ROLE OF PRECIPITATION IN THE PROCESS OF CLEANING THE ATMOSPHERE FROM AEROSOL

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Precipitation cleaning of the urban atmosphere is studied based on the data of observations in 1981-1996 at the Meteorological Observatory of Moscow State University (MSU), city of Moscow.

Precipitation plays an important role cleaning the atmosphere from aerosol. Precipitation cleaning effect on the optical properties of hazes, while having been addressed recently by a number of authors,[1,4,6] is still poorly understood. The measurement data available do not always agree. The precipitation cleaning effect in the atmosphere, under urban conditions, is most difficult to evaluate. The point is that the precipitation cleaning, although actually reducing impurity concentrations, is only efficient in regions out of the direct impact from the anthropogenic sources of aerosol. Even intense and abundant precipitation makes little change to the level of atmospheric pollution, maintained by nearby sources, that quickly restores just after the rainfall.

The meteorological observatory at MSU (MOMSU) is located on the territory of MSU botanical garden, in a "cleanB southwestern district of Moscow, far removed from large industrial enterprises of the city. Therefore, conclusions drawn from analysis of many-year observations at MOMSU reflect only general features of the precipitation washing out of aerosols.

To characterize the aerosol load of the atmosphere, we use aerosol optical thickness of the atmosphere, τ_a, for the effective wavelength of solar spectrum, $\lambda_0 = 550$ nm. The value τ_{a,λ_0} is calculated using measurement data on the direct integrated solar radiation.[9] Since the processes of cleaning the atmosphere are mainly related to precipitations, their chemical composition and mineralization may also serve as indicators of the air pollution. This enables one to make combined analysis of the two variables using data of observations in 1981−1991 from MOMSU observatory.

Monitoring of chemical composition of atmospheric precipitation has been performed at MOMSU since 1982. As part of the World Meteorological Organization (WMO) program, each sample in an individual snowfall or rainfall is analyzed for the presence of hydrocarbonate (HCO_3^-), sulfate (SO_4^{2-}), chloride (CL^-), calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), ammonium (NH_4^+), as well as the acidity index of precipitation pH, defined as the negative logarithm of concentration of hydrogen ions in the solution.[2,3]

For the observation period, the acidity of precipitation varied in a wide range, from the minimum pH of 3.20 in June, 1987 to the maximum of 9.65 in November, 1983. The annually mean pH values varied between 4.6 and 5.2, being lower than the background value for nonpolluted precipitation (pH = 5.6). The acidity of samples depends on the phase state of precipitation. Most samples have pH values between 4 and 6 for rainfalls and between 6 and 8 for snowfalls. During several years, samples with pH < 5.0 (strong acidity) were observed in 25 to 64% of all rain events in a season, and samples with pH < 4 in 2 to 6% of cases, except in 1987 when percentage of such rain events was 25%.[5]

Concentrations of Na^+ and K^+ ions change insignificantly during a year, while the general mineralization of precipitation and SO_4^{2-}, Ca^{2+}, and Mg^{2+} ions exhibits spring-summer maxima. As long-term observations at MOMSU show, the atmospheric aerosol optical thickness is also maximum in these same months. Figure 1 presents annual behavior of concentration of some chemical species and aerosol optical thickness, averaged over the entire period of observations. The annual variations of the average concentrations for all species studied, except for H^+, with the April maxima, closely match those of τ_{a,λ_0}. The April peaks in mineralization of the precipitation and τ_{a,λ_0} are caused by seasonal increase in the atmospheric aerosol content owing to natural aerosols. After snow thawing, the air temperature increases thus favoring convection and, hence, entrainment of many soil particles, not fixed by vegetation, into the atmosphere. The summer maximum is associated with dominating air masses coming from south and rich in natural aerosol.

More detailed analysis of the relation between monthly values of concentrations of chemical species and monthly values of τ_{a,λ_0} performed both on monthly and yearly basis gave no positive results, primarily due to different meteorological conditions under which the two variables have been measured. Characteristics of the atmospheric aerosol turbidity can also be measured when the Sun limb is not screened by clouds. At the same time, precipitation falls from clouds, overcast or broken. Besides, washing out of aerosols by precipitation takes place in the under-cloud layer, while the atmospheric aerosol optical thickness characterizes the turbidity of the entire atmospheric column.[8]

The efficiency of aerosol washing out from the atmosphere depends on such factors as the amount, duration, and intensity of precipitation, as well as on the precipitation regime and meteorological conditions. Regular precipitation is more effective than the occasional ones.
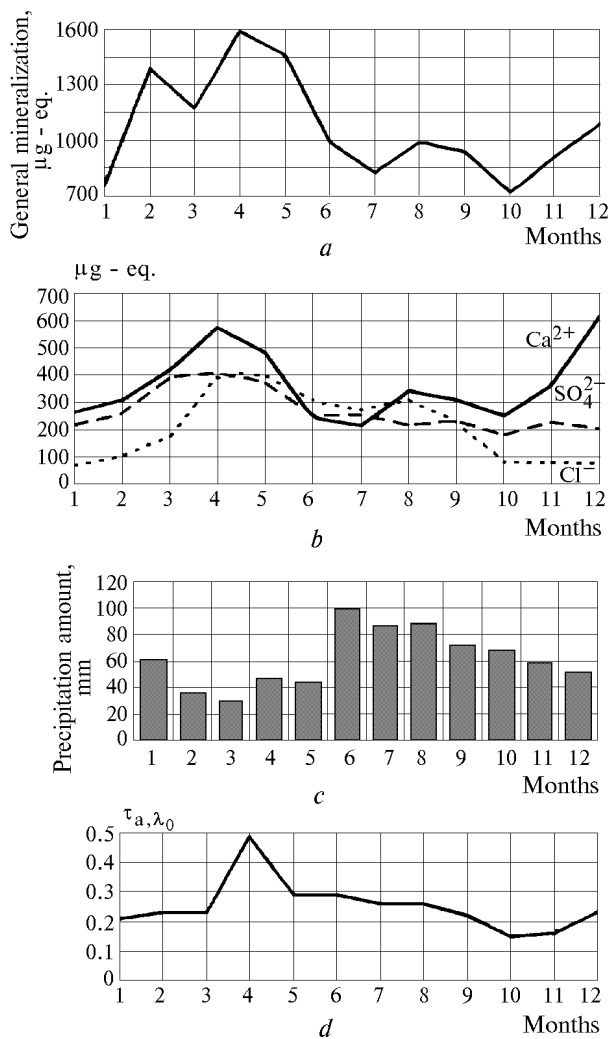
amount reaching 50−60-mm precipitation per month, and then saturates. The cleaning is proportional to the duration of precipitation: 2-h long rain removes dust completely, while 6-min rain (of the same intensity) removes only 18% of the dust. For a given rainfall rate and wind velocity of 3 m/s, no dust was detected in 8 cases of 20 for north winds and in only two cases for south winds.[7] The highest values of $\tau_a\lambda_0$, admixture concentrations, as well as increased acidity of precipitation in the data acquired at MOMSU during a year, occur when south winds dominate. This is because the air masses from Southern Russia and Middle Asia contain more anthropogenic aerosol, and because these air masses are additionally contaminated by anthropogenic aerosol during their passage over south-eastern part of the city where its biggest industrial plants are located. As an example, Table I presents atmospheric aerosol optical thickness before and after the frontal and air-mass precipitation of different duration, amount, and intensity.

Considerable decrease in τ_{a,λ_0} after the passage of frontal precipitation may be due to the change in dominating air mass after the front passage, and not only due to the precipitation cleaning effects. However, it is due to most extended and abundant rainfall that maximum changes in τ_{a,λ_0} occur.

Major factor of precipitation cleaning is the rainfall duration. From May 25 to 27, 1990, the atmospheric aerosol optical thickness has decreased by a factor of 15 after 20 hours of rain, and only by a factor 5 after extremely abundant rain on June 6−7, 1991. The air-mass precipitation caused a factor of 1.5−2 change in τ_{a,λ_0} value measured before and after the rain. The most long-duration and abundant rain on August 10, 1990, has resulted in cleaning of previously very polluted air mass, from the pre-rain τ_{a,λ_0} value, of a factor of 3 larger than the monthly mean value, to the post-rain τ_{a,λ_0} value, not exceeding the average over many years. Thus, a significant dependence of τ_{a,λ_0} on the precipitation duration, amount, and intensity is observed only in the warm season, that is when the atmospheric aerosol concentration is maximum.

FIG. 1. *Annual behavior of general mineralization, concentration of chemical elements, precipitation amount, and* τ_{a,λ_0}, *in Moscow (1982−1991).*

Based on the data of observations at MOMSU, the washing out becomes essential at the precipitation

TABLE I. *Comparison of pre- and post-rain* τ_{a,λ_0} *values.*

Pre-rain date	τ_{a,λ_0}	Rain date	Duration, h	Amount, mm	Post-rain date	τ_{a,λ_0}
		Frontal precipitation				
30.04.90	0.28	1.05.90	2	2.8	2.05.90	0.06
10.05.90	0.14	11 − 12.05.90	2	0.7	12.05.90	0.05
16.05.90	0.22	16 − 18.05.90	7	8.9	19.05.90	0.08
25.05.90	0.46	25 − 26.05.90	20	25.8	27.05.90	0.03
6.06.91	0.50	6 − 7.06.91	7	78.1	8.06.91	0.09
		Air-mass precipitation				
6.07.90	0.79	06.07.90	1	3.1	7.07.90	0.54
10.08.90	0.64	13 − 16.08.90	26	72.9	17.08.90	0.25
18.06.91	0.57	18 − 19.06.91	4	11.5	19.06.91	0.31
5.07.91	0.12	6.07.91	1	0.4	7.07.91	0.08

To study the dynamics of precipitation washing out of aerosol from the atmosphere, we acquired fractional (in the same rain and of equal volumes) samples. Analysis of these samples showed that most ions typically decrease in concentration with time. Figure 2 presents the histograms of concentrations of sulfates, nitrates, chlorides, and ammonium in the successive fractions of rainfall on June 6, 1991. As seen, the concentration of chloride-ion is small in all fractions and being practically the same. This is because this rain was abundant (78.1 mm, an absolute maximum over 37-year observation period), so that samples of individual fractions have already been sufficiently diluted at the time of observations. That is why the chloride concentration rapidly saturated to the value characteristic of clean precipitation at the background stations. Also low are the concentrations of other ions. On 18 June, 1991, the amount of precipitated rainwater was close to the average value and rain was not too long; so only 3 fractions were acquired, and the samples were small in volume, so that concentrations of all ions were higher in them (Table II).
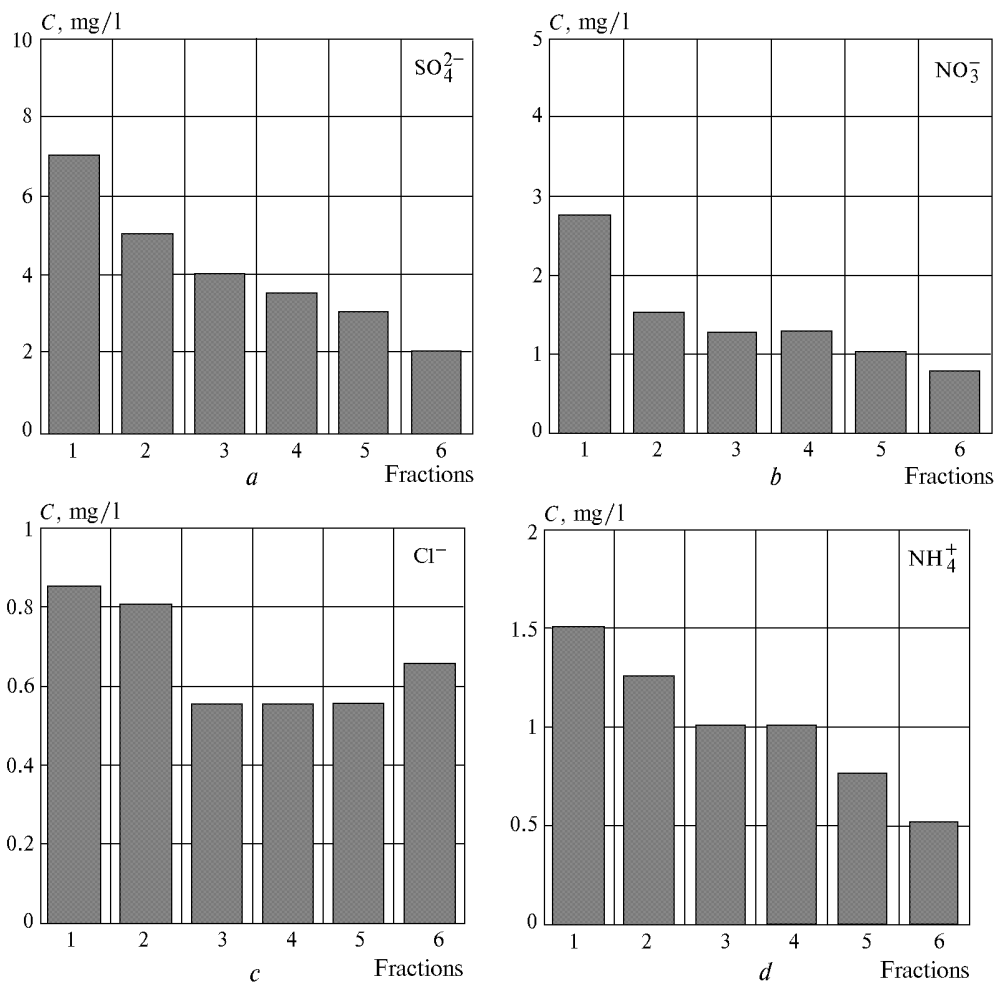

FIG. 2 Concentrations of sulfates (a), nitrates (b), chlorides (c), and ammonium (d) in the successive rainwater fractions, June 6, 1991.

TABLE II. Chemical composition of rainwater fractions.

Fraction number	Time of sampling, min	Ion concentration, mg/l				
		HCO_3^-	SO_4^{2-}	Cl^-	NO_3^-	NH_4^+
Constant-rate rainfall						
1	35	0	15.0	4.1	2.7	2.2
2	40	0	11.2	2.7	2.3	1.1
3	57	0	8.1	2.4	2.1	0.9
Variable-rate rainfall						
1	8	1.4	2.3	3.5	0.9	1.6
2	45	6.0	16.2	18.0	7.1	6.5
3	10	1.2	2.0	3.2	0.6	1.5

It is seen that the cleaning effect was maximum during the initial 35 min into the rain. However, by the time of taking the third fraction, the atmosphere was far from being clean, because the post-rain τ_{a,λ_0} value exceeded the many-year mean value. The precipitation rate was fairly constant in these cases.

The situation is different when precipitation rate varies during the sampling time (see Table II). Then, given the same amount of the precipitated rainwater as in the above case (10.9 mm), it took different times to collect equal-sized samples. Initially, the rain was heavy, with large and rapidly falling drops. So it took us about 8 s to gather the first fraction. Then, the rainfall rate substantially decreased, and it took us 45 min to collect the second fraction. The smaller droplets have larger total area and longer lifetimes in the atmosphere, thus absorbing more aerosol, what explains higher ion concentrations in the second fraction. Finally, the rain intensified again, so that we could collect the third fraction as rapid as the first one. However, because of again small lifetimes of large drops and the sufficiently cleaned atmosphere, during the measurements, aerosol concentration in the third fraction is lower than in the first one.

With concentrations of all ions in the atmospheric precipitation having been known, it is possible to estimate its salt content. In the region under study, 95 to 160 kg/ha of salt falls out annually, with the main contribution coming from sulfates (30−40%).
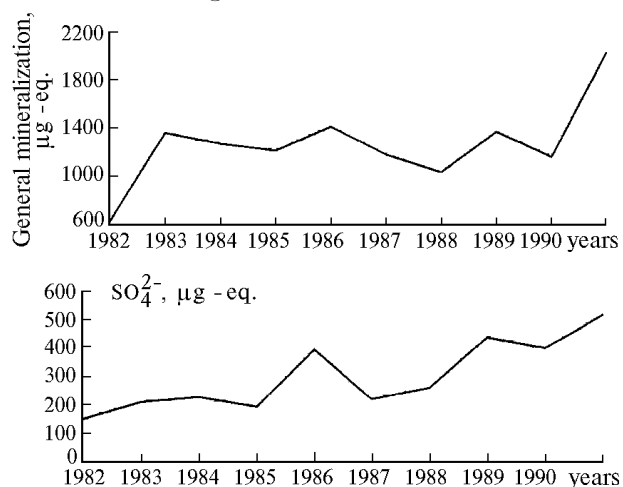

FIG. 3. *Many-year variations of the total mineralization of precipitation*, SO_4^{2-}.

Analysis of annual variations of some ions showed no any tendency to increase in concentration. However, there is a substantial growth of sulfates in recent years, these being the main industrial pollutants in precipitation, which, together with the positive trend in τ_{a,λ_0},[10] is indicative of the growing anthropogenic influence on aerosol pollution of the atmosphere over the city (Fig. 3).

Thus we can state that the aerosol optical thickness of the atmosphere and composition of atmospheric precipitated water bear information on the aerosol pollution of the atmosphere. The comparison made between the values of τ_{a,λ_0} measured before and after the rain, as well as chemical composition of the fractional samples of precipitation, show that the duration and intensity of precipitation are the factors that determine the cleaning effect of precipitation. Since washing out of atmospheric admixtures is different for different admixtures, further study of this problem needs for measurements of chemical composition of dry aerosol.

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