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Assessment of wet sulphur deposition over the former USSR

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ABSTRACT

The data obtained by different national and international networks of precipitation chemistry monitoring the territory of the former Soviet Union (FSU) have been generalized. Analyses of long-term trends of sulphur concentrations in precipitation for a 30-year period showed that the changes did not correlate with the dynamics of anthropogenic sulphur emissions into the atmosphere both over the FSU territory and Europe as a whole. Sulphur concentrations in precipitation reveal an obvious seasonal dependence with the spring maximum while deposition intensity has its maximum during summer time. All the FSU territory has been divided into 20 regions, and the mean values of sulphur concentration in precipitation and wet deposition have been estimated for each of them. The highest values were characteristic of the south-western part of the country and lowest ones of the remote areas of NorthEast. In the 80s, about 6.0 Tg of sulphur were annually wet deposited over the European territory of the FSU, about 3.0 Tg over the territory of Kazahstan and Central Asia, about 5.7 Tg in Siberia and the Russian Far East. The total wet sulphur deposition made up 60–90% of the anthropogenic sulphur emissions into the FSU atmosphere.

1. Introduction

Assessments of sulphur deposition with precipitation from the atmosphere form the basis of our knowledge of the atmospheric sulphur cycles both at the global and regional levels. The bulk of anthropogenic sulphur does not leave the continental atmosphere and settles down to the surface within hundreds of kilometers from its sources (Rayboshapko, 1983). Thus, Galloway and Whelpdale (1987) indicated that only 25–30% of anthropogenic sulphur is transported beyond the North American continent into the Atlantic Ocean. It is quite natural that a still greater portion of anthropogenic sulphur should settle down on the Eurasian continent since the bulk of sulphur is released into the atmosphere over the western periphery of the continent and transported inland by the prevailing easterly air flows.

Available estimates indicate that by the late 1980s, the continental man-made sulphur emission was 15 Mt/yr in Western and Central Europe (Iversen et al., 1990), from 8 (GOSCOMSTAT, 1990) to 13 Mt/yr (Ryaboshapko, 1990) in the USSR, 10 in China, 0.6 in Japan, 0.8 Mt/yr in the Korean Peninsula (Kato and Akimoto, 1992). The spatial emission distribution over the European part of the former Soviet Union (FSU) was studied within the framework of the international EMEP programme (Iversen et al., 1990). Fig. 1 shows the distribution over the Asian part of FSU with a resolution of $5 \times 5^\circ$ (based on the data of GOSCOMSTAT, 1990). As it follows from EMEP data and Fig. 1, the bulk of atmospheric sulphur emissions over FSU was associated with the western and central regions. Taking into account the pattern of the global atmospheric circulation, it can be stated a priori that the largest part of sulphur emitted deposited directly over FSU whose area exceeded $22 \cdot 10^6$ km^2.

The first episodic observations of precipitation chemistry in Russia data back to the end of the last century. Regular observations were initiated in

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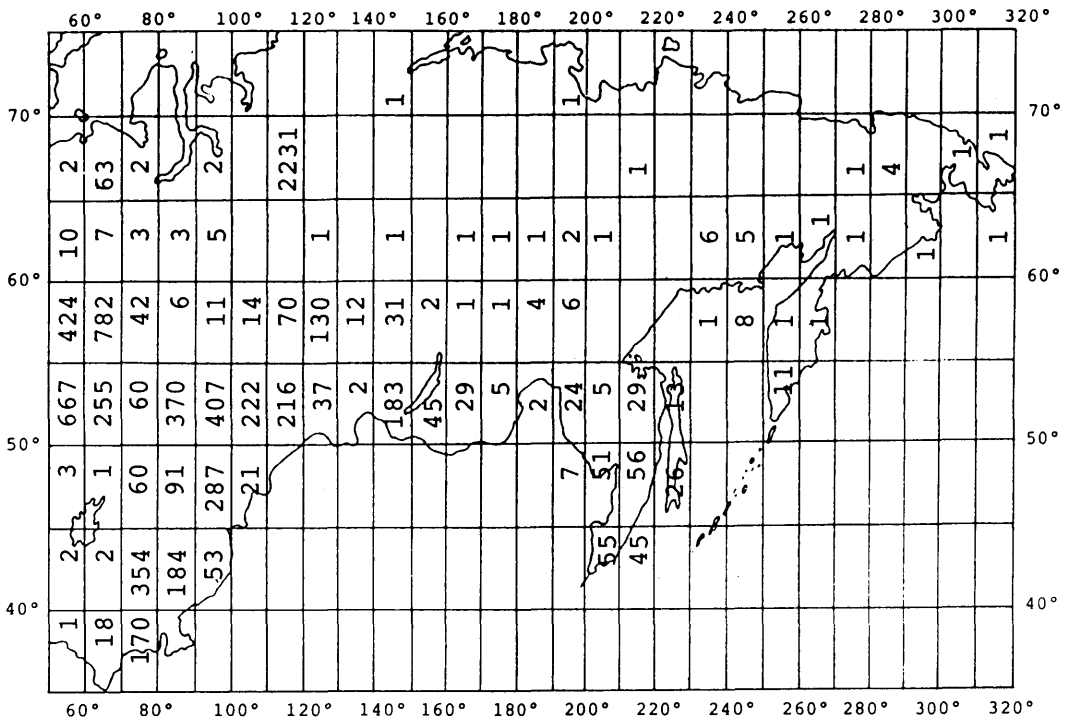

Fig. 1. Anthropogenic sulphur emission distribution over the FSU territory in 1989, *1000 ton SO_2/a.

1957 when Ye. Selezneva and V. Drozdova laid the foundation of precipitation chemistry monitoring in the USSR. The establishment of an international BAPMoN programme gave a powerful impetus to the development of the network. Later on, two more networks for precipitation pollution monitoring were established under the auspices of GEMS and EMEP international programmes. Since the late 1970s, a snow cover chemistry observation system has been operating which consists of a few hundred sampling sites. Besides observations carried out at the national level, a number of regional programmes were implemented in Siberia, the Far East, the Urals and Georgia.

Data accumulated have been partly summarized in monographs (Drozdova et al., 1964; Andreev and Rozhkova, 1971; Chernjaeva et al., 1978; Vasilenko et al., 1985). However, most of them are contained in separate articles and technical reports of various institutions. The data obtained are not equivalent from the viewpoint of sampling and analysis methodology used. In a number of cases, methodological mistakes are so significant that the information is to be discarded. Thus, data on sulphur concentrations and deposition over FSU are rather scattered, contradictory and practically inaccessible for an English speaking audience. The present paper makes an attempt to summarize data on sulphur content in precipitation available in the Russian literature and to estimate wet deposition levels for individual regions and the FSU as a whole.

It is well-known that a part of the precipitation sulfate is of marine origin. If we assume that chloride and sodium ions are solely of marine origin, the contribution of marine sulfate can be calculated based on known ion ratios in sea salt. Such an operation is quite applicable to coastal areas, but it becomes rather conditional for the continental atmosphere where sodium and chloride in precipitation can be not only of marine but additionally technogenic and eolian origin. Never-

theless, we shall use here the aforementioned technique for "excess sulphur" calculation which allows us to give a more adequate idea of the deposition of biogenic and industrial sulphur.

2. Characteristics of observation programmes and networks

When analyzing information on precipitation chemistry, estimates of its representativity were based on the availability of data not only on sulphur concentration, but also on all other major ions. In that case, it was possible to assess the ion balance and, hence, to obtain an objective criterion for data selection. Another important factor was the availability of information on the sampling and analysis methodology used by the authors.

The present paper is devoted to regional and global sulphur concentration and deposition levels. On that basis, we only considered stations seemingly not affected by the influence of local atmospheric pollution sources. It is quite natural that no clear-cut criteria can exist here. Nevertheless, whenever a station caused any doubt, we did not consider it. Proceeding from the above, we shall only deal with the background stations below.

By the end of the 80s, about 300 stations of precipitation chemistry monitoring were in operation in the FSU within the three programmes mentioned above. About 150 of them can be considered as background programmes. The GEMS programme was emplemented at 15 of them and the EMEP programme at 11 of them.

According to the BAPMoN and GEMS sampling protocol, precipitation should be collected in the course of precipitation events and poured into a common container to obtain an averaged sample. Sampling duration of one averaged sample is one month at BAPMoN stations and 10 days at GEMS stations. To prevent dry deposition effect on sample composition at BAPMoN and GEMS stations, a precipitation collector should be opened by an operator just before precipitation starts and closed just after the event is over. Before being sent to an analytical laboratory, the samples are stored at the GEMS stations at 4°C, and at the BAPMoN stations at room temperature.

At EMEP stations, open bulk collectors are used. The collector surface is thoroughly cleaned every day irrespective of the fact of whether there was a precipitation event the day before or not. The procedure significantly reduces the contribution of dry deposition to the collector's surface (especially during long dry periods). At the EMEP stations, the samples are stored at -7°C.

The GEMS and BAPMoN samples before 1990 were analyzed at the analytical laboratories for all major ions using classical manual analytical chemistry methods. The technique cannot be referred to high accuracy methods. For strongly diluted solutions (such as precipitation), the accuracy of a single measurement of any major ion can vary within $\pm 30\%$. The deviation from the ion balance typical of the leading laboratory is 10%.

All samples collected at the Soviet part of the EMEP network were analyzed at a specialized laboratory. Sulphur content in precipitation was analyzed by the thorin method or by ion chromatography techniques. In the first case, the analytical error could not exceed 15%, in the second it was as low as 5%.

The quality of precipitation composition measurements is under permanent control provided by the Main Geophysical Observatory (Petrenchuk and Lavrinenko, 1982). Quality assurance of the data obtained at the Soviet BAPMoN and GEMS was recently made by Artz and Lavrinenko (1993) in comparison with the data from three USA BAPMoN stations. The authors have examined the quality by four different techniques and obtained satisfactory results. Quality assurance of the EMEP data is provided by the Chemical Coordination Center of EMEP by systematic intercomparisons and special experiments.

In addition to the national level networks, some regional networks were used to study precipitation chemistry. Such researches can be exemplified by a study carried out in 1970–1976 in the South Urals (Chernjaeva et al., 1978). A network consisting of 44 sampling stations was established, covering an area of 659 km^2. Samples were collected by a container which was only exposed during a precipitation event. In dry periods, the container was closed by a lid and stored indoors. 2004 mean monthly samples were analyzed over the whole working period of that regional programme. Sulfate in precipitation was measured by turbidimetry, the relative error being about 10%.

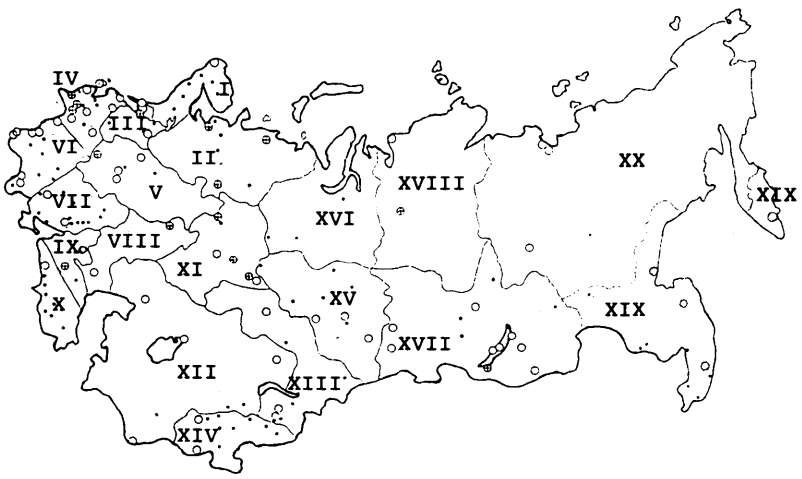

Fig. 2. Precipitation chemistry network on the FSU territory.

Monitoring period: • 3 years; ○ 3–10 years; ⊕ more than 10 years

XVI: region number.

Other regional programmes of varying duration were implemented in a similar way, though they involved less sampling sites. As a rule, the objective of the programmes was to determine the degree of precipitation pollution and to quantify pollutants and nutrients coming from the atmosphere (Votintsev, 1954; Denisov, 1956; Denisov and Bugaev, 1956; Girenko, 1959; Agaev and Stepanov, 1964; Fotiev, 1964; Gabrieljan and Bozojan, 1964; Yakusheva and Brazhnikova, 1964; Supatashvili, 1968; Bashmakova et al., 1969; Davydov, 1969; Khasanov and Rahmatullina, 1969; Masilunas and Paulukjavichus, 1969; Davtjan and Vardanjan, 1970; Andreev and Rozhkova, 1971; Gadzhieva and Bezhaev, 1972; Matveev et al., 1976; Votintsev and Khodjer, 1981; Astratov et al., 1986; Kharkevich, 1986; Ivanov and Kashin, 1989; Mikhalenok, 1989; Shopauskiene et al., 1992).

The above-mentioned networks of observational stations differ in their density and do not provide a uniform coverage of the country. Fig. 2 shows the location of the stations the data from which formed the basis of the present work. The figure divides the stations into three categories through duration of the observation period, which may be indicative of the representativity of the information obtained. It can be seen that the highest network density is typical of the European territory while the vast north-eastern regions of the country are not covered at all by the observations.

3. Long-term trends

44 stations conducted observations over the FSU for more than 10 years (some of them are affected by local pollutant sources and therefore are not shown in Fig. 2). Data from such stations allow one to establish the presence or absence of long-term trends in precipitation chemistry and assess the variability of the annual values. Based on data from Petrenchuk (1979), Izrael et al. (1989) established that in the western regions of the country (Lithuania, Latvia, Bjelorussia) there was an increase in precipitation sulfate of 2.3% a year between 1958 and 1976. Shopauskiene et al. (1992) performed a detailed analysis of long-term trends for all major precipitation ions at 3 Lithuanian stations. According to them, there was a slight reduction of sulfate concentrations in precipitation (not more than 2% a year) in 1981–1990. To some extent, the reduction can be related to changes in the type of agricultural activity. A detailed analysis of long-term trends in

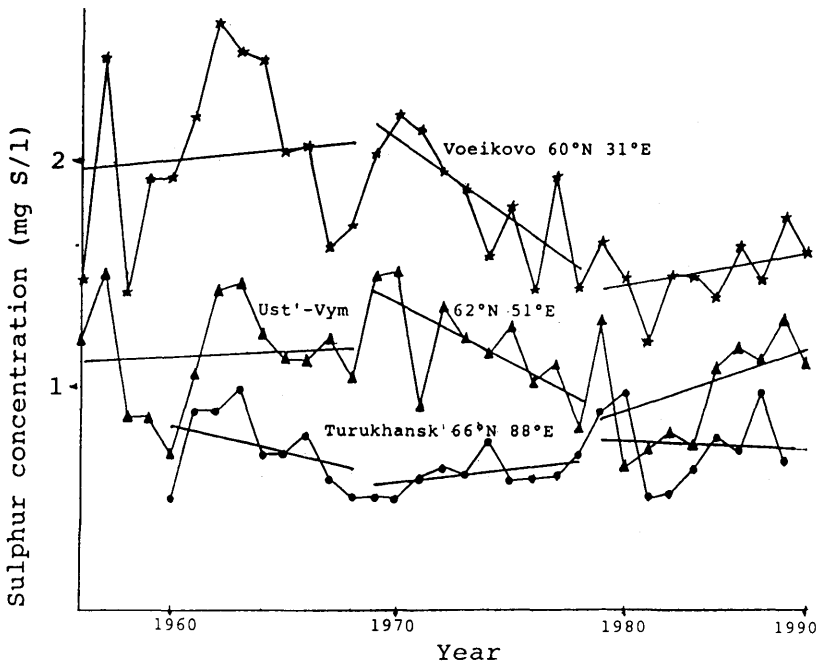

Fig. 3. Long-term variations of sulphur concentration in precipitation.

precipitation acidity at the BAPMoN stations was carried out by specialists from the Main Geophysical Observatory (Monthly Data, 1989): more-or-less stable trends were not identified.

Fig. 3 presents excess sulphur concentrations obtained at three BAPMoN stations over a 30-year observation period. The stations reflect situations in regions cardinally different from the geophysical viewpoint: the highly industrialized Leningrad District, the relatively clean region in north-eastern Europe and the practically background area in Siberia. Straight-line sections represent a linear regresion equation for every 10 years. It follows from the figure that there are no clear-cut trends in the content of excess sulphur in precipitation. Nevertheless, one can notice a certain similarity of long-term concentration variations for two stations located in Europe, confirmed by a rather high correlation factor between them: 0.59. Most likely, these are variations of the climatic nature.

It would be interesting to compare long-term data on sulphur levels in precipitation with estimates of long-term changes in anthropogenic sulphur emission over FSU and Europe as a whole. Fig. 4 presents such estimates taken from Ryaboshapko (1990) and Mylona (1993). It follows from the figure that there was an intensive emission growth (about 5% a year) in the USSR during the 1960s. The USSR anthropogenic emission reached its maximum in the mid-70s and then a decrease trend appeared. In the 1980s, emission in the USSR was practically stable (13 Tg S/yr). In Western and Central Europe, the maximum in emission was reached by the end of the 70s, and after that, a rather sharp reduction was observed.

If one suggests that precipitation composition over the European part of the FSU was determined mainly by Soviet emission, it is quite easy to account for practically unchanging precipitation chemistry during recent years. However, it is difficult to interpret data of the 1950s–1960s when there was an intensive growth of anthropogenic sulphur emission both in the USSR and in Europe as a whole.

Analysis of long-term observation series indicates that sulphur concentration in precipitation can vary significantly from year to year. Thus, for stations presented in Fig. 3, individual mean annual values between 1981 and 1990 (a period of

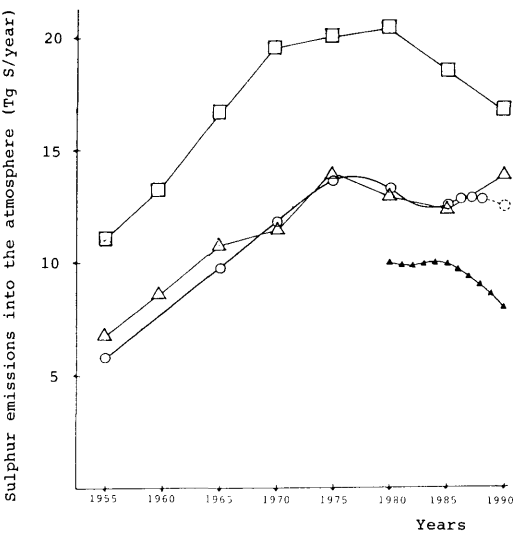

Fig. 4. Anthropogenic sulphur emissions into the atmosphere over the USSR and Europe.

□ — Europe without USSR (Mylona, 1993)
△ — USSR (Mylona, 1993)
○ — USSR (Ryaboshapko, 1990)
▲ — USSR (GOSCOMSTAT, 1991).

a relatively stable anthropogenic emission in the USSR) can differ from the 10-year mean by 40%. Sulphur deposition values are still more variable, which is related to a significant variability of precipitation amount at an individual point. In some years, the sulphur deposition level can differ from the 10-year mean by a factor of 2.

4. Seasonal variations of concentrations and deposition

Since the atmospheric input of anthropogenic sulphur varies during the year, it can be expected to influence the seasonal dynamics of sulphur atmospheric concentrations and deposition with precipitation. Afinogenova and Galperin (1985) noted a significant inhomogeneity of emission during the year: the December maximum exceeds the June minimum over the European part by the factor of 1.7. Emission variations are best correlated in the phase with changes in the light day duration and somewhat lag behind the annual temperature variations. It can be assumed that for Siberian regions, the amplitude of the seasonal emission variations would be still larger than for the European territory.

Figs. 5 and 6 present the mean monthly concentrations and deposition of sulphur over the whole observation period at three BAPMoN stations, 2 EMEP stations and 1 GEMS station. Boxes denote mean standard deviations from the mean weighted value (let us remember, however, the variations of concentrations and deposition are not described by the normal distribution), and whiskers denote the least and highest values. The BAPMoN stations can give an idea of changes in the spatial nature of seasonal variations within the latitudinal zone between 60° and 65° from the Leningrad District in the west to central Siberia in the east. The EMEP stations stretch in the meridional direction from the Kola Peninsula to Western Ukraine.

It follows from the figures that sulphur concentrations in precipitation are actually subject to seasonal variations; however, the concentration maximum does not coincide with the emission maximum in time. In all cases, the concentration maximum is shifted to spring. It is typical of temperate climate zones (Voeikovo, Berezina, Svitjaz) to have the maximum in March. At stations with an Arctic climate (Turukhansk, Janiskoski), the maximum is shifted to late spring. At the same time, the amplitude of concentration variations corresponds to the variations of anthropogenic emissions over FSU. Meszaros (1974) tried to explain such a shift of maximum concentrations by risen oxidation of sulphur dioxide by ozone during a spring ozone maximum. Let us note a rather high year-to-year variability of the mean monthly concentration. For instance, the March concentrations at Voeikovo varied from the least value of 0.55 mg S/l to a maximum value of 7.1 mg S/l.

Unlike concentrations, the deposition values are totally different from the annual variations of the anthropogenic emission. As follows from Figs. 5 and 6, we should rather speak about the summer sulphur deposition maximum. The summer maximum is especially pronounced at the Arctic station of Janiskoski. Spring and autumn maximums can also occur at the northern stations of Ust-Vym and Turukhansk. In all cases, the intensity of sulphur deposition with precipitation in winter is lower than that in summer. Such a seasonal variation pattern is mainly determined by the fact that the wash-out efficiency of sulphur dioxide, and to a

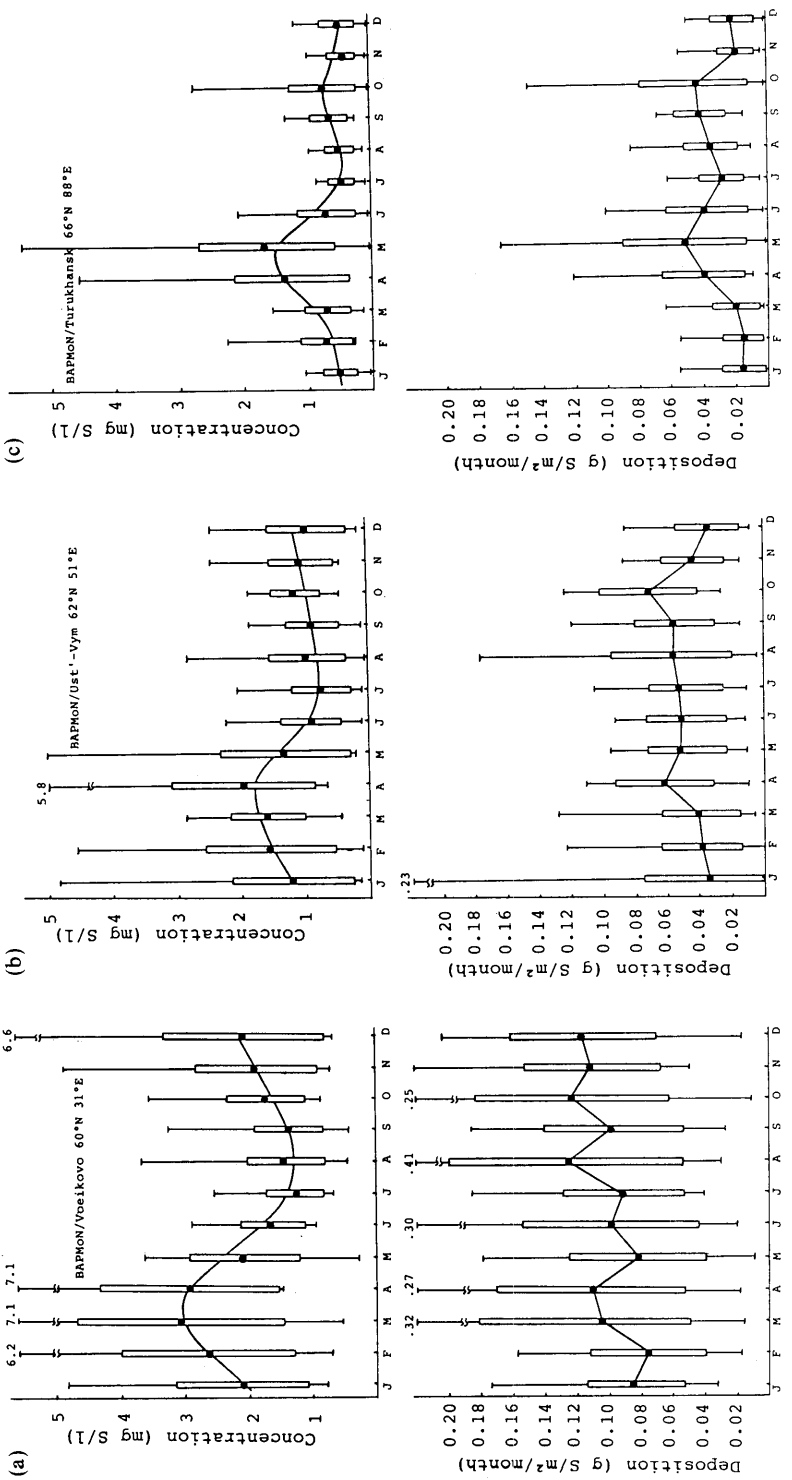

Fig. 5. Seasonal variations of sulphur concentration in precipitation and wet deposition at BAPMoN stations.
(a) BAPMoN/Voeikovo
(b) BAPMoN/Ust'-Vym
(c) BAPMoN/Turukhansk.

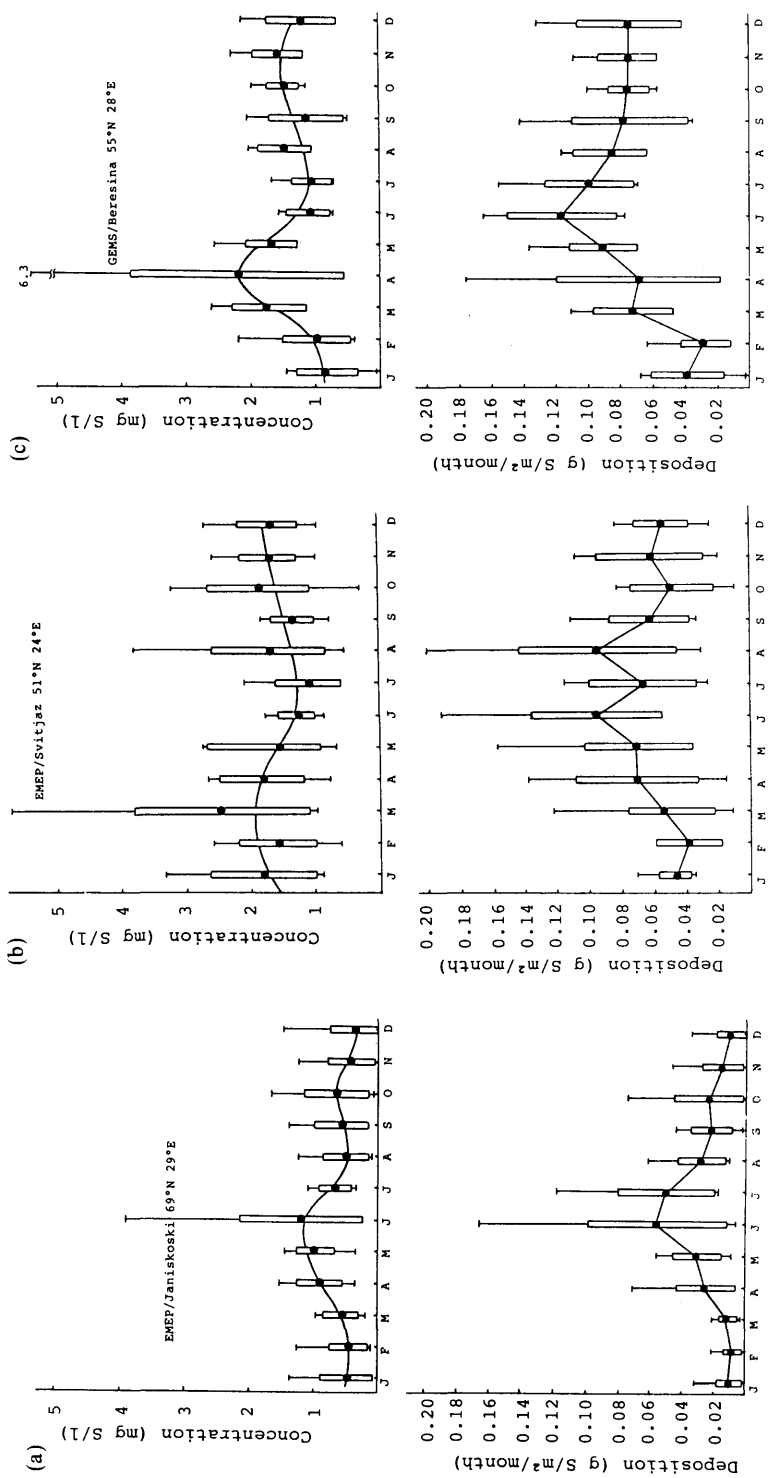

Fig. 6. Seasonal variations of sulphur concentration in precipitation and wet deposition at EMEP and GEMS stations.
(a) EMEP/Janiskoski
(b) EMEP/Svitjaz
(c) GEMS/Beresina.

lesser extent that of sulfate, is much less for snow than for rain.

5. Concentrations and deposition of sulphur-bearing substances over various regions of the country

Attempts to estimate sulphur deposition intensity over rather large regions are always connected with the following dilemma: one can use observational data obtained either during a certain year or within a rather long period. In the first case, the assessments obtained can be correlated with anthropogenic emission in that particular year, but the integral assessment error can be quite high due to a considerable interannual variability of deposition. In the second case, we can use smoother deposition values at individual points, but the error can result from the long-term variability of the emission. The present paper employs the second approach using the means of sulphur concentrations measured in 1981–1990 to estimate sulphur deposition in individual regions. To justify the approach selected, we can say that at that period, the anthropogenic emission over the whole FSU varied much less than possible interannual variations of concentrations and depositions.

To obtain integral deposition assessments, the whole territory of the country was divided into separate regions (mainly based on administrative divisions). Such a division is rather conditional from the geophysical viewpoint. However, it allows us at least, to avoid mistakes in determining the areas of individual regions. The division is aimed at obtaining sulphur concentration and deposition levels averaged for a given region. Below, observation data will be considered in detail for large geographic zones.

Sulphur deposition with precipitation in an individual region was calculated from the estimated value averaged for all stations of the given region, taking into account their "weight" contributions determined by the number of samples taken at a station. In some cases (e.g., the Caucasus region), we tried to use generalized deposition data obtained within separate regional programmes.

5.1. European territory of FSU (ET-FSU)

Sulphur levels in precipitation over ET-FSU vary rather considerably, depending mainly on the intensive anthropogenic emission both in Western and Central Europe and in the country itself. Several large industrial source regions can be identified here: the Kola Peninsula, Leningrad District and North-Eastern Estonia, Moscow/Tula industrial region, Eastern Ukraine, Ural industrial region. The Caucasus and Ural regions were included in ET-FSU as a whole.

Drozdova et al. (1964) noted a general trend of sulphur concentration decrease in precipitation from north to south. The latitudinal concentration gradient is less pronounced. Against such a background, we can single out individual spots of very high concentrations, e.g., in the Donbass region, and relatively low concentrations, e.g., in the North Caucasus foothills.

The lowest sulphur concentrations and deposition are typical of northern regions I and II (see Fig. 2) where the concentrations vary within 0.6–2.6 mg S/l and deposition within 0.2–1.2 g $S/m^2/yr$. The highest concentration and deposition levels are observed in the south-western part of ET-FSU (Western Ukraine and Moldavia). Most likely, this fact reflects the effect of long-range atmospheric transport of sulphur compounds from Central European industrial areas. Concentrations observed here can be as high as 7 mg S/l with an average value of 2.4 mg S/l. Similarly high levels (up to 5 mg S/l) are typical for the Donbass region (Eastern Ukraine) where the density of anthropogenic sulphur emission is exceptionally high (about 26 g $S/m^2/yr$ for Donbass as a whole).

Region VIII (Middle and Lower Volga) is not covered by observations. Data are only provided by two stations located in different physico-geographical conditions. The 1966–1970 data obtained by Matveev et al. (1976) reflect the situation in the arid Caspian Lowland which is characterized by the absence of local anthropogenic sources and the presence of intensive eolian weathering of solonchak soils in the Kalmyk steppe zone. Another station is located in a zone with a much higher precipitation level. However, precipitation composition can be affected here by anthropogenic emissions from a relatively large urbanized area in Tataria.

The values obtained in the Caucasus are most variable. This is primarily related to the difference in stations elevation and to a highly inhomogeneous distribution of precipitation intensity in mountains. As a result, sulphur concentration

in precipitation varies from 5.3 mg S/l in the Apsheron Peninsula (Agaev and Stepanov, 1964) to 0.04 mg S/l on the Elbrus slopes at 3,400 m a.s.l. (Matveev, 1964). On the Armenian Highland, the concentration varies from the trace levels to 1.6 mg S/l (Gabrieljan and Bozojan, 1964). Deposition also varies within a very broad range since precipitation on the western Caucasian slope reaches several meters a year, while in the eastern areas, it can be as low as 300 mm/yr. To assess a general situation in the Caucasian region, we used estimates by Supatashvili (1968), who summarized data on 768 samples from various locations in Georgia. If Georgian conditions are assumed typical of the whole region, the mean sulphur concentration in precipitation can be taken to be equal 1.1 mg S/l and deposition can be considered 1.2 g $S/m^2/yr$.

Table 1 gives generalized data on all ET-FSU regions indicating the annual amount of sulphur deposition in a region. When comparing data from Table 1 and Fig. 2, it becomes clear that there is a marked trend of a deposition reduction northwards and a weak trend of an eastward reduction. For the Ural region, our estimate is quite close to an earlier estimate by Chernjaeva et al. (1978): 0.9 Tg S/yr. We estimate the total amount of sulphur deposition in ET-FSU as 6 Tg S/yr. The accuracy assessment of the values obtained for individual regions is rather conditional and subjective.

If we assume that the emission from the part of FSU covered by EMEP is 5 Tg S/yr (Iversen et al., 1990), it implies that there should be significant natural or external anthropogenic sources. It is more likely that the official assessments of anthropogenic sulphur emissions into the atmosphere over FSU are considerably underestimated (Ryaboshapko, 1990; Mylona, 1993).

5.2. *Kazakhstan and Central Asia*

The bulk of monitoring stations are concentrated in foothill areas where one can observe increased precipitation and the highest population density. Vast areas in deserts and semi-deserts where the annual precipitation amount does not exceed 200 mm are not practically covered by observations. Northern and North-Eastern Kazakhstan can be referred to the steppe and forest-steppe zone which makes it rather peculiar from the viewpoint of eolian sulphur emission contribution to precipitation composition. The territory considered was conditionally divided into 3 regions: deserts and semideserts (region XII), steppe and forest-steppe (region XIII) and mountains (region XIV).

Sulphur concentration in precipitation in desert and semi-desert areas can be quite high: up to 10 mg S/l, demonstrating the obvious effect on the eolian weathering of solonchak soils on precipitation chemistry. At the same time, total annual deposition with precipitation can be rather low. The above can be exemplified by the desert stations where the annual deposition with precipitation is only 0.1 g S/m^2.

The Aral Sea area is a special case with its marked effect of anthropogenic activity on precipitation chemistry. Excessive water use for

Table 1. *Wet sulphur deposition over ET-FSU regions*

Region	area (10^{12} m^2)	Concentration (mg S l^{-1})	Deposition intensity (g S m^{-2} yr^{-1})	Deposition (Tg S yr^{-1})
I	0.317	0.8 (±0.1)	0.4 (±0.05)	0.13 (±0.016)
II	1.149	1.4 (±0.2)	0.6 (±0.1)	0.69 (±0.115)
III	0.197	1.7 (±0.2)	1.25 (±0.15)	0.25 (±0.03)
IV	0.294	1.9 (±0.2)	1.3 (±0.15)	0.38 (±0.044)
V	0.748	2.2 (±0.3)	1.3 (±0.2)	0.97 (±0.15)
VI	0.440	2.4 (±0.3)	1.4 (±0.2)	0.62 (±0.088)
VII	0.470	3.8 (±0.5)	1.9 (±0.3)	0.89 (±0.141)
VIII	0.536	2.5 (±0.5)	1.1 (±0.2)	0.59 (±0.107)
IX	0.265	1.8 (±0.5)	1.1 (±0.3)	0.29 (±0.08)
X	0.276	1.1 (±0.4)	1.2 (±0.5)	0.33 (±0.138)
XI	0.824	1.9 (±0.2)	1.05 (±0.1)	0.87 (±0.082)
total	5.516			6.01 (±1.11)

Table 2. *Wet deposition of sulphur in Kazakhstan and Central Asia*

Region	area (10^{12} m^2)	Concentration (mg S l^{-1})	Deposition intensity (g S m^{-2} yr^{-1})	Deposition (Tg S yr^{-1})
XII	2.590	4.0 (±2.0)	0.84 (±0.42)	2.18 (±1.09)
XIII	0.989	1.5 (±0.3)	0.47 (±0.09)	0.46 (±0.09)
XIV	0.480	1.4 (±0.3)	0.74 (±0.16)	0.36 (±0.08)
total	4.059			3.00 (±1.26)

irrigation over the last 30 years has resulted in a catastrophic reduction of the sea area and exposure of marine sediments enriched in a salt of a specific composition with an increased sulfate content (Ryaboshapko, 1983). Due to an intensive eolian weathering, local precipitation is highly enriched in sulfate. In the late 1960s, Khasanov and Rahmatullina (1969) estimated the mean sulphur concentration on the Aral coast as 4.7 mg S/l and deposition as 0.49 g S/m^2. In the first half of the 80s, the values increased to 6.5 mg S/l and 0.91 g S/m^2/yr, respectively. The highest mean annual sulphur concentration of 13.3 mg S/l was recorded here in 1989 while the deposition was 1.2 g S/m^2/yr.

In Northern and Eastern Kazakhstan, the precipitation amount is much higher and there are no conditions for a significant eolian weathering. The most representative data for the northern part of the region were obtained over an 8-year observation period at a station of biospheric reserve named Borovoe. However, we cannot exclude a possible influence of local anthropogenic sources. The eastern part of the region is characterized by rather diverse physicogeographical conditions which are reflected in the variability of observed concentrations.

Foothills and mountain areas are characterized by highly variable concentrations and annual precipitation intensity. Rather high concentrations can be observed in Central Asian valleys which is accounted for by a considerable population density, high levels of air pollution, intensive agricultural activity and specific climatic conditions in the valleys with stable atmospheric inversions. Precipitation chemistry above the inversion level differs greatly from that in the valleys. According to Nikolaenko (1989), sulphur concentration in Central Asian precipitation was 4.1 mg S/l at heights from 250 to 700 m and only 1.7 mg S/l at 700–2,500 m. Some stations are located at high elevations and their data can reflect the chemical composition of the free troposphere rather than that of the boundary layer.

Table 2 presents generalized data for Kazakhstan and Central Asia. Once again, we would like to emphasize two features of the regions considered. Firstly, initial data are quite variable, which can result in an estimate error. Secondly, a significant though unquantifiable part of sulphur is related to the eolian emission of solonchak soils with a high sulfate content.

5.3. Siberia and the Far East

The whole territory of Siberia and the Far East is divided into 6 regions differing both in physicogeographical conditions and the degree of industrialization. The monitoring network density in southern regions is relatively high, while eastern Yakutia and Chukotka are not covered by observations at all. Researchers were mostly interested in the hydrochemical peculiarities of precipitation in the Baikal area where observations have been conducted for the last 4 decades at several stations.

Far East region XIX differs drastically from the rest in its climatic conditions. Regional precipitation and chemistry are largely affected by the Pacific Ocean and have a markedly seasonal character. The highest concentrations of excess sulphur in precipitation are recorded at coastal stations, where the deposition intensity can reach 2.7 g S/m^2/yr.

The highest population density and degree of industrialization are typical of region XV. At the same time, the most powerful anthropogenic source of sulphur (the Norilsk Mining and Metallurgy Complex) is located in region XVIII. The annual atmospheric sulphur emission from the complex is about 1.1 Tg. Precipitation chemistry observations have never been conducted in the

Table 3. *Wet deposition of sulphur over Siberia and Far East*

Region	area (10^{12} m^2)	Concentration (mg S l^{-1})	Deposition intensity (g S m^{-2} yr^{-1})	Deposition (Tg S yr^{-1})
XV	1.154	1.7 (±0.2)	0.90 (±0.11)	1.04 (±0.13)
XVI	1.273	0.9 (±0.2)	0.43 (±0.10)	0.55 (±0.13)
XVII	2.326	0.9 (±0.2)	0.40 (±0.09)	0.93 (±0.21)
XVIII	1.797	1.0 (±0.4)	0.42 (±0.17)	0.75 (±0.31)
XIX	1.612	0.9 (±0.2)	0.80 (±0.18)	1.29 (±0.29)
XX	4.604	0.8 (±0.2)	0.24 (±0.06)	1.10 (±0.28)
total	12.766			5.66 (±1.35)

vicinity of the source. It can be assumed that relatively high sulphur concentrations in precipitation at a station situated at 500 km distance northwestward are partly related to the impact of the complex. On the other hand, Vavilov Glacier chemistry studies (Severnaya Zemlya Archipelago) have not revealed any impact of that source (Astratov et al., 1986). Data from region XVIII stations are rather contradictory, which cannot avoid affecting the accuracy of mean concentration estimates.

Table 3 presents estimated sulphur concentrations in precipitation and deposition levels for Siberia and the Far East. It follows from the table that total sulphur deposition over the territory is practically equal to that over ET-FSU. However, the mean deposition intensity is only 40% of the European one. It is quite likely that a considerable part of sulphur deposition in region XIX has a biogenic marine origin.

6. Discussion and conclusions

Generalization of data from Tables 1–3 shows that on the whole, the annual excess sulphur deposition with precipitation over FSU in the 1980s was 14.7 ± 3.6 Tg. The value was about 60–90% of the anthropogenic emission over the FSU (25 Tg S/yr according to Ryaboshapko, 1990 and 16 Tg S/yr according to GOSCOMSTAT, 1990). Earlier, Vasilenko et al. (1988) reported that total sulphur deposition over the USSR territory was 13.8 Tg/yr. It is very close to our estimate, but Vasilenko et al. (1988) based their figure on snow samples integrated over the winter period and did not distinguish (at least, for the winter period) wet and dry deposition. Thus, it seems to use that their value may be an underestimate, if we consider it as total sulphur deposition.

We cannot answer the question: what part of sulphur in precipitation had an anthropogenic origin. The anthropogenic contribution was evidently dominant over the European part of the FSU. Here, in western regions, the deposition intensity exceeded that in remote Siberian regions by 8 times, while the concentration was 5 times higher.

The situation with the general atmospheric sulphur budget over the whole FSU still remains uncertain. According to various estimates, anthropogenic sulphur transport to FSU from Central and Western Europe in the 1980s varied from 2.3 to 4.2 Tg (Brukhanov et al., 1982; EMEP/MSC-E, 1986; Izrael et al., 1987). 2.6–2.9 Tg out of that amount deposited over the European part of the FSU (EMEP/MSC-W, 1984; Izrael et al., 1987). Taking into account the pattern of the global atmospheric circulation, it can be stated a priori that the atmospheric transport of anthropogenic sulphur from China, Korea and Japan must be rather insignificant. The natural sulphur flux from the Atlantic Ocean to FSU is absolutely unknown. The assumption that the flux could be quite perceptible is confirmed by rather high levels of excess sulphur deposition with precipitation at the Pacific coast (Region XIX). Finally, as estimated by Ryaboshapko (1983), several million tons of eolian sulphur are released into the atmosphere in FSU arid zones, primarily in Central Asia.

The removal part of the budget is represented by advection beyond the boundaries of the territory considered, by wet and dry deposition. The anthropogenic sulphur advection flux to the west has been estimated using model calculations and airborne measurements as 0.4–0.8 Tg S/yr

(EMEP/MSC-E, 1986; Izrael et al., 1989). Some part of the flux can return to the FSU. The latest assessments show that about 0.25 Tg of "Soviet" sulphur deposited in the European countries in the late 1980s (EMEP/MSC-E, 1992). Some estimates indicate that 0.97 Tg S/yr were transported from FSU to the Arctic (Whelpdale and Barrie, 1989). The figure seems to be an underestimate since only the direct sulphur emission into the Arctic atmosphere over FSU was no less than 1.5 Tg S/yr (Tuovinen et al., 1993). It is evident that the bulk of the Arctic sulphur does not settle down on the underlying surface, but returns to the moderate latitudes, since its residence time in the Arctic atmosphere is quite long (Barrie and Hoff, 1984). Transport across other boundaries has not been studied on a serious scientific basis. A significant though a rather uncertain part of anthropogenic sulphur reaches the underlying surface due to dry uptake. Tuovinen et al. (1993) estimated that the contribution of dry uptake to the budget of sulphur input from the atmosphere over the Kola Peninsula varies within 50–90%. Based on the Whelpdale (1992) assessments, it can be assumed that the contribution for the whole FSU varies within 20–50%.

The above means that our knowledge of the atmospheric sulphur budget in such a large world region as the FSU is far from being certain. The uncertainty is aggravated by the pattern of long-term trends in sulphur deposition with precipitation, which is difficult to interpret from the viewpoint of available data on anthropogenic emission dynamics. We believe that many questions can be answered by developing numerical models of the atmospheric cycles of sulphur compounds and other anthropogenic and natural substances on the scale of the whole Northern Hemisphere.

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