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Assessment of metallic trace elements contamination in a mining area of the processing of Djebel Onk phosphate ore (Algeria)

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Abstract

Research relevance. Phosphate mining activities are one of the main sources of contamination by heavy metals since they harmfully affect the soil and lead to the degradation of the ecosphere.
Research aim. This study aims at characterizing and evaluating the metallic contamination of soils in the vicinity of the Djebel Onk mine in the town of Bir El Ater (Wilaya of Tébessa) in eastern Algeria, and particularly focusing on estimating the spatial variability of this contamination and the extent of the contaminated area.
Methodology. The physicochemical characteristic (pH) was determined by pH-meter, whereas AAS was used for the determination of heavy metals (Pb, Cu, Zn, Cd) in the examined soil
Research results. The results reveal that soils that are close to the mine site studied and even those that are far away are heavily contaminated with heavy metals — lead (Pb), zinc (Zn), copper (Cu) and cadmium (Cd); the results also point out to a high variability of concentrations not only between sampling sites but also within the same mine site. The analysed soil pollution index is generally high even for soils sampled downstream more than 30 km from the mine site, whereas it is extremely high on the surface of the tailings slopes, underlining the fact that tailings are considered perennial sources of heavy metal contamination in their current state.
Conclusion. The unhealthy area affected by metal pollution from mining sites in the Djebel Onk is very large as a result of the dispersion, by wind and water transport, of residual pollutants from the mine wastes abandoned on site. Remediation measures must be put into place to immobilize the pollutants and limit their spread to the environment

***Keywords*:** Soil contamination, Algeria, heavy metals, phosphate mine, pollution index.

Introduction

The global development of industrialization has led to the massive use of several types of minerals alongside significant growth in mining activities. These mining activities are considered to be highly pollutant as well as their impact on the environment and health are on the scale of their economic importance [1–7]. Most mining pollutants are harmful to the flora and fauna of the terrestrial and aquatic environments. In addition to acid leachate and mine tailings, which cause deterioration of ecosystems, trace metal elements (TMe) cause even more harm on the physiological functions of living organisms [8–13].

At low doses, metals such as iron, zinc, chromium and copper are necessary for everyday life. However, when they are at high doses, they become toxic and can cause significant functional complications and serious health problems. In addition, some metals such as lead or arsenic are highly toxic and are classified as "hazardous air pollutants" [2, 14].

Several studies have looked at TMe pollution of mine sites and their surroundings and the impact of these activities on the environment [4, 15–26].

These studies have shown that pollution by TMe can affect the ecosystem irreversibly if adequate protective measures are not taken promptly. As a result of erosion in contaminated areas, TMe can be dispersed in river systems or as aerosols several kilometers from the sites of origin. This diffuse contamination makes the phenomenon even more devastating and difficult to control [27–29].

Mining is one of the fundamental pillars of the Algerian economy, and the metals sector has developed substantially since 1931. Algerian phosphate production had exceeded 1.3 tons in 2001, representing 3.5% of world production [30–34]. In this context, Algeria's mines have made a significant contribution to national phosphate production. Nevertheless, mining activities have led to a profound modification of the region's landscape and its contamination by treatment, by-products and rejected wastes.

The present study mainly focuses on assessing the degree of metallic contamination (Pb, Zn, Cu and Cd) in non-rhizospheric soils (bare soils) in the vicinity of mines in order to

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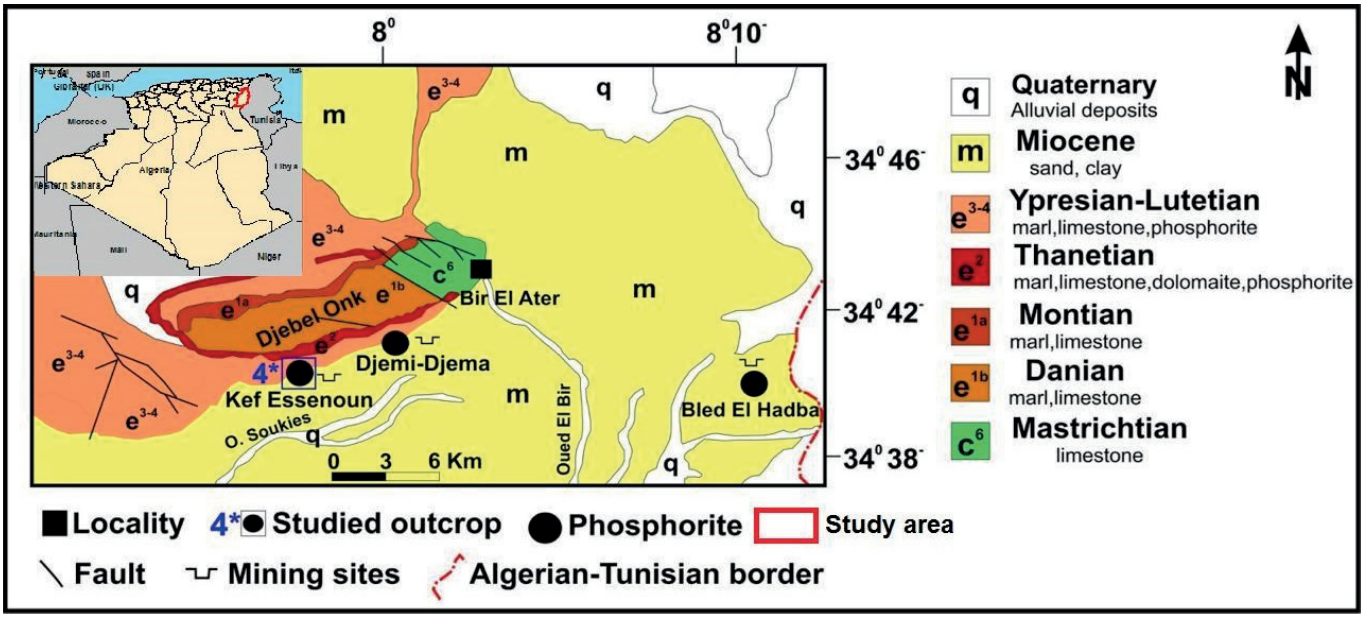

Figure 1. Geological sketch map of Djebel Onk (Tébessa) [40].
Рисунок 1. Геологическая эскиз-карта Джебель Онка (Тебесса) [40].

identify multi-element contamination and its spatial variability in the vicinity of mines, upstream and far downstream. The difference between the concentrations measured in soils that are close to the mining centers and in soils that are far enough away allows the detection of possible contamination and the estimation of the extent of the area affected by the metal contamination.

Material and method

Study area: situation and characterization

Algerian phosphates were discovered at Boughari [35], a little before the Gafsa phosphates, 1885. It is only at the end of 1906 and the beginning of 1907 that L. Jouleau discovered the Djebel Onk deposit in the south east Constantinois and underlined the lithological, stratigraphic and structural analogies with the Gafsa Basin phosphates, he then provided detailed lithological sections of the phosphate deposits of the Djebel Onk zone and published the first chemical analyses of the ore.

The Djebel Onk region is located in the southern part of Algeria, precisely 100 km south of the wilaya of Tébessa and 20 km from the Algerian-Tunisian border. This region constitutes the natural geographical limit between the high plateau of Constantine and the Saharan region. The massif of Djebel Onk forms a 20 km long limestone ensemble which culminates at 1198 m of altitude in Djebel Tarfaya. This massif constitutes the eastern extremity of the Nememcha Mountains which plunges towards the East of the Aurès Massif. The lowest altitudes at the foot of Djebel Onk are about 635 m [31–34, 36–39]. The Djebel Onk region has a series of seven deposits that are undergoing appraisal; three are adjacent to exploitation (Djemi Djema East and West and Kef Es Sennoun) and three are 6 to 35 km away (Djebel Onk North, Bled el Hedba, Oued Betita) (Fig. 1). Mining at the level of the Djebel Onk mining district (active exploitation since 1965) has inevitably led to changes in the natural environment and caused numerous disturbances with negative landscape impacts and risks for the local residents: Deep quarries, galleries, shafts, down pipes and surface installations have been abandoned without any safety measures; slag heaps that harm the aesthetics of the environment and degrade the landscape; and finally, unvegetated tailings disposal sites, weighing several million tones, are abandoned in open areas near agricultural land, on the course and bed of the Soukias valley and on the banks of its tributaries.

Sample collection and analysis

Non-rhizospheric soils (bare soils) and mine tailings were sampled during spring in the Djebel Onk mining district (Fig. 2) on the tailings slopes for the mining centre, in the vicinity of the slopes and at varying distances from the slopes (1, 2, 3, 4, 5, 6, 7, 8, 9).

Sampling was also carried out downstream more than 10 km away from the mining district in Djebel Onk (1 and 2). A reference sample was also taken in station (3 and 4) 30 km upstream of the mining area away from any source of pollution. Soils were sampled in non-agricultural areas using a plastic hand bucket.

The source of pollution in this area is mainly washing waste forming large sand dikes. These sands are easily transported by winds and precipitation in particulate form. Furthermore, some of these dikes have been covered by a (rejections) substrate corresponding to the residual materials directly from the ore extracted during the operation of the mine. Stations 5, 6 and 8 corresponding to the oldest dikes in the Djebel Onk area are located on an uncovered dike.

Attempts have been made to reforest the area around the processing plant, but due to high dust emissions, the forest massif located in the axis of the prevailing winds could not withstand this pollution (stations 7 to 9).

In every station, each soil sample is a composite sample of 5 sub-samples taken at random from an area of 5–5 m (0–15 cm deep). The samples taken were placed in clean plastic bags and stored at low temperature. Profiles of the main soil types encountered in the study area were studied to determine the main soil types in the region.

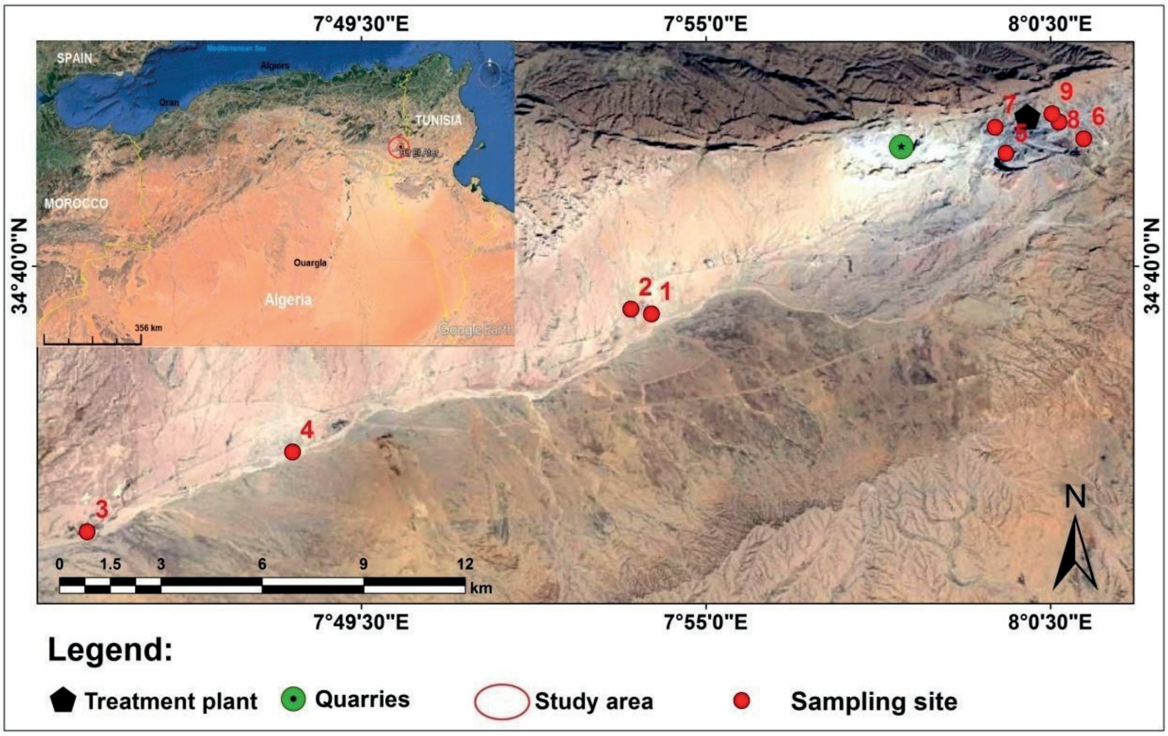

Figure 2. Geographical location of the regions studied and location of the sampling stations.
Рисунок 2. Географическое расположение исследуемых регионов и расположение станций отбора проб.

In the laboratory, particle size analysis was performed for the soil samples collected from the tailings piles, the physico-chemical characteristic (pH) was determined by Eutech instruments pH510 type pH-meter.

All the soil samples taken were dried at a temperature of 80 °C in a clean room in ambient air and then crushed in an electric FRITSCH Pulverisette mill. The mineralization protocol for the preparation for the determination of heavy metals (Pb, Cu, Zn, Cd) in the examined soils is the one recommended by Chiffoleau [41].

The determination of metals in soils was carried out at the Unit on Analyses and Technological Development in Environment, Scientific and Technical Research Centre in Physico-Chemical Analyses CRAPC Bou Ismail, Tipaza (Alegria) on soil mineralization by flame atomic absorption spectrometry (type 240FS AA AGILENT TECHNOLOGIES 200) for lead (Pb), copper (Cu), cadmium (Cd) and zinc (Zn).

The validity of the analytical method was verified by internal control using two certified reference samples: IAEA-433, BCSS-1 (sediments); and by external control using intercalibration exercises (IAEA).

Results and discussion

pH measurements

The results obtained show neutral to weakly acidic pH values ranging from 6.7 to 7.7 (Table). Of all the samples, only two samples showed slightly acidic pH values at the 3 site.

The soils in the vicinity of the Djebel Onk mining centre are raw mineral soils generally covered with a thin film of tailings, especially around the slopes and in the direction of the prevailing winds. Similar results were reported [44] for soils near a lead mine in the USA. Saidi reports pH values between 6, and 7.7 in the mining district of Zaide in eastern Morocco [45]. However, [5] found pH values between 3.1 and 8.4 in the mining areas of Cabezo Rajao in Spain.

Several authors have shown that soil pH plays a major role in the solubility and bioavailability of metals [46–50].

In the present study, many sites had alkaline pH levels, which generally result from the presence of carbonates in the soil. These carbonates can be an important reservoir for soil TMe. According to [46], cadmium, copper, lead and zinc have a high affinity to carbonates. In addition, several authors have shown that pH is a factor controlling the solubility and/or retention of metals in soil [47, 48]. According to [51], several metals are relatively more mobile at acidic pH and oxidizing conditions, whereas they are strongly retained under alkaline and reducing conditions. Thus, in environments where lead concentrations are relatively high, carbonates in the form of hydro-cérusite and cérusite are the major regulators of the distribution of this element in soils and surface waters [52].

Heavy metal content in soils

The results of heavy metals (Pb, Zn, Cu, Cd) analysis in soils gathered from the different stations studied are shown in Table 1. The mining region of Djebel Onk is characterized by grades ranging from 11.92 to 136.64 mg/kg for Zn, 3.60 to 24.60 mg/kg for Cd, 5.40 to 115.20 mg/kg for Cu and 8 to 36 mg/kg for Pb. According to the results, the soils at the Djebel Onk mine site and even those located 30 km downstream from the mine site in the Djebel Onk area (3 and 4) are generally characterized by high heavy metal contents. The grades are particularly high in the soils of the 5, 6, 7, 8 and 9 mine tailings. Comparison between the four sites shows the variability in metal levels between soils at the four sites as well as between soils at the same mine site. In general, metal contents in soils are very high in 5, 6, 7, 8 and 9, high in 1 and 2 and moderately

pH values, levels of metals (mg/kg) and pollution index in soil samples.
Значения pH, содержание металлов (мг/кг) и индекс загрязнения в пробах почвы.

Station	Site	pH	Concentrations in mg/kg				IP
			Zn	Pb	Cd	Cu	
1	Agricultural soil	7.7	44,48	16	8,40	115,2	1,07
2	Agricultural soil under olive trees	7.3	50	24	6,20	99,6	0.87
3	Sediment	6.7	13,06	16	4,40	14	0.45
4	Sediment	7.1	11,92	26	3,60	12,2	0.41
5	Processing residues	7.4	129,68	8	24,60	49,8	2.30
6	Processing residues	7.6	136,64	24	24,00	57,2	2.32
7	Soil under olive tree next to the plant	7.5	97,62	20	19,60	5,4	1.78
8	Processing residues	7.2	122,56	12	22,20	23,4	2.04
9	Vegetated soil (under olive tree next to the plant)	7.7	136,02	36	21,60	69	2.18
Normal levels of uncontaminated soil [42]			90	35	0.35	30	
Limit values soil Alegria (M. B. 12.04.1995)*			200	100	2	50	
Soil limit contents France [43]			300	100	2	100	

*January 12, 1995 – Government Order regulating the use on or in soils of sewage sludge or sludge from septic tank sludge treatment plants (M. B. 12.04.1995)

high in 3 and 4, but variations in contents are recorded from one soil to another at the Djebel Onk mine site, depending on the distance from the slopes and the position in relation to the direction of runoff and prevailing winds. The grades of Pb, Zn and Cd at Djebel Onk decreased (but still higher than normal) from the pour to the residue at station 1, which is more than 10 km from the pour.

The variability in metal levels between soils at the mine sites studied and even between soils at the same mine site appears to be a characteristic of mining pollution in the region. Indeed, according to [53, 54], total metal levels in soils near mining and smelting areas are high and vary greatly among sites.

On the other hand, heavy metal levels in soils at the mining centre and even far away (3 and 4), for most samples and metals (Pb, Zn, Cu and Cd), are much higher than the levels recorded for the station soil (3 and 4) in which the metal levels studied, with the exception of Cd and Cu in 4, are below or close to the global averages for uncontaminated soils, as they far exceed the normal global averages for uncontaminated soils given by Bowen (1979) and exceed the limit values for these metals in soils in Algeria (M. B. 12.04.1995) and France [43] (Table). These are soils that certainly require remediation work.

The maximum levels of heavy metals in the soils studied were recorded in the surface soils of the slopes (7 and 9). These levels are very high compared to the levels recorded for the soil (4) and exceed them up to ≈12 times, ≈ 2 times, ≈ 7 times, ≈ 6 fois, respectively for Zn, Pb, Cd and Cu. These results indicate that these tailings, abandoned without rehabilitation, are important sources of trace metal elements that can have long-lasting contaminant effects in the immediate vicinity of mine sites and even further away in the directions of runoff and prevailing winds.

In fact, tailings from the slopes in the study site, consisting of fine, homogeneous particles of homogeneous size, containing a very low percentage of organic matter, non-vegetated and of low physical stability, are subject to wide dispersion in the environment as a result of water erosion in rainy periods and wind erosion in dry periods. The Djebel Onk is swept by very violent winds that reinforce the drought in the region and cause dust clouds in summer that can last for several days. The flight of fine particles from the mine tailings in significantly suspended solids and toxic flying dust rich in TMe reach the surrounding and even distant soils, which are then contaminated.

During rainy periods, the frequent thunderstorms in the Djebel Onk region result in groundwater runoff and flooding. The slopes leach and release dissolved and especially particulate metals in suspension. The transport of the residues is inevitable due to the impermeable marno-dolomitic substrate that the slopes are built on. Sandy discharges rich in heavy metals reach nearby and distant watercourses and soils.

As a result, due to the spread and entrainment of contaminants by winds and water far away from the mine site, the area affected by metal pollution from the Djebel Onk mining area is very large. The negative impacts of the tailings disposal area at the Djebel Onk mine are not limited to the localized impacts on the landscape, but also include the long-lasting impacts related to the transport of these heavy metal-rich tailings. It is therefore necessary to limit the negative environmental impact of the tailings piles without stabilization in the mining area. Remediation measures must be taken to immobilize the residual metal pollution in these discharges and limit the dispersion of contaminants following wind and water transport responsible for the extension of the contaminated zone beyond the metal deposit mining sites.

On the other hand, metallic contamination of the soils in the mining region studied is critical and can affect the quantities of heavy metals drawn from these soils by plants and induce toxicity. Indeed, given the fact that the critical soil metal content is defined as the value above which toxicity is possible and that the values above which soil is critical are 8 mg/kg Cd, 125 mg/kg for Cu and 400 mg/kg for Pb and Zn, there is at least one metal above the critical level in all soils sampled and at least one metal (Cd) above the critical level in the tailings soils. On the other hand, according to [4], excessive concentrations of TMe in soils influence the uptake of these elements by plants; as they can be toxic to plants as well as animals, they can affect agricultural production as well as the safety quality of food for human consumption.

Soil Metal Pollution Index

Heavy metal contamination of soil surfaces, particularly at mine sites, is associated with a mixture of contaminants rather than a single metal [4]. Thus, the concept of a soil pollution index (PI) has been introduced in many studies to identify multi-element contamination resulting in increased metal toxicity [3, 4, 49, 55–57]. This is a criterion for assessing the overall toxicity of contaminated soil. According to [49], the soil pollution index is calculated from the average of the ratios of metal concentrations in soil samples to the limit guideline values. These limit values correspond to the assumed tolerable levels in soil suggested by Kloke [58]. Thus, the pollution index is calculated by the following equation:

$$\frac{\left(\frac{Cd}{3}+\frac{Cu}{100}+\frac{Pb}{100}+\frac{Zn}{300}\right)}{4}$$

and an IP greater than 1 corresponds to polluted soil.

Within the framework of the present study, the pollution index was calculated for the different soils sampled at the Djebel Onk site; the results are reported in Table 1.

The results obtained reveal PI values that vary between 0.41 for the station (4) and 2.32 (soils from treatment residues). With the exception of the station soils (2, 3 and 4), all soil samples collected at the Djebel Onk mine site and even the furthest soil (1) collected 10 km away have a PI greater than 1, confirming polymetallic soil contamination throughout the downstream area of the mine sites.

The extremely high PIs recorded for the tailings soils (5, 6 and 8) underline the extremely harmful nature of these substrates abandoned without rehabilitation, which are a perennial source of heavy metal contamination for the environment and agriculture in the region. The tailings dams of Djebel Onk have the highest polymetallic contamination in the study area (5 = 2.30, 6 = 2.32, 8 = 2.04). They are followed by Vegetated Soil (under olive tree next to the plant) with a PI equal to 2.18, then 7 and 1 with a PI equal to 1.78 and 1.07, respectively.

Conclusion

The evaluation of the degree of heavy metal (Pb, Zn, Cu, Cd) contamination of soils in the vicinity of the Djebel Onk mining centre in Algeria generally shows that the metal contents of soils in the vicinity of the mining districts and even far away are high compared to the contents of the soil far away or to the contents considered normal in the literature for uncontaminated soils.

Maximum metal levels are recorded in soils from tailings impoundments that show polymetallic contamination, indicating that tailings impoundments are an important perennial source of heavy metals on the surface of mine sites. These polymetallic contaminants are thus disseminated by water and wind pathways from the tailings and reach nearby and even distant soils via wind and water erosion and deposition mechanisms.

The results of the study converge towards a finding of greater or lesser contamination by heavy metals in an area that extends beyond the mining area itself. Remediation measures must be taken in order to immobilize the residual metallic pollutants from the mining discharges and limit their transport, which is responsible for the extension of the contaminated zone beyond the mining sites.

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Оценка загрязнения металлическими микроэлементами в горнодобывающем районе по переработке фосфатной руды Джебель Онк (Алжир)

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Аннотация
Актуальность исследования. Добыча фосфатов является одним из основных источников загрязнения тяжелыми металлами, поскольку они наносят вред почве и приводят к деградации экосферы.
Цель исследования. Это исследование направлено на характеристику и оценку металлического загрязнения почв в окрестностях шахты Джебель Онк в городе Бир-эль-Атер (Вилайя Тебесса) на востоке Алжира, и, в частности, на оценку пространственной изменчивости этого загрязнения и степени загрязнения.
Методология. Физико-химическую характеристику (pH) определяли с помощью pH-метра, а для определения тяжелых металлов (Pb, Cu, Zn, Cd) в исследуемой почве использовали AAS.
Результаты исследований. Результаты показывают, что почвы, расположенные близко к исследованному руднику, и даже те, что находятся далеко, сильно загрязнены тяжелыми металлами - свинцом (Pb), цинком (Zn), медью (Cu) и кадмием (Cd); результаты также указывают на высокую изменчивость концентраций не только между участками отбора проб, но и в пределах одного рудника. Анализируемый индекс загрязнения почв, как правило, высок даже для почв, отобранных ниже по течению более чем в 30 км от рудника, тогда как на поверхности хвостохранилищ он чрезвычайно высок, что подчеркивает тот факт, что хвостохранилища в их нынешнем состоянии считаются многолетними источниками загрязнения тяжелыми металлами.
Вывод. Нездоровая зона, пораженная загрязнением металлами горнодобывающих предприятий в Джебель-Онк, очень велика в силу процессов рассеивания ветром и водным транспортом остаточных загрязнителей из отходов шахт, оставленных на участке. Необходимо принять восстановительные меры для иммобилизации загрязняющих веществ и ограничения их распространения в окружающей среде.

Ключевые слова: Загрязнение почвы, Алжир, тяжелые металлы, фосфатный рудник, индекс загрязнения.

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