# **Review of Huijnen et al.: Quantifying uncertainties due to chemistry modeling - evaluation of tropospheric composition simulations in the CAMS model.**

The manuscript reports an evaluation of tropospheric reactive gases, including ozone and precursors, in three chemistry versions of the IFS: CB05BASCOE (CBA), MOZART (MOZ) and MOCAGE (MOC). The three configurations use the same meteorology, emissions, transport and deposition schemes. Therefore, the differences in the model results should be basically due to differences in the chemical schemes, namely gas phase chemistry, heterogeneous chemistry, photolysis and chemical solver.

Overall, the manuscript is well-written, the presentation quality is good and the authors give credit to related work. As motivated in the introduction, this work documents the diversity in global model simulations caused by chemistry, unlike many other papers which focus on the impact of emissions or other model parameterizations. This is a useful exercise that falls very well within the scope of the journal.

My only concern is that I miss some in depth discussions about the potential reasons for the differences in the performance of the three model configurations considering what is different in them (i.e. the chemistry). The "Main comments" below include some suggestions on how to improve this. These are not really major comments. I hope that some of them are useful, but I am aware of the complexity involved in comparing different chemistry schemes. There are also some minor and technical corrections which need attention. I will fully support the publication of the manuscript in GMD once these issues have been addressed.

### Main comments

(1) Section 2.1.4 (Key differences in chemistry modules, including Table 1) provides a relatively detailed overview of the differences in the three chemical mechanisms. All this information is very valuable but not completely helpful. A simple summary comparing the complexity of the different processes in the three configurations could be useful to get a broader picture of the main differences. For instance:

1.1. Complexity of organic chemistry: MOCAGE (more extended lumping) ~ MOZART (more explicit) > CB05BASCOE

1.2. Complexity of tropospheric inorganic chemistry: MOCAGE (e.g. includes HONO) > MOZART ~ CB05BASCOE

1.3. Complexity of tropospheric heterogeneous chemistry (HO2 & N2O5): CB05BASCOE ~ MOZART > MOCAGE.

Table 1 indicates that MOCAGE has as many heterogeneous reactions as the other schemes, but that information is not very helpful. I assume those reactions are only relevant for the conversion of SO2 to H2SO4 in the troposphere (lines 139-140) and for stratospheric chemistry, because on lines 179-180 the authors indicate that "Heterogeneous reactions of HO2 and N2O5 on aerosol ... not in the IFS(MOCAGE) version considered here".

1.4. Is there a two-way coupling between MOZART / CB05BASCOE oxidants and CAMS aerosol fields? In other words, are the oxidants passed to the aerosol module to produce aerosols, depleted in that processes and then passed back to the chemistry schemes? Or does the coupling only consist of using CAMS aerosols for heterogeneous chemistry in the two chemistry modules? This information is not clear in the main text and is relevant e.g. to evaluate differences in NOx and HNO3.

1.5. Complexity of photolysis: Is it more complex in CB05BASCOE because it includes the impact of aerosols? Anyway, if there is no easy way to compare complexity of photolysis among the three configurations one can leave this out.

1.6 Complexity of stratospheric chemistry: CB05BASCOE > MOZART ~ MOCAGE

1.7 Chemical solver: CB05BASCOE uses Rosenbrock (differences in NOy chemistry and up to ~20% less O3 regionally). It would be interesting to know if the "differences in the N2O5 chemistry" indicated by the authors (line 199) often result in smaller amounts of the species evaluated here (NO2 and HNO3). If not clear, please leave this out.

Summarising, although I understand how hard it is to compare different chemistry schemes, a new table or some bullet points similar to those indicated above could be used to better see the main differences in the three model configurations. Afterwards, one could elaborate on the potential implications for the representation of the main tracers evaluated here (see next comment).

(2) Some additional comments in different parts of the text could be helpful to try to understand the model differences. As mentioned above, I do not expect the authors to be able to explain absolutely everything, but hope that some of these comments will be useful and easy to include in the text:

2.1 Section 4 (Assessment of inter-model differences). The comparison of NOx annual mixing ratios (Figure 2, lines 330-333) could easily be improved:

"MOZ and CBA are overall similar, but MOC is showing higher values over the NH high-latitudes (>60°N) and also at altitudes below 900 hPa in the tropics". First, a minor correction: I clearly see differences in the lower to mid-troposphere from the tropics to the high latitudes, i.e. not only for latitudes >60°N and below 900 hPa in the tropics.

"This is likely related to the fact that in this version of IFS(MOCAGE) the coupling with the aerosol module has not yet been established, contrary to CBA and MOZ". Do you mean that the lack of heterogeneous hydrolysis of N2O5 in MOC will probably raise the NOx concentrations (because that reaction produces the reservoir species HNO3, which is efficiently removed from the atmosphere by dry and wet deposition, resulting in a removal of NOx)? If so, please clearly indicate this. Otherwise, you should explain which other processes might be involved. For instance, is there aerosol nitrate production, removing NOy from the gas phase, in CBA and MOZ?

2.2 The last paragraph of Section 4 (Assessment of inter-model differences) tries to relate the differences in NOx-O3-OH in the models. Overall, the authors point to faster chemical production of O3 in MOCAGE, counterbalanced by fast loss by reaction with OH and HO2. They also suggest a relatively short lifetime of O3 in MOCAGE and conclude the paragraph with "Such differences in oxidation capacity naturally have important implications for understanding differences in the performance of NMHCs, as discussed in the next sections". A couple of questions:

\* The authors are probably right, but does the C-IFS have any diagnostics that can be used to quantitatively compare budgets/lifetimes of O3 due to different processes in the three configurations? That would be useful.

\* That paragraph should also include a comment for CO. A few lines above (lines 326-328), the authors compare CO in the three configurations (CBA >> MOZ > MOC), "suggesting mainly differences in oxidizing capacity". I think they could be more specific. I am aware that there is no straightforward comparison of the CO and OH levels in the models, because different VOC degradation schemes will impact the CO mixing ratios (as correctly indicated on first paragraph of Section 5.2). Nevertheless, the authors could relate the higher (lower) CO levels in CBA (MOC) to the lower (higher) OH levels seen for the same models in Figure 2. I will come back to this below (comment on Section 5.2).

2.3. Section 5.1 provides a detailed evaluation of ozone. However, after so many comparisons, it is a bit hard to conclude what we have learnt. A summary of the model performance, even if the reasons for the model differences need to be better investigated, might help.

For instance, I know there are some specific comments for MOZ in the first paragraph of the section (e.g. "MOZ simulates too high O3 concentrations ... Here it is worth mentioning that recent updates to reaction probabilities and aerosol radius assumptions in the heterogeneous chemistry module in IFS(MOZART) significantly improved O3 concentrations particularly in the NH"), but a summary mentioning that at the end would be useful.

Some conclusions might be that modelled ozone is often within the uncertainty of the selected sondes (Figure 6), but that MOZ tends to yield higher O3 levels everywhere in the mid-to-upper troposphere (500-300 hPa) and everywhere in the northern hemisphere (Figure 5). Somewhere one should also mention that this configuration yields the highest biases (15.9 ppb), but at the same time the best weighted correlations, when compared to aircraft data (Table 4). Then, one could say again that the biases have partly been corrected in recent versions with improved heterogeneous chemistry.

Finally, the authors could try to repeat Figure 5 but showing accumulated errors (e.g. RMSE) instead of biases. If that provides any further information to document the performance of the three configurations then it should be included.

### 2.4. Section 5.2 (Carbon monoxide).

The authors include a useful introduction about all the processes impacting CO levels (emissions and VOC degradation) and then comment several plots comparing models with different datasets (MOPITT, surface, aircraft profiles). Overall, I am rather happy with this section, but again it would be nice to finish it with a paragraph summarising what we have learnt from these comparisons.

For instance, from Figures 7-9 (MOPITT & surface CO) one can see that CBA is the configuration with the highest CO levels, yielding the best comparisons (lowest negative biases) in the northern hemisphere but positive biases in the southern hemisphere (Figure 9). The authors should mention that this may partly be related to the fact that this scheme is the one with the lowest OH levels (Figure 2). I would say this is one of the main outcomes from these comparisons.

Towards the end of this section the authors could indicate whether modelled CO is within a given range for the three schemes (they mention 10% only for the comparisons with aircraft observations, but in the abstract and conclusions they indicate 20%). That range (expressed as a percentage or ppb) could be compared to the overall model error for this species (when compared e.g. with surface observations) to get an idea about the fraction of that error that could be attributed to differences in the chemistry. Here or in the "Conclusions" section one could even point to the need of producing some model diagnostics in the future to understand to what extent the differences in the atmospheric mixing ratios of CO arise from differences in CO degradation (by reaction with OH) or differences in CO production (from VOC degradation) in the chemistry schemes.

2.5. Section 5.3 (formaldehyde). It is nice that the authors use weighted biases to show that the simulations are within the range of uncertainty of the few observations available. The correlations are also quite decent. However, this section is rather short and could probably benefit from some improvements and/or discussions.

Lines 506-508 say: "Formaldehyde is important as the most ubiquitous carbonyl compounds in the atmosphere (Fortems-Cheiney et al., 2012). It is mainly formed through the oxidation of methane, isoprene and other VOC's such as methanol (Jacob et al., 2005), while its oxidation and photolysis is responsible for about half of the source of CO in the atmosphere".

First, a couple of minor comments: You should mention that HCHO is not only secondarily formed but also directly emitted. In the first sentence, do you mean "one the most ubiquitous carbonyl compounds"?

I am not sure that the end of the second sentence is right. There the authors mention that the degradation of HCHO is half the source of CO in the atmosphere. Somewhere else (lines 448-449) they mention that "Approximately half of the CO burden is directly emitted, and the rest formed through degradation of methane and other VOCs". Are the authors implying that most of the CO formed (not emitted) in the atmosphere comes from HCHO? Isn't that too much? Before making that statement, the authors should check some references or compare the production terms of CO in the chemistry schemes. If HCHO is so important in terms of CO production, could one try to relate the model performance for both species? For instance, it is possible to relate the fact that CBA is the simulation with the highest CO mixing ratios (which I previously attributed to the low OH in that model) to the negative HCHO biases found for the same scheme in this section?

### 2.6. Section 5.4 (Ethene, C2H4).

The authors have mentioned the main sources of this species and have provided some insights into the reasons for the model performance. Most of this section looks good to me, but I have some comments:

\* The last paragraph says: "Furthermore, interesting is the comparatively large difference present between the simulations at high latitudes (e.g. SONEX, Newfoundland), where the largest relative differences in modelled OH have been found, (see also Sec. 4), illustrating the importance of OH for explaining inter-model differences". Could one explicitly mention that the higher C2H4 mixing ratios in CBA (red in Figure 13, right) compared to the other two schemes are probably related to the lower OH levels in the same scheme?

On the other hand, I am surprised that CBA is the scheme with the largest negative biases for C2H4 in Table 4 (-6.28 ppt). It does not look like that from Figure 13. Is that because the figure only shows data for some specific campaigns and locations? If so, my previous comment might not be valid.

\* Lines 531-532: "As the underestimation appears to be ubiquitously distributed this suggests that C2H4 decomposition is too strong, or that the model versions miss chemical production term (e.g., Sander et al., 2018)". This conclusion looks fine, but they should change "miss chemical production term" to "miss some chemical production terms".

Somewhere in this section it would be worthwhile to briefly mention (without all the explanations given below) that one should not expect to get this species right because model and observations are not really collocated in time. I understand that model data are monthly averages (considering both daytime and night-time data) while observations were probably collected at daylight. In addition, model and aircraft observations are from different years. All this is relevant if we keep in mind all the processes affecting the mixing ratios of this species. Many of those processes have already been mentioned by the authors: terrestrial biogenic emissions (probably with strong diurnal cycle), which should be much larger in Newfoundland (Figure 13 right, high C2H4 mixing ratios) than in Christmas Islands and Tahiti (oceanic environment, Figure 13 left and centre, low C2H4); diurnal cycle of anthropogenic emissions (again source probably much higher in Newfoundland); biomass burning sources (with strong interannual variability); upward lifting and subsequent horizontal transport of plumes (not sure if seen on observations from Figure 13 left for Christmas Islands), and relatively short lifetime, mainly controlled by its reaction with OH (higher levels at daylight than at night-time).

### 2.7 Section 5.6 (Nitrogen dioxide, NO2).

Overall, this section is fine too, but I miss more detailed explanations and some integration with the rest of the text towards the end of the section. In addition, providing maps with biases would be helpful to see the differences between the model configurations and OMI (this is also indicated in a minor comment).

For instance, around lines 582-586, the authors mention the lower NO2 levels for CBA than for the other two models, over polluted and clean regions in the northern hemisphere. Then, around the same lines they say: "This all suggests a relatively short NOx lifetime in CBA compared to MOZ and MOC, which in turn helps to explain the lower O3 over the NH-mid latitude regions as modelled with CBA (see Figure 5)". A couple of comments on this:

\* In Section 2.1.4 the authors mentioned some implications of the use of the Rosenbrock solver (in CBA) for NOx and O3, but I think that is hardly mentioned again in the rest of the text. Shouldn't one mention it here?

\* CBA has less NOx, O3 and OH, but more CO, than the other model configurations. Not sure if all these facts are related. Shouldn't this be mentioned somewhere?

### 2.8. HNO3 (also related to NOx).

There is a good introduction relevant to HNO3 around lines 601-604: "It has a very high solubility and therefore tends to be scavenged by precipitation very efficiently, providing an effective sink for the NOx family. Furthermore, it can act as a precursor for nitrate aerosols (Bian et al., 2017). HNO3 concentrations are therefore expected to show amongst the largest variation between the simulations, as the production and the sink terms can largely differ due to uncertainties in the parameterizations". However, later on, the authors are not very specific about the potential reasons for the model differences they find. They just add some general sentences like these ones:

Lines 612-613 (for HNO3): "The discrepancies between the model versions can be mainly attributed to differences in NOx lifetimes and nitrate aerosol formation"

Lines 653-655 (in the conclusions): "For instance IFS(MOCAGE) tends to predict significantly higher NOx and HNO3 concentrations in the lower troposphere compared to the other two chemistry versions".

Can one relate some of the model differences to the lack of coupling of MOCAGE chemistry with the aerosol module? I assume this is not simple. For instance, the inclusion of heterogeneous loss of N2O5 in CBA and MOZ should remove NOx from the system but form HNO3, which would not explain the high HNO3 in MOC. On the other hand, if there is a two-way coupling between the chemistry and the aerosol modules, CBA and MOZ will provide oxidants (e.g. HNO3) to the aerosol model to form aerosols (e.g. nitrate) and the oxidants will be depleted in that process. This could counteract the production of HNO3 by heterogeneous reactions. Are all these processes taking place in CBA and MOZ but not in MOC?

And is wet deposition done exactly in the same way for the three configurations? Otherwise, it should have an impact on the HNO3 mixing ratios.

#### Minor comments

\* line 22: "compared against a range of aircraft field campaigns, ozone sondes and satellite observations". Surface observations should also be mentioned here (even if only used for CO).

\* Lines 25-26: "Most of the divergence in the magnitude of NMHCs can be explained by differences in OH concentrations". Why only NMHCs? Shouldn't one mention CO too?

\* Chemical names, formulae and abbreviations are not used in a consistent way throughout the text. There are some examples here, but this list is not exhaustive at all (the authors should revise the whole text):

"PAN" should be changed to "peroxyacetyl nitrate (PAN)" on line 135.

"polar stratospheric clouds (PSC)" needs to be mentioned only once (for B05BASCOE). In the description of the other two configurations the authors can simply use "PSC".

It should be "VOCs" and not "VOC's". The same applies to "NMHC's" and "CFC's". Please, also spell out VOC and CFC.

In particular, special attention is required in Section 2.2 (Emissions). There the authors use capitals for names such as "Olefins", "Aldehydes", "Butenes" or "Parafins", but that is not needed.

The names "methane" and "CH4" are used in different parts of the text, but the authors never indicate "methane (CH4)". All names of chemical species should be given together with their formulae the first time they appear in the text.

They authors use the name "acetylene", while the IUPAC name (ethyne, C2H2) might be more appropriate.

When possible they should indicate both the name and the formula for all the species in Table 2 (otherwise, please give both name and formula in the main text). There should be a reference to this table from the very beginning of section 2.2.

\* Section 2.2. Please indicate whether the non-anthropogenic emissions (e.g. biogenic VOCs and NO, NO from lightning) are prescribed or interactive.

\* The authors have indicated the experiment IDs in Table 3. This and other information they provide will be very useful to ensure traceability. I have also noted that in the "Data Availability" section they say: "The model simulation data as used in this work can be obtained upon request from the corresponding author". Are the model experiments from Table 3 stored on ECMWF's servers and accessible to other users? If that is the case, the data availability section could be rewritten.

\* In Section 2.3 (Model configuration and meteorology) the authors mention that "the model was sampled ... every three hours. These are used to compute monthly to yearly averages. Standard deviations are computed to represent the model variability for a specified range in time and space". This is fine but, after reading section 3 (Observational datasets), it is not clear how the model output and observations are sampled for many of the comparisons shown in the text. I assume that the data are always collocated in space but not completely collocated in time. For instance, aircraft observations are from different years. Please indicate for all datasets whether they are for the simulated year (i.e. 2011) or for another period. Then indicate whether all comparisons are made using model and observations from the same month (even if the data are from different years).

\* Section 3.1 (Aircraft measurements). The authors used the compilation of Emmons et al. (2000) as well as TOPSE and TRACE-P data. If it is not too complicated, they could also indicate the names of all the campaigns used, the period covered and so on. In Figure 1 they indicate that "Each field campaign is represented by a different colour" but they do not mention the names. Later, some of those names appear on the profile plots (Figure 11, 12, ...). I simply think it would be helpful to know the geographical location / time of the year for the comparisons between model and observation. This would also be great to ensure traceability.

\* Section 3.2 (In situ observations). I wonder myself whether "Near-surface CO and ozonesondes" could be a more appropriate name, because I assume that most of the aircraft measurements in the previous section will also be in situ.

\* Table 4 (summary of bias and correlation coefficients of models vs. aircraft observations) is a bit confusing:

The authors first mention correlation coefficient (R) but then they indicate R-squared both in the caption and the header. Please be consistent.

"Bias and R2a is in pmol/mol, (except for CO and O3)". R or R2 cannot have units. In addition, it might be simpler to write "Bias in pmol/mol (nmol/mol for CO and O3)" and remove the note below the table.

The authors show methyl hydroperoxide (CH3OOH) in the table because there are some aircraft observations, but this species is hardly discussed in the text. Why is that? I think there is only one comment about this species around lines 372-374 ("... while model versions have more difficulties with CH3OOH"), but it is a little bit out of place there. The negative biases can be high for this species but the correlations are fine.

The concept of weighted biases and correlations has been introduced on page 15, but shouldn't the weighted vs. non-weighted values be discussed in some parts of the text? For instance, in the case of C2H4, the three simulations have similar values of R (0.54-0.58) but rather different values for weighted R (0.03-0.39); why is that? In other cases, such as for CO and C2H6, the values of weighted R are higher than those of R. So the weighting can improve or deteriorate the correlations. Summarising, it might be good to explain why the values of R sometimes change so much, not always in the same direction, after weighting.

\* Around lines 395-397 the authors say "MOZ simulates too high O3 concentrations ... Here it is worth mentioning that recent updates to reaction probabilities and aerosol radius assumptions in the heterogeneous chemistry module in IFS(MOZART) significantly improved O3 concentrations particularly in the NH".

It might be worth to mention whether the assumptions made in the heterogeneous chemistry are very different in MOZART and CB05BASCOE. Are the uptake coefficients very different in both configurations? Are they constant or parameterized? And which are those aerosol radius assumptions, considering that both configurations use the same CAM aerosol fields?

By the way, the changes mentioned for O3 in the NH, if they are partly consequence of updates in the heterogeneous uptake of N2O5, might have some seasonality and not be so relevant all year round (see e.g. Tie et al., 2003, which used a very simple parameterization of that process). Or do the "recent updates to reaction probabilities" refer to reactions in the gas phase?

Tie, X., L. Emmons, L. Horowitz, G. Brasseur, B. Ridley, E. Atlas, C. Stround, P. Hess, A. Klonecki, S. Madronich, R. Talbot, and J. Dibb: Effect of sulfate aerosol ontropospheric NOx and ozone budgets: Model simulations and TOPSE evidence, J. Geophys. Res., 108(D4), 8364, doi:10.1029/2001JD001508, 2003.

\* Figure 4 and Lines 405-406 (comparison with ozonesondes): "However, it should be noted that in the SH regions this evaluation is less representative because there are very few observations".

I see that the number of sites within the different latitude ranges is shown later (Figure 5), but some additional information about the geographical data coverage for this and other datasets would be useful (e.g. in Section 3, Observational datasets). The authors should also indicate if the ozonesonde dataset is for 2011 or corresponds to the climatology for a longer period. And why not including a map (in a supplement if you prefer) indicating the location of all ozonesonde sites, with a different colour to highlight those shown in Figure 6?

By the way, this sentence in the caption of Figure 4:

"Tropospheric ozone profiles of volume mixing ratios (ppbv) against by model versions CBA (red), MOC (blue) and MOZ (green) against sondes (black) over six different regions" should be rewritten to something like:

"Tropospheric ozone profiles (volume mixing ratios, ppbv) from model versions CBA (red), MOC (blue) and MOZ (green) as well as sondes (black) over six different regions"

\* Figure 5. Please draw a line at 0 to better distinguish positive and negative biases. I would also add a symbol to better identify the mean bias within each latitudinal band.

\* Lines 454-455 (for modelled O3 vs. MOPITT CO, Figures 7 and 8): "low bias over the NH during April (further analysed in, e.g., Stein et al., 2014), as well as a low bias over Eurasia during August". Need to change "low bias" to "negative bias". In addition, I would say that in August the negative biases occur not only over Eurasia, but over most continental areas of the northern hemisphere.

The authors could also be a bit more quantitative and provide spatial correlations between modelled and observed CO for each of the maps on Figures 7 and 8. They could even show maps with biases in a supplement if some of the biases I mention are not so clear.

By the way, apart from Stein et al. (2014), the authors could also cite this previous paper regarding the low CO bias over the NH:

Shindel et al., Multimodel simulations of carbon monoxide: Comparison with observations and projected near-future changes, 2006, https://doi.org/10.1029/2006JD007100.

\* Lines 563-565: This sentence is a bit awkward: "Considering its strong diurnal cycle, due to the fast photolysis rate, here only daytime values have been used to construct the averages, because the observations from the various field campaigns were equally conducted in daylight conditions".

It would be convenient to split all this information into two separate sentences. Moreover, the authors should mention not only the fast photolysis rate but also the strong diurnal cycle of anthropogenic emissions.

\* Lines 578–582: "Figures 16 and 17 evaluate tropospheric NO2 using the OMI satellite observations. The simulations deliver generally appropriate distributions ... Another interesting finding is a relatively strong negative bias over ...". It is a bit hard to see the biases in the figures. It would be helpful to show analogous maps with those biases (in a supplement).

\* Line 600: "Compared to the trace gases previously analysed, Nitric acid is not primary emitted but is purely photochemically formed". Reformulate this sentence, because O3 is not primarily emitted. Also, change "Nitric" to "nitric".

## **Technical corrections**

\* line 28: Remove comma in "Other, common".

\* line 56: "correlation between tropospheric composition with ENSO conditions". Change to

"relationship between tropospheric composition and ENSO conditions" or

"dependence of tropospheric composition on ENSO conditions"?

\* line 57: arctic --> Arctic

\* lines 65-66. Change "and, afterwards, their chemistry modules technically integrated" to ". Afterwards, their chemistry modules were technically integrated".

\* line 85: "which is further expanded" --> "which are further expanded"

\* line 130: I think Lacressonnière et al. 2012 is missing in the reference list.

\* lines 139-140: "and 2 for the troposphere for the aqueous oxidation reaction of sulfur dioxide into sulfuric acid"-->"and 2 for the aqueous oxidation reaction of sulfur dioxide into sulfuric acid in the troposphere"

\* Line 186: Brasseur et al. (1998) is missing in the reference list.

\* line 222: "This has consequences for the description of the various emissions". Do you mean "partitioning" instead of "description"?

\* Line 246: "experiment ID's" --> "experiment IDs"

\* Table 3. Remove "in" from "Specifications of the experiments in evaluated.

\* Line 324: No need comma before "over the NH extra-tropics"

\* Lines 326, 327 and probably other parts of the text: Please change "highest" to "the highest"

\* The expression "associated to" is used three times on page 13 as well as in other parts of the text. I think the use of "associated with" is preferable.

\* Line 367: Need space before parenthesis in "deviation(Jöckel".

\* line 389: compare --> compares

\* "Southern Hemisphere (SH)" is spelled out on line 404, but this should have been done before (around line 393).

\* Figure 9: "Izaa" should be changed to "Izaña" (or "Izana" if the "ñ" is problematic) in one of the panels.

\* Figure 10: "monthly mean surface CO by observations (GAW network) and by the model simulations" --> "monthly mean surface CO as derived from observations (GAW network) and model simulations"

\* line 482: "phae" --> "phase"

\* caption of Figures 12, 13, 14, 18 and 19: "Comparison simulated" --> "Comparison of simulated"

\* page 27: The reference "Aydin et al., 2014" is mentioned twice. I believe this paper is from 2011, as indicated in the reference list.

\* lines 549-550: "the correlation showed acceptable values for all versions (R2 >0.7)". The authors should explicitly refer to the "weighted correlation" rather than just "correlation".

\* lines 562-563: "due to its photochemical balance with nitrogen oxide". Do you refer to NO? If so, please change "nitrogen oxide" to "nitrogen monoxide" or "nitric oxide".

\* line 569: Need comma or semicolon before "hence".

\* Caption of Figure 16: "Tropospheric NO2 from OMI" --> "Tropospheric NO2 columns from OMI"

\* Caption of Figure 17: Remove ", and the model biases with respect to this".

\* line 646: Change "is" to "are" in "the averaged differences for O3 (CO) is"

\* Reference list: Benedetti et al. (2009) is included twice.

\* Reference list: I think that Horowitz et al. (2003) is not mentioned in main text.

\* The main text includes these four references: Madronich and Flocke, 1997; Madronich and Flocke, 1999; Madronich et al., 1989; Madronich, 1987. They are somewhat different in the reference list. Please double-check.