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Reply on CC2

Juan Cuesta et al.

Author comment on "Ozone pollution during the COVID-19 lockdown in the spring of 2020 over Europe, analysed from satellite observations, in situ measurements, and models" by Juan Cuesta et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-785-AC3, 2022

Responses to reviewers on
the manuscript "Ozone pollution during the COVID-19 lockdown in the spring 2020 over Europe analysed from satellite observations, in situ measurements and models" by Cuesta et al.

We would like to thank the reviewers for their suggestions that have improved the clarity of the paper. All their remarks have been addressed in the Revised Manuscript (RM). Their main suggestions have been followed by:
(i) Shortening and simplifying the paper and the figures (as requested by Referees #1 and #2), through suppressing from the main manuscript the results of the version C2 of the CHIMERE model, only showing results for only one period (1-15 April) and mainly in terms of daily averages. A single exception to this is one figure, which is important for comparing the results of the current paper with other publications. Some of the withdrawn results have been moved to the supplement as suggested by referee #1.
(ii) An additional figure (Figure 8) is added that briefly describes the link with the variability of ozone at the free troposphere and the link with the stratosphere, as well as the associated meteorological conditions at the free troposphere (as suggested by Referee #2).
(iii) A new figure (Figure 7) addressing the point stated by Amir Souri concerning the influence of meteorological conditions in near surface ozone over the Iberian Peninsula
(iv) Two additional Figures (Figs. 4b and 5d) and Table 3 show the model smoothed by the averaging kernel of the satellite approach and the direct comparison with respect to satellite data, as requested by Referee #1.

These main points are described in detailed in the following paragraphs, as well as all additional and specific points remarked by the Referees. All other points have also been clarified and addressed thoroughly

Comment from Amir Souri:

The manuscript caught my eye because the analysis focused here is on the same time period, the same area, and the same atmospheric compounds as those in Souri et al. [2021]. Our draft was cited more than 16 times in the current manuscript, extensively expressing a strong degree of agreement, particularly in terms of the surface ozone enhancement over central Europe. Nonetheless, there are two striking differences in this study compared to those in Souri et al. [2021], Ordóñez et al. [2020], and Barré et al. [2021], both of which were not thoroughly justified. A disagreement is far more interesting than an agreement, but if it results from a negligent model (or faulty data), it should be rectified. Two substantial differences follow:

1. Surface MDA8 measurements based on UV photometry (which are highly accurate) show a large reduction in the Iberian Peninsula (called southwestern Europe in the manuscript). This tendency coincided with Souri et al. [2021] and Ordóñez et al. [2020]. The studies of Souri et al. [2021] and Ordóñez et al. [2020] came to the same conclusion showing that the large reduction of surface ozone was due to meteorology. In particular, Souri et al. [2021] showed that the anthropogenic factor played a very marginal role in shaping the decline over the area. The major reason behind the reduced ozone is assumed to be cloud causing photochemistry to dampen, shown in Figure 2 in Ordóñez et al. [2020] and the last column of Figure 6 (the ratio of photolysis rate below clouds to a clear-sky) in Souri et al. [2021]. Our analysis using TROPOMI NO2 showed that the frequency of the satellite observations is 2.5-3 times as large in April 2019 as those in April 2020 due to more overcast in the latter. The numerically resolved P(O3) values (Figure 12 in Souri et al. [2021]) were found to be relatively low over the Iberian Peninsula suggesting that the anthropogenic emissions are not sufficiently high enough to become the main driver of ozone anomalies observed by surface measurements. This manuscript, on the other hand, claims that 4.5 ppbv out of 5.0 (90%) of the reduction in the MDA8 surface ozone is solely due to the emissions (Table 3). How did the models perform with respect to cloud optical thickness and cloud fraction? It may be worth calculating J values (an approximate value can be derived based on
https://www.cmascenter.org/cmaq/science_documentation/pdf/ch14.pdf)

Clarified and a figure added. We appreciate this interesting remark. For clarifying this aspect, we have added an additional figure in the RM (Fig. 7) presenting the meteorological conditions used by CHIMERE including ozone photolysis rates, surface temperatures and winds, and mixing boundary layer heights. Ozone photolysis rates are calculated according to cloud cover within the CHIMERE model package code. The new figure shows the changes in the meteorological conditions between 2020 and 2019 that can be related with the negative anomaly of surface ozone over the Iberian Peninsula simulated by CHIMERE in 2020 with respect to 2019 (using in both cases with the same standard emission inventories, Fig. 4b). This reduction in 2020 is clearly co-located with reduced ozone photolysis rates, which is associated with enhanced cloudiness. However, other meteorological conditions likely produce the opposite effect: enhanced surface temperatures and lower windspeeds in 2020 are expected to favor ozone production and shallower mixing boundary layer heights to inhibite turbulent vertical mixing thus inducing a relative enhancement of surface ozone concentrations in 2020. These effects are expected to compensate between them and thus CHIMERE simulations suggest that the anomaly of ozone surface concentration in 2020 associated with meteorological conditions is only moderate over the Iberian Peninsula (-2.4% for the southwestern region in Table 3).
This is added in the RM (lines 434-447) as "Other factors significantly affecting simulated concentrations of ozone and its precursors are clearly linked to the meteorological fields used by the model. This is shown in terms of changes on 2020 with respect to 2019 of ozone photolysis rates, surface temperatures and winds, and mixing boundary layer heights used by CHIMERE (Figure 7). Two distinct behaviors are clearly observed over the continent north of 44°N and over the Iberian Peninsula. North of 44°N, anticyclonic conditions prevailing in 2020

induced clearer sky conditions (thus enhancements of ozone photolysis rates), higher surface temperatures and lower windspeeds, which clearly favor photochemical production of ozone. This explains the frank positive anomaly of surface ozone over this region visibly simulated by CHIMERE, accounting (Fig. 3a) or not (Fig. 4a) for the emission changes during the lockdown. Over the Iberian Peninsula, reduced ozone photolysis rates (Fig. 6a) associated with enhanced cloudiness in 2020 is likely at the origin of the meteorology-associated decrease of ozone concentrations (Fig. 3b). However, other meteorological conditions likely produce the opposite effect: enhanced surface temperatures and lower windspeeds in 2020 are expected to favor ozone production and shallower mixing boundary layers to inhibit turbulent vertical dilution of ozone, thus inducing a relative enhancement of surface ozone concentrations in 2020. These effects are expected to compensate between them, explaining the moderate reduction of ozone simulated by CHIMERE over this region (-2.4% for the southwestern region in Table 5)."

2. Souri et al. [2021] and Barré et al. [2021] observed a large reduction from in-situ and TROPOMI tropospheric NO2 columns measurements over central Europe in March-April-May 2020 with respect to a reference (e.g., 2019). Figure 8 shows a substantial enhancement of NO2 over Germany in this manuscript, which strongly contradicts two other studies. This discrepancy has not been well justified. While we did see some disagreement between the satellite and the surface observations, it is highly unlikely for the surface measurements to be substantially different among all these studies. Please double-check the data (their validity flag) or your code to see if this is caused by a bug. If this is true (which is extraordinary), please dedicate a paragraph to discuss why.

Clarified. We have double-checked the validity of the in-situ surface dataset used in Figure 8 and flags indicate that they are valid measurements for background stations (of all categories urban, suburban, and rural). Positive NO2 anomalies in 2020 over Germany are particularly observed during the period 1-15 April (Figure 8) but are less marked for the average over the whole month of April (as analyzed by Souri et al. 2021 and Barré et al; 2021). They are also simulated by CHIMERE C2, at the surface (Fig. 8e of the original manuscript) and particularly evident for total columns (Figure 9d of the original manuscript). Using meteorology-only anomalies (with the same inventories), CHIMERE simulations show such anomalies strong positive anomalies very clearly (not shown). They are also observed in TROPOMI measurements of Souri et al. 2021 (figure 4), which show a full horizontal coverage. We think that the reason why this is not observed in the surface in situ figures from Souri et al. 2021 and Barré et al; 2021 is likely related to the period of analysis (the whole month of April for the previous studies and 1-15 April for the current) and also the choice of the in-situ stations which is probably much more restrictive is those publications and in the current one. Since the CHIMERE model and TROPOMI measurements also show positive anomalies of NO2 over Germany in 2020 as compared to 2019 and during the period 1-15 April, the surface in situ measurement we show are consistent with them.
We have added the following statement in the supplement of the RM "Additionally, surface measurement for some stations over Germany show a positive anomaly of NO2. This seems to be simulated by CHIMERE C2 at the surface and particularly evident for total columns, likely linked with meteorological conditions. These NO2 enhancements are particularly seen in the period 1-15 April, which could partly explain why they are not depicted by Souri et al. (2021) and Barré et al, (2021) for the whole month of April (as well as the particular choice of the in-situ stations for each of these studies).".