CORRELATION BETWEEN ICE-FORMING PROPERTIES AND SIZE OF PARTICLES IN THE ATMOSPHERIC AEROSOL

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This paper presents analysis of the results obtained during simultaneous measurements of concentration of atmospheric ice nuclei, concentration of Aitken nuclei, and size spectra of aerosol particles in the range from 0.01 *to* 10 μm. *The measurements have been performed in Moscow region in 1993–1997. The data of 15-month observations demonstrate that concentration of ice nuclei not always correlates predominately with that of large aerosol particles. In many cases, correlation between concentrations of ice and Aitken nuclei predominates, or one can see that concentration of ice nuclei correlates with that of large and small particles simultaneously. There were periods of full absence of correlation between concentration of ice nuclei and dispersity of atmospheric aerosol. The periods coincided with time of frequent changes in weather and synoptic conditions.*

Some experimental data demonstrate that ice-forming nuclei of the atmospheric aerosol are mostly large particles.[1] This was most clearly demonstrated by Georgi and Kleinjung[2] with the results of a series of daily three-time measurements of concentration of ice nuclei (IN), Aitken nuclei (AN), and particles larger than 0.3 μm in size. The measurements have been being carried out during a month at the Kleiner Feldberg mountain, 20 km to the north of Frankfurt on Main. Concentration of ice nuclei well correlated with that of large particles (correlation coefficient value being about 0.87). At the same time, there were no any correlation observed between the ice nuclei and Aitken nuclei.

Higher content of ice nuclei in the coarse-disperse fractions of atmospheric aerosol was found in measurements of IN in the free atmosphere.[3,4] Similar results well agree with the physical knowledge about ice-forming ability as a function of the basic nucleus size. However, the variety of chemical composition of aerosol, especially in the industrial regions, as well as variations of the weather and synoptic conditions during the longitudinal measurements can, evidently, make the relationship between the ice-forming ability and size of atmospheric particles too complicated.

A year long series of aerosol measurements has been carried out in Moscow region by the Central Aerological Observatory (CAO) in 1987. This series has revealed correlation neither between concentration of ice nuclei and total concentration of submicron aerosol, nor between the concentration of IN and that of large particles with the diameter from 0.2 to 1 μm.[5]

According to analysis we have performed of two series of measurements carried out in 1993 (Ref. 6), the correlation between ice-forming ability of atmospheric aerosol and size of aerosol particles considerably varies being different in different periods and seems to be different in different seasons. The data of systematic measurements compiled in CAO on ice nuclei characteristics and on the atmospheric aerosol allow one to analyze correlation between the ice nuclei and particles' size based on a larger data set.

Aerosol measurements had been performed during 11 years at CAO, Dolgoprudnyi, Moscow region, Russia since 1987. We have chosen from this series the systematic measurements from one to several-month duration. In these series, the concentration of IN was measured simultaneously with the total concentration of Aitken nuclei (Scholtz's counter), size spectra of particles in the range from 0.01 to 1 μm (a TSI 3030 electrostatic analyzer), and the size spectra of large nuclei in the range from 0.3 to 100 μm (a PKZV–906 device). The device PKZV–906 has been periodically tested and adjusted by the manufacturer. We have chosen, from these measurement sessions, the data acquired during the periods of its most reliable operation. All in all, 15 months were selected for the analysis. Those include two half-year (January-June) series of measurements in 1994 and 1997, and three-month measurements in summer and fall of 1993 and 1994.

The measurements have been carried out during working days from 9 to 10.30 a.m. Atmospheric air was sampled at a height of 12 m above the Earth's surface. The air was pumped into an aerosol reservoir of 1 m^3 volume inside a laboratory from which it was then sampled by all the measurement devices. Concentration of ice nuclei was measured with a cloud device "SALYA"[7] at a constant temperature of −20°C. The total number of simultaneous measurements of disperse and ice-forming characteristics of atmospheric aerosol involved in the analysis equals to 323.

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The data obtained have been analyzed on a monthly basis. For each month we have determined the coefficients of correlation between the concentration of ice nuclei and that of atmospheric aerosol particles measured in 8 channels (size range from 0.01 to 1 μm) of the TSI–3030 analyzer, as well as between the concentration of ice nuclei and total concentration of aerosol particles measured with this instrument. Similarly, the correlation coefficients between the IN concentration and concentration of large nuclei, the total one and in 6 size ranges (from 0.3 to 10 μm) have also been determined from the data acquired with a PKZV–906 device. The data acquired in the size range from 10 to 100 μm, though available, have been excluded from the analysis due to small number of such particles in atmospheric aerosol ensembles. The coefficient of correlation between the IN concentration and the concentration of Aitken nuclei has also been calculated. These measurements have been done with a Scholtz counter.

The correlation coefficient $R \geq 0.5$ was conventionally taken as the threshold value of significant correlation. So the cases with the correlation coefficient $R \geq 0.5$ have been first isolated and involved in correlation analysis. The frequency of detecting and the distribution of $R \geq 0.5$ values over the particle size demonstrates that during 15 months of the measurements considered, significant correlation between IN concentration and concentration of aerosol particles of various sizes existed during 12 months of the total duration of 15 months. During the other months, all the correlation coefficients were less than 0.5. Significant correlation between the IN concentration and the concentration of particles having sizes above 0.3 μm was found among 10 months. For 8 months the correlation have been revealed between the IN concentration and either the particles from the size range of the TSI–3030 instrument (0.01–1 μm) or the Aitken nuclei. In 8 of the 10 months, the correlation was observed simultaneously between the IN and the number of large particles of one or other size from the range accessible to a PKZV–906 device (0.3–10 μm) and the concentration of particles from the size range from 0.01 to 1 μm. In 2 months, IN concentration correlated only with the concentration of large particles.

There were no a month when IN correlated with the particles from the size range $0.01 < d < 1$ μm while not correlating with large particles. The concentration of ice nuclei correlated simultaneously with the concentration of Aitken nuclei, concentration of large particles, and particles of 0.01 to 1 μm size in 5 months. In 3 months no correlation has been revealed between the IN concentration and the concentration of Aitken nuclei while, at the same time, their concentration correlated with the concentration of large nuclei from the measurement range of PKZV–906 and small ones from the range of the TSI–3030. During 1 month the concentration of IN correlated with the concentration of Aitken nuclei ($R = 0.51$) while, at the same time, showing no significant correlation with the large particles and particles with the size $0.01 < d < 1$ μm.

Comparison of the correlation coefficients demonstrates that, during 7 of 12 months, the significant correlation was the highest between the IN number and large particles (PKZV–906). During 5 months the correlation was the highest between the IN and fine particles (from the measurement range of the TSI–3030 instrument or Aitken nuclei). The maximum values of the coefficient of correlation between the IN and particles of different sizes were 0.87 for large particles (PKZV–906), 0.87 for particles from the size range of TSI–3030, and 0.75 for the Aitken nuclei. On the whole, consideration of correlation coefficients greater than 0.5 did not reveal any distinct correlation between the ice nuclei and coarse fraction of the atmospheric aerosol, though certain preferred correlation between the concentration of ice nuclei and large particles has been reaveled.

Taking into account that the ranges of particles' size detectable with the PKZV–906 and TSI–3030 devices partially overlap, the following step was to determine the mean coefficients of correlation between the IN and the concentration of particles in the size ranges out of the overlap, that means from 0.02 to 0.3 μm (measured with a TSI–3030 and called the medium-sized particles), and from 0.5 to 10 μm ("large" particles measured with a PKZV–906 instrument).

Coefficients of correlation between the IN concentration and the concentration of aerosol particles in these size ranges, and those for concentrations of Aitken nuclei are presented in Fig. 1 for the half-year series of measurements in 1994 and 1997.

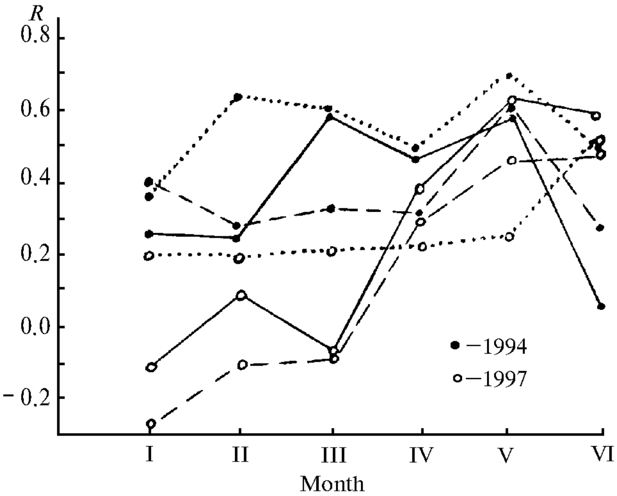

FIG. 1. Coefficients of correlation between the IN concentration and the concentration of Aitken nuclei (dotted lines), medium-sized (dashed lines), and large (solid lines) particles in half-year series of measurements in 1994 and 1997.

Figure 2 presents maximum values of the correlation coefficients of IN observed within the same ranges of particles' size.

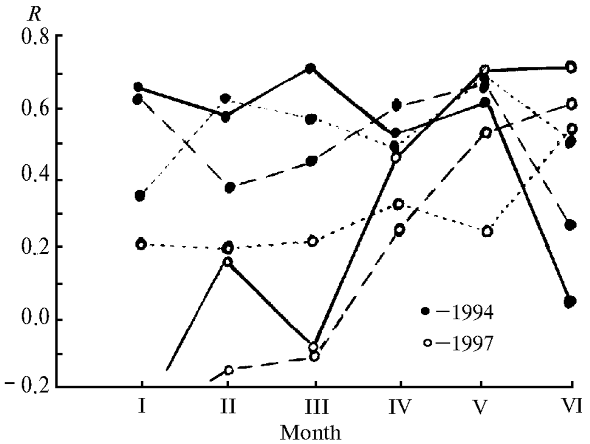

FIG. 2. Maximum values of the coefficients of correlation between the IN and aerosol particles in the ranges of large- (solid lines) and medium-sized (dashed lines) particles. Dotted lines correspond to the coefficients of correlation between the IN and the concentration of Aitken nuclei.

The Table I presents similar mean and maximum values of the correlation coefficients for months of measurements in 1993 and 1994. Figures 1 and 2 and the Table present all the correlation coefficients including the values of R less than 0.5.

TABLE I. The mean and maximum values of the coefficients of correlation between the IN concentration and the concentrations of particles from different size ranges according to measurements in 1993 and 1994.

Particles	Values	1993			1994
		Month			
		VII	X	XI	X
Large	Mean	0.64	0.74	−0.21	0.82
	Maximum	0.71	0.79	−0.27	0.87
Middle	Mean	0.8	0.8	0.47	0.57
	Maximum	0.82	0.87	0.64	0.82
Aitken nuclei		0.75	0.65	0.40	0.69

Analysis of the correlation coefficients that are presented in Figs. 1 and 2 and in the Table allows to draw certain conclusions. The correlation between the ice nuclei and atmospheric aerosol particles considerably varies both during a year and from year to year. There are months (mainly during summer and fall) when correlation between the IN number and concentration of aerosol particles of different size is high. At the same time, one can observe rather long periods when the IN concentration has, in fact, no relation to the disperse composition of aerosol (1997, January–March).

Predominant correlation of ice nuclei with large aerosol particles is a regularly observed fact. Although the correlation between the concentration of IN and that of the aerosol particles was the highest for large particles (1997, May–June; 1994, October), the coefficients of correlation between the IN and concentration of particles of different sizes proved, on the whole, to be rather close to each other. In some periods (e.g., 1994, January–May), the correlation between the IN concentration and that of the Aitken nuclei was, on the average (see Fig. 1), even higher than with the concentration of particles of large sizes.

The agreed behavior of the coefficients of correlation between the IN and concentration of particles from different size intervals that may be seen in Figs. 1, 2, and in the Table, especially with the large- and medium-sized ones, is of a particular interest.

During the periods when high correlation occurred between the IN and large nuclei, significant correlation has also been observed between the ice nuclei and the concentration of medium-sized and small particles (1993, July, October; 1994, March, May, October; 1997, May–June).

Close coincidence of the coefficients of correlation between the IN and the concentration of particles of different sizes can be explained by the presence of correlation between the concentration of particles from different parts of the size spectrum. Indeed, analysis of the correlation between the concentration of Aitken nuclei, and large- and medium-sized particles demonstrates that this explanation is reasonable in more than a half of the cases. Thus, the absence of a pronounced correlation between the IN and large particles and the presence of a simultaneous correlation between the IN and large and small atmospheric nuclei can only partially be taken as an indication to a uniform distribution of the ice nuclei over the size spectrum of atmospheric aerosols.

The periods when no correlation occurred between the ice nuclei and the particles from all size ranges are also of certain interest. To find an explanation of such situations, we have analyzed the weather and synoptic conditions in the months with high and low correlation between the IN and aerosol particles. According to this analysis, the correlation between the ice nuclei and disperse composition of the atmospheric aerosol is influenced by the stability of the synoptic and weather situations. The coefficients of correlation between the IN and aerosol particles (first of all, large ones) decrease in months of high cyclone activity with frequent change of air masses, winds, and precipitation. In this respect, it is instructive to compare the data obtained June of 1994 and in June of 1997.

As seen from Figs. 1 and 2, the coefficients of correlation between the IN and the concentration of aerosol particles of all sizes were high enough. In June, 1994 no correlation has, in fact, occurred between the IN and the concentration of large- and medium-sized particles. In 1997 the anticyclone situation with stable weather without precipitation prevailed during this month. In June, 1994 there happened four cyclones and changes of air masses. Precipitation of different intensity occurred on 13 days of the 20 ones considered.

Similar dependence of the correlation between the IN and the concentration of aerosol particles on the synoptic situations has also been revealed for other months when the measurements have been carried out. Variations of the total aerosol concentration and of the ice nuclei content in aerosol that are caused by changes in the air masses and precipitation obviously mask the connection between the ice nuclei and size spectrum of the atmospheric aerosol. The seasonal behavior of the correlation coefficients with a minimum in winter months seems to be explained by same causes.

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