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Quasi-biennial and quasi-triennial oscillations in atmospheric methane

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सार - 1983-92 के दौरान सम्पूर्ण विश्व में वायुमंडलीय मैथेन की वृद्वि की गति को कई स्थानों पर मापा गया और इसका स्पेकट्रल विश्लेषण किया गया। कई स्थानों पर महत्वपूर्ण अर्ध-द्वैवार्षिक (2-3 वर्ष) और अर्ध-त्रैवार्षिक (3-4 वर्ष) दोलनों का पता चला है किन्तु सभी स्थानों पर ये दोलन साथ-साथ नहीं देखे गए हैं। इनकी आवर्तिताएँ 2.10-2.44, 3.1-3.5 और 3.9-4.8 वर्षों के समूह के रूप में पाई गई है और कुछ स्थितियों में यह आवर्तिता 50 हेक्टापास्कल भूमध्य क्षेत्रीय पवन की 2.54 वर्ष और / या इनसो के ~ 2.30 वर्ष और ~ 4.5 वर्ष के अनुरूप थीं।

ABSTRACT. The growth rates of atmospheric methane measured at several locations distributed over the globe during 1983-92 were subjected to spectral analysis. Significant Quasi-biennial (2-3 years) and Quasi-triennial (3-4 years) oscillations were noticed at several locations but not simultaneously at all locations. The periodicities were bunched at 2.10-2.44, 3.1-3.5 and 3.9-4.8 years and, in some cases, matched with the 2.54 year periodicity of 50 hPa equatorial zonal wind and/or ~ 2.30 year and ~ 4.5 year periodicities of ENSO.

Key words — Quasi-biennial oscillation (QBO), El-Niño and Southern Oscillation (ENSO), Atmospheric methane

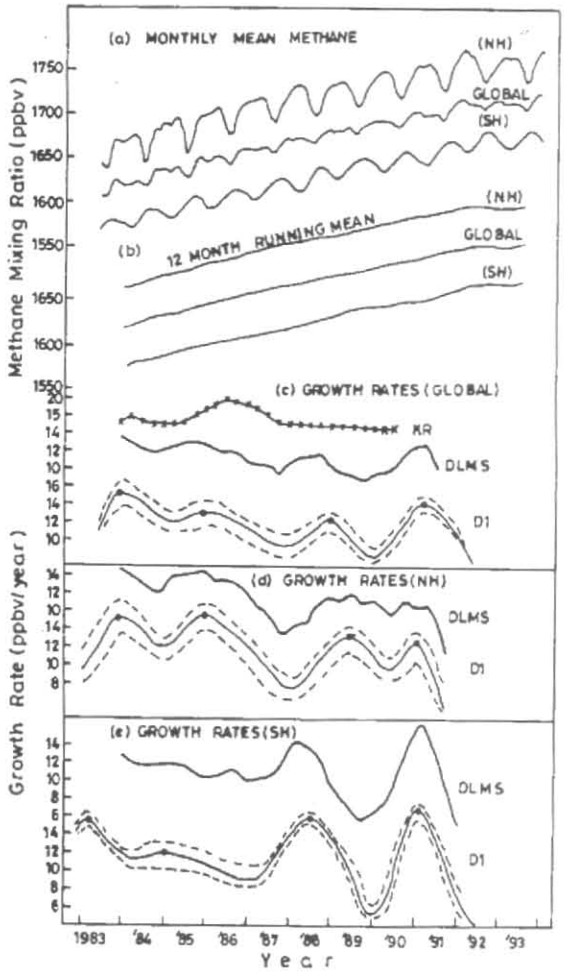
1. Introduction

The Climate Monitoring and Diagnostics Laboratory (CMDL) of the National Oceanic and Atmospheric Administration (NOAA), Air Resources Laboratory, Boulder, Colorado, USA, has been making meticulous measurements of several trace elements such as carbon dioxide, carbon monoxide, methane, ozone, surface aerosols, water vapour, nitrous oxide and halocarbons. Monthly means and results and analysis are reported in their summary reports and in several publications. In a recent publication (Kane, 1994), it was shown that the 12-month running means of most of these elements showed conspicuous fluctuations of a Quasi-biennial and Quasi-triennial nature. However, the peaks were not always well defined and the spacing varied in a very large range (20-50 months), probably because of complicating factors for individual locations. In some cases, the data length was very short. For example, for methane (CH_4) measured by the Carbon Cycle Division (led by P.P.Tans), we could use data only for 1984-90, for five locations BRW, MLO, SMO, CG and SPO. Recently, Dlugokencky *et al.* (1994a,b; henceforth referred to as D1, D2) presented results of methane measurements at 36 fixed sites and from regular shipboard samplings centered on 5° latitude intervals between 45°N and 35°S in the Pacific Ocean between North America and New Zealand (see details in Steele *et al.*, 1987, and also gave global and hemispherical averages for 1983-92. Bakwin *et al.* (1994) updated the analysis upto 1993. Also, Dlugokencky *et al.* (1995; henceforth referred to as D3) presented a detailed, updated (upto 1993) analysis specifically for atmospheric methane at Mauna Loa and Barrow. Another group at the Oregon Graduate Institute of Science and Technology, Portland, Oregon USA presented results for methane measured at six globally distributed locations and discussed their environmental implications (Khalil and Rasmussen, 1990, 1993; Khalil *et al.*, 1993).

Most of these studies fit the data with a quadratic polynomial which represents long-term trend and a series of four harmonics which represents the average seasonal

cycle and, when these are eliminated, the residuals are digitally filtered to determine inter-annual variability (Steele *et al.*, 1992). An oscillatory structure is often seen. There was a large fluctuation in the growth rate in 1988-89, opposite in sign for the northern and southern

hemisphere, indicating a ~10% increase in interhemispheric transport. It was attributed to the La Niña event. However, a similar fluctuation occurred two years later (1991-92), during an El Niño. Thus, these fluctuations may have some other origin besides ENSO.



Figs. 1(a-e). (a) Monthly means of global and hemisphere (NH = Northern Hemisphere, SH = Southern Hemisphere) methane, (b) 12-month running means, (c) Global growth rates (KR = Khalil and Rasmussen, DLMS = Present analysis of data of D1, Dlugokencky *et al.*, 1994 a) for methane, (d) Growth rates for northern hemispheric (NH) and (e) southern hemispheric (SH) methane

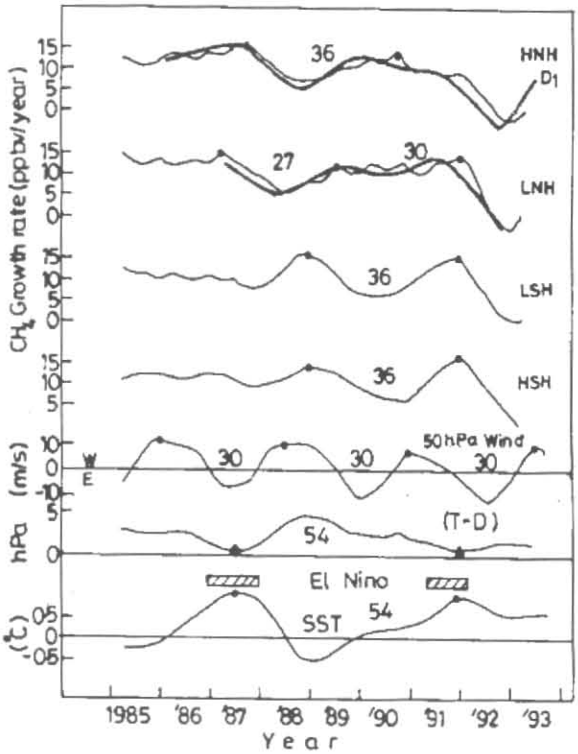


Fig. 2. Methane growth rates for northern high (HNH) and low (LNH) latitudes and southern low (LSH) and high (HSH) latitudes (thin line, present analysis; thick line, D1, Dlugokencky *et al.*, 1994a results) and plots of 12 month running means of 50 hPa zonal wind, Southern Oscillations Index represented by Tahiti minus Darwin atmospheric pressure difference (T-D) and equatorial eastern Pacific sea surface temperature SST. Hatched rectangles indicate moderate El Niños

Since there are ~ 4-5 maxima during 1983-93, possible QBO and/or QTO (Quasi-biennial and/or Quasi-triennial oscillations) are indicated. In this communication, the QBO and QTO are identified and their latitude dependence examined from the methane data at several globally distributed locations.

2. Data

Methane data were obtained mostly from Trends 93 (1994) as follows:

(*i*) Khalil and Rasmussen (henceforth referred to as KR) Oregon Graduate Institute for Science and Technology, Portland, Oregon 9721, USA. Data for Global CH_4, (average of Alaska, Cape Grim, Samoa, Cape Meares, Mauna Loa and Cape Kumukahi, South Pole and Mawson), Trends 93, page 271 and data for Cape Meares alone, Trends 93, page 364.

(*ii*) Dlugokencky Lang, Masarie and Steele (henceforth referred to as DLMS), ERL (Environmental Research Laboratories of NOAA/CMDL), Boulder, Colorado 80303-3328 USA and DAR (Division of Atmospheric Research, CSIRO, Aspendale Victoria, 3195, Australia). Data for 23 fixed sites and 14 shipboard Pacific Ocean latitudes and their global averages, Trends 93, page 265 and pages 274-350.

(*iii*) Brunke (CSIR, AIM-EMA TEK, Stellenbosch 7600, South Africa) and Scheel, Seiler (IFU, D-82467, Garmisch-Partenkirchen, Germany) (henceforth referred to as BSS). Data for Cape Point, Trends 93, page 355.

Basic data were monthly means. Data having many gaps were not used. A few missing values were interpolated by taking the means of the values for the previous and succeeding months. All data were from air samples analysed by gas chromatographs. Details of sampling and measurement procedures are given, for example, in D1, D2. Global CH_4 data were available from Trends 93 (pages 265 and 271), hemispheric data were provided privately by Dr. Dlugokencky.

3. Procedure of analysis

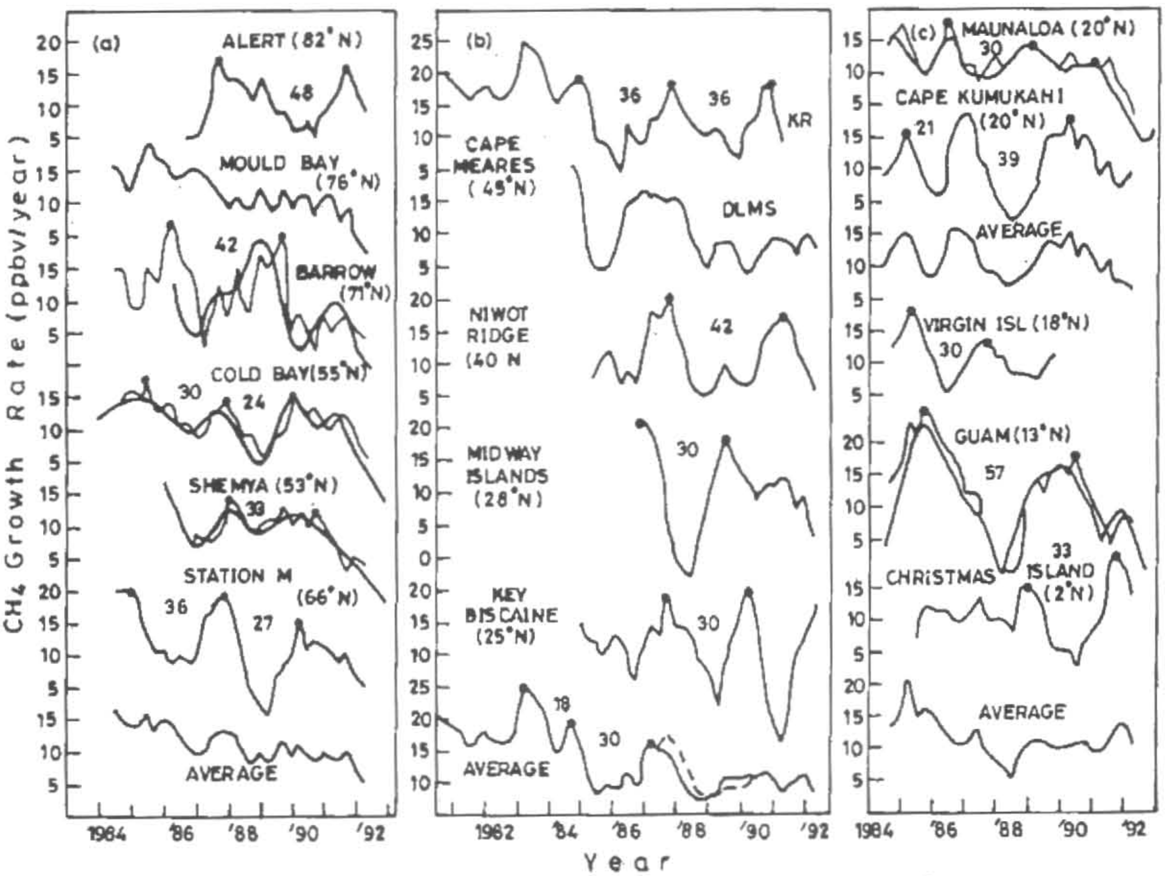
Fig. 1 illustrates the procedure of analysis. The second plot in Fig. 1(a) is for monthly means of global methane as given by DLMS in Trends 93 (page 265) and further data from private communication. Two types of variations are clearly seen *viz.* (*i*) a seasonal wave and (*ii*) a long-term uptrend. The latter has been discussed in detail in earlier papers (D1, D2 *etc.*), including the rapid decrease in growth rate in recent years. The seasonal variation is surprising as, on a global scale, the opposite seasonal cycles of the northern and southern hemispheres are expected to cancel out. However, as mentioned in D1, the seasonal cycle in northern hemisphere is larger than that in the southern hemisphere, thus, leaving a residue in their average. The first and third plots in Fig. 1(a) show the monthly means for northern hemisphere (NH) and southern hemisphere (SH) (as given in D1), obtained by combining data from individual locations weighted by the cosine of the station latitude. As can be seen, the amplitude of the NH seasonal cycle is almost double that of SH. Also, the mean value for NH is ~ 80 ppb more than that for SH, mainly because the methane source is mostly (75%) in the Northern Hemisphere. In NH, the maxima are broad and the minima are sharp, occurring mostly in July. In SH, maxima and minima are almost equally broad.

As mentioned earlier, most of the other workers fitted a quadratic polynomial to represent the long-term trend and a series of four harmonics to represent the seasonal cycle, subtracted these from the original values, digitally filtered the residuals and obtained the growth rates. We have adopted a simpler procedure *viz.* eliminating the seasonal cycle by calculating running averages over successive 12 month periods. Fig. 1(b) shows the 12-month running means centered 3 months apart (4 values per year). The plots are almost straight lines, but not quite. Small wiggles are superimposed, which represent the short-term inter-annual variability. To separate the same, other workers subtracted statistically estimated long-term trends. We adopted another procedure *viz.* obtaining the growth rates as differences of 12 month running means centered 4 seasons apart. For example, Fig. 1(b) has 4 values : 1987 (1), 1987 (2), 1987 (3), 1987 (4) and 4 values : 1988 (1), 1988 (2), 1988 (3), 1988 (4). The differences 1988 (1) minus 1987 (1), 1988 (2) minus 1987 (2), 1988 (3) minus 1987 (3) and 1988 (4) minus 1987 (4) give the growth rates as 4 values per year. Fig. 1(c) (thick line marked as DLMS) shows the growth rates so obtained for global methane for the DLMS data. Regarding error bars, the DLMS data have been subjected by the authors (D1, D2) to rigorous editing and selection and the precision of measurements is stated as ±0.2%. The monthly means are probably accurate to ±1 ppbv (in ~ 1700 ppbv). The 12-month running means would have still better accuracy and the growth rates obtained by us as differences of these 12-month running means should be accurate to less than 1 ppbv. It would be interesting to compare these with the growth rates obtained by D1, D2 with much more rigorous, sophisticated statistical methods. Their result is reproduced in Fig. 1(c), marked at D1 with $\pm\ \sigma$ uncertainties (dashed lines) determined by those authors (D1, D2) from the bootstrap method. As can be seen, the growth rate DLMS obtained by our simple methodology compares well with their growth rate (D1), certainly within their $\pm\ \sigma$ limits. It may be reasonable to presume that the error bars on our results are also of the same order *viz.* ~ ±1 ppbv / year in growth rates of ~10 ppbv/year.

The global methane monthly means have been reported by another group also *viz.* The Oregon Graduate Institute for Science and Technology and Khalil and Rasmussen have presented the monthly means in Trends 93 (page 271). The growth rate for these data are shown in Fig. 1(c) as the plot marked KR. These data are an average of six globally distributed locations *viz.* Alaska, Cape Grim, Samoa, Cape Meares, Mauna Loa (and Cape Kumukahi) and South Pole (and Mawson). The plot KR differs considerably and significantly from the plots DLMS and D1, leaving in doubt whether these global growth rates are really representative. In the growth rate plots DLMS and D1 (NOAA data, analysed by two different methods), there are four maxima during 1983-92), (marked by dots on the D1 plot) with spacings ~ 24, 36, 26 months (QBO, 2.0, 3.0, 2.2 years).

4. Hemispheric growth rates

D1 and Bakwin *et al.* (1994) have presented growth rates for northern hemisphere (NH) and southern hemisphere (SH) separately. Figs. 1(d&e) show the growth rates obtained by our method (thick lines, marked



Figs. 3(a-c). Growth rates of methane for : (a) Northern high latitudes (53°-82°N), (b) Northern middle latitudes (25°-45°N) and (c) Northern low latitudes (2°-20°N). Thin lines, present analysis. Thick lines, results of D1, D2, D3 (Dlugokencky *et al.*, 1994a,b; 1995)

DLMS) and those reported in D1 and Bakwin *et al.* (1994), (thin line, escorted by dashed lines showing $\pm\sigma$ limits). The DLMS plots match very well with the D1 plots, again indicating that our simple methodology gives results very similar to those obtained by the sophisticated method. However, as noted by D1, the patterns of NH and SH growth rate interannual variability are very different from each other. In particular, during 1987-89, the NH growth rate decreased while the SH growth rate increased. Bakwin *et al.* (1994) estimated that there was an enhancement of ~12% over average interhemispheric transport during 1988-89, Steele *et al.* (1992) attributed it to the La Niña event. From the D1 plot in Fig. 1(d), the NH growth rates have 4 peaks, almost similar to those for global growth rate [D1 in Fig. 1(c)]. From the D1 plot of Fig. 1(e), the SH growth rates also have 4 peaks. The first two (during 1983-85) had a spacing of ~2 years, same as that for NH plot but occurred ~1 year earlier. The latter two (during 1988-92) had a larger spacing (~30 months, 2.5 years). Whether this was due to the La Niña event, is a moot question. In our method using running means, half year data are lost on either end and hence, 1992 data are not studied fully. However, D1 have pointed out that for the southern hemisphere, the maximum in growth rate in 1991 did not correlate with a local minimum in the northern hemisphere growth rate or a La Niña; it occurred during an El Niño. Thus, the effects of La Niña or El Niño on the interhemispheric transport do not seem to be well defined.

5. Semi-hemispheric growth rates

Since the growth rates in the northern and southern hemispheres were different, D1 sought additional information from analysis of semi-hemispheric averages. Since their main interest was in the long-term growth rates, they applied a low-pass filter to remove much of the inter-annual variability. However, in D3, the growth rates of Barrow and Mauna Loa are compared with HNH and LNH. Fig. 2 shows the growth rates for high (H) and low (L) latitudes in the northern (HNH, LNH) and southern

(HSH, LSH) hemispheres. The plots for the southern hemisphere low (LSH) and high (HSH) latitudes are similar and show two clear peaks with a spacing of ~36 months (3.0 years). The plots for the northern hemisphere low (LNH) and high (HNH) latitudes are not alike and while some peaks are seen, the peak separations are different, 36 months for HNH and 27 and 30 months for LNH. The thick lines represent the HNH and LNH given in D3 and compare well with our thin line plots, indicating again that our simple method gives results similar to those obtained by using sophisticated methods. In any case, these plots show that the inter-annual variability might have finer latitude dependence. Hence, data for individual locations need to be examined. The bottom plots in Fig. 2 refer to two phenomena well-known for having QBO and QTO *viz.* Stratospheric low latitude zonal wind and ENSO (El Niño/Southern Oscillation). The plots show 50 hPa zonal wind (Venne and Dartt, 1990 updated), Southern Oscillation Index (SOI) represented by Tahiti minus Darwin atmospheric pressure (T-D), (Meteorological Data Reports) and equatorial eastern Pacific SST (sea surface temperature, Dr. Angell, private communication), with El Niños (EN) interposed as rectangles (Quinn *et al.*, 1987). As expected, the moderate El Niños of 1987 and 1991 coincided with (T-D) minima and SST maxima. During 1985-93, the 50 hPa wind had 4 (westerly) maxima with spacings 30, 30, 30 months while (T-D) had two minima and SST had two maxima with a spacing of ~54 months. The peaks in CH_4 do not seem to match with any of these.

6. Growth rates at individual locations

Fig. 3 shows plots of growth rates for individual locations. Fig. 3(a) refers to the northern high latitude locations Alert (82°N), Mould Bay (76°N). Barrow (71°N), Cold Bay (55°N), Shemya Island (53°N) and Station M (66°N). For Barrow, Cold Bay and Shemya Island, the superposed thick lines are the growth rates obtained by sophisticated methods reported in D1, D2, D3 and match well with our thin line plots. However, the various plots at different locations differ considerably from each other.

As mentioned in D3, the CH_4 time series are a combination of three primary factors *viz.* long-term trends due to small imbalance between sources and sinks; annual cycles due to seasonality in atmospheric transport and in some sources and the photochemical sink; short-term variations (days to weeks) due to variations in transport at the sampling sites. As 12 month running means are used, the latter two factors are most probably eliminated. Detailed information on temporal changes in CH_4 sources and sinks is not available. But some possibilities are: changes in atmospheric [HO] which change the magnitude of the chemical sink (Madronich and Granier, 1992; Prinn *et al.,* 1992), changes in emissions from cattle and other ruminant animals and rice agriculture (Khalil and Rasmussen, 1993), and changes in emissions from fossil fuel sources (Steele *et al.*, 1992; D1, D2, D3). There is no particular reason to suspect instrumental vagaries. Hence, the diversity of Fig 3(a) should be indicative of highly localized variability of sources and sinks, with no pattern representative of large areas. In some individual plots, distinct peaks are observed with spacings in the range (24-48) months, thus, involving QBO, QTO and probably larger periodicities. These may or may not be random, but certainly are not persistent in time or space, so that the average plot (Fig. 3a, bottom) shows virtually no clear peaks, only a decreasing trend. We conclude, therefore, that in the high latitudes of the northern hemisphere, there is no overall, consistent pattern of QBO and QTO in the inter-annual variability.

Fig. 3(b) refers to middle latitudes of the northern hemisphere. For Cape Meares (45°N), two data sets were available, one from the Oregon group (KR, 1979-91, Trends 93, page 364) and the other from the NOAA/CMDL group (DLMS, 1983-92, Trends 93, page 297). For the overlapping period 1983-92, the two plots match with each other but only roughly. The plots for Niwot Ridge (40°N) and Key Biscayne (25°N) also compare reasonably with the Cape Meares plots. But the plot for Midway Islands (28°N) is very different. The bottom plot of Fig. 3(b) shows the average and the dashed portion represents the average excluding Midway Islands but is not very different from the full line. The average does not show prominent QBO; only a decreasing trend is indicated.

Fig. 3(c) refers to low latitudes of the northern hemisphere. The top plot is for Mauna Loa (20°N) with our result (thin line) matching very well with the result of D2 (thick line). However, both these differ considerably from the second plot for Cape Kumukahi (20°N). These two sites are on the island of Hawaii, only 90 km apart but separated in altitude by 3394 m and D1 have examined the vertical gradients and seasonal cycles and found some differences. However, 12 month running means are not expected to show great differences. The match between the thin line and thick line for Mauna Loa shows that errors related to methodology are less than 2 ppbv/year of growth rate. The difference between the growth rate patterns of Mauna Loa and Cape Kumukahi is much larger, (8-10 ppbv/year), throwing in doubt the credibility of these patterns as representative of that region. The third plot in Fig. 3(c) is the average of Mauna Loa and Cape Kumukahi and shows much reduced variability.

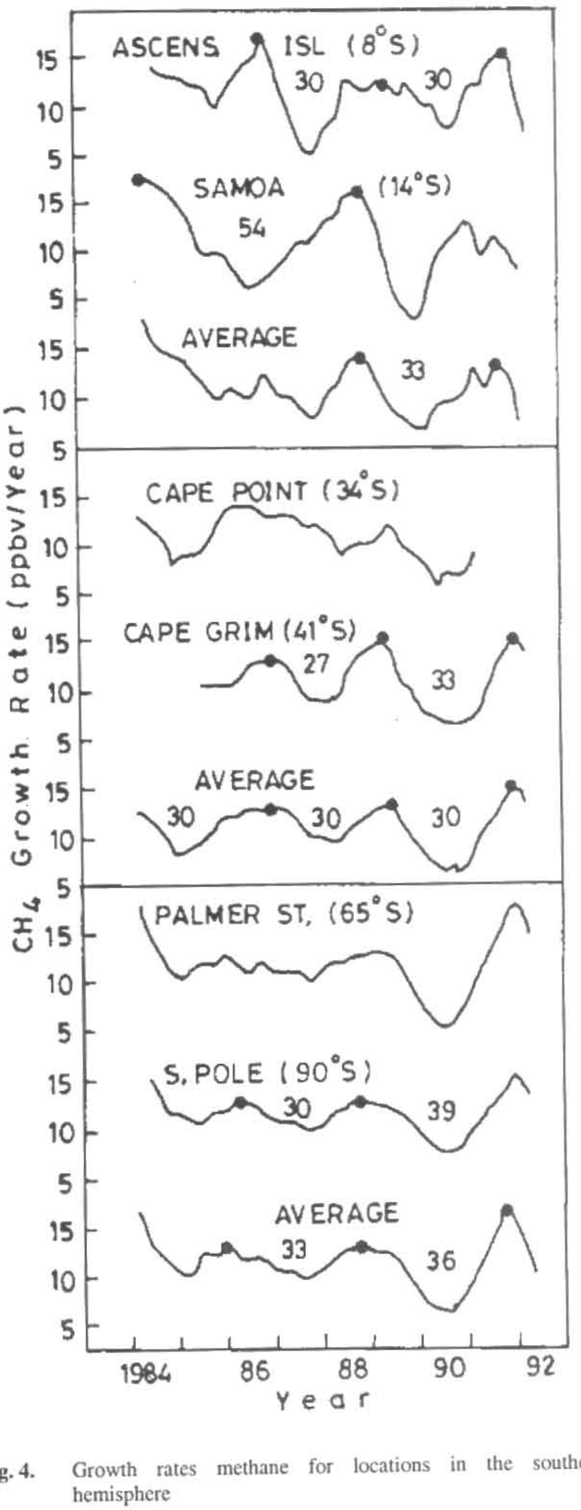


Fig. 4. Growth rates methane for locations in the southern hemisphere

The lower part of Fig. 3(c) shows growth rates for Virgin Islands (18°N), Guam (13°N) and Christmas Island (2°N). For Guam, our plot (thin line) is very similar to that of D2 for Guam (thick line). But both these differ considerably from the plots for Virgin Islands and Christmas Island. The bottom plot is the average and shows no clear QBO peaks; only a sharp decrease in the first half and a steady level in the latter half, is indicated.

Fig. 4 shows growth rate plots for locations in the southern hemisphere. The top two plots are for Ascension Island (8°S) and Samoa (14°S) and are not similar. The average (third plot) shows a small variability. The fourth and fifth plots are for Cape Point (34°S) and Cape Grim (41°S) and are dissimilar. Their average (sixth plot) shows a small variability but peak spacings of ~30 months. The seventh and eighth plots for Palmer Station (65°S) and South Pole (90°S) are fairly similar and their average (ninth plot) shows peak spacings of 33 and 36 months. Thus, in the southern hemisphere, the averages do show some residual QBOs. But the stations are very few. Also, peaks at different southern latitude belts (low, middle, high) do not tally completely, and the overall average has very little QBO.

In many plots in Figs. 3 and 4, individual locations show peaks with spacings in a wide range (21-57 months), which could be random or indicative of intermittent QBO and QTO. It is necessary to have quantitative statistical estimates to check whether any of these are statistically significant.

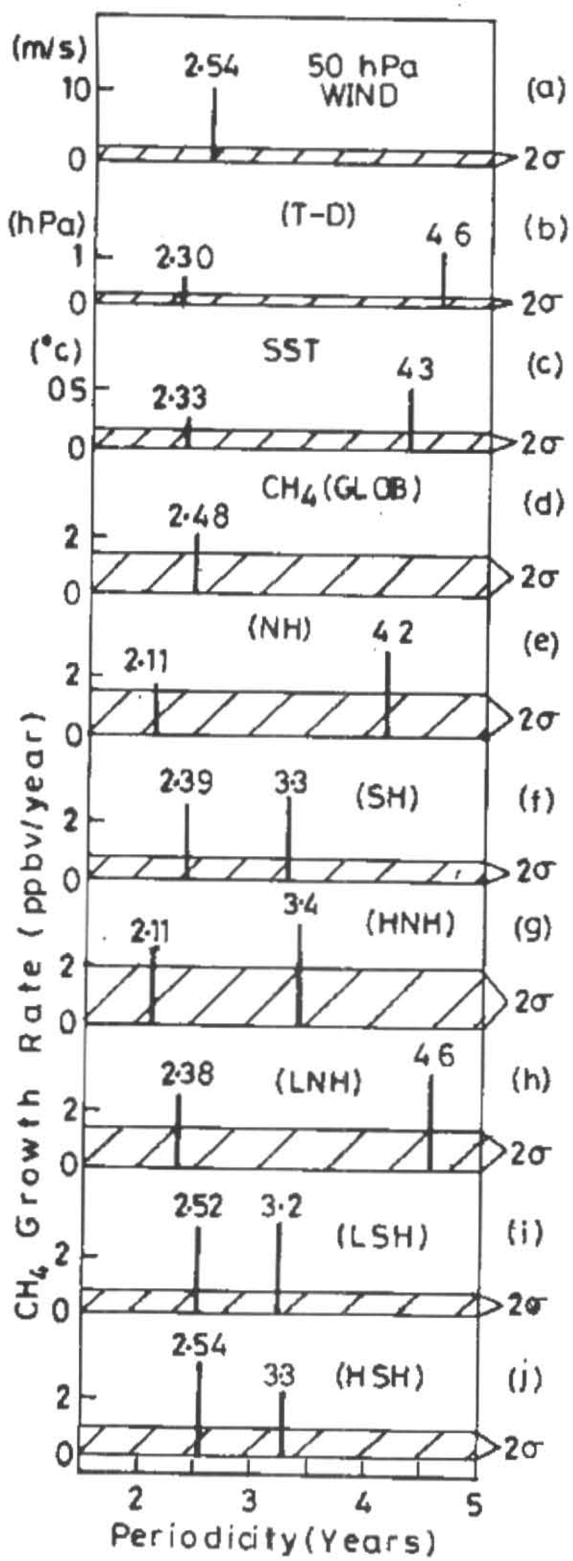
7. Power spectrum analysis

All the series mentioned above were subjected to MESA (Maximum Entropy Spectral Analysis), Burg, (1967); Ulrych and Bishop (1975) which is much more accurate than the conventional BT (Blackman and Tukey, 1958) method. In BT, only frequencies $f/2$m, (f=1,.....m) can be studied where the lag m is recommended to be ~25% of data length. In MESA, an equivalent parameter is LPEF (Length of the Prediction Error Filter). With low LPEF, small periodicities are resolved. Higher LPEFs resolve larger periodicities; but smaller periodicities suffer peak splitting. A compromise LPEF is ~50% of data length. In MESA, the amplitude (or Power) estimates are unreliable (Kane, 1977; Kane and Trivedi, 1982). Hence, MESA was used only for detecting the peaks T_k (k=1 to n) and the T_k for every series were used in the expression:

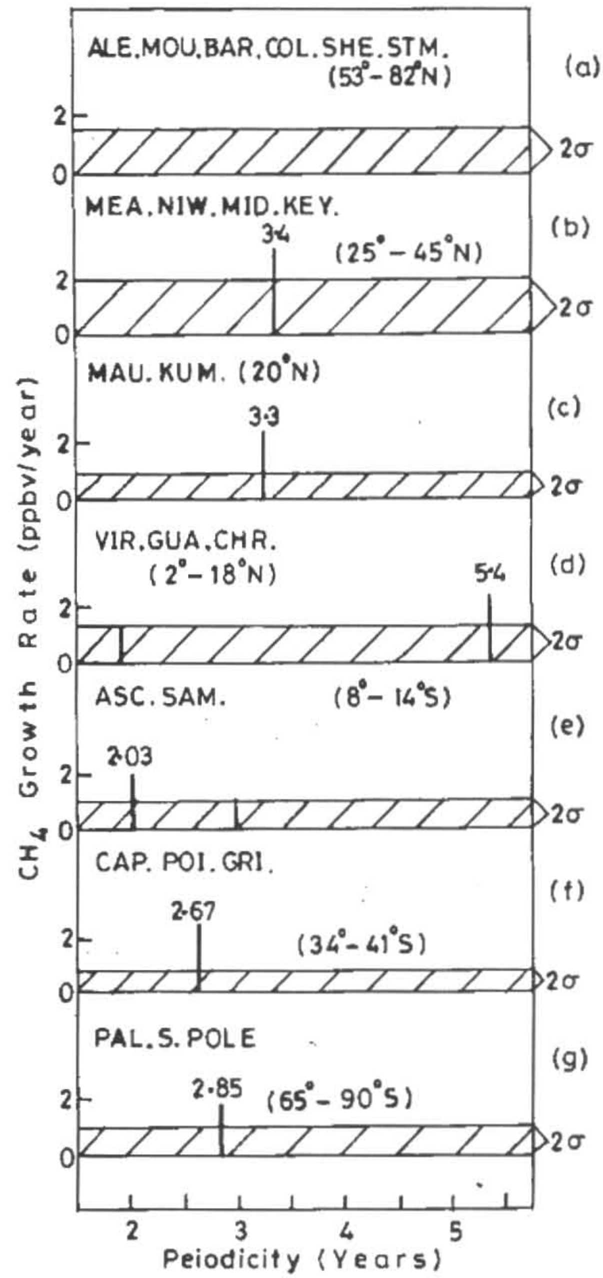
$$f(t) = A_0 + \sum_{k=1}^{n} \left[a_k \mathrm{Sin}\left(2\pi t / T_k\right) + b_k \mathrm{Cos}\left(2\pi t / T_k\right)\right] + E$$

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

where $f(t)$ is the observed time series and E is the error factor. A Multiple Regression Analysis (Bevington, 1969)



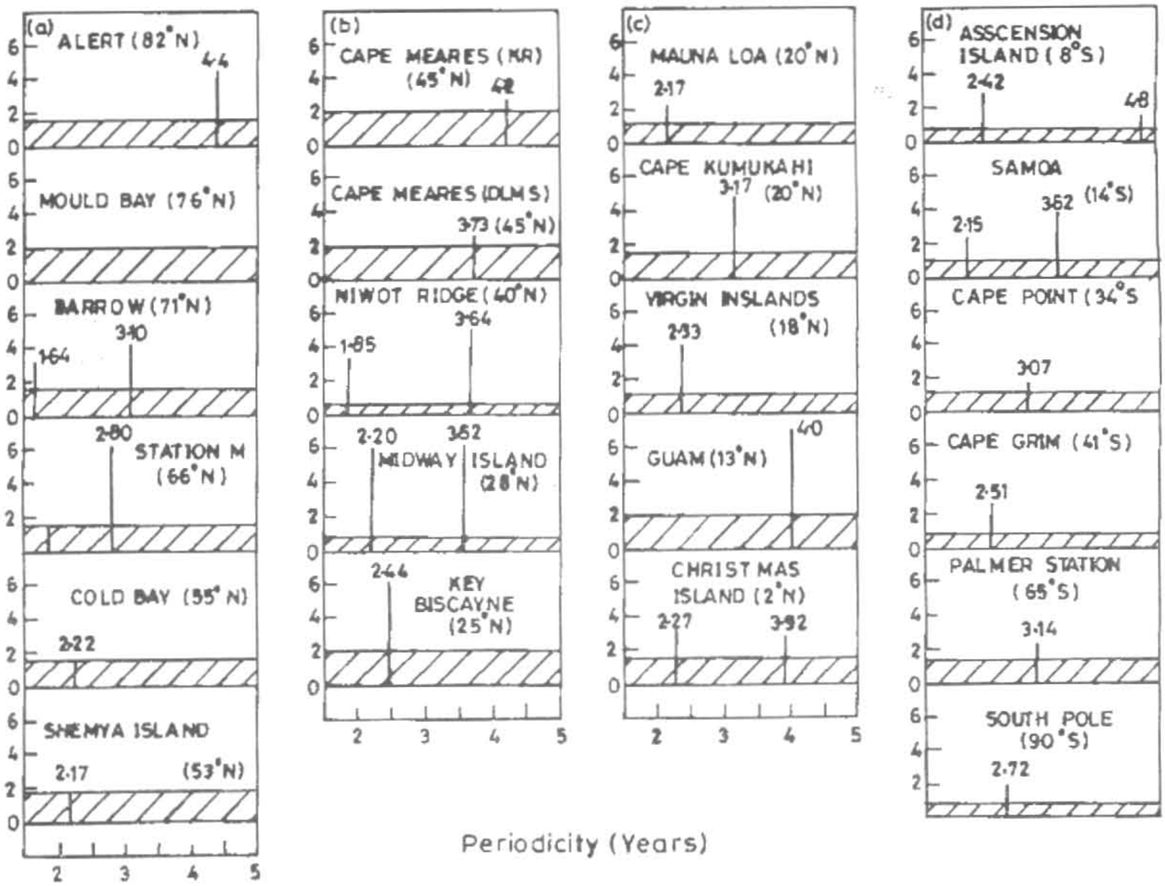
Figs. 5(a-j). Spectra (amplitudes of the various periodicities obtained by MESA and MRA) for (a) 50 hPa wind, (b) (T-D), (c) SST, (d) Global methane growth rates and (e-j) hemispheric and semi-hemispheric methane growth rates. The hatched portion indicates 2σ limits



Figs. 6 (a-g). Same as Fig. 5, regional methane growth rate averages : (a) 53°-82°N, (b) 25°- 45°N, (c) 20°N , (d) 2°-18°N, (e) 8°-14°S, (f) 34°-41°S, (g) 65°-90°S, from locations as indicated

was then carried out to estimate A_0, (a_k, b_k) and their standard errors (by a least-square fit). From these, r_k and their standard error σ_k (common to all r_k, in this methodology) was estimated and r_k exceeding $2\sigma_r$ were accepted as significant at a 95%(a priori) confidence level.

Fig. 5 shows the amplitudes of the various periodicities. Fig. 5(a) refers to the 50 hPa zonal wind and



Figs. 7 (a-d). Same as Fig. 5, for methane growth rates at individual locations in : (a) 53°-82°N, (b) 25°-45°N, (c) 2°-20°N and (d) 8°-90°S

shows one prominent QBO peak at 2.54 years, agreeing with the ~30 months spacing seen in Fig.2 (the hatched markings in Fig. 5 indicate the 2σ limit). Fig. 5(b) refers to the Southern Oscillation Index (T-D) and shows a prominent peak at 4.6 years and a minor peak at 2.30 (QBO), the former agreeing roughly with the 54 month spacing seen in Fig. 2. Fig. 5(c) refers to the equatorial eastern Pacific SST and shows a prominent peak at 4.3 years and a minor peak at 2.33 years (QBO). Thus, whereas (T-D) and SST resemble each other and have prominent peaks near 4.5 years and minor QBO (~2.30 years), these are different from the single major QBO peak of the 50 hPa wind at ~2.50 years.

Figs. 5(d-j) refer to CH_4 at different latitude zones. Thus, Fig. 5(d) refers to global methane and has only one significant peak at 2.48 years. On the other hand, northern hemisphere (NH, Fig. 5e) shows a major peak at 4.2 years and a minor, barely significant QBO at 2.11 years, while southern hemisphere (SH, Fig. 5f) shows major peaks at 2.39 years (QBO) and 3.3 years (QTO). MESA is very accurate at low periodicities and the difference between NH and SH is significant and substantial and neither of the two resembles 50 hPa wind or (T-D), SST completely. Figs. 5 (g-j) refer to semi-hemispheric methane (HNH, LNH, LSH, HSH). Whereas the southern semi-hemispheres (LSH, HSH) show similar results (QBO, ~2.50 years and QTO, ~3.2 years), the northern semi-hemispheres show very different results (HNH, 2.11 and 3.4 years; LNH, 2.38 and 4.6 years) from each other and from the southern hemisphere. Thus, the northern hemisphere methane characteristics should be highly variable with latitude, while southern latitude characteristics are not very much latitude dependent.

Fig. 6 shows the spectra for regional averages. Fig. 6(a) refers to the average of high northern latitude locations *viz.* Alert, Mould Bay, Barrow, Cold Bay, Shemya Island and Station M (53° -82°N). There is no significant periodicity in this average, indicating that there

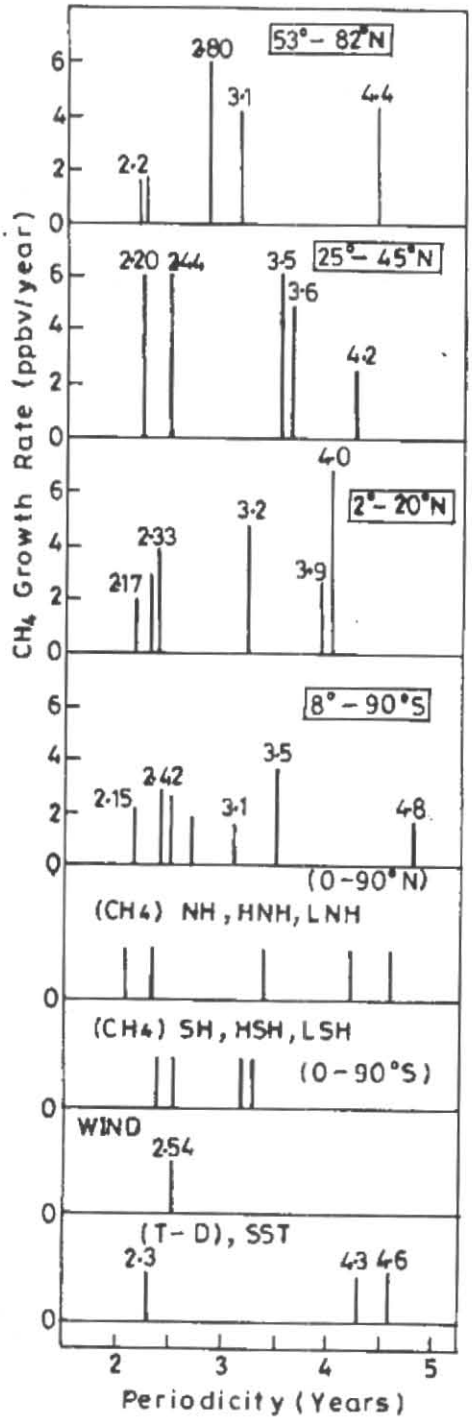


Fig. 8. Same as Fig. 5, for methane growth rates at individual locations superposed for four latitude ranges 53°-82°N, 25°-45°N, 2°-20°N and 8°-90°S, values for northern and southern hemisphere methane (0-90°N and 0-90°S) and spectra for 50 hPa wind and (T-D), SST

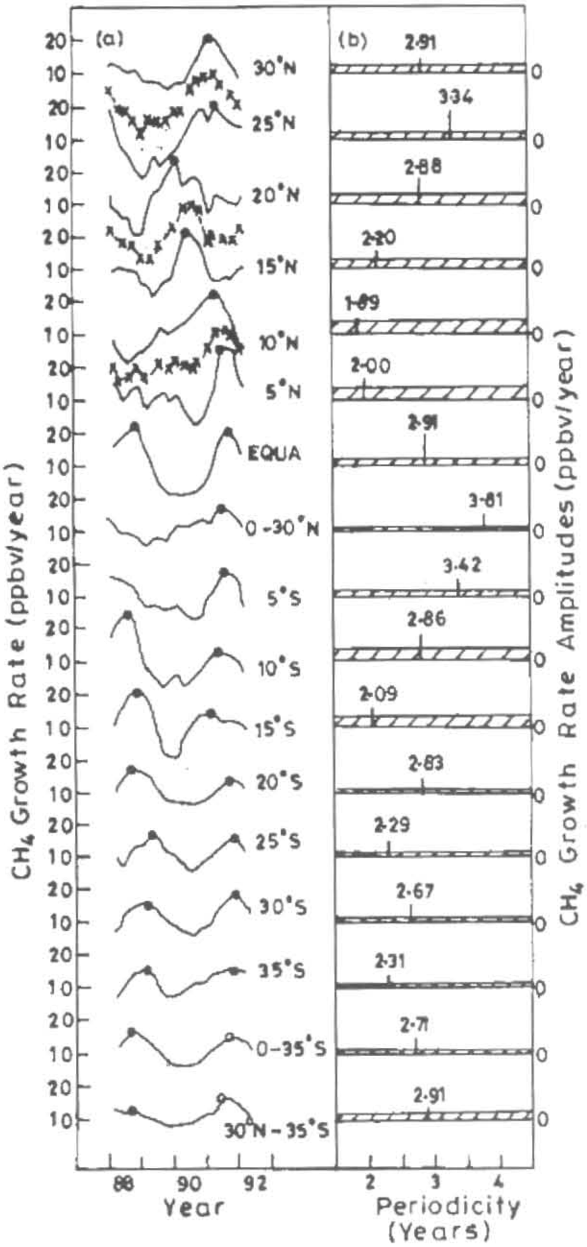
is no pattern for this region as a whole (as was obvious from the bottom plot of Fig. 3a). We do not know what locations were selected by D1, D2, D3 for the NH and HNH groups, which showed significant periodicities at 2.11, 3.4 and/or 4.2 years [Figs. 5(e & g)]. Since, for averaging, the weighted data by cosine of station latitude, the contribution from these high latitude stations must have been very small.

Fig. 6(b) refers to the average of northern mid-latitude locations *viz.* Cape Meares, Niwot Ride, Midway Islands and Key Biscayne (25°-45°N). There is a significant peak at 3.4 years. Fig. 6(c) refers to the average of Mauna Loa and Cape Kumukahi, and also shows a significant peak at 3.3 years. Thus, these northern middle latitude locations have a significant QTO at ~3.3 years. For lower northern latitudes (Fig. 6d), there is no significant peak except at 5.4 years, not a QBO or QTO but probably reflecting the non-uniform long-term trend (see Fig. 3c, bottom plot).

Figs. 6 (e-g) refer to the southern hemisphere and show significant QBO peaks near 2.00 years (a biennial oscillation) for the average of southern low latitudes *viz.* Ascension Island and Samoa, 2.67 year for the average of southern middle latitudes *viz.* Cape point and Cape Grim and 2.85 years for the average of southern high latitudes *viz.* Palmer Station and South pole.

Fig. 7 shows the spectra for individual locations. Fig. 7(a) refers to locations in (53°-82°N) belt. Alert (4.4 years), Barrow (3.10 years) and Station M (2.80 years) peaks are highly significant. Cold Bay (2.22 years), Shemya Island (2.17 year) are barely significant and Mould Bay has no significant peaks. In Fig. 7(b) for (25°-45°N), Cape Meares (KR and DLMS) show different peaks, at 3.73 and 4.20 years. Niwot Ridge and Midway Island have peaks near 3.60 years as also near 2.00 years, while Key Biscayne has one strong peak at 2.44 years. In Fig. 7(c) for (2°-20°N), Mauna Loa (2.17 years) and Cape Kumukahi (3.17 years) show radically different patterns, though the two locations are very near to each other. Virgin Islands (2.33 years) and Christmas Island (2.17 years) have similar QBO while Guam (4.00 years) and Christmas Island (3.92 years) have similar QTO. In Fig. 7(d) for the southern hemisphere, QBO peaks are seen at 2.15, 2.42, 2.51, 2.72 years and QTO peaks at 2.07, 3.14, 3.52 years and a peak at 4.8 years with no preference for any latitude in particular.

Fig. 8 shows a consolidated picture for the four latitude zones of Fig. 7. In general, peaks bunch at (2.10-2.44) years, (3.1-3.5) years and (3.9-4.8) years. The lower half of Fig. 8 shows the occurrences in the northern (NH, HNH, LNH) and southern (SH, HSH, LSH) hemisphere methane and in 50 hPa wind and ENSO (T-D, SST). Whereas some methane QBO match with wind or ENSO QBO, the (3.1-3.5) year methane peaks have no match in



Figs. 9 (a&b). (a) Methane growth rates for Pacific Ocean sites, 30°N-35°S. Thick lines are averages for 0-30°N and 0-35°S (b) Spectra (amplitudes of the various periodicities obtained by MESA and MRA) for the methane growth rate series at the Pacific ocean sites

wind or ENSO. Even though the data used for spectral analysis are seasonal points (4 points per year), the values mentioned here are averages for a roughly 10 year period (about 40 data points), and trials with artificial samples have shown that the periodicities are accurate to better than ± 0.05 years. Hence, 2.10 is certainly different from say, 2.40, and both should have different origins.

In the data set, analysed so far, there is a sharp decline in the growth rate during 1991-92. However, the wavy structures (QBO, QTO) occur before this period. The papers D1 and Bakwin *et al.* (1994) refer to 1993 data and show a substantial recovery of the growth rates in 1993 for the northern and southern hemisphere averages in general and for Barrow and Mauna Loa in particular.

8. Pacific ocean data

Air sampling of CH_4 for the NOAA/CMDL program has been carried out since December 1986 by the merchant vessel Southland Star and since May 1990 also by Wellington Star. The ships sail from Los Angeles southward, passing near Christmas Island, American Samoa and Fiji en route to Auckland, New Zealand and then back passing Hawaii en route to Seattle. Data are available for 35°S to 30°N at 5° latitude intervals. The frequency of sampling is ~3 weeks per latitude per ship, with a minimum of two longitudes for a given latitude. Longitudinal variations of CH_4 are considered negligible and monthly means for different latitudes are given by DLMS in Trends 93 (pages 322-327).

Fig. 9(a) shows the growth rates. The data length (1987-92) is rather small and in the method of running averages, one year of data are lost. Hence, only ~2 waves of QBO can be detected. In the northern hemisphere, the growth rate patterns varied considerably with latitude. For 30°N and 25°N, the patterns had a clear maximum in the middle of 1991. (The dashed plot is an average of 30°N and 25°N). However, for 20°N and 15°N, the maximum occurred about an year earlier (the dashed plot in between is an average of 20°N and 15°N). For 10°N and 5°N, the maximum occurred later, in the middle of 1991. At equator, one maximum was in the middle of 1991 while another occurred almost 3 years earlier, by the end of 1988. The thick plot for average of 0° -30°N shows a small variation but a clear maximum in the middle of 1991. In the southern hemisphere, patterns are more uniform and similar at different latitudes *viz.* A maximum in 1988-89 and then another in 1991-92, also reflected in the thick plot for average 0-35°N, and to a smaller extent in the overall average (30°N-35°S) in the bottommost plot of Fig. 9(a).

Fig. 9(b) shows the spectra, with peaks at (2.88-3.34) years for 30° -20°N, (1.89-2.20) years for 15°-5°N, (2.86-3.81) years for 0° -10 °S and (2.09-2.83) years for 15°-35°S. There is a latitudinal variation, with higher periodicities near equator and again at high northern latitudes, but not so much at southern high latitudes. It would be interesting to check whether these patterns were maintained for data for 1993 onwards.

9. Conclusions and Discussion

The growth rates of methane at different locations for 1983-92 were subjected to Maximum Entropy Spectral Analysis. In general, QBO (2.10-2.44 years), QTO (3.1-3.5 years) and larger periodicities (3.9-4.8 years) were significant. But there was no clear latitude dependence. There was occasional similarity with the 50 hPa low latitude zonal wind QBO (2.54 years) and/or with Southern Oscillation Index QBO (~2.30 years) and larger periodicity (4.3-4.6 years).

As mentioned in D1, the methane concentration at each location is a result of sources, sinks and atmospheric transport, integrated over various temporal and spatial scales. In the high latitudes of the southern hemisphere, there are no significant sources of CH_4 and it is very well mixed over wide areas. In tropical regions, wetlands, grazing livestock, termites, biomass burning, chemical destruction and transport, all contribute to seasonal cycles. The northern hemisphere is the source of ~75% of global methane (Fung *et al.*, 1991) and variations can occur due to all the factors mentioned above. However, the major changes are seasonal. Our analysis uses 12 month running means and hence, these seasonal effects should be obliterated. For studying growth rates and long-term trends. D1, D2, D3 have used sophisticated methods of analysis. However, earlier Steele *et al.* (1987) had used 12 month running means to remove seasonal cycle and to determine the trend. This method has the advantage of simplicity but has the major drawback of losing 6 months of data at each end of the record. Mitchell *et al.* (1966) have pointed out other potential problems with the running means. However, as stated in D1, results of trends calculated with the running mean are almost identical to those calculated by the sophisticated methods. Our growth rate plots are also similar to those in D1, D2, D3 and Bakwin *et al.* (1994). Hence, the differences in growth rate patterns at the various locations cannot be an artifact of the data analysis methods. For long-term trends, D1 note that the smoothed growth rate has decreased atleast 3 times faster in the high northern latitudes (30^0-90°N) than in the other three semi-hemispheres and attribute this to changes in highly populated, industrialized regions, related to fossil fuel use. There is also evidence that rates of increase in emissions from cattle, domestic animals, and rice agriculture have declined (Khalil and Rasmussen, 1993). As mentioned in D1, the dramatic changes in 1992 could also be related to the global cooling and changes in precipitation patterns resulting from the eruption of Mount Pinatubo in mid-1991; because methane emissions from wetlands are strongly dependent on soil moisture content and surface temperature. (The eruption of Mt. Pinatubo in June 1991 resulted in a decrease in surface air temperature of 0.7°C in the northern hemisphere in 1992, Dutton and Christy, 1992). The large changes in 1988-89 are considered to be consistent with an increased inter-hemispheric transport during the La Niña event at that time (Steele *et al.*, 1992). In this scenario, the methane QBO and QTO observed intermittently in both space and time seems to be an enigma. Which sources and/or sinks or transport mechanisms could have these characteristics? This needs further exploration. The data length used here is also small. Further data need to be analysed to see which of these patterns have persisted, if any.

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