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## ANALYSIS OF MERCURY CONTENT INSIDE MINING WASTE DUMP – CASE STUDY IN THE UPPER SILESIA IN POLAND

Mercury is ranked third on the Substance Priority List, an index of substances determined to pose the most significant potential threat to human health compiled by the Agency for Toxic Substances and Disease Registry. This element is activated with the extraction of hard coal and accumulated in the natural environment or re-emitted from the waste deposited on dumping grounds. So far, studies on mercury content have focused on the analysis of the dumps surface and the adjacent areas. In this paper, the detection of mercury content inside mining waste dumping grounds was analysed. The recognition of mercury content in the profile of the mining waste dump is important in terms of the dismantling of the facility. The dismantling may pose a risk of environmental pollution with mercury due to the possibility of increased fire risk, re-emission, and the transfer of xenobiotics to another place. In this paper, the study of mercury content in the mining waste dump profile was presented. The research demonstrated that there is no significant relationship between the mercury content and the sampling depth. The mercury content in the mining waste was determined based on the rank and origin of hard coal only. Therefore, intensive efforts should be undertaken to identify the environmental hazards arising from the dismantling of mining waste dumps and to adopt measures to prevent these hazards.

**Keywords:** mine waste; mercury; waste dumping ground; hazard pollution; waste dump dismantling

## 1. Introduction

Nowadays, heavy metals, including mercury, pose a particular threat to the environment [1,2]. The problem of environmental pollution with mercury arouses interest due to the variability of the forms in which the element occurs, its high biological and chemical activity and its

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impacts on the biosphere. Once introduced to the environment, mercury remains there forever, changing only the place where it has been disposed first [3]. In the literature, there is a lack of research on mercury content inside dumping grounds where waste from hard coal mining industry is deposited despite the fact that such studies are important for dismantling or reclamation works [4-6]. The research conducted so far has focused on the analysis of the surface of the dumping grounds and the areas adjacent to them [7-9]. The purpose of the research was to identify mercury decomposition inside the dump of mining waste after it has been deposited for many years. According to the United Nations Environment Programme, coal combustion is the second most common source of mercury emissions in the world (average emission of 474 Mg/year), after handicraft- and small-scale gold production (average emission of 727 Mg/year). The share of coal burning in global mercury emissions is about 24%, while the handicraft- and small-scale gold production is about 37% [10]. In Poland, among the various types of anthropogenic activities that affect the pollution of the environment with mercury compounds, it is coal extraction, processing and energy utilization that have the major environmental impact [11-14]. The most significant source of mercury re-emission from mining waste is its ignition resulting in the emission of gases which are the products of oxidation and gasification of coal and cause a nuisance for humans and the environment [15,16]. Studies on the emission of mercury vapours from dumps that caught fire were investigated by Michalska et al. [17]. It was found that the disorganized emission of mercury vapour from mining waste dumps displaying thermal activity was a typical phenomenon, amounting to 8.64 g of mercury emission per year from the surface of the studied testing grounds (12 m$^2$). In Poland, a large difference in the mercury content in bituminous coal can be observed. The mercury content in Polish coals varied from 1 to 758 ppb [18,19]. By way of comparison, in the USA it varied from 27 ppb to 682 ppb, in Russia from 70 ppb to 120 ppb, in Australia from 50 ppb to 100 ppb, whereas in China from 60 ppb to 80 ppb and even to 154 ppb [20,21]. The varied content of mercury in bituminous coal is associated with the location of accompanying rocks, the types of circulating water and temperature as well as the age and rank of coal. Mercury in bituminous coal is mainly connected with mineral substances (65-70%). The remaining part is related to the organic compounds or occurs in an unbound form [22,23].

The concentrations of Hg in water from mine waste leaching varied widely from <0.001 to 760 µg of Hg in leachate/g of the sample [24]. The content of mercury in mining waste coming from hard coal preparation processes (i.e. heavy liquids, jigs and flotation) ranges from 55 ppb to 401 ppb. It was confirmed that the mercury content in the waste from coal extraction and processing is significantly higher than the mercury content in raw coal. Mercury content in the tested waste occurs in the range from 70 ppb to 270 ppb with a maximum value of 380 ppb [7,25,26]. The hazards of soil and water contamination caused by mining waste disposal from Polish mines were studied. The testing ground included two mining waste dumping grounds from which 40 environmental samples were collected. Based on the conducted research, mercury concentration in the soil around the dumps was found to be at the level of 368-447 ppb, whereas in the sludge from ditches surrounding the dump it ranged from 0.460 to 2.14 ppm. In addition, mercury content in the soil around the mining waste dumping ground of one of Polish hard coal mines located in the Upper Silesian Region was examined. The mercury concentrations in the soil around the mine waste dumps were in the range of 9-155 ppb (the average 58.5 ppb) [17]. The analysis of the content of mercury in the surface layer of the soil in the vicinity of the selected mining waste dumping grounds conducted by Klojzy-Karczmarczyk and Mazurek [28] demonstrated

that the mercury content varied from 20 ppb to even over 180 ppb (the average 78 ppb). Moreover, Klojzy-Karczmarczyk and Mazurek [29] studied also the degree of mercury contamination of the subsurface soil layer in the vicinity of a conventional coal-fired power plant. Based on the conducted studies, it was found that the mercury content of the tested soils varied widely from about 10 ppb to over 100 ppb (75 ppb on average). The highest content of mercury was detected more than 2 km away from the power plant emitter. The determined mercury content did not exceed the permissible levels of mercury for the soil (0.5 mg/kg) as defined in the Ordinance of the Minister of the Environment of September 9, 2002 on Soil Quality Standards and Land Quality Standards (Journal of Laws No. 165, item 1359) [29]. The development of mine waste dumping facilities constitutes a side effect inextricably linked to mining operations and poses a serious threat to the environment; therefore, the recognition of its negative environmental impact is an important issue. This study focused on selected anthropogenic terrain with a uniform material from long-stored mining waste deposits.

## 2.  Materials and Methods

The research subject is a mining waste dump located in Upper Silesia, Poland encompassing a total surface of 71 ha. The research area is located in south-western Poland (see Fig. 1)
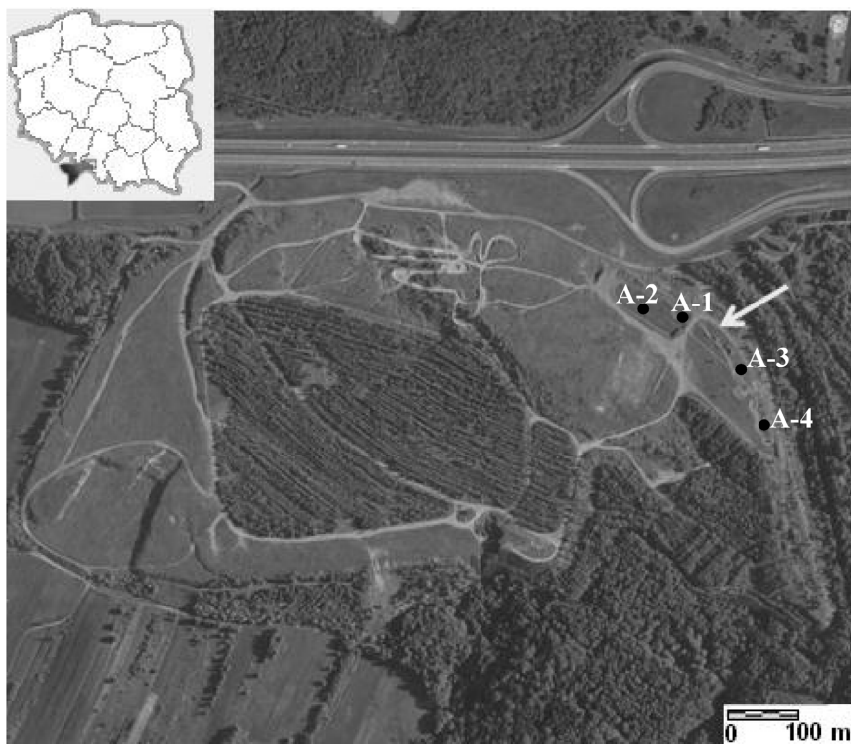


Fig. 1. Studied mine waste dump

The extractive mine waste has accumulated at the mine waste dump since 1979. From December 1999 on, the highest part of the dump has been the northern part, with the third level of the dump formed with an elevation from 286m asl (above sea level) up to 296m asl and the surface of about 6.5 ha. The basic part of the dumping ground is built at the level of 274 m asl covering an area of 30.79 hectares. The water management of above-surface structures is exclusively formed by precipitation water and classified as the so-called precipitation-retention economy. Since May 1999, the storage of mining waste from hard coal mining industry has been suspended at the dumping facility. In the fourth quarter of 2000, reclamation works were started.

Currently, reclamation works are not continued at the site. The dump is an alternative source of raw materials used inter alia in road engineering, engineering works such as the construction of flood embankments, canal embankments, river embankments and water reservoirs, etc., or in the construction sector as mortar, concrete, building ceramics and filling or direct building material; that is why the dismantling of the facility is a prospective operation of the commune of Godów in order to remove the threat to the environment. The commune as the owner may transfer the land title (e.g. the tenancy agreement or the lease agreement) to the entrepreneur, who, after addressing the Marshal of the Silesian Voivodeship, obtains a permit for the extraction of waste [30] and their processing [31].

Planning of sampling was carried out in accordance with the guidelines contained in the standards PN-ISO 10381-1: 2008 and PN-ISO 10381-5: 2009. The samples used in the study were taken from 4 drillings into the waste dump. The test holes were made between 05.07.2016 and 07.07.2016. A "dry" rotary drilling system was used with a H20SG drilling rig including a 100 mm diameter screw auger and a fork-type picker produced by WAMET Sp. z o.o. (Fig. 2) The openings were drilled in the north-western part of the dump (see Fig. 1). The depth of the test openings ranged from 14 to 20 m, which enabled sampling to the floor of the dump. The samples were taken from individual drillings from two-meter depth intervals 0-2, 2-4, 4-6 m, etc. In total, 32 samples were selected for testing (A 1/1 ÷ A 1/10; A 2/1 ÷ A 2/6; A 3/1 ÷ A 3/10 and A 4/1 ÷ A 4/6).



Fig. 2. a) H20SG car drilling rig by WAMET Sp. z o.o., b) Field works

Openings A1 and A3 were drilled to the depth of 20 m. The embankment of the mining waste was drilled to the depth of 19.5 m. From the depth of 19.5 m to the bottom of the well, sandy clay was drilled in a hardening state. Borehole A2 was drilled to the depth of 14 m. To the depth of 13 m the embankment of the mining waste was drilled. To the depth of 13 m to the bottom of the borehole, sandy clay was drilled in a hardening state. The A4 opening was drilled to the depth of 14 m. To the depth of 12.6 m the embankment of the mining waste was drilled. From the depth of 12.6 m to the bottom of the well, sandy clay was drilled in a hardening state.

The total mercury content of the samples collected was determined by means of a fully automated MA-2000 Nippon Instrument Corporation system (Fig. 3) for measuring the amount of mercury in solids, gases and liquids in the 0-1000 ng measurement range and the detection limit of 0.002 ng. Dry air (0.5 $dm^3$/min) was used as the carrier gas in the analyser. The samples were introduced into the combustion pipe where they were subjected to thermal decomposition at the temperature of 900°C. The mercury released during combustion was absorbed on a gold-plated deposit. The resulting amalgam was then heated to release the concentrated mercury that reaches the measuring system. The samples were also tested for the determination of selected parameters such as ash content, silicon oxide, aluminium oxide or trace elements. The tests were carried out by the method of fluorescence X-ray dispersion spectrometry. The spectrometer of the ZSX Primus II series by Rigaku was used for this purpose, equipped with a system with an anode X-ray tube. In case of the ash content the determination was performed using the weighing method according to the internal test procedure. During the experimental works, 32 samples from 4 openings were analysed.



Fig. 3. Ma-2000 mercury analyser

## 3.  Results and discussion

Total mercury content, determined in all tested samples varied from 31 ppb (opening A2, depth range 0-2 m) to 123 ppb (opening A4, depth range 10-12 m), while its average value was 56 ppb (see Fig. 2).
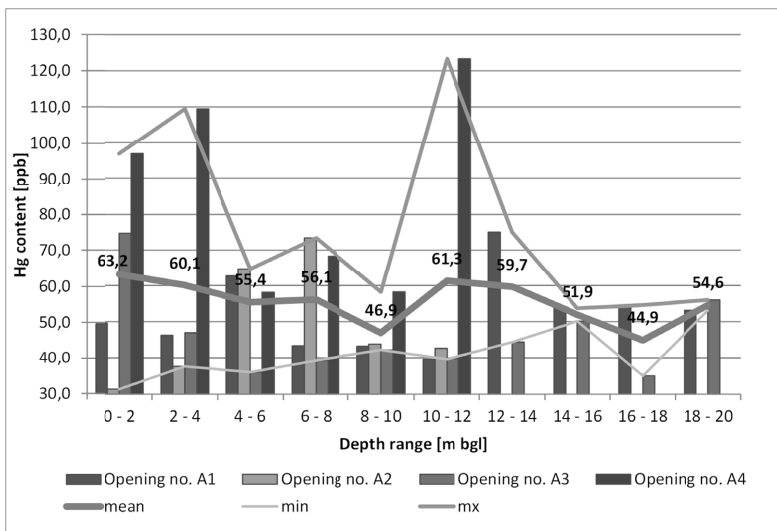
Fig. 4. Content of mercury in individual openings

In samples collected from the shallowest depth, i.e. at the depth of 0-2 m (Fig. 4) the total mercury content was from 31 ppb (opening A2) to 97 ppb (opening A4), reaching an average of 63 ppb. In samples taken from the depth range of 10 m up to 12 m (Fig. 3), the mercury content ranged from 39 ppb (opening A1) to 123 ppb (opening A4), reaching an average of 61 ppb. In the samples taken from the deepest layers, i.e. 18-20 m, the mercury content was measured at 53 ppb (opening A1) to 56 ppb (opening A3), reaching an average of 54 ppb (see Fig. 5).
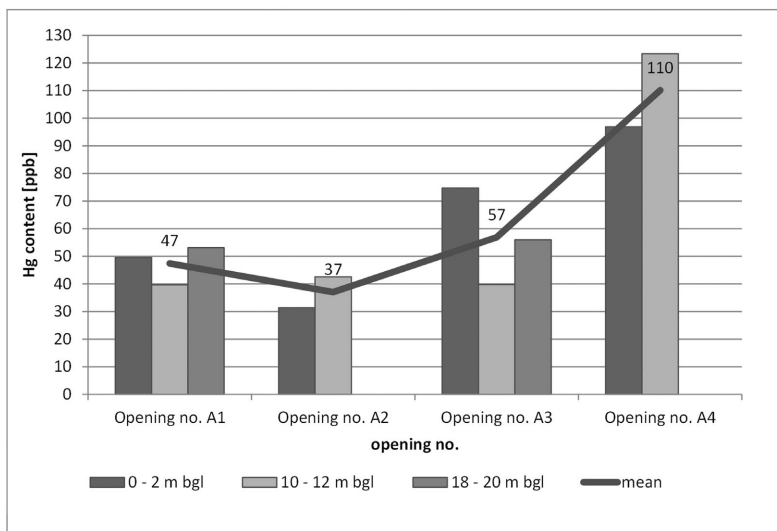


Fig. 5. Mercury content in selected depth ranges

The statistical analyses were performed with the Statistica 13 PL analytical tool. The population structure was analysed by the descriptive measures shown in Table 1.

Basic statistics of mercury content in samples

| Parameter | Value |
|---|---|
| Number of samples | 32 |
| Minimum value | 31 |
| Maximum value | 123 |
| Mean | 56 |
| Std. error | 4 |
| Variance | 458 |
| Standard Deviation | 21 |
| Median | 50 |
| 25 prcntil | 42 |
| Kurtosis | 3 |
| Skewness | 2 |

With regard to mercury content in samples coming from all openings, there is a noticeable difference between the arithmetic average and the median. There is also a large gap between the results obtained (around 92 µg / kg dry weight).

Nonparametric analyses were used due to the rejection of the hypothesis on normal distribution of mercury (see Fig. 6). The Kruskal-Wallis test were used. A statistical significance criterion was established as p = 0.05.
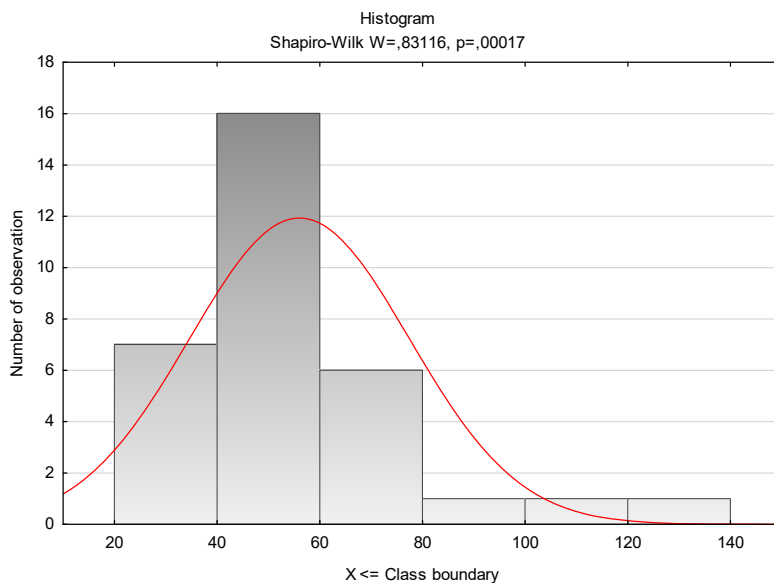


Fig. 6. Histogram of total mercury concentration in samples

The results for mercury showed that there are no significant differences in the average amount of this element depending on the sampling depth. The results are shown in Table 2 where it can be observed that p value is 0.9737, which confirms the fact that groups are not significantly different.

TABLE 2

Kruskal-Wallis test for the sampling depth

| Depend: Mercury (ppb) | Kruskal-Wallis ANOVA by Ranks; Mercury (ppb) Independent (grouping): sampling depth Kruskal-Wallis test: H (9, N = 32) = 2.741477 p = ,9737 | | | |
|---|---|---|---|---|
| | Code | Valid N | Sum of Ranks | Mean Rank |
| from 0 to 2 m | 1 | 4 | 75.00 | 18.75 |
| from 2 to 4 m | 2 | 4 | 64.00 | 16.00 |
| from 4 to 6 m | 3 | 4 | 74.00 | 18.50 |
| from 6 to 8 m | 4 | 4 | 69.00 | 17.25 |
| from 8 to 10 m | 5 | 4 | 53.00 | 13.25 |
| from 10 to 12 m | 6 | 4 | 54.00 | 13.50 |
| from 12 to 14 m | 7 | 2 | 42.00 | 21.00 |
| from 14 to 16 m | 8 | 2 | 36.00 | 18.00 |
| from 16 to 18 m | 9 | 2 | 22.00 | 11.00 |
| from 18 to 20 m | 10 | 2 | 39.00 | 19.50 |

We analyzed also whether there is a significant difference in the average amount of mercury depending on the location where a given sample was taken. Again, we had two hypotheses: the null hypothesis stated that there is no significant difference in the average quantities of this element depending on the sampling location, whereas the alternative hypothesis stated the opposite. The results for mercury (Table 3), under the same conditions as in previous analyses, showed that there is a significant difference in the average amount of this element depending on the location in which the samples were taken (p value is 0.0105).

TABLE 3

Kruskal-Wallis test for the sampling location

| Depend: Mercury (ppb) | Kruskal-Wallis ANOVA by Ranks; Mercury (ppb) Independent (grouping): sampling location Kruskal-Wallis test: H (3, N = 32) = 11.24621 p = ,0105 | | | |
|---|---|---|---|---|
| | Code | Valid N | Sum of Ranks | Mean Rank |
| Opening A1 | 1 | 10 | 167.0 | 16.7 |
| Opening A2 | 2 | 6 | 78.0 | 13.0 |
| Opening A3 | 3 | 10 | 119.0 | 11.9 |
| Opening A4 | 4 | 6 | 164.0 | 27.3 |

Taking into consideration that the Kruskal-Wallis test gives statistically significant results, a post-hoc test was conducted. In Figure 7, it can be seen that the average mercury content in opening A4 differs significantly from the average content in opening A2 and opening A3. This may indicate the heterogeneity of the waste deposited on the dump.
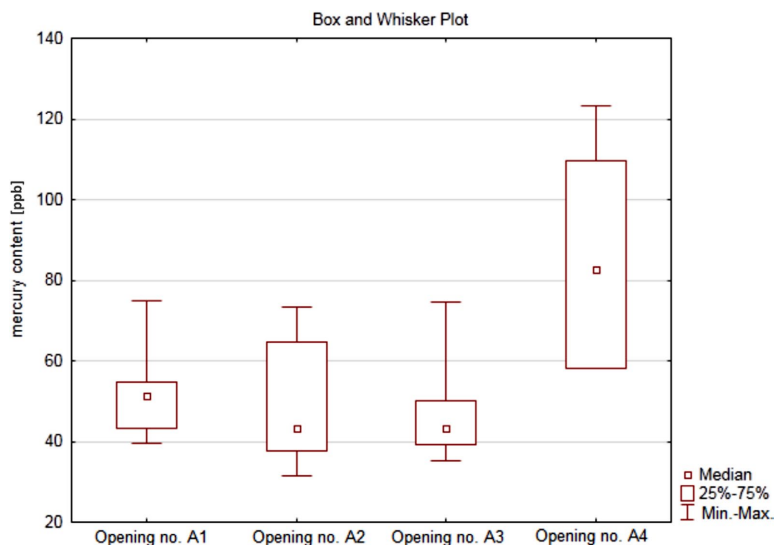
Fig. 7. Box and Whisker Plot for mercury

The results of the laboratory analyses show that the mining waste deposited in the dump is characterized by a high ash content, ranging from approximately 67.3% to 89.7% (81.2% on average) (Table 4). The dominant component of the waste is silicon oxide (SiO2), the content of which ranges from 35.5% by weight. % up to 51.7 wt.% and aluminium oxide ($Al_2O_3$), the content of which ranges from 14.6 wt.%. % up to 22.8 wt.% The analysed waste is characterized by the values of the Al2O3 / $SiO_2$ ratio in a narrow range from 0.347 to 0.492, with an average of 0.41, which indicates that in mineralogical terms they are similar to the mixture of quartz + kaolinite + illite in the ratio 1: 1: 1. Another component showing significant contents is iron oxide ($Fe_2O_3$) ranging from 5.5 wt.%. % up to 7.5 wt.% The tested waste is also characterized by a fairly high content of potassium oxide ($K_2O$), ranging from 2.1% by weight. % up to 3.2 wt.%, which proves the presence of high illite content among clay minerals (Table 5).

TABLE 4

Ash content in the studied mining waste samples

| Sample no. | Ash content [wt.%] | Sample no. | Ash content [wt.%] | Sample no. | Ash content [wt.%] | Sample no. | Ash content [wt.%] |
|---|---|---|---|---|---|---|---|
| A-1/1 | 76,5 | A-3/1 | 83,5 | A-1/9 | 83,6 | A-3/9 | 80,6 |
| A-1/2 | 81,6 | A-3/2 | 81,7 | A-1/10 | 83,4 | A-3/10 | 76,7 |
| A-1/3 | 76,8 | A-3/3 | 83 | A-2/1 | 89,7 | A-4/1 | 78,4 |
| A-1/4 | 80 | A-3/4 | 83,2 | A-2/2 | 88,3 | A-4/2 | 80 |
| A-1/5 | 80,6 | A-3/5 | 82 | A-2/3 | 84 | A-4/3 | 79 |
| A-1/6 | 83,2 | A-3/6 | 82,2 | A-2/4 | 84,7 | A-4/4 | 71,8 |
| A-1/7 | 82,2 | A-3/7 | 83,2 | A-2/5 | 83,5 | A-4/5 | 67,3 |
| A-1/8 | 82,9 | A 3/8 | 83,6 | A-2/6 | 85 | A-4/6 | 75,8 |

Trace elements complement waste research. The highest levels, above 100 ppm, were found for barium (Ba), molybdenum (Mn), rubidium (Rb), strontium (Sr), vanadium (V) and zinc (Zn). A lower share, usually several dozen ppm, was shown by: chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), while the share of a few ppm was shown by: arsenic (As), cadmium (Cd), cobalt (Co), molybdenum (Mo), antimony (Sb), tin (Sn), thallium (Tl) and tungsten (W).

TABLE 5

$Al_2O_3/SiO_2$, $Fe_2O_3$, $K_2O$ content in the studied mine waste dump samples

| Sample No. | $Al_2O_3/SiO_2$ | $Fe_2O_3$ | $K_2O$ | Sample No. | $Al_2O_3/SiO_2$ | $Fe_2O_3$ | $K_2O$ |
|---|---|---|---|---|---|---|---|
| | [wt.%] | | | | [wt.%] | | |
| A-1/1 | 0,416 | 7,02 | 2,41 | A-3/1 | 0,411 | 6,79 | 3,21 |
| A-1/2 | 0,415 | 6,49 | 2,65 | A-3/2 | 0,423 | 6,18 | 3,07 |
| A-1/3 | 0,41 | 6,78 | 2,35 | A-3/3 | 0,421 | 6,69 | 3 |
| A-1/4 | 0,412 | 6,32 | 2,6 | A-3/4 | 0,425 | 6,1 | 3,07 |
| A-1/5 | 0,417 | 6,32 | 2,71 | A-3/5 | 0,412 | 6,19 | 3,02 |
| A-1/6 | 0,409 | 6,8 | 2,79 | A-3/6 | 0,412 | 6,92 | 3,05 |
| A-1/7 | 0,418 | 7,5 | 2,63 | A-3/7 | 0,409 | 6,57 | 3,1 |
| A-1/8 | 0,417 | 6,56 | 2,69 | A 3/8 | 0,415 | 5,93 | 3,21 |
| A-1/9 | 0,416 | 6,74 | 2,74 | A-3/9 | 0,403 | 5,96 | 2,94 |
| A-1/10 | 0,413 | 6,7 | 2,73 | A-3/10 | 0,41 | 5,67 | 2,88 |
| A-2/1 | 0,492 | 6,26 | 2,17 | A-4/1 | 0,403 | 6,65 | 2,56 |
| A-2/2 | 0,347 | 5,55 | 2,82 | A-4/2 | 0,402 | 6,49 | 2,58 |
| A-2/3 | 0,383 | 6,5 | 2,81 | A-4/3 | 0,403 | 6,72 | 2,55 |
| A-2/4 | 0,386 | 6,42 | 2,79 | A-4/4 | 0,409 | 5,94 | 2,32 |
| A-2/5 | 0,402 | 6,72 | 2,73 | A-4/5 | 0,411 | 5,7 | 2,14 |
| A-2/6 | 0,357 | 6,21 | 2,68 | A-4/6 | 0,402 | 6,59 | 2,49 |

# 4. Conclusions

The study of mercury content in the mining waste dump profile indicates that there is no significant relationship between the mercury content and the sampling depth. This may indicate a lack of uniform migration into the dump profile and the disposal of mining waste from various mines and deposits.

We have proved that the sampling site affects the content of mercury in the mining waste dump. This may indicate the heterogeneity of the waste deposited on the dump.

During the potential dismantling of the dump, the mercury contained in the deposits of mine waste will be subjected to deposition in the processes of washing out, deflation and mechanical transport to the destinations.

The dismantling of mining waste dump may pose a risk of mercury pollution of the environment due to the possibility of an increase in fire risk, re-emission, and the transfer of xenobiotics to another place. Depending on the recovery method, mercury along with other xenobiotics can be immobilized in the concrete mass, and mining waste can be used for example in the cement or engineering industries. Bearing in mind the above, intensive efforts should be made to iden-

tify the environmental hazards arising from the dismantling of mining waste dumps as well as to effectively prevent the occurrence of these hazards. The results of the research indicate the necessity to undertake the determination of mercury content in dumps, which will have an impact on the determination of the level of environmental risk and the proper preparation of the land reclamation process.

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