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Atmospheric Chemistry and Physics Discussions

Halogen Occultation Experiment (HALOE) and balloon-borne in situ measurements of methane in stratosphere and their relation to the quasi-biennial oscillation (QBO)

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Abstract

Measurements of methane have been made from various observational platforms in the atmosphere. In this article, we have compared four high precision balloon-borne measurements from Hyderabad (17.5° N), India in the period of 1987 and 1998 with a part of HALOE/UARS global observations available since 1991. All the balloon measurements correspond to boreal spring (March and April) but belong to different years. A comparison shows fairly good agreement with each other. The strongest effect of dynamical influence on methane vertical profiles is shown to be resulting from the quasi-biennial oscillation (QBO) in the stratosphere, and that has been consistently derived from both the measurement techniques. It is observed that the QBO signal in CH_4 anomaly exhibits interhemispheric asymmetry caused by the tropics to midlatitude circulation in the stratosphere. A mechanism is suggested to explain how and to what extent the methane vertical profiles over Hyderabad and higher latitudes could be modulated by the prevailing QBO winds in the tropics. We have also discussed how the same mechanism would affect ozone distribution in the stratosphere quite differently.

1. Introduction

Methane (CH_4) plays important role in chemistry and radiative balance of the lower and middle atmosphere. It has been estimated that typically about 97% of CH_4 oxidation by OH takes place in the troposphere (Lawrence et al., 2001), and this gas generates 20 times more radiative forcing compared to that of CO_2 on per molecule basis (IPCC, 1996). In the stratosphere chemical reactions with O^1D and Cl dominate CH_4 destruction. Its abundance has increased rapidly since 1950s to the present day (Etheridge et al., 1998). However, from the beginning of 1990s a dramatic decline in CH_4 increase rate has been observed globally (Dlugokencky et al., 2001). Since its emission rate is changing very slowly and its total photochemical loss rate in the lower/middle stratosphere is quite small, anomalies in its stratospheric distribution bear footprints of dy-

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namics, such as quasi-biennial oscillation (QBO), annual and semi-annual oscillations etc. (see Baldwin et al., 2001, for a review). It is established that the QBO constitutes a major part of the variabilities in CH_4 and O_3 anomaly with a time period of about 27–30 months in the stratosphere (Tung and Yang, 1994; Randel et al., 1998). The QBO phase in O_3 anomaly changes at around 15° on either sides of the equator and the higher latitude anomaly extends as deep as 60° latitude (Randel and Wu, 1996). However, the latitudinal extent and interhemispheric differences of QBO influence on CH_4 anomaly have not been very well understood due to shorter temporal coverage of data on the record. Only recently, extensive discussions on observations of the QBO in HALOE CH_4 have been made (Ruth et al., 1997; Randel et al., 1998; Dunkerton, 2001). Here we take advantage of some of these understandings in the explanation of observed variabilities in CH_4 distributions over a tropical site in India.

The measurements of CH_4 in the stratosphere have been started with the advent of appropriate technology, and records are available since early 1970s by using balloon-borne cryogenic collection of air samples (Ehhalt, 1980; Fabian, 1981), and from the satellite platforms only during the past two decades (Jones and Pyle, 1984; Russell et al., 1993). But quality data from a tropical latitude using balloons, and continuous timeseries measurements from the satellite of CH_4 are available since about past 10 years only (Borchers et al., 1989; Russell et al., 1993; Lal et al., 1994, 1996). These observations provide us a unique opportunity to compare two different kind of datasets spreading over a decade, coverring about 4 complete QBO cycles. The satellite data sets are also being utilized extensively to study the influence of various dynamical, and chemical composition changes in the stratosphere. However, no significant usage of satellite observations together with in situ measurements in lower and middle stratosphere could be found in the literature. It could also be noted that though there have been some discussion on the differences in behaviour of vertical distribution of longlived gases over Hyderabad, India (Lal et al., 1996; Patra et al., 1997, 2000), no detailed study has been carried out so far.

Here we have attempted to find possible cause for the differences in the vertical

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profiles observed over the tropics using balloon-borne techniques during March–April period of four different years that are well separated by an interval of about 4 years. The satellite observations over this location and its dynamical neighbourhood will be used to support our findings. In the next section details about the data sources and analysis procedure are given, followed by the results and discussion section. The major conclusions of this study are listed in Sect. 4.

2. Data and Analysis

In the period of 1987 to 1998, four balloon flights have been conducted from the National Balloon-launching Facility (NBF) at Hyderabad (17.5° N, 78.6° E), India on (1) 27 March 1987, (2) 9 April 1990, (3) 16 April 1994, and (4) 18 April 1998. The first two balloon flights were conducted jointly by Max Planck Institute for Aeronomy (MPAE), Lindau and Physical Research Laboratory (PRL), and the latter two flights by PRL only. The payload (balloon gondola) containing a 16-tube cyro-sampler alongwith its control unit were flown with the help of large volume (~150 000 m^3) Pyrex rubber balloon filled with hydrogen gas. The ambient air samples were collected at 15 separate altitudes in the upper troposphere and stratosphere during both ascent and descent of the balloon. Typically, a ceiling altitude of about 35 km was attained during the experiments. The sampling tubes were then brought to the laboratory and analysed for several chemical constituents using GCs coupled with flame ionization detector (for CH_4 and CO), electron capture detector (SF_6, N_2O, chlofluorocarbons, Halons etc.) and mass spectroscopy (for hydrogenated halocarbons). The calibration for absolute concentration of CH_4 was done with respect to the standard gases supplied by Linde gas, UK and NIST, USA. The precision of GC analyses is estimated to be within 5 ppbv (1σ), and the errors in absolute concentration are estimated to be in the range of 2–3% (1σ). The results from these balloon flights have been published earlier (Borchers et al., 1989; Lal et al., 1996; Patra et al., 2000; Lal and Sheel, 2000), but no systematic comparison has been made so far in terms of interannual variations and with satellite

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data.

The vertical profiles of CH_4, constructed from 4 different balloon flight experiments, during boreal spring at an interval of about four years, are shown in Fig. 1a. As argued using the stratospheric measurements of SF_6 over India in a previous study (Patra et al., 1997), the dynamical condition on the measurement day of 16 April 1994 represents a normal day at 17.5° N, that is typical of the tropical stratosphere (hereinafter referred to as "normal" profiles). The 1994 profiles of SF_6, N_2O, CF_2Cl_2 etc. also match fairly well with two different 2-dimensional model simulations (Patra et al., 1997, 2000). The location of the measurement station is subtle in a sense that the tropical upwelling region in stratosphere is located over Hyderabad during northern hemisphere (NH) summer (May–June–July), and in NH winter (December–January–February) the upwelling region moves to the southern hemisphere (SH). Thus sometimes the balloon travel close to the stratospheric "surf zone", and measured vertical distributions often deviate from the normal profile. The observations on 27 March 1987 and 9 April 1990 are the examples of disturbed profiles from the "normal" profile. It can be noted that the magnitude of deviation as well as the altitude of occurrence could vary largely with time. The characteristics of the profile observed on 18 April 1998 exhibit relatively better match with the 1994 profile compared to the other two. This also suggests that the transition from winter to summer in the stratosphere takes place during late March to early April period at 17.5° N. We have checked the horizontal distributions of monthly mean geopotential heights (z) from the NCEP/NCAR reanalysis datasets (Kistler et al., 2001), which commonly show that the gradient in z starts around 30° N in April at 50 mb height. This behaviour of z was quite similar at 10 mb pressure layer during April. However, in March the location of z gradient region shifts over to lower latitudes and lie over Hyderabad.

Figure 1b shows the zonal wind compoment as obtained from the NCEP/NCAR reanalysis at Hyderabad. It is wellknown that the distribution of CH_4 in the troposphere is rather well-mixed and does not depend on dynamics to a greater extent. The main features in this plot are observed in the stratospheric heights: 1) The 1994 profile is

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uniformly higher in methane above 20 km than other profiles – from the wind profiles it is seen that magnitude of easterly winds (also the easterly wind shear) was maximum in 1994 compared to other years of measurements. 2) This also readily shows that in 1990 the minimum in methane in the 25–33 km layer is related to the westerly wind shear and downwelling in the deep tropics and subtropics as methane decreases upward. The upward increase in methane above 32 km in 1990 is probably related to lofting in QBO easterly wind shear; some indication of strong easterly shear in the wind profile is appearing above 27 km. In 1987 the minimum in methane near 20–26 km is related to a westerly wind shear regime and downwelling. Then there appears to be lofting in easterly shear zone and the reversal in wind shear above 30 km seem to be causing the very low CH_4 concentration at 33 km.

At the same time, global coverage of several chemical constituents are available since the launch of HALogen Occultation Experiment (HALOE) onboard the Upper Atmosphere Research Satellite (UARS) in September 1991. Though the UARS mission itself began with an expected lifetime of about 2–3 years, this satellite and many of the instruments onboard continued to make equally good quality measurements till 2002. The datasets are being used for various other kinds of study; e.g. deriving trends in CH_4 in the stratosphere, timeseries correlation of CH_4 with other constituents such as ClO_x, and also to create four dimensional regular grid tracer distributions (Froidevaux et al., 2000; Patra and Santhanam, 2001). For comparison with the balloon-borne measurements of CH_4, we have created the representative vertical profiles for Hyderabad during March–April months in the complete HALOE operation period. First the HALOE CH_4 profiles (Level 2, SPF formatted, both sunrise and sunset), observed between March 15-April 15 of each year and in the 16–19° N latitude band (Hyderabad location ± 1.5°), are selected and the values are interpolated to 30 pressure levels in the height region of 100.0–1.0 mb. Then CH_4 concentrations at each pressure level are averaged to produce one vertical profile representing the boreal spring of each calendar year, which are depicted on Fig. 2 (error bars, 1σ, are also shown). While the average values depict the interannual variations in CH_4 distributions, the spread of the error bars

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indicate the variability in observed concentrations during March/April months of a particular year. It is apparent from the diagram that depending on dynamical condition in our study region, the characteristics of the vertical profiles change from year to year. For instance, the 1997 and 2001 profiles show clear deviation from the other profiles by an amount larger than the error bars, and also the altitude of maximum deviation lies in the range of 25–35 km. The 1995 and 1993 profiles also show discernible difference from the normal profiles, though the intensities and altitude regions are different. We have also used the 4-dimensional CH_4 dataset produced by Patra and Santhanam (2001) later in this work.

It is wellknown that the long-lived trace gases (e.g. CH_4, N_2O, O_3) and aerosol distributions in the stratosphere are closely linked with zonal wind variations in the tropics (Randel et al., 1998; Jones et al., 1998; Tung and Yang, 1994; Hitchman et al., 1994). Some studies also highlighted the interaction between seasonal, subennial and quasi-biennial oscillations in the meteorological parameters to explain the observed variations in CH_4 (Ruth et al., 1997; Dunkerton, 2001). In Fig. 3, we show the altitude-time variation of zonal winds anomaly in the period of 1985 and 2002 as revealed from the NCEP/NCAR reanalysis data (Huesmann and Hitchman, 2001). This diagram shows the temporal extent and altitude distribution of QBO phase and intensity at the equator. As it is discussed above only two out of four balloon-borne observations of CH_4 have overlapping measurements using HALOE. To compare the balloon data with satellite observations, we have identified the QBO phases from this diagram that year 2001 and 1997 closely match with 1987 and 1990 boreal spring conditions, respectively. This search was needed because the QBO is an irregular oscillation, and its period has varied between 30 months in the period of 1980–1992 and that being 27 months in the earlier decades (Tung and Yang, 1994). In addition the QBO and annual cycle are known to have interplay that influence the constituent transport and distribution due to upwelling motion in the equatorial lower stratosphere (Mote et al., 1996; Hinsta et al., 2000). During this selection process, the observed vertical profiles of CH_4 from Hyderbad were also kept in mind to ensure the best possible suitability.

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3. Results and Discussion

In this diagram (Fig. 4), a comparison of CH_4 vertical profiles obtained from the balloon-borne measurements and the satellite observations are shown. Since the HALOE observations were non existing before 1991, we have used 2001 and 1997 HALOE profiles, respectively, corresponding to the balloon observations in 1987 and 1990 as discussed above. We choose to show the best possible matches between the disturbed in situ measurements of CH_4 from Hyderabad on March 1987 and April 1990, and the profiles from HALOE data. For the simultaneous measurement periods in 1994 and 1998, the profiles of HALOE CH_4 in the latitude band of $17.5 \pm 1.5^{\circ}$ are presented. The CH_4 concentrations in the upper troposphere are found to be about 1.64, 1.68, 1.68, 1.67 ppmv, from the balloon measurements, respectively, for the years 1987, 1990, 1994, and 1998 (ref. Fig. 1). On a few occasions similar concentrations are also found in the lower stratosphere due to intense upwelling across the tropical tropopause. It can be noticed here that the CH_4 increase rate in upper troposphere has been fairly stabilized in the period of 1990-1998, which agrees well with the observations on the ground (Dlugokencky et al., 2001). The more interesting features can be seen in the middle stratospheric altitudes. The CH_4 concentrations at around 30–32 km altitude in 1990 and at 34 km in 1987 are only about 60% of those observed in 1994 and 1998 in the same altitude regions. Such large changes cannot be explained by the changes in atmospheric CH_4 burden, solar irradiance variations, or shift in the abundances of reactive radicals in the stratosphere (i.e. chemistry). It is also apparent that such events are not rare in this part of the tropical stratosphere, and are captured by the satellite measurements quite frequently, which has finer temporal resolution.

Here we argue that the dynamical conditions over Hyderabad in March–April 2001 and 1997 were similar to those experienced during the balloon-borne experiments in March 1987 and April 1990, respectively, based on the zonal wind distribution in the stratosphere. This comparison between the balloon and satellite measurements is shown in Fig. 4. On this diagram individual profiles observed by the HALOE are shown

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because some earlier studies suggested that the monsoon structures in the upper troposphere could impose zonal asymmetries in lower 15–20 km of the stratosphere, while above that level quasi-stationary planetary waves can cause zonal asymmetries in the meteorological fields (e.g. Quiroz et al., 1975). As seen from the legends, the HALOE observations had large zonal coverage within about a day in a narrow latitude band. However, no gross feature could be extracted in order to find any zonal asymmetry in the CH_4 distributions. This is probably because of the fact that CH_4 is well mixed in the troposphere and no significant zonal gradient is found in the lower stratosphere due to its long lifetime (~10–100 years). In general, the balloon measurement closely matches with the HALOE observations, except in 1994, when the balloon observed CH_4 mixing ratios are higher by 10–15% than an average HALOE profile at all altitudes. Eventhough for 1987 and 1990 balloon measurements there are no contemporaneous HALOE observations, the match between two compared datasets for different years is unexpectedly good. The HALOE observations in 2001 and 1997 reproduce the CH_4 concentrations at most altitudes and successfully capture the low values at 34 km in 1987 and around 30–32 km in 1990 balloon data, respectively. The 2001 HALOE CH_4 observations also have a tendency to create a kink at 25 km, similar to that could be seen more prominently in 1987 balloon profile.

The obvious next step is to relate the observed CH_4 anomalies with the known dynamical oscillations of the tropical stratosphere, the QBO being the most dominant one. It should be made clear that rest of the study uses the 4-dimensional CH_4 distribution produced by Patra and Santhanam (2001). In Fig. 5, the time-height crosssections of CH_4 anomalies are plotted at four latitudes (5°, 15°, 25°, 35°) in both the hemispheres. An oscillation in the CH_4 anomalies with similar time period like that of the QBO (~27 months) can be seen in the whole height range of tropical stratosphere. The most surprising finding is the latitudinal extent of the QBO features in CH_4 anomalies, which is in contrary to the presence of in-phase QBO signals in the zonal winds as well as in ozone distribution; that generally is restricted to about 15° on either side of the equator (Swinbank and O'Neill, 1994; Baldwin et al., 2001). It also suggests that the height

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distribution of phase propagation of CH_4 anomaly (ascending in nature) to be opposite in phase with the zonal wind anomaly (descending). A possible explanation for this is the difference in QBO winds effect on transport of longlived trace constituents from the tropical stratosphere to the higher latitudes. It has been shown in previous study that the descending westerlies in the tropics enhance transport out of the tropics in the lower stratosphere, whereas the descending easterly winds enhance such transport in the middle stratosphere (Jones et al., 1998). Because the descending limbs of zonal wind anomalies are already out-of-phase to a great extent by about 14 months between the lower (e.g. 50 mb or ~20 km) and middle (e.g. 5 mb or ~35 km) stratosphere near the equator, and the "out of the tropics" transport acts oppositely in the two QBO wind regimes. It could be possible to draw a changeover line at around 8 mb for this regime shift from the time-height plots of CH_4 anomalies (see Fig. 5). It may be relevant to point out here that the Fig. 5 is quite resembling to the time-height plot of water vapour anomaly in the stratosphere, depicting the "tape recorder effect" (Mote et al., 1996), except an important difference that the variation of water vapour in stratosphere is closely linked to the entrainment of dry and wet tropospheric air through the tropical tropopause, and has a periodicity of about a year.

The CH_4 anomaly distribution with QBO periodicity is far more prominent in the case of NH than that in the SH (compare left and right panels on Fig. 5), which could be caused by the asymmetry of the low to middle latitude circulation anomaly (Kinnersley, 1999). He also suggested that strong cross-equatorial flow occurs near the shear zone if the zonal wind anomaly is easterly. From a plot (not shown here) of zonal winds from the NCEP/NCAR reanalysis (Kistler et al., 2001) as a function of time and latitude at 10 mb, it can be seen clearly that the latitude extent of QBO easterlies are larger in the NH compared to that in the SH. Thus the CH_4 anomalies in the NH are also larger compared to the anomalies corresponding to SH stratosphere. The strength of CH_4 concentration change with height is in good agreement with that has been observed for the zonal winds, with the maximum in the middle stratosphere to being weak in the lower and upper stratosphere (see Fig. 5 and 3). Though the NCEP/NCAR reanalysis

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winds as well as the model assimilated winds of the UKMO (Swinbank and O'Neill, 1994) show that the QBO signal in the zonal winds does not generally exist beyond the tropics, but the CH_4 anomalies in the NH stratosphere do bear the QBO signal well into the midlatitudes as shown in Fig. 5. The latitude-time plot apparently does not decipher opposite QBO phases between tropics and extratropics (or midlatitudes) (see also Randel et al., 1998; Dunkerton, 2001), which is contrary to that has been observed for O_3 distribution (Randel and Wu, 1996).

The difference between CH_4 and ozone anomaly distribution is mainly arising from the dissimilarity in their source/sink regions, and hence the influence of stratospheric transport mechanisms. The source of CH_4 in the stratosphere is located at the tropical tropopause; whereas ozone is produced largely in the middle to upper stratosphere region, spreading over a greater latitude region around the equator. The gross picture of latitude-altitude crosssections of CH_4 distribution is relatively simple in the stratosphere; the mixing ratio decreases with the increase in both latitude and height. On the other hand the ozone mixing ratio decreases in either side of the equator only at the middle stratosphere, forming a peak around 1 mb height over the tropics. While the ozone mixing ratio increases with latitude at the base as well as in the top layers of the stratosphere (Fig. 6). Thus transport of O_3, in the lower and upper stratosphere (below 30 mb and above 3 mb, respectively), due to horizontal mixing (from low to high mixing ratio) and cross-equatorial advection (tropics to extratropics) counter each other. This mechanism does not allow the in-phase QBO signal in O_3 distribution to propagate much across the latitudes. The equator to midlatitude transport in the stratosphere works quite differently for CH_4 since the effect of meridional advection and horizontal mixing adds up. The distribution of aerosols adds another dimension to constrain the understanding of stratospheric transport. Hitchman and coworkers (e.g. Hitchman et al., 1994, and references therein) diagnosed tropical reservoir regions with an "upper" and "lower" transport regime, which divide the stratosphere around 22 km in the tropics.

The processes of stratospheric transport around the tropical region has been schematically represented in Fig. 7 during both phases of the QBO, based on the two

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distinct trace species, i.e. CH_4 and O_3, as far as their sources, sinks and distributions in the stratosphere are concerned. For this diagram we have assumed the tropical upwelling at the base of the stratosphere to be constant over a QBO cycle as that signal in CH_4 distribution is not very strong in lower stratosphere (ref. Fig. 5). However, the strength of the meridional transport due to advection is assumed to be variable, which is believed to be forcing the QBO signal in tracer distribution. The downwelling (lofting) during the westerly (easterly) phase of QBO wind shear helps to produce the prominent "rabbit ears" in the CH_4 distribution in the upper stratosphere. In addition, the "return arm" of tropical QBO circulation over midlatitude, which is where the air descend (ascend) in the extratropics during easterly (westerly) equatorial shear (Baldwin et al., 2001), is considered to produce the mid- and high latitude O_3 anomaly. Thus the resulting anomaly in the subtropics is out of phase with that in the tropics. Because the CH_4 mixing ratio decreases sharply with increase in latitude and altitude both, effect of the return arm of the equatorial QBO is not very significant (ref. Fig. 6). These transport processes in the tropics and subtropics could explain the differences in altitude and latitude variations in CH_4 and O_3 anomaly. Though this discussion is restricted to the trace gases alone, but is in compliance with the transport models derived from the aerosol distribution in the stratosphere (Hitchman et al., 1994). Lastly, if an analogy is drawn from the aerosol distribution in the tropics and on basis of the CH_4 and O_3 distributions in the stratosphere, it could be suggested qualitatively that major part of O_3 transport into the subtropics should be occurring in the middle/upper stratosphere, where as the "out of tropics" transport of CH_4 would be far more stronger in the lower part of stratosphere.

4. Conclusions

A comparison between balloon-borne measurements from Hyderabad (17.5° N), India in March–April months of different years in the period of 1987–1998, and the HALOE/UARS measurements of CH_4 during 1991–2002 has been made. Following

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are the major conclusions drawn from this study:

1. The measurements show good agreement, within and between the observation techniques, in lower stratospheric mixing ratios of CH_4, which are relatively less disturbed by the prevalent meteorology and largely influenced by the troposphere-stratosphere exchange.

2. We show that the interannual CH_4 variability in the middle stratosphere over Hyderabad during 1987, 1990, 1994 and 1998 are caused by the stratospheric dynamics, primarily governed by the quasi-biennial oscillations. In general, this relationship would provide useful information for planning the future balloon flight experiments from Hyderabad.

3. Our analysis of HALOE CH_4 data suggests that the effect of QBO on the transport of tracers in the stratosphere are asymmetric across the equator and this signal could penetrate as deep as the midlatitudes, particularly in the northern hemisphere.

4. A mechanism for opposite QBO influence on CH_4 and O_3 mixing ratios beyond $\approx 15^\circ$ latitude has been described qualitatively, which is primarily arising due to the differences in their stratospheric distribution.

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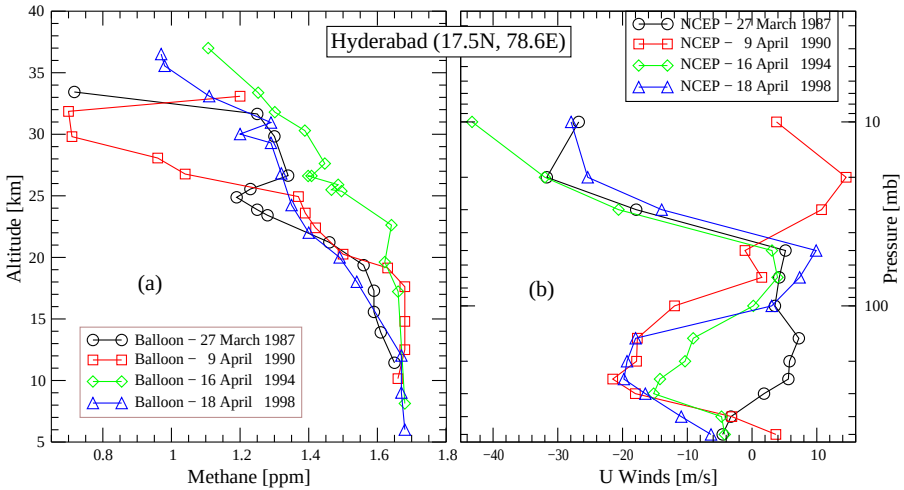

Fig. 1. Vertical profiles of CH_4 obtained by analyzing the ambient air collected during the cryo-sampler experiments in boreal spring of 1987, 1990, 1994 and 1998. The 1987 and 1990 profiles are obtained from the PRL-MPAE collaborative programme (Borchers et al., 1989), and 1994 and 1998 profiles are from the PRL conducted experiments (Lal et al., 1996).

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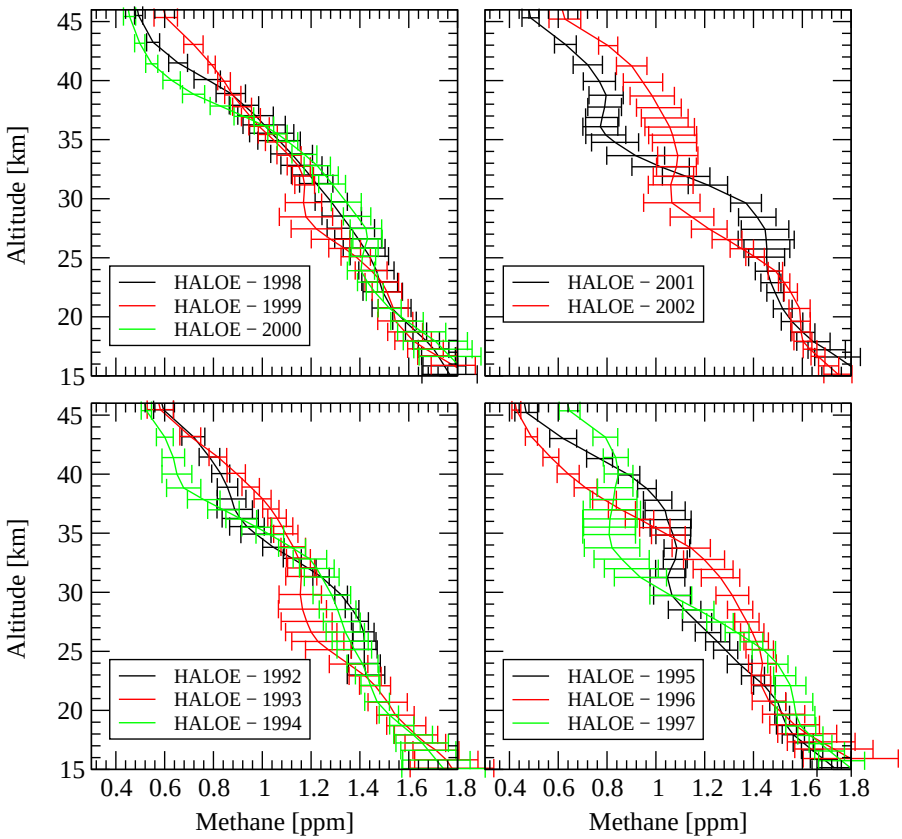

Fig. 2. Observed HALOE CH_4 profiles during 15 March–15 April of each year in the period of HALOE operation. All the available (sunset and sunrise both) profiles in the latitude 17.5±1.5° N band are averaged.

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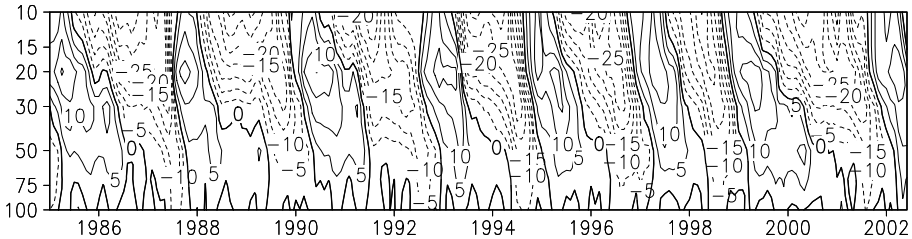

Fig. 3. Temporal variations in monthly mean zonal wind anomalies (m/s) with respect to pressure (mb), produced by the NCEP/NCAR reanalysis project over the equator. The contour interval is 5 m/s; negative wind speed represents the Easterlies (denoted by dotted lines), and the Westerly phase of QBO is seen as the positive winds (solid lines). Note that the height extent of QBO is approximately from 100 mb to 10 mb, and above 3 mb (not shown here due to the lack of NCEP/NCAR reanalysis data beyond 10 mb height) dominance of the semi-annual oscillation (SAO, westerly near the equinox and easterly near the solstice) over QBO is apparent (Swinbank and O'Neill, 1994).

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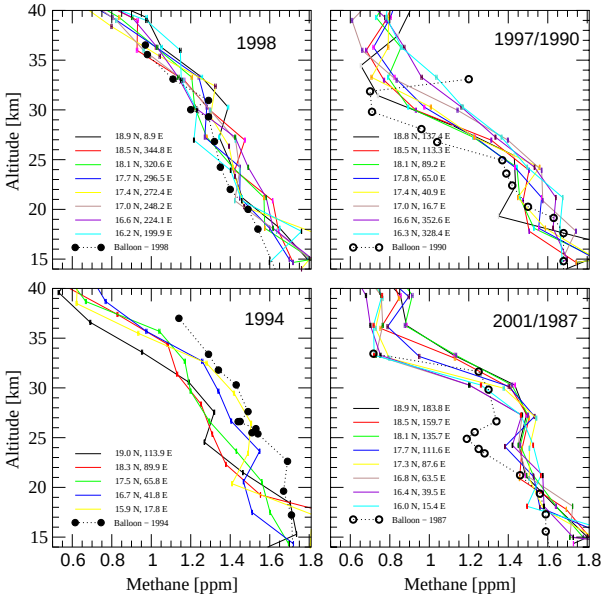

Fig. 4. Showing the match between the remote sensing observations with in situ measurement of CH_4. The comparison of 1994 and 1998 balloon measurements (filled circle) with the HALOE CH_4 profiles are shown as they were observed in the respective years (panels on the left). Whereas for 1987 and 1990 balloon data (open circle), we have chosen the HALOE profiles from the years 2001 and 1997, respectively, for comparison (right panels) due to the lack of contemporaneous HALOE measurements, which are selected based on the meteorology of the stratosphere. The time and type of all HALOE measurements can be designated as: 1) 5 April (22:17 GMT) to 6 April (04:43 GMT) in 1994 (Sunrise), 2) 12 April (17:36 GMT) to 13 April 04:50 GMT) in 1998 (Sunset), 3) 17 April (09:03–20:17 GMT) in 1997 (Sunset), and 4) 27 March (05:52–17:05 GMT) in 2001 (Sunset). The HALOE profiles are chosen as close to balloon measurement dates as possible irrespective of sunset or sunrise data retrieval.

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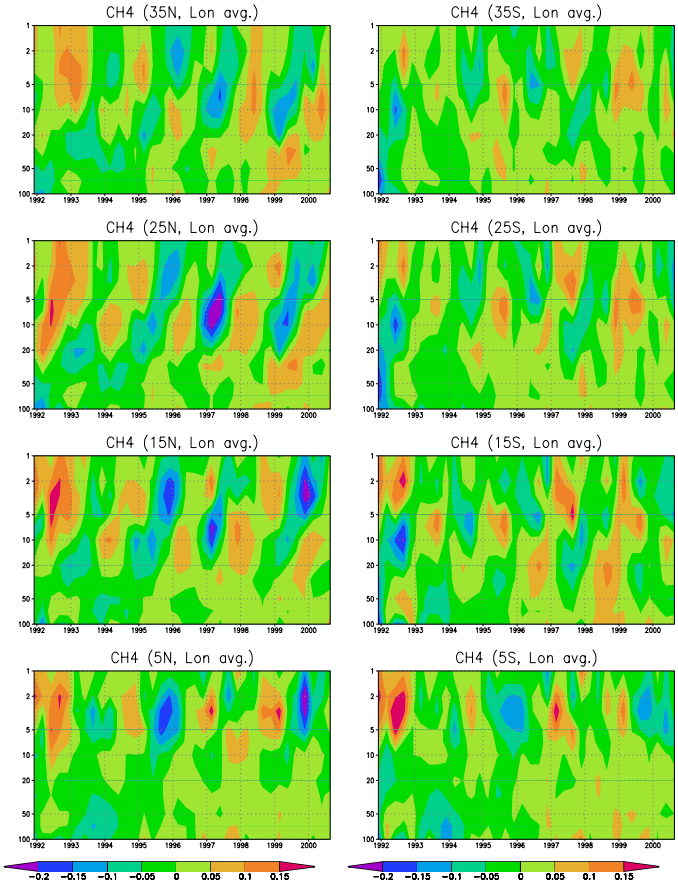

Fig. 5. Time-height variations of HALOE CH_4 anomalies in the stratosphere at various latitudes. Note the out-of-phase relation of CH_4 anomaly in the stratosphere and zonal wind anomalies in the tropics (see Fig. 3).

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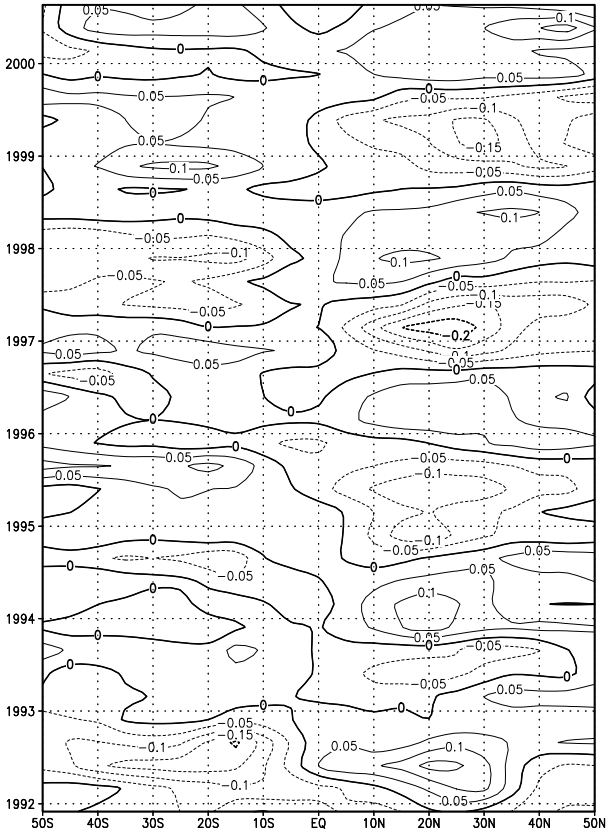

Fig. 6. Latitude-time crosssection of CH_4 anomaly showing the detailed latitudinal extent of QBO signal at 10 mb pressure layer. It can be noticed that the CH_4 anomalies in NH and SH are separated by a time lag of about one year.

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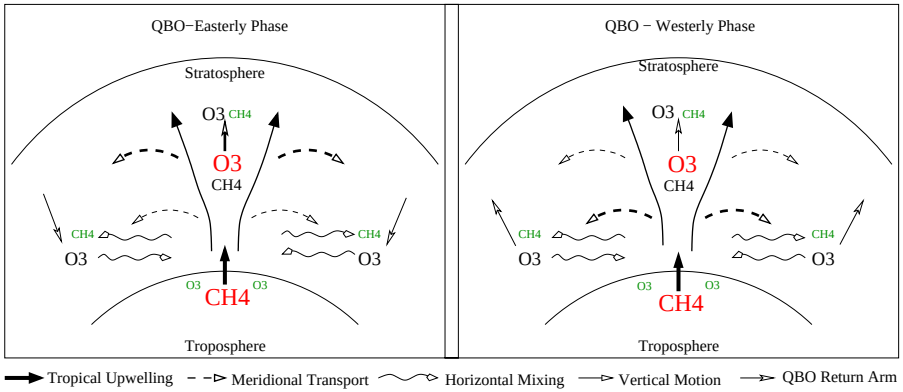

Fig. 7. Schematic diagram of the stratospheric dynamics in two phases of the QBO. Different arrows have been used to indicate prevailing dynamical components such as tropical upwelling, out of tropics (meridional) transport by the mean meridional circulation, horizontal mixing arising from the concentration gradient (from high to low), ascending (descending) QBO return arms over midlatitude during westerly (easterly) QBO phases, and the downward (upward) motion in the tropics that produce "double peak" structure in the stratosphere. The size of CH_4 and O_3 symbols are varied to represent areas of high to low (by big, medium and small sizes) values of mixing ratios, which inturn determines the direction of transport due to horizontal mixing. Similarly, thickness of arrow indicates the changes in strength of the meridional transport during two QBO phases.

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