Article AR2178a DOI: HTTPS://DOI.ORG/10.23910/1.2021.2178a

International Journal of Bio-resource and Stress Management

Crossref

February 2021

Print ISSN 0976-3988
Online ISSN 0976-4038

IJBSM 2021, 12(1):040-046

Review Article *Natural Resource Management*

Studies of Soils and Vegetation on Non-ferrous Metallurgy Slag Dumps

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Citation: Zolotova, 2021. Studies of Soils and Vegetation on Non-ferrous Metallurgy Slag Dumps. International Journal of Bio-resource and Stress Management 2021, 12(1), 040-046. HTTPS://DOI.ORG/10.23910/1.2021.2178a.

Data Availability Statement: Legal restrictions are imposed on the public sharing of raw data. However, authors have full right to transfer or share the data in raw form upon request subject to either meeting the conditions of the original consents and the original research study. Further, access of data needs to meet whether the user complies with the ethical and legal obligations as data controllers to allow for secondary use of the data outside of the original study.

Conflict of interests: The authors have declared that no conflict of interest exists.

Funding: The research was conducted with the kind and supports from the Zavaritsky Institute of Geology and Geochemistry, Ural Branch of Russian Academy of Sciences (in the framework of the state assignment No AAAA-A18-118052590028-9).

Abstract

The metallurgical industry is one of the major pollution sources of natural ecosystems. Now the slag dumps of non-ferrous industries occupy huge areas all over the world. The purpose of this literature review was to assess the knowledge degree of the soils and vegetation formed on the non-ferrous metallurgy slag dumps. Most of the research was carried out for the dumps of the copper-smelting (including old dumps) and lead-zinc industries, the dumps of the nickel and aluminum industries have been studied to a lesser extent. The composition of non-ferrous metallurgy slags, the issues of soil pollution with heavy metals, their bioavailability were discussed. The influence of heavy pollution on the biodiversity of pioneer plant communities on the slag dumps of non-ferrous metallurgy and the floristic composition for abandoned copper ore deposits are noted. The experience of Russian scientists in the reclamation of an aluminum sludge dump and Chinese scientists in the reclamation of zinc production slag dumps are considered. The possibility of introducing waste from the copper smelting slag recycling waste into natural ecosystems was discussed. The analysis of literature revealed gaps in knowledge about the gradual formation of the soil and vegetation on man-made landscapes, about the plant biodiversity in conditions of heavy pollution, ways of their adaptation, and the heavy metals accumulation by different plant species.

Keywords: Industrial dump, heavy metals, pollution, metallurgical slags

1. Introduction

The intensive development of the metallurgical industry leads to an increase in the areas of disturbed natural landscapes, pollution of the air, surface and ground waters, soils and vegetation (Norgate et al., 2007; Masloboev et al., 2014; Iles, 2016; González-Fernández et al., 2018). Pollution sources are gas emissions, waste water, slag and sludge dumps of industries. Quite a lot of research is aimed at assessing the impact of airborne emissions from metallurgical industries on biotic components of ecosystems (Serbula et al., 2013; Bergman and Vorobeichik, 2017; Nesterkov, 2019). At the same time, insufficient attention is paid to the study of the formation of soils, vegetation and microorganisms on the slag dumps.

Now a huge amount of industrial mineral waste has been accumulated (Petlovanyi et al., 2019; Alimbaev et al., 2020). Dumps of non-ferrous metallurgy slag are the most dangerous due to the high content of heavy metals (Dudka and Andriano, 1997). Toxic elements from slag are involved in biogeochemical cycles and lead to pollution of water bodies, decrease

Article History

RECEIVED in 12th January 2021 RECEIVED in revised form 16th February 2021 ACCEPTED in final form 25th February 2021

in soil fertility, degradation of vegetation and diseases of animals and people (Khorasanipour and Esmaeilzadeh, 2016; Gabasiane et al., 2019). The influence degree of the metallurgical slag dump on the environment depends on the granulometric, mineralogical and chemical composition of industrial waste and a complex of natural factors (Piatak et al., 2014). Old metallurgical slags are considered more hazardous for the environment than modern ones, as they contain more potentially toxic elements and were disposed of uncontrollably (Kierczak et al., 2013). The dangerous practice of placing metallurgical slags in the lowlands of small towns was in the past. For example, in Russia, the technozems of the Chusovoi city contain barium – 1000 mg kg^{-1} and chromium – 2000 mg kg^{-1} (Vodyanitskii et al., 2010).

Non-ferrous metallurgy slags are sent to dumps after preliminary granulation or in a hot state. Subsequently, slags are a parent rock for the formation of technogenic soil (technozem). The correct description and classification of these soils is a difficult issue (Sobocka et al., 2017). The composition, physical and chemical properties of the slag determine the soil properties, which in turn determines the ecological composition of the plants and the rate overgrowing of the dump. Climatic conditions determine only the species composition of pioneer plants (Shilova and Loginova, 1974). However, self-overgrowing of dumps is an extremely slow process and it has been mainly studied for dumps of various deposits, including copper ore (Zheleva et al., 2012).

The purpose of this literature review to assess the knowledge degree of the soils and vegetation formed on the non-ferrous metallurgy slag dumps.

2. Dumps of Copper Smelting Slag

Chile, Peru, China, USA, Congo, Australia, Zambia, Mexico, Russia are the major countries in copper mine production worldwide (Statista, 2020). The largest enterprises of the copper-smelting industry in Russia are the holding "Ural Mining and Metallurgical Company ("UMMC") (Sredneuralsk Copper Smelter, Mednogorsk Copper and Sulfur Plant, Svyatogor Smelter, "Uralelektromed" Copper Refinery), holding «Russian Copper Company» (Aktyubinsk Copper Company, "Karabashmed", "Uralgidromed", Kyshtym Copper Electrolytic Plant, Novgorod Metallurgical Plant, and other), as well as "Norilsk Nickel" – the world market leader for palladium and high-grade nickel and one of the largest producers of platinum, cobalt and copper.

The mineral basis of the copper smelting slag is fayalite ($2FeO \cdot SiO_2$), and the composition of minor minerals depends on the original ore, the metallurgy technologies, the time and conditions under which the slag was stored. For example, the Karabash copper smelter ("Karabashmed") black slags of the Soviet period are composed mainly of olivine-pyroxene aggregate with a significant glass content and the constant presence of chromite, wustite, and various sulfides of the Cu-Fe-S and Pb-Ni-S systems (Erokhin et al., 2019). The Polevskoy copper smelter slags (smelter operated until 1931) include technogenic silicate glass, pyroxene, magnetite and minerals related to ferrites. The rock-forming elements content in this slag: Si> 10%, Al> 5%, Fe> 5%, Ti – 0.106%, Mg – 0.29%, Ca – 0.37%, K – 0.23%, Na – 0.32% (Makarov et al., 2018).

Geochemical features of the technogenic soil and plants growing spontaneously on an old slag dump of the Polevskoy copper smelter (Middle Urals, Russia) were studied (Zolotova and Ryabinin, 2019). An environmental assessment of the dump was carried out. The maximum permissible concentration of total forms of dangerous chemical elements (MPC) are regulated in Russia by state documents for environmental objects (for soil this is GN 2.1.7.2041-06) and are used for ecotoxicological assessment of soil. The most significant excess of MPC for all regulated elements (Figure 1) in the soil formed on the slag dump of the Polevskoy copper smelter are observed in the fine soil (slag particles less than 1 mm), which makes up more than a third of the mass of technogenic soil and is a sorption geochemical barrier. It has been confirmed that in the conditions of unlimited supply of elements released from slag, plants reach the upper threshold of accumulation. The highest values of the biological absorption coefficient for the aboveground plants part were found for selenium, potassium, calcium and phosphorus; for plant roots and mosses – for selenium and aluminum (Zolotova and Ryabinin, 2019).

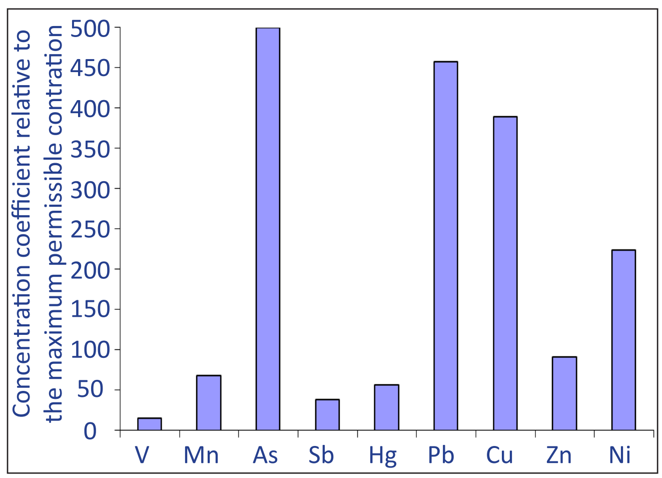

Figure 1: Environmental assessment of fine soil fractions of the technogenic soil of the old slag dump of the Polevskoy copper smelter (Middle Urals, Russia) (Zolotova and Ryabinin, 2019)

Polish scientists (Kierczak et al., 2013) studied the influence of older dumps (14th-16th century) of pyrometallurgical copper slag (porous and cast) on environmental objects. The studied dumps are located in the forests and riverbeds of the Rudava Yanovickie region. Chemical analysis of soils revealed excess of environmentally acceptable standards for the content of copper (up to 4000 mg kg^{-1}), zinc (up to 1500 mg kg^{-1}), arsenic (up to 300 mg kg^{-1}) and lead (up to 200 mg kg^{-1}) (Kierczak et al., 2013).

In the literature, we did not find data on the species composition and vegetation structure of slag dumps, but the floristic composition was studied for abandoned copper ore deposits (Zheleva et al., 2012; Turisova et al., 2016). Phytocenotic studies of the Elatsite copper-porphyry deposit dumps (Bulgaria) revealed 55 plant species, of which 32 are weeds. The dominant species are *Tussilago farfara* L. and *Silybum marianum* (L.) Gaertn. *Hypericum perforatum* L. and *Lamium purpureum* L. are relatively common (Zheleva et al., 2012). The heavy metals content in the technogenic soil of old abandoned deposit Pieski (Slovakia) remains high (copper concentration from 933.40 to 1485.40 mg kg^{-1}), under these conditions 156 taxa of vascular plants were found. The most common species are *Acetosella vulgaris*, *Agrostis capillaries* and *A. Stolonifera*, *Arabidopsis arenosa*, and *Festuca rubra* (Turisova et al., 2016).

Studies of the microbiological situation in technogenic soils formed on slag dumps have not been found in the literature. Attempts to assess the effect of pyrometallurgical slags on the rhizosphere microorganisms diversity were made in a pot experiment on growing sunflower (Helianthus annuus) on a 50% mixture of agricultural soil and granulated copper smelting slags (Agnello et al., 2018).

3. Slag Dumps of Lead-Zinc Production

The major countries in lead and zinc mine production are China, Australia, Peru, USA, Mexico, India, Russia, Bolivia, Kazakhstan (Indian Minerals Yearbook, 2019). The lead-zinc industry in Russia is represented by the Belovsky Zinc Plant (part of the “SIBPLAZ” inter-industry holding), the Chelyabinsk Electrolytic and Zinc Plant, the Sadonsky Lead-Zinc Plant, “Dalpolimetal”, “Electrozinc”, “Ryaztsvetmet”, and other.

Slags of the lead-zinc production “Dalpolimetal” (Primorsky Territory) are classified by mineral composition as medium iron (Fe_2O_3 – 14.0-25.0 wt.%), magnesian (MgO – 8.0-16.0%), alumina (Al_2O_3 – 4.0-11.0%), relatively rich in calcium oxide (CaO – 9.0-20.0%), and contains SiO_2 from 18.0 to 33.0% by weight. The heavy metals concentration varies within (n × 10-3, wt.%): Pb – 2.0-16.0; Zn – 2.0-13.0; Cu – 0.1–2.0; Mn – 0.01–1.0 (Zemnukhova and Falaleeva, 2011). Studies on the formation of soil and vegetation cover on these dumps (and near them) have not been found.

Chinese scientists conducted comprehensive studies on reclamation of zinc production slag dumps (Luo et al., 2018, 2019, 2020). *Arundo donax*, *Broussonetia papyrifera*, *Robinia pseudoacacia* and *Cryptomeria fortune* were planted. The studies were carried out 5 years after reclamation. It was found that the restoration of the vegetation cover plays an important role in changing the physicochemical properties of the slag substrate, for example, such as humidity, pH; there is an increase in the accumulation of nutrients and a decrease in the bioavailability of heavy metals (Cu, Zn and Cd), with the exception of lead – its mobility increases (Luo et al., 2019). An increase in the number and diversity of rhizosphere bacteria was recorded in the technogenic soil after reclamation, and the content of available forms of zinc and cadmium had the greatest effect on their composition (Luo et al., 2018).

Special mention should be made of studies aimed at assessing the impact of extreme heavy metal pollution (and other properties of technogenic soils) on the structure and biodiversity of pioneer plant communities on post-smelting dumps (Osyczka and Rola, 2013; Rola et al., 2015). The study area is an Upper Silesian industrial region in southern Poland. Scientists divided the studied lichens, mosses and vascular plants into three groups using modern statistical methods: (i) species that are resistant to pollution and are more productive at higher concentrations of heavy metals; (ii) species growing only in less polluted areas; (iii) plants that are indifferent to heavy metal pollution and are abundant in all dumps. The first group mainly includes lichens (for example, of the genus Cladonia (Osyczka and Rola, 2013). Increased concentrations of heavy metals negatively affect the biodiversity of vascular plants. The authors conclude that lichens are effective pioneer species in the non-ferrous metallurgy slags dumps and are an important element of natural vegetation restoration, which should be take into account during reclamation (Osyczka and Rola, 2013; Rola et al., 2015).

Other scientists (Houben et al., 2013) studied the slag dump of zinc production, where the vegetation spontaneous recovered, and made some conclusions. First, the metal leachability increased in the revegetated soils, in particular, due to the higher release of organic anions. Secondly, the metals mobility depends on growing plant species. The highest leachability of Cd was found in the soil covered by *Agrostis tenuis*, while the highest leachability of both Zn and Pb was observed in the soil below *Armeria maritima*. They concluded that, when using pioneer plants for phytostabilization purposes, preference should be given to pseudo-metallophyte over hyperaccumulator species (Houben et al., 2013).

4. Slag Dumps of Nickel Production

Indonesia, Philippines, Russia are considered the world leaders in nickel production (Indian Minerals Yearbook, 2019). Large enterprises for the processing of copper-nickel ores in Russia are the Kola Mining and Metallurgical Company (“Norilsk Nickel”: “Severonickel” and “Pechenganikel”), the inter-industry holding “SIBPLAZ” (“Ufaleinickel”), the South Ural Nickel Combine.

The mineral matrix in the slags of the “Severonikel” smelter (Monchegorsk, Murmansk oblast) is represented by calcium aluminosilicate $CaO \bullet 2Al_2O_3 \bullet SiO_2$, and the technogenic ore phases are pyrrhotite, whose structure includes nickel $(Fe, Ni)_9S_8$, and zinc spinel (ganite $ZnO \bullet Al_2O_3$) (Shadrunova et al., 2013). Chemical analysis of nickel production slags revealed the presence of sulfur (5-10%), chromium (0.4%), nickel (0.1%), copper (0.2%), and cobalt (0.05%) impurities

(Parshina and Korelskiy, 2008). The elements leachability strongly depends on the slag age, the maximum values are noted for 15-year-old waste and amount to 10-20 mg l^{-1} for chromium, 7-11 mg l^{-1} for sulfur, 5-8 mg l^{-1} for copper, 1.5-2 mg l^{-1} for nickel and 0.5-1.5 mg l^{-1} for cobalt. The formation probability of acidic drainage water increases with the operation time of the dump, due to the oxidation of sulfur in the hypergenesis zone, and reaches a maximum in 15 years, then remains unchanged due to the increase in fracturing of the newly received waste. “Severonikel” slag dumps form acidic drainage waters (pH = 3), which form technogenic halos with an area of 58 km^2 and pollution streams 15-20 km long (Parshina and Korelskiy, 2008).

Weathering of nickel smelting slag (3 kinds of slag) was studied for dump produced during reworking of lateritic Ni ores in Szklary (Lower Silesia, southwestern Poland) (Kierczak et al., 2009). The slags have been exposed to atmospheric conditions for 30-80 years, those occurring in the dump are not affected by weathering, and small vitreous slag fragments occurring in nearby agricultural fields have only thin (<100 µ m) crusts due to weathering. Some potentially toxic elements are concentrated in silicates: diopside is enriched in Cr (up to 2.3 wt.% Cr_2O_3), forsterite in Ni (up to 1.7 wt.% NiO), and melilite in Zn (up to 0.7 wt.% ZnO), but their reactivity was found to be limited in the alkaline soils (Kierczak et al., 2009).

Environmental monitoring studies of the soil and plants in forest ecosystems located in the impact zone of nickel production conducted (Korelskiy, 2013; Lyanguzova et al., 2016). The dependence of the level of contamination of the upper horizon of Podzols Rustic (Al–Fe-humus podzols) with heavy metals on the distance from the “Severonickel” was studied in the medium-aged pine stands (Lyanguzova et al., 2016). In the buffer zone of the smelter, the concentrations of Ni and Cu exceed background values by 8-17 times; in the impact zone, by 50-100 times. Firm bounding of heavy metals in the organic horizon coupled with their continuing aerial input did not allow the beginning of the soil self-purification process, which might last for decades and centuries (Lyanguzova et al., 2016).

5. Slag Dumps of Aluminium Production

China, Russia, Canada, India, United Arab Emirates, Australia, Norway, Bahrain, Saudi Arabia, USA are the ten largest aluminium producers in the world (Indian Minerals Yearbook, 2019). The aluminium industry of Russia was represented by enterprises of the “RusAl” holding (Achinsk Aluminium smelter, Krasnoyarsk Aluminium smelter, Novokuznetsk Aluminium smelter and other) and “SUAL” holding (Irkutsk Aluminium smelter, Ural and Bogoslovsky aluminium smelters, and other), these holdings in 2007 year merged with Swiss commodity trader “Glencore International” and formed the world’s largest aluminium company “United Company Rusal”.

The mineralogical basis of the Ural and Bogoslovsky aluminium smelters red mud consists of iron and aluminium-containing minerals–hydrogoethite, limonite, chamosite, pyrite, natrolite, etc. (Shilova and Loginova, 1974). Comparative analysis of the individual elements content (including those toxic to plants: Ni, Co, Pb, S) in red mud and in soil showed significant excess (1-2 magnitude orders). The red mud has a strongly alkaline reaction aqueous medium and a high content of harmful salts. Scientists (Shilova and Loginova, 1974) noted a single-species phytocenosis from *Suaeda corniculata* (C. A. Mey.) Bge. on the Ural aluminium smelter dump, however, vegetation covered an extremely insignificant part of the dump. Subsequently, they conducted an experiment on the settlement of the red mud dump with perennial grasses and legumes, and concluded that biological reclamation of this dump is possible only after root reclamation, i.e. the roots of cultivated plants should be isolated from the negative influence of aluminium red mud (Shilova and Loginova, 1974).

The toxicological studies of light gray forest soils and plants in the zones affected by slag dumps of aluminium smelter casting in the Oryol region are presented (Stepanova et al., 2020).

The elements absorption by plants growing on an abandoned site of an aluminium smelter (Smokey Mountain Smelters, Knoxville, Tennessee USA), where slag waste was dumped, has been studied (Abercrombie et al., 2011). ICP analyses indicated the highest slag metal concentrations were 223,000 mg kg^{-1} Al, 281 mg kg^{-1} As, 132 mg kg^{-1} Se, and 2910 mg kg^{-1} Cu. Pteris cretica accumulates Al in high concentrations, but not As. Metal concentrations in plants grown on slag were lower than controls grown in uncontaminated soil, suggesting low metal availability or root exclusion mechanisms (Abercrombie et al., 2011).

When analyzing the literature, I came across the interesting study on the phytoremediation of effluents from aluminum smelters using aquatic plants: *Typha latifolia*, *Lemna minor*, *Nuphar variegatum* and *Potamogeton epihydrus* (Goulet et al., 2005). L. minor had the highest Al uptake rate (0.8–17 mg Al g^{-1} d^{-1}). However, because T. latifolia (cattails) yielded the highest biomass, it was responsible for 99% of the Al uptake, largely in its root tissue (Goulet et al., 2005).

6. Metallurgical Slag Recycling Waste

The dumps of the nonferrous industry cause significant damage to the environment, which is why the recycling of metallurgical slags and competent reclamation of dumps are so important (Dudeney et al., 2013; Jain et al., 2016). Currently, non-ferrous metallurgy enterprises are improving technologies and introducing methods for recycling waste slag. New types of mineral waste appear, they have properties different from the original metallurgical slag. As an example, I considered the cast slag recycling waste from the Sredneuralsk copper smelter (Middle Urals, Russia).

In Russia, the first successful attempts to process dump cast slag as an unconventional source of copper date back to the 90s of the 20th century. The technology consists in grinding cast slag followed by flotation extraction of copper concentrate. The resulting copper concentrate with a solids content of 50-60% and a copper concentration of 10 to 25% through pipelines enters the batching department of the copper smelter, and magnetite-containing sands are filtered and stored in dry dumps – open storage warehouses (Makarov et al., 2010). The copper smelting slag recycling waste ("technical sand") is finely dispersed material with a dimension of 0.05 mm and its properties are poorly understood (Kotelnikova and Ryabinin, 2018). The experiment to assess the elements mobility from the copper smelting slag recycling waste into brown forest soils (Haplic Cambisols) under the canopy of pine forests and on clear-cut areas in the southern taiga district of the Trans-Ural hilly-foothill province (Middle Urals) was carried out (Zolotova et al., 2021). It was found that the waste after being in the soil for two years loses 11% of its mass. Most of the chalcophilic elements are involved in the biogeochemical cycle. The content of zinc, arsenic, cadmium, selenium decreases most strongly. The difference in the elements migration from "technical sand" into brown forest soils of two forest types and clear-cuttings (determined according to the genetic forest typology) was revealed (Figure 2). Mostly for all chalcophilic elements, the maximum migration was noted for the soils under the berry pine forest with linden, and the minimum – for the soils of the cowberry shrub pine forest. It was found that a single surface application of 1 kg m^{-2} of the copper smelting slag

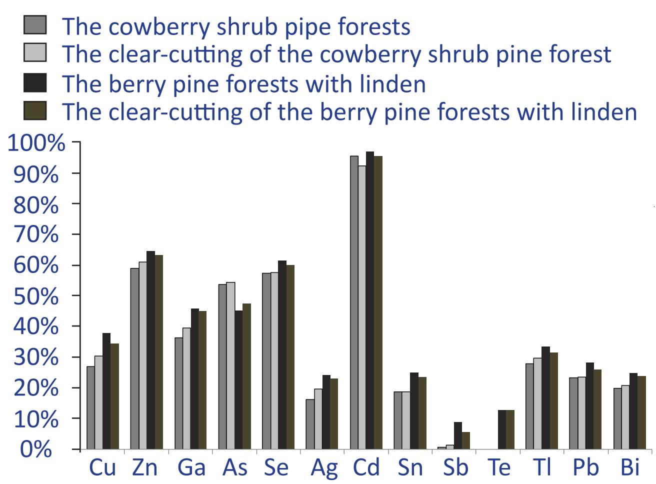

Figure 2: Changes in the content of chalcophilic elements (% of the elements content in the initial slag) of the copper smelting slag recycling waste ("technical sand SUMZ") after two-year stay in the humus horizon of brown forest soils of two forest types and clear-cuttings in the Trans-Ural hilly foothill province (Middle Urals, Russia) (Zolotova et al., 2021)

recycling waste to forest soils in the autumn period did not affect the qualitative composition of the herbaceous layer (dominant and diagnostic species) of the studied forest types and corresponding clear-cuttings in the next spring-summer period (Zolotova et al., 2021).

The development of methods for introducing metallurgical slag recycling waste into the soil of natural ecosystems would make it possible to solve the extremely urgent problem of recycling industrial waste. However, all this is possible only with a thorough and comprehensive study of slag recycling waste, and provided there is no negative impact on the environment.

7. Conclusion

Monitoring studies of non-ferrous metallurgy slag dumps are necessary for the purposes of sustainable development and environmental safety of the regions. The analysis of the conducted research revealed gaps in knowledge about the gradual formation of the soil and vegetation on man-made landscapes, about the plant biodiversity in conditions of heavy pollution, ways of their adaptation, and the heavy metals accumulation by different plant species.

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