Response to Reviewer #2

General:

I appreciate your careful review and your insight into the analyses that I have undertaken. Please refer to my response to Reviewer #1, regarding my substitute of the term Brewer-Dobson circulation for the net circulation and also of my agreement about the combined transport and loss terms for a chemical tracer such as methane. Tables A1 and A2 will replace original Tables 1 and 2, and Table A3 is new. I have also appended new Figure A1 and important revisions to a number of the other Figures, especially Figures 9 and 10.

Thank you for reminding me of the work of Rosenlof (2002), which is an important precursor to my study. Primary differences between our two studies are: (1) I consider HALOE data from 1992 to 2005, (2) I include interannual terms in my MLR model, (3) my binning of the data is different, and (4) I am supplying estimates of uncertainty for my analyzed trends. With regard to item (3), I will provide more details in the revised manuscript about how I analyzed the HALOE methane data. In particular, I do not combine the HALOE SS and SR profiles into narrow (5 degree wide) latitude bins and in terms of monthly zonal averages, as does Rosenlof (2002) and many other investigators. Instead, I generate separate bin averages of SS and of SR measurements and retain the exact times for those SS and SR bin-averaged points. This choice provides more frequent sampling and at exact latitude crossing times for a better determination of the dominant semi-annual term from the HALOE occultation measurement opportunities at the low latitudes. However, my approach does mean that the SS and SR points in the data series are not evenly spaced in time. It does not alter the MLR trend coefficients appreciably and only affects the analyzed errors of the coefficients of the interannual terms in a minor way. This choice also means that fewer profiles are available for the averages of each of the SS and SR points of my time series, such that I needed to widen my latitude bins to 10 degrees rather than the 5 degree averages of Rosenlof. I require that there be at least 5 profiles for each of my SS and SR average points. Because adjacent profile samples are separated by about 24 degrees of longitude and because the zonal variations of methane are periodic and relatively small compared to the true zonal mean value, the average of 5 profiles is representative of the mean. One exception is in the instance of disturbed (i.e., wave-1) polar winter vortex conditions, but that instance is normally present in the data time series for only several weeks to a month or so in each year. My analyzed linear trends are not changed significantly, when I require 7 rather than 5 profiles for my bin averages.

For the attached plot of methane trends in pressure-altitude versus latitude space (Figure A1) that I will include in my revised manuscript, I considered HALOE methane profiles within 14 overlapping, 20-degree wide bins centered at latitudes from 65S to 65N. In general, my analyzed trends (in %/decade) are larger than those of Rosenlof (her Figure 4, 2002). The rather large trends at middle latitudes are primarily because variations of methane with latitude are larger for a 20-degree wide bin than for the 5-degree wide bin of Rosenlof. Dark shading in Figure A1 is where the trend term is present with greater than 90% confidence from the MLR analysis model; lighter shading denotes where that confidence is between 70% and 90%. Where analyzed trends are highly significant (shaded regions), the reduction in bin width from 20 to 10 degrees yields analyzed trends do not differ by much in the tropics (10 degrees), at the high latitudes (60 degrees), and even at 35 degrees (see the corresponding values in the pairs of trend profiles in Table A3 that are marked by asterisks). This finding is partly because the meridional gradient of the zonally-averaged methane is small in those regions (see Table A1) and contributes very little to the analyzed trends. Figure A1 shows that the trends at 10 degrees are increasing within this so-called tropical pipe region; the trends are zero to decreasing in the high latitude region of 60 degrees. The analyzed trends are smaller in Table A3 at 35N, where the meridional gradients of methane are large and when I use a 10-degree wide versus a 20-degree wide bin. However, their respective trends still agree closely, where the term coefficients are significant (at 5 to 50 hPa). The trend profiles at the middle latitudes are positive, and they indicate a robust and consistent character for their net Equator-to-Pole or BDC circulations in each hemisphere, at least during the period of the HALOE measurements. Note also that the trends are asymmetric and greater in the northern than in the southern hemisphere in Figure A1 from 50 hPa to about 10 hPa.

Accordingly, I will replace Table 2 with attached Table A2 and Figures 6 and 7 with updated ones (also attached) for the revised manuscript. They show trends for the BDC based on the methane profiles at 10±5 degrees (tropical) and at 60±5 degrees (extratropical) latitudes for both the southern and northern hemispheres. Their 2σ errors are shown in those Figures for pressure levels, where the trend terms are highly significant. The trends in revised Figures 6 and 7 differ from those in the original manuscript because of the new, more narrowly-defined bin widths; the uncertainties for those trends are somewhat larger than before though, presumably because of the slightly fewer samples for the new results.

Specific:

P24187/L10: I agree and will change the wording.

P24188/L28ff: I should have said that there are clear vertical variations of CH_4 at the low latitudes that are related to the phase of the QBO.

P24189/L11ff: I agree that the width of latitude bins is an issue at middle latitudes of the upper stratosphere where CH_4 has large gradients. But how to incorporate information on average latitude for each data point of the time series is not straightforward to do. Instead, I have focused on examining how the analyzed trend terms are changed by altering the width of the latitude bins. Please refer to the figures and discussion in my response to General above. Variations with longitude are small, as can be seen in the plot images at the HALOE Website (click on Browse Images on the left menu, followed by selecting 'longitude versus pressure cross section' for SS or SR CH_4 for a month and year; click on a day to see the image).

P24189/L14ff: As is customary, semiannual and annual terms are part of the MLR model. A Fourier analysis was applied to the residual or difference of the MLR model from the data time series for a given latitude and pressure altitude, in order to identify significant periodic, interannual terms that remained. For the revision manuscript I have more carefully determined that up to three interannual terms are present: an 838-dy (27.5-mo) or QBO1 term, a small amplitude, 718-dy (23.5-mo) biennial or QBO2 term, and a 690-dy (22.6-mo) sub-biennial (SB) term, whose period arises from the difference interaction between QBO and annual terms. The relative amplitudes of these three interannual terms vary with latitude and altitude. The 22.6-mo term agrees closely with the anti-symmetric EOF identified by Dunkerton (his Eq. (5.2), JAS, 2001) for the subtropical latitudes from a shorter HALOE methane data series. In addition, at higher northern latitudes of the middle stratosphere there is a weak, decadal-scale variation in the data series, whose amplitude seems to be characterized by a difference interaction between the QBO and sub-biennial terms (or a 10.6-yr period). That term is not part of the MLR models in the present study, however. The primary purpose of identifying and including interannual terms in the MLR model is so that their significant structures are accounted for as part of analyses for linear trend terms. It is also noted that the time series of methane at 50 hPa has a slightly non-linear (quadratic) character, whose maximum value occurs in 1999. This character is presumably reflective of an episodic increase in the methane source gas that is present in measured, ground-level time series. However, no similar non-linear character is apparent in the CH_4 data series at the next higher level of 30 hPa.

P24190/L17: net seasonal heating—I will say instead that the methane in Figures 3 and 4 exhibits a strong seasonal cycle due to the circulation in the meridional plane that dynamically balances the differential heating between equator and pole (or in the upper stratosphere from summer to winter hemisphere). That circulation is termed the diabatic circulation, while the BDC takes into account the effects of meridional mixing, too (Andrews, et al., 1987).

P24190/L21: width of bins—I have replotted Figure 3 (attached, but now for 10 hPa), based on the 20˚-wide bins used to generate the distribution of the trends in Figure A1. The anomalously low points in the series at 55˚S are still not accounted for by the AO or SAO terms. This circumstance is because those perturbations are present in the data for only a short time (few weeks) in late winter/early spring, most likely related to the time of the final warming in the southern hemisphere. HALOE does not sample often enough for the MLR model to resolve those perturbations. If such perturbations do not occur at an end point of the time series, they have little to no effect on the analyzed trend coefficient.

P24191/L10ff: mixing non-periodic—Figure 4 has been regenerated based on analyses at 15˚S and at 15˚N latitude, each having bin widths of 20 degrees. The same general features are present, as in original Figure 4, although there are fewer "outliers" than before in the data series at 15˚N. This result is mainly due to the somewhat narrower bin width of the present data series. Effects of the QBO forcing are present at 15˚N; however, the amplitude of the annual cycle seems to be dampened at 15˚N, at least compared with that at 15˚S. As shown by Rosenlof (her Figure 1, 2002), the HALOE sampling at low latitudes did not change by much at all from 1991 to 2001, and the reduced sampling thereafter affected both the southern and northern hemisphere observations in approximately the same way each year (see the HALOE Website, click on Coverage in the left menu, and then select a given year). You are correct to say that the effects of mixing can be periodic, but perhaps not with the same intensity and/or extension to 15±10˚N each year. To my mind, the HALOE CH4 time series are well-suited for examining and comparing such variations for the northern versus the southern hemispheres, although I am not focusing on that aspect of the CH4 time series in this manuscript.

P24191/L23: I shall explicitly say that I am taking into account a 2-yr delay for the surface methane source gas to reach the tropical middle stratosphere when choosing to consider a time frame of 1990-2003 for the source time series. Methane from remote boundary layer sites was globally-averaged by Dlugokencky et al. (see GRL, 2001 and their Figure 1 in GRL, 2009). They showed that the de-seasonalized CH4 mixing ratios continued to grow during this period, although at rates that were slower from 1992 to 1999 (3.0 %/decade) and from 1999 to 2005 (near zero) than from 1983 to 1992 (4.7 %/decade). Exceptions are the episodic increases in 1991 and 1998 that were followed by sharp decreases the next year. By taking endpoint values from their global time series at 1990 and 2003, I estimated an overall trend of 3.1±0.7 %/decade. That trend becomes slightly larger, if I consider 1988 as my early-year end point. The globally-averaged, 1-σ uncertainties from the station data are of the order of 0.6 ppbv/yr, giving the 2-σ value of 0.7 %/decade above. A separate indication of the CH4 source term to the stratosphere is obtained from what I find in the tropics from my analysis of the HALOE data. To that end, I include a revised Figure 5 that shows CH_4 time series at 50 hPa for 5˚S and for 5˚N that are part of the trend results in Figure A1. Their linear trends and 2σ uncertainties are 2.8±2.8 and 3.3±3.4 %/decade, respectively, or essentially the same values as from the tropospheric time series, although the tropospheric data have a much smaller uncertainty. The two tropical time series in Figure 5 have a maximum in late 1992 and a somewhat non-linear (quadratic) character thereafter with a maximum in 1999. Both maxima follow the episodic increases in the troposphere by about a year. Average HALOE trends (and 2σ) values are 1.7±2.1 and 3.1±2.1 %/decade for 5˚S and 5˚N at 30 hPa in Figure A1, but those data time series exhibit very little non-linear character (not shown).

P24192/L26: to detect: I'll rephrase to say "…to detect an acceleration of the hemispheric …"

P24193/L1: See my response to comment about P24191/L23. I propose adding the $2\text{-}\sigma$ error estimate of ±0.7 %/decade to the captions of Figures 6 and 7.

P24193/L18: I will remove this sentence from here. But see my reply to your comment for P24189/L14ff. Checking for any remaining structure in the residuals is a standard acceptance test of a given MLR model. However, if the remaining structure is episodic, rather than periodic, it is often difficult to assign it to a specific forcing function.

P24194/L1: I will delete "correct" and use your suggested phrase, which I agree is much more to the point.

P24195/L10: I conducted my own analysis of the HALOE CH_4 time series at 2 hPa for 1992 to 1997, and I find a large negative trend similar to that of Nedoluha et al. (1998) and to a lesser extent to that of Randel et al. (JGR, 1999). In each case our trends are due to the time span of the time series. Although there are slightly fewer HALOE samples for the time series from 2001 to 2005, Figure 8 shows that the data points are still adequate for defining even the SAO term in the CH_4. I will change the sentence at L10 to say that I obtain a much different, weakly positive trend in Figure 8, when I consider the entire time span of 1992-2005.

P24195/L21ff: SR and SS images across latitude zones can be found on the HALOE Website by clicking the menu "Browse Images", followed by displaying a "pressure vs. latitude cross section" for CH4 for a selected month and year. It is impractical for me to show a number of the images and discuss them in the paper. However, I shall refrain from referring the reader to the HALOE Website in the revised manuscript, and in this instance will point the reader to similar plots that were published by Randel et al. (1998), Ruth et al. (1997), and Shu et al. (2013).

P24196/L6ff: Your interpretation is what I meant to convey with this sentence. Since I am not explicitly accounting for the occurrences of SSWs in my MLR model, their effects are still present in the residual and affect the analyzed trend term.

P24196/L25: You are correct that the ratio strictly applies only if the source of H2O to the stratosphere is constant, and it has been shown by others that this is not the case. My comparisons of the H2O and CH4 trends are meant to indicate that they are qualitatively about right for the middle stratosphere to lower mesosphere. For better clarity, I have replotted my findings of the trends in terms of ppmv/decade in revised Figures 9 and 10. I show their mutual trends at 35 degrees latitude, in order to be representative of the southern and of the northern hemispheres and to verify that the analyzed CH4 trends are reasonable. Plotting the trends in terms of ppmv/decade is analogous to that shown in Randel et al. (Figure 6, JGR, 1999), although I avoid including a profile for (H2O + 2CH4). Even so, an approximate 2:1 relationship can be found in the upper stratosphere, at least within the uncertainties of the trends. Seasonal and interannual variations of H2O are large in the tropical lower stratosphere, so one must account for them with good accuracy. It is likely though that the trends in H2O in that region are affected by the more episodic ENSO (?) forcings that are not accounted for in my MLR model. This circumstance affects any anti-correlations between H2O and CH4 during entry through the tropical tropopause and ascent to the middle stratosphere. I clearly find the stepwise decrease in HALOE H2O discussed by Fueglistaler (2012), especially in the tropical lower stratosphere. For instance, I find slight increases in tropical H2O in 1999 to early 2001 at 50 hPa, followed by a sharp decline after that (not shown). In fact, it would be more appropriate to use a piecewise linear fit to the H2O time series data at those altitudes. I find that a change in H2O also occurs about a year later at 10 hPa, although it is dampened considerably at middle latitudes and is less pronounced in the northern than in the southern tropics (not shown). An episodic change for the source of H2O to the lower stratosphere is a primary reason why I do not focus on H2O as a tracer of the BDC throughout the stratosphere.

P24197/L29ff: Figure A1 shows that the linear trends are positive through most of the northern lower stratosphere, while zero to negative in southern hemisphere. It may be that we are seeing a net transport of low values of CH_4 from the southern polar vortex region toward middle latitudes at 30 to 50 hPa.

P24198/L8: I agree that the age-of-air spectrum ought to be similar in both hemispheres, at least that part due to the diabatic circulation. However, the trends in H_2O at 10 hPa are much weaker at middle latitudes of the northern versus the southern hemisphere (c.f., H_2O trends in revised Figures 9 and 10), and any possible sampling issues for the HALOE profiles occurred in a very similar way for each hemisphere. I tentatively conclude that we must be seeing a more important role of wave forcings and mixing for the BDC within the northern hemisphere. I also plotted the H_2O time series at 10 hPa and at 60 degrees latitude of each hemisphere (not shown). Yet, a sudden change due to the shift in the entry level H_2O from 2000/2001 is not apparent at that latitude for either hemisphere. Of course, it may be that the HALOE measurements did not extend for quite long enough to see that change, at least according to independent estimates of age-of-air (Hegglin et al., Nature Geoscience, 2014).

P24198/L19ff: You are correct to conclude that significant periodic, interannual variability is being accounted for in the MLR models, but episodic forcings due to SSWs or their equivalent are not part of the analysis.

Figure 12: Units of residual are HCl (ppbv) and CH_4 (ppmv); I will add them to the captions.

Figures 11 and 12: I presume you meant to say Figures 12 and 13. I will combine them.

Additional References:

Andrews, Holton, and Leovy, Middle Atmosphere Dynamics, 1987.

Dlugokencky et al., Geophys. Res. Lett., 28, 499-502, 2001.

Hegglin, et al., Nature Geoscience, doi:10.1038/ngeo2236, 2014.

Randel, et al., J. Geophys. Res., 104, 3711-3727, 1999.

Table A1. Mean CH_4 mixing ratio profiles (ppmv).

P (hPa)	60S	35S	10S	10N	35N	60N
0.4	0.20	0.23	0.23	0.24	0.24	0.20
0.5	0.21	0.24	0.25	0.26	0.26	0.20
0.7	0.22	0.28	0.29	0.31	0.30	0.22
1.0	0.23	0.31	0.36	0.40	0.36	0.26
2.0	0.28	0.44	0.57	0.62	0.49	0.35
3.0	0.37	0.56	0.73	0.77	0.56	0.41
5.0	0.53	0.74	0.95	0.96	0.68	0.52
7.0	0.65	0.86	1.11	1.11	0.79	0.63
10.0	0.78	0.98	1.26	1.24	0.90	0.75
20.0	0.98	1.15	1.45	1.42	1.09	0.97
30.0	1.10	1.22	1.52	1.51	1.19	1.06
50.0	1.23	1.35	1.58	1.56	1.34	1.21

Table A2. Southern and northern hemisphere tropical and extratropical CH_4 trend profiles (% decade^{-1}) and the associated confidence intervals (CI in %) for their presence in the MLR models.

Pressure (hPa)	60±5˚S, CI	10±5˚S, CI	10±5˚N, CI	60±5˚N, CI
0.4	-5.3, 75	8.6, 5	7.7, 34	-29.6, 98
0.5	-7.2, 72	9.4, 7	8.7, 33	-25.2, 93
0.7	-11.0, 83	11.0, 13	1.5, 18	-28.0, 92
1.0	-13.3, 89	5.9, 13	-4.2, 59	-22.5, 78
2.0	-6.2, 60	5.0, 21	1.2, 67	-3.7, 22
3.0	-5.4, 82	15.4, 48	8.7, 11	-3.3, 71
5.0	-3.7, 90	17.4, 97	20.4, 91	-6.3, 99
7.0	-0.7, 57	13.3, 99	12.2, 97	-11.3, 99
10.0	1.0, 7	6.9, 96	9.8, 99	-19.5, 99
20.0	4.0, 69	1.8, 93	10.0, 99	-8.4, 97
30.0	0.1, 7	1.5, 85	3.8, 99	0.3, 2
50.0	-4.1, 62	1.8, 70	3.3, 99	3.8, 84

Table A3. Methane trend profiles (%/decade). Asterisk denotes a trend term having a confidence interval (CI) $\geq$ 95%.

P (hPa)	10N ±10	10N ±5	35N ±10	35N ±5	60N ±10	60N ±5
0.4	10.2	7.7	23.5	12.3	-8.5	-29.6
0.5	9.6	8.7	24.9	8.6	-10.7	-25.2
0.7	3.1	1.5	23.8	4.5	-17.8	-28.0
1.0	-0.6	-4.2	21.3	2.0	-19.0	-22.5
2.0	5.1	1.2	23.4	7.8	-10.3*	-3.7
3.0	8.2	8.7	25.4	15.5	-7.2	-3.3
5.0	16.3	20.4	15.7*	13.8	-8.3*	-6.3*
7.0	11.5*	12.2*	7.7	7.0	-14.2*	-11.3*
10.0	7.4*	9.8*	4.2	1.3	-21.3*	-19.5*
20.0	8.0*	10.0*	9.7*	5.7*	-6.7*	-8.4*
30.0	4.3*	3.8*	10.7*	8.6*	0.9	0.3
50.0	3.4*	3.3*	6.5*	6.2*	3.4*	3.8