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ANTHROPOGENIC SOILS POLLUTION WITHIN THE LEGNICA–GŁOGÓW COPPER DISTRICT

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Abstract. The industrial complex of the Legnica–Głogów Copper District (LGCD) with two copper smelters, tailings ponds, a few shafts, and copper ore reloading facilities are the main sources responsible for pollution of soils with heavy metals. A geochemical soil survey was conducted in the LGCD in 1996. The survey was arranged with a 1x1 km grid pattern while the field work in the regions of particular interest (of copper smelting industry and copper mining) followed the more detailed grid pattern of 500x500 m. Samples were leached with *aqua regia*; next, using the ICP-AES method, determinations of Ag, Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Mg, Ni, P, Pb, S, Sc, Sr, V, and Zn concentrations were made. As far as Hg was concerned, its concentration was measured using the CV-AAS method. Sieve analysis and laser method were employed to define grain-size composition of soils.
The geochemical survey of the LGCD area soils revealed that occurrence of elements such as Al, Co, Cd, Mg, Ni, Sc, Sr, Ti, and V was mainly related to the structure of geological basement. Mining, ore processing, and copper ore smelting were the main sources of anthropogenic pollution of soils by copper and lead as well as by silver arsenic, zinc, cobalt, and nickel (to smaller degree). Other local sources of soil pollution were towns with associated industry, transport system, and local emissions of dust and gases from coal burning households and local heating plants. Geochemical anomalies that are related to the copper industry cover vast areas with high copper and lead concentrations in the "Głogów" and "Legnica" copper smelters environs. Increased content of other elements, such as silver, arsenic, zinc, cobalt, and nickel occurs within copper-lead anomalies areas. Urban soils in Legnica and Głogów, in the areas affected by smelters, are significantly polluted with copper and lead.

Key words: anthropogenic pollution, geochemical mapping, soils, southwestern Poland.

Abstrakt. Kompleks przemysłowy Legnicko-Głogowskiego Okręgu Miedziowego (LGOM-u), na który składają się kopalnie rud miedzi, dwie huty miedzi, zakłady przeróbcze i osadniki odpadów poflotacyjnych, jest głównym źródłem zanieczyszczenia gleb tego rejonu. Opróbowanie gleb na terenie LGOM-u przeprowadzono w 1996 r. stosując gęstość podstawową 1x1 km oraz zagęszczenia do siatki 500x500 m w rejonach hut i obszarach górnictwa miedziowego. Próbki gleb trawiono wodą królewską, a następnie oznaczano w nich zawartość Ag, Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Mg, Ni, P, Pb, S, Sc, Sr, V i Zn metodą ICP-AES. Analizy Hg przeprowadzono metodą CV-AAS, a oznaczenia składu granulometrycznego wykonano metodą sitową w połączeniu z laserowym pomiarem wielkości cząstek. Badania geochemiczne ujawniły, że spośród analizowanych pierwiastków można wydzielić te, które związane są przede wszystkim ze składem chemicznym skał macierzystych (Al, Co, Cd, Mg, Ni, Sc, Sr, Ti i V).
Wydobycie rud miedzi, ich przeróbka i procesy hutnicze są głównym źródłem zanieczyszczenia gleb miedzią i ołowiem, szczególnie w bezpośrednim sąsiedztwie hut. W glebach na terenie rozległych anomalii miedzi i ołowiu wokół hut obserwuje się również podwyższone ilości srebra, arsenu, cynku, kobaltu i niklu. W obszarach miejskich kontaminacja pochodzi również z emisji innych gałęzi przemysłu, transportu, elektrociepłowni i palenisk domowych. Gleby miejskie Legnicy i Głogowa w obszarach narażonych na emisje z hut są znacznie wzbogacone w miedź i ołów.

Słowa kluczowe: zanieczyszczenia antropogeniczne, kartografia geochemiczna, gleby, południowo-zachodnia Polska.

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INTRODUCTION

A geochemical soil survey conducted in the Legnica –Głogów Copper District (LGCD) in 1996 was intended to provide a general assessment of actual chemical state of soils in this region. The industrial complex of the LGCD with copper mines as well as copper smelters "Głogów" and "Legnica" are the fundamental sources responsible for environmental pollution of this area. The contamination is exclusively of anthropogenic character since deep-seated copper ores cannot manifest themselves by a chemical aureole on the surface.

Pollution of soils with heavy metals within the LGCD comes from dust emission of smelters, copper ore reloading facilities, and heat and power generating plants. Results of geochemical soil survey published earlier focused on immediate neighbourhood of smelters and occurred to be limited to small range of studied elements (Roszyk, Roszyk, 1975, 1976; Kabata-Pendias *et al*., 1981; Drozd *et al*., 1984; Andruszczak *et al*., 1986; Szerszeń *et al.,* 1986; Roszyk, Szerszeń, 1988a, b; Szerszeń *et al*., 1991; Czuba *et al.,* 1995; Strączyński, Andruszczak, 1995). According to the mentioned above authors opinions, metal-rich dust is the main source of pollution. The most severe and most arduous dust emission occurred in the early years of this district development.

Another serious problem arises from discharge of sewage into the surface reservoirs, which is responsible for pollution of surface waters and water sediments as well. Brine waters from mines dewatering are discharged to the Odra river.

FIELD WORK

A general sampling grid density was 1×1 km; in the vicinity of copper smelters and copper mining this grid was replaced by sampling pattern 500×500 m. In both cases a hand penetrometer was used to collect a sample from the depth of 0–20 cm. A total number of soil sampling sites reached 5,567. The weight of a mixed sample (5 sub-samples) was approx. 2 kg. When collecting sub-samples, attention was paid to not to mix soils of visibly different grain size

CHEMICAL ANALYSES

Soil samples were dried at a room temperature, then by quartering they were subdivided into two sub-samples: one for chemical analysis, and second for sieve analysis. Sub-samples of grain size <2 mm for chemical analyses were ground in an agate ball-grinder to obtain grain size of <0.063 mm. Samples were leached with *aqua regia* for 1 hour at temperature of 95°C, in a thermostatic aluminium block; then using the ICP-AES method, concentrations of Ag, Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, Mg, Ni, P, Pb, S, Sc, Sr, V, and Zn were determined. As for Hg, its concentrations were measured with the use of CV-AAS method. Chemical elements determinations were carried out on 5,677 samples, totalling in 139,175 results. Measurements of pH were performed in aquatic environment following standard adopted in soil science (Kardasz, Kamińska, 1987). Sieve analysis and laser method were employed to define grain-size composition of soils.

PARENT ROCKS OF SOILS

The surveyed area is located within the Fore-Sudetic Block, covered mostly with Cainozoic sediments. It is a structural unit of an old basement, cropping out on the surface in scarce localities only, in the southern and western segments of the investigated area (Mojski, 1977; Mojski, Sawicki, 1995, 1996). Tertiary sediments occur on the surface in small patches, mostly in the southern part of the area. The Miocene is composed mostly of clayey sediments containing lenses of brown coal. The Pliocene is represented by sands and gravels. Exposures of Tertiary basalts and their tuffs are known in the region of Legnica–Złotoryja (Mojski, Sawicki, 1995). Quaternary sediments form uniform cover over almost entire area, and penetrate deeply the Sudetes Mts. along river valleys. They are mostly of glacial and glaciofluvial origin (tills, gravel and sands) as well as of fluvial sedimentation (sands, alluvia) of Holocene age. Valleys of the Odra river and its tributaries are filled with sediments of accumulation terraces. Connected with these valleys are also occurrences of aeolian sands that sometimes form dunes (Mojski, Sawicki, 1996). The largest area ofthe peat occurrence lies in the Szprotawa valley. Peat are classified as lowmoor sedge-moss ones (Mojski, 1977; Kowalkowski *et al*., 1994).

The type and content of the present-day soil cover are governed by lithologic differentiation of rocks in the basement. Soils that developed on parent rocks described so far represent several types. Buff, brown, and rusty-brown soils are dominant in the region (Szerszeń *et al*., 1995). Of secondary importance are rusty and subsolic soils. Peaty soils (peat) developed within waterlogged areas. The mentioned soil types differ from each other in vertical profile and in grain-size composition, which is an important factor for their applicability to cultivation and to their ability of toxic substances accumulation.

CONSTRUCTION OF GEOCHEMICAL MAPS

The sampling point co-ordinates, field data, and laboratory data were included into one database. The field data constitute a basis for consecutive separation of sub-sets applicable to statistical calculations according to different environmental criteria, such as chemical elements concentrations in cultivated soils, in forest soils or in urban soils (Table 1).

Generation of geochemical maps was based on set of sampling points of known co-ordinates and tested elements content attributed to each one. A triangulation method was used along with a *Surfer* programme. A method of pixels with their side equal to 500 m was employed to construct the maps. The levels of chemical elements contents were chosen following the division into percentiles (15, 25, 50, 75, 90, 95, 97, 99 and 100%).

A simplified base map was used for the construction of geochemical maps. It shows elements of topographic situation such as main roads, railway lines, main rivers, boundaries of communes, outlines of towns and villages, and location of mine shafts and individual plants.

Table 1

Statistical parameters of some elements and acidity in soils

Soils	Parameters	As ppm	Cd ppm	Cu ppm	Hg ppm	Pb ppm	Zn ppm	pH
All soils n = 5,567	a b c	<5 6,000 <5	<0.5 97.3 <0.5	1 86,470 16	<0.05 5.13 0.06	<5 56,000 21	4 16,300 31	3.0 9.1 5.8
Cultivated soils n = 2,857	a b c	<5 126 5	<0.5 7.8 <0.5	3 1,398 18	<0.05 2.74 0.06	<5 1,231 22	7 1,303 35	3.5 9.1 6.3
Forest soils n = 1,282	a b c	<5 68 <5	<0.5 3.5 <0.5	1 2,224 10	<0.05 1.97 <0.05	<5 780 16	4 661 16	3.0 8.0 4.1
Industrial area soils n = 134	a b c	<5 6,000 8	<0.5 97.3 <0.5	4 86,470 71	<0.05 5.13 0.10	9 56,000 57	14 16,300 68	3.5 8.0 6.8
Urban soils n = 182	a b c	<5 31 6	<0.5 2.5 <0.5	7 577 40	<0.05 0.72 0.12	7 1,139 50	11 512 79	4.3 8.0 7.0
Soils of Poland[1] n = 10,840	a b c	<5 3,444 <5	<0.5 253.3 <0.5	<1 6,401 5	<0.05 7.55 <0.05	<3 16,972 13	<1 91,110 35	2.1 9.7 6.1
Cultivated soils of Poland[1] n = 4,899	a b c	<5 168 <5	<0.5 16.7 <0.5	<1 2,190 5	<0.05 4.75 <0.05	<3 2,113 12	<1 2,140 34	2.8 9.3 6.4

a – minimum, b – maximum, c – median, n – number of samples
[1] Lis, Pasieczna, 1995

RESULTS

There is very characteristic differentiation of surveyed soils with respect to their grain-sizes composition. Soils containing a high percentage of the finest fractions (<0.02 mm and 0.10–0.02 mm) occur in the northern part of the surveyed area (hills, mostly built up of glacial till), and in the Odra river valley where their parent rocks are loam, mud, and Holocene sands. In the south, soils of similar grain-size distribution developed from different lithologic formations of various age. An increased content of aluminium is their common feature.

Soils enriched with sand dominate, above all, in the central part of the area where glaciofluvial sands and gravel are dominant in their basement. Similar soils occur in the extreme northern fragment of the map where they have also developed on glaciofluvial sands and gravel.

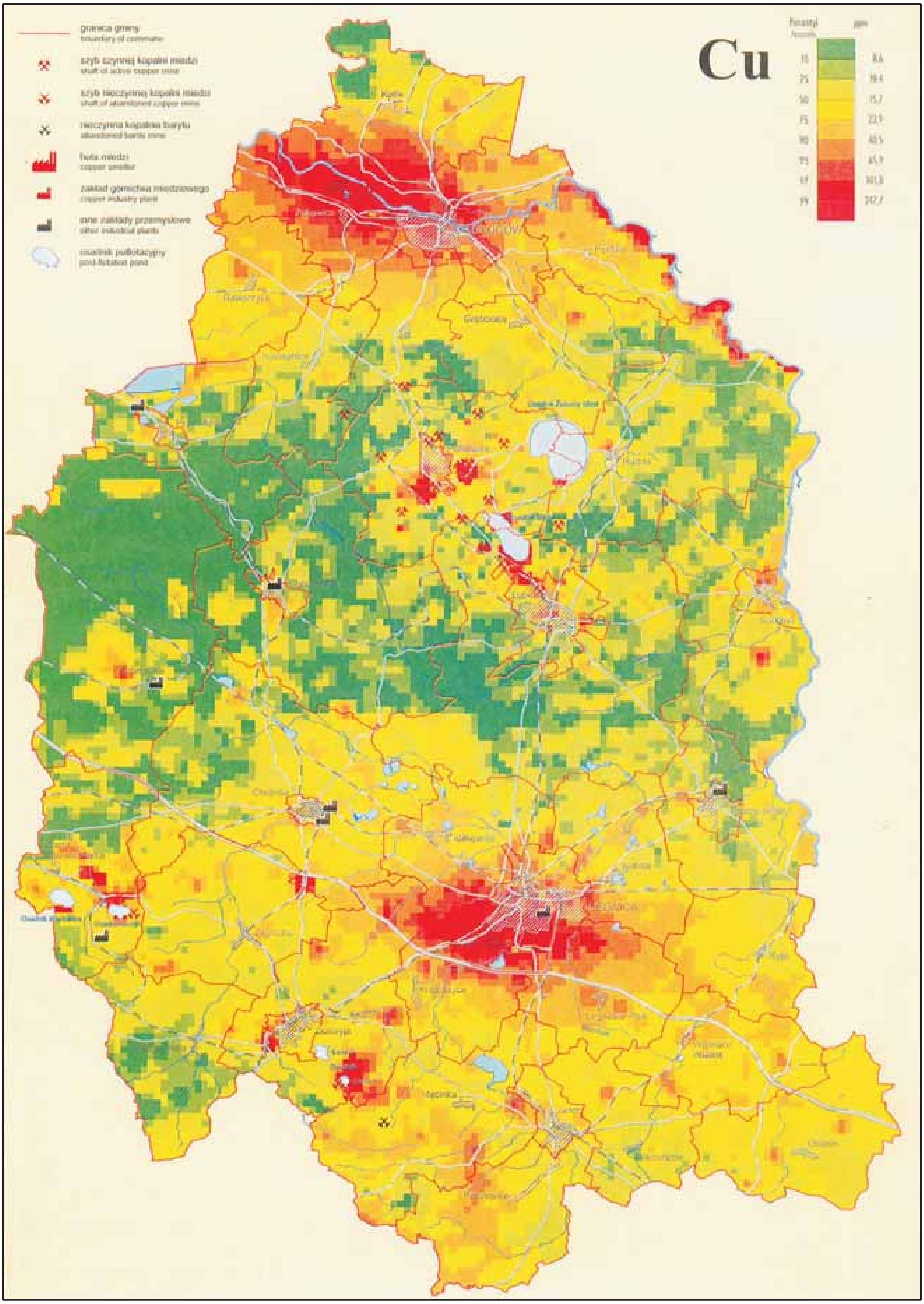

Fig. 1. Copper content in soils (after Lis *et al.*, 1999)

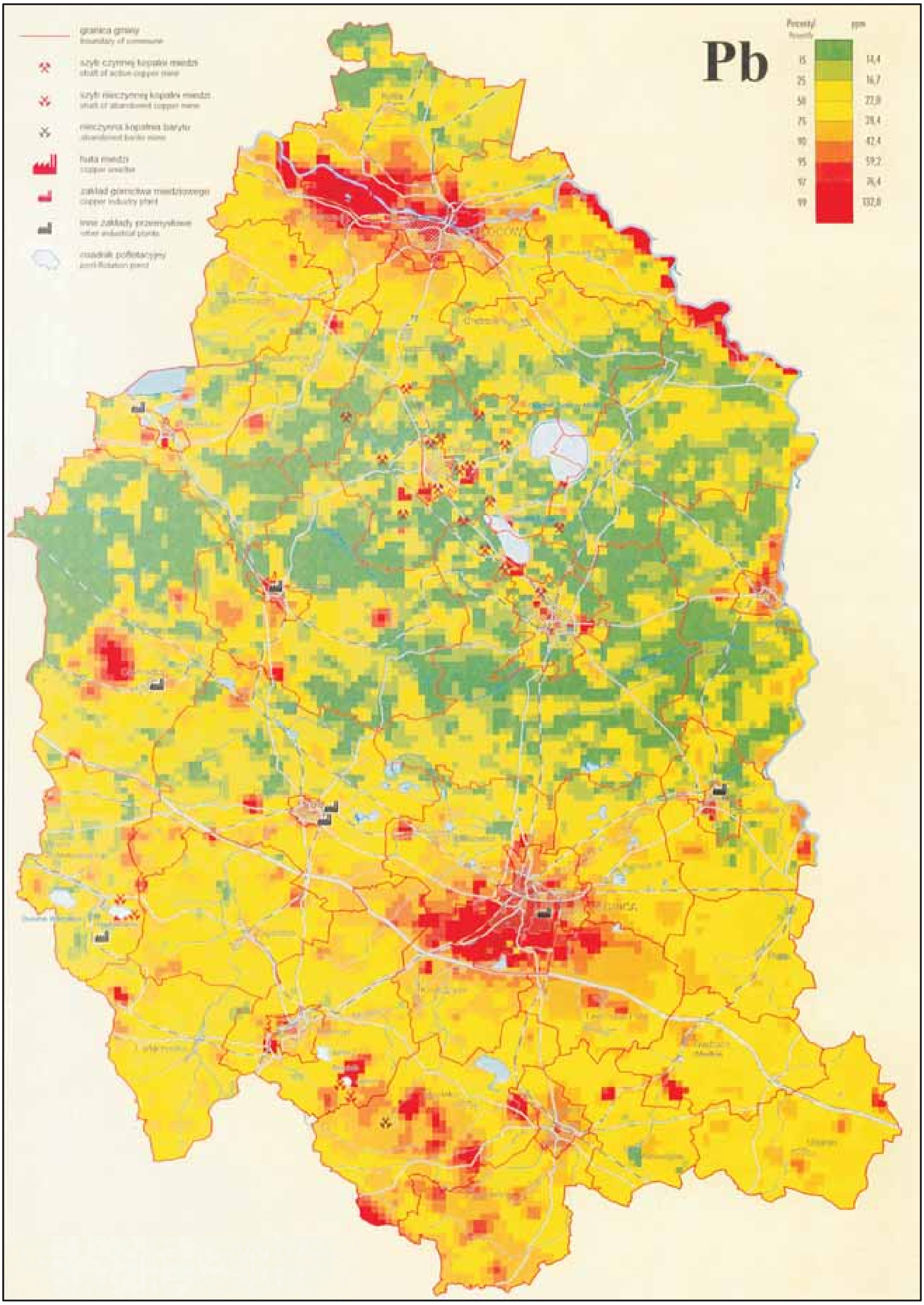

Fig. 2. Lead content in soils (after Lis *et al.*, 1999)

The soils reaction is also very differentiated. Markedly distinguishable are very acidic forest soils. A mosaic distribution of soil reaction is observed within those regions where agriculture is a dominant type of land use. Larger towns distinguish themselves by neutral soils, and also by frequent occurrences of alkaline soils. Elevated pH values in urban and industrial soils are connected with fallout of alkaline dust. This phenomenon was repeatedly observed over the whole Poland (Lis, 1992; Lis, Pasieczna, 1995, 1998, 1999; Pasieczna *et al*., 1996).

Spatial distribution of elements inherited from parent rocks permits to trace differentiation of geochemical background and, if necessary, to distinguish local anomalies of the elements content. Elements contained in soils within the surveyed area, and characterising rocks of geological basement, include Al, Co, Cr, Mg, Ni, Ti, and V. Increased contents of these elements were observed in the south-western part of the Sudetes Mts. and their foreland areas. In the north, increased contents of Al, Co, Cr, Mg, Ni, Ti, and V are noted in soils developed on glacial till. The central part of the studied area is characterised by low and very low concentrations of the afore-mentioned elements in soils that were formed mainly on sandy and sandy-gravely glaciofluvial sediments.

Some elements (As, Fe, P, Pb, S, and V) are very mobile and easily concentrated in soils of suitable properties. This concerns soils of high sorption capacity and rich in organic matter (peaty soils and peat). In these soils, the above mentioned elements develop distinct and sometimes very intensive geochemical anomalies (so called “false anomalies”). They are of natural origin but they are not connected with the presence of ore minerals in the basement or with rocks enriched with the discussed elements.

Mining, ore treatment, and copper ore smelting are the main sources of anthropogenic soils contamination by Cu (Fig. 1) and Pb (Fig. 2) around the Głogów and Legnica smelters, as well as to smaller degree, in the neighbourhood of copper ore processing plants between Polkowice and Lubin. A Cu anomaly around the “Legnica” smelter took a shape of parallel-elongated ellipse (Fig. 1). Maximum Cu content reaching 26,101 ppm was noted in soils within the smelter premises. In the industrially developed areas of the Legnica region, the Cu contents in soils are varying between 15 and as much as 26,101 ppm. Of smaller size, though of similar magnitude (>85 ppm) is a Pb anomaly (Fig. 2). In the industrially developed areas, the Pb contents is changing between 23 and 5,185 ppm. As far as other elements are concerned (Ag, As, Cd, Co, Hg, Mn, Zn), their abnormal concentrations in soils are situated within the smelter premises. Those urban soils that are partly situated within an area of smelters influence exhibit a distinct copper-related pollution (from 14 up to 577 ppm). As regards other elements, lead and zinc only were noted in individual samples in concentrations higher than tolerable.

The copper anomaly (>50 ppm) around the “Głogów” smelter has a shape of an ellipse elongated from ESE to WNW which follows a general course of the Odra river. Soils within the industrially developed area in the Głogów region contain Cu in the range of 40 to 86,470 ppm. Considerably smaller is a lead anomaly (>85 ppm) of similar shape. The Pb contents (Table 1) within the industrial areas are contained in the range of 33 ppm up to as much as 56,000 ppm. Urban soils in Głogów, lying partly within the area of the smelter influence, are markedly polluted with copper (from 35 to 237 ppm).

CONCLUSIONS

A survey of soil geochemistry in the LGCD area indicates that the enriched contents of some elements (such as Al, Co, Cr, Mg, Ni, Sr, Ti, and V) are connected, above all, with basement geology. As a result of migration process in the environment, a group of elements such as As, Fe, P, Pb, S, and V was subject to concentration in organic (peaty) soils and peat.

Areas of higher lead and copper contents around the “Głogów” and “Legnica” smelters express geochemical anomalies that are connected with the copper industry. Elevated contents of other elements, such as silver, arsenic, zinc, cobalt, and nickel occur within the copper-lead anomalies in smaller areas around the smelters, too. Small anomalies of elements of similar origin can also be found near the ore processing plants in the Lubin and Polkowice regions. No soils pollution by metals was traced in areas close to the “Gilów” and “Żelazny Most” tailing waste ponds. However, question on these ponds impact on groundwater remained beyond the scope of this study. It should only be noted here that samples collected from the surface layer (0–20 cm) of the inactive “Gilów” pond have revealed quite substantial amount of metals.

A separate question, not too closely connected with the of the copper industry activity, concerns the pollution of alluvial soils with elements such as silver, arsenic, barium, cadmium, chromium, copper, mercury, lead, zinc, phosphorus, and sulphur within the Odra river valley. Discharge of the Upper Silesia mine water may be the pollution source. This could be evidenced by a high barium content observed in alluvial soils in the Odra river valley, downstream of main rivers confluence that drains the Upper Silesia area. The discharge of municipal sewage and industrial effluents to upper course of the Odra river could be another source of those elements.

As the surveyed soils contain high concentrations of extremely toxic elements (such as mercury and cadmium, for instance), a further detailed geochemical survey of alluvial soils seems to be necessary. Soils under agricultural cultivation do not indicate, in general, that they are polluted with metals. However, it also seems necessary to conduct a further detailed soil survey in those communities that are situated in the smelters vicinity.

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