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2	Technical Note:
3	Multi-year Changes in the Brewer-Dobson Circulation from HALOE Methane
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11	Abstract. This study makes use of Halogen Occultation Experiment (HALOE) methane (CH ₄) in
12	a search for multi-year changes in the Brewer-Dobson Circulation (BDC). Changes in CH4 are
13	determined for three, successive 5-yr time spans from 1992 to 2005, and there are significant
14	differences in them. There is a clear separation for the changes in the northern hemisphere near
15	30 hPa or at the transition of the shallow and deep branches of the BDC. The CH4 changes are
16	positive and large in the shallow branch following the eruption of Pinatubo, but they then
17	decrease and agree with tropospheric trends in the late 1990s and early 2000s. CH ₄ decreases in
18	the upper part of the deep branch from 1992 to 1997 or following the eruption of Pinatubo. CH_4
19	continues to decrease in the deep branch in the late 1990s but then increases in the early 2000s,
20	although those changes are small compared with the seasonal and interannual variations of CH4.
21	Multi-year changes are due, in part, to wave forcings during El Nino Southern Oscillation
22	(ENSO) of 1997-1998 and beyond and to episodic, sudden stratospheric warming (SSW) events
23	during both time spans. It is concluded that time series of HALOE CH4 provide effective tracer
24	diagnostics for studies of the nature of the BDC from 1992 to 2005.
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28 **1. Introduction**

29 Global-scale stratospheric transport is characterized in each hemisphere by a seasonal Brewer-30 Dobson circulation (BDC), consisting of upward transport in the tropics, poleward transport to higher latitudes, and descent in the polar vortex region (e.g., Butchart, 2014). Model studies 31 indicate that there are also multi-year changes in the BDC in response to increases in the 32 greenhouse gases (GHG) and to dynamical forcings during El Nino/Southern Oscillation 33 (ENSO) events, but where the effects of those forcings may differ within the shallow (lower) and 34 deep (upper) regions of the BDC. Remsberg (2015) reported that the distributions of methane 35 36 (CH₄) from the Halogen Occultation Experiment (HALOE) provide tracer diagnostics for 37 changes in the BDC. The present study is a refinement of his initial analysis and gives some insight on mechanisms for changes in the BDC. Section 2 is a brief description of the methane 38 39 data and the analysis approach for them. Section 3 presents the results of the analyses in terms of changes in the distribution of CH_4 for three successive 5-yr time spans. Qualitative 40 attributions are also considered for those changes. Section 4 summarizes the findings from this 41 exploratory study. 42

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2. Data and Analysis Method

HALOE obtained sunrise (SR) and sunset (SS) occultation measurements across latitude zones 45 throughout its mission of October 1991 to November 2005. The present study considers zonal 46 averages of CH₄ for nine latitude zones and at twelve pressure levels (0.4 to 50 hPa), for a total 47 of 108 separate time series. A minimum of 5 profiles gives representative zonal averages for 48 each latitude zone; averages are based on many more profiles in most instances. Figure 1 shows 49 example time series from zonal averages of the SR and SS measurements at specific pressure 50 levels and in three different latitude zones. Figure 1(a) is the time series for the 10 hPa level at 51 30°N latitude, and there is a clear quasi-biennial oscillation (QBO) signal in the data. Figure 52 1(b) is for 10 hPa at 30°S, where there is a combination of annual (AO), semi-annual (SAO), and 53 OBO signals. One can also see that seasonal and interannual variations are much larger than the 54

longer-term changes. Figure 1(c) is for 2 hPa at 45°N, where CH₄ decreases gradually in the
early to middle 1990s but where it also has large amplitudes in early 2002 and 2004.

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58 The analysis of CH₄ for this study is in the manner of Remsberg (2015) with the following modifications. The nine latitude zones are from 60°S to 60°N with a spacing of 15° and no 59 overlap. The latitude bins are a bit narrower than before (15° versus 20°) but still provide 60 representative sampling, even at $\pm 45^{\circ}$ latitude from 2000 to 2005 when the samples from 61 HALOE are limited. To look for secular trends in CH₄, multiple linear regression (MLR) 62 analysis was applied to the CH₄ time series, as separated into three, 5-yr time spans that overlap 63 64 by one year (July 1992 to June 1997; July 1996 to June 2001; and July 2000 to June 2005). The beginning and end months of July and June, respectively, were selected to avoid large excursions 65 in CH₄ at the end points of time series for the northern hemisphere during the dynamically active 66 67 winter season. Data prior to July 1992 were not used, to avoid issues related to variable solar lock-down procedures for the HALOE sun sensor and because of significant extinction from 68 interfering aerosols following the Pinatubo eruption of June 1991. The analyses also do not 69 include the period after June 2005, when HALOE operations were limited. 70

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An initial MLR analysis was applied to the 13-yr time span of the HALOE measurements for a 72 73 range of pressures and latitudes but using only AO and SAO terms. Time series residuals from those runs were analyzed for interannual cycles, yielding significant terms with periods of 882 74 75 days (~29-month or QBO-like) and of 690 days (22.6-month or sub-biennial (SB)). Those two 76 terms were highly significant for many of the latitude/pressure time series, so they were included 77 along with the seasonal terms for the MLR model. The 5-yr (or 60 month) time span is equivalent to two complete QBO cycles and avoids biases in the MLR trends due to that periodic 78 79 term. A biennial (718-dy) term was also indicated for the subtropics, but it was not uniformly present elsewhere and was not retained for the model. A linear term completes the final MLR 80 81 model; the analyses also correct for lag-1 autoregressive (AR1) effects. The MLR model fit to 82 the data points is shown by the oscillating solid curve for July 1996 to June 2001 in each panel of Fig. 1, and the combination of the constant and linear terms is the dashed line. One can see that 83 the seasonal and interannual variations have large amplitudes compared with the overall 5-yr 84

trend line, such that even minor changes from year to year can affect the linear changes.

Although the MLR fits and trends are based on analyzed AR1 values for each case, the MLR

curves in Fig. 1 are based on AR1 = 0 and give maximum amplitudes for the periodic terms.

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The sensitivity of the trend coefficient to the approximate QBO term of the MLR fit was 89 determined for Fig. 1(a) (30°N, 10 hPa), where a QBO cycle shows clearly. Specifically, the 90 91 length of the QBO cycle was altered (28 months versus 29.5 months) as well as the length of the 92 time span for the MLR analysis (58 months rather than 60 months). The resulting trend coefficients in each case differ by less than 6% from the one of Fig. 1(a). Figure 1(c) focuses on 93 94 the upper stratosphere, where CH₄ decreases from 1992 to 1997 or from one year after the 95 Pinatubo eruption. The 5-yr trend is less negative from 1996 to 2001 and then is positive from 2000 to 2005, punctuated by two winter maximums in early 2002 and 2004. 96

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The distribution of the average CH₄ (its constant term) is shown in Figure 2 for the time span of 98 99 July 1996 to June 2001. Tropical entry-level values extend upward and are transported poleward in each hemisphere. CH₄ decreases with altitude and latitude, due to the relatively slow chemical 100 101 conversion of CH_4 to water vapor (H₂O) and molecular hydrogen (H₂) in the upper stratosphere (Brasseur and Solomon, 2005). That decay of CH₄ is nearly symmetric between the two 102 hemispheres. The primary purpose of Fig. 2 is to show the vertical and meridional gradients of 103 CH₄ that are acted upon by the BDC, generically considered as hemispheric, net circulations 104 105 composed of tropical ascent, poleward transport, plus descent in the polar vortex region. 106 Although the CH₄ distributions for the other two 5-yr time spans are like that of Fig. 2, there are 107 small but distinct differences in the 5-yr changes in CH₄ for the three successive time spans.

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Distributions of the linear terms (% change / 5-yr) from the zonally averaged CH₄ data are shown and discussed in Section 3 for each of the three periods of July 1992 to June 1997, July 1996 to June 2001, and July 2000 to June 2005. Notably, there is good continuity for the trends with pressure and latitude, indicating that each distribution is meaningful and related physically to multi-year changes for the large-scale BDC. Mechanisms giving rise to the changing CH₄ are related to external (volcanic) and/or wave forcings followed by radiative and/or chemical
relaxations therefrom. The changes in CH₄ are also compared with estimates of the stratospheric
net circulation that have been diagnosed and reported by other researchers.

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3. Multi-year changes in CH₄

119 *(a) July 1992 to June 1997*

Figure 3 shows that CH₄ decreased in the upper stratosphere and lower mesosphere from July 120 1992 to June 1997 or from one year after the Pinatubo eruption of June 1991. The shading 121 122 indicates where the trends are robust, the dark shading having a confidence interval (CI) of greater than 90% and the light shading having CI between 70 and 90%. Note that there are 123 small, positive trends in CH₄ within the lower stratosphere, due to its tropospheric trends of ~0.4 124 % / yr (or 2.0 % for this 5-yr period) (Dlugokencky et al., 2009). Changes of the CH₄ 125 126 distributions across the 5-yr time span represent where there were accelerations of the BDC (positive changes of greater than the tropospheric trends of ~ 2.0 %) or decelerations of the BDC 127 (changes of less than ~ 2.0 %). 128

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Negative changes in CH₄ in the upper regions of Fig. 3 imply that there was an overall 130 weakening of the deep branch (above the ~20-hPa level) of the stratospheric BDC during this 5-131 yr period. Those negative changes are more pronounced at middle latitudes of the northern than 132 of the southern hemisphere, indicating that there was ascent of CH₄ within the deep branch of the 133 134 BDC in the northern subtropics due to external forcings from the Pinatubo eruption near 15°N followed by a relaxation toward lower values thereafter. In fact, separate, zonal mean cross 135 sections of HALOE CH₄ (not shown) reveal that the 0.8 ppmv contour of CH₄ occurred at ~4 136 hPa in November 1991 but had risen to ~2 hPa by February 1992, most likely a response of the 137 BDC to winter wave forcings (e.g., Russell et al. 1999). Thereafter, the CH₄ values that had 138 been lofted to higher altitudes underwent a gradual decline over time. Sudden stratospheric 139 warming (SSW) events also tend to accelerate the deep branch of the BDC and mix middle 140 latitude and polar air; that mixing flattens the contours of zonal average CH₄ mixing ratio. 141

However, there were no SSW events in the northern hemisphere during 1992 to 1997 (Choi etal., 2019).

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A more traditional indicator of changes in the BDC is stratospheric age-of-air (AoA), where 145 negative AoA indicates acceleration and positive AoA implies a deceleration of the BDC. 146 Pitari et al. (2016) estimated that AoA decreased in the middle to upper stratosphere by ~0.5 to 147 148 0.7 yr during 1991-1992, due mainly to ascent following the eruption of Pinatubo. Fig. 3 indicates a decline of CH₄ (and presumably an increase in AoA) from July 1992 onward. 149 Methane is not a perfect tracer, however, as it has a chemical lifetime as short as only a few 150 151 months at 45 km (~1.5 hPa) and then lengthening to 6 months and longer at 55 km and above 152 and at 40 km and below (Brasseur and Solomon, 2005). The relatively short lifetime of CH₄ at 1.5 hPa means that even the seasonal variations of CH_4 are dampened at that level. The near-153 zero changes for CH₄ near 15°S and 2 hPa in Fig. 3 may imply that there was still some transport 154 of CH₄ to that region from the tropics after July 1992. 155

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The 5-yr changes in Fig. 3 also indicate that there was an accumulation of CH₄ at ~20 to 30 hPa at middle latitudes of both hemispheres during this period, in reasonable accord with a net poleward transport of tropical CH₄ at the top of the shallow (below the ~20-hPa level) branch of the BDC. The tropical trend of 3 to 4 % at 20 to 30 hPa is half that at middle latitudes (8 %), although it is still larger than the tropospheric trends for CH₄ of ~2.0 % for this 5-yr period.

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Figure 4 gives more detail about the effects of the Pinatubo eruption on CH₄ in the lower 163 stratosphere. Fig. 4(a) is for 15°N, 50 hPa and shows an initial increase in CH₄ in 1991 to the 164 middle of 1992, followed by decreasing values through 1993. HALOE CH₄ values are of the 165 order of 1.55 ppmv in 1992, declining to 1.45 ppmv in 1993, and then increasing again. 166 Independent CH₄ measurements at ground level are between 1.70 and 1.75 ppmv (Dlugokencky 167 et al., 2009). As an aside, HALOE CH₄ values for SR in Fig. 4(a) are consistently larger than for 168 169 SS. Those differences are likely due to uncorrected detector hysteresis effects for tropical SR measurements just above cloud tops; they decrease at 30 hPa and are negligible at 20 hPa. Diallo 170

et al. (2017) reported that AoA decreased during the first six months following the eruption of

172 Pinatubo due to tropical upwelling. Then, AoA increased from early 1992 to spring 1993

between 20°S and 30°N and from 20 to 27 km (~50 hPa to 15 hPa), implying a deceleration of

the shallow branch of the BDC during that time. The HALOE SR and SS CH₄ variations are in

accord with the changes in AoA from 1991 to 1993 in the shallow branch of the BDC.

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Figure 4(b) is the HALOE CH₄ time series for 45°N, 30 hPa, and it shows a gradual increase of CH₄ for 1993 to 1997. Yet, Diallo et al. (2017) reported increases in AoA for 1993 at tropical and middle latitudes due to meridional mixing, followed by decreases in mixing and AoA through 1997. Fig. 3 suggests that there was an accumulation of CH₄ at middle latitudes between ~20 and 30 hPa, due in part to that mixing trend. It may also be that there was an overall slowdown in the BDC during this 5-yr period, which was absent of SSW events and any enhanced descent of CH₄-poor, polar air plus its subsequent mixing to middle latitudes.

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185 *(b) July 1996 to June 2001*

Figure 5 shows the 5-yr CH₄ changes for 1996 to 2001, when there were several SSW events— 186 187 on 15 December 1998, 25 February 1999, and 20 March 2000 (Choi et al., 2019). The negative trends in the upper stratosphere are smaller in the northern hemisphere and larger in the southern 188 hemisphere than in Fig. 3, suggesting that there was tropical ascent but also increased mixing of 189 CH₄ to higher latitudes, related in part to SSW activity. Those changes are also where the 190 191 chemical loss of CH₄ to H₂O and H₂ may be a factor. It is apparent that there was greater meridional transport of CH₄ from the tropics to middle latitudes and an accumulation of CH₄ at 192 \sim 10 hPa in both hemispheres during 1996 to 2001. Those positive trends are at a level of the 193 stratosphere where the conversion of CH₄ to H₂O and H₂ is not as effective. 194

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196 There was a major warm ENSO event in 1997-1998 that altered wave forcing effects on CH₄ and

197 for the BDC. Randel et al. (2009) and Calvo et al. (2010) reported enhanced upwelling in the

tropics and an acceleration of the BDC at that time. Diallo et al. (2019) reported that ENSO

leads to the overall strengthening of the shallow branch of the BDC in the extratropics. It maybe that enhanced poleward transport in the shallow branch is why the CH₄ changes are more

200 of that enhanced pereviate transport in the shanow orange is why the Orig enanges are more

nearly zero in the tropics and agree closely with tropospheric trends that were smaller after 1995 (or $\sim 1.0 \% / 5$ -yr) (Dlugokencky et al., 2009). There is a clear separation at ~ 30 hPa in the sign

of the changes in the shallow versus the deep branch of the BDC in the northern hemisphere.

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205 The 1997-1998 warm ENSO event occurred near solar minimum, for which Calvo and Marsh (2011) also found enhanced wave forcing in the middle and upper stratosphere. That activity 206 leads to acceleration of the BDC and poleward transport of CH₄ to the extratropics. Barriopedro 207 208 and Calvo (2014) also found connections between ENSO and SSW events, although the exact 209 effects depend on the relative sequence of those events. Since major SSWs within 1996-2001 occur in December 1998, February 1999 and in March 2000, it is likely that they merely led to 210 further accelerations of the BDC. As an example, Tao et al. (2015) gave details about how the 211 SSW of 2009 led to an acceleration of the BDC. Their analyses may support the present finding 212 of increases in CH₄ in the extratropics near 10 hPa in Fig. 5. However, more focused studies of 213 the relative roles of SSWs and ENSO on the results of Fig. 5 are beyond the scope of the present 214 exploratory study. 215

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217 *(c) July 2000 to June 2005*

There was even more SSW activity in the northern hemisphere during the 5-yr span from 2000 to 218 2005 (on 11 February 2001, 2 January 2002, 18 January 2003, and 7 January 2004, according to 219 Choi et al., 2019). The distribution of changes in CH_4 in Figure 6 includes the net effect of those 220 episodic SSW events. There was an increase in CH₄ at upper altitudes, where the effect of SSWs 221 222 may have also led to greater poleward transport of CH₄ to higher latitudes. As before, an SSW event accelerates the deep branch of the BDC, bringing more CH₄ to high altitudes and greater 223 meridional transport to higher latitudes. At the stratopause (~1 hPa) and in the lower mesosphere 224 even small changes in CH₄ mixing ratio translate to relatively large percentage changes. Those 225 changes are from negative to positive from Fig. 5 to Fig. 6 and are rather uniform across latitude. 226 On the other hand, the changes near 10 hPa and at middle latitudes of the northern hemisphere 227

are weaker now than in Fig. 5. Fig. 1(a) indicates that this change may be a consequence, in part,
of large seasonal amplitudes for CH₄ in early 2001 and in 2005 or near the end points of the 5-yr
period from July 2000 to June 2005.

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In the southern hemisphere there was an anomalous SSW event on 22 September 2002, leading to a splitting of the polar vortex (Newman and Nash, 2005). The CH₄ changes from Fig. 5 to Fig. 6 at 10 hPa and 30°S were likely altered by that event (c.f., the time series segments in Fig. 1(b) for those two 5-yr periods). Note that there is no clear separation of the shallow and deep branches of the BDC for the southern hemisphere in Fig. 6.

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238 Figure 7 provides a clearer picture of what occurred from 2000 to 2005. Fig. 7(a) is a time series of CH₄ at 45°S and 20 hPa, and it shows pronounced annual cycles in CH₄. A peak seasonal 239 value occurs in 2001, and it may by influencing the overall analyzed trend for that time span. On 240 the other hand, there is little indication of a change in CH₄ at the time of the anomalous SSW 241 event of September 2002. Fig. 7(b) shows the corresponding CH₄ time series at the Equator and 242 20 hPa, where CH₄ variations are forced primarily by the QBO. There is a clear decrease in CH₄ 243 in 2001 compared to the maximum at 45°S in Fig. 7(a). Fig. 7(b) also shows that tropical QBO 244 signals are nearly absent in CH₄ from 1996 to 2000. Bönisch et al. (2011) reported that tropical 245 upwelling increased after 2000 and accelerated the shallow branch of the BDC. Similar studies 246 based on variations in CH₄ may be helpful in determining the nature of the shallow layer of the 247 BDC both prior to and after 2000. 248

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4. Summary findings

The present study is an analysis of the distributions of HALOE CH₄ for indications of secular changes in the BDC. Linear trends in CH₄ were determined for three, successive 5-yr time spans, and there are significant differences between them. There is a clear separation of the deep and shallow branches of the BDC at about 30 hPa in the northern hemisphere in each time span. Although the changes for CH₄ in the shallow branch are rather large following the eruption of

Pinatubo, they agree well with tropospheric trends for CH₄ during the late 1990s and early 2000s. There are decreasing changes in the upper part of the deep branch of the BDC in the early to middle 1990s, indicating a decline of CH₄ from one year after the eruption. CH₄ changes in the middle and upper stratosphere differ markedly for the early 2000s compared to those of the late 1990s, although those differences are small compared to the seasonal and interannual variations of CH₄. In addition, the seasonal changes within the deep branches differ in each hemisphere, perhaps due to episodic SSW events and to wave forcings during ENSO.

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In terms of multi-year changes for the BDC, it appears that during the period of 1992 to 1997 264 265 there was acceleration of the shallow branch and deceleration of the deep branch. However, 266 those implied changes in the BDC may be anomalous because of the large perturbation to the CH₄ distribution in 1991 from the Pinatubo eruption. During 1996 to 2001 the changes in the 267 shallow branch were nearer to zero, while decreasing trends persisted in the deep branch. Yet, it 268 269 also appears that there was acceleration of the poleward transport and mixing at middle latitudes 270 within the layer from \sim 30 hPa to \sim 7 hPa during that 5-yr period. Then, there was a deceleration in the shallow branch and acceleration in the deep branch of the BDC during 2000 to 2005. The 271 implied BDC also differed markedly in the two hemispheres over that final 5-yr span. It is 272 concluded that time series of HALOE CH₄ provide effective tracer diagnostics for studies of the 273 secular nature of the BDC from 1992 to 2005. 274

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276 *Data availability*. The HALOE V19 profiles are at the NASA EARTHDATA site of EOSDIS,

and its website is <u>https://disc.gsfc.nasa.gov/datacollection/UARHA2FN_019.html</u> (Russell et al.,

278 1999).

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280 *Competing interests.* The author has declared that there are no competing interests.

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361 Figures

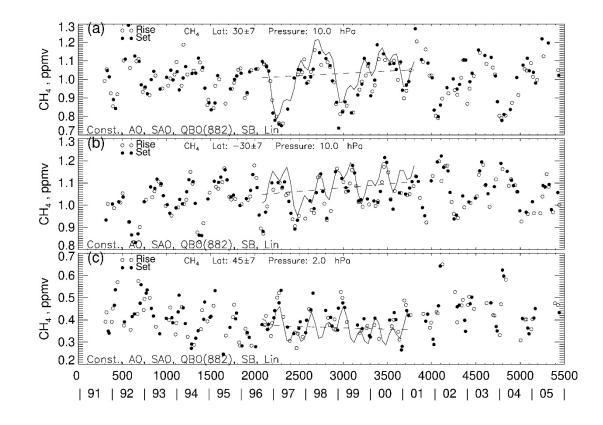
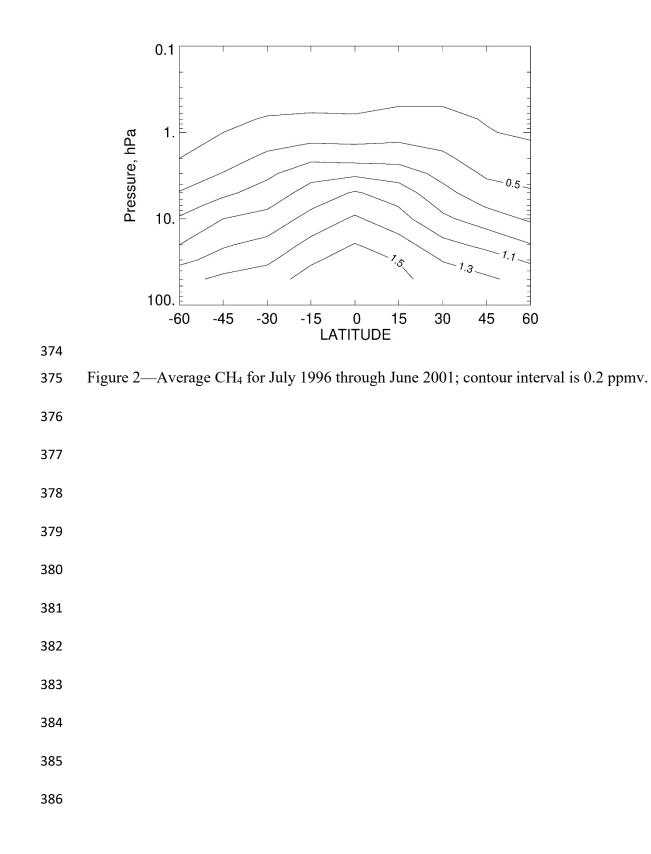


Figure 1—Time series of HALOE CH₄ (a) 30°N and 10 hPa, (b) 30°S and 10 hPa, and (c) 45°N and 2 hPa. MLR fit for July 1996 through June 2001 is the solid curve, and its linear trend is the dashed line. Day numbers on the abscissa are from 1 January 1991. Model terms are listed at bottom left. The Pinatubo eruption occurred in June 1991.



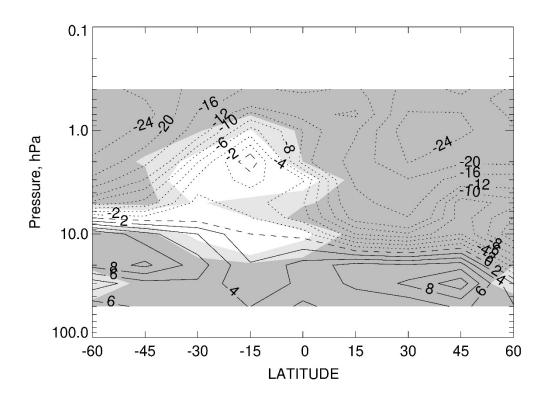
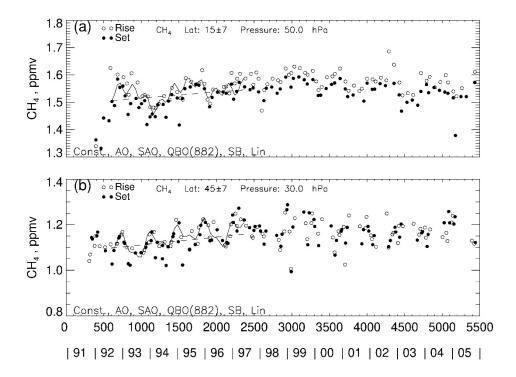
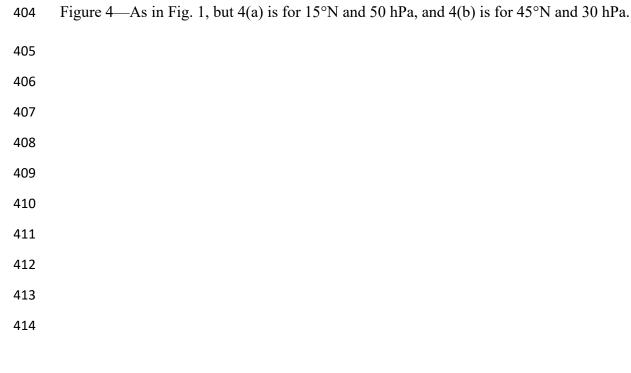
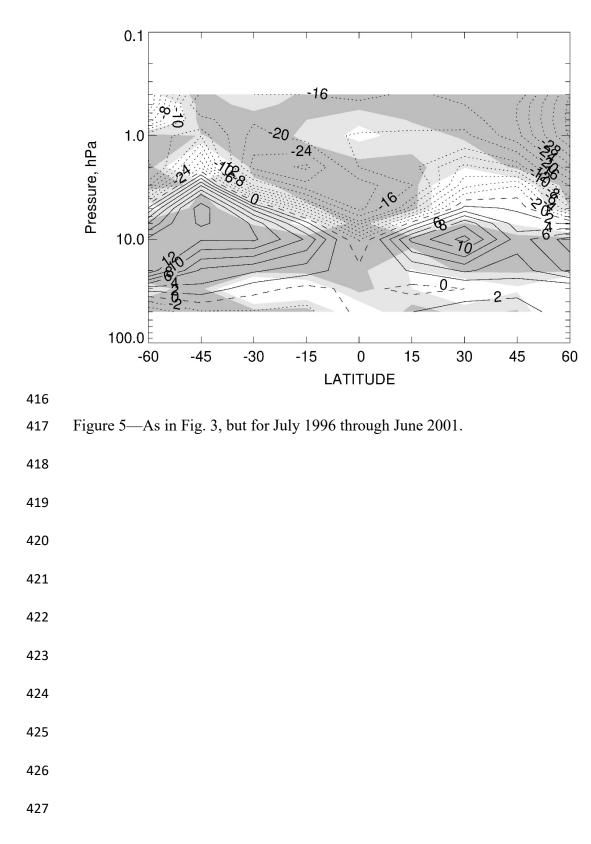


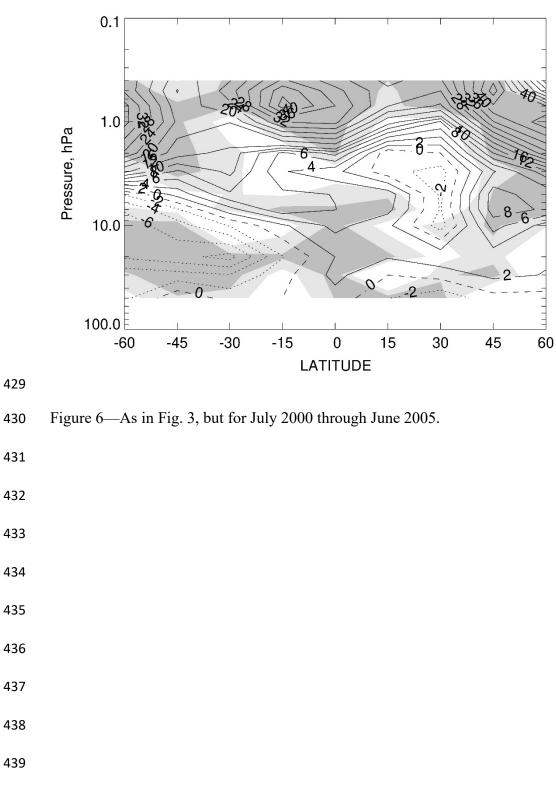
Figure 3—Changes in CH₄ for July 1992 through June 1997 (in % / 5-yr); positive changes are solid, negative changes are dotted, and zero is dashed. Contour interval is 2 % within ±12 % but 4 % outside that range. Dark shading shows where the confidence interval (CI) for the trends is greater than 90 %, and light shading shows where CI is between 70 and 90 %.

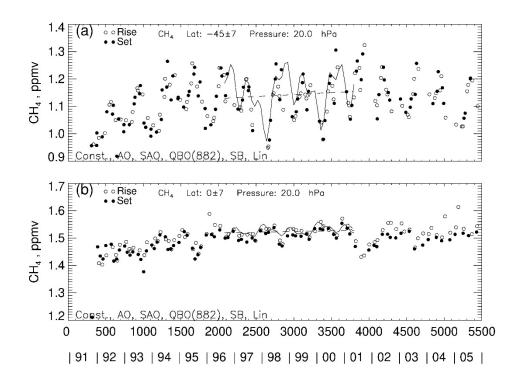












443 Figure 7—As in Fig. 1, but 7(a) is for 45°S and 20 hPa, and 7(b) is for Eq and 20 hPa.