27 July 2016

Manuscript Title: *Evaluation of Water Vapour Assimilation in the Tropical Upper Troposphere and Lower Stratosphere by a Chemical Transport Model* **by Payra et al.**

RESPONSES TO THE REVIEWERS

We would like to thank the reviewers for their insightful comments that were helpful in improving substantially the presentation and contents of the revised manuscript. We hope we have addressed appropriately all issues raised by the reviewers. The reviewers' comments are repeated below in blue and our responses appear in black.

We have inserted this sentence in the acknowledgments:

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We finally would like to thank the two anonymous
reviewers to their fruitful comments.
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The following changes have been made in the revised manuscript.

Anonymous Referee #1

This paper reports a technique for assimilation of MLS water vapour in the UTLS into the MOCAGE chemical transport model. In principle this is a fruitful line of research - as is well known measurement of water vapour in this region of the atmosphere has many technical difficulties. Furthermore there still remains significant uncertainty over the relative importance of different processes that potentially affect water vapour and determine the variations in its concentration on daily to decadal timescales.

→ Specific changes have been made in response to the reviewers' comments and are described below. The page numbers correspond to the revised manuscript.

Point-to-point response:

1) However after reading this paper I was left wondering what exactly had been gained by the assimilation process described. To me it seemed as if the overall outcome was that in 'MLS space' (a term which was poorly explained) the assimilated water vapour fields matched the MLS observed water vapour fields. So the assimilation simply seemed to provide a systematic (and perhaps very satisfactory) way of interpolating the MLS observations in space and time.

→ The reviewer points out several issues that are linked to the terminology employed in our paper as for instance the terms 'MLS space', 'MIPAS space' and 'model space' and the general well-known method of assimilation that is, in the 3D-FGAT method, not just a clever interpolation method. Details are written in line no 429 to 431 (page no 19) of the revised manuscript. Also in the first version of the paper, we wanted to avoid writing the basic equation of the cost function J but, taking into account the comments from the two reviewers, we have updated it for more clarity. This will also help us better characterize the terms “background”, “forecast”, “free run”, etc. for which some comments are addressed by the two reviewers. Consequently, we have reformulated the entire subsection related to the assimilation.

The assimilation system used here to incorporate MLS H_2O observations in MOCAGE, is the VALENTINA system, which was initially developed in the framework of the ASSET (ASSimilation of Envisat daTa) project (Lahoz et al., 2007a), and has been used in numerous atmospheric chemistry data assimilation studies (Massart et al., 2009; El Amraoui et al., 2010; Barret et al., 2012). It is developed jointly by Météo-France and CERFACS (Centre Européen de Recherche et de Formation Avancée en Calcul Scientifique). Herein, we used a 3D-FGAT formulation (3D-Variational in the First Guess at Appropriate Time variant; Fisher and Andersson, 2001). For variational systems, the assimilation method is based on the minimization of the cost function, J, that can be formulated using the notation of Ide et al. (1997):

$$J(x) = J_b + J_o \tag{1}$$

$$J_b = \frac{1}{2}\left[\mathbf{x}(t_0) - \mathbf{x}^b(t_0)\right]^T \mathbf{B}^{-1}\left[\mathbf{x}(t_0) - \mathbf{x}^b(t_0)\right]$$

$$J_o = \frac{1}{2}\sum_{i=0}^{N}\left[\mathbf{y}^o(t_i) - \mathbf{H}_i\left(\mathbf{x}(t_i)\right)\right]^T \mathbf{R}_i^{-1}\left[\mathbf{y}^o(t_i) - \mathbf{H}_i\left(\mathbf{x}(t_i)\right)\right].$$

J_b is the misfit to the background state, and J_o represents the misfit to the observations. $\mathbf{x}^b(t_0)$ and $\mathbf{y}^o(t_i)$ are the background state at the initial time t_0 and the observation at time t_i, respectively. **B** and **R** are the background and the observation error covariance matrices, respectively. $\mathbf{x}(t_i)$ is the model state at the observation time, t_i, and represents the propagation of the initial state, $\mathbf{x}(t_0)$, by the model operator, **M**:

$$\mathbf{x}(t_i) = \mathbf{M}_i\mathbf{x}(t_0). \tag{2}$$

$\mathbf{H}_i$ is the observation operator, generally non-linear, which maps the model state $\mathbf{x}(t_i)$ to the measurement space where $\mathbf{y}^o(t_i)$ is located. The subscript i refers to time and N is the number of time steps in the assimilation window $[t_0 , t_N]$.

Since we are interested in the study of the diurnal cycle of H_2O in the tropical tropopause based on the work from Carminati et al. (2014), we have setup VALENTINA with an assimilation window of 1 hour to assimilate MLS H_2O observations. Although

> the VALENTINA system has the capability to include the effect of the averaging kernel, which takes into account vertical variations of the sensitivity of the retrieval to the actual H_2O mixing ratios, we will not use this opportunity in the present study (see section 4). In VALENTINA, the background error covariance matrix (B) formulation is based on the diffusion equation approach (Weaver and Courtier, 2001) and can be fully specified by means of a 3D standard deviation field (diagonal of B) and 3D fields of horizontal (L_x and L_y) and vertical (L_z) local correlation lengths. This assimilation technique has already produced good quality results compared to independent data sets, especially for O_3 and CO (see e.g., Abida et al., 2016; El Amraoui et al., 2010; Claeyman et al., 2011).

We have inserted a new reference:

Ide, K., Courtier, P., Ghil, M., and Lorenc, A.: Unified notation for data assimilation: Operational, sequential and variational. J. Meteor. Soc. Japan, 75, 181–189, 1997.

→ Regarding the terminology, a great care has been taken to define the terms "MLS, MIPAS and model spaces" in the abstract, in the core of the manuscript and in the conclusion by inserting such a sentence:

> The studies have been performed within 3 different spaces in time and space coincidences with the MLS (MLS space) and MIPAS (MIPAS space) observations and with the model (model space) outputs and at 3 different levels: 121 hPa (upper troposphere), 100 hPa (tropopause), and 68 hPa (lower stratosphere).

2) To me the potential gain of assimilation is that it acts as a kind of filter on a given set of observations - selecting (and gaining value from) those aspects of that set that are not in strong conflict with other observations and the underlying model. Here the 'other observations' are meteorological observations that determine the state of the ARPEGE model and hence provide a kind of the lower boundary condition for the MOCAGE model - but the evidence presented in the paper seems to imply that they play very little role in determining the concentrations of water vapour resulting from the assimilation.

→ The potential gain of assimilation is not in a sort of filtering/selection of observations against model or in a sort of a clever interpolation of observations in another observation/model space. The potential gain of assimilation is presented in Figure 4. It clearly shows that the data assimilation is an analysis technique in which the observed information is accumulated in the model state. This information is then spread on a large scale through dynamical processes to build a more consistent analysis. We confess we did not discuss long enough the importance of the outcomes of this Figure in the previous version of the manuscript. So, we have revisited the associated paragraph.

Figure 4 shows the temporal evolution of Observations-minus-Forecast (OmF) during the long-run (1st August 2011-31 January 2012) assimilation experiment at three MLS pressure levels: 121, 100, and 68 hPa. The MLS assimilated observations minus their forecast-equivalent values are averaged over the tropics (30°S-30°N) for each hour. The time evolution of the zonally-averaged OmF at the 3 levels is a key indicator of the potential gain of the assimilation tools. Indeed, the 3DFGAT assimilation does not just act as a clever filter or interpolation of observations, it also plays at improving the background knowledge as observations are injected in the system as time evolves. This is clearly indicated in Figure 4.

In August 2011, the background state is by definition set to the free model. The model forecast is initially high biased with respect to MLS observations at 121 hPa (about –4 ppmv) and at 100 hPa (about -2 ppmv) whilst it is unbiased at 68 hPa with a variability of ±0.1 ppmv (Fig. 4). The OmF magnitude decreases gradually with time over the whole long-run experiment time period. It takes about four months of assimilation, by December 2011, to reach a model forecast state with minimum values of OmF reduced to: ±0.2 ppmv at 121 hPa, ±0.1 ppmv at 100 hPa and ±0.05 ppmv at 68 hPa. This means that, by December 2011, the background state is no longer set to the free model but rather close to the observations. This emphasizes the extreme difficulty of constraining MOCAGE H_2O field, which is marked by important biases, when assimilating only MLS measurements.

3) Alongside this, the model used as a basis for the assimilation apparently simply treats water vapour as a tracer, with for example, no loss through condensation. This is clearly an unsatisfactory model for water vapour in the tropical UTLS. So my view is that the authors provide a clearer justification for the procedure they have chosen. Why, for example, have they essentially imposed a discontinuity - across 135 hPa - in the influence of ARPEGE on the one hand and MLS on the other. Would it not be better to have some kind of blend of the two in a transition region. Why did they not include some kind of saturation criterion in the MOCAGE model for water vapour?

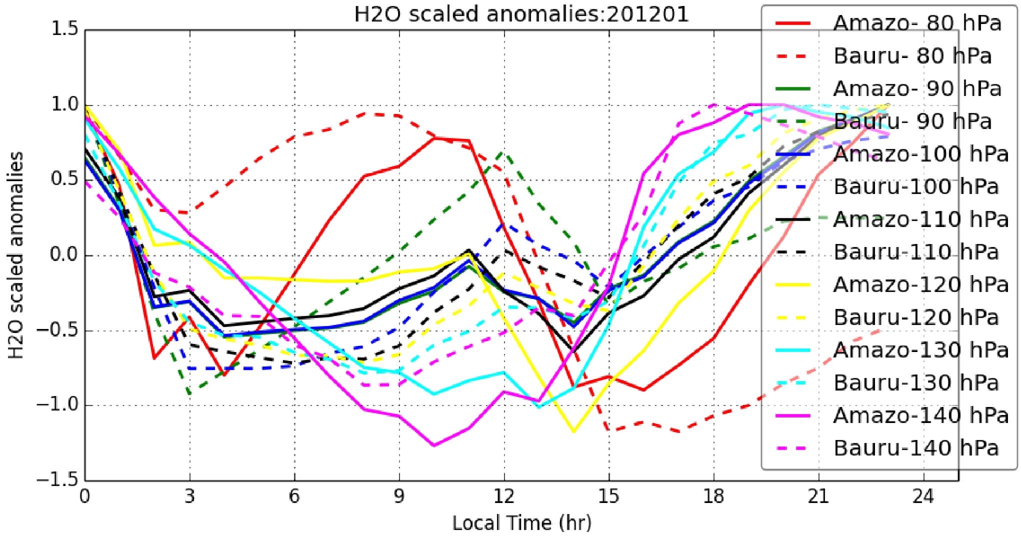
→ Initially, the MOCAGE model was set up to treat water vapour as a chemical compounds only in the stratosphere. Consequently, “tropospheric H2O” was constrained by ARPEGE and “stratospheric H2O” was calculated by the standard chemical kinetic reactions available in the stratosphere. There were no need of neither supersaturation, nor microphysics in the model. The transitional region between the “tropospheric H2O” and the “stratospheric H2O” was initially defined by the 10-ppmv limit. For mixing ratios less than 10 ppmv, H2O was treated by MOCAGE. For mixing ratios greater than 10 ppmv, H2O was treated by ARPEGE.

It was clearly impossible to use such an unique criterion to study H2O in the TTL. Two important parameters had to be taken into account from 1) the observations, and 2) the models. 1) Regarding the observations, we knew from Carminati et al. (2014) that, although MLS H2O measurements are worthwhile using down to 300 hPa, essentially 3 independent layers need to be studied in the TTL at 121, 100 and 83 hPa. This meant we needed to use MOCAGE in a configuration at least down to 121 hPa, but not too low, because of the presence of clouds, of supersaturation, and microphysical processes that are not present in the model. The trade-off between observation and model constraints has been to choose a pressure layer of 135 hPa as a transition between ARPEGE and MOCAGE. This is indeed a simple procedure but by considering the factual scenario. So it is already giving interesting results. But this is a procedure that could be improved in the future, as suggested by the reviewer: transitional region, microphysics, … We have thus updated the text in order to explain the reasons why we chose the 135-hPa transitional region.

> However, to achieve the goal of our study, namely to constrain MOCAGE H_2O as chemical species by actually using MLS observations at 121, 100 and 83 hPa, we have modified this initial treatment by considering a transition level at 135 hPa. Tests have shown that 135 hPa was the optimum transition level since, for transition pressures greater than 135 hPa, the impact of ARPEGE H_2O onto the assimilated fields in the upper troposphere/tropopause layer was negligible. In conclusion, i) for pressures greater than 135 hPa, H_2O is calculated directly from ARPEGE specific humidity, and ii) for pressures less than 135 hPa, the H_2O distribution is fully controlled by MOCAGE via the chemistry and transport schemes. This has the main advantage of being very simple to run but has the main drawback to produce unrestricted supersaturation in the upper troposphere/tropopause layer (see section 5.2).

4) The authors should also make it clearer what in their view has been gained from the assimilation. (Is it any more than spatial and temporal interpolation of the MLS fields?)

→ We have indeed presented in detail what has been gained from the assimilation in our study, both in the reply to the comments from 1) to 3) from the Reviewer#1 and in the new version of the manuscript. In addition, as underlined in the conclusions, the analyses will be used to assess the impact of the continental convective activity on the diurnal cycle of H2O in the tropical UTLS above the Southern American continent (Amazonia vs. Bauru, Brazil) with a temporal resolution of 1 hour (see **Figure R1**).



<u>Figure R1.</u> Relative diurnal cycle of H_2O analyses (%) from MOCAGE-VALENTINA above Bauru, Brazil (17-27°S; 44-64°W) and above Amazonia (0-10°S; 55-75°W) at different pressure levels from 140 to 80 hPa in January 2012.

There are many improvements that could be made to the paper on a detailed level – as set out in comments below.

5) l23: 'hPa hPa' > 'hPa'
→ Done

6) l34: 'In the MLS space', 'in the model space', etc - I found it difficult to guess what precisely you meant by the term 'space' - use some more straightforward terminology?
→ We modified the incriminated sentence into:

> The studies have been performed within 3 different spaces in time and space coincidences with MLS (hereafter referred to as MLS space) and MIPAS (MIPAS space) observations and with the model (model space) outputs and at 3 different levels: 121 hPa (upper troposphere), 100 hPa (tropopause), and 68 hPa (lower stratosphere) in January and February 2012.

7) l40: 'prevent to assess' - this is an example of a minor problem with English grammar (MPEG) - should be 'prevent assessment'.
→ Done

8) l52: 'and transported from one place to another on the globe' > 'and to transport it from one place to another on the globe' (MPEG)
→ Done

9) l53: ‘Unlike other greenhouse gases’ > ‘unlike some other greenhouse gases’, or perhaps the sentence is confused by ‘additional water vapour’ - ‘additional’ with respect to what?

→ We have clarified this sentence into:

> Unlike some other greenhouse gases, the contribution of anthropogenic sources to the atmospheric water vapour is negligible (IPCC, 2007).

We have inserted a new reference:

Intergovernmental Panel on Climate Change, Working Group I: The Physical Science Basis, 2.5.6 Tropospheric Water Vapour from Anthropogenic Sources, Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis, K.B. Averyt, M. Tignor and H.L. Miller (eds.), Cambridge University Press, 2007.

10) l61: Seems odd to give Panwar et al., 2012 as a reference for low concentrations of stratospheric water vapour. The fact that stratospheric water vapour concentrations are low has been known for a long time (not just since 2012) and over the years there have been several review papers on this topic (e.g. see reference list of Randel and Jensen 2013).

→ Reference modified from Panwar et al. (2012) into Randel and Jensen (2013)

11) l80: ‘Heggling’ > ‘Hegglin’ (!)

→ Done

12) l94: ‘Around the tropopause, large gradients in H2O and interplay of transport processes between troposphere and stratosphere, mainly due to rapid change in H2O by deep convection’ - re deep convection - are you thinking specifically of the tropical tropopause? Even here it is not clear to me that deep convection is the most difficult process to understand nor the main mechanism for generating large gradients.

→ Yes indeed, we were referring to the tropical tropopause and to the difficulty for the modelled deep convection to penetrate into the uppermost troposphere/lowermost stratosphere. We have reformulated the sentence into:

> Around the tropical tropopause, large vertical gradients in H_2O and interplay of transport processes between troposphere and stratosphere, mainly due to rapid change in H_2O by deep convection reaching the uppermost troposphere/lowermost stratosphere, are highly challenging for an accurate representation of H_2O in global models.

13) l122: ‘The changes mostly impacted H2O fields over this period’ > ‘The changes over this period which have had most impact on H2O fields’ (MPEG)

→ Done

14) l140: 'there is no lower stratospheric wet bias as suggested in previous studies' of course these studies were not of ERA-I - so needs to be amended to something like 'in studies of earlier ECMWF analysis or re-analysis fields'.

→ Done

15) l243: 'bias' in this case presumably means 'difference between MLS and frost-point hygrometer' - i.e. at 83hP and 100Pa MLS is showing larger water vapour concentrations than the frost-point hygrometer? Please clarify.

→ We clarified this point and modified the sentence into:

> At 83 and 100 hPa, statistically significant biases from 0.1 to 0.3 ppmv (from 3 to 8%) were found, with MLS showing larger water vapour concentrations than the frost-point hygrometer.

16) l249: 'following the three independent vertical layers in the TTL' - do you mean 'vertical levels' - i.e. you are considering MLS-derived water vapour mixing ratios for the precise levels - or do you mean finite layers (each centred on one of these levels?

→ We have clarified this point by modifying the sentence into:

> With a methodology approaching that of Carminati et al. (2014), we will consider in the following the 3 independent vertical layers in the TTL, for which the most representative averaging kernels peak at 121 hPa for the upper troposphere (UT), 100 hPa for the tropopause (TP), and 68 hPa for the lower stratosphere (LS). See for instance Fig. 3 of Carminati et al. (2014) for a representation of the 3 vertical layers.

17) l332: It would be useful to know a bit more about the physics of water vapour in ARPEGE. Is there condensation immediately saturation is reached, or is some limited supersaturation allowed?

→ A new sentence has been inserted, together with a new reference.

> The condensation scheme is based on the probability density function from Smith (1990). Supersaturation is not allowed by the physics but can be created by horizontal advection and then removed by the physics.

Smith, R. N. B.: A scheme for predicting layer clouds and their water content in a general circulation model. Q. J. R. Meteorol. Soc., 116, 435-460, 1990.

18) l356: 'capability to include the effect of the averaging kernel' - this is the first time you have used the term 'kernel' - do you mean the MLS averaging kernel? If yes then I am confused by your previous reference to particular layers (or levels). Again please clarify - perhaps the assimilation uses the MLS observations directly via the averaging kernel, but your subsequent analysis is level-based? (This may be a distraction since later - l386 - you seem to say that you do NOT use MLS averaging kernels - in which case it would have been better to make that clear at ~l356.

→ Yes indeed the MOCAGE-VALENTINA system is able to assimilate individual observations together with their associated averaging kernels as it is performed routinely when assimilating nadir-viewing measurements of carbon monoxide from MOPITT (El Amraoui et al., 2014), or ozone from IASI (Emili et al., 2014), etc. The assimilation system simply requires the pair observation and its corresponding averaging kernel. Unfortunately, in the case of MLS observations, only seasonally-averaged and zonally-averaged averaging kernels are provided to the scientific community. There is no individual averaging kernel associated to each individual MLS observation. We have nevertheless performed some tests considering the seasonally- and zonally-averaged averaging kernels. The assimilation test with averaging kernels shows that assimilated water vapour fields were highly unstable (see **Figure R2**). This is the reason why, in the present paper, we present the assimilation of MLS H2O data without considering averaging kernels.

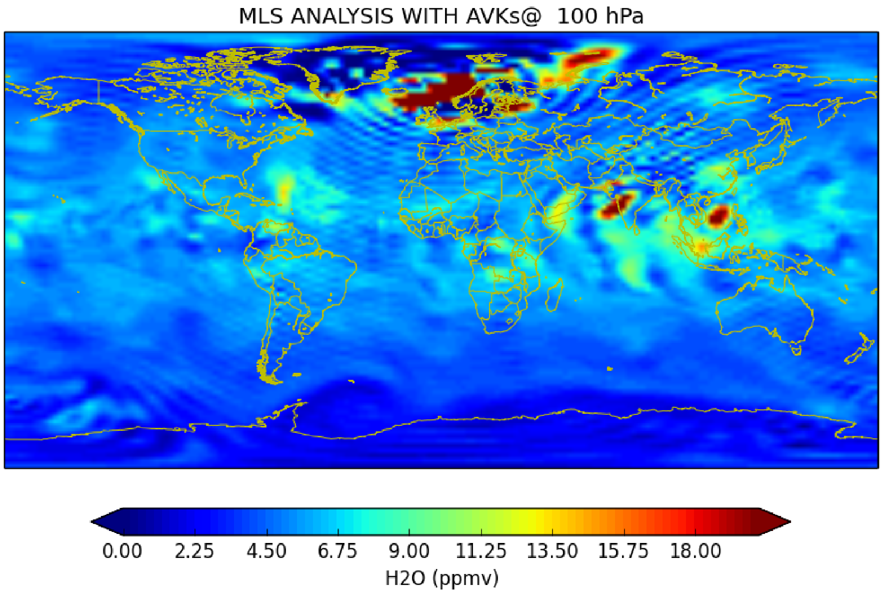


__Figure R2.__ Global-scale distribution of H_2O analyses from MOCAGE-VALENTINA at 100 hPa taking into account the MLS averaging kernels showing instabilities propagating at high latitudes.

El Amraoui, L., Attié, J.-L., Ricaud, P., Lahoz, W. A., Piacentini, A., Peuch, V.-H., Warner, J. X., Abida, R., Barré, J., and Zbinden, R.: Tropospheric CO vertical profiles deduced from total columns using data assimilation: methodology and validation, Atmos. Meas. Tech., 7, 3035-3057, doi:10.5194/amt-7-3035-2014, 2014.

Emili, E., B. Barret, S. Massart, E. Le Flochmoen, A. Piacentini, L. El Amraoui, O. Pannekoucke, and D. Cariolle, Atmos. Chem. Phys., 14, 177-198, doi:10.5194/acp-14-177-2014, 2014.

→ The description of the VALENTINA assimilation system has been entirely rewritten in the section 3 (see the reply to the comment 1) from the reviewer#1). The ambiguity linked to the use or not of averaging kernels is no longer present.

18) l386: As noted above you seem to say here that averaging kernels were not use - i.e. you use MLS estimates of concentrations on particular levels? Please confirm. (Of course, the MLS estimates on the levels in the end come from averaging kernels – do you understand why the use of averaging kernels in the assimilation causes problems?)
→ As stated above, we have not used averaging kernels in our study. The incriminated sentence has been modified into:

> Although the MOCAGE-VALENTINA system is able to take into account the averaging kernels, we have not used the MLS H_2O averaging kernels in our study because we found unrealistic values in some regions of the globe. Our system associates one averaging kernel to one measurement. But in the case of MLS observations, only seasonally- and zonally-averaged averaging kernels are provided and this might produce instabilities.

19) l377: Is this 'simple parametrisation' based on previous experience?
→ Yes indeed, it is based on the work presented in El Amraoui et al. (2014). We modified the sentence into:

> For this study, we used a simple parameterization for the B matrix, consistently with the analysis presented in El Amraoui et al. (2014).

We have inserted a new reference:

El Amraoui, L., Attié, J.-L., Ricaud, P., Lahoz, W. A., Piacentini, A., Peuch, V.-H., Warner, J. X., Abida, R., Barré, J., and Zbinden, R.: Tropospheric CO vertical profiles deduced from total columns using data assimilation: methodology and validation, Atmos. Meas. Tech., 7, 3035-3057, doi:10.5194/amt-7-3035-2014, 2014.

20) l403: To clarify - the 'free run' is (within MOCAGE) simply treating water vapour as a conserved chemical species?
→ Yes indeed. For pressure less than 135 hPa, the model free run field comes from MOCAGE where H_2O is treated as a chemical species. For pressure greater than 135 hPa (as shown on Fig. 3), the model MOCAGE field comes from the ARPEGE model. We have modified the sentence into (see also the comment 20) from Reviewer#2 and the comment 3) from the Reviewer#1):

> On 1 December 2011 at 00:00 UTC, we perform a free model simulation (without assimilating MLS observations) that is initialized by the obtained analysis state.

21) l410: 'Three levels will be studied in detail: 121 hPa (UT), 100 hPa (TP) and 68 hPa (LS).' - this repeats exactly what you have said earlier.
→ We removed this sentence.

22) l419: I'm still unclear on what exactly you mean by 'in the MLS observation space'. Also where does the background profile' in Figure 5 come from? (Indeed where do the background fields displayed in many subsequent Figures come from?)
→ Clarification relative to the terms 'MLS space', 'MIPAS space' and 'model space' has been made on the sentence starting l411:

> Because we used different data sets calculated or measured at different times and locations not necessarily consistent within all the data sets, the analyses will be presented within 3 spaces in time and space coincidences with MLS (hereafter referred to as the MLS space) and MIPAS (the MIPAS space) observations and with the model outputs (the model space).

→ The background field is a component of the assimilation system (see equation 1). We have explained in detail what we call background (see the reply to the comment 1) from the Reviewer#1).

23) l453: This now explains what is meant by 'MLS observation space' etc. This explanation should have come much earlier.
→ We now have explained this term earlier, so we removed the portion of the sentence '(namely in time and space coincidence with MLS observations)'.

24) l483: To be explicit, when you say 'cannot cope with supersaturation' I think you mean 'allows supersaturation, i.e. does not impose any kind of saturation condition'. Clarification would be helpful.
→ We clarified the sentence and used the term "allows unrestricted supersaturation". See also the comment 25) from Reviewer#2.

25) l509: "At 68 hPa (Fig. 12), the background and the MLS analyses (~4 ppmv) are very consistent with the MLS observations (Fig. 1), whilst ARPEGE is much drier (< 2 ppmv) and the Free run is much wetter (> 6 ppmv). The assimilation system behaves nominally in the lower stratosphere since the background is no longer affected by the Free Run even outside of the assimilation window when and where the MLS observations are taken into account." - You have previously (Figures 5 and 6) shown that there is excellent agreement between MLS and MLS analyses. So aren't you simply repeating that point.
→ Yes indeed, you are right. But in that case (section 5.3), our conclusions are drawn in the model space (in time and space coincidence with the model outputs) although, up to the section 5.2, results were mainly presented in the MLS space (in time and space coincidence with MLS observations). In the validation section (section 6), this point will be treated again showing that, in the MIPAS space (in time and space coincidence with MIPAS observations), at 68 hPa in the LS, the assimilation system behaves nominally.

26) l663: “Sensitivity studies show the great improvement on the H2O analyses in the tropical UTLS when assimilating spaceborne measurements of better quality particularly over the convective areas.” - I assume that this refers to the work reported in 7.2 – but in reading that section I didn’t get any clear sense of ‘great improvement’ - only that there were differences in the analyses when different data versions were used. So 7.2 should be clearer on where exactly the improvement is identified.

→ Indeed, the incriminated sentence is probably too short to summarize the sensitivity studies performed in the section 7 that includes the sections 7.1 and 7.2. We have thus rephrased the sentence into :

> Two sensitivity studies are performed. 1) We investigate the impact of some periods with no measurements onto the assimilated fields. We check that the background field tends to be redirected towards the free run, losing the memory of the MLS-driving information injected in the assimilation system whatever the pressure considered. 2) We investigate the impact of using two versions of the MLS data (V3 and V4) on the assimilation fields, V4 showing an improvement in the cloud screening and first guess estimation compared to V3. In the tropical UTLS, the difference between the two analyses is significant, particularly over the convective areas in the upper troposphere/tropopause layer where the presence of clouds is prominent.

27) l971: Figure 7 and subsequent Figures. Why do you have the boxes (which I guess correspond to the geographic regions defined for Figure 6) marked only on the ‘Free Run’ panel? This seems odd when the ‘Free Run’ is not actually a case included in Figure 6.

→ The incriminated Figures (7-12) have been modified by inserting the geographical boxes.