

## Seasonal Cycles and QBO Variations in Stratospheric CH<sub>4</sub> and H<sub>2</sub>O Observed in UARS HALOE Data

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### ABSTRACT

Measurements of stratospheric methane (CH<sub>4</sub>) and water vapor (H<sub>2</sub>O) are used to investigate seasonal and interannual variability in stratospheric transport. Data are from the Halogen Occultation Experiment (HALOE) on the *Upper Atmosphere Research Satellite* (UARS) spanning 1991–97. Profile measurements are binned according to analyzed potential vorticity fields (equivalent latitude mapping), and seasonal cycles are fit using harmonic regression analysis. Methane data from the UARS Cryogenic Limb Array Etalon Spectrometer and water vapor from the Microwave Limb Sounder are also used to fill in winter polar latitudes (where HALOE measurements are unavailable), yielding complete global seasonal cycles. These data reveal well-known seasonal variations with novel detail, including 1) the presence of enhanced latitudinal gradients (mixing barriers) in the subtropics and across the polar vortices, 2) strong descent inside the polar vortices during winter and spring, and 3) vigorous seasonality in the tropical upper stratosphere, related to seasonal upwelling and the semiannual oscillation. The observed variations are in agreement with aspects of the mean meridional circulation derived from stratospheric meteorological analyses. Interannual variations are also investigated, and a majority of the variance is found to be coherent with the equatorial quasibiennial oscillation (QBO). Strong QBO influence is found in the tropical upper stratosphere: the double-peaked “rabbit ears” structure occurs primarily during QBO westerlies. The QBO also modulates the latitudinal position of the tropical “reservoir” in the middle stratosphere.

### 1. Introduction

Important information on the transport and overall flow of mass in the stratosphere can be obtained by analysis of long-lived chemical constituents. Two such constituents are methane (CH<sub>4</sub>) and water vapor (H<sub>2</sub>O). Methane is produced by biotic activity near the earth's surface, is transported into the stratosphere in the Tropics, and chemically destroyed (oxidized) above 35 km. The photochemical lifetime of CH<sub>4</sub> below 40 km is >100 days, so the stratospheric distribution is deter-

mined mainly by the circulation. Methane measurements from satellites have been used in a number of studies to deduce stratospheric circulation features (e.g., Jones and Pyle 1984; Solomon et al. 1986; Gray and Pyle 1986; Holton and Choi 1988; Stanford et al. 1993; Russell et al. 1993a; Kumer et al. 1993; Bithell et al. 1994; Schoeberl et al. 1995). Likewise, water vapor has been used to deduce the global stratospheric circulation (Brewer 1949) and examine details of tropical (Mote et al. 1995, 1996) and global (Lahoz et al. 1996a; Rosenlof et al. 1997, hereafter RO97) transport. The ability to correctly simulate CH<sub>4</sub> and H<sub>2</sub>O is a useful benchmark for numerical models of the middle atmosphere (e.g., Prather and Remsberg 1992).

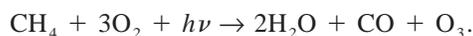
The objective of this study is to analyze a long record (1991–97) of satellite CH<sub>4</sub> and H<sub>2</sub>O measurements in order to quantify the seasonal cycle and interannual vari-

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ability in stratospheric transport. The primary data are from the Halogen Occultation Experiment (HALOE) on the *Upper Atmosphere Research Satellite (UARS)*. To fill in polar regions not sampled by HALOE, we use  $\text{CH}_4$  measurements from the Cryogenic Limb Array Etalon Spectrometer (CLAES) and  $\text{H}_2\text{O}$  data from the Microwave Limb Sounder (MLS) instruments on *UARS* obtained during 1992–93. These combined data provide a complete global sample of the seasonal cycles of  $\text{CH}_4$  and  $\text{H}_2\text{O}$ . Interannual variations are studied based on HALOE data alone.

The overall structure and variability of  $\text{CH}_4$  is tightly coupled with  $\text{H}_2\text{O}$  in the stratosphere. This is because a principal source of stratospheric  $\text{H}_2\text{O}$  is from the oxidation of  $\text{CH}_4$ , via a series of chemical reactions that may be summarized (following Remsberg et al. 1984) by



This yields two molecules of  $\text{H}_2\text{O}$  for every one of  $\text{CH}_4$ . LeTexier et al. (1988) examined the chemistry in more detail and noted that some intermediate species may not be converted to  $\text{H}_2\text{O}$ , but that molecular hydrogen ( $\text{H}_2$ ) could be produced. A conserved quantity in these chemical reactions is the total number of hydrogen atoms:

$$H_{\text{total}} = 2[\text{H}_2 + \text{H}_2\text{O} + \beta \times \text{CH}_4],$$

where  $\beta$  is a so-called chemical yield factor. Examination of data and model results have shown that  $\text{H}_2$  is nearly constant and  $\beta \sim 2.0$  over much of the stratosphere (Jones et al. 1986; Garcia and Solomon 1994; Dessler et al. 1994; Harries et al. 1996a; Remsberg et al. 1996). Thus the quantity  $D = \text{H}_2\text{O} + 2 \times \text{CH}_4$  is an approximately conserved parameter. The result of this is that spatial gradients of  $\text{CH}_4$  are approximate mirror images (with opposite sign) of those in  $\text{H}_2\text{O}$ , so that stratospheric transport variations are echoed in opposing signals in these two constituents.

There are two regions where  $\text{H}_2\text{O}$  data provide details of transport not seen in  $\text{CH}_4$ . First, a strong seasonal cycle is imparted to  $\text{H}_2\text{O}$  at the tropical tropopause from seasonal temperature variations, and this annual cycle is observed to propagate vertically in the tropical lower-middle stratosphere (Mote et al. 1995, 1996). Second, strong dehydration is observed in the Antarctic polar vortex during winter and spring (due to extreme cold temperatures), and this signal provides an opportunity to study transport across the edge of the vortex (Russell et al. 1993a; Tuck et al. 1993; Pierce et al. 1994; RO97). The seasonal cycle  $\text{H}_2\text{O}$  data presented here provide novel detail and global perspective to these features.

## 2. Data and analyses

### a. HALOE data and equivalent latitude mapping

The primary data analyzed here are HALOE V18 vertical profile measurements of  $\text{CH}_4$  and  $\text{H}_2\text{O}$ . The HALOE instrument is described in Russell et al.

(1993b), and the  $\text{CH}_4$  and  $\text{H}_2\text{O}$  data are discussed in detail in Park et al. (1996) and Harries et al. (1996b), respectively. We use HALOE level 3a data, which are available on 16 standard *UARS* pressure levels spanning 100–0.32 mb (approximately 16–56 km), with a vertical spacing of about 2.5 km. The time period analyzed here covers November 1991–March 1997.

HALOE is a solar occultation instrument that obtains 15 sunrise and 15 sunset measurements per day, each near the same latitude but spaced  $\sim 24^\circ$  apart in longitude. The latitudinal sampling progresses in time, so that much of the latitude range  $60^\circ\text{N}$ – $\text{S}$  is sampled in 1 month. One method to analyze the latitude–height structure of these data is to construct zonal means by simple longitudinal averaging. However, this has the disadvantage of averaging airmasses that may be physically distinct, in particular those inside versus outside the polar vortex [for cases when the vortex is elongated or not centered over the pole; see the example Plate 1 of Schoeberl et al. (1995)].

An alternative method is to average data along potential vorticity (PV) contours, which acts as an approximate vortex-following coordinate (McIntyre and Palmer 1983; Burchart and Remsberg 1986; Schoeberl et al. 1989, 1992, 1995; Norton 1994; Manney et al. 1995). For the analyses presented here we have derived PV from daily wind and temperature fields output from the United Kingdom Meteorological Office (UKMO) stratospheric data assimilation system (Swinbank and O’Neill 1994). The utility of PV versus zonal averaging is illustrated in Fig. 1, showing HALOE  $\text{CH}_4$  measurements at 10 mb during September–October 1993 plotted versus latitude and versus PV. (Here and throughout the rest of this work the PV coordinate is expressed in terms of “equivalent latitude,” i.e., the latitude of an equivalent PV distribution arranged symmetrically about the pole.) The distribution of  $\text{CH}_4$  versus latitude in Fig. 1 shows a wide scatter over high southern latitudes, due to sampling inside (low  $\text{CH}_4$ ) and outside (high  $\text{CH}_4$ ) of the polar vortex. Conversely, the  $\text{CH}_4$  shows strong correlation with PV, with pronounced separation of vortex interior data (see also Bithell et al. 1994; Schoeberl et al. 1995). In a similar manner, HALOE  $\text{H}_2\text{O}$  data are found to be correlated with the polar vortex (Pierce et al. 1994; RO97) and associated PV fields.

Figure 2 shows an example of the PV mapping of HALOE  $\text{CH}_4$  for data during January 1994. The PV mapping is done on each *UARS* pressure level separately (one can choose to perform the PV mapping using potential temperature as a vertical coordinate, but this involves vertical interpolations before and after mapping, and gives nearly identical results to mapping on pressure levels used here). Data are analyzed between  $80^\circ\text{N}$  and  $80^\circ\text{S}$  (as available for each month) on a  $4^\circ$  equivalent latitude grid. There are two advantages to the PV mapping: 1) data that are physically distinct remain separated; and 2) there is an increase in the effective latitude range of the data, as measurements inside the vortex

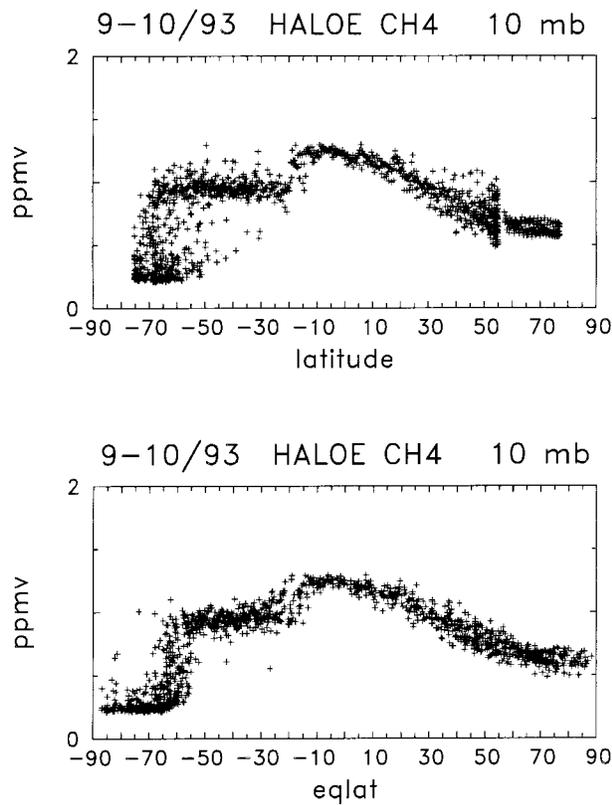


FIG. 1. Scatter diagrams of HALOE  $\text{CH}_4$  observations at 10 mb during September–October 1993 plotted as a function of latitude (top) and potential vorticity (PV) (bottom). Potential vorticity is expressed in terms of equivalent latitude.

correspond to high equivalent latitude (note the NH data shown in Fig. 2). The equivalent latitude data (over  $80^\circ\text{N–S}$ ) are binned into monthly samples prior to further analyses; we note these are not true monthly means, as data at different latitudes are observed at different times during each month. We require at least three data points at a particular altitude and equivalent latitude or else the corresponding monthly sample is left blank.

#### b. Seasonal cycle fits

Seasonal cycles are estimated from the monthly binned data by the fitting of seasonal harmonics at each (equivalent) latitude and pressure altitude. This method works well for the irregularly sampled HALOE measurements [similar analyses are used by McCormick et al. (1993)]. Examples are shown in Fig. 3 for  $\text{CH}_4$  and  $\text{H}_2\text{O}$  data at 10 mb and  $28^\circ\text{S}$ . These show the monthly sampled equivalent latitude data (as “plus” signs), together with the harmonically varying seasonal cycles (the repeating smooth curves). The seasonal cycle at each location is determined using a time mean, plus annual and semiannual harmonics fit by least squares regression. The linear trend (determined over January 1993–December 1996) is removed prior to these sea-

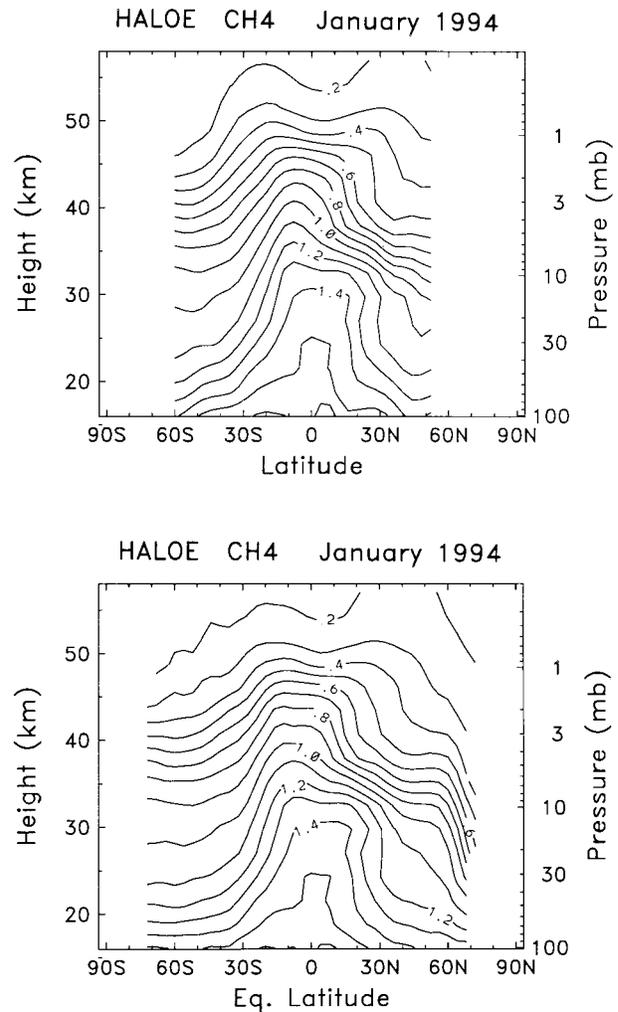


FIG. 2. Comparison of HALOE  $\text{CH}_4$  observations during January 1994 calculated using zonal averages (top) and averages along PV contours (equivalent latitude mapping—bottom).

sonal fits, and it is also omitted from the interannual analyses discussed below. Note the opposing seasonality (and interannual variations) in the  $\text{H}_2\text{O}$  and  $\text{CH}_4$  data in Fig. 3; this is a feature observed throughout the stratosphere in general, due to conservation of the quantity  $D = \text{H}_2\text{O} + 2 \times \text{CH}_4$  (as discussed above).

Over high latitudes of both hemispheres, HALOE data are not available during several months in winter (the equivalent latitude PV mapping helps fill in this void in the NH, but less so during SH winter when the vortex is typically centered over the pole). For such high (equivalent) latitudes where data are unavailable each year, the seasonal cycle is only fit over months that data are available, and the remaining months are left blank [specifically, we require at least two years of data (out of five) for each individual month]. An example is shown in Fig. 4 for  $\text{CH}_4$  data at  $72^\circ\text{S}$ , where no seasonal cycle is fit during April–August.

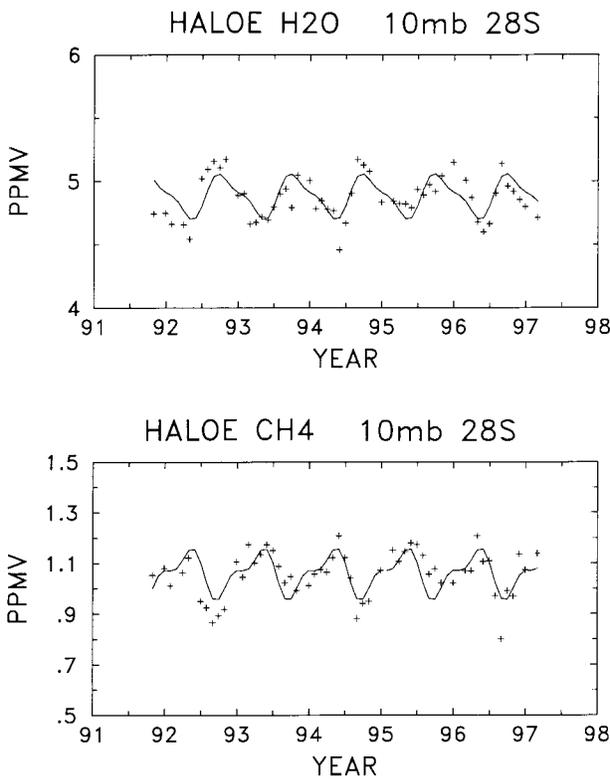


FIG. 3. Time series of HALOE H<sub>2</sub>O and CH<sub>4</sub> data at 10 mb, 28°S. Crosses denote monthly mean data, and thin lines denote (repeating) seasonal cycles fit by harmonic regression.

### c. Addition of CLAES CH<sub>4</sub> and MLS H<sub>2</sub>O data

In order to fill in the high latitude voids in the seasonal cycle estimates of CH<sub>4</sub>, we include CH<sub>4</sub> observations from the CLAES instrument on UARS (Kumer et al. 1993; Roche et al. 1996). These data span a little more than one year (January 1992–April 1993), but offer more continuous sampling than the HALOE data, in particular observing the polar winter regions. The data used here are from the version 7 (V7) retrieval. The CLAES CH<sub>4</sub> data are mapped into equivalent latitude space and monthly averaged. Direct comparison of the CLAES and HALOE data shows reasonable agreement in the latitudinal structures, although there is an offset (bias), with CLAES approximately 20% higher than HALOE (as discussed in Roche et al. 1996). We remove this bias for the analyses here by simply adjusting the CLAES data (using a latitude-independent multiplicative factor) to match the HALOE measurements over a region adjacent to the missing HALOE data. This matching is done using a seasonal cycle fit of the CLAES data (over 1992–93) together with the seasonal cycle HALOE data (1991–97), in order to obtain smooth transitions. The adjusted CLAES data and fit seasonal cycle are included in the time series at 72°S shown in Fig. 4. Note the CLAES data are used only in the sea-

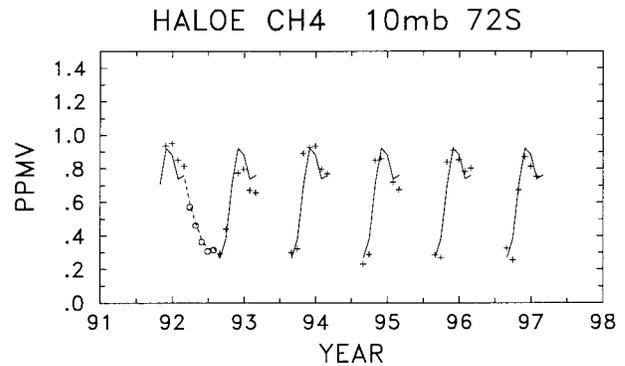


FIG. 4. Time series of CH<sub>4</sub> data at 10 mb, 72°S. Crosses and solid lines denote HALOE observations and fit seasonal cycle; note data are only available during September–February for each year. Circles and dashed lines during 1992–93 show adjusted CLAES data and seasonal cycle fit.

sonal cycle estimates, since the short CLAES record precludes interannual analyses.

In a similar manner, we augment the HALOE seasonal cycle H<sub>2</sub>O results using MLS data in polar regions. Stratospheric H<sub>2</sub>O data are derived from MLS 183-GHz measurements, as described in Lahoz et al. (1996b). We use the version 4 (V4) retrieval for the analyses here, with time period spanning November 1991–April 1993. The seasonal cycle fit of the equivalent latitude MLS data are adjusted (by a small amount, ~5%) to smoothly match the HALOE seasonal cycle fit.

### d. Calculation of residual mean circulation

Part of the results here include comparisons of the constituent seasonal cycles with estimates of the mean meridional circulation derived from United Kingdom Meteorological Office stratospheric analyses (Swinbank and O'Neill 1994). Specifically, we calculate monthly average components of the residual mean meridional circulation ( $v^*$ ,  $w^*$ ) using the coupled transformed Eulerian-mean (TEM) thermodynamic and continuity equations (as in Solomon et al. 1986; Rosenlof 1995). Radiative heating rates were calculated from an accurate radiative transfer code (Olague et al. 1992), incorporating UKMO temperatures, HALOE ozone, water vapor and methane, and climatological distributions of other trace gases. The overall circulation features are very similar to previous calculations (e.g., Gille et al. 1987; Eluszkiewicz et al. 1996). Aspects of constituent transport are studied based on the TEM constituent continuity equation [Andrews et al. 1987, their Eq. (9.4.13)]:

$$\frac{\partial \bar{\chi}}{\partial t} = -\bar{v}^* \bar{\chi}_y - \bar{w}^* \bar{\chi}_z + \nabla \cdot \mathbf{M} + \bar{S}. \quad (1)$$

Here  $\bar{\chi}$  is the zonal-mean constituent mixing ratio,  $\bar{v}^*$  and  $\bar{w}^*$  are components of the TEM residual mean cir-

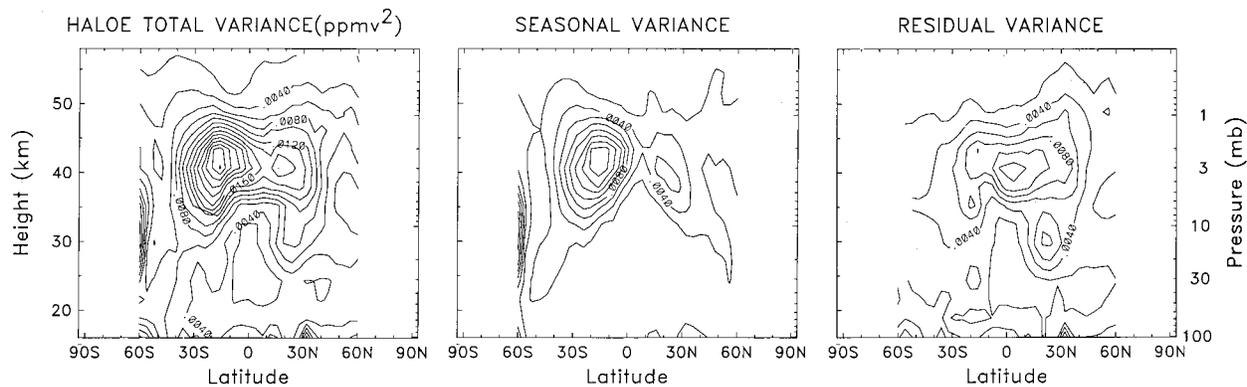


FIG. 5. Components of temporal variance in HALOE  $\text{CH}_4$  data over 1991–97. Shown are total variance (left), seasonal cycle variance (middle), and residual interannual variance (right). Contours in all panels are  $0.002 \text{ ppmv}^2$ .

ulation (discussed above),  $\nabla \cdot \mathbf{M}$  is an eddy transport term, and  $\bar{S}$  is a chemical source or sink term. This formalism has been used to study constituent transport in three-dimensional model simulations by Randel et al. (1994) and Strahan et al. (1996).

### 3. Seasonal variability

The overall temporal variance of the HALOE  $\text{CH}_4$  data during 1991–97 is shown in Fig. 5, along with the component attributable to the harmonic seasonal cycle, and the residual (i.e., that attributable to interannual variance). Only HALOE data are used in these calculations, which span  $60^\circ\text{N}$ – $\text{S}$ . Variance in all components peaks in the middle-upper stratosphere (over 30–50 km) over low latitudes. The overall variance is split fairly evenly between the seasonal and interannual components, but the spatial patterns are distinct (note the seasonal variance has a minimum over the equator, while the interannual variance has a maximum there). A majority of the interannual variance in Fig. 5 is associated with the QBO, as discussed below. Variance patterns in  $\text{H}_2\text{O}$  data (not shown) reveal upper stratospheric maxima similar to that in Fig. 5, with additional maxima in the Tropics over 20–30 km due to both seasonal and interannual components.

#### a. Global seasonality

Figure 6 shows the global structure of  $\text{CH}_4$  in January, April, July, and October, derived from the seasonal cycle fits. The overall structures are similar to those shown previously (e.g., Luo et al. 1995; Park et al. 1996), but the synthesis of many years of data here allows the seasonality to be studied in some detail. Contours bulge upward in the Tropics and move latitudinally with season in the upper stratosphere, reflecting variations in the upward transport circulation in the Tropics (as shown below). There is a double-peaked pattern in the upper stratosphere in April related to the seasonal subtropical upwelling and the

tropical semiannual oscillation (SAO); a similar double-peaked structure is not seen in October (the SAO is discussed in more detail below). Relatively strong horizontal gradients are seen in the subtropical lower stratosphere near  $20^\circ$ – $30^\circ \text{N}$ – $\text{S}$  throughout much of the year, and strong gradients are also associated with the polar vortex in both hemispheres during winter–spring. These regions of enhanced gradients are associated with differential vertical advection (upward in the Tropics and downward in polar winter) and relative minima in horizontal mixing (so-called mixing barriers). The SH winter–spring polar vortex is particularly robust in these data, due to strong downward flow inside the vortex and small transport across the vortex edge (Russell et al. 1993a; Schoeberl et al. 1995; Fisher and O’Neill 1993; Manney et al. 1994; Sutton 1994). In contrast, the  $\text{CH}_4$  contours are relatively flat over midlatitudes during winter–spring, showing evidence of more rapid mixing associated with the planetary waves (McIntyre and Palmer 1983, 1984; Leovy et al. 1985). There are relatively weak vertical gradients in high latitudes over 20–30 km during SH summer (January), possibly a remnant of very low (vortex)  $\text{CH}_4$  following the SH final warming.

The corresponding seasonal cycle patterns in  $\text{H}_2\text{O}$  are shown in Fig. 7. The overall patterns are approximate mirror images of the  $\text{CH}_4$  data in Fig. 6, including the seasonal movement of the upper-stratospheric maxima and appearance of a double peak in April. These similar patterns reflect the fact that the  $\text{H}_2\text{O}$  increase with altitude is due to  $\text{CH}_4$  oxidation, so that total hydrogen is conserved (see references in introduction). This is demonstrated with the data here by calculation of the quantity  $2\text{CH}_4 + \text{H}_2\text{O}$ , shown for the October distribution in Fig. 8. Note the contour interval is identical between Figs. 7 and 8, so that the relative lack of spatial structure in  $2\text{CH}_4 + \text{H}_2\text{O}$  (cf.  $\text{H}_2\text{O}$  or  $\text{CH}_4$  alone) demonstrates that to first order  $2\text{CH}_4 + \text{H}_2\text{O} \approx \text{constant}$  for these seasonal cycles. Two locations where this approximation does not hold

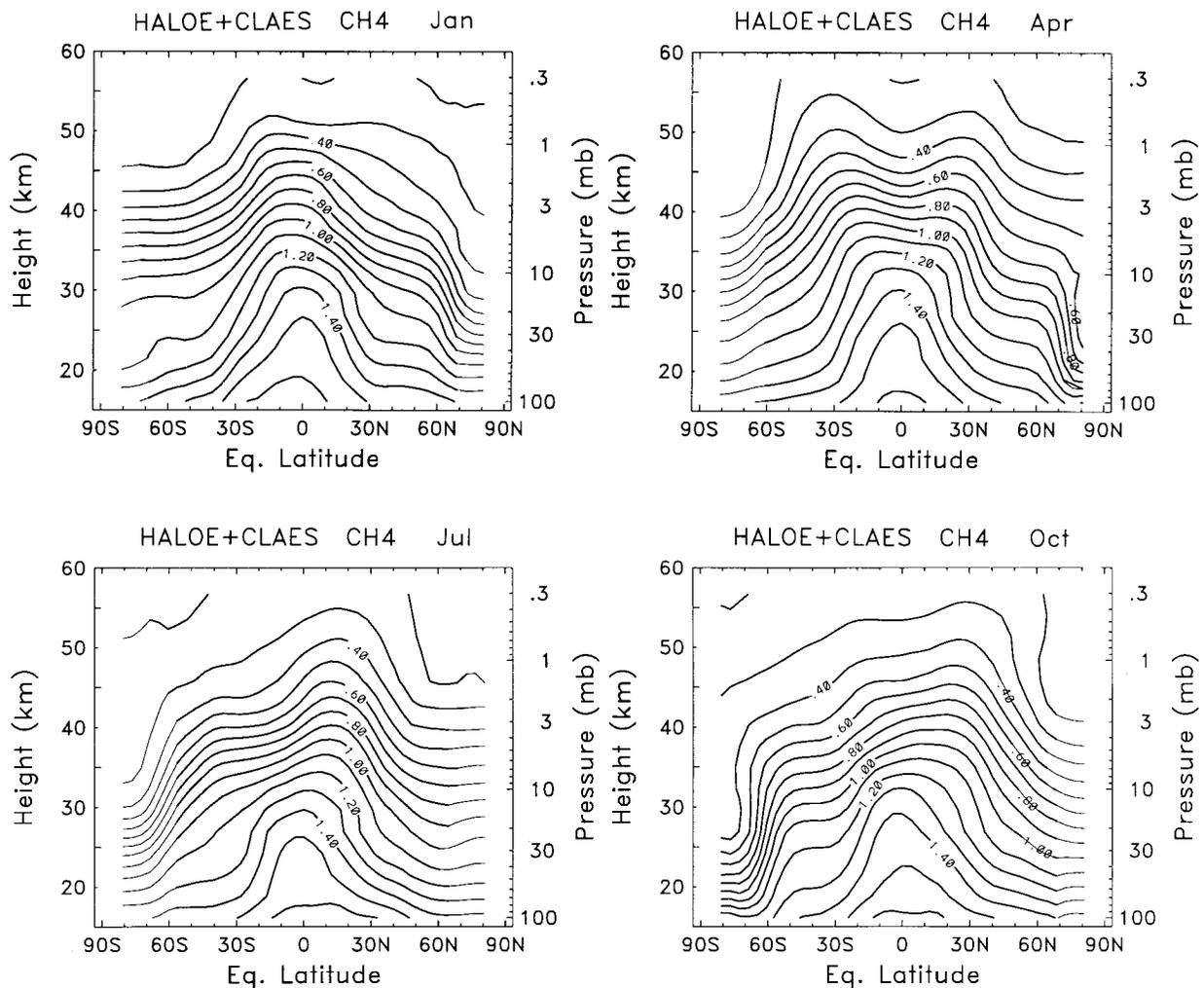


FIG. 6. Seasonal cycle estimates of  $\text{CH}_4$  in January, April, July, and October. Contour interval is 0.1 ppbv.

are 1) in the tropical lower stratosphere, and 2) over Antarctica in winter–spring, as discussed in the introduction.

The latitude–time evolution of  $\text{CH}_4$  at 68, 10, and 2.2 mb is shown in Fig. 9. There is a slight latitudinal shifting of the tropical maximum region in the lower stratosphere (68 mb) toward the respective summer hemisphere. Low  $\text{CH}_4$  is observed in the springtime high latitudes of both hemispheres; these low polar values in the NH have received less attention than those in the SH (e.g., Russell et al. 1993a; Schoeberl et al. 1995), but appear very similar in these equivalent latitude data. These overall variations of  $\text{CH}_4$  are in reasonable agreement with seasonality in the mean vertical circulation at 68 mb shown in Fig. 10 (calculated as discussed in section 2d). The patterns of high tropical  $\text{CH}_4$  follow the upward velocity variations over low latitudes; note the weak tropical maximum during NH winter and the similar latitudinal movement of the contours (the  $w^* = 0$  contour is

included in the lower panel in Fig. 9). Furthermore, there are springtime maxima in downward motion over both poles, in agreement with the corresponding minima in  $\text{CH}_4$ .

The  $\text{CH}_4$  seasonal cycle in the middle stratosphere (10 mb) shows weakening of the midlatitude gradients during winter–spring in both hemispheres; this effect is more obvious in the SH. Similar midlatitude evolution at 10 mb is seen in the seasonal variation of nitrous oxide ( $\text{N}_2\text{O}$ ) measured by CLAES (Randel et al. 1994). At 10 mb strong  $\text{CH}_4$  gradients are seen across both polar vortices beginning in autumn, and the larger and stronger SH vortex is echoed in these  $\text{CH}_4$  measurements.

Seasonality in the upper stratosphere (2.2 mb) is very different from the lower levels, showing a strong semiannual oscillation (SAO) with distinct maxima in the respective summer subtropics. These maxima are very similar to the patterns of vertical velocity at 2.2 mb (shown in Fig. 10), with the  $\text{CH}_4$  maxima lagging the

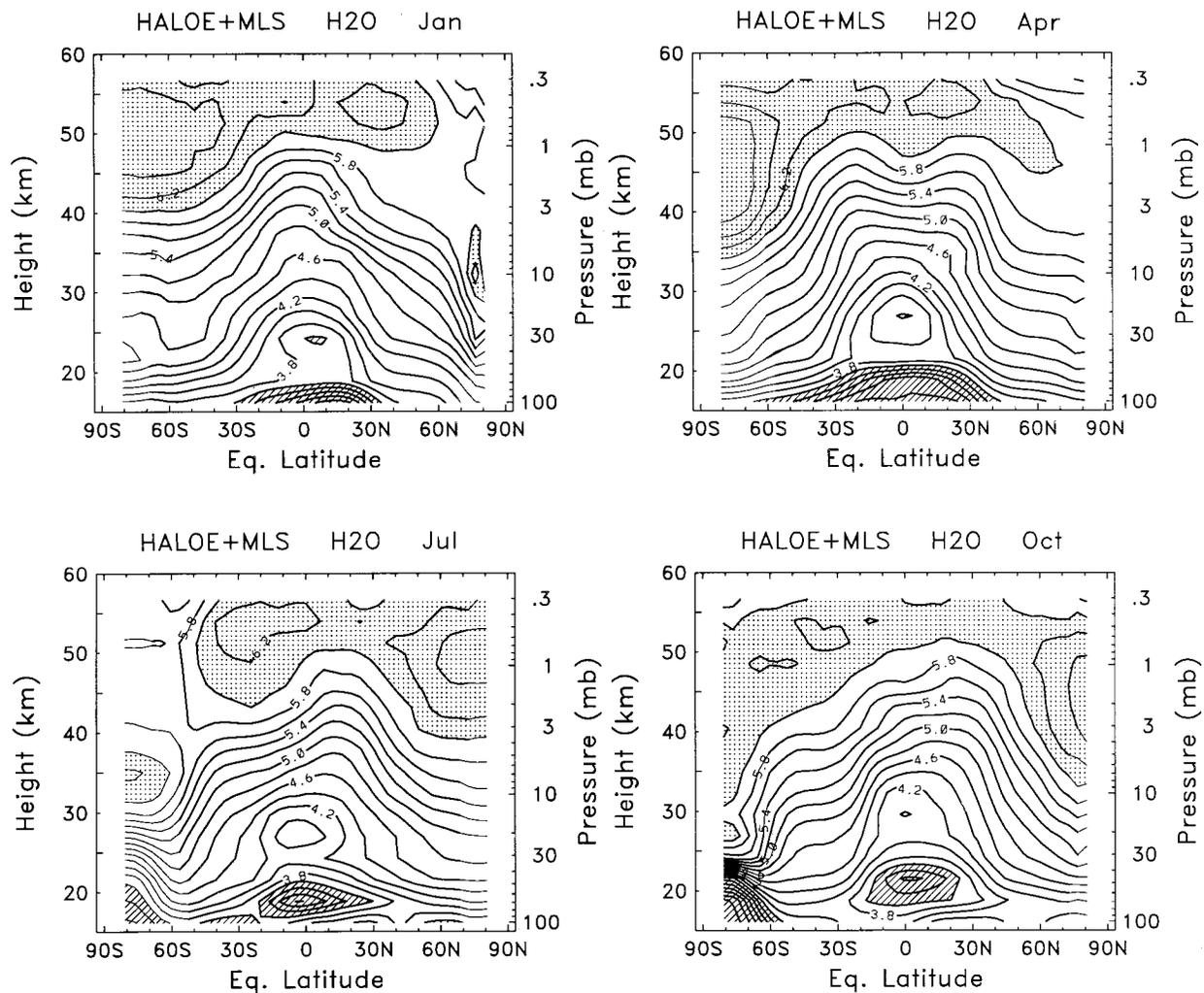


FIG. 7. Seasonal cycle estimates of  $\text{H}_2\text{O}$  in January, April, July, and October. Contour interval is 0.2 ppmv. Values below 3.6 ppmv are hatched, and values above 6.0 ppmv are stippled.

upward motion by approximately 1 month. A double-peaked latitudinal structure in  $\text{CH}_4$  is seen over April–June.

Figure 11 shows the altitude–time variation of  $\text{CH}_4$  at  $76^\circ\text{N}$  and  $\text{S}$ ; note the time axes have been shifted in order to directly compare midwinter variations over both poles. The overall evolution is similar between hemispheres, with regular downward movement of the isolines during autumn and winter, followed by rapid elevation of the contours in spring (associated with break-up of the polar vortices and mixing in of midlatitude air). Somewhat lower  $\text{CH}_4$  values are observed in the SH middle and upper stratosphere in winter, and the springtime transition is more dramatic and somewhat later in the SH; these aspects are due to the larger and more isolated SH polar vortex. The 0.3–0.8 isolines show consistent downward movement in the SH middle stratosphere during autumn and winter with a rate of  $-1.0$  to  $-1.5$   $\text{km month}^{-1}$ ; there is little downward

movement in the lower stratosphere until spring (September–October), when values near  $-1.5$   $\text{km month}^{-1}$  ( $-0.6$   $\text{mm s}^{-1}$ ) are inferred. These latter values are in excellent agreement with the downward velocities derived from the HALOE  $\text{CH}_4$  data by Schoeberl et al. (1995), and with the TEM vertical velocities shown in Fig. 10 (see also Rosenfield et al. 1994). The NH middle stratosphere data (e.g., the 0.5 contour) yields descent rates near  $-1.5$   $\text{km month}^{-1}$ , similar to the SH. In the NH lower stratosphere the data in Fig. 11 shows a relative maximum in November–December (see also the lower panel of Fig. 9) that is inconsistent with the downward circulation in Fig. 10. This isolated feature is entirely due to the use of CLAES data during one year (1992), and its reality is suspect.

Figure 12 compares the observed wintertime variation of zonal-mean methane ( $\bar{\chi}$ ) near the South Pole ( $76^\circ\text{S}$ ) with a transport calculation based solely on the mean vertical velocity  $w^*$ :

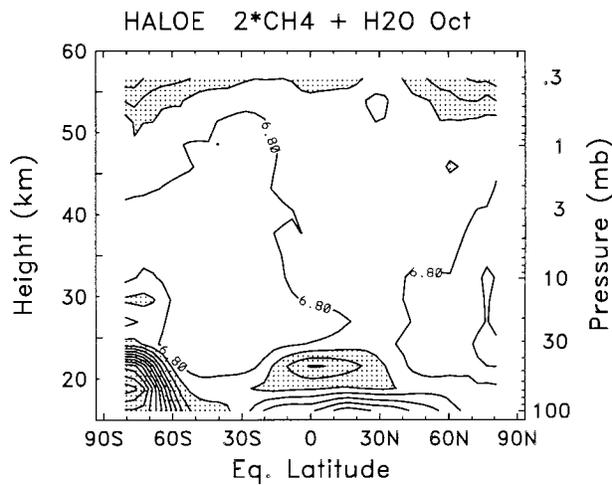


FIG. 8. Meridional cross section of the quantity  $\text{H}_2\text{O} + 2\text{CH}_4$  in October, derived from the seasonal cycle  $\text{H}_2\text{O}$  and  $\text{CH}_4$  data (Figs. 6 and 7). Contour interval is 0.2 ppmv, and values less than 6.6 ppmv are shaded.

$$\bar{\chi}(t) = \bar{\chi}(\text{March}) - \int_{\text{March}}^t \overline{w^* \chi_z} dt. \quad (2)$$

This is derived from Eq. (1), neglecting the  $\overline{v^* \chi_y}$ ,  $\nabla \cdot \mathbf{M}$ , and  $\bar{S}$  terms. This is a reasonable approximation inside the Antarctic winter vortex, where  $\overline{w^*}$  transport dominates  $\overline{v^*}$ , the chemical lifetime is very long ( $\bar{S}$  is small), and eddy mixing with the vortex exterior is small (Bowman 1993; Chen et al. 1994; Strahan et al. 1996). March is chosen for the starting time as this is near the formation of the SH polar vortex, and the idea here is to test how well calculated descent rates in the vortex agree with observations (the comparisons go through October, just prior to vortex breakup). The comparisons in Fig. 12 show quite reasonable agreement at both the 10- and 46-mb levels. There is relatively strong downward motion in autumn (March–May) at 10 mb, followed by a relative minimum in  $\overline{w^*}$  and more constant  $\text{CH}_4$  in winter. At 46 mb there is relatively little downward motion until late winter–spring (see also Figs. 9 and 10). The approximate agreement in these comparisons provides a confirmation of the heating rates and vertical velocities derived from the meteorological analyses, and also supports the notion of vortex isolation in the SH (e.g., Bowman 1993; Chen et al. 1994; Manney et al. 1994; Bacmeister et al. 1995). Similar calculations in the NH (beginning in September) are shown in Fig. 13; here there is poor agreement, with the observed  $\text{CH}_4$  changes much less than those calculated from  $\overline{w^*}$ . This is likely due to stronger meridional transport into and less isolation of the NH polar vortex.

Both NH and SH data in Fig. 11 show  $\text{CH}_4$  minima (below 0.1 ppmv) near the stratopause during summer (see also the top panel in Fig. 9). These minima are due to rapid photochemical destruction of  $\text{CH}_4$  over polar regions during summer (see Fig. 17 below); the cal-

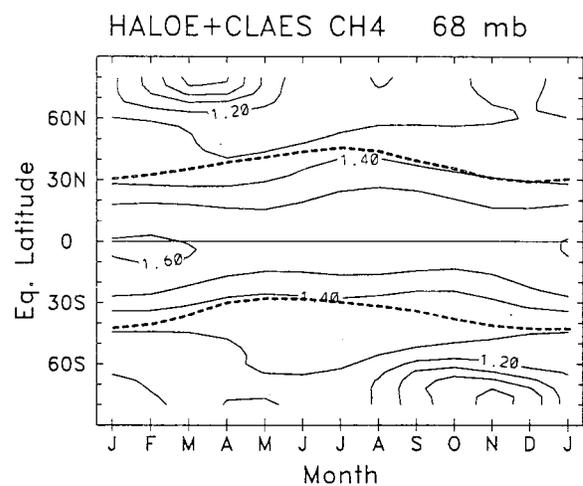
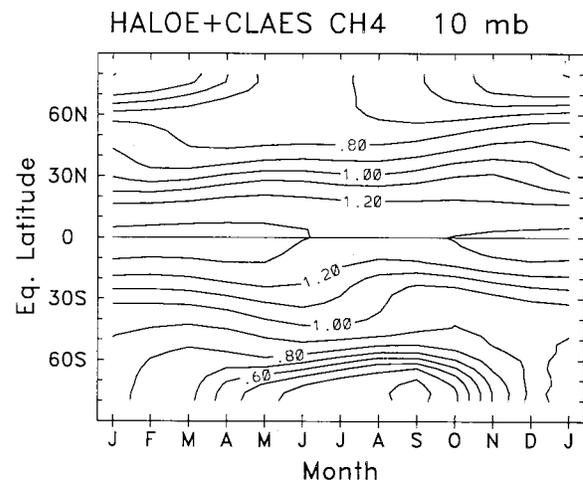
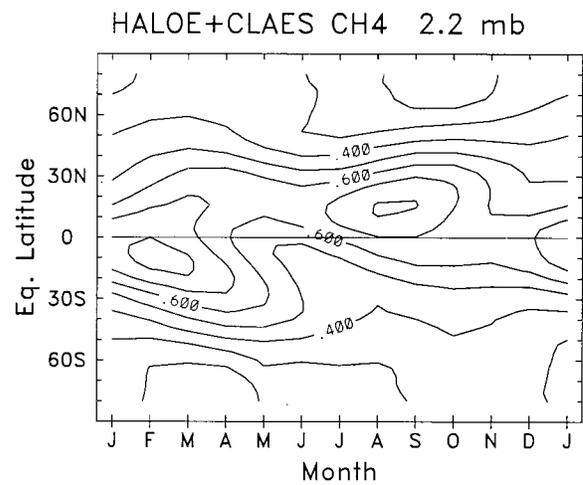


FIG. 9. Latitude–time sections of seasonal cycle variations in  $\text{CH}_4$  at 2.2 mb (top), 10 mb (middle), and 68 mb (bottom). Contour interval is 0.01 ppmv. The dashed lines in the lower panel are the  $\overline{w^*} = 0$  contours (from Fig. 10).

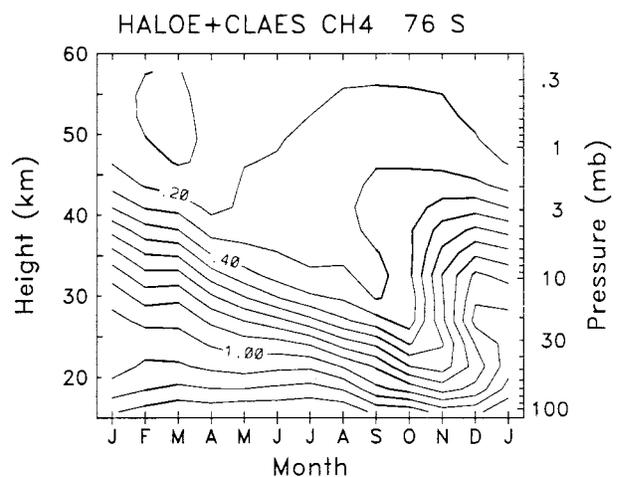
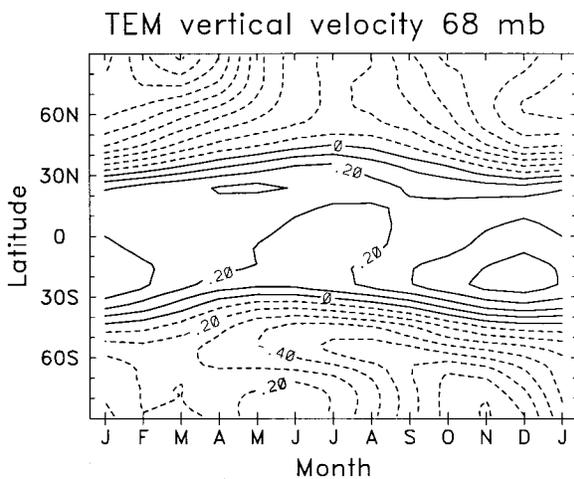
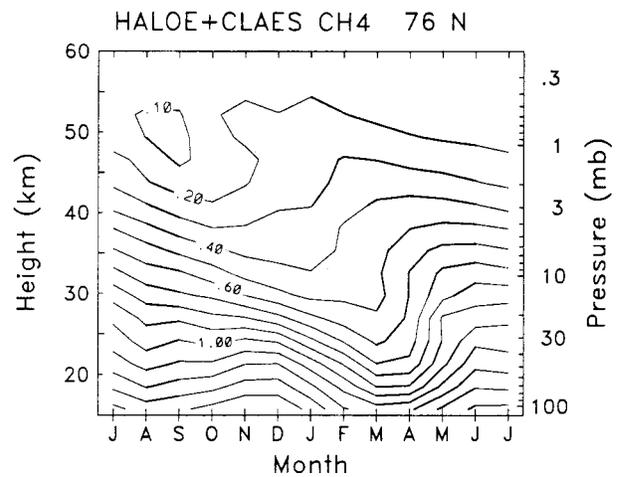
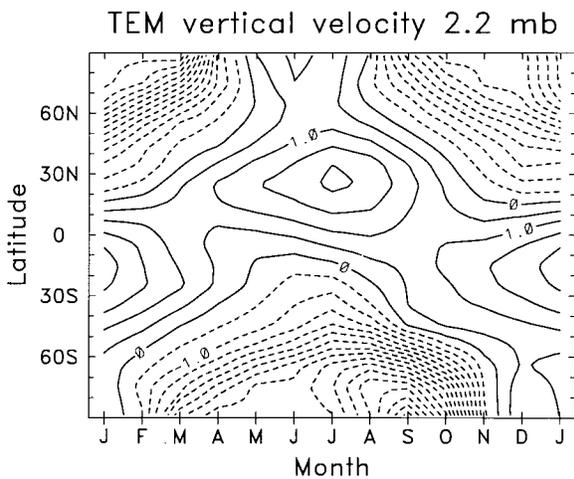


FIG. 10. Latitude–time sections of residual mean vertical velocity ( $w^*$ ) derived from UKMO data at 2.2 mb (top) and 68 mb (bottom). Contour intervals are 0.5 and 0.1  $\text{mm s}^{-1}$ , respectively, and positive values denote upward motion.

FIG. 11. Altitude–time sections of seasonal cycle variations in  $\text{CH}_4$  at  $76^\circ\text{N}$  (top) and  $76^\circ\text{S}$  (bottom). Contour interval is 0.1 ppmv. Note the respective time axes are shifted by 6 months between the two panels.

culated photochemical lifetime of  $\text{CH}_4$  in this region is near 1 month [see Fig. 1 of Solomon et al. (1986)].

### b. Tropical variations

The altitude–time variations of both  $\text{CH}_4$  and  $\text{H}_2\text{O}$  over the equator are shown in Fig. 14, which also includes diagrams with the respective time means removed at each altitude to highlight seasonality. There is a strong upward propagating annual variation in  $\text{H}_2\text{O}$  in the lower stratosphere, associated with the seasonal variation of tropical tropopause temperatures and the freezing out of  $\text{H}_2\text{O}$  in the tropical upwelling region [see extensive discussions in Mote et al. (1995) and (1996)]. A nearly identical pattern is seen in the total hydrogen  $2\text{CH}_4 + \text{H}_2\text{O}$ , as illustrated in a different manner in Fig. 15. Here the individual zonal means over  $4^\circ\text{N–S}$  are plotted, along with the associated harmonic fits,

for pressure levels 100–10 mb (16–32 km). A strong annual cycle that progresses upward with time is observed over 100–32 mb; this cycle can be traced to 22 mb using the harmonic analysis, although there is substantially more interannual variability at this level. At 15 and 10 mb (29–32 km), the annual harmonic is relatively weak, and a semiannual component dominates at these levels (and above). The upward propagation of the annual signal over 100–32 mb (16–24 km) takes approximately 11 months; this corresponds to a vertical velocity of  $\sim 0.7 \text{ km month}^{-1}$  ( $\sim 0.25 \text{ mm s}^{-1}$ ), in reasonable agreement with the tropical TEM vertical velocities in Fig. 10.

Figure 16 shows the space–time structure of both  $\text{H}_2\text{O}$  and  $2\text{CH}_4 + \text{H}_2\text{O}$  at 100, 68, and 46 mb. While the annual cycles at 68 and 46 mb are approximately centered over the equator, variations at 100 mb are maxi-

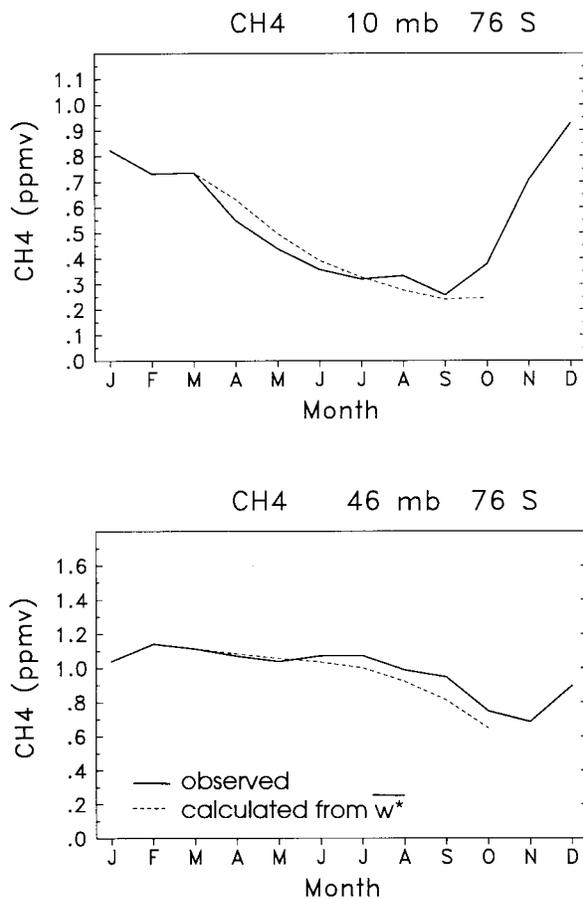


FIG. 12. Solid lines show seasonal cycles of  $\text{CH}_4$  at  $76^\circ\text{S}$ , 10 mb (top), and 46 mb (bottom). Dashed lines show  $\text{CH}_4$  estimated from  $w^*$  transport [Eq. (2)] between March and October.

imum near  $15^\circ\text{--}30^\circ\text{N}$ . Lowest values are observed near  $15^\circ\text{N}$  in February–March, although the minimum 100-mb temperatures occur near the equator (e.g., Newell and Gould-Stewart 1981; Tuck et al. 1993). The maximum during NH summer (August–September) is even more asymmetrical, with maximum near  $20^\circ\text{--}30^\circ\text{N}$ ; this structure has little relation to zonal-mean temperatures. There is also a seasonal variation in meridional transport at 100 mb inferred in Fig. 16, with relatively little spreading of the low subtropical values into the NH during NH winter (note the strong gradients near  $30^\circ\text{N}$ ), but stronger propagation of the NH summer maximum into high latitudes (with a time lag of order three months between  $30^\circ$  and  $60^\circ\text{N}$ ). There is much less spreading of this maximum into SH midlatitudes. Based on analyses of these same HALOE  $\text{H}_2\text{O}$  data, RO97 discuss the asymmetry seen in Fig. 16 and note the meridional propagation of maximum values into NH midlatitudes. The asymmetry of these patterns (particularly the NH summer maximum) contribute to the higher values of  $\text{H}_2\text{O}$  observed in the NH versus SH lower stratosphere, as reported by Kelly et al. (1990). These data suggest the NH–SH asymmetry in the lower stratosphere is not

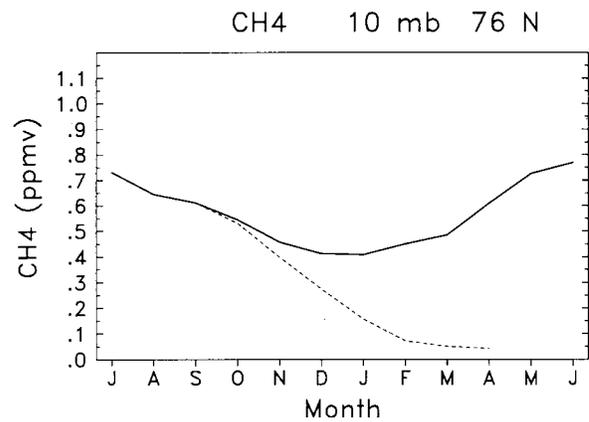


FIG. 13. Observed  $\text{CH}_4$  variations at  $76^\circ\text{N}$  and 10 mb (solid line) and that calculated from  $w^*$  transport (dashed line).

due to asymmetric transport of equatorially centered maxima, but to dynamical processes in the NH subtropics (such as the Indian monsoon during NH summer). The enhanced meridional spreading of the  $\text{H}_2\text{O}$  and  $2\text{CH}_4 + \text{H}_2\text{O}$  at 100 mb compared to levels at and above 68 mb agrees with the results of Trepte et al. (1993), who present evidence for rapid meridional flow between Tropics and midlatitudes below 20 km, but not above this altitude. Similar results were inferred from the aerosol observations in Grant et al. (1994). This behavior is consistent with the tropical vertical velocity profile discussed by RO97: there is a minimum in  $w^*$  near  $50\text{--}70$  mb, such that only 20% of the upward mass flux at 100 mb continues upward across this level (with the remaining 80% moving laterally to either hemisphere). The data in Fig. 16 also provide convincing evidence that there is not strong mixing of the dehydrated SH polar vortex air into midlatitudes following vortex breakup (as also noted by RO97).

The  $\text{H}_2\text{O}$  data at 68 and 46 mb in Fig. 16 show minimum values in the Tropics and relatively strong horizontal gradients near both  $30^\circ\text{N}$  and  $30^\circ\text{S}$ , similar (but opposite) to the  $\text{CH}_4$  latitudinal structure seen in Fig. 9. The lack of such gradients in the quantity  $2\text{CH}_4 + \text{H}_2\text{O}$  (i.e., the 46-mb plot in Fig. 16) implies that it is the downward transport of photochemically aged air in the extratropics, plus a lack of rapid meridional mixing with the Tropics, that is responsible for these latitudinal  $\text{H}_2\text{O}$  gradients. Seasonal variations in both  $\text{H}_2\text{O}$  and  $2\text{CH}_4 + \text{H}_2\text{O}$  show that the upward propagating tropical annual cycle is confined to  $\sim 30^\circ\text{N--S}$ .

Inspection of Fig. 16 and the time series in Fig. 15 shows an attenuation of the amplitude of this annual cycle over  $100\text{--}32$  mb (in both  $\text{H}_2\text{O}$  and  $2\text{CH}_4 + \text{H}_2\text{O}$ ) from approximately 1.5 to 0.7 ppmv; the attenuation is most rapid between 100 and 68 mb. Above 68 mb, where the tropical signal appears isolated from midlatitudes, the annual amplitude decreases from 1.1 to 0.7 ppmv between 68 and 32 mb. Supposing that this attenuation is due entirely to the mixing in of extratropical air (with

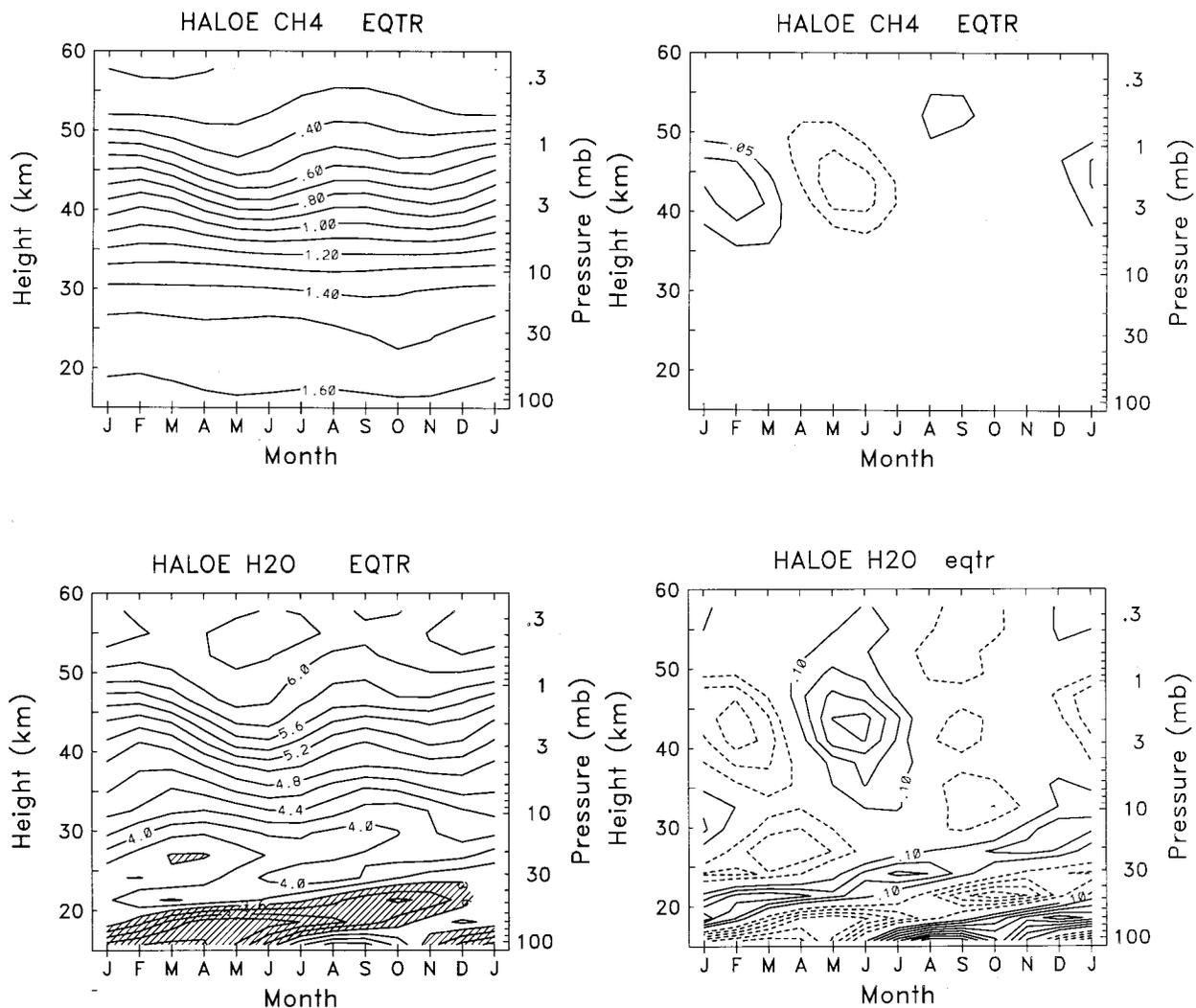


FIG. 14. Altitude–time sections of seasonal cycle variations in  $\text{CH}_4$  (top) and  $\text{H}_2\text{O}$  (bottom) over the equator. Left panels show full fields, while right panels show seasonal anomalies (calculated by subtracting the time means at each pressure level). Values below 3.6 ppmv in the lower-left panel are shaded.

very small annual cycle in  $2\text{CH}_4 + \text{H}_2\text{O}$ , of order 0.1–0.2 ppmv; see Fig. 16), an estimate of the mixing time  $\tau$  may be estimated from

$$\frac{dA}{dt} = \frac{A - A_0}{\tau},$$

where  $A$  is the amplitude of the tropical annual harmonic,  $A_0$  is the midlatitude value, and  $d/dt$  is the derivative following the upward propagating signal. This is an approximation to Eq. (1), where the meridional and eddy transport terms are approximated as a mixing process with timescale  $\tau$ . Using  $dA/dt = (0.4 \text{ ppmv per } 10 \text{ months})$  between 68 and 32 mb, and  $(A - A_0) \sim (0.6 \text{ ppmv})$  averaged over 68–32 mb, gives an estimate of  $\tau \sim 15$  months. This is a lower limit on the meridional mixing time for air entering the Tropics (30°N–S) from

middle latitudes above 19 km, in order to maintain the observed tropical annual cycle. The estimate of 15 months is in reasonable agreement with that derived by Volk et al. (1996) from aircraft constituent measurements, and by Schoeberl et al. (1997) using *UARS* constituent data.

The upper stratosphere signal in Fig. 14 is dominated by a semiannual oscillation (SAO), with a much stronger amplitude during the first half of the year. This is a well-known aspect of the SAO (e.g., Dunkerton and Delisi 1988), related to stronger extratropical wave driving in the NH winter (as compared to the SH winter). Variations in  $\text{H}_2\text{O}$  echo those in  $\text{CH}_4$ , such that the total hydrogen  $2\text{CH}_4 + \text{H}_2\text{O}$  is nearly constant throughout the seasonal cycle. Dynamics of the SAO is discussed further in the next section.

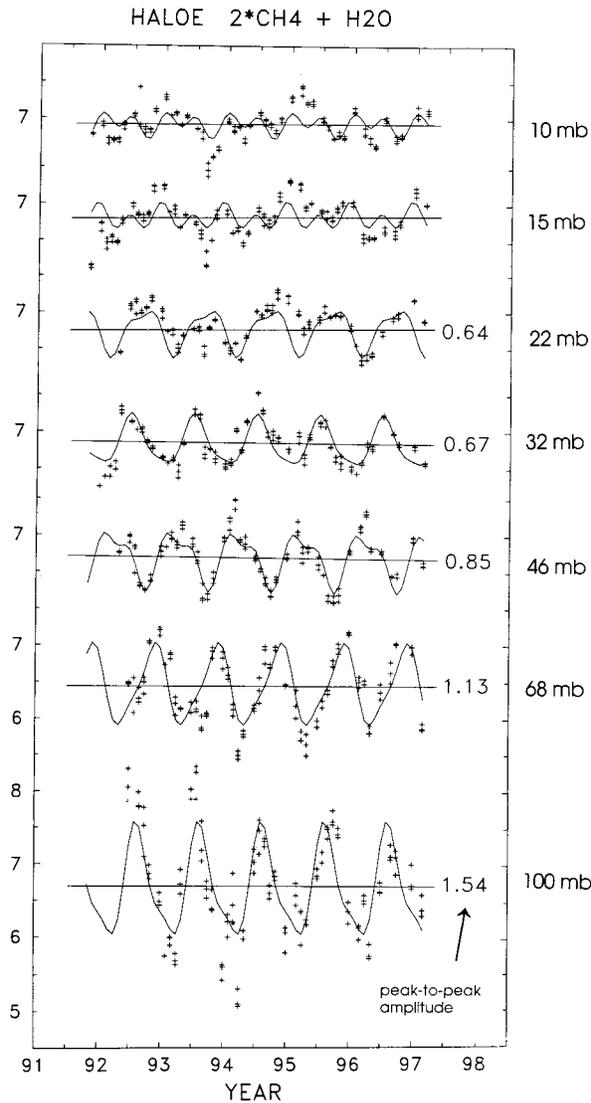


FIG. 15. Time series of  $\text{H}_2\text{O} + 2\text{CH}_4$  over  $4^\circ\text{N}–4^\circ\text{S}$  during 1991–1997, for pressure levels 100 mb to 10 mb. Crosses show monthly means and thin lines show (repeating) seasonal cycle fits at each level.

### c. Global budgets and estimates of eddy transport

In this section we examine details of the constituent continuity equation [Eq. (1)] applied to the observed seasonal cycle variations in global  $\text{CH}_4$ . For these calculations we use the monthly average residual mean circulation ( $\overline{v^*}$ ,  $\overline{w^*}$ ) derived from UKMO data (e.g., Fig. 10), and the chemical sink term  $S$  is approximated by a linear relaxation expression

$$\overline{S} = -\frac{\overline{\chi}}{\tau},$$

with  $\text{CH}_4$  lifetimes ( $\tau$ ) taken from the model results of Garcia and Solomon (1983). The  $\nabla \cdot \mathbf{M}$  term in Eq. (1) is a complicated quantity involving eddy covariances, and not derivable from the data here. We calculate this

term as a residual to the balance in Eq. (1); it thus contains the actual eddy transports plus any errors in the explicitly calculated terms.

Figure 17 shows the structure of the individual terms  $\partial\overline{\chi}/\partial t$ ,  $-\overline{v^*}\overline{\chi}_y$ ,  $-\overline{w^*}\overline{\chi}_z$ ,  $\overline{S}$ , and  $\nabla \cdot \mathbf{M}$  (calculated as a residual) for the seasonal  $\text{CH}_4$  budget, for statistics averaged over January–February. Transport by the residual mean circulation is associated with large positive tendencies in the tropical upper stratosphere (due to upward and northward transport of relatively high  $\text{CH}_4$ ) and negative tendencies over winter polar regions (due to downward transport of low  $\text{CH}_4$ ). The parameterized chemical sink  $\overline{S}$  is an important component of the budget in the tropical upper stratosphere, with relatively large values also over the summer (SH) high latitudes (this photochemical destruction leads to the very low  $\text{CH}_4$  summer pole amounts observed in Fig. 11). The calculated residual ( $\nabla \cdot \mathbf{M}$  term) shows a north–south dipole pattern over the Tropics and NH, with opposite sign to the mean meridional circulation transport. Although this is a residual calculation, the structure of this field shows characteristics very similar to  $\nabla \cdot \mathbf{M}$  calculated explicitly using general circulation model output in Randel et al. (1994) (see their Fig. 8). An important point is that these eddy transport effects extend well into the Tropics in both model calculations and the residual estimate here. Note also that the tendency term  $\partial\overline{\chi}/\partial t$  is a relatively small part of the overall budget, so that accurate simulation of  $\partial\overline{\chi}/\partial t$  will be sensitive to small uncertainties in calculation of the other budget terms.

Figure 18 shows altitude–time plots of several  $\text{CH}_4$  seasonal budget terms over the equator [ $-\overline{v^*}\overline{\chi}_y$ ,  $-\overline{w^*}\overline{\chi}_z$  and  $\nabla \cdot \mathbf{M}$  (residual)]. The chemical sink  $S$  (not shown) is nearly constant over the equator throughout the year, and the observed tendency  $\partial\overline{\chi}/\partial t$  (not shown) is a relatively small component. Transport by the mean meridional circulation ( $\overline{v^*}$ ,  $\overline{w^*}$ ) exhibits a pronounced semiannual variation in the upper stratosphere, with maxima near the solstices. Transport by both mean circulation components is important in this variation: the  $\overline{w^*}$  transport is a maximum during NH winter, while the  $\overline{v^*}$  component exhibits a dominant maximum during SH winter (along with a weaker NH winter peak). These enhanced  $\overline{v^*}$  transports during both solstices are due to strong flow toward the winter hemisphere (see Fig. 17) combined with enhanced meridional constituent gradients near the equator (see Fig. 6). The budget residual ( $\nabla \cdot \mathbf{M}$  in Fig. 18) also shows a strong semiannual variation at the equator for altitudes above approximately 30 km. Although some component of this may be due to budget uncertainties, this semiannual seasonality is consistent with enhanced planetary wave transport in the respective winter hemispheres reaching into the Tropics (as discussed above). The strong semiannual forcing (by both mean flow and eddy components) is likely responsible for the dominance of semiannual behavior for the tropical constituent fields above  $\sim 30$  km (see Figs. 14 and 15).

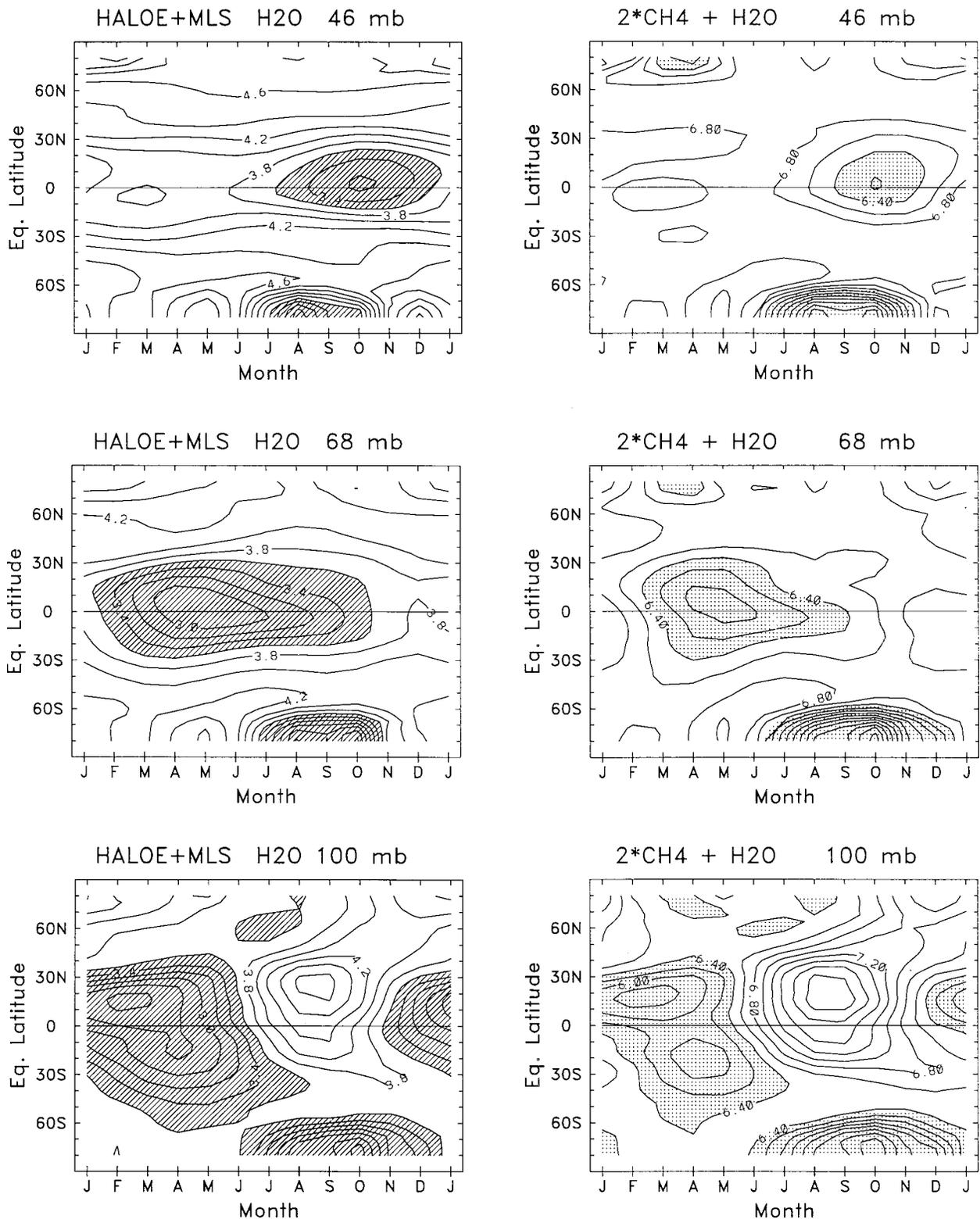


FIG. 16. Latitude–time plots of seasonality in H<sub>2</sub>O (left) and H<sub>2</sub>O + 2CH<sub>4</sub> (right) at 100 mb (bottom), 68 mb (middle), and 46 mb (top). Contour interval is 0.2 ppmv in each panel.

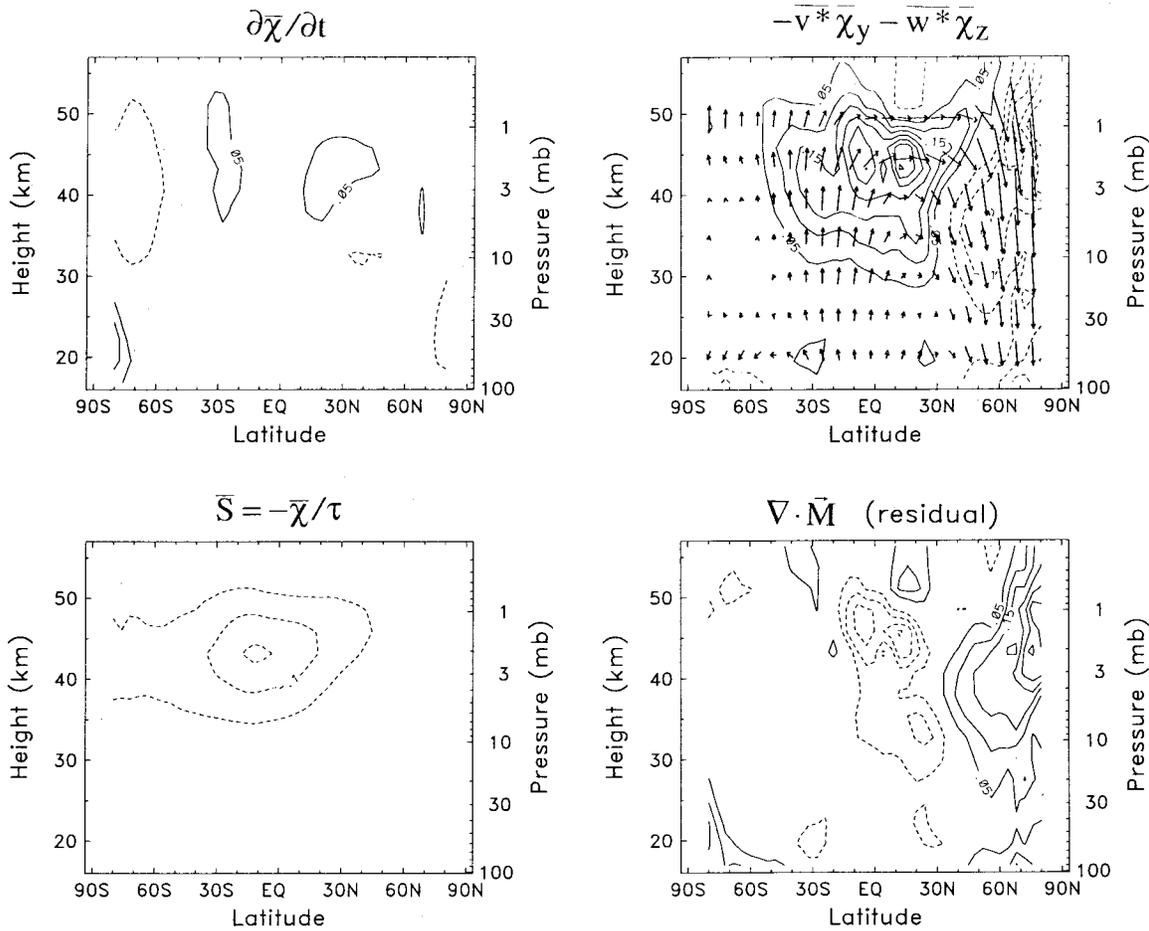


FIG. 17. Meridional cross sections of individual terms in the seasonal  $\text{CH}_4$  continuity equation [Eq. (1)], for averages during January–February. Contour interval in all panels is  $0.05 \text{ ppmv month}^{-1}$ , with zero contours omitted. Vectors in the upper right-hand panel denote components of the residual mean circulation ( $v^*$ ,  $w^*$ ). The  $\nabla \cdot \bar{\mathbf{M}}$  term (lower-right panel) is calculated as a residual.

#### 4. Interannual variability and the QBO

##### a. Observations

This section focuses on interannual variations in  $\text{CH}_4$  and  $\text{H}_2\text{O}$  over 1991–97, defined as deviations from the seasonal cycles defined above (with linear trends also removed). Figure 19 shows altitude–time sections of  $\text{CH}_4$  anomalies over the equator (average over  $10^\circ\text{N}$ – $10^\circ\text{S}$ ); here data gaps have been interpolated and the monthly anomalies smoothed slightly in time. Largest  $\text{CH}_4$  anomalies are observed in the upper stratosphere, over 35–45 km, with an approximate 2-yr periodicity; these values represent approximately  $\pm 10\%$  deviations from the time mean. The approximate 2-yr periodicity in  $\text{CH}_4$  anomalies suggests a link with the equatorial quasibiennial oscillation (QBO), and this is confirmed by comparison with the UKMO equatorial zonal wind anomalies shown in Fig. 20a (with the  $\text{CH}_4$  anomalies superimposed). Specifically, the  $\text{CH}_4$  anomalies over 35–45 km are approximately in phase with the zonal wind anomalies near 30 km.

Figure 21 shows the  $\text{H}_2\text{O}$  interannual anomalies over the equator. Maxima are observed in the upper stratosphere with very similar (but oppositely signed) patterns to the  $\text{CH}_4$  anomalies, reflecting conservation of  $2\text{CH}_4 + \text{H}_2\text{O}$  on interannual (as well as seasonal) timescales. There are also significant  $\text{H}_2\text{O}$  anomalies in the lower and middle stratosphere; there are relative maxima over 27–33 km with an approximate 2-yr periodicity, but these are not observed in the  $\text{CH}_4$  data.

Space–time structure of coherent QBO variations in  $\text{CH}_4$  may be isolated in an optimal manner using singular value decomposition (SVD) (Randel and Wu 1996). This involves manipulation of the covariance matrix between  $\text{CH}_4$  anomalies (as a function of latitude and altitude) and zonal wind anomalies (over 10–70 mb), and yields modal spatial structures that explain optimum fractions of covariance between these fields. Figure 22 shows the spatial structure of the first two modes derived from this analysis (termed SVD1 and SVD2); patterns show the location (and phase) of anomalies in  $\text{CH}_4$  that are coherent with the QBO zonal wind

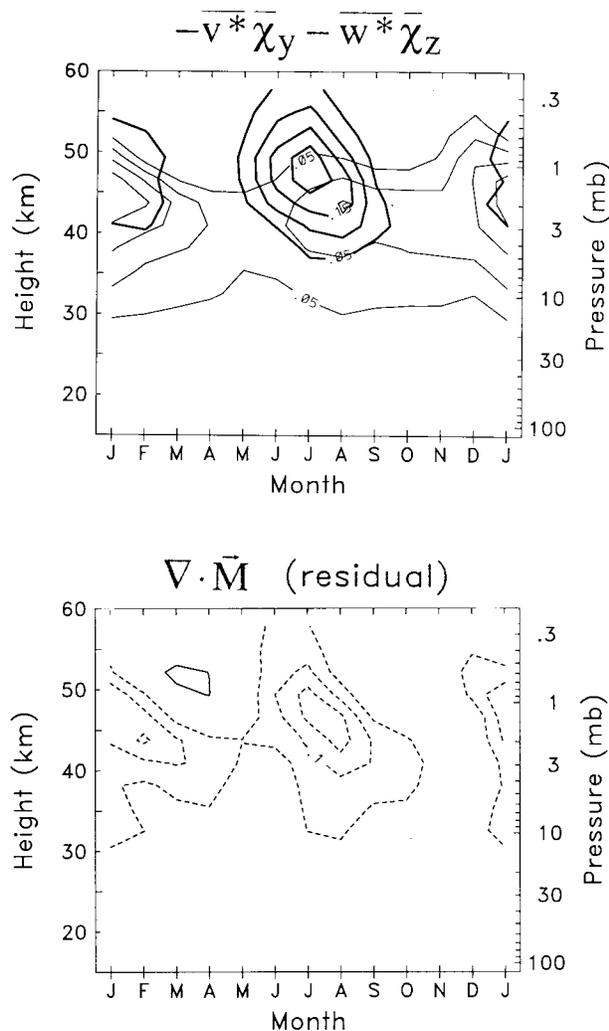


FIG. 18. Altitude-time sections of terms in the seasonal  $\overline{\text{CH}_4}$  continuity equation [Eq. (1)] over the equator. Shown are  $-\overline{v^* \chi_y}$  (top panel, heavy lines),  $-\overline{w^* \chi_z}$  (top panel, light lines), and  $\nabla \cdot \overline{\mathbf{M}}$  (calculated as a residual—bottom panel). Contour interval in both panels is  $0.05 \text{ ppmv month}^{-1}$ , with zero contours omitted.

anomalies. (Two such modes are typical for a propagating oscillation, analogous to a harmonic sine and cosine Fourier decomposition.) The spatial structure of SVD1 shows a maximum in the upper stratosphere over  $35\text{--}45 \text{ km}$ , centered in the Tropics over approximately  $30^\circ\text{N--S}$ . This structure corresponds to the equatorial anomalies seen in Fig. 19. The structure of SVD2 shows a narrow maximum over the equator at  $40\text{--}45 \text{ km}$ , and several maxima over  $\sim 10^\circ\text{--}40^\circ$  in both hemispheres [note these modes are spatially (and temporally) orthogonal by construction]. These extratropical patterns are qualitatively similar to QBO variations observed in column and profile ozone and  $\text{NO}_2$  data (Bowman 1989; Randel and Wu 1996), and deduced from CLAES  $\text{N}_2\text{O}$  data by O'Sullivan and Dunkerton (1997). The QBO patterns in Fig. 22 are largest near gradients in the back-

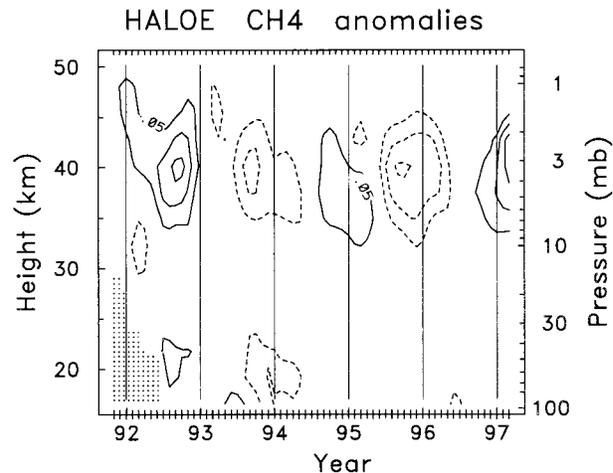


FIG. 19. Altitude-time section of interannual anomalies in  $\text{CH}_4$  over the equator. Contour interval is  $0.05 \text{ ppmv}$ , with zero contours omitted. The vertical lines denote January for each year.

ground  $\text{CH}_4$  structure (included in Fig. 22), as anticipated for anomalies due to transport variations. Note the similarity of these QBO patterns with the total interannual variance map shown in Fig. 5.

Temporal evolution of the QBO patterns in  $\text{CH}_4$  is revealed by projection of the full monthly  $\text{CH}_4$  anomalies onto the spatial patterns in Fig. 22. Figure 23 shows a phase-space plot of the projections of SVD1 versus SVD2 over the entire time record 1991–97. Nearly regular counterclockwise phase progression is observed as the QBO anomalies project successively onto the patterns in Fig. 22. This nearly regular QBO phase variation in  $\text{CH}_4$  data is analogous to the harmonic QBO variations in dynamical quantities discussed by Wallace et al. (1993), and the ozone variations shown in Randel and Wu (1996).

Figure 24 shows  $\text{CH}_4$  data observed during April for four years (1993–96); these time periods are indicated in Fig. 23 and correspond to opposing projections of SVD1 and SVD2. These plots demonstrate that there is a strong effect of the QBO in modulating the double-peaked structure in upper-stratospheric  $\text{CH}_4$ : data during April 1993 and 1995 (westerly QBO winds in Fig. 20a) exhibit a pronounced double peak, whereas April 1994 and 1996 (easterly QBO winds) do not.

Figure 25 shows latitude-time series at  $3 \text{ mb}$  of the full  $\text{CH}_4$  field and the detrended anomalies (which exhibit a clear QBO dependence). Time periods of enhanced double-peak structure are also indicated in Fig. 25. The QBO anomalies show maxima centered near the equator, with a suggestion of propagation into the subtropics, particularly into the NH. This latitudinal propagation near  $3 \text{ mb}$  is associated with successive projections of SVD1 and SVD2 (Fig. 22). The timing of these anomalies is such that large NH subtropical maxima occur during NH winter–spring; this is the time of seasonal maximum in SH subtropics, and this com-

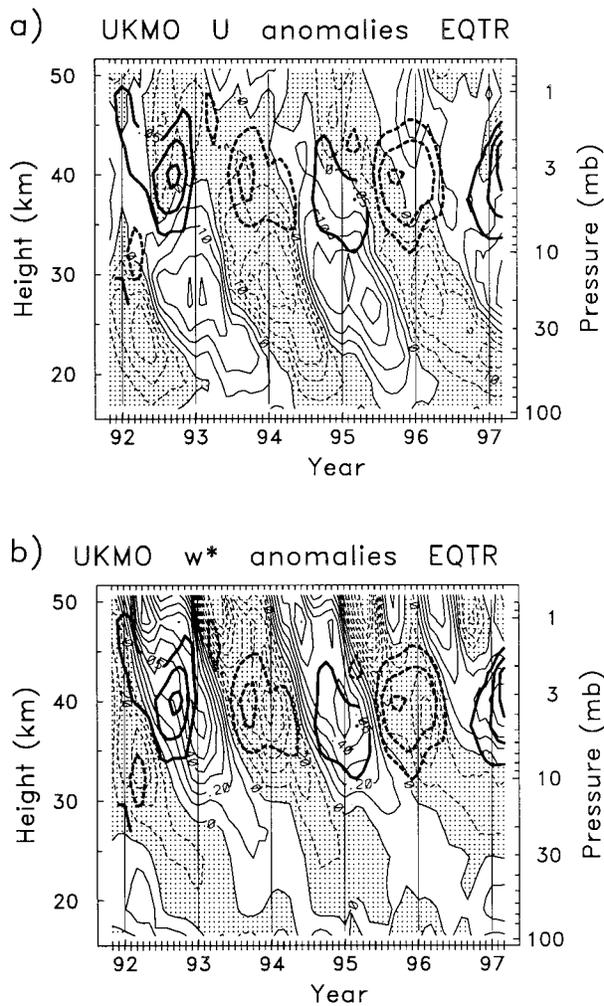


FIG. 20. Altitude–time sections of interannual anomalies in (a) zonal-mean zonal wind and (b) QBO-filtered vertical velocity  $w^*$  over the equator, derived from UKMO analyses. Contour interval is 5 m s<sup>-1</sup> and 0.1 km month<sup>-1</sup>, respectively. Heavy contours near 40 km are CH<sub>4</sub> anomaly isolines (from Fig. 19).

bination leads to double-peaked structure for the positive QBO anomalies in NH spring 1993 and 1995. The details of this behavior clearly depend on the phase relationship between the QBO and the seasonal cycle, and this is something that will change on a decadal timescale (Gray and Dunkerton 1990). We note that relatively small anomalies are observed over the equator during these times; the double-peak structure (or its absence) in this time sample is mostly due to the presence of subtropical anomalies, particularly in the NH (corresponding to a positive projection of SVD1 + SVD2, i.e., Fig. 23).

Figure 26 shows the latitude–time evolution of CH<sub>4</sub> at 10 mb throughout 1991–97, for both the full-field and interannual anomalies. The anomalies at this altitude are very small at the equator but large over the subtropics of each hemisphere (somewhat larger in the NH), with a strong QBO time dependence. These interannual

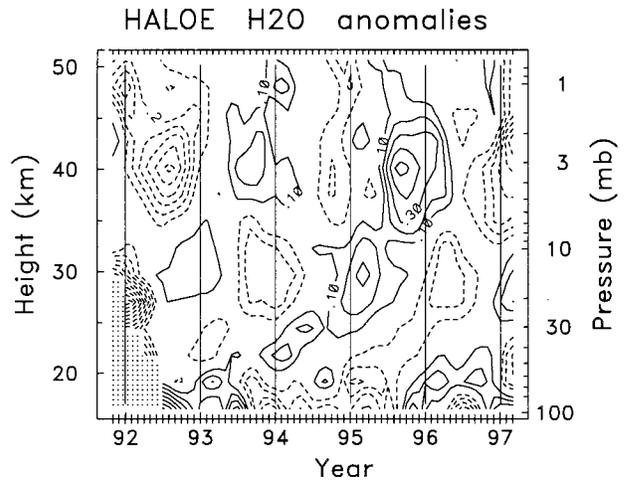


FIG. 21. Altitude–time sections of interannual anomalies in H<sub>2</sub>O over the equator. Contour interval is 0.1 ppmv, and zero contours are omitted.

anomalies are associated with a latitudinal shifting of the tropical maximum region in CH<sub>4</sub>, as seen in the upper panel of Fig. 26. The maximum is shifted toward the NH during QBO easterlies (near 30 mb) enhancing NH subtropical gradients, and toward the SH for QBO westerlies (decreasing these gradients). Similar CH<sub>4</sub> patterns are observed over approximately 22–10 mb (27–32 km), but not below 30 mb; these anomalies are associated with strong projection onto SVD2 in Fig. 22. Note the clear effect of these anomalies in the cross sections in Fig. 24, especially on the vertical gradient structure in NH midlatitudes. This is remarkable evidence of QBO modulation of the latitudinal position of the tropical maximum region (Trepte and Hitchman 1992) and the interaction with extratropics.

Figure 27 shows the latitudinal structure of H<sub>2</sub>O at 10 mb throughout 1991–97, for both the full field and the interannual anomalies. Large anomalies are observed in the NH subtropics, mirroring the CH<sub>4</sub> patterns in Fig. 26. Unlike the CH<sub>4</sub> data, the H<sub>2</sub>O anomalies extend across the equator, giving rise to the equatorial anomalies centered near 10 mb seen in Fig. 21. The overall structure of H<sub>2</sub>O at 10 mb shows latitudinal movement of the tropical minimum region and modulation of subtropical gradients on the QBO timescale, very similar to that seen for CH<sub>4</sub> in Fig. 26.

#### b. QBO budget calculations

The observed QBO anomalies in CH<sub>4</sub> and H<sub>2</sub>O likely result from transport variations associated with the QBO. Although a detailed budget analysis of QBO transport is beyond the scope of this work, there are some aspects of coupling with QBO circulation anomalies that deserve mention. Figure 20b shows an altitude–time section of the QBO anomalies in residual mean vertical velocity ( $w^*$ ) over the equator derived from UKMO data (Randel

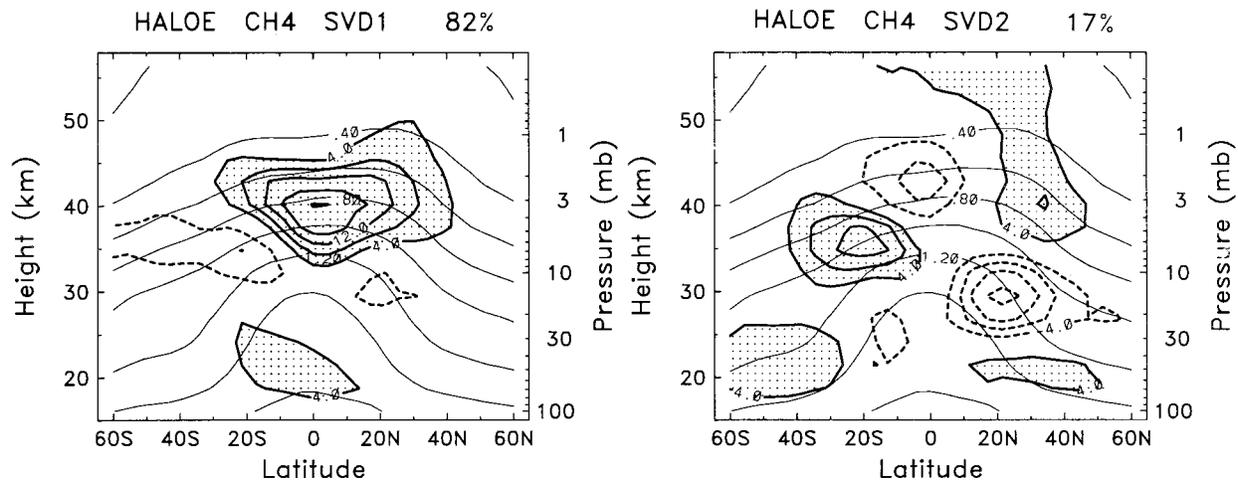


FIG. 22. Heavy solid and dashed lines with shading show the spatial structure of QBO SVD1 (left) and SVD2 (right) for CH<sub>4</sub>. Contours show local mixing ratio variations with arbitrary units. Light lines denote background annual mean CH<sub>4</sub> structure.

et al. 1997). These data have been filtered to isolate QBO variations, but the QBO patterns are clearly evident in the full interannual anomaly fields (see W. Randel et al. 1998, manuscript submitted to *J. Atmos. Sci.*). These  $w^*$  anomalies are maximum near regions of largest vertical shear of the zonal-mean wind (Fig. 20a)—which are also regions of largest QBO temperature anomalies—and exhibit similar downward propagation in time. These QBO anomalies in  $w^*$  are of order 10%–20% of the time-mean background values; largest  $w^*$  anomalies are observed in the upper stratosphere (35–45 km), although largest QBO zonal winds are seen in the lower-middle strato-

sphere. Comparison between the vertical velocity anomalies and the equatorial CH<sub>4</sub> anomalies (also included in Fig. 20b) shows strong correlation in the upper stratosphere, with upward velocities associated with positive CH<sub>4</sub> maxima, and vice versa.

The observed space–time coherence between QBO anomalies in  $w^*$  and CH<sub>4</sub> in the equatorial upper stratosphere (Fig. 20b) is puzzling in light of Eq. (1). If vertical transport is the dominant effect (a reasonable assumption for equatorial QBO variations), the continuity equation [Eq. (1)] suggests a correlation between  $w^*$  and  $\partial\bar{\chi}/\partial t$ , rather than between  $w^*$  and  $\bar{\chi}$  as observed. An estimate of the budget based on Eq. (1) (not shown) results in large residuals with an approximate QBO periodicity, with a phase relationship such that the residual is a (negative) maximum during QBO westerly winds. One possible explanation is that eddy transport effects are modulated by the QBO zonal wind structure. Although this is speculative, based on variations in the residual to Eq. (1), the in-phase coherence between  $w^*$  and CH<sub>4</sub> (or H<sub>2</sub>O) anomalies near 40 km appears to be a robust result (leading to these large residuals).

Figure 28 shows a latitude–time diagram of the QBO anomalies in  $w^*$  at 10 mb. Together with alternating anomalies over the equator (over 10°N–S), this figure shows large anomalies in the NH subtropics that are approximately out of phase with those at the equator. These coupled patterns are likely a manifestation of the meridional circulation cells anticipated theoretically for the QBO (e.g., Plumb and Bell 1982), although the meridional scale is somewhat wider than in idealized models and there is substantial NH–SH asymmetry in these observations. The subtropical  $w^*$  maxima at 10 mb bear strong resemblance to the CH<sub>4</sub> and H<sub>2</sub>O anomaly patterns shown in Figs. 26 and 27. Comparison of the time evolution shows the circulation anomalies lead the constituent patterns by up to a quarter cycle (as expected for transport variations), al-

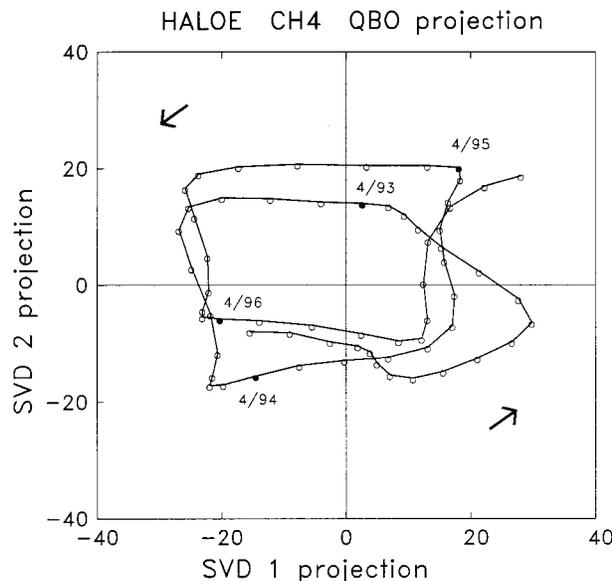


FIG. 23. Phase–space diagram of the projection of the monthly global CH<sub>4</sub> anomalies onto spatial structures SVD1 and SVD2 (shown in Fig. 22). Time progression corresponds to counterclockwise orbit transits, with locations during several months noted.

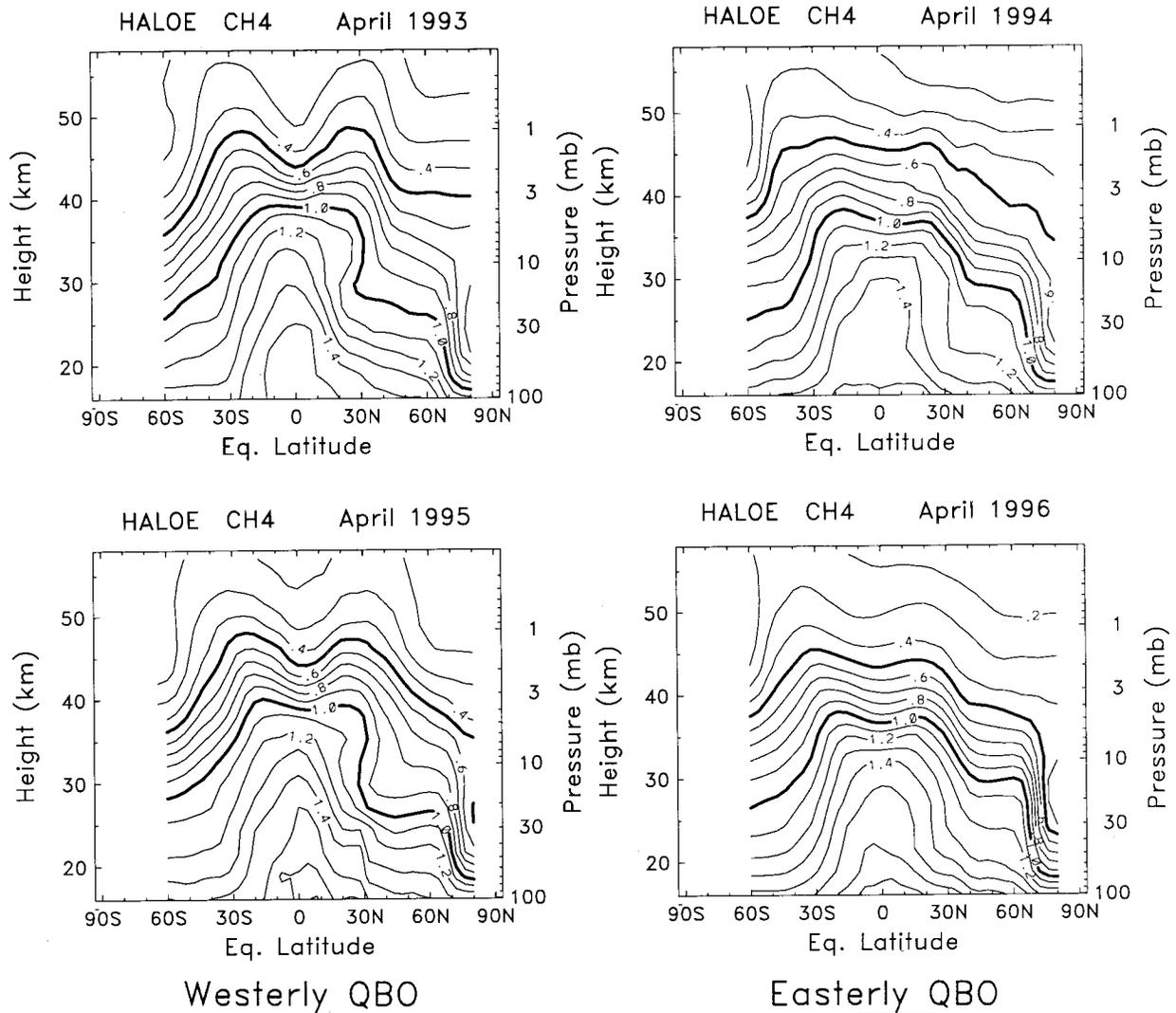


FIG. 24. Meridional cross sections of  $\text{CH}_4$  during April 1993, 1994, 1995, and 1996 (time periods denoted in Fig. 23). Note the double-peaked upper stratosphere patterns in 1993 and 1995.

though the anomalies in early 1994 are more nearly in phase. The similarity in these patterns suggests that advection by the mean meridional circulation is one important component for forcing the observed subtropical QBO constituent variations. Model calculations furthermore suggest that eddy transport effects are important for modulation of the subtropical mixing barrier (Polvani et al. 1995; O'Sullivan and Chen 1996).

## 5. Summary and discussion

High quality observations of stratospheric  $\text{CH}_4$  and  $\text{H}_2\text{O}$  from HALOE provide a near-continuous global record of seasonal and interannual variations in stratospheric transport. Motivated by the observed strong correlation between these tracers and derived potential vorticity (PV) fields, we have mapped the HALOE data into equivalent latitude

(PV) coordinates. This has the dual advantages of separating physically distinct measurements and increasing the effective latitude sampling of the data (see Fig. 2). Seasonal cycles are then constructed by harmonic regression analysis of monthly binned data. To obtain complete global seasonal cycles, we have included CLAES  $\text{CH}_4$  and MLS  $\text{H}_2\text{O}$  data over polar winter regions (with smooth transitions obtained by matching the data over midlatitudes). These global seasonal cycle data allow novel study of details in the seasonal cycle of stratospheric transport. These data will also be useful for initializing or studying transport characteristics in stratospheric models.<sup>1</sup>

<sup>1</sup> These seasonal cycle datasets (and similar ones for other HALOE constituents) are available to potential users via anonymous FTP from NCAR (contact the author at randel@ucar.edu).

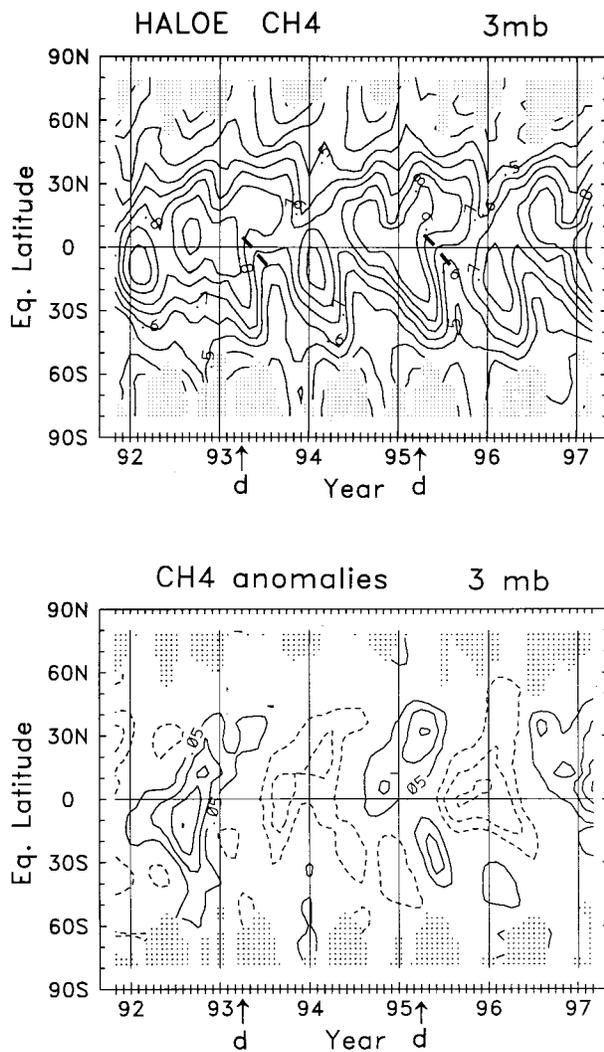


FIG. 25. Latitude–time sections of  $\text{CH}_4$  at 3 mb during 1991–97. Shown are the full field (top) and the detrended interannual anomalies (bottom). Dashed lines and arrows denote periods of double-peaked (d) latitudinal structure.

The seasonal cycle of  $\text{CH}_4$  in the lower stratosphere shows characteristics that have similar space–time structure to vertical velocity fields derived from meteorological analyses. This is a reasonable finding, as  $\text{CH}_4$  is strongly vertically stratified. The enhanced subtropical gradients observed in  $\text{CH}_4$  coincide with the region of weak vertical motions, that is, where the tropical upwelling changes to middle–high-latitude downward motion. Furthermore, a seasonal latitudinal shifting of both the tropical  $\text{CH}_4$  and vertical velocity fields is observed, with isolines displaced slightly into the respective summer hemispheres. This behavior suggests that tropical upwelling plays a large role in formation of the subtropical gradients for tropospheric source gases; winter hemisphere wave activity is also likely to be important (Polvani et al. 1995; Waugh 1996).

Polar regions in the lower stratosphere of both hemi-

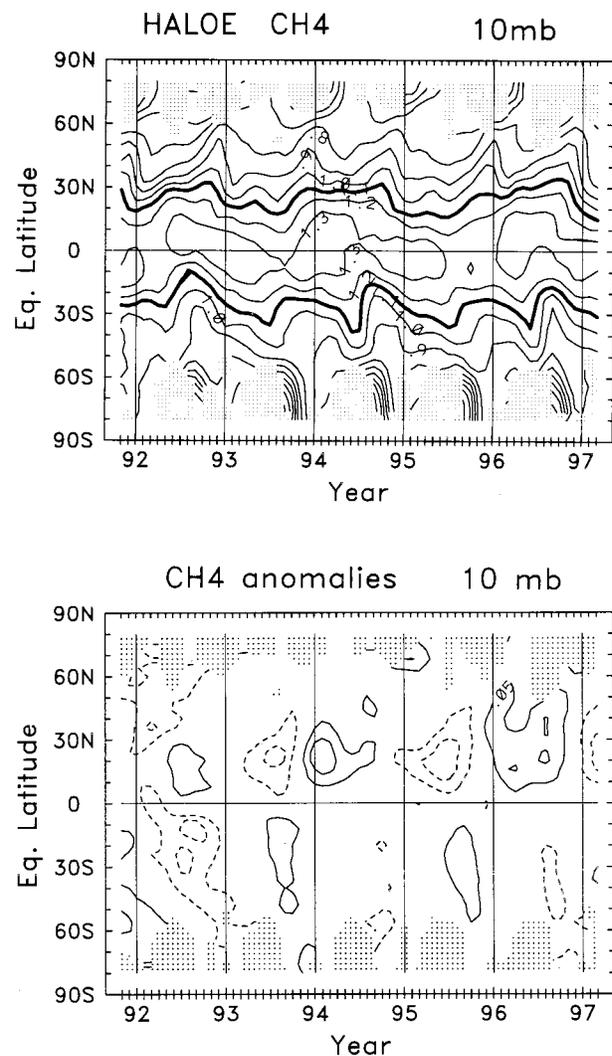


FIG. 26. Latitude–time sections of  $\text{CH}_4$  at 10 mb over 1991–97, for the full field (top—contour interval 0.1 ppmv) and interannual anomalies (bottom—contour interval of 0.05 ppmv, with zero contours omitted).

spheres show very similar seasonality in  $\text{CH}_4$ , with very low values during spring (March–April in the NH and October–November in the SH). Such low values in the SH vortex have been discussed by Russell et al. (1993a) and Schoeberl et al. (1995) and provide evidence of relatively unmixed descent from high altitudes. The similar low values observed in the NH data here are due to having organized the observations according to the polar vortex structure (i.e., zonal means do not show such low values). Good agreement with vertical transport calculations (Fig. 12) confirms isolation of the SH polar vortex and demonstrates that the vertical velocities (derived primarily from radiative heating codes) are reasonably correct. Poor agreement in the NH (Fig. 13) suggests less isolation of the NH polar vortex.

The harmonic seasonal cycle fits of the HALOE water

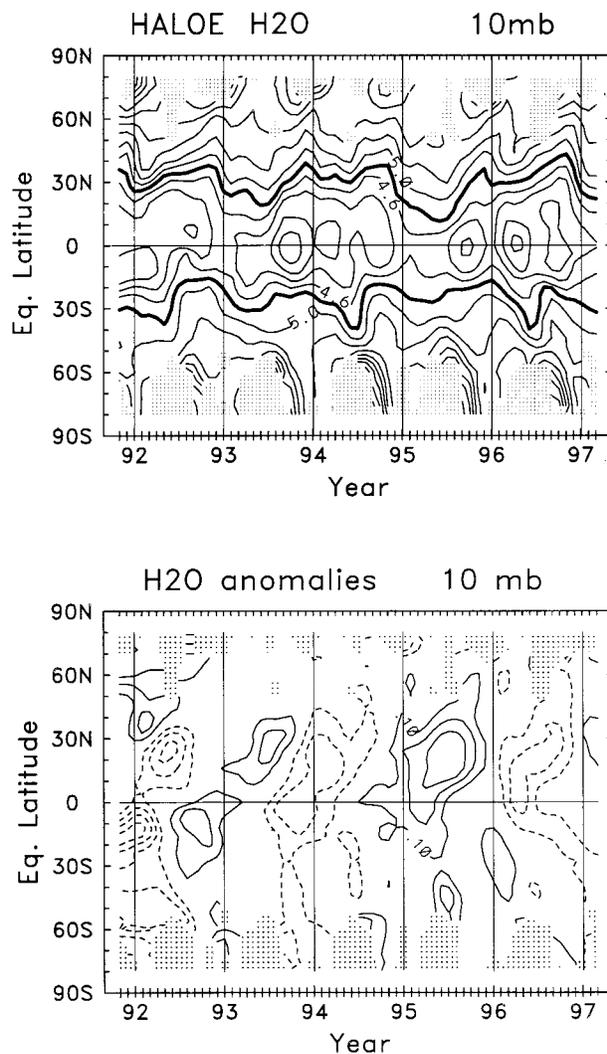


FIG. 27. Latitude–time sections of  $\text{H}_2\text{O}$  at 10 mb over 1991–97, for the full field (top—contour interval 0.2 ppmv) and interannual anomalies (bottom—contour interval of 0.1 ppmv, with zero contours omitted).

vapor data provide a clear view of the upward-propagating annual cycle in the Tropics (Figs. 15–17). The annual cycle is dominant over 100–22 mb (up to  $\sim 27$  km); above this altitude the seasonal variation is primarily semiannual. The lack of propagation of the annual cycle above 27 km suggests there is somewhat stronger horizontal mixing above this altitude, with less isolation of tropical air (i.e., this is the upper level of the tropical “reservoir”). This is consistent with enhanced tropical mixing above 10 mb noted by Dunkerton and O’Sullivan (1996). Two mechanisms for enhanced mixing with a semiannual timescale are 1) mean circulation advection, and 2) the tropical extension of winter hemisphere planetary wave effects.

The annual cycle in water vapor at and above 68 mb is centered over the equator, whereas seasonality at 100

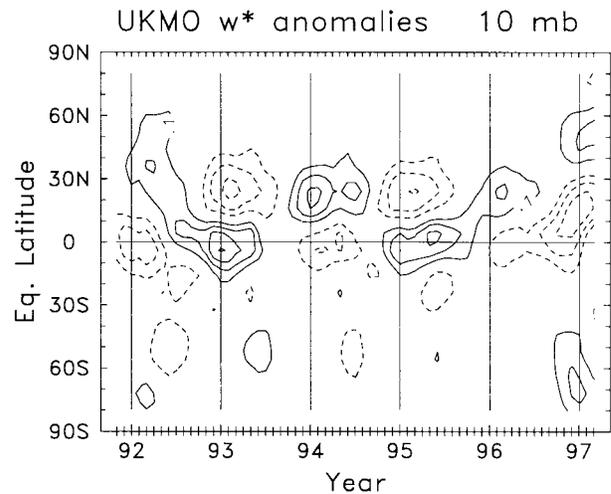


FIG. 28. Latitude–time section of interannual anomalies in TEM vertical velocity ( $\overline{w^*}$ ) at 10 mb. These data have been filtered to isolate components coherent with the zonal wind QBO, and multiplied by  $\cos(\text{latitude})$  to accentuate tropical features. Contour interval is  $0.1 \text{ km month}^{-1}$ , with zero contours omitted.

mb shows relative maxima in the NH subtropics. Regions of high  $\text{H}_2\text{O}$  at 100 mb during NH summer are particularly shifted toward the NH, and there is evidence of strong transport into NH middle and high latitudes (Fig. 16). The asymmetry of these patterns (both in location of the tropical extrema and the latitudinal propagation) contributes to a significantly wetter lower stratosphere in the NH compared to the SH (Kelly et al. 1990; RO97). There is little meridional spreading of the tropical annual cycle above 68 mb beyond  $30^\circ\text{N-S}$ ; the slow weakening of the tropical annual cycle with altitude over 68–22 mb is further evidence of tropical isolation, and these data infer an exchange time of order 15 months for mixing of midlatitude air into the Tropics. These data also demonstrate that there is little influence of Antarctic dehydration in SH midlatitudes following vortex breakup (as also noted from the analyses of RO97).

The data here show strong seasonality in the tropical upper stratosphere, for both  $\text{CH}_4$  and  $\text{H}_2\text{O}$ , with the characteristics of a semiannual cycle forced by alternating upwelling in the respective summer subtropics. This semiannual signal in  $\text{H}_2\text{O}$  was also discussed by McCormick et al. (1993) and Eluszkiewicz et al. (1996). A double-peaked latitudinal structure is evident in the climatology during April–June, but this is strongly modulated by the phase of the QBO.

A global budget analysis of the seasonal cycle  $\text{CH}_4$  data using UKMO residual mean circulation estimates and parameterized photochemical loss rates showed large residuals, which we attribute (at least in part) to eddy transport effects. The patterns of these eddy transport tendencies show a north–south dipole pattern, with positive tendencies in the winter stratosphere and negative tendencies in low latitudes (extending across the

equator). These eddy transports, which are derived as a residual here, are very similar to the explicit eddy transport calculations performed using general circulation model output in Randel et al. (1994). Other studies have also suggested that the effects of planetary waves reach low latitudes (Polvani et al. 1995; Waugh et al. 1996; O'Sullivan 1997). There is a clear semiannual variation of the eddy transports (residuals) in the Tropics, and this is consistent with the amplification of large-scale wave transports in the respective winter hemispheres.

A number of model and diagnostic studies have been used in an attempt to explain the double-peaked constituent patterns in the tropical upper stratosphere (e.g., the April patterns in Figs. 6 and 7) (Gray and Pyle 1986, 1987; Hamilton and Mahlman 1988; Choi and Holton 1991; Sassi et al. 1993; Kennaugh et al. 1997). The mechanism for the double peak in these studies is downward motion over the equator (associated with the westerly shear zone of the dynamical SAO during equinox) coupled with upward motion in subtropics. A common feature of these studies, however, is an underestimation of the strength of the double peak, which is usually attributed to a lack of resolved downward motion over the equator. However, the balance of terms in the budget analyses here suggests that the vertical mean circulation does not primarily determine the tropical behavior. Transport by the  $\bar{v}^*$  circulation contributes large positive SAO tendencies in the tropical upper stratosphere during both solstices, due to strong flow toward the winter hemisphere and enhanced meridional constituent gradients near the equator. Chemical relaxation is a relatively large budget component, but exhibits little seasonality in the Tropics. There are also relatively large residuals in the tropical budget during solstice seasons, and these may be at least partly attributable to winter hemisphere eddy transport effects that reach the Tropics.

A majority of the interannual variance over the 1991–97 time sample analyzed here is found to be coherent with the QBO; aspects of QBO variations in HALOE  $\text{CH}_4$  have also been shown in Luo et al. (1997) and Ruth et al. (1997). The structure of the QBO signal in  $\text{CH}_4$  exhibits a maximum in the tropical upper stratosphere over 35–45 km. This is somewhat higher than the QBO signals observed in ozone (~20–38 km) and nitrogen dioxide (~28–35 km) (Zawodny and McCormick 1991). These differing vertical structures are attributable to differences in the respective background means (e.g., Chipperfield and Gray 1992); the strongest background gradients in  $\text{CH}_4$  are observed over 35–45 km.

We find that the QBO exerts a strong influence on appearance of the double-peaked “rabbit ears” structure in the tropical upper stratosphere during NH spring: the double peak occurs primarily when QBO westerlies are present near 10 mb. The double-peaked structure in the mean seasonal cycle (Figs. 6 and 7) represents a time average over both phases of the QBO. This has implications for numerical simulation of the stratopause SAO and its effects on tracer distributions (e.g., Sassi et al.

1993), suggesting that details of the winds in the lower stratosphere may be important. Kennaugh et al. (1997) have recently presented a modeling study of this QBO influence on the double peak.

The observed equatorial  $\text{CH}_4$  (and  $\text{H}_2\text{O}$ ) QBO anomalies over 35–45 km are observed to be strongly correlated with anomalies in the residual mean vertical circulation  $\bar{w}^*$  (Fig. 20b). This is a curious result in light of the anticipated dominance of vertical transport in this region, as the continuity equation [Eq. (1)] suggests a correlation between  $\bar{w}^*$  and  $\partial\chi/\partial t$  (not  $\bar{\chi}$ ). Budget calculations show a large residual with a QBO periodicity, and one possible explanation is that eddy transports in the tropical upper stratosphere are modulated by QBO winds (with maximum negative tendencies during QBO westerly winds). This is not unreasonable in light of observational (Dunkerton and Baldwin 1991; Baldwin and Dunkerton 1991) and modeling (O'Sullivan and Chen 1996) studies showing QBO modulation of planetary wave effects in both subtropics and midlatitudes.

These constituent data also show evidence of a latitudinal shifting of the tropical maximum region and modulation of subtropical gradients in the middle stratosphere (~27–32 km) associated with the QBO. There is a shift of approximately  $10^\circ$ – $15^\circ$  lat correlated with the QBO winds near 30 mb, with movement toward the NH during QBO westerlies. The sharpening of the NH subtropical gradients during QBO easterlies is similar to the patterns observed in stratospheric aerosol by Hitchman et al. (1994) and Grant et al. (1996). Examination of QBO anomalies in  $\bar{w}^*$  at 10 mb shows relatively large subtropical maxima that are approximately in quadrature with the constituent anomalies, suggesting that advection by the mean circulation may be important [presumably in addition to eddy transports, e.g., Polvani et al. (1995) and O'Sullivan and Chen (1996)]. One caveat to interpretation of the QBO results here is that there is a certain phasing of the QBO and seasonal cycle for this relatively short record (~ two QBO cycles); details of the QBO in constituent transport may evolve as this phase relationship changes slowly in time. Overall there is reasonable agreement between the space–time patterns of QBO variations in  $\text{CH}_4$  and  $\text{H}_2\text{O}$  throughout the stratosphere. One region of apparent difference is over the equator near 10 mb, where anomalies are seen in  $\text{H}_2\text{O}$  but not  $\text{CH}_4$ . Details of these differences await explanation.

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