

Article

Frequency Dependence of the Correlation between Ozone and Temperature Oscillations in the Middle Atmosphere

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Abstract: This study investigates the frequency dependence of the correlation or anticorrelation of ozone and temperature in the middle atmosphere. The anticorrelation of ozone and temperature plays a role for a possible super recovery of upper stratospheric ozone in the presence of man-made cooling of the middle atmosphere due to increasing carbon dioxide emissions. The correlation between lower stratospheric ozone and temperature indicates the dependence of lower stratospheric temperature trends on the ozone evolution in addition to greenhouse gas emissions. Ozone and temperature measurements of the microwave limb sounder (MLS) on the satellite Aura from 2004 to 2021 are utilized for Bern (46.95° N, 7.44° E) at middle latitudes and for the equator region. The time series are bandpass filtered for periods from 2 days to 5 years. The correlation coefficient depends on the period of the oscillation in temperature and ozone. The strongest correlation and anticorrelation are found for the annual oscillation. The anticorrelation between ozone and temperature in the upper stratosphere is about -0.7 at a period of two days and -0.99 at a period of one year. Thus, the temperature dependence of the ozone reaction rates also leads to an anticorrelation of ozone and temperature at short periods so that ozone can be considered as a tracer of planetary waves. At the equator, a dominant semiannual oscillation and an 11 year solar cycle are found for nighttime ozone in the upper mesosphere. The semiannual oscillation (SAO) in ozone and temperature shows a strong correlation indicating a dynamical control of the ozone SAO in the upper mesosphere. The SAO in the equatorial nighttime values of ozone and temperature is possibly due to a semiannual modulation of vertical advection by the diurnal tide.

Keywords: ozone; temperature; oscillations; annual oscillation; semiannual oscillation; correlation; middle atmosphere; Aura/MLS



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1. Introduction

Ozone and temperature are linked in the middle atmosphere. The positive vertical gradient of temperature in the stratosphere is a consequence of the absorption of solar ultraviolet radiation by stratospheric ozone, which heats the stratosphere. The sources and sinks of odd oxygen (atomic oxygen and ozone) are described by the Chapman cycle [1]:



where M represents a collision partner that is inert. In the lower and middle stratosphere, the atomic oxygen that is released by photodissociation of ozone immediately recombines with molecular oxygen to again form ozone.

Trends of lower stratospheric temperature partly depend on trends of lower stratospheric ozone. It was found that the increase in stratospheric ozone has decelerated the

stratospheric cooling since the late 1990s [2]. A correlation analysis showed that the lower stratospheric temperature is mainly regulated by ozone changes [2]. Observations show that the ozone decrease in the stratosphere above Antarctica is correlated with a stratospheric temperature decrease in the years before 2000 [3]. Later (2000–2014), the Antarctic ozone trend becomes positive and also the temperature increases. This mirrored pattern in the changed trends before and after 2000 indicates the healing of the Antarctic ozone hole, which was generated by man-made emissions of chlorofluorocarbons (CFCs). Radiative calculations indicated that ozone increases have contributed to Antarctic warming of the lower stratosphere over 2000–2014, but dynamical changes that are likely due to internal variability over this relatively short period also appear to be important [3]. Solomon et al. concludes that the coupling of dynamics, chemistry, and radiation is important for the full understanding of the causes of observed stratospheric ozone and temperature changes [3].

A correlation between large-scale ozone and temperature variations in the tropical lower stratosphere across a wide range of timescales was found by [4]. Their simple model approximately explained the observed (T/O_3) amplitude and phase relationships, including sensitivity to timescale and altitude, and highlighted distinct balances for “fast” variations (periods < 150 days, controlled by transport across background vertical gradients) and “slow” coupling (seasonal and interannual variations, controlled by radiative balances).

Alternating advection of ozone-rich, warm air parcels and ozone-poor, cold air parcels explains the correlation of ozone and temperature in planetary wave-like oscillations in the lower stratosphere in winter observed above a mid-latitude station [5]. At mid-latitudes, the planetary wave-like oscillations are strongest during the winter season in the lower stratosphere. The cold polar vortex contains ozone-poor air. Planetary waves can shift the polar vortex to a mid-latitude station, so that a correlation of low ozone and low temperature would be recorded. If the polar vortex goes back to high latitudes, a coincident increase in lower stratospheric ozone and temperature would be recorded by a mid-latitude station. At Bern, we found periods of about 20 days in the stratospheric ozone oscillations during winter [6,7]. These ozone oscillations are related to planetary waves and sometimes to sudden stratospheric warmings, which can also shift or deform the stratospheric polar vortex [5,8–11]. The ozone oscillations would be reduced or would vanish in the zonal mean at mid-latitudes. Thus, in the present study we look for correlations of ozone and temperature oscillations above the mid-latitude station Bern, which is most familiar for us.

In the upper stratosphere, the correlation changes into an anticorrelation of ozone and temperature since the ozone reaction rate coefficients of Equations (2) and (4) are temperature-dependent. The reaction rate coefficient of Equation (2) decreases with an increase in temperature [12–14]. Thus, the ozone generation is lower for higher temperatures. In addition, the ozone decrease via Equation (4) is enhanced when the temperature increases [13–15]. Thus, the reactions in Equations (2) and (4) are both leading to less ozone for high temperatures so that an anticorrelation of ozone and temperature exist in the upper stratosphere. Generally, the ozone distribution is controlled by dynamical processes in the lower stratosphere since the lifetime of odd oxygen (atomic oxygen and ozone) is of weeks to months while the upper stratospheric ozone distribution is controlled by photochemical processes because of the short lifetime of odd oxygen in the upper stratosphere, which is of the order of hours [13].

Stratospheric cooling due to increasing greenhouse gas emissions [16] may induce a super recovery of ozone in the upper stratosphere [17]. Model simulations indicated that the sensitivity coefficient of the ozone response to temperature decreased as chlorine increased in the stratosphere [18]. Thus, simultaneous measurements of ozone and temperature are valuable to estimate in how far the ozone increase can be attributed to the decrease in chlorine in the upper stratosphere after the ban of chlorofluorocarbons emissions by the Montreal Protocol in 1987 [18]. Further, an indirect radiative effect (cooling) due to short-lived halogens should now be incorporated into climate models to provide a more realistic natural baseline of Earth’s climate system [19].

In the upper mesosphere, the upwelling and downwelling of the secondary ozone layer that appears only during nighttime generate a correlation between temperature and nighttime ozone. The upwelling is associated with adiabatic cooling and vertical advection of ozone-poor air from below while the downwelling is associated with adiabatic heating and vertical advection of ozone-rich air from above. Thus, the nighttime ozone fluctuations in the mesopause region are under dynamical control [20,21]. Nighttime ozone in the upper mesosphere observed by the microwave limb sounder on the satellite Aura (Aura/MLS) also showed an 11-year oscillation that is in phase with the solar cycle [20].

The annual oscillation (AO) belongs to the natural oscillations in stratospheric ozone that were observed at northern mid-latitudes [22]. In the upper stratosphere, ozone is decreased during summer due to the higher temperature. Between 4 and 25 hPa, ozone is increased in summer, which is due to enhanced production of odd oxygen in summer due to enhanced solar short wave radiation. The ozone AO reaches an amplitude of about 1.1 ppmv (or 16%) at Bern in Switzerland [22]. Below 30 hPa, a further change of the phase of the ozone AO occurred, which was not discussed by [22].

The semiannual oscillation (SAO) mainly occurs in the tropics, partly because the Sun traverses the equator two times per year at equinox. In the stratosphere, it was found that the amplitudes of the temperature and ozone SAO are maximal at the equator [23]. The ozone and the temperature SAO are anticorrelated in the upper stratosphere and correlated in the lower stratosphere. The phase structure reveals that the temperature SAO descends faster than the ozone SAO [23].

The frequency dependence of the correlation between ozone and temperature oscillations has not been investigated yet. It is an open question if the correlation coefficients for high frequency fluctuations are similar to those of low frequency fluctuations of ozone and temperature. This knowledge is important for using ozone as a tracer of planetary waves and for the assessment of trends in ozone and temperature. Some experiences with other geophysical datasets would suggest that high frequency oscillations of two time series are uncorrelated while their low frequency oscillations may follow a seasonal cycle and would be correlated. The coincident observations of ozone and temperature in the middle atmosphere by the satellite instrument Aura/MLS offer a great opportunity to investigate the correlation of ozone and temperature oscillations at different places. In the following, we focus on a place at mid-latitudes in order to investigate planetary-wave-like oscillations, and we analyze the equatorial middle atmosphere, which is interesting because of the strong quasi-biennial oscillation and the semiannual oscillation [24,25].

Section 2 describes the dataset and the data analysis of the Aura/MLS ozone and temperature observations. Section 3 shows the results at mid-latitudes (Bern and its surrounding). Section 4 shows the results in the equatorial middle atmosphere (SAO and 11 year oscillation).

2. Dataset and Data Analysis

2.1. Dataset

The present study uses ozone and temperature profiles from the Microwave Limb Sounder (MLS) on the NASA satellite Aura, which was launched in 2004 [26]. Aura has a sun-synchronous orbit with two overpasses around noon and midnight at a given place per day. Level 2 data of the retrieval version 5 are analyzed in the following, and the data screening described by [27] was applied. The vertical range of the profiles, used here, is from 260 hPa to 0.001 hPa. The accuracy of the temperature profiles is about 1 K in the stratosphere and about 3 K in the mesosphere [28]. The accuracy of the ozone profiles changes from about 5–10% in the stratosphere to about 100% in the mesosphere [27]. In spite of the large error at upper altitudes, the ozone profiles contain valuable information about the secondary ozone layer, which appears during nighttime around the mesopause [20].

The study focuses on two regions. Firstly, atmospheric profiles are selected in a surrounding of Bern (46.95° N, 7.44° E). The surrounding is $\pm 3^\circ$ in latitude and $\pm 10^\circ$ in longitude around Bern. The restriction to a certain place at mid-latitudes is useful for the

study of regional ozone and temperature variations due to planetary wave-like oscillations and movements of the polar vortex. A zonal mean would smooth out many regional variations. Secondly, the equatorial region is selected. Here, all longitudes are considered in the average, and a latitude range of $\pm 2^\circ$ around the equator is taken. The time interval of the selected Aura/MLS data is from June 2004 to December 2021.

2.2. Data Analysis

The frequency dependence of the ozone and temperature series are analyzed by means of bandpass filtering. The time series are filtered with a digital non-recursive, finite impulse response bandpass filter. Zero-phase filtering is ensured by processing the time series in forward and reverse directions. A Hamming window has been selected for the filter. The number of filter coefficients corresponds to a time window of three times the central period, so that the bandpass filter has a fast response time to temporal changes in the data series. The variable choice of the filter order permits the analysis of wave trains with a resolution that matches their scale. The bandpass cut off frequencies are at $f_c = f_p \pm 10\% f_p$, where f_c is the cut off frequency and f_p is the central frequency. Further details about the bandpass filtering are provided by [6]. The correlation coefficients of the filtered ozone and temperature series are well determined because of the long time interval of about 17 years. We only consider statistically significant correlations with p -values less than 0.05. For visualization of the long period oscillations of the solar cycle, the ozone time series were filtered by a 5 year lowpass filter with related characteristics as the described bandpass filter. The mean seasonal behavior of the time series is obtained by sorting the data for the day of the year (DOY) and taking the mean.

3. Results

3.1. Results around Bern at Mid-Latitudes

Considering all the ozone and temperature data pairs observed by Aura/MLS around Bern, a scatter plot is drawn in Figure 1 for the upper stratosphere at the pressure level $p = 2.15$ hPa. The straight line has a gradient of $\Delta O_3/T = -0.068$ ppmv/K and an anticorrelation of $r = -0.92$. This anticorrelation is expected for the upper stratosphere. The straight line fit is a good choice, though Stolarski et al. investigate the relation between logarithmus of ozone concentration versus inverse of temperature, which is theoretically more justified [18]. However, in the present study, the simple gradients $\Delta O_3/T$ and correlations T and O_3 are computed since we want to have the same data analysis procedure at all heights in the middle atmosphere.

Next, the correlation coefficient of ozone and temperature is computed at all altitudes and shown by the black curve in Figure 2. Nighttime ozone data are used for a better interpretation in the mesosphere. It is evident that there is a change between height regions of correlation (lower stratosphere, mesosphere) and anticorrelation (upper stratosphere) whereby the areas of correlation are due to a dynamical control of ozone and the area of anticorrelation is related to the photochemical control of ozone. Dynamical control means that transport processes of ozone-rich air in the lower stratosphere (usually from the south of Bern) or ozone-poor air from the polar region induce correlated temporal variations in the ozone and temperature time series at Bern. It is often observed at Bern that a sudden stratospheric warming can shift the polar vortex towards Bern so that suddenly ozone-poor and cold air from the polar vortex is above Bern [5,8,29,30]. Moreover, a double tropopause at mid-latitudes can indicate an exchange of stratospheric ozone-rich air and tropospheric ozone-poor air [31]. Of course, the correlation is not always present. There are exceptions of the rule that ozone-rich air is warmer than ozone-poor air in the lower stratosphere. For example, in late winter and spring, it happens that ozone-rich air is downwelling from the middle stratosphere into the lower stratosphere at high latitudes. In this case, the polar stratospheric air that is transported to mid-latitudes is ozone-rich but relatively cool because of less solar radiation in the polar region. According to [13], the regions of dynamical and photochemical control are related to the lifetime of odd oxygen. The

region of photochemical control agrees with the region of anticorrelation of temperature and ozone in Figure 2. The anticorrelation is due to the temperature-dependent ozone photochemistry in the upper stratosphere where the lifetime of odd oxygen is about 1 h, as already explained in the introduction.

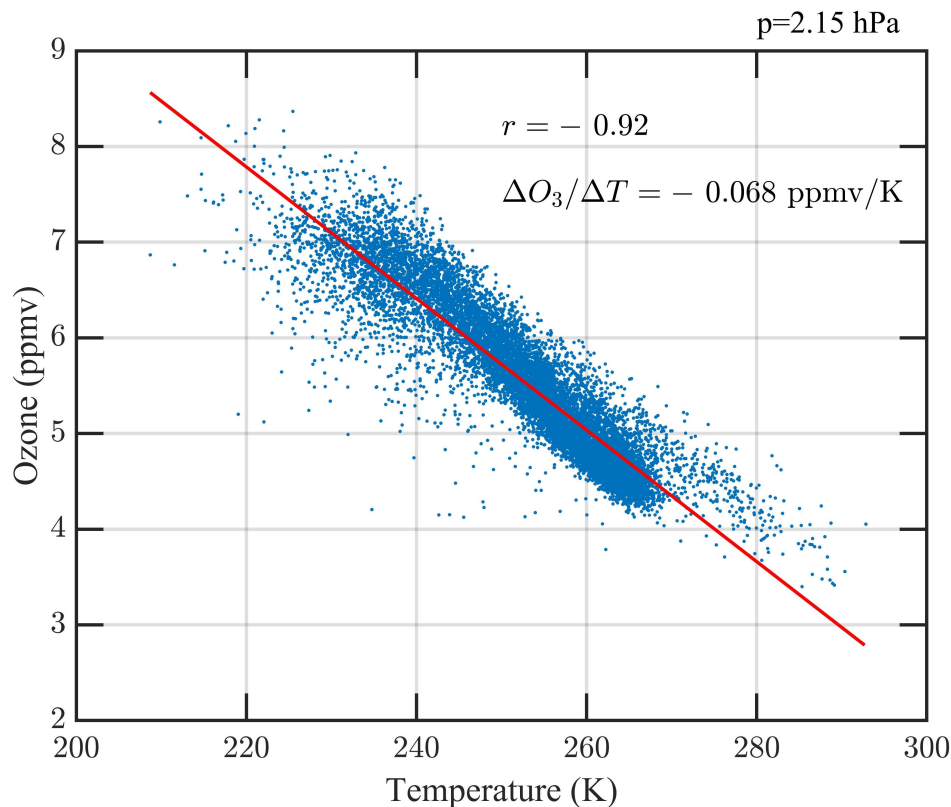


Figure 1. Scatter plot of ozone versus temperature above Bern observed by Aura/MLS in the upper stratosphere ($p = 2.15 \text{ hPa}$) above Bern from 2004 to 2021.

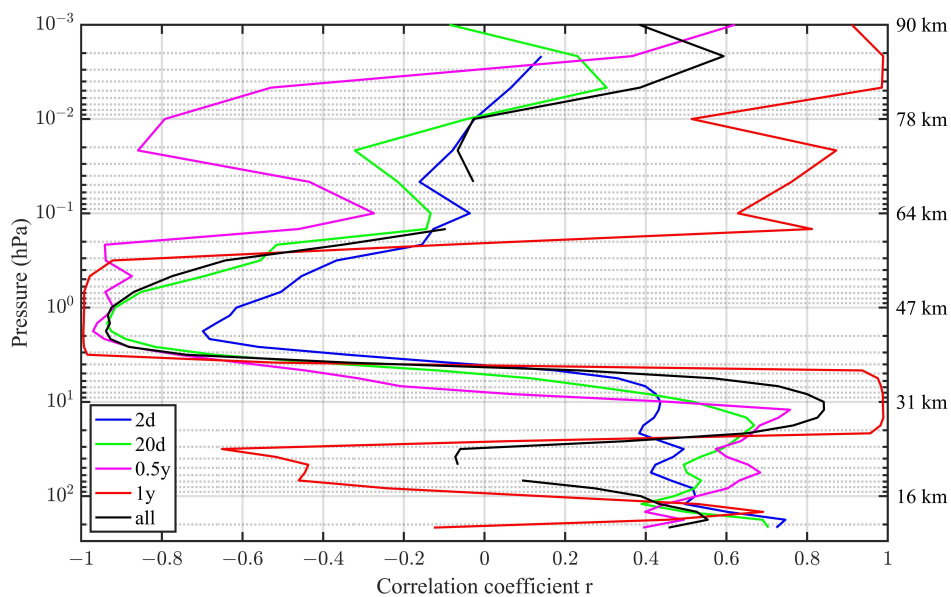


Figure 2. Correlation of ozone and temperature (black). The other colors are for the correlation profiles of the bandpass filtered oscillations of ozone and temperature. The bandpass periods are 2 days, 20 days, 0.5 year, and 1 year. The diagram shows the statistically significant correlation coefficients with p -values less than 0.05.

The correlation coefficient can be also calculated for bandpass filtered oscillations of ozone and temperature. Figure 2 shows the results for the 2 day oscillation, 20 day oscillation, SAO and AO. The correlation of the AO reaches the highest values, almost 1 and -1 . It is also evident that the correlation of the AO turns into an anticorrelation at about 20 km height, which is different to the periods of 2 days, 20 days and 0.5 years.

The anticorrelation in the upper stratosphere also occurs at the short period of 2 days but a bit reduced. Some authors supposed that photochemistry might be too slow for an anticorrelation at short periods, but they did not quantify this statement [5]. Considering the lifetime of odd oxygen in the upper stratosphere, which is about 1 h, planetary wave-like oscillations should occur in ozone. It would be interesting if the anticorrelation in the upper stratosphere is also valid for periods of less than 2 days. The coarse temporal sampling of Aura/MLS at a given place does not permit this investigation. Here, the coincident observations of a temperature and ozone radiometer at Bern may reveal more [32–34].

The gradients $\Delta O_3/T$ are shown in Figure 3 as function of pressure level. The gradients are switching between positive values (correlation) and negative values (anticorrelation). The gradient of the SAO becomes strong at the mesopause, which is due to a strong ozone SAO at upper mesospheric altitudes.

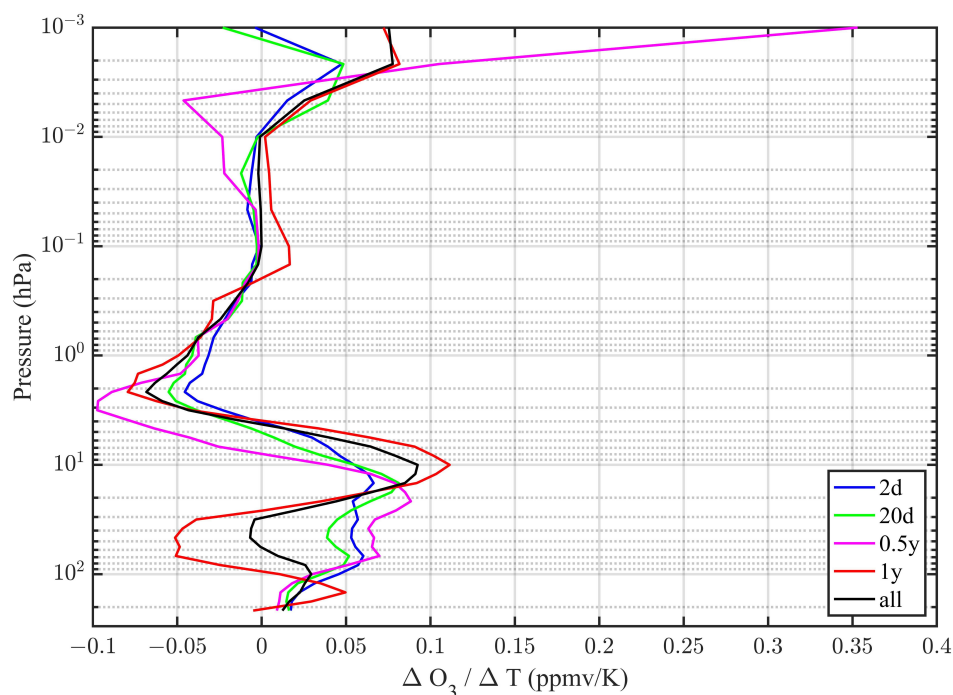


Figure 3. Gradient of the ozone versus temperature linear regression line. Black is for the unfiltered series. The other colors are for the gradients of the bandpass filtered oscillations of ozone and temperature. The bandpass periods are 2 days, 20 days, 0.5 years, and 1 year.

The spectra of r and $\Delta O_3/T$ are shown in Figure 4 in the upper stratosphere ($p = 2.15$ hPa). The anticorrelation of ozone temperature is a bit reduced at shorter periods. This might be due to more atmospheric noise at shorter periods. However, one can say that the anticorrelation also exists at a period of 2 days so that ozone can be used as a tracer of short period temperature waves in the upper stratosphere. As expected, the anticorrelation is strongest at a period of about 1 year. There is an unexpected reduction of the anticorrelation at a period of about 1500 days (ca. 4 years). Thus, it remains unclear if long period oscillations or trends of ozone and temperature have a strong anticorrelation or not.

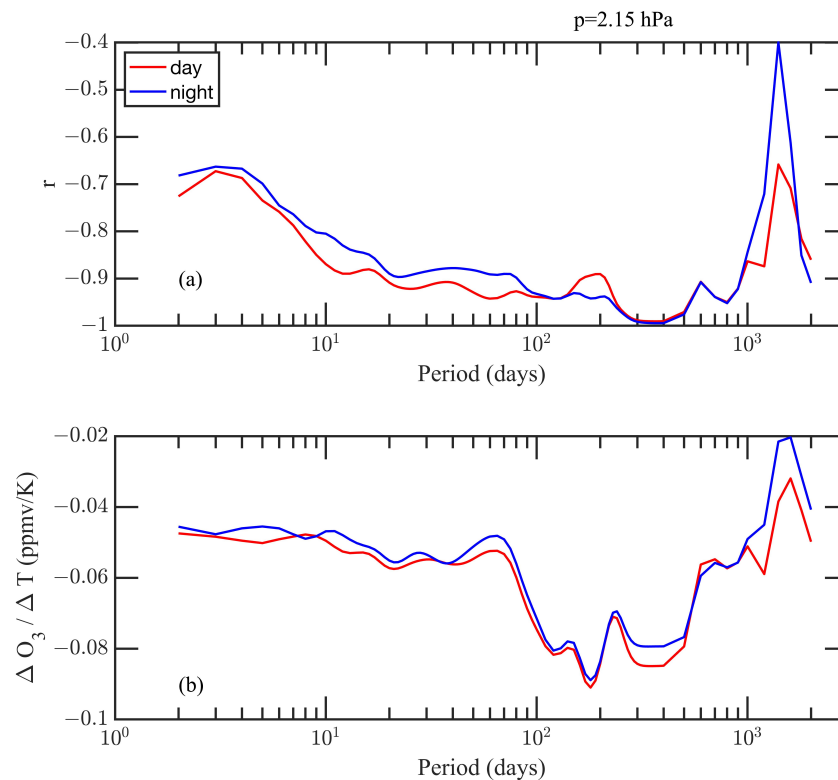


Figure 4. Frequency dependence of the correlation r (a) and the gradient $\Delta O_3/T$ (b) for daytime (red) and nighttime (blue) ozone in the upper stratosphere ($p = 2.15$ hPa).

The spectra of r and $\Delta O_3/T$ are shown in Figure 5 in the lower stratosphere ($p = 38.31$ hPa). There is a positive correlation of about 0.5 for all periods from 2 days to an half year. This correlation can be explained by the fact that ozone-rich temperature has usually a higher temperature in the lower stratosphere. In the spectrum region of the annual oscillation the correlation switches to an anticorrelation of about $r = -0.5$. The reason could be related to the maximum of total column ozone in the winter polar region and to the subsequent transport of polar, cold ozone-rich air to mid-latitudes in spring [35]. This annual transport of ozone may induce an anticorrelation of ozone and temperature observed in the lower stratosphere above a mid-latitude station. At lower periods, the highest correlation of about 0.8 is reached at a period of about 4 years so that it is likely that long-term trends of ozone and temperature are correlated. It is reasonable that increasing ozone values lead to higher temperatures in the lower stratosphere. As expected, there is no significant difference between the spectral curves of daytime and nighttime ozone in the stratosphere.

The close relationship between the annual oscillations in temperature and ozone is shown in Figure 6. It is surprising how fast the correlation between ozone and temperature turns into an anticorrelation at pressure levels above 4 hPa. This phase change is not so rapid at the equator (not shown here). In the lower stratosphere (below 30 hPa) there is a phase reversal of the ozone AO with positive values in late winter. In the upper mesosphere (beyond 0.01 hPa), there is a clear correlation between ozone and temperature with negative values in summer. This is due to the gravity-wave-induced upwelling in summer, which leads to adiabatic cooling. The nighttime ozone is under dynamical control and ozone-poor air from lower altitudes is vertically advected into the upper mesosphere during summer. There is a region in the lower mesosphere from 0.2 hPa to 0.01 hPa where ozone does not show a significant annual oscillation.

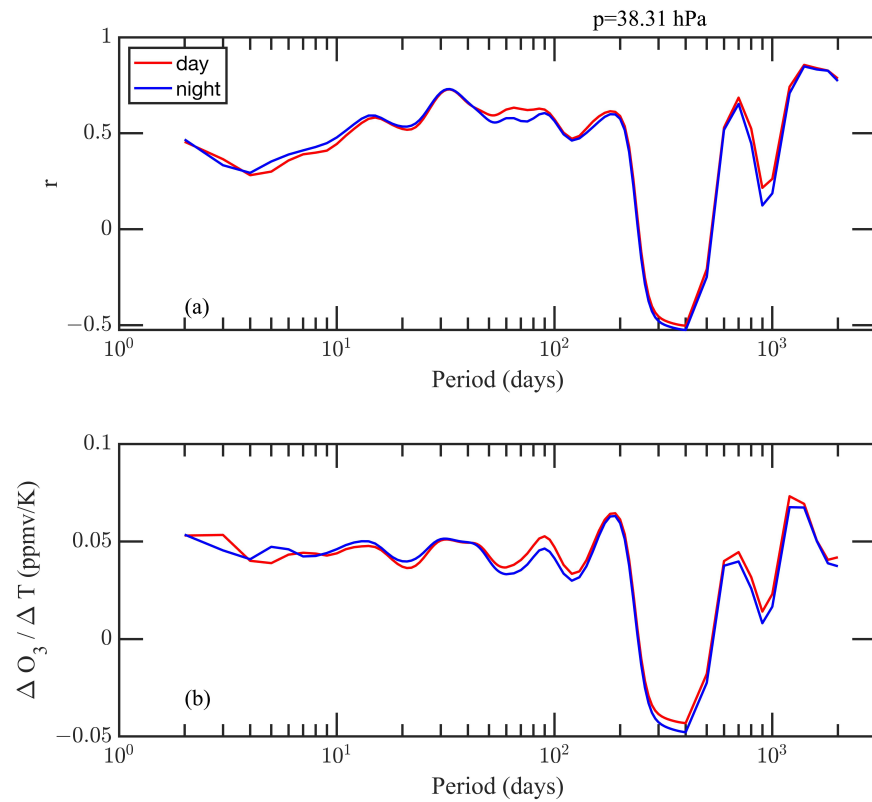


Figure 5. Frequency dependence of the correlation r (a) and the gradient $\Delta O_3 / \Delta T$ (b) for daytime (red) and nighttime (blue) ozone in the lower stratosphere ($p = 38.31$ hPa).

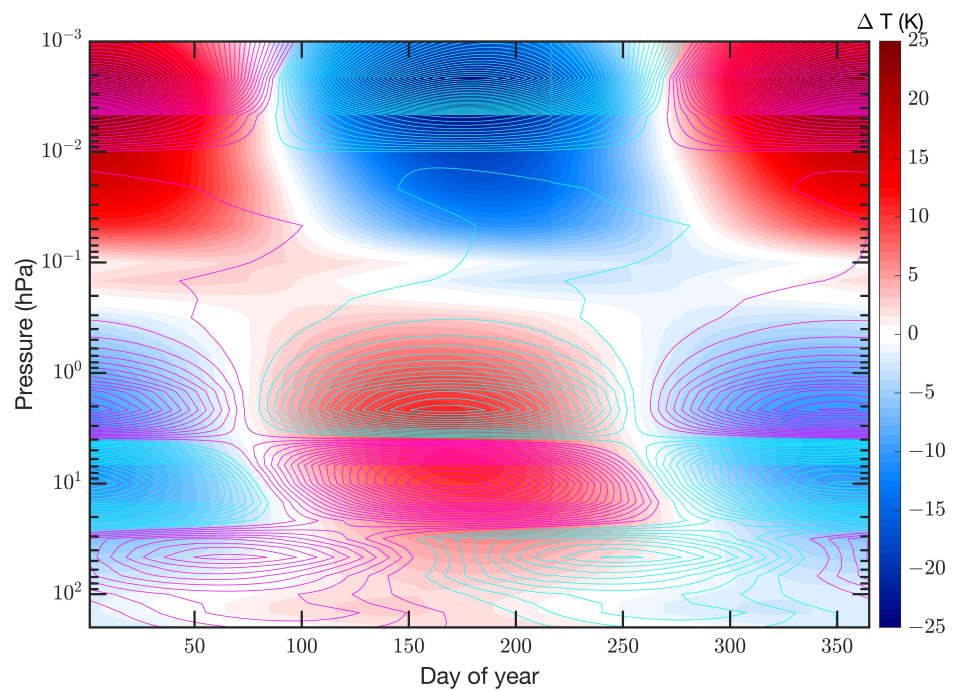


Figure 6. Climatology of the annual oscillation in nighttime temperature (color shading) and nighttime ozone (contour lines with spacing of 0.05 ppmv. Cyan for negative values and magenta for positive values).

3.2. Results at the Equator

It is certainly interesting to investigate the ozone and the temperature semiannual oscillations at the equator. The climatology of the SAOs in temperature and ozone is shown in Figure 7. It is evident that the SAO has a phase progression below 0.01 hPa. Between 1 hPa and 30 hPa, the phase progression of the ozone SAO is stronger than the phase progression of the temperature SAO. Fadnavis and Beig [23] reported this in the opposite manner, but maybe they meant the vertical steepness of the phase fronts (and not the phase progression) of the SAOs. As in the case of the AO at mid-latitudes, there is a region in the lower mesosphere (0.5 hPa to 0.01 hPa) where the ozone oscillation is small while the temperature SAO is maximal. Thus, the photochemical control does not transfer the temperature SAO into the ozone SAO here. Beyond 0.01 hPa, there is a dynamical control of nighttime ozone leading to a strong correlation of the temperature and ozone SAO. The mesospheric SAO in the nighttime values is mainly due to the strong semiannual modulation of the diurnal tide at the equator, which shows maximal downwelling during nighttime in April and October [36].

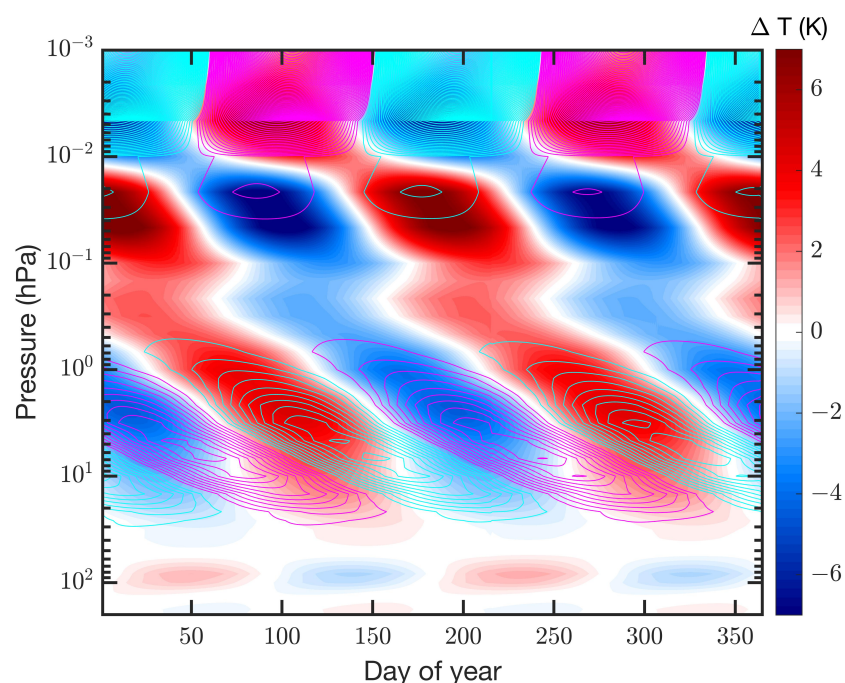


Figure 7. Climatology of the semiannual oscillation in nighttime temperature (color shading) and nighttime ozone. (Contour lines with spacing of 0.05 ppmv. Cyan for negative values and magenta for positive values).

Using the global Aura/MLS observations during nighttime, the signatures of the strong downwelling in nighttime ozone and nighttime temperature at $p = 0.0046$ hPa in the upper mesosphere in April (2005 to 2021) are shown in Figure 8. The results are similar to the results of [36] obtained from TIMED/SABER observations of atomic oxygen and temperature. The retrieval of atomic oxygen was described in [37]. Atomic oxygen and ozone in the mesosphere are in a photochemical equilibrium, and the detailed discussion of the effect of vertical advection of atomic oxygen by the diurnal tide in the equatorial mesosphere [36] can be applied to nighttime ozone. The enhanced values of ozone and temperature in the Southern polar region are due to downwelling by the residual mean meridional circulation.

The signal of the 11 year solar cycle in nighttime ozone at the mesopause was investigated by [20] using Aura/MLS observations. Since the Aura time series is not long enough for bandpass filtering, we apply here a 5-year-lowpass to the equatorial nighttime ozone data in order to emphasize the periods longer than 5 years (Figure 9). The lowpass ozone

series are subtracted by the mean nighttime ozone value at each pressure level. In the upper mesosphere, the filtered ozone oscillation is in phase with the solar cycle, which had a maximum in 2014. This finding confirms the study of [20]. The relative ozone change per solar flux unit is about 40%/100 sfu at 0.001 hPa (sfu: solar flux unit of 10.7 cm solar flux). This magnitude and also the increase in the nighttime ozone response in the upper mesosphere is in a rough agreement with an observational study by [38] who showed that nighttime ozone in the upper mesosphere and beyond is most sensitive to the solar cycle. Please see [20] for a comparison between the Aura/MLS results and solar cycle modeling studies.

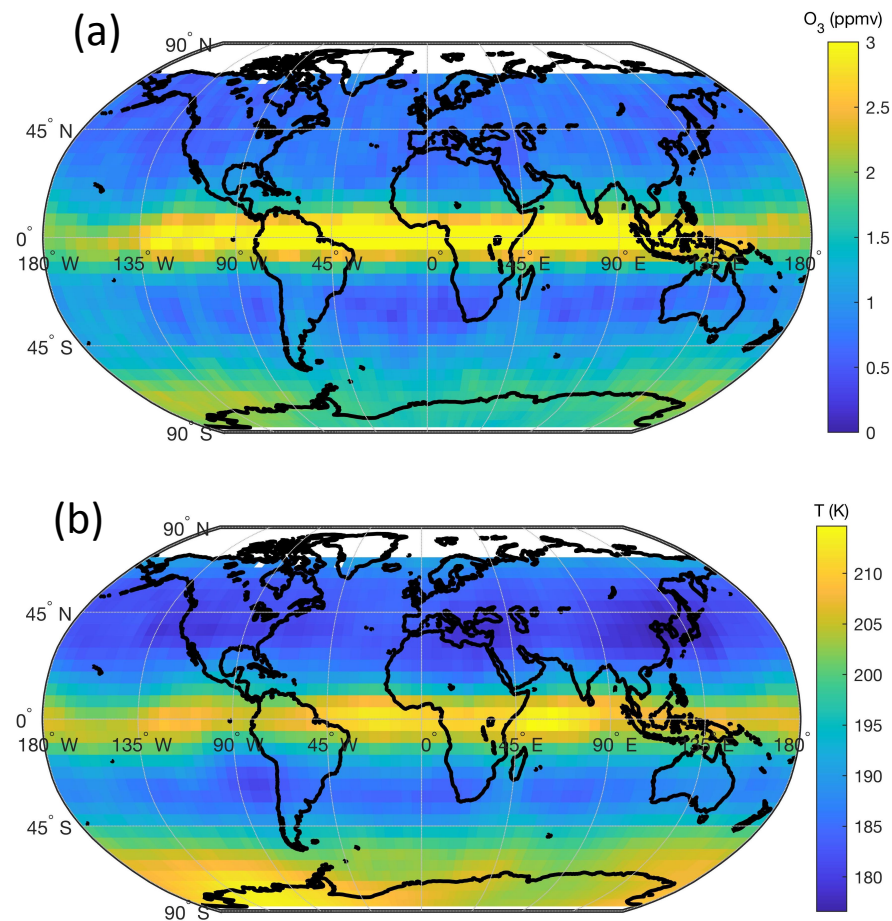


Figure 8. Climatology of nighttime ozone (a) and nighttime temperature (b) in April (years 2005 to 2021) at $p = 0.0046$ hPa in the upper mesosphere observed by Aura/MLS. Missing values of nighttime observations are in white.

There is also an ozone variation in the stratosphere at about 10 hPa, which has a phase lag of about two years with respect to the solar cycle. Generally, the altitude variation of the nighttime ozone variation in Figure 9 agree well with [38] who also found two regions where the ozone response (in ppmv) to the solar cycle is relatively strong: in the mid-stratosphere and in the upper mesosphere.

We also tried lowpass filtering of the temperature, but there are too many temperature oscillations with periods greater than 5 years, so that an attribution to the solar cycle becomes more difficult than in case of ozone.

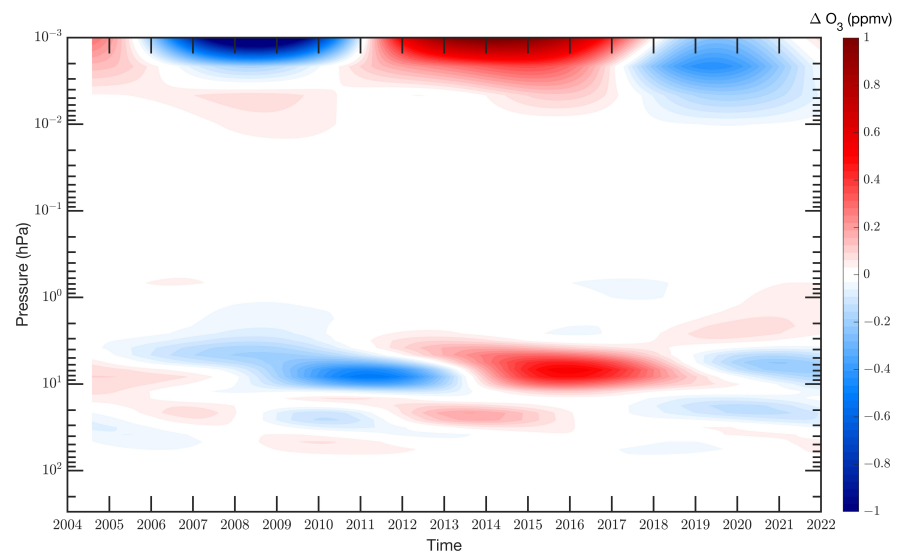


Figure 9. Result of 5 year-lowpass filtering of nighttime ozone (subtracted by mean nighttime ozone profile) at the equator for the search of an 11-year solar cycle signal in ozone.

4. Conclusions

The frequency dependence of the correlation between ozone and temperature was investigated for the first time. The study focused on Aura/MLS observations at mid-latitudes. The anticorrelation of ozone and temperature in the upper stratosphere is also found for short periods down to 2 days (with statistically significant anticorrelations greater than -0.4). However, it remains unclear as to how far the anticorrelation is also valid for periods greater than 4 years and long-term trends of temperature and ozone in the upper stratosphere.

In the lower stratosphere, it seems that the correlation of ozone and temperature is valid at periods greater than 2 years, suggesting a correlation of ozone and temperature trends (correlation coefficient is about 0.8 for a period of 4 years at the pressure level 38 hPa). The annual oscillation (AO) shows an anticorrelation of ozone and temperature in the lower stratosphere (correlation coefficient is about -0.5). This anticorrelation is not found at other frequencies which showed a correlation. It is supposed that yearly transport of cold and ozone-rich polar air to mid-latitudes may explain this anomaly of the correlation of the AOs.

The mid-latitude AOs and equatorial SAOs of nighttime ozone are strongly correlated with those of the temperature in the upper mesosphere beyond 0.01 hPa. The correlation is due to a dynamical control of nighttime ozone influenced by upwelling and downwelling of the secondary ozone layer. The equatorial SAO is possibly due to vertical advection by the diurnal tide, which is associated with enhanced values of nighttime ozone and nighttime temperature, particularly in April and October. The 11-year ozone variation in the equatorial upper mesosphere seems to be in phase with the solar cycle (as already reported by [20]). In the stratosphere at 10 hPa, there seems to be a 2 year phase lag of the ozone variation with respect to the solar cycle.

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