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Comment on acp-2022-347

Anonymous Referee #1

Referee comment on "Diagnosing ozone–NO_x–VOC sensitivity and revealing causes of ozone increases in China based on 2013–2021 satellite retrievals" by Jie Ren et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-347-RC1, 2022

This study combined satellite observations of NO2 and HCHO with surface air quality measurements over China to characterize ozone formation sensitivities and its long-term changes. A methodology largely consistent with Jin et al. (10.1021/acs.est.9b07785, 2020) were applied to harmonize long-term data from OMI and TROPOMI, and to diagnose the ozone sensitivity regimes. Then changes of satellite NO2 and HCHO were used to interpret the recent ozone increases, which is mainly attributed to NO2 reductions while VOC changed little in the context of the VOC-limited regime. Two examples showing ozone responses during COVID-19 lockdown were discussed to complementally support their arguments.

While the topic is definitely within the scope of ACP and the methodology & results are clearly written, easy to follow and overall sound, I have moderate reservations regarding publishing the manuscript at its present form. The main issue is about its novelty which I will outline later. I will support the publication of this paper, if the following concerns can be adequately addressed.

Major points:

- Novelty. The whole idea of using satellite observations of HCHO and NO2 to identify an indicator of ozone formation regime and to interpret ozone changes, as the authors also described, has been proposed and applied for ~two decades. In particular, I can easily list two recent ACP papers that essentially used the same idea, almost the same data and processing, applied to the same domain (China) and overall consistent time period (since 2010s), and came up with consistent conclusions: Wang et al. (10.5194/acp-21-7253-2021, 2021) and Li et al. (10.5194/acp-21-15631-2021, 2021). I merely found substantially novel/additional insights from this manuscript relative to these two papers. In the next round the review, the authors should highlight the unique points in their manuscript relative to these two papers to justify that their paper is substantially novel.

- Potentially unnecessary data processing. The idea of "harmonization" of OMI and TROPOMI data might be borrowed from Jin et al. (10.1021/acs.est.9b07785, 2020), but it is odd to be used here for 2013-2021, which is already fully covered by OMI. The "harmonization" process will introduce potentially more uncertainties, especially considering the short time period (thus the climatological differences between the two sensors and the adjustments based on that will be more contaminated by meteorological anomalies). Furthermore, I do not see any unique insights/interpretations that are only available at <20 km spatial scale in the manuscript. If the "harmonized data" is still used in the revision, the authors should evaluate how different the results become if only using OMI data, and justify that these differences are strong and due to the unique information from TROPOMI.
- The COVID-19 analysis. Section 3.4 is confusing to me. First, the main topic is to discuss long-term changes (and maybe relevance with emission regulations), I believe this short-term ozone responses to NO2 and HCHO do not support the long-term analysis before. Second, the Chinese lockdown is during February and March, 2020. The authors selected April for Beijing, and May for Chengdu, why? Indeed, both NO2 and HCHO do not show the "COVID-typical" reductions to me. Third, can the current analysis for one month in three years, each year with their unique meteorological conditions/variations, really support the attribution of these ozone responses to be driven by emissions? Including more years that potentially envelope possible meteorological variabilities seem more reasonable to me.
- Strong arguments about potential PM effects. The authors concluded that "Our study highlights that the root cause of ozone increase in major regions is the significant reduction of NOx alone without effective control of VOC and not the concurrent decreases in the PM2.5 level as suggested in previous studies". However, their results cannot support this argument. PM2.5 and NOx decrease simultaneously during the investigated period, therefore ozone increases are also associated with PM2.5 reductions. Whether the chemical regime is NOx-limited or NOx-saturated does not rule out the PM effects, since uptake of HO2 will affect both regimes according to Li et al. (2019). If the authors would like to retain their strong arguments that PM is not affecting the ozone production, they will need to validate that ozone at similar HCHO and NO2 level (e.g. bins in Figure 3a, with meteorological effects also minimized/normalized) stays the same over time.

Minor points:

1) The "x" in "NOx" should be a subscript.

2) Line 63-64: "highly controversial" due to one paper finding inconsistency over one site?

3) Beijing and Chengdu are selected to look at COVID-19 effects (Figure 7), and Beijing,

Chengdu and Guangzhou are used to investigate optimal emission regulation (Figure 8). Why are these cities selected? Can they represent other cities?

4) Line 81: The "reference state" was at 273 K before September 2018, and at 298 K afterwards. Is this factor considered? Should be included in the introduction.

5) Line 97-103: please provide more detailed introduction of the data. How the re-gridding is done? What is the temporal resolution of data used?

6) Line 118: Please provide a map of the 9 regions for people unfamiliar with geography of China.

7) Figure 3a: Some isopleth lines will greatly help guide the audience.

8) Figures 2/4/5/6: Are the annual maps really necessary? Maybe one map for each phase (2013, 2019, 2021) will be enough?

9) Figure 7: Without e.g. a modeling framework to isolate each contribution, just listing all the monthly-average numbers of these parameters cannot support the discussion in Section 3.4

10) Figure 8 and its relevant discussion: It is unusual to introduce more results in the last section. Please consider re-organize.

References:

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Li, R., Xu, M., Li, M., Chen, Z., Zhao, N., Gao, B., and Yao, Q.: Identifying the spatiotemporal variations in ozone formation regimes across China from 2005 to 2019 based on polynomial simulation and causality analysis, Atmos. Chem. Phys., 21, 15631–15646, https://doi.org/10.5194/acp-21-15631-2021, 2021.

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