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Atmospheric Chemistry and Physics Discussions
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Interactive comment on "Equatorial middle atmospheric chemical composition changes during sudden stratospheric warming events" *by* O. Nath and S. Sridharan

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Response to Reviewer-1's comments

We thank the Reviewer for his/her comments, which greatly helped us to improve the manuscript. Our point-by-point response to his/her comments is given below.

The paper of Nath and Sridharan reports some changes in atmospheric tracers (H2O,CH4 and O3), recorded by MLS/Aura and MIPAS/Envisat, which developed at equatorial stratospheric latitudes roughly in correspondence with the occurrence of strong SSW events (i.e. 2004, 2009 and 2012) in northern polar regions. In the case

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of O3 the authors attributed such changes to the weakening of BDC and to chemistry because loss processes for O3 are less efficient at cold temperatures. Moreover, they suggest that the low temperatures favoured the shift in the ratio of O3/O toward O3. Then, the reduction of atomic oxygen abundance affected the methane oxidation which resulted in decreased (increased) water vapor (methane) abundance. Even if the topic is potentially interesting, the present study is too qualitative and still very preliminary. The whole introduction focus on chemical changes occurred at Polar Regions and no reference (excepted two studies of the same authors) deal with equatorial regions.

As per our best of knowledge, changes in equatorial chemical compositions during the major SSW events have not been reported except a few, namely, Sridharan et al., 2012, Nath et al. 2015, which showed only enhancement of ozone during the SSW events. The other papers (Wrotny et al., 2010; Stolarski et al, 2012 for example) focussed on long-term variations of chemical constituents over equator, whereas the present manuscript report for the first time the short-term variations of chemical constituents namely ozone, CH4 and water vapour during the disturbed polar winter times and gives possible explanations for those variations. The variations are not just seasonal, as they occur closely during the SSW events, the time of occurrence of which varies from early winter to late winter.

The analysis shows only some time series of the investigated constituents over the equator under the period of interest and the figures are not well organized (e.g. these results could easily be constrained in 3 figures, one for each SSW).

In the revised manuscript, the figures are made organized.

The discussion on the possible mechanisms which cause such changes reduces to few lines and it is not convincing.

As suggested by the Reviewer, we expanded the discussion part.

The temperature dependence of the odd oxygen loss reaction is more effective in upper stratosphere, say around 1-2 hPa (e.g. Stolarski et al., JGR, 2012). For example, the panels ‘d’ and ‘f’ of Fig. 1 suggest the occurrence of these changes at about 45 km around DOY 30. Nevertheless the O3 increase at these altitudes seems to last only about a week in both 2009 and 2012 while variability in H2O and CH4 encompasses many weeks. O3 variations at lower altitudes are probably related to dynamics. Therefore, I believe that the potential shift in the ratio of O3/O toward O3 cannot explain such changes.

We agree with the Reviewer and we are taking back our hypothesis of the temperature dependence of ozone variation to be responsible towards the observed decrease in water vapour volume mixing ratio. As the Reviewer pointed out, the reaction rate (of the order 10-33) is too slow to be effective in short term scale variations.

As Reviewer suggested, we considered dynamics also to explain the changes in the chemical composition. In the revised manuscript, we showed that the CH4 increase is due to tropical upwelling by relating CH4 variations with residual vertical velocity over equator.

Although the study deals with changes in tracers and the chemical conversion of CH4 to H2O with altitude is supposed to be slow, the authors did not take into account the possibility that such variability could arise also from dynamics (e.g. upwelling). This study cannot disregard this additional element.

We agree with the Reviewer that tropical upwelling plays a major role in transporting methane to the higher heights. As per the Reviewer’s suggestion we have calculated residual meridional circulation as well as residual vertical velocity over equator which are shown below- As expected, we have seen enhanced northward flow between 20°S and 80°N during the build-up to the SSW event (R1fig1a). And the residual vertical velocity is also high around 2 hPa pressure level over equatorial region which suggests tropical upwelling of methane in upper stratosphere. In the below figure we have plotted the methane volume mixing ratio and w* at 45 km height (R1fig1b) to show the

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simultaneousness of the variation-enhancement of CH4 VMR and increase in residual vertical velocity. We have shown it at 45 km because the observed variations in methane and water vapour are found to be more around 45 km (R1fig2). The source of water vapour in upper stratosphere is none other than methane oxidation which goes via two ways (Brasseur and Solomon, 2006): CH4 + OH => CH3 + H2O rate constant= 6.5x10-15 [cm3/molecule s]......................[1] CH4 + O (1D) => CH3 + OH rate constant= 1.13x10-10 [cm3/molecule s]...................[2] And for destruction of water vapour in stratosphere, the reaction is: H2O+O (1D) => OH+OH rate constant= 2.3x10-10 [cm3/molecule s]..............[3] [All the reaction rates are for the temperature range 200-300 K] The residual meridional circulation becomes stronger just before the peak day of warming which results in upwelling of methane from troposphere. The lifetime of methane in stratosphere is ranging from few months to years. So the methane which reaches the upper stratospheric height gets accumulated there for time being and takes much time to get oxidized to water vapour. In upper stratosphere the source of water vapour is nothing but oxidation of methane. The oxidation of methane can happen via two ways: reaction with OH radicals and reactions with O (1D) (reactions mentioned above). Both the reactions are very slow. Whereas the destruction of water vapour molecules in stratosphere occurs by reaction with O (1D). The rate of the reaction 3 is almost double of that of reaction 2. The respective residence time of H2O, CH4 and O (1D) in upper stratosphere (40-50 km) are few days, few months [Brasseur and Solomon, 2005] and few sec [Seinfield and Pandis, 2006]. In the upper stratosphere, the time scales for formation of ozone (O3) from atomic oxygen and oxygen molecules (O + O2 + M => O3 + M, where M is the third body) and photolysis of O3 (O3 + hÊŃ => O + O2, k=10-3 s-1) are short compared to the time scales for reactions such as recombination of atomic oxygen and O3 (O + O3=> 2O2, rate constant of the order 10-12[k=(8x10-12)exp(-2060/T) cm3molecule-1s-1]). The rate of the O (1D) production reaction is of the order 10-3. Moreover the lifetime of O (1D) is around $\sim$10-7 s. The excited state atomic oxygen gets stabilized very fast by reacting with neutral molecules like N2, O2. Out of the two methane oxidation reactions, that one via hydroxyl radical

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pathway is much slower than that via O (1D). If we compare all the reaction rates of the three reactions the oxidation of methane by hydroxyl radical can be neglected and in between the production reaction and destruction reaction of water vapour by O (1D), the later one is faster. So over all a reduction in water vapour volume mixing ratio can be observed during SSW. Moreover, the authors did not discuss the variability of CH4 and H2O in context of their respective satellite climatology preventing the possibility of e.g. to compare such changes with years not affected by SSWs and to assess the actual significance of such variability. In the abstract as well as in the figures, the authors highlight the connection between the SSW occurrence and the chemical changes at low latitudes. However they did not do any attempt to further explore this potential link. Overall, in the present form this study is below the scientific standards of ACP. Therefore, unless you address the above-mentioned weaknesses, I cannot recommend the manuscript for publication.

We thank the Reviewer for this suggestion. We used MIPAS CH4 data (R1fig3a) of years 2006-07 to 2010-11 and MLS H2O data (R1fig3b) for the years 2004-05 to 2010-11 to obtain the composite daily mean for the months December-February. We can see from the figure that the CH4 enhancement and H2O decrease are more during day number around 60-70. However, during SSW events, these changes are shifted and are observed during when SSW events occurred. It may be noted that every year there is a sudden stratospheric warming occurring in the northern hemisphere during the late winter, either major or minor. With the available CH4 and H2O data, we cannot bring climatology of CH4 variations, as most of the winters are disturbed.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C11530/2016/acpd-15-C11530-2016-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 15, 23969, 2015.

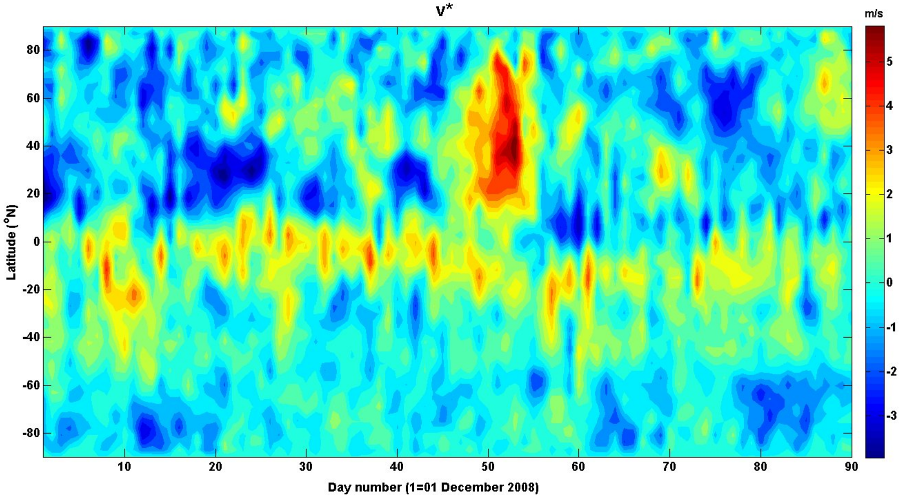
v*
m/s
5
4
3
2
1
0
-1
-2
-3
80
60
40
20
0
-20
-40
-60
-80
Latitude (°N)
10
20
30
40
50
60
70
80
90
Day number (1=01 December 2008)

Fig. 1.

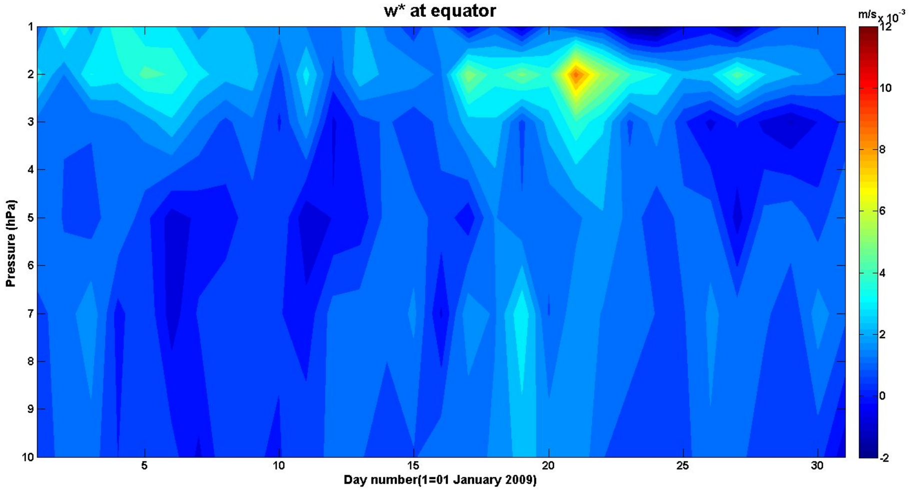

Fig. 2.

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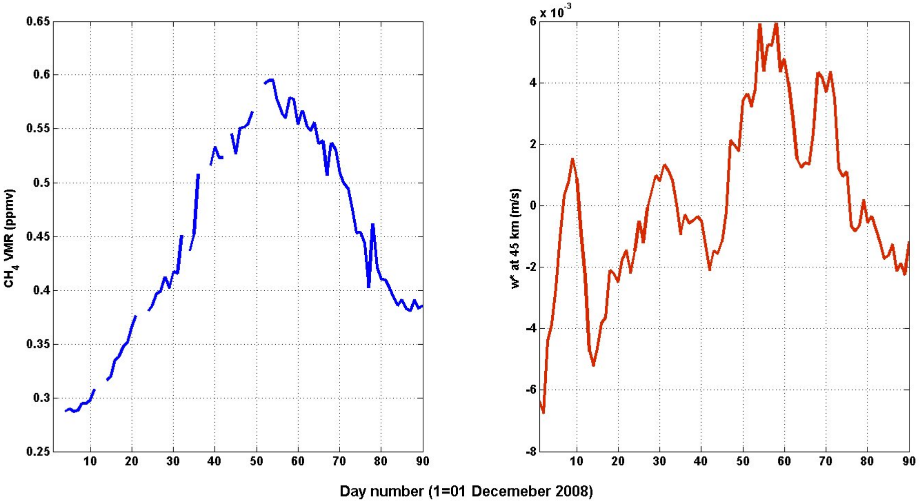

Fig. 3.

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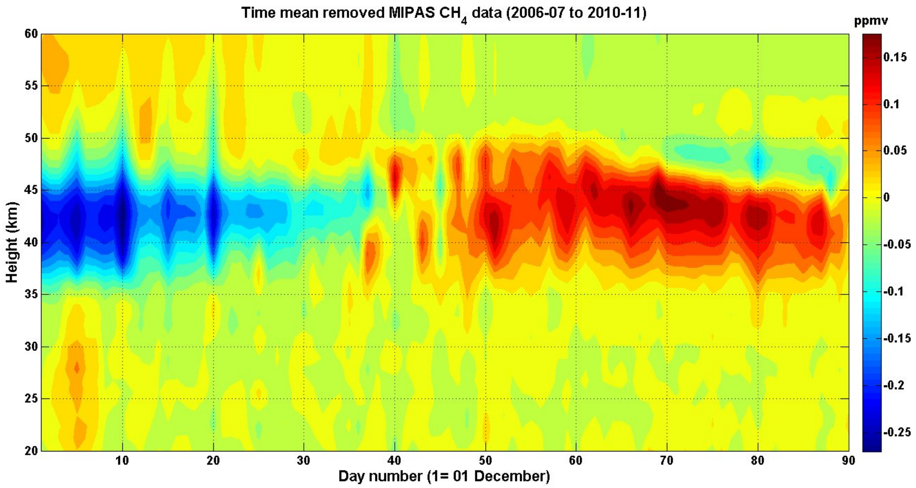
Time mean removed MIPAS CH$_4$ data (2006-07 to 2010-11)
ppmv
0.15
0.1
0.05
0
-0.05
-0.1
-0.15
-0.2
-0.25
Height (km)
60
55
50
45
40
35
30
25
20
10
20
30
40
50
60
70
80
90
Day number (1= 01 December)

Fig. 4.

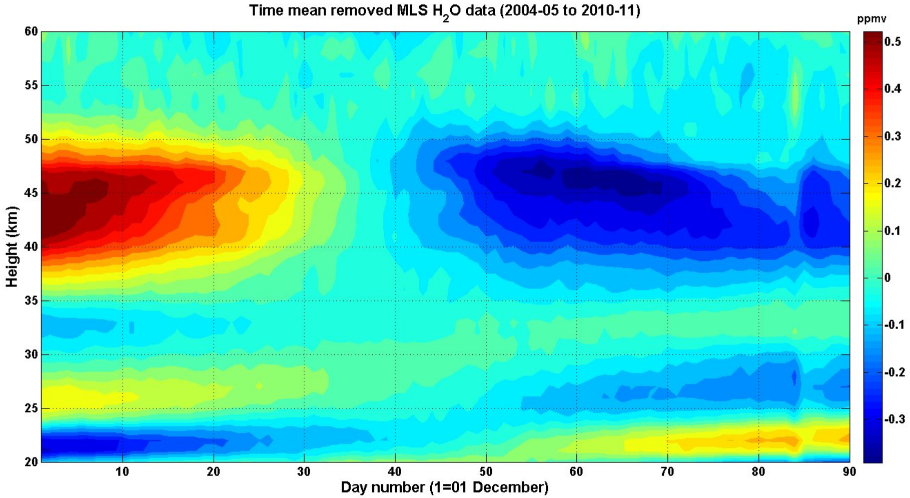

Fig. 5.

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