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Research Article

MERCURY AND ALUMINIUM IN ARABLE SOILS IN THE AREA OF INFLUENCE OF BROWN COAL MINE AND POWER PLANT BASED ON THE EXAMPLE OF THE BOGATYNIA REGION (SOUTHWESTERN POLAND)

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ABSTRACT
This study presents research on the content of mercury and aluminium in arable soils within the range of influence of the Coal Mine and Power Plant Turów (Bogatynia region). The results were compared for reference with the region of Zgorzelec, which is situated outside the range of emission and with the results of tests conducted by other authors. The tests did not show a statistically significant increase in the mercury level in arable land from the range of influence of the Coal Mine and Power Plant Turów, neither in comparison to soils from Zgorzelec region. It is noticeable that the maximum level of mercury in the analysed area was nearly twice as high as in the values obtained in soils from industrial area in Opole and 25 times lower than the maximum mercury level found in the soils of Jelenia Góra District. On the other hand, tests of aluminium content showed a significant, nearly 3 times higher level of this metal in the range of direct influence of the Coal Mine and Power Plant Turów in comparison with the Zgorzelec region. The noted values were higher than in soils from industrial area in Opole.

Keywords: soil, mercury, aluminium, contaminated soils, industrial contamination.

INTRODUCTION

Diffuse pollution is one of the main threats for soil. This type of contamination is characteristic for strongly industrialized areas. One of the major sources of this type of contamination is opencast and underground coal mining, the connected usage of the mined material in energy production and the storage of industrial waste. Regions where large quantities of coal are mined and processed are located throughout the world, mainly in China, the USA, Australia, Indonesia and South Africa. In Europe the problem exists, among others, in Germany and Poland (Tables 1 and 2). These results in the fact that mining and energy production areas are more exposed to soil contamination, in comparison to other regions. In spite of the promotion of renewable energy sources, the interest in the usage of coal as the main energetic resource is not decreasing [6]. It is believed that coal will continue to play the main role in the global structure of energy sources as the power consumption continues to increase. Moreover, in spite of the application of new technologies limiting the emission of harmful substances, the problem of contamination resulting from the mining and processing of coal and its impact on the environment will continue to worsen.

The problem presented in this study is significant also due to the obligations resulting from

Table 1. Coal in electricity generation in 2012 according to the statistics by World Coal Association [30]

Country	Coal in electricity generation (%)
Mongolia	98
South Africa	94
Poland	86
PR China	81
Australia	69
India	68
Israel	59
Indonesia	44
USA	43
Germany	43
UK	29
Japan	27

Table. 2. Top 10 coal producers in 2012 according to the statistics by World Coal Association [30]

Country	Coal production (Mt)
PR China	3549
USA	935
India	595
Indonesia	443
Australia	421
Russia	359
South Africa	259
Germany	197
Poland	144
Kazachstan	126

Poland's membership in the EU and the implementation of the IPCC (Integrated Pollution Prevention and Control) Directive, including the creation of *European Pollutant Release and Transfer Register (E-PRTR) and EMEP* (European Monitoring and Evaluation Programme). Mercury is considered as a priority harmful substance, which was included in the Water Framework Directive.

One of the areas prone to contamination connected with the mining and processing of brown coal in energy production in Poland is the Bogatynia region. Sources of gaseous and dust pollutants are two main facilities on the Polish side, i.e. Coal Mine and Power Plant Turów, but also some plants located on the territory of the Czech Republic, and, until recently, on the German side of the border. It is estimated that cross-border pollutants inflowing to Poland account for as much as 60% of the contamination. Integrated environmental activities that were started in mid-1990s led to a decrease in the emission of air pollutants from power plants located in this region by approximately 60-70%, which has undoubtedly contributed to the improvement of the condition of the natural environment [22].

The aim of the conducted study was to determine the total content of mercury and aluminium in samples of arable soils collected within the influence range of the Coal Mine and Power Plant Turów, to compare the obtained results with Zgorzelec region, located outside the range of emitters, with results obtained by other authors for this region, the geochemical background of Poland and with results obtained for areas that were subject to similar types of anthropogenic pressure in other countries. The research hypothesis assumed that there is a connection between the emission of pollutants from the Coal Mine and Power Plant Turów and the content of mercury and aluminium in the Bogatynia region and that the obtained results should be significantly higher than the values obtained in the Zgorzelec region and the geochemical background of Poland.

MATERIAL AND METHODS

The study was conducted in the years 2008-2011 as part of the evaluation of content of metals in abiotic and biotic elements of the environment in Southwestern Poland. Soil samples originated from two regions:

- Bogatynia (localities: Bogatynia, Działoszyn, Bratków, Ręczyn, Wyszków, Wolanów), encompassing the area of influence of the Coal Mine and Power Plant Turów.
- Zgorzelec (localities: Jerzmanki, Łagów, Jagodzin, Gronów, Sławnikowice), situated outside the range of direct influence of emitters.

A total of 64 soil samples, collected at the depth of 0.0 to 0.3 m b.g.s. (below ground surface) and from 0.3 to 0.6 m b.g.s. were analysed (Figure 1).

The AAS method with amalgamation of cold mercury vapour with the use of the mercury analyser MA 2000 was used to determine the content of total mercury was determined directly in air dry mass of soil. In order to determine the content of total aluminium in soil 0.5 g of weighed quantity was subject to wet mineralisation in aqua regia with use of hypotension microwave system. In the obtained mineralisate, the content of total aluminium was determined with the ICP-AES method, with use of plasma spectrometer Varian-

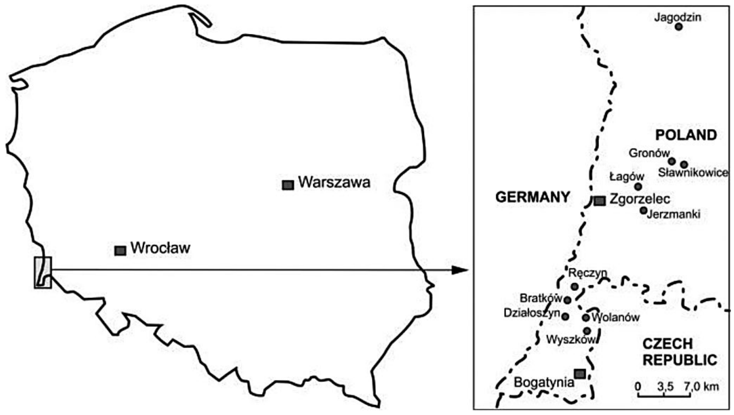

Figure 1. Location of sampling areas

Liberty 220. The obtained results were subject to statistical analysis, including the calculation of arithmetical mean and standard deviation. The t-student test was used for the purposes of statistical analysis. The value $p \leq 0.05$ was assumed as statistically significant difference. The analyses took into account the physical and chemical properties of samples: the content of organic matter and grain size distribution, which are the key factors influencing the content of heavy metals in soils.

RESULTS AND DISCUSSION

Basic characteristic of soil samples

Soils from both regions are characterised by an acidic pH (Table 4). The pH values measured in 1M KCl for all analysed samples fell within the pH range from 4.70 to pH 6.22. while measured in H_2O fell into the range from pH 3.42 to pH 6.42. In both analysed regions the content of organic matter was higher at the depth from 0.0 to 0.3 m and it fell within the range from 2.83 to 7.60 %, which is beneficial for the neutralisation of heavy metals. At the depth from 0.3 to 0.6 m the average content of organic matter varied from 2.52 to 4.62 % (Table 3). Soils from the Bogatynia region are medium and heavy silty soils (silt loam) with a high content of silt fraction, whereas soils in the proximity of Zgorzelec include very light, light and medium soils (sand, loamy sand, sandy loam), in which a high content of sand fraction was found [9, 15, 21].

Mercury

Mercury belongs to the group of chemical elements that are characterised by particularly strong chemical and biological activity and variability of the forms of existence [5]. It is one of the strongest environmental poisons and its toxicity depends on the form.

In Poland there are no deposits of mercury ore. The main sources of this element in raw materials mined in our country are brown coal, copper ore and hard coal (Table 4). The average mercury content in Polish brown coal is 332 ppb. In the Turów deposit maximum values reaching 950 ppb were noted. Dutch research showed a similar

Table 3. Results of the analysis of physical and chemical properties and grain size distribution of soil samples [9]

Research area	Depth (m b.g.s.)	Parameters	pH		Organic matter (%)	Fraction content (%)		
			H_2O	1M KCl		A	B	C
Bogatynia Region	0.0–0.3	range	5.38–6.32	4.73–6.22	3.63–7.60	23–29	56–68	8–12
		mean	5.80	5.64	5.76	25.67	63.00	9.67
	0.3–0.6	range	5.58–6.42	4.81–6.05	2.52–4.62	15–26	58–72	9–13
		mean	6.02	5.46	3.79	21.00	65.00	11.33
Zgorzelec Region	0.0–0.3	range	5.51–5.96	4.70–5.32	2.83–6.71	52–98	2–41	0–7
		mean	5.63	5.02	5.30	78.80	16.40	3.60
	0.3–0.6	range	3.42–6.01	4.88–5.42	2.54–4.18	51–98	1–39	0–9
		mean	5.65	5.15	3.53	81.80	13.60	3.20

Explanation: A - Sand fraction 2.0-0.05 mm; B - Silt fraction 0.05-0.002 mm; C - Clay fraction <0.002 mm.

Table 4. Sources of mercury in raw materials mined in Poland [12]

Source	Participation in mercury yield [%]
Brown coal	48.4
Copper ore	29.2
Hard coal	19.4
Zinc ore	1.6
Clay raw materials	0.8
Carbonate raw materials	0.6
Total	100

mercury content in brown coal imported from Poland – 350 ppb. Compared to the values obtained in brown coal originating from other countries these are very high: Germany – 160 ppb, the USA – 140 ppb. The lowest content was noted in coal from Columbia and Russia, i.e. 60 ppb [12].

According to the data of PSE S.A. [23], the total installed capacity of hard coal power plants in Poland in the year 2011 was 20 152 MW, and of brown coal power plants – 9630 MW. The coal mine Turów mined 861 tonnes of coal from the beginning of its operation to 2010, while 881 million tonnes were mined in the Bełchatów mine, 553 million tonnes in Konin and 187 million in Adamów. Brown coal-based power plants account for 26% and hard coal power plants account for 54% of the installed capacity in the National Energetic System. The analysis of the composition of brown coal from the Turów coal mine presented hereinabove, together with the amount of coal combusted in the power plant Turów, combined with the additional exposure to cross-border contamination points to a potential threat of contamination of soils in this region with mercury.

Mercury is released to the environment mainly during the combustion of coal, which naturally contains some quantities of this element. Mercury is usually found in coal in combination with sulphur (65–70%), and the other 30–35% are connected with the mineral ash and organic fraction of coal [4]. Global mercury emission from natural and anthropogenic sources ranges from 4400 to 7500 tonnes per year, where the anthropogenic emission accounts for as much as 3500 tonnes per year. In Europe the main source of contamination, accounting for approximately 69% is the combustion of fuels, and 6% is caused by metallurgy of non-ferrous metals. The emission of mercury into the air in quite high in Poland, and the highest amounts of mercury (over 60%) are released as a result of combustion of hard coal and brown coal [19, 24]. Mercury bound in the Earth's crust or in fuels does not pose a significant threat. Only when released it becomes highly mobile and remains in the environment permanently. It is one of the elements characterised by the highest accumulation coefficient, i.e. the relation between the quantity of metal obtained from natural resources to the amount for which demand exists [5]. Natural mercury content in soils varies within the range from 0.05 to 0.3 mg Hg kg^{-1} and the acceptable value in industrial areas, minerals and communication areas (lands in the C group) is 30 mg Hg kg^{-1} dry mass in superficial layers of soil [24, 26]. This element accumulates mainly in the upper layer of soils, and its concentration decreases with the increase in depth [27]. In soil solution it is found in a form of ions, where it is subject to methylation processes with the participation of microorganisms, and the emerging methylated derivatives become more available for plants. High concentrations of mercury were noted in clayey soils (where the sorption of this metal is the strongest) and in peat soils. The pH of soils is an important factor influencing its distribution. In soils with a pH from slightly acidic to alkaline it is strongly tied to large- and small-particle humus substances which leads to its immobilisation, limits the migration of contaminants to the root system of plants and thus into the circulation of matter [1]. In acidic soils it is tied mainly to organic matter and the lowering of the pH value results in an increased availability of organic mercury compounds to plants and to the release of its toxic forms, even if originally they existed in a non-available form. Factors influencing the transformation of this metal in soil also include the content of organic matter, soil structure, mineral composition or the solubility of soil solution [14, 24]. Study conducted in the area of the Bogatynia commune in 2002 showed the presence of mercury in arable soils in the amount from 0.01 to 0.17 mg Hg kg^{-1} whereas in the proximity of Działoszyn these amounts reached even 0.85 mg Hg kg^{-1} [22].

In own analyses (Table 5), in samples of soil collected from the depth from 0.0 to 0.3 m, originating from the area directly influenced by the Power Plant and Coal Mine the maximum mercury level amounted to 0.1028 mg Hg kg^{-1} and it was nearly twice as high as that obtained by Kusza et al. [10] in soils from industrial area in Opole, although 25 times lower than the maximum content of this metal found in the soils of Jeleniogórski District, adjacent to the region of Zgorzelec and

Table 5. Content of mercury in soils

Research area	Parameter	Mercury (mg Hg kg^{-1})	
		Depth (m b.g.s.)	
		0.00–0.30	0.30–0.60
Bogatynia Region	range	0.042–0.102	0.043–0.073
	mean ± SD	0.067±0.025	0.062±0.013
Zgorzelec Region	range	0.033–0.078	0.018–0.061
	mean ± SD	0.059±0.017	0.040±0.017

Explanation: SD – standard deviation.

Bogatynia [8]. Similar mercury contents for the analysed area were obtained by Pasieczna [20]. The average mercury content in world soils is 0.06 mg Hg kg $^{-1}$ [13], in the research presented by Li [11] the mercury content varied from 0.24 for agricultural soils to 0.35 mg Hg kg^{-1} for soils in China, 3.13–3.82 mg Hg kg^{-1} for mining areas in China and Iran and 52.9 mg Hg kg^{-1} for contaminated areas in the proximity of a mercury mine in Spain. In the context of the presented mercury content in the soils of agricultural and industrial areas, the values obtained in the Bogatynia region are low, they fall into the range determined by Polish geochemical background and average mercury content in world soils.

Aluminium

Aluminium is a basic component of soils. It is a part of the composition of most minerals, where it is usually tied to hardly soluble, non-toxic aluminium silicates or creates amorphous hydroxides, characterised by a high capacity to absorb trace metals, among others. Only 0.1% of aluminium contained in soils has a soluble or exchangeable form, and the content is strictly linked to the type of soil and to its physical and chemical properties [25]. Particularly high levels of this metal are found in soils with a high content of the fraction below 0.02 mm, as well as these characterised by a lower content of organic matter. This dependence was noted during the analyses of hydrogenic soils and peat-muck soils originating from river valleys in the region of Grójec and the upper course of Liwiec river [7, 18]. The highest aluminium content, i.e. 3.38 g Al kg^{-1}was found in muck soils, the second highest – in the peat layer (2.71 g Al kg^{-1}), and the lowest, i.e. 1.5 g Al kg^{-1} in the mineral bed. Moreover, it was observed that in the deeper situated layers the quantities of this metal are significantly lower, which is also connected with lower organic matter content. The factor that strongly determines the increase in the activity of aluminium and its compounds is the pH of soils. As aluminium is easily soluble in acidic environment, the most significant processes occur in soils with a pH ranging from 4.0 to 4.5 [2]. In particularly acidic soils, with a pH ranging from 3.0 to 4.0 they may additionally replace other, useful cations from the absorption complex, which results in the decrease in the content of nutrients necessary for plant development in soils. High aluminium content, through its antagonistic activity, also contributes to the decrease in the content of calcium and magnesium in soil [17]. The increased content of this metal is particularly dangerous in the surface-adjacent layers of soils, as it causes both poor rooting of plants and a weakening of the development of the root system. High content of available forms of aluminium in arable soils leads to an increase of the quantities of this metal in plants, in particular in their underground parts. This leads to disturbances in the transport and collection of nutrients (such as phosphorus, potassium, calcium and natrium) from soils, along with a simultaneous increase in the collection of iron and manganese [2]. The increase in the activity of aluminium is also caused by anthropogenic factors, connected with human activity, which have a significant influence on soil acidification process [16]. Although one of the major sources of this metal is the chemical industry, some quantities of it are also released during the combustion of coal and in the metallurgical industry. Inorganic forms of aluminium are released with particular intensity in regions exposed to the deposition of strong, inorganic acids, in the proximity of mines, deposits of pyrite waste and means of transport emitting nitrogen and sulphur oxides. As a result, alkaline cations are replaced by hydrogen and aluminium cations, what causes a further increase in soil acidity [28]. Additionally, the acidification of the environment by acid rains increases the activity of aluminium,

Table 6. Content of aluminium in soils

Research area	Parameter	Aluminium (g Al kg^{-1}) Depth (m b.g.s.) 0.00–0.30	Aluminium (g Al kg^{-1}) Depth (m b.g.s.) 0.30–0.60
Bogatynia Region	range	19.32–32.42	21.80–30.20
	mean ± SD	24.56±5.65*	25.50±3.50*
Zgorzelec Region	range	3.07–16.02	3.46–17.80
	mean ± SD	7.71±5.16*	8.56±5.88*

Explanation: SD – standard deviation; * statistically significant differences at $p \leq 0.05$

stimulates the process of dissolution of its compounds and of silty minerals. The analyses of aluminium content in soils collected at the depth from 0 to 30 cm in Opole showed that the soils contained from 3.44 g Al kg^{-1} to 14.8 g Al kg^{-1} and that its mobility was strongly connected with the pH of soils [3]. These values are lower than those obtained for the range of influence of the Coal Mine and Power Plant Turów that are discussed in this study. One of the regions where soils are exposed to the emission of numerous toxic metals, similarly to the Zgorzelec and Bogatynia region is the Silesian Beskids [31], where the influence of contamination originating both from the Czech Republic and Upper Silesia is noticeable. The aluminium content in soil determined by the authors fell into the range from 23.2 to 50.4 g Al kg^{-1} and they were higher than those obtained as a result of our analyses of soils in the Zgorzelec and Bogatynia regions. Certainly the local and cross-border contamination overlapping in these regions and the physical and chemical properties of the soil itself may significantly influence the adsorption of this metal. The conducted study of aluminium content (Table 6) showed a significant, nearly 3 times higher level of this metal in the range of direct influence of the Coal Mine and Power Plant Turów in comparison with the Zgorzelec region, regardless of the depth from which the samples were collected.

The mean content of aluminium in soil samples collected from the depth from 0.0 to 0.3 m in the Bogatynia region was 24.56 g Al kg$^{-1.}$ whereas in the Zgorzelec region – 7.71 g Al kg$^{-1.}$ and in samples collected from the depth from 0.3 to 0.6 m, respectively: 25.50 and 8.56 g Al kg^{-1}. For soils in the industrialized area of Hamburg the value 6.0 g Al kg^{-1} was obtained, and in Bangkok 13.8 g Al kg^{-1} [29]. Slightly higher aluminium content in samples of soil collected from the depth from 0.30 to 0.60 m in both analysed regions confirm the relation between accumulation and depth, as well as the results of tests obtained by other authors. In the context of the presented literature survey the aluminium content in the analysed area may be considered as increased.

CONCLUSIONS

The authors did not note a significantly higher mercury level in arable soils in the range of influence of the Coal Mine and Power Plant Turów, in comparison with soils from the Zgorzelec region, neither for samples collected from the depth from 0.0 to 0.3 m nor for those collected from the depth from 0.3 to 0.6 m, in spite of the disadvantageous, acidic pH of soils and higher content of small grain fractions in the soil. The obtained results do not differ from the geochemical background in Poland and they are similar to the results for the analysed area presented in literature. No influence of industrial contamination on the increase in mercury content in arable lands in the discussed area was found in the tests.

The significantly higher level of aluminium content in the range of direct influence of the Coal Mine and Power Plant Turów in comparison with the Zgorzelec region, regardless of the depth from which the samples were collected, is connected with the emission of contaminants and with the physical and chemical properties of soils – mainly the acidic pH caused by the emission of nitrogen and sulphur oxides. The aluminium content in soils in the Bogatynia region was higher than in soils from industrial area in Opole and lower than in soils originating from the Silesian Beskids.

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