Organization of monitoring of the greenhouse gases and of the components oxidizing the atmosphere over Siberia and some results obtained. II. Aerosol composition

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Data of long-term monitoring of the aerosol fine fraction in the ground atmospheric layer and free atmosphere have been analyzed. It is shown that despite the maximum in the generation of aerosol-forming substances in summer, the minimum in concentration of nanoparticles in the ground layer is observed. This can be explained by their transport to higher layers, because their concentration inversely depends on the height of the mixing layer.

Introduction

Multilevel system for monitoring of greenhouse gases and components oxidizing the atmosphere developed under the international cooperation project and some results obtained by now on gas composition of air over Siberia were described in the first part of this paper.[1]

Let us remind that monitoring is being carried out under three programs and extension of the study on a wider area is supposed to be done later on. The first program includes detection of greenhouse gas emissions in Iksinskoe (Bakcharskoe) marsh. Here chambers are mounted, from which air is pumped through two sensors sensitive to the methane and carbon dioxide.

The second program is devoted to monitoring of the flux in the atmospheric boundary layer. Spatial distribution of the greenhouse gases was measured with the help of towers located at the Karasevo station, Tomsk Region; Berezorechka village, Tomsk Region, the Noyabrsk town, Demyanskoe village, Tyumen Region; Igrim village. Each of the towers is equipped with identical sets of instruments. Measurements have been carried out continuously with the intakes mounted at two heights.

The third program is aimed at the study of spatial distribution of the components under study. Vertical distribution of greenhouse gases in the atmospheric boundary layer is measured from onboard an AN-2 aircraft. The flights are performed weekly. AN-30 aircrafts are used for experiments on assessment of the regional contribution to the global balance. Sampling and measurements of the air gas composition are carried out on the 20ies of every month depending on the weather. The sounding site was chosen southwest of Novosibirsk for the urban plume not to affect the instruments. Measurements are performed over the pine woods near Zavyalovo and Ordynskoe settlements. The height range is 500–7000 m. The project YAK-1 has been started in 2006, in which the measurements have been carried out with the same instruments along the route from Novosibirsk to Yakutsk and back to Novosibirsk at pre-selected heights of flight.

Along with the measurements of gas components at a number of measurement sites, aerosol content is monitored. The results on the disperse composition of aerosol are given in this paper.

1. Seasonal behavior of the fine aerosol fraction in the ground atmospheric layer

Earlier we have analyzed the diurnal behavior of the fine aerosol fraction in the ground layer.[2,3] In this paper, we consider seasonal dynamics of the fraction.

Summer maximum is expected in the seasonal behavior by analogy with the diurnal behavior of concentration of the nucleation aerosol subfraction, which evidently reflects aerosol generation processes. However, quite reverse situation is evident from data presented in Fig. 1. The minima of monthly mean concentrations of both fine aerosol and particles of the nucleation mode fall on summer months. Another characteristic feature of the annual behavior is that one of the maxima is observed in spring, which can be considered characteristic since it is pronounced on all the curves depicted in Fig. 1. Quite similar annual behavior is characteristic of the submicron aerosol $0.1 < d < 1.0$ μm.[2]

Assessments of particle generation rates[3] point out to the fact that nucleation processes do not weaken in summer while the particle production rates insignificantly exceeds those in winter. Then, because of the locality of *in situ* processes, one can assert that the intensity of fine aerosol generation has no strong seasonal dependence. First, this seems to be

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illogical since the source of both primary aerosol and substances which are precursors of secondary aerosol is the underlying surface. This means that in summer there is larger amount of substances to generate fine aerosol. Remind, that according to the classical nucleation theory[4,5] the production rate of stable nuclei strongly depends on the air temperature, i.e., at the same concentration of vapor of aerosol forming substance (VAFS) the nucleation rate is higher at low temperatures. On the other hand, plenty of VAFS in summer results in nucleation acceleration and to the increase of clusters mobility followed by the decrease of their sizes. The probability of formation of aerosol nanoparticles essentially decreases then, as the nuclei quickly sink to atmospheric aerosol.

Thus, if the behavior of aerosol generation remains relatively stable within a year, then aerosol concentration in the surface layer is to be defined by the volume within which the aerosol is dispersed, i.e., by the height of the mixing layer. According to airborne sounding data for Western Siberia,[6,7] the annual behavior of the mixing layer height is reverse and shows its maximum in summer. This means that the surface concentration in summer is to be diluted due to the turbulent redistribution of aerosol particles in a large air volume of the atmospheric boundary layer. The smearing of the maximum in the diurnal behavior of the total number concentration of the fine aerosol most likely takes place due to the above circumstance[3] since the height of the mixing layer in summer varies noticeably during a day. To check up this fact, data on vertical distribution of aerosol particles in the troposphere are required. Such data have been obtained from onboard an AN-30 "Optik-E" instrumented aircraft.

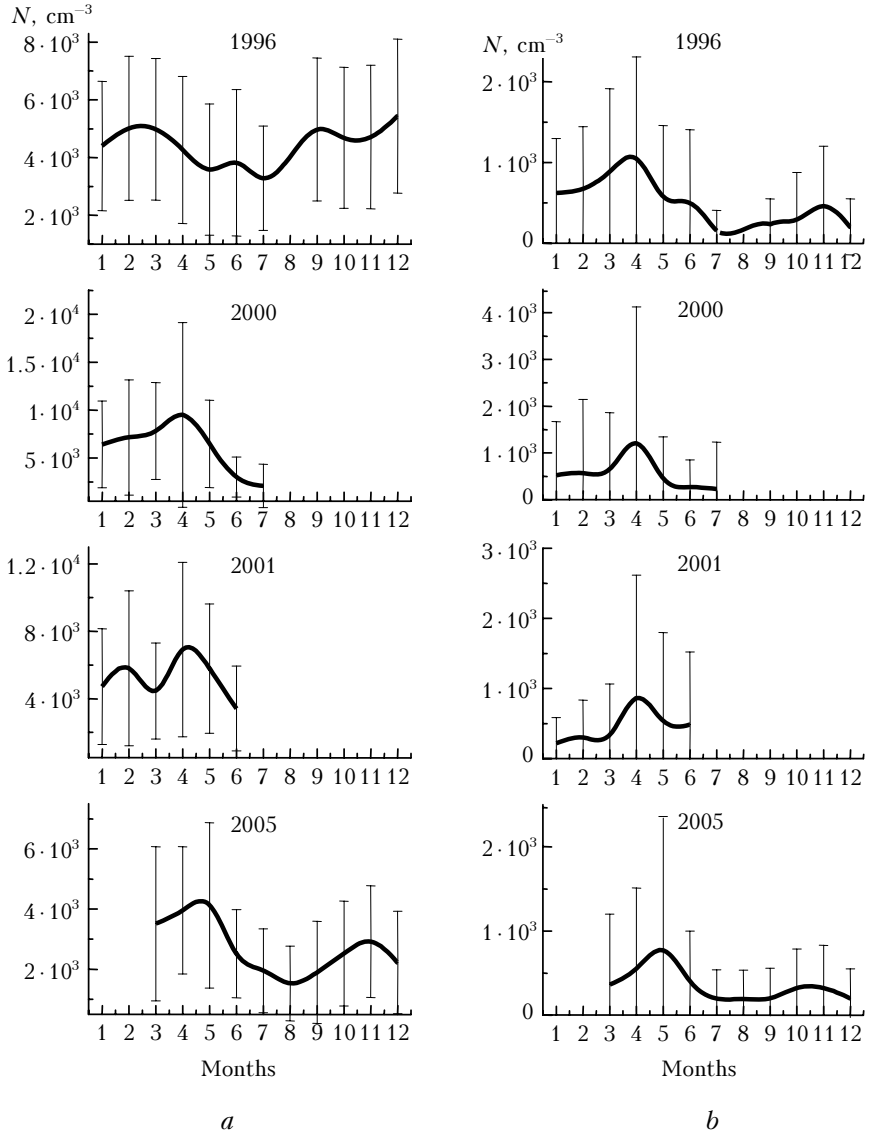

Fig. 1. Annual behavior of the total number concentration of fine aerosol (*a*) and particles of the nucleation mode (*b*).

2. Seasonal dynamics of the vertical distribution of nanoparticles

First, consider seasonal behavior of the concentration of fine aerosol at different heights in the atmosphere (Fig. 2).

As is seen from Fig. 2, one of the concentration minima at the height of 500 m is observed in summer months as in the case with the surface layer aerosol. The characteristic feature in this case is sharp decrease of the concentration in winter because of a barrier layer arising due to temperature inversions, observed during winter flights. As a result, the aerosol generated in the surface layer cannot reach the height of 500 m.

At the height of 7000 m the concentration maxima of particles with $d_p < 70$ nm were clearly pronounced only in the annual behavior of 2003 and 2005. In 2004, the concentration at this height varied insignificantly around $\sim 10^2$ cm^{-3} during the year.

Concentration of aerosol particles with $d_p < 70$ nm and $d_p > 70$ nm at the height of 3000 m reflect the dynamics of the boundary layer, i.e., the possibility that aerosol penetrates from bottom layers due to vertical mixing. Similar picture is to be observed at other heights in the atmospheric boundary layer.

3. Discussion

Return to the question on whether the summer minimum in the surface and boundary layers is due to the change of the mixing layer height only. For this, it is necessary to calculate the aerosol column density, the upper boundary of which is to be the mixing layer height. The heights of both the mixing and atmospheric boundary layers were determined with the use of meteorological parameters recorded with on-board instruments of the AN-30 "Optik-E" instrumented aircraft. At present the technique that uses virtual potential temperature Θ_v (Ref. 8) is commonly used for this purpose:

$$\Theta_v = T_v \left(\frac{p_0}{p_H}\right)^{R/c_p} = T(1 + 0.608q)\left(\frac{p_0}{p_H}\right)^{R/c_p}, \quad (1)$$

where p_0 is the ground-level pressure, p_H is the pressure at the height H, T_v is the virtual temperature, K, T is the ambient air temperature (K) at the height H;

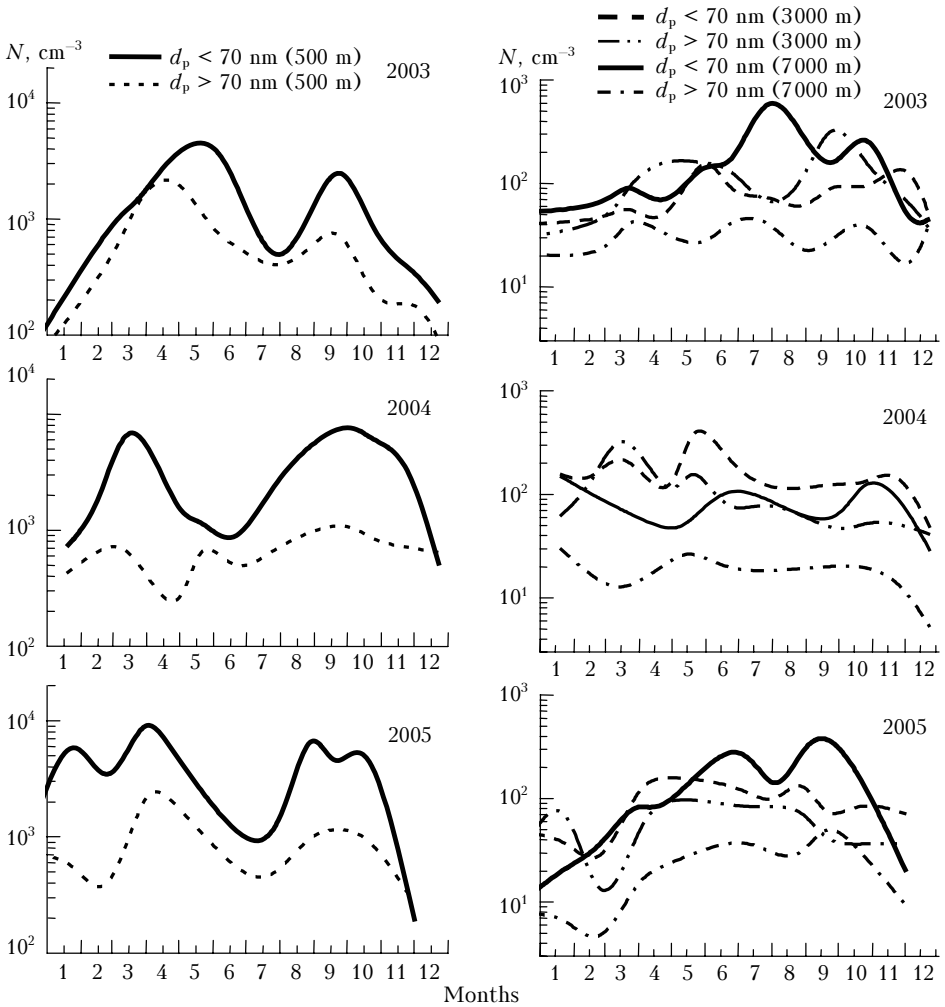

Fig. 2. Concentration of aerosol particles at different heights in the troposphere over the south of Western Siberia.

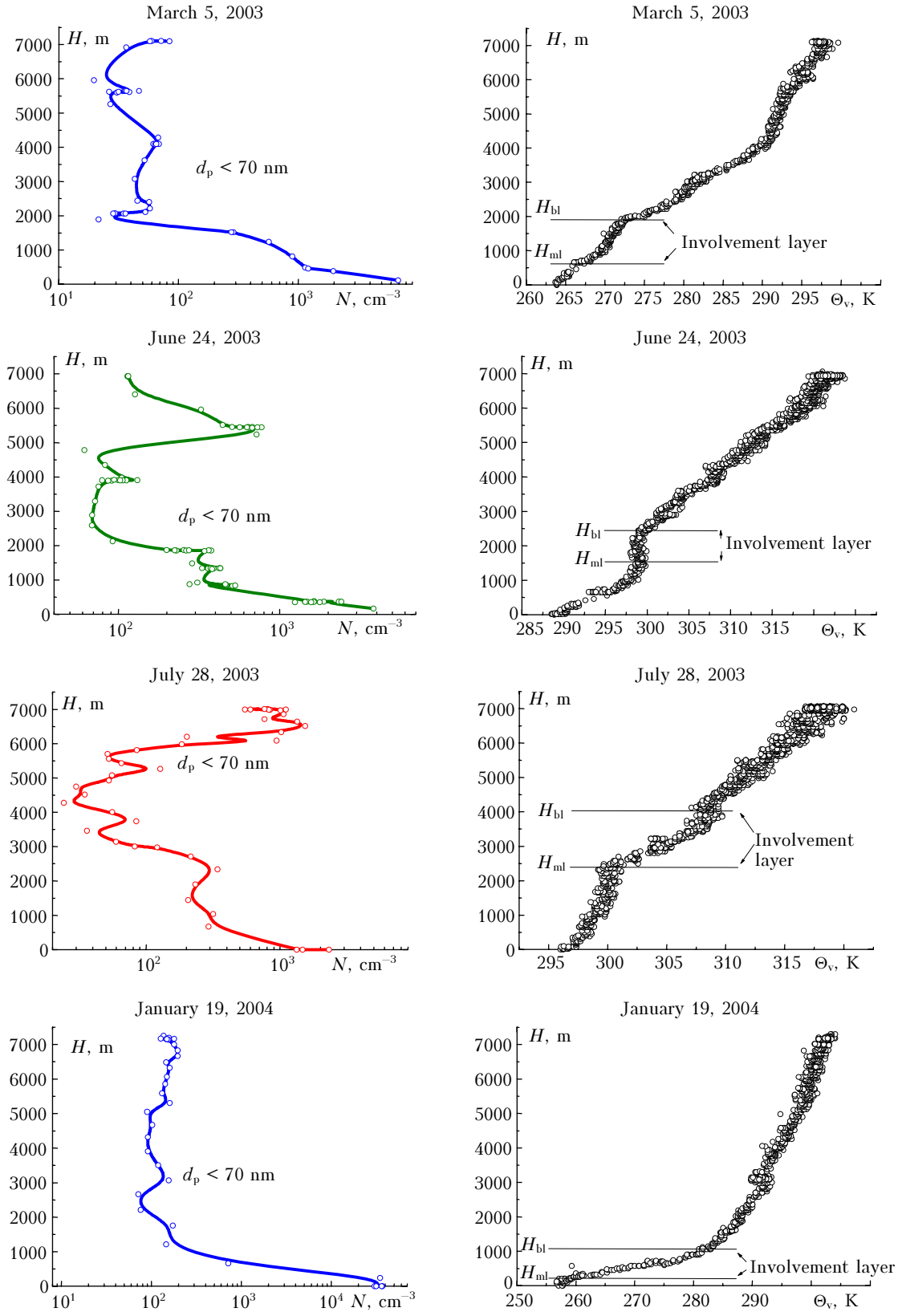

Fig. 3. Vertical profiles of aerosol particles concentration ($d_p < 70$ nm) and virtual potential temperature (H_{ml} is the height of mixing layer; H_{bl} is the height of boundary layer).

R is the specific gas constant of dry air (287.05 J/(kg·K)), c_p is the specific heat at constant pressure (1005 J/(kg·K)), q is the water vapor mass fraction (grams per 1 g of dry air) calculated by temperature and relative humidity:

$$q = 0.622\frac{rhE(T)}{p}, \qquad (2)$$

where rh is the relative humidity (0–1), $E(T)$ is the pressure of the saturated water vapor, p is the pressure.

Calculated results on the virtual potential temperature are shown in Fig. 3. The heights of mixing and boundary layers are determined by points of inflection in the vertical profile of virtual temperature. Figure 4 shows the seasonal dynamics of the mixing layer height determined from the profiles of Θ_v.

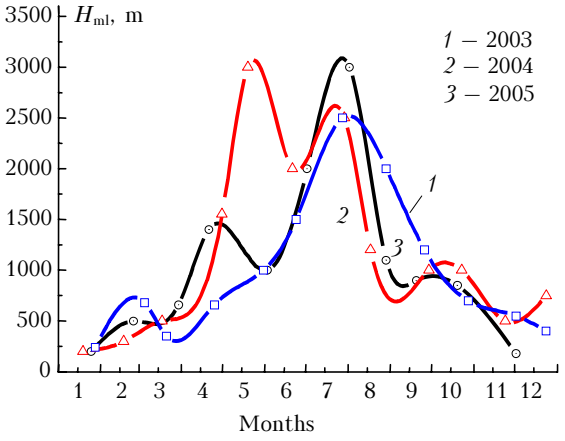

Fig. 4. Seasonal trend of the mixing layer height.

Now calculate the integral aerosol content in a vertical air column of 1 cm^2 cross section. Seasonal dynamics of the integral aerosol content in the mixing layer is shown in Fig. 5 which proves our suppositions concerning the summer maximum. However, there is a seasonal dependence in the annual behavior of aerosol content. As in the case with the number concentration, spring and fall maxima are pronounced here. This means that increasing vapor release of aerosol forming compounds takes place in these seasons.

If an additional VAFS emission in spring can be explained by evaporation at snow melting and vegetation activity of Siberian forests evolving photochemical active substances similar to ozone, the cause of the autumn maximum is not yet evident.

It is most probable that the secondary maximum, observed in fall, is caused by the reverse biological process of putrefaction. It is known,[4,5,9,10] for example, that the main sources of gaseous sulfur and nitric oxide compounds in the background land areas are microbial products due to the decay of plant residues. In support of this, a complex experiment on studying spatiotemporal variability of aerosol and aerosol precursor gases is to be carried out as well as physical-chemical characterization of VAFS.

Another peculiarity of the seasonal behavior of the integral aerosol content is the fact that its absolute minimum has been observed in December due to the minimum influx of solar radiation, required for photochemical transformations, and low moisture content.

Conclusion

The performed analysis of the seasonal dynamics of the fine aerosol fraction in the surface and atmospheric boundary layers has shown that the content of nanoparticles in the atmosphere is determined by the balance between the aerosol forming substances and the transport of the particles formed to higher atmospheric layers. Hence, the minimum in concentration of fine aerosol in the surface layer is observed in summer when the VAFS generation reaches its maximum. This concentration reversely depends on the height of the mixing layer.

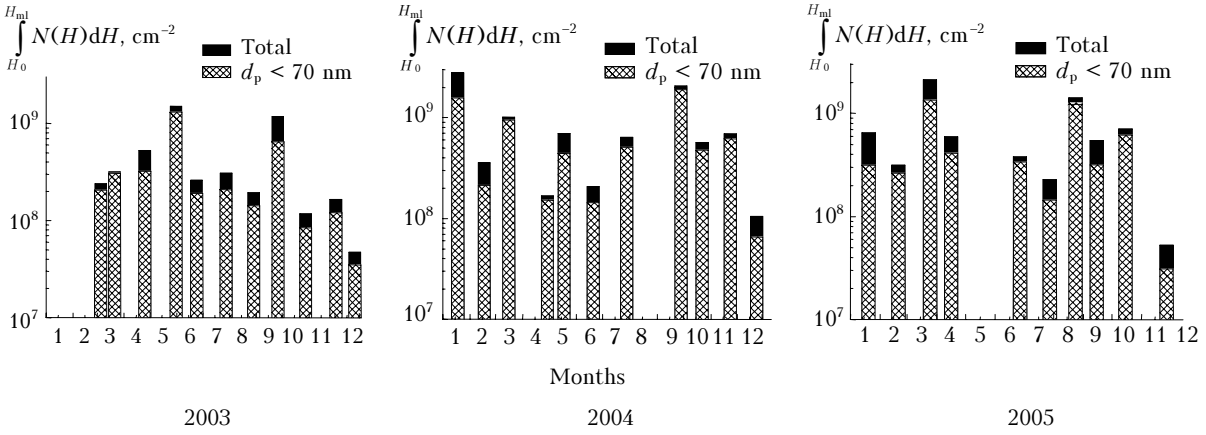

Fig. 5. Integral aerosol content in a vertical air column of 1 cm^2 cross section.

Acknowledgements

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