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Reply on RC2

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-AC4, 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 Some of the withdrawn results have been moved to the supplement as suggested by referee #1.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.

Responses to comments from Referee #2

Overview

The paper deals with the influence of the COVID pandemic on ozone pollution over Europe during the general lockdown in spring 2020. In my opinion, the manuscript is generally scientifically sound, presenting a lot of interesting information and analysis regarding in-situ and satellite measurements as well as modelling simulations during the lockdown period and it deserves publication in ACP, in principle.
Although the manuscript content is interesting, I think that the presented information to too dense making the paper reading rather difficult. For this reason, I would encourage the authors to try to reduce the length of the paper and be focused on the most essential points. I would suggest indicatively to reduce the modelling part by presenting only one modelling simulation including, if possible, the more recent emission inventory as well as the more important vertical mixing (see also comments below).

Agreed and manuscript shortened. As suggested by both referees, we have substantially shortened the main RM, limiting the description of the results in terms of one model setup (the CHIMERE C1 simply called CHIMERE), one period (1-15 April with the greater changes in pollutant emissions during lockdown) and one main metrics for surface ozone (daily averaging). These aspects are more detailed in the second response to referee #1. The CHIMERE model setup C1 is chosen as it shows a clearly better agreement with observations. Unfortunately, this setup is based on a version of the CHIMERE code which is not compatible with the most recent emission inventory. Setting up CHIMERE C1 with such inventory would require a very heavy coding effort. On the other hand, the paper shows the influence of vertical mixing in the atmosphere for the two model setups is clearly larger than that from the year of the emission inventory. The C1 setup uses a vertical mixing scheme which provides simulations in better agreement with observations of both NO2 and O3 surface concentrations.

In addition, the use 8h-average maximum daily surface ozone concentrations would be sufficient and more appropriate as this parameter is more representative than the corresponding daily averages for comparison with the free-tropospheric satellite measurements.

Clarified. Additional results provided in the RM (Table 2) show that IASI+GOME2 retrievals of LMT ozone provides overall consistency with both daily averages and MDA8 (maximum daily average over 8h) of surface ozone. Daily averages of surface ozone show the same variability of IASI+GOME2 data and an average difference (or shift) of about – 8 ppb (being satellite-derived concentrations lower). On the other hand, MDA8 surface ozone concentrations present a variability 20 % larger than IASI+GOME2 retrievals, a larger average difference in absolute values of – 12 ppb and a slightly higher correlation. This last might be associated with the larger variability of MDA8 data. Regional positive and negative anomalies between 2020 and 2019 over Europe are very similar for both MDA8 and daily averages (see below). We have chosen to avoid redundancy and only show results in terms of daily averages of surface ozone (whose variability and average concentrations are closer to the satellite data).
We have added the following clarification in the RM (lines 195-197) "Surface MDA8 concentrations are closely linked with the daily maximum that occurs within the mixing boundary layer. These values show larger variability than satellite data and their average values present greater differences than with respect to surface daily averages. Therefore, these last ones are used for comparisons with IASI+GOME2."

General comments

Some remarks regarding the analysis of observational data are presented below, which I think that they would improve the clarity of the interpretation of the results:

As the IASI+GOME2 satellite measurements are most sensitive at 2-3 km height, it has to be noted that based on relatively recent publications (Kalabokas et al., 2013; Doche at al., 2014; Zanis et al., 2014; Akritidis et al., 2016; Kalabokas et al., 2017, Gaudel et al., 2018), the variability of free tropospheric ozone over Central Europe and even more over the Mediterranean basin could be better understood if the variability of synoptic meteorological conditions, affecting especially vertical ozone transport are taken into account. This process would allow the assessment of the influence of either higher tropospheric layers, usually richer in ozone, or boundary layer, usually poorer in ozone. Based on the above, I would suggest examining, at least, the corresponding charts of Geopotential height, vector wind speed and omega vertical velocity for the lockdown period as well as their anomalies relatively to the average long-term climatology. By checking these charts, it comes out that in fact the two examined periods in April 2020 were very different from the meteorological point of view than the corresponding periods for April 2019 as higher atmospheric pressures and temperatures associated with enhanced downward vertical transport and indicating strong tropospheric influence to the boundary layer and to the surface were observed over most of the European continent, with the exception of its southwestern and southeastern parts. In these areas (Iberian Peninsula and Eastern Mediterranean) upward air movements were observed suggesting boundary layer influence to the free troposphere, which are usually associated with lower ozone concentrations. I think that this information might help explaining better the differences in ozone levels observed in Tables 2 and 3, over the examined European areas.

Agreed and two new figures added. We agree that the variability of ozone at the free troposphere may also be an important factor influencing near surface ozone, while being modulated by vertical mixing in the troposphere. For assessing its influence on the ozone anomaly between 2020 and 2019, we have added a new figure in the RM showing IASI+GOME2 measurements of ozone anomalies at the upper troposphere and at the stratosphere, the tropopause height and geopotential heights and winds. These new elements suggest that near-surface ozone is likely influenced by a rather large-scale reduction of ozone in the free troposphere in 2020 as compared to 2019. This reduction is partly associated with a large-scale reduction of ozone precursor emissions over the northern hemisphere (linked to lockdowns in numerous countries during that period) in consistency with Steinbrecht et al. (2021) and over northern Europe also due to less abundant stratospheric ozone.

The influence of meteorological conditions was discussed in the original manuscript and the RM extends further these discussions. The enhanced anticyclonic conditions in 2020 with respect to 2019 are particularly seen north of 44°N by increased geopotential heights and lower windspeeds at 850 hPa. This situation favor subsidence and thus vertical advection of airmasses from the free troposphere down to the atmospheric boundary layer. As remarked by referee #2, two distinct behaviors are clearly observed over southern Europe (particularly the Iberian Peninsula) and other European countries further north, respectively corresponding to reductions and enhancements of surface ozone in 2020 as compared to 2019. This is shown in Figure 3b of the RM in terms of ozone anomalies simulated in 2020 and 2019 with the same emission inventory. This reduction of surface ozone over the Iberian Peninsula is probably linked to a clear reduction of insolation (enhanced cloudiness). However, this is likely limited by the effect of enhanced surface temperatures and lower windspeeds that would favor ozone production and by inhibited turbulent vertical mixing associated with less deep boundary layer heights. Inversely, the ozone enhancement in Central Europe is associated with a prevailing enhancement of

photochemical production of ozone in clearer sky conditions, higher temperatures and lower windspeeds. Higher boundary layer heights in Central Europe are expected to enhance turbulent vertical mixing, thus reducing surface concentrations, and limiting the net enhancements of these concentrations.

These remarks are provided in the RM as (lines 434-465) : "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).

Furthermore, the variability of ozone at the free troposphere may also be a significant factor influencing near surface ozone, depending on vertical mixing. The enhanced anticyclonic conditions in 2020 with respect to 2019 are particularly seen north of 44°N by increased geopotential heights and lower windspeeds at 850 hPa (Fig. 8a). This situation favor subsidence and thus vertical advection of airmasses from the free troposphere down to the atmospheric boundary layer. This is less clearly noted over the Iberian Peninsula and Eastern Mediterranean, where a transition between lower and higher geopotential heights is seen (Fig. 8a). Ozone anomalies at the upper troposphere are depicted by IASI+GOME2 retrievals between 6 and 12 km in Figure 8c. They mainly reveal an overall reduction of ozone concentrations in 2020 with respect to 2019, particularly over the North Sea and the Central Mediterranean. This is probably related with the large-scale reduction of free tropospheric ozone in 2020 observed by Steinbrecht et al. (2021), mainly related with the lockdowns-associated drop of precursor emissions over the northern hemisphere. Downward mixing of these ozone poorer airmasses probably contributes to the large-scale reduction of ozone observed at the LMT by IASI+GOME2 and its negative shift with respect to surface concentrations (Figs. 1 and 2a). Indeed, the only geographically coincident patterns observed both at the LMT and the upper Troposphere are the ozone reductions of ozone over the Mediterranean and the North Sea.

At the upper troposphere, a near zero variation is observed over North-eastern Europe and an ozone enhancement over Western Iberian Peninsula (Fig. 8c). This last one is probably associated with coincident lower tropopause heights (Fig. 8b), thus with a relatively larger contribution of stratospheric ozone. Over the North Sea, the reduction of upper tropospheric ozone at 6-12 km of altitude is strengthened by a depletion of stratospheric ozone occurring in 2020 (see in Fig. 8d as ozone anomalies with respect to 2019)."

Specific comments

Figures 1, 2: As mentioned above, given the best sensitivity of the IASI+GOME2 satellite at 2-3 km altitude, I would suggest using mid-day ozone concentrations (like MDA8 used in later Figs), instead of morning ozone corresponding to the satellite passage time, for a more representative comparison between in situ surface and free tropospheric satellite ozone measurements, as at mid-day the tropospheric influence to the boundary layer gets its maximum, minimizing at the same time the effects of NOx titration and dry deposition on ozone concentrations.

Clarified. We appreciate this suggestion. We have remarked a good consistency between IASI+GOME2 data of LMT ozone and both MDA8 and daily averages of surface ozone. We have preferred to use daily averages of ozone concentrations as they include the mi-day variations and show a better match with IASI+GOME2 satellite retrievals of LMT ozone, in terms of variability and mean values. This aspect is better discussed in the second answer to referee #2.

In relation to the above comments and given the high meteorological variability between the examined years, the comparison of the year 2020 with the 3-4 previous years would be more representative, instead of 2019 alone. It could be at least shown for the surface in-situ measurements, as I understand that for satellite measurements it would be a heavy task.

Clarified. We agree that it could interesting to do such comparison with several years before 2020. However, we expect redundancy with respect to our current results comparing 2020 and 2019. This is shown by the comparison of our results and those from Ordonez et al., 2020, that use a 5-year period as reference. Both datasets show a clear consistency in terms of anomalies of surface ozone between 2020 and 2015-2019 and those between 2020 and 2019, differing by some large-scale shift in the background. Moreover, comparing 2020 with several years before would require a too heavy task of running the CHIMERE model and processing multispectral satellite data.