Macroscale estimate of an anthropogenic lead emission in the territory of Russia

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We have calculated the amount of lead emission from vehicles and power stations (thermal power stations (TPSs) and state regional power stations (SRPSs)) operating on coal in the territory of Russia in 1990. The emissions from vehicles were estimated based on the amount of consumed gasoline doped with lead (68–80% of the total consumed gasoline). Lead emissions from power stations were calculated based on the performance characteristics of the equipment of the stations using the lead emission coefficients for the combustion of fuel of various types. The theory of mass balance was used to calculate these emission coefficients. The obtained results, though uncertain to some extent, can be used to estimate the pollution loading in large territories.

Introduction

The progression of the modern world cannot be conceivable without use of metals; however, simultaneously with this, the problem of waist recovery becomes more and more urgent. From year to year an increasing amount of toxic gases and aerosols is emitted by enterprises and enters the environment through the atmosphere. Being gradually spread in the atmosphere, toxic gases and aerosols reach the territories far from the pollution sources and inevitably penetrate the trophic systems of animals and humans. Many base metals, because of their toxicity, are recognized as global pollutants. Their participation in biochemical cycles, accumulation, and redistribution within individual components of the environment against the background of significant anthropogenic emissions disturb the natural balance in the biosphere and pose threats to the environment and humans. In this connection, much recent attention has been given to the problem of pollution of ecosystems and the biosphere by the base metals.

One of the most toxic metals, most dangerous for the environment, is the lead. The amount of lead entering the air basin from anthropogenic sources is less than its amounts entering water areas and soil; however, the emission of base metals into the atmosphere causes ecological problems not only on local, but also on regional and global scales because of long-range transport of pollutants from the emission sources and their subsequent sedimentation and accumulation in soil and water.

The lead enters the atmosphere with aerosols of natural and anthropogenic origin. Relative contributions of the main sources of lead, in percent, are illustrated by Fig. 1 based on the data of Refs. 1 and 2. Vehicles are the main anthropogenic sources of lead. According to global estimates,[1–5] up to 210–250 thousand tons of lead enter the atmosphere per year in the form of highly dispersed aerosols in the combustion of gasoline in vehicles. This makes 62–76% of its total anthropogenic emission and exceeds almost 18 times its natural content in the atmosphere.

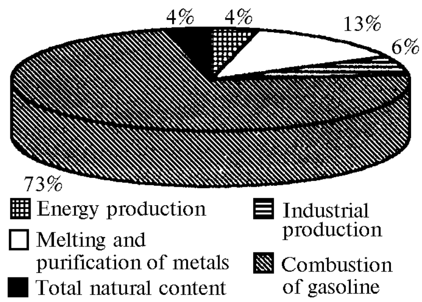

Fig. 1. Main sources of atmospheric lead.

The Pb emission by the enterprises of electric power industry is about 4% of its total content. However, in spite of the fact that in the combustion of fuel only microamounts of lead enter the atmosphere, even such small loading may bring significant threat to natural ecosystems and public health. In addition, because aerosols are discharged into the atmosphere from high smoke stacks (higher than 100 m), they are spread over large territories and hence determine the global and regional background levels of pollution in case of long-range pollutant transport in the atmosphere. However, an analysis of the existing statistical data[6–9] has revealed that insufficient attention is given to the emissions of base metals by enterprises of electric power industry, and the amounts of emissions of these substances are obviously underestimated.

When estimating the regional level of air pollution by the base metals, it is impossible to evaluate experimentally the contribution of each source. Therefore, a problem arises to use approximate methods for estimating concentrations of microelements in emissions. In the present paper, methods for calculating the atmospheric lead emission in the combustion of coal at power stations and of gasoline in vehicles are considered, and the results of corresponding calculations are presented.

Lead emissions by the enterprises of electric power industry

Unfortunately, now there is no universal technique for calculating emissions of the base metals by industrial enterprises. Therefore, to evaluate the atmospheric emission of base metals in the combustion of coal at enterprises of electric power industry, we developed and used a method based on a balance of element mass in the combustion of coal, that is, on the difference of the element mass shoveled in a furnace with coal, removed from the furnace with cinders, and caught by ash filters.

In the combustion of fuel in boilers of power stations, a part of ashes comprised in the coal shoveled into the furnace goes to cinders and is then removed from the boiler; one more part leaves a combustion chamber as volatile ashes and is partly precipitated on electrofilters. The efficiency of electrofilters of the power stations of the USSR varied from 85 to 99%, with average efficiency 92%. Thus, not all ashes are precipitated on the filter; from 1 to 15% of volatile ashes, depending on the filter efficiency, are emitted as solid particles into the atmosphere together with superheated gases. In the combustion of fossil coal, significant part of toxic microelements comprised in ashes is precipitated on solid particles and is subsequently emitted into the atmosphere.

The parameter that characterizes the amount of an element entering the atmosphere in the combustion of a fuel is the emission factor k, which depends on fuel type and regime of fuel combustion and specifies the mass of an individual pollutant comprised in the emission when a unit mass of fuel has been burnt out. Hence, for power stations operating on coal, it is necessary to take into account types of furnaces, filters, and smoke stacks. Difficulties associated with balancing of the element mass in the fuel combustion at a thermal power station (TPS) are as follows: first, consumed coals and hence volatile ashes have different chemical compositions; second, different regimes of coal combustion are used.

Under the auspices of the Ministry of Energy, about 300 power stations were in operation in the USSR in 1990. It is extremely difficult to evaluate the operating efficiency of these enterprises and their environmental effects using any classification or unification, because the data on the equipment, fuel, regimes of fuel combustion, and filtering of emissions are practically lacking or are available only as individual instructions and references in different reference books and reports. Based on an analysis of the available statistical and performance characteristics, we established that the main regime of fuel combustion in the territory of the USSR was coal-dust heating with liquid or solid (dry) cinder removal. It should be noted that the method of cinder removal is the salient feature of different combustion regimes. In particular, this is manifested in the proportion between cinders and volatile ashes, which in case of dry (solid) cinder removal are in the ratio 10–15% of cinder to 85–90% of ash, whereas in case of liquid cinder removal they are in the ratio 30–40% and 60–70%, respectively. For convenience, below we use the coefficients K_1 and K_2 that specify the fractions of coal ashes in the form of volatile ashes and cinders, respectively.

Taking into account that the proportion between ashes and cinders is the characteristic parameter for each combustion regime and using simple analytical formulas, based on the experimental data on the base metal concentrations in ashes of the consumed coal ($C_{\text{coal ash}}$), cinders (C_{cinder}), and volatile ashes ($C_{\text{vol ash}}$), borrowed from Ref. 10, we calculated the fractions of base metal comprised in cinders (A) and volatile ashes (B)

$$A = K_2 \times C_{\text{cinder}} / C_{\text{coal ash}},\ B = K_1 \times C_{\text{vol ash}} / C_{\text{coal ash}}. \quad (1)$$

With the known content of an element in the consumed coal, its amount (R_{el}) emitted into the atmosphere with ashes of outgoing gases was calculated, disregarding the efficiency of filters, by the formula

$$R_{\text{el}} = M(1 - A - B), \quad (2)$$

where M is the element mass comprised in the consumed coal. To calculate the realistic emission factor for an element, the efficiency of filters (η) must be introduced:

$$k_{\text{el}} = R_{\text{el}} \times (1 - \eta). \quad (3)$$

In our calculations the average values of the parameters A and B for lead, according to the data of Ref. 10, were set equal to 0.04 and 0.35, respectively, and $R_{\text{el}} = 0.61M$. The calculated lead emissions are given in Table 1 as functions of the filter efficiency.

Table 1. Average annual lead emissions (in t/yr.) by power stations as functions of their output electrical power.

Parameter	Output electrical power, MW					Total
	> 1200	800–1200	400–800	200–400	< 200	
Number of power stations with the indicated output electrical power	31	5	40	48	4	128
Total mass of consumed coal, Mt	81.2	6.2	38.3	21.5	0.6	147.7
Produced electrical energy, Billion kW·h	377.9	26.0	110.7	62.7	3.4	580.7
Average annual Pb emission by a single power station with the given output electrical power as a function of the filter efficiency, t/yr.						
85	11.55	5.43	4.20	1.97	0.67	
92	11.18	5.26	4.09	1.91	0.65	
99	10.63	5.00	3.89	1.82	0.62	

Estimate of lead emission by vehicles

Procedures developed in the USSR for calculating the emission of harmful substances from vehicles, including lead, are based on the use of specific atmospheric emissions of substances per unit run of automobiles of different classes.[11–13] Their use is convenient and justified when emissions of harmful substances are evaluated in a small territory (town, settlement, etc.), where fairly reliable data can be collected on the intensity of traffic. Unfortunately, already for a region the reliability of statistical data used for calculations casts some doubt. To obtain large-scale estimates of atmospheric lead emissions in large territories, it is most convenient to calculate the amount of consumed fuel with the use of lead emission factors. In so doing, the fact is used that the lead is not produced in the combustion of gasoline, but is doped to it and is emitted into air in proportion to the amount of consumed fuel. The given approach has already been used for a long time for estimating the atmospheric lead emissions from vehicles both abroad and in our country.[1,14,15] Just it was used in the present paper.

The input data on the consumption of automobile gasoline were the data on the fuel balance in the territory of the USSR (collected for republics, krai, and regions), according to which the total consumption of automobile gasoline in 1990 was 61 271 thousand tons. To calculate the atmospheric lead emission factor, it is necessary to know its content in gasoline and its behavior while the automobile motor is running.

Practically all foreign authors assumed that, as a whole, 75% of lead comprised in gasoline is emitted into the atmosphere.[1,16] According to experimental studies, about 80% of lead mass comprised in gasoline enters the air with exhaust gases; the remaining lead is precipitated on electrodes, insulators, and exhaust systems of automobiles. These data are valid only for automobiles with runs of up to 15 000 km (first year of running). For automobiles with greater runs the emission of lead with exhaust gases is increased up to 90% of its amount comprised in gasoline because of reduced efficiency of its sedimentation on exhaust systems of automobiles and partial detachment of its accumulations.[11,17]

In the USSR the automobile fleet comprised 51 automobiles per 1000 inhabitants in 1990, that is, there were 14.8 million automobiles[18] (A_t). In 1989, 1217.6 thousand automobiles (A_f) were produced, and their average runs in 1990 were less than 15 thousand km (see Ref. 19); hence, the weighted-mean lead emission factor for vehicles (if we take the fleet renewal factor in 1990 in the same proportions for all regions) is

$$K_e = [0.9(A_t - A_f) + 0.8A_f]/A_t = 0.89. \quad (4)$$

The lead is emitted into the atmosphere with the exhaust gases in the form of aerosol particles, up to 80% of which have diameters less than 5 μm and do not settle on the ground for a long time.[11,20] Hence, to evaluate the fraction of lead emission that participates in the atmospheric transport and dispersion, it is necessary to introduce the sedimentation factor $K_s = 0.8$.

Exhausts in the combustion of gasoline are caused by wide application of lead dopes, being antidetonation agents, to produce automobile gasoline with high-octane number. According to the data published in the literature (see Refs. 21–23), the amount of consumed fuel (in thousand tons) was calculated for each type of gasoline, and then for its part subject to ethylation. In so doing, the percentage of lead comprised in AI gasoline was refined. The results are summarized in Table 2 (the percentage of ethylated gasoline for a given gasoline type is indicated in parentheses).

Table 2. Structure of gasoline consumption in the USSR in 1990.

Gasoline type	Characteristic	Lead content, g/liter	Consumption, thousand tons	
			Total	Including ethylated
A-72	Nonethylated	0.013	3554	0 (0%)
A-76	Nonethylated	0.013	49139	35396.6 (72)
	Ethylated	0.170		
AI-93	Nonethylated	0.013	8578	4548.5 (53)
	Ethylated	0.370		
AI-95	Nonethylated	0.013		
Total			61271	39945.1 (65)

The total content of lead in gasoline consumed in the territory of the former USSR in 1990, calculated from the data of Table 2, was $Q = 10{,}781$ t (for an average gasoline density of 0.74 kg/liter, being unnormalized parameter[24]). Hence, the total atmospheric lead emission due to gasoline consumption in the territory of the USSR in 1990 is

$$Q_a = Q_\mathrm{r} K_e K_\mathrm{s} = 7676 \text{ t}. \qquad (5)$$

The spatial distribution of lead emission from vehicles in the territory of the USSR was evaluated for 150×150 km squares of the extended EMEP grid in proportion to the intensity of traffic. For this purpose, in squares of the grid the intensity of traffic was evaluated in balls, from the density and importance of the automobile roads[25] (see Table 3).

For each administrative region, the spatial distribution of consumed fuel was calculated by the formula

$$Q_\mathrm{sq} = Q_\mathrm{r}\, B_i\, P_i \Big/ \left(\sum_j B_j\, P_j \right), \qquad (6)$$

where Q_sq and Q_r are amounts of gasoline consumed in squares inside the region and in the entire region, respectively; B_i is the intensity of traffic, in balls; P_i specifies the fraction of the ith square area that belongs to the examined region; the subscripts j denote summation over the squares that belong to a given region. The atmospheric lead emission factors differ for the territory of the USSR owing to different types of gasoline consumed in different regions. Practically complete lack of data and estimates in this respect has forced us to assume that all nonethylated gasoline (except for A-72 gasoline), purer from the ecological viewpoint, is consumed by modern motors, irrespective of gasoline types, in squares with the intensity of traffic $B = 3$ to 5 balls, whereas all AI-93 high-octane gasoline – in squares with $B = 4$ to 5 balls. Based on these assumptions, the weighted-mean atmospheric lead emission factors for vehicles were calculated separately for squares with different intensities of traffic. The results are summarized in Table 4.

Table 3. Criteria for estimating the density of automobile roads.

Ball (B)	Criteria
0	The data for this territory are lacking or in the given square there are no paved roads
1	There are at least one to two state or republican roads, without state highways
2	There is at least one state highway or rare network of other state and republican roads
3	There are one to two state or republican highways with rare network of other state and republican roads or dense network of state and republican roads without highways
4	There are one to two state highways converging toward a town and other state and republican roads or one state highway with a dense network of state and republican roads
5	There are two or more state highways converging toward a city and other state and republican roads or two to three small centers of convergence of state highways and other state and republican roads in one square

Table 4. Results of calculating the parameters of lead emission from vehicles.

Intensity of traffic	Gasoline consumption, thousand tons	Presence of nonethylated gasoline	Lead emission factor, g/t	Total atmospheric lead emission
1–2	16789.6	No	160.0	2686
3	21306.6	Yes	72.8	1551
4–5	23174.8	Yes	148.4	3439

Results and conclusions

We have calculated the atmospheric lead emission in the territory of the USSR in 1990 from two main anthropogenic sources: in the combustion of gasoline in vehicles and coal at power stations. In the present paper, the main attention has been given to the distribution of the atmospheric lead emission over different economic regions of Russia, which is illustrated by Figs. 2 and 3. For Siberia (see Fig. 4) the distribution of lead emissions over administrative regions is illustrated.

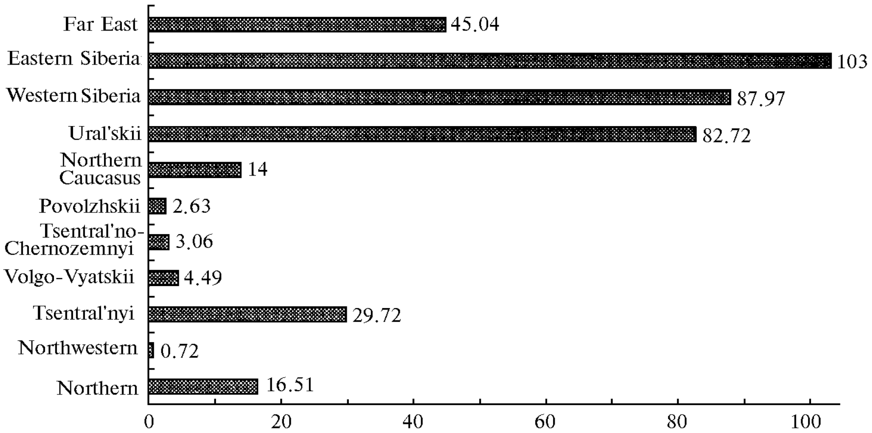

Fig. 2. Distribution of atmospheric lead emissions in the combustion of coal at the enterprises of electric power industry over the economic regions of Russia (t/yr.).

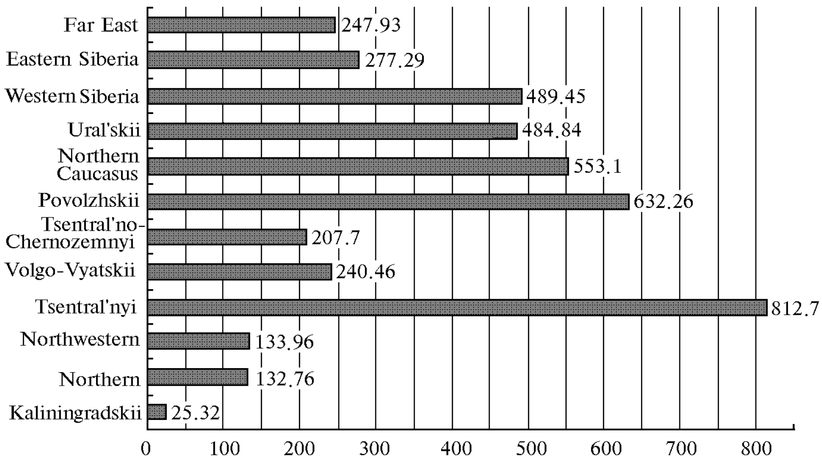

Fig. 3. Distribution of atmospheric lead emissions from vehicles over the economic regions of Russia (t/yr.).

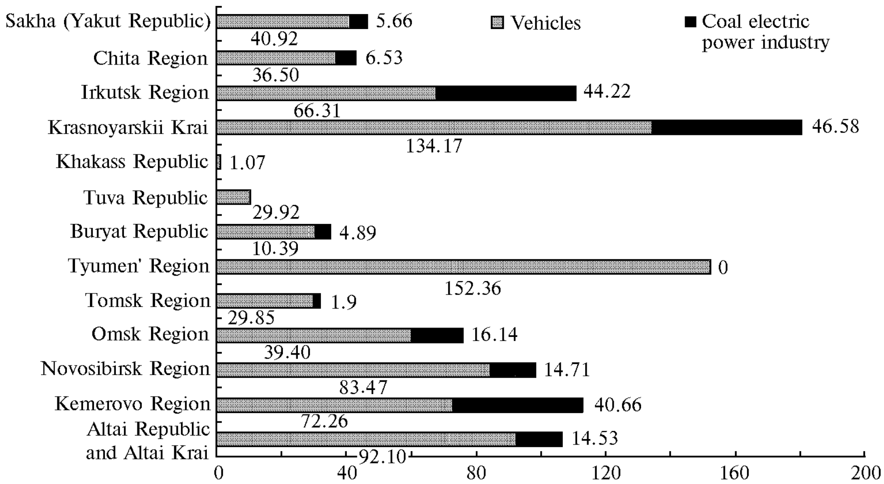

Fig. 4. Distribution of atmospheric lead emissions from vehicles and power stations operating on coal over administrative regions of Siberia (t/yr.).

The amount of lead emitted by power stations in the combustion of coals for average efficiency of filters of 92% was 630.7 t for the entire territory of the USSR in 1990, including 390 t emitted in the territory of Russia. The distribution of atmospheric lead emissions over the economic regions of Russia is illustrated by Fig. 2. From the enterprises of electric power industry most lead was emitted into the atmosphere in the territories of Eastern Siberia, Western Siberia, and Ural'skii region. The lead emission from vehicles in the territory of the USSR in 1990 was 7.7 thousand tons, including 4.23 thousand tons emitted in the territory of Russia and only 766 t – in the territories of Western Siberia and Eastern Siberia. The greatest lead emission from vehicles was in the Central Region of Russia (see Fig. 3).

Of course, these results are uncertain to some extent and cannot be considered as absolutely true. However, based on the results of comparison with the existing statistical data,[7,9,18] we can conclude that the procedures for calculating anthropogenic emission of lead, used in the present paper, are acceptable for analytical estimation of macroscale emission, and the results obtained with their help describe adequately the realistic situation.

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