Updated Reply to Comments of Reviewer 1 (in italics)

This study investigates variability in stratospheric CH4 as measured by HALOE during the 1992-2005 time period. This is a follow up to a previous study on this topic by the same author. The trends over three 5-year periods are found by multiple linear regression with the expected terms included to account for stratospheric variability. The focus on 5-year periods rather than the entire 13-year period as in the author's previous paper is understandable due to the variability in the 5-year trends. Reexamination of available measurement data of this kind to try to infer more information is useful and important.

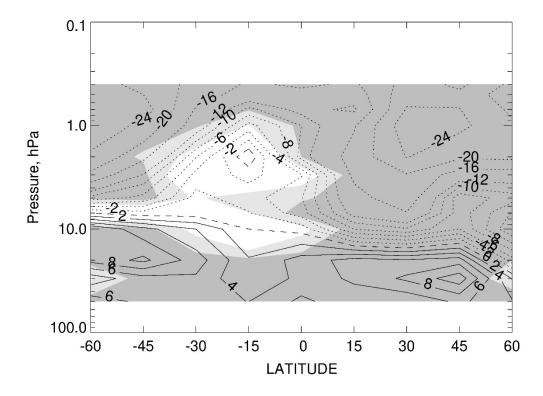
While I support this study and applaud the brevity, the discussion of the trends is confusing and needs some rewriting before publication. I have listed some specific instances below where I was unclear about the discussion and I would suggest that at least these points be addressed. But I would also encourage the author to review all of Section 3 for clarity and consistency. The summary section is written clearly so that does help get the main points across in the end.

Thank you for your careful review of the manuscript and for pointing out several inconsistencies about my interpretations of the findings from Figures 3-5 (now Figs. 3, 5, and 6). I have added more details to Section 2 about my analysis method, and I have rewritten Section 3 to address your concerns about the contradictions that you noted. I have also added one figure and one reference to Subsection 3a.

## Specific comments

Line 77: The 5-year time span chosen to evaluate trends likely doesn't eliminate all bias due to the QBO since the QBO has a variable period that is almost never exactly 2.5 years at all latitudes and altitudes. How different would Figures 3-5 look if you started the time series in June or August? Your only mention of this is in lines 187-9. Related to this, it would be helpful to show the statistical significance of the trends in Figures 3-5.

I tested the sensitivity of the trend coefficient from the MLR fit in Fig. 1a (30°N, 10 hPa), where a QBO cycle shows clearly. For the testing I changed the length of the QBO cycle (28 months versus 29.5 months) and then separately by considering a time span of 58 months rather than 5yr (or 60 months) for the MLR analysis. The resulting coefficients differed by less than 6% from the trend coefficient of Fig. 1a. I conclude that the 5-yr trend distributions in Figs. 3-5 (now 3, 5, and 6) are robust with respect to my approximations of the QBO cycle. On your related suggestion, I added shading to indicate the significance of the trends, where dark shading is where the confidence interval (CI) for the trends is greater than 90% and where light shading is for CI between 70 and 90% (see revised Fig. 3 below).



Lines 108-112: I don't understand the reasoning here. The trends are negative throughout the upper stratosphere during this period and you state that the 'ascent within the deep branch of the BDC occurred mainly in the northern subtropics' where the trends are most negative. Ascent will bring relatively large mixing ratios to essentially any region of the stratosphere as shown by Figure 2. You actually state this further down in lines 131-132 when referring to the lower stratosphere. And then further down in lines 142-3 you state that the 'negative changes in CH4 in upper regions of Fig. 3 imply that there was a slowdown in the deep branch of the BDC.' This is what I would conclude as well but doesn't seem consistent with the earlier statements. Some clarification is necessary.

*I moved the two sentences from lines 142-145 to the end of the first paragraph of Section 3, giving better continuity of the findings for the upper stratosphere.* 

Lines 128-9: Wouldn't it be easier to infer BDC changes if you removed the tropospheric growth rate before doing the MLR analysis? If you had a period with no BDC changes there would still be CH4 trends in the stratosphere due to the tropospheric growth rate.

You are correct that there should be a residual increase in stratospheric  $CH_4$ , due to its positive trends within the troposphere. However, I am not confident of the exact tropospheric trend curve to use for such a correction. Nevertheless, the  $CH_4$  trends in Figs. 3-5 (now Figs. 3, 5, and 6) are much larger (or smaller) and override tropospheric trends throughout much of their pressure/latitude domains.

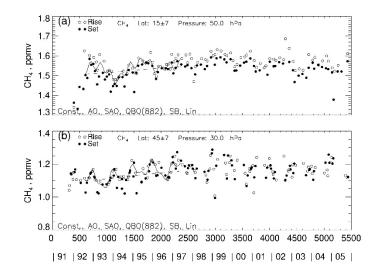
Lines 140-1: Positive trends in the tropical lower stratosphere should indicate stronger upwelling and yet you say they indicate a reduced tropical upwelling. Again, some clarification is necessary.

The sentence beginning on line 140 is incorrect; thank you for pointing that out. I have revised it in Subsection 3a.

Lines 151-3: Here you state that the SSWs led to greater ascent of CH4 in the tropical upper stratosphere where it was chemically destroyed leading to negative trends. Whereas, in the next section (Lines 183-6) you state that the SSWs in the later period 'accelerates the deep branch of the BDC bringing more CH4 to high altitudes' resulting in a positive trend. These seem to be inconsistent statements.

I corrected those inconsistencies and revised the sentence at line 151. I also modified the sentence starting on line 183.

Finally, I added the following figure to the revised manuscript to show details of the effect of the Pinatubo eruption on  $CH_4$  in the lower stratosphere, in addition to Figure 3. The top panel is for 15N, 50 hPa and shows an initial increase in  $CH_4$  in 1991-mid1992, followed by decreasing values through 1993. Those variations are in accord with changes in AoA in the shallow branch of the BDC, as reported by Diallo et al. (2017). The bottom panel is for 45N, 30 hPa and shows the steady increase of  $CH_4$  from 1992 to 1997 as a companion to the trends in Figure 3.



Updated Reply (in italics) to comments of Reviewer 2:

This technical note uses methane (CH4) profile data from the Halogen Occultation Experiment (HALOE) to diagnose changes in the Brewer-Dobson Circulation (BDC). The author analyzes CH4 trends for three 5-year time spans from 1992 to 2005 and finds significant changes in CH4 trends, particularly in the Northern Hemisphere (NH) near 30 hPa, which is a transition layer between the shallow and deep branches of the BDC.

The author finds that CH4 changes were positive and large in the shallow branch following the eruption of Mount Pinatubo, but they then decreased and agreed with tropospheric trends in the late 1990s and early 2000s. In the upper part of the deep branch, CH4 decreased from 1992 to 1997, following the Pinatubo eruption. CH4 continued to decrease in the deep branch in the late 1990s, but then increased in the early 2000s, although the changes were small compared with the seasonal and interannual variations of CH4.

The author concludes that these multi-year changes in CH4 trends were due, in part, to wave forcings during the El Niño Southern Oscillation (ENSO) of 1997-1998 and beyond, and to episodic sudden stratospheric warming (SSW) events during both time spans. The author also concludes that time series of HALOE CH4 provide effective tracer diagnostics for studies of the nature of the BDC from 1992 to 2005.

Overall, this is a well-written and informative manuscript. I recommend it for publication, with the following suggestions:

Thank for you careful review of the manuscript and for your suggested changes. However, with regard to items 1 and 3 below, I do not wish to expand my Note but to urge others to conduct their own analyses of the HALOE CH4 data for comparison.

1. Limitations of using multi-variate regression model to detect short-term trends

The author should highlight the limitations of using a multivariate regression model to detect short-term trends in CH4. A major limitation is that multivariate regression models can be sensitive to the choice of explanatory variables and the model structure. Additionally, short-term trends can be difficult to distinguish from interannual variability. Authors should also mention that overall tropsopheric CH4 trends are non-linear (hiatus and then rapid increase).

At the suggestion of Reviewer 1, I altered the QBO term in my model for the latitude and pressure level of Fig. 1a and found only very minor differences of the analyzed trend coefficient. While there may be other regressors, I did examine the MLR time series residuals for each pressure level and latitude zone but did not find any significant periodic structure in them.

2. Role of OH chemistry in CH4 loss

The author should discuss the role of OH chemistry in controlling CH4 loss rates. Changes in OH concentrations can have a significant impact on CH4 trends. For example, the eruption of Mount Pinatubo injected sulfur dioxide into the stratosphere, which led to the formation of sulfuric acid aerosols altering OH concentrations (e.g Branda et al., 2014). Authors should also discuss importance of this pathway.

Bândă et al., (2015), The effect of stratospheric sulfur from Mount Pinatubo on tropospheric oxidizing capacity and methane, J. Geophys. Res. Atmos., 120, 1202–1220, doi:10.1002/2014JD022137.

The effects of  $SO_2$  and aerosols on the production of OH and the loss of tropospheric CH4 appear to be secondary, especially when compared with the lower stratosphere trends of  $CH_4$  in Figures 3 and new Figure 4. In addition, those secondary effects do not extend past 1992.

3. Comparison with gap-free data

The author could compare the results from the raw HALOE data with the gap-free stratospheric CH4 profile data constructed by Dhomse and Chipperfield (2023). This comparison would provide additional insights into the accuracy and reliability of the results.

Dhomse Sandip S. (2022). TCOM-CH4: TOMCAT CTM and Occultation Measurements based daily zonal stratospheric methane profile dataset [1991-2021] constructed using machine-learning (1.0) [Data set]. Zenodo. <u>https://doi.org/10.5281/zenodo.7293740</u>

Dhomse and Chipperfield report that their gap-free, zonal mean CH<sub>4</sub> profiles agree to within ~10% with those of the original HALOE dataset. I conclude that it is very unlikely that separate analyses of their gap-free dataset using my same regressors will yield trends that are qualitatively different from what I am showing. Therefore, I do not wish to expand my Note to include additional analysis results for my three, 5-yr time spans. Others may want to consider those gap-free HALOE data and also to extend their analyses to 2012 and to the present day using the MIPAS and ACE data, respectively.

Minor comments:

Line 147: Change "July 1996" to "July 1997".

The year 2996 in the sub-head is a typo and should be 1996; the second, 5-yr time span has a one-year overlap with that of 1992-1997 of Figure 3. The third 5-yr time span also has a one-year overlap with that of 1996-2001 of new Figure 5.