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VARIABILITY OF MERCURY CONTENT IN COAL MATTER FROM COAL SEAMS OF THE UPPER SILESIA COAL BASIN

ZMIENNOŚĆ ZAWARTOŚCI RTĘCI W MATERII WĘGLOWEJ POKŁADÓW GZW

The process of identifying and documenting the quality parameters of coal, as well as the conditions of coal deposition in the seam, is multi-stage and extremely expensive. The taking and analyzing of seam samples is the method of assessment of the quality and quantity parameters of coals in deep mines. Depending on the method of sampling, it offers quite precise assessment of the quality parameters of potential commercial coals. The main kind of seam samples under consideration are so-called "documentary seam samples", which exclude dirt bands and other seam contaminants. Mercury content in coal matter from the currently accessible and exploited coal seams of the Upper Silesian Coal Basin (USCB) was assessed. It was noted that the mercury content in coal seams decreases with the age of the seam and, to a lesser extent, seam deposition depth. Maps of the variation of mercury content in selected lithostratigraphic units (layers) of the Upper Silesian Coal Basin have been created.

Keywords: coal matter, Upper Silesia Coal Basin, seam samples, mercury

Użytkowanie węgla związane jest z emitowaniem do atmosfery wielu zanieczyszczeń. Jednym z nich, szczególnie niebezpiecznym dla zdrowia i życia ludzi, są związki rtęci. Zanieczyszczenie to szczególnie łatwo rozprzestrzenia się w środowisku i nie ulega biodegradacji. Ocenę jakości węgla prowadzi się na etapie wytwarzania produktów handlowych węgla i ich użytkowania, jak i na etapie przygotowania i prowadzenia eksploatacji. Przykładem tego drugiego jest pobieranie i badanie próbek bruzdowych z pokładów węgla. W zależności od zastosowanych metod ich pobierania dają one możliwości bardziej lub mniej dokładnej oceny parametrów jakościowych, możliwych do uzyskania węgli handlowych. W pracy analizowano zawartości rtęci w pokładach GZW, bazując na standardowo pobieranych przez służby geologiczne kopalń węgla kamiennego, próbkach pokładowych dokumentacyjnych. Są to próbki materii węglowej zawartej w pokładach węgla. Pozyskane próbki poddano kolejnym stadiom rozdrabniania i pomniejszania, aż do uzyskania próbek analitycznych według normy (PN-G-04502:2014-11, 2014). W próbkach analitycznych, poza standardowym zakresem analiz, oznaczono dodatkowo zawartości rtęci. Każda pobrana próbka posiada przyporządkowane współrzędne X, Y, Z, określające miejsca jej pobrania. Pozyskane dane wykorzystano do wyznaczenia zależności statystycznych oraz zaproponowano mapy

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zmienności zawartości rtęci w materii węglowej grup pokładów, opracowane za pomocą oprogramowania SURFER®. Rozkład zawartości rtęci w uzyskanych próbkach przedstawiono graficznie w postaci histogramu na rysunku 2. Rozkład danych jest niesymetryczny i charakteryzuje się dodatnią asymetrią. Obliczona wartość średnia zawartości rtęci wynosi 52,5 µg/kg, a wartości dolnego i górnego kwartyla wynoszą odpowiednio 23 i 70 µg/kg. Na rysunku 3 przedstawiono rozrzut zawartości rtęci w zależności od wartości współrzędnej Z (głębokość pobrania próbki), a na rysunku 4 zobrazowano rozrzut zawartości rtęci w zależności od podziału litostratygraficznego karbonu produktywnego GZW, wyrażonego przez numery pokładów. Na obu rysunkach widoczna jest tendencja zmniejszania się zawartości rtęci w materii węglowej zawartej w pokładach wraz z przechodzeniem do starszych warstw i ogólnie głębiej zalegających, chociaż głębokość zalegania pokładów nie jest pochodną wyłącznie wieku pokładów węgla. W obu wypadkach wartości współczynnika R^2 są małe i nie upoważniają do wykazania istnienia zależności statystycznej. Następnie obliczono średnie wartości zawartości rtęci w materii węglowej zawartej w poszczególnych warstwach – grupach pokładów, a wyniki obliczeń zestawiono w tabeli 1, i na rysunku 5. Dane z tabeli 1 i rysunku 5 uwypuklają tendencje zmian zawartości rtęci w próbkach pokładowych w zależności od głębokości pobrania próbek i budowy litostratygraficznej karbonu produktywnego GZW. Obliczona wartość współczynnika korelacji pomiędzy średnimi zawartościami rtęci w poszczególnych grupach pokładów GZW, a numerami grup pokładów, będących liczbowym odwzorowaniem ogniw litostratygraficznych GZW, jest duża i wynosi 0,86. Populację wyników zawartości popiołu w analizowanych próbkach pokładowych bruzdowych dokumentacyjnych, przedstawiony na rysunku 6, w postaci histogramu. Obliczona wartość średnia zawartości popiołu wynosi 7,64%, a wartości dolnego i górnego kwartyla wynoszą odpowiednio 4,44% i 8,90%. Na rysunkach 7 – 10 przedstawiono prawdopodobne przebiegi izolinii zawartości rtęci w materii węglowej w pokładach warstw libiąskich i łaziskich (Rys. 7), orzeskich (Rys. 8), rudzkich (Rys. 9) i siodłowych (Rys. 10).

Słowa kluczowe: materia węglowa, GZW, próbki pokładowe, rtęć

1. Introduction

Coal is the pillar of energy security in many countries of the world. Despite the fact that coal has the most negative environmental impacts of all the fossil fuels, its utilization is actually increasing at the highest rate in comparison to that of crude oil and natural gas (BP Statistical Review, 2014). In many countries, coal usage for electricity generation dominates the usage of other primary energy carriers. For example in the USA, around 38% of electricity is produced from coal; in Germany, 46%; in China, 76%; in India, 71%; in Australia, 69%; in Poland, around 84%, and in South Africa, as much as 93% (IEA Statistics 2014 a, b).

The catalogue of environmental contaminants emitted during the utilization of coal, addressed by preventative measures, is already quite extensive and is still growing. A contaminant often discussed in recent years is mercury (UNEP 2010, http://www.mercuryconvention.org/Convention/tabid/3426/Default.aspx). This is due to the threat posed to human health and life by the presence of mercury in the environment (Mercury Study Report to Congress, 1997), this is particularly concerning because mercury compounds are not naturally biodegradable. Moreover, this contaminant easily spreads throughout the environment and the utilization of coal largely contributes to the anthropogenic mercury emissions to the atmosphere on a global scale (UNEP, 2013).

Identification of the types and levels of contaminant content in coal is a multi-stage process, particularly in the case of deep mines. During classification of a coal deposit, it is also important to identify the types and concentrations of contaminants present in coal seams. Once exploitation has started, the contaminants contained in the coal, including mercury, are analyzed and assessed at several levels with increasing accuracy. Seam samples and technological samples of the raw coal are taken and analyzed. The obtained information is used for multiple purposes including informing policy on reduction of emissions of different pollutants. An example of the use of such information is *fuel switching* (UNEP, 2010, Deborah et al., 2006), proven in the case of the reduction of SO_x emissions.

Technological analyses of raw coal, aimed at the assessing the suitability of certain cleaning processes to the coal, allow prediction of the extent to which levels of various contaminants in commercial coals can be decreased through cleaning. The cleaning processes allow for, among other things, reducing the mercury load carried by coal to its users. Literature data show that this effect varies depending on the technological characteristics of the raw coal (Pavlish, 2003; Das et al., 2013; Dziok et al., 2014; Ozbayoglu, 2011; Ozbayoglu, 2013; Pyka et al., 2010; Quick et al., 2002; Mastalerz et al., 2005; Wang et al., 2009; Wang et al., 2006a; Wang et al., 2006b; Swaine, 2000; Huggins et al., 2009).

Mercury content in the used coal is the first factor which determines the mercury emissions to the atmosphere (UNEP, 2010) as well as the type and cost of appropriate preventive measures. Both the mercury emissions to the atmosphere and the associated preventive measures are currently the objects of regulatory, analytic, research and implementation works (Sloss, 2012, http://www.mercuryconvention.org/Convention/tabid/3426/Default.aspx, http://www.mos.gov.pl/artykul/7_archiwum/23417_polska_podpisala_konwencje_w_sprawie_rteci.html).

It is crucial that information on the mercury content of the coal corresponds to the commercial coal. The use of some other information, for example the mercury content of coal seam samples, can lead to impractical conclusions (Pyka et al., 2015). Information on coal contaminants, obtained at various stages of the coal life cycle, have to be adequately interpreted and used. In the case of analyses of mercury contamination of coal seams, the contaminant content of the raw coal is not necessarily representative of the environmental threats posed by the emissions from burning the coal. The reason for this could be the fact that the values indicating the amount of mercury in coal seams are higher than the values of the amount of mercury in commercial coal products. We are speaking here on the impact of coal cleaning. In the literature, remarks can be found that suggest the application of coefficients which decrease the mercury content determined during the coal seam examination stage, before the obtained information is used for the assessment of the mercury emission threat. The coefficients should reflect the degree to which the mercury content in the coal is reduced during coal cleaning (Demir et al., 1998; Toole-O' Neil et al., 1999). The above occurs in the analyses of coal seams throughout their entire cross sections together with the contaminants in the seams. Seam samples can be taken by various methods and for various purposes which determine the quality of the material in the sample including the levels of contaminants.

This article presents the results of an assessment of the mercury contamination in coals exploited in Poland in the Upper Silesian Coal Basin based on the results of analyses of routinely taken seam samples. This analysis is limited to the USCB area where mainly steam coals are exploited. Identification of the degree of mercury contamination and its changes from selected USCB coal seams was conducted with respect to its lithostratigraphic structure. The described works are based on the results of analyses of 315 unit documentary channel seam samples. A graphical representation (map) of the differentiation of this contaminant has been prepared based on specialist modelling and computer software. Particular attention was paid to ensuring the representativeness of the information on mercury content in the coal seam samples, which is the derivative of the applied technique, and in particular, of the methodology of sampling. The term "coal matter" will henceforth refer to only clean coal matter from a coal seam and is not representative of the material quality of the whole seam.

2. Scope and methodology of research

The object of this analysis is information on the mercury content of the “coal matter” of coal seams in active steam coal mines of the USCB. The information was obtained from the analysis of the documentary channel seam samples routinely, and in large numbers, taken in the USCB collieries. Samples were taken in the frames of the standard sampling design used in the respective collieries and the scope of the analyses was broadened by the non-routinely analyzed mercury content. Details on standards, and the methods of sampling of coal seams described in them, are given in previous work (Pyka et al., 2015). All samples were taken in 2014. The mining areas from which the seam samples originate are shown in Figure 1.

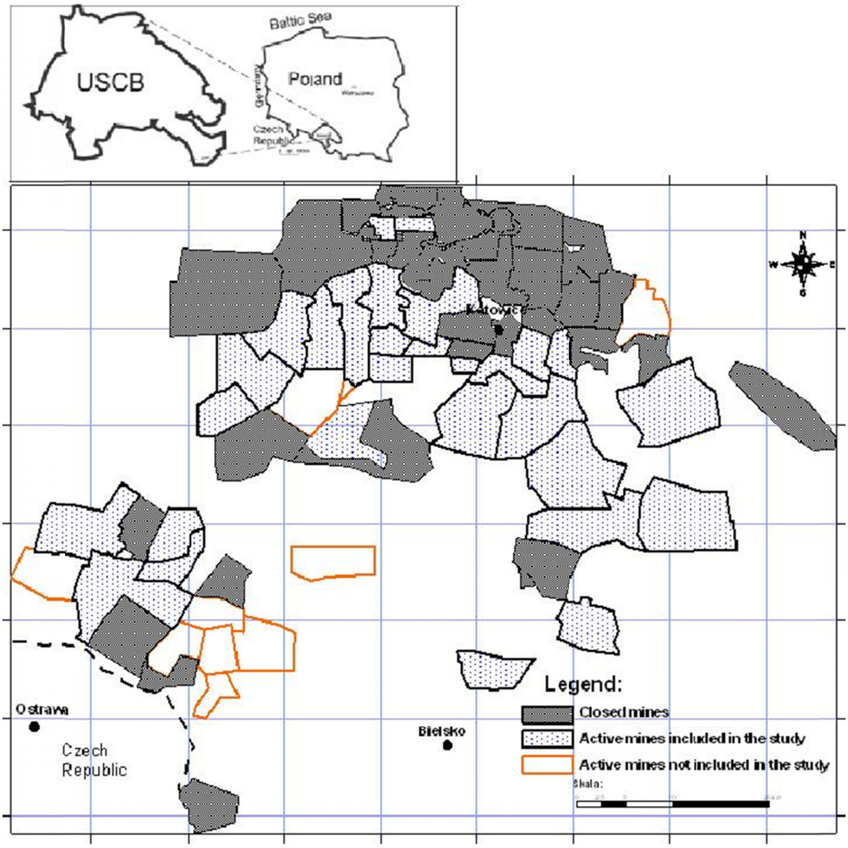

Fig. 1. Map of the Upper Silesian Coal Basin upon which the analyzed mining areas are marked (Own elaboration based on Jureczka et al., 2009)

Samples were taken from the collieries producing steam coal of the type 31-34 according to the Polish standard (PN-82/G-97002 1982). The 31-34 coal types, according to the above mentioned standard, are the counterparts of the subbituminous and bituminous coals with high volatile matter content (high volatile bituminous) defined as subbituminous as well as para- and ortho-bituminous according to the international classification (Economic Commission for Europe, 1998). The samples were taken from seams mined during the sampling or made accessible in such a way that the channel samples were taken in the frames of the existing seam block development.

Channel samples were subjected to subsequent stages of grinding and reduction in order to obtain analytic samples in compliance with the standard (PN-G-04502:2014-11, 2014). In the analytic samples, besides the standard scope of analyses, the ash and mercury content were determined. These parameters were determined in the laboratories of Główny Instytut Górnictwa (Central Mining Institute – GIG) accredited by the Polish Centre for Testing and Certification. The ash content was determined according to the standard (PN-ISO1171:2002), and the mercury content according to an accredited procedure (SC-1.PB.23, 2012). All the determination results used in this article are provided for the air dried basis of coal fuel.

Each sample was assigned X, Y and Z coordinates which define the sampling locations. The obtained data was processed in the first instance by applying statistical methods using Statistica software to first order and then eliminate gross errors.

Maps of the variation of mercury content in the carboniferous matter of the seam groups were made using the SURFER® software. The software uses the cringing method to determine a regular grid based on geostatistical methods. The input data for the interpretation of the mercury concentration maps were the coordinates X and Y, the locations of the sampling points and the results of mercury content determination. Construction of the maps is intuitively based on a linear variogram model for describing the parameters of mercury content for which the values, depending on the distance and the directions of change are determined. The SURFER® software contains procedures which allow adjustment of the variogram model input data, i.e. location of the sampling points and determination of the mercury content, through interpreting the isoline of the mercury content in the analyzed area.

The variogram characterizes the space of the continuity of the whole data set; this is used for the preparation of isoline maps. The idea of the variogram is based on the assumptions of geostatistics, assuming that the value of a function in the near vincinity is not random but strongly correlated.

3. Results and discussion

3.1. Characteristics of the variation of mercury content in the samples

Initial analysis of the data by the Statistica package indicated that of the set of 315 collected results, three were found to be outliers and were removed. The remaining data are presented graphically in Figure 2. The distribution of the data is characterized by positive asymmetry, indicating the occurrence of a relatively small number of samples with mercury content significantly larger than the mean value of the population. The calculated mean value of mercury content equals 52.5 μg/kg and the values of the lower and upper quartiles equal 23 and 70 μg/kg, respectively.

For the purposes of further analysis and, in particular, for the visualization of data on the maps, a partial averaging of the determination results was performed due to the closeness of the sampling locations of some of the samples. The results of determination and sampling coordinates grouped in an area of about 500×500 m were replaced by one mean value. After the calculations were made, the input data set used in further analyses amounted to 109 values (results of determination and coordinates of the sampling locations).

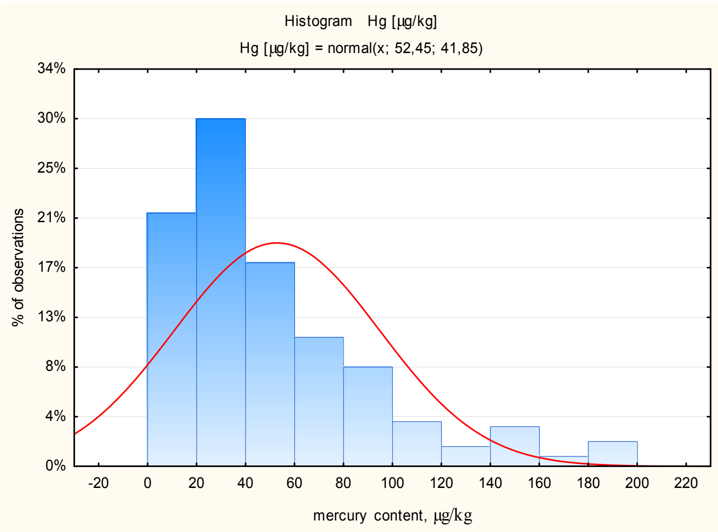

Fig. 2. Histogram showing the mercury content in the analyzed samples from the documentary seams

Figure 3 presents the distribution of the mercury content according to Z coordinate value (depth at which the sample was taken). A certain correlation can be seen showing that a decrease in mercury content in the documentary seam samples occurs alongside an increase in the depth at which the sample is taken (Z coordinate). The data, however, is dispersed and the calculated correlation coefficient, R^2, is very small and does not provide evidence of a significant statistical correlation of the analyzed variables.

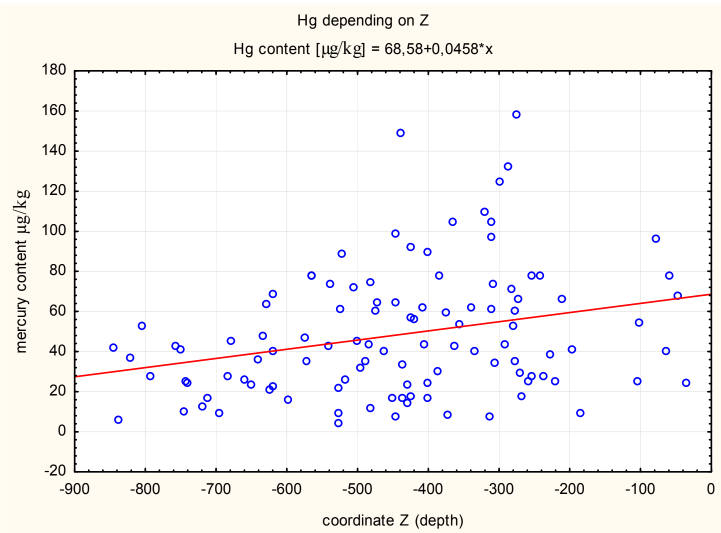

Fig. 3. Scatter plot showing mercury content in the samples versus Z coordinate

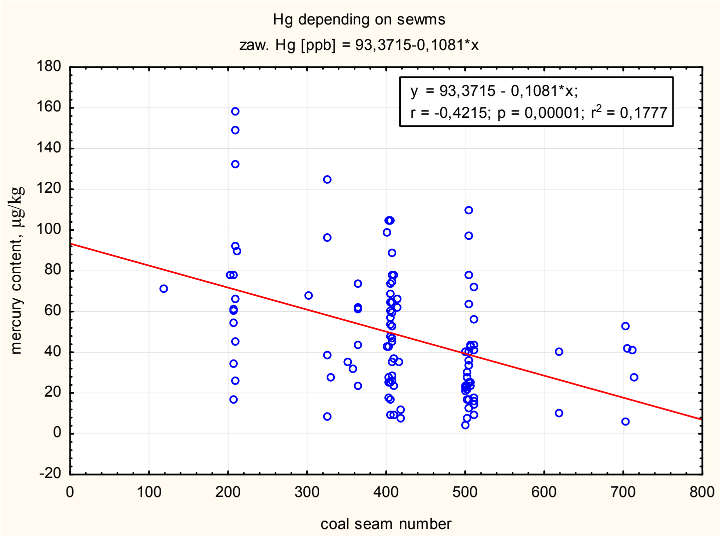

Fig. 4. Scatter plot showing mercury content in different seam groups (Table 1)

Figure 4 shows the mercury content according to the lithostratigraphic division of the productive carboniferous system of the USCB expressed, in this case, through seam numbers. Also in this case, mercury content in the seams can be observed to be lower in older beds. The value of the R^2 coefficient is also low in this case, indicating the absence of strong statistical dependence between the analyzed variables.

In general, it can be stated that together with the increase in seam number, seam age and, most often, depth of deposition also increase. This, however, is not a rule regarding the tectonic disturbances nor other productive carboniferous system disturbances in USCB and therefore the correlations observed in Figures 3 and 4 are weak. High variability of mercury content in the respective beds (seam groups) and respective seams as seen in Figure 4 and wide intervals of the determined mercury content (i.e. 15 µg/kg to 160 µg/kg for seam 209) can also be observed. This also contributes to the weakness of the correlations discussed above.

In a further analysis, the mean values of the mercury content in the respective beds were calculated. The results of these calculations are presented in Table 1. Figure 5 shows the aver-

TABLE 1

Mercury content in the beds (seam groups)

Beds (seam groups)	**Mercury content Hg[a], µg/kg**	**Number of analyzed points**
Libiąż Beds (100)	71,7	1
Łaziska Beds (200)	76,2	16
Orzesze Beds (300)	53,4	13
Ruda Beds (400)	49,3	38
Saddle Beds (500)	35,3	34
Poręba Beds (600)	24,9	2
Jakłowice Beds (700)	33,9	5
Mean value	49,2	—

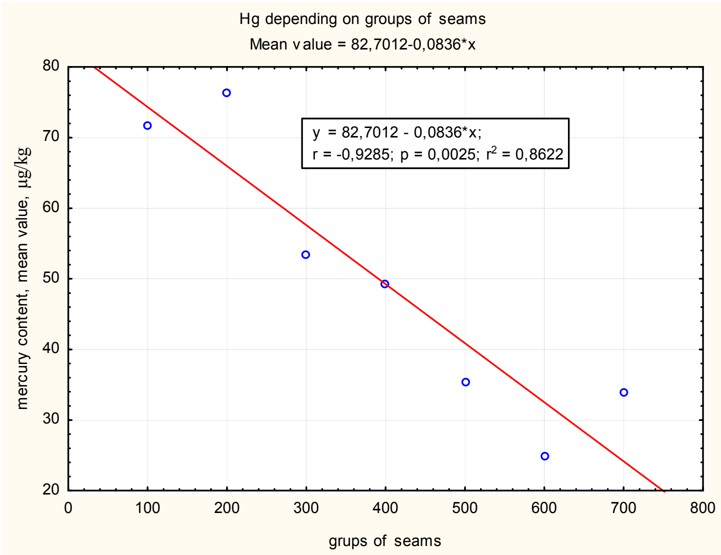

Fig. 5. Scatter plot showing the mean value of mercury content against the beds (seam groups) of the USCB

age mercury content as a function of lithostratigraphic division units (beds) of the productive carboniferous system of the USCB (for which the means were calculated).

Analysis of the data from Table 1 and Figure 5 highlights the trends in the mercury content of the seam samples with sampling depth and lithostratigraphic structure of the productive carboniferous system of the USCB. The calculated value of the correlation coefficient between the mean mercury content in the respective seam groups of the USCB and the numbers of the seam groups (figure mappings of the lithostratigraphic units of the USCB) is high and equals 0.86. Based on the above stochastic relationship, a generalization can be made that average mercury content in the carbonaceous matter of the USCB seams clearly decreases together with the age of the productive carboniferous system deposits of the USCB.

3.2. Characteristics of the variability of ash content in the analyzed samples

Ash content in the analyzed documentary channel seam samples is presented in Figure 6. The distribution of the ash content in the data is, similarly to the distribution of the mercury content, asymmetrical and characterized by positive asymmetry which indicates the presence of a relatively few samples with ash content significantly higher than the mean value. The calculated mean value of ash content equals 7.64% and the values of the lower and upper quartiles equal 4.44% and 8.90%, respectively.

The majority of the samples possessed low ash content – over 80% of the samples had ash content below 10%. Ash content in the documentary channel seam samples can be compared only to the ash content of the fully cleaned coarse and medium-sized coals produced by the steam coal mines in the USCB. These are products with particle size above 20 mm. Average ash content in these coals equals 6% and the maximal value is around 14%. Since the coal in these

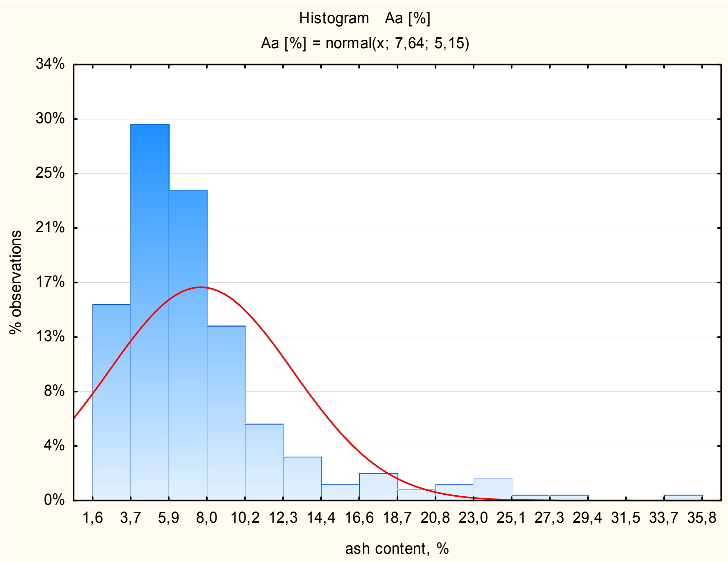

Fig. 6. Histogram showing ash content in the analyzed documentary seam samples

documentary channel seam samples is raw coal, the maximal noted ash content is higher – even up to 30%. Average ash content in the remaining part of steam coal production in the collieries producing these types of coal in the USCB is significantly higher and equals around 19%, with lower and upper quartiles equal to 13% and 27%, respectively. This shows that the pure "coal matter", which we are dealing with in most cases of the documentary channel seam samples, does not fully correspond to the quality of the commercial steam coal products from the USCB mines.

3.3. Maps of the variability of mercury content in the carboniferous matter of the beds (seam groups) of the USCB

Due to the spatial distribution of the data obtained for the respective beds, a common map for the Libiąż Beds (100) and Łaziska Beds (200) and separate maps for the Orzesze, Ruda and Saddle Beds were made. The Libiąż and Łaziska Beds belong to one lithostratigraphic unit i.e. Krakow Sandstone Series. For seams of the Poręba and Jakłowiec Beds no maps were made due to the very small number of sampling points.

Figure 7 presents the probable distribution of mercury content in the "coal matter" in the Libiąż and Łaziska Beds i.e. in the youngest lithostratigraphic unit of the USCB. They are located and currently mined in the eastern part of the USCB. The highest mercury content, equaling around 100 μg/kg to 160 μg/kg, can be observed in the central part of the analyzed beds. Based on interpolation, increased mercury content can be expected in the South-East part of the analyzed area.

Changes in mercury content of the "coal matter" of the Orzesze Beds, located in the central and eastern part of the USCB, are presented in Figure 8. An increase in mercury content in those beds can be observed towards the South, from the lowest values of around 10 mg/kg up to over 100 mg/kg. The highest mercury content can be observed in the western part of the mine deposits as well as in the eastern part of the active mine deposits.

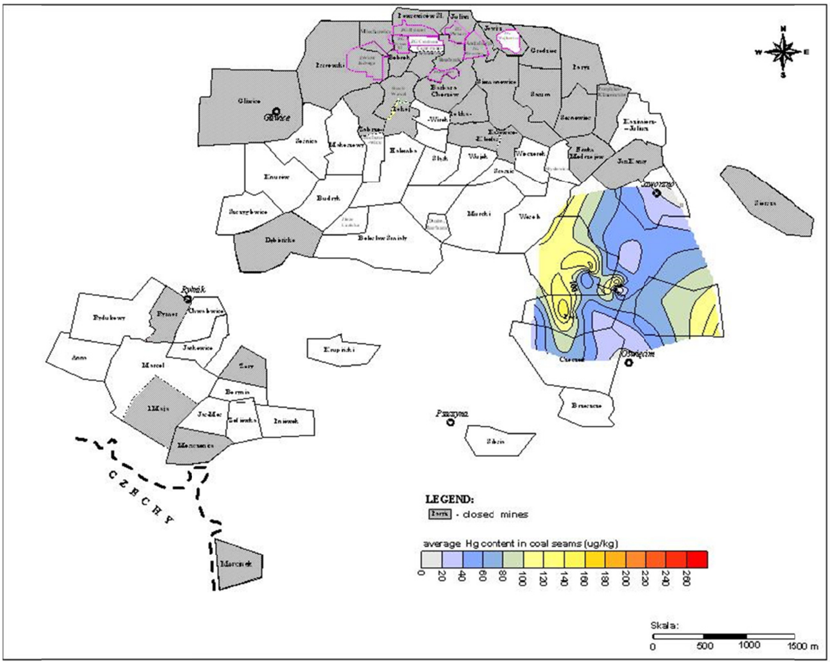

Fig. 7. Map of the variability of mercury content in the Libiąż and Łaziska Beds

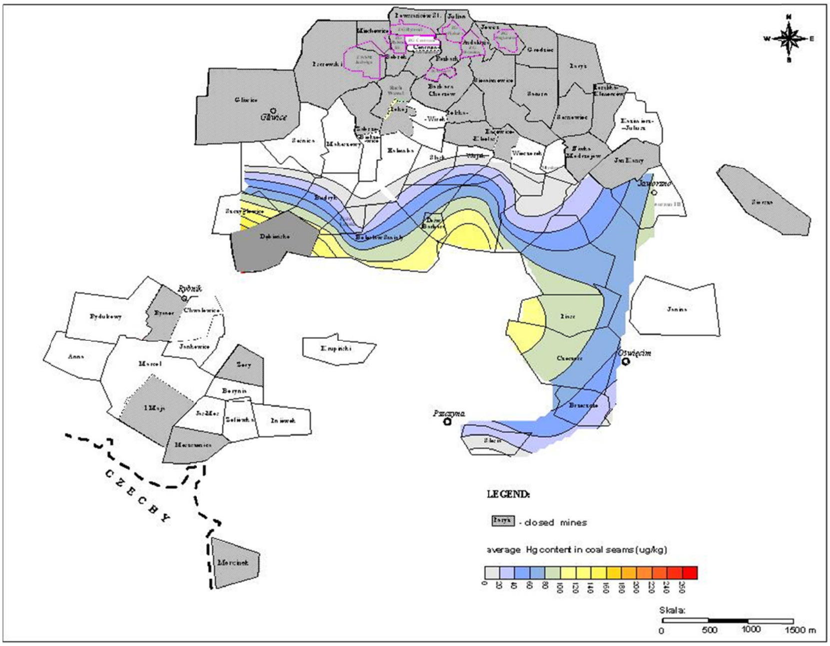

Fig. 8. Map of the variability of mercury content in the “coal matter” of the Orzesze Beds

Average mercury content in the "coal matter" of the mined Ruda Beds equals around 50 mg/kg, with the distribution of values ranging from 10 mg/kg to over 100 mg/kg. On the prepared map (Figure 9) an increase in mercury content can be observed towards the South-West part (SW) of the USCB. In these areas the mercury content in the "coal matter" in the Orzesze beds most probably exceeds 150 mg/kg. In the North-West (NW) area of the map an anomaly, which reaches 120 mg/kg, can be observed; most probably this is the result of an interpolation error. Generally in the North, the mercury content equals around 80 mg/kg.

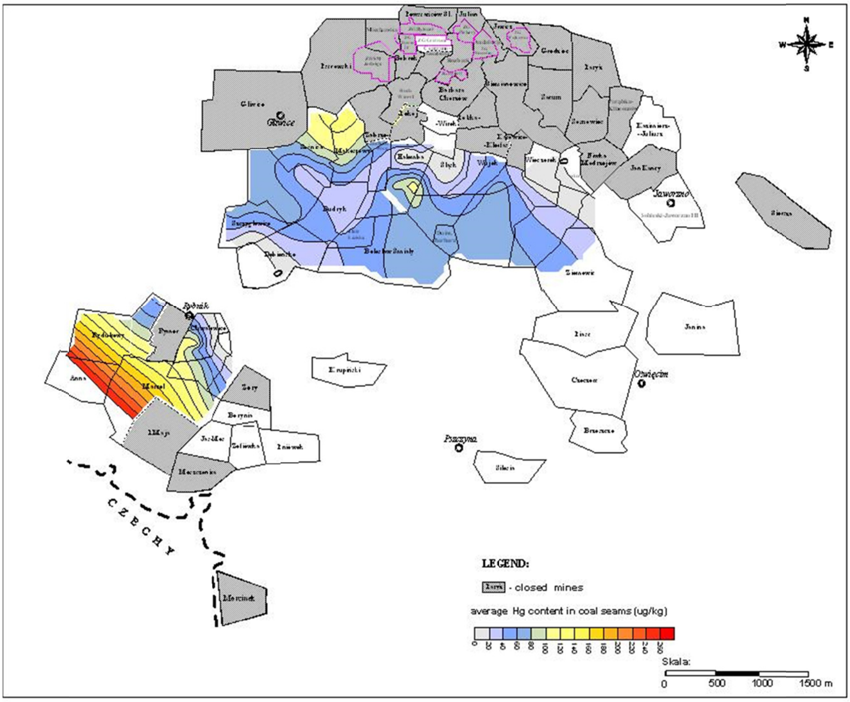

Fig. 9. Map of the variability of mercury content in the "coal matter" of the Ruda Beds

Average mercury content in the "coal matter" of the mined Saddle Beds equals around 35 mg/kg, with the minimal and maximal values equal to 5 mg/kg and over 100 mg/kg, respectively. A map, based on the experimental data, presenting the variability of mercury content in the Saddle Beds is presented in Figure 10. The highest mercury content, reaching up to 100 mg/kg, was observed only locally in the South-West (SW) region of the USCB.

Mercury content in the fully cleaned products of the collieries producing steam coal in the USCB equals around 76 mg/kg, and in raw smalls around 120 mg/kg. Here we are dealing with a different case to that described in the literature (Demir et al., 1998; Toole-O' Neil et al., 1999), in which the mercury content in the analyzed seam samples is equal to or lower than the mercury content in the commercial products comprising the separated coal. This shows that the objective of seam sampling, and thus the sampling technique, and in particular the decision of whether or not to reject the contaminants in the seams influences the assessment of mercury contamination in commercial coal. Since the mercury content in the "coal matter" of the USCB seams is generally low (as confirmed by the prepared maps which are a generalization for large areas of the USCB) it can be expected that the rejected contaminants of the seams, as well as part of the raw coal from the coal cutting with floor and roof ripping, must be characterized by higher mercury content than the "coal matter". This is confirmed by the above presented results of the analyses of mercury content in commercial smalls, which are characterized by relatively high mercury content in comparison to the mercury content of the seam. Raw commercial coal products (uncleaned) must contain contaminants from the exploited coal seams.

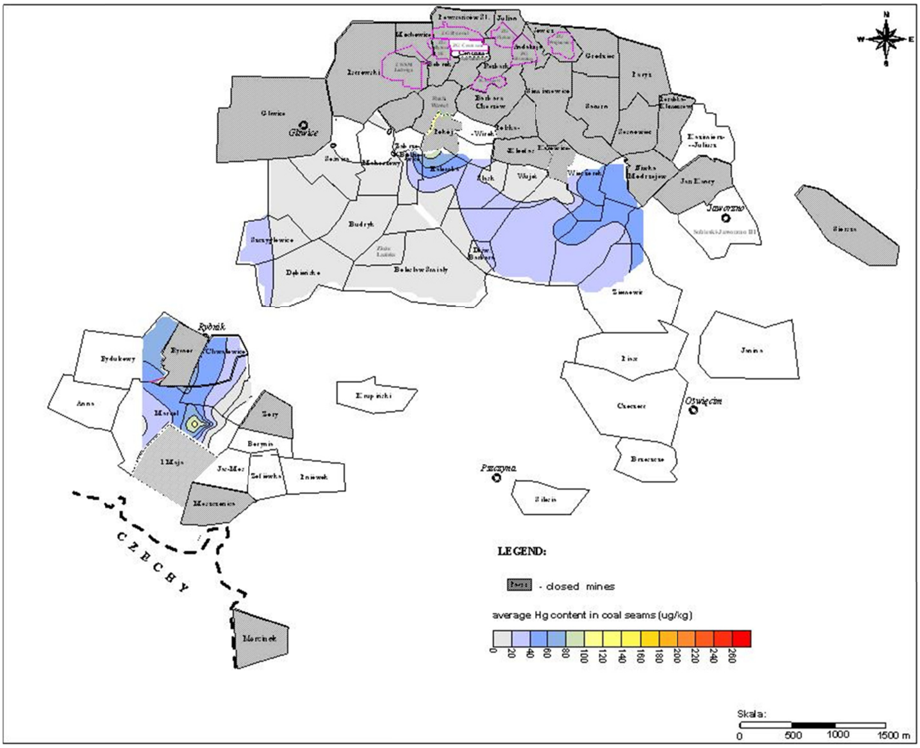

Fig. 10. Map of the variability of mercury content in the "coal matter" of the Saddle Beds

4. Conclusions

1. The documentary channel seam samples most commonly sampled in the USCB mines can be defined as composed in the majority of pure coal matter. This is supported by the low ash content observed in those samples. Average ash content equals 7.64%, with lower and upper quartiles equal to 4.44% and 8.90%, respectively. The distribution of ash content is characterized by positive asymmetry.
2. Average mercury content in the "coal matter" of the Upper Silesian Coal Basin seams is low and equals 52.5 μg/kg, with lower and upper quartiles equal to 23 mg/kg and

70 μg/kg, respectively. The distribution of mercury content is characterized by right-sided asymmetry.

3. Significantly high distribution of mercury content in the respective beds and series (seam groups) of the productive carbonaceous system of the USCB was identified.
4. Analysis of the relationship between mercury content and depth at which the sample was taken, as well as lithostratigraphic division of the productive carbonaceous system of the USCB, expressed through the seam numbers, does not allow any statistical relationships to be defined; however, averaged mercury content in the seam groups, analyzed in relation to the lithostratigraphic division of the USCB expressed in the seam groups, show a strong tendency to decrease with the age of the productive carbonaceous system, as can be observed on the elaborated maps.
5. Interpretation of the results of the determination of mercury content in the mined seams of the productive carbonaceous system of the USCB, through applying the isoline maps and the cringing method allow for a ”smoothing” of the noted large dispersion of the results. From the elaborated maps, it follows that the “coal matter” is characterized in the majority of the analyzed seam areas by a low mercury content and only locally can places with increased mercury content be found.
6. The elaborated maps confirm the decrease in the mercury content in the respective seam groups which is, most probably, related to the sedimentation process in the area of the USCB. The youngest coals are characterized by a relatively high mercury content reaching as much as 140 μg/kg and the mercury content in the oldest analyzed coals from the Saddle Beds are characterized by clearly lower mercury content – up to 40 μg/kg on average.

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