Control of the Dynamics of Tropospheric Ozone through the Stratosphere

P. N. Antokhin and B. D. Belan

V.E. Zuev Institute of Atmospheric Optics, Siberian Branch, Russian Academy of Sciences,
pl. Akademika Zueva 1, Tomsk, 634021 Russia
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Abstract—We consider two possible mechanisms of stratospheric control of the ozone concentration and dynamics in the troposphere. The first mechanism is implemented through the total-ozone-caused modulation of the ultraviolet radiative flux, incoming to the troposphere and initiating therein the photochemical processes. The second mechanism acts through direct transport of stratospheric ozone, which will "trigger" the photolysis and initiate these same processes of ozone generation, but now in the troposphere. Both of these mechanisms of stratospheric ozone control of the near-ground ozone concentration are shown to be apparent near Tomsk. It is noteworthy that the control via ultraviolet flux determines the amplitude modulation, and the transport from the stratosphere to the troposphere drives the temporal modulation.

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INTRODUCTION

Ozone, as a trace gas, plays an important and multisided role in the atmosphere. The stratosphere contains up to 90% of ozone, which is of enormous significance in the protection of the biosphere from hard ultraviolet radiation. Ozone performs a negative function in the troposphere, where it is one of the most toxic gases exerting a depressing effect on biological objects and, in particular, on human beings. Being a most powerful oxidant, ozone causes destruction of many materials. Moreover, with regard to its contribution to the greenhouse effect, tropospheric ozone is ranked fifth behind water vapor, carbon dioxide, methane, and nitrous oxide. All these negative properties of ozone in the troposphere necessitate a detailed study of its spatiotemporal variations and a better understanding of the mechanisms of its generation and sinks.

Ozone scientists had long and firmly believed that the main source supplying ozone to the troposphere is ozone transport from overlying layers and, more specifically, from the stratosphere, where it is generated under the influence of solar ultraviolet radiation [1]. And, it was as late as the 1960s when scientists realized that a considerable amount of ozone may also be formed photochemically, directly from precursor gases [2]. Crutzen and Zimmermann underline [3] that the photolysis of the ozone itself, available in the troposphere, initiates a chain of photochemical reactions, which sometimes lead to smog formation. This latter warrants considering two possible mechanisms behind stratospheric control of the ozone concentration and dynamics in the troposphere: (a) through modulation of ultraviolet radiative flux incoming to the tropo-sphere and initiating photochemical processes there, and (b) through the direct transport of ozone from the stratosphere that will "trigger" photolysis and initiate these same processes of ozone generation, but now in the troposphere.

Manifestation of both these mechanisms near Tomsk is analyzed in this work.

FIRST MECHANISM

Variations in the total ozone (TO) content must be primarily translated to variations in UV radiative influx to the lower troposphere, where ozone is photolyzed and formed under its influence [4]. Madronich [5] considered this process and found that the influx of UV-B radiation not only changes in magnitude, but also changes spectrally: its maximum shifts from 310 toward 315−330 nm. This, in turn, should decrease the photolysis rate, which is maximal at 305−307 nm [2]. The subsequent measurements of UV-B radiation and their comparison with TO variations showed that the interrelation between the two is well discernible on long timescales and not always stable on short timescales [6−8]. Then, a direct comparison was used to show that TO decrease (increase) is accompanied by growth (decrease) of the near-ground ozone concentration (NOC) [9]. A relevant conclusion is that this mechanism of stratospheric control determines the quantity of the "photochemical" addition to ozone concentrations in the troposphere.

This conclusion was first checked by Belan et al. [10] by using the TOR station measurement data on the atmospheric composition near Tomsk [11] and satellite measurement data on TO [ftp://jwocky.gsfc.nasa.gov].
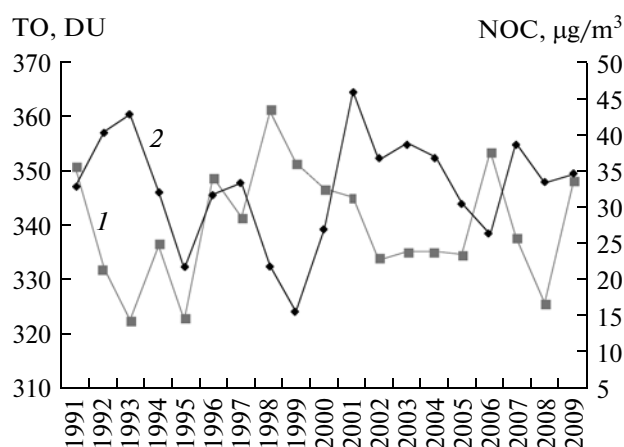
Fig. 1. The annual average total ozone content (curve *1*) and the near-ground ozone concentration (curve *2*) near Tomsk.

Bondarenko et al. [12] and Bazhenov [13] showed that the satellite measurements correlate well with direct TO measurements performed with the help of a M-124 ozonometer. This comparison showed that the TO and near-ground ozone concentration vary in antiphase. The available observation series has become longer (Fig. 1) since the publication of work [10].

From Fig. 1 it can be clearly seen that the TO decrease (increase) is accompanied by growth (decrease) of the near-ground ozone concentration. Six nodes, at which these time series are in antiphase, can be identified over a period of 19 years, with the periods of antiphase occurrences occasionally varying from one year to six years. Evidently, TO plays a leading role in this process.

SECOND MECHANISM

This mechanism essentially consists of ozone transport from the stratosphere to the troposphere in a Brewer–Dobson cell [14]. As Dutsch [15] showed, ozone is mainly produced in the tropical upper stratosphere, with the main ozone source region located between 10° S and 35° N during the northern hemisphere summer and between 38° S and 12° N during the northern hemisphere winter. Ozone thus generated is afterward transported by meridional and descending vertically ordered air motions in the system of the Brewer–Dobson circulation [16, 17].

Fabian et al. [18] were among the first to estimate the parameters of the Brewer–Dobson circulation. They found that the meridional transport velocity is 0.4 cm/s, and the velocity of the descending motion is about 0.1 cm/s. Modern estimates [19, 20] indicate that the velocity shows annual behavior and depends on specific features of the general circulation of the atmosphere. Calculations with the use of models of different degrees of complexity [21–23] try to predict the trend of variations in tropospheric ozone due to variations in the parameters of the Brewer–Dobson circulation.

Still another process of ozone transport from the stratosphere to the troposphere, caused by tropopause "breaking" and folding in the jet stream zone, is the ozone transport through tropopause folds; it was discovered as early as the 1960s, based on radioactivity and back trajectory data [24]. The results of this process were recorded later with the help of aircraft experiments [25] and continue currently with aircraft sensing work [34–36]. Even more information comes from lidars, capable of fixing the phenomenon with finer spatiotemporal resolution [26, 27].

The essence of this phenomenon is that a jet stream is surrounded by spiral circulation which transports ozone from the stratosphere to the troposphere in its descending branch; while on another side of the axis, in its ascending branch, it transports water vapor, aerosol, and other trace gases from the troposphere to the stratosphere [28, 29]. It is noteworthy that the circulation changes its direction along the stream, thus ensuring both balance of the momentum and conservation of thermobaric gradients sustaining the existence of the jet stream [30, 31].

The literature had long mentioned the descending branch of the spiral circulation, in which ozone is transported, seemingly because of the lack of reliable instrumentation for measurement of other constituents of air. At present, researchers have accumulated a significant data which confirm that the ascending branch exists and transports trace gases, namely, water vapor [32, 33], methane, nitrous oxide, etc. [34], from the troposphere to the stratosphere. In discussion of any phenomenon in the atmosphere, it is important to know its frequency of occurrence and power, i.e., the significance of the process as a whole. Such data are collected in work [35], which, in particular, showed that tropopause folds in the Northern Hemisphere are most numerous during December–January, when up to 700 events per month are recorded. The lowest frequency is recorded in June–July and does not exceed 400 events per month. These intrusions are much fewer in the Southern Hemisphere. The frequency of their occurrence is maximal during the cold period from April to June and equals 340–360 events per month; and it is minimal during summer (in December) and equals 260 events per month.

If these two processes of ozone transport from the stratosphere to the troposphere are compared in the systems of the Brewer–Dobson circulation and vertical jet-stream circulation, the ozone flux seems to be largely accounted for by the former. Firstly, this source acts permanently, though changing in intensity with time. Secondly, it encompasses almost the entire globe. In comparison, the vertical circulation around a jet stream develops only in separate regions and
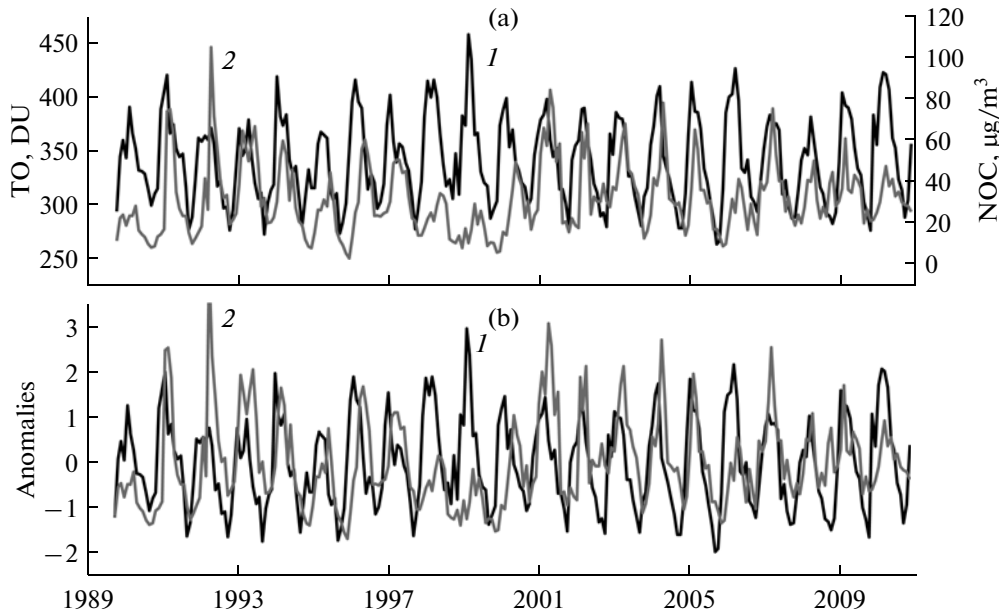
Fig. 2. Long-term monthly average behavior of (a) TO (curve *1*) and NOC (curve *2*) and (b) their anomalies.

changes its direction along the stream: from thermally direct circulation, favoring the intrusion of ozone-rich stratospheric air to the troposphere, to thermally indirect circulation, when ascending (descending) motions occur on the cyclonic (anticyclonic) side of the stream, and the reverse process occurs, i.e., a decrease in ozone in the upper troposphere. Thus, the vertical jet-stream circulation ensures only local ozone transport from the stratosphere to the troposphere. On average, on long timescales and on a net basis globally, the contribution of the vertical jet-stream circulation should vanish because, as dictated by the continuity of the atmosphere, there should be a preservation of balance: the air transported from the stratosphere to the troposphere should be equal in amount to tropospheric air returning to the stratosphere.

The velocity of the descending motions in the Brewer−Dobson circulation system is a few tenths of cm/s; therefore, it is reasonable to consider the emergence of its effect on timescales comparable to time intervals required for the air to descend from ozonopause altitude, which is close to the tropopause level [36, 37], i.e., on timescales from a few days to two weeks. No turbulent transport from tropopause level to the atmospheric boundary layer can occur in most of the standard situations [38]. The analysis will be based on daily and monthly average ozone concentrations, measured at the TOR station during the period from 1990 to 2011 [11, 12], and on the total ozone data for this same region, available on the NASA website (ftp://jwocky.gsfc.nasa.gov). Figure 2 shows the long-term behavior of TO and NOC and their anomalies.

To calculate the anomalies, the observation time series were processed to remove the average values,

341.5 DU and 32.1 $\mu g/m^3$, respectively. The results were normalized to standard errors of these time series.

From Fig. 2a it can be seen that, in principle, the time dynamics of the near-ground ozone concentration matches, with some delay, the TO variations, although the amplitudes of their oscillations differ during separate time periods. Moreover, the plots indicate that the delay period may vary from one to a few months. After conversion to relative units (Fig. 2b), it becomes clear that the time oscillations are comparable. It is noteworthy that TO variations are prior to NOC dynamics, suggesting that TO variations are prior to those of its tropospheric component.

We will use time series analysis methods to obtain information on the dynamics of delay in these time series. Correlation methods [39−41], spectral Fourier analysis [42, 43], and wavelet transforms [44, 45] are most often used at present to study the time lag between atmospheric processes. In contrast to correlation methods, the spectral methods can be used not only to estimate the time lag, but also to study the time behavior of this lag.

ANALYSIS RESULTS AND DISCUSSION

The time series of the daily average TO and NOC, for which the calculation was made, had the length of 4834 pairs. Preliminarily, the initial data were examined for spikes, and then were processes to remove suspicious measurements. This was done using the median criterion [40, 41].
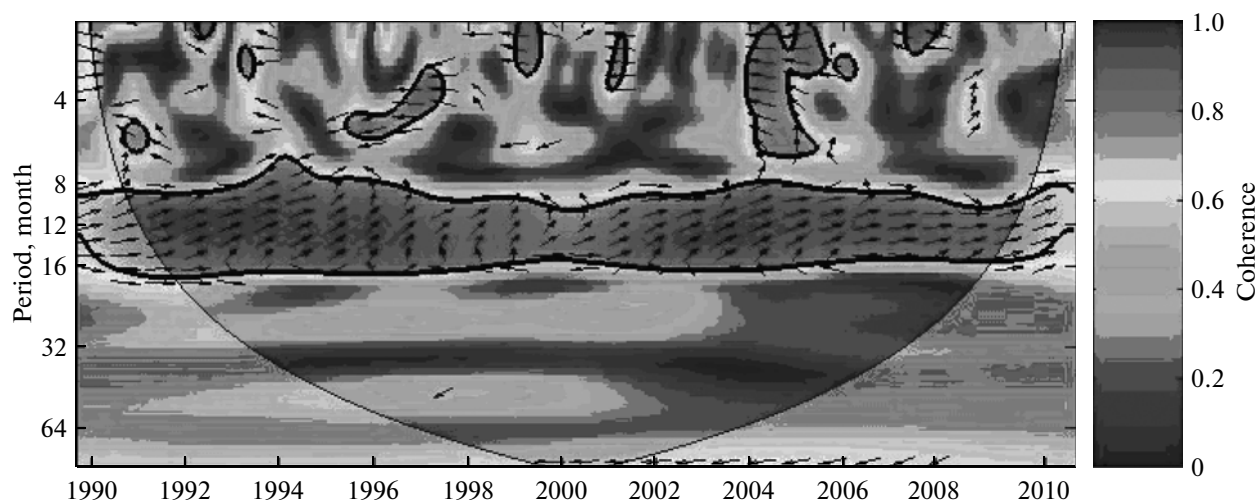
Fig. 3. Wavelet spectrum of coherence of NOC and TO.

The coefficient of the pair correlation r between time series was 0.317 ± 0.013. The Student–Fisher test showed that this coefficient was significant at 0.95 level. The coefficient of the pair correlation confirms that TO and NOC variations correlate at zero time lag. Therefore, if r is calculated for a time lag close to 1–2 months, its values should increase. For this, we calculated the cross-correlation function and obtained the following result. The cross-correlation function peaks not at zero lag, but rather at a lag of 28 days, and is equal to 0.455 ± 0.111. As compared to zero lag, the coefficient of the pair correlation has increased by 30%. It can be preliminarily concluded that NOC is delayed from the TO by approximately 1 month. When a 1-month lag between annual variations in TO and TOC is specified, the coefficient of the pair correlation increases to 0.94 ± 0.10.

At the next stage, we calculated the Fourier cross-spectrum of the monthly average TO and NOC. The time series consisted of 255 pairs, and the time period was from 1989 to 2010. The phase cross-spectrum
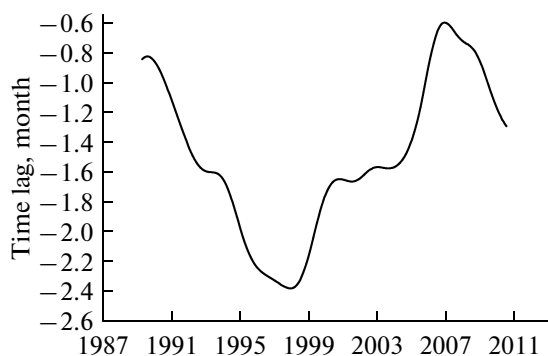

Fig. 4. Variations in the phase of the cross-wavelet spectrum for 12-month period.

exhibits the phase difference between the processes for the time period of 1.94 months; that is, TO variations are approximately two months ahead of NOC variations.

Importantly, the Fourier transform presumes that the time phase difference between processes does not change. When TO and NOC time variations are modulated, the Fourier transform provides an average over the entire time period. In this regard, we now proceed to cross-wavelet analysis, elucidating the joint behavior of the processes in time and during a certain time period.

Figure 3 shows the cross-spectrum of coherence of NOC and TO. In this figure we clearly see a group of periods which corresponds to the annual behavior. This group is statistically significant throughout the time interval addressed. In this band of periods, the arrows point from left to right and upward, indicating that TO and NOC oscillate in-phase, but with a phase shift.

The phase shift gradually increases starting from 1995, reaches a maximum in 1999, then starts to decrease, and in 2002 reaches the value close to that already taken in 1995. We will plot separately the variations in the joint phase for a 1-year period of oscillations, to study in more detail the behavior of the time lag.

The phase difference between TO and NOC for this period is shown in Fig. 4; here, the minus sign indicates that NOC variations lags behind TO variations.

It can be seen that the delay time of variations in NOC relative to TO may range from 0.6 to 2.4 months in different years. The maximum (minimum) time lag of 2.4 months (0.6 months) was in 1998 (2007); the average is 1.4 months (about 42 days).

Therefore, considering that the average ozonopause altitude is 10–11 km [36, 37], the average velocity of transport of stratospherically derived ozone to
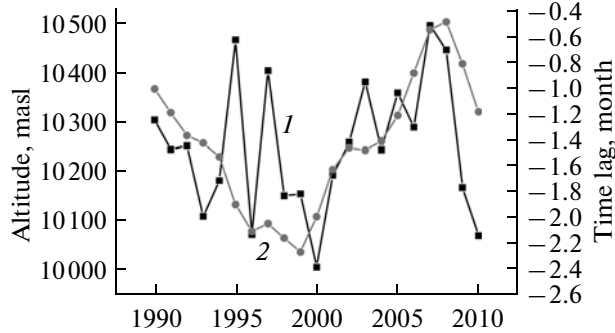
Fig. 5. Annual average tropopause altitude over Novosibirsk (curve *1*) and time lag between TO and NOC (curve *2*).

the near-ground atmospheric layer will vary from 0.64−0.71 to 0.16−0.18 cm/s. This estimate spans a somewhat wider range than reported in [18]: 0.1−0.4 cm/s. It also falls within the range of velocities of ozone deposition onto the underlying surface: 0.01−1.0 cm/s [46]. Thus, even in the absence of photochemical ozone generation in the troposphere, the net ozone balance will not be disturbed. Hence, there must be a correlation between the ozonopause altitude and the near-ground ozone concentration.

There are no ozone monitoring stations in our region; therefore, we will take into consideration the fact that tropopause and ozonopause are located at nearly the same altitudes and interrelated [36, 37]. The tropopause altitude was analyzed using radio sensing data available on http://weather.uwyo.edu for two sites of aerological observations closest to Tomsk: Kolpashevo and Novosibirsk.

Figure 5 compares the annual average tropopause altitudes (Novosibirsk) and time lag between the near-ground ozone concentration and total ozone content.

It can be seen that the time lag decreases (increases) with increasing (decreasing) tropopause altitude. The correlation coefficient between them is 0.42 at a 0.99 significance level. The differences between the curves of long-term variations are well pronounced in 1995 and 1997, which requires extra study.

At first glance, this is a paradoxical result because, as would be expected, the lower the tropopause and ozonopause altitudes, the shorter the time required for stratospheric ozone to reach the near-ground air layer while descending. However, all takes its rightful place if we recall that the tropopause is higher over an anticyclone and lower over a cyclone [47, 48]. It is well known that descending motions prevail in anticyclones, with velocities of a few cm/s [48]; conversely, ascending motions are more intense in cyclones than in anticyclones. Therefore, the descending motions will be added to the descending branch of the Brewer−Dobson circulation in the case of a high tropopause

under anticyclonic conditions. Conversely, ascending motions will be subtracted from the descending branch of the Brewer−Dobson circulation in the case of a low tropopause under cyclonic conditions. As a result, longer time periods will be required for the stratospheric ozone to be supplied to the near-ground air layer, and this supply will even be screened out in case of intense ascending motions.

Summarizing the work as a whole, we note that both mechanisms of the stratospheric ozone control of the near-ground ozone concentration appear near Tomsk. It is noteworthy that the control through the ultraviolet flux determines the amplitude modulation, and the transport from the stratosphere to the troposphere drives the temporal modulation.

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