

# Estimation of heavy metal emissions from coalfired power plants in Russia

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#### Abstract

The idea of our study was to calculate trace element (Pb, Cd, Hg, Cu) emission into the atmosphere from coal combustion. Emission of the heavy metal was calculated using balance method based on the principle of element mass balance during coal firing. Experimental data of element concentration in the coal, slag and fly ash were adopted for this calculation. Also four others methodologies suggested by different authors were utilized for estimation of element emissions from energy coal combustion. Comparison of the different methods of calculation was made. As a result of this study total amounts of atmospheric heavy metal emissions on the territory of former Soviet Union were evaluated.

### **1** Introduction

Last decades more and more attention having been paid to the problem of ecosystem pollution by heavy metals. Many trace elements are registered now like global pollutants due to their toxic nature. Their negative influence to the environment is caused by accumulation in different ecosystem components and increased involving into biochemical cycles. Atmosphere is a main way of pollutant transport from emission sources to background territories where heavy metals are deposed onto water and plants. Heavy metal emissions into the atmosphere cause certain global environmental problems due to a long life-time of these elements, its long-term transport in the atmosphere and increasing rate of accumulation in the environment even at most remote territories.

Moreover, heavy metals have evidently entered to the human food chains, although an influence of global ecosystem pollution by heavy metals on human health is not well known yet.

The most part of trace elements comes into atmosphere with natural and man-made aerosols. The main sources of natural aerosols in the atmosphere are soil erosion and weathering of mountain rocks, volcanic and space dust, forest firing smoke and others. Major antropogenic sources of toxic elements are fossil fuel combustion, mining, industrial processes and waste incineration. The antropogenic flow of heavy metals to the atmosphere is about 94-97% of the total (Rovinskiy, et.al. [1]).

An inventory of emission sources should be the first step in developing of control strategy and modeling of global and regional cycles of trace elements. Experimental measurement are the most accurate methodology to determe pollutant concentration in industrial emission flows. This is absolutely necessary to investigate a problem of local pollutant effect on the surrounded areas. For the problem of long-term effect of heavy metal emissions from many industrial sources on large territory it is impossible to obtain experimental measurements of all sources and estimation methodology of heavy metal emissions from industrial sources are nessesary.

As a first step of total estimation of heavy metal emissions from industry coal-fired power plants were chosen. Calculation of element mass balance during coal combustion is a very difficult problem. There are many different kinds of coals used on power plants there as well as different technologies of coal firing. Many authors (Pacyna [1], Brumsuck [3], etc) applied a technique of emission factor for the calculation of element emission. Emission factor was defined as a mass of element in the emission per single fuel unit combustion. It is depended on both the type of fossil fuel and combustion technology. It is necessary to take into account type of a furnace and purifying efficiency. One of the general characteristic of coal firing technology is a type of slag removal: dry-bottom or wet-bottom (Brumsuck [3]).

There is no, unfortunately, a unified methodology of heavy metal emission calculation. There are few useful methods which are described in "The Atmospheric Emission Inventory Guidebook" (McInnes [4]) and usually are adopted and used in certain studies including calculation of heavy metal emission from Russian power plants. However, combustion technologies and coals used in Russia are strongly differed from European standard and the results seem to be doubtful.

The experimental data on heavy metal concentration in coal, slug and fly ash during coal combustion on Russian power plant were used for the calculation based on the principle of element mass balance. We have, also, calculated heavy metals emission using all well-known methodologies suggested by different authors and the comparison of the results was made.

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## 2 Methods of calculations

During coal combustion in power plants furnaces hard coal ash is removed both as a slag and as a fly ash. Further fly ash pass through purification equipment where a part of it is deposed. Efficiency of dust control equipment is varied from 85 to 99% with average for Russian power plants of 92%.

Thus, from 1 to 15 per cents of fly ash are not deposed on purification equipment, but emitted into the atmosphere as fine particles. There are a number of toxic elements in the coal ash. A part of these elements is emitted into the atmosphere with fine particles.

There are three main combustion technologies used in Europe: dry bottom boiler (pulverised coal), grate firing and fluidised bed boiler (MaInnes [4]). Pulverised coal is More close to the technology used in Russia. However, there are two types of boiler which are used on Russian power plants: Dry Bottom Boiler and Wet Bottom Boiler. Principle difference of these technologies is temperature of coal combustion. Combustion technology with low temperature (900 - 1100°C) corresponds to Dry Bottom Boiler and combustion technology with high temperature (>1400°C) - Wet Bottom Boiler.

#### 2.1 Element balance method

Presented calculation method is based on element mass balance during coal combustion. According to technical features of pulverised burning boiler 10-15% of coal ash is removed from boiler as slag ( $K_2 = 0.15$ ) and 85-90% leave boiler as a fly ash ( $K_1 = 0.85$ ) for dry slag removing. In the case of wet slag removing  $K_2$  and  $K_1$  are equal 0.40 and 0.60 correspondently.

Based on experimental data on heave metal concentration in coal ash, slag and fly ash Egorov et. al [5] offered to calculate emission factor as a difference between mass of element in coal ash and total mass of element in slag and fly ash,

$$R = Z (C_{\text{coal ash}} - K_2 \times C_{\text{slag}} - K_1 \times C_{\text{fly ash}}),$$
(1)

In this equation Z is the quantity of ash per 1 ton of coal (g),  $C_{\text{coal ash}}$  is element concentration in ash contained in coal,  $C_{\text{slag}}$  is element concentration in discharged slag,  $C_{\text{fly ash}}$  is element concentration in flying ash Equation (1) can be used for the calculation of total amount of element emissions into the atmosphere in the case of 100% efficience of purification equipment:

The part of element mass that comes from coal into slag and fly ash can be calculated using simple analitic equalations:

$$A = K_2 \times C_{\text{slag}} / C_{\text{coal ash}}$$
(2)

$$B = K_1 \times C_{fly ash} / C_{coal ash}$$
(3)

where A is a part of element removing with slag, B is a part of element which is adsorbed by the fly ash and deposed in the control equipment. In this case mass of element emitted with outflow gases can be calculated if the element amount in coal is known,

$$R_{el} = M (1 - A - B)$$
(4)

In this equation  $R_{el}$  is mass of element emitted with outflow gases per ton of burned coal, M is element mass per ton of coal.

Previous equations (1-4) are correct if the efficiency of purification equipment is 100%. A real efficiency is varied from 85 to 99% and an average efficiency of purification equipment used on Russian power plants is 92%. Thus, a part of flying ash is released into atmosphere as aerosols. K<sub>1</sub> coefficient was recalculated for different values of control equipment efficiency.

It is known that the part of coal ash. The total amount of ash that left boiler as a fly ash is equal  $K_1$ . Only a part ( $\eta$ ) of it is catched by a control equipment. The other (1- $\eta$ ) is emitted into the atmosphere.

$$K_1 = K_{1,1} + E$$
 (5)

$$\frac{K_{\perp}}{E} = \frac{\eta}{1 - \eta} \tag{6}$$

in these equations  $K_{1,1}$  is a part of fly ash catched by control equipment, E is a part of fly ash emitted into atmosphere,  $\eta$  is an efficiency of control equipment. From the equations (5) and (6),

$$K_{1,1} = K_1 \times \eta \tag{7}$$

Modified values of  $K_{1,1}$  coefficient are presented in the Table 2.1.1.

Table 2.1.1: Part of fly ash catched by control equipment for different efficiency of control equipment.

Type of	Control Equipment efficiency, (%)					
Boiler	85	92	99			
Dry Bottom	0.72	0.78	0.84			
Wet Bottom	0.51	0.55	0.59			



Using equation (3) coefficient B can be recalculate,

$$B = K_{1,1} \times C_{\text{fly ash}} / C_{\text{coal ash}}$$
(8)

Results of coefficient B calculation, presented in Table 3.1.2. were used for the calculation of element mass emitted into atmosphere according to equation (4).

#### 2.2 Calculation methods used for the comparison

Others calculations of heavy metal emission from coal-fired power plants were made using methodologies suggested by different authors. The results of these computation were used for the comparison of different calculation methodologies. There are three different methodologies presented in McInnes [4] for determination of heavy metal emission from coal-firing power plants. They are based on

1. fuel composition,

2. fly ash composition,

3. fly ash concentration in clean gas.

The choice of the methodology depends on data availability. Calculations further were made according to the 1-st and 2-d methodologies. Available data were not enough to determe emission factor according to 3-d methodology.

The calculation according to the first way of emission factor determination was produced using different values of main coefficients. Meanings of such coefficients as fraction of ash leaving the combustion chamber as particulate matter, enrichment factor and fraction of heavy metals emitted in gaseous form were taken from 1.1 McInnes [4], 1.2 experiment Egorov et.al. [5] and 1.3 Shpirt [6]. We have also calculated heavy metal emission according to the second way of emission factor determination using different meaning of main coefficients, such as fly ash emission factor of raw gas and heavy metal concentration in fly ash of raw gas, from 2.1 McInnes [4], 2.2 experiment Egorov et.al. [5] and 2.3 Pacyna [2]. Pacyna [2] calculated emission factor taking into account energy production instead of mass of fired coal used in all others methods. In addition we have also calculated heavy metal emission using emission factors suggested by Shpirt [6].

## 3 Materials and data

#### 3.1 Experimental data

An experimental data presented by A.P.Egorov et al.[5] were used in this study. The concentrations of heavy metals (lead, vanadium, chromium, nickel, cooper and arsenic) were determined in hard coal (received from DONBASS and KUZBASS), removed slag and flying ash set down in the control equipment. Average contents of different metals in coal ash, slag and fly ash precipitated

on the control equipment are presented in Table 3.1.1. According to the data from Table 3.1.1. coefficients A and B from equation (2) and (8) were calculated. Results are presented in the Table 3.1.2.

Table 3.1.1: Average contents of different metals  $(n \times 10^{-4} \% \text{ of mass})$  in coal ash (CA), slag (S) and fly ash precipitated on the control equipment (FA).

Element	СА	S	FA			
	Dry Botto	om Boiler				
Cu	196	122	131			
Pb	173	40	66			
Wet Bottom Boiler						
Cu	138	97	115			
Pb	Pb 126		55			

Table 3.1.2: Part of element adsorbed by the fly ash deposing on the control equipment (coefficient B) and removing with slag (coefficient A) for different metals.

Element	Efficienc	y of control equi	pment, %	Coefficient A
	85			
Cu	0.48	0.09		
Pb	0.27	0.32	0.04	
Cu	0.42	0.28		
Pb	0.22	0.24	0.26	0.11

#### 3.2 Data base on power plant

There were about 300 power plants which worked in frame of Energy Ministry of USSR in 1990. They produced more than 95% of total energy production. Only 128 of them used coal as a fuel. Distribution of coal combustion and electric energy productions by power plants with different capacity is presented in Table 3.2.1.

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Table 3.2.1: Coal consumption and electric energy productions by power plants with different capacity.

Parameters		Electric capacity, MW								
	>1200	800-	400-	200-	<200	Total				
		1200	800	400						
Number of stations	31	5	40	48	4	128				
Coal consumption, Mt	81.16	6.16	38.3	21.47	0.61	147.71				
Coal-fired energy production, PJ	1359.51	93.43	398.24	225.74	12.34	2094.59				

#### 3.3 Data on element content in coals

Data on element content in coals of different coal-fields were presented by Shpirt [6]. Average element content in coal were used. They were 7 g/t for Pb, 0.5 g/t for Cd, 0.05 g/t for Hg and 10 g/t for Cu.

#### 4 Results

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The results of calculations by suggested balance method are presented in Table.4.1. Emission of Pb and Cd was calculated for different control equipment efficiency (CEE), but only one value of emission factor was used for mercury. We believed that the 90% of mercury contained in coal released in the atmosphere as a gas phase vapour and that this value is not depend on control equipment efficiency.

Heavy metal emission was also calculated according to the methodologies described in part 2.2 of this paper. Emission factors used for this calculation are presented in Table 4.2 and results are presented in Tables 4.3. and 4.4.

### 5 Conclusions

Heavy metals emissions from coal fired power plants were calculated according to the suggested element mass balance methodology and well-knowen methodologies suggested by different authors.

Heavy metals emissions calculated according McInnesa [4], Pacyna [2] and Shpirt [6] are very strongly depend on control equipment efficiency. Emissions of elements from power plants with 85% purification efficiency are 15 times higher than for  $\eta$ =99%.

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Table 4.1: Pb, Cd and Hg emissions to the atmosphere from coal-firing power plants with different electric capacity according to element mass balance calculation methodology (t/y)

CEE,	Elect	ric capac	ity of pov		Total			
%	>1200	800-	400-	200-	<200	Mean	Min	Max
		1200	800	400				
Pb								
85	357.92	27.16	168.93	94.69	2.7	651.4		
92	346.56	26.3	163.57	91.68	2.61	630.72	413.6	816.8
99	329.51	25.01	155.53	87.17	2.48	599.7	-	-
Нg								
-	1.46	0.11	0.69	0.39	0.01	2.66	-	-

Table 4.2: Emission factors (g/t) used for calculation of Pb and Cd emission according to different methodologies.

Calculation methodology	CEE		Pb		Cd		
is based on	%	mean	min	max	mean	min	max
balance methodology	85	4.41	3.08	5.6	-	-	-
balance methodology	92	4.27	2.8	5.53	-	-	-
balance methodology	99	4.06	2.52	5.46	-	-	-
fuel composition	85	4.9	4	5.9	0.35	0.23	0.46
fuel composition	92	2.6	2.1	3.1	0.18	0.12	0.24
fuel composition	99	0.31	0.25	0.39	0.02	0.01	0.03
fly ash composition	85	0.98	0.95	1	-	-	-
fly ash composition	92	0.52	0.5	0.54	-	-	-
fly ash composition	99	0.065	0.063	0.068	-	-	-
Shpirt [6] coefficients	85	5.3	3.2	6.6	0.38	0.22	0.48
Shpirt [6] coefficients	92	3.1	1.4	5.6	0.22	0.15	0.3
Shpirt [6] coefficients	99	0.42	0.21	0.7	0.04	0.02	0.05

Values of emissions calculated according to coal composition, fly ash composition and Shpirt [6] coefficients for different CEE have dispersion of 38 - 42%, 7.8-7.2% and 66-72% correspondently.

Results of balance methodology calculation are very closly correspond to other values of heavy metals emissions in the case of low control equipment efficiency. However, the influence of purification equipment on atmospheric emissions of trace elements is much less in the case of mass balance calculation than in previous methodologies.

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CEE,	Elect	tric capacity of power plant, MW			MW		lotal			
%	>1200	800-	400-	200-	<200	Mean	Min	Max		
		1200	800	400						
	Ca	lculation	methodol	ogy base	d on fuel	composit	ion			
85	397.69	30.18	187.7	105.21	3.0	723.78	595.6	877.1		
92	211.02	16.01	99.6	55.82	1.59	384.05	307.7	462.4		
99	25.16	1.91	11.88	6.66	0.19	45.79	37.2	57.1		
	Calc	ulation n	nethodolo	gy based	on fly as	h compos	ition			
85	79.54	6.04	37.54	21.04	0.6	144.76	140.3	147.7		
92	42.2	3.2	19.92	11.16	0.32	76.81	73.9	79.8		
99	5.28	0.4	2.49	1.4	0.04	9.6	9.3	10.0		
	Calcu	lation me	ethodolog	y based c	n Shpirt	[6] coeffi	cients			
85	433.48	32.9	204.6	114.68	3.27	788.92	472.7	974.9		
92	248.84	18.88	117.45	65.83	1.88	452.88	206.8	827.2		
99	329.51	25.01	155.53	87.17	2.48	599.7	-	-		
	Ca	alculation	methodo	logy sug	gested by	Pacyna [	[2]			
-	59.76	4.31	16.89	8.03	0.15	89.2	82.9	115.6		

Table 4.3: Pb emission calculated according to different methodologies.

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Table 4.4: Cd and Hg emissions calculated according to different methodologies.

CEE,	Elect	Electric capacity of power plant, MW To					Total	
%	>1200	800-	400-	200-	<200	Mean	Min	Max
		1200	800	400				
				Cd				
	Ca	lculation	methodol	ogy base	d on fuel	composit	ion	
85	28.41	2.16	13.41	7.51	0.21	51.7	33.97	67.95
92	14.61	1.11	6.9	3.86	0.11	26.59	17.7	35.45
99	1.62	0.12	0.77	0.43	0.01	2.95	1.48	4.43
	Calcu	lation me	thodolog	y based c	on Shpirt	[6] coeffi	cients	
85	30.84	2.34	14.56	8.16	0.23	56.13	32.5	70.9
92	17.86	1.35	8.43	4.72	0.13	32.5	22.2	44.3
99	3.25	0.25	1.53	0.86	0.02	5.91	3.7	7.4
	Calculation methodology suggested by Pacyna [2]							
-	0.42	0.03	0.12	0.06	0.001	0.628	0.502	0.754
	Hg,	Calculati	on metho	dology sı	uggested	by Pacyn	a [2]	
-	0.42	0.03	0.12	0.06	0.001	0.628	0.502	0.754

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#### Air Pollution

Emissions of elements from power plants with 85% purification efficiency are only 8% higher than for  $\eta$ =99%. Dispersion of results for mass balance calculations is about 60%.

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