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Effects of the stratospheric quasi-biennial oscillation on long-lived greenhouse gases in the troposphere

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Abstract. An analysis is presented of results of an extended integration with a global general circulation model that includes treatment of a long-lived tropospheric trace constituent as well as a momentum source that forces a realistic stratospheric quasi-biennial oscillation (QBO). It is shown that the dynamical QBO may modulate stratosphere-troposphere exchange in such a manner as to produce a QBO in global-mean tropospheric tracer mixing ratio. For the growth rate of tropospheric methane this transport-induced QBO is expected to have a peak-to-peak amplitude of 1–2 ppbv yr^{-1}, which, while modest compared with the full range of observed variability in growth rate, is still significant. The observed methane growth rate time series during 1983–1999 is shown to be consistent with the predicted QBO effect, although the record is definitely dominated by other interannual variations.

1. Introduction

The study of the observed interannual variability in the concentrations of greenhouse compounds such as CO_2, N_2O, and CH_4 should help elucidate their sources and sinks, and thus can play an important role in improving the understanding of climate change. As an example, the solid curve in Figure 1 shows the growth rate in global mean surface concentration of CH_4 estimated from weekly air sample flask data from 36 stations worldwide [*Dlugokencky et al*., 1998]. The growth rate has exhibited a complicated pattern of interannual variability over a range of timescales. Thus far, the observed variations have not been explained in detail, but some factors that have been suggested include tropospheric climate changes on various timescales, leading particularly to variations in tropospheric water vapor and OH concentrations [*Khalil and Rasmussen*, 1986], variations in biological production and biomass burning [*Lowe et al*., 1997], variations in UV flux due to stratospheric ozone changes or volcanic aerosol loading [*Bekki et al*., 1994; *Dlugokencky et al*., 1996], or changes in stratosphere-troposphere exchange due to volcanic aerosol loading of the stratosphere [*Schauffler and Daniel*, 1994]. Tropospheric CO_2 and N_2O concentrations also display somewhat irregular interannual variations that likely have complicated causes [*Khalil and Rasmussen*, 1992].

The present paper explores the role of the stratospheric quasi-biennial oscillation (QBO) in modulating the interannual variations of long-lived tropospheric constituents. Two possible effects are considered. One involves the QBO modulation of stratosphere-troposphere exchange and, in particular, the flux of air through the tropical tropopause into the stratosphere. Figure 2 illustrates the basic idea. The variations in zonal-mean zonal wind in the QBO are known to be accompanied by changes in zonal-mean temperature and hence changes in the mean meridional diabatic transport circulation [e.g., *Reed*, 1965; *Plumb and Bell*, 1982]. The peak westerly shear in the descending QBO regimes corresponds to the maximum downward QBO anomaly in the meridional circulation. Similarly, eastward shear regions should be accompanied by anomalously high mean upwelling. The amplitude of the zonal wind and temperature QBO is maximum near 40 hPa and decreases in the lowermost stratosphere. However, the QBO still has nonzero amplitude near the tropical tropopause. Thus the QBO modulation of upwelling should influence the transport of air from the troposphere through the tropical tropopause. When there is peak westerly (easterly) mean flow shear in the lowermost stratosphere, the upwelling through the tropopause should be at its minimum (maximum). The air leaving the troposphere near the equator will be high in concentration of CH_4, N_2O, etc. and will be replaced by air entering the troposphere from the stratosphere at higher latitudes that will have lower concentration of these species. Thus the effect of westerly (easterly) shear near the tropopause will be to decrease (increase) the rate of transport loss of the tropospheric inventory of these compounds. Of course, there may be some delay (of the order of a few months) for the effects of these near-tropopause fluxes to mix throughout the global troposphere, and thus affect the global mean near the surface that is reported in the CH_4 measurements in Figure 1. The effects of QBO modulation of temperature and transport near the tropical tropopause on stratospheric water vapor have been considered recently by *Giorgetta and Bengtsson* [1999].

The second possibility for QBO influence on tropospheric greenhouse gases is through the QBO modulation of the stratospheric ozone. It is known that the QBO changes the total ozone column near the equator by ~5–10% peak-to-peak [e.g., *Tung and Yang*, 1994]. This should result in a QBO modulation of ultraviolet (UV) flux into the troposphere that can cause direct effects on the chemical loss rates for constituents in the troposphere.

The remainder of this paper is organized as follows. Sections

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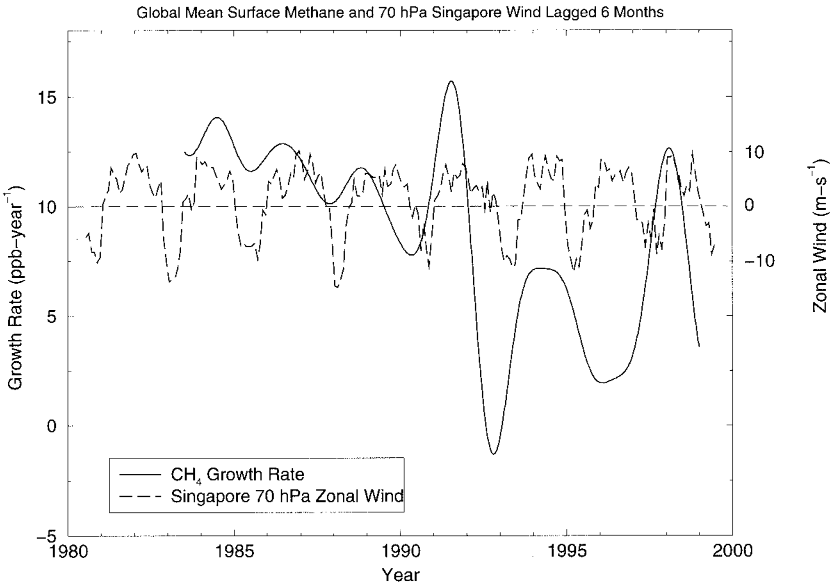

Figure 1. Time series of rate of change of global-mean surface methane mole fraction (solid curve) and the monthly mean zonal wind at 70 hPa measured at Singapore (dashed curve). The methane data are from the global flask air sampling network described by *Dlugokencky et al.* [1996, 1998]. The wind data are lagged 6 months (e.g., the wind values for January 1984 are plotted on the graph at July 1984).

2 and 3 consider the QBO transport effects on long-lived tropospheric trace constituents in the context of general circulation model (GCM) experiment. Section 4 examines the observed global mean CH_4 record in light of the GCM results and also briefly considers the possible role of QBO modulation of stratospheric ozone. Conclusions are summarized in section 5.

2. SKYHI Model Formulation and the QBO Experiment

The SKYHI model [*Fels et al.*, 1980; *Hamilton et al.*, 1995] is a comprehensive global general circulation model that solves the governing equations on a latitude-longitude nonstaggered grid using second-order horizontal differencing. The version used here is that described by *Hamilton* [1998] and has 3°–3.6° latitude-longitude resolution and 40 levels in the vertical from the ground to 0.0096 hPa ($\sim$80 km altitude). As is the case with almost all GCMs [e.g., *Hamilton*, 1996], the standard version of the SKYHI GCM, when run at this resolution, does not spontaneously produce a QBO. The dynamics of the model version used in the present project have been modified from that of the control version (described by *Hamilton et al.* [1995]) by imposition of a zonally symmetric zonal momentum source that is designed to force a QBO in the zonal-mean circulation [*Hamilton*, 1998]. The resulting QBO in mean wind and temperature in the model is documented by *Hamilton* [1998]. The imposed QBO is realistic in terms of the amplitude of the temperature and mean wind variations, and in terms of its vertical and meridional modulation. The imposed QBO is slightly idealized, however, in that it has a period of exactly 27 months and regular downward phase progression, rather than the somewhat irregular period and phase progression seen in the real QBO [e.g., *Naujokat*, 1986].

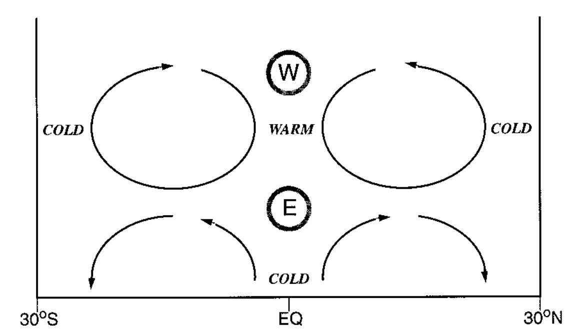

Figure 2. Schematic view of the QBO influence on the mean meridional circulation in the tropical stratosphere. "E" and "W" refer to the maximum easterly and westerly mean-zonal winds, and the peak warm and cold anomalies are also marked. Adapted from *Reed* [1965]. For present purposes, one can imagine the equatorial tropopause being located just above the lower edge of the panel.

As explained by *Hamilton* [1998], the initial fields for this experiment were taken from the end of a multiyear control run. Then the QBO momentum source was turned on, and the model was integrated for 48 years. In the present paper the analysis was restricted to the last 46 years. In the integration the sea surface temperatures (SSTs) were prescribed with seasonally varying, but annually repeating, climatological values.

The version discussed here includes a prognostic calculation of the concentration of N_2O. The implementation of the N_2O "chemistry" is extremely simple. The model was initialized with uniform N_2O mixing ratios several model years before the integration analyzed here. The integration proceeds with no source of N_2O and a linear sink (representing photolysis and reactions with atomic oxygen) with a prescribed destruction rate that depends only on latitude and pressure (a meridional section of the prescribed destruction rates is shown by *Hamilton and Mahlman* [1988]). The calculation of the advective tendency of tracer concentration is performed with a centered Eulerian flux scheme which is second order in the horizontal and fourth order in the vertical. This scheme conserves total

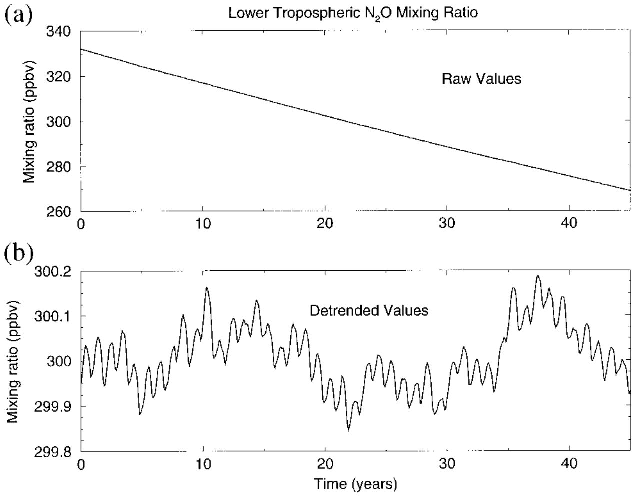

Figure 3. (a) Time series of lower tropospheric N_2O mixing ratio for 45 years of SKYHI model integration. The values plotted are for averages between 60°S and 60°N and roughly 700–800 hPa. Results are rather insensitive to the precise altitude and latitude limits employed in calculating the mean. (b) The mixing ratio time series when an exponentially decaying trend is removed.

mass to machine precision in the absence of sources and sinks, and thus is particularly well suited to simulating the small long-period changes in tracer concentration of present interest. The scheme is not positive definite, of course, but no "filling" of negative mixing ratios is used in the present simulation of N_2O.

The result is a simulated N_2O field that has some realistic features in terms of its variation with height and latitude [e.g., *Hamilton and Mahlman*, 1988]. However, the simulated inventory of N_2O decreases constantly with time, as seen in Figure 3a which shows the lower tropospheric mixing ratio averaged over 60°S–60°N in each month for 45 years of the integration (see the figure caption for details of the diagnostic calculation). The 60°S–60°N band was chosen for these calculations to be roughly similar to the latitude coverage that could be expected from an extensive worldwide observational network, but if the mean is taken over the whole globe, the results are almost identical. Figure 3b shows the mixing ratios when a best fit exponential decrease over the 45 years is removed from the data. A seasonal cycle and interannual variability on various timescales are both evident. It is possible that the very long-term (~30 year period) variation seen reflects some remaining transient adjustment to the initial conditions in the N_2O field.

3. Results From the GCM Experiment

The tropospheric mixing ratios shown in Figure 3b exhibit a seasonal cycle as well as interannual variations on a wide range of timescales. Figure 4a shows the rate of change of the detrended lower tropospheric N_2O timeseries in Figure 3b. These growth rates also clearly display a seasonal cycle and interannual variations. Figure 4b shows the growth rates with the long-term mean seasonal cycle removed, and Figure 4c shows a 12-month running mean of the same quantity. The time evolution seen in Figure 4c is rather complicated, but a variation with roughly quasi-biennial timescale does stand out: there are 18 distinct peaks (and 18 minima) in the 45 year time series shown.

The correlation between the QBO and the tropospheric N_2O growth rates in the simulation is demonstrated explicitly in Figure 5 which shows the smoothed growth rates of Figure 4c together with the equatorial zonal-mean zonal wind at 70 hPa. The wind values have been lagged by 6 months, and with this lag there is a positive correlation between the two curves, with all but two of the 20 individual maxima in the wind curve being matched by peaks in the growth rate (the two exceptions being in year 18 and in year 25). The correlation coefficient of the two monthly time series shown in Figure 5 is 0.49. If the

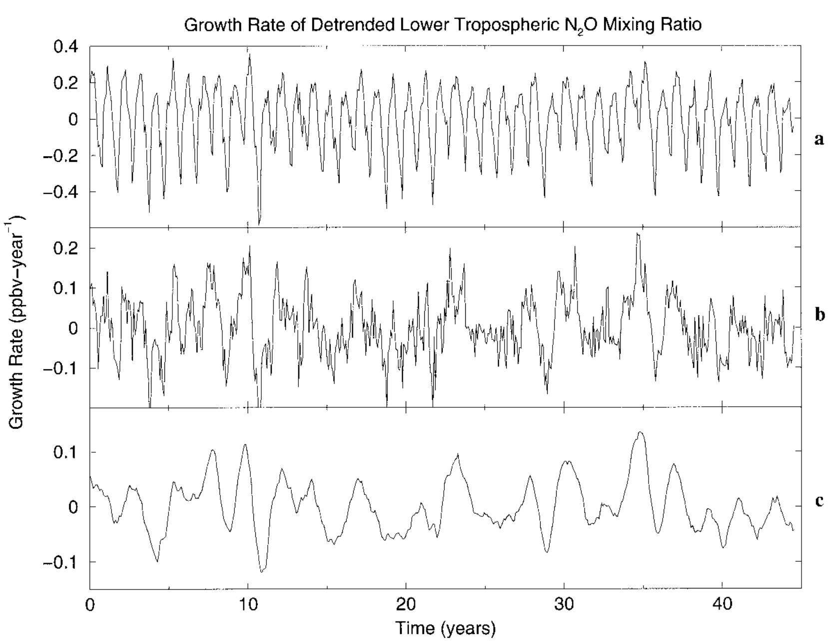

Figure 4. (a) N_2O mixing ratio "growth rate," that is, the rate of change of the detrended time series of Figure 3b. (b) The growth rate time series with the 45-year-mean seasonal cycle removed. (c) The 12-month running mean of the deseasonalized growth rate time series.

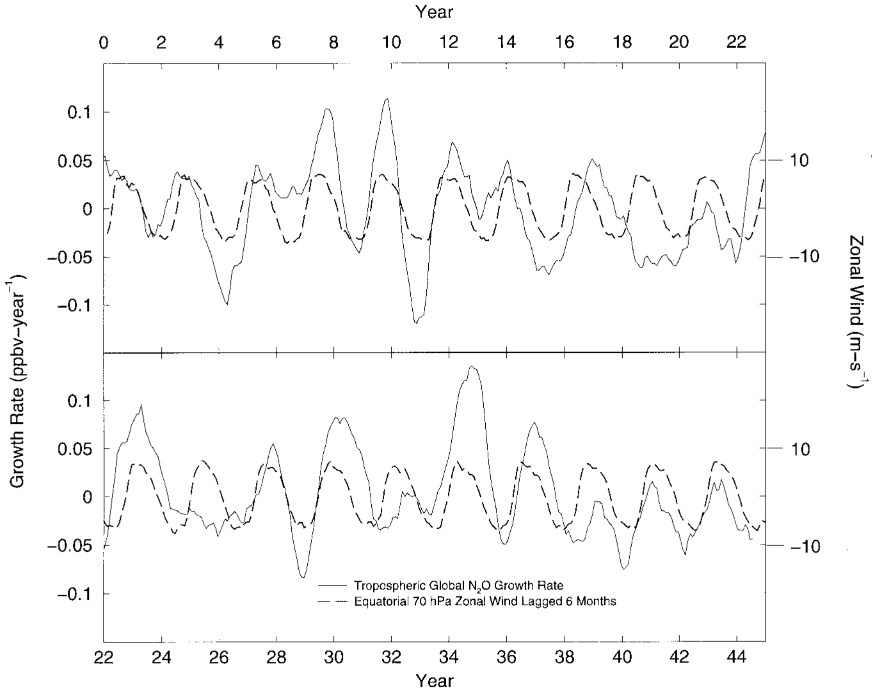

Figure 5. The 12-month running mean of the detrended, deseasonalized N_2O growth rate time series (solid curve) compared with the time series of monthly mean zonal-mean equatorial zonal wind at 70 hPa in the SKYHI model simulation (dashed curve). The wind values are lagged by 6 months.

smoothed time series is regarded as representing roughly 45 independent samples, this value is significantly different from zero at the 0.001 level. The growth rate of mean lower tropospheric N_2O mixing ratio appears to be modulated in such a way that it tends to be a maximum (minimum) about 6 months after the wind in the lower stratosphere reaches its maximum westerly (easterly) value. The QBO variations in the wind right at the tropopause (e.g., 100 hPa) are much smaller, and so the 70 hPa zonal mean wind is a reasonable measure of QBO-related shear in the lowest ~1–2 km of the equatorial stratosphere. Thus the results for the GCM are in basic agreement with the conceptual model for QBO transport modulation of tropospheric trace constituents described earlier in section 1.

The connection between tropical upwelling and the tropospheric N_2O mixing ratios is made more directly in Figure 6, which compares the N_2O growth rates with anomalies in the diabatic heating rates averaged between 12°S and 12°N and for 92 hPa (actually an average of values at model levels at 103 and 81 hPa). The heating rates are 12-month running means and are lagged by 6 months. The diabatic heating is a good indicator of the upward mass transport by the mean meridional circulation [e.g., *Andrews et al.*, 1987]. The heating rates are plotted with positive values downward in Figure 6, and so the apparent positive correlation of the two curves indicates a tendency for tropospheric N_2O growth rates to be near minimum about 6 months after the maximum diabatic heating (and hence peak mean upwelling) at the base of the stratosphere near the equator.

While the effects of the tropical mean upwelling are clearly visible in the growth rate time series, there are other complicated interannual variations as well. As noted earlier, any such variations reflect interannual changes in the model transport and must be generated spontaneously by the model dynamics. Earlier studies have shown that the SKYHI model does simulate a significant degree of interannual dynamical variability, even when run with annually repeating SSTs [*Hamilton*, 1995]. These other spontaneously generated variations are apparently strong enough on occasion to completely mask the expected QBO peaks in growth rate (notably in years 18 and 25 of the simulated record), and must account generally for the irregularity in the QBO cycle-to-cycle behavior of the growth rates. The nature of these variations has not been diagnosed, but most likely they reflect anomalies in extratropical stratosphere-troposphere exchange. The present simulation provides both an estimate of QBO effects and also an indication of the magnitude of other spontaneously generated transport influences on the global mean concentrations of long-lived tropospheric trace gases. However, note that given the use of annually repeating SSTs, the present results might be taken as a lower bound for the magnitude of the interannual transport effects.

Another possible mechanism for QBO transport modulation of the stratosphere-troposphere exchange could arise through the tropical QBO effect on the extratropical stratosphere. In particular, *Holton and Tan* [1980] showed from observations a statistical tendency for the Northern Hemisphere extratropical stratospheric circulation to be more disturbed (less zonal) in boreal winters when there are easterlies near the 50 hPa level at the equator. This QBO-modulated extratropical circulation anomaly could in turn affect the exchange of mass across the extratropical tropopause. The SKYHI model version discussed here actually captures the observed statistical Holton-Tan effect quite well [*Hamilton*, 1998]. However, in both the observations and the model results the tropical QBO effect on the extratropical stratospheric circulation is far from predictable on a winter-by-winter basis. The QBO correlation with tropospheric N_2O growth rates seen in Figure 5 is much more predictable from cycle to cycle, and this favors the simple QBO

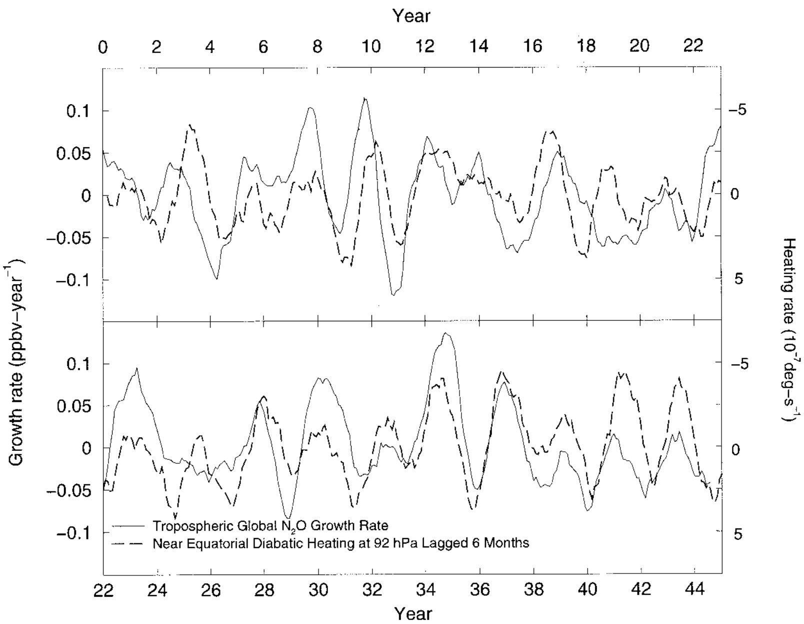

Figure 6. The 12-month running mean of the detrended, deseasonalized N_2O growth rate time series (solid curve) compared with the 12-month running mean of the zonal-mean radiative heating rate anomalies averaged over 12°S–12°N at 92 hPa (dashed curve). The heating rates are lagged 6 months and are plotted with positive values downward (see axis labels on right-hand side).

modulation of tropical upwelling as the most plausible explanation for the result.

The annual cycle of N_2O growth rate in Figure 4a also makes an interesting comparison with the QBO. The equatorial tropopause temperature in both the SKYHI model [see *Pawson et al.*, 2000, Figure 2] and observations [*Yulaeva et al.*, 1994] undergoes a significant seasonal cycle, with coldest temperatures in January. This should be accompanied by a seasonal cycle of diabatic transport with maximum upwelling in January. If the same 6-month lag applies to the seasonal cycle, then minimum in global lower tropospheric N_2O growth rate should occur 6 months later (around July). Inspection of Figure 4a shows that in fact the minima in the growth rate generally occur about July. The peak-to-peak amplitude of the annual cycle in tropopause temperature is 5°C in both observations and the model, which is roughly an order of magnitude larger than that in the QBO as imposed in the model. Thus the larger amplitude of the annual cycle in N_2O growth rate relative to that of the QBO is understandable, and both variations in N_2O are perhaps largely ascribable to the same basic transport mechanism involving modulation of tropical upwelling.

The magnitude of the QBO in global-mean surface N_2O in the model appears to be of the order of 0.1–0.2 ppbv yr^{-1} peak to peak. The existing observed estimates of global-mean N_2O do not appear adequate to detect this variation. The more extensive observations of CH_4 allow a better chance of detecting a QBO transport effect, and these observations are considered in the next section.

4. Application to the Observed Methane Record

The SKYHI model experiment did not explicitly include CH_4 as a tracer, but the overall behavior of CH_4 should resemble that of N_2O. Both tracers have long lifetimes in the troposphere and chemical sinks in the stratosphere, so they should share a similar phase relation between the dynamical QBO and any QBO in tropospheric mixing ratios. The magnitude of the QBO effect will depend on the actual mixing ratios themselves, and most particularly on the contrast between the tropospheric and lower stratospheric mixing ratios. In the box model calculations of *Schauffler and Daniel* [1994], perturbations purely to stratospheric-tropospheric exchange rates have an effect on tropospheric CH_4 volume mixing ratios that is a factor of 11 larger than that on N_2O volume mixing ratios (scenario C in their Tables 5 and 6). Using that estimate together with the SKYHI N_2O results would suggest that the QBO in tropospheric CH_4 growth rate should be ~1–2 ppb yr^{-1}.

Figure 1 shows the global-mean methane growth rate estimated from surface observations together with the observed time series of 70 hPa monthly mean zonal wind at Singapore (1.3°N, 104°E). The wind series clearly shows the QBO, although there is some more irregularity in this single station data than in the zonal-mean equatorial wind in the model experiment. As in Figure 5, the wind values have been lagged 6 months. There is a degree of correspondence between the two curves in Figure 1, with the CH_4 growth rate maxima in mid-1984, early 1986, late 1988, mid-1991, early 1994, and late 1998 all being roughly coincident with peak westerly phases of the lagged Singapore wind series. On the other hand, the westerly phase in 1996 has no corresponding peak in the CH_4 growth rate. There is some resemblance in Figure 1 to the behavior of the SKYHI model N_2O growth rate and equatorial wind curves, but clearly the observed CH_4 growth rate is affected by more dramatic irregular interannual variations, notably the large drop in late 1992 and subsequent recovery. The curves have a positive correlation coefficient of 0.19; the small value reflects the large non-QBO-related CH_4 variations. However, the observed record seems to be at least consistent with the expected transport-driven QBO of 1–2 ppb yr^{-1} superimposed on other larger, but less regular, changes.

Another possible mechanism coupling the stratospheric QBO with tropospheric CH_4 is via the QBO modulation of stratospheric ozone. The QBO changes the total ozone column near the equator by ~5–10% peak to peak, and this should result in a QBO modulation of UV flux into the equatorial troposphere. Calculations with a photochemical model by *Thompson et al*. [1989] suggest that a 1% decrease in column ozone may result in 0.6–0.9% increase in tropospheric OH concentration. Reaction with OH is the main atmospheric sink for CH_4, thus minimum ozone near the equator should result in enhanced destruction of CH_4 and reduced growth rates of the tropospheric CH_4 inventory, at least near the equator. A complication is that the QBO in total column ozone extends well beyond the equator and reverses sign near 15° latitude in both hemispheres, so that the QBO in global-mean ozone column is quite small [e.g., *Tung and Yang*, 1994]. Thus the effects in the region poleward of 15° will compensate to some extent for the low-latitude effects, although given the stronger incident UV flux, the low-latitude effects would be expected to dominate the influence on the global-mean CH_4.

As mentioned in the Introduction, the effects of all observed ozone variations during 1979–1992 were introduced into a photochemical model (with no interannual variations in transport) in work reported by *Bekki et al*. [1994]. They show their calculated growth rate time series for global-mean CH_4 relative to the same quantity calculated without any interannual ozone column variations (see their Figure 3). These results do show some roughly biennial behavior with peaks in the growth rate occurring near January 1981, February 1982, January 1984, December 1986, June 1987, March 1989, and March 1991. The mean period of these oscillations is significantly under 2 years, however, so they do not appear to indicate a systematic effect of the tropical stratospheric QBO. These roughly biennial variations in the Bekki et al. calculations are also not very well correlated with the fluctuations in the observed CH_4 growth rate.

A direct comparison of the observed CH_4 growth rate record with the total ozone columns measured by spectrometers on the Nimbus 7 and Adeos satellites is given in Figure 7. There is an apparent correlation over at least part of the record, with minima in CH_4 growth rate corresponding with maximum ozone column in early 1985, mid-1987, early 1990, and perhaps in early 1992. Note, however, that this is exactly the opposite phase relation expected for CH_4 growth rate from the purely photochemical influence of ozone column variations. To the extent that a QBO cycle can be discerned in the observed CH_4 record, it must represent predominantly a transport effect rather than an ozone column photochemical effect.

Another issue that arises in interpreting apparent QBO variability in the CH_4 record is possible confounding with signals really due to the Southern Oscillation (SO). The influence of the SO on tropospheric temperature and humidity could have an effect on CH_4 chemistry. The SO has a broader and redder spectrum than the QBO, but there is some overlap, and the possibility of confounding QBO and SO signals, at least for limited periods, is real. As part of the present investigation, the familiar SO index (pressure difference between Darwin and Tahiti) was plotted together with the CH_4 growth rate for the 1983–1999 period (not shown). It is apparent that, while there may possibly be some connection between the two time series, the quasi-biennial timescale variations that appear in the CH_4 record have no clear counterpart in the SO index time series.

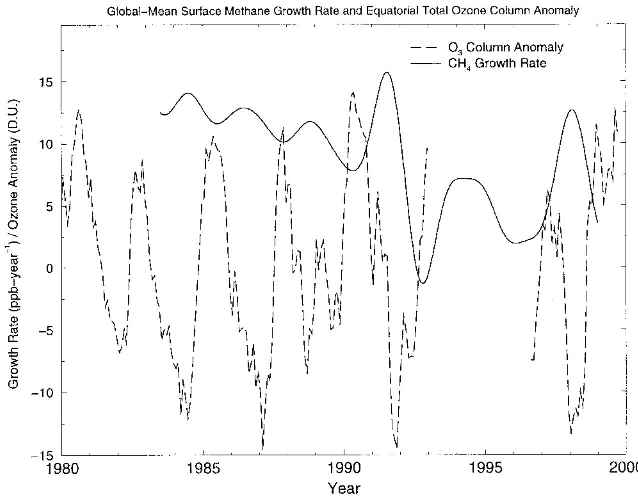

Figure 7. Methane mole fraction growth rates (solid curve) compared with anomalies in the total column of ozone averaged over 10°S–10°N as measured by total ozone mapping spectrometer instruments deployed on Nimbus 7 (to 1992) and Adeos (after August 1996). The ozone anomalies are computed relative to the seasonal mean over the total record of each satellite.

5. Conclusion

The present paper has examined the observed global-mean surface CH_4 record in light of an extensive GCM experiment that studied the QBO transport effect on a tracer with a long tropospheric lifetime. The model results suggest that the transport effects of the stratospheric QBO may systematically modulate global-mean tracer concentration in the lower troposphere. The effects are predicted to have a modest, but not negligible, effect on the growth rate of tropospheric CH_4, and the observed record over the last 2 decades seems consistent with the predicted QBO effect. The model simulation of N_2O also displays interannual variations unrelated to the QBO, but produced by spontaneous (i.e., generated purely within the atmosphere) variations in transport. These irregular transport-generated variations appear to be of roughly the same magnitude as the systematic QBO. This suggests that such spontaneous transport effects can also play a significant, although far from dominant role, in determining the interannual variations of tropospheric CH_4.

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