šefčík, 1999). Only 3 out of 14 sampling sites (sites 3, 5, and 14) corresponded to this range, while the others contained high to extremely high total Mg concentrations as result of anthropogenic enrichment.

Available Mg far exceeded very high content for texturally medium soils (> 0.255 g kg^{-1}) at all sampling sites, even at sites 13 and 14, which according to the data referred by Turčan Consulting (1992) were minimally affected by Mg-rich, alkaline dust deposition. However, sampling sites 13 and 14 are located in the direction of predominant winds, behind the magnesite processing factories.

Results achieved in this study indicated that Mg-rich, alkaline dust caused long-lasting soil degradation. The evidence is the relationship of our results with the findings of Turčan Consulting (1992), who during 10 years (1980–1990) measured the deposition of alkaline dust in the affected area. Locations assigned by Turčan Consulting (1992) as having the highest dust deposition (> 41–25 g m^{-2} 30 $days^{-1}$), corresponded to sampling sites 6–9, where still nowadays (after 40 years) we found the highest Mg total but mainly Mg available (Figures 1 and 2). These sampling sites also had the highest pH values and carbonate content. Unfortunately, since 1990 no detailed spatial research of alkaline dust deposition in the affected locality has been carried out.

According to Hronec (1992), natural leaching in the soil-climatic conditions of Slovakia can reduce total Mg content in soil on a yearly basis by 26–34 kg ha^{-1}, provided that additional Mg-rich alkaline dust does not enter the soil. However, Brozmanová (2018) stated that there are still up to 20 tons of particulate matter yearly emitted into the environment from magnesite processing factories every year. However, this quantity represents only 0.25% compared to the situation in 1970, when 7,846 t $year^{-1}$ were emitted. These values have proven that adopted dust reduction measures are more effective. Conversely, Bobro & Hančulák (1997) stated that although there is no longer a massive supply of magnesium to the soil, the supply is still active and it is likely that soils will not be able to get rid of the excess of this element through natural processes.

Before the intensification of production in magnesite processing factories, the initial pH of local topsoil was 5.5–6.5 (Hronec et al., 1992). At present, in deteriorated areas, neutral to alkaline soil pH prevails (Figure 2d). Since increased pH and carbonate content more or less copied the localities heavily loaded by alkaline dust deposition and correlated with the Mg content in the soil, it can be concluded that in addition to MgO, $Mg(OH)_2$, $4MgCO_3.(MgOH)_2.4H_2O$, soil degradation is also dominated by $MgCO_3$, i.e. magnesite. Our assumption was confirmed by data published by Baluchová et al. (2011), who investigated the mineralogical composition of dust fallout from 2006–2008 in the Jelšava region. They identified magnesite as the dominant mineral (>60%), while periclase had variable content, dolomite presented <10%, and calcite <5%. Furthermore, Baluchová et al. (2011) stated that beside the magnesite processing plant, an important source of magnesite in alkaline dust could be mining, as well as abandoned surface mines. Conversely, the chemical composition of alkaline dust fallout reported by Šály and Mindáš (1995) showed a 35%–50% dominance of amorphous MgO, and 10%–20% of other minerals (periclase, dolomite, and calcite). This information showed that the chemical composition of alkaline dust has changed over time, as confirmed Baluchová et al. (2011). They reported that a decreasing proportion of periclase and an increasing proportion of magnesite in dust particles indicate that dust-reduction measures in Jelšava and Lubeník are effective.

Considerable spatial differences in the concentration of Zn, Cu, Pb, and Ni at studied sites (Figure 4) might be due to the changing atmospheric pressure and other meteorological factors during the deposition of alkaline dust in the soil. The quantity of total and available forms of analyzed heavy metals was lower than limit values reported by the U.S. Environmental Protection Agency (1993). Therefore assayed soil was classified as unpolluted.

In studied locality, no significant linkage between Mg and monitored heavy metals were found. However, during the processing of magnesite, trace amounts of some elements (Cu, Ni, As) were emitted together with Mg emissions into the atmosphere (Hronec et al., 1992). Potentially toxic elements (Zn, Cu, Cr and especially Mn) are directly bound to the emitted dust and pollute soil and other components of the environment (Fazekašová et al., 2017). Hančuľák & Bobro (2004) reported that in 1999, the alkaline dust in Jelšava contained 394,500 ppm Mg, 13,100 ppm Ca, >1 ppm Cd, 75 ppm Cu, 5 ppm Ni, >1 ppm Pb and 400 ppm Zn. Increased concentrations of Zn, Cu, Cd, Ni can be attributed mainly to alkaline dust fallout, but also to fuel oil used in the past.

The mobility and availability of metals are influenced by number soil properties and processes. Despite not all of them are equally important for each metal, but some properties are of greater importance than others, in particular: the quantity and quality of organic matter, soil texture, pH, sorption capacity, the forms in which cations occur, oxidation–reduction potential, and the activity of microorganisms and concentrations of macro- and micronutrients (Ashworth & Alloway 2004; Chojnacka et al. 2005).

4.2 Influence of Mg-rich alkaline dust deposition on soil organic matter and enzymatic activity

In soils affected by high amount of Mg-rich, alkaline dust deposition, the microbial activity, biomass production is limited, original vegetation is replaced by vegetation resistant to high alkalinity, Mg concentration, unfavourable and macro- and micro-nutrients ratio (Kautz et al., 2001; Blanár et al., 2019). Results in this study also demonstrated the disruption of soil biological properties as well as soil organic matter quantity and quality. The lowest quantities of total and labile organic carbon were found in sampling sites the most loaded by total, but mainly available Mg (Figures 2a, b and 5a). Despite the C_T content was lower in sites the most polluted with alkaline deposition, the relationship between C_T and Mg was not significant (Table 1). Similar relationships were confirmed also by Fu et al. (2011) and Yang et al. (2012). On the other side, significant negative correlation between the C_L and the available Mg suggest, that in localities containing high excess of available Mg, lower stock of newly formed organic matter prevailed. Since plants are the main source of fresh organic matter, their shortage resulted in low stock of labile soil organic matter, mainly in the areas the most affected by excess of available Mg as well as high alkalinity.

The quantity of labile organic carbon was significantly related with soil microbial activity. According to Lemanowicz (2019), dehydrogenases together with catalase activities provide information regarding microbial activity in soil. Alkaline and acid phosphatases catalyse the hydrolysis of organic phosphorus compounds and their transformation to inorganic phosphorus (Nannipieri et al., 2011). The activity of all studied enzymes significantly decreased with higher content of available Mg what proved that soil microbial activity was negatively influenced by excess of available Mg. Decline in acid phosphatase activity with increased alkalinity was in agreement with research of Dick et al. (2000) who stated that the optimum pH of soil for the activity of acid phosphatase is 4.0–6.5, while for alkaline phosphatase it is 9.0–11.0. Błońska et al. (2016) consider pH as dominant factor affecting the total microbial abundance and activity of enzymes.

Accordingly with our research, Yang et al. (2012) observed significant decrease in microbial biomass carbon and nitrogen, and potential net N mineralization rate with increased soluble Mg content and pH values. Bartkowiak et al. (2017) and Lemanowicz (2018) highlight the role of enzymatic activity as an early indicator of changes in the intensity of microbial processes as response on soil degradation. Sufficient content and quality of soil organic matter is important for intensive microbial activity. Moreover, organic matter protects and immobilizes enzymes. It stabilizes the protein structure of enzymes, decreases their sensitivity to negative changes caused by environmental factors (Zhang et al., 2015).

A significant rise in enzymatic activity was associated with an increase in both total and labile soil organic carbon content (Table 1). Thus, in addition to filters that effectively capture alkaline emissions, one of the most important measures for enhancing the enzymatic activity of soil degraded by alkaline dust deposition is the enrichment of soil with organic matter, as was confirmed by our results.

4.3 Reclamation and land use possibilities around magnesite processing plants

Reclamation methods of land degraded by Mg-rich, alkaline dust deposition from magnesite processing plants have already been suggested (Holobradý, 1981; Hronec et al., 1992). However, their implementation only seems to be more effective currently, as alkaline emissions have decreased by 99.75% compared to 1970 (that is, to 20 tons of particulate matter per year) (Brozmanová, 2018). Therefore, an effective revitalisation of the affected area could be started by procedures already known from the past.

Classical methods suggest that from the most affected areas the impermeable Mg-rich crust should be mechanically removed, milled, and provided not containing excess concentration of heavy metals or other pollutants it can be used as a good magnesium fertilizer on acidic or sandy soils. Holobradý (1981) suggested use chemical reclamation at each locality where the available Mg exceeded 2,000 mg kg^{-1}. The dose of ameliorative matter should be calculated based on the concentration of available Mg in the soil. In practise, the reclamation was based on a mechanical loosening of the soil with concurrent incorporation: 10–50 t ha^{-1} of gypsum, or 10–50 t ha^{-1} of citric-gypsum (waste from citric acid production), or 2,000 L ha^{-1} of sulphite leaches (pulp waste containing $Ca(HSO_3)_2$), or ground sulphur. After the above-mentioned chemical melioration, soluble magnesium sulphate is formed and gradually leached out of the soil by rainwater. To increase soil microbial diversity and biological activity, it is recommended to incorporate 40–50 t ha^{-1} of farmyard manure (FYM) every 3–4 years. In case of shortage the FYM, it is possible to use composts, or recently recommended manure-biochar, compost-biochar composite composts (El-Naggar et al., 2015). The effect of biochar on reclamation of soils degraded by excess of magnesium was not studied yet. However, enhancing the soil with biochar can improve chemical and physical soil properties and hence stimulate biological activity (Beesley et al., 2011). Therefore, it is important to study the effect of biochar on soil degraded by alkaline dust deposition. On the other side, biochar enriched by Mg hydr(oxid) was stated as having the potential to prevent phosphorus leaching from organic soils (Riddle et al., 2019).

Similar problem with excess of Mg, but coming from irrigation water was solved by Vyshpolsky et al. (2008; 2010). They highlighted positive effect of phosphogypsum application (by-product of the phosphate fertilizer industry) at a dose of 4.5 t ha^{-1}, before the snowfall, every 4–5 years for optimizing the ionic balance of soil with heavily exceeded levels of Mg^{2+} in Southern Kazakhstan. Wang et al. (2015b) successfully decreased Mg content in soil samples using anionic polyacrylamide and calcium dihydrogen phosphate and controlled leaching of soil columns.

More recent methods include biological reclamation, which involves the growing of Mg hyper-accumulating plants that, after composting, could be used as an organic fertilizer naturally enriched with Mg. This method can be used at localities with the content of Mg less than 2,000 mg kg^{-1}. Markert (1992) in Parzych & Astel (2018) stated that in general, the natural Mg content in the dry plant biomass is 1,000–3,000 mg kg^{-1}. Despite the plants with higher Mg accumulation that have been identified, they did not grow in the soil with excessive Mg content: *Stellaria nemorum* (L.) 5,716±746 mg kg^{-1}, *Urtica dioica* (L.) 5,127±581 mg kg^{-1}, *Caltha palustris* (L.) 4,965±602 mg kg^{-1} (Parzych et al., 2018). Higher Mg accumulation was identified in plants growing in affected area and forming large monocultures: *Elytrigia repens* (L.) 21,208 mg kg^{-1}, *Phragmites australis* (Cav.) Trin. 6,860 mg kg^{-1} and *Agrostis stolonifera* (L.) 5,419 mg kg^{-1} (Fazekaš et al., 2018). Of these plants, only *Phragmites australis* was characterised by high biomass production, that is 12.7 t ha^{-1} of dry matter (Demko et al., 2017). Therefore, its use as a source of biomass bio-forticated by Mg for compost production can be considered. Effective phytomeliorative removing of excess Mg^{2+} from lightly Mg-contaminated soil was demonstrated by Wang et al. (2014) using *Aneurolepidium chinense* (Trin.) and

Puccinellia distans(Jacq.) Parl. with the application of $Ca(H_2PO_4)_2$. H_2O. They stated that planting *A. chinense* and*Elymus dahuricus* (L.) with the application of $Ca(H_2PO_4)_2$. H_2O could accelerate the vegetation restoration in moderately and severely Mg-contaminated soil.

Hronec et al. (1992) suggested that land containing <1,000 mg kg^{-1} of available Mg could be converted gradually into arable land. As already mentioned, in the past, the land near factories was used for agricultural purposes. However, after soil contamination with alkaline dust, especially during the period 1958–1984, these soils were excluded from agricultural use. Based on the available Mg content (Figure 2b), we highlight the possibility of reusing the land at sampling sites 4, 5, 13, and 14 (Figure 1) for agricultural production. To accelerate the removal of excessive Mg, we recommend use the phytoremediation.

At sites with content of available Mg higher than 1,000 mg kg^{-1} (where only limited species of Mg tolerant vegetation grow), care must be taken to maintain the vegetation covering the soil (sites 1, 2, 3, and 6–11). According to the Shannon Index, plant diversity on the studied sited was extremely low (0.0) to middle low (1.5) (Fazekaš et al., 2018). It is necessary to maintain a favourable state of natural vegetation, for example by mulching of meadows, thereby limiting the spread of invasive plants, as well as avoiding the removal of aboveground plant biomass. Sufficient plant biomass is necessary to increase the storage of soil organic matter, which is an important factor in increasing the biological and enzymatic activity (Mganga et al., 2019; Nyawade et al., 2019; Wei et al., 2019), even in soils deteriorated by alkaline dust deposition (Table 1). Feeding cattle with biomass produced on deteriorated areas is not appropriate due to the plants dusting. Consequently, many diseases that threaten animals occur (nervous, respiratory and digestive disorders, diarrhoeas, weight loss, disruption of the sexual cycle, miscarriages) (Hronec et al., 1992; Machín & Navas 2000). In addition, biomass is of low nutritional value, as in the most affected areas dominate plants:*Elytrigia repens* , *Agrostis stolonifera* , *Puccinellia distans* , *Chenopodium glaucum* (L.), invasive *Solidago canadensis* (L.), and recently also *Phragmites australis* , known as invasive in some alkaline sites (Bart et al., 2006).

An interesting use of deteriorated area could be the growing of plants for energy purposes. A prospective plant is *Phragmites australis* , which is abundant in humid locations with pH above 9 (Huttmanová et al., 2015) and has spontaneously appeared in the locality only recently. Natural production in Slovakia is 12.7 t ha^{-1} of dry matter with high-energy storage of 221.622 GJ ha^{-1}(Demko et al., 2017). Therefore, it is more profitable to use*Phragmites australis* for direct biomass combustion, or production of biofuel pellets, than for the production of biogas and methane. Alternatively, Suhai et al. (2016) stated that this plant species is a sustainable and renewable resource for the production of bioethanol.

At present, when the presence of Mg-rich, alkaline dust in the soil has been significantly reduced, the application of these measures can offer a more lasting positive result compared to the previous period, when the high fallout of alkaline dust had not allowed successful land reclamation in the vicinity of magnesite processing plants. Subsequently, gradually returning the soil and landscape in the affected area to a more productive state will be possible.

5 Conclusions

Mg-rich, alkaline dust causes long-lasting soil degradation. The evidence is the relationship of findings reached in this study with the results of dust deposition measured during 10 years, 40 years ago.

Sites close to factories 3–4 folds exceeded the natural regional background content of total Mg in topsoils. Available Mg 3–68 fold exceeded very high content for texturally medium soils (>0.255 g kg^{-1}) at all grassland sampling sites, even at distance of 10 km from factories, along the direction of the alkaline emissions spreading. Areas close to factory contained up to 14.4–17.4 g kg^{-1} of available Mg. Studied heavy metals (Zn, Cu, Pb and Ni) did not exceeded limit values and their concentration was not in linkage with the content of Mg, as well as enzymatic activity.

Higher excess of available Mg caused significant increase of soil pH (up to 9.39) and worsened conditions for the growth of vegetation. Since plants are the main source of fresh organic matter, their shortage resulted in low stock of labile soil organic carbon (0.50–0.96 g kg^{-1}) and consequently weaker enzymatic activity. Thus, in addition to effective, alkaline emissions capturing filters, an important measure supporting the microbial activity of affected soil is enrichment by organic matter.

At present, when the entry of Mg-rich, alkaline dust into the soil has been significantly reduced (by 99.75% compared to year 1970), the application of measures can bring more lasting positive results than in the past.

Nevertheless, classical methods are still the most effective for reclaiming the most affected areas. The impermeable Mg-rich crust should be mechanically removed, milled, and provided not containing excess concentration of heavy metals or other pollutants it can be used as a magnesium fertilizer. Soil with available Mg exceeded 2,000 mg kg^{-1} has to be treated chemically, by incorporating gypsum compounds, sulphur, as well as high doses of farmyard manure, or composts, manure-biochar, compost-biochar composite composts.

Recent methods are applicable to less affected areas. These involve the growing of Mg hyper-accumulating plants that, after composting, can be used as an organic fertilizer enriched with Mg, or growing plants that can be used for energy purposes. In the studied area, among naturally occurring vegetation, only *Phragmites australis* is characterised by high biomass production.

Sampling sites, where the available Mg decreased under the critical level of 1,000 mg kg^{-1}, can be reused for agricultural production. Over the whole affected area, care must be taken to maintain a favourable state of natural vegetation. Consistent application of measures will enable a gradual return of the soil and landscape to a more productive state.

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Conflict of Interest Statement

The authors declare that they have no conflict of interest.

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Figure legends

FIGURE 1 Map of distribution of sampling sites 1–14 and magnesite processing factories in Lubeník (A) and Jelšava (B). Data on the amount of alkaline emissions deposition are based on findings of Turčan consulting (1992).

FIGURE 2 Mean ± standard deviation of a) total and b) available magnesium, c) total and available calcium, d) pH_{H2O} and carbonates content (CO_3^{2-}) in soil affected by alkaline dust deposition in the vicinity of magnesite processing factories; sampling sites correspond with that in Figure 1

FIGURE 3 The dynamic changes in a) total and b) available magnesium, c) total and d) available calcium content, e) pH_{H2O} and f) carbonates content depending on the direction of alkaline emissions spreading

FIGURE 4 Mean ± standard deviation of a) total and b) available zinc, copper, lead and nickel in soil affected by alkaline dust deposition in the vicinity of magnesite processing factories; sampling sites correspond with that in Figure 1

FIGURE 5 Mean ± standard deviation of a) total (C_T) and labile (C_L) soil organic carbon, b) activity of alkaline and acid phosphatases, c) dehydrogenases and d) catalase, in soil affected by alkaline dust deposition in the vicinity of magnesite processing factories; sampling sites correspond with that in Figure 1

FIGURE 6 The dynamic changes in a) total (C_T) and b) labile (C_L) organic carbon depending on the direction of alkaline emissions spreading

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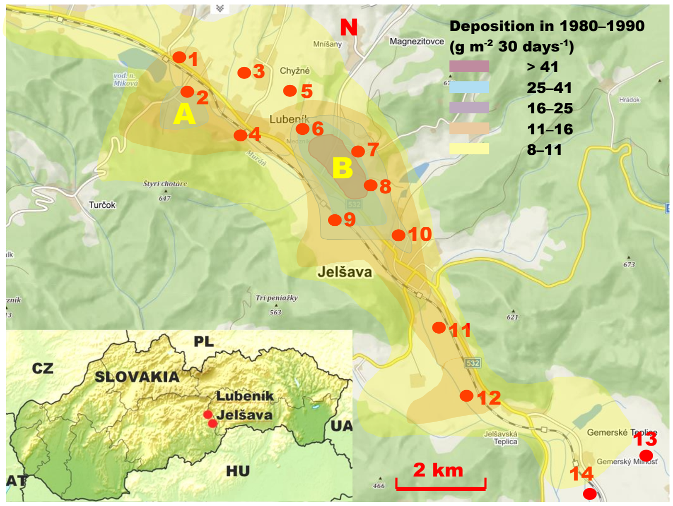
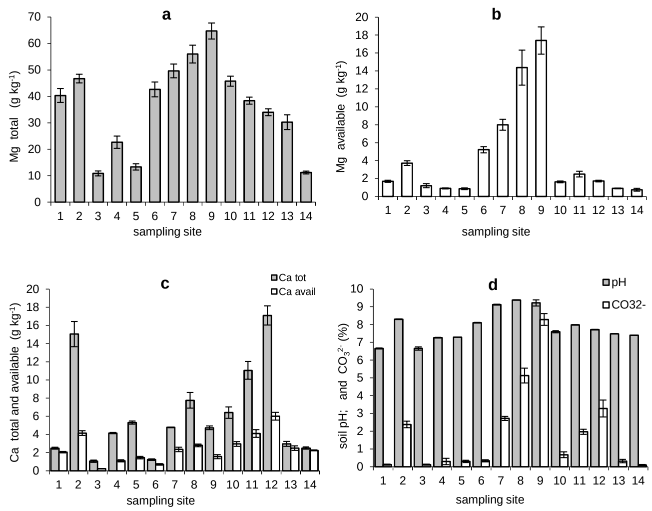
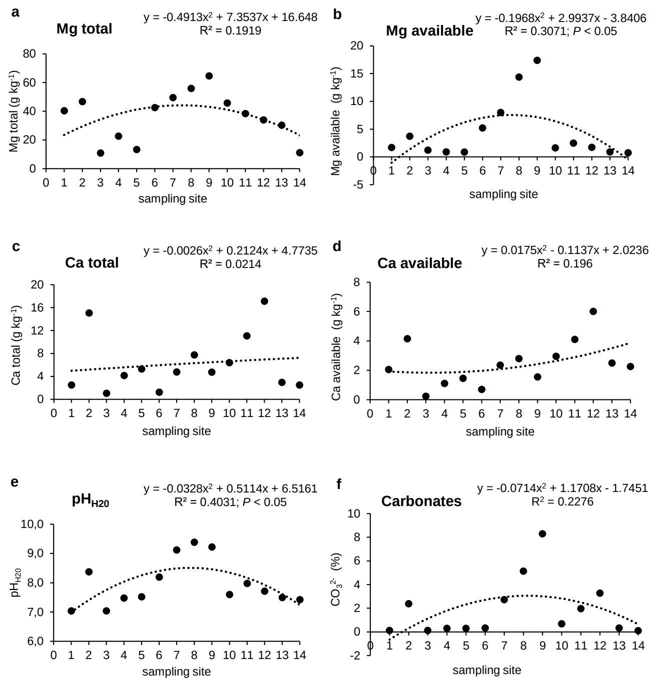
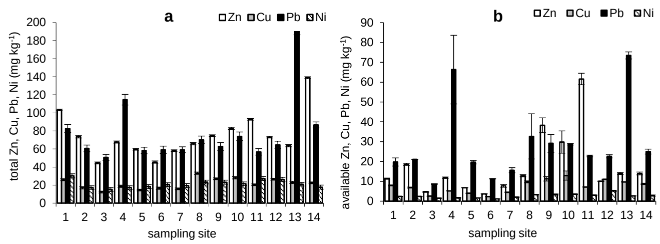
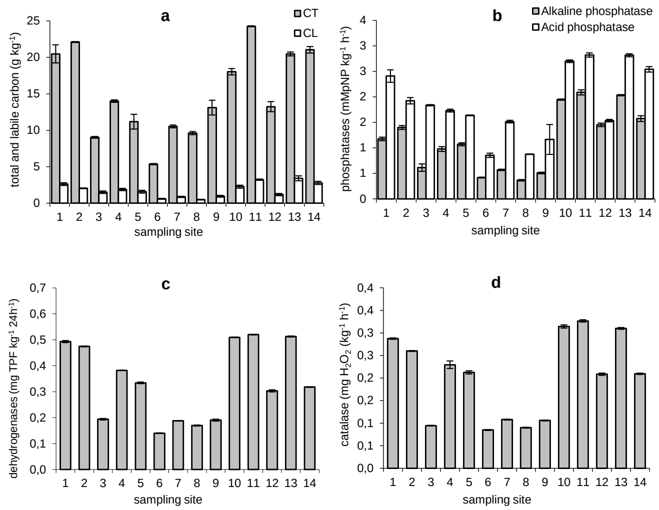
`Table1.docx` available at https://authorea.com/users/323526/articles/455790-secondary-enrichment-of-soil-by-alkaline-emissions-the-specific-form-of-anthropogenic-soil-degradation-near-magnesite-processing-factories-and-possibilities-of-land-management

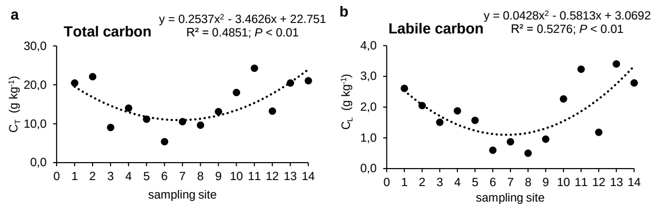

N
Deposition in 1980–1990
(g m-2 30 days-1)
> 41
25–41
16–25
11–16
8–11
1
2
3
4
5
6
7
8
9
10
11
12
13
14
A
B
Lubeník
Jelšava
Chyžné
Magnezitovce
Mníšany
Turčok
Štyri chotáre
Tri peniažky
Gemerské Teplice
Jelšavská Teplica
2 km
CZ
PL
SLOVAKIA
Lubeník
Jelšava
UA
AT
HU


a
Mg total (g kg⁻¹)
sampling site
b
Mg available (g kg⁻¹)
sampling site
c
Ca tot
Ca avail
Ca total and available (g kg⁻¹)
sampling site
d
pH
CO32-
soil pH; and CO3 2- (%)
sampling site


a
Mg total
y = -0.4913x² + 7.3537x + 16.648
R² = 0.1919
Mg total (g kg⁻¹)
sampling site
b
Mg available
y = -0.1968x² + 2.9937x - 3.8406
R² = 0.3071; P < 0.05
Mg available (g kg⁻¹)
sampling site
c
Ca total
y = -0.0026x² + 0.2124x + 4.7735
R² = 0.0214
Ca total (g kg⁻¹)
sampling site
d
Ca available
y = 0.0175x² - 0.1137x + 2.0236
R² = 0.196
Ca available (g kg⁻¹)
sampling site
e
pHH20
y = -0.0328x² + 0.5114x + 6.5161
R² = 0.4031; P < 0.05
pHH20
sampling site
f
Carbonates
y = -0.0714x² + 1.1708x - 1.7451
R² = 0.2276
CO3²⁻ (%)
sampling site


a
Zn Cu Pb Ni
total Zn, Cu, Pb, Ni (mg kg-1)
sampling site
b
Zn Cu Pb Ni
available Zn, Cu, Pb, Ni (mg kg-1)
sampling site


a
CT
CL
total and labile carbon (g kg-1)
sampling site
b
Alkaline phosphatase
Acid phosphatase
phosphatases (mMpNP kg-1 h-1)
sampling site
c
dehydrogenases (mg TPF kg-1 24h-1)
sampling site
d
catalase (mg H2O2 (kg-1 h-1)
sampling site



a
Total carbon
y = 0.2537x² - 3.4626x + 22.751
R² = 0.4851; P < 0.01
C_T (g kg⁻¹)
sampling site
b
Labile carbon
y = 0.0428x² - 0.5813x + 3.0692
R² = 0.5276; P < 0.01
C_L (g kg⁻¹)
sampling site