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Physicochemical and geophysical investigation of soils from former coal mining terrains in southern Poland.

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ABSTRACT: To investigate the influence of the long-term activity of a coal mine on soils in its immediate vicinity, mineral compositions, heavy metal (HM) concentrations and hydrocarbon concentrations were determined for samples collected from one of coal mines in the USCB, southern Poland. The ESEM method was used to analyse the mineral compounds of industrial soils (IS) and elevated Zn, Pb and Ba contents in relation to the legislative limits for contaminated sites were found. The highest HM quantity (up to 1589 mg·kg-1) was measured for Zn. Among hydrocarbons only mineral oils and PAH showed elevated concentrations, however, their values did not exceed the permissible limit of the standard for IS. HM concentrations in the areas of former coal mining activities were compared with values for urban soils. In addition, geophysical methods, such as geoelectrical and refraction methods, were used to determine bearing capacity and thickness of the anthropogenically altered surface soil layer. Research methods applied were very useful for preliminary assessment of the environment in which mining activities took place over many years.

1 INTRODUCTION

The Upper Silesia Coal Basin (the USCB) in southern Poland is one of the largest regions in Europe where the natural environment has been changed as a consequence of coal exploitation on a large scale. In the 1980s the annual coal output averaged 190 – 200 M tons with 70 coal mines running (Helios-Rybicka, 1996), whereas today 30 mines are still productive with an exploitation of 90 m tons of coal per year. In the USCB there are ca. 700 km^2 coal fields under the direct influence of exploitation and ca. 1000 km^2 are regarded as being under its indirect impact (Cabala et al., 2004a).

When Poland became a member of the European Union, economic policy on the industrial structure of towns connected with mining activities has had to be significantly changed and new mechanisms of management and market economy have been introduced. Particularly important for further town development are sites on which mining plants with shafts and coal processing plants had been built and places in which fuel, waste materials and debris had been stored (Cabala et al., 2004b). These terrains might be affected for more than a hundred years as a result of mining operations. A short time after closing of any coal mine, post-mining areas including all ancillary facilities were placed under the supervision of local authorities. These terrains seem to be interesting in respect to the further development of towns which have until now been perceived as typically mining and industrial ones. These terrains are from several to tens of hectares in size and are situated near city centres. In respect of infrastructure they can be classified as having great potential. However, the town development strategy does not disclose full information as to rules and regulations for rehabilitation and revitalisation of post-mining areas. That is why it poses a problem as to how such land should be developed in a proper way. In general, recreation grounds, green belts, Scientific and Technological Parks, light industrial development zones have been designed on such areas. Although investors also take interest in these areas, as they could have great possibilities for flourishing towns and businesses, they are limited because of the scant knowledge of environmental conditions. Soil and water contaminated with heavy metals and hydrocarbons owing to mining activities has not been sufficiently examined. In addition, the properties of the bearing layer have not been recognised yet. At present it is very essential to develop the most effective methods for a comprehensive evaluation of the environmental state and to establish potential threats for future users of these terrains.

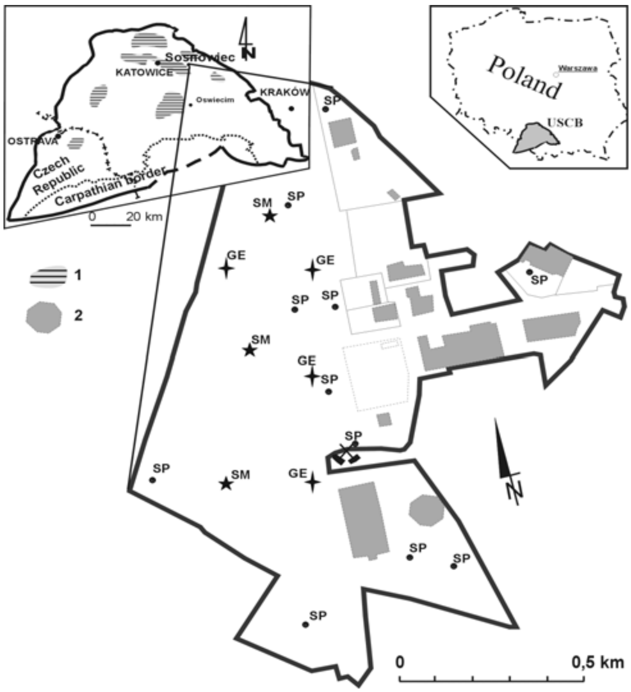

Figure 1. Location of mining areas in the Sosnowiec coal field in the USCB. 1 – areas of intensive coal exploitation, 2 – industrial infrastructure, SP – sampling points, GE - sites of geoelectrical sounding, SM - sites of seismic study.

2 MATERIALS AND METHODS

2.1 Study area & soil sampling

The research was carried out in Sosnowiec in the post-mining area of the ‘Niwka-Modrzejów’ coal mine which was closed in 1999 (Fig. 1). In the study area the mining activities started in 1909. Plant sites were rebuilt several times from that time on. This resulted in the formation of an anthropogenically altered surface layer with the thicknesses from dozens of centimetres to several metres. The primary substrate is comprised of Quaternary glaciofluvial sands and alluvial deposits lying over Triassic carbonate rocks. Geophysical methods such as vertical electric sounding (VES) and shallow refraction seismic were used to evaluate thicknesses and to determine the degree of surface layer alteration. Chemical analyses were made on 11 IS samples taken at depths of 0.2 m and 2 m, respectively. Two-kilogram samples were averaged and ground. The laboratory research including chemical analyses and the ESEM method was carried out in the Faculty of Earth Sciences, University of Silesia, Sosnowiec.

2.2 Geophysical investigations

VES was performed with the use of the geoelectrical equipment TERRAMETR SAS 4000 together with a MULTIMAC system in four measurement positions. The Schlumberger electrode system was applied in the study with a maximal spacing of 81 m. The data were interpreted to obtain information about the thicknesses of layer differing in electrical resistivity. Refraction measurements were done in three positions with the seismic equipment TERRALOC mk 6+. Anisotropy of seismic properties for altered layers was determined on the basis of data obtained from azimuth seismic prospecting. Seismograms were interpreted to estimate the longitudinal wave velocity in the study area.

2.3 Chemical analyses

The trace element content was determined with a Unicam SOLAAR M6 spectrometer (AAS). The hydrocarbons were analysed with an Agilent Tech gas chromatograph, type 6890, series II, in conjunction with a mass spectrometer, type 5973 Network. The gathered data and spectrum processing were performed with the Hewlett Packard Agilent Technologies Chemstation programme. Bituminous substances were extracted and separated into fractions with the use of preparative thin-layer chromatography (PLC). Column chromatography (CC) was used to perform the extraction of the petrol fraction.

2.4 ESEM investigation

BSE images and EDS analyses were done using a Philips XL 30 ESEM /TMP environmental scanning microscope with an EDS analytical attachment (type EDAX Sapphire). From the IS substrata mineral grains bigger than 1 mm were removed. Using magnifying binoculars, mineral grains were separated according to their shape and colour. Samples for SEM analyses were prepared and lay on the carbonic strips (1x1.5 cm) and then they were glued on aluminium discs.

3 RESULTS AND DISCUSSION

3.1 Industrial soil properties

The largest thickness of the anthropogenic layer (up to 2.1 m), which was found using geophysical research methods, occurs in the central part of the mining plant in the vicinity of shafts and also in the area of former industrial buildings (Fig. 1). In the northern direction the thickness of the layer diminishes up to ca. 1.5 m. In the peripheral zones of the plant the thickness does not exceed 0.5 m.

The IS, which occur on anthropogenic embankments, include a considerable quantity of concrete, debris, bricks and coal processing waste. In general, there is no soil layer which would enable a lush vegetation development. Ruderal plants and sparse species of grasses appear sporadically. The soil layer (0.2 – 0.4 m), which makes vegetation possible can only be found in the vicinity of administrative buildings of the coal mine where the surface layer has not been strongly altered.

3.2 Mineral composition

The main mineral groups, which are included in studied IS, were determined using the ESEM method and EDS analyses (Tab. 1). The mineral content comprises quartz, clayey minerals and carbonates. The surface layer of the anthropogenic embankment is characterized with high contents of coal, carbonates, oxides, iron hydroxides, slags, metal-bearing sulphides and carbonates (Tab. 1). The mineral components of the soils are characterized by different forms, e.g., crystalline, amorphous or spheroidal. A significant part of aluminosilicate aggregates have spheroidal forms, typical of pyrolysis products. Traces of melting are found in sintered carbonic and aluminosilicate aggregates which derive from metallurgical waste, slags and ash. The industrial soils show neutral or slightly alkaline reaction (pH 6-7).

The presence of secondary crystalline calcium carbonate is characteristic (Fig. 2) These carbonates are connected with anthropogenic embankments abounding with debris and carbonate aggregates. Lower values of pH (4.2-6) are typical of layers consisting of carbonates and Carboniferous rock waste materials including carbonaceous schist and organic matter. The applied methods enabled the recognition of the following metal-bearing minerals: blue lead, lead carbonate, $PbCO_3$ (Fig. 3); zinc sulphides, ZnS (Fig. 4); spinels with Al, Fe, Mg, Zn, Fe, iron oxides, Fe_2O_3, iron sulphates, $FeSO_4*nH_2O$; barium sulphate, $BaSO_4$ (Fig. 5).

In the investigated IS here, only hydrated iron sulphates have been found in small quantities. No Ca sulphate has been identified. Leaching of sulphate ions in the soils studied is limited. Unstable Fe sulphides have not been identified either. Among minerals which include Fe, stable oxides and hydroxides have been recognized and their appearance points to the limited ability to leach Fe into soil solutions.

Table 1. Mineral occurrence in IS in the area of the coal mine.

Main mineral groups				
Aluminosilicate:	Fe hydroxide	Quartz	Organic matter	Sphalerite
K;	Fe oxide	Calcite	Carbon & aluminosilicate	Cerussite
Ca,Mg,Fe;	Fe & Al spinel	Dolomite	Agglomerate	Barite
Ca,Mg,Fe,Zn;	Magnetite	Ankerite	Aluminosilicate slag	Zircon
Fe,Mn	Fe sulphate	Si oxide (enamel)	Cellulose with Si	Phosphate REE
K feldspar			(wood with Si)	
Kaolinite, Illite			Amorphous silica	
Hydrohalloysite			Wood carbonaceous	

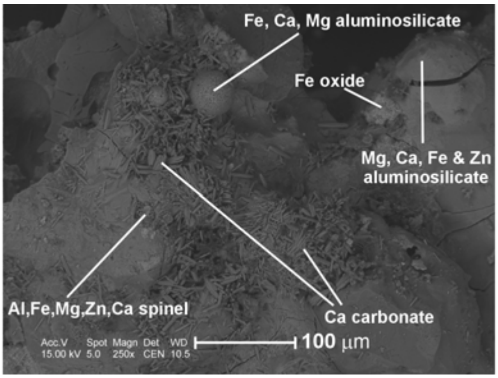

Figure 2 Metal-bearing mineral aggregates in industrial. soils BSE image.

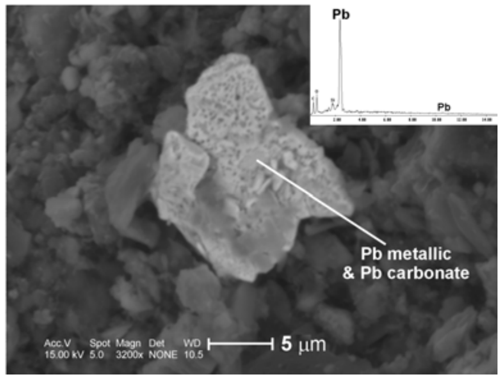

Figure 3 Metal-bearing mineral components in industrial soils. BSE image.

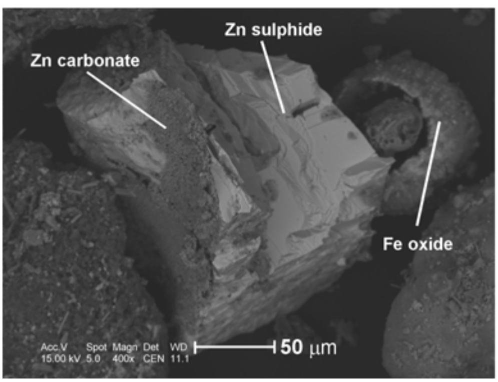

Figure. 4 Metal-bearing, crystalline mineral components in industrial soils. BSE image.

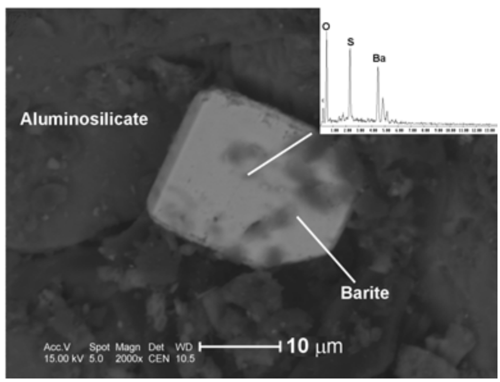

Figure. 5 Crystalline barite grains in industrial soils. BSE image.

4 HEAVY METALS

ESEM methods enabled the recognition of enhanced Zn, Pb and Ba contents occurring in the IS under investigation. AAS analyses confirmed the high concentrations of these metals (Tab. 2). The relatively high HM quantity, which exceeds the admissible value in industrial areas, was only measured for zinc (Fig. 6). The other concentrations of HM are contained in the permissible range of the standard for farm and urban areas. As and Hg contents are lower than 2 $mg \cdot kg^{-1}$ and Mo and Sn are less than 10 $mg \cdot kg^{-1}$. The highest Zn and Pb quantities were measured in the central part of the mining plant in the vicinity of the shaft.

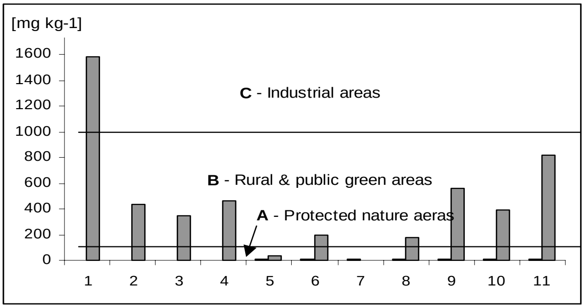

Figure 6. Zn concentrations in IS in the area of the mining plant. Horizontal lines indicate the permissible values according to the Polish Law.

Table 2 Descriptive statistics for HM in IS samples

n=11	Zn	Pb	Cd	Ni	Cu	Co	Ba	Cr
Mean	456.6	86.0	0.7	12.6	31.7	8.5	145.9	15.4
Median	391.0	49.0	0.1	8.0	30.0	9.0	150.0	5.0
Max	1589.0	454.0	2.6	31.0	84.0	12.0	300.0	64.0
Min	2.0	2.0	0.1	2.0	2.5	2.4	5.0	1.0
Std dev	442.8	127.9	0.9	11.2	24.5	2.9	103.1	20.5

5 HYDROCARBONS

In samples under investigation enhanced quantities of carbonaceous substances were found. They are connected with carbonaceous schists, slags and other post-mining waste which were used for hardening the surface of the area. The contents of light hydrocarbons (C6-C12) are low, whereas mineral oils (C12 – C35) and PAH show considerably higher values. However, hydrocarbon concentrations measured in the industrial soils do not exceed the permissible values. Surface layers (0-0.2 m) were endangered most by the influence of industrial organic contaminations. The concentration of organic contaminations decreases with depth. It might be connected with both washing out by infiltration waters and PAH degradation by bacteria. Chlorinated hydrocarbons, tetrahydrofurane, pyridine, cyclohexane and cresol have not been identified in the samples. Concentrations of benzene, ethyl-benzene, toluene, xylene, styrene, naphthalene, phenanthrene and chrysene are relatively low.

6 ELASTIC PROPERTIES OF LAYERS

Seismic measurements carried out in the study area show that the value of longitudinal wave velocity in the surface layer averaged ca. 500 m/s. It confirms that the consolidation of this medium is not very strong. The layer shows certain anisotropy of elastic properties. The velocity changes from 350 m/s to 700 m/s according to the azimuth of the measuring profile. The highest velocities have been found in the NNE and the lowest ones in the NNW direction. The anisotropy of elastic properties that probably results from the directional arrangement of the remains of the dormant mine’s infrastructure is a feature that distinguishes the anthropogenically altered layer from the dry sand beds in which the velocity of seismic waves is similar. Seismic data revealed that under the altered layer there occurs a more consolidated one in which the velocity of longitudinal waves is ca. 1400 m/s. This is probably a bed of moist sand, which is confirmed also by the VES investigations results (the specific resistivity of this bed amounts to ca. 150 ohmmeters).

7 SUMMARY AND CONCLUSIONS

IS on areas of dormant coal mines are characterised with various concentrations of HM and hydrocarbons. The concentrations are within the limits of standards for IS on industrial terrains. The highest levels were measured for Zn: the pollution with this metal is higher on the study area than in urbanised areas of large European cities (Tab. 3). The concentrations of Pb, Cu, Ni and Cr are similar to values determined for town soils. Higher concentrations of zinc are related to the presence of aluminosilicates and spinels containing zinc absorbed in their structure. Other sources of zinc are sulphides of Zn present in materials used for the construction of embankments. High levels of lead are connected with the presence of this metal in various elements of the old mine's technical infrastructure. Barium is present in a stable sulphate of barium, i.e. barite.

Table 3 Mean metal content (mg kg –1) in urban soils

City	Zn	Pb	Cu	Ni	Cr	References
Torino	183	149	90	209	191	Biasioli et al., 2006
Madrid	210	161	72	14.1	75	De Miguel et al., 1998
Napoli	251	262	74		11	Imperato et al., 2003
Warsaw	140	53	25		13	Pichtel et al., 1998
Sosnowiec*	456	86	31.7	12.6	15.4	Present study

* - IS, area of a dormant coal mine.

Poland’s legislation (Journal of Laws Dz.U. 165 item 1359, 2002) defines permissible concentrations of HM. They are similar to standards used in other EU countries and in the case of certain metals are even stricter (Tab. 4). In the polish legislation the permissible HM concentrations depend on the water permeability of soils and this often makes a univocal classification of a soil difficult.

Table 4 Legislation limits for contaminated sites in Italy and Poland. Pseudototal content (mg kg-1).

Element	Public and private green areas	Commercial and industrial areas	Protected nature areas	Rural & public green areas	Industrial areas
	Italy (in Biasioli et al., 2006)		Poland (Journal of Laws Dz.U. 165 item 1359, 2002)		
Zn	150	1500	100	350	1000
Pb	100	1000	50	200	600
Cu	120	600	30	150	600
Ni	120	500	35	100	300
Cr	150	800	50	190	500

The level of hydrocarbon contamination does not exceed standards for industrial areas. The main source of hydrocarbons in the IS are slags, ash and coal processing waste used in the embankments construction and paving the surface of the plant’s area. During several tens of years from the formation of industrial soils part of the hydrocarbons was subject to natural biodegradation. The results of geophysical studies point to a fact that the anthropogenically altered surface layer from 0.5 to 2.1 m thick is characterised with poor elastic properties and thus cannot be regarded as a bearing layer for buildings construction. The underlying layer which is better compacted and has much better elastic properties may be used for the construction of building foundations should the mining plant area be developed.

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