Author comments in reply to the anonymous referee on "Simultaneous assimilation of satellite NO_2, O_3, CO, and HNO_3 data for the analysis of tropospheric chemical composition and emissions" by K. Miyazaki et al.

We want to thank the referee for the positive words, helpful comments and suggestions. We have revised the manuscript according to the comments, and hope that the revised version of the manuscript is now suitable for publication. Below are the referee comments in italics with our replies in normal font.

Reply to Referee #2

It is an regretful omission of the paper, that authors did not discuss the benefit (or drawback) of including the emissions in the state vector for the realism of the assimilated concentration fields itself. It would have been very interesting to present a comparison of an assimilation run without emissions in the state vector (not included in the paper), with the full DA run and the emissions inversion run (both included in the paper). In particular the concentration profiles in the PBL and lower troposphere should be different, if optimised emissions are used.

In response to this request we conducted a new assimilation run without emissions in the state vector (the fixed-emission assimilation run). The results are discussed in Section 5.3 in the revised manuscript.

The following sentences have been added:

Section 3.3:

"In the emission inversion run, only surface emissions are optimized (i.e., without concentrations in the state vector). In the fixed-emission assimilation run, only concentrations are updated from the data assimilation (i.e., without emissions in the state vector). The emission inversion run and the fixed-emission assimilation run have been compared with the full assimilation run in which both the concentration and emission are updated. The comparison allows us to understand the relative importance of the emission optimization and the direct concentration adjustment in the simultaneous assimilation (see Sect.~5.3)."

Section 5.3 (Relative importance of the emission and concentration optimization on the tropospheric ¥chem{O_3} amount analysis):

"Fig. 15 shows the relative importance of the emission inversion and the direct concentration assimilation on the vertical ¥chem{O_3} profiles. The emission inversion largely changes the ¥chem{O_3} profiles in the PBL, especially below 900 hPa. This demonstrates the importance of optimizing ¥chem{O_3} precursors fields in correcting the near surface ¥chem{O_3}. The obvious

impact in the PBL, with a mean difference of up to 15 $¥%$, is found in the tropics and at northern mid-latitudes in July, associated with changes in biomass burning and anthropogenic emissions, respectively. The regional differences are more pronounced over the northern mid-latitudes polluted regions, central Africa, and south America in July, with a maximum difference of 30 $¥%$ (figure not shown). Even in the free troposphere, the ¥chem{O_3} analysis is significantly affected by the emission changes through vertical transport of ¥chem{O_3} and its precursors. However, the direct concentration adjustment dominates the changes in the ¥chem{O_3} profiles in the free troposphere in the combined data assimilation. The simultaneous adjustment of the emissions and the concentrations is thus a powerful approach to optimize the whole tropospheric ¥chem{O_3} profiles. Note that the sum of these two individual effects mostly explains the difference between the full assimilation run and the control run (figure not shown). The ¥chem{O_3} changes estimated from the sum of these two individual effects in the tropical troposphere are an exception to this rule in that they are slightly ($¥sim$ 15 $¥%$) larger than the changes estimated from the full assimilation run. This may indicate too large emission adjustments and resultant ¥chem{O_3} productions in the emission inversion run. The spatial pattern of the changes in the ¥chem{O_3} profiles obtained from emission inversion run and the fixed-emission assimilation run is very different (figure not shown), confirming the independent adjustments realized from the emission and concentration optimizations."

Section 6:

"The emission optimization dominated the changes in the ¥chem{O_3} profiles in the PBL in the tropics and at northern mid-latitudes, whereas the direct concentration adjustment was much more important in the free troposphere. This reveals the importance of the simultaneous assimilation of multiple satellite data sets for the tropospheric ozone budget and profile analyses."

Abstract:

"The simultaneous adjustment of the emissions and concentrations is a powerful approach to correcting the tropospheric ozone budget and profile analyses."

Improving emission data sets by inverse methods (top-down) is a fascinating research topic. One of the important question is whether the optimised emissions compensate for other model errors, e.g. in the chemical mechanism. The authors should have discussed this point in more detail. Do they believe that the optimised emissions from the inversion run or the one from the full DA provide a better estimate of the emissions. The evaluation of an independent model run using the optimised emissions could have clarified this. Further, is the DAS, which is built on the assumption of unbiased model and observations, able to correct emission biases or does it only minimise the variance of the emission error (i.e. by introducing an improved variability)

The impact of the optimized emissions has been discussed in more detail using the emission inversion run and the fixed-emission assimilation run in the revised manuscript (please see my reply to your previous comment above). It is expected that the emissions optimized from the full assimilation run provide a better estimate than those from the emission inversion run because of the reduced errors in the concentrations of various chemical species. However, the emissions estimated by the simultaneous assimilation can still be affected by model errors. To discuss this limitation, the following sentences have been added:

"It is expected that the simultaneous data assimilation provides a better estimate of the emissions than the inversion run because the concentration assimilation may reduce some of the model errors. However, this will not be the case for all model errors. For instance, errors in boundary layer venting or deposition may be compensated in our assimilation system by (incorrectly) changing the emissions."

Use of an independent model run is outside of the scope of this paper.

Please see my reply to your specific comment about the unbiased assumption below.

The paper is very long and could be shortened or re-arranged for better readability. On occasions, one has to go through a lot of text until one reaches the truly interesting pieces of information. The authors should consider if the general description of the considered species in the introduction can be omitted or shortened. The presentation of the observations used for assimilation (2.1) or evaluation (2.2) contains a lot of detail, which is important in general, but can be found elsewhere and is not so much of interest for the focus of the paper. Moving this section into an appendix might improve readability. The presentation of the evaluation of the data assimilation experiments is very long. The reader is easily convinced that CHASER-DAS overall improves the match with independent or assimilated observation. Providing more and more numbers is not needed. The evaluation statistics are already provided in tables.

Based on the reviewer's comment, the manuscript length has been shortened to improve readability. Although moving sections 2.1 and 2.2 into an appendix may improve readability, these sections contain important information for understanding the data assimilation results. I therefore think that the sections should be in the main text. Instead, some sentences, which might be too general or not really relevant to the conclusion of this study, have been removed from the manuscript. The following sentences have been removed:

Section 1:

"Because of its long lifetime in the troposphere, ¥chem{CO} is a~good tracer of the long-range transport of polluted air."

"Oxidation of ¥chem{NO_x} to ¥chem{HNO_3} and aerosol ptake of ¥chem{NO_2}, ¥chem{NO_3}, ¥chem{N_2O_5} dominate ¥chem{NO_x} loss in the troposphere."

Section 2:

"The detailed process of the conversion from simulated ¥chem{NO_2} fields into tropospheric ¥chem{NO_2} columns using the averaging kernel is"

"¥citet{Miyazaki2012} demonstrated that the super-observations help to improve the analysis."

"which was launched in July 2004 into a~polar sun-synchronous orbit with an ascending equator crossing time of 13:40¥,LT,"

" The MOPITT retrieval algorithm is based on the Maximum A~Posteriori (MAP) solution ¥citep{Deeter2010}."

"The vertical sensitivity of the nadir retrieval varies from scan to scan, depending primarily on atmospheric temperature, clouds, and trace gas amounts."

"derived from Model for Ozone and Related Chemical Tracers (MOZART) CTM simulations"

"TES total columns have also been compared with the observations from OMI and MLS ¥citep{Osterman2008}, confirming a~similar high bias both in the troposphere and stratosphere."

"The quality of TES ¥chem{O_3} retrievals in the troposphere has been evaluated using ozonesonde measurements."

"Since the retrieval is based on measurements in the TIR spectral range, the actual vertical resolution of a~retrieved profile depends on the thermal contrast between the surface and the atmosphere."

"Results from MOZART were used to create a~monthly mean climatology used as the a~priori."

"¥chem{CO} mixing ratio profiles are retrieved at nine standard MOPITT pressure levels and at the surface for clear sky measurement."

"Compared to version 4, the version 5 product offers improved long-term stability and more accurately accounts for random errors due to geographical noise ¥citep{Deeter2011}."

"The presence of thick clouds (i.e. ice particles in convective cores) can lead to unphysical values in version 3.3 data."

"The error characteristics of version 3.3 ¥chem{O_3} data may not be significantly different from those of version 2.2 in the UTLS ¥citep{Froidevaux2008}. In contrast, those of ¥chem{HNO_3} in version 3.3 have been greatly improved over those in version 2.2; e.g. a~low bias throughout the stratosphere has been largely eliminated, with a~typical precision (bias uncertainty) of 0.7--1.2¥,¥unit{ppbv} (0.5--1.0¥,¥unit{ppbv}) from 215 to 32¥,¥unit{hPa} ¥citep{Livesey2011}."

"The TES ¥chem{CO} parameters describing the retrieval vertical information in the troposphere are much improved after the optical bench warm up in early December 2005, resulting in better alignment of the instrument and increased signal to noise."

Section 5

"The comparison against the MOPITT ¥chem{CO} shows significant improvements due to the data assimilation."

"The comparison against the lower tropospheric (at 700¥,¥unit{hPa}) TES ¥chem{O_3} fields shows much less improvement, because of the low sensitivity of TES to lower tropospheric ¥chem{O_3}. The TES ¥chem{O_3} sensitivity is reduced greatly in the lower troposphere especially due to the presence of clouds and makes it difficult to improve the analysis."

"The data assimilation also greatly improves the agreement with MLS ¥chem{O_3} data, mostly due to the assimilation of the MLS ¥chem{O_3} data itself."

"a~spatial correlation increase of up to 0.06"

", by about 70--85¥,{¥%} and 40--45¥,{¥%}, respectively"

"The data assimilation also removes the positive model bias (about 70¥,{¥%}) in January."

"The data assimilation improves the spatial correlation ($¥sim$0.04) and the RMSE (by 20--35¥,{¥%}) compared to TOC data obtained from the independent MLS/OMI data."

"The most pronounced corrections in the simulated ¥chem{O_3} is found in the tropics in January and in the Southern Hemisphere in both January and July in the UTLS."

Specific comments:

p 16133 l 17: be more specific about "large" uncertainties, provide references and numbers

Jaegle et al. (2005) and Zhao et al. (2011) have been cited. The following sentence has been added: "¥citet{Zhao2011} estimated the uncertainties of a bottom-up inventory of Chinese anthropogenic NOx emissions to be -13$¥%$¥sim37$¥%$."

p 16133 l 25: provide references

Lamsal et al. (2011) added.

p 16134 l 16: SAGE II and OSIRIS provided early NO2 observations

This sentence is about tropospheric NO2 columns, whereas OSIRIS and SAGE II measured NO2 profiles in the upper troposphere and stratosphere. To make this clearer, "tropospheric NO2 concentrations" has been replaced with "tropospheric NO2 column concentrations".

p 16135 l 15: provide more detail on the differences of CHASER-DAS with other chemical data assimilation systems presented in the literature. This includes the mulit-species vs. uni-variate approaches.

The following sentences have been added:

"However, most recent satellite data assimilation systems optimize either the concentration of a very limited number of chemical species ¥citep[e.g.][]{Mallet2005, Parrington2008, Flemming2009, Elguindi2010} or emissions ¥citep[e.g.][]{Muller2005, Kopacz2010, Hooghiemstra2011}. Only a few advanced studies ¥citep{Hanea2004, Elbern2007} have demonstrated that the simultaneous optimization of multiple chemical states including emissions is an effective way to improve air quality near the surface using surface in-situ observations."

p 16135 l 18: "stiffness" is a numerical property of a system of ODS representing the chemical mechanism. Stiffness is caused by the different time scales and causes numerical instability. It can not be argued that this stiffness "damps out" perturbation or that it the reason that CTM simulations depend not only on initial conditions. The initial conditions are less important because of the boundary conditions (i.e. the fluxes) and because of the chemical equilibrium.

The sentences have been replaced with:

"A large part of the atmospheric chemical system is not sensitive to the initial conditions because of the chemical equilibrium, which is different from the chaotic system involved in the numerical weather prediction ¥citep{Constantinescu2007, Lahoz2007b}, but is sensitive to the model parameters (e.g. emission, chemical reaction rate, and deposition velocity) and processes (e.g. chemical reaction equation, wet and dry deposition, and atmospheric transport)."

p 16135 l 25: please provide reference for the claim that uncertainty of emissions is main source of model error

Mallet and Sportisse (2005) added.

p 16135 l 25: The theory of the KF assumes a bias free model (BLUE). The emissions inventories are not only uncertain in terms of variability but they are "biased", .i.e. the most probably have a constant off-set. Please explain better how a KF system based on BLUE can fix the emission bias.

In CHASER-DAS, the emissions are not changed by the model, and therefore there is no systematic bias. The initial emissions are biased, but this bias is efficiently removed during a short spin-up period. After this period, the changes made to the emissions by the analysis out to zero. In addition, the background error covariance is continuously inflated by the covariance inflation technique during data assimilation. Therefore, the emission bias can be reduced gradually by using the state-augmentation

approach in out system. To explain this, the following sentence has been added to Section 3.3.2:

"Because of the absence of any forecast model (i.e., model bias) to the emissions, and of the use of the background covariance inflation, initial bias in the a priori emissions can be reduced gradually through the data assimilation cycle using the state-augmentation approach, as discussed by ¥citet{Lin2008}."

p 16136 l 15: Please mention other DA systems, which optimise concentrations and model parameters such as Elbern et al. (2007 et al) and Hanea et al. (2004)

Mentioned.

p 16137 l 7: is yo-yb the satellite - analysis difference (?) yb is defined as the model field in observations space

It has been replaced with "satellite-model difference"

p 16137 l 13: "for a large part" - please explain or omit

Omitted.

p 16137 l 17: consider moving section 2.1. and 2.2 into an appendix

Please see my reply to your general comment above.

p 16138 l 18: (=2.8 degree), please provide resolution in km, omit "="

"about 300 km in the equator" added.
"=" omitted

p 16140 l 6: MOPITT also measures in the 2.2-2.4 microm interval

Added.

p 16143 l 25: "both peak" is unclear - does this mean they have a maximum ?

Yes. The sentence has been replaced with "The TES and MOPITT retrievals both have a maximum

sensitivity mainly from 300 to 800¥,¥unit{hPa}”

p 16146 l 15: be precise, convective cloud top height is the parameter used in this parameterization

“deep convection” has been replaced with “convective cloud top height.”

p 16147 l 3: Please summarise briefly differences between LETKF and the more common EnKF.

The following sentence has been added:
“Because of the large state vector size and the large number of grid cells in a global CTM, the computational advantages of the LETKF over the original EnKF is important for global tropospheric chemistry data assimilation.”

p 16147 l 8: Please explain how the ensemble X is obtained (variation of which model parameter)

This is explained in Section 3.3.1.

p 16147 l 16: Please explain how Pa is projected in time (this is the key feature of any Kalman Filter), Please explain how the model error is taken into account, and if not how deflation of Pa is avoided.

The following sentences have been added:
“The background error covariance ($¥mathbf{P}^{b}=¥mathbf{X}^{b}(¥mathbf{X}^{b})^T$) tends to underestimate the true background error covariance because of model errors and sampling errors ¥citep{Houtekamer1998}. To prevent the covariance underestimation, the covariance inflation technique (with a covariance inflation parameter of 5 $¥%$) is applied at each analysis step, as in ¥citet{Miyazaki2012}.”
“An ensemble simulation with the new analysis ensemble is then used to predict the background error covariance $¥mathbf{X}^{{b}}$ in the next forecast step.”

Further details are described in Hunt et al. (2007) and Miyazaki et al. (2012), as noted in the manuscript.

p 16148 l 11: explain choice of period and its length

The following sentences have been added:
“The March 2006 experiment was used to validate against the INTEX-B aircraft data, while the January

and July 2007 experiments were used to compare the seasonal difference in the data assimilation performance."

p 16148 l22: see comment on stiffness above

Corrected.

p 16149 l 11: Does cib include all species including the very short-lived like OH?

No. Cib includes only the predicted (transported) chemical species. Short-lived species like OH are not included in the state vector. The following sentences have been added to explain this.

"…and the concentrations of all the predicted (i.e., transported, total 35) chemical species, ${c}$, are optimized at all the models grid cells for each data assimilation cycle. The concentrations of radical and members of family species are not included in the state vector. The data assimilation influences their concentrations through the chemical coupling during the forecast."

p 16149 l 19: It is not clear how the localisation, i.e. the suppression of certain covariance values, was derived from the sensitivity experiment. It seems more a practical but ad-hoc choice. Please clarify, and provide more detail. The choice of this localisation might be useful for other chemical DAS.

The following sentences have been added:

"The optimization of the variable localization was based on a comparison against satellite data. If the data assimilation significantly deteriorated the agreement with at least one of the data used for the data assimilation and the validation, variable localization was applied to reduce the deterioration by considering dominant chemical processes, as will be further described in Section 4.2. "

p 16150 l 10 ff: this paragraph would be better placed in section 3.3

Moved.

p 16150 l 27: please mention that the covariance deflate because no model error is assumed.

The previous sentence has been rewritten as:

"However, because a~model error term is not implemented during the forecast step, the background error covariance can be continuously deflated and underestimated during the data assimilation."

p 16151 l 5: Was the error specification of the emissions constrained in any other way apart from the initial error value? Were large day-to-day changes damped (by using a "red noise" error model) Were points with low emissions (say over the oceans) restricted to maintain low emissions by the DA system.

In CHASER-DAS, the a posteriori emission error (i.e., the analysis ensemble perturbation) is measured by the LETKF at every analysis step, given the background error covariance and the observations. The analysis spread can be very small in some cases owing to effective corrections by the data assimilation and owing to the absence of any forecast model. To prevent covariance underestimation in our system, the analyzed standard deviation (i.e. background error) is artificially inflated back to a minimum predefined value at each analysis step in our system, as described in the manuscript. As a result of the data assimilation and the covariance inflation, the analysis spread can no longer be strongly influenced by the initial error setting after some assimilation cycles. Because of the inflation applied to the emissions and the daily observations (e.g., by OMI), the data assimilation represent short-term (i.e., day-to-day) variations of the emissions. The data assimilation is also able to increase the emissions gradually, even over the oceans, because of the inflation applied at each analysis step, if there are enough large systematic differences between the model and observations.

To explain this, the following sentences have been added or rewritten as follows:

Section 3.3.2:

"Although the optimized emissions (i.e., the analysis mean) and the uncertainty (i.e., the analysis spread) are not strongly sensitive to the choice of the initial error after some assimilation cycles (e.g. several weeks) because of the analysis applied for both the mean and spread fields and the inflation, convergence is generally attained faster in the case for larger initial uncertainties."

Section 5.2.1:

"As a result of the data assimilation and the covariance inflation, the mean a posteriori error for the surface ¥chem{NO_x} emissions typically ranges from 12 to 60 $¥%$, with smaller relative errors over polluted areas than over clean areas. The mean differences between the a priori and the a posteriori emissions are generally larger than both the a posteriori error and the variability (i.e., standard deviation) of the a posteriori emissions estimated during the analysis period."

p 16152 l 9: The term "localisation length" is not clear - is it the horizontal correlation length ? Is there an assumption about the vertical correlation length?

The term is the horizontal localization length, which is noted in the revised manuscript.

The following sentence has been added:

"The physical vertical localization length was set to $ln P$ [hPa] $=$ 0.2 based on sensitivity experiments (results not shown)."

p. 16153 l 9: Figure 3 is a very interesting figure. It is not clear how the "global mean" was constructed. Is it the spatial means of the covariance for each grid point or is the covariance of the global mean state vector. Is there a distinction between day and night grid points?

The global mean is the spatial means of the covariance for each grid point. The following sentence has been added to the figure caption:

"The global mean of the covariance estimated for each grid point is plotted."

Both day and night grid points were used in the analysis.

p 161f3 l9: Why does Figure 3 not contain OH - this would have been very interesting as well.

The background error covariance shown in Figure 3 does not contain OH (non-transported species), because the state vector used contains only the predicted (transported) chemical species. This is clearly described in the revised manuscript.

p 16153 l 10 ff: This discussion is interesting but should not only focus on the positive correlations. Because of the chemical nature there are also many negative correlations between the species (as many as positive) These should also be discussed. For instance the strong negative correlation between Ox and ethylene should be explained.

The following sentences have been added:

"Negative correlations are also found between reactive species. For instance, $r=$-0.63 between ¥chem{O_x} and ¥chem{C_2H_4} (ethene) at the surface results from the removal of ¥chem{C_2H_4} as a result of the fast reaction with ¥chem{OH} and ¥chem{O_3} ¥citep{Sawada1986}."

p 16154 l 9 - 15: Where is this statement backed up - is there a figure available?

The statement was based on OSE plots (not shown in the manuscript, since the plots do not add unique or important information). To avoid confusion for readers, the sentences have been shortened and rewritten as follows:

"The OSEs confirm that the assimilation of each species data set has a~strong influence on both assimilated and non-assimilated species through the use of the inter-species error correlation and through

the chemical coupling provided by the model forecast. The assimilation of OMI ¥chem{NO_2} data provides some changes in ¥chem{O_3} and ¥chem{CO} concentrations, whereas the assimilation of TES ¥chem{O_3} data has some effects on ¥chem{NO_2} fields, as will be shown in Sect.~5.1.2."

p 16156 l 25: chi-square tests are a large class of methods. The chi-square diagnostics by Menard and Chang (2000) is motivated by chi-square testing.

The sentence has been rewritten as "A~quantitative criterion for the choice of the background error is a chi-square ($¥chi^2$) test, the $¥chi^2$ diagnostics ¥citep[e.g.][]{Menard2000}."

p 16164 l 15 - 25: I do find this reasoning a bit far fetched. It remains speculative and assumes a perfect representation of the chemistry by the model. The assimilation of MLS HNO3 is clearly not efficient enough to improve HNO3 overestimation in the lower and mid troposphere (Figure 13). I do not see that adding more NOx (more lightning or more convection?) will help to reduce the positive OH bias.

It might be true that insufficient model performance, along with insufficient observational constraint, obstruct improvement of various chemical fields. To note this, the following sentence has been added:
"There are many other factors in the chemical transport processes affecting the overall model performance. They may obstruct further improvements by the data assimilation."

p 16166 l 1-5: The emissions are part of the state vector. If I understand the DA method correctly, the optimised emissions are available for each analysis time step and each surface grid point. It would therefore be interesting not only to discuss the bias but also the temporal and spatial variability of the optimized emissions. Is this variability, w.r.t. to time and space, large compared to the bias? Is there a temporal trend of the emissions within the time frame? By including the emissions in the state vector, they can simple vary much more than the emissions from the static inventories.

The emissions are optimized for each analytics time step and each grid point, and they vary with time and space. The temporal variability has been evaluated from the standard deviations of the a posteriori emissions during the analysis period, as shown in Tables 6, 7, and 8 and as discussed in Section 5.2 in the revised manuscript. The standard deviation is generally smaller than the bias.

The table captions have been rewritten as follows:
"The 15-day means (from the 16--30 of each month) and the standard deviations (from the means) of the global and regional surface ¥chem{NO_x} emissions obtained from the a~priori emissions and the a~posteriori emissions."

The following sentence has been added:

"The mean differences between the a priori and the a posteriori emissions are generally larger than both the a posteriori error and the variability (i.e., standard deviation) of the a posteriori emissions estimated during the analysis period."

There is sometimes a trend-like variation in the a posteriori emissions, probably due to the seasonal migration of the emissions. However, the analysis period is not long enough to detect such a trend.

p 16168 l3: provide reference

Hooghiemstra et al. (2011) added.

p 16168 l15: Table 8 shows much lower numbers for new inventories despite increased emission inversion results for Europe. Please discuss this further.

The following sentence has been added in response to this comment:

"The newer inventories show lower emission values than the a priori emissions over Europe, whereas the data assimilation further increases the emissions from the a priori emissions. In contrast, the a posteriori emissions are significantly larger than both the a priori emission and the newer inventories over Eastern China. Over Eastern United States, the data assimilation decreases the emissions; however, the newer inventories show even lower emissions. These results imply different error characteristics in the different bottom-emission inventories."

p 16170 l 15: The low CO bias might also be caused by an overestimation of OH. Increasing emissions alone seems not the entire solution.

(p. 16168?)

The sentences have been rewritten as follows:

"The underestimated ¥chem{CO} fields might be mostly attributed to an underestimation of the surface ¥chem{CO} emissions along with an overestimation of ¥chem{OH}. Correspondingly, the assimilation of MOPITT data largely increases the surface ¥chem{CO} emissions in the northern extratropics both in January ($+$66¥,{¥%}) and July ($+$25¥,{¥%})."

p 16173 l25: please also mention the deterioration of some of the fields by DA in the lower troposphere (see Figure 13)

The sentence has been rewritten as follow:

“The results confirm that the assimilated satellite data have highly valuable information about the tropospheric chemical processes, although further improvements are required for the lower tropospheric processes.”

p 16174 l 16: Please discuss the benefit and drawbacks of (i) emission inversions, (ii) concentration field DA and (iii) combined concentration field emission DA

The last part of the paragraph has been rewritten as follows:

*“*As a~result of the simultaneous optimization, the tropospheric ¥chem{O_3} burden is increased by 5.6¥,{¥%} in July, with almost equal contributions from the emission optimization and the direct adjustment to the concentration fields. The emission optimization dominated the changes in the ¥chem{O_3} profiles in the PBL in the tropics and at northern mid-latitudes, whereas the direct concentration adjustment was much more important in the free troposphere. This reveals the importance of the simultaneous assimilation of multiple satellite data sets for the tropospheric ozone budget and profile analyses.”

p 16174 l 20: “somewhat different” - please be more specific

“somewhat” deleted. The specific description is given in the next sentence.

p 16175 l12: What are the conclusion of the paper w.r.t to model improvements - just changing emissions?

This paragraph describes that the analysis increment information obtained from the simultaneous data assimilation is valuable for improving the model performance in future studies. Further research is needed to improve the model performance using the data assimilation results.

This assumes that emissions are the only source of the model error, which is perhaps not true.

The emissions are not the only model error source, as discussed in the manuscript. The analysis spread estimated reflects not only the emission uncertainty, but also errors in the model input data (differences in the observed and forecasted concentrations) and errors in the measurements assimilated in the simultaneous data assimilation system, as described in the manuscript.

The optimised emission from the full assimilation run and the emission inversion run differ - why.

This has been already discussed in Section 5.2 and described in the concluding section.

Is a model run with the optimised emissions as good as the full DA run ?

No. The direct concentration adjustment by the data assimilation also plays important roles in improving the various chemical states in the simultaneous data assimilation. This is described in Section 5.3 and the concluding section in the revised manuscript.

p 16204 figure 2: clarify 260 hPa / 220 hPa

The following sentences have been added:
"Assimilation of MLS ¥chem{O_3} and ¥chem{HNO_3} data affects the concentrations only above 260 and 220 hPa, respectively."

p 16205 , figure 3: Mention averaging period, please clarify Ox, why is OH not included.

The averaging period has already been mentioned (15-20 July 2007). The background error covariance contains the concentration of only the predicted (transported) species. This is explained clearly in the revised paper. The following sentence has been added to the figure caption.
"¥chem{O_x} is the sum of ¥chem{O_3} and ¥chem{O(^{1}D)}, and ¥chem{NO_x} is the sum of ¥chem{NO}, ¥chem{NO_2}, and ¥chem{NO_3}."

p 16197: table 4: explain localisation length

Explained.

p 16201, Table 8: are the lower values for newer emissions for Europe and E-USA correct?

Yes. The larger a priori emissions might be related to the fact that the 1995 and 2000 emission inventories are extrapolated to create the a prior emissions for the simulation years 2006–2007. This procedure may give spurious results for certain regions, as mentioned in the manuscript.

p 16216, figure 14: Why are there no lightning emissions over the oceans?

There are lightning emissions over the oceans. However, the emission value is much weaker over the oceans than over land, and the monthly mean value is generally below the lowest colour scale over the oceans in this figure.