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Discussions

Interactive comment on "Correcting model biases of CO in East Asia: impact on oxidant distributions during KORUS-AQ" *by* Benjamin Gaubert et al.

Anonymous Referee #1

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In this article, CO MOPITT data are assimilated in a global coupled chemistry-climate model (CAM-Chem) and the resulting distributions of CO and other compounds are evaluated against aircraft measurements from the KORUS and ARIAs campaigns conducted in May-June 2016 above South Korea and Northern China. In addition, the impact of different emission datasets on the model performance is evaluated with CAM-Chem using dynamical fields nudged to GEOS-FP analysis. Both the MOPITT assimilation and the comparisons with aircraft data suggest a substantial underestimation of CO emissions over Eastern China, consistent with several previous studies. Furthermore, the emission update improves (somewhat) the model performance for ozone and HOx, based on aircraft measurements (for O3) and box-model calculations (for HOx) constrained by measurements from the KORUS campaign. The paper is gener-

ally well-written, and the methodology is well described. A rather detailed discussion is provided, which covers not only the influence of emissions but also aspects related to dynamics, which I appreciated. The conclusions of the study appear mostly valid, although with some exaggeration of the benefits of assimilation and some oversimplication regarding specific aspects.

Major comments

- Most importantly, the role of model biases for NOx was clearly overlooked. The paper claims a good agreement against aircraft NOx data, whereas Fig. S2 shows a strong model underestimation (> factor of 2) of both NO and NO2 at all levels except the surface. This aspect warrants more discussion and possibly additional sensitivity calculations as it might have a strong impact on ozone, HOx and therefore CO and VOCs.

- l 106-115 "it has become evident (...) that several standard inventories of CO emissions in China are still too low (Tang et al 213; Yumimoto et al. 2014)" The two references cited are inappropriate for this statement. The first concerns only Beijing; and in the second study, the optimized CO emissions were found to be lower than their a priori inventory. Furthermore, Elguindi et al. (2020) showed that two widely used inventories (MEICv1.3 and REAS 2.1) have higher emissions than most top-down estimations. Therefore, I recommend to nuance the claim made repeatedly in the article that inverse modelilng suggest higher emissions than bottom-up inventories. Clearly, things are more complex as there is considerable variability among models (as well as among bottom-up inventories), and this should be acknowledged. I would be curious to know how the bottom-up emissions used in Cam-Chem compare with those intercompared by Elguindi et al. (2020). The top-down emissions determined in in this study using MOPITT CO depend on OH levels not only over S. Korea (for which we have partial constraints from KORUS) but also over East Asia and elsewhere. The related uncertainties deserve some discussion in this manuscript.

Minor comments

- the reference Feng et al. 2020 is cited (line 112) but is missing

- l 136-137 Note that De Smedt et al. (2015) (https://doi.org/10.5194/acp-15-12519-2015) reported negligible trends over Beijing and PRD over 2005-2016. Wang et al. (2015) (www.atmos-chem-phys.net/15/1489/2015) also reported decreased NMVOC traffic emissions over Beijing since 2002

l 142-145 "Ozone production and accumulation.." I don't see the link between this sentence and the rest of the paragraph

l 194-195 "This suggests an underestimation of emissions" Where does this come from?

l. 201-202 "two episodes of the transport phase": unclear

l. 230-231 "A high correlation between organic aerosol and CH2O, which is a characteristic of the importance of primary and secondary sources": very unclear, rephrase

l. 289 Where does this threshold of 20 ppbv comes from? How sensitive are the results to this value?

l. 317 Does the FINN inventory includes the emissions due to to agricultural residue burning, very high over the North China Plain in June ? Could an underestimation of those emissions in the model account for the low bias of CAM-Chem CO wrt MOPITT and aircraft ? It could be interesting to investigate whether the low bias is highest in June, compared to May and July. By the way, I don't think that the duration of the assimilation run is mentioned in the manuscript. What days or months of data are used in the assimilation? More generally the assimilation setup could be more detailed.

l. 335-336 "They found missing PFT data over Seoul and a large sensitivity in PFTs to changes in temperature": unclear, I don't understand what is meant here.

l. 341 "to determine a best fit to the observations...": the model does not match well

the observations for e.g. methanol, acetaldehyde, methyl hydroperoxide.. In fact, the high emissions lead to a worsening of the vertical gradient for some species. Please rephrase. The sensitivity test is not uninteresting but it fails to prove that biogenic emissions have a moderate impact on CO.

l. 381 I suppose that the assimilation of MOPITT influences not only emissions but also meteorology. If so, this should be mentioned and maybe shortly discussed.

l. 442 Does this implies that e.g. NOx anthropogenic emissions are updated as well, similar to CO?

l. 443 "strongly correlated": or completely correlated?

l. 479 "We compare our emission estimates": confusing. In this paragraph you sometimes refer to emissions and sometimes to the the model-calculated concentrations. Please re-structure this section to clearly state what is compared to what.

l. 485 Please define what is a "tag"

l. 489 The CAMS inventory is not mentioned in Table 2

l. 504 Define DACOM

l. 511 "Because of the reduction of the CO in the middle troposphere (...) the RMSE in MOPITT-DA is reduced...": I don't see that, please check.

l. 534-535 "if true errors in the VOCs are not correlated to CO, only noise will be introduced": sorry, unclear.

l. 545 replace "or" by "and/or"

l. 555 " bias in OH leads to correlated errors between CO and alkanes that can be mitigated by including these species in the state vector": I understand, but even if the VOCs would not be the state vector, their concentrations would be modified (improved) by the optimization.

l. 589 Only a small part of the HO_2 underestimation is explained by the CO underestimation

l. 595 Note that the reduced HCHO formation is due to lower OH, entailing reduced VOC oxidation rate

- Note that the $J(NO_2)$ underestimation could play a role in the O_3 underestimation.

- Does the model have biogenic CO emissions? Dry deposition of CO?

- l. 627-628: Note that this overestimation could be partly due to an overestimation of biogenic VOC emissions over Southern China and Southeast Asia (cf. Souri et al., ACPD 2020).

l. 638 "the spatial patterns of the prior emissions are important..": nevertheless, the posterior differences (Fig 5f) are larger than the prior differences (Fig 5c) suggesting that the prior emission differences are not crucial.

l. 645-657: This increase is not at all visible on Fig. 5g

l. 652 'China' –> 'Central China'

l. 657 I suppose those numbers are for Northern China, right?

l. 684 'smaller': by how much?

l. 685-687 The posterior CO is still 50 ppbv below MOPITT-DA. Why is that, what does this tell us? I think this deserves more attention.

l. 697 I recommend moderating the claim of better O_3 match.

l. 'It means that CO acts..." Of course, this is expected. Delete "and is more consistent with the observations"

l. 699-700 "10 ppbv in the free troposphere": are you sure? I see this in the PBL, not the FT.

l. 710 Make a reference to Table 4 and mention that the average is for all levels.

l. 716-717 "such as errors in transport or chemistry": strange statement! How well does the assimilation matches MOPITT CO? If the match is very good (which I expect), then the discrepancy is due to a combination of measurement or representativity issues.

l. 738-740 "This suggests that weather patterns and direct anthropogenic emissions explain most of the CO variability during the campaign": I'm not so sure, since the posterior model run does not perform very well. The fact that the MOPITT assimilation run performs well indicate that the assimilation of initial conditions is important, which does not really tell mcuh about the reasons for CO variability. Is there a temporal evolution of CO emissions during the campaign period in the MOPITT-DA run?

l. 766 "for higher altitudes": at what altitudes?

l. 767 I would rather say "of stratospheric influence" or something like that.

l. 782-791: As mentioned above, the model discrepancies for NOx are very relevant for the discussion. I recommend to display the NOx comparisons for the separate phases. In Fig S2, the NOx are clearly much too low between 850 and 650 hPa, which might explain the HO_2 biases. Note that the good agreement for OH might therefore be fortuitous, since it could be affected by higher NOx.

l. 803-804 "chemical production and loss via OH reaction from emissions..." unclear

l. 926 There is a net loss of HOx for all non-zero gamma values...

l. 932-933 "with a more pronounced effect in the upper part of the boundary layer with less influence on the $OH+NO_2+M$ reaction while..." unclear

l. 935 "this exact effect" also unclear

Technical comments/language

- l. 187 "looked"??

l. 247 Capitalize "we"

l. 277-278 "Their analysis of the aerosol pollution was mostly located below..." unclear, please rephrase

l. 309 Delete "Therefore, "

Equation 4 and following text: please use consistent notations/fonts for mathematic expressions

- l. 523 A parenthesis is missing

l. 633 remove final 's' in Plains

l. 642 & 643: '6' should be '5'

l. 645 Compare –> Compared

l. 653 The sentence is weird. The verb should be after the parenthesis. I suggest deleting 'is two times higher than'

l. 661 replace 'than for' by 'and'

l. 691 'above 900 hPa' is ambiguous, rephrase

l. 695 The layer?

l. 712 'means' –> "implies'

l. 763 "for" –> "to"

l. 767 'from' –> 'of'

- at various places in the manuscript and supplement, write "methanol" and not "Methanol", etc. for all species.