Optical monitoring of unperturbed ozonosphere at Siberian Lidar Station

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The paper presents some results of analysis of the data of lidar sensing of the ozonosphere in February and March of 1996–1999 period. As the initial data for analyzing the ozone vertical distribution (OVD) and the temperature vertical distribution (TVD) in the stratosphere the night-mean profiles were used in the altitude range from 13 to 35 km. Analysis also involves the data on nitrogen peroxide vertical distribution (NO_2VD) in the 5 km layers in the stratosphere obtained by means of a twilight spectrophotometer as well as the data on the total ozone content (TOC) based on the ozonometric observations. A higher ozone content observed in 1996 and 1998 was accompanied by a temperature rise in the middle stratosphere. High correlation between the OVD and TOC in the lower stratosphere was found. The correlation analysis of OVD and TVD revealed a significant positive correlation in the 18–25 km layer as well as a significant negative correlation in the 13 km layer. The calculated altitude distribution of trends in the ozone content and the temperature in the stratosphere indicates a marked decrease of ozone and cooling of the atmosphere at altitudes of 22–30 km as well as an increase in the ozone content in the lower stratosphere, at the heights from 14 to 16 km. Trends in the NO_2 content in 5 km layers in the stratosphere also point to a statistically significant decrease in the NO_2 content within the layer from 30 to 35 km.

Introduction

In the early 1980's the Director of the Institute of Atmospheric Optics Academician V.E. Zuev has formulated a task of developing stationary lidars based on large-size receiving mirrors for laser sounding of the stratosphere and mesosphere. The first version of such a lidar with a 1-m-diameter mirror was constructed in 1985.[1] Since 1986 this lidar has been used for regular monitoring of the optical state of the stratospheric aerosol layer (SAL) over Tomsk.[2] In 1989 this lidar was equipped with the UV–channels for laser sounding of stratospheric ozone.[3] In 1990 a multichannel stationary lidar with a 2.2-m-diameter receiving mirror has been put into operation. This lidar facility has been mounted inside a specially designed building. Using this lidar the multifrequency lidar sensing of stratospheric aerosol was performed and the data were obtained not only on the optical but also on microstructure characteristics of aerosol in the altitude range from 10 to 30 km.[4] Using this lidar the observations were made of the behavior of the vertical distribution of stratospheric ozone especially after the Mt. Pinatubo eruption in 1995.[5] And finally, this lidar has made it possible to make temperature measurements in the altitude range up to 80 km.[6]

For the purpose of complex investigation of the ozone layer transformation on the basis of knowledge gained in one of the main research areas of the Institute of Atmospheric Optics a long-term scientific program on research in the stratospheric and tropospheric ozone (SATOR) has been unfolded in 1991.[7] In the framework of this program two basic experimental measuring complexes were constructed. One of these complexes is the TOR-station for integrated study of ozone forming and ozone destructing processes in the atmospheric boundary layer[8] and the Siberian Lidar Station (SLS) intended for integrated study of transformation of stratospheric ozone layer (SOL).[9] The measuring complex of Siberian Lidar Station consists of stationary lidars with the receiving mirrors of 1 m and 2.2 m in diameter and also of a sun spectrophotometer and a twilight spectrophotometer. The optical arrangement of the stratospheric measuring channels of the complex is depicted in Fig. 1. This lidar and spectrophotometric measuring complex enables us to perform practically on one and the same spatiotemporal scale the optical monitoring of basic characteristics of the stratosphere, which well represent the SOL behavior, namely, the ozone (OVD), temperature (TVD), aerosol (AVD) and nitrogen peroxide (NO_2VD) vertical distributions, and the total ozone content (TOC). A combined analysis of variability of these characteristics with a strongly inhomogeneous spatiotemporal structure gives a great deal of information about SOL transformation due to dynamic and photochemical processes in the atmosphere.

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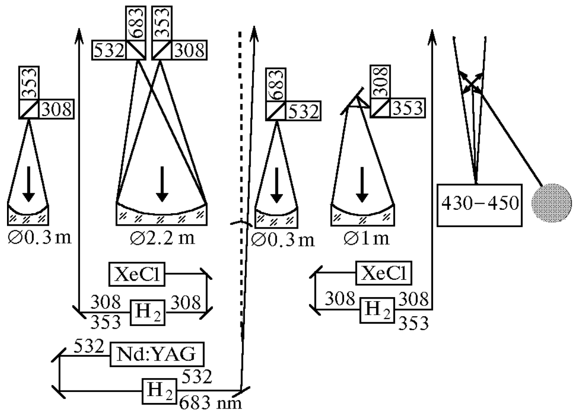

Fig. 1. Optical arrangement of the lidar–spectrometric complex of the Siberian Lidar Station (56.5°N; 85.0°E); the operating wavelengths are denoted by figures.

The unique composition and parameters of the basic experimental measuring complex of the Siberian Lidar Station and a realizable complex approach to optical monitoring of the ozonosphere determined an official inclusion of the SLS by the order of the Ministry of Science of the Russian Federation in the List of Unique Scientific–Research and Experimental Setups of the National Significance (reg. No. 01–64).

Up to now the data of long-term observations over aerosol vertical distribution (14 years), ozone vertical distribution (11 years), NO_2 vertical distribution (5 years), and total ozone content (7 years) have been collected at the SLS. The results of investigations were summarized in a number of publications.[10–16] A considerable period in these temporal variations is connected with the effect produced on the stratosphere by the products of the most powerful in this century explosive eruption of the Mt. Pinatubo on the Philippine Islands in June 1991. This eruption has determined an extended period of a depression state and subsequent reconstruction of the stratospheric ozone layer observed over Tomsk during a four-year period (Fig. 2). The state of the ozonosphere can be considered "unperturbed" only from summer 1995 and up to now.

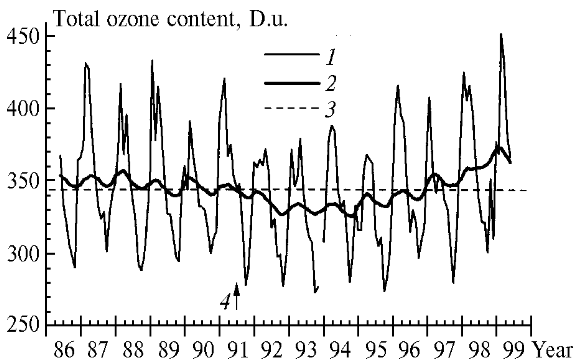

Fig. 2. Time dependence of TOC over Tomsk from June 1986 to June 1999 based on the data of ground-based (M–124) and spaceborne (TOMS) measurements: monthly mean values (*1*); 26 monthly sliding mean (*2*); long-term mean (344 Dobson's units) (*3*); the instant of Mt. Pinatubo eruption (*4*).

For a more sophisticated treatment of the processes forming the present–day state of the "unperturbed" ozonosphere, we have statistically processed the data of optical monitoring of OVD, TVD, NO_2VD in the stratosphere, and TOC. We excluded from the consideration the high–amplitude seasonal oscillations in time variations, which make analysis difficult, and used only winter periods (February–March) corresponding to maximum variability and maximum ozone content in the annual variation. The results of that statistical analysis and their discussion make up the subject of this paper.

Results of lidar observations of OVD and TVD

As the initial data for statistical analysis we used the night mean profiles of OVD and TVD obtained from a series of individual lidar profiles. The profiles of OVD and TVD were obtained within the altitude range from 13 to 35 km with a spatial resolution from 100 to 400 m, and measurement time, characterizing the profile averaging, from 1.5 to 3 hours. In the subsequent discussion for statistical analysis the profiles of OVD and TVD were averaged over the 2-km-thick layers. "esides the above analysis we have also analyzed the data of twilight spectrophotometric measurements from which NO_2VD was reconstructed with the averaging over 5-km thick layers at altitudes ranging from 0 to 60 km (the data were analyzed only at altitudes from 15 to 30 km).

The data on OVD and TVD were obtained using an UV–DIAL–lidar with a 1-m-diameter receiving mirror at the wavelengths of 308/353 nm and 353 nm, respectively, and the data on the NO_2VD were obtained using a twilight spectrophotometer at the wavelengths from 430 to 450 nm. The instrumentation was a part of the measuring complex at Siberian Lidar Station (Fig. 1) and was described in detail elsewhere in the literature.[17,18]

The summarized results of lidar sensing of the ozonosphere obtained in 1996–1999 in the form of the mean vertical ozone profiles over the periods from February to March are shown in Figs. 3 and 4. Here the curves of rms deviation are given indicating the ozone variability as well as the variability of the model ozone distribution (the Krueger model)[19] and temperature (the midlatitude model).[20] As is seen from Fig. 3, for all the obtained mean profiles of the OVD a stable, in altitude, ozone maximum (at about 20 km) was observed. As compared with the model distribution, in the upper part of the profile we observed reduced ozone content and below the ozone maximum – the enhanced ozone content. Such a descend of ozone layer was observed previously when analyzing the lidar sensing data obtained in winter and in summer 1998.[13,14] This altitude decrease characterizes the regional peculiarity of the OVD over Tomsk.

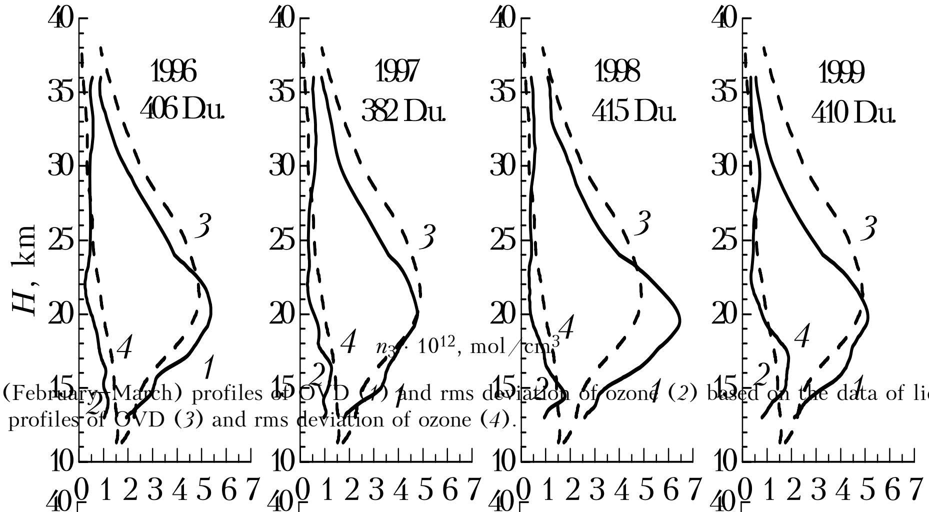

Fig. 3. The mean (February–March) profiles of OVD (*1*) and rms deviation of ozone (*2*) based on the data of lidar observations in 1996–1999, model profiles of OVD (*3*) and rms deviation of ozone (*4*).

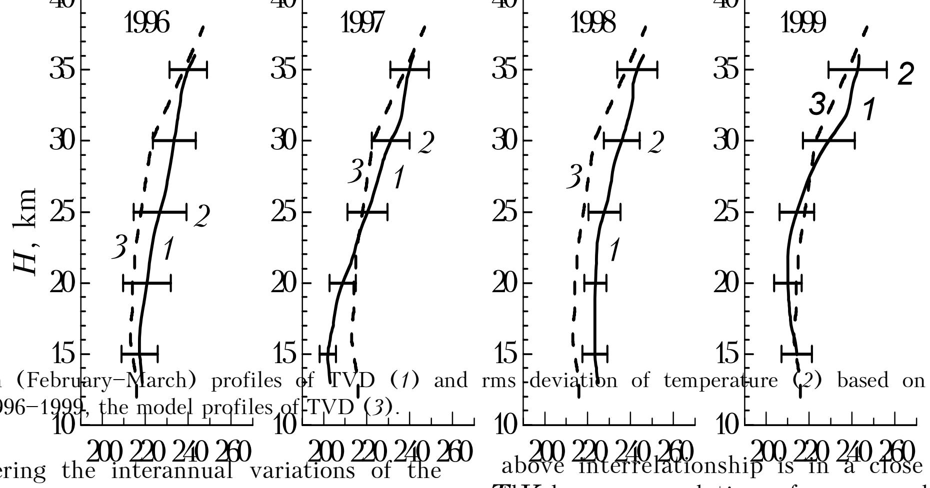

Fig. 4. The mean (February–March) profiles of TVD (*1*) and rms deviation of temperature (*2*) based on the data of lidar observations in 1996–1999, the model profiles of TVD (*3*).

In considering the interannual variations of the OVD the typical, for midlatitude stratosphere, periodic quasi–two–year variations of the ozone content are clearly seen, being most obvious in the ozone maximum. Thus in 1996 the ozone concentration here was $5.5 \cdot 10^{12}$ mol/cm^3, in 1997 the ozone concentration dropped down to $4.5 \cdot 10^{12}$ mol/cm^3, in 1998 the ozone concentration reached $6.3 \cdot 10^{12}$ mol/cm^3, and finally, in 1999 the concentration again decreased to $5 \cdot 10^{12}$ mol/cm^3. The above-mentioned ozone maximum variations correlate well with the TOC variations during the same periods. The values of TOC variations are shown in Fig. 3. Analysis of the variability of OVD has shown that, as could be expected, the maximum by magnitude and repetition ozone variations are observed in the layer from 12 to 20 km in the lower stratosphere. It should be also noted that the rms deviation values obtained are in a good agreement with the model ones.

In the vertical temperature distribution in Fig. 4 considerable interannual differences are observed at altitudes below 20 km as well as the quasi–two–year cyclicity can be followed up. ” y comparing the TVD and OVD (see Figs. 3 and 4) it is easy to verify that higher ozone content corresponds to enhanced temperatures (1996 and 1998) and vice versa. The above interrelationship is in a close agreement with the known correlation of ozone and temperature in the lower stratosphere.[21]

Results of correlation analysis of OVD, TVD, and TOC

The calculated results on correlation between the 2-km layer of OVD and TOC are shown in Fig. 5. It can be seen from the figure that at the significance level of the correlation coefficient at 0.3 with the confidence coefficient 0.95% the lower part of ozone layer correlated with TOC most strongly (curve *2*). This result differs slightly from the previously obtained result of correlation analysis carried out based on the laser sounding data obtained in summer and winter 1998.[13,14] In this case the region with the maximum positive connection between OVD and TOC was observed in the upper part of ozone layer above the maximum. Such a difference is not contraction since in that case particular events were found presenting the atmospheric processes of those periods. In this case we consider an average period where the peculiarities of separate periods are smoothed out and the overall regularity is revealed.

If one considers in Fig. 5 the mean profiles of the coefficient of correlation between the OVD and TOC, it should be mentioned that in the lower part of the OVD profile both maximum variability of the ozone content and maximum correlation between TOC and OVD are observed. Hence, one can conclude that it is just this layer in OVD at altitude from 13 to 20 km, which mainly determined the TOC variability thus confirming once more this well-known fact (see, for example, Ref. 23).

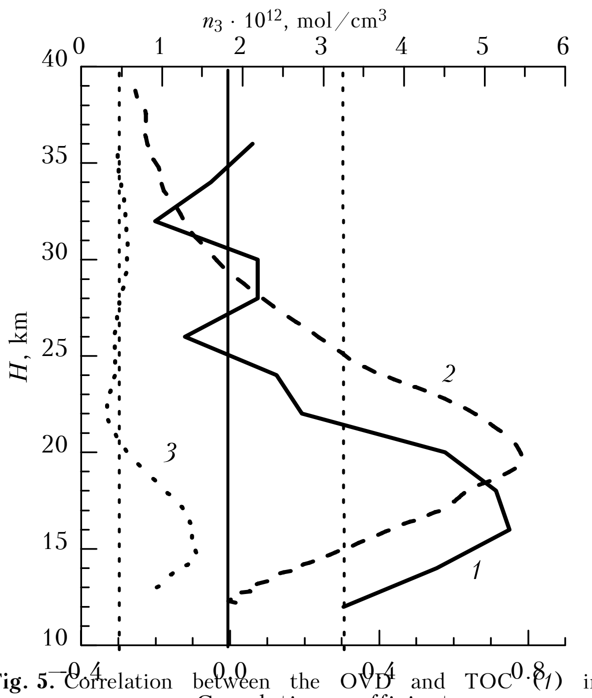

Fig. 5. Correlation between the OVD and TOC (*1*) in February–March and mean profiles of OVD (*2*) and rms deviation of ozone (*3*) based on the data of lidar observations in 1996–1999.

In Fig. 5 (curve *1*) an altitude range close to 26 km can be observed where the correlation coefficient is not statistically significant but, nevertheless, it shows certain regularity in the OVD due to the atmospheric processes involved in its formation. In the midlatitudes this range is a transition region with a complex small-gradient structure of ozone field[24] where the dynamic and photochemical processes affect the formation of OVD,[25] namely, at altitudes up to 22–24 km the dynamic processes become dominating in the OVD formation being responsible for the horizontal–vertical transport of ozone, while at altitudes above 28–30 km the photochemical processes dominate in the OVD formation. As a result a marked decrease is observed in the altitude behavior of the coefficient of correlation between the OVD and TOC at heights near 26 km.

The negative correlation at altitudes higher than 30 km (Fig. 5, curve *1*) shows the peculiarity of horizontal ozone distribution in the middle and upper stratosphere, which has a marked zone structure and a horizontal gradient directed toward lower latitudes (in the lower stratosphere the gradient is directed to the North Pole). Therefore, at large-scale reconstructions of baric field encompassing the troposphere and the stratosphere, with the increasing ozone content in the lower stratosphere and, respectively, increased ozone content in the middle stratosphere, the decrease of any correlation between OVD and TOC near 35 km altitude is indicative of the photochemical processes dominance. Owing to these processes the ozone content rapidly recovers and is in photochemical equilibrium.

The altitude behavior of the coefficient of correlation between the layered ozone content and the temperature is shown in Fig. 6. The two altitude intervals can be isolated with a significant amount of correlation (more than 0.3) near 13 km with the negative correlation and in the range of 18–25 km with the positive correlation. The significant negative correlation shows an essential dependence of the ozone and temperature on the direction of a meridian component of atmospheric transport and on type of circulation in the troposphere and lower stratosphere when the advection of cold in a back front of a cyclone (front of anticyclone) is followed by an increase in the ozone content and, on the contrary, the advection of heat in the front of a cyclone (back front of an anticyclone) is followed by a decrease in the ozone content. This is especially true for high, vast baric formations propagating in the lower stratosphere. Significant positive correlation in the range of the main ozone maximum represents a well-known relations among geopotential, temperature, and ozone content in the stratosphere taking into account vertical transfer – the Dobson–Normal–Ried principle,[21,23] according to which the descending flows bring down from above the air rich with ozone to lower and middle stratosphere. In this case the air within these flows is adiabatically heated. In turn, the ascending flows decrease the amount of ozone in the lower and middle stratosphere and are adiabatically cooled. This is conditioned by the fact that in midlatitudes the ozone mixing ratio grows with the altitude reaching its maximum at 32–34 km altitude and then decreases. ” ecause ozone is a long–lived component in the lower stratosphere, it can be accumulated there. In the upper stratosphere, at altitudes higher than 35 km, the lifetime of the ozone molecule is less than one day. Therefore ozone occurs there in a photochemical equilibrium. The equilibrium rapidly recovers or breaks when there is a lack or an excess of ozone, respectively. In Fig. 6 of particular interest is the altitude range close to 15 km (although statistically insignificant because of small length of a series but interesting from the physical viewpoint), where the layer is observed in the altitude behavior of the correlation coefficient, in which the sign of correlation is changed and the coefficient of correlation takes nearly zero value. As is seen from Figs. 3 (curve *1*) and 5 (curve *3*) in this altitude range secondary ozone maximum is observed. In this case the fine layered ozone structure is manifested and here the ozone variability is most strong. This altitude range in the midlatitudes represents an intermediate interval between tropospheric and stratospheric circulations, (in the troposphere the atmospheric motion occurs mainly in the form of vortexes, in the stratosphere – in the form of long planetary waves) where the meridian component of atmospheric transfer is observed. This component is unstable in the velocity and direction. Under the effect of the meridian component due to intrusion in the vertical ozone distribution a thin layered structure is formed. With the decreasing length of a planetary wave (the increase of the wave number) that is observed at blocking processes in the troposphere, the increase of meridian circulation as well as the strengthening of interlatitude exchange are observed, which result during winter–spring period in midlatitudes in a wide change of ozone content both in the OVD within the lower stratosphere and in TOC as a whole. The decrease and the change of the sign of the coefficient of correlation between OVD and TVD at altitudes above 30 km in the middle stratosphere, although the correlation coefficient is statistically insignificant, depend on the same Dobson–Norman–Ried principle and on the decrease of the ozone mixing ratio with altitude in the upper stratosphere.

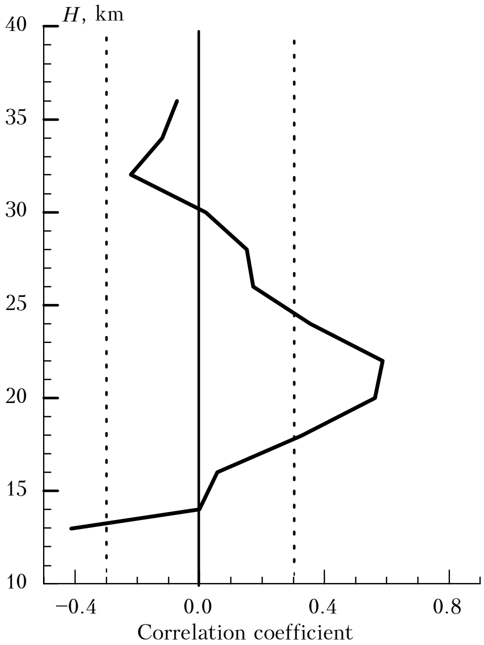

Fig. 6. Correlation between OVD and TVD in February–March calculated from the data of lidar observations in 1996–1999.

Variations in the altitude behavior of the gradient of the coefficient of correlation between the OVD and TVD in the middle stratosphere in the layer from 22 to 28 km is most likely connected with the presence of the transition region in OVD, while horizontal gradients in the field of temperature do not change their direction with altitude variation. And because the short-period variability of ozone due to horizontal advection exceeds variations of the ozone content owing to vertical flows, a small variation in the altitude behavior of the correlation coefficient occurs in the layer from 22 to 28 km.

Now we discuss the results presented in Figs. 7 and 8 concerning the analysis of interlayer correlation between the OVD and TVD performed using data of laser sounding of ozone and temperature in February–March 1996–1999. The autocorrelation functions between the layers averaged over 2 km have been calculated.

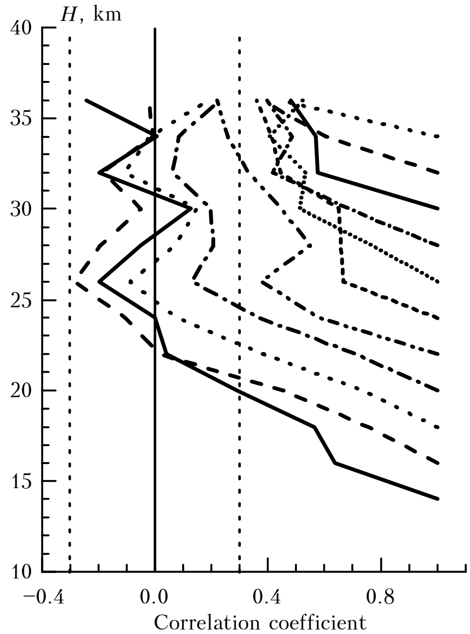

Fig. 7. Interlayer correlation of OVD in February–March based on the data of lidar observations in 1996–1999.

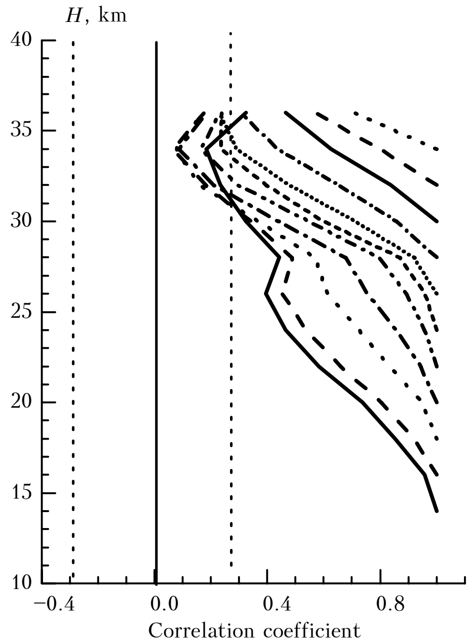

Fig. 8. Interlayer correlation of TVD in February–March based on the data of lidar observations in 1996–1999.

Figure 7 shows the correlation functions of the OVD where three different groups of isocorrelates can be distinguished, which characterize certain atmospheric processes at different altitudes. The first group of isocorrelates includes the layer up to 20 km, the second group includes the layer from 20 to 26 km, and the third group includes the layer located above 26 km.

In the first group a uniform and fast decrease of correlation is observed; at the same time in the layer from 14 to 18 km the correlation coefficients change sign at altitudes ranging from 22 to 26 km. The levels of this group have variable near-zero correlation with the levels higher than 26 km. Such a behavior of the autocorrelation points to a marked nonzonality of the OVD in the lower stratosphere up to 20 km during the considered period and reflects clearly a mechanism of horizontal-vertical ozone transport, which will be discussed below.

In the third group the altitude behavior of correlation in the layer above 26 km has a significant correlation of 0.4-0.6 with a stable altitude behavior of the correlation coefficient at high density of isocorrelates. High density of isocorrelates, especially above 28 km, shows a marked zonality and low variability of the OVD in the middle stratosphere, that depends on a significant weakening of the impact of dynamic processes on the ozone and the increase of the photochemical effects role.

In the second group the inversion is observed in the altitude behavior of a significant correlation coefficient for 22 km level in the layer from 26 to 28 km. On the whole, the layer from 22 to 26 km in the OVD is a transition one and according to Ref. 24, has a complex structure at comparatively small horizontal gradients in the field of ozone. For this reason the variability of atmospheric circulation in the above layer results in significant variations in the ozone field.

The mechanism of horizontal-vertical transport of ozone forming the OVD, in which the above-mentioned peculiarities of interlayer correlation are observed is as follows. With the advection of an arctic air mass, which is rich with ozone in the lower stratosphere while having low ozone content in the middle stratosphere, the increase of ozone content in the lower stratosphere and the decrease of ozone content in the upper stratosphere are observed. On the contrary, with the advection of a tropical air mass having low ozone content in the lower stratosphere while a higher one in the middle stratosphere, a decrease of the ozone content below and an increase in the upper layer occur.

The presence of ascending flows results, according to Ref. 23, in an extra decrease of ozone in the lower and middle stratosphere, especially in the 24-26 km layer, as well as in a slight increase of its content in the 26-30 km layer. The presence of descending flows tends to an extra significant increase of ozone in the lower stratosphere and a slight increase of the ozone in the middle stratosphere; in this case in the upper stratosphere the ozone content practically does not change because of its fast photochemical recovery.

As compared with the interlayer correlation of the OVD the behavior of correlation of the TVD shown in Fig. 8, is different. In this case two groups of isocorrelates may be distinguished. The first group includes the layer up to 26-28 km, the second group includes the layer above 28 km.

In the first group we observe a gradual fall off of the correlation with altitude increasing up to 26-28 km and the inverse behavior for a layer of 14-16 km in the altitude range between 26 and 28 km with a subsequent decrease to a level of insignificance (less than 0.3 for 95% probability) at an altitude above 30 km (although above 35 km the level of significance of correlation with the layer at 14 km layer is achieved). From the comparison of the behavior of isocorrelates and the results of numerical simulation of distributions of the ozone temperature and mean zonal wind for winter in midlatitudes,[25] which are in close agreement with the results of observations, it follows that such an altitude distribution of interlayer correlation reflects peculiarity of the TVD in the lower stratosphere. The zonal distribution of the TVD is typically characterized by small horizontal and vertical gradients. In the middle stratosphere a sharp increase of vertical and horizontal gradients is observed in the vertical temperature distribution and vertical distribution of zonal wind due to the influence of stratospheric jet flow in the region of the stratopause during the transition between polar night and day. In our case this increase of gradients of the isocorrelate manifests itself at heights from 26-28 km by the increase of correlation between the 14-16 km layer and the layer at 26-28 km height as well as by a marked variation of its altitude behavior starting with 28 km height. Thus the second group of isocorrelates can be constructed, that shows the TVD in the middle stratosphere of the midlatitude winter in the altitude range of the stratospheric jet flow periphery and, as a result, the break of the TVD zonality is observed there.

Trends of TOC, OVD, TVD, and NO_2VD in the stratosphere

As investigations of TOC have shown (see the results presented in Fig. 9 in the form of time series of monthly mean values measured from June 1995 to June 1999), a stable tendency toward an increase in the total ozone content is observed during a five-year period both for annual mean and for monthly mean values in February and March. The estimated ozone trends show that the TOC increased throughout this period at a rate of 12 ± 5 D.u./year (or $3.4 \pm 1.4\%$ yearly), and at a rate of 5 ± 10 D.u./year (or $12 \pm 2.5\%$ yearly) according to February-March data. In the first case the trend is statistically significant within 98% probability, while in the second one it is statistically insignificant.

A detailed analysis of reasons for the TOC behavior observed can be found in Ref. 14. In this case it is necessary to compare the TOC trend with the trends calculated for the OVD and TVD shown in

Figs. 10 and 11. The trends in TOC, OVD, and TVD were evaluated with the use of linear regressive analysis without the account of quasi-two-year cyclicity.

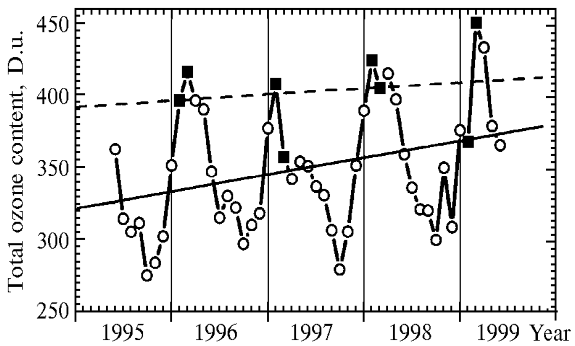

Fig. 9. Time dependence of monthly mean values of TOC from June 1995 to June 1999 based on data of ozonometric observations: linear trend 12 ± 5 Dobson's units/year (3.4 ± 1.4% yearly) is shown by solid curve as calculated using data of the entire series; the trend of 5 ± 10 Dobson's units/year (1.2 ± 2.5% yearly) shown by dashed line was calculated using the data obtained in February-March period indicated by solid marks.

As seen from Fig. 10, a negative statistically significant trend is observed in the OVD in the middle stratosphere at altitudes ranging from 27 to 32 km with the maximum about −3% a year at 30 km altitude. At the same time there is observed an increase of the ozone content in the lower stratosphere. The statistically significant value less than 3% a year of the trend is observed at 16 km altitude. "y this it is meant that the TOC increase revealed is due to the ozone growth in the lower stratosphere where, as far as we know, the amount of ozone is larger.

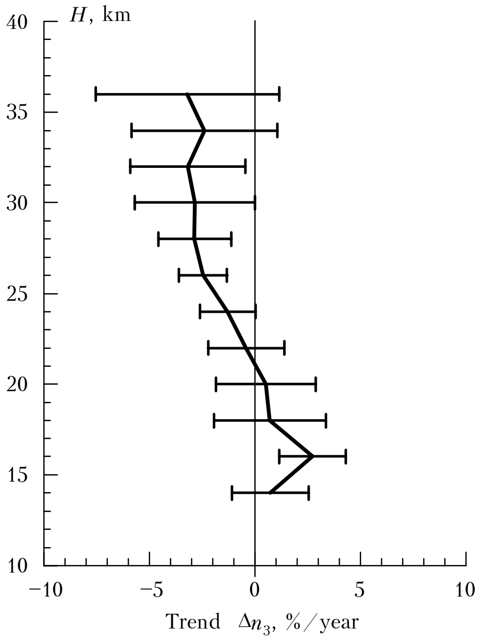

Fig. 10. Altitude distribution of the trend of ozone concentration and its standard deviations in the stratosphere in February-March 1996-1999 according to data of lidar observations.

In the TVD shown in Fig. 11 a negative trend is also observed in the upper part of the lower stratosphere and in the bottom part of the middle stratosphere. The maximum statistically significant temperature decrease to −1 K a year is observed at 25 km altitude. In the upper part of the middle stratosphere an opposite tendency is observed, namely, the temperature rise, whose maximum statistically significant value equals 0.4 K a year at altitude about 34 km. Analysis of the trends in the OVD and TVD obtained allows the following conclusions to be drawn. A slight statistically insignificant rise in temperature, occurring in recent years in winter and spring over Western Siberia, and the increase of the ozone content in the lower stratosphere and the temperature drop as well as the ozone content decrease in the middle stratosphere may be related to the strengthening of the "residual" (or nonadiabatic, Stokes) meridian circulation due to the strengthening of the vortex component of atmospheric circulation. The strengthening of vortex transport is connected with activation of blocking processes in the troposphere and slowing down of long planetary waves in the stratosphere.

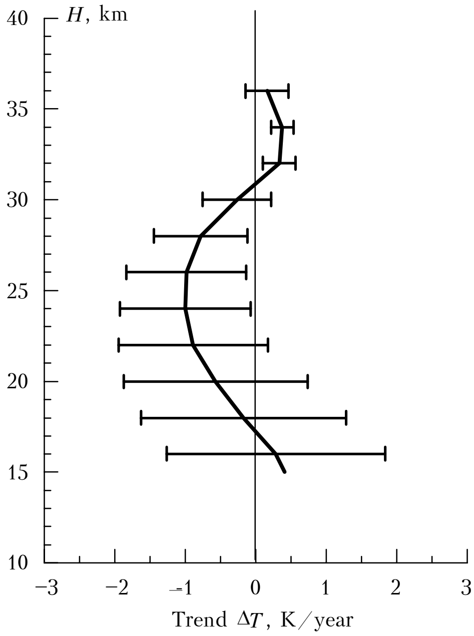

Fig. 11. Altitude distribution of the temperature trend and its standard deviations in the stratosphere in February-March 1996-1999 according to data of lidar observations.

The suggestion has been made that there exists interrelation between a secular solar cycle and a long-term variation of the general circulation of the atmosphere,[27,28] which, in particular, manifests itself in variations of parameters of centers of action of atmospheric circulation (Azores and Hawaiian anticyclone, Icelandic and Aleutian systems and other constant and seasonal centers of action). The strengthening of meridian circulation and blocking processes is precisely determined by strengthening of the influence of these centers of action or their pairs on the character of atmospheric transport and, respectively, on the change of content of conservative components in the atmosphere, such as ozone and stratospheric aerosol.

In the troposphere and lower stratosphere the strengthening of meridian circulation is followed by a local increase of the value of a potential velocity vortex connected with the strengthening of descending flows in the lower stratosphere, which may cause both the influx of ozone from above and its adiabatic heating, as well as advection of cold in the troposphere. In midlatitudes the strengthening of convergence of zonally averaged turbulent heat flux occurs in the middle stratosphere due to slowing down of a long planetary wave in the below laying layers. The heat flux is compensated for by cooling when broadening (owing to ascending motions).[25] In general, as is shown in Ref. 26, the temperature variations due to wave perturbations in the stratosphere in winter may have an opposite behavior.

The existence of the opposite trends in the OVD and TVD in the middle stratosphere in the layer above 30 km may be conditioned by the inverse connection between the temperature and ozone concentration in a photochemically active layer of the stratosphere (higher than 30–32 km),[25] i.e., the temperature rise owing to wave perturbations will cause a compensating decrease of photochemical ozone formation that agrees also with the NO_2VD behavior for the same period. The time behavior of the NO_2VD exhibits negative trends. From all the trends shown in Fig. 12 the trend in the 30–35 km layer is statistically significant. According to known photochemical bond of the odd nitrogen series, chloride and oxygen in the middle and upper stratosphere,[25,26] the decrease in the ozone content causes an increase in the chlorine oxide quantity usually bonded by nitrogen peroxide, which is in photochemical equilibrium with the chlorine oxide. Therefore, to establish the heat balance, a larger amount of nitrogen peroxide is consumed. At the same, a pronounced feedback between the temperature and the nitrogen peroxide content exists due to the temperature dependence of the oxidation reactions of nitrogen oxide and peroxide in the nitrogen catalytic exchange.

There exists an alternative mechanism explaining the decrease of the ozone and nitrogen peroxide content in the photoactive stratospheric layer associated with the change of the photochemically active solar emittance resulting in the decrease of photochemical formation of both the ozone and nitrogen peroxide. However, the interconnection of the variability of solar activity and the atmospheric circulation is too intricate to grasp and should be thoroughly checked. And finally, the variations of atmospheric circulation and thermodynamic regime of the stratosphere are connected with the other climatic factors, e.g., with the increase of concentration of greenhouse gases in the atmosphere. Note that practically all the interconnections in the atmosphere between its components and parameters are reversible and intersecting that makes their analysis and a search for regularities difficult. Hence, our conclusions regarding the trends of OVD and TVD to establish the heat balance to be true to life need for a larger amount of nitrogen peroxide. At the same time, a pronounced feedback between the temperature and the nitrogen peroxide content exists due to the temperature dependence of the reactions of oxidation of nitrogen oxide and peroxide in the nitrogen catalytic exchange.

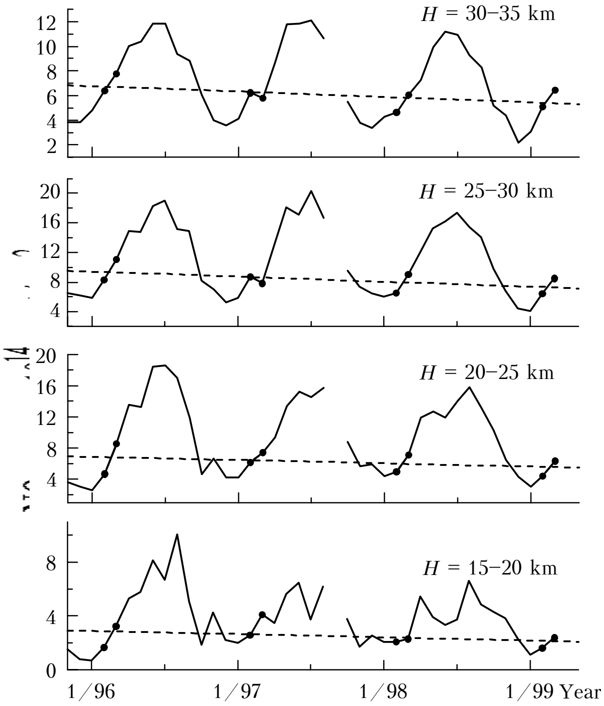

Fig. 12. Time dependence of monthly mean values of NO_2 content in the 5 km layers in the stratosphere from November 1995 to March 1999 according to data of spectrophotometric observations: linear trends are calculated based on the data for February–March shown by dots; the trend value in the 30–35 km layer, statistically significant at 85% level, equals $(0.44 \pm 0.27) \cdot 10^{14}$ mol $\cdot$ cm^{-2} year^{-1}.

Hence our conclusions relative to the trends of the OVD and TVD are disputable and should be discussed later on.

Conclusion

Unfortunately, a long-term influence of the volcanic emissions on the ozonosphere has resulted in a significant reduction of the data bulk and, respectively, statistical provision of the experimental data in our time series characterizing its "unperturbed" state. However, based on general analysis of all the experimental results obtained we can arrive at some qualitative conclusions.

1. The observed cooling of the stratosphere following the cooling of the mesosphere[29] is associated

with the long-term climatic change of atmospheric circulation, which may likely be caused by a secular cycle of the solar activity.

2. The long-term climatic variations of the general circulation of the atmosphere are specifically manifested in the oscillations of pairs of centers of action of the atmosphere such as the north–Atlantic, north–Pacific, and south oscillations. The change of the strength of these centers influence on the atmospheric circulation appears in the change of both intensity of transport of conservative components in the lower stratosphere (e.g., ozone and aerosol) and in formation of thermodynamic conditions favorable for the photochemical and/or dynamic depletion of the stratospheric ozone.[25,30,31] The existence of a positive trend in the behavior of the ozone content in the lower stratosphere in recent years tends to the increase of transport of arctic air masses associated with the activation of blocking processes in the troposphere and wave processes in the lower stratosphere.

3. The variation of photochemically active solar emittance must immediately affect photochemical processes in the middle and upper atmosphere. These data agree with the observational data on NO_2 content in the stratosphere, which is one of the most photoactive trace gases in the atmosphere, and the ozone content in photochemically active layer in the middle atmosphere, what indicates to the presence of negative trends.

The above results are in a good agreement with the recently[32] proposed hypothesis on the influence of a secular cycle of solar activity on the behavior of the ozonosphere, according to which at the turn of the XXth and the XXIst centuries the increase of the total ozone content in the Earth's atmosphere should be observed.

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