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Quasi-biennial and quasi-triennial oscillations in the growth rates of atmospheric chlorofluorocarbons 11 and 12

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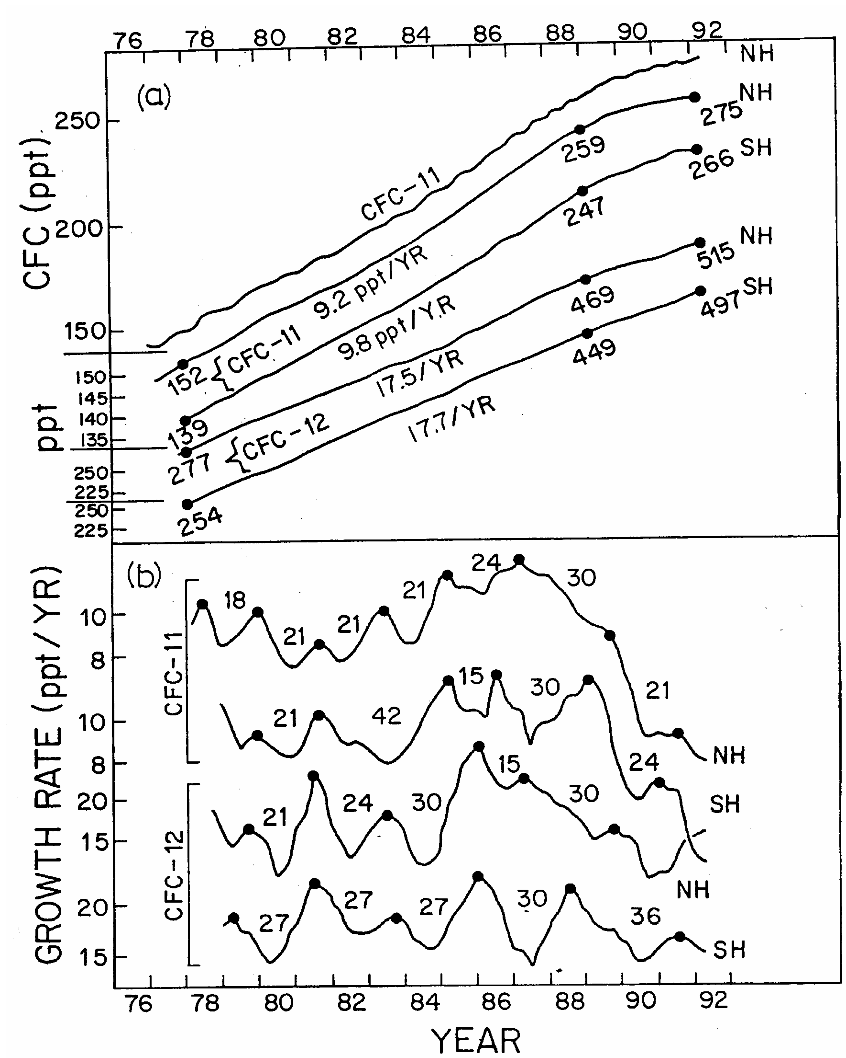
सार - 1977 से 1992 तक की अवधि के सी.एफ.सी.- 11 और सी.एफ.सी.-12 के बारहमासी निरंतर माध्यों की जाँच की गई है। पहले के शोधकर्त्ताओं द्वारा किए गए प्रेक्षणों के समान ही वर्ष 1977-1988 के दौरान इन यौगिकों में तीव्र गति से उत्तरी गोलार्द्ध में ~70 प्रतिशत और दक्षिणी गोलार्द्ध में ~77 प्रतिशत की रैखिक बढ़ोतरी पाई गई है। वर्ष 1988 से लेकर 1992 तक उत्तरी गोलार्द्ध के सी.एफ.सी.- 11 में वृद्धि की गति अपेक्षाकृत धीमी थी। इस पैटर्न में क्यू.बी.ओ. (अर्द्धवार्षिक दोलन) और क्यू.टी.ओ. (अर्द्धत्रैवार्षिक दोलन) की प्रधानता थी। विभिन्न श्रृंखलाओं के स्पेक्ट्रल विश्लेषणों से नीचे लिखी बातों का पता चला है। 50 हैक्टापास्कल गति की निम्न अक्षांशीय क्षेत्रीय पवनें 2.58 वर्षों की अवधि में एक प्रमुख क्यू.बी.ओ. की अधिकतम और इसी तरह से (क्यू.बी.ओ.) 5.1 वर्षों की अवधि में 2.00 की अपेक्षाकृत कम अधिकतम पाई गई है। ताहिती माइनस डार्विन वायुमंडलीय दाब (T-D) द्वारा प्रस्तुत किए गए दक्षिणी दोलन इंडैक्स के अनुसार ये 4.1 वर्षों में प्रमुख रूप से अधिकतम और 2.31 वर्षों में अपेक्षाकृत कम अधिकतम पाई गई है। दक्षिणी गोलार्द्ध में सी.एफ.सी.-11 की 3.7 वर्षों में केवल एक उल्लेखनीय अधिकतम होता है जो प्रायः 4.1 वर्ष (T-D) के अधिकतम से मामूली रूप से मेल खाते हैं। सी.एफ.सी.-12 की प्रमुख क्यू.बी.ओ. दोनों गोलार्द्ध में (2.16-2.33वर्ष) अवधि में और अलग-अलग स्थानों के लिए सी.एफ.सी.-11 (1.95-3.07 वर्ष) की रेंज में क्यू.बी.ओ. के नाममात्र के महत्व का दक्षिणी गोलार्द्ध में क्यू.टी.ओ. (3.6 वर्ष) में पता चलता है। जबकि (1.86-2.38 वर्ष) की अवधि की रेंज में सी.एफ.सी.-12 की प्रबल क्यू.बी.ओ. का पता चला है। सी.एफ.सी.-11 और सी.एफ.सी.-12 समय श्रृंखलाओं के आकलनों में स्पेक्ट्रल विशिष्टताओं की भिन्नताएँ उनके जीवन कालों (44 और 180 वर्ष), स्रोत उत्सर्जन गति और संचालन प्रक्रिया से प्रभावित होती है।

ABSTRACT. The 12-monthly running means of CFC-11 and CFC-12 were examined for 1977-1992. As observed by earlier workers, during 1977-1988, there was a rapid, almost linear increase of these compounds, ~70% in the northern and ~77% in the southern hemisphere. From 1988 up to 1992, growth rates were slower, more so for CFC-11 in the northern hemisphere. Superposed on this pattern were QBO, QTO (Quasi-Biennial and Quasi-Triennial Oscillations). A spectral analysis of the various series indicated the following. The 50 hPa low latitude zonal wind had one prominent QBO peak at 2.58 years and much smaller peaks at 2.00 (QBO) and 5.1 years. The Southern oscillation index represented by (T-D), Tahiti *minus* Darwin atmospheric pressure, had a prominent peak at 4.1 years and a smaller peak at 2.31 years. CFC-11 had only one significant peak at 3.7 years in the southern hemisphere, roughly similar to the 4.1 year (T-D) peak. CFC-12 had prominent QBO (2.16-2.33 years) in both the hemispheres and a QTO (3.6 years) in the southern hemisphere. For individual locations, CFC-11 showed barely significant QBO in the range (1.95-3.07 years), while CFC 12 showed strong QBO in the range (1.86-2.38 years). The difference in the spectral characteristics of CFC-11 and CFC 12 time series is attributed to differences in their lifetimes (44 and 180 years), source emission rates and transport processes.

Key words – CFC-11, CFC-12, QTO, QBO, ENSO.

1. Introduction

The National Oceanic and Atmospheric Administration's (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL) at Boulder, Colorado, USA, has been collecting CFC-11 (CCl_3F) and CFC-12 (CCl_2F_2) data from flask samples and *in situ* instruments located at several locations in latitudes ranging from 83° N to 90° S. Details are given in the various CMDL summary reports (Thompson *et al*., 1994). In Kane (1994), we had reported results for the interannual variability of several trace elements measured by CMDL and had indicated the existence of QBO (Quasi-Biennial Oscillations). However, in some cases, the indications were not very clear, probably because the data were for individual stations which could be affected by localized circulation



Figs. 1(a&b). (a) Three-monthly averages for CFC-11 in the northern (NH) hemisphere and 12-month running averages of CFC-11 and 12 in the northern (NH) and southern (SH) hemispheres and (b) Growth rates (yearly). The numbers indicate spacings (in months) between successive maxima

patterns. Also, few latitudes were examined and growth rates were not studied. Earlier, Elkins *et al.* (1993, 1994) presented the NOAA data for global and hemispheric monthly means of CFC-11 and 12 for 1977-1992 and discussed the characteristics of their growth rates, particularly the recent decrease of the growth rates, as also their relationship with ENSO events. They also mentioned that there was a good correlation between the maximum of easterly winds of the equatorial stratosphere and the drop in the southern hemisphere growth rates for both CFC. Cunnold *et al.* (1994) used an independent data set (ALE/GAGE) of both CFC and fitted an annual cycle and a 29-month (2.4 years) oscillation and estimated their amplitudes for the period 1978-1991. Earlier, Prinn *et al.* (1992) had noted a QBO in another CFC (methyl chloroform). QBO and ENSO effects have been reported earlier for stratospheric ozone also (Zerefos *et al.*, 1992). In this note, a detailed analysis is presented about the interannual variability of CFC-11 and CFC-12, specially in the QBO and QTO (Quasi-Biennial and Quasi-Triennial Oscillation) ranges.

2. Hemispheric CFC data

Weighting data from individual stations by the cosine of the station latitude, Elkins *et al.* (1993, 1994) generated global and hemispheric means. The data were in the form of monthly means. Since these showed considerable seasonal variations, data were first combined into 4 quarters (January to March; April to June; July to

September; October to December). The top plot in our Fig. 1(a) shows the variation of Northern Hemisphere CFC-11 (4 values per year). As can be seen, there is a large (almost linear) increase from 150 ppt (part per trillion) in 1977 to 260 ppt in 1988, after which the growth slowed down. However, there are still some short-term wiggles, suggesting that a seasonal variation may still exist. To eliminate this possibility, 12 monthly running averages were made from the quarterly data. The next four plots in Fig. 1(a) show the variation of the 12 monthly means (centered 3 months apart) for northern (NH) and southern (SH) hemisphere CFC-11 and CFC-12. During 1984-85, there was a gap in CFC-12 values, which was filled by comparing with CFC-11 values before and after the gap. In the beginning of 1978, the CFC-11 values were 152 and 139 ppt for NH and SH respectively. For CFC-12 also, SH values were smaller than NH values. This is entirely because a major part of the industrial CFC is produced in the northern hemisphere, escapes into northern atmosphere and later spreads into the southern hemisphere with a mean interhemisphere exchange time of ~1 year (Jacob *et al*., 1987; Prather *et al*., 1987; Cunnold *et al*., 1994). The CFC-12 values for both hemispheres (277 and 254 ppt) were almost double than those for CFC-11. The relative abundances depend upon source strengths and atmospheric lifetimes and transport processes. Cunnold *et al*. (1994) estimated the equilibrium lifetimes of CFC-11 and CFC-12 as 44 and 180 years respectively and, using the same in a two-dimensional model, estimated the global releases of CFC-11 and CFC-12 in 1990 as 249 million and 366 million kg respectively.

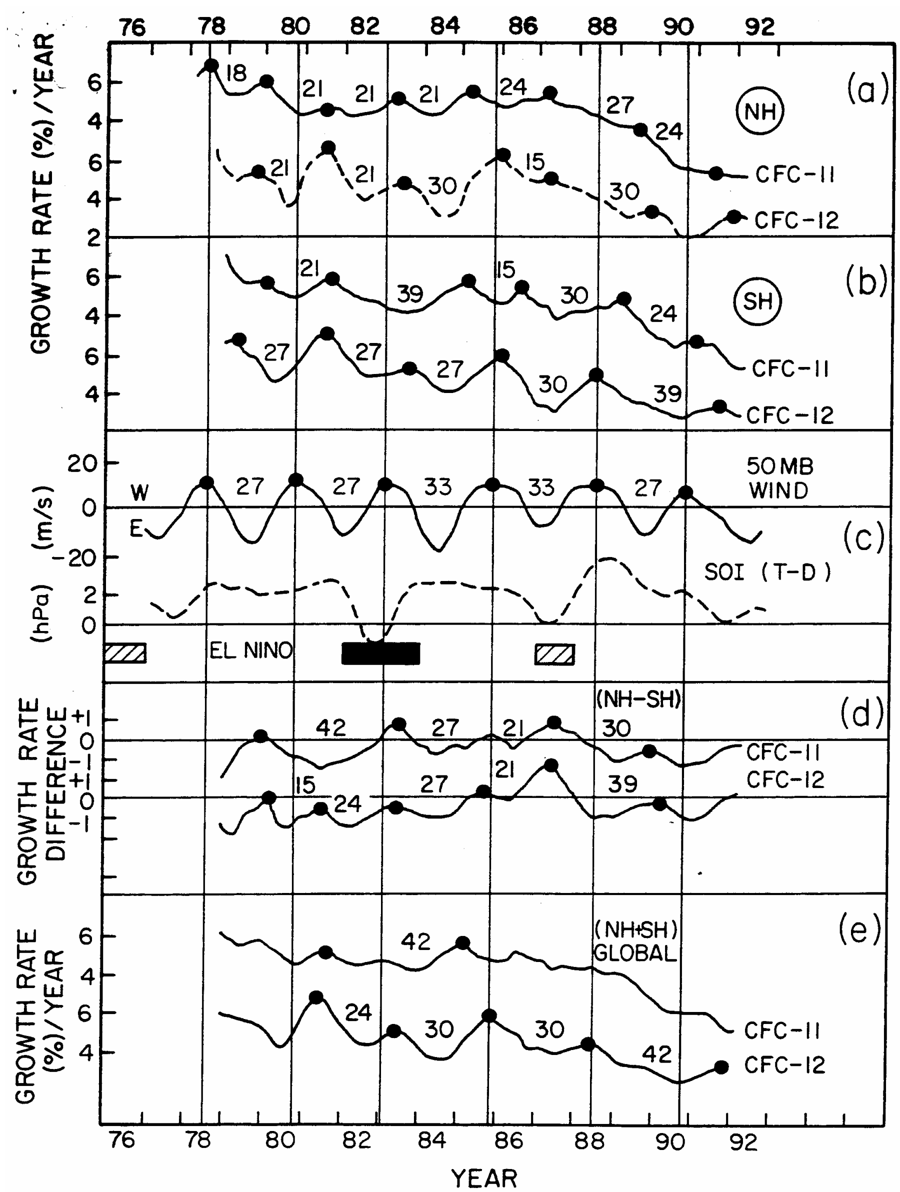
To estimate the yearly growth rate, the 12-monthly running means starting each quarter were subtracted from similar averages starting at the same quarter of the following year. For example, 12-monthly mean starting at the July to September quarter of 1979 was subtracted from that starting at the July to September quarter of 1980 and so on. The yearly growth rates so obtained are shown in Fig. 1(b). After 1987, CFC-11 shows highly reduced growth rates in both hemispheres, but more so in the northern hemisphere, indicating a decrease in industrial production (obeying the Montreal Protocol) in the northern hemisphere where most of the material is used and emitted, as also due to the curtailed use of CFC-11, specially in the aerosol propellant and foam blowing applications in 1990 (Elkins *et al*., 1993). The growth rates for CFC-12 have slowed less rapidly. For detailed discussions see Cunnold *et al*. (1994); Prinn *et al*. (1995).

In the earlier period (1977-1987), the growth rates in both the hemispheres and for both CFC-11 and 12, show clear quasi-periodic variations. The numbers indicate the spacing (in months) between successive maxima (marked by big full circles) and vary in a wide range (15-42 months). Spectral analysis with amplitudes and their errors is discussed later.

3. Results from visual inspection

Since the concentrations of CFC-11 and 12 are different (CFC-12 almost double of CFC-11), the growth rates in ppt are misleading. Also, the concentrations rapidly changed with time. Hence, a more appropriate measure is the percentage growth rate *i.e*. the yearly growth rate expressed as a fraction of the concentration for that year. Fig. 2(a) shows the percentage growth rates for the northern hemisphere for CFC-11 and 12. As can be seen, the percentage changes for CFC-11 are not similar to those of CFC-12. In CFC-11, 7 cycles occur in 156 months while in CFC-12, 6 cycles occur in 144 months, yielding different periods of ~22 and ~24 months. These differences could be because, firstly, the lifetimes of CFC-11 and CFC-12 are different (44 and 180 years) and CFC-12 lasts longer and secondly, their emissions are from different uses. CFC-11 is used in packing materials and foams and is emitted to the atmosphere faster than CFC-12 which, is used in refrigerators and air-conditioners.

Fig. 2(b) shows similar plots for the southern hemisphere. Here again, CFC-11 and CFC-12 plots are dissimilar. Both show 5 cycles but occurring in 129 and 150 months, yielding average spacings of ~26 and ~30 months. The SH spacing is larger than that for NH. Fig. 2(c) shows plots for two atmospheric phenomena *viz*. stratospheric wind and Southern Oscillation Index (SOI). To eliminate seasonal variations, 12-monthly running means were calculated for CFC and the same procedure was adopted for wind and SOI data. Fig. 2(c) shows a plot of 12-monthly running means of 50 hPa zonal wind, as given by Venne and Dartt (1990) and updated from meteorological data reports. A plot of SOI (Southern Oscillation Index) represented by the Tahiti (18^{o} S, 150^{o} W) *minus* Darwin (12^{o} S, 131^{o} E) mean sea level pressure difference (T-D) is also shown (Parker, 1983, updated). The 50 hPa wind has a fairly regular QBO with 5 peaks occurring during ~150 months, yielding an average spacing of ~30 months (2.5 years). This matches with CFC-12 of SH. The westerly wind maxima (marked by vertical lines) seem to be roughly at or slightly before the maxima of CFC-12 of SH. This is roughly the same as the tallying of the easterly wind maxima with SH-CFC minima, reported by Elkins *et al.* (1993). The SOI index, whose minima are known to be associated with El Niño events (warm waters along Peru-Ecuador coast), has a rough spacing of 3-5 years, not in the QBO range. In our Fig. 2(d), the differences of the growth rates of NH and SH shown for CFC-11 and 12, are not similar. For the 1986-87 moderate El Niño, Elkins *et al*. (1993) interpret the (NH-SH) peak as an inhibition of the southward



Figs. 2(a-e). Percentage growth rates for CFC-11 and CFC-12 for:
(a) The northern hemisphere (NH),
(b) The southern hemisphere (SH),
(c) Plots of 12-monthly running averages of 50 hPa wind and southern oscillation index (Tahiti *minus* Darwin atmospheric pressure). Rectangles represent El Niños,
(d) Difference (NH-SH) of northern and southern hemisphere growth rates and
(e) Global growth rate (NH+SH). Vertical lines mark 50 hPa westerly wind maxima. Dots mark maxima and numbers indicate spacings (in months) between successive maxima

transport, which is borne out by the decreased growth rate (lower SH) in Fig. 2(b). For the stronger 1982-83 El Niño, Elkins *et al.* (1993) mention a weaker effect, though we do see peaks, stronger in CFC-11. A serious problem is however in 1979-80, when we do see an equally strong peak in the (NH-SH) plot; but there was no El Niño. Prinn *et al*. (1992), also observed an anomalous behavior of methyl chloroform during 1979-1980. In between El Niño years, there are often years of La Niña (cold water events along the Peru-Ecuador coast). During this (NH-SH), should decrease. This seems to have happened during the La Niña of 1988-1989 [Fig. 2(d)].

To remove the effects of inter-hemispheric transport, the plots for northern and southern hemispheres were combined. The resulting plots for global CFC-11 and

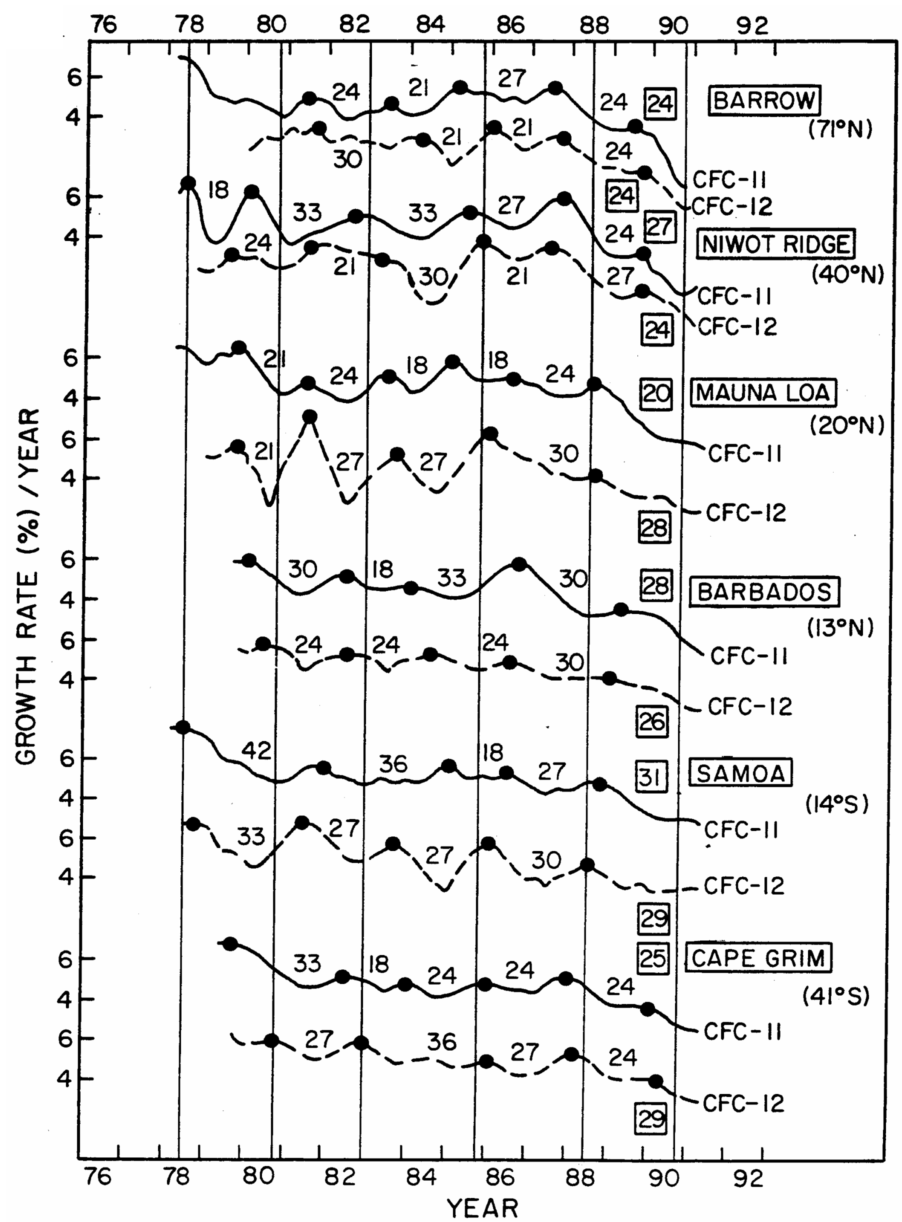


Fig. 3. Percentage growth rates for CFC-11 and CFC-12 for various locations *viz*. Barrow (71° N, 157° W), Niwot Ridge (40° N, 106° W), Mauna Loa (20° N, 156° W), Barbados (13° N, 59° W), Samoa (14° S, 171° W) and Cape Grim (41° S, 145° E). Vertical lines mark 50 hPa westerly wind maxima. Dots mark maxima and numbers indicate spacings (in months) between successive maxima. Numbers in rectangles indicate average spacing

CFC-12 are shown in Fig. 2(e). Whereas CFC-11 shows no distinct peaks, CFC-12 shows at least 4 clear peaks.

Since the difference in the CFC characteristics in the northern and southern hemispheres is striking, it would be interesting to check whether the characteristics are different for different latitudes. For this, data (Elkins *et al*., 1994; Prinn *et al*., 1994) at the locations Barrow (71° N, 157° W), Niwot Ridge (40° N, 106° W), Mauna Loa (20° N, 156° W), Barbados (13° N, 59° W), Samoa (14° S, 171° W) and Cape Grim (41° S, 145° E) were analyzed and the results are shown in Fig. 3. For Barrow in the extreme north, the plots of CFC-11 and CFC-12 differ considerably, but the average spacing is the same (24 months). The average spacings are 27 and 24 months at Niwot Ridge, 20 and 28 months at Mauna Loa, 28 and

26 months at Barbados, 31 and 29 months at Samoa and 25 and 29 months at Cape Grim. Thus, there is no obvious latitude dependence and CFC-11 and CFC-12 show dissimilar spacings. All these results are from visual inspection, which can be highly subjective. The purpose of our Figs. 2 and 3 is mainly to indicate the existence of peaks. Rigorous, statistical estimates were obtained by a spectral analysis, as described in the next section.

4. Power spectrum and correlation analysis

To get quantitative estimates of the possible periodicities, a power spectrum analysis was carried out. In the analysis of Cunnold *et al*. (1994), a QBO period of 29 months was assumed. If, in reality, the period were not 29 months (or any other pre-determined number), the amplitude estimates would be unrealistic. Hence, a different approach was attempted. The method used was MESA (Maximum Entropy Spectral Analysis, Burg, 1967; Ulrych and Bishop, 1975) which detects periodicities much more accurately then the conventional BT (Blackman and Tukey, 1958) method. Also, unlike BT where only certain frequencies (*f* / 2m, *f* = 1, ……, m) can be investigated where the lag m is recommended to be ~25% of the data length, MESA can be obtained for any frequency in the range 0-0.5 (folding frequency) and at any chosen frequency steps. However, the power estimates in MESA are not reliable (Kane, 1977, 1979; Kane and Trivedi, 1982). Hence MESA was used only for detecting possible peaks T_k (k = 1, …...., n), with a LPEF (Length of the Prediction Error Factor) set at 50% of the data length (see details in Kane, 1977, 1979). The T_k so obtained were used in the expression:

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

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

where $f(t)$ is the observed series for any parameter and E the error factor. A multiple regression analysis (Bevington, 1969) was then carried out to estimate A_o (a_k, b_k) and their standard errors (by a least-square fit). From these, the amplitudes r_k and their standard error σ_r (common to all T_k in this methodology, see Bevington, 1969) could be evaluated. Any r_k exceeding $2\sigma_r$ was significant at a 95% (a priori) confidence level.

Fig. 4 shows the results. In Fig. 4(a), the top plot for 50 hPa wind shows one prominent peak at 2.58 years (31 months) and smaller peaks at 2.00 and 5.1 years. Incidentally, we are using only 50 hPa wind data, for the following reasons. Low latitude stratospheric zonal wind does have a vertical structure. But the changes with altitude are small. For example, at 30 hPa, the wind spectra are exactly the same as at 50 hPa, except that the maxima and minima occur ~3-4 months earlier at 30 hPa. In principle, all stratospheric levels can affect atmospheric circulations. But, for tropospheric elements, the lower stratosphere may be more relevant.

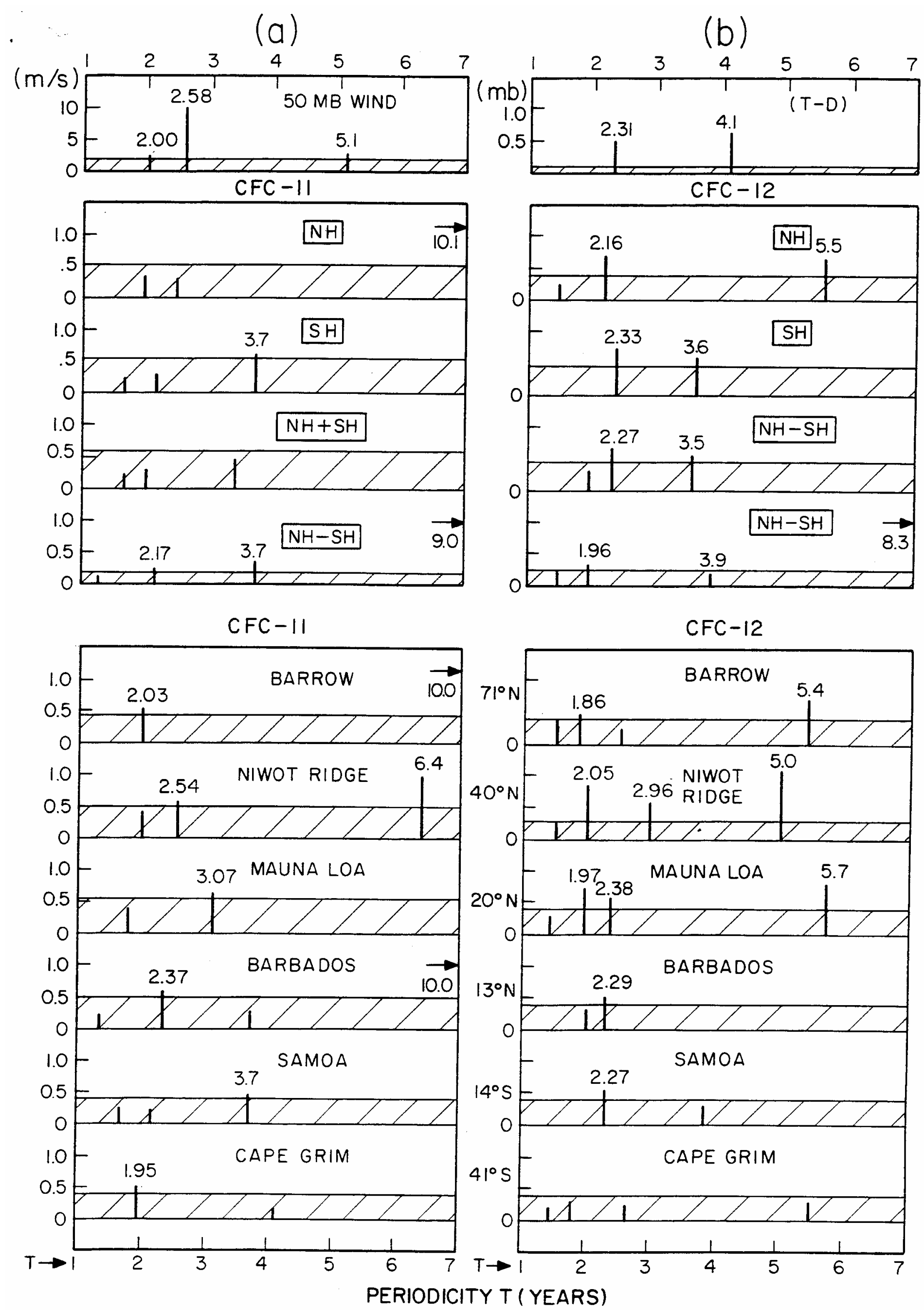
The top plot in Fig. 4(b) shows spectra for the Southern Oscillation Index, represented by (T-D) *i.e*. Tahiti *minus* Darwin atmospheric pressure. Here, the prominent peak is at 4.1 years, though a peak in the QBO region (2.31 years) is also seen.

The other plots in Fig. 4(a) are for CFC-11 and in Fig. 4(b) for CFC-12. The second plot in Fig. 4(a) for northern hemisphere (NH) shows no significant peaks (exceeding the 2σ limit, marked by the hatched portions) except a peak near 10 years, which reflects crudely the long-term CFC-11 trend seen in Fig. 2(a). The next plot for southern hemisphere (SH) shows a barely significant peak at 3.7 years, roughly similar to the 4.1 year peak of (T-D) (Fig. 4b, top). The plot for global CFC-11 (NH+SH) shows no significant peaks, while the difference (NH-SH) shows a significant peak at 3.7 years and a barely significant peak at 2.17 years.

The behaviour of CFC-12 is quite different [Fig. 4(b)]. NH shows strong peaks at 2.16 and 5.5 years, SH at 2.33 and 3.6 years, global CFC-12 (NH+SH) at 2.27 and 3.5 years. The difference (NH-SH) shows barely significant peaks at 1.96 and 3.9 years. Thus, QBO (2.16-2.33 years) is strong in CFC-12 in both hemispheres and QTO (3.6 years) barely significant in the southern hemisphere. However, this QBO range is not similar to the 50 hPa wind QBO (2.58 years); but the QTO has a rough similarity with the ENSO.

The plots in the bottom part of Fig. 4 refer to CFC at individual location, Fig. 4(a) for CFC-11 and Fig. 4(b) for CFC-12. For CFC-11, Barrow shows a peak at 2.03 years, Niwot Ridge at 2.54 years and Mauna Loa at 3.07 years, giving an impression of larger periods at lower latitudes. However, Barbados shows a barely significant peak at 2.37 years, Samoa at 3.7 years and Cape Grim at 1.95 years. Thus, there is no consistent latitude dependence.

For CFC-12 [Fig. 4(b)], all locations except Cape Grim show strong QBO in the range (1.86-2.38) years, while Niwot Ridge shows a strong peak at 2.96 years also. Three locations show peaks in the range (5.0-5.7) years; but these probably reflect the uneven long-term changes. Thus, QBO seems to be a strong feature in CFC-12 only. It is dissimilar to the 50 hPa wind QBO, though a similarity with ENSO QBO (2.31 years) it indicated.



Figs. 4(a&b). Amplitudes of the periodicities obtained by a Maximum Entropy Spectral Analysis (MESA) and Multiple Regression Analysis of the various time series for 1979-1991

(a) Top plot, 50 hPa low latitude zonal wind; Next, CFC-11 for the northern hemisphere (NH), southern hemisphere (SH), (NH+SH), (NH-SH) and at various individual locations and

(b) Top plot, Southern Oscillation Index represented by Tahiti *minus* Darwin atmospheric pressure difference (T-D); Next, CFC-12

Number indicate periodicities in years, significant above the 2σ limit (shown hatched)

Elkins *et al.* (1993) mention a good correlation between the maximum of easterly winds of the equatorial stratosphere (QBOs) and the drop in the southern hemisphere growth rate for both CFCs. In our Fig. 2, average CFC (SH) in Fig. 2(b) shows only a rough relationship with 50 hPa wind as mentioned earlier.

Cunnold *et al*. (1994) mention that the 29-months cycle had the largest amplitude for Cape Matatula, American Samoa. In our case, Samoa does have a QBO at 2.27 years [~27 months, Fig. 4(b)]; but it is barely significant. Instead, Niwot Ridge and Mauna Loa show a very strong biennial oscillation (~2 years).

To check possible correlations between CFC and the 50 hPa wind and (T-D), a cross-correlation analysis was conducted. All correlations between CFC and 50 hPa wind were very low (0.3 or less), indicating very poor relationship. Correlations with (T-D) were higher, (~0.5 ± 0.16), indicating some possibility of a relationship, though not a straightforward one.

5. Conclusion and discussion

The 12-monthly running means of CFC-11 and CFC-12 concentrations for 1977-1992 were examined for interannual variability. The following was noted:

(a) In agreement with Elkins *et al*. (1993) and Cunnold *et al*. (1994), there was a rapid, almost linear CFC increase from 1977 to 1988, ~9.2 ppt/yr for CFC-11 and ~17.5 ppt/yr for CFC-12 in the northern hemisphere and ~9.8 ppt/yr for CFC-11 and ~17.7 ppt/yr for CFC-12 in the southern hemisphere. From 1988 onwards, the growth rates decreased considerably, most probably due to the action taken by industries to curtail production and use of CFC, in accordance with the Montreal Protocol.

(b) Superposed on this general pattern, the yearly growth rate showed fluctuations in the Quasi-Biennial (24-36 months) and Quasi-Triennial (36-48 months) ranges. The spacings between maxima were irregular; but CFC in the northern hemisphere seemed to have shorter average spacings of 20-22 months compared to 26-30 months in the CFC of the southern hemisphere.

(c) A spectral analysis revealed the following:

(*i*) The 50 hPa low latitude zonal wind had one prominent QBO peak at 2.58 years and smaller but significant peaks at 2.00 and 5.1 years. The Southern Oscillation Index (T-D) *i.e*., Tahiti *minus* Darwin atmospheric pressure had a major peak at 4.1 years and a minor but significant QBO peak at 2.31 years.

(*ii*) CFC-11 had only one barely significant peak at 3.7 years, similar to the 4.1 years peak in (T-D). On the other hand, CFC-12 had a strong QBO (2.16-2.33 years) in both the hemispheres, which did not match with the 50 hPa wind QBO (2.58 years) but matched with the minor (T-D) QBO (2.31 years). Also, CFC-12 had a QTO (3.6 years) in the southern hemisphere.

(*iii*) CFC-11 at individual locations had barely significant QBOs in the range (1.95-3.07) years. CFC-12 had a strong QBO in the range (1.86-2.38) years and Niwot Ridge had an additional peak at 2.96 years. Cape Grim had no significant QBO.

(*iv*) In a data set of ~12 years, the peaks like 9 years are not very meaningful and probably reflect the long-term trends and their abrupt changes from 1988 onwards.

Thus, ENSO relation with CFC in the southern hemisphere in general, seems to be genuine. In the stratosphere, strong easterly winds can enhance vertical transport upward from the stratosphere (Plumb and Bell, 1992) and concurrently decrease the north to south tropospheric exchange (Webster and Holton, 1982). During ENSO events, westerly winds in the upper troposphere are suppressed and reduce inter-hemispheric transport (Trenberth, 1991). But the QBO in CFC-11 is barely significant and the QBO of CFC-12 of some locations is of a biennial nature (~2.00 years) and not like the stratospheric wind QBO (2.58 years) but more like the ENSO QBO (2.31 years). The origin of this almost biennial QBO needs further investigation. In the case of stratospheric ozone, Hamilton (1989) and Gray and Dunkerton (1990) report annual synchronization of the ozone QBO, while Krzyscin (1994) shows that ozone variations can be accounted for by using nonlinear terms coupling wind QBO and ENSO, wind QBO and solar activity and ENSO and solar activity. A similar scheme may be applicable to variations in the troposphere. The CFC growth rates depend on the imbalance between sources and sinks. Unlike CO_2 or CH_4 where the sources have geophysical origins, CFC sources are mainly of industrial origin and are not likely to have periodicities like QBO. Thus the QBO and QTO of CFC may come through the sinks, *viz*. stratospheric photolysis, where the stratospheric ozone QBO might play some role. Also, at specific locations, atmospheric transports (both inter-hemispheric and stratosphere-troposphere transports) would have the same effects as of sources/sinks and could create phase and amplitude differences between CFC NH and CFC SH. The different lifetimes and emission processes of CFC-11 and CFC-12 would cause differences in their transports and hence, in their QBO characteristics. Since CFC-12 has a longer lifetime, it stands a better chance of participating in the atmospheric circulations of longer time-scales (QBO). A closer network of observations operating for a long time may throw some light on this problem. However, the possibility remains that QBO of CFC may have an altogether different origin. For 500 hPa winds at 30-35^{o} S, Trenberth (1980) reported a QBO which was out-of-phase with the 50 hPa wind (Kane, 1992). ENSO has also a QBO, but Trenberth (1980) indicated that it was not related to the 50 hPa wind

QBO. Rasmusson *et al*. (1990) identified two dominant time-scales of ENSO variability, *viz*. a biennial mode and a lower frequency mode of period 4-5 years. They considered the biennial mode as a fundamental element, strongly phase locked to the annual cycle. It is tempting to suggest that the CFC-12 at some locations is roughly showing this biennial mode. However (T-D) for 1979-1992 does not show a biennial mode but shows a periodicity of 2.31 years.

It must be remembered that even though wind QBO and ENSO are obvious mechanisms, several other phenomena *e.g*. modulation of semiannual oscillation by QBO (Kennaugh *et al*., 1997) could be operative. Complex interrelated feedback processes may generate semi-periodic components, which may modulate the transport of tracers and the effects of individual mechanisms may get obscured.

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