LIDAR AND SPECTROPHOTOMETRIC OBSERVATIONS OVER THE OZONE LAYER IN THE STRATOSPHERE OVER TOMSK

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This paper presents some results of long-term complex observations of the stratospheric ozone layer over Tomsk (south–west of Siberia). The observations were performed at the Institute of Atmospheric Optics, Siberian Branch of the Russian Academy of Sciences. Variability of stratospheric ozone is considered in relation to variability of the stratospheric aerosol, nitrogen dioxide, temperature, and synoptic processes. Attention is paid to the problem of lidar data calibration.

INTRODUCTION

Regular lidar observations of the stratosphere over Tomsk were started at the Institute of Atmospheric Optics (IAO), Siberian Branch of the Russian Academy of Sciences, in 1986, when the first stationary lidar built around the receiving telescope with the mirror 1 m in diameter was put into operation.[1] First, only the stratification of stratospheric aerosol was the subject of sensing.[2] Later on, since 1988, the sensing of vertical distribution of stratospheric ozone has been additionally conducted.[3] In 1990 the lidar station with the receiving telescope with the mirror of 2.2 m in diameter was put into operation at the IAO,[4] which was then joined with the first large-size lidar into the Siberian Lidar Station (SLS) at the IAO. In 1994 at the SLS the synchronous measurements were conducted together with the spaceborne LITE lidar aimed at making intercomparison of results.[5] In 1995 the SLS obtained first results on laser sensing of the tropospheric ozone.[6] Regular spectrophotometeric observations over the total ozone content (TOC) have been conducted at the SLS since 1993 with the use of the standard calibrated M-124 ozonometer. Since 1995 we are also conducting the twilight (morning and evening) observations of the NO_2 total content and vertical distributions.[7]

At present the SLS conducts, at cloudless nights, the complex lidar sensing of the ozone and aerosol vertical distribution and temperature in the stratosphere, operating in the regime of regular measurements, and the observations of TOC and NO_2 in the regime of practically routine observations. In the near future we plan to have additional channels for measuring TOC and NO_2 using a lunar spectrophotometer and the ozone vertical distribution using a twilight spectrophotometer.

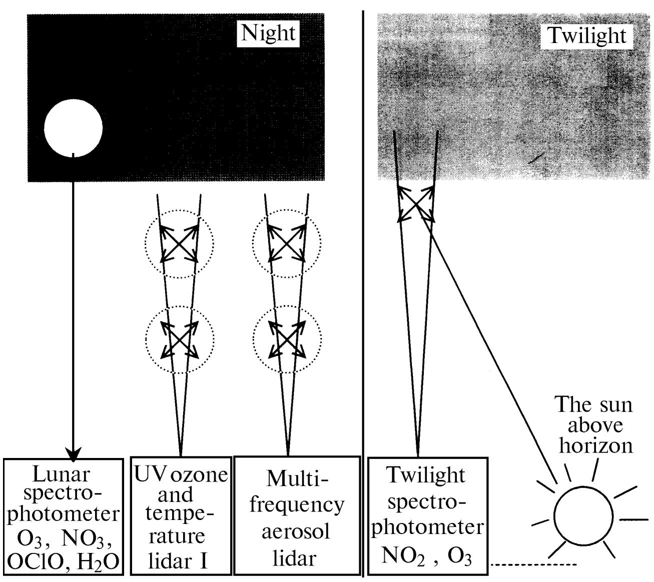


FIG. 1. The SLS ozone measuring complex: τ_a is the aerosol optical depth, T is the temperature, P is the pressure, V_V is the wind velocity, and V_d is the wind direction.

Figure 1 schematically shows the SLS complex for ozone measurements. This complex makes the instrumental basis for the initiative SATOR Program at IAO to study the stratospheric and tropospheric ozone.

SOME RESULTS OF LIDAR DATA CALIBRATION

The specifications of the SLS and methods used for sensing stratospheric constituents and parameters are described in Refs. 8–10. The peculiar feature of the SLS is its capability of simultaneously sensing ozone, temperature, and aerosol, including its microstructure, with the use of optical and spectral separation of lidar returns both with a single receiving telescope and several telescopes at the same time.

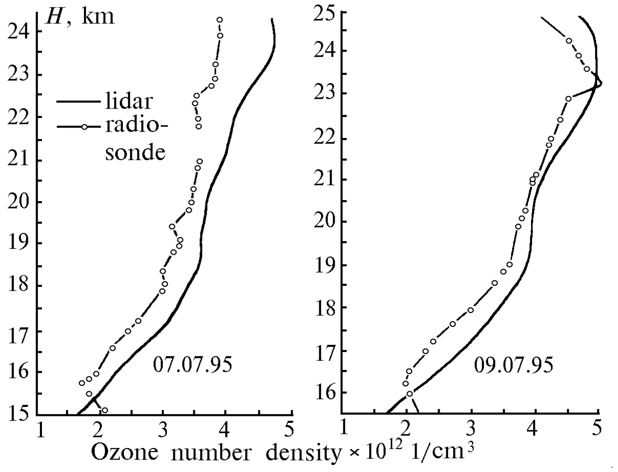


FIG. 2. Temperature profiles obtained with the lidar and a baloon.

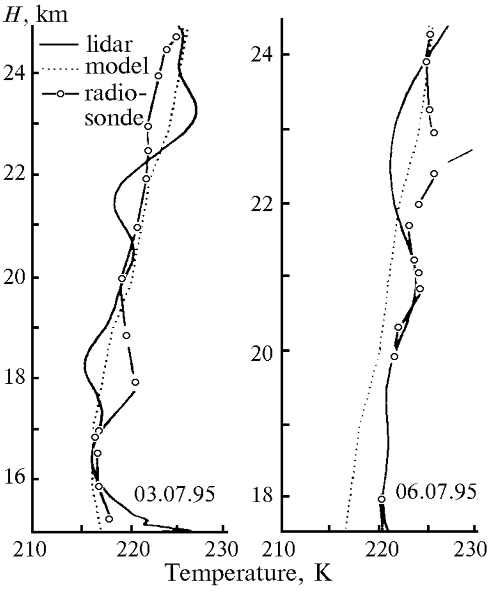


FIG. 3. Ozone and temperature profiles.

Optical calibration of all lidar signals is done using signals from altitudes no less than 30 km, where, with no aerosols, they are formed only due to Rayleigh scattering, whose cross section is proportional to the density of atmospheric air at these altitudes.[11]

Calibration of temperature and ozone profiles measured with the lidar by the results of direct measurements of temperature and ozone concentration with the use of baloons was done in July 1995 within the framework of SATOR-95 measuring campaign. Altogether nine baloons were launched. We succeeded in comparing in three cases. The results of the comparison are shown in Figs. 2 and 3. One can see good agreement between the lidar and baloon data.

Close agreement is also observed when comparing the integrated values of stratospheric ozone concentration obtained with the lidar with TOC values measured using a calibrated M-124 device certified at Voeikov Main Geophysical Observatory in 1994.

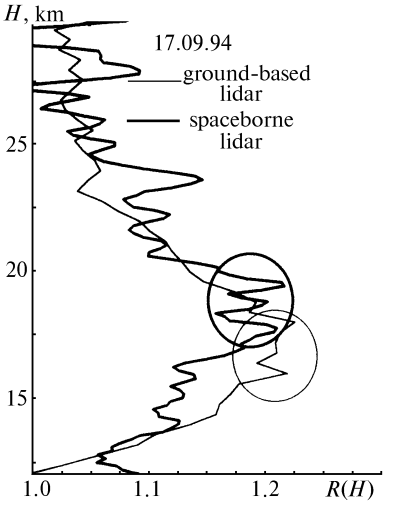


FIG. 4. Scattering ratio profiles obtained in September 1994 with the ground-based and spaceborne lidars.

A peculiar kind of intercalibration was done for the lidar aerosol channel during the correlation measurements synchronously with the LITE spaceborne lidar in September 1994 (Fig. 4). The results of intercoparison of laser sensing data on stratospheric aerosol[5] demonstrate good correlation. In the marked regions, practically the same aerosol stratification was measured, and the height difference for these regions agrees well with the difference in tropopause heights over Tomsk and Altay, above which the Shuttle flew that time.

STRATOSPHERIC OZONE AND TEMPERATURE

The behavior of stratospheric ozone and temperature is known to have a positive correlation. This is connected with the fact that heating of the stratosphere directly depends on the concentration of ozone, the main absorber of UV solar radiation in the middle atmosphere, as well as on the influence of common dynamical factors, like vertical flows, on the vertical distribution of temperature and ozone. In addition, the higher is the temperature, the higher are the rates of ozone-producing chemical reactions.

Interesting situation, characterizing the correlation between the vertical distribution of ozone and temperature profile, was observed in February 1996. Figure 5 shows the night profiles of ozone and temperature obtained from lidar sensing data. Thus, in early February (05.02.96) moderate ozone content in the stratosphere corresponds to the vertical temperature profile close to the model one. Sudden appearance of a large amount of ozone in the 15–25 km layer detected on February 12 was followed by the temperature rise by 10 K in this layer. Then, on 20.02.96, the decrease in ozone content in the 15–25 km altitude range took place simultaneously with the temperature decrease at these altitudes. The lidar temperature profile in this case was very close to the model one. Very high ozone content in the extended 15–30 km layer was noticed on February 22. Significant increase in the stratospheric ozone content was observed against the background of sharp temperature increase in the 10–30 km layer mentioned. On February 28 a decrease in the ozone concentration and at the temperature profile coming back to the model one took place.

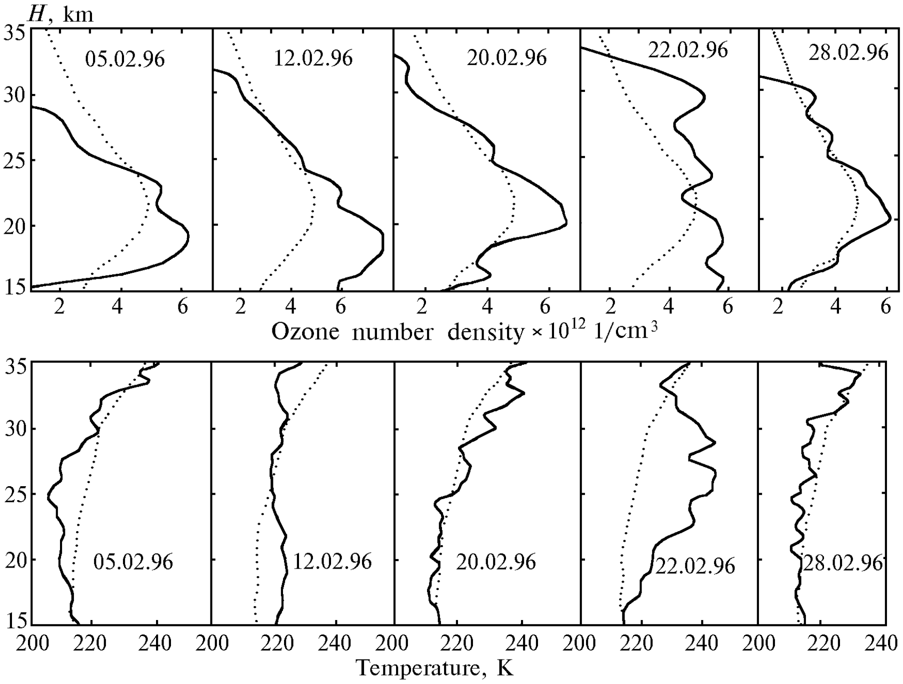


FIG. 5. Dynamics of the ozone and temperature profiles observed in February 1996.

Sufficiently strong positive correlation between the temperature and ozone concentration vertical profiles (see Fig. 5) is indicative of a significant role of the dynamical processes in the middle atmosphere.

Oscillations of the circumpolar cyclonic vortex about its mean (statistical average) position, that result in phase shift (oscillations) of stationary planetary waves, govern the intensity of vertical motions and rearrangement of thermobaric fields in the lower atmosphere. Consequently, combinations of different dynamic conditions in the lower and middle atmosphere will determine different shapes of vertical profiles of temperature and ozone.

The analysis of synoptic data demonstrates that high values of ozone concentration in the middle stratosphere and total ozone content, on the whole, on February 12 and 22, to which the higher temperature corresponded, were caused by the influence of the leading part of the height pressure trough belonging to the circumpolar cyclone. The circulation had a well pronounced zonality. Typical of such a situation are descending motions of cold air at the cyclonic side from the height frontal surface. Then, as a result of lowering, the adiabatic increase in temperature and ozone inflow from above occur.

The situations taking place on February 12 and 22 differ from each other due to somewhat different position of the circumpolar vortex, which caused more intense descending motions in higher layers and, correspondingly, increase in temperature and ozone concentration. However, in the lower stratosphere similar position of vortex caused weak ascending motions, while in the stratosphere it resulted in descending motions with formation of a warm region of high pressure.

Vertical distribution of temperature and ozone on February 5, 20, and 28 (especially on 20 and 28) is characteristic of the middle position of the circumpolar vortex. Lower values of temperature and ozone concentration in the middle stratosphere on February 5 were due to mostly ascending motions in this layer, while in the troposphere in these days a high warm anticyclone was observed.

STRATOSPHERIC OZONE AND MT. PINATUBO

Very strong eruption of Mt. Pinatubo at the Philippines occurred, as known, on June 15, 1991. Already two weeks after that (June 29) first traces of eruptive clouds appeared in the stratosphere over Tomsk at altitudes of 13–16 km, which reached their maximum intensity by July 9 (Fig. 6).

At that time the behavior of vertical profiles of stratospheric ozone was characterized by a pronounced negative correlation with aerosol profiles. This also show seen in the behavior of the total ozone content which dropped by almost 40% during three days (from 7 to 9 of July).

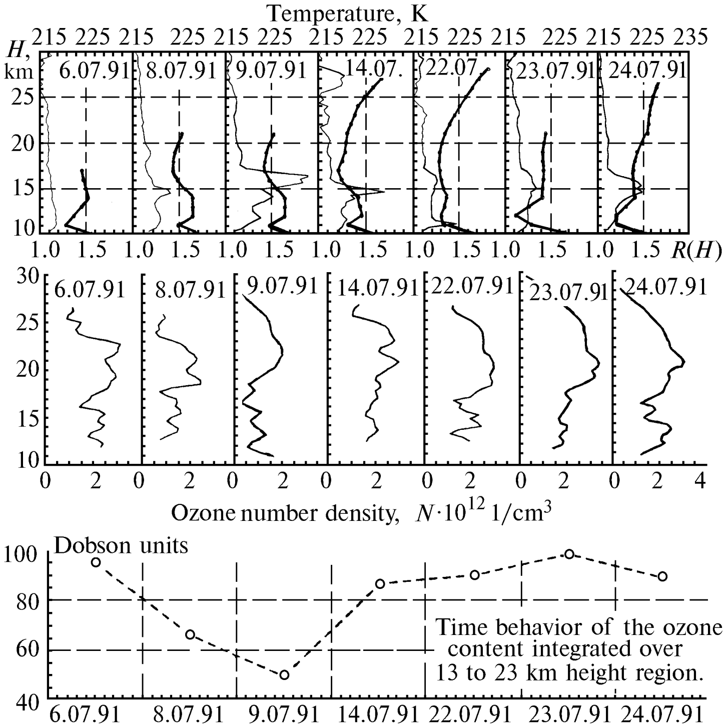


FIG. 6. Total ozone content in the 13–23 km *altitude range and the vertical profiles of ozone, scattering ratio, and temperature in July 1991 in the early period of appearance of the products of Mt. Pinatubo eruption over Tomsk.*

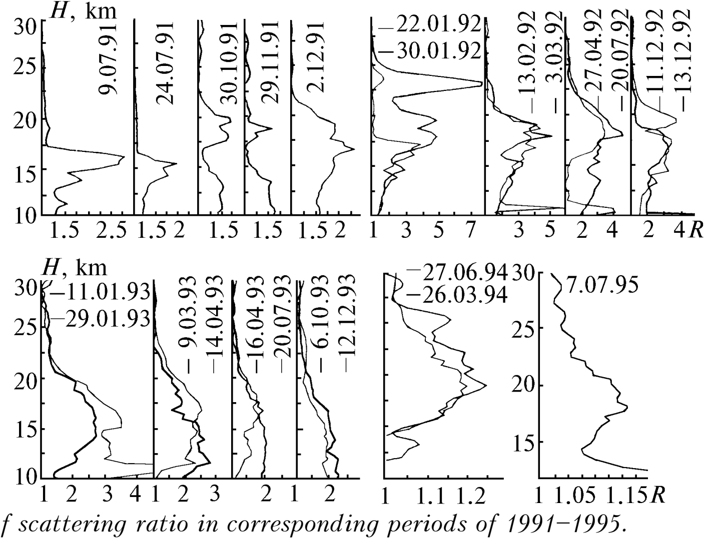


FIG. 7. Typical profiles of scattering ratio in corresponding periods of 1991–1995.

Such a behavior of stratospheric ozone can be explained by both heterogeneous reactions of ozone with aerosol constituents and homogeneous reactions with gases emitted during the eruption, for example, hydrogen (H_2). The latter mechanism will be described in more detail below, in Section «Stratospheric ozone and stratospheric clouds.»

Figure 7 shows some episodes with most typical profiles of aerosol stratification from October 1991 to July 1995. As one can see, maximal scattering ratios $R(H)$ were recorded in later January–February 1992. Minimal, as compared to the analogous period of the preceeding year, ozone concentrations in the stratosphere also were observed in the same period (Fig. 8). «Eating away» of ozone in the lower part of the profile is the unique manifestation of «the ozone depression». The presence of coincident maxima in the profiles of R and ozone at an altitude of 21–22 km is indicative of the meridional transpot, at these altitudes, of tropical air masses into mid-latitudes. On the whole, both these facts are responsible for formation, over the territory of Russia, of migrating structures similar in their characteristics to ozone holes (Fig. 9).

In spring of 1992 $R(H)$ remained high. However, the behavior of ozone and aerosol vertical profiles became practically independent, although the influence of aerosol overburden of the stratosphere on the formation of vertical distribution of stratospheric ozone, especially in the lower atmosphere, was seen even in winter–spring of 1993. The aerosol particle size spectrum in July 1991, being compared to that in April 1992, demonstrates a significant enlargement of particles (Fig. 10). On the whole, this was found to lead to an increase in the effective aerosol surface, with which the ozone molecules could interact, and consequently to an increase in the activity of heterogeneous reactions.

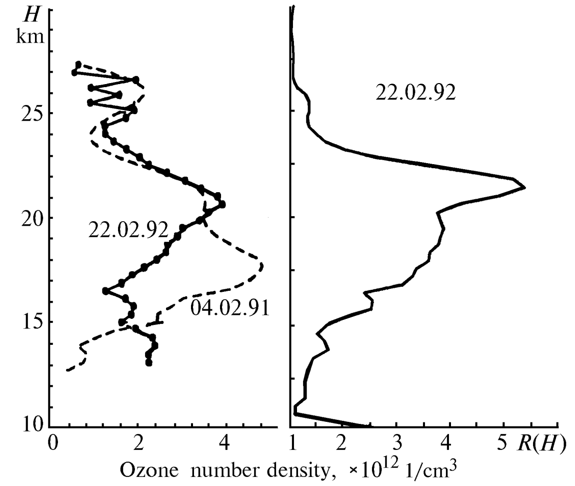


FIG. 8. Vertical profiles of ozone and scattering radio in February 1992 and the typical ozone profiles for February 1991.

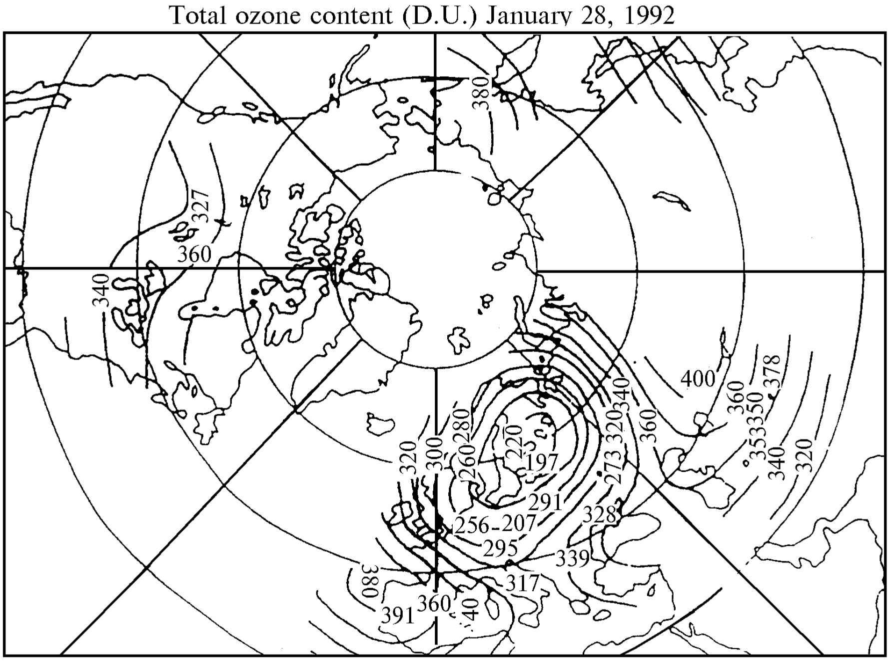


FIG. 9. The field of stratospheric ozone illustrating the formation of the type of ozone hole.

In general, the aerosol overburden of the stratosphere by eruptive aerosol has been observed by summer of 1995, during five years after the Pinatubo eruption. The behavior of the total ozone content in that period was characterized by the abnormal negative trend (Fig. 11), which is not directly connected with the stratospheric aerosol, because, as was mentioned above, the aerosol–ozone interactions became insignificant as early as in 1992. Thus, another factor should affect the TOC abnormal trend. Postvolcanic depression of NO_2

in the stratosphere can be considered as one of such factors (Fig. 12). The positive trend of the total content of NO_2 in the stratosphere in 1992 to 1995 was found to lead to strengthening of ozone cycle with «odd nitrogen NO_x», and consequently to an increase in TOC. When the NO_2 total content came to its standard level, values of TOC in the stratosphere also became normal.

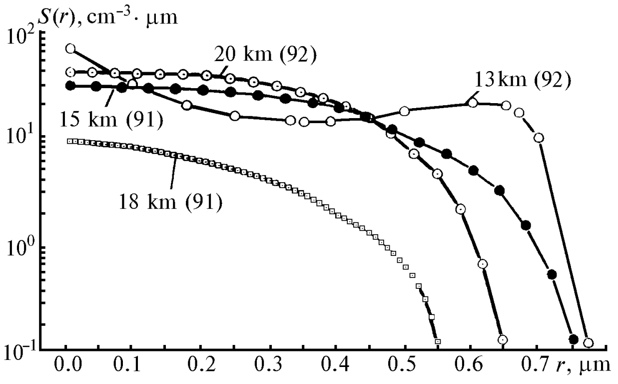


FIG. 10. Size spectra of aerosol particles of volcanic origin in July of 1991 and 1992.

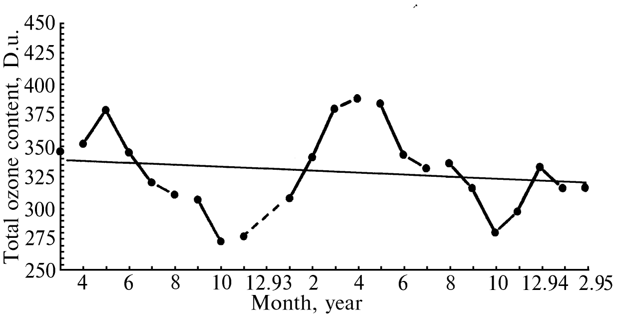


FIG. 11. Time behavior of monthly average TOC over Tomsk for the period from March 1993 to February 1995. Straight line is for linear trend.

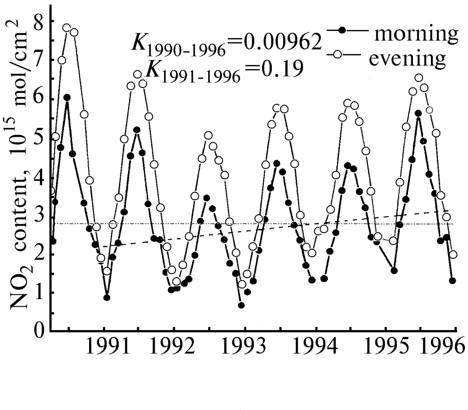


FIG. 12. Time behavior of the nitrogen dioxide total content.

One more factor, governing the TOC behavior during the postvolcanic period, is the temperature regime in the stratosphere that has changed during this period, and, as a consequence, circulation regime, in particular, strengthening of meridional component and invasion of tropical air masses into mid-latitudes in winter period. On the other hand, the TOC behavior for the entire period of observations well correlated with the variability of the baric atmospheric formations, while its distribution over years well agrees with the quasi-two-year cycle (Fig. 13).

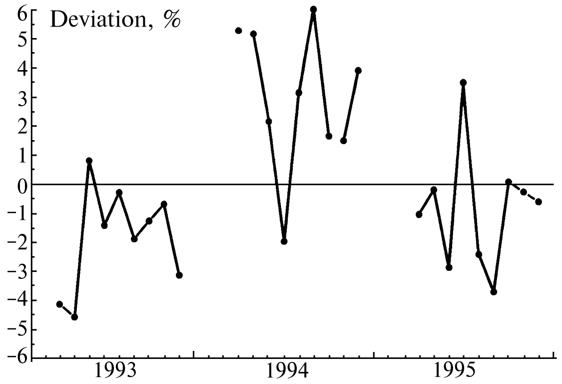
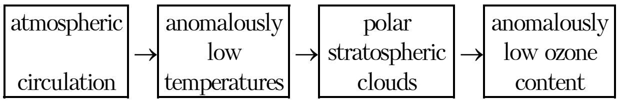


FIG. 13. Deviation of the monthly average TOC (March-November) from the average values for Tomsk in 1993–1995.

STRATOSPHERIC OZONE AND STRATOSPHERIC CLOUDS

The processes of the ozone layer destruction and their connection with the stratospheric clouds were first considered in the models of the ozone hole formation over the Antarctic.[12] According to this scheme, that has found a wide use now, the anarctic air masses locked up inside the circumpolar vortex underwent an intense cooling. When stratospheric temperature drops below −70° C, polar stratospheric clouds (PSCs) are formed. At low temperatures, at the surface of ice crystals the bonds of molecule NO_2, reservior of ClO, break thus releasing chlorine which then actively participates in the processes of catalytic ozone destruction. Thus, the cause–and–effect relationship of the processes can be presented by the following scheme (*a*):



A fresh original view of the correlation between the processes of ozone layer destruction and PSCs formation was put forward by Syvorotkin in Ref. 13. According to his scheme, due to degasation of the Eath the lightest gaseous fractions, hydrogen H_2 and methane CH_4, may be emitted into the stratosphere. In the stratosphere, hydrogen is oxidized by ozone producing water vapor and oxygen:

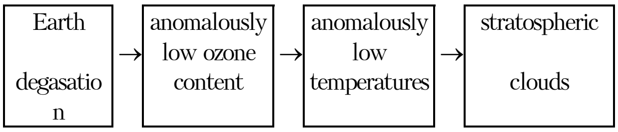
$$H_2 + O_3 \rightarrow H_2O + O_2. \qquad (1)$$

The relation (1) thus is indicative of the processes of ozone destruction and supersaturation of stratospheric layers with water vapor, that can stimulate the formation of stratospheric clouds. As to the methane emission into the stratosphere, according to Syvorotkin it should lead to blocking of the chlorine cycle of catalytic ozone destruction because of the reaction

$$CH_4 + Cl \rightarrow CH_3 + HCl. \qquad (2)$$

producing the molecule–reservoir of hydrogen chloride.

One important factor universally accompanying stratospheric clouds is absent in the scheme by Syvorotkin, namely, anomalously low temperatures. Although it should be a logic consequence of the reaction (1). Naturally, heating of the stratosphere is directly related to the ozone content, because ozone is the main absorber of solar radiation in the stratosphere. Consequently, the ozone layer destruction should result in stratosphere cooling down. This, in turn, under conditions of supersaturated water vapor according to Eq. (1) should stimulate the processes of crystal formation. Taking into account the above-said, we can construct the following cause–and–effect relationship (*b*):



When comparing schemes (*a*) and (*b*) it is seen that if one observes the stratosphere in the period of ozone anomalies, three main factors should be observed: low ozone concentration, low temperature, and stratospheric clouds. However, different character of the cause–and–effect relationships is responsible for different temporal sequence of events. Just this moment can allow the schemes (a) and (b) to be experimentally tested.

During regular lidar observations of the stratosphere over Tomsk at the SLS of the Institute of Atmospheric Optics since 1986, as rare episodes the abnormal increase of the scattering ratio *R* (the ratio of the total atmospheric backscatter to the molecular backscatter) was detected that disappeared some days later. Thus, for example, Fig. 14 shows vertical profiles of *R*, recorded at wavelength of 1064 nm in January 1995. The maximal values of *R* recorded that time were far higher than that on other days of the same year. Figure 15 demonstrates the results of our observations of the total ozone content, with an M-124 spectrophotometer, and temperature in the stratosphere. One can see from the figure that the situation in the stratosphere evolved in the following way. On January 21 the minimal TOC values were observed, then TOC became to progressively increase. Minimal temperature in the stratosphere was recorded a little bit later, on January 24. At the same time the stratospheric clouds were observed. As seen, such an evolution corresponds more closely to the scheme (*b*).

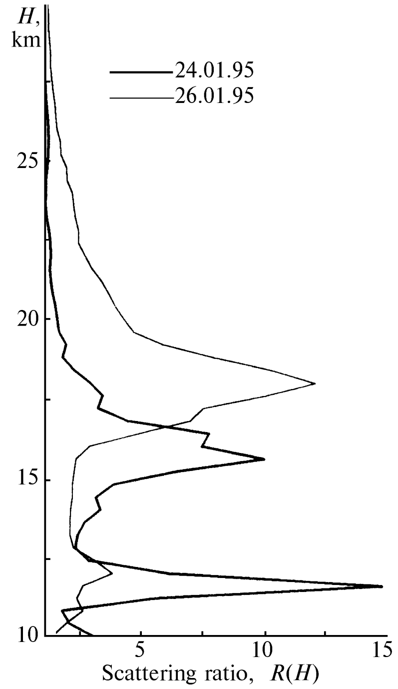


FIG. 14. Vertical profiles of scattering ratio at wavelength λ = 1064 nm *in January 1995.*

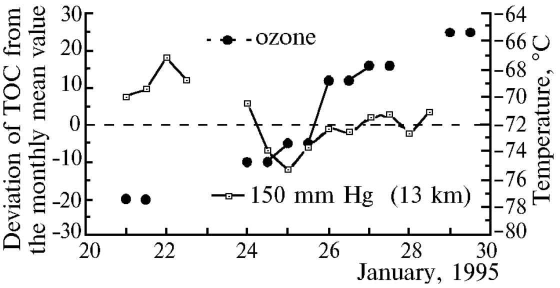


FIG. 15. Time behavior of the total ozone content and temperature at the corresponding millibar surface (baloon data) in January 1995.

Certainly, it is incorrect to draw a conclusion based on a single case. However, these results set us considering more closely the data series on lidar sensing of ozone and aerosol we obtained, in particular, in July 1991, during the first month after Mt. Pinatubo eruption. Well pronounced negative correlation between the ozone and aerosol profiles was then observed in the lower stratosphere. It may be a consequence of the scheme (*b*). At least, in this case the minimal temperatures also were recorded with a time lag with respect to the minimal values of the total ozone content in the stratosphere.

STRATOSPHERIC OZONE SITUATIONS WITH MINIMAL CONTRIBUTION OF DYNAMICAL FACTORS

As known, the ozone variability is due to superposition of two circumstances: atmospheric chemistry and dynamics, and the contributions from them can hardly be separated. Since the anthropogenic factor in the processes of ozone layer destruction is related to the chemistry of the stratosphere (for example, the chlorine cycle of catalytic ozone destruction stimulated by freons), then it can be studied most adequately when the contribution from the dynamic processes is minimal. Such a situation for mid-latitudes is usually characterized by a stable temperature stratification with no meridional component in the circulation of the stratosphere. One more criterion of similar situation can be the analysis of the relative behavior of stratospheric ozone and the components of its nitrogen cycle, in particular, NO_2. Since in dark time of a day NO_2 quickly reacts with O_3:

$$NO_2 + O_3 \rightarrow NO_3 + O_2$$

with the following reactions producing N_2O_5, then one can expect that under conditions of stable stratosphere well pronounced negative correlation should be observed in the behavior of NO_2 and O_3. Figure 16 shows the time variability of the night ozone content and the evening NO_2 content in the 25–30 and 25–35 km stratospheric layers, respectively, obtained using the lidar method and the twilight specrophotometric method. One can separate here at least one interval for February 24 (55th day) to March 13 (73rd day), 1996, when the opposite behavior of O_3 and NO_2 is observed. On other days their behavior is uncertain. The results of synoptic analysis indicate that in this time interval the stratosphere was characterized by a stable zonal transport of air masses. Thus, the complex analysis of data of lidar and spectrophotometric observations at the same point allows one to pictorially separate out the sections with minimal contribution of dynamic factors.

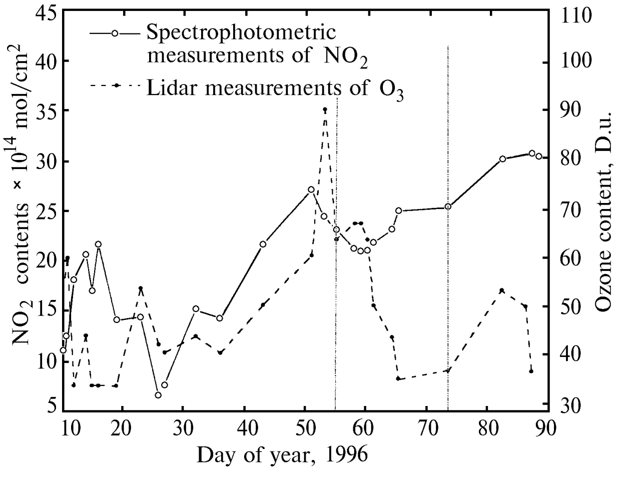


FIG. 16. Time behavior of nitrogen dioxide and ozone (from lidar data) in 1996.

CONCLUSION

Many-year complex observations of the ozone layer over Tomsk at the SLS of the Institute of Atmospheric Optics Sb RAS have shown that:

– Anomalous behavior of the stratospheric ozone held during five years after the Mt. Pinatubo eruption. Therefore it is practically impossible to draw some conclusions about tendencies manifesting themselves in the ozone layer during that period. This is apparently also true for the problem of detection of the numerous ozone holes in spring and winter of 1995 over East Siberia.

– Ozone total content and vertical distribution behave, as a rule, in good agreement with the circulation processes and the behavior of baric formations.

– In the chemical transformations of ozone, the cycle of interactions with odd nitrogen NO_x is well pronounced. For more detailed study of the chemistry of stratosphere we are planing to arrange the channels of spectrophotometric measurements of the NO_3 and OClO total content in the near future. It should be emphasized that the information about NO_3 should serve as a peculiar kind of indicator of the chlorine cycle of stratospheric ozone, sine the ratio $NO_2/O_3/NO_3$ changes markedly at production of molecules–reservoirs $ClONO_2$.

– On the whole, many-year observations give no grounds to the statement of the prevailing part of anthropogenic factor in the destruction of ozone layer of the Earth's atmosphere.

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