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Biosafety evaluation for some remediants and their effectiveness in detoxication peaty soils with heavy metals

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Abstract. Contamination soils with heavy metals is a problem with high interest, because contamination with heavy metals affect food chains and human health by intake and accumulation in living beings. Although all of these methods not quite effective for recovering contaminated environments, however they are still good options for recovering contaminated soils. In this research evaluated the recovering potential in different concentrations and combinations of dolomite with additives like zerovalent iron, Fe-Mn concretions, iron powder, ferrihydrite and iron nanoparticles. With these treatments, achieved reduction of concentration of all heavy metals founded (Co, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) and also reduction in toxicity, confirmed with a biotest with Daphnia magna. These findings confirms that the optimal remediants are nanoparticles with biochar and iron powder with and without biochar.

Keywords: remediation soils, heavy metals, biotest, ecotoxicity, soil's contamination

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Оценка биобезопасности некоторых ремедиантов и их эффективности при детоксикации торфяных почв тяжелыми металлами

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Аннотация. Почвенное загрязнение тяжелыми металлами – крайне важная проблема, поскольку этот вид загрязнения влияет на пищевые цепи и в конечном итоге на человеческое здоровье из-за аккумуляции в живых организмах. В настоящее время существуют различные технологии и методы восстановления экосистем. Хотя эти методы не всегда достаточно эффективны для устранения загрязнения окружающей среды, они вполне приемлемы для почвенной ремедиации. В исследовании был протестирован восстановительный потенциал доломита в различных концентрациях, в сочетании с такими добавками, как ноль-валентное железо, конкреции Fe-Mn, железный порошок, ферригидрит и наночастицы металлов. Предложенная схема восстановления почвы привела к снижению концентрации всех тяжелых металлов в почвенных пробах (Co, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn), а также уменьшению токсичности, что было подтверждено биотестом с Daphnia magna. Эти исследования показывают, что оптимальными ремедиантами являются наночастицы с биочаром и железный порошок с биочаром и без него.

Ключевые слова: ремедиация почв, тяжелые металлы, биотест, экотоксичность, почвенное загрязнения

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Introduction

The contamination soils by heavy metals is a serious problem that has increased due to human activities, such as mining, metallurgical industry, automobile's emissions, indiscriminate waste disposal activities, agriculture and natural discharge of some aquifers. Although it does have a lot of benefits for society, thanks to economic development, there are also a lot of risks for environment [1; 3; 6; 9; 11; 14].

Heavy metals are a group of toxic elements with high biological and industrial importance, that could be uptake by food crops and vegetables, affecting food chains and as a result human health by intake of contaminated food. In general, heavy metals have a specific density > 5 g/cm^3, that is why their excessive accumulation in soils may cause soil deterioration and generate other environmental problems, like disturbances in soil microbiota and alterations in the metabolic activities of these microorganisms [4; 7; 12].

Since the heavy metals tends to accumulate themselves in the soil's surface, it is make them available to be absorbed by plants roots. Plants absorb and accumulate these elements in aerial parts, such as stems, foils and fruits. While moving through trophic chains heavy metals may be affecting on biosafety and food quality [4; 7; 12].

The heavy metals could be released to environment by natural processes (like weathering of underlying bedrock) or by human exploitation (refining and mining, pesticides using, incorrect disposal of batteries, solid wastes irrigation with wastewater, use of fertilizers and exploitation of industrial tanneries). Oppose to organic contaminants, the heavy metals are hard to be biologically, chemically or physically decomposed. Therefore, the remediation of soils contaminated with heavy metals only could be achieved by isolation or altering their mobility, solubility or toxicity, through changes in their chemical valence, favoring their dissolution or chelation [4; 7–9; 16].

One of the techniques that is used to remediate contaminated soils is the use of biochar, a carbon rich, porous, purpose-produced charcoal, manufactured during pyrolysis of organic residues such as animal wastes, crop residues, municipal wastes, wood and biosolids. Using biochar changes soil features, like microbiota and their physical and chemical properties, or increase of soil pH, that cause precipitation and immobilization of heavy metals. With the addition of biochar to a contaminated soil increases the population and microbial activity, and also the seed germination and plant growth. Besides, this technique contributes to processes of immobilization/mobilization of different kinds of heavy metals, with benefits to environmental and economic aspects. Therefore, biochar is a helpful method to remediate polluted with heavy metals soils [2; 7; 17].

Despite that there are different methods and technologies used in remediation processes, all of them have not the same effectiveness to recover contaminated environments, especially those polluted with heavy metals. Since not all of the remedial substances and technologies that are used in order to recover contaminated environments and ecosystems have the same potential, it's necessary to evaluate their effectiveness and biosafety in the environment, in this case, in soils.

To immobilize toxic heavy metals with sorbents in acidic peat soils, pre-treatment with acid-reducing soil conditioners is generally recommended. In particular, the variant of treatment with dolomite powder was tested on samples of polluted peat soil [13]. It seems important to address the issue of how pre-treatment of soils and altered soil properties effect the efficiency of subsequent detoxification with metal-containing sorbents.

The purpose of this work was to study the effectiveness and biosafety of tillage substances to determine the ecotoxicity and remediation effect.

Materials and methods

Object and methods

Polluted peat soil was collected at an industrial wasteland 0,7 kilometers from a copper-nickel plant (67°55′70″ N, 32°51′50″ E). This area is a part of a northern taiga subzone located in Kola peninsula, Russia. Samples of soil were collected from a depth of 0–20 cm, from 10 equidistant points. Total sampling area reach 400 m^2. Soil samples were later dried and mixed.

Experiment consisted of 8 variants: control (polluted soil), soil with dolomide and 6 types of remidiants. Remidiants used in the experiment were Biochar and a vary of ZVI (zerovalent iron) additives such as Fe-Mn concretions, iron powder, ferrihydrite and iron nanoparticles.

Dolomite is a carbonate mineral $CaCO_3*MgCO_3$. It is widely used to treat acidic soils by leaching them. It also improves soil structure and water regime. We have compared the effect of two doses – 3 and 10% dolomite on the detoxification polluted soils using biochar and several metal-containing sorbents.

Biochar is a coal of plant origin that results after pyrolysis of biomass. It is widely used as a fertilizer. Biochar improves water regime, microbial life and reduces nutrient leaching. Thus, perfecting conditions for plant growth.

Fe-Mn concretions is a natural source of iron and manganese. They are commonly found in peaty soils. They consist of Fe and Mn oxides and form a red-brown spheres.

Iron powder is a ground ZVI. Iron nanoparticles is also ground ZVI but the size of particles is much smaller therefore surface is higher than than regular iron powder. Iron nanoparticles can aggregate and cannot be used without other additives. Nanoparticles were mixed with biochar.

Ferrihydrite is a mineral $5(Fe^{3+})_2O_3*9H_2O$. It can be found in soil.

Results and discusion

In this study have been obtained two kind of results: in first place we have results for chemical analysis and in second place, in order to determine toxicity of soils after treatment.

Chemical analysis soils:

In order to determinate the concentrations of heavy metals in soil samples this analysis was performed by the extraction method. In Table 1 shows the values of heavy metals' founded in the soil's samples.

Table 1. **Average values for heavy metals' concentration (mg/kg) and values pH in soil's samples**

Soil's samples		Heavy metals' concentration (mg/kg) and pH									
Series	**Remediant**	**Cd**	**Co**	**Cr**	**Cu**	**Fe**	**Mn**	**Ni**	**Pb**	**Zn**	**pH**
A	Soil without treatment	0.21	13.39	0.08	161.01	75.98	111.04	381.00	0.00	7.32	4.65
B1	Dolomite 3%	0.00	0.77	0.04	11.65	16.94	9.28	17.04	0.00	0.63	5.84
C1	Biochar + iron nanoparticles	0.00	0.48	0.05	8.58	15.36	6.97	9.79	0.00	0.35	5.92
D1	Biochar	0.00	0.66	0.02	12.41	14.58	7.92	15.49	0.03	0.52	5.85
E1	Fe-Mn concretion	0.00	0.58	0.02	14.42	16.33	114.44	19.27	0.00	0.63	5.82
F1	Biochar + Fe-Mn concretion	0.00	0.54	0.03	11.54	15.07	104.53	17.93	0.00	0.62	5.80
G1	Iron's Macroparticles	0.00	0.56	0.00	9.37	18.29	6.61	11.31	0.00	0.36	5.91
H1	Biochar + Iron's Macroparticles	0.00	0.52	0.02	7.93	16.04	6.67	9.95	0.00	0.35	5.88
B2	Dolomite 20%	0.00	0.25	0.04	12.18	16.64	4.83	6.72	0.00	0.38	7.16
C2	Biochar + iron nanoparticles	0.00	0.15	0.04	8.42	17.28	3.10	4.41	0.00	0.24	7.12
D2	Biochar	0.00	0.25	0.06	12.73	16.24	4.39	6.72	0.00	0.46	7.13
E2	Fe-Mn concretion	0.00	0.14	0.03	9.67	11.96	13.11	5.11	0.04	0.33	7.13
F2	Biochar + Fe-Mn concretion	0.00	0.12	0.06	11.10	14.95	9.36	4.94	0.00	0.33	7.14
G2	Iron's Macroparticles	0.00	0.13	0.02	8.54	14.74	2.37	4.16	0.00	0.26	7.20
H2	Biochar + Iron's Macroparticles	0.00	0.14	0.02	9.91	18.36	2.24	4.46	0.04	0.22	7.34

For a more visual distribution of elements in soil samples, the next graphs show the distribution of heavy metals in samples of treated and uncultivated soil with the addition of 20% dolomite and 3% dolomite, respectively: Figure 1 and 2.

According to the graphics, there are high differences between samples with treatments (B-H 1 and 2) and sample control, without them (A). In soil without treatment, there are high concentrations of nickel, copper, iron and manganese. These high levels of copper and nickel can explain the contamination by a metallurgical factory, to 700 m from sampling point. The presence of iron and manganese can be explained by soils natural compounds, because in peat soils there are high amounts of those metals in the form of ferruginous-manganese nodules, also solid accumulations of iron and manganese.

Even after the addition of dolomite, in both cases the concentration of heavy metal ions is significantly reduced. When adding 20% dolomite, the ion concentration is less than when adding 3% dolomite. In Figure 1, it's shown that with the addition of 3% dolomite, there are two peaks of manganese's concentration, probably by the addition of ferruginous-manganese modules. With the use of 20% dolomite doesn't reported the same behavior, probably because of the higher pH levels.

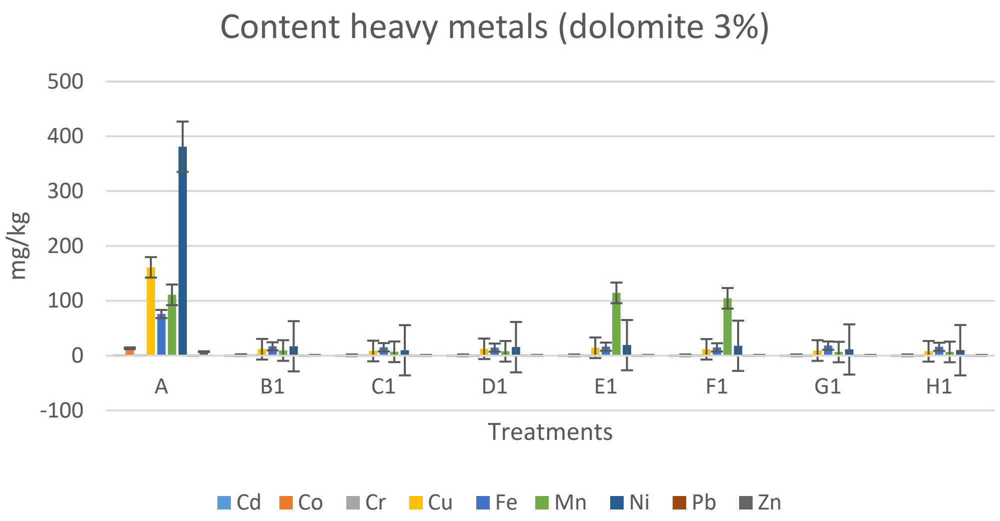

Figure 1. Average content of heavy metals in soil samples with 3% dolomite
Source: compiled by the authors.

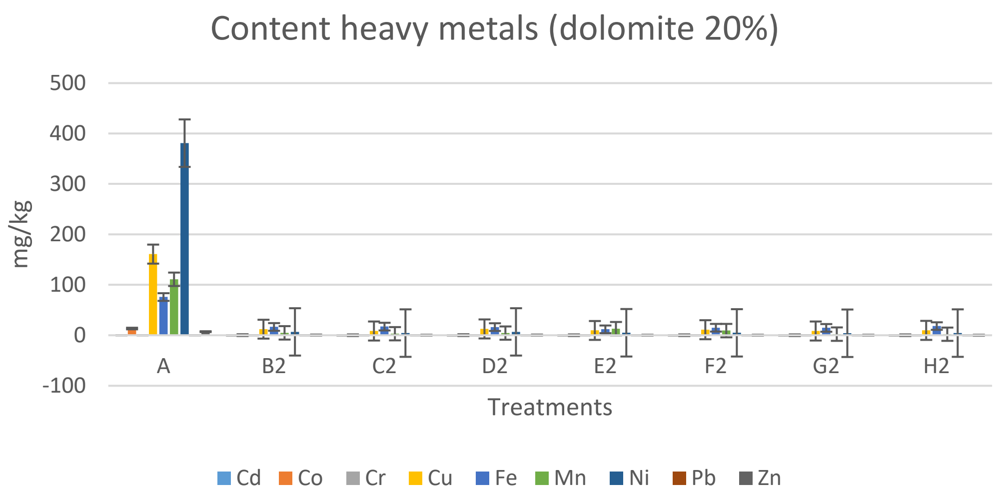

Figure 2. Average content of heavy metals in soil samples with 20% dolomite
Source: compiled by the authors.

It follows from Figure 3 that only dolomite and its quantity affect the acidity of the samples, since with the addition of 3% dolomite, the pH of the samples averaged 5.86, and with the addition of 20% dolomite was 7.2.

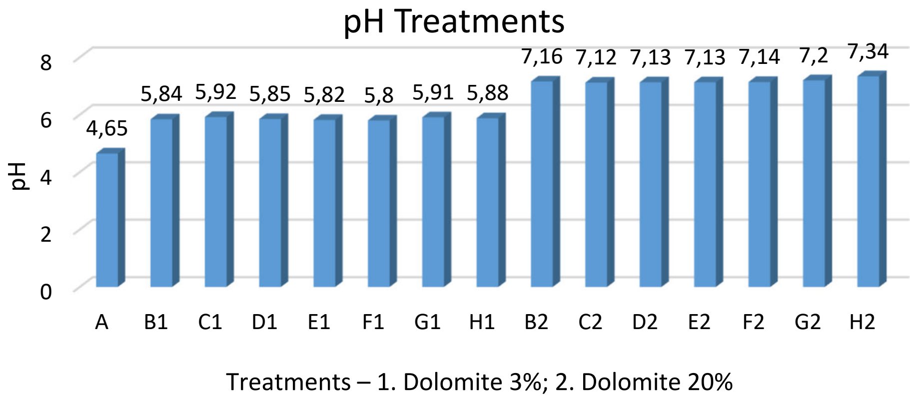

Figure 3. Values pH for soils with different treatment
Source: compiled by the authors.

Results of biotest with hydrobionts

The test on hydrobionts was carried out in 4 repetitions. The toxicity of the sample is assessed based on the survival of the organisms. The results are resumed in Table 2.

Table 2. Survival organisms *Daphnia magna* in aquatic extracts soil's samples without and with remediators

Sample	Code sample	Survival organisms, %
Soil without treatment	A	0
Dolomite 3%	B1	25
Biochar + iron nanoparticles (Dolomite 3%)	C1	100
Biochar (Dolomite 3%)	D1	40
Fe-Mn concretion (Dolomite 3%)	E1	75
Biochar + Fe-Mn concretion (Dolomite 3%)	F1	55
Iron's Macroparticles (Dolomite 3%)	G1	100
Biochar + Iron's Macroparticles (Dolomite 3%)	H1	100
Dolomite 20%	B2	64
Biochar + iron nanoparticles (Dolomite 20%)	C2	75
Biochar (Dolomite 20%)	D2	85
Fe-Mn concretion (Dolomite 20%)	E2	90
Biochar + Fe-Mn concretion (Dolomite 20%)	F2	100
Iron's Macroparticles (Dolomite 20%)	G2	100
Biochar + Iron's Macroparticles (Dolomite 20%)	H2	75

The Figure 4 shows the proportions of organisms that survive to treatments.

According to results, in the samples treated with dolomite 20% was an increase in pH, these values were higher than samples with dolomite 3%. In general, the pH values for treatments with dolomite 20% were in average 7.2 (neutral), while with dolomite 3% were 5.87 (weak acid).

In toxicity analysis, it's have been determined the level of toxicity, according to percentage of survived organisms *D. magna* in aquatic extracts, obtained from soil's samples; While the percentage of survived organisms is high, it's still considered high efficiency in remediation soils, with reduction of toxicity. As can it see in the graphic, treatments with dolomite 20% show that there were better levels remediation as with dolomite 3% as were, mainly in use only dolomite and dolomite's combination with biochar, with iron-manganese's concretion and combination of biochar with iron-manganese's concretion. Moreover, treatments with dolomite 3% mixed with iron nanoparticles and biochar with iron nanoparticles shown higher levels than those treatments with the dolomite 20%; in the case of dolomite mixed only with iron nanoparticles, both treatments had the same effectiveness.

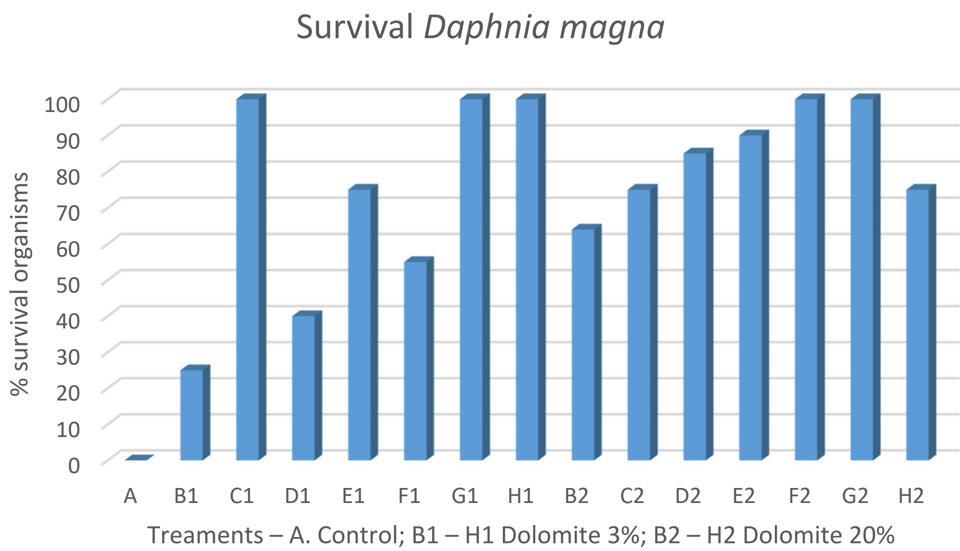

Figure 4. Percentage organisms *D. magma* survivor to treatments with dolomite
Source: compiled by the authors.

Some authors like Machado [10] says that iron's nanoparticles are efficient to degrade contaminants in soils, like halogenated hydrocarbons, polychlorinated biphenyls and also heavy metals. In this research, we found that addition of iron helps to improve the quality of soil, at least, with reduction/elimination toxicity in soil, according to the results obtained with biotests.

Furthermore, Galdames [5] says that remediation treatments with iron nanoparticles have disadvantages, like a short lifetime, due to their structural changes, surface passivation and environmental risks associated with the second metal, namely, the metal causing contamination, especially for iron, due to the toxicity of the obtained by-products and reactivity decrease if they are not eliminated. However, these nanoparticles have a high reactivity, due to their surface area, an important characteristic for rapid degradation contaminants. Moreover, these authors also say that iron's nanoparticles it's an effective decontaminant agent. In this research, we can see the potential in reduction of concentration heavy metals, although not in the same proportions for all the treatments including this additive with dolomite in different concentrations.

By other hand, in the use of dolomite contributes to fixation heavy metals in soils, authors like Zhang & Lu [17] says that dolomite helps to prevent biological transfer of heavy metals like Cd or Cr from soils, to plants, using like main mechanism immobilization, the surface adsorption. Also, Vrînceanu [17] says that addition of dolomite helps to reduce significantly the concentration of heavy metals in soil, mainly lead and zinc; for this investigation, was found that dolomite helps to recover soil of contamination, reducing concentrations of heavy metals.

Conclusions

The problem of chemical pollution of soils, particulary pollution with heavy metals, has many solutions. Within the studied variants of sorbents, the optimal remediants were determined simultaneously of adding dolomite at different concentrations, which is for different pH. For an acidic environment, the optimal remediants are nanoparticles with biochar and iron powder with and without biochar. For a neutral environment, the optimal remediants are ferromanganese nodules with biochar and iron powder without biochar. Thus, among those examined, an optimal remediant was found that works in both cases—iron powder without the addition of biochar.

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