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THE USE IN AGRICULTURE OF CALCAREOUS AMENDMENTS AND THEIR INFLUENCE ON THE CHEMICAL PROPERTIES OF THE REDDISH LUVOSOIL FROM THE MOARA DOMNEASCA STATION, SOUTHEASTERN ROMANIA

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Abstract

The increasing volume of waste generated by the steel industry, with repercussions given by environmental pollution and the removal from the agricultural circuit of significant areas of land that is being used for waste storage, requires the identification of efficient solutions for recycling this waste, while also requiring the use of elements with beneficial effects on the soil and on the cultivated plants. The metallurgical industry produces various residues, and some of them, such as steel slag, can be used successfully in the agricultural activity. Steel slag can have a beneficial influence on the physico-chemical properties of the soil and on the agricultural production. In order to mitigate the environmental consequences caused by the disposal of large quantities of slag in landfills and in order to reduce transport costs for its disposal, the steel industry encourages the sustainable use of slag in various fields of application, such as agriculture. Research conducted in 2020, on the experimental field at the "Belciugatele" research station/"Moara Domneasca" development farm, followed the influence of various calcareous amendments on the chemical properties of reddish luvosoil from this area. During the experiment, different doses of dolomite, calcium carbonate and two types of slag from the steel industry were applied and changes in the reaction of the soil and the content of macroelements and microelements in the soil were monitored.

Key words*: soil, amendments, slag, chemical properties, pollution.*

INTRODUCTION

The use, in agriculture, of solid metallurgical waste such as steel slag, has become very important for its recycling and recovery, which would lead to a reduction of overall environmental pollution, while also contributing to soil improvement and higher yields.

Every year, in Europe, over 40 million tonnes of iron and steel slag are produced as a result of industrial processes (Branca & Colla, 2012; Yildirim & Prezzi, 2011).

Based on its properties, slag is classified as a non-hazardous waste and can be stored, but this procedure requires a large area of land for storage. There is always the risk that various components of the slag may be leached into the soil's profile and contaminate the water table, and implicitly, groundwater.

Therefore, it is necessary to research these types of wastes that could be used as valuable secondary raw materials in other areas of the economy, such as agriculture.

The main chemical components that slags contain in their composition and which are important for their use in agriculture are CaO, MgO, SiO_2, P_2O_5, Fe and MnO. Concentrations of elements in slag vary greatly depending on the raw materials used, the type of steel manufactured, furnace conditions and other aspects (Yi et al., 2011; Shi, 2004).

It was shown that, due to the high calcium content, slag can help increase the soil's pH and can help mobilize nutrients, which leads to an increased agricultural production (Branca & Colla, 2012). Balanced plant nutrition is one of the main factors affecting their growth and yield. The application of industrial waste as a fertilizer or as an amendment to correct the soil's pH reaction has become a common practice in agriculture (Liu et al., 2002; Yang & Zhang, 2005).

The application of steel slag was studied on acidic soils, cultivated in the no-tillage system, with excess of Al^{3+}, and after 27 months from the application of slag in doses of 2 t/ha, 4 t/ha and 8 t/ha (Fernandes et al., 2018), without incorporation, an increase in the degree of saturation in bases was recorded, with V% = 70%, at a depth of 20-40 cm, when the dose of 4 t/ha of slag was applyed, while an increase in the pH value was also recorded, from a pH of 4.6 in the non-fertilized variant to a pH of 5.6 in the variant where 4 tonnes of slag/ha were applied. During the same period, the effects of $CaCO_3$ on the soil were observed only at a depth of 0-20 cm.

Due to the high level of CaO and MgO and also due to the high pH level, that can reach up to 12.5, the repeated application of steel slag can make the soil excessively alkaline, which can reduce the bioavailability and the plant's uptake/absorption of macroelements such as P and microelements, such as Fe, Cu and Zn and can impede their growth and overall yields (Chand et al., 2015).

MATERIALS AND METHODS

Because slag is rich in Ca and other fertilizer components such as Mg, Fe, N, P, K, in our research we aimed to evaluate the impact of the application of two types of steel slag (furnace slag - LF and converter slag - CV) (Tables 1 and 2) on the soil's quality while also aiming to compare their influence on the soil's chemical properties, with the influence of amendments used traditionally, respectively calcium carbonate and dolomite.

The steel slag used in this study, as a potential source of certain nutrients beneficial to both soil and plants, is a by-product of the steel industry and comes from the ArcelorMittal Galati Steel.

Table 1. Composition of different types of slag from Romania, ArcelorMittal Galati

	Fe	SiO_2	CaO	MnO	MgO	Al_2O_3	P_2O_5
	%	%	%	%	%	%	%
A-LF	2.38	15.55	48.52	6.12	3.50	16.63	0.50
B-LF	0.98	11.89	52.56	0.56	4.15	22.28	0.29
C-LF	11.28	7.88	40.46	5.93	5.11	22.54	0.60
D-CV	19.09	13.25	50.80	3.05	1.48	1.85	1.71
E-CV	1.17	12.09	49.65	0.98	5.20	24.04	0.31
F-CV	5.55	7.54	46.71	2.25	6.75	24.07	0.50

The most significant oxide components of slag are: CaO (usually 46-52.5%), Al_2O_3 (2-22.5%), SiO_2 (7.5-15.5%) and MgO (3.5-6.8%). The slag generated by the furnace has a Fe content between 1% and 19%, while the MnO content is between 0.5% and 6.2% and less than 2% of P_2O_5 (Table 1).

LF slag (ladle furnace slag) is a secondary metallurgy slag resulted from the process of refining steel slag generated in the different primary unit (electric arc furnace).

The most usual applications of dusty LF slag are focused, at present, on the following areas (Popescu et al., 2016):

- The reintroduction into the steelmaking units: electric furnace slag frothing, converter with oxygen blast-furnace desulfurization or ladle (Zhao et al., 2015);
- The use as an additive of clinker in the cement industry (Setien et al., 2009; Shi, 2004);
- Used for acid mine water treatment (Mark & Gutta, 2009);
- Used as a fertilizer in agriculture, because of trace elements found in slag, which may act as micronutrients (Bonenfant et al., 2008), or a neutralizer for acidic soils (Smallfield, 2000).

The current steelmaking process is based on the Basic Oxygen Steelmaking process, where a basic slag is produced in the Linz-Donawitz converter. The LD slag contains about 1-3 wt% of P_2O_5, which is too low to be used as a phosphate fertilizer, but, at the same time, it is too high to be used in the BF or recycled in the sinter plants.

Because blast furnace slag contains the fertilizer components CaO, SiO_2, and MgO, it is used as calcium silicate fertilizer. In addition to these three components, steelmaking slag also contains components such as FeO, MnO, and P_2O_5, and is used for a broader range of agricultural purposes, including dry field farming and pastures. Its alkaline property can also correct soil acidity (Humaria, 2014).

Table 2. Chemical composition of slag used in the experiment

Materials	Na	Ca	Mg	Cd	Cr	Cu	Ni	Mn	Pb	Zn	Fe
	%	%	%	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	%
LF	0.28	11.7	5.43	U/nd*	7.61	2.39	4.88	9084	0.3	6.3	0.08
CV	0.023	18.5	1.62	U/nd*	725	20.1	3.68	28164	3.75	40.8	12.2

*U/nd - undetectable by the test method used

The concentrations of heavy metals in the slag were analyzed by atomic absorption spectrophotometry (AAS), from extracts with wet mineralization, ICPA Methodology (1983), Chapter 14 pt 134.

The study was conducted at the "Belciugatele" research station/"Moara Domnească" development farm belonging to the University of Agronomic Sciences and Veterinary Medicine of Bucharest (USAMV Bucharest), where an experimental field which included 9 variants in 3 repetitions V1 (control), V2 (carbonate calcium - 3 tonnes/ha), V3 (dolomite - 3 tonnes/ha), V4 (LF-slag 1 tonne/ha), V5 (LF-slag 3 tonnes/ha) , V6 (LF-slag 5 tonnes/ha), V7 (CV-slag 1 tonne/ha), V8 (CV-slag 3 tonnes/ha), V9 (CV-slag 5 tonnes/ha) where the slag was incorporated immediately after application.

The aspect of the slag used in this study is presented in Figures 1 and 2.

Figure 1. LF slag (foto. 2019)

Figure 2. CV slag (foto. 2019)

RESULTS AND DISCUSSIONS

The type of soil found in the area of research is the reddish luvosoil, that had, before the application of the slag, the following chemical characteristics (Tables 3 and 4), where we can observe in the surface horizon a pH of 5.98, a humus content of 1.95, Nt of 0.110 %, P_{AL} 41 mg/kg, K_{AL} 200 mg/kg, Ca^{2+} 2.98 me/100 g and a heavy metal content below the maximum permissible limits.

Table 3. The main chemical properties of the reddish luvosoil in the reseach area

	Tests performed						
Depth	pH	Humus	Nt	P_{AL}	P_{AL}[1]	K_{AL}	Ca^{2+}
cm	pH units	%	%	mg/kg			me/100 g
0-20	5.98	1.95	0.110	41	41	200	2.98
20-40	5.70	1.78	0.102	29	29	146	11.83

[1]recalculated values in accordance with the soil´s pH

Table 4. The main chemical properties of the reddish luvosoil in the reseach area

	Tests performed								
Depth	Cd	Cu	Co	Cr	Mn	Ni	Pb	Zn	Fe
cm	mg/kg								%
0-20	0.21	22.6	12.4	21.5	863	30.6	19.1	64.5	2.839
20-40	0.15	21.9	11.6	21.7	785	33.2	17.1	67.0	3.106

The analysis of the soil's pH reaction at a depth of 0-20 cm shows a very significant increase in the pH value when applying a dose of 3 tonnes of LF slag per hectare, respectively 6.03, compared to 5.96 in the control variant, as shown in Table 5.

Table 5. The influence of the applied amendments on the soil's pH reaction at a depth of 0-20 cm

Variant	Soil's pH		Difference		Significance
		%		%	
V1-control	5.96	100	Mt	-	
V2-$CaCO_3$	6.03	101.11	0.06	1.11	***
V3-$CaMg(CO_3)_2$	5.96	100.05	0.00	0.05	-
V4-LF 1 t/ha	5.85	98.21	-0.10	-1.78	ooo
V5-LF 3 t/ha	6.03	101.11	0.06	1.11	***
V6-LF 5 t/ha	5.88	98.71	-0.07	-1.28	ooo
V7-CV 1 t/ha	6.10	102.29	0.13	2.29	***
V8 CV 3 t/ha	6.09	102.18	0.13	2.17	***
V9-CV 5 t/ha	6.51	109.16	0.54	9.16	***
LSD 5% = 0.02 *LSD 1% = 0.03* *LSD 0.1% = 0.04*					

At a depth of 20-40 cm, the increase in the value of the soil's pH reaction is very significant in all experimental variants, when compared to the control variant. The highest increase was recorded in the variant where dolomite was applied, with a pH of 6.11, compared to a pH value of 5.71 in the control variant. For the variants where LF slag was applied, the soil's pH reaction was of 5.74 for the variant with 1 tonne of slag per hectare, 5.97 for the variant where 3 tonnes of slag per hectare were applied and 5.92 for the variant where 5 tonnes of LF slag were applied per hectare, as detailed in Table 6.

Table 6. The influence of the applied amendments on the soil's pH reaction at a depth of 20-40 cm

Variant	Soil's pH		Difference		Significance
		%		%	
V1-control	5.71	100	Mt	-	
V2-$CaCO_3$	6.02	105.36	0.30	5.36	***
V3-$CaMg(CO_3)_2$	6.11	107.05	0.40	7.05	***
V4-LF 1 t/ha	5.74	100.58	0.03	0.58	***
V5-LF 3 t/ha	5.97	104.55	0.26	4.55	***
V6-LF 5 t/ha	5.92	103.61	0.20	3.61	***
V7-CV 1 t/ha	6.17	107.99	0.45	7.99	***
V8-CV 3 t/ha	6.19	108.45	0.48	8.45	***
V9-CV 5 t/ha	6.24	109.27	0.53	9.27	***
LSD 5% = 0.03 *LSD 1% = 0.05* *LSD 0.1% = 0.07*					

The application of CV slag determined an increase of the soil's pH reaction at both the depth of 0-20 cm and the depth of 20-40 cm in all variants where CV slag was applied. The most significant increase was recorded at a depth of 0-20 cm, when a dose of 5 tonnes per hectare was applied, where the pH value increased to 6.5, as compared to the value of 5.96 in the control variant. At a depth of 20-40 cm, also when applying a dose of 5 tonnes per hectare, the highest increase of the soil's pH reaction was registered, respectively a value of 6.24, compared to 5.71 for the control variant.

The application of steel slag in the experiments undertaken on the reddish luvosoil from Moara Domnească determined an increase of the soil's pH reaction and of the main chemical properties of the soil, in the first 3 months from the application, as shown in Tables 7 and 8.

Table 7. The influence of the applied amendments on the chemical properties of the reddish luvosoil from Moara Domneasca at a depth of 0-20 cm

Treatment	Humus	Nt	P_{AL}	K_{AL}	Ca^{2+}	Mg
	%	%	mg/kg	mg/kg	me/100 g	me/100 g
V1-Control	2.01	0.106	35	159	2.98	17.1
V2- $CaCO_3$	1.78	0.107	35	174	12.75	18.2
V3- $CaMg(CO_3)_2$	1.95	1.110	41	200	3.27	18.4
V4-LF 1 t/ha	2.07	0.106	41	123	10.05	18.2
V5-LF 3 t/ha	2.13	0.227	51	204	15.58	20.0
V6-LF 5 t/ha	2.01	0.115	70	166	11.55	18.2
V7-CV 1 t/ha	1.78	0.110	46	182	16.84	18.9
V8-CV 3 t/ha	2.07	0.117	43	160	12.68	19.8
V9-CV 5 t/ha	1.84	0.109	85	130	17.64	20.1

The application of the amendments did not lead to an increase of the humus content in the soil, the highest value was registered when applying a dose of 3 tonnes of LF slag per hectare, respectively 2.13% compared to 2.01% in the control variant.

The total nitrogen content of the soil registered increases in most experimental variants, the highest value being registered in the variant where 3 tonnes of LF slag were applied per hectare, respectively 0.227%.

The increase of the phosphorus content in the soil was recorded in all experimental variants. The highest values were recorded when applying the dose of 5 tonnes per hectare, both in the case of LF slag (70 me/kg) and in the case of CV slag (85 me/kg).

Significant increases in the soil's potassium content were recorded in most experimental variants compared to the variant where no calcareous amendments were applied, the highest value being recorded in the variant where the dose of 3 tonnes of LF slag were applied per hectare.

In the case of calcium, there were increases in all experimental variants, the highest value being recorded where converter (CV) slag was applied on the soil in a dose of 1 tonne per hectare.

As for the case of calcium and magnesium, there were increases in all experimental variants compared to the control variant, where the value was of 17.1 me/100 g.

The largest increases were recorded when applying LF slag in a dose of 3 tonnes/ha (20 me/100 g) and when applying CV slag at a dose of 5 tonnes/ha (20.1 me/100 g).

Table 8. The influence of the applied amendments on the chemical properties of the reddish luvosoil from Moara Domneasca at a depth of 20-40 cm

Treatment	Humus	Nt	P_{AL}	K_{AL}	Ca^{2+}	Mg
	%	%	mg/kg	mg/kg	me/100 g	me/100 g
V1-Control	1.60	0.092	37	160	3.21	19.4
V2- $CaCO_3$	1.78	0.102	29	146	11.83	20.6
V3-$CaMg(CO_3)_2$	1.78	0.109	53	144	4.11	20.4
V4-LF 1 t/ha	1.95	0.115	39	132	2.98	20.2
V5-LF 3 t/ha	2.01	0.121	37	186	5.43	19.1
V6-LF 5 t/ha	1.95	0.107	44	144	19.75	20.4
V7-CV 1 t/ha	1.30	0.085	12	148	24.79	20.0
V8-CV 3 t/ha	1.66	0.095	19	148	1.49	20.5
V9-CV 5 t/ha	1.54	0.092	30	138	3.47	21.8

After 3 months from application, the influence of the amendments on the depth of 20-40 cm, as compared to the depth of 0-20 cm, is not significant, with the only exception being in the case of calcium, where an increas can be observed in most experimental variants, when compared to the control variant. The highest increase, of 19.75 me/100 g, was registered when applying a dose of 5 tonnes of furnace slag (LF) per hectare and of 24.79 me/100 g, when applying converter slag (CV) in a dose of 1 tonne per hectare.

The analyzes performed, in the experimental field, on the reddish luvosoil, in order to determine the soil's content in heavy metals, show that all the heavy metals analyzed had a content below the maximum allowable limits (Table 9).

Table 9. The heavy metal content of the soil at Moara Domneasca at a depth of 0-20 cm

Heavy metals	Cd	Cu	Co	Cr	Mn	Ni	Pb	Zn	Fe
				mg/kg					
V1-Control	0.19	22.6	12.2	21.8	788	32.40	17.0	64.3	2.767
V2- $CaCO_3$	0.18	22.5	11.5	21.4	821	31.57	21.8	68.6	2.846
V3- $CaMg(CO_3)_2$	0.19	21.2	13.0	25.8	849	41.20	19.6	69.7	2.922
V4-LF 1 t/ha	0.13	20.6	10.5	22.3	77	32.65	20.9	61.9	2.700
V5-LF 3 t/ha	0.10	23.6	10.8	20.2	802	38.39	21.1	66.9	2.783
V6-LF 5 t/ha	0.11	22.7	10.5	22.8	842	34.93	20.9	67.8	2.812
V7-CV 1 t/ha	0.19	20.4	14.3	24.7	812	37.70	19.7	65.8	2.873
V8-CV 3 t/ha	0.18	22.6	13.9	30.6	848	48.00	17.3	64.5	3.062
V9-CV 5 t/ha	0.32	21.3	13.6	28.2	852	44.70	19.4	68.9	2.977
Maximum heavy metal allowable limit for soil	3	100	-	100	-	50	50	300	-

CONCLUSIONS

Our research highlighted that slag from steel processing plants can be used as an amendment to correct the soil's pH reaction.

A high CaO content, of 50%, may recommend the use of these by-products of the steel industry for soil improvement.

In addition to the calcium intake, this waste also brings an important contribution of microelements in the soil, necessary for the growth and development of crops.

There were significant increases in the value of the soil's pH reaction both at a depth of 0-20 cm and at a depth of 20-40 cm when applying the converter slag (CV) in a dose of 5 tonnes/ha.

After 3 months from the application of these residues, no accumulations of heavy metals was found in the soil, even for the maximum applied doses.

The results of research on the use of slag from the steel industry in agriculture have shown that the correct application of these residues brings benefits to the soil's chemical properties, by increasing the pH value of acidic soils, by increasing the nutrient contents of phosphorus, calcium and magnesium, and all these aspects have a contribution in achieving higher crop yields.

Environmental risk assessment based on the application of slag requires further investigation, which is an opportunity for both the steel and fertilizer industries to promote the economy/preservation of natural resources, in order to reduce CO_2 emissions, to prevent the accumulation of large amounts of waste, and to bring social awareness in regards to the sustainability of some industrial activities.

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