Lin Tan:

Dear Clara,

Many thanks for this interesting study. Below I would summarize my understanding about your work and provide some comments if you find them useful.

The study aims to investigate the changes in the vertical distributions of atmospheric species over Europe before and during the COVID-19 pandemic. The pre- and intra-pandemic vertical distributions were measured by 3 aircraft campaigns: UTOPIHAN campaigns in 2003/2004, the HOOVER campaigns in 2006/2007, and the BLUESKY campaign in 2020. The model ECHAM5/MESSy2 Atmospheric Chemistry (EMAC) is run in a pre-pandemic scenario (also known as the no-lockdown scenario); the model data subsampled along with the flight tracks of the three campaigns areused to compare with the observations. The model data are first validated against HOOVER and are found to reproduce the HOOVER observations, including the trends. Then, assuming that the pre-pandemic atmospheres remain the same, this study compares the intra-pandemic observations by BLUEKSY with hypothetical pre-pandemic BLUESKY measurements constructed using the model data. A major finding is that in addition to the significant drop in major pollutants at the surface that are related to car exhausts such as NO_x and CO, there is also a significant drop in NO_2 in the upper troposphere at 10 km, which is likely due to the reduced air traffic. Nonetheless, this study finds that the production rate of O_3 in the upper troposphere remains unchanged despite the NO_x change. Another major finding of this study is that the chemistry regimes in the upper troposphere might have changed from a VOC-limited chemistry in the pre-pandemic era to a NO_2-limited chemistry in the intra-pandemic era.

Dear Lin,

Thank you very much for your feedback and your time to read our manuscript. We found your comments really helpful and have implemented your suggestions in the manuscript draft.

I have a few minor comments and hopefully you may find them helpful:

If I understand it correctly, in both Figures 2 and 3, there is only one model simulation: the ECHAM5/MESSy2 Atmospheric Chemistry (EMAC) that was run in the no-lockdown scenario. But Figures 2 and 3 may give an impression that there were different simulations separately for HOOVER and BLUESKY. Similarly, calling the subsampled model data on the BLUESKY flight path as BLUESKY-NL also made me think that there was another BLUESKY campaign before the lockdown. Would something like EMAC(on HOOVER path) and EMAC(on BLUESKY path) be clearer?

Thank you for pointing this out. Figure 2 shows the comparison of the vertical profiles for HOOVER for the measured data in orange and the modeled data along the flight track in blue. Figure 3 shows only modeled data generated from EMAC along the flightpaths of each individual campaign. Two simulations were run on the BLUESKY path: the lockdown and the no-lockdown scenario. We have tried to clarify this in the caption of the Figures.

Figure 2 Lines 3 f.: Blue colors show modeled data by EMAC along the HOOVER campaign flight track (Model) and orange colors show experimental data (Experiment).

Figure 3 Lines 2 ff.: All data shown here are from EMAC model simulations along the flight track of each research campaign. Two separate simulations were run on the flight path of BLUESKY, one with lockdown (BLUEKSY) and one with business-as-usual emissions (BLUESKY-NL).

Since you found that there was more NOx in the upper troposphere before the pandemic, have the possible self-contamination due to the NOx emission of the aircraft itself been removed or calibrated in order to establish the robustness of the NOx decrease from the pre-pandemic era to the post-pandemic era?

We can exclude self-contamination due to the flight trajectories. Usually self-contamination only occurs when flying narrow curves with simultaneous advection or performing return flights on the same flight trajectories, both of which were avoided during the campaign.

Figure S4–S6 are important results of this study. Especially, Figure S4 demonstrates the impact of air traffic in the model, which is one of the two major conclusions of this study. I strongly think that these 3 figures should be put in the text. The x-axis range of Figure S5 could probably be either re-adjusted or re-plotted using the log scale for better data representation.

Thank you for this suggestion. Figure S5 (now Figure S7) shows the same vertical profiles as Figure 3d, just on a different x-axis scale. We have added some note for clarification. We have added Figures S4 and S6 and some explaining text to the main manuscript.

Figures S7: NO2 vertical profiles during UTOPIHAN, HOOVER, BLUESKY with and without lockdown scenario as in Figure 3d of the main manuscript, but showing the full x-axis range.

Lines 241 ff.: The observed NO reduction in the upper troposphere can be attributed to reduced air traffic which we show in Figure 4. In addition to the vertical profiles of NO for BLUESKY (red) and BLUESKY-NL (yellow), we present the modeled BLUESKY-NL scenario without aircraft emissions in blue. In the lower troposphere, where aircraft emissions do not play a significant role, this profile is identical to the BLUESKY-NL scenario. In the upper troposphere, it is very similar to the BLUESKY scenario (including the air travel restrictions), showing that reduced air traffic causes the observed NO decrease.

Lines 289 f.: Modeled P(O3) via NO2 photolysis and measured P(O3) via reaction of NO with O3, OH and HO2 are in good agreement which we show in Figure 6.

The conclusion “While the NOPR did not change under lockdown conditions due to compensating effects in the NOx chemistry, we can expect impacts on tropospheric ozone from changes in VOCs (including CH4) relevant for future emission scenarios.” Maybe a little more justification may help support this statement. For example, the impact of aviation NO_2 on O_3 and CH_4-related species in the upper troposphere and lower stratosphere during the pre-pandemic era have been discussed previously, e.g.

Khodayari, A., Tilmes, S., Olsen, S. C., Phoenix, D. B., Wuebbles, D. J., Lamarque, J.-F., and Chen, C.-C.: Aviation 2006 NOx-induced effects on atmospheric ozone and HOx in

Community Earth System Model (CESM), Atmos. Chem. Phys., 14, 9925–9939, https://doi.org/10.5194/acp-14-9925-2014, 2014.

Khodayari, A, Seth C. Olsen, Donald J. Wuebbles, Daniel B. Phoenix, Aviation NOx-induced CH4 effect: Fixed mixing ratio boundary conditions versus flux boundary conditions, Atmospheric Environment, 113, 135-139, https://doi.org/10.1016/j.atmosenv.2015.04.070, 2015.

I think by adding some discussions of these literature may help strengthen your study. In addition, have you tried changing upper tropospheric CH_4 in the ECHAM5/MESSy2 model and test its impact on upper tropospheric O_3?

We have added some text on these literature suggestions in the text. Thank you also for the suggestion of investigating the impacts of CH4 on O3 in the UT, which we agree to be an interesting topic to study, but we believe to be beyond the scope of this study.

Lines 342 ff.: The effects of NOx aircraft emissions on O3 and CH4 have been previously discussed for pre-lockdown conditions in Khodayari et al. (2014) and Khodayari et al. (2015) who present increased methane loss rates and a shorter lifetime as a response to increased OH concentrations from aviation as well as higher ozone production rates. Having investigated NOPR in this study, lockdown effects on CH4 loss in the UT induced by reduced air traffic could be subject to future studies.

Overall, this is a very interesting study. Thank you for your work and good luck!

Lin Tan

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