Response to Review #2

We thank reviewer #2 for the review, the insightful comments to the paper and for his/her endurance to read the long paper.

We will respond to the review point by point. The reviewer's comments are included in bold italics

Major comments;

1. An important advantage of this study is the simultaneous analysis of trace gases and aerosols within the same data assimilation framework. However, it is unfortunate that their interactions were not considered in the current setting. More discussions on their potential would still be useful. I suggest discussing this topic in an additional section, for instance, how much changes in trace gas concentration can be expected using the analyzed aerosol fields, and if these changes bring further improvements in the trace gas analysis (and vice versa). Although the paper is already very long, presenting several sensitivity calculation results could be helpful.

We fully agree that interactions between chemistry and aerosol within a data assimilation frame work is an important topic. However, its study will be more the focus of ongoing and future work and it is not the result of the work presented here. In the current version of the manuscript we mention the prospects in the conclusion section (L 1155).

In the CAMSiRA (and MACCRA) no interaction between aerosol, chemistry and meteorology was simulated. The only potential interaction would be the impact of the tropospheric ozone assimilation on CO and vice versa. As reported in Inness et al. (2015) the applied system does not show a strong inter-species synergy, in particular as no NO2 retrievals were assimilated in CAMSiRA. A further explanation for the lack of synergies is that no adjoin and tangent linear of the chemistry scheme was applied and that no species-to-specie background error covariances were considered in our 4D-VAR approach.

In the next version of the CAMS system, the impact on assimilated aerosols and ozone in the radiation scheme, the impact of aerosol on photolysis rates and on some heterogeneous reaction (N2O5, HO2) will be considered.

To clarify that the assimilation system used for the paper does not present these interactions to a large extend, we add the following statement in section 2.4 (C-IFS data assimilation).

"A further potential interaction between the assimilated species could be introduced by the adjoint and tangent linear representations of the chemical mechanism and the aerosol module as part of the 4D-VAR approach. The applied tangent linear and adjoin formulation of C-IFS accounts only for transport processes and not the sources and sinks of atmospheric composition. Because of this limitation and the lack of aerosol-chemistry-meteorology feedbacks in the C-IFS version used in this study, interactions among species and with the meteorology as part of the assimilation are not represented in CAMSiRA. "

2. As the system was developed at meteorological operational centers, the authors may want to discuss more about the contribution of the CAMS interim reanalysis to meteorological and climate activities. This discussion would be useful to many readers in understanding how the composition and aerosol reanalysis will be helpful in wide research fields.

In the current version of the manuscript we mention applications of the re-analysis of atmospheric composition, such as boundary condition for regional models and trace-gas climatologies in the introduction and in the conclusions.

We can report that new trace-gas climatologies for ozone and aerosol were compiled from CAMSiRA and implemented in the new cycle of the operational ECMWF NWP model. In particular the reduced ozone bias in the upper stratosphere and mesosphere led to an improved skill in temperature forecasts in this region. See https://software.ecmwf.int/wiki/display/FCST/Implementation+of+IFS+Cycle+43r1

A report/paper is in preparation but not yet ready to be cited.

An other application of CAMSiRA is the analysis of trends, which we demonstrate on the example of CO surface data (Figure 6). Finally, the evaluation of model runs would be a new application for AC re-analysis data. However, we would leave it (within the scope of the paper) to the reader to decide if there is enough confidence that CAMSiRA is well suited for this purpose.

3. In Section 3, the differences in CO between the systems are primarily explained by surface emissions. There could also be clear differences in OH and natural CO sources by oxidation, which may explain the CO differences.

When discussing the global patterns of the differences between CAMSiRA and CR we actually come to the conclusion (L 337) that " … global chemical loss and production of CO as well as problems with the large scale transport. … " and less the CO emissions itself are the reason for the biases of the model.

We find it difficult to distinguish with the discussed model runs to clarify in detail if emissions and distribution of CO pre-courser species such as VOCs and CH4 or a reduced CO lifetime because of higher OH values are more likely the reasons for the identified CO biases. In any case we conclude that the CO emissions are not the sole reason for the CO biases.

We mention this in the conclusion of the paper (L 1128) but will refine the statement to :

"However, the rather zonally homogeneous CO differences between CR and CAMSiRA suggest that not only biases in the fire emissions but also of the CO lifetime and chemical production as well as the CO transport need to be investigated further. "

Specific comments:

L. 394: "Owing to the hemispheric…". This sentence is not clear.

We reformulate as follows:

"Because of the hemispheric influence, i.e. the hemispheric reduction in CO, the CO trend in CR over Eastern China became negative in the middle troposphere."

L.404-407: It is surprising that, even after correcting the concentration by data assimilation, the influence of different emission data is so large. Does this mean that the observational constraints are insufficient to remove the influence of a priori model errors? Further discussions would be helpful.

In the current approach the surface emissions are not changed by the assimilation of CO observations. This has been identified as a topic for future developments in the conclusions (see L 1185).

The missing total agreement with the observations at the time of the analysis is also caused by the relative size of the observation and background error statistic. The background error for CO is calculated using an ensemble of forecasts, which only accounts for the variability in the transport (winds) and not for the uncertainty of the emissions. The background error at the surface is therefore most likely underestimated leading to an "over-confidence" in the model as part of the assimilation process.

We will add at line (L 274, Section C-IFS data assimilation)

"However, the ensemble did not account for the uncertainty of the emissions, which leads to an underestimation of the background error for CO."

And we will ad in the section on recommendations (L 1187)

"A promising development is to enable the correction of emissions with the C-IFS data assimilation system based on observations of atmospheric composition. This could also improve the analysis of tropospheric ozone as ozone precursor emissions would be corrected. An intermediate step in this direction is to better account for the emission uncertainty in the model background error statistics. "

L. 437-440: How does the bias vary with year?

The data coverage of the MOZAIC/IAGOS data varies a lot so that a robust conclusion for the year-to-year variability would be difficult to obtain. However, we discuss the agreement of the trends for surface CO observations in section 3.4. and show a good correspondents in the observed, modelled (CR) and assimilated trends (CAMSiRA).

A conclusion of the discussion of the inter-annual variability of the CO burdens (section 3.2) over Europe and North-America is that there is better agreement between CR and CAMSiRA at the end of the period. This could indicate that the biases of the anthropogenic emissions decrease from 2003 to 2015.

L. 798-799: "However, the change of..." This sentence is not clear

We reformulate as follows:

"It is not caused by the change of the assimilated MLS version (from V2 to V3.4) because this took place already at the beginning of 2013 (see Table 2)."

L. 1008-1010: How long was the spin-up period?

The MACCRA was started on the 1.12.2002 (Inness et al. 2013)

We add " …and the short spin-up period of only 1 month"