# The possibilities for reducing mercury, arsenic and thallium emission from coal conversion processes

**Piotr Burmistrz, Faustyna Wierońska, Marta Marczak and Dorota Makowska**

AGH University of Science and Technology, Faculty of Energy and Fuels,
A. Mickiewicz Avenue 30, 30-059 Krakow, Poland

wieronsk@agh.edu.pl

**Abstract**. According to The European Parliament and The Council (Decision No. 1386/2013/UE), the emission of air pollutants during the last years has decreased, but still constitute a serious problem in many parts of Europe. Apart from sulfur oxide, nitrogen oxide and volatile organic compounds (VOCs), among air pollutants are also elements, which have a negative impact on environment and human health and life. During coal combustion and coking coal processes, a part of these elements are released into the atmosphere. For this reason, coal processing is one of the main sources of their emissions to the environment. Particular attention should be paid to the emission of such elements as mercury (Hg), arsenic (As), thallium (Tl) and their compounds, which are characterized by very high ecotoxicity. In this article the current standards and regulations on emissions of mercury, arsenic and thallium into the atmosphere for selected coal conversion processes as well as opportunities to reduce these emissions were presented.

## 1. Introduction

The amount of coal mined in Poland in 2016 amounted to over 130 million tons, 75% of which were coals combusted in the power industry, and more than 9% of coals were used to produce coke [1]. Coal, in addition to the main elements (C, H, O, N, S), contains the so-called ecotoxic elements. During the combustion and coking processes of coal, some of these elements are released into the atmosphere, causing that coal processing processes are one of the main sources of their emission to the environment. According to the European Parliament and the Council (1386/2013/EU), emissions into the air have declined in recent years, however, it is still a problem in many parts of Europe and requires taking measures to reduce emissions in particular: sulfur oxides, nitrogen oxides, volatile organic compounds (VOCs) and ecotoxic elements such as mercury (Hg), arsenic (As), thallium (Tl) and their compounds.

Mercury is a highly toxic heavy metal and the human body does not show physiological demands on it. This element, together with arsenic, has been classified by the US Environmental Protection Agency (US EPA) as dangerous air pollutants [2]. Hg and As show the most adverse environmental impact (Group I), while Tl is classified in Group III with slightly lower toxicity according to Swaine [3]. However, many sources compare toxicity of thallium to mercury or lead [4]. Mercury, arsenic and thallium are elements with a very high accumulation ratio [5]. Moreover, arsenic according to International Agency for Research on Cancer data has a proven carcinogenic effect [6]. The above elements are included in the list of dangerous substances set out in the Regulation of the Minister of Health of September 28th, 2005 in Poland [7].

The US Environmental Protection Agency adopted in 2012 a law introducing limitations in the emission of ecotoxic elements to the environment. These standards have been included both existing and newly built coal-fired power plants. Limits have been set for ecotoxic elements, including for mercury and arsenic [8]. Currently in Poland and in the European Union there are no specific limits regarding the emission of ecotoxic elements to the environment for combustion or coking coal. The only limits that have been introduced concern the certain industry sectors, including incineration and waste co-incineration plants in accordance with Directive 2010/75/EU and the Ordinance of the Minister of the Environment of the Republic of Poland of 4 November 2014 (table 1).

**Table 1.** Emission limit values of TE for plants incinerating or co-incinerating waste according to the Regulation of the Minister of the Environment of November 4, 2014.

| Trace element (TE)/ sum of TEs | Content of TE in flue gases (mg/Nm$^3$) |
|---|---|
| Cd + Tl | 0.05 |
| Hg | 0.05 |
| Sb+As+Pb+Cr+Co+Cu+Mn+Ni+V | 0.5 |

In addition, Poland is required to register quantities of ecotoxic elements (including mercury and arsenic) released into the air, water and soil on the basis of the arrangements approved during the Geneva Convention in 1979 on the long-range transboundary atmospheric pollutants transport [9] and in accordance with Regulation of the European Parliament and the Council 166/2006 [10]. The National Center for Emissions Management (KOBiZE) is responsible for developing the Polish report. According to data from KOBiZE and the European Environment Agency (EEA), Poland was the largest mercury emitter (18.6%) to the environment and one of the largest arsenic emitters (23.2%) from the EU-28 countries in 2015.

However, the legislative situation regarding the emission of ecotoxic elements to the environment is going to change, because in 2017 the European Commission adopted conclusions on the best available techniques BAT for large combustion plants [11]. The standards for $CO_2$, $SO_2$, $NO_x$ and particulate matter PM emissions are going to be tightened, as well as mercury emission limits will been introduced. The mercury emission limit values varies from 1 to 10 μg/Nm$^3$, depending on the total thermal power delivered in the fuel and the type of fuel (hard coal, brown coal) (table 2). Furthermore, the BAT conclusions introduce the obligation to monitor emissions of ecotoxic elements, including arsenic and mercury to air (BAT 4) in accordance with EN 14385, EN 13211, EN 14884 standards, which involve the determination of these elements directly in the exhaust gas.

The standards regarding the emission of a total amount of Tl and Cd to the environment for co-incineration of waste with coal are also tightened (5-12 μg/Nm$^3$ for installations <300 MW, 5-6 μg/Nm$^3$ for installations ≥ 300 MW), biomass and peat (<5 μg/Nm$^3$). The above requirements and standards must be met by 2021. In the future, similar regulations are expected for other ecotoxic elements, including for arsenic and thallium. At present, the coke industry must meet the environmental requirements included in the IPPC Directive and BAT standards [12, 13].

**Table 2.** Levels of mercury emissions into the atmosphere from combustion process of hard coal and brown coal defined in BAT conclusions [11].

| Combustion plant total rated thermal input ($MW_{th}$) | BAT-AELs (μg/Nm$^3$) Yearly average or average of samples obtained during one year | | | |
|---|---|---|---|---|
| | New plant | | Existing plant | |
| | Coal | Lignite | Coal | Lignite |
| < 300 | < 1-3 | < 1-5 | < 1-9 | < 1-10 |
| ≥ 300 | < 1-2 | < 1-4 | < 1-4 | < 1-7 |

## 2. Content and occurrence of mercury, arsenic and thallium in coal

The mercury content in coals is relatively low, from several dozen to several hundred μg/kg. The average mercury content in Polish subbituminous coal ranges from 25 to 300 μg/kg, while in Polish lignites varies from 100 to 450 μg/kg [14-17]. Mercury is present in coal both in a form associated with a mineral matter (mainly pyrite), an organic matter, e.g. in sulfuric connections and in the form of native mercury [18].

Arsenic in coal is mainly associated with pyrite. It can also occur as organic arsenous ($As_{org}$) and in combination with silicates [19]. The content of arsenic in coal is reported according to various literature sources within 0.5-80.0 mg/kg [20], 0.3-16.6 mg/kg [21] and 0.3-11.0 mg/kg [22]. There are also coals containing even up to 0.4% of As [23]. Its quantity in Polish bituminous coals and lignites is respectively 0-40 mg/kg [24] and 5-15 mg/kg [25].

Thallium in coal is mainly associated with pyrite. The content of this element in coal is in the range from 0.01-3 mg/kg [20, 26, 27]. A high concentration of thallium was found in lithotypes containing significant amounts of arsenic or sulfur (up to 26 mg/kg) [26]. In Polish hard coals and brown coals, the thallium content is 0.2-5.3 mg/kg and 0.2-2.4 mg/kg [28], respectively.

## 3. The possibilities for reducing mercury, arsenic and thallium emission from coal conversion processes

### *3.1. Coal combustion processes*

The limitation of the emission of ecotoxic elements to the atmosphere from combustion processes can be divided into two groups of methods: primary (so-called pre-combustion) and secondary (post-combustion). The first group includes, among others, coal cleaning, descaling (dry separation), thermal preparation, chemical and biological methods, as well as selective coal mining [29 - 33]. Due to the connection of mercury, arsenic and thallium with coal mineral matter (mainly with sulfides) [16, 34, 35], coal cleaning processes achieve relatively high efficiency in removing these elements from coal. In the case of mercury, they allow to remove from 10 to 78% of this element [36 - 38]. The efficiency of arsenic removal in these processes ranges from 35% to 83% [39, 40], and from 41% to even 92% for thallium [41]. Research conducted on a pilot installation for dry separation (air concentrating table) also indicate the effectiveness of this method in removing mercury from coal [42]. Whereas, mercury associated with an organic matter can be successfully removed by thermal preparation. The best results in the removal of mercury from coal can be achieved using a hybrid method combining coal cleaning with thermal preparation [43].

The document Ref. Ares (2017) 1248230-09/03/2017 presents the BAT for reduction of mercury emissions, which can be used to reach the lower range of BAT standards for combustion of bituminous coal and lignite. These techniques are divided into two groups. The first includes the co-benefit methods, dedicated to removing other contaminants, but during which ecotoxic elements are removed by the way. Among these methods can be distinguished: selective catalytic reduction of nitrogen oxides (SCR), electrostatic precipitators (ESP), bag filters (FF), wet flue gas desulfurization (WFGD), dry/semi-dry flue gas desulfurization (D/SDFGD). The second group of methods includes special techniques of reducing mercury emission, in which one can distinguish among others: application of carbon sorbent (e.g. activated carbon or brominated activated carbon) injected into flue gases, use of halogenated additives in fuel or injection into the furnace.

In the gas formed after the combustion of coal, three speciation forms of mercury can be distinguished: oxidized mercury ($Hg^{2+}$), mercury associated with fly ash particles ($Hg_{(p)}$) and elemental mercury - metallic mercury ($Hg^0$). Due to the insolubility in water, durability and the ability to carry over long distances, the most undesirable form of mercury is $Hg^0$. The other two forms can be effectively removed in the wet desulfurization installation, as well as in dedusting devices such as ESP or FF [11, 31, 44] (table 3). According to Wang's research [45] the efficiency of the ESP in the mercury removal from flue gas varies from 6 to 46% and depends on the $Hg_{(p)}$ share in the flue gases and the efficiency of dust removal in the ESP. The use of SCR, through the catalytic oxidation of mercury $Hg^0$ to $Hg^{2+}$, can cause removal of 34% to 68% of the $Hg^0$ contained in the exhaust. On the other hand, the installation for removing sulfur oxides by the wet method can remove 15.3% - 80% of mercury from flue gas (including up to 97% of $Hg^{2+}$ coming from the flue gas to FGD [45, 46]. The results of the research prove that the efficiency of flue gas cleaning from mercury depends largely on the applied exhaust gas treatment system and its effectiveness as well as on the occurrence of mercury in the exhaust gas, which is a consequence of the chemical composition of the coal burned and the type of the boiler [44].

**Table 3.** The effectiveness of mercury removal depending on the Air Pollution Control Devices (APCD).

| Boiler | Coal (Hg content) | Air Pollution Control Devices | Effectiveness of Hg removal | Reference |
|---|---|---|---|---|
| PCB 200 MW | hard coal (233±12 μg/kg) | ESP+WFGD | 68% | [45] |
| PCB 600 MW | hard coal (142±38 μg/kg) | ESP+WFGD | 70% | |
| PCB 300 MW | anthracite (174±1 9 μg/kg) | ESP+WFGD | 81% | |
| PCB 600 MW | brown coal (35±19 μg/kg) | ESP+WFGD | 28% | |
| PCB 100 MW | hard coal (385±113μg/kg) | ESP+CFB-FGD+FF | 66% | |
| PCB 165 MW | brown coal, (17±5 μg/kg) | SCR+ESP+WFGD | 37% | |
| PCB 350 MW | hard coal (38.6 μg/kg)<br>hard coal (33.7 μg/kg)<br>brown coal (36.2 μg/kg) | PCB (low-emission burners) + SCR + combination of ESP/FF + WFGD | 59% - 73% | [46] |
| PCB 370 MW | hard coal (66±9 μg/kg) | SNCR + ESP + WFGD | 72% - 84% | [15] |
| PCB 225 MW | hard coal (100±15 μg/kg) | ESP + WFGD | | |
| PCB 370 MW | brown coal (596±99 μg/kg) | ESP + WFGD | | |

PCB - pulverized-coal boiler; ESP - electrostatic precipitator; WFGD - wet flue gas desulfurization; CFB-FGD - circulating fluidized bed flue gas desulfurization; FF - fabric filter; SCR - selective catalytic reduction; SNCR - selective non-catalytic reduction.

Among the active methods, injection of powdered activated carbons and/or brominated activated carbons is the most commonly used. The effectiveness of mercury removal by means of the above method can range from 50% to over 90% (depending on the solution used) [47, 48]. However, with the increase in the mercury removal efficiency, the associated costs increase. The unit cost of removing mercury from flue gas using powdery activated carbons may amount to 40,000 - 90,000 $/kg [48].

Arsenic contained in coal goes into gas state in coal combustion process. The most common compounds of arsenic in gas are $As_{(g)}$, $As_2O_{3(g)}$. At high temperatures that prevail in boilers arsenic can also form solid As-Ca compounds ($Ca_3(AsO_4)_2$ and $Ca(AsO_2)_2$). Arsenic compounds are sorbent on fly ash particles when a flue gas temperature drops and are almost entirely removed from fly ash in an ESP or FF [19, 49]. The efficiency of cleaning the arsenic associated with ash separated on the electrostatic precipitator can reach up to 99.95%. According to Zhao [50] SCR is able to remove up to 29% of arsenic, which may be related to the condensation of arsenic compounds, in particular $As_2O_3$ in the micropores of the $V_2O_5$ catalytic converter [51, 52]. The efficiency of WFGD in purifying flue gas from arsenic can reach up to 71% according to Aunela-Tapola's research [53]. Depending on the treatment system used, the efficiency of removing As from gas can range from 98.85 - 99.9% [11, 54].

Thallium, same as arsenic, goes into the gas phase during coal combustion processes and then condenses on the fly ash particles. It is estimated that the amount of thallium released into the environment may be 700 - 2500 μg/m$^3$ [26]. On the other hand, according to of research [53], the use of an ESP in connection with the semi-dry desulfurization and bag filter installations results in lowering the Tl emission to the environment below 2.35 μg/m$^3$. Significantly lower emissions were obtained with the use of an ESP and wet desulfurization plant (table 4).

**Table 4.** Emissions of arsenic, mercury and thallium to the environment, depending on the on the APCD.

| Boiler | Coal | Air Pollution Control Devices | TE emission As | Hg | Tl | Reference |
|---|---|---|---|---|---|---|
| PCB 660 MW | hard coal | SCR + ESP + WFGD + WESP | 0.01 μg/m$^3$ | - | - | [50] |
| PCB 160 MW | hard coal | PCB (low-emission burners) + ESP + S-DFGD + FF | <2.58 μg/m$^3$ | 1.88 μg/m$^3$ | <2.35 μg/m$^3$ | [53] |
| PCB 113 MW | hard coal | PCB (low-emission burners) + ESP + S-DFGD + FF | <2.03 μg/m$^3$ | <0.11 μg/m$^3$ | <1.98 μg/m$^3$ | |
| PCB 350 MW | hard coal | PCC (low-emission burners) + SCR + combination of ESP/FF + WFGD | 0.11 μg/m$^3$ | - | - | [55] |
| PCB 350 MW | brown coal | PCC (low-emission burners) + SCR + combination of ESP/FF + WFGD | 0.20 μg/m$^3$<br>0.25 μg/m$^3$ | - | - | |
| PCB | hard coal | ESP + WFGD | 2.3 μg/m$^3$ | 8.8 ng/m$^3$ | 0.1 μg/m$^3$ | [56] |
| PCB 660 MW | - | SCR + ESP + WFGD + WESP | 0.01 μg/m$^3$<br>0.05 μg/m$^3$ | - | - | [54] |

PCB - pulverized-coal boiler; ESP - electrostatic precipitator; WFGD - wet flue gas desulfurization; FF - fabric filter; SCR - selective catalytic reduction; WESP - wet electrostatic precipitator; S-DFGD - semi-dry flue gas desulfurization.

### 3.2. *Coal coking processes*

The reduction of mercury emissions from coking processes can be obtained by using coal enrichment processes, which will allow the removal of between 21% - 84% of mercury contained in the fuel before the coking process [57]. In the coal coking process, the ecotoxic elements contained in coal pass into coke: 6.0% - 23.4% for Hg, 51.6% - 97.21% for As and 31.3% - 99% for Tl. Such broad ranges may be caused by various forms of occurrence of the above elements in coal [13]. During coal coking, the ecotoxic elements may be emitted to the environment during coke oven cell filling, coke pushing out from the chamber, during the process of quenching coke and burning coke oven gas in heating channels of the battery. The content of arsenic in the gas collected during charging of the coking chamber, pushing out coke and burning coke oven gas were respectively: 1.7 μg/m$^3$, 76.8 μg/m$^3$ and 2.3 μg/m$^3$ [58]. According to the requirements presented in the BAT conclusions adopted for coke installations, the limitation of the above fugitive emission of dust and thus ecotoxic elements to the environment can be obtained by filling coking chambers using low-emission filling systems and by using special exhaust hoods during coke pushing out from chamber. It is also recommended to use of dry coke quenching with the removal of coking dust using dedusting devices [59].

## 4. Conclusion

The paper reviews the regulations and standards for mercury, arsenic and thallium emissions to the environment from coal combustion and coking processes as well as the possibilities of reducing their emissions to the environment.

- In the BAT conclusions adopted in 2017, in addition to the tightening of the $CO_2$, $SO_2$, $NO_x$ and PM emission standards, mercury emission limits have been introduced for new and existing coal combustion plants, which will apply in the EU power industry from 2021.
- In order to meet mercury emission standards for combustion processes, it will be important to use both primary and secondary methods. In the case of insufficient effectiveness of passive secondary methods (installation for catalytic selective reduction of $NO_x$ and $SO_2$, PM removal in ESP or FF) additional methods will be required, e.g. the most commonly used dusty sorbent injection.
- Due to the comparable toxicity of arsenic and thallium to mercury and the European Union's environmental policy conducted in the future, standards for other ecotoxic elements may be expected to be introduced.
- Currently, there are no regulations regarding the emission of ecotoxic elements to air from coal coking processes.
- The emission of arsenic during the coke ejection process can amount to as much as 76.8 μg/m$^3$. This is a value much higher than the arsenic emission to the environment from combustion processes.

### Acknowledgment
Work carried out in the framework of the statutory AGH No. 11.11.210.373

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