On the effect of volcanic aerosol on variations of stratospheric ozone and NO_2 according to measurements at the Siberian Lidar Station

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Variations of stratospheric ozone and nitrogen dioxide (one of the key components of catalytic nitrogen cycle of ozone destruction) after powerful eruption of Pinatubo volcano (June, 1991), as well as after relatively weak eruption of Rabaul volcano (October, 2006) are considered on the basis of the measurements obtained at the Siberian Lidar Station (SLS) of the Institute of Atmospheric Optics SB RAS in Tomsk (56.5°N, 85.0°E). It is pointed out that the artificial increase of the stratospheric aerosol content, suggested recently for compensation of the global green-house effect, may result in the uncontrollable distortion of stratospheric chemical balance and destruction of the stratospheric ozone layer.

Introduction

The state of the stratospheric ozone layer, where more than 85% of total atmospheric ozone is concentrated, is prone to following factors: chemical processes, atmospheric dynamics, the temperature, solar activity, powerful volcanic. An expressed depression of the stratospheric ozone is observed after the powerful explosive volcanic eruptions, when the sulfur-containing gases are emitted directly in the stratosphere and form there sulfate aerosol (on the average, 75%-solution of sulfuric acid in water). A considerable global decrease of ozone was observed during several years after eruptions of most powerful in the XXth century volcanoes Agung (March, 1963), El Chichon (March–April, 1982), Pinatubo (June, 1991).[1–5] The Pinatubo eruption caused the largest decrease (10–15%) of total ozone content (TOC) in the atmosphere of moderate and high latitudes of the Northern Hemisphere. The most pronounced ozone depression up to 30% was recorded in the central part of the Pinatubo aerosol layer (15–20 km). The depression is a result of heterogeneous chemical reactions of ozone destruction on the enlarged surface of stratospheric aerosol (SA), formed during several months after the eruption. Besides, after the powerful volcanic eruptions the radiation-temperature regime and general circulation of the atmosphere are disturbed that, in turn, affects the altitude and spatial variations in the stratospheric ozone distribution.

The level of the stratospheric ozone content was decreasing greatly up to the middle of the 1990s, especially in the region of poles. From the end of the 1990s up to the present time practically universally[6] the termination of the ozone layer destruction or even weak trend of TOC growth are observed, especially obvious in the moderate and high latitudes of the Northern Hemisphere. Simultaneously, the decrease of the content of ozone-destructive chlorine and bromine compounds in the stratosphere[6] is observed, which sometimes is associated with realization of the international Montreal Protocol (1987) and subsequent agreements, which limit the production and emission in the atmosphere of ozone-destructive compounds.

However, the long life time of Freons, ejected in the atmosphere, (more than 80 years) does not explain rapid paces in the modern-day reconstruction of the ozone layer. Thus, it is necessary to consider all factors affecting the variation of the ozone layer. The period of recorded TOC variations coincides with the long-term period of the absence of high-power explosive eruptions (the last-named is the Mt. Pinatubo volcano in June, 1991), which since 1997 is characterized by minimal level of background content of stratospheric aerosol for more than 30 years of regular observations with the use of the present-day ground-based and space means of investigations.

Based on the measurement data, obtained at the Siberian Lidar Station of the Institute of Atmospheric Optics SB RAS in Tomsk (56.5°N, 85.0°E), the mechanisms of variations of stratospheric ozone and components of ozone cycles in the stratosphere, both after the powerful Mt. Pinatubo eruption and after a comparatively weak Rabaul volcanic eruption (October, 2006) are considered in this paper.

1. Instrumentation

The lidar measurements of characteristics of the stratospheric aerosol layer (SAL) and stratospheric ozone, spectrophotometric measurements of TOC in the atmosphere, the total content and vertical

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distribution of nitrogen dioxide in the stratosphere are under consideration. The integral coefficient of aerosol backscattering B_π^a in the altitude range H from 15 to 30 km is determined from data of SAL laser sensing at a wavelength of 532 nm. This quantity characterizes the total aerosol loading of stratosphere. The vertical distribution of stratospheric aerosol can be represented in the form of the altitude profile of the scattering ratio $R(H)$ – the ratio of the sum of coefficients of aerosol backscattering and molecular scattering to the latter. The measurement error $R(H)$ varies from 3 to 4% within a 10–20 km altitude range and increases up to 6% at 30 km altitude. The observational error B_π^a is no more than 5%. The SAL characteristics at the Siberian Lidar Station have been measured since 1986.

The TOC measurements have been made since 1993 using a calibrated M-124 ozonometer. Time series of these observations are supplemented with data obtained with the space instrumentation TOMS,[8] which well correlate.[7] The vertical distribution of ozone concentration between 12 and 35 km were measured by the differential absorption lidar at 308 and 353 nm wavelengths. Such measurements have been conducted at the SLS since 1989.

The nitrogen dioxide has been measured since 1996, using an automated spectrophotometer,[9] which records spectra of solar radiation, scattered at zenith, in the wavelength range of 430–450 nm. The spectra obtained at twilight, when the solar zenith angle varies from 83 to 96°, are used for calculating the nitrogen dioxide content at oblique paths; in this case the measurement error does not exceed 5%. Then solution of the inverse problem gives the NO_2 content in the 5 km atmospheric layers.

The total NO_2 content in the vertical atmospheric column can be found as a sum by the corresponding layers. The present paper considers the total NO_2 content in the 10–50 cm column. The analysis of the morning and evening observations has shown the regular excess of the evening values by 25%. This reflects a slow transformation of NO and NO_2 to N_2O_5 during night with the subsequent photodissociation of N_2O_5 to NO_2 and NO at daytime. The morning measurements reflect the variations of NO_2, which occur mainly under the effect of dynamic atmospheric processes, while evening measurements show mainly changes caused by photochemical processes.

2. Results of measurements

2.1. Period of stratospheric aerosol disturbance after Pinatubo eruption, subsequent relaxation, and background state

Figure 1 shows the general dynamics of variations of SA and TOC by measurements in Tomsk, supplemented by TOMS data for the period before 1993. Prior to the Mt. Pinatubo eruption the ozonosphere was subjected to a series of weaker explosive eruptions of volcanoes. The ozone depletion is clearly seen in the period of SA increased content after the Mt. Pinatubo eruption. The marked ozone depletion was recorded in the troposphere layers with increased content of volcanic aerosol.[5] In spring of 1993 the marked layers of volcanic aerosol were not observed yet, and general increased aerosol content was retained in the lower stratosphere. Figure 2 shows an example of a typical altitude ozone distribution (curve *1*) and SA (curve *2*) in that period.

The SA distribution is given as the scattering ratio at a wavelength of 353 nm. At altitudes below 20 km (the area A in Fig. 2), where the increased aerosol content remains, the decreased ozone content is observed as compared with our empirical model of the background ozone distribution (curve *3*), built by measurements for a period 1996–2005. At the altitude of 24 km (the area B in Fig. 2) the ozone depletion is less expressed as compared with the long-term background mean in period 1996–2005. Possible radiation chemical processes, which are responsible for such altitude variations of ozone content, will be considered in Chapter 3.

Pre-Pinatubo values of SOC characteristics were observed at the end of 1996, and since 1997 these values even slightly lowered. The TOC values were restored by 1997. A so-called new background period under conditions of long-term absence of high-power volcanic eruptions has been observed since 1997. Aerosol content of the stratosphere during this period was minimal and depended on the background non-volcanic component of stratospheric aerosol. From measurements at the SLS from 1999 to September, 2006 the trend of variations of B_π^a was statistically insignificant, i.e., (-1 ± 4)% yearly[10] that, as a whole, agrees with the data of the balloon and lidar measurements at other observation points[11] of the Northern and Southern Hemispheres, which also show the absence of marked long-term variations in the level of content of the background non-volcanic component of the stratospheric aerosol.

Linear regression of a series of TOC values over Tomsk for the period of the SAL background state in 1996–2005 was measured as deviations of means for a month from the similar means for this period. Thus, a weak, statistically insignificant negative yearly trend (-0.22 ± 0.16)% is seen. For NO_2 series of the same period the trends are also statistically insignificant (-0.75 ± 0.44)% yearly for morning measurements and $(+0.25 \pm 0.39)$% yearly for evening measurements.

The NO_2 observational data in Tomsk were compared with data of spectrophotometric measurements of NO_2, NO, and HNO_3 at the station Kitt Peak (32°N, 112°E)[12] in the Northern Hemisphere. During the period from 1992 to 2002 the trends of values of evening observations normalized relative to the means for the period were the following: for the total NO_2 content in the stratosphere: $(+0.52 \pm 0.32)$% yearly, for NO $(+1.56 \pm 0.45)$% yearly. The trend for NO_2, like in Tomsk, was positive and more pronounced, because it reflected variations of NO_2 since 1992, i.e., included the period of relaxation of increased content of volcanic aerosol. During this period along with the decrease of SA content the increase of NO_2

content was observed. The total content of HNO_3 decreased from March, 1992 to the early 1996 by 20% and then it was relatively stable.

Thus, the presented results of observations show that since 1997, during the period of constant background SA content, TOC values of total content of NO_2, as well as NO and HNO_3 are relatively stable and do not undergo considerable long-term variations.

2.2. Variations of stratospheric ozone and NO_2 after the Rabaul eruption

A long period, free of volcanic eruptions, was interrupted by the Rabaul eruption on October 7, 2006 (Papua, New Guinea: 4.3°S, 152.2°E). The altitude of volcanic product emission was 18 km. Already on October 17, 2006 the traces of the emission were recorded over Tomsk in the form of the aerosol scattering increase in the lower stratosphere (Fig. 1*a*). A detailed dynamics of the development of aerosol perturbation in stratosphere over Tomsk was described in Refs. 10 and 13. The maximum aerosol content of the stratosphere was recorded late in January and early in March, 2007, when mean values of B^{a}_{π} were equal to $3.2 \cdot 10^{-4}$ sr^{-1} and increased ones reached $4.6 \cdot 10^{-4}$ sr. During the background period of 1997–2006 the mean values of B^{a}_{π} were about $1.5 \cdot 10^{-4}$ sr^{-1} and did not exceed $2.5 \cdot 10^{-4}$ sr^{-1}, which is maximal winter aerosol content in the stratosphere. As a whole, mean values of B^{a}_{π} after the Rabaul eruption were more than twice higher than many-year mean values of the background period. Many-year mean background values B^{a}_{π} over Tomsk returned to summer–autumn of 2007.

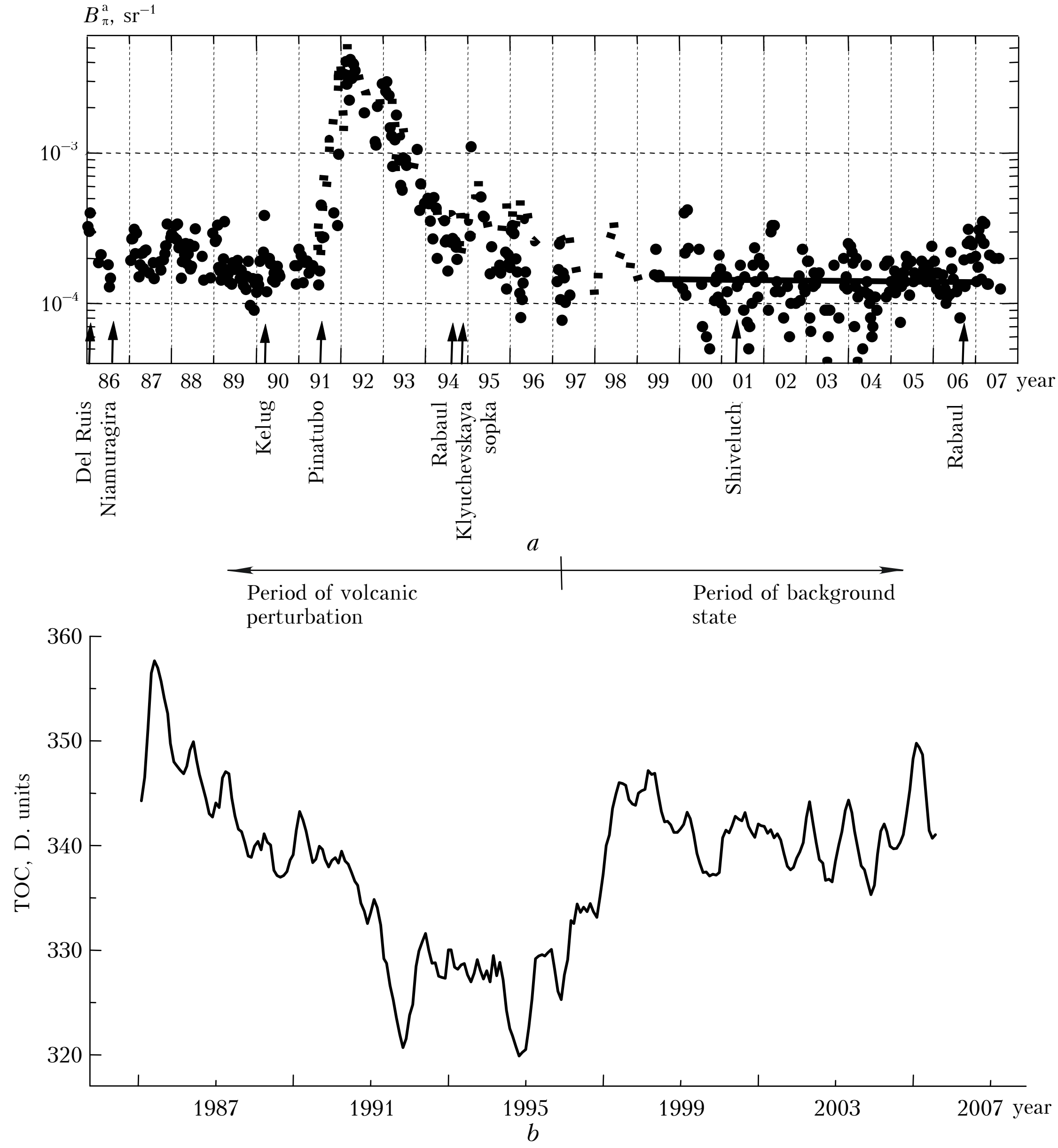

Fig. 1. Time behaviors of integral coefficient of aerosol backscattering within 15–30 km (volcanic explosive eruptions are shown by arrows) (*a*) and semiannual mean values of TOC, smoothed by a two-year sliding mean (*b*).

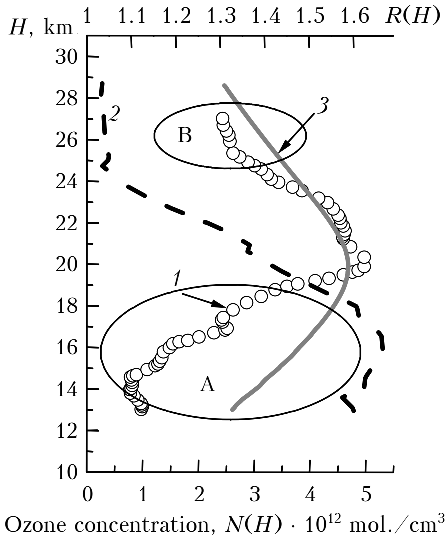

Fig. 2. Altitude distribution of ozone concentration from lidar measurements on April 12, 1993 (curve *1*); simultaneously measured vertical profile of the scattering ratio (*2*); the mean empirical model of background distribution of ozone from lidar measurements in 1996–2005 (*3*).

The character of aerosol perturbation of the stratosphere over Tomsk changed periodically. Typical examples of altitude SA distribution in the form of profiles $R(H)$ are given in Fig. 3.

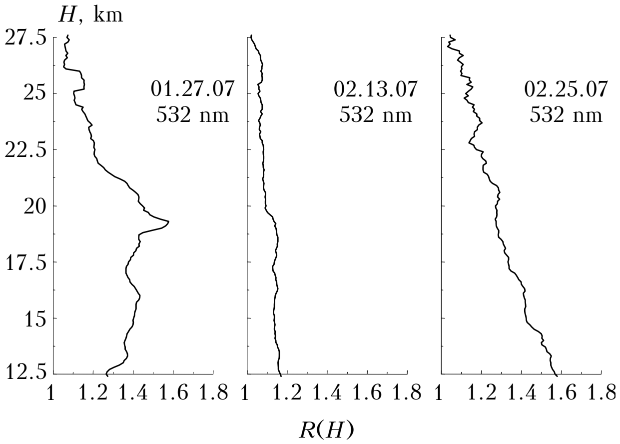

Fig. 3. Profiles of the scattering ratios.

A noticeable aerosol layer at 19 km altitude was observed on January 27, 2007; the state of SAL close to background was observed on February 13, 2007; the increased aerosol content at the lower stratosphere was observed on February 25, 2007.

Figure 4 shows the time dynamics of variations of B_{π}^{a} values, TOC, and NO_2 content for the observation period of maximum content of volcanic stratospheric aerosol late in January – early in March, 2007. The values of B_{π}^{a} in Fig. 4*a* are given for particular measurement days. The number of days is determined by conditions of cloudless sky, under which lidar measurements in the stratosphere are possible. The time dependence of TOC and NO_2 content is presented for the same dates of observations. For TOC (Fig. 4*b*) we used data of TOMS [Ref. 8] for the Tomsk site. Figure 4*c* shows the values of the total content of NO_2 in a 10–50 km column for evening measurements. As a whole, figure 4 shows that to the short-term periods with increased SA content on January 27 and February 21–27, correspond to the periods with the decreased content of ozone and nitrogen dioxide, and for February 14 – vice versa.

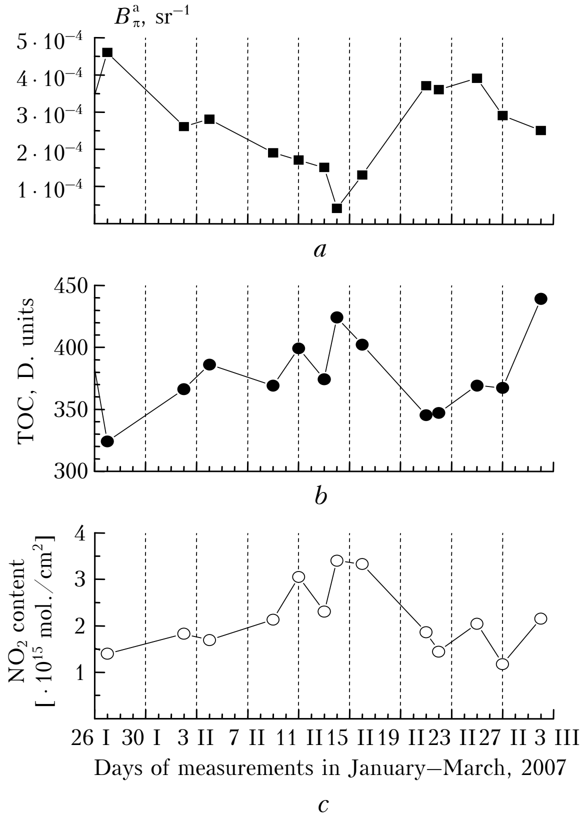

Fig. 4. Time dynamics of variation of B_{π}^{a} (*a*), TOC (*b*), and NO_2 (*c*) late in January – early in March, 2007.

The correlation between TOC or NO_2 content and the content of volcanic aerosol, based on data of Fig. 4, is shown in Fig. 5. The statistically significant negative correlation is observed. The interconnection of aerosol and TOC is determined by the correlation coefficient $R = -0.73$ at a probability density $P = 0.004$. For aerosol and NO_2 content $R = -0.77$ at $P = 0.002$.

The calculations have shown that for the same periods of SA background content in 2000–2006 the similar correlation was not observed and short-term variations of TOC and NO_2 occurred casually under the action of dynamic factor of the atmosphere. The correlation appears, beginning from a definite level of the increased aerosol content. In our case, after the Rabaul eruption the correlation between B_{π}^{a} and TOC is observed at the level of SA content, which is characterized by the values of the scattering ratio $R(H) \sim 1.4$–1.6 in the lower stratosphere and mean values $B_{\pi}^{a} \geq 3 \cdot 10^{-4}$ sr^{-1} (for the background level of

SA content $R(H) \sim 1.1$ and $B_\pi^a \sim 1.5 \cdot 10^{-4}$ sr^{-1}). An important characteristic of volcanic explosion except for the ejection altitude is the amount of the ejected sulfur dioxide, the main source of formation of the sulfuric acid aerosol. At Rabaul eruption 0.125 Mt of SO_2 was ejected,[14] for comparison, at high-power El Chichon volcanic eruption there was 8.1 Mt of SO_2 and at Mt. Pinatubo 17–20 Mt.[11]

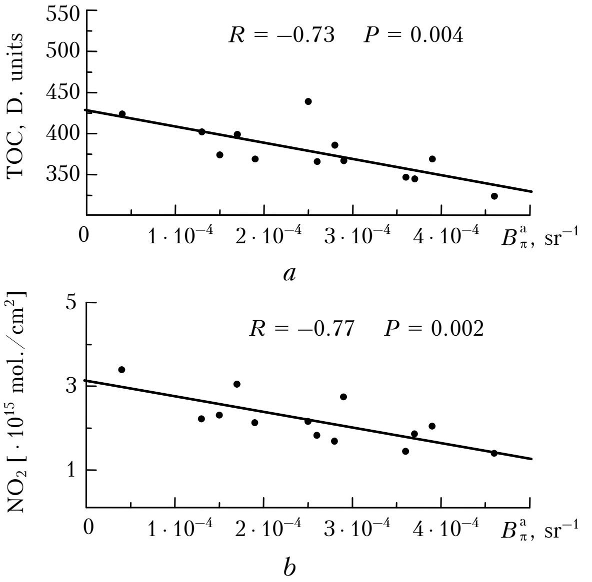

Fig. 5. Correlation between the volcanic aerosol content and TOC (*a*) for NO_2 (*b*) based on data of Fig. 4.

Figure 6 shows the altitude variations of NO_2 content.

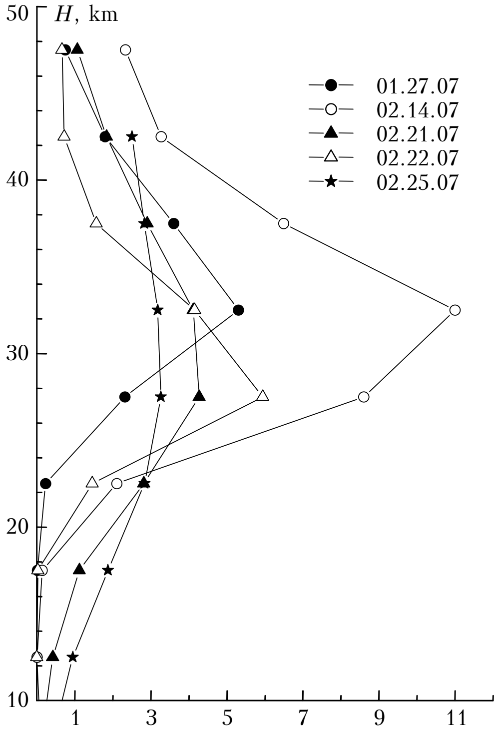

Fig. 6. Altitude profiles of NO_2 content for measurement days.

During measurement days of January 27 and February 21, 22, and 25, when the increased values of SA content were noticed, the decrease of NO_2 content as compared to February 14 was observed, when the B_π^a values were minimal (see Fig. 4). Note that the basic NO_2 layer is localized at altitudes about 20–40 km; at these altitudes the basic variations of NO_2 content occur at variations of SA content. The increased content of volcanic aerosol itself is observed in the lower stratosphere up to altitudes of 25 km (see Fig. 3). The reasons for such altitude variations of NO_2 content will be discussed in Chapter 3.

Thus, even after comparatively weak explosive volcanic eruptions with a short-term increase of SA content the decrease of total ozone and NO_2 content takes place.

3. Discussion of observational results: specificity of ozone cycles in the presence of volcanic aerosol layers

The surface area of sulfuric acid SA after high-power volcanic eruptions increases by several tens of times. After the Pinatubo eruption the specific area of aerosol particle surface almost during 3 years was within 10–30 mm$^2 \cdot$ m^{-3}, while its background typical values are ≤ 1 mm$^2 \cdot$ m^3 [Refs. 2 and 3]. Laboratory experiments, modeling of SA and ozone with inclusion of chemical, radiation and dynamic processes, full-scale measurements of characteristics of SA and gas components of ozone cycles have shown that of primary importance in the ozone balance at the increase of the SA content are the following heterogeneous reactions on the aerosol surface[4,15,16]:

$$N_2O_5 + H_2O \rightarrow 2HNO_3, \quad (1)$$

$$ClONO_2 + H_2O \rightarrow HNO_3 + HOCl. \quad (2)$$

The reaction (1) converts nitrogen from a relatively reactive molecule N_2O_5 to the HNO_3 reservoir with a lifetime of several weeks or months. The decrease of N_2O_5 leads to the decrease of ozone destruction through reducing NO and NO_2 (nitrogen components of catalytic cycle of ozone destruction). At the same time, the ozone destruction is amplified due to the decreased rate of conversion of active chlorine to the reservoir components through reactions with nitrogen oxides. The reaction (1) is almost independent of the temperature and is effective in a wide range of H_2O content in the sulfuric acid aerosol.

The $ClONO_2$ is the basic time reservoir of chlorine in the stratosphere. The reaction (2) converts chlorine from the reservoir to ozone-destructive components as a result of HOCl photolysis. The reaction (2) is more effective at low temperatures, sensitive to the aerosol composition (H_2SO_4:H_2O), and responsible for greater activity of chlorine in the lower stratosphere.

At altitudes higher than 25 km of the ultimate importance is the catalytic cycle of ozone destruction using the nitrogen oxides. As a result of different variations of reaction efficiency (1) and (2) for different conditions of the stratospheric state the decrease of the level of ozone content depends on the latitude, altitude, season, temperature, etc. As a whole, after high-power volcanic eruptions the total decrease of ozone and nitrogen dioxide is observed.

During the initial period after high-power volcanic eruptions (Pinatubo in our case), when aerosol layers and the total increased content of SA are observed up to the altitudes of 30 km, the reduction of ozone and NO_2 contents mainly results from heterogeneous reactions (1) and (2). In case of increased aerosol content in the lower stratosphere after low-power volcanic eruptions (Mt. Rabaul) or during a period of relaxation of aerosol perturbation of the stratosphere after Mt. Pinatubo eruption (spring, 1993) the reaction (2) is of primary importance in the ozone destruction. The reaction (2) is more effective at low temperatures of the lower stratosphere. The NO_2 content increase with simultaneous decrease of HNO_3 content[12] take place due to the decrease of efficiency of the reaction (1), because the stratosphere is cleared of volcanogenic aerosol.

In the stratospheric layers, located higher than the volcanogenic aerosol layer boundary, the radiation backscattering effect of the volcanogenic aerosol layer manifests itself (so-called albedo-effect). The amplification of albedo-effect leads to the increase of radiation field at altitudes of photochemical ozone generation higher than 25 km, with growth of the role of photochemical reactions of ozone destruction by the known Chapman cycle:

$$O_3 + h\nu\ (\lambda < 1100\ \text{nm}) \rightarrow O + O_2, \qquad (3)$$

$$O_3 + O \rightarrow 2O_2, \qquad (4)$$

where $h\nu$ ($\nu = c/\lambda$) is the light quantum energy; h is the Planck constant; c is the velocity of light; λ is the wavelength. The formation of a supplementary atomic oxygen through the reaction (3) leads to amplification of the efficiency of ozone destruction by nitrogen oxides via reactions of the catalytic cycle:

$$NO_2 + O \rightarrow NO + O_2, \qquad (5)$$

$$NO + O_3 \rightarrow NO_2 + O_2, \qquad (6)$$

$$\overline{\text{Total: } O_3 + O \rightarrow 2O_2.}$$

In this case, the increase of O and decrease of O_3 in reactions (5) and (6) result in NO_2 decrease. Finally, the amplification of albedo-effect from the layers of volcanogenic aerosol leads to the increase of the importance of photochemical reactions destructing ozone and NO_2 at altitudes higher than the boundary of the layer of volcanogenic aerosol.

Such characteristics of altitude variations of ozone and NO_2 content are clearly demonstrated in Figs. 2 and 6 under conditions of enhanced content of volcanogenic aerosol up to conventional altitude boundary of a layer about 24 km. In Fig. 2 higher than this boundary (area B), we observe the decrease of the ozone content related to the long-standing mean background content. Figure 6 shows that higher this boundary the decrease of NO_2 content relative to the measurements of February 14, 2007 with the background of SA content is observed. The decrease of ozone and nitrogen dioxide occurs mainly as a result of reactions (3)–(6). Figure 2 shows the decrease of ozone in the layer of increased content of volcanogenic aerosol below 20 km (area A), which takes place mainly due to heterogeneous reaction (2).

Conclusion

After high-power Mt. Pinatubo eruption (June, 1991) the marked ozone depletion (up to 10% TOC decrease) was observed at the Siberian Lidar Station in Tomsk over a period of 5 years.

Since 1996, over a period of background SA content, the values of TOC and total NO_2 content are relatively stable and do not undergo significant long-term variations.

After the Mt. Rabaul eruption (October, 2006) during a period of maximal aerosol stratospheric perturbation in late January – early March, 2007, the statistically significant negative correlation was observed between the variations of SA content and TOC, between the content of SA and NO_2. The interrelation between the SA content and the TOC is determined from the correlation coefficient $R = -0.73$ at the probability density $P = 0.004$. For the content of SA and NO_2: $R = -0.77$ at $P = 0.002$.

In the stratospheric layers with increased content of volcanogenic aerosol the ozone destruction occurs as a result of a preferred effect of heterogeneous ozone-destructing reactions on the enlarged aerosol surface. At altitude higher than the layer of increased aerosol content under the action of the solar radiation scattering effect the above layer enhances the role of photochemical reactions of ozone destruction and reactions of catalytic cycle of ozone destruction by nitrogen oxides.

Note that in recent years the artificial increase of the stratospheric aerosol content for compensation of global warming was proposed,[17] which due to the albedo-effect would decrease the ground temperature. To lower the tropospheric temperature by 1–2°C, it is proposed to eject about 0.6 Mt of aerosol in the stratosphere during 2 years; to do this, it is necessary to burn about 0.2 Mt of sulfur in the stratosphere. In this case no harmful ecological results are expected, including falling sulfur on the Earth's surface. However, it is neglected that the increase of SA content will lead to uncontrollable disarrangement of all chemical balance of the stratosphere and destruction of stratospheric ozone layer, which protects the Earth's biosphere against the short-wave solar UV-radiation. Such artificial increase of stratospheric aerosol content can reduce to zero the appearing stabilization and growing of TOC, including all the efforts and expenses on realization of the Montreal Protocol and subsequent agreements.

Acknowledgements

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