

THE OCCURRENCE OF SELECTED TRACE ELEMENTS IN GRAIN FRACTIONS OF DUST EMITTED FROM POWER, COKE AND CEMENT PLANTS

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Abstract: The emission of dust from power and industrial sources introduces a lot of contaminants into the air, including compounds of trace elements contained in fuels and raw materials. They are contained in respirable dust particles, creating hazard to human health. The results of investigations into the occurrence of selected trace elements in PM₁, PM_{2.5} and PM₁₀ fractions of dust emitted from coal-fired boilers equipped with air protection devices such as cyclone, electrofilter, wet and dry-scrubbing FGD plant have been presented. Dust emitted from a coke battery (battery heating) and rotary kiln for cement manufacture was also subjected to research. The research material was taken by means of a cascade impactor, enabling a fraction of different grain size dust to be separated from a stream of dust collected in an electrofilter. The ICP-AES method (of atomic emission spectrometry (AES) with plasma excitation) was used to determine the trace elements after prior mineralization of samples by microwave method. The results of measurements and analyses were presented by determining the ranges of trace elements occurrence in flue dust and emission factors in PM_{2.5}. It was found out that big utility boilers and rotary kilns in the cement industry which are equipped with air protection devices meeting BAT requirements do not contaminate the air with dust and dust-related trace elements in the amounts that could create hazard. Excessive emission of dust, including a respirable fraction is still observed in the case of municipal heating plants equipped solely with mechanical dust separators (cyclones). Coke battery heating does not pose danger due to small range of influence.

INTRODUCTION

Poland and many countries belonging to the former eastern block saw their economies' restructuring, which caused liquidation or reduction of the technologically outdated and power-consuming branches of industry. The changes resulted in the emergence of new branches or modernization of raw materials branch and power industry, which have maintained their important position in the national economy and have good prospects. There



is a need to investigate the emission of dust and other contaminations from the currently operating units in power plants and municipal heating plants burning hard and brown coal, coke industry and cement industry. The previous studies are no longer useful due to technological progress and changes to the raw materials' base. Examples of changes which have taken place in the power industry include the improvement of coal quality, low-emission combustion technologies, flue gas desulfurization and high-efficient electrofilter; in the coke industry liquidation of outdated production facilities, better airtight sealing of coke batteries, waste gas dedusting as well as coke dry cooling, in the cement industry – widespread application of the dry production method, partial replacement of coal with waste fuel, higher efficiency of flue gas cleaning.

The aim of investigations is to provide information on the size and characteristics of the emitted dust, which is necessary to evaluate the threat to populations in industrial areas. In particular, it is important to investigate the share of respirable fraction dust and the content of trace elements compounds, including heavy metals. Twelve of them have been identified as hazardous air pollutants. They include: Sb, As, Be, Cd, Cl, Cr, Co, Pb, Mn, Hg, Ni and Se. Emitted into the air from natural and anthropogenic sources, they are subject to wet or dry deposition, exerting a harmful influence on biological functions in the human organism, where they penetrate chiefly through respiratory tracks, but also via the alimentary system, participating in particular links of food chain. The amount of emission is recorded on a national, continental and global scale, which requires continuous updating.

AIR CONTAMINATION WITH TRACE ELEMENTS' COMPOUNDS

Most trace elements are chemically very active and their low concentrations play an important role in the development and functioning of human organisms. However, in the case of higher concentrations they may inhibit biological processes or become toxic compounds. The higher solubility of a given metal salts, the higher probability of metal toxicity increase, which may lead to acute as well as chronic poisoning both as a result of exposure at a workstation and in the polluted environment.

Harmfulness of trace elements contaminating the environment in great measure results from their biochemical and biological properties, which influence the following phenomena:

- susceptibility to bioaccumulation:
 - from water environment, e.g. Hg, Cd, Pb, Cu, Zn, Sr;
 - from the soil, e.g. Cd, Zn, B, Sn, Cs, Rb;
- concentration in biolites due to geological processes, e.g. Ba, Be, V, Se, B, Ge, Rb, La, Ce, Pb, F, Zn, U;
- absorption from alimentary system, e.g. Hg, Cd, I, Zn, B;
- penetration through placenta to fetus, e.g. Cd, Hg, Pb, Zn;
- penetration through the biological barrier blood-brain, e.g. Hg, Pb, B, Al;
- formation of connections with sulfidrilic groups of proteins, e.g. Hg, Pb, Se, Cd;
- damaging of nucleic acids' chain (especially RNA and DNA), e.g. Cu, Zn, Cd, Hg, Ni [7].



The harmful effect that trace elements, in particular heavy metals which are natural micro-components of the earth crust, exert on biological functions is enhanced by their ability to move and accumulate between particular environment components. In most cases, trace elements are contained in the emitted dust, less frequently they occur as vapors or volatile compounds of these elements. Deposition processes usually take place when aerosols with metals contained in them are washed out with rainfall or when they drop due to the gravity force.

In densely populated, urbanized and industrialized areas the size of contamination emissions is mainly influenced by anthropogenic sources – facilities in which mineral resources and fossil solid fuels are used and processed. The dust emitted due to mechanical processes: grinding, classification, mixing and transport most frequently do not differ from the processed raw materials in terms of chemical composition. On the other hand, in high-temperature processes fine grains are enriched with trace elements. Vapors of some of them after cooling condense mainly on the finest dust particles, the specific surface area of which is the biggest [15]. This frequently results in the enrichment of fine particles by trace components' condensation, which has been confirmed on many occasions [3, 8, 16].

Dust emission from anthropogenic sources in Poland has been systematically abated due to economy modernization and improvement of waste gas dedusting technology. The structure of dust emission given in Table 1 [6] is characteristic of the countries which have recently followed the path of market economy.

Table 1. Emission of dust according to the type of activity in Poland in 2006

Type of activity	Emission in thousands of Mg
Total	447.5
Combustion processes in the sector of energy production and transformation	45.2
Combustion processes in the municipal and housing sector	179.0
Combustion processes in industry	49.1
Production processes	17.6
Extraction and distribution of fossil fuels	36.5
Road transport	55.3
Other vehicles and devices	12.7
Waste management	17.3
Agriculture	32.5
Other sources	3.2

Anthropogenic dust is mainly composed of mineral particles with the dimensions ranging from 0.1 to 100 μm – these are chiefly products of combustion processes (ash, soot, metal salts etc.) and other high-temperature processes (metallurgy, lime and cement industry etc.).

Fine particles of anthropogenic origin which form suspended dust (PM_{10}) pose the biggest threat to human health as they occur in high concentrations in densely populated



areas characterized by high industrial activity and due to the accumulation of metals having toxic properties and dangerous organic compounds, chiefly polycyclic aromatic hydrocarbons and dioxins. The major reason, however, is respirable fraction ($PM_{2.5}$) contained in suspended dust, which creates health hazard [2].

The inhaled dust deposits in pulmonary alveoli and penetrates into the blood. This is estimated to be the cause of many general diseases, lung function deficiencies, asthma, bronchitis and different lung, heart and other organs diseases. Children, who use up 50% more air than adults per body mass, are particularly exposed. In the USA children account for 25% population, but the share of children in asthma incidence reaches 40% (according to EPA) [2]. It is estimated that every year 15 000 infants die there for this reason [14].

The emission of selected trace elements in Poland is recorded and similarly to dust emission, it is observed to be falling (Tab. 2), which concurs with the global tendency (Fig. 1) [19].

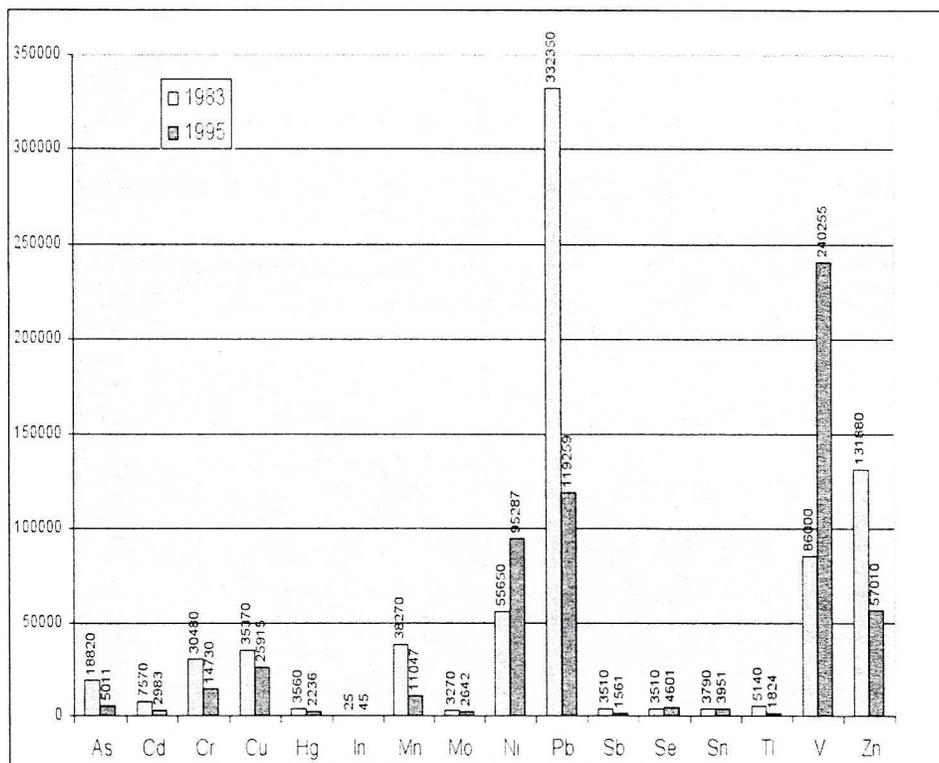


Fig. 1. Changes in the global emission of heavy metals [Mg/a]

In consequence, the relationship between the emission of dust and trace elements from anthropogenic sources reveals the effect that main economy branches using or processing big streams of resources and raw materials exert on the air quality. That is why utility, industrial and municipal boilers and coke and cement industry facilities have been chosen as the subject of investigations. The importance of these branches is confirmed by figures: Polish power industry uses 53.1 million Mg/a of hard coal and 60.2 million Mg/a

Table 2. Total emission of heavy metals in Poland in 2006 according to the type of activity

Specification	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
	Emission [Mg]							
Total	46.3	42.2	46.8	344.7	21.3	177.5	524.2	1303.2
Including:								
Professional power houses and CHPs	2.8	0.2	3.5	8.9	8.3	7.3	10.7	26.1
District heat plants	1.4	2.2	1.9	7.7	0.8	7.5	13.6	55.1
Municipal heat plants	1.0	1.6	1.4	6.0	0.2	5.4	10.4	41.5
Housing and services	13.1	20.1	15.6	68.9	1.0	66.2	115.9	465.2
Combustion processes in industry, boilers, turbines and engines	1.1	1.7	1.4	6.0	0.5	7.4	10.4	42.0
Production processes	1.0	2.6	9.3	18.3	1.4	6.8	90.1	171.4
Road transport	0.0	0.3	2.0	2.9	0.0	4.9	17.5	0.0
Waste incineration	0.0	0.1	0.0	0.1	0.1	0.0	1.4	0.9

of brown coal, coke industry products 9.7 million Mg/a, and the cement industry – 14.7 million Mg/a (the data provided by [5]).

Owing to the development of dedusting and waste gas cleaning technologies, the excessive air contamination, among others with dust and trace elements compounds contained in dust particles, may be effectively prevented. However, even the most effective dust removal devices are not perfect: separation efficiency for a given type of device drops as the dust particles decrease. As a result, when the total efficiency of electrofilter exceeds 99%, separation efficiency of dedusting a fraction with particle size distribution below 0.4 μm does not exceed 95%. In consequence, the mass fraction $\text{PM}_{2.5}$ in the emitted dust increases even up to 50%.

The efficiency of capturing a number of trace elements (in solid phase) is close to the total dedusting efficiency. In the case of some elements (manganese, beryllium, chromium), enrichment is not high, slightly higher enrichment has been observed for antimony, lead, nickel and selenium, whereas the highest level of enrichment was found in the case of arsenic and cadmium [21]. In fly ash captured in a bag filter, more trace elements were found compared to flue dust arrested in an electrofilter [4].

A relative drop in the efficiency of fine fractions dedusting is lower in the case of bag filters. However, even when PTFE membranes are applied, the efficiency of filtration drops as grain size decreases [1]. Widespread application of flue gas desulfurization has also contributed to dust emission reduction. An FGD plant, most frequently using the wet lime method, placed behind the electrofilter, at the same time serves as the second stage dedusting, the total efficiency of which reaches 80%. Behind the FDG plant the mass fraction $\text{PM}_{2.5}$ exceeds 90%, and the mass share of submicron fraction increases to 50%. However, this only refers to residual emission of flue gas, which accounts for maximum 0.05% of the total amount of ash produced in the process of coal combustion. Dust penetrating through FGD plant contains a considerable fraction (ca 50%) of gypsum and calcium. There is a proposal to call it *fly dust*, which better corresponds to the actual state than *fly ash*. Its composition slightly depends on the composition of coal mineral substance and the content of ash in coal [17]. Nevertheless, trace elements deposited on fine dust

particles penetrate and escape into the air [18]. The emission factors for low volatility elements depend directly on the efficiency of flue gas dedusting and desulfurization [23]. In the assessment of the impact exerted by power facilities using hard or brown coal, the emission of $PM_{2.5}$ fraction is of great importance due to the content of dangerous substances: PAH (Polycyclic Aromatic Hydrocarbons), dioxins and heavy metals, and due to this dust ability to penetrate into an organism via the respiratory tract.

With reference to utility and heating boilers, it may be stated that the application of an effective electrofilter and wet desulfurization radically reduces threat caused by the emission of trace elements [17, 20].

ASSUMPTIONS OF THE STUDY

The proved harmfulness and even toxic effect that the compounds of trace elements, in particular heavy metals, have on people, encourage investigations into the occurrence of these substances in dust emitted from power and industrial facilities. This refers to installations which have maintained their strong position in the national economy after a change of the economic system in Poland. They include utility boilers and municipal heating plants based on hard and brown coal combustion, coke and cement industry. The previous national studies devoted to these problems are out of date now due to restructuring changes in the economy, economic progress and change of raw materials base.

The assessment of the size and character of emission has been based on measurements and investigations into selected representative installations by analyzing the dust samples taken during full-scale industrial technological processes in production facilities.

In hard or brown coal-fired power and heating plants the air quality is influenced by fuel circulation, which includes fueling, flue gas desulfurization and cleaning, removal of fly ash, slag from under the boilers and waste from flue gas desulfurization installations. Waste gas emission is of crucial importance due to the size of stream and height of chimney dischargers, enhanced by the effect of dynamic and thermal upheaval. Application of dedusting devices (electrofilters and bag filters) the efficiency of which exceeds 99% allows dust concentration in waste gas to be reduced to maximum 50 mg/m^3 . In power plants which annually burn several to several dozen millions Mg of coal, the removed waste gas is cleared of sulphur dioxide by wet or dry scrubbing method, which results in a further reduction of dust concentration to several mg/m^3 .

The amount and composition of the compounds of selected trace elements contained in dust emitted from coal-fired boilers have been assessed by measuring basic parameters of waste gas as well as the amount and composition of dust penetrating through dedusting and desulfurization installation, at the sample-taking point (Tab. 3) placed behind these installations, before the waste gas is introduced into the atmosphere by chimney dischargers.

In the coke industry the flue gas from coke gas combustion for coke battery heating is emitted in an organized way through a chimney discharger having the height of several dozen meters. Considerable noxiousness for the environment is caused by emission, which results from the fact that the coke process is not fully hermetized.

The amount and composition of the compounds of selected trace elements contained in dust emitted from coke battery heating have been assessed by measuring basic parameters of flue gas as well as the amount and composition of dust at the sample-taking

Table 3. Characteristics of the investigated power/heating plants

Facility	Type of boiler	Type of furnace	Power	Fuel	Flue gas dedusting		Flue gas cleaning	
					DD ¹ P ³	η [%]	FGD ² P ³	η [%]
Heating plant Nowy Wirek	WLM 5	mechanical stocker	5.8 MW _t	hard coal	Cyclone P	89	none	
PEC Gliwice	WR 25	mechanical stocker	32 MW _t	hard coal	EF P	99	none	
PEC Gliwice	WP 70	pulverized-fuel fired furnace, tangential furnace	81 MW _t	hard coal	EF P	99	none	
Power plant Opole	BP 1150	pulverized-fuel fired furnace, tangential furnace	360 MW _e	hard coal	EF	99.6	wet lime P	95
Power plant Belchatów	BB 1150	pulverized-fuel fired furnace, tangential furnace	360 MW _e	brown coal	EF	99.6	wet lime P	95
Power plant Siersza	OP 380	pulverized-fuel fired furnace, tangential furnace	120 MW _e	hard coal	EF	99.6	dry-scrubbing FW P	87 > 99.6
Power-heating plant Nowa	OPG 230	pulverized-fuel fired furnace, tangential furnace	steam: 230 Mg/h	hard coal waste gases	EF P	99	none	

DD¹ – dedusting device, FGD² – flue gas desulfurization plant, P³ – sample-taking point

point (Tab. 3) before the outlet where flue gas is introduced into the atmosphere through chimney dischargers.

In the cement industry the main source of flue gas emission is a rotary kiln, where a raw mix is heated, carbonates decompose, and calcium oxide reacts with acid oxides and material sintering, which results in clinker synthesis. Furnace gases cleaned in high-efficient electrofilters, with the content of dust reaching a maximum of several milligrams in a cubic meter, escape into the atmosphere through a high chimney discharger. Grinding of sintered clinker with additives is the last procedure. Waste gas from mills is cleaned in high efficiency bag filters, and next introduced into the atmosphere through a low or medium height chimney discharger. The residual emission of dust produced in the process of clinker grinding as well as packaging and shipment of cement is of marginal significance.

The amount and composition of the compounds of selected trace elements contained in dust emitted from a rotary kiln for clinker have been assessed by measuring basic parameters of waste gas as well as the amount and composition of dust passed through an electrofilter at the sample-taking point placed behind the dedusting system, before the outlet where waste gas is introduced into the atmosphere through a chimney discharger.

AIM AND SCOPE OF THE STUDY

The aim of the study [11] is to extend knowledge on the sources and size of emission of selected trace elements that have been identified as dangerous air pollutants in Poland,

which in consequence will allow basing the national register of these substances' emissions on scientific grounds, thus making the continental and global inventory more precise. In particular, the aim of investigations is to identify a potential threat posed to people by respirable dust containing these substances, emitted from selected industrial facilities. The scope of the study includes selected trace elements in flue gas emitted by power boilers fired with hard and brown coal, from coke production installations (coke battery) and cement production facilities (rotary kiln). They include: antimony, chromium, cadmium, cobalt, manganese, nickel, lead, selenium and strontium.

CHARACTERISTICS OF THE INVESTIGATED INSTALLATIONS

Characteristics of the investigated power facilities

Investigations were carried out for utility power plant and municipal heating plant widely used in Poland. They include the following boilers, listed according to the increasing degree of emission reduction:

- hard coal-fired boiler with a mechanical stocker and cyclone for flue gas dedusting (Nowy Wirek heating plant),
- hard coal-fired boiler with a mechanical stocker and electrofilter for flue gas dedusting (PEC Gliwice WR 25),
- hard coal-fired boiler with a pulverized coal furnace and electrofilter for flue gas dedusting (PEC Gliwice WP 70),
- hard and brown coal-fired boilers with a pulverized coal furnace and electrofilter for flue gas dedusting as well as wet lime FGD plant (Opole and Bełchatów power plants),
- hard coal-fired boiler with a pulverized coal furnace and electrofilter for flue gas dedusting as well as dry scrubbing FGD plant and a bag filter to purge flue gas of dry desulfurization products (Siersza power plants),
- boiler fired with hard coal and waste gases with a pulverized coal furnace and gas burners equipped with electrofilter for flue gas dedusting (Nowa power-heating plant). The characteristics of these installations have been given in Table 3.

Characteristics of coke installations

The coke battery number 5 at Coke Plant Przyjaźń in Dąbrowa Górnicza was subjected to investigations. At that time it was the most modern coke battery in Poland. Coke Plant Przyjaźń is the second biggest Polish coke plant, producing over 25% of coke in Poland, its annual production capacity reaching 2.5 millions Mg of coke. The characteristic of this facility has been given in Table 4.

Characteristics of cement production installations

Dyckerhoff Polska Sp. z o.o. belongs to Buzzi Unicem Group, which has a leading position among building materials producers around the world. In Poland, where it sells over a million Mg of cement annually, it is included in the group of ten biggest cement producers. The investigations were carried out in a modern Cement Plant Nowiny near Kielce, belonging to Dyckerhoff Polska Sp. z o.o. The characteristics of this installation have been given in Table 5.

Table 4. Characteristics of coke battery number 5 in Coke Plant Przyjaźń

Specification	Unit	Size
Number of chambers	piece	2 x 38
Dimensions of cold chamber: length, height, width	[mm]	15040 5500 410
Chamber capacity	[m ³]	33.91
Capacity of coal charge in chamber	[m ³]	30.3
Dry coal charge in chamber	[Mg]	21.63
Coking cycle	[h]	15–16
Yield from 1 Mg of dry charge: dry coke, anhydrous tar, crude benzol, crude gas $W_d = 17.6 \text{ MJ/m}^3$ (conventional)	[kg/Mg] [m ³ /Mg]	767.1 36 010.4 355.0
Dry coke produced in chamber	[Mg]	16.59
Daily production of dry coke	[Mg]	1924.4
Annual production of dry coke	[Mg]	702400

Table 5. Technical and technological data of rotary kiln in Cement Plant Nowiny (Dyckerhoff)

Specification	Unit	Size
Length	[m]	65
Diameter	[m]	4.6
Slope	[%]	3
Kiln revolutions	[rev./min]	2.5
Production of clinker	[Mg/d]	2100
Fuel: coal, heavy oil	[Mg/h]	12 6
Stream of gases	thousands of m ³ /h	3200–3700
Initial temperature of gases	[K]	1473–1673
Oxygen content in gases	[% vol.]	8.5–11
Dedusting device		electrofilter
Dust emission	[kg/h]	1.14–2.33

METHODOLOGY

Measurement of the emitted dust grain size composition and sample taking

Grain size composition of the emitted dust was measured by determining the mass of dust arrested in particular stages of the cascade impactor, introduced into the flue gas conduit behind the dedusting device. A six-stage impactor Andersen Mark III with a final filter which captures the finest dust grains was used. The masses of separated grain fractions were measured by means of Mettler Toledo scales with 2 μg accuracy. In order to average the results for each facility and to compare granulometric composition of dust measured

in particular facilities, the results of all measurements were referred to the same, unified fraction ranges: < 1 , $1-2.5$, $2.5-10$ and > 10 μm . The method of measuring, interpreting the results and determining the emission factors of dust granular sizes has been described in a publication devoted to similar investigations [13].

Determination of trace elements in dust

Before trace elements were determined by ICP AES method, the samples were pulped in a mineralizer with focused microwave energy. Filter papers with the examined dust were placed inside a reaction vessel and 15 cm^3 of HNO_3 was added. After closing the vessel, the microwave system worked for 60 minutes, using 600 W output. The process temperature was 483 K, pressure in the autoclave was 15 bar. Next the samples were transferred to an induced plasma spectrometer with optical detection. During the analysis, optimized measurement parameters were applied to the spectrometer.

RESULTS

Emission factors EF_{PM1} , $EF_{PM2.5}$ and EF_{PM10} from the investigated facilities

On the basis of the measured mass streams of the total emitted dust and fuel or fuel chemical energy consumed as well as the product obtained (coke, cement), the emission factors EF_{PM1} , $EF_{PM2.5}$ and EF_{PM10} in kg/Mg were calculated. The emission factors are given in Table 6.

Table 6. Emission factors EF_{PM1} , $EF_{PM2.5}$ and EF_{PM10} from the investigated plants

Facility	EF_{PM1}	$EF_{PM2.5}$	EF_{PM10}
	[kg/Mg]		
Heating plant Nowy Wirek WLM 5	1.809	2.925	7.983
PEC Gliwice WR 25	0.124	0.223	0.344
PEC Gliwice WP 70	0.106	0.217	0.818
Power plant Opole BP 1150	0.023	0.046	0.197
Power plant Belchatów BB 1150	0.042	0.060	0.130
Power plant Siersza OP 380	0.002	0.004	0.120
Cement Plant Nowiny	BDL	0.020	0.078
Coke Plant Przyjaźń	0.009	0.019	0.028
	[kg/GJ]		
Power-heating plant Nowa OPG 230	0.013	0.018	0.088

BDL – below detection limit

Concentration of selected trace elements in the emitted dust

When measuring the emitted dust granular composition by means of a cascade impactor, 340 samples were obtained for trace elements determination. The mass of individual samples was small: from a part to a few mg. In order to ensure a necessary concentration of analytes, the samples containing grains within a similar diameter range: $0-1$ μm , $1-2.5$ μm and > 2.5 μm were mineralized jointly. The concentration of the investigated trace elements in granulometric compositions of the emitted dust has a very wide range. As an example, a full range of the measured concentrations [ppm] and a range limited to values

found in 80% of the examined samples of dust emitted from boilers have been presented in Table 7.

Table 7. A full range of concentrations measured [ppm] and a range limited to values found in 80% of samples of dust emitted from the investigated boilers

Element	Average	Median	Minimum	Maximum	P ₁₀	P ₉₀
	concentrations [ppm]					
Cd	7.16	BDL	BDL	44.10	BDL	28.03
Co	23.48	12.81	BDL	82.08	BDL	65.13
Cr	1169.59	575.49	51.99	7184.12	68.31	2054.30
Mn	648.96	246.08	BDL	3179.94	68.86	1450.62
Ni	2449.76	1048.33	BDL	16374.83	160.67	6177.76
Pb	74.94	30.06	4.66	514.04	6.71	235.66
Sb	63.77	BDL	BDL	421.04	BDL	270.79
Se	353.09	149.84	BDL	2366.48	BDL	696.15

BDL – below detection limit

Emission factors of selected trace elements from the tested facilities

The measured concentrations of selected trace elements were used to calculate emission factors of these substances from the examined installations. To this end, we used the emission factors of dust $W_{PM_{2.5}}$ (Tab. 6) determined on the basis of measurements taken in the investigated installations and the data on the content of a given trace element related to $PM_{2.5}$ fraction in the emitted dust, expressed in mg/kg. Trace elements content was calculated by multiplying $PM_{2.5}$ mass fraction in a unit of total dust mass by the concentration of a given trace element in $PM_{2.5}$ fraction contained in the emitted dust composition. The obtained results for five boiler installations have been given in Table 8. On the basis of dust emission factors and elements contents in $PM_{2.5}$ fraction, the emission factors (EF_i) for particular trace elements in respirable fraction – $PM_{2.5}$ emitted by boiler installations, coke battery heating facility and a rotary kiln (Tab. 9) were calculated. Emission factors

Table 8. Contents of selected trace elements in PM 2.5 emitted from the investigated boilers

Element	Heating plant Nowy Wirek WLM 5	PEC Gliwice WR 25	PEC Gliwice WP 70	Power plant Opole BP 1150	Power plant Siersza OP 380
	contents [$\mu\text{g/g}$]				
Cd	BDL	7.14	BDL	7.45	BDL
Co	16.42	22.49	1.62	12.31	0.31
Cr	547.49	393.53	217.15	102.79	22.31
Mn	347.39	74.35	68.05	191.23	16.92
Ni	712.61	610.12	331.07	137.46	63.84
Pb	35.80	16.80	4.79	30.53	3.90
Sb	118.21	23.92	8.77	BDL	BDL
Se	151.21	84.13	19.33	45.14	BDL

BDL – below detection limit

for Nowa power-heating plant were not determined as its boiler fired with such fuel has an individual character. Due to lack of more space in this study, data on trace elements' concentration in granular fractions of the emitted dust and mass granular fractions in a unit of total dust mass have not been included.

Table 9. Emission factors of selected trace elements contained in $PM_{2.5}$ emitted from the investigated plants

Element	Plants							
	Heating plant Nowy Wirek WLM 5	PEC Gliwice WR 25	PEC Gliwice WP 70	Power plant Opole BP 1150	Power plant Bełchatów BB 1150	Power plant Siersza OP 380	Cement Plant Nowiny	Coke Plant Przyjaźń
[mg/Mg]								
Cd	0.0	1.6	0.0	0.3	0.4	0.0	0.0	0.3
Co	48.0	5.0	0.4	0.6	0.7	0.0	0.4	3.1
Cr	1601.4	87.8	47.1	4.7	145.4	0.1	6.2	92.4
Mn	1016.1	16.6	14.8	8.8	59.2	0.1	4.6	33.5
Ni	2084.4	136.1	71.8	6.3	325.7	0.3	9.9	185.0
Pb	104.7	3.7	1.0	1.4	0.7	0.0	0.7	6.5
Sb	345.8	5.3	1.9	0.0	0.9	0.0	0.1	6.7
Se	442.3	18.8	4.2	2.1	22.6	0.0	1.0	13.6

DISCUSSION

In this study it has been assumed that the tested plants affect the environment due to the emission of this part of dust which is not arrested in flue gas dedusting installations and escapes into the air, carrying the deposited contaminants, among others trace elements. That is why the determined emission factors of dust and selected trace elements as well as technical factors influencing their size are noteworthy. Respirable dust is especially important due to its effect on people. It was found out that boilers fired with hard coal and not equipped with FGD plant emit dust with 12% to 20% of PM_1 fraction. $PM_{2.5}$ fraction accounts for 20–35%. In flue gas from boilers fired with brown coal, the PM_1 and $PM_{2.5}$ fraction is the biggest, reaching 32% and 46% respectively. The application of a filter bag in the case of dry-scrubbing desulfurization method considerably reduces the PM_1 fraction to 1% and $PM_{2.5}$ to 2%. A boiler fired with coal and metallurgical gases, equipped with an electrofilter (Nowa power-heating plant) was also subjected to investigations – it was found that PM_1 and $PM_{2.5}$ fractions were similar to those in the above discussed first group of boilers. This results from the dominant fraction of mineral substance contained in coal in the formation of fly ash in such a furnace.

Flue gas emitted due to battery coke firing is produced as a result of refined coke gas combustion. PM_1 and $PM_{2.5}$ fractions are high, reaching 31% and 67% respectively. It should be emphasized that flue gas from battery firing is not dedusted.

Flue gas from a rotary kiln in a Cement Plant is produced as a result of hard coal and used car tyres burning, which are utilized in this way, thus reducing the consumption of coal. Flue gas contains fly ash produced in the process of coal and tyres burning, as well as particles of clinker – a product and small amounts of pulverized mineral resource.

Flue gas is dedusted by means of a high-efficient electrofilter. $PM_{2.5}$ fraction reaches 25%, while PM_1 fraction is very small (below 1%).

The determined emission factors refer to different installations, which vary in their effect on the environment. Particularly important are emission factors from power and heating boilers which are used throughout the country and considerably affect the condition of air. It is easy to notice that big power boilers equipped with FGD plant are characterized by low dust emission factors. FGD plants provide the second stage of dedusting and together with electrofilters furnished behind the boiler effectively reduce dust emission. Application of a bag filter in desulfurization installations using dry-scrubbing method reduces the emission factors of PM_1 and $PM_{2.5}$ (respirable dust) to a very low value. Also boilers equipped with wet FGD installations have low PM_1 and $PM_{2.5}$ emission factors. A comparison of emission factors determined for a boiler equipped with a mechanical stocker, dedusted with a cyclone (heating plant Nowy Wirek WLM 5) and for a boiler with a mechanical stocker, dedusted by means of an electrofilter (PEC Gliwice WR 25) confirms a possibility to radically reduce dust emission with an electrofilter installed on a boiler with mechanical stocker. $PM_{2.5}$ emission factor is 13 times lower after a cyclone has been replaced by an electrofilter.

Emission factors of dust from a rotary kiln in the Cement Plant, which is fired with coal and used car tyres prove that modern electrofilters effectively protect the air against contamination. Respirable dust emission factors are low.

Low emission factors of dust produced in the process of coke battery firing indicate that emission from this source has a slight local effect.

In order to assess the scale of hazard to population due to emission of dust containing dangerous substances, the emission factors of selected trace elements from the tested facilities have been calculated. Similarly to the emission factors of dust granular compositions, considerable differences are observed in the case of emission factors for selected trace elements, depending on the type of furnace and air protection devices furnished on the boilers. The emission factors range from tenth parts of mg to a few grams in relation to 1 Mg of fuel burnt. For obvious reasons the highest values of emission factors have been recorded for a boiler with a mechanical stocker, where flue gas is dedusted by means of a cyclone. Much lower emission factors are recorded in the case of boilers equipped with electrofilters, low emission factors – for dust boilers with electrofilters and dry-scrubbing FGD plant, and the lowest – in the case of dust boilers with electrofilters and dry-scrubbing FGD plant with a bag filter. For the elements considered dangerous, the emission factors of respirable dust $PM_{2.5}$ [mg/Mg] have the following ranges: antimony 0–346; cadmium 0–1.6; chromium 0.1–1601; cobalt 0–48; lead 0–105; manganese 0.1–1016; nickel 0.3–2084 and selenium 0–442.

Emission factors of $PM_{2.5}$ produced in the process of coke battery firing are quite high considering the fact that gaseous fuel is burnt. On the other hand, one should bear in mind that flue gas emitted by a battery is not dedusted.

Emission factors of $PM_{2.5}$ produced in the process of clinker firing are quite low for chromium, manganese and nickel, their values reaching: 6.2; 4.6 and 9.9 mg/Mg of clinker.

EVALUATION OF POTENTIAL THREATS CAUSED BY THE EMISSION OF TRACE ELEMENTS CONTAINED IN RESPIRABLE DUST

When considering a potential threat posed by trace elements contained in respirable dust, a number of sources, i.e. dust emitting installations, activities of these sources and the prospects of their presence in the changing economies should be taken into account. There is a high probability that in Poland coal will continue to be used in professional and municipal power houses until the second half of the century. Power plants which burn hard coal are located throughout the country and several million people live within their range of influence. Coke plants are located in the central part of the Silesian Province, and the production of coke will probably decrease. Coke plants emit dust through medium height emitters or are a source of small range disorganized emission. The prospects of cement industry are stable. Due to high efficiency of flue gas dedusting the effect of modern cement plants on the environment is not big. That is why in the evaluation of potential hazards to the population; the attention was focused on boiler houses fired by hard coal. To this end, calculations of PM_{10} and $PM_{2.5}$ dust emissions from power plants, combined heat and power plants (CHP) and municipal heating plants were made on the basis of dust emission factors (Tab. 6) and previously described studies [12].

The aim of calculations is to prove that widespread application of modern flue gas dedusting electrofilters and wet FGD in power plants and CHP plants as well as replacing the cyclones with flue gas dedusting electrofilters in municipal heating plants may considerably reduce the emission of respirable dust and trace elements contained in it.

The calculations have been based on the following assumptions:

- boilers in all power plants and CHP plants are equipped with EF and wet FGD plant and show $PM_{2.5}$ emission factor equal to the one determined for power plant Opole;
- boilers in all municipal heating plants are equipped with cyclones and show $PM_{2.5}$ emission factor equal to the one established for heating plant Nowy Wirek WLM 5 (option 1);
- boilers in all municipal heating plants are equipped with EF and show $PM_{2.5}$ emission factor equal to the one determined for PEC Gliwice WR 25 (option 2).

The results of calculations given in Table 10 prove that out-of-date solutions in a form of boilers with a mechanical stocker and cyclone working as a dedusting device result in PM_{10} and $PM_{2.5}$ emissions disproportionate to the amount of coal burnt. PM_{10} and $PM_{2.5}$ emissions from these boilers are 9 and 7.5 times higher compared to the emissions in the subsector of power plants and CHP plants, burning nearly 9 times bigger amounts of coal.

Table 10. Estimated PM_{10} and $PM_{2.5}$ emissions from power plants and CHPs in Poland after applying more effective technologies of air protection

Type of source	Annual (2004) consumption of coal in millions of Mg	Air protection installation	Annual emission [Mg]	
			PM_{10}	$PM_{2.5}$
Utility boilers and CHPs	50.90	EF + wet FGD	1170.8	2341.5
Municipal heating plants Option 1	5.97	C	10799.7	17462.3
Municipal heating plants Option 2	5.97	EF	740.3	1331.3

Practical application of emission factors also enables an annual assessment of the emission of a selected element from a particular boiler installation. To this end, model calculations have been made. Table 11 quotes the calculated emission of selected elements from a boiler equipped with a mechanical stocker (WLM 5) and cyclone working as a dedusting device (using the example of heating plant Nowy Wirek) and from a pulverized-fuel fired boiler (BP 1150) equipped with EF as a dedusting device and wet FGD plant (example – power plant Opole). The conducted simulation of calculations has shown that basic (and obsolete in actual fact) boiler equipment with air protection devices results in excessive emission of dust and its dangerous components. As a result, a small boiler burning 15 000 Mg of coal in a season contaminates the air much more than a big boiler burning 600 000 Mg of coal. It has to be added that the calculations were based on the emission factors which were measured for technical conditions and fuel used in the above mentioned facilities.

Table 11. Seasonal emission of selected trace elements from two boilers with the older and technologically advanced installations of air protection

Pollutants	WR 25		BP 1150	
	Coal consumption [Mg/h]			
	5		200	
	Calorific value [MJ/kg]			
	21		23	
	Boiler efficiency* [%]			
	82		91.7	
Emission of selected pollutants				
	[mg/GJ]**	[kg/season]	[mg/GJ]**	[kg/season]
Cd	0	0	0.013	0.180
Co	2.286	0.720	0.026	0.360
Mn	48.386	15.242	0.383	5.280
Ni	99.257	31.266	0.274	3.780
Pb	4.986	1.571	0.061	0.840
Sb	16.467	5.187	0	0
Se	21.062	6.635	0.091	1.260

* – boiler efficiency is a measure of losses in the process of converting the chemical energy contained in fuel to heat energy,

** – chemical energy of fuel

The calculations refer to a period of 3000 h, which corresponds to a heating season. They confirm the conviction that obsolete boiler installations need to be promptly withdrawn and prove that the emission factors of dust and trace elements contained in it, which have been determined on the basis of measurements and analyses, allow us to initially estimate the size of the investigated substances' emission in the scale of the country and the effect of a particular type of emission source on the emission size. They may be used to assess the level of dangerous substances in the air and amounts of dangerous substances absorbed by people via respiratory tract.

In order to initially estimate the degree of people's exposure to the harmful or dangerous components of respirable dust, calculations were made on the basis of these substances' contents in dust (Tab. 8) and the level of respirable dust measured in a big town

in the Upper-Silesian Agglomeration. In a heating season the high level of $PM_{2.5}$ in the air is influenced by the emission of dust from numerous sources, also from municipal heat plants and professional power plants. Three options have been considered. In the option A an identical, definite amount of heat for the heating needs of a town is provided by a local heating plant using boilers with mechanical stockers equipped with cyclones as dedusting devices; boilers used in the option B are similar, but equipped with electrofilters; and in the option C the heat is supplied by a CHP plant using a boiler with a pulverized-fuel fired furnace equipped with an electrofilter and wet FGD plant. For the purposes of evaluation it is important to estimate correctly the level of respirable dust from the considered boilers in the ground layer of air.

For this purpose, the measurement and computational data from our own research and literature sources were used. In the previously unpublished investigations which were carried out in Bytom in the Upper-Silesian Agglomeration, a maximum annual concentration of $PM_{2.5}$ in the air, caused by the emission of dust from the local heating plant in this town was determined. It reaches $0.2795 \mu\text{g}/\text{m}^3$. These boilers with mechanical stockers are equipped with cyclones, so they correspond to the option A [13].

If these boilers were equipped with electrofilters in place of cyclones, and the activity of the sources, fuel, technical parameters of the heating plant as well as the conditions of waste gas emission and diffusion in the air did not change, then it might be assumed that in the option B the annual concentration of $PM_{2.5}$ in the air due to dust emission from a modernized local heating plant in this town would amount to $0.0213 \mu\text{g}/\text{m}^3$ (compare $PM_{2.5}$ emission factors in Table 6). The emissions and environmental impact of PM_{10} and trace elements from a modern coal-fired power plant equipped with ESP and wet FGD plant [17]. The option C evaluated the effect of a boiler with a pulverized-fuel fired furnace, electrofilter and FGD plant, which works in a combined system, produces heat (thus replacing the complex of local heating plant) and additionally produces electric energy for other recipients. It was assumed that the dust emitted from this boiler is responsible for $0.01 \mu\text{g}/\text{m}^3$ maximum annual concentration of $PM_{2.5}$ in the air on the territory of the discussed town [17]. To be precise, it has to be added that in the above quoted publication the concentration of dust is PM_{10} , in which the fraction of $PM_{2.5}$ is more than 90%. On the basis of the presented data, an average amount of $PM_{2.5}$ absorbed by a town inhabitant in a heating season may be estimated. If during 24 h the amount of inhaled air is 7 m^3 , and the heating system works at full power for 125 days, then in subsequent options the intake of $PM_{2.5}$ will reach 0.2446 mg (option A), 0.0186 mg (option B) and 0.0086 mg (option C). It must be remembered that in the option C only ca 30% of the boiler's capacity produces heat – the remaining power is used to produce electric energy for other recipients. If the existing outdated boilers are replaced by modern boilers with EF and FGD with required power, then the amount of $PM_{2.5}$ will be thirty times lower.

With regard to the amounts of selected trace elements in the inhaled air, it has to be stated that they are very small. This is confirmed by the example of nickel contained in $PM_{2.5}$ emitted from a boiler in the option A. The measured amount of nickel: $712.61 \mu\text{g}/\text{g}$ is the highest among all the examined elements in the considered three types of boiler installations. (Tab. 8) The concentration of nickel in the air due to boiler emission in the option A reaches $0.1992 \text{ ng}/\text{m}^3$, and a seasonal intake absorbed by a town inhabitant is $0.1743 \mu\text{g}$. In the case of the remaining technical options and elements, the amounts are smaller.

With reference to human health hazard, an important factor taken into consideration is the number of particles. According to the literature data [17], one gram of fly ash from coal combustion is composed of $6.35 \cdot 10^{10}$ particles. Hence, the seasonal amounts in mg which have been previously calculated for the three options, if expressed as a number of particles, are as follows: $15.5 \cdot 10^6$ option A $1.18 \cdot 10^6$ option B and $0.55 \cdot 10^6$ option C.

Certainly, this does not change the fact that the estimated amounts of $PM_{2.5}$ from power installations which are the subject of investigations are a small fraction of mass or the number of respirable dust particles inhaled by the population. Nevertheless, fly ash produced in the process of coal burning is 100 or even 1000 times more enriched with some trace elements compared to their average concentration in the earth crust. The presence of numerous toxic substances in dust is probably accompanied by the synergic effect.

Respirable dust comes from many sources, which should be subject of further investigations. A big role in the level of respirable dust is played by secondary dusting, which should be limited by taking better care of roads and streets. In the case of building, leveling and road works, waste utilization, mineral resources mining and other activities, the reduction of dust emission may be offered by technical, technological and logistic solutions as well as better organization of works.

CONCLUSIONS

It has been found out that big utility boilers equipped with air protection devices, fulfilling BAT requirements do not introduce dust and dust-related trace elements into the air in the amounts which could be considered dangerous.

Municipal heating plants using only mechanical dust collectors (cyclones) continue to pose danger. The conducted computational simulation has shown that this results in an excessive emission of dust and its dangerous components.

Application of electrofilters in place of cyclones in municipal heating plants may bring about a radical reduction of respirable dust emission and multiple reductions of selected trace elements.

It has been found out that rotary kilns used in the cement industry, which are equipped with air protection devices and fulfill BAT requirements, do not introduce dust and dust-related trace elements into the air in the amounts that pose threat to humans.

The calculated amounts inhaled via respiratory tract by people living within the range of influence of the outdated municipal heating plants may pose threat, given the synergic effect related to the simultaneous presence of dangerous substances. We should also take into consideration the emission due to coal burning in house stoves, which are quite common in unmodernized parts of towns.

New coke plants fulfill the air protection requirements related to emissions due to battery heating.

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WYSTĘPOWANIE WYBRANYCH PIERWIĄSKÓW ŚLADOWYCH WE FRAKCJACH ZIARNOWYCH PYŁU EMITOWANEGO Z INSTALACJI ENERGETYCZNYCH, KOKSOWNICZYCH I PRODUKCJI CEMENTU

Emisja pyłu ze źródeł energetycznych i przemysłowych powoduje wprowadzenie do powietrza wielu zanieczyszczeń, w tym związków pierwiastków śladowych zawartych w paliwach i surowcach. Znajdują się one w ziarnach pyłu respirabilnego, co stwarza zagrożenie dla zdrowia ludzi. Przedstawiono wyniki badań nad występowaniem wybranych pierwiastków śladowych we frakcjach PM₁, PM_{2,5} i PM₁₀ pyłu emitowanego

z kotłów spalających węgiel i wyposażonych w urządzenia ochrony powietrza takie jak cyklon, elektrofiltr, instalacje odsiarczania spalin metodą mokrą wapieniową i metodą pól suchą. Badano też pył emitowany z baterii koksowniczej (opalenie baterii) i z pieca obrotowego do wypalania cementu. Materiał badawczy został pobrany za pomocą impaktora kaskadowego umożliwiającego wydzielenie ze strumienia spalin odpylonych w elektrofiltrze frakcji pyłu o różnej wielkości ziarna. Do oznaczenia pierwiastków śladowych: Cd, Co, Cr, Mn, Ni, Pb, Sb i Se wykorzystano metodę atomowej emisyjnej spektrometrii o wzbudzeniu plazmowym (ICP-AES) po uprzedniej mineralizacji próbek metodą mikrofalową. Przedstawiono wyniki pomiarów i analiz określając zakresy występowania pierwiastków śladowych w emitowanym pyłu i wskaźniki emisji w $PM_{2,5}$. Stwierdzono, że kotły energetyczne o dużej mocy i piece do wypalania klinkieru w przemyśle cementowym wyposażone w urządzenia ochrony powietrza odpowiadające wymogom BAT nie wprowadzają do powietrza pyłu i pierwiastków śladowych związanych z pyłem w ilościach stanowiących zagrożenie. Nadal nadmierną emisję pyłu, w tym frakcji respirabilnej, wykazują komunalne obiekty energetyczne stosujące jedynie odpylacze mechaniczne (cyklony). Opalenie baterii koksowniczych nie stanowi zagrożenia ze względu na niewielki zasięg oddziaływania.