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# SPLIT OF MERCURY BETWEEN PRODUCTS OF COAL CLEANING VERSUS MERCURY EMISSIONS REDUCTION

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**Abstract:** The article deals with the analysis of the split of mercury present in raw coals between commercial products and wastes in 21 Polish collieries producing hard steam coal (Upper Silesia Coal Basin). The coal cleaning constitutes the first step in the reduction of mercury emissions in coal utilisation (mainly combustion) processes by decreasing the charge of mercury in the commercial products in comparison to the raw coal. The ratio of this reduction depends, first of all, on the technological characteristics of raw coal, as well as on the range of the applied coal cleaning method. The charges of mercury in exploited raw coals are split in coal preparation processes (mainly coal cleaning processes) between commercial products and waste products. The mercury content in commercial products has been analysed together with the emissions from coal combustion processes. In the second case, tools for the reduction of emissions have already been employed. Characteristics of waste products, in particular the mercury content, have been under consideration to a less extent so far. Data presented in the article allows for better, broadened with the waste products, analyses, understanding and assessment of all environmental mercury originated risks, arising from coal production, including coal cleaning. Presented data generate also the need for discussion of such terms like: "mercury reduction in commercial coal products" and "mercury emissions reduction", as the result of coal cleaning processes.

***Keywords:*** *hard coal, mercury charge, coal cleaning, mercury charge split, mercury emissions reduction*

## 1. Introduction

The legal regulations on the reduction of mercury emissions of the coal utilization (mainly combustion) processes are already a fact (UNEP, 2013; Sloss, 2012). Further regulations are in preparation (Best Available, 2013), including the executive acts of the Minamata convention, which most probably will encompass not only the mercury emissions to the atmosphere but also other aspects, for example the gold artisanal production, as well as other potential sources of mercury transferred to the environment. Coal production and utilization are counted among those types of economic activity as a result of which large quantities of mercury enter the environment. It is estimated that in 2005 around 50% of mercury emissions originated

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from coal combustion (UNEP, 2008). At the same time the high uncertainty of the mercury emission estimates is emphasized (UNEP, 2008) together with the observed decrease in the emissions. For example in Europe between 1980-2005 the mercury emissions decreased by almost 80% (Pacyna et al., 2009). Broadening of knowledge on the mercury charge in the Polish coals has enabled, among other, to recalculate and decrease the values of the reported mercury emissions (Ochrona Srodowiska, 2013; Paczosa, 2014). All this points out the necessity of conducting further analysis of the anthropogenic sources of mercury emissions to the environment. Such a necessity was defined in the Minamata Convention itself, for example in Article 9 (UNEP, 2013). A significant source of mercury emissions to the environment can be the coal mining waste and in particular the coal cleaning waste. If the mercury emissions during the utilization of cleaned coal (clean coal concentrates) are to be lower than during the utilization of raw coal, then a question should be asked on the mercury split in the coal cleaning processes and on the degree of mercury pollution in the waste from these processes.

Cleaning processes are commonly applied for improving coal quality before utilization. Their application is supported by the need to minimize the pollutants charge in the used coal, which becomes of particular importance for long-distance transport of coal (Couch, 1999). Utilisation of the cleaned coal, characterized by high quality parameters, increases also the efficiency of the conversion of the chemical energy of coal to other forms of energy and decreases the costs of countermeasures to the negative environmental impact of energy production as well as the exploitation costs in power plants (Couch, 1999).

The coal cleaning methods used for removal of various pollutants and adaptation to the end-user needs, including the reduction of mercury in coal, provide another advantage. The decrease of the charge (mass) of the pollutants in coal concentrates in comparison to the raw coal must be accompanied by the transfer of a part of the charge of the pollutants to the waste. Literature provides data on the mercury content in raw coals and after cleaning (Best Available Technique, 2013; Bialecka et al., 2012; Dziok et al., 2014; Matalerz et al., 2005; Ozbayoglu, 2011; Ozbayoglu, 2013; Pyka et al., 2010a; Toole-O’Neil et al., 1999; UNEP, 2010; Zajusz-Zubek et al., 2014). Some of the publications contain data on the mercury content in the coal cleaning waste (Dziok et al., 2014; Bialecka et al., 2012; Huggins et al., 2009; Ozbayoglu, 2013; Wang et al., 2009; 2006a; 2006b) but practically no literature contains the mercury mass (charge) split between the cleaned coal and waste.

The article provides the results of the split of the mercury mass between the commercial products and waste for 21 Polish collieries producing hard steam coal and the weighted average of the split as a function of the coal cleaning range. The presented data indicate the scale of the environmental threat following the extraction and utilisation of the coal polluted by mercury. The data confirm that the threats cannot be identified solely with the mercury emissions from the coal utilisation processes.

The industrial cleaning processes of coal and other raw materials are not ideal. Their unavoidable, typical features are the losses of the carbonaceous matter to the waste. The presented data in combination with the information on the calorific value of the raw coal, cleaned coal well as the "calorific value" of the waste (i.e. energy balance of energy contained in the raw coal – the feed for the cleaning processes) allow for starting discussion on the method of an objective assessment of the reduction of mercury emissions as a result of mercury charge change in coal during coal cleaning.

## Impact of coal cleaning on reduction of mercury content and charge in coal

In the UNEP (2010) document it is shown that the classic coal cleaning methods lead to around 30% decrease of the mercury charge (mass) in coal after cleaning. The application of advanced, chemical coal cleaning methods (e.g. the *K-fuel* method) can lead to a decrease of mercury charge in coal even by 70%. In another literature source, which reviews the current, already quite rich American experiences (Pavlish, 2003), various degrees of mercury content reduction in cleaned coals for various deposits– starting with practically none up to around 80% is discussed. Thus, the variability in the coal susceptibility to the reduction of mercury content as a result of coal cleaning is significant. This is confirmed by the results of other researchers (Das et al., 2013; Dziok et al., 2014; Feeley et al., 1994; Lopez-Anton et al., 2006; Ozbayoglu, 2011; Pyka et al., 2010a; Quick et al., 2002; Zajusz-Zubek et al., 2014).

It is worth pointing out that the decrease of the mercury contamination in coal after coal cleaning processes, similarly to the reduction of other contaminants, has two dimensions. The first one is the assessment of mercury content in coal before and after cleaning. The second one is the assessment of the mercury charge in coal before and after coal cleaning. For example Ozbayoglu (2013), discusses removal of mercury from raw coal in the cleaning process with the "reduction rate" of the mercury at around 53%, even though the mercury content in some of the cleaned coals, obtained in unit processes applied to various particle size fractions, was higher than in the raw coal. This is a classic example, where coal cleaning leads to the reduction of the contamination of the commercial product with mercury. The technological characteristics of the mining product indicate the lack of possibility to decrease the contaminant concentration in the cleaned coal in relation to the content (concentration) of a given pollution in the feed for cleaning processes. Similar results of industrial research for several Polish hard coal collieries were presented by Dziok et al. (2014). The analyses of the technological characteristics of several Polish hard coals (Pyka et al., 2010a) indicated that the coal cleaning in the full particle size range always leads to concentrates with lower mercury content than in the feed. It gives an incentive to some theoretical considerations. It can be assumed that during the coal cleaning processes the total energy contained in the raw coal is transferred to the commercial products as well as the whole mercury is transferred to the commercial products. The chemical energy of the coal fuel is not lost with the waste, but also the waste is not contaminated with mercury. Despite the higher calorific value of the commercial products (pure waste was separated with no energy loss) their mercury emissions will remain the same as for raw coal, since we are dealing here with the same amount of energy and the same mercury charge (waste free of mercury). In practice the contaminants from raw mining product concentrate most often in the waste, but their discharging results in energy losses. This makes the assessment of the impact of the cleaning processes on the reduction of contaminants' emissions in coal utilization processes much more complex. Moreover, the information on the mercury content in coal and its reduction as the result of coal cleaning is incomplete. The lack of information on the mass balance of the coal cleaning process does not allow to directly estimate how much the mercury mass (charge) directed with coal to the end users was decreased.

Literature provides various methods of assessing the efficiency of decreasing the mercury content in coal cleaning processes (Dziok et al., 2014; Lopez-Anton et al., 2006; Toole-O'Neil et al., 1999; Wichlinski et al., 2013). The simplest are based on the difference between the mercury content in the cleaned coal and the raw coal. An example of including the improvement of the calorific value of the cleaned coal in relation to raw coal was proposed by Tool-O'Neil et al. (1999) in the form of the mercury reduction index:

$$\eta_{\mathrm{Hg2}} = \frac{\frac{Hg_{\mathrm{feed}}}{Q_{\mathrm{i\_feed}}} - \frac{Hg_{\mathrm{concentrate}}}{Q_{\mathrm{i\_concentrate}}}}{\frac{Hg_{\mathrm{feed}}}{Q_{\mathrm{i\_feed}}}} 100\ \% \tag{1}$$

where: $\eta_{\mathrm{Hg2}}$ – mercury reduction index, [%],
$Hg_{\mathrm{feed}}$ – mercury concentration in the feed, [μg/kg],
$Q_{i\_\mathrm{feed}}$ – calorific value of the feed, [kJ/kg],
$Hg_{\mathrm{concentrate}}$ – mercury concentration in the cleaned coal, [μg/kg],
$Q_{i\_\mathrm{concentrate}}$ – calorific value of the cleaned coal, [kJ/kg].

The reference to the calorific value of coal before and after the cleaning process, assumed in Eq. 1 in practice leads to an overestimation of the assessment of the reduction of mercury content in coal. The equation does not include the inevitable energy loss connected with the waste in the cleaning processes. For the purpose of objectivity, the energy loss related to waste in the cleaning processes should be included and the reduction should be estimated based on some constant reference, at the best constant quantity of energy in coal before and after cleaning. (Pavlish, 2003; Smolka et al., 1999). Taking the above into consideration the equation should, in our opinion, be supplemented with coefficient $\eta$, which would include the energy loss in waste since the commercial products composed of the cleaning products of the given raw coal lot will always "carry" lower energy than the raw coal lot:

$$\eta_{Hg2} = \frac{\frac{Hg_{\mathrm{feed}}}{Q_{i\_\mathrm{feed}}} - \frac{Hg_{\mathrm{concentrate}}}{Q_{i\_\mathrm{concentrate}}}}{\frac{Hg_{\mathrm{feed}}}{Q_{i\_\mathrm{feed}}}} \eta \cdot 100\ \% \tag{2}$$

where: $\eta$ is the energy loss in the waste.

The quoted publications (Dziok et al., 2014; Ozbayoglu, 2013, Pyka et al., 2010a), allow for drawing another significant practical conclusion. The "behavior" of mercury in the cleaning processes is a function of the particle size. The results of the tests conducted for one, narrow size fraction, are more often than not unrepresentative and do not allow to generalize on the mercury reduction in cleaning processes in the full particle size range of raw coal. Results of tests conducted on samples of heavily grounded (e.g. below 0.5 mm) coal as well as attempts of generalization on the reduction of mercury contamination of the commercial coal in the cleaning processes adopted for the full particle size range can be found in literature (Feeley et al., 1994; Guangqian et al., 2013; Ozbayoglu, 2011; Lopez-Anton et al., 2006; Quick et al., 2002)

Literature data show that the mercury content in the waste from coal cleaning is, similarly to that of the cleaned commercial coal, variable (Dziok et al., 2014; Bialecka et al., 2012; Ozbayoglu, 2011; Ozbayoglu, 2013; Pyka et al., 2010b). Generally, these values are not significantly larger than for raw coal. The mass of the waste produced in the cleaning processes is relatively large. In Poland in 2013 it was around 35 teragrams (Tg) at the coal production around 76 Tg (Paszcza et al., 2014). Thus, the contamination with mercury is a problem (Pyka et al., 2010b). Similarly to the small mercury content in the coal used on a mass scale, also the small mercury content in the waste translates into large masses (charges) of mercury being exposed to the environment.

## Testing program and methodology

Samples of commercial coals of selected intermediate products (commercial products components) and waste were taken from different collieries. In order to ensure the representativeness of the data, samples of commercial products (components) were taken over the period of at least one week. The samples for analysis applying Polish Standards (2014) were prepared from individual as well as combined samples.

The basic quality parameters and the mercury content were determined according to the certified internal procedure elaborated at GIG (Poland) (No. SC-1.PB.23) applying the cold-vapor atomic absorption spectrometry. In the case of the waste, most mercury analyses were performed on the analytical samples obtained from the collieries.

For each colliery, from several to several dozens of mercury samples of the commercial product, or their basic components were analyzed. In the case of complex commercial product, the composition system was used. From 1 to 3 analysis of mercury in the waste were performed. In order to eliminate the accidental sampling errors, the results of the laboratory tests of the basic quality parameters of the commercial products and waste were verified through comparison with the data from the online monitoring of production quality. The obtained quality data were combined with the information on the production of respective commercial products and waste per year. Due to unavailability, during the preparation of the publication, of annual data on year 2014 production, for the purpose of calculation of the mercury charge between the commercial products and waste, the data on the 2013 production were used.

The above calculations and analyses were prepared for 21 collieries producing steam coal, located in the Upper Silesian Coal Basin (USCB) in Poland. For the selected collieries the assessment of the split of energy in the raw coal between the commercial products and waste in coal cleaning process was performed. These values were used for assessment of the real reduction of the mercury content (as well as mercury emissions) resulting from coal cleaning.

## The results and discussion

Figure 1 presents the results of the analysis of the split of the mercury charge in 21 steam coal mines located in the USCB in Poland. The split of the mercury mass between the commercial products and waste is provided in percent. The analyzed mines can be divided into two groups:

- collieries 1–7 (Fig. 1), in which only the coarse and middle sized coals (coarse and medium coal size fractions) are cleaned i.e. ROM coal with the particle size above 20 mm,
- collieries 8–21 (Fig.1), in which to a smaller or larger extent also the fines are cleaned, i.e. ROM coal with the particle size below 20 mm in dedicated cleaning processes.

Colliery 6 belongs to the group of collieries in which raw fines are not cleaned. There the cleaning of coarse and middle sized coals results in transfer of only 4% of the mercury charge to the waste. The same is for colliery 4, in which over 30% of the mercury charge can be found in the waste. The fact that coal cleaning technology for the coarse and medium size fractions is similar in each of the collieries and concerns more or less the same part (mass) of the ROM coals shows a strong influence of the technological characteristics of ROM coal on the efficiency of mercury reduction.

Colliery 17 belongs to the group of collieries in which the coal cleaning concerns also the fines in which only around 15% of the mercury charge in the raw coal is directed to the waste. Despite the fact that for none of the analyzed collieries the raw fines are cleaned as a whole, in the case of two collieries (numbers 8 and 21) over 50% of the mercury charge in the raw coal is transferred to the waste. This indicates a potential for reduction of the mercury content and charge in the cleaning processes in the analyzed Polish steam coal collieries as well as the scale of contamination of the waste with mercury.

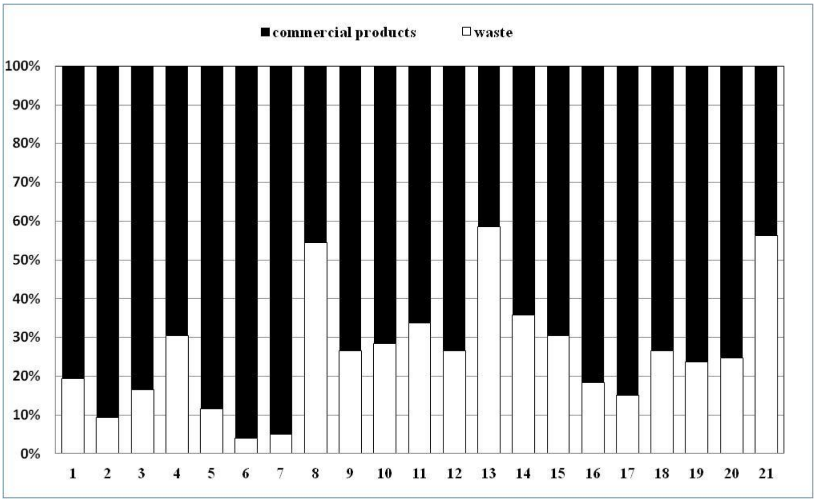

Fig. 1. Split of mercury between commercial products and waste, expressed in % in 21 of analysed collieries

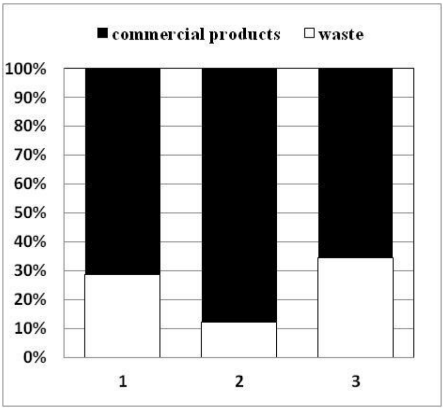

Fig. 2. Weighted mean split of mercury between commercial products and waste for 21 collieries (1) and divided between the mines in which only the coarse and medium coal size fractions are cleaned (2) as well as the fractions themselves and partly the smalls (3)

Data in Fig. 1, cumulated for all the collieries as well as divided into the two assumed colliery groups have been recalculated including the coal mass and the waste produced/generated in the analysed collieries. The results of these calculations are presented in Fig. 2. The mean mercury charge (mass) presented in Fig. 2 is the weighted mean and illustrates in particular the impact of coal cleaning on the split of the mercury charge.

In the collieries in which only the coarse and middles sized coals are cleaned, on average around 12% of the mercury charge from the raw coal is transferred to the waste. In the collieries which also clean fines, but not in the full range, on average almost 35% of the mercury charge from raw coal leaves the cleaning plant together with waste. The part of mercury charge contained in raw coal removed with the waste for all of the analyzed collieries equals, on average, around 29%. The mercury charge in the waste, directly and objectively illustrates the additional environmental threats resulting from coal production in Poland, which should not be identified only with mercury emissions to the atmosphere.

The split of mercury charge in raw coal between the cleaned coal and waste should not be directly identified with the reduction of mercury in coal, estimated for the purpose of demonstrating the limiting of the mercury emissions through coal cleaning. Not only the mercury charge is carried with the waste, but also unavoidable losses of the energy contained in raw coal. The losses deprive us of a constant basis for treating the mercury charge split equally to the reduction of mercury emissions. There is a lack a constant basis in the form of the same amount of energy in the raw coal and the commercial products of the coal (Smolka et al., 1999). In the earlier analyses, conducted for raw coals from several of the USCB collieries (Smolka et al., 1999), it has been estimated that depending on the range of coal cleaning the energy losses can be:

- additionally from around 6.4 to over 10%, when only the coarse and middles sized coals are cleaned,
- additionally from around 6.5% to over 14% when also the fines are cleaned.

The results were taken from modeling of the coal cleaning processes which include their randomness but in the industrial processes the energy losses can be even higher. The reduction of the mercury content in coal, despite the presented significant amounts of mercury being transferred to coal waste, should be assessed including the energy losses and thus it is reasonable to apply for example in Eq. 1 coefficient $\eta$ (Eq. 3) with the value smaller than 1, which includes the energy losses in the waste:

$$\eta = \frac{100 - \text{energy losses}}{100} \tag{3}$$

In practice the value of coefficient $\eta$, after being introduced into Eq. 1 results in a decrease of the value of the estimated mercury content reduction as a result of coal cleaning. Assuming the values above we see a decrease, by around 6%, when only the coarse and medium coal size fractions are cleaned, and up to over 24%, when the coarse and medium coal size fractions are cleaned together with the fines.

## Conclusions

The charge of the contaminants contained in raw coal is divided in the coal cleaning processes between the commercial products and waste. In the case of mercury contained in coal, the mercury charge in the raw coal for several of the analyzed collieries is divided into the commercial products and waste on average in the ratio around 71%/29%.

Coal cleaning is an efficient method of reduction of mercury emissions to the atmosphere during its utilization. The split of mercury charge between the products and waste, especially in case of the collieries in which only the coarse and middles sized coals are cleaned, depends on coal processing technological.

The difference between the split of the mercury charge into the commercial products and waste in collieries cleaning only the coarse and middles sized coals and the collieries cleaning also the fines shows that broadening of the scope of coal cleaning leads to efficient decrease of the mercury charge in commercial coal. Thus, we obtain the confirmation that in Poland coal cleaning is a very important element of the mercury emission reduction strategy.

The mercury charge (mass) in commercial coal is the basis for the assessment of the threat of mercury emissions. These charges can be matches or even exceeded by the mercury charge in the waste from coal cleaning. This comprises another environmental issue which needs to be analyzed, and surely even addressed by preventive measures.

The results of the assessment of the mercury charge/ content in coal and in the final outcome also mercury emissions as the result of coal cleaning, conducted applying the currently used approaches do not provide fully objective information. These results suffer from the lack of a comparison basis such as the constant energy of the raw coal, partially lost with the waste. The energy losses in the waste do not allow for a direct comparison of the mercury charge in raw coal before cleaning and in the coal commercial products produced from this raw coal. These losses can exceed 24% of the energy contained in the raw coal and the mercury reduction effect in the products, in reference to the content/charge of mercury in the raw coal. Thus, also the mercury emissions reduction resulting from coal cleaning will be decreased at the same level.

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