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Lead distribution in coal and coaly shales in the Upper Silesian Coal Basin

Lead in the Upper Silesian Coal Basin is mainly of inorganic origin. Its average content, in coal and coaly shales of the paralic series, is higher in the western part of the basin. In the limnic series, it is higher in the eastern part of the basin. Great differentiation between the quantity of lead of organic and inorganic origin in individual beds in the Upper Silesian Coal Basin has been observed. A great contrast between the influence of organic and inorganic matter on the average content of this element has been ascertained. It testifies the great dynamics of lead accumulation and dispersion. Differences of lead content in the beds are correlated to the distribution of maximal extensive accumulation and dispersion of lead.

INTRODUCTION

The Upper Silesian Branch of the Polish Geological Institute made 24 boreholes during the period 1975–1988 in the area of the Upper Silesian Coal Basin. They were made within the framework of examination project of the deep horizons of Carboniferous coal-measures. Detailed recognition of the geological conditions of coal occurrence, in the coal-bearing series to the base of the paralic series, was a one of the goals of this project. The boreholes were placed in areas without coal mining and in the neighbourhood of these areas, where coal was being exploited in the upper part of the coal-bearing series. They reached depths of 2300 m, often to Upper Visean sediments.

This paper summarizes an elaboration of the geochemical data collected during realization of the project (A. Różkowska, 1989). The major goals of this paper are: (1) establishing the rules of lead content changes in the Upper Silesian Coal Basin in both organic and inorganic matter and in coaly shales, (2) determining the impact of these

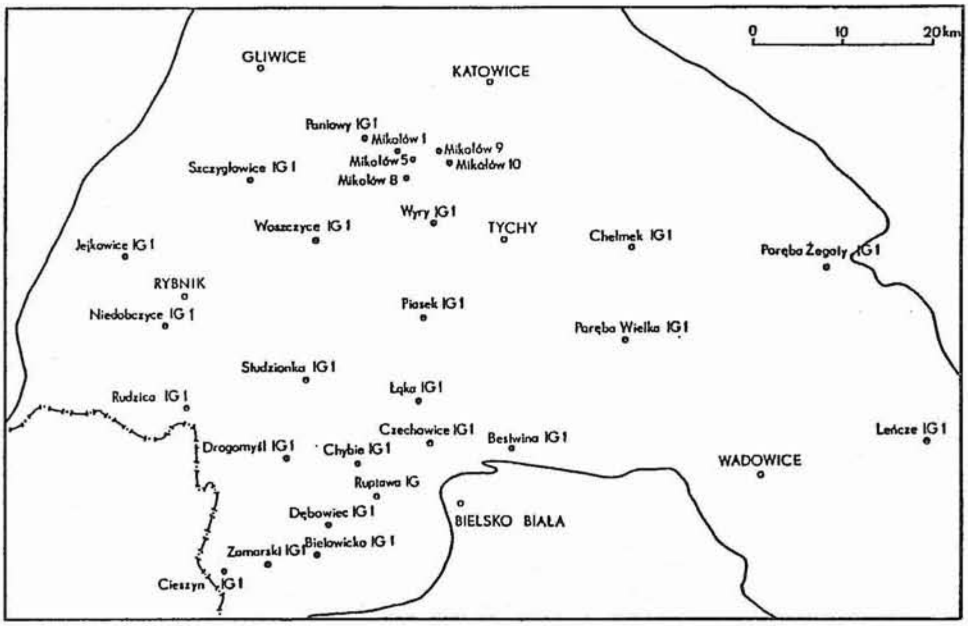

Fig. 1. Location of boreholes in the area of the Upper Silesian Coal Basin
Rozmieszczenie otworów wiertniczych na obszarze GZW

substances on lead accumulation and (3) reconstruction of lead accumulation and dispersion processes in the Upper Silesian Coal Basin.

METHODS OF EXAMINATION AND RESULTS PREPARATION

28 boreholes (Fig. 1) were sampled. One homogenized sample represented each bored bed of coal and coaly shales. The samples were ashed at a temperature of 525°C. Lead content was determined after X-ray fluorescence in 1053 samples of coal and 23 samples of coaly shales from cores of the boreholes made by the Upper Silesian Branch of the Polish Geological Institute, and in 201 samples of coal and 3 samples of coaly shales from cores of the boreholes made by the Katowice Geological Enterprise in the Mikołów area[1].

The quantitative relation of average lead content in coal and coaly shales to quantity of organic and inorganic matter (determined after ash-content measurement) was defined by means of geochemical examinations. The proportional content of these substances influencing average lead content in the studied rocks was also defined. The values mentioned above were calculated after the "distributary function of concentration" by M.

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[1] The detailed analytical results are presented in borehole geological documentations in the Archives of the Polish Geological Institute, Upper Silesian Branch.

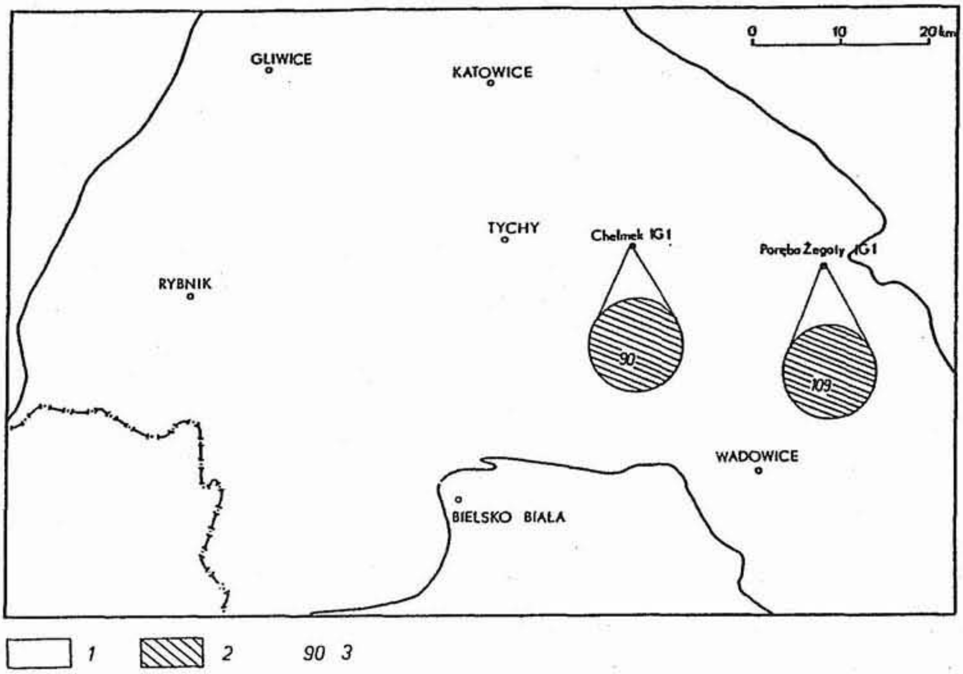

Fig. 2. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Libiąż Beds

1 — organic matter; 2 — inorganic matter; 3 — lead content in organic and inorganic matter (in ppm)

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw libiąskich.

1 — substancja organiczna; 2 — substancja nieorganiczna; 3 — zawartość (ppm) ołowiu w substancji organicznej lub nieorganicznej

Marczak (1985) which shows the relation of lead content to ash content. Separated calculations for two geological units were made:

1. Limnic and paralic series of the Upper Silesian Coal Basin subdivided to two parts: the eastern one (boreholes: Chybie IG 1, Chełmek IG 1, Poręba Wielka IG 1, Poręba Żegoty IG 1, Czechowice IG 1, Bestwina IG 1, Leńcze IG 1) and the western one (other boreholes presented in Fig. 1). This subdivision is based on the results of litho- and biostratigraphical research of the studied units [2], supervised by A. Kotas. The results of calculation are presented in Table 1.

2. The beds: Libiąż, Łaziska, Orzesze, Załęże, Ruda, Poręba, Jaklovec, Hrušov, Petřkovice, Anticlinal Beds and Upper Visean rocks — in selected boreholes.

The results have been presented in Figs. 2–11. The lead content in both organic and inorganic matter, and in coaly shales (in ppm) has been placed in the corresponding part of a circle diagram, and the proportional contribution of these substances in the formation

[2] These materials are presented in borehole geological documentations in the Archives of the Polish Geological Institute, Upper Silesian Branch.

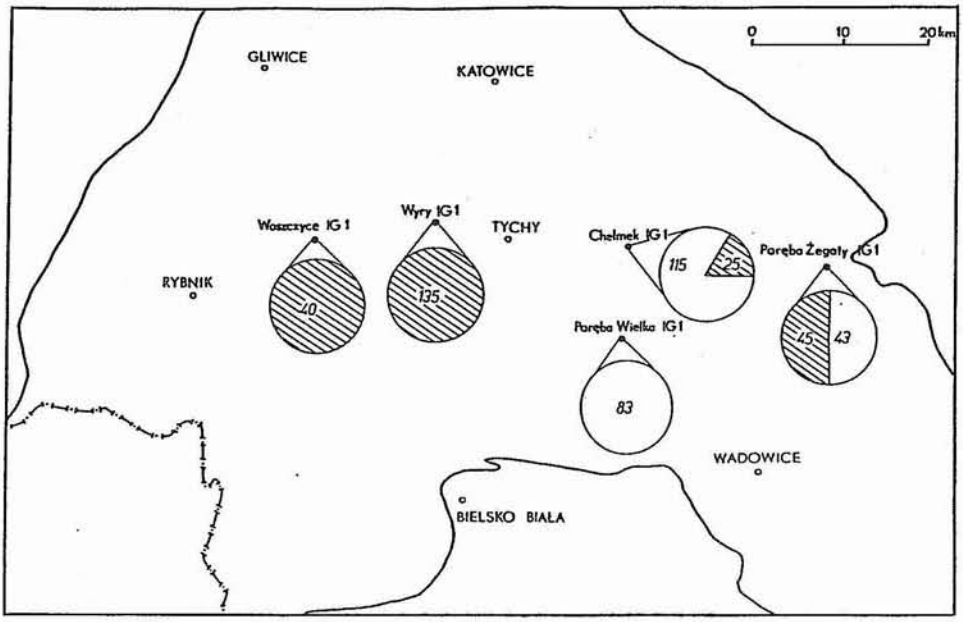

Fig. 3. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Łaziska Beds

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw łaziskich

Objaśnienia jak na fig. 2

of average lead content in coal and coaly shales (100%) has been marked with hatching on the diagram, assuming the whole circle area as a 100% content. Considering the great distance between the boreholes and the well-known great changeability of measured values in coal seams, isolines of these values have been not drawn.

Table 1

Average lead content in coal and coaly shales of the Upper Silesian Coal Basin related to organic and inorganic matter

Series	Average content in ppm (100%)	Proportional control of lead accumulation	
		organic matter	inorganic matter
Limnic (west)	27.5	12	88
Limnic (east)	39.4	0	100
Paralic (west)	29.8	39	61
Paralic (east)	25.4	0	100

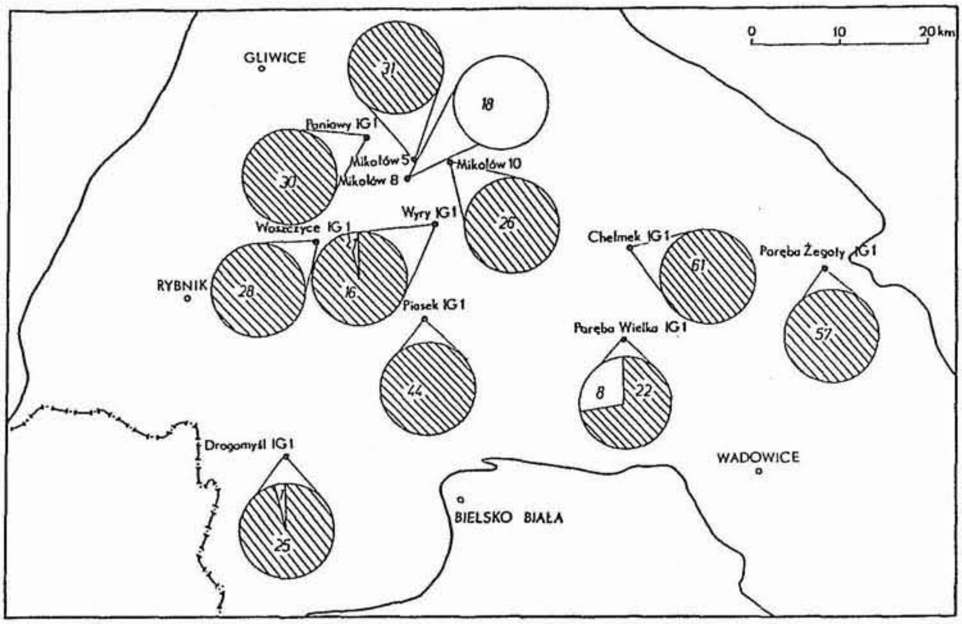

Fig. 4. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Orzesze Beds

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw orzeskich

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GEOCHEMICAL INTERPRETATION OF THE RESULTS

In the paralic series, lead content in coal and coaly shales is higher in the western part of the Upper Silesian Coal Basin. In the limnic series it is lower in the same part of the basin (Table 1). In both these facies, total lead quantity (in the eastern part), or the majority of it (in the western part), is of inorganic origin. Organic matter played a subordinate role in lead cumulation only in the western part of the Upper Silesian Coal Basin. The contribution of this matter to lead concentration in coal and coaly shales was bigger in the paralic facies than in the limnic one.

Two facts show the extensive dynamics of lead accumulation and dispersion. They are: (1) the great differentiation of lead of organic and inorganic origin in individual beds of the Upper Silesian Coal Basin and (2) the varying influence of both organic and inorganic matter on the average lead content in coal and coaly shales (Figs. 2–11). In the Visean sediments and in the beds: Petřkovice, Hrušov and Poręba, inorganic lead occurs mostly in coal and coaly shales in the external part of the area covered by these units. Organic lead occurs mostly in the internal part (Figs. 8, 10, 11). In the Jaklovec Beds, excluding the areas of Czechowice and Poręba Żegoty, organic matter is a main carrier

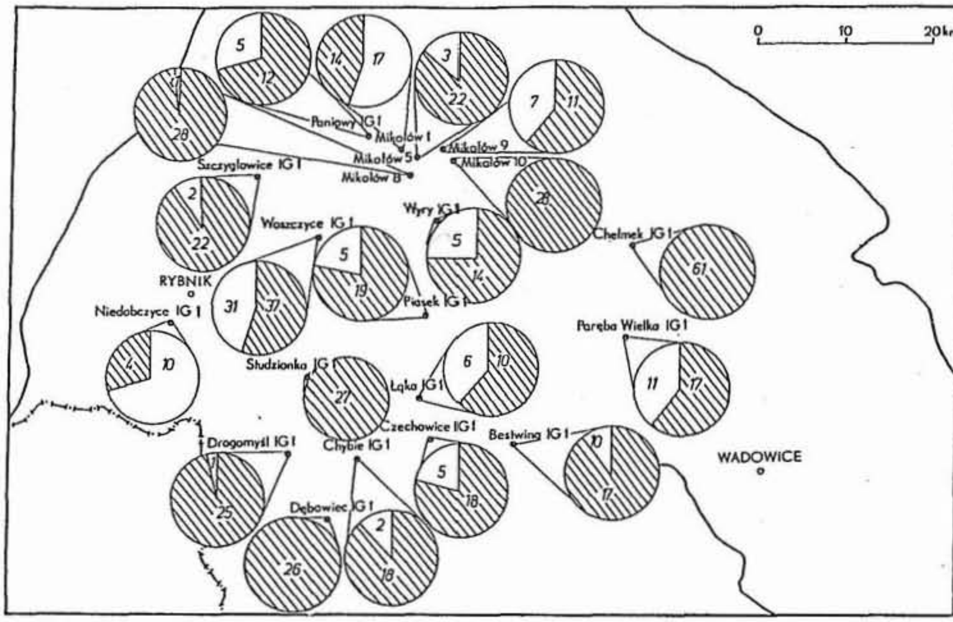

Fig. 5. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Załęże Beds

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw załęskich

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of this element (Fig. 9). The content of inorganic lead also increases regularly in the coal and coaly shales of limnic facies of all the beds from the Anticlinal Beds to the Libiąż one (Figs. 2–7). Lead related to both the organic and inorganic matter only occurs in the Łaziska Beds in the eastern part of this area (organic lead) and in the western one (inorganic lead).

ORGANIC LEAD

Differentiation of organic lead content in coal and coaly shales has been related in a few cases to the volume of peat-forming plants and their differing abilities to absorb chemical components of this element. However, the papers of some authors were analysed during preparation of this paper from this point of view. There were papers concerned with (1) the origin of the formations of the Upper Silesian Coal Basin and transport directions of clastic material (R. Gradziński et al., 1961; *Karbon Górnośląskiego Zagłębia Węglowego*, 1972; K. Bojkowski, 1978), (2) the distribution of volcanic and magmatic centres in the Upper Silesian Coal Basin and its close vicinity during the time of formation of coal seams and coaly shale beds (J. Borowski, 1968; S. Bukowy, S. Cebulak, 1971; F.

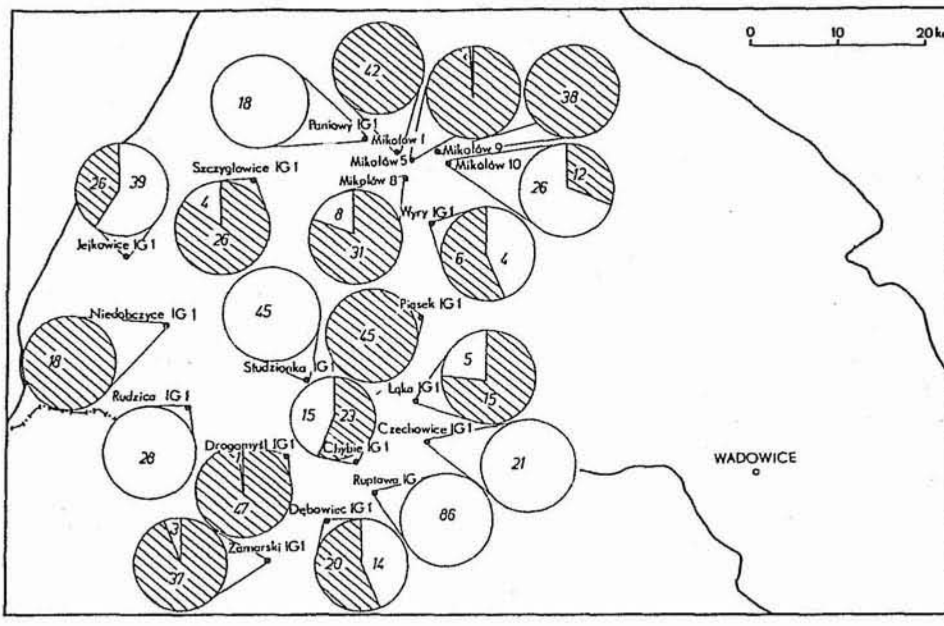

Fig. 6. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Ruda Beds

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw rudzkich

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Ekiert, 1971*a*; O. Juskowiak, 1971; H. Pendias, 1971; W. Ryka, 1971, 1974) and (3) the occurrence of rocks rich in lead-bearing minerals in the area of alimentation (S. Bukowy et al., 1964*a, b*; K. Piekarski, 1971; F. Ekiert, 1971*b*). This analysis allows us to conclude than some locations, where plants contained more lead than plants in neighbouring areas, could have occured in the Upper Silesian Coal Basin. Higher lead content was caused by absorption of lead-rich solutions. Those locations (Figs. 2–11) could be placed in areas as follows: (1) Dębowiec and Ruptawa — during the formation of the coal and coaly shales of the Upper Visean and the Petřkovice Beds; (2) Cieszyn, Leńcza and Poręba Żegoty — during formation of the Hrušov, and Jaklovec Beds, (3) Drogomyśl and Ruptawa — during formation of the Anticlinal Beds and (4) Chełmek, Poręba Wielka and Poręba Żegoty — during formation of the Łaziska Beds.

Lead adsorption and absorption abilities of coal-forming matter substantially influenced lead content in coal and coaly shales of the Upper Silesian Coal Basin. These abilities depended only partially on the composition of peat-forming Carboniferous phytocoenosis. They depended mostly on pH-value in the peat-forming environment, which was controlled by factors of clastic material accumulation: its petrographical differentiation and transport extent. Alkalization of the sedimentary environment, caused by supplying alkaline terrigenous and volcanic material, could cause an increase of lead

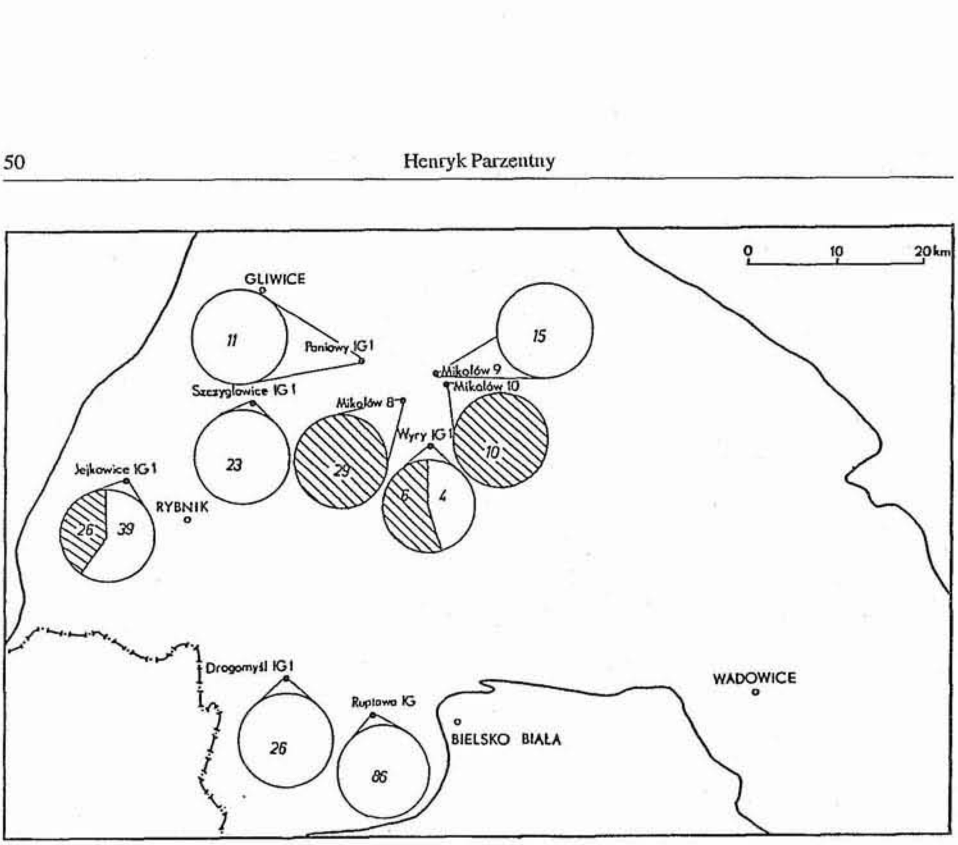

Fig. 7. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Anticlinal Beds

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw siodłowych

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adsorption by plant remains and lead absorption by peat-bog solutions (with the origin of soluble humates and fulvonates). The Kraków — Lubliniec area, containing small occurrences of alkaline rocks, was an alimentary area of terrigenous sediments (S. Bukowy, 1984). These sediments could have been transported during the deposition of the Ruda to the Libiąż Beds (K. Bojkowski, 1978). Moreover, syngenetical alkaline volcanic and magmatic events were observed in the Puńców, Marklowice, Sośnica and Czechowice areas in the rock complex from the Hrušov to the Załęże Beds (C. Gaebler, 1909 and others). Apart from these areas, the organic lead content in coal and coaly shales could also possibly have increased in the Łaziska Beds in the Poręba Wielka, Chełmek and Poręba Żegoty areas (Fig. 3). Surface water and groundwater and also clastic material rich in carbonates supplied from the Ordovician, Silurian, Devonian and Lower Carboniferous rocks of the Cracow — Lubliniec area (S. Bukowy, 1984), could also alkalize an environment. This process probably acted at the time of increasing of sedimentation rate, i.e. during the deposition of the rock complex from the Ruda to the Libiąż Beds (K. Bojkowski, 1978).

The activity of the processes mentioned above probably caused the increase of organic lead content in coal and coaly shales mostly in the eastern and south-eastern parts of the Upper Silesian Coal Basin and, particularly, in the Chełmek area (Figs. 2–6).

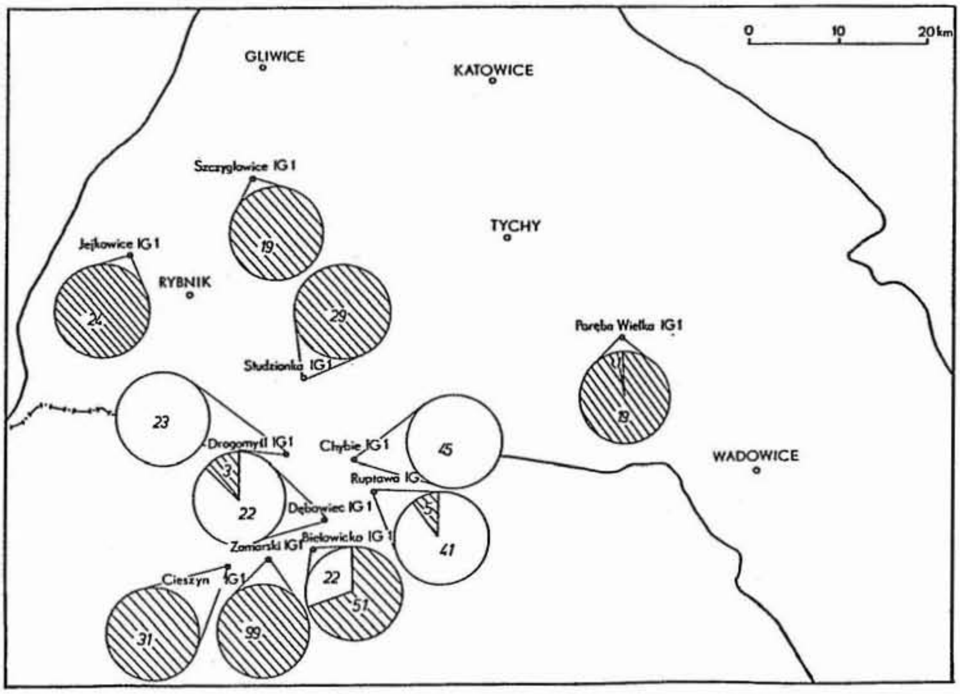

Fig. 8. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Poręba Beds

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw porębskich

Objaśnienia jak fig. 2

The significance of sorption for lead concentration in coal and coaly shales is difficult to define uniquivocally after the data processing of Figs. 2–11. In the areas of the coal basin where organic lead was supplied extensively, processes of inorganic lead accumulation were active at the same time. Mostly lead desorption probably acted in the peat-forming basin, because peat solutions usually showed acidic reaction. In those conditions, lead could became a component of authigenic minerals forming during the stage of diagenesis of coal-forming material or it could be sorbed by auto- and allogenic minerals common in peat-forming matter.

INORGANIC LEAD

The occurrence of inorganic lead in coal and coaly shales depended mostly on the extent of the supply of lead-rich clastic material. However, alkaline material, supplied in small quantity only, occurs in the Upper Silesian Coal Basin relatively rarely. Acidic sediments are the main components of coal measures. After J. Judović (1978), lead-bear-

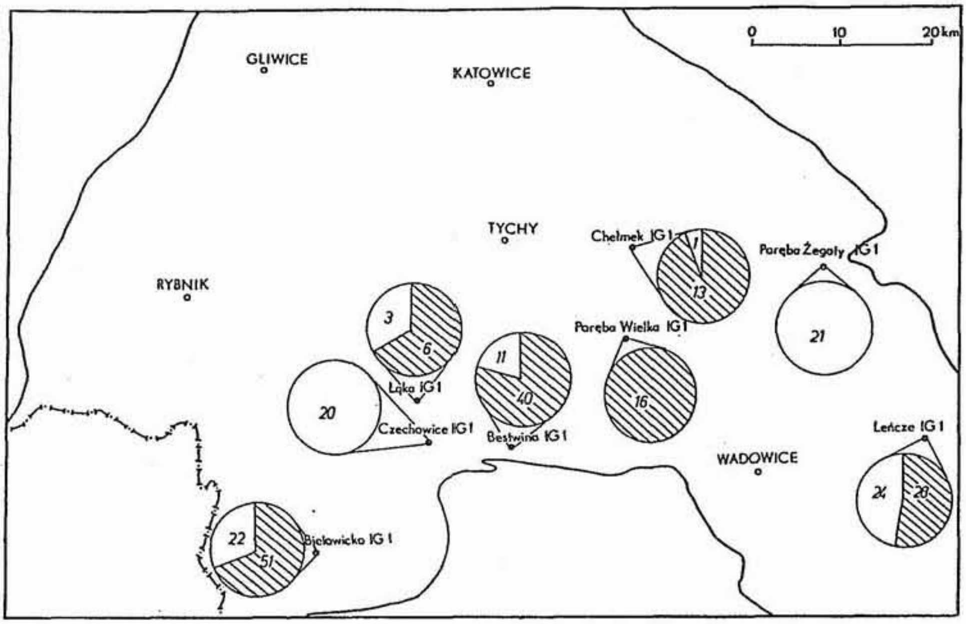

Fig. 9. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Jaklovec Beds

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw jaklowieckich

Objaśnienia jak na fig. 2

ing minerals, both of the rock-forming and ore-bearing groups, could be partly dissolved or be preserved in the barren intercalation in authigenic form in the peat-bog of acidic reactions. After the known data, these minerals enter into the composition of the rocks at the margin of the Upper Silesian Coal Basin in the Kraków — Lubliniec area (S. Bukowy et al., 1964*a*, *b*; K. Piekarski, 1971; F. Ekiert, 1971*b*). If transport directions of clastic material after K. Bojkowski (1978) have been fair, an important part of the lead in coal and coaly shales of the rock complex from the Ruda to the Libiąż Beds (in contact with the area mentioned above) is of inorganic origin. It is important to remember that part of the inorganic lead could be sorbed by the peat-forming plants (J. Judović, 1978) and the quantity of sorbed element depended on the pH-value of peat-bog solutions. After known results of investigations, coal and coaly shales occurring near the boundary of the extent of the coal measures could contain lead of volcanic origin.

A significant part of lead in coal and coaly shales of the Upper Silesian Coal Basin is inorganic-diagenetic in origin. This element originated in the process of bioaccumulation and was contained in organic matter, and that part sorbed by organic matter, was connected probably in chemical complexes forming humates, fulvonates and other organic compositions. After common opinion, an important part of the organic/metallic connections were

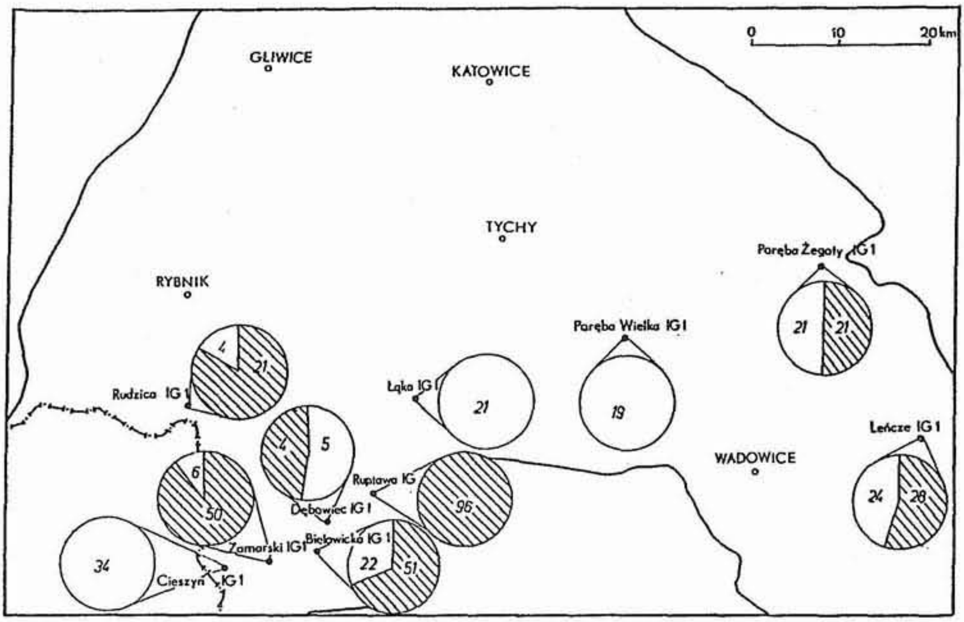

Fig. 10. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Hrušov Beds

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw gruszowskich

Objaśnienia jak na fig. 2

destroyed during the geochemical stage of coal origin (P. Zubović et al., 1960). Liberated ions of trace elements met gases and solutions rich in carbon oxides, sulphur ions and carbonates and they could have formed authigenic carbonate and sulphide minerals in favourable conditions. Pseudomorphoses of sulphides after carbonates and of carbonates after sulphides, described by M. Smyth (1965) and recently also by the author (H. Parzentny, 1992), originated in the same circumstances. Clay minerals able to sorb lead from circulating water solutions, also originated during coalification (G. W. De Vore, 1959; D. Carrol, H. C. Starkey, 1960; A. C. Newham, G. Brown, 1966; A. Bolewski et al., 1970). Diagenetic lead-bearing minerals are, at present, most frequent in all the formations in the western part of the Upper Silesian Coal Basin and in the Marginal Beds in the eastern part of the basin.

ORGANIC AND INORGANIC LEAD

The migration of lead-rich solutions was a main factor controlling the content of organic and inorganic lead in coal and coaly shales. This process probably acted during the whole period of formation of coal seams and coaly shales beds (up to this time). It

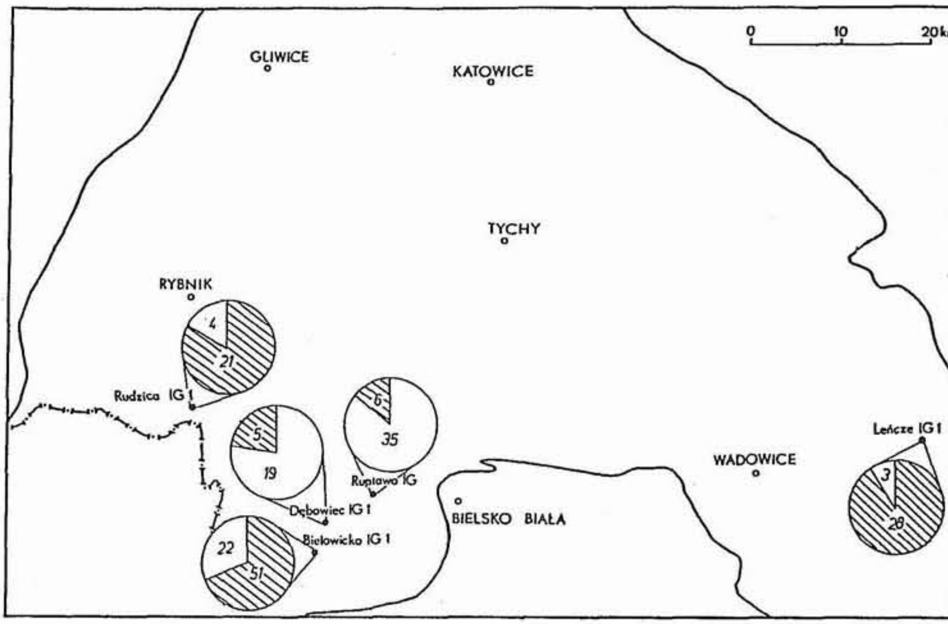

Fig. 11. Lead content in organic and inorganic matter and proportional contribution of these substances to control of average content of this element in coal and coaly shales of the Petřkovice Beds and Visean sediments

Explanations see Fig. 2

Zawartość ołowiu w substancji organicznej i nieorganicznej oraz procentowy udział tych substancji w kształtowaniu średniej jego zawartości w węglu i łupkach węglowych warstw pietrzkowickich i wizenu

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acted in the whole coal basin, however, particularly in the fault zones. The solutions of a buried peat-bog containing lead compositions could change their pH several times during the penetration of rock series with coal seams and intercalations (J. Judović, 1978). The character of these changes was influenced by the chemical composition of the coal-bearing and overlying series and it controlled part of the processes of sorption and desorption as a way of lead cumulation in organic and inorganic matter. This process acted without distinct extreme of intensity (Figs. 2–11).

The solutions migrating during and after formation of lignite and hard coal either disolved lead and other elements or caused the accumulation of mineral agglomerations. After common opinion, ore and carbonate epigenetic mineralization was most significant in the eastern part of the Upper Silesian Coal Basin. E. Jensch (1887) first drew attention to the supposed character of this mineralization of coal-bearing formations and their enrichment in lead. However, analysis of Figs. 2 and 3 did not confirm these conclusions. This result is probably caused by too low a frequency of measuring points (boreholes). After the author's observations, the other data class showed epigenetic mineralization is of an origin related to the Triassic carbonate-zinc-lead sediments in the Cracow sandstone series and — more rarely — in the underlying series, too. The neighbourhood of ore-bearing rock bodies (Olkusz area) and the hydraulic connection of the coal basin with these ore deposits played the main part in this process. Carbonate-ore-bearing mineral associations observed in coal contain a great quantity of zinc, lead and other trace elements (H. Parzentny, 1992).

The absence or presence of the Tertiary overburden and horizontal differentiation of the coal-bearing series controlled further migration of water solutions across the coal-bearing formation. The first relation, this connected to an overburden has not been noticed in the presented material due to the lack of data from the Main Anticline (uncovered hydrogeologically) and from the area placed further to the north. However, organic lead is more frequent in coal and coaly shales in highly permeable rock series (Figs. 3–7). Differentiation of organic and inorganic lead content in studied coal and coaly shales seems not to be related to vertical and horizontal hydrochemical zonality presented by A. Różkowski and K. Przewłocki (1985). However, such a relation has been observed for chlorine and phosphorus distribution (A. Różkowska, 1987; A. Różkowska, H. Parzentny, 1990).

Reduction/oxidation processes have concentrated or diluted undefined parts of lead. These processes were activated after erosion of the overburden and coal-bearing series and as the result of mining activity. Many examples of the oxidation of sulphides to sulphates influenced by oxygen (J. Kubisz, 1964; J. Kuhl et al., 1970; W. Gabzdyl, A. Kopiec, 1970), or the activity of bacteria of genus *Thiobacillus* (B. Cwalina, Z. Zawada, 1988; B. Cwalina et al., 1989; B. Cwalina, Z. Dzierżewicz, 1989) are known in the Upper Silesian Coal Basin. This process, mainly active on a local scale only, usually caused the dispersion of lead.

CONCLUSIONS

1. Lead in coal and coaly shales of the Upper Silesian Coal Basin is mostly of inorganic origin.

2. Lead content in coal and coaly shales of the paralic series is higher in the western part of the Upper Silesian Coal Basin, and of the limnic series is higher in the eastern part of the basin.

3. Quantity of organic and inorganic lead in the individual beds of the Upper Silesian Coal Basin is strongly differentiated. Great changeability in the influence of organic and inorganic matter on average lead content shows the high dynamics of processes of lead accumulation and dispersion.

4. Sorption of lead by phytogenic matter at the peat and lignite coalification stages and desorption of lead by infiltrating water were the two main processes responsible for

present organic lead content in coaly and coaly shales. A part of the lead inorganic components has been supplied by terrigenous material and a part of them is of dia- and epigenetic origin.

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ROZMIESZCZENIE OŁOWIU W WĘGLU I ŁUPKACH WĘGLOWYCH GÓRNOŚLĄSKIEGO ZAGŁĘBIA WĘGLOWEGO

S t r e s z c z e n i e

Badaniami objęto 1254 próbki węgla kamiennego i 28 próbek łupków węglowych z 28 otworów wiertniczych, wykonanych na obszarze GZW (fig. 1) w latach 1975–1988 przez Oddział Górnośląski PIG w Sosnowcu. W popiele tych próbek oznaczono zawartość ołowiu metodą fluorescencji rentgenowskiej. Przez rozwiązanie równania wyrażającego zależność zawartości pierwiastka w popiele od zapopielenia węgla i łupków węglowych określono jaka część średniej zawartości ołowiu pochodzi z substancji organicznej, a jaka z nieorganicznej. Obliczenia wykonano dla całego zbioru danych reprezentujących zagłębie z podziałem na facje, na część wschodnią i zachodnią (tab. 1) oraz oddzielnie dla każdej grupy warstw (fig. 2–11).

Interpretacja wyników prowadzi do stwierdzenia, że w GZW występuje ołów pochodzenia głównie nieorganicznego. Jego zawartość w węglu i łupkach węglowych serii paralicznej jest większa w części zachodniej niż w części wschodniej GZW, zaś w serii limnicznej — większa w części wschodniej niż zachodniej. Stwierdzono duże zróżnicowanie ilości ołowiu pochodzenia organicznego i nieorganicznego w obrębie poszczególnych warstw, a także duże zróżnicowanie wpływu substancji organicznej i nieorganicznej na zawartość tego pierwiastka w węglu i w łupkach węglowych, świadczące o dużej dynamice procesów gromadzenia się i rozpraszania ołowiu. Różnice zawartości po rozciągłości warstw są zbieżne z rozkładem działania procesów o największej intensywności gromadzenia bądź rozpraszania ołowiu. Obecna zawartość ołowiu pochodzenia organicznego jest wynikiem głównie sorpcji tego pierwiastka przez materiał pochodzenia roślinnego w stadium torfu i węgla brunatnego, a także desorpcji ołowiu pod wpływem wód infiltrujących. Z kolei nieorganiczne związki omawianego pierwiastka częściowo dostarczył materiał terygeniczny, a częściowo są pochodzenia dia- i epigenetycznego.