STUDY OF DISPERSED AEROSOL COMPOSITION AND CONCENTRATION OF MINOR GAS COMPONENTS IN TEMPERATE AND ARCTIC AIR MASSES

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The data are presented on the dispersed aerosol composition and concentration of minor gas components in the surface layer of the atmosphere for temperate and Arctic air masses obtained in an expedition far from large sources of anthropogenic emissions (Chik settlement, 20–26 February 1996). Interrelation of aerosol characteristics with minor gas components and meteorological parameters is discussed.

Atmospheric aerosols variously affect the Earth's radiative balance. Backscattering (reflection back into space) and absorption of solar radiation by aerosol particles decrease the portion of energy received by the surface.[1] This is so-called "white house" effect or the direct aerosol effect resulting in cooling of the Earth. Another mechanism (indirect aerosol effect) is that the aerosol particles affect the radiative balance taking part in cloud formation. From the data of Global Historic Climate Network (GHCN), the maximum rate of warming, about 3°C/century, was observed over the course of the past century in the Siberian region, whereas the average global rate of warming was 0.4°C/century. Thus, Siberia is the important and peculiar object from the climatological viewpoint. At the same time, the Siberian aerosol is understood much less than the aerosol in other regions.

POSITION OF OBSERVATION POINT AND METEOROLOGICAL CONDITIONS

Investigations were carried out on the basis of the Mobile Ecological Laboratory of the EKOS group of the Institute of Chemical Kinetics and Combustion of the SB RAS. The Laboratory was capable of performing continuous twenty-four-hour measurements of a number of aerosol and gas components in the atmosphere.

The aerosol particle size spectrum was measured with a complex comprising the AZ-5 photoelectric counter and the DSA diffusion aerosol spectrometer built around a screen diffusion battery. The following characteristics of the aerosol particle size spectrum are considered in the present paper:

– number density of the coarse (larger than 0.7 μm) aerosol particle fraction (AZ-6);

– total surface area of particles larger than 0.7 μm;

– number density of the finely dispersed aerosol particle fraction, between 2 and 300 nm (DSA);

– number density of aerosol particles between 2 and 4 nm (the ultradisperse fraction). The minimum particle size was determined by a threshold of a condensation coarsening device and the maximum particle size – by aerosol settling on screens of the first cascade of the battery.

The mass concentration of ammonium sulfate aerosols was measured with a thermodenuding system[2] and a fluorescent SO_2 analyzer. The same analyzer was used to measure sulfur dioxide gas component.

The challenges of the expedition were to investigate the dispersed aerosol composition and the concentration of minor gas components in air masses and to eliminate, where possible, the effect of anthropogenic emissions of Novosibirsk. To attain these ends, the position of observation point was chosen in the region of Chik settlement.

Chik settlement is 30 km to the west of Novosibirsk; the measurement point was 4 km to the north of Chik. The observation site was relatively far from localities (Chik settlement was the closest locality) and hence the density of sources of anthropogenic emissions was low.

Meteorological conditions during our expedition were as follows (Fig. 1). From February 20 to 22, the southwest wind was observed with a typical speed of 2 m/s, corresponding to the temperate air mass (in accordance with classification of Ref. 3). From February 24 to 26, the north wind was blown with an average wind speed of 3 m/s, corresponding to the Arctic air mass.[3] The validity of this classification was confirmed by the data of the Western Siberian Administration on Hydrometeorological Service for that period. These data indicated that the temperate air mass passed through the Southern Ural region to the measurement point and the Arctic air mass passed through the Dikson peninsular. The air mass changed for one and a half day on February 22–23. The enhanced ozone concentration (see Fig. 1) caused by accumulation of ozone from underlying air layers due to intensification of vertical transfer is typical of atmospheric changes of this type.

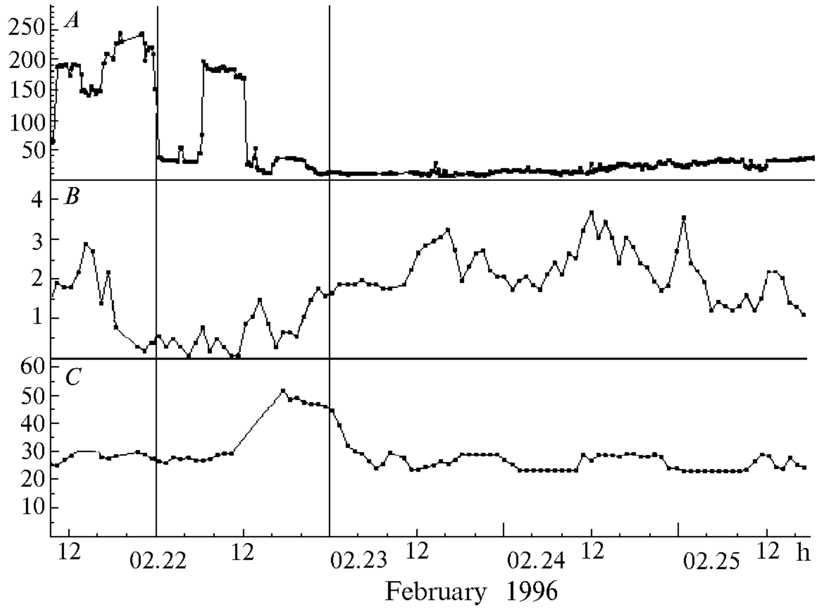


FIG. 1. Meteorological parameters during the expedition: A is the wind direction, in deg*; B is the wind speed, in* m/s*; C is the ozone concentration, in* ppb*, in Chik.*

AEROSOL MEASUREMENTS

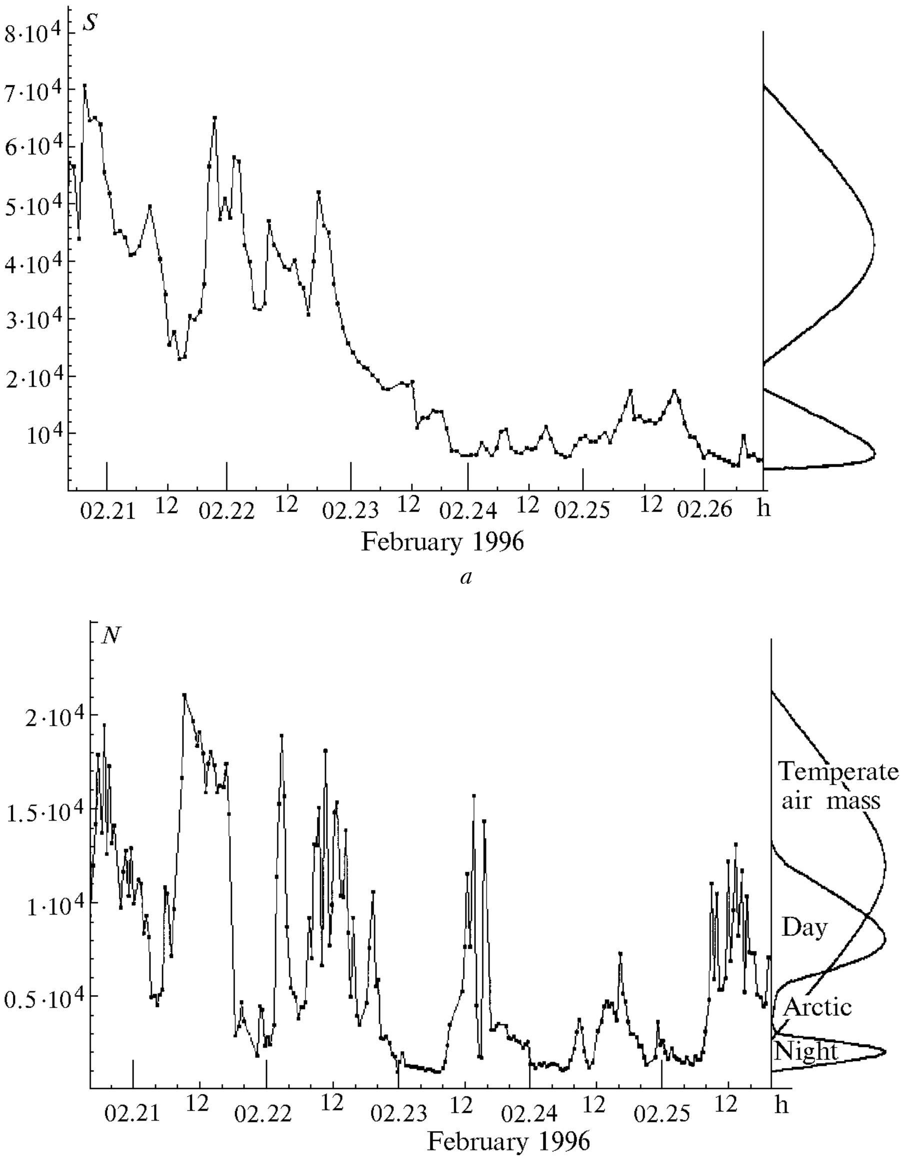
Change of air masses affected the number density and the dispersed composition of aerosol particles. For the temperate air mass (February 20–22) the characteristic number density of coarse particles was 35 cm^{-3}; in the transition period (February 22–24), the particle number density decreased uniformly and fell to its background value in the Arctic air mass being equal to 4 cm^{-3} that increased in the daytime, that is, possessed diurnal variations.

The total (integral) surface area of particles of this fraction was 40 $\mu\mathrm{m}^2/\mathrm{cm}^3$ (Fig. 2*a*) for the temperate air mass and 5 $\mu\mathrm{m}^2/\mathrm{cm}^3$ for the Arctic air mass and varied in the same manner. It increased for the Arctic air mass up to 13 $\mu\mathrm{m}^2/\mathrm{cm}^3$ in the daytime.

Sharp stochastic variations of the number density of the finely dispersed particle fraction of about 11 000 cm^{-3} were observed over a period of passage of the temperate air mass; the particle number density decreased to a background level of 2 000 cm^{-3} already in the transition period with sharply pronounced diurnal variations to 15 000 cm^{-3}. Frequency spectra for the temperate air mass and daytime and background (nighttime) Arctic air masses are shown in Fig. 2*b*.

The average number density of the ultradisperse fraction (2–4 nm) was 40 cm^{-3} for the temperate air mass and reduced to a background value of 10 cm^{-3} by the beginning of the transition period with daytime maxima up to 180 cm^{-3}.

A peculiarity of the particle number density for this fraction in the Arctic air mass is its lower level without significant maxima. After the sunset the number density of these particles decreased monotonically for 3–5 hours.



a

b

FIG. 2. Variations of the total specific surface area of aerosol particles (S, μm^2/l*) larger than* 0.7 μm *(a) and of the particle number density N, in* cm^{-3}*, of the finely dispersed particle fraction with their frequency spectrum in Chik.*

MEASUREMENTS OF THE CHEMICAL COMPOSITION AND THE CONCENTRATION OF GASEOUS COMPONENTS

The mass concentration of ammonium sulfate (Fig. 3) and its main product – sulfur dioxide – was measured simultaneously with the parameters of aerosol particles.

An analysis of the obtained data indicates the diurnal behavior of ammonium sulfate concentration against the background of its synoptic variations (the curve was drawn through nighttime points). The duration of the daytime maximum of ammonium sulfate concentration was (10 ± 20%) h. A start of the increase of sulfate concentration was delayed by 3–4 h relative to the solar illumination. The daytime concentration maximum was observed when the illumination started to decrease (at 15 h, LT).

It also should be noted that the rate of the concentration increase was 9 times greater in daylight than at night.

In comparison with the number density of aerosols that changed by an order of magnitude when the air mass was changed, the concentration of ammonium sulfate changed insignificantly (the relative standard deviation was 25%). The concentration of sulfur dioxide behaved similarly to the ammonium sulfate concentration. Its average level was 4–6 ppb over the entire measurement period.

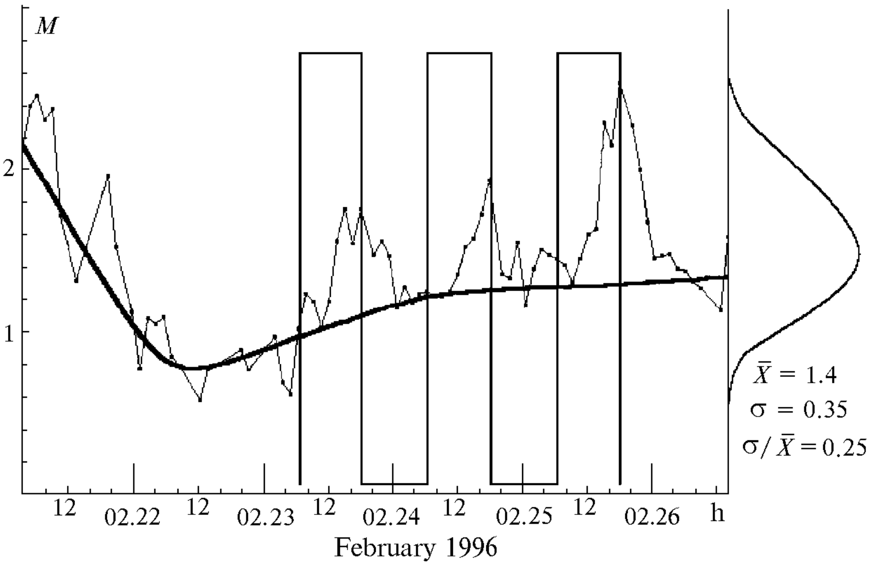


FIG. 3. Mass concentration of ammonium sulfate in aerosol particles (M, μg/m³) with their frequency spectrum in Chik (daytime is shown by the meander).

DISCUSSION OF RESULTS

Ammonium sulfate aerosols

An analysis of the atmospheric pressure during the expedition time showed that on February 12–13 it sharply increased up to its maximum value (770–775 mm Hg) recorded in winter of 1996. In the next 5–6 days, the pressure preserved its high level, which was a record for this winter. The pressure started to decrease from February 19 with an average gradient of about 5 mm Hg/day and from February 22 the pressure gradient became positive and equal to 3 mm Hg/day. We note that the behavior of ammonium sulfate in these days practically coincided with the atmospheric pressure behavior. Their minima agree to within 12 hours.

Selection of clear–air conditions against the criterion of total specific surface area of aerosol particles

During the expedition, the total specific surface area of particles larger that 0.7 μm decreased from 45 to 5 μm²/cm³ (see Fig. 2) when the air masses changed. As known from the literature, the same specific surface area of aerosols was observed practically over the entire Pacific Ocean.[4] Weber and McMurry[5] reported that on Hawaiian islands they observed formation of new finely dispersed particles (3–4 nm) from the gaseous phase in clear–air masses with a heterogeneous particle surface area of about 4 μm/cm³. We may assume that in our situation with the same particle surface area homogeneous mechanisms of particle formation dominate. This allows us to classify the period of passage of the Arctic air mass as the clearest–air situation.

Daytime maxima of finely dispersed particle number density under clear–air conditions

For the temperate air mass, we observed the maximum number density of particles larger than 300 nm at random any time of the day (see Fig. 2*b*). To the contrary, the increase of the particle number density in the Arctic air mass was clearly correlated with the intensity of illumination. This correlation may be due to photochemical processes, for example, due to the reaction of sulfur dioxide with OH radicals in sunlight.[6]

Correlation of ultrasmall particle number density with illumination

A scheme of formation of ultrasmall particles assumes that in the clear–air atmosphere particles may grow in sunlight from gas molecules forming large number of dimers, smaller number of trimers, and so on.[7]

This process continues until the particle size reaches its quasistable value. Our instrumentation was capable of recording particles starting from a diameter of about 2 nm. We selected the ultradisperse aerosol fraction (2–4 nm) as an indicator of formation of new particles through homogeneous condensation from the gas phase. It is likely that we observed this process in Chik under clear–air conditions (on February 24–26, see Fig. 4). The small particle number density sharply increased after the sunrise and the particle average size decreased[8] from 37 nm at 5 a.m. to 11 nm at 1 p.m. (Fig. 5). The number density of the ultradisperse particle fraction maximized in the daytime and reduced to zero at night. After the sunset the rate of the particle number density decrease was not so high as the rate of the particle number density increase in the morning. Particles decreased their number for 2–4 h.

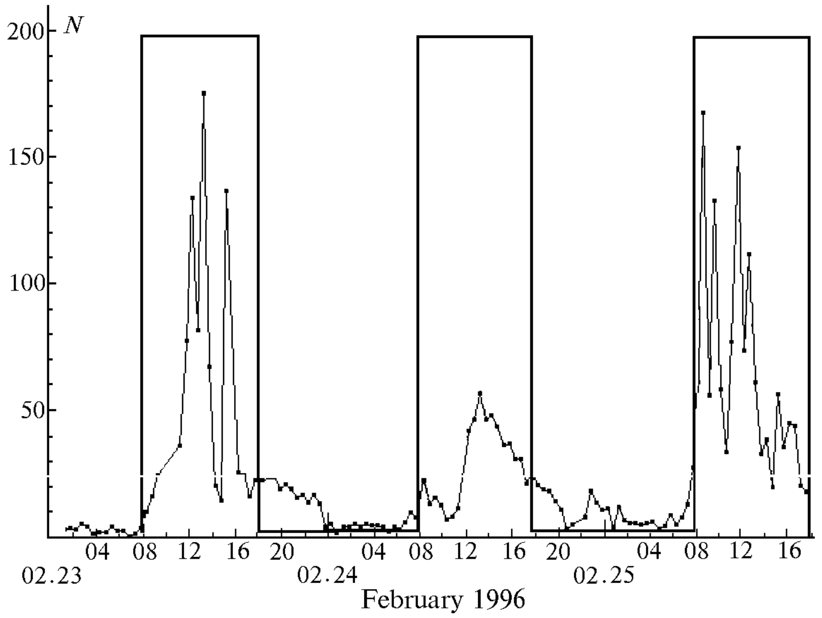


FIG. 4. Number density of ultrasmall particles N, in cm^{-3}*, in the Arctic air mass (daytime is shown by the meander).*

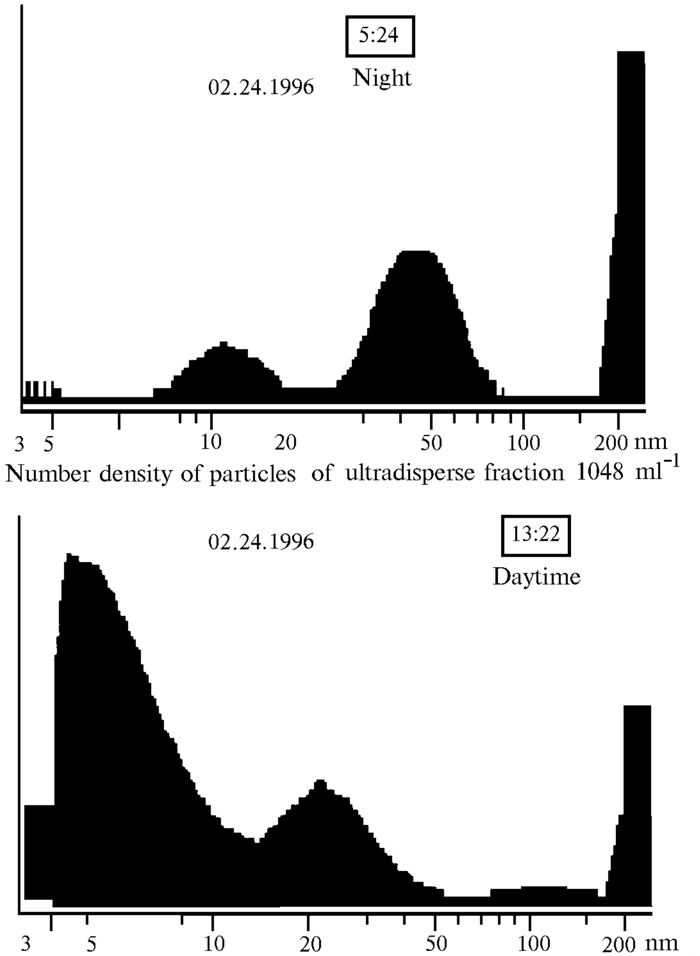


FIG. 5. Size distribution of particles of the finely dispersed fraction at night and in the daytime.

Considering the behavior of the number density of finely dispersed particles and their insignificant heterogeneous surface area, we may suggest that the daytime increase of the ammonium sulfate concentration in the atmosphere[9] is connected with homogeneous formation of new particles with predominant sulfate composition.[9] Clarke[4] who organized expeditions in clear–air masses in the Pacific Ocean was also inclined to believe that such was the case.

CONCLUSION

As a result of our measurements during the expedition, we have distinguished the Arctic and temperate air masses against their meteorological parameters, measured the aerosol characteristics and concentration of minor gas components typical of these air masses and of the period of their change. It was found that the parameters of the aerosol phase (number density, specific surface area, and so on) differed by an order of magnitude for the examined air masses, whereas variations of the concentration of the ammonium sulfate aerosol did not exceed 25%. Relying on this, we set up the hypothesis that ammonium sulfate is well mixed in the Northern hemisphere.

The clearly pronounced diurnal variations were established for some measurable components of the atmosphere in the Arctic air mass (for example, for the number density of small particles, concentration of ammonium sulfate, etc.) It was found that the rate of the increase of the sulfate concentration in daylight was 9 times greater than at night.

Our results together with an analysis of few data obtained in clear–air regions of the Earth (the Pacific Ocean, Hawaiian islands) showed that the sulfate aerosol was largely formed through homogeneous condensation in the Siberian Arctic air mass.

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