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SHORT COMMUNICATION

Heavy metals contamination of post-mining mounds of former iron-ore mining activity

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Abstract

The main question of the present study is how much are the former post-mining mounds (PMM) - the ancient remnants of mining of a provisional nature located in forest areas-contaminated by heavy metals (HM). To investigate if the PMM contain HM, we collected 15 soil samples from PMM and, for comparison, 15 from the surroundings - all these samples (30) were collected from a depth of 5–30 cm by soil auger. To find how deep the contamination of HM goes, we did 4 soil profiles to the depth 100 cm in four randomly selected PMM. In every soil profile, 4 soil samples were collected (16 soil profile samples). In total, 46 soil samples were analysed. Concentrations of HM were measured using spectrophotometry. Our results indicate the following: (1) PMM are not much contaminated by HM - only two (Fe and Mn) from nine HM significantly exceed the limits - the order of abundance of the studied HM is as follows: Fe > Mn > Cr > Zn > Ni > Cu > Pb > Co > Cd; (2) PMM are more contaminated by heavy metals than their surroundings; (3) within PMM, overburden is much more contaminated by HM than paleosols; (4) the rate of penetration of HM into the depth of the soils (into paleosols) is reduced due to the properties of the overburden of PMM.

Keywords Clay ironstones · Not transformed areas · Overburden · Paleosols · Soil profile · Soil samples

Introduction

Heavy metals naturally present in the environment, but most emissions of these metals are due to variety of economic activities, e.g. industry (Contreras-Tereza et al. 2021), application of fertilizer (e.g., Rashid et al. 2023; Salem et al. 2020), or traffic density (Cesur et al. 2021; Cetin and Jawed 2022; Sevik et al. 2020). All these human activities have negative impact on nature environment (e.g., Aricak et al. 2020; Bozdogan et al. 2019; Pekkan et al. 2021; Siddique et al. 2021; Yücedağ and Kaya 2019; Zhao et al. 2022).

One of the world's largest industries is mining. The exploitation of mineral resources has long been a tradition all over the world and has resulted in serious environmental impacts (Aradhi et al. 2023; Camizuli et al. 2018; Mwesigye et al. 2016; Wahsha et al. 2019). These effects are determined by the time and methods of exploitation and type of material exploited (Khan et al. 2016; Verma et al. 2021; Yan et al. 2018). One of the unwanted results of mining is heavy metal (HM) pollution (e.g., Adewuyi and Osobamiro 2016; Chileshe et al. 2019; Dusengemungu et al. 2022; Hasimuna et al. 2021). Nowadays, in many post-mining sites the HM concentration has reached an alarming proportion that merits attention because it is a problem of an ecological, evolutionary, nutritional, and environmental nature (Abiya et al. 2019; Jaishankar et al. 2014; Lasheen et al. 2022; Nagajyoti et al. 2010; Peša 2021).

Various studies have shown that soils of mining areas are seriously polluted by HM (Bu et al. 2020; Xin et al. 2022) and all remnants of mining activities present a perpetual danger of moving and transforming toxic elements (Assabar et al. 2023; Fazekašová and Fazekaš 2020). Higher concentrations of HM in soils have been documented at a variety of sites associated with, inter alia, Pb–Zn mining (Mayanna et al. 2015; Potra et al. 2017; Woch et al. 2015), or Ag-Pb mining (e.g., Cabała et al. 2020) A great deal of attention

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has been paid to the currently existing large area mining which is considered environmentally loaded and unhealthy, but former post-mining sites also deserve attention.

The former post-mining fields emerged about 300 years ago (eighteenth century) in mines of iron-ore (clay ironstones) of a still provisional nature. The iron ore was extracted by hand (without machines, so without any additional pollution), and the method consisted of digging many narrow (0.5–1.5 m wide), fairly deep (from several metres to several tens of metres) shafts, whose shaft top looked like an ordinary well with a windlass. A single shaft was most often operated by three miners. One worked in the shaft, two others brought up the output to the surface using ropes (Podgórska 2019). On a mining field, the shafts were dug out very close to one another. The pieces of iron ore were picked up from the excavated material while the unwanted tailings were left around the shafts. Owing to this practice, up to this day old mining fields are dotted by hundreds of mounds near the abandoned shafts (Podgórska 2015, 2016).

The remnants of former mining activity, post-mining mounds (PMM), look like small hips (0.3–3.5 m high) of excavated substrate (unprocessed material, extracted from deep rock layers) left behind by the miners (Fig. 1A). Because of this, they are different from commonly occurring heaps of post-processing mining waste (e.g., Wang et al. 2017; Wu et al. 2021).

Taking into account the current state of knowledge about the post-mining areas in the context of contamination with heavy metals (HM), the authors set three research hypotheses about the remnants of mining of a provisional nature, post-mining mounds (PMM): *i*) PMM are heavily polluted by various kinds of HM; *ii*) PMM are more contaminated by HM than their surroundings; *iii*) the HM occur in the same amount on the PMM and at the depth of 1 m of the paleosols soil profile.

To verify the above-mentioned hypotheses, the authors set two main goals: (*i*) to investigate if PMM are generally contaminated with HM and if the potential content of HM is higher than in the surroundings; (*ii)* to find how severe the contamination of heavy metals in the soil profile is.

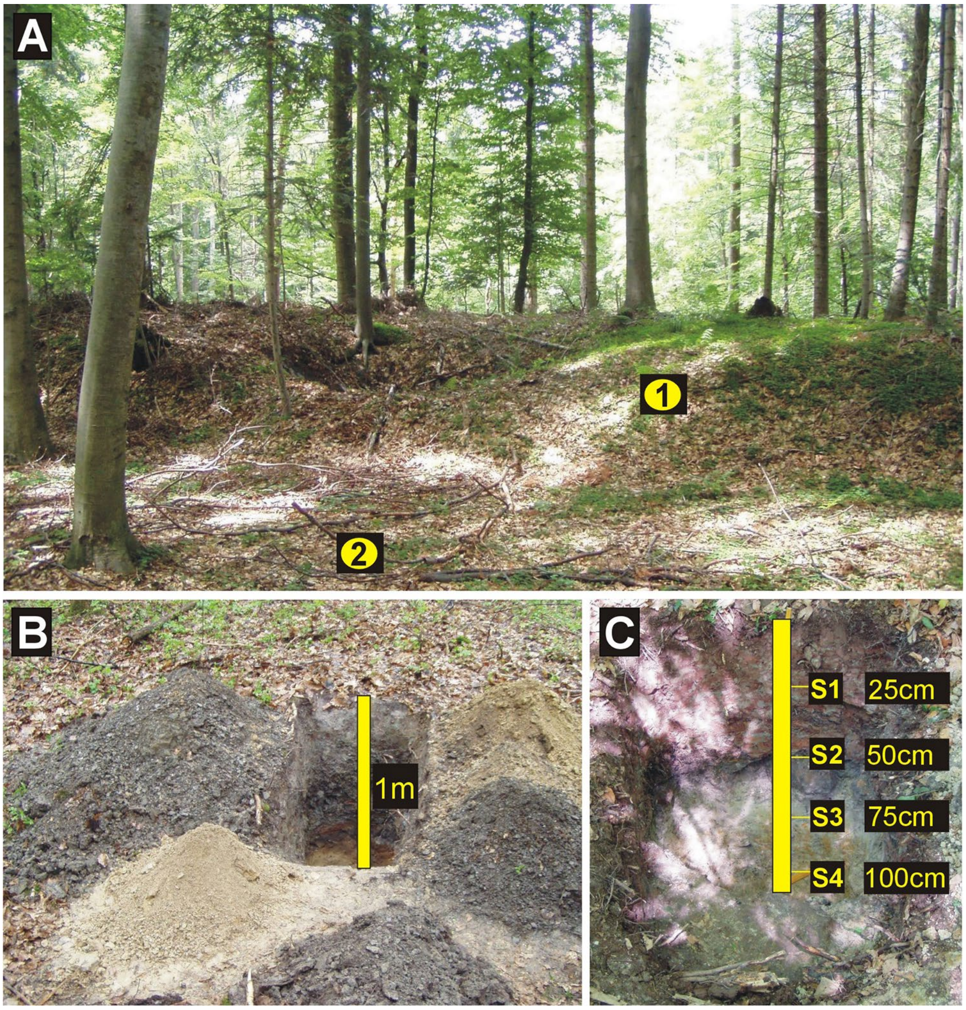

Fig. 1 A sample PMM (**A**) with an indication of the place where a sample soil profile was taken (1), and the surroundings of PMM (2); **B** a model scheme of the vertical distribution of sampling in the soil profile (**C**): S1-overburden, S2–S4-paleosols (Photo: M. Podgórska)

Materials and methods

Study area

The research was carried out in a forested area in the northern foreland of the Świętokrzyskie Mountains (Poland, Central Europe) on the remnants of former iron-ore mining activity (focused on clay ironstone mining) – former iron-ore post-mining mounds (PMM) located in eighteenth century post-mining fields. From the viewpoint of its geology, the study area occupies the northern, Mesosoic periphery of the Paleozoic core of the Świętokrzyskie Mountains. The northern part of the study area is built of Lower Jurassic forms, whereas the southern part of the area is built of Triassic formations. In both formations, ore-bearing horizons occur. In the lithological-stratigraphic profiles performed in both types of formations, three principal horizons stand out: the under ore-bearing horizon (made of compact sandstones), the ore-bearing horizon (made of clays and shales, marls, and clay ironstones), and the above-ore-bearing horizon (made of sandstones with clay intercalations) (Kleczkowski 1970). As a result of old mining activities, in places where mining shafts were excavated, there was a reversal of the distribution of rock layers which occurred due to the transfer of the deeper formations onto the surface. At present, in the places where the PMM occur, the formations of above-ore-bearing horizons (actually paleosols) are covered by the formations of ore-bearing horizons (actually PMM) (Podgórska 2019 and the literature cited there). It is worth saying that all post-mining fields under study are located in the large forest complexes away from any human settlements (Podgórska 2018) and consequently they are not contaminated by current management.

Field sampling and data analysis

To investigate if the PMM contain HM, we collected 15 soil samples from the PMM and, for comparison, 15 from the surroundings (areas not transformed by mining) - 10 m away from each PMM. All these samples (30) were collected from a depth of 5–30 cm by soil auger.

To find how deep the contamination of HM goes, we did 4 soil profiles to the depth 100 cm in four typical, randomly selected PMM. Every soil profile was dug on the PMM slopes by spade. In every soil profile, 4 soil samples were collected and sectioned (Fig. 1B) - the first soil samples (S1) were collected from the overburden (from a 25 cm depth) and the last samples from paleosols at about 25 cm intervals: S2 (50 cm), S3 (75 cm), and S4 (100 cm). Therefore, from four soil profiles 16 soil profile samples were collected.

Soil samples (in total 46 soil samples) were collected with a polyethylene scoop and stored in plastic bags. The soil samples were air-dried and passed through a 2 mm plastic sieve to remove gravel and rocks, put in plastic bags, then sent to the Environmental Research Laboratory of the Department of Environment Protection and Modelling (UJK, Poland) for analysis. Concentrations of 9 heavy metals (Cd, Cr, Cu, Co, Fe, Mn, Ni, Pb, Zn) were measured using an ICP-MSTOF (OptiMass 9500 GBC) spectrometer (Producer: GBC Scientific Equipment Pty Ltd). Despite the fact that these two used methods (classical method of soil science - collection of soil samples during field studies and laboratory analysis of soil samples with the use modern equipment - spectrometer) are not innovative they allowed to examine the contents of HM of post-mining mounds in an objective way - they were previously used in several similar studies (Matini et al. 2011; Świercz et al. 2023). The obtained results were compared to the limit values of heavy metal content in soils (Table 1) according to European (Rademacher 2003) and Polish norms (Regulation of the Polish Minister of Environment 2002).

To find the difference between PMM and their surroundings, nonparametric U-Mann-Whitney test was performed. The nonparametric Kruskal–Wallis H test was applied to check whether there were significant differences of HM content between the soil horizons. Pearson's correlation coefficients (r) were used in order to detect the relationships among the HM and the depth. All the results of the conducted analyses were considered as statistically significant at $p \leq 0.05$.

Table 1 The limit values for heavy metals (HM) according to European (Rademacher 2003) and Poland norms (Regulation of the Polish Minister of Environment 2002)

HM	Fe	Mn	Cr	Zn	Ni	Cu	Pb	Co	Cd
Reference values of Poland (mg/kg)	–	–	150	300	100	150	100	20	4
Reference values of Europe (mg/kg)	30,000	450	130	250	85	140	150	50	2

Results and discussion

Difference of content of heavy metals between the post-mining mounds and its surroundings

In the PMM, much higher contents of the analysed HM (Cd, Cr, Cu, Co, Fe, Mn, Ni, Pb, Zn) were found than in their surroundings (Table 2). Statistically significant differences between PMM and their surroundings were observed in the case of eight HM (Table 2). However, in the PMM, only three HM exceeded the limits - these were iron (range: 52,203.5–543,880.8 mg/kg), manganese (range: 680.2–27,637.4 mg/kg), and chromium (range: 58.4–220.6). In the surroundings, only iron exceeded the limits (range: 2928.4–118,390). The contents of remaining HM were still within the normal range (Table 2). For both the PMM and the surroundings, the order of abundance of the studied HM was as follows: Fe > Mn > Cr > Zn > Ni > Cu > Pb > Co > Cd.

Difference in the heavy metal content between overburden and the paleosols within PMM

The most polluted layer was the overburden (S1)-the material of which the PMM are built, exploited from the deepest layers (from the ore-bearing horizon) during former mining activities. In this layer, all of the 9 analysed HM were found (Table 3). From among the paleosols, the S2 sample was the most polluted. As in the overburden, in the S2 sample all of the HM occurred. In the S3 sample as well as the deepest sample (S4), 8 analysed HM occurred-cadmium was not found (Table 3).

The analysis showed that there is a negative correlation ($r = -0.70$, $P = 0.002$) between the depth and the HM

Table 2 Difference of the HM content between the PMM and the surroundings. U - value of the U-Mann-Whitney test

HM (mg/kg)	PMM	Surroundings	U
Fe	283,816.2 (52,203.5–543,880.8)	40,331.4 (2928.4–118,390)	U = 14.0****
Mn	9974.9 (680.2–27,637.4)	115.1 (2.9–345.6)	U = 0.0****
Cr	104.1 (58.4–220.6)	94.3 (43.7–150)	U = 109.0 ^{ns}
Zn	30.7 (15.1–48.9)	12.5 (1.7–24.6)	U = 8.0****
Ni	28.1 (2.3–77)	3.4 (1–7.4)	U = 32.0***
Cu	18.9 (3.2–46.8)	3.6 (1.7–5.1)	U = 22.0***
Pb	13.3 (2.7–56.4)	4 (0.5–13.6)	U = 56.0*
Co	5.1 (2.2–14.8)	0.7 (0.1–1.9)	U = 0.0****
Cd	0.1 (0.1–0.2)	0.05 (0–0.1)	U = 42.0**

*$0.05 \geq P \geq 0.01$; **$0.01 > P \geq 0.001$; ***$0.001 > P \geq 0.0001$; ****$P < 0.0001$; *ns*-not statistically significant

Table 3 Difference of the HM content between the soil horizons (mean–range); S1–S4 - successive soil samples collected from different depths of the soil profiles (S1-25 cm, S2-50 cm, S3-75 cm, S4-100 cm) and the total content of HM in the whole soil profiles of the PMM. H - value of the Kruskal–Wallis H test

HM (mg/kg)	overburden		paleosols		H
	S1	S2	S3	S4	
Fe	256,512.8 (523,880.8–62,203.5)	22,193.9 (40,659.8–12,658.4)	14,734.7 (31,732.2–4301.1)	42,898.3 (65,190.1–17,614.5)	H = 12.44**
Mn	7281.3 (26,637.5–760.2)	100 (246.3–0)	12.6 (30.1–0)	0	H = 13.22**
Cr	100.5 (208.4–60.9)	191.6 (260.6–162.8)	181.3 (212.8–159.1)	56 (67.4–35.3)	H = 9.31*
Zn	29.8 (48.9–18.6)	5.7 (11.7–2.5)	3.2 (6.4–0.5)	5.4 (9.6–2.1)	H = 11.41**
Ni	24.8 (68.4–2.3)	4 (7.8–2.6)	2.2 (3.4–1.1)	2.8 (4.5–1.4)	H = 6.06 ^{ns}
Cu	16.2 (37.9–4.8)	4.9 (8.4–3)	2.9 (3.2–2.7)	5.5 (8.6–3.6)	H = 10.65*
Pb	11.2 (44.9–2.9)	5.7 (14–1)	1.1 (1.8–0.6)	1.5 (2.6–1.9)	H = 9.92*
Co	5.01 (11.9–2.2)	0.4 (0.7–0.2)	0.3 (0.5–0.1)	0.2 (0.3–0.2)	H = 11.61**
Cd	0.2 (0.2–0.1)	0.07 (0.1–0)	0	0	H = 10.42 *

*$0.05 \geq P \geq 0.01$; **$0.01 > P \geq 0.001$

^{ns}Not statistically significant

content of all the studied samples of all the soil profiles - the overburden (S1) and paleosols (S2, S3, S4) - with their contamination decreasing substantially with soil depth. Statistically significant correlations were found for 8 of the 9 HM (Table 4), and the highest one was for manganese ($r = -0.91$, $P < 0.0001$). Only for chromium (Cr) was the correlation not statistically significant (Table 4).

Discussion

Our results show, surprisingly, that PMM are not as much contaminated by HM as the first hypothesis posits, so the first working hypothesis seems to be rejected. Only three from nine HM exceeded the limits (iron, manganese, and chromium). Additionally, only iron and manganese significantly exceeded the limits (Tables 1 and 2). The high values of these HM are connected with the kind of rocks of the ore-bearing horizon which were brought to the surface by miners with the object of extraction-clay ironstones, where iron and manganese occur naturally (Podgórska 2018, 2019). In the other locations where ironstones were exploited (e.g. in the Republic of Slovakia), iron is also considered as one of the most serious pollutants (Fazekašová and Fazekaš 2020). Moreover, in Pb–Zn post-mining sites, Pb and Zn have the highest values, because these metals simply occur naturally in the ore-bearing horizon (e.g. Oyebamiji et al. 2018; Potra et al. 2017; Świercz et al. 2023; Wang et al. 2018).

The contamination of the other HM in the PMM was very low. This is an unexpected situation, because other post-mining areas are usually highly contaminated by many HM, even if we are dealing with ancient activities (e.g. Assabar et al. 2023; Cabała et al 2020; Camizuli et al. 2018; Demková et al. 2017; Fashola et al. 2016; Rożek et al. 2015). The low contamination of the other HM in the PMM may be connected with the not very invasive, provisional methods of extraction. In the study area, iron ore was extracted by hand (without machines so without additional contamination) (Podgórska 2019). Additionally, the fact that PMM are not heavily polluted by HM allowed for a significant increase in species richness within them (Podgórska 2015, 2016).

It is also worth emphasizing, that PMM are covered by ancient forests (Podgórska 2018), which, to a large extent, restrict the spread of current contamination from other sources, e.g. agricultural activities, that we are facing at other extraction sites (e.g. Alengebawy et al. 2021; Jin et al. 2019; Ma et al. 2022).

Even if the content of many HM in the PMM do not exceed the limits, the obtained findings show that the PMM are more highly contaminated by heavy metals than their surroundings (not transformed areas). This confirms the second working hypothesis. A similar situation was observed by Strzeleczek et al. (2017) on the remnants of mid-forest iron ore excavations, where not transformed areas in the vicinity of mounds had a lower content of HM.

The last working hypothesis seems to be rejected, because the PMM are much more contaminated by HM than paleosols. On the PMM, the most polluted layer is the overburden, with the highest contamination of iron and manganese. In many post-mining polluted sites, the most polluted layer is the 'top layer' (e.g. Camizuli et al. 2018; Wahsha et al. 2019), as is the case in our study area. And it is well known that the top layers can strongly affect HM contamination of the local soils (e.g. Bu et al. 2020; Chen et al. 2018). The mobility of HM in the substrate primarily depends on its physicochemical characteristics (e.g. Sarapulova et al. 2017). We discovered that the HM contamination decreased substantially with soil depth. This is a different situation than in other post-mining sites, where in many cases an increase in HM concentration with increasing soil depth was observed (e.g. Huang et al. 2017). The decrease in the HM concentration observed on the PMM may be caused by the physical properties of the PMM (overburden) - we are dealing here with a poorly permeable substrate with a very high proportion of clayey particles (Podgórska and Jóźwiak 2020), which significantly hinders the penetration of HM.

Conclusion

(1) Ancient remnants of former iron-ore mining made of overburden (PMM) are not very highly contaminated by heavy metals (HM). Only two (iron and manganese) from nine HM significantly exceed the limits. The high values of these HM are connected with the kind of rocks of the ore-bearing horizon, where iron and manganese occur naturally.

(2) Even if the content of many HM in the PMM do not exceed the limits, the obtained findings show that the PMM are more highly contaminated by heavy metals than their surroundings (not transformed areas). For

Table 4 Pearson's correlation coefficients (r) for the HM content and the depth of soil profiles of the PMM

HM	Fe	Mn	Cr	Zn	Ni	Cu	Pb	Co	Cd	Total
Depth	− 0.68**	− 0.91****	0.06	− 0.79***	− 0.70**	− 0.64*	− 0.59*	− 0.83***	− 0.69*	− 0.70**

*$0.05 \geq P \geq 0.01$; **$0.01 > P \geq 0.001$; ***$0.001 > P \geq 0.0001$; ****$P < 0.0001$

both the PMM and the surroundings, the order of abundance of the studied HM is as follows: Fe > Mn > Cr > Zn > Ni > Cu > Pb > Co > Cd.

(3) The overburden are much more contaminated by HM than paleosols within PMM. The rate of penetration of HM into the depth of the soils (into paleosols) is reduced due to the properties of the overburden of PMM.

(4) The obtained results shed new light on the ancient mining activity. The findings reveal that the remnants of former mining activity - the ancient post-mining mounds are slightly contaminated by heavy metal. This, in turn, might suggest that they do not pose a threat to the natural environment. In addition, all of them are covered with natural forests which were formed as a result of centuries - long succession, so they do not need any reclamation treatments.

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Declarations

Conflict of interests The authors declare that they have no conflict of interest.

Competing interests No competing interests have been declared.

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