MAUSAM, 51, 2 (April 2000) 163 – 168

551.510.41 : 546.172.5

Interannual variability of atmospheric Nitrous Oxide (N_2O)

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(*Received 27 October 1998, Modified 17 August 1999*)

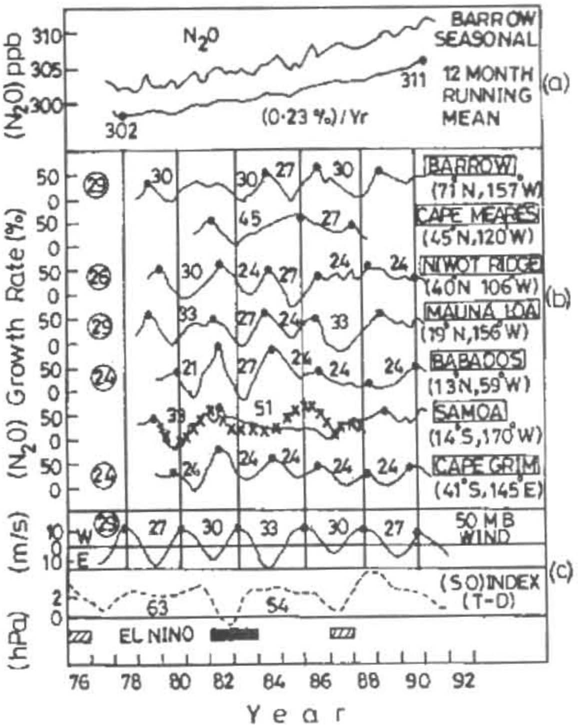
सार - 1977 से 1991 की अवधि के दौरान नाइट्रॅस ऑक्साइड (N_2O) के सात स्थानों से मापे गए 12 महीनों के निरंतर माध्यों का उपयोग एम.ई.एस.ए. (अधिकतम एंट्रॉपी अनुक्रम विश्लेषण) की वार्षिक (प्रतिवर्ष 4 मान, तीन महीनों के कालखंडों में (वृद्धि दर क्रम का प्रतिशत ज्ञान करने के लिए किया गया है। इस स्पेक्ट्रा से वर्षों के परास (रेंज) (2.04-2.38) में अद्ध-द्विवार्षिक दोलन की अवधियों तथा लगभग 4.0 वर्षों के अर्द्ध-त्रिवार्षिक दोलन की अवधियों सहित उल्लेखनीय क्य.बी.ओ. (अद्ध-द्विवार्षिक दोलन) तथा क्यू.टी.ओ. (अर्द्ध-त्रिवार्षिक दोलन) का पता चला है। ये 2.31 वर्ष की अवधि के दोलनों से मेल खाते हैं। किन्तु ये ताहिती माइनस डार्विन समुद्र तल वायुमंडलीय दाब अंतर (टी-डी) के द्योतक 50 हेक्टापास्कल की निम्न अक्षांशीय क्षेत्रीय पवन 2.58 वर्षों के अर्द्ध-द्विवार्षिक दोलनों से मेल नही खाते हैं।

ABSTRACT. The 12-monthly running means of N_2O measured at seven locations during 1977-91 were used for obtaining the yearly percentage growth rate series (4 values per year, centered 3 months apart), which were subjected to MESA (Maximum Entropy Spectral Analysis). The spectra revealed significant QBO and QTO (Quasi-biennial and Quasi-triennial oscillations) with QBO periods in the range (2.04-2.38) years and QTO periods near 4.0 years. These do not resemble the QBO of 2.58 years of the 50 hPa low latitude zonal wind but do resemble the QBO of 2.31 years and the 4.1 year periods of the Southern oscillation phenomenon, represented by Tahiti minus Darwin sea level atmospheric pressure difference (T-D).

Key words – Quasi-biennial oscillation (QBO), El Nino and Southern Oscillation (ENSO), Atmospheric Nitrous Oxide.

1. Introduction

Measurements of atmospheric trace elements are being carried out by the Climate Monitoring and Diagnostics Laboratory (CMDL) of National Oceanic and Atmospheric Administration (NOAA), Air Resources Laboratory, Boulder, Colorado, USA as also by other groups under the Atmospheric Lifetime Experiment (ALE) programme followed by the Global Atmospheric Gases Experiment (GAGE) programme. Using the CMDL data, Kane (1994) showed that most of the trace elements had an interannual variability involving QBO and/or QTO (Quasi-biennial and Quasi-triennial oscillations). For Nitrous oxide (N_2O), Montzka *et al.* (1992) reported a growth rate of ~ 0.6 ppb/yr during 1977-85, which increased to ~ 1.1 ppb/yr during 1988-91. Since 1991, the growth rate of N_2O seems to have decreased to ~ 0.5 ppb/yr and the annual values seem to have leveled off in 1993 (Thompson *et al.*, 1994). Using 10 years of ALE-GAGE N_2O data (1978-88), Prinn *et al.* (1990) estimated trends in the northern and southern hemispheres. They concluded that the major trends and the latitudinal distributions were consistent with the hypothesis that stratospheric photo dissociation is the major atmospheric sink for N_2O; but the N_2O increases were not caused solely by increases in anthropogenic N_2O emissions (fossil fuel combustion etc.) but were probably due to a combination of growing tropical source (land disturbance) and a growing northern mid-latitude source (fertilizer use and fossil fuel combustion). They also conducted a multiple regression analysis which included cycle (annual, quasi-biennial) terms and concluded that a statistically significant QBO was evident at Barbados and Tasmania but otherwise, both annual and quasi-biennial oscillations were weak or insignificant at all sites. These were statistical, average results for an assumed period of 2.25 years (27 months) for the QBO. Recently, Montzka *et al.* (1992) and Prinn *et al.* (1994) have published N_2O data. In



Figs. 1(a-c). (a) Seasonal and 12-month running means of N_2O at Barrow (71°N, 157°W), (b) Yearly growth rate of N_2O at various locations. Numbers represent spacing(months) between successive maxima and (c) 12-month running means of 50 hPa low latitude winds and Southern Oscillation (SO) index represented by Tahiti minus Darwin atmospheric pressure difference (T-D). Vertical lines mark the 50 hPa westerly wind maxima. Rectangles indicate El Nino events. Some doubtful peaks are marked halfway between two clear peaks

this note, we propose to examine in detail the interannual variability of these N_2O series, with emphasis on the QBO.

2. Data

Montzka *et al*. (1992) have given the monthly means or N_2O (ppb) from the Flask programme, for the locations Barrow (71°N, 157°W), Niwot Ridge (40°N, 106°W), Mauna Loa (19°N, 156°W) and Samoa (14°S, 170°W) for 1977-91, while Prinn *et al*. (1994) have given monthly mean N_2O (ppb) for Barbados (13°N, 59°W), Cape Grim (41°S, 145°E), Cape Mears (45°N, 120°W) and Samoa (14°S, 170°W) for 1978-91. The procedure of analysis is illustrated in Fig. 1. An examination of the various data showed that there was considerable annual (month to month) variation. To reduce the same, seasonal means for JFM, AMJ, JAS and OND were obtained. In Fig. 1(a), the top plot shows the seasonal means for Barrow. As can be seen, some seasonal variation is still evident. To eliminate the same, running averages over four consecutive seasons were evaluated, thus yielding basically 12-month running means, centred at March end, June end, September end and December end (4 values per year). These are plotted for Barrow in the second plot. The plot is now fairly smooth. Starting from a value of ~303 ppb in 1978, the N_2O values at Barrow rose to ~311 ppb in 1991, a rise of 9 ppb in 13 years, yielding an average growth rate of ~0.23%/yr. To obtain the growth rate of every year, every 12-month mean was subtracted from the 12-month mean centred 4 seasons later and the difference was expressed as percentage of the N_2O value at the middle of that year. The percentage growth rates so obtained (4 values per year) are shown in Fig. 1(b) for the various locations. Instead of showing steady long-term increases or decreases, the growth rates seem to have QBOs. The numbers indicate spacings (in months) between successive peaks.

3. QBO characteristics by visual inspection

The QBOs at different locations seem to have different characteristics. The peaks are not always clearly seen. So, some indefinite peaks are assumed to be half way between two clearly identifiable peaks. For Barrow, the spacing is 27-30 months and the average spacing (circled number at the left) is ~29 months. The same is true for Mauna Loa where the spacing range is larger (24-33 months) but the average is ~ 29 months. In the case of Niwot Ridge and Barbados, there is ambiguity in the last three peaks. Only Cape Grim shows clear peaks.

Among atmospheric parameters, the stratospheric low latitude zonal winds are well known to have a prominent QBO (Reed *et al*., 1961). Fig. 1(c) shows a plot of 12-month running means of 50 hPa wind, obtained as an average of four stations *viz*. Gan (0.7°S, 73.2°E), Balboa (8.9°N, 79.6°E), Singapore (1.4°N, 103.9°E) and Canton (2.8°S, 171.7°W) given by Venne and Dartt (1990), updated from Meteorological Data Reports. The vertical lines mark the westerly wind maxima and the spacing between successive maxima is 27-33 months, with an average spacing of ~29 months. Thus, the average spacing for N_2O at Barrow and Mauna Loa matches that for 50 hPa wind. The individual N_2O growth rate maxima seem to match roughly with 50 hPa easterly wind maxima (westerly wind minima). Similarly, N_2O growth rate minima seem to match with 50 hPa westerly wind maxima (vertical lines). However, the matching is not perfect and could as well be accidental. If individual maxima or minima of N_2O are compared with those of the 50 hPa wind, there is some indication of a match in some cases, with slight phase shifts, but the phase shifts are not always consistent.

For Niwot Ridge, the spacing is 24-30 months with an average spacing of ~26 months while for Barbados, the spacing is 21-27 months with an average of ~24 months. At Barbados, there is one big QBO wave during 1982-84, which probably gave the significant QBO amplitude reported by Prinn *et al.* (1990). At Cape Grim (Tasmania), the spacing is very regular and the average is 24 months. Thus, the average spacing of 2.25 years (27 months) assumed by Prinn *et al.* (1990) is appropriate only for Niwot Ridge, if at all. Even the 50 hPa wind does not have this spacing (during 1976-92).

The data for Cape Mears are shorter and the QBO pattern is very irregular. For Samoa also, the QBO pattern is very irregular. Here, the full lines represent the CMDL Flask program data (Montzka *et al.*, 1992) while the crosses represent the gas chromatographic measurements of Prinn *et al.* (1994). Firstly, the two plots do not tally, which is very disconcerting. Secondly, both show irregular QBO (spacing 33, 51 months). Thus, for some reason (including probably doubtful data), the QBO is very obscure at Samoa. At stations like Barbados and Niwot Ridge, there are perturbations on the QBO pattern.

Another phenomenon of meteorological interest is the Southern Oscillation (SO). A simple index representing the same is the Tahiti (18°S, 150°W) minus Darwin (12°S, 131°E) atmospheric pressure difference (T-D), obtained from Parker (1983) (updated from Meteorological Data Reports) and plotted in Fig. 1(c). Here, (T-D) minima in 1976, 1982-83 and 1987 are associated with El Ninos (warm water events in the Peru-Ecuador coast) marked as rectangles. However, these have spacing of ~50-60 months and do not seem to be related to the N_2O growth rates. Incidentally, Rasmusson *et al.* (1990) have identified two dominant time scales of ENSO (El Nino-Southern Oscillation) variability *viz.* a biennial mode (24 months) and a lower frequency mode of period 4-5 years. It is tempting to compare the N_2O QBO at Barbados and Cape Grim with this biennial mode.

4. Power spectrum analysis

So far, QBO characteristics were examined crudely by visual inspection. A finer, rigorous examination by spectrum analysis is advisable. To detect periodicities, all these series were subjected to a power spectrum analysis, using MESA (Maximum Entropy Spectral Analysis, Burg, 1967; Ulrych and Bishop, 1975), which detects periodicities much more accurately than the conventional BT method (Blackman and Tukey, 1958). Similar to the parameter lag m in BT, MESA has a parameter called LPEF (Length of the Prediction Error Filter), which can be chosen. With low LPEF, only low periodicities are resolved. Larger LPEF resolve larger periodicities, even those approaching the data length, but the errors are larger and the low periodicities show peak-splitting. An LPEF of ~50% of the data length is generally adequate and was used in the present analysis.

MESA has a drawback viz., the power estimates are not reliable (Kane and Trivedi, 1982). Hence MESA was used only to identify the possible periodicities T_k, which were then used in the expression:

$$f(t) = A_0 + \sum_{k=1}^{n}\left[a_k \operatorname{Sin}\left(\frac{2\pi t}{T_k}\right) + b_k \operatorname{Cos}\left(\frac{2\pi t}{T_k}\right)\right] + E$$

$$= A_0 + \sum_{k=1}^{n} r_k \operatorname{Sin}\left(\frac{2\pi t}{T_k} + \phi_k\right) + E \qquad (1)$$

where $f(t)$ is the observed series and E the error factor. A multiple regression analysis (MRA, Bevington 1969) was then carried out to obtain the best estimates of A_0, (a_k, b_k) and their standard errors, by a least square fit. From these, r_k and their standard error σ (common for all r_k in this methodology) can be calculated and any r_k exceeding 2σ would be significant at a 95% (a priori) confidence level.

Fig. 2 shows the result of MESA. The top plot is for 50 hPa wind and shows one very prominent peak at 2.58 years, ~31 months, slightly larger than the average spacing of ~29 months seen in Fig. 1(c). (The hatched portion represents 2σ limit). The difference could be due to the presence in the wind spectra of additional small peaks at 2.0 and 5.1 years. The second plot is for (T-D). Here, there is a major peak at 4.1 years (49 months) and a minor peak at 2.31 years. Thus, in (T-D), the major peak is a QTO (Quasi-triennial oscillation) but there is a minor QBO also, whose period (2.31 years) is smaller than the wind QBO period (2.58 years) and the two are probably unrelated.

The other plots in Fig. 2 refer to the spectra of N_2O growth rates at individual stations. The following may be noted:

(a) Most of the locations have a peak near 1.60 years (~ 20 months).

(b) Most of the locations have a prominent QBO peak in the range (2.04-2.38) years, somewhat similar to the minor 2.31 year period in the (T-D), but different from the wind QBO (2.58 years). Thus, relationship between 50 hPa wind and N_2O does not seem to exist. Of course, there is no particular reason to expect such a relationship, as N_2O is tropospheric.

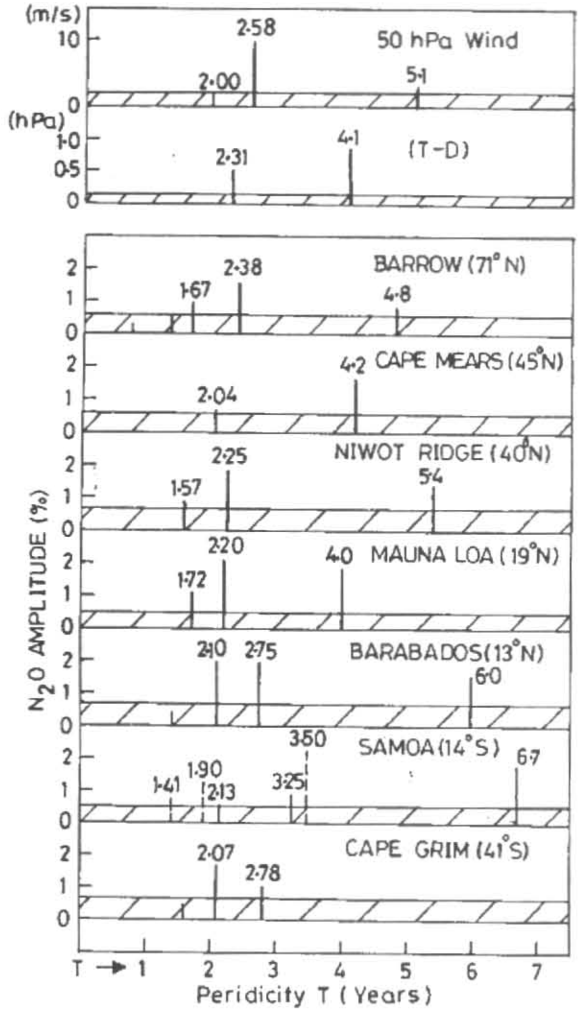


Fig. 2. Amplitudes of the various periodicities T, detected by a Maximum Entropy Spectral Analysis of the various series. Top plots are spectra for the 50 hPa low latitude zonal wind, and the Southern Oscillation index Tahiti minus Darwin sea-level atmospheric pressure difference (T-D). The other plots are for the spectra of N_2O series at various locations. The hatched portion represents the 2σ limit and amplitudes exceeding the same (numbers indicating periodicities T in years) are significant at a 95% (a priori) confidence level

(c) Four locations have a peak in the range (4.0-5.4) years. The 50 hPa wind does have a peak at 5.1 years, but it is barely significant. The (T-D) has a prominent peak at 4.1 years. Thus, the N_2O peaks at Barrow (4.8 years), Cape Mears (4.2 years) and Mauna Loa (4.0 years) may be related to the (T-D) peak.

(d) Some locations have peaks at larger periods, *e.g.*, Niwot Ridge at 5.4 years, Barbados at 6.0 years and Samoa at 6.7 years. In a short data sample of only ~12 years, any long term trend may make some contribution to these periods. Hence, these may not be authentic. Some contribution must have come because of the decrease of the growth rates since 1991 onwards, as mentioned in the Introduction.

(e) Barbados (13°N) and Cape Grim (41°S) have additional peaks at 2.75 years and 2.78 years.

(f) The longer Samoa series has a barely significant peak at 2.13 years and a significant peak at 3.25 years. The Prinn *et al.* (1994) Samoa data (dashed lines) show significant peaks at 1.41years (17 months), 1.90 years (23 months) and a strong peak at 3.50 years, somewhat different from the peaks in the other Samoa series. The N_2O results for Samoa are somewhat confusing. Samoa is often used as a typical equatorial station. The only prominent QTO peak here seems to be ~3.2-3.5 years, different from the ~4 year N_2O peak at other locations, and the (T-D) peak. The reason for this difference is not clear. Data quality may be doubtful.

5. Conclusions and discussion

The 12-month running means of N_2O were used to obtain percentage yearly growth rates at the locations Barrow (71°N), Cape Mears (45°N),Niwot Ridge (40°N), Mauna Loa (19°N), Barbados (13°N), Samoa (14°S) and Cape Grim (41°S). A spectral analysis of the time series of all these for the period 1977-91 (four values per year of the growth rates, centered 3 months apart) revealed a small but significant (exceeding 2σ limit) peak at ~ 1.65 years (~ 20 months) at some locations, strong QBO peaks in the range (2.04-2.38) years at all locations, and significant additional peaks at ~ 2.75 years at Barbados and Cape Grim. Strong larger periodicity peaks were observed at Barrow (4.8 years), Cape Mears (4.2 years), Niwot Ridge (5.4 years), Barbados (6.0 years) and Samoa (6.7 years). Some of these may be reflections of changes in long-term trends. The 50 hPa low latitude zonal wind had one prominent peak at 2.58 years, which did not match with any of the N_2O peaks. Thus, a relationship between stratospheric wind and tropospheric N_2O seems to be non-existent. It is not expected also, as tropospheric N_2O is due to biospheric sources. On the other hand, the Southern Oscillation represented by Tahiti minus Darwin atmospheric pressure difference (T-D) at sea level, has a prominent peak at 4.1 years and a less prominent but significant peak at 2.31 years, (a QBO, different from the wind QBO of 2.58 years). Thus, the N_2O peaks seem to be similar to the (T-D) peaks.

Soon after the discovery of the QBO in the stratospheric low latitude zonal wind (Reed *et al.* 1961;

Veryard and Ebdon, 1961), Funk and Garnham (1962) reported a similar QBO in equatorial total ozone column and Angell and Korshover (1962) in middle and high latitude ozone. At low latitudes, the stratospheric wind, ozone and stratospheric temperature seem to have almost exactly similar QBO, with a period of ~30 months. However, at middle and high latitudes, the QBO characteristics change considerably. Yang and Tung (1995) reported that the ozone QBO signal was strongest in the middle and high latitudes, mainly in the winter-spring season and had additional peaks at ~8 and 20 months, besides the 30 month peak. Several studies link the equatorial wind QBO with the extra-tropical stratosphere in winter (*e.g.*, Holton and Tan, 1980,1982; Van Loon and Labitzke, 1987; Dunkerton and Baldwin, 1991) and demonstrate that the polar vortex tends to be colder and more intense in the west phase of equatorial wind QBO. For lower altitudes (tropopause, troposphere), the extension of the wind QBO is obscure. Angell and Korshover (1983) presented a study of the seasonal and annual changes of global tropospheric temperature and Angell (1990) studied their El-Nino association. Trenberth (1980), Yasunari (1989) and Kane (1992) compared the QBO of different parameters in the troposphere and stratosphere. Recently, Kane and Buriti (1997) studied the spectra of temperature series at five atmospheric levels and seven climatic zones and reported that for stratosphere and tropopause, the temperature variations near the equator and near the North Pole had some resemblance with the equatorial stratospheric wind. But for troposphere and surface, the temperature variations, especially those near the equator, resembled those of equatorial eastern Pacific sea-surface temperature (SST), which forms a part of the ENSO (El-Nino/Southern Oscillation) phenomenon. For most trace gases, ocean is an important source or sink, depending on temperature. Convective activity associated with SST could modulate the atmospheric Hadley cell circulation as well as interaction between equatorial wind QBO and extra-tropical planetary waves. Gray and Chipperfield (1990) and Chipperfield and Gray (1992) gave an interactive radiative-dynamical-chemical model of the atmosphere, which predicts QBO in the time series of temperatures and chemical species in the troposphere and stratosphere. Kane (1994) made a preliminary attempt to identify such QBO in some trace gases. In this communication, QBO is revealed in the N_2O series at various locations and the QBO and QTO characteristics resemble those of the ENSO phenomenon rather than those of the stratospheric wind.

Acknowledgements

This work was partially supported by FNDCT Brazil under contract FINEP 537/CT.

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