

## LIDAR OBSERVATIONS OF THE STRATOSPHERIC OZONE AND AEROSOL IN TOMSK (56°N, 85°E) FOLLOWING THE ERUPTION OF THE PINATUBO VOLCANO

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Received June 3, 1993*

*The vertical profiles of the stratospheric ozone and aerosol in the presence of thick aerosol formations of volcanic origin in the stratosphere measured at the high-altitude lidar sounding station in Tomsk are presented. The experimental data and the factors causing the vertical stratification of the observed parameters are analyzed. The role of the aerosol cycle in the ozone destruction and the significance of the dynamic processes in the formation of the vertical distribution of the ozone concentration are discussed.*

### INTRODUCTION

Many years observations of the stratospheric aerosol have shown that during a sufficiently long period following the volcanic eruptions the properties and characteristics of the stratospheric aerosol layer are mainly determined by the aerosol of volcanic origin. This allows one to conclude that strong volcanic eruptions emitting a huge amount of gas and dust into the atmosphere can cause the climate changes on the global scale.<sup>1-3</sup>

Volcanic eruptions disturb the physicochemical balance in the atmosphere, causing thus sporadic perturbations in its heat-radiative, chemical, and optical structure, especially in the upper troposphere and low stratosphere where the main transport of air masses and occurrence of the baric formations take place. It is also known that just in these regions there is the layer with maximum ozone content and where dense aerosol layers are formed.

The observation of volcanic emissions give a unique possibility of studying the effect of the stratospheric aerosol on the atmospheric structure and its interaction with other atmospheric constituents in more detail. However, it is not so easy to isolate the effect of the aerosol layers of the volcanic origin against the background of short-term variability of the atmospheric structure. Therefore, it is of interest to reveal the variability of the atmospheric parameters rather than temporal behavior of their mean values.<sup>4</sup>

In connection with recent developments of the lidar methods of studying the atmosphere their use in practice of routine lidar observations of the stratospheric aerosol gives a new effective means of acquiring a valuable information about aerosol layers of volcanic origin aiming at the creation of atmospheric models and numerical calculations.<sup>5,6</sup>

Lidar observations of the stratospheric aerosol at the high-altitude lidar sounding station in Tomsk have been performed systematically since 1986 (see Refs. 6 and 7). Observations conducted after a powerful eruption of the Pinatubo volcano occurred in Philippines in June, 1991 provided a unique possibility of studying the dynamics of the stratospheric aerosol of the volcanic origin and

transformations of its microphysical properties<sup>8</sup> and of determining the relation between the stratospheric aerosol and ozone, the observations of which were also performed at that time. This relation is of special interest and importance since the influence of aerosol on the ozone is uncertain because of its strong variability and a complicated interaction of the ozone with other stratospheric constituents. Besides, the presence of natural short-period variability of the content and distribution of the ozone in the atmosphere also results in difficulties in determining the effect of stratospheric aerosol on the ozone. Therefore an essential increase of the aerosol content in the stratosphere occurring after volcanic eruptions and resulting in an increased importance of the aerosol cycle of the ozone destruction allows one to study this process in more detail. However, the mechanism of interaction between the aerosol and the ozone is not yet clear at certain because the sulphuric acid, the drops of which mainly make up the volcanic aerosol, does not directly interact with the ozone. Note, that in Ref. 9 the possibility of direct destruction of the ozone molecules on the surface of the volcanic aerosol containing the aluminium oxide is also considered. Besides, indirect schemes involving heterogeneous losses of the hydroxyl molecules on the surface of the sulphuric acid aerosol are considered, such losses lead to enhancement of the nitric oxides content which, in turn, initiate the nitrogen cycle of the ozone destruction.<sup>5,10</sup> Thus, it follows from the above discussion that simultaneous measurements of the ozone and aerosol, which would allow one to obtain, in addition, the vertical distribution of these parameters, are of undoubt interest.

The measurements were made using lidars described in Refs. 11 and 12. The experimental error and the spatial resolution of aerosol measurements were less than 5% (at the height of 30 km and even less at lower heights) and 400 m, respectively. The same parameters in ozone sounding were 3% and 1 km at the height of 15 km, 7% and 1.5 km at the height of 20 km, and 15% and more than 2.5 km at the height of 25 km. The deterioration of the spatial resolution in these measurements is determined by the regularization algorithms used in the data processing.

### OPTICAL CHARACTERISTICS OF THE AEROSOL AT THE WAVELENGTH $\lambda = 532$ nm

Optical parameter of the atmosphere, which is most frequently measured at lidar sensing of the stratosphere, is the so-called scattering ratio  $R(H)$  (see Ref. 11)

$$R(H) = 1 + \beta_{\pi}^{\alpha}(H) / \beta_{\pi}^m(H), \quad (1)$$

where  $\beta_{\pi}^{\alpha}(H)$  and  $\beta_{\pi}^m(H)$  are the aerosol and molecular backscattering coefficients.

The lidar observations of the height and temporal transformations of this parameter conducted at the high-altitude lidar sounding station in Tomsk following the eruption of the Pinatubo volcano till March, 1992 were analyzed in Ref. 13. A portion of vertical profiles  $R$  obtained in Ref. 13 are depicted in Fig. 1a. Results of further investigations of the vertical stratification of the aerosol are shown in Fig. 1b. The profiles shown represent practically every month being chosen arbitrarily within a month.

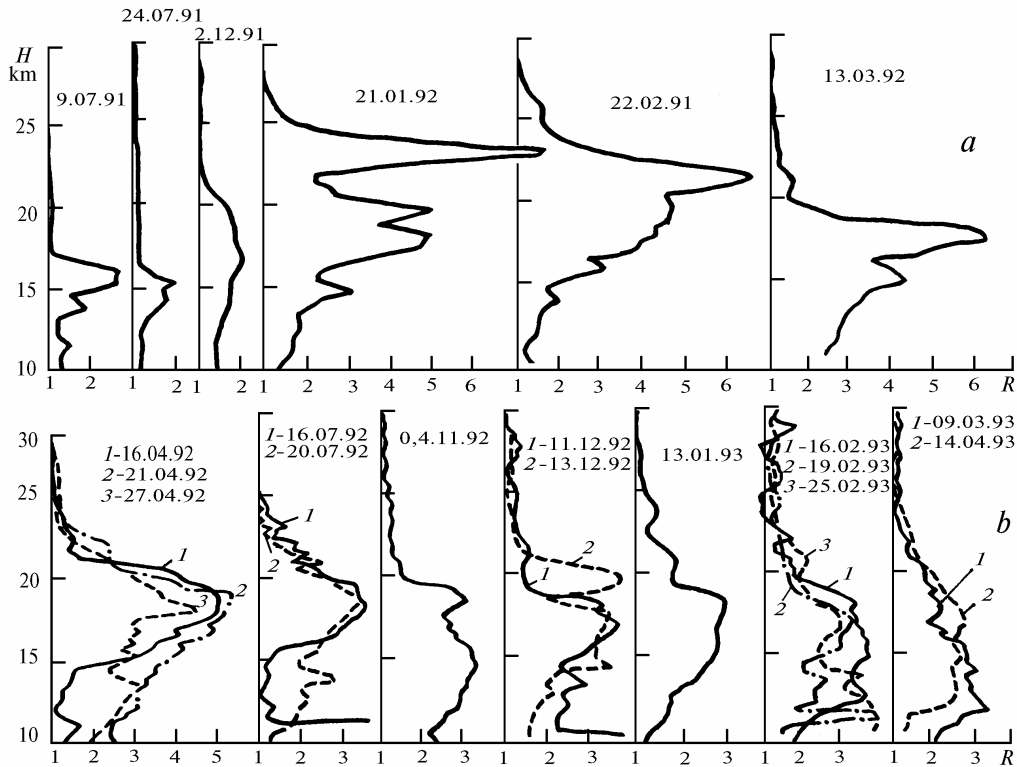


FIG. 1. Scattering ratio profiles at the wavelength  $\lambda = 532$  nm.

Common features in the altitude behavior dynamics of the scattering ratio for a given set of profiles are the descent of the maximum of  $R$  and its stabilization at the level of  $R = 3$ , whereas on January, 1992 the maximum values of  $R$  reached 8 (see Ref. 13). However, the scattering ratio  $R = 3$  is certainly higher than the scattering ratio ( $R = 1.15$ ) observed for the background aerosol content in this stratosphere.

When the spring, winter, and fall profiles of  $R$  are compared, no successive transformations are observed regardless of the time interval between the providing measurements (Fig. 1b). The profiles  $R$  show quite abrupt changes from night to night. Such a behavior can be indicative of the fact that the aerosol formations of the volcanic origin have nonuniform spatiotemporal structure even a long time after the eruption. The aerosol stratification over an observation point is obviously the result of such inhomogeneities motion. Therefore it can be stated that the stratospheric aerosol of volcanic origin is entrained, at least its major part, into the general motion of air mass.

The assumption of this kind agrees with the character of circulation processes at middle latitudes in the Northern Hemisphere. During winter in middle latitudes the eastward transfer and the high wind velocities increasing with the altitude are observed throughout the stratosphere. In spring a reconstruction of the thermobaric fields occurs, the wind velocity decreases and the meridional wind components with large variability in their directions become dominating. The main feature of the summer circulation of the stratosphere is the presence of so-called velopause. It appears as a result of changing in the wind direction from the western to the eastern (practically  $180^\circ$  change of direction), it is characterized by small wind velocities (some meters per second) and is localized at the height of 20 km. During the fall the reconstruction of circulation processes in the stratosphere also takes place in the direction that is quite opposite to the spring one.

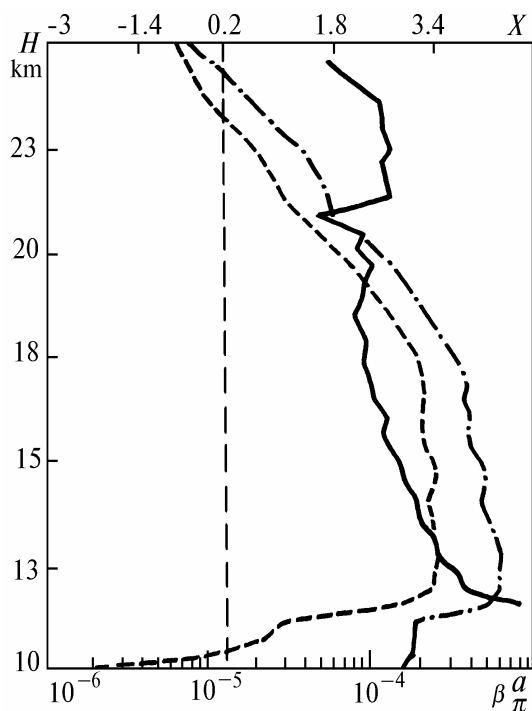


FIG. 2. Altitude behavior of the parameter  $X$  (solid line) and the aerosol backscattering coefficients at wavelengths  $\lambda = 532$  nm (dot-dashed line) and  $\lambda = 683$  nm (dashed line).

Returning to Fig. 1b once more, we would like to note that weak winds in the region of the velopause (17–22 km) well explain the stability of the July profile behaviors of the scattering ratio there, whereas stronger winds at lower altitude result in their noticeable diversity. However, during the period under consideration the variations of vertical stratification caused by the air mass transport were supplemented by the change of scattering properties of the volcanic aerosol. The latter circumstance can be caused by physicochemical processes resulting in aerosol microstructure modifications. Figure 2 shows the height behavior of the parameter  $X$  observed on April 14, 1993, obtained with the use of the wavelengths 532 nm (the second harmonics of Nd:YAG laser) and 683 nm (SRS conversion of  $\lambda = 532$  nm by  $H_2$ ). The physical meaning of this parameter and the formula for its determination were described in Ref. 14 in detail. It can be seen that  $X(H)$  decreases with increasing height practically up to 19 km. Such a behavior indicates that the number of aerosol particles optically active in the spectral region 532–683 nm increases with increasing the height. A comparison of  $X(H)$  depicted in Fig. 2 with that reported in Ref. 14 on April 27, 1992 shows that not only vertical stratification (Fig. 1b) has been changed but also the scattering properties of the volcanic aerosol. One more parameter, in addition to the scattering ratio, became recently widely used to characterize the aerosol

content of the stratosphere. It is the integrated backscattering coefficient  $\beta_\pi^\alpha$

$$\beta_\pi^\alpha = \int_{15 \text{ km}}^{30 \text{ km}} \beta_\pi^\alpha(H) dH. \quad (2)$$

Its temporal behavior during the whole period of functioning of the high-altitude sounding station in Tomsk is presented in Fig. 3. It is seen that till the summer of 1991 the values  $\beta_\pi^\alpha$  in the series of observations are close to each other with a pronounced season behavior and even small negative trend. Beginning from the summer of 1991 rather sharp increase in the values of  $\beta_\pi^\alpha$  is observed with their maximum in winter of 1992. This temporal behavior of  $\beta_\pi^\alpha$  reflects the process of penetration of the eruption products of the Pinatubo volcano into the middle latitudes that leads to the aerosol formation.

### STRATOSPHERIC OZONE AND VOLCANIC AEROSOL

Lidar observations of the stratospheric ozone performed in Tomsk since 1989 became especially interesting after the eruption of the Pinatubo volcano. The ozone observations aimed at investigation of the role of aerosol in the ozone destruction started from the very first observation of the eruptive cloud over Tomsk.<sup>13</sup> Some results of this study are presented in Fig. 4 in the form of the ozone concentration profiles, which were calculated using the methods described in Ref. 14, and simultaneously obtained profiles of the scattering ratio at  $\lambda = 353$  nm. Synoptic situation at that time (July, 1991) was described in Ref. 8 in detail. The depicted series of profiles begins with the profile on July 6, 1991, when the stratosphere over the Western Siberia was only slightly perturbed by the volcanic aerosol.<sup>8</sup> All the succeeding profiles of  $R$  at heights below 16 km have the  $R$  values essentially exceeding the background ones. In addition, some of these profiles show pronounced aerosol layers which correspond to the local minima in the ozone concentration profiles (Fig. 4). Such situations are clearly seen on July 9, 14, 20, and 24. However, these layers influence on the integral ozone content much weaker. Figure 5b, which presents the temporal behavior of the integral ozone content according to the lidar data, demonstrates a rather deep minimum corresponding to the presence of the aerosol layer only on July 9. As for other days, only insignificant variations are observed. In these cases the minima in the temporal behavior of the integral ozone content do not necessarily coincide with the presence of aerosol layers (Fig. 5b). Besides, it is seen that the increase of the altitude range of integration diminishes the amplitude of these variations. They are not observed at all in the temporal behavior of the total ozone content obtained using an M-124 ozonometer (Fig. 5a).

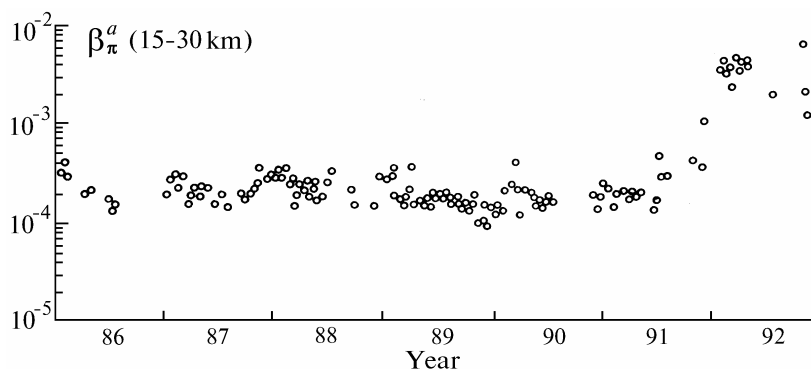


FIG. 3. Temporal behavior of the integral aerosol backscattering coefficient  $\beta$  at  $H = 15\text{--}30$  km.

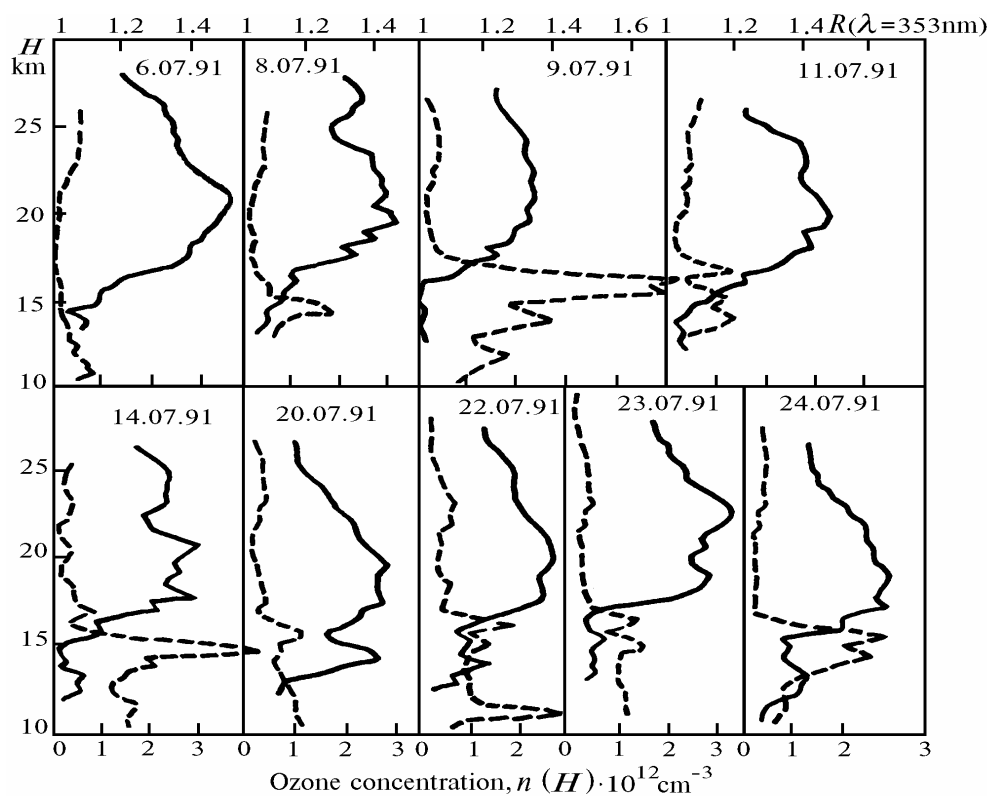


FIG. 4. Vertical ozone concentration profiles (solid line) and the scattering ratio profiles (dashed line) for some days.

Moreover, a comparison of the temporal dependencies of the lidar sounding data, i.e.,  $\beta_{\pi}^{\alpha}$  and data of satellite measurements of the total ozone content during the presence of the most powerful aerosol formation in the stratosphere (winter, 1992) demonstrates their total independence (Fig. 6). An explanation of this fact can be smoothing of local variations in the ozone and aerosol contents at different altitudes when integrating, as is seen in Fig. 6. Thus, the investigations of the role of the aerosol cycle in the ozone destruction based on the data on the integral characteristics of ozone and aerosol are less informative.

Moreover, the data presented in Fig. 6 were not synchronized that hinders adequate comparison of the

experimental data in the case of strong dynamics of the stratosphere over the Western Siberia in winter time. The role of stratospheric dynamics, i.e., the transport of air masses due to atmospheric circulation processes, in the formation and variability of the stratospheric ozone layer is illustrated by Fig. 7. This figure shows the ozone profiles measured on April, 1992, when the stratosphere contained dense aerosol formations (see Fig. 1b). Note that two ozone profiles are presented for each night, the time required to measure one profile was approximately 20 min, and the interval between the ending of one measurement and the beginning of the next one was about 30 min. Their identical behaviors (of two profiles for one night) within the limits of experimental error are indicative of the representativity of the obtained data.

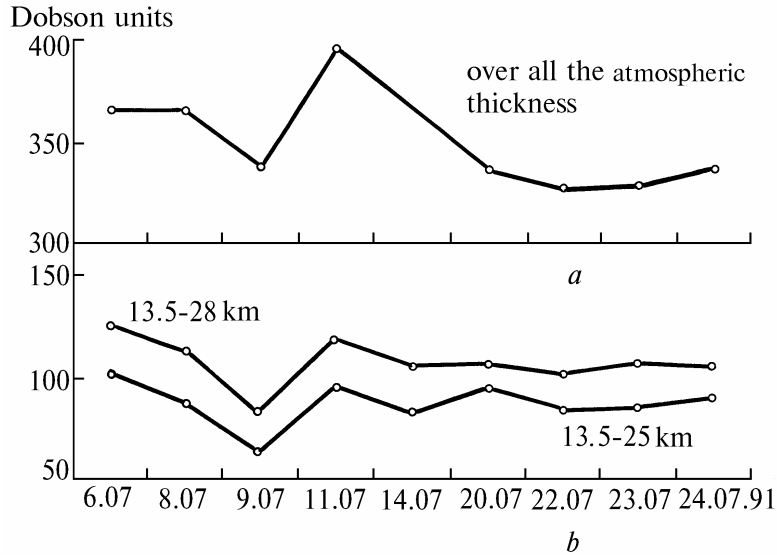


FIG. 5. Temporal behavior of the total ozone content (in Dobson units).

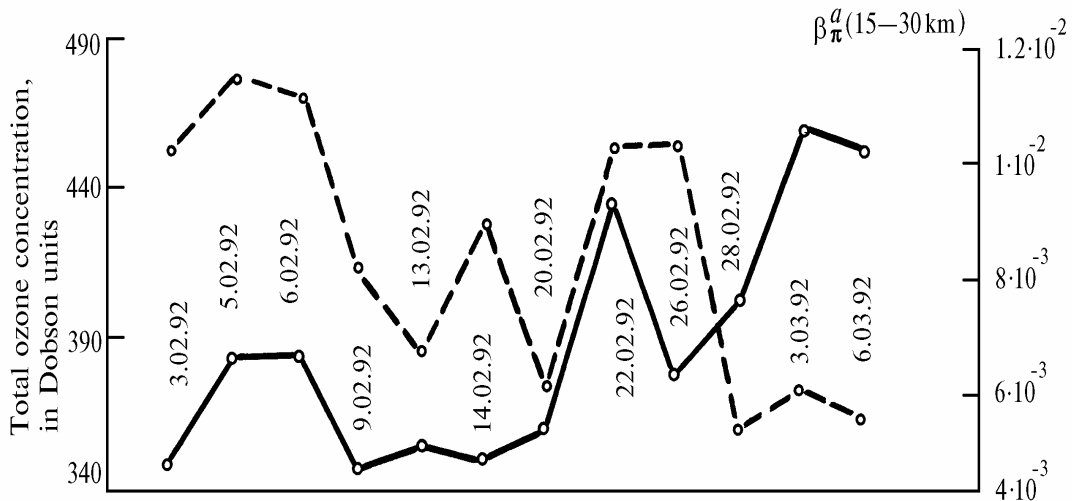


FIG. 6. Temporal behavior of the integral aerosol backscattering coefficient (lidar data, dashed line) and the total ozone content (satellite data, solid line).

The vertical profiles of the ozone shown in Fig. 7a allow one to note that on April 21, 1992 at the altitudes of maximum values of the scattering ratio (15–20 km, see Fig. 1b) the observed ozone concentration was less than the model one. On the other hand, the ozone concentration at these altitudes obtained from lidar data on April 27, 1982 essentially exceeds the model values. Analysis of the atmospheric situation concerning the aerosol component (see Fig. 1b) shows that the scattering ratio on 27.04.92 is smaller compared to that observed on 21.04.92. However, the observed decrease of  $R$ , most likely due to a decrease of the aerosol concentration, could not cause an increase in the ozone concentration shown in Fig. 7b. The occurrence of such a vertical ozone distribution with maximum concentrations at heights being not characteristic of the Western Siberia in spring

time was the invasion of the Arctic air masses into the middle latitudes of the Northern Hemisphere. The Arctic vertical ozone distribution is characterized by larger maximum ozone concentrations as compared with that in mid-latitudes and their localization is at heights of 15–16 km.

Figure 8 shows some results of sounding of the stratospheric ozone obtained on April, 1993. These vertical ozone profiles also show smaller values of the ozone concentrations as compared with the model ones in the region where the maximum scattering ratio occurs. The difference between the model and measured profiles at heights above 20 km can be explained by both the error in empirical profiles and physical processes taking place in the ozonosphere at the moment.

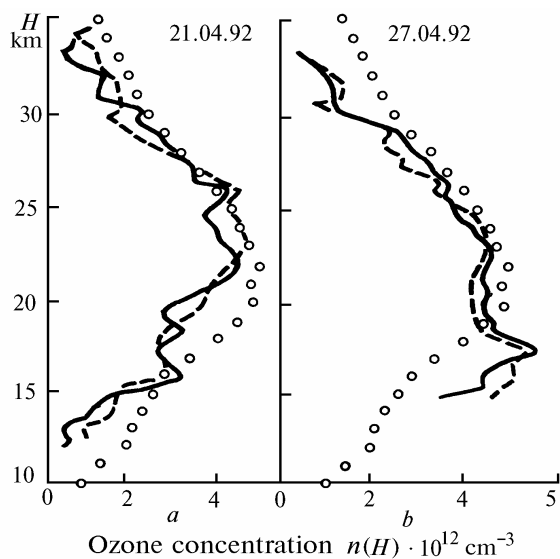


FIG. 7. Vertical profiles of the ozone concentration on April, 1992 (solid and dashed lines) and that according to the Kruger model (circles).

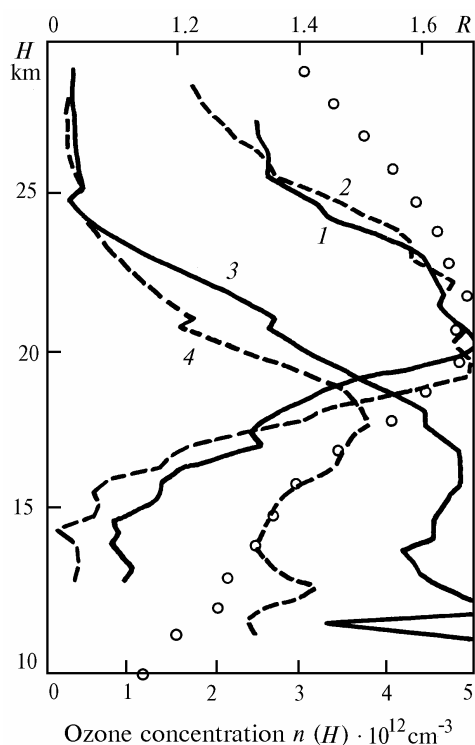


FIG. 8. Vertical profiles of the ozone concentration (12.04.1993, curve 1 and 13.04.1993, curve 2), scattering ratio (12.04.93, curve 3 and 13.04.93, curve 4) and that according to the Kruger model (circles).

## CONCLUSION

Thus, the eruption of the Pinatubo volcano resulted in formation of dense aerosol layers in the stratosphere, the backscattering coefficient of which was several times as large as that of molecular scattering. Two years after the eruption the aerosol content in the stratosphere is far above its background. In the presence of dense aerosol formations of volcanic origin in the stratosphere, correlation between local minima of the ozone profiles and maxima in the scattering ratio profiles is observed. However, the important role in the formation of the ozonosphere on regional scales as well as in the redistribution of stratospheric ozone over the latitudes belongs to the atmospheric circulation processes.

The research has been supported by the Russian Fundamental Science Foundation (Project 93-05-9383).

The authors acknowledge Dr. G.M. Kruchenitskii for the data on the total ozone content, M.V. Grishaev, A.V. Nevzorov, and E.V. Sharabarin for the help in organization and performance of the experiments.

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