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Research Article

The Influence of a Practical Remediation Medium on the Relationship of Exchange Reaction in Soil to Hazardous Lead and Inorganic Forms of Nitrogen

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The objective of this research was to analyse soil and garden compost as a remediation medium at the foothill´s locality of the Tribeč Mountains, Southwestern Slovakia, to determine the pH/KCl, Pb, NO_3^--N, NH_4^+-N contents in soil and soil affected by compost (the ratio 1 : 1), to evaluate the statistical significance of pH/KCl in relation to Pb and inorganic forms of nitrogen, and to find whether garden compost affects the monitored parameters. The analyses were carried out using Atomic Absorption Spectrometry, and Colorimetric and Potentiometric methods. Ascertained values of exchange reactions in soil ranged from 5.74 to 6.83 and exchange reactions in soil affected by garden compost ranged from 6.78 to 7.98. According to the calculated indices of dependencies, the development of pH/KCl values can be evaluated as moderately dynamic. The obtained results were evaluated statistically using the SAS 9.4 software method by the Spearman's correlation coefficient. The results in the observed three-year period showed that NO_3^--N contents in soil had reached 14.73 mg.kg^{-1}, NH_4^+-N 9.50 mg.kg^{-1}, N_{in} 23.05 mg.kg^{-1}, Pb 67.38 mg.kg^{-1}, soil affecting by remediation medium – garden compost (the ratio 1 : 1) showed that NO_3^--N contents had reached 23.93 mg.kg^{-1}, NH_4^+-N 26.42 mg.kg^{-1}, N_{in} 42.63 mg.kg^{-1}, Pb 64.71 mg.kg^{-1} dry matter. Statistical dependence was high (P <0.01), which was demonstrated for soil pH : soil + compost Pb, soil + compost pH : soil + compost Pb. The garden compost influenced pH/KCl, reduced Pb levels, and increased the proportion of inorganic forms of nitrogen in soil.

Keywords: exchange reaction, plant nutrition inorganic compounds, remediation medium, mathematical relationship of parameters, toxic element

1 Introduction

Many ways of element movement such as complexation, adsorption/desorption, coagulation, biological uptake and their opposite processes, are strongly dependent on pH. It has been scientifically proven that pH has significant effect on the specification of chemical elements and thus it directly affects the bio accessibility and the potential toxicity of metal contaminants (Khun et al., 2008).

One of the most significant environmental problems of our time, which affects every part of the environment, is the contamination of agricultural land by toxic chemical elements and the different representation of necessary nutrients and macro-, microelements in soil. Various chemical, physical, and biological processes are constantly in progress in soil and their influence causes intense changes in soil properties. Consequently, these changes have a great impact on fertility and organisms (Koren & Bisesi, 2003).

Lead (Pb) is an unessential trace element in plants that is ubiquitous in the environment. Lead has high affinity to clay minerals of illite-smectite type in acidic medium but its affinity to the formation of organic complexes increases in carbonate area (alkaline conditions) (Wang et al., 2006).

According to Khun et al. (2008), individual elements are generally more mobile in the acidic reaction of specific medium. It is a generally accepted rule that potential accessibility of potentially risky elements and toxic elements is inversely proportional to the value of soil reaction (Čéry et al., 2007). Acidic pH of environment is found in areas affected by intensive anthropogenic activity and mining activity (mining ores containing

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sulphur - pyrites, galena). This state is often accompanied by atmosphere pollution (e.g. SO_2), which is formed from burning of fossil fuels. Due to the fact that risk elements can be significantly toxic even at small concentrations and a large part of them may be accumulated in living organisms, their presence in soil and in plants can pose potential risk of negative changes to the quality of the environment. Anthropogenic activities such as disposal of urban pollution, smelting, mining, metal manufacturing, combustion engines, and exhaust gases enhance the concentration of dangerous heavy metals in the environment and are carcinogenic to human health (Morais et al., 2012). The result of such toxic influence is a reduction of organic matter decomposition (Niño-Savala et al., 2019). In crops plants, the toxicity of dangerous heavy metals such as lead reduces uptake and translocation of nutrients and water, increases oxidative damage, disrupts plant metabolism, and inhibits plant morphology and physiology (Haider et al., 2021). It also interferes with the transformation of nitrogen compounds. The root crops and the leafy vegetables affected by dangerous heavy metals pose one of the most serious health risks for humans (Tlustoš et al., 2007).

Zhang et al. (2008) reported that nitrate and ammonium nitrogen (NH_4^+-N) are the main forms of nitrogen that can be directly absorbed by plants. The NH_4^+-N is the primary decomposition product of plant and animal organic nitrogenous substances and nitrates serve as the basis for metabolic assimilation processes (Liu et al., 2015). Yli-Viikari et al. (2007) reported that the nitrogen balance is identified as the main agri-environmental indicator. Improper interventions on soil lead to gradual soil degradation, which reduces soil fertility and increases the high-risk accumulation of toxic elements in soil (Ying et al., 2018). From the point of view of the demand of quality foodstuffs, cultivation in alternative media is preferred in growing conditions. Compost is such substance as it is a complementary alternative to planting nutrition, as well as a remediation medium that affects the bioavailability of chemical elements (Khan et al., 2017). The main objectives of this paper are: to determine the pH/KCl in soil and soil affected by remediation medium - garden compost (the ratio 1 : 1, the best scientifically proven division according to the valid agrochemical practice), Pb contents in soil and in soil affected by garden compost, to determine the NO_3^--N, NH_4^+-N contents in soil and in soil affected by garden compost, to evaluate the statistical significance of dependencies of monitored parameters using by the SAS Software, to evaluate the statistical significance of monitored pH/KCl in relation to the Pb and inorganic forms of nitrogen, and to find whether garden compost affects the Pb content and the selected forms of nitrogen. We will learn about the positive or negative impact of the selected remediation medium in garden agri-environmental conditions, and about how the garden compost influences the lead content and inorganic forms of nitrogen.

2 Material and Methods

2.1 Study area, creation of the garden compost

Agricultural land such as a garden area is still not sufficiently explored for the contents of chemical elements, while they are playing an important role in growing quality crops. The study area is located at the foothill´s of the Tribeč Mountains, Southwestern Slovakia (48° 20´ 15´´ N, 18° 06´ 30´´ E, 236 m above sea level). The location belongs to the corn production area, to a warm and slightly dry climate region, where the average temperature during the growing season is 16.4 °C and the average annual precipitation is 561 mm. The garden compost as a remediation medium was created based on agrochemical practice and valid legislation of Slovakia (Act No. 79/2015 Coll.) for the production and processing of biodegradable organic waste materials. Ingredients for compost as a remediation medium were: mowed grass, biowaste from households, plant-based waste, weeds, sawdust, straw, dry grass, shrubs, bark, tree leaves and conifer needles, soil (Rendzic Leptosol), slaked lime (the materials used were applied in exact percentage shares, the compost area was 4 m^2, the maximum height of one ingredient was up to 10 cm, the moisture, grain size, and , above all, process hygiene were maintained. The maximum composting height was 1.20 m). The composting process was put through the stages of decomposition, transformation, synthesis, stabilization, and maturation processes. The process time was at least 60 days and no more than 12 months from the start date (Urminská et al., 2019).

2.2 Experimental soil analysis

The methodology for sampling the soil was conducted in accordance with the valid Slovak legislation on agricultural land (Decree of the MARD of Slovakia No. 59/2013 Coll,, NAFC, 2019). Sampling of soil (Rendzic Leptosol) was carried out regularly at monthly intervals and was monitored over the three years. It was carried out by a drilling probe placed in the centre of a circular shape with the radius of 10 m, the depth of 20 cm, and in addition, the sampling of 5 separate samples weighing 0.5 kg was randomly done at the surface soil horizon from the sites for chemical analysis (NAFC, 2019). The samples were homogenized and dried at 40 °C for 48 hours. Subsequently, they were grounded with a soil grinder VEB ThurmZG1 (VEB Elektromotorenwerke Kreis Zwickau DDR) to a fine grain fraction (less than 2.0 mm).

The analysis of inorganic forms of nitrogen of leachate samples was carried out using 1% K_2SO_4 extracts, NO_3^--N were carried out using 2,4 phenoldisulfonic acid, and N-NH_4^+ was carried out using the Nessler´s reagent using the Colorimetric method (Spectroflex 6100, WTW company). Lead analysis of leachate samples was carried out using aqua regia (HNO_3 + HCl 1 : 3) using the Atomic Absorption Spectrometry method (VarianAA240Z, Australia). Analysis of samples in leachate to find out values of exchange reactions in soil and in soil affected by garden compost was carried out by machine pH meter JENWAY 3510. It was calibrated at 3-point pH calibration (buffer) (pH 4, 7, 9). Values of exchange reaction in soil were determined by the Potentiometric method in aqueous suspension and in 1 mol.dm^{-3} from the solution KCl according to the methodology by Fiala et al. (1999).

2.3 Statistical analysis

Statistical methods are very important evaluation indicators of scientific observation.They represent an integral part of the results. The obtained results were statistically evaluated using SAS 9.4 (North California USA) (Benda Prokeinová, 2014). SAS provided all the activities related to performing risk analysis and selecting control samples. The Spearman's correlation was used to check the relationship between selected parameters in topsoil and topsoil with compost (Stehlíková, 1999).

3 Results and Discussion

The results showed that pH/KCl in soil varied from 5.74 to 6.83, and in soil affected by compost (the ratio 1 : 1) from 6.78 to 7.98. The NO_3^--N contents in soil varied from 2.15 to 14.73 mg.kg^{-1} and in soil with compost 8.15–23.93 mg.kg^{-1}. The NH_4^+-N contents in soil were 3.84–9.50 mg.kg^{-1} and in soil + compost 14.26–26.42 mg.kg^{-1}. The N_{in} contents in soil were 6.89–23.05 mg.kg^{-1} and in soil + compost 24.20–42.63 mg.kg^{-1}. Lead contents in soil were 48.51–67.38 mg kg^{-1} and in soil + compost 36.19–64.71 mg.kg^{-1} (limit for Pb in soil is 115.0 mg.kg^{-1} of dry matter, Decree of the MARD of Slovakia No. 59/2013 Coll.). The values of analytically detected lead were below the levels of toxicity. However, lead occurrence in the values of several tens of milligrams detected in areas is a negative signal and the measured lowest pH value 5.74 also contributes to the environmental risks. Concentration is crucial if it reaches high value exceeding the risk limit for organisms under current valid legislation. The populated area, the land utilized by intense agricultural and gardening activities, and the land affected by transport emissions and domestic waste emerge in the fore. Lead as a non-essential element affecting living organisms is a highly risky element because it remains in individual components of the environment in an unchanged concentration. Another negative phenomenon regarding the environment is that it has highly exhibited significance and positive dependency with pH, other chemical elements as As, Cd, Cu, Zn, and Hg. Garden soil contains a wide variety of chemical elements. From beneficial micro and macro elements to risk toxic elements. They are accumulated in soil and subsequently in cultivated crops, some are physiologically important and some cause undesirable effects in biogeochemical cycle, and thus, have a direct relationship with agricultural production and human health (Morais et al., 2012; Onistratenko et al., 2016).

Table 1 Average pH/KCl and Pb, NH_4^+-N, NO_3^--N, N_{in} (mg.kg^{-1} of dry matter) in soil and soil with compost (the ratio 1 : 1) during the three year-long monitored period

n.	pH/KCl S	pH/KCl SC	Pb S	Pb SC	NH_4^+-N S	NH_4^+-N SC	NO_3^--N S	NO_3^--N SC	N_{in} S	N_{in} SC
1	6.06	7.67	59.17	50.20	4.68	16.40	6.39	11.92	11.07	28.33
2	6.23	7.49	61.08	54.13	4.58	16.71	6.00	11.63	10.58	28.34
3	6.02	7.33	56.04	49.18	4.69	17.97	4.99	12.99	9.69	30.97
4	6.61	7.63	52.80	39.02	4.85	15.80	3.68	11.46	8.53	27.26
5	6.19	7.72	53.12	40.21	5.15	15.04	3.48	12.37	8.62	27.41
6	6.26	7.74	54.09	42.17	7.40	19.43	4.02	11.56	11.42	31.00
7	6.41	7.76	62.03	53.99	8.55	22.28	4.91	13.64	13.47	35.91
8	6.32	7.52	60.17	56.41	8.36	23.31	6.60	14.85	14.96	38.16
9	6.37	7.68	58.04	43.80	7.13	18.10	9.30	16.67	16.43	34.77
10	6.28	7.84	52.76	38.98	8.56	19.23	11.12	18.83	19.68	38.06
11	6.08	7.84	57.14	45.06	7.21	18.50	10.24	17.52	17.44	37.00
12	5.95	7.86	56.33	42.93	5.92	21.45	7.95	13.99	13.87	35.44

n – months; S-soil; SC-soil + compost (the ratio 1 : 1); N_{in} – inorganic forms of nitrogen (NH_4^+-N + NO_3^--N)

The presence of toxic heavy metals generally results into a negative impact on the organisms (Onistratenko et al., 2016). Fertile forms of nitrogen are irreplaceable for plant growth. At high contents, they cause undesirable effects for organisms (Yli-Viikari et al., 2007). Soil nitrogen criterion (agricultural nutrient reserve) reached a good to a high value after application of compost (Fecenko & Ložek, 2000). The average contents of the selected chemical properties are shown in Table 1.

Soil agroecosystem with acidic pH character (pH <7) and dynamic change of pH in the environment may cause significant release of parts from chemical elements as well as release of risk elements from garden soil into the ecosystem. This can result in contamination of soil ecosystem and its surroundings. Many chemical, biochemical, and physical ways of element movement are significantly dependent on pH. It has been scientifically proven that pH has significant effect on the specification of chemical elements and thus directly affects the bio accessibility and the potential toxicity of chemical contaminants (Tlustoš et al., 2007; Khun et al., 2008). As regards soil exchange reaction, the acidic pH environments occur in the areas with an intense anthropogenic activity. Lead is less soluble and less bio accessible when it is present in the form of crystalline minerals than when it adsorbed on the mineral surfaces. However, if the pH reduces, it can be easily desorbed on the mineral surfaces. Tlustoš et al. (2007) reports that lead admissibility is considerably affected if the pH is changed from 5 to 7. Mobility proportion decreased as much as by 20%. However, if the pH drops below 4, its mobility increases. The toxicity of Pb is significant for each environmental component (Rehman et al., 2017). Scientific works prove that most intensively, it is taken up by the plants growing in the acidic soils, which are poor in humus (Peng et al., 2009; de O. Pinto et al., 2016; Khan et al., 2017). Our results agree with de O. Pinto et al. (2016). In soil affected by compost (the ratio 1 : 1), significant changes in contents were noted. It was shown that compost significantly affected pH/KCl and selected forms of nitrogen and Pb. In most agrochemical cases, compost as a remediation medium reduces the concentrations of hazardous heavy metals (Khan et al., 2017). Organic amendments such as compost and bio-solid waste effectively reduce the availability of toxic heavy metals to plants, even in highly polluted soils (Borgulat et al., 2018). Fulvic acid and humic acid are important compounds that have the potential to change the soil's physical and chemical properties and transform pollutants in contaminated soils. Functional groups such as carboxyl and phenolic-OH groups that are associated with fulvic acid and humic acid enable the acids to interact with pollutants to form high metal complexes and thus play a key role in changing the metals' bioavailability, transport, and solubility in the soils. The accumulation of toxic heavy metals in plants depends on the genotype (Haider et al., 2021). According to Cuske et al. (2016), significant statistical correlations were found between harmful heavy metals and organic amendments. The statistically highly significant correlation between pH/KCl Pb content and the selected nitrogen parameters during the three-year monitored period is presented in Table 2. High statistical dependence (*P* <0.01) was demonstrated for soil pH : soil + compost Pb and soil + compost pH : soil + compost Pb. Statistical dependence at (*P* <0.05) was demonstrated for soil pH : soil Pb, soil + compost Pb, soil NO_3^--N, soil NH_4^+-N, soil N_{in}. Soil + compost pH : soil Pb, soil NH_4^+-N, soil N_{in}, soil + compost N_{in}, soil + compost Pb, soil + compost NH_4^+-N, soil NO_3^--N, soil + compost NO_3^--N. We confirmed the fact that the chemical components of the soil as well as the substances we have selected interact with each other. The fact was also confirmed statistically. Exceedingly high significance was determined for soil and compost, which correspond

Table 2 Spearman´s coefficient showing the statistical correlations between pH/KCl and Pb, NH_4^+-N, NO_3^--N, N_{in} in soil and in soil with compost (the ratio 1 : 1) during the three-year monitored period

Relationship		The first	Year	The second	Year	The third	Year
		pH/KCl S	pH/KCl SC	pH/KCl S	pH/KCl SC	pH/KCl S	pH/KCl SC
Pb	S	0.74580 ns	0.17358*	0.62300*	0.79430*	0.55673ns	0.23750ns
	SC	0.32274*	0.91172**	0.39881**	0.76743*	0.87245ns	0.62398**
NH_4^+-N	S	0.54014*	0.41532*	-0.00893ns	-0.32760ns	0.12963ns	-0.07019ns
	SC	0.33957ns	0.02398ns	-0.16090ns	-0.62109*	0.08480ns	-0.04623
NO_3^--N	S	0.12530ns	0.28117ns	-0.22328ns	0.00067ns	-0.44122*	-0.40702*
	SC	0.00701ns	0.09987ns	-0.14427ns	-0.06841ns	-0.18564ns	-0.33088*
N_{in}	S	0.34558*	0.45945*	-0.16782ns	-0.20599ns	-0.24010ns	-0.21373ns
	SC	0.16263ns	0.08313ns	-0.16930ns	-0.44130*	-0.08524ns	-0.16556*

significance level: **P* <0.05; ***P* <0.01; ns – not significant; S – soil; SC – soil + compost to the long-term scientific patterns (de O. Pinto et al., 2016). It has been shown that where Pb contents decrease due to the addition of compost, the selected forms of nitrogen increased. The Spearman's coefficients confirmed a positive correlation for Pb parameters and negative correlation for some forms of inorganic nitrogen parameters. This fact confirms that the contents pH/KCl : Pb and some forms of NO_3^--N, NH_4^+-N, N_{in} during the years were dependent as the Table 2 documents.

An interesting positive correlation was demonstrated for the relationship between pH soil + compost: Pb soil + compost in the first observed year (*P* <0.01), pH/KCl soil : Pb soil + compost in the second observed year and pH/KCl soil + compost : Pb soil + compost in the third observed year. During the monitored period, the relationship between soil affected by compost and the detected concentration of lead and pH/KCl in the soil affected by compost came to the fore every year. This fact documents the importance of applied compost in influencing selected chemical substances in the soil. These selected substances belong to the important substances that need to be monitored for the quality of soil. The significance level (*P* <0.05) was demonstrated for the relationship between pH/KCl : inorganic forms of nitrogen (Table 2). The results showed that applied garden compost would ensure sufficient nitrogen nutrition for plants in small growing conditions. An important high correlation was found between the pH/KCl in soil and soil + compost : Pb soil + compost (*P* <0.01). The Pb contents in the soil were significantly positively affected by the addition of compost (Bradl, 2005). If organic fertilizers are used in practice properly, we can increase crop production and improve the quality of the environment, reducing the availability of toxic elements (Liu et al., 2015). According to Kinley et al. (2010), the addition of organic fertilizers to the soil supports the increase in the availability of nitrogen in the soil, especially of nitrate nitrogen. Liu & Haynes (2012) found an increase in the content of nitrate and nitrogen due to fertilization with organic fertilizers. The compost had a statistically highly significant effect on the pH/KCl, Pb contents, and inorganic forms of nitrogen. It is very important to look for suitable and inexpensive remediation media for environmental problems (Shahid et al., 2017). The garden compost can be such a medium. Very interesting data were found regarding the relationship between pH/KCl, Pb contents in soil, in soil with the garden compost, and inorganic forms of nitrogen during the observed three years. The exchange reaction is a significantly important agrochemical key factor.

4 Conclusions

The results of the studies showed that garden compost affected pH/KCl, reduced Pb levels, and increased the proportion of inorganic forms of nitrogen in the soil. The scientific fact has been confirmed that the occurrence of dangerous lead in the soil environment is significantly influenced by soil pH. This is also confirmed by our research during the monitored three-year period with a high statistical dependence. The use of biodegradable substances is currently a priority in improving the environmental quality of soil and waste management. The use of garden compost as an environmentally, economically available remedial medium is a suitable alternative for the conditions of Slovak gardening practice, as it is a suitable alternative improving the quality of soils. The issue of mutual relations of chemical substances in the components of the environment is still highly relevant; preferably, especially from the point of view of demand for quality cultivated crops. A practical and affordable remediation medium (garden compost) contributes to the quality of crops and affects chemical elements and hazardous heavy metals present in the soil environment.

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