Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2021-479-RC1, 2021

Reponse to Reviewer Comments on acp-2021-479

Anonymous Referee #1

Referee comment on "The Effects of the COVID-19 Lockdowns on the Composition of the Troposphere as Seen by IAGOS" by Hannah Clark et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-479-RC1, 2021

Review of acp-2021-479 "The Effects of the COVID-19 Lockdowns on the Composition of the Troposphere as Seen by IAGOS"

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Summary and General Comments:

The authors report IAGOS measurements from Frankfurt airport of ozone (data since 1994) and CO (data since 2001) during the MAM 2020 COVID lockdowns. In addition, IASI SOFRID CO satellite data, and ECMWF (boundary layer heights, FLEXPART trajectories) are used in supporting analyses. In general, the authors show increases in March and May 2020 surface layer ozone, little change in March and May 2020 free-tropospheric ozone, and decreases in MAM surface layer CO.

This analysis is a worthwhile and appreciated effort to quantify the effects of COVID emissions changes on the trace gases ozone and CO – there are few published studies that use in-situ profile measurements during the spring 2020 period that aim to accomplish this.

My main concern with this manuscript is that it is difficult to tell how robust, particularly for the surface layer ozone increases, the 2020 anomalies truly are. There are drastically different periods used to calculate the 2020 anomalies for ozone (1994-2019) and CO (2016-2019). The reasoning behind using the 4-year baseline for CO is the decreasing trend since measurements began in 2001. This makes sense. However, there is also a clear increasing trend in surface layer ozone since 1994 (Figure 3b). One assumes that this is at least partly the result of decreasing titration of ozone by NO from long-term NOx emissions reductions. If using a 2016-2019 baseline period to calculate surface layer ozone anomalies, the March 2020 positive anomalies may disappear entirely, and the May anomalies will likely be reduced substantially. 1994-2002 appear to have a strong influence on the 2020 positive ozone anomalies. The results are also shown for months with approximately half of the typical number of profiles. There are no statistics presented on confidence intervals/p-values to confirm the significance of the results and whether they fall outside of expected recent interannual variability.
The results presented here underscore the difficulty of quantifying COVID-related air pollution changes from a single location. The vast majority of published studies of surface and satellite data use dozens to hundreds of locations to bolster their results. It is why Steinbrecht et al. (2020) required dozens of ozonesonde stations to show a convincing decrease in NH free-tropospheric ozone from sonde measurements.

>We have addressed the reviewer's comments in two ways. First, we have changed our reference period for ozone to 2016-2019 as the reviewer suggested. In addition we have added confidence limits to all of the individual monthly bar plots that we discuss calculated on the basis of Student's t-test. This is such that we can now account for the differing number of profiles in each month. The ozone anomaly in the surface layer in May lies outside the expected recent interannual variability even when the smaller sampling is considered.

Minor Comment: Were NOx measurements also available on these flights (Berkes et al., 2018; https://amt.copernicus.org/articles/11/3737/2018/amt-11-3737-2018.pdf)? Even if only recent years are available, you could calculate Ox = ozone + NO2. If Ox is about the same in 2020 as past years, and NO2 or NOx is lower, that would further support the argument of reduced NO titration leading to increased ozone in 2020. At the very least, the profile data should be combined with nearby surface NOx data to confirm the NO titration argument, rather than leave it to speculation.

>Unfortunately, we do not have the NOX data available from IAGOS for these flights as the instrument did not fly during this period. The decrease of NO2 at Frankfurt from surface stations and satellites has already been documented by Barré et al 2021. Barré et al controlled for meteorological factors and the estimates of lockdown-induced NO2 changes for Frankfurt were -24% and -33% based on TROPOMI observations and surface stations respectively. We infer therefore that the positive anomaly in ozone at night is linked to this drop in NO2 at Frankfurt and the consequent reduction in ozone titration. We add reference to this article in the text.

To summarize, I suggest the following analyses in addition to other topics raised in the line-by-line comments:

- Re-assess the ozone results with the same baseline period as CO of 2016-2019
  >We have changed our reference period for ozone to 2016-2019 as the reviewer suggested.
- Produce a more robust statistical analysis indicating the significance of observed ozone and CO anomalies. This is important because only one location is being analyzed, and there is a lot of noise, interannual variability, and underlying long-term ozone trends in the data.

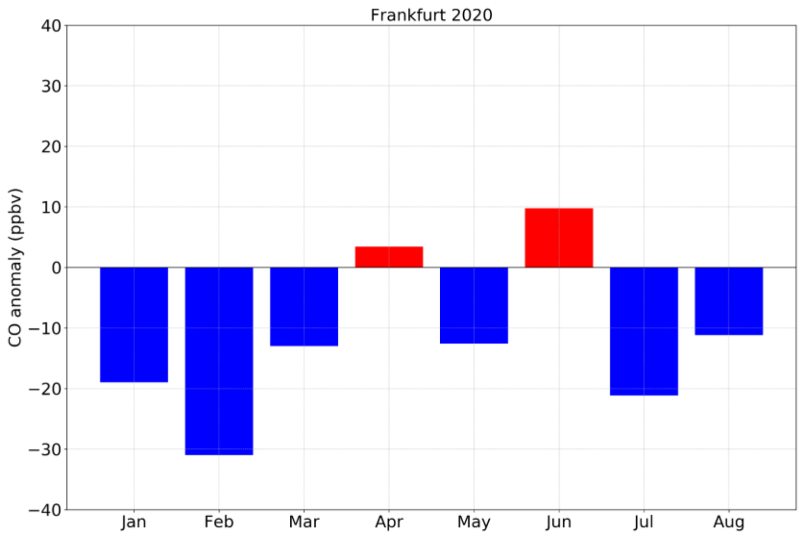
  >We have added confidence limits and the interannual variability to all of the individual monthly bar plots that we discuss calculated on the basis of Student's t test. This means we can identify which anomalies were significant once sampling has been accounted for.

- Attempt to incorporate nearby surface NOx/NO2 measurements to confirm the reduction in NO titration of surface layer ozone (or IAGOS NOx if available).

  >As already mentioned above, we do not have the NOX data available from IAGOS for these flights as the instrument did not fly during this period. The decrease of NO2 at Frankfurt from surface stations and satellites has already been documented by Barré et al 2021. Barré et al controlled for meteorological factors and the estimates of lockdown-induced NO2 changes for Frankfurt were -24% and -33% based on TROPOMI observations and surface stations respectively. We infer therefore that the positive anomaly in ozone at night is linked to this drop in NO2 at Frankfurt and the consequent reduction in ozone titration.

- Integrate boundary layer CO to account for changes in boundary layer height that potentially reduce the surface layer CO mixing ratios in 2020.

>We integrated the CO over the boundary layer as the reviewer suggested. This leads to a negative anomaly of 30ppbv in February and 12ppbv in March and May which can be ascribed to the drop in emissions. We have updated the text accordingly.



Recommendation:

This paper could be considered for publication in AMT if the authors present more compelling evidence that the IAGOS ozone and CO data collected in MAM 2020 were directly influenced by COVID-related emissions changes, and not simply a result of interannual variability, long-term trends in ozone (surface layer increases) and CO (decreases), and meteorological factors (e.g. boundary layer heights). I recommend Major Revisions that include an assessment of the statistical significance of the results.

Specific/Technical and Line-by-Line Comments:

Line 26: Cite Liu, F. et al. (2020) paper for China TROPOMI NO2 decreases: https://advances.sciencemag.org/content/6/28/eabc2992

>Done

Line 27: Cite Duncan et al. (2016) paper, which describes the relationship between economic downturn and NOx emissions/OMI NO2 satellite measurements: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1002/2015JD024121

>Done

Line 26 and/or Line 197: Cite Goldberg et al. (2020) paper, which controls for meteorological variability when examining COVID-related TROPOMI NO2 decreases, https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2020GL089269

>Done on line 33

Line 79: Change "balloon and sonde measurements" to "balloon-borne ozonesonde measurements"

>Done

Line 110: Stylistic comment, suggest to remove "life"

>Done

Line 120: Small typographical error "1° horizontal"

>Done

Line 125: Please define GFAS acronym

>Done

Line 127: "anthropogenic *sources*"

>Done

Line 144: How many profiles in total are averaged into the MAM 1994-2019 ozone climatology? Similarly, if there were no profiles in April 2020, how many of the 84 profiles were from March and May 2020? It might be more proper to indicate March/May rather than MAM in the text and figures. (Also see General Comment about the chosen baseline period for ozone).

>The MAM profile plots now clearly state that this is a March+May average for both the 2020 and the reference period. We have added this to the figure caption. We have also added the number of profiles (220) to the caption.

Line 181: change reservoir to emissions

>Done

Line 189: "in the amount of NO as evidenced by the TROPOMI satellite measurements of NO_2"

>Done

Line 198: I don't understand what is meant by "but that the photochemical effects from NOx were dominant." Is this just referring to reduced titration of ozone from NO? Please clarify.

>We have clarified this in the text. The authors found changes in NOX that could not be explained by meteorology alone, and that were a result of the emissions reductions. All found that there were important and differing impacts of meteorology, but that there were changes in NOx that were unattributed to the meteorological conditions and linked to falling emissions during the lockdowns.

Figures 4 and 5: Is it correct that there are 7 nighttime profiles and 13 daytime profiles in May 2020? How robust is the result of a 41% increase in nighttime surface layer ozone from 7 profiles?

>It is correct that the number of nighttime profiles is 7 and the number of daytime profiles is 13. We now see a 30% increase in the ozone with respect to the shorter reference period. We have added 95% confidence limits to figures 4 and 5 to show how robust the increases were despite the smaller number of profiles.

Line 232: change “seen by balloons and sondes” to “observed by ozonesondes”

>done

Line 235-236: change “balloon and sonde” to “ozonesonde”

>done

Line 236: Suggest to add: “For example, there was no notable decrease in free tropospheric ozone in the sparsely-sampled Southern Hemisphere.”

>done

Line 245: Change “inflexion” to “inflection”

>Done

Figure 8: (Similar to comment for Line 144) Please indicate how many CO profiles are available for MAM 2016-2019

>We have added the number of profiles to the figure caption (300 profiles).

Line 265: Change “biased low” to “anomalously low”

>Done

Line 270: Now I see that there are CO profiles for April 2020, so it would be helpful to indicate if the ozone instrument was inoperable in April 2020 (or whatever the cause is).

>The ozone instrument was not working during April 2020. We have added a line in the text on this.

Paragraph near Line 280: Is all of this discussion necessary? Isn’t the boundary layer height simply calculated at the grid point closest to the Frankfurt airport, where all of the surface layer data are collected? The ECMWF output is a fairly coarse 1° resolution, so I would assume this is the case. Please correct me if I am wrong.

>Yes it is approximately the case. The ECMWF data is interpolated to the position of the aircraft. It will have travelled about 20-50km from the airport over the first 2000m of the atmosphere (Petetin et al 2018). We used a bilinear interpolation in space using a distance weighting from the 4 nearest grid cells to the aircraft position, and a linear interpolation in time. Unless the aircraft is in the centre of the grid cell then the 4 surrounding cells are used. We have added this in the text.

Line 284: Why not be consistent in definitions of day and night as for ozone (10:00-18:59 UTC and 00:00-09:00/19:00-23:59 UTC)?

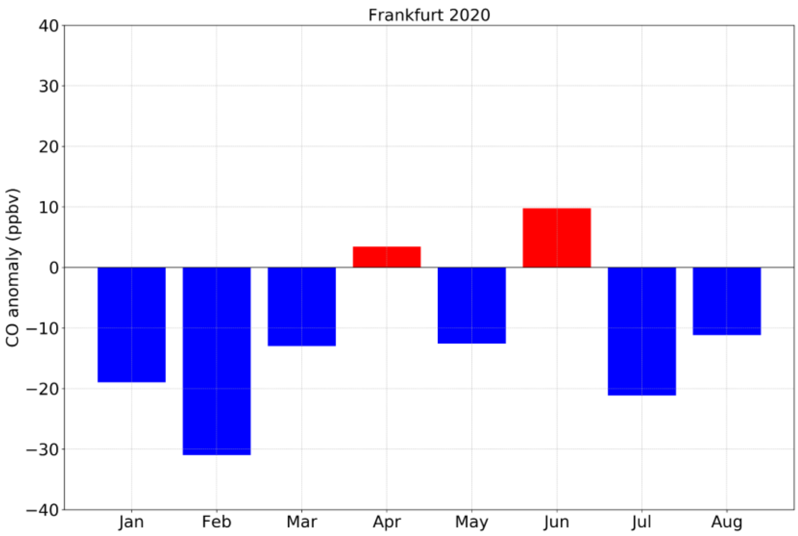
>The referee is right that we did not use the same definition of daytime and nighttime for ozone as for CO, but the objectives of the investigation are not the same. For the ozone, we wanted to account for any bias introduced by an uneven sampling over the diurnal cycle of ozone. For CO we wanted to check the influence of the depth of the boundary layer.

We based our definition of the maximum and minimum phases of the diurnal cycle on the diurnal cycle of ozone over Frankfurt as shown by Petetin et al 2016a. The diurnal cycle of ozone depends on UV chemistry and dynamics. This was perhaps not well explained so we have added clarification in the text about this.

For CO we verified the impact of the depth of the boundary layer on the anomalies of CO as Peuch et al. 2020 found that the boundary layer was anomalously high during the period of interest. The night and daytime heights of the boundary layer are based on dynamics but not on UV chemistry.

Lines 289-290: Given the relatively long lifetime of CO, a simple check on whether decreased CO concentrations near the surface are a result of dilution in a deeper boundary layer would be to integrate the boundary layer CO content. This will simplify discussion and may lead to more convincing results.

>We integrated the CO over the boundary layer as the reviewer suggested. This leads to a negative anomaly of 30ppbv in February and 12ppbv in March and May. We are now more confident that this the result of a drop in emissions. We have updated the text accordingly.



Line 302: Why are you also excluding fire sources of CO? What does that have to do with lockdown decreases in emissions?
>We can say a bit more about the fire sources of the CO. It should be noted that SOFT-IO does not calculate a background value for CO. It is adapted to analysing the origin of plumes that are well defined against the background. This is not our case here. We have quantified the absolute contribution from the biomass burning as requested. However, because the anthropogenic emissions are not updated for the COVID period, we cannot give the relative contributions of biomass burning and anthropogenic emissions. Since the biomass burning contribution decreased in 2020, we expect a higher contribution from anthropogenic sources. We have added some discussion about this in the text related to line 302 and line 336 in the comment below.

Line 304: Please indicate in the text that the trajectories terminate at Frankfurt in the surface layer (>950 hPa).

>We have clarified in the text that the trajectories terminate at the aircraft position within the surface layer.

Figures 12 and 13: Please number the regions on the map and legends so it is easier to identify the source regions. It's a bit difficult to distinguish some of the colors.

>We have added texture to some of the areas to help distinguish the regions.

Lines 336-337: Including information from MACC-City fire source CO should help confirm this hypothesis.

>Yes, in the reference period, the CO from fire sources contributed to 20% of the total, so anthropogenic sources were the primary contribution to CO over Europe. In 2020, absolute amounts from biomass burning have decreased, suggesting a greater contribution from anthropogenic sources. We have added some comments in the text regarding this.

Line 363: The sign in front of 2 ppbv and 1% should be negative, and actually -1.8 ppbv to be consistent with the previous text (Line 315). > *DONE*