Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-864-RC1, 2020

Atmospheric Chemistry and Physics
Discussions
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Interactive comment on "On the relationship between tropospheric CO and CO_2 during KORUS-AQ and its role in constraining anthropogenic CO_2" *by* Wenfu Tang et al.

Anonymous Referee #1

Received and published: 4 November 2020

The manuscript "On the relationship between tropospheric CO and CO2 during KORUS-AQ and its role in constraining anthropogenic CO2" by Tang et al. presents analysis based on the CO, CO2 and 14CO2 data collected during the KORUS-AQ campaign over South Korea. It compares simulations of CO, CO2 and FFCO2 concentrations from a global transport model to the data and presents inversions of the FFCO2 emissions in East Asia (Eastern China, Korea and Japan) using this transport model and these data.

The prospect of the joint analysis of CO2, CO and 14CO2 data supported by transport model simulations and of their joint assimilation in an inversion system is very

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Discussion paper

promising. Sometimes, the manuscript nearly reaches interesting insights on this topic. However, in a general way, the study and the manuscript fail to exploit the potential of such analysis. I think that more work and thoughts are needed to produce a paper that deserves publication and that this goes beyond what is usually done for "major revisions".

Here are some of my main concerns regarding this manuscript:

1) It is often very difficult to follow because the writing and the reasoning are not structured and rigorous enough. A large amount of sentences are confusing because of the lack of clarity, precision and explanations. The reading of the long series of statistics lacks of hierarchy. The use of simulations or datasets that are not much exploited in the analysis does not help. For instance, efforts are needed to follow, in section 3, whether or why under or over estimations of concentrations are supposed to highlight the under or over estimation of local, regional or global sources and sinks. The reading of section 4 is even more difficult.

Furthermore, despite being relatively long, the text goes too fast on some of the critical parts of the reasoning like the rationale for the study in the introduction and the justification for the use of specific inversion configurations and parameters.

I think that many assumptions and analysis are debatable and that the manuscript shows a lack of hindsight on the topic and results of this study.

2) The specific scope and the objectives of sections 3 and 4 are not clear. This manuscript is the 5th one analyzing the CO2 and/or CO data from the KORUS-AQ campaign using transport models (after Tang et al. 2018, Halliday et al. 2019, Tang et al. 2019a and Gaubert et al. 2020, ACPD). How do section 3 and 4 draw on these previous publications and bring new learning ?

Furthermore, opposed to what is claimed repeatedly (e.g. in the title, the abstract, the beginning of section 1.1, the beginning of the conclusion...), I hardly see how these sections feed the configuration (in particular the error covariance matrices) or the analysis of the inversions in section 5.

The introduction does not help much. Until the beginning of section 1.1, this is a collection of very general (and sometimes misleading) statements about the monitoring CO_2 anthropogenic emissions using atmospheric data. Section 1.1 fails to bring clear specific context, rationale and objectives for this study. Lines 150-154 look like a summary of the activities rather than a list of objectives. Lines 154-172 attempt to distinguish this new study from the previous ones by pointing to practical differences that are hardly convincing. Lines 769-770 and 775-776 claim that the set-up of matrix Se follows values of errors on CO and CO_2 and $errR_{CO,CO2}$ from section 4 but I do not see how. Anyhow, the errors and the correlation of errors in CO and CO_2 modeled concentrations are driven by the atmospheric transport, the surface fluxes (and other source and sinks), and to errors in both the transport model and in the modeled sources and sinks. I do not see why it should be used to characterize transport model errors only. The authors seem to miss the links between their statistics of model-data differences and Sa (see below my general comment about the key role that this matrix should have played).

3) There is a critical lack of proper discussion on the spatial extent and scales that are suitable for the analysis of the data. Parts of sections 3 to 5 attempt to distinguish the influence of Eastern Asia or the rest of the world vs that of Korea, l340-350 discuss the sources overflown during the campaign and the flights over the West Sea are said to be designed to capture “China pollution outflow”. Some sentences even raise (too late) some concerns associated to the coarse spatial resolution of CAM-chem. However, in general, and in particular in the title, introduction and section 2, there is no real reasoning regarding the observation footprints and regarding the modeling and inversion domain and resolution that are suited for these data.

The introduction mixes all inversion scales and all types of observation networks. Nothing is said about the wind fields during the campaign. The comparisons of a single global coarse resolution model to TCCON individual sites, to OCO-2, to MOPITT data or to the aircraft data are brought together without consideration for the differences between these observation datasets in terms of flux and process representativeness. The statistics obtained here are sometimes compared with results from other campaigns in different regions or with different coverage, or from "state-of-the-art" models (without mentioning whether they are global or regional). The contrast between the spatial extent of the regions for which the total emissions are rescaled by the inversion and the local (nearly vertical) reading of the data footprints at l340-350 is questioning.

Given the spatial extent of the KORUS-AQ campaign (less than 8°x8°) and its density of data over South Korea, the use of a global 1° resolution transport model with coarse vertical resolution to analyze it is not obvious and sounds like a step backward compared to the previous publications on this campaign (which used higher resolution models). From what I understand, the model is interpolated at each observation location and all the statistics are derived over the ensemble of observation locations. What can be the meaning of such statistics at a resolution much finer than that of the model ?

4) The usual concept for the co-assimilation of CO and CO2 is that the signal from misfits between modeled and measured CO can be used to add constraint on the inversion of FFCO2 emissions because uncertainties in the FFCO2 emissions are connected to uncertainties in FFCO emissions. This usually translates into two options for the inversion: (a) rescaling activity levels underlying both FFCO and FFCO2 emissions rather than FFCO and FFCO2 emissions separately or (b) rescaling separately FFCO and FFCO2 emissions accounting for positive correlations between their respective prior uncertainties. The key challenge for joint CO-CO2 inversions is usually thought to be the uncertainties and high spatial and temporal variations in the CO/CO2 emission ratios i.e. in (a) the uncertainties and variations in the CO and CO2 emission factors to be multiplied by the common activity indices to get emissions and in (b) the level and variations of the positive correlation between the prior uncertainties in FFCO and

FFCO2 emissions. The authors of this study control separately FFCO and FFCO2 emissions but neglect the correlations between their respective prior uncertainties or acknowledge that they have no idea about how to parameterize them, cutting the critical connection between FFCO2 and FFCO (see lines 819-830). The configuration of Sa is actually made all the more difficult by using two different inventories for prior FFCO2 and FFCO emissions, which undermines the ability to rely on tights connections between these emissions (especially since the CO inventory has been multiplied by 2 to better fit the data before the inversion).

The authors even assume that these correlations are negative, i.e. that the combustion efficiency could be the main source of uncertainty in both FFCO2 and FFCO emissions. However, the prior FFCO and FFCO2 emissions are based on different inventories (and FFCO emissions have been multiplied by 2), so that, in principle, the ratios between these emissions should not correspond to an assumption on this efficiency. In a more general way, such an assumption is quite surprising. Uncertainties in the level of activity at national, and even more at regional to local scales should be a dominant source of uncertainty in FFCO2 emission inventories (especially at sub-annual temporal scales). Such a driver of uncertainty in FFCO2 emissions raises correlations with uncertainties in FFCO emissions. Even if uncertainties in CO emission factors are one of the main sources of uncertainties in FFCO emissions, their counterpart in FFCO2 emissions (the generation of plus or minus CO2 depending on the combustion efficiency) can hardly balance this driver. The assumption of negative correlations between uncertainties in FFCO2 and FFCO prior emissions is further weakened by various discussions in sections 3 and 4 that point to common underestimation or overestimation of sources in both the CO2 and CO inventories.

The idea that Se could ensure the expected connection between FFCO2 and FFCO is not relevant. Se prevents the inversion from overfitting the data and limits the corrections to the prior emissions. Correlations between CO2 and CO transport modeling errors help the inversion better filter such errors when deriving FFCO2 and FFCO emissions but this has a very indirect and weak impact on the connection between these emissions. One could think that the results from the inversions in section 5 contradict these statements. However, I assume that the consistency between FFCO2 and CO2-CO inversions and their divergence from CO2 inversions is intrinsically linked to the very low dimension of these inversions (with 4 or 8 unknowns). Various assumptions and settings of these inversion raise issues (see below) and I thus have the feeling that this result is not robust.

The general topic of the CO-CO2 correlations that should be at the heart of this paper is complex and I feel that the authors missed it.

5) Some concerns about the inversion configuration:

- I don't understand how global inversions (even if they focus on East Asia) assimilating CO2 (with or without CO) can behave correctly when inverting scaling factors for FFCO2 emissions only and keeping other CO2 fluxes fixed. The "posterior" biogenic fluxes from CTE and CAMS bear large uncertainties. The problem is enhanced by the lack of spatial resolution of the inversion, which does not help distinguish the patterns from anthropogenic vs biogenic fluxes in the data.

- I do not understand the rationale behind removing observations above 3 km (the "localization purposes": l791-792 refer to "previous section" but I do not see where sections 3 and 4 help understand this choice). The inversions are global, and data above 3 km should be helpful for constraining ROW.

- I have some doubts regarding the way the FFCO2 simulations are compared to gradients of FFCO2 from gradients in 14CO2 observations: I believe that the authors should have computed, in the FFCO2 simulations, the gradients to the location of the 14CO2 background sites rather than introduce a "ffCO2 offset".

6) There is a lack of consideration for the inventories. As mentioned above, the fact that the FFCO2 and FFCO inventories are quite independent strongly hampers the in-

terpretation of the model-data comparisons and the reading of CO/CO2 ratios in the model. The EDGAR inventory, which plays a critical role in this study (this is the FFCO2 emission inventory behind the simulations in sections 3 and 4 and the prior emission inventory in section 5) is not even named in the main text, the authors speaking about CTE and CAMS fluxes (which combine natural fluxes optimized through global inversions with the EDGAR inventory) only. Differences between the CAMS analysis and CAM-Chem simulations are not supported by insights on the emission maps behind these two simulations. The FFCO2 emissions from the inversion are hardly compared to official inventories (and the CO emissions from the inversions are not discussed).

7) I could draw a long list of secondary issues in specific paragraphs or sentences (and even in the mathematical notations and equations) but I restrain myself to the main ones raised above

In conclusion, I think that the scope and configuration of the analysis should be rethought, and that the presentation of such a study should be strongly improved. I thus recommend this manuscript to be rejected and re-submitted after a deep revision.

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