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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Pollution trace gas distributions and their transport in the Asian monsoon upper troposphere and lowermost stratosphere during the StratoClim campaign 2017" *by* Sören Johansson et al.

Anonymous Referee #2

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Johansson et al. presented atmospheric trace gas measurements of HNO3, O3, PAN, C2H2 and HCOOH from the GLORIA instrument collected during the recent StratoClim campaign. These are valuable measurements from the Asian Summer Monsoon region and should be published in time. I have a few major comments. Number one being the use of language in writing. I agree with the other reviewer. Please have the manuscript edited by a professional writer before submission of revision. The other two major comments are related to the discussion in Section 4, pages 14-15 and the model evaluation approach in Sections 5 & 6. See below for details. I found the model evalu-

ation and bias assessment being the most problematic weakness of this study. These concerns need to be addressed before the paper should be accepted for publication.

P2, L6 & L12: Santee, 2007 → Santee et al., 2007.

P2, L16-17, what's the name of the aircraft campaign?

P2, L22-24. The logical connections of these two sentences and the previous section seems to be amiss. What's the relationship between radiative heating rates and transport in reanalyses with trace gases? Observations are sparse, so what? How do observations help? The last sentence in the next paragraph (atmospheric chemistry models . . .) is out of place. It fits much better in this paragraph, instead.

P2, L24-28. These three sentences seem to repeat themselves in various ways. It can be easily condensed into a single sentence but capture all essential elements. Please revise.

P2, L33. How about change to ". . . we use the NMVOC measurements from GLORIA collected during StratoClim to address two important science objectives."?

P3, L1. Which two models? EMAC and CAMS? Please specify. And I am sure this is not the "first evaluation" of these two atmospheric chemistry models.

P3, L7-8, you can simply say "Section 5 presents . . . & Section 6 discusses ..."

P3, L11. You probably should list the names of the five targeting species here.

P3, L22: → (MIPAS, von Clarmann et al., 2009)

P3, L25. "within the transport . . ." → "to examine the transport . . ."

P3, L30. WMO, 2019 → This one is actually "WMO, 2018"

P4, L1. Catalytic reactions with nitrogen oxides is a sink of ozone in the stratosphere and to some extent in the upper troposphere. Primary sources of ozone is the troposphere is in situ production of NOx +HOx, and NOx + peroxide radicals from VOC

oxidation, hence indicator of polluted air masses.

P4, R1. To be more accurate, you should change "->" to "<->"

P4, P17. Change to "the lifetime of PAN is very short at lower altitudes due to rapid thermal decomposition"

P4, L27. Maximum tropospheric mixing ratios of a few ppt for C2H2? Are you sure it is not a few ppb?? Check Xiao et al., (2007).

P7, L11. With regard to a better → with a better

P7, L14. Can you describe what are the NMVOC emission sources used in EMAC? When you say 50% and 100% additional emissions, do you mean from all emission sources, e.g. biomass burning, biofuel, fossil fuel, etc., and globally or just over Asia?

P8, L3-4. I find this sentence very awkward, with no clear description of what was actually done.

P7-8. In sections 2.3, could you also provide the details on which year, time period of the model simulations that were conducted?

P8. Section 2.3.5 is out of place. This is observations and it should be listed in Section 2.1 or Section 2.2, not in the modeling subsection.

P9, L3 & Figure 2. I think it is more accurate to say these are colored boxes are "air masses" of interest, rather than "regions" of interest. Also in figure 2 caption, add "shows" after "the green line". I find the green line very hard to see. A thick solid dark gray line would be much better. It also distinguishes its functionality from the color boxes.

P9, L21. Discriminate -> distinguish

P9, L27. As for Pan –> Similar to PAN

P9, L28. Delete "as for PAN"

P9, L32. -> there is a minor local maximum with VMRs up to 100 pptv.

P10, Figure 2. I find all panels very noisy, which is not surprising due to the large errors in GLORIA measurements as listed in Table 1. I would suggest average the measurement samples to larger temporal and vertical bins. This way you can average down the noise and illustrate the discussed features much better. In the present form, these features are barely distinctive from the surrounding background air masses. This is particularly problematic from C2H2 and HCOOH.

P11, L12 and hereafter. Please use the proper terminology: Henry coefficient should be changed to Henry's Law Constant.

P11, L14-15. You used two "while" in one sentence, grammatically not correct.

P12, Figure 3 and the corresponding discussion. (a) In the text, the relevant discussion uses km as a unit while the y-axis only shows pressure. Please add the corresponding km on y-axis. (b) The cyan box in TRACZILLA show likely convective influences while ATLAS shows none. Why the two models are showing such different results? And how can the GLORIA measurements help in assessing which back trajectory model is more accurate. Also, overall, I can see TRACZILLA shows more convective influences that ATLAS. How can you assess which one is more accurate?

P14-P15, the discussion on various air mass signatures. For clarity and easy-to-follow purposes, I highly recommend you assemble all this information into a table. In the table, please list the type of targeting air masses, altitude at which they are sampled, surface regions where they were originated from, average measured HNO3, O3, PAN, C2H2, HCOOH concentrations within these colored boxed, transport time since they left the surface, etc. Second, please add a summary discussion on the different chemical signature of airmasses from different regions, e.g. the purple/blue box air from the marine background vs. the orange/red box air from China, etc.

Sections 5 & 6. I found the observation-model comparison and evaluation a major

weakness of this study. Neither CAMS nor EMAC produces well the observed features and gradients of all five species. This is particularly the problem for C2H2 and HCOOH. I also have problems with the brutal way of increasing NMVOC emissions by 50% and 100%. I don't see any improvements in model performance with such approach. By matching with observations better in a few patchy spots, you are also creating huge biases in other places (Figure 6) for all three species. PAN, C2H2 and HCOOH can be emitted and/or formed from various sources, i.e. anthropogenic emissions and biomass burning emissions being the highly relevant sources. The differences in the regional distributions of these sources can have a dominant impact on tropospheric distribution of these gases after they are being lofted and formed during transport. A proper way to address this model bias is to adjust the emission strength of these individual sources in separate runs and assess how do the resulted distribution change. This way, one can potentially assess the sources of these biases. You are only presenting analysis of one single flight. Therefore, such model sensitivity simulations can be easily conducted within a few days. The new model results and the corresponding discussion should be included in the revised manuscript before the paper is moving forward for publication.

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