Pollution of coals of the Upper Silesian Coal Basin from the perspective of the European registers of the release and transfer of pollutants to air - the case of chlorine

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Abstract: The regulation WE 166 (2006) of the European Parliament and European Council on the establishment of the European Register of Release and Transfer of Pollutants presents the limiting values of pollutants to the atmosphere and values from mining operations and power plants. Chlorine and its compounds constitutes a significant pollutant. Taking into account the chlorine content of selected coals (up to 2.69% in the ashes) and the limiting values of the releases into the atmosphere one should perform a detailed analysis of its release from coal mines and power plants to the environment.

1. Introduction

Hard coal is mostly an organic substance comprised of carbon, hydrogen, oxygen, nitrogen, sulphur and insignificant amounts of phosphorus. Besides the dominating content of organic substance, coal also includes mineral substance, that is, inorganic impurities found after incineration in the form of ash [e.g. 1]. The mineral components found in coal may be syngenetic – formed simultaneously to the coal substance or epigenetic – accumulated after the coal seam was formed. It has been determined that trace elements occur in the inorganic coal substance. Their type and content varies significantly. In some coals, certain elements, such as: Ge, As, B, Be, exhibit concentrations multiple times higher than in the earth crust [e.g. 2,3,4]. Concentrations exceeding the crustal abundance are also exhibited at times by: Ba, Bi, Co, Cu, Ga, La, Li, Pb, Hg, Mo, Ni, Sc, Se, Ag, Sn, V, Y and Sr. These elements include ones that constitute environmental pollutants. Threshold values for their release to air, water and soil were determined in the EC Regulation No. 166 of the European Parliament and Council dated January 18th 2006, regarding the establishment of a “European Pollutant Transfer and Release Register.” Based upon this regulation, the entities releasing the substances to environment, including mining plants, CHP plants and other coal combustion systems, shall be required to control and to follow a series of requirements regarding the release of pollutants. Considering the above information, this publication presents the components of coal that are pollutants. Using the example of chlorine, its identification in the coal mines of the Upper Silesian Coal Basin was described while giving consideration to the threshold values of release to air and water. Chlorine was also selected due to the negative impact of that element on the machines used in coal processing by the formation of tarnish (chlorine corro-
sion) on heating devices. Moreover, chlorine is an undesirable component of coking coal as 40-60\% of chlorine remains in the coke [5].

2. Components of coals in view of the “European pollutant transfer and release register” to air

Table 1 presents the components included in the coals of the Upper Silesian Coal Basin that simultaneously constitute pollutants, the release of which to air, water and soil is limited based on the European Pollutant Transfer and Release Register (Regulation No. 166/2006 of the European Parliament and Council).

| No. | Pollutants                                      | Threshold value for release to air, kgs/year |
|-----|------------------------------------------------|-------------------------------------------|
| 1   | Carbon monoxide                                | 500000                                    |
| 2   | Carbon dioxide                                 | 100000000                                 |
| 3   | Nitrogen oxides (\(\text{NO}_x/\text{NO}_2\))  | 150000                                    |
| 4   | Total nitrogen                                 |                                           |
| 5   | Total phosphorus                               |                                           |
| 6   | Arsenic and arsenic compounds                  | 20                                        |
| 7   | Cadmium and cadmium compounds                  | 10                                        |
| 8   | Chromium and chromium compounds                | 100                                       |
| 9   | Copper and its compounds                       | 100                                       |
| 10  | Mercury and its compounds                      | 50                                        |
| 11  | Nickel and its compounds                       | 50                                        |
| 12  | Lead and its compounds                         | 200                                       |
| 13  | Zinc and its compounds                         | 200                                       |
| 14  | Chlorium (as total Cl)                         |                                           |
| 15  | Chlorium and its inorganic compounds (as HCl)  | 10000                                    |
| 16  | Fluorides (as total F)                         |                                           |
| 17  | Fluorine and its inorganic compounds (as HF)   | 5000                                     |
| 18  | Suspended particulate matter (PM\(_{10}\))    | 50000                                    |

3. Content of chlorine in the coals of the Upper Silesian Coal Basin

Chlorine – similarly to nitrogen and sulphur – is an element that is commonly occurring in hard coal. In various coals the content is different and it depends on the conditions of formation. It was determined that the content of chlorine increases along with the depth of the deposit and the level of water salinity.

As provided by T. Mielecki [6], the mean content of chlorine in the coals of the Upper Silesian Coal Basin is 0.01\% (from 0 to 0.025\%) in case of saddle beds (limnic type), while in the case of paralic beds, it reaches 0.175\%, usually not exceeding 0.1\%. J. Kuśmierska [6] exhibited that in case of certain coals, the chlorine content reaches 0.3\%. On the other hand, A. Różkowska [7], while analysing the chlorine content in coals from core samples, found that the chlorine content was from 63 to 18871 g/Mg. The highest contents of chlorine are found in the coals of the eastern and south-eastern parts of the Upper Silesian Coal Basin (4000-9000 g/Mg), while the lowest contents are found in the south-western part (400-700 g/Mg). As provided by A. Strugała [5], the chlorine content in coals varies in the range from 0.02 to 0.4\%, whereas M. Kosewska and K. Wróblewska [8] have exhibited that the amount of chlorine in coking coals used as a charge is within the range from 0.02 to 0.4\%. Based on the tests of coal samples from the south-western part of the Upper Silesian Coal Basin, it has been found that the content of chlorine in ash after combustion reaches 4.08\%. The results of chlorine content in ash after combustion of coals from the Cracow sandstone series (KSP), mudstone series (SM), the Upper Silesian sandstone series (GSP) and the paralic series (SP), have been presented in tables from 2 to 7.
Table 2. The content of chlorine in the ashes of selected coals from the Cracow sandstone series.

| Lithostratigraphic series | Coal seam | Cl, % |
|---------------------------|-----------|-------|
| KSP                       | 206       | 1.26  |
| KSP                       | 206       | 0.51  |
| KSP                       | 209       | 2.69  |

Table 3. The content of chlorine in the ashes of selected coals from the Mundstone series (the Orzesze layers).

| Lithostratigraphic series | Coal seam | Cl, % |
|---------------------------|-----------|-------|
| SM                        | 308       | 1.12  |
| SM                        | 348       | 0.79  |
| SM                        | 361       | 0.19  |
| SM                        | 364/1     | 4.08  |

Table 4. The content of chlorine in the ashes of selected coals from the Mudstone series (the Ruda layers).

| Lithostratigraphic series | Coal seam | Cl, % |
|---------------------------|-----------|-------|
| SM                        | 403/3     | 2.65  |
| SM                        | 404/4     | 0.15  |
| SM                        | 405/2     | 0.44  |
| SM                        | 409/4     | 1.68  |

Table 5. The content of chlorine in the ashes from selected coals of the Upper Silesian sandstone series.

| Lithostratigraphic series | Coal seam | Cl, % |
|---------------------------|-----------|-------|
| GSP                       | 502/2     | 0.42  |
| GSP                       | 503       | 0.37  |
| GSP                       | 505       | 0.68  |
| GSP                       | 507       | 0.23  |
| GSP                       | 510/1     | 0.25  |

Table 6. The content of chlorine in the ashes from selected coals of the Paralic series (the Porębskie Layers).

| Lithostratigraphic series | Coal seam | Cl, % |
|---------------------------|-----------|-------|
| SP                        | 600       | 0.91  |
| SP                        | 626/1     | 0.05  |
Table 7. The content of chlorine in the ashes from selected coals of the Paralic series (the Jaklowieckie layers).

| Lithostratigraphic series | Coal seam | Cl, % |
|--------------------------|-----------|-------|
| SP                       | 712/2     | 0.68  |
| SP                       | 713/1     | 0.96  |

The use of hard coal, consisting mostly in its combustion, results in the release of the chlorine present in the coal, most often in the form of hydrochloride, which is a compound that is aggressive to power and processing machines. Considering this issue, the results of the chlorine content tests of coals from various lithostratigraphic series and giving consideration to the threshold value in line with the regulation No. 166/2006 of the European Parliament and Council amounting to 10000 kgs/year, the content of chlorine (hydrochloride) released to air as gas and as particulate matter during the combustion in CHP plants should be analyzed.

4. Summary
Chlorine is a common component in the coals of the Upper Silesian Coal Basin and – at the same time – is a significant pollutant. This is why the threshold values for its release to air and water have been specified in the regulation (EC) No. 166/2006 of the European Parliament and Council in the established “European Pollutant Release and Transfer Register.” Tests have exhibited that the chlorine content in ash after the combustion of selected coals of the Upper Silesian Coal Basin reaches 2.69%. Considering these results, detailed analyses of conditions of disposal of chlorine from coal mines and power plants should be conducted.

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