Sulfur and Nitrogen Budgets in the Territories of the Former Soviet Union

Invited contribution

A. Ryaboshapko[1], S. Paramonov, S. Gromov, O. Afinogenova, H. Rodhe, E. Kjellström, L. Gallardo-Klenner and D. Möller

[1]*Institute of Global Climate and Ecology, Glebovskaja str., 20-B, Moscow 107258 Russia*

The global atmospheric cycles of many pollutants, especially sulfur and nitrogen compounds, have been strongly affected by human activity during the last decades. The Soviet industry made a significant contribution to the world-wide pollutant emissions. It is impossible to carry out any global or hemispheric assessment of the atmosphere pollution conditions without considering the situation in the former Soviet Union (FSU). In spite of the current economic crisis, the countries of the FSU are still playing a significant role in atmospheric pollution on a hemispheric scale. Major objectives of our work were:

* compilation and check of information on emissions of sulfur and nitrogen compounds into the atmosphere over the FSU;
* compilation of information obtained in the FSU on the chemical composition of the atmosphere and atmospheric precipitation;
* calculation of atmospheric budgets of sulfur and nitrogen;
* exposure of problems and elaboration of recommendations for the FSU countries, for WMO and EMEP concerning data reliability and quality.

Nowadays, anthropogenic emissions are the most important items of atmospheric budgets of S and N compounds. Data on the emissions officially presented in the FSU were obviously underestimated. During the 1970s to 1990s some alternative attempts were made to estimate independently total SO_2 and NO_x emissions in the FSU. Serious discrepancies between official data and data calculated by independent experts were found (Table 1).

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Table 1: Estimates of annual SO_2 and NO_x emissions in the FSU.

Information source	Year	SO_2 emission, Mt S	NO_x emission, Mt N
Brodsky, 1977	1972–1973	11.8	–
Lübkert, 1987	1980	–	4.1
Ryaboshapko, 1990	1985	12.4	4.4
Berlyand, 1991 (official data)	1985	9.75	1.73
Berlyand, 1991 (official data)	1990	7.9	2.03
Mylona, 1993	1985	12.6	–
Benkovitz *et al.*, 1996	1985	15.4	5.4

In this work, fuel quality of about 500 oil-fields and 65 coal geological provinces were considered to estimate anthropogenic sulfur emissions. Export of fossil fuels, primary and secondary desulfurisation measures, SO_2 retention by flying ash and other processes which could determine SO_2 emissions were taken into account. In addition to fossil fuel combustion, other most important anthropogenic sources were estimated. The calculations showed that total sulfur emissions were 12.6 and 11.3 Mt S/a in 1985 and 1990, respectively, with an uncertainty of ± 17 %. The difference between our and official data is larger than the uncertainty of our data.

To calculate NO_x emissions, fuel consumption statistics and emission factors were used taking into account peculiarities of the Soviet economy. NO_x emissions amounted to 3.83 ± 0.80 and 4.10 ± 0.86 Mt N/a. So, the official values are roughly 2 times lower than ours and they are about 1.6 times lower than the low limits of our estimates.

Ammonia emissions originate from different and widely distributed sources. All known ammonia sources, including human metabolism, have been considered in our work. An approach based on emission factors was used. Total emission values made up 3.9 ± 1.2 and 3.8 ± 1.1 Mt N/a. Major contributions to ammonia emission come from animals, more than 80 %.

For model assessments of any long-term changes, *e.g.* climatic changes due to sulfate aerosols, it is important to know historical trends in anthropogenic sulfur emissions. At the beginning of this century, only 0.3 Mt S/a was emitted into the atmosphere. The emission maximum (13.6 Mt S/a) was achieved in the mid-1970s.

Model calculations of atmospheric transport of S and N compounds need knowledge of the spatial distribution of anthropogenic sources. In this work, S and N emissions were distributed over the FSU territory with a 1° × 1° geographical degree resolution. The highest emission densities were in the Ukraine, central European Russia and the Ural mountain region. The most intensive SO_2 source in the world was located in Norilsk City in the Arctic (~ 2 Mt SO_2/a).

Sulfur and nitrogen have been circulating through the atmosphere in all geological epochs. There are many natural sources of these compounds to the atmosphere. The most important sources for the FSU are listed in Table 2. Some natural sources can contribute significantly to the atmospheric budget.

Table 2: S and N natural emissions into the atmosphere (in brackets: uncertainty range), Mt/a.

Source	Oxidised sulfur	Oxidised nitrogen	Reduced nitrogen
Volcanism	0.6 (0.3–0.9)	0	0
Aeolian weathering	8.7 (4.4–13.1)	0	0
Biogenic processes in soil	0.3 (0.2–0.4)	1.0 (0.7–1.3)	3 (1.3–7)
Biomass burning, forest fires	0.08 (0.04–0.12)	0.02 (0.01–0.03)	0.08 (0.04–0.12)
Lightning	0	0.12 (0.06–0.18)	0
Total	9.7 (4.9–14.5)	1.1 (0.8–1.5)	3.1 (1.3–7.1)

The first observations of the chemical composition of precipitation water in the Russian Empire date back to the beginning of this century. Regular monitoring of S and N compounds in rainwater was initiated in the FSU in the mid-1950s. By the late 1980s, about 300 monitoring stations were carrying out observations of precipitation pollution. The highest precipitation pollution levels were observed in the south-western part of the FSU (2.4 mg $S(SO_4^{2-})$/L; 1.0 mg $N(NO_3^-)$/L; 1.5 mg $N(NH_4^+)$/L). The lowest S and N concentrations in precipitation were observed in north-eastern Siberia (0.4 mg $S(SO_4^{2-})$/L; 0.1 mg $N(NO_3^-)$/L; 0.2 mg $N(NH_4^+)$/L).

The mean annual wet deposition of S and N has been estimated separately for 21 physical-geographical regions. It was assumed that within each region both precipitation amounts and concentrations in precipitation were homogeneously distributed. The calculations showed that wet deposition fluxes amounted to

14.7 (10–19) Mt S/a, 2.5 (1.8–3.2) Mt $N(NO_y)$/a and
5.4 (3.8–7.0) Mt $N(NH_4^+)$/a.

Dry deposition has been estimated separately for 43 regions based on mean pollutant concentrations, prevailing surface type, agricultural practices, seasonal variations of concentrations and properties of underlying surfaces. Due to a large variability of all parameters the uncertainty of calculations is high. It was estimated that the dry deposition fluxes over the FSU between 1980 and 1990 amounted to 6.9 (3.2–10.6) Mt S/a and 2.3 (0.9–3.8) Mt $N(NO_y)$/a.

Alternative estimates of dry and wet deposition fluxes in the FSU have been obtained using a global 3-D tracer transport model MOGUNTIA (Feichter *et al.*, 1996). Such an approach gives a possibility to check the consistency between emissions and deposition fluxes and estimate deposition fluxes in areas where no observations are available. In addition, advective fluxes from a region and to a region can be estimated.

For sulfur, aeolian emission has not been considered in model calculations while for nitrogen both anthropogenic and all possible natural emissions have been used.

Comparison of calculated and observed values reveals a very good correspondence for nitrogen. For sulfur, the measured values are usually higher. This fact suggests that a significant additional natural flux of sulfur should be included in model calculations. According to our estimates about 40 % of sulfur emitted within the FSU could have a natural origin, mainly from aeolian soil weathering. There is another evidence of possible importance of natural sulfur sources. Even in the beginning of this century, when anthropogenic sulfur emission was tens of times lower than nowadays, wet deposition of sulfur over European Russian territory made up at least 25–30 % of current levels.

All estimated input and output fluxes for 1985 are shown in Table 3 which shows that the atmosphere over the FSU was practically balanced: the sums of S and N input fluxes were practically equal to the sums of output ones. The net advective fluxes across the borders of the FSU made up not more than 10 % of the sums of the sources. However, the output advective fluxes accounted for about 1/3 of total national emissions.

Table 3. Fluxes of S and oxidized N over the FSU in 1985.

Fluxes	Oxidised sulfur		Oxidised nitrogen	
	Observed	Modelled	Observed	Modelled
Anthropogenic emission	12.6	15.0	3.8	4.4
Biogenic emission from soils	0.3	0.03	1.0	0.42
Biomass burning, forest fires	0.08	0	0.02	0
Lightning	0	0	0.12	0.12
Volcanoes	0.6	0.36	0	0
Oceans	–	0.15	0	0
Aeolian soil weathering	8.7	–	0	0
Sum of sources	22.3	15.6	4.9	4.9
Wet deposition	14.7	7.7	2.5	2.4
Dry deposition	6.9	6.7	2.3	2.2
Sum of sinks	21.6	14.4	4.8	4.6
Advection to the FSU (*)	–	2.2	–	1.1
Advection from the FSU (*)	–	3.5	–	1.5
Net advective flux	–	1.3	–	0.4

(*) Anthropogenic emissions only.

A very important point is that emissions from the FSU domestic sources cannot be less than the difference between total deposition and import flux. Thus, for oxidised nitrogen, domestic emission cannot be lower than 4.6–1.1 = 3.5 Mt N/a. This suggests that the official estimates of anthropogenic emissions were underestimated by a factor of 2. We can recommend to the governments of the new independent states of the FSU to check the methods of emission evaluation.

Another important conclusion is that more attention should be given to natural sulfur emission sources. Such sources can give a significant contribution to sulfur atmospheric loads that are not connected with acidification problems. It can lead to overestimation of acidifying deposition. So, natural sources should be taken into consideration in the EMEP modelling evaluations of sulfur transport and deposition in Europe, especially in eastern and southern regions of Europe.

The balance approach showed that the FSU was a net source both for sulfur and nitrogen compounds: the outcoming fluxes exceeded the incoming ones. The main part of sulfur and nitrogen compounds was transported to the Arctic and the Pacific Ocean.

In this work, S and N atmospheric loads were estimated in the regions where no observations were available. Such regions cover huge forest territories of Siberia. The simulated nitrogen deposition can be used in studies related to the carbon cycle in the taiga zone of the FSU. Besides, the calculated deposition fields can be used for improving the configuration of monitoring networks.

Data on anthropogenic emissions from different branches of the FSU economy, data on spatial emission distribution, data on S and N concentrations in air and precipitation are available in the electronic form at the Institute of Global Climate and Ecology.

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