

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

Nora Pollakova<sup>1</sup>, Ján Hamar<sup>1</sup>, Vladimír Simanský<sup>1</sup>, Agata Bartkowiak<sup>2</sup>, and Joanna Lemanowicz<sup>2</sup>

<sup>1</sup>Slovak University of Agriculture in Nitra

<sup>2</sup>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). Since 1984, adopted measures have improved the emissions situation; however, this has resulted in insufficient attention being paid to soil properties. Therefore, 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 the highest excess of magnesium in the sampling sites near both factories. Total and available Mg significantly affected soil pH, carbonate content, and conductivity, while the content of monitored heavy metals was not related to Mg. In localities with higher excess of available Mg, lower stock of newly formed organic matter prevailed with consequently weaker enzymatic activity. Therefore, enrichment by organic matter provides a measure to support the biological 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 soil degradation.

## 1 Introduction

Slovakia is rich in natural crystal magnesite, and belongs to the largest producer of magnesite in the world behind China, Russia, North Korea and Turkey. Production is localised at two sites: Jelšava (Slovak magnesite processing plant) and Lubeník (Slovmag). Mining of magnesite and its subsequent processing to refractory materials is a very dusty process 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 developed rapidly around the world during the past thirty 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 & Mindáš, 1995). Near magnesite processing factories, the maximum permitted concentration of  $150 \text{ t km}^{-2}\text{year}^{-1}$  (i.e.  $12.5 \text{ g m}^{-2}$  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 more than 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 acid (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 (Kautz et al., 2001; Mihál et al., 2015). In closer surroundings of magnesite processing plants, considerable deposition of Mg-rich, alkaline dust has led to the collapse of indigenous 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 influencing ecologically important soil functions (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 ( $48^{\circ}60'$ – $48^{\circ}66'$  N, and  $20^{\circ}15'$ – $20^{\circ}27'$  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 average annual rainfall at the site is 728 mm, and the average annual temperature is  $8.2^{\circ}\text{C}$ . 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 ( $\text{g m}^{-2}$  30 days<sup>-1</sup>) 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 ( $30 \text{ m} \times 30 \text{ m}$ ) were designated, and from these, networks of  $10 \text{ m} \times 10 \text{ m}$  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 combined to form a 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 through 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 ( $\text{CO}_3^{2-}$ ) volumetrically using 10% HCl; electrical conductivity (EC) using a Conductometer HANNA DiST 6; hydrolytic acidity by method of Kappen in solution of  $1 \text{ mol dm}^{-3}$   $\text{CH}_3\text{COONa}$  (dry soil to solution ratio 1:2.5) and was quantified with  $0.1 \text{ mol dm}^{-3}$  NaOH; the content of available magnesium and calcium were analysed by method of Mehlich III (Mehlich, 1984); total soil organic carbon ( $\text{C}_T$ ) was analysed by the Tyurin method (Kononova, 1966) by sample oxidation in the mixture of  $\text{K}_2\text{Cr}_2\text{O}_7$  and  $\text{H}_2\text{SO}_4$ ; and labile carbon ( $\text{C}_L$ ) i.e. oxidisable by  $5 \text{ mmol dm}^{-3}$   $\text{KMnO}_4$  in an acidic medium of  $2.5 \text{ mmol dm}^{-3}$   $\text{H}_2\text{SO}_4$  according to Loginow et al. (1987).

Total concentrations of heavy metals and content of total magnesium and calcium were assayed after mineralisation of the samples in the mixture of HF and  $\text{HClO}_4$  (Crock & Severson, 1980); and available forms of heavy metals were extracted with  $1 \text{ mol dm}^{-3}$  HCl according to Rinkis method (Novozamsky et al., 1993). The total contents and available forms were determined by applying the method of atomic absorption spectroscopy with a PU 9100X spectrometer (Philips).

Activities of some redox and hydrolytic soil enzymes were determined according to standard methods, namely dehydrogenase activity (DEH) [E.C.1.1.1] was assayed with the Thalmann method (1968), and catalase activity (CAT) [E.C. 1.11.1.6] by the Johnson & Temple method (1964). Alkaline phosphatase (AlP) [E.C. 3.1.3.1] and acid phosphatase (AcP) [E.C. 3.1.3.2] were performed with the Tabatabai & Bremner (1969) method.

## 2.4 Statistical analysis

Each analysis was completed in three replications. Results shown in Tables 1, 3, and 5 represent the average values (mean  $\pm$  SD). One-way analysis of variance (ANOVA) and the least significant difference (LSD) method 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, electrical conductivity, hydrolytic acidity, 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 properties and content of heavy metals in soil

Alkaline dust generated during magnesite processing contains beside magnesium also the calcium, therefore Table 1 presents the total and available contents of Ca and Mg 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<sup>-1</sup>).

Consequently, the available Mg 3–68 fold exceeded very high content for texturally medium soils (> 0.255 g kg<sup>-1</sup>) at all grassland sampling sites, even at distance of 10 km from factories. The dynamics of changes in total as well as available Mg content in soil (depending on the direction of prevailing winds and therefore alkaline emissions spreading) are clearly documented in the quadratic polynomial trend (Figure 2c and d). While both forms of Ca showed no trends (Figure 2a and b), 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 the prevailing 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, carbonate content, and the lowest hydrolytic acidity (Figure 2e and f).

Sites situated in close proximity to both sources of pollution recorded the highest values of soil pH<sub>H2O</sub> 7.60–9.39. With increased distance from the factories, pH values, together with Mg content, demonstrated a decreasing tendency (pH<sub>H2O</sub> 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 cause statistically significant increase of soil pH. Accordingly, no significant relationship was found between total Mg and Ca and also between available Mg and Ca (Table 2). As expected, the lowest hydrolytic acidity (H) was observed in areas most affected by alkaline dust deposition (Table 1). Increased carbonate concentration (CO<sub>3</sub><sup>2-</sup>) 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 (Tables 1 and 2). Conversely, there was no linkage between carbonate content and total or available calcium. Both total and available Mg (as well as carbonates) contributed to the significant increase in soil conductivity ( $r = 0.787$ ;  $0.870$  and  $0.781$ ;  $P < 0.001$ ).

Table 3 displays the contents of total and available zinc, copper, lead and nickel on studied sites. The contents of analysed metals varied by sampling sites. This was also confirmed by analysis of variance. Table 4 clearly demonstrates that the increase of available Ni content was associated with an increase in the total Ca content ( $r = 0.663$ ;  $P < 0.01$ ) not Mg ( $r = 0.335$ ;  $P > 0.05$ ). In addition, the content 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 content 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 content and altered quality of soil organic matter. Total organic carbon (C<sub>T</sub>) was in range 5.4–24.3 g kg<sup>-1</sup>, i.e. low content predominated (Table 5).

The dynamics of changes in total as well as labile carbon (C<sub>L</sub>) content depending on the direction of alkaline emissions spreading is documented in the quadratic polynomial trend (Figure 3). The lowest contents of C<sub>T</sub> and especially C<sub>L</sub> 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 (Table 1; Figure 2). Although the C<sub>T</sub> content was lower in the localities most polluted with alkaline dust fallout (Table 5), there was no significant relationship between C<sub>T</sub> and total and available Mg (Table 2). Conversely, there was a significant negative correlation between the labile fraction of organic matter (C<sub>L</sub>) 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<sub>H2O</sub> values ( $r = -0.602$ ;  $P < 0.05$ ), as shown in Table 2.

The results of the ANOVA test (Table 5) showed a significant effect on the changes in activity of study enzymes for the sampling sites. The highest activity of DEH (0.520 mg TPF kg<sup>-1</sup>24h<sup>-1</sup>), catalase (0.327 mg H<sub>2</sub>O<sub>2</sub> kg<sup>-1</sup>h<sup>-1</sup>), AIP (2.087 mMpNP kg<sup>-1</sup>h<sup>-1</sup>) and AcP (2.819 mMpNP kg<sup>-1</sup>h<sup>-1</sup>) were reported in the soil

collected from sites 11 and 8, which also contained the highest quantity of  $C_L$ .

We determined a significant decrease in soil enzymatic activity because of increased Mg content (Table 2). In particular, depending on the available Mg content, the alkaline phosphatase, acid phosphatase, dehydrogenase and catalase activities significantly decreased ( $r = -0.613$ ;  $r = -0.640$ ;  $r = -0.574$ ;  $r = -0.610$ ;  $P < 0.05$ ). Moreover, the activity of acid phosphatase was negatively influenced by increased  $pH_{H_2O}$  ( $r = -0.608$ ;  $P < 0.05$ ). 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<sup>-1</sup> (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 enhancement.

Available Mg far exceeded very high content for texturally medium soils ( $> 0.255$  g kg<sup>-1</sup>) 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 the prevailing 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 ( $> 41\text{--}25$  g m<sup>-2</sup> 30 days<sup>-1</sup>), corresponded to sampling sites 6–9, where still nowadays (after 40 years) we found the highest Mg total but mainly Mg available (Table 1). These sampling sites also had the highest pH values, carbonate content, and the lowest hydrolytic acidity. 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<sup>-1</sup>, 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<sup>-1</sup> 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 (Table 1). 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)<sub>2</sub>, 4MgCO<sub>3</sub>·(MgOH)<sub>2</sub>·4H<sub>2</sub>O, soil degradation is also dominated by MgCO<sub>3</sub>, 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 content of Zn, Cu, Ni and Pb at studied sites (Table 3) might be due to the changing atmospheric pressure and other meteorological factors during the deposition of alkaline dust in the soil. The values of the heavy metals under study classify the analysed soils as unpolluted. All soil samples had lower values of total and available heavy metal content than limits reported by the U.S. Environmental Protection Agency (1993).

In studied locality, no significant linkage between Mg and monitored heavy metals were found (Table 4). 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 (Fazekášová et al., 2017). Hančulák & 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.

Mobility and availability of metals are controlled by many chemical and biochemical processes in soils. Not all are of the same importance for each metal, and this largely depends on many soil properties such as: organic matter content; the content of clay fraction; 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). Disruption of soil biological properties as well as soil organic matter content and quality demonstrated also results in this study. The lowest contents of total and labile organic carbon were found in sampling sites the most loaded by total, but mainly available Mg (Tables 1, 5). 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 2). 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 content of labile soil organic matter significantly related with soil microbial activity. According to Lemanowicz (2019), activity of the dehydrogenases could be considered a good indicator of the oxidative metabolism in soils, and together with catalase are used to give information on the microbial activities 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 and for alkaline phosphatase is 9.0–11.0. According to Błońska et al. (2016), pH is a major factor affecting the total microbial count 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. The enzymatic activity is an early indicator of changes in the level of intensity of biological processes and the level of soil degradation, and it is usually correlated with its physical and chemical properties (Bartkowiak et al., 2017; Lemanowicz, 2018). Organic matter offers a protective function towards enzymes, which are thus immobilised. It has a positive effect on the stability of protein structure, decreasing the sensitivity to negative changes triggered by environmental factors (Zhang et al., 2015).

A significant increase in enzymatic activity was associated with an increase in both total and labile soil organic carbon content (Table 2). 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 used as a good magnesium fertilizer on acidic soils. Holobradý (1981) suggested use chemical reclamation at each locality where the available Mg exceeded 2,000 mg kg<sup>-1</sup>. Ameliorative matter dose should be calculated based on the available Mg content in the soil. In practise, the reclamation was based on a mechanical loosening of the soil with concurrent incorporation: 10–50 t ha<sup>-1</sup> of gypsum, or 10–50 t ha<sup>-1</sup> of citric-gypsum (waste from citric acid production), or 2,000 L ha<sup>-1</sup> of sulphite leaches (pulp waste containing Ca(HSO<sub>3</sub>)<sub>2</sub>). After the above-mentioned chemical melioration, soluble magnesium sulphate is formed and gradually leached out of the soil by rainwater. To increase soil microbial biodiversity and biological activity, it is recommended to incorporate 40–50 t ha<sup>-1</sup> of farmyard manure every 3–4 years. 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 phosphorous fertilizer industry) at a dose of 4.5 t ha<sup>-1</sup>, before the snowfall, every 4–5 years to optimize the ionic balance of soil with heavily exceeded levels of Mg<sup>2+</sup> 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<sup>-1</sup>. 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<sup>-1</sup>. 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<sup>-1</sup>, *Urtica dioica* (L.) 5,127±581 mg kg<sup>-1</sup>, *Caltha palustris* (L.) 4,965±602 mg kg<sup>-1</sup> (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<sup>-1</sup>, *Phragmites australis* (Cav.) Trin. 6,860 mg kg<sup>-1</sup> and *Agrostis stolonifera* (L.) 5,419 mg kg<sup>-1</sup> (Fazekas et al., 2018). Of these plants, only *Phragmites australis* was characterised by high biomass production, that is 12.7 t ha<sup>-1</sup> 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<sup>2+</sup> 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<sub>2</sub>PO<sub>4</sub>)<sub>2</sub> · H<sub>2</sub>O. They stated that planting *A. chinense* and *Elymus dahuricus* (L.) with the application of Ca(H<sub>2</sub>PO<sub>4</sub>)<sub>2</sub> · H<sub>2</sub>O could accelerate the vegetation restoration in moderately and severely Mg-contaminated soil.

Hronec et al. (1992) suggested that land containing  $<1,000 \text{ mg kg}^{-1}$  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 (Table 1), 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  $1,000 \text{ mg kg}^{-1}$  (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 investigated sites was extremely low (0.0) to middle low (1.5) (Fazekas 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 content 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 (Tables 1 and 5). 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  $12.7 \text{ t ha}^{-1}$  of dry matter with high-energy storage of  $221.622 \text{ GJ ha}^{-1}$  (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

This study concluded that Mg-rich, alkaline dust causes long-lasting soil degradation, which is manifested mainly near both magnesite processing factories, and the excess of available Mg along the direction of the prevailing spreading of alkaline emissions.

Dominant influence of excessive content of total and available magnesium compounds on soil chemical properties was confirmed by a significant increase of soil pH, carbonate content, and conductivity, while total and available Ca did not affect the mentioned characteristics. On the contrary, Ca was positively related to available Cu and Ni.

Negative relationship between the labile fraction of soil organic matter and available Mg suggests that in more affected localities weaker formation and stock of newly formed organic matter prevails, and consequently lower 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.

Research of soil enzymatic activities are essential, as these processes reflect the degree and size of contamination of the natural environment or any revisions by humans. It is important that the high enzymatic



activity in soil was observed during a period of two to three consecutive years. This may indicate that the soil condition has become more stable.

At present, when the entry of Mg-rich, alkaline dust into the soil has been significantly reduced (by 99.75% compared to 1970), the application of measures can bring more lasting positive results than in the past, when the high deposition of Mg dust did not allow successful land reclamation.

Nevertheless, classical methods are still the most effective for reclaiming the most affected areas. It is suggested that the impermeable Mg-rich crust is mechanically removed, milled, and use as a magnesium fertilizer. Subsequently, the soil where the available Mg exceeded 2,000 mg kg<sup>-1</sup> has to be treated chemically, by incorporating gypsum, phosphogypsum, citric-gypsum, sulphite leaches, anionic polyacrylamide and calcium dihydrogen phosphate as well as high doses of farmyard manure.

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<sup>-1</sup>, 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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