

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-121

Anonymous Referee #1

Referee comment on "Unraveling Pathways of Elevated Ozone Induced by the 2020 Lockdown in Europe by an Observationally Constrained Regional Model using TROPOMI" by Amir H. Souri et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2021-121-RC2>, 2021

Souri and co-authors present an analysis of lockdown-induced changes in NO₂, HCHO, and O₃ over Europe based on a data assimilation approach involving TROPOMI measurements (NO₂, HCHO) and the WRF-CMAQ model. An advantage of the approach is that it explicitly accounts for meteorological influences and so forth in assessing the causes of AQ changes during the COVID period.

The paper topic is suitable for ACP and will make a useful contribution to the literature. There are some methodological and science comments and questions that I feel should be addressed before publication; these are listed below. In a number of cases things are described in a confusing way and need to be clarified. Finally, in many places the writing can be clearer or is overly wordy. For example, many of the paragraphs are about a page long and cover multiple topics, which really does not help communication. Once these issues are addressed I recommend publication in ACP.

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Science comments. Numbering refers to line numbers.
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1. The abstract is very long (almost 500 words!) which partly defeats the point of an abstract. I suggest reducing it by approximately half.

56. "Earth's atmosphere has exponentially become more polluted during previous decades". Too vague / sweeping. What do you mean by "previous decades"? Some parts of the world have become significantly less polluted (for PM and ozone) over the last 2-3 decades. Elsewhere I do not think the word "exponentially" is necessarily accurate.

78. "whereas the concentrations of several secondarily formed compounds such as ozone increased due to emissions and/or meteorology". This is not universally true; e.g., see Bekbulat et al., <https://doi.org/10.1016/j.scitotenv.2020.144693>.

767. For reports, please list references in a way that readers can readily access them, e.g. with a doi or persistent URL.

138-139. RMSE already has a definition; what you are reporting here is not the RMSE.

148-150. "There are challenges..." Unclear what you mean here. Please rewrite for clarity.

153. "we uniformly scale up NO₂ pixels by 25% based on the low bias determined by Verhoelst et al. [2021] while considering the potential reduction in the bias through the use of higher spatial resolution trace gas a priori profiles." Not clear what is meant here by "while considering". How was this considered? Do you mean it was considered by choosing 25% rather than the median value of 37% reported by Verhoelst? Or is something different being implied here? Please clarify.

153. The choice of a 25% bias correction for NO₂ seems a bit arbitrary. As I understand it, the argument being presented is as follows: "the bias was reported previously to vary from -23 to -51%, with a median of -37%. But the use of higher-resolution shape factors here should reduce the bias. So, we use +25%." I agree that higher-resolution shape factors will reduce the bias, but there is no quantification of that effect here, so the 25% value seems to be pulled out of a hat. There is also the fact that the TROPOMI bias was shown previously to vary between rural and polluted environments, but this is not accounted for here. Overall, there needs to be either a more rigorous justification for the bias correction being employed, and/or some sensitivity analysis to quantify the degree to which this assumption affects the results.

183-187. A similar comment applies to HCHO. I appreciate that the authors pay close attention to uncertainty and bias in the satellite data. But in the end the employed corrections are chosen a bit haphazardly from the range of reported biases. How can this choice be better justified, or if it is necessarily a little arbitrary, how can the impact of that assumption be quantified?

156. "we set the RMSE to 1.1×10^{15} molec/cm² in clear regions and 3.5×10^{15} in moderately to highly polluted regions." This is confusing because at this point in the text we don't know what is meant by "set the RMSE". We learn later that these values will be used to populate the error covariance matrices for the inversion; please clarify that here so the reader understands what is happening.

199-202. Does this mean that you only use the dark blue product? Please clarify.

The TROPOMI retrievals do not account for aerosols in the scattering weights. Yet I presume that aerosol loadings over Europe changed between the COVID and reference period. To what degree does this bias the retrieval differences and therefore the NO₂ and HCHO comparisons between these periods?

236. "We nudge moisture, wind and temperature fields toward the reanalysis data used only outside of the PBL layer." Wording is unclear, as is the reason for doing this. Please clarify.

239. "Extensive model evaluations based upon surface observations show a striking correspondence". The model temperature bias appears to be 50% smaller in 2020 than 2019 (0.8 vs 1.2 degrees). Does this have any impact on the model interpretation of changes between years? For example, assessing changes in anthropogenic VOCs relies on distinguishing changes in biogenic emissions which depend exponentially on temperature.

239. PBL height is a major factor for model performance in simulating AQ-relevant species. How well does the simulation capture measured PBL depths over your domain?

242. Please state the time resolution at which you are optimizing emissions. I guess there is a single 3-month mean value being derived for each grid cell but unless I missed it I

don't think this is stated anywhere.

250. Please state how the Jacobian is calculated. Is there a finite difference run for every model grid cell, each tracer, and each iteration?

252. "In terms of the prior errors, we use the numbers reported in Souri et al. [2020a]." Since this is an important aspect of the inversion please briefly summarize here.

257. "here we iterate Eq 1 3 times." How do you know this is sufficient? As you know the emission-concentration relationship for NO_x in particular is highly non-linear. Do you employ a test for convergence?

275. "faster vertical mixing due to larger sensible fluxes (more diluted columns due to stronger advection in higher altitudes)". This is a little convoluted. Faster vertical mixing by itself wouldn't change the column amount, and faster winds during summer (really?) would only smear the columns.

277-280. Wording is quite awkward here.

281. "pronounced decreases", please clarify that you mean in 2020 vs 2019.

282. "In contrast, we see negligible reductions..." actually some of the regions mentioned seem to show a clear March increase.

296. "suggests an abrupt hiatus in the ongoing reduced NO_x emissions". Unclear if this means the emissions went into hiatus or the reduction went into hiatus.

317. "but also stems from the fact that isoprene reactivity significantly increases by rising temperature [Pusede et al. 2015]." This is a bit oddly worded; I think you simply mean that OH is increasing seasonally along with isoprene emissions.

335-337. These evaluation statistics should be displayed in SI in a table or figures.

341-344. "However, in practical terms, the magnitude of these anomalies is not as drastic as the ratio of observation to model ratio because of the consideration of observational errors and chemical feedback [Souri et al., 2020a], which always leaves some doubt about the practicality of direct mass balance methods." I am unsure what the authors are trying to say here.

358-360. The optimization naturally improves the simulation of HCHO with respect to TROPOMI, that is the whole point of the optimization. Does it also improve the simulation with respect to independent observations?

358-378. This paragraph is really unclear; I had to read it multiple times to try and parse what is being argued. It sounds like you're arguing that the chemistry changed the emissions. Please rewrite.

395. "Horizontal transport (shown as wind vectors) plays a critical role in explaining the spatial variations in emissions downwind." Why would wind affect the emissions?

397-418. This section is all quite speculative and unconvincing. It does not appear that there is much required information conveyed here, recommend deleting.

409. "This in turn will provide an opportunity for the volume of air to become dispersed". Poor wording. The VOC lifetimes do not affect how a "volume of air is dispersed".

422. "Unfortunately we limit the analysis to NO₂ due to the lack of routinely measured HCHO observations." The HCHO data are ultimately being used to constrain VOC emissions; so are there VOC measurements that can be used for this purpose?

440. "The surface measurements reinforce the less pronounced reduction in NO₂ in northern Germany and UK, although the magnitudes are not as large as those suggested by the model." This is not clear from the figure. For example the observations suggest that decreases over the UK in April and May are quite large compared to the rest of Europe.

492-496. "This apparent discrepancy is caused by the differences in boundary and initial conditions which are not quantifiable by the process analysis and would require additional sensitivity test." Is it just the ICs and BCs, or is it that these processes being examined are not strictly independent and additive?

Equation 6 is incorrect (the wrong rate constant is indicated).

544. "This analysis strongly coincides with Lee et al. [2020] and Wyche et al. [2021] who observed roughly constant O₃+NO₂ concentrations over the UK before and during the lockdown 2020." With this in mind, why not actually just show the modeled Ox = O₃ + NO₂ change (and measured change too, if available)? This seems like the most direct way to make this point.

572. "The reduced anthropogenic VOC emissions were a result of two key assumptions: the reduced NO_x emissions in NO_x-rich areas increased HCHO made from VOCs (evident in larger Jacobians derived from the regional model), and TROPOMI HCHO suggested a negligible difference in HCHO concentration between the two years." Again the wording here is really confusing. It appears to be arguing that changes in NO_x emissions and in the ensuing chemistry changed the actual VOC emission rates. I think I know what is meant (i.e., that these factors change the emission rates that one infers for a given HCHO level) but it really needs clearer description.

Minor edits.

73. "atmospheric composition" not "compositions"

78. "particulate matter"

157 and 185. "clean regions" and "clean areas" rather than "clear"

177. "mainly located" or "predominantly located"

437. "by only considering grid cells"