Response to Reviewers

We thank the reviewers for the constructive comments and suggestions which have helped us improve the manuscript. Below we give full detailed answers to each issue raised by each reviewer. Our response is in blue, to differentiate from the comment which is in black. Furthermore, we include any new text added in the manuscript in red, to facilitate this second revision.

To summarize the main changes to the manuscript, we would like to point out:

1. We have expanded the discussion section to include 1) a more detailed description of the two different photochemical regimes that describe the sensitivity of O3 to its precursors, 2) a new figure entitled “O3 concentration as a function of VOC/NOx concentration.”, 3) more description of the trajectory experiments using the FLEXPART-WRF model and 4) further justification of the chemical scheme chosen for the simulations.
2. We have rewritten the main text to clarified that changes in the oxidation capacity are related to O3 concentrations given that VOC and CO oxidation by OH are the initial reactions for ozone formation and we have expanded the discussion of the oxidation capacity.
3. We have added more references to support the main text when introducing the tropospheric ozone and O3 photochemical regimes.

Response to comments of Reviewer 1

This is a very nice analysis that provides a lot of useful information and insight regarding the production of ozone associated with the reduction of anthropogenic emissions during the COVID-19 pandemic, as well as the changes in the chemical regime associated with it.

Response: We thank the reviewer for his/her comments. Below are our point-by-point replies to each specific comment raised by Reviewer 1.

Specific comments

Line 22: Add more recent references as Fleming et al (2018), Sillman et al (2021)

Response: Thank you for suggesting more references. These two references have been added to the updated manuscript.

This section now reads: Tropospheric ozone (O3) is a radiatively active gas that acts as an oxidizing agent and a surface pollutant in urban areas, where it is a major component of photochemical smog and causes a number of respiratory health effects (Sillman, 2003; Anenberg et al., 2010; Fleming et al., 2018; Sillmann et al., 2021).

Line 70: remove 70

Response: We have corrected this typo.

Section 2: some of the discussion belongs to Introduction.

Response: Thank you for this comment. We have moved some of the discussion from Section 2 to Section 5.3 as suggested by Reviewer 2 (see next comment). We think that the rest of the text belongs to this section because describes our case study.

Lines 174-185: the discussion could be part of the supplementary material.

Response: Thank you for this suggestion. We have moved some of the discussion from Section 2 to Section 5.3 as suggested by Reviewer 2 which we found more appropriate than the Supplementary material as you suggest because that is where the discussion of the trajectory experiments is described.

The revised Section 5.3, now reads:

Figures 9 and 10 show the trajectories of the air masses arriving at the monitoring stations on the selected days, which were modelled with the Lagrangian particle dispersion model FLEXPART-WRF (Brioude et al., 2013). This version of the Lagrangian model works with the WRF mesoscale meteorological model, with the same parametrization as the WRF-Chem model (see section 3.1). The transport model has been run in backwards mode, which means that what is represented in each plot is the residence time, at each grid cell of the map, for the air masses arriving at each site. Twenty-four-hour back trajectories were calculated for each day at a release time of 16 h and with a grid cell size of 0.03 x 0.03 degrees. Figures 9 and 10, show that the air masses on the 3rd of April and 22 of May were transported from the AMB to rural areas such Montseny and the Vic Plain, and we can see an influence from the bottom layers (0-300 m) and the upper layers (300-2000 m) at the different sites. The air masses on the 6th of April were channelled from the AMB northwards to Montseny, the Vic Plain and the Pyrenees. The air masses on the 26th of May were also transported from the AMB northwards to Montseny, the Vic Plain and the Pyrenees, but the air masses that arrived at the surfaces of these locations had strong local components and larger influences from the upper layers.

Line 193: Fig 2 is refereed first time after Figs. 3 and 4

Response: Amended. We have changed the number of Figures in the updated manuscript: Figure 2 is now Figure 4 and Figures 3 and 4 are Figures 2 and 3, respectively.

Section 3.1: Mar et al (2016), Im et al (2016) showed that RADM2 underestimates the O3 concentration when compared to other chemical mechanisms. A discussion about the choice of chemical mechanism would be beneficial since it looks like the Authors obtained the right answers for the wrong reasons.

Response: Thank you for pointing this out. The chemical mechanism RADM2 has been successfully used in several studies of air quality in Europe (Im et al., 2015; Tuccella et al., 2011, Badia et al., 2021). In particular, the RADM2 chemical mechanism has been used in Badia et al., 2021 over the Metropolitan Area of Barcelona.

From Mar et at., (2016):

- *Model biases for O3 in both the MOZART and RADM2 simulations are in line with biases found in other regional modeling studies for Europe.*

- *The temporal correlation with hourly measurements for O3 in this study are also in line with other regional modeling studies of O3 for Europe.*

From Im et al (2016):

- *All models capture, reasonably well, the shape of the domain-averaged annual diurnal cycle of O3 over both domains, while the sub-regional temporal variability are simulated from moderate to good depending on the season and the sub-region that the particular model is configured for.*

Having said that, we have expanded the description of the choice of the chemical mechanism.

Section 3.1, lines 195-198: The chemical mechanism RADM2 has been broadly used in modeling studies of the air quality over Europe (Im et al., 2015; Tuccella et al., 2011, Badia et al., 2021) and its model biases for NO2 and O3 are inline with other air quality modelling studies over Europe (Im et al., 2015, Mar et at., 2016). In particular, the RADM2 chemical mechanism has been used in Badia et al., 2021 over the AMB.

Section 3.3: Please check the numbers in the Tables, not always the MB=MM-OM

Response: Thank you for pointing this. The numbers have been checked and updated in the manuscript.

Lines 301-314: A lot of this information should go to the Figures caption (e.g. “The dots in the lower row represent the land use for each grid cell, which is the key to understanding how industrial, open urban, compact urban, water, agriculture, natural open and forestland uses influenced the O3 regimes”)

Response: Thank you for this comment. We have rewritten this part and moved information to the Figures caption.

Section 5.1, lines 301-303: In addition, the land use is the key to understanding how industrial, open urban, compact urban, water, agriculture, natural open and forest land uses influenced the O_3 regimes (see Figure S11 and Table S9 in the Supplement for more detail on the land use classification).

Figure 4 caption: Modelled O3 concentrations (top panels) for 30 March to 12 April (only weekdays) and 18 to 30 May (only weekdays) for both simulations, BAU (left panels) and COVID (right panels), over the AMB area during the morning (6-8 UTC). Each dot of the top row corresponds to the O3 concentration difference (ppb) of one grid cell of the AMB at the surface level. The dots in the lower row represent the land use for each model grid cell.

Line 315: please specify the land-use categories that belong to “green areas”.

Response: Thank you for pointing this out. Green areas (forest, natural open and agriculture) are described later in the text (line XX). However, we have rewritten the text to clarify that in line 315 we are talking about “urban forest”:

Section 5.1, lines 318-320: Overall, without any reduction in emissions (BAU simulation), this analysis indicates that in urban forests far from anthropogenic sources and influenced by high biogenic VOC emissions, the photochemical regime of O3 formation is NOx-sensitive in the mornings and afternoons.

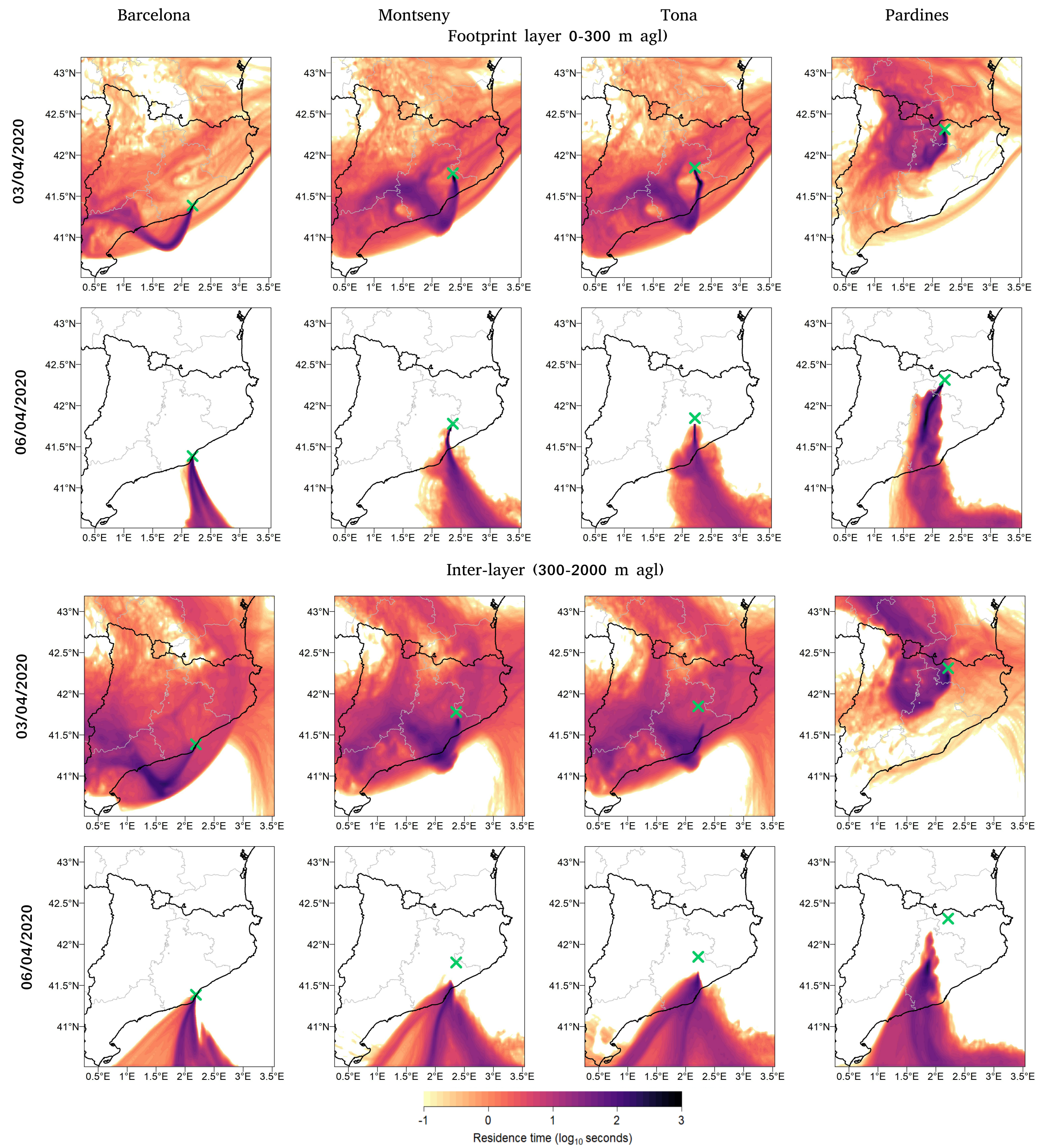
Section 5.1, lines 321-322: Consequently, we found a transition to a VOC-limited regime in green areas (forest, natural open and agriculture) in the evenings.

Figures 3-4: Increase the size of the cross and explain what it represents.

Response: We have increased the size of the cross and the add more information into Figures 3-4 caption.

Figure 3-4 caption: Simulated air parcel trajectories at the footprint layer (0-300 m agl, top panels) and interlayer (300-2000 m agl, bottom panels) for days 3 and 6 of April at 16 h at the four sites (from left to right): Barcelona, Montseny, Tona (Vic plain) and Pardines. The location of each site is shown with a green cross.

In the updated manuscript, Figures 3-4 are:



Barcelona
Montseny
Tona
Pardines
Footprint layer 0-300 m agl)
03/04/2020
06/04/2020
Inter-layer (300-2000 m agl)
03/04/2020
06/04/2020
43°N
42.5°N
42°N
41.5°N
41°N
0.5°E
1°E
1.5°E
2°E
2.5°E
3°E
3.5°E
-1
0
1
2
3
Residence time (log_{10} seconds)

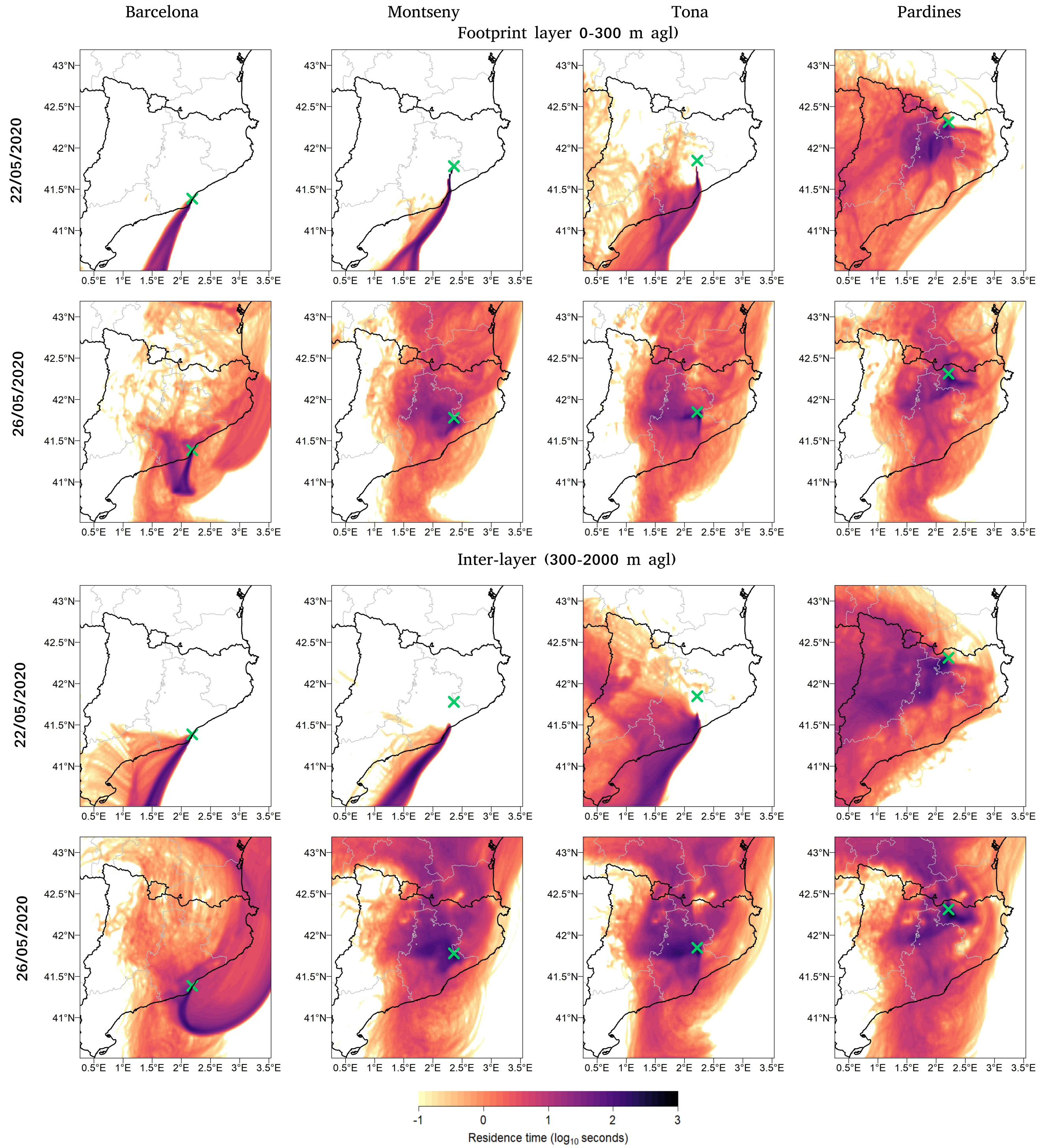
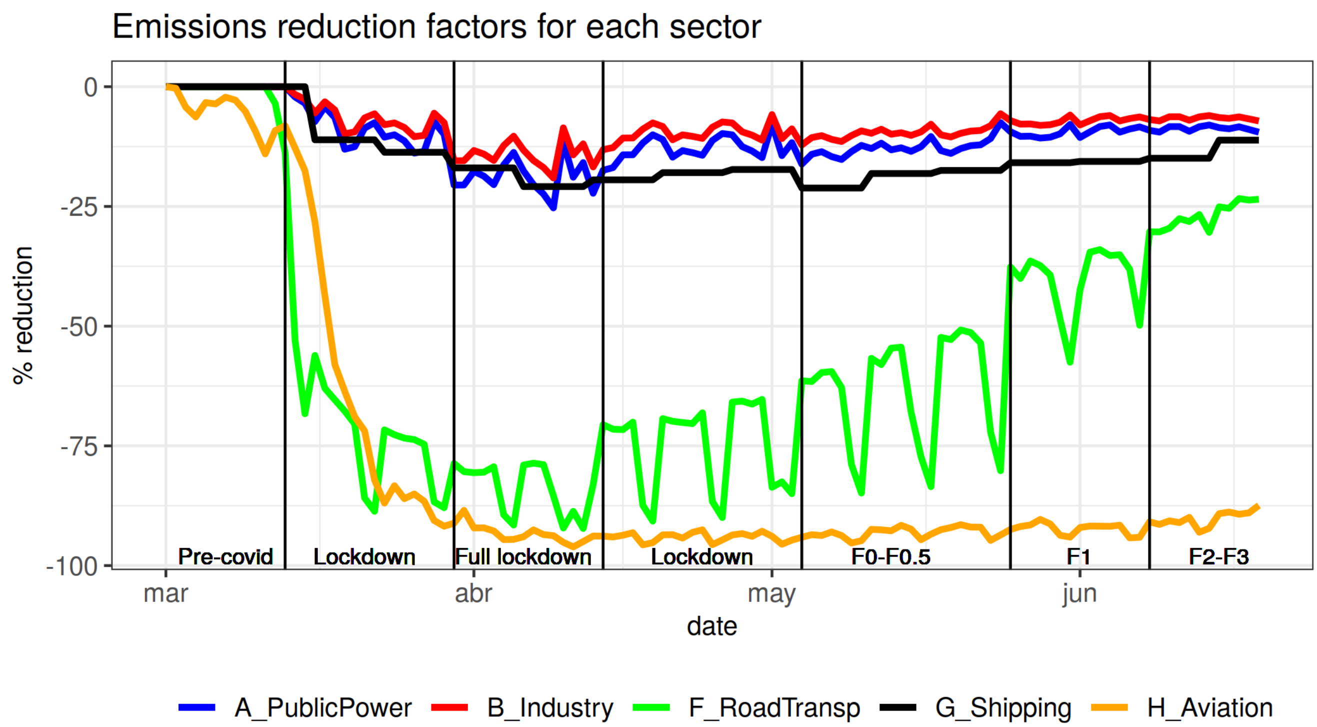
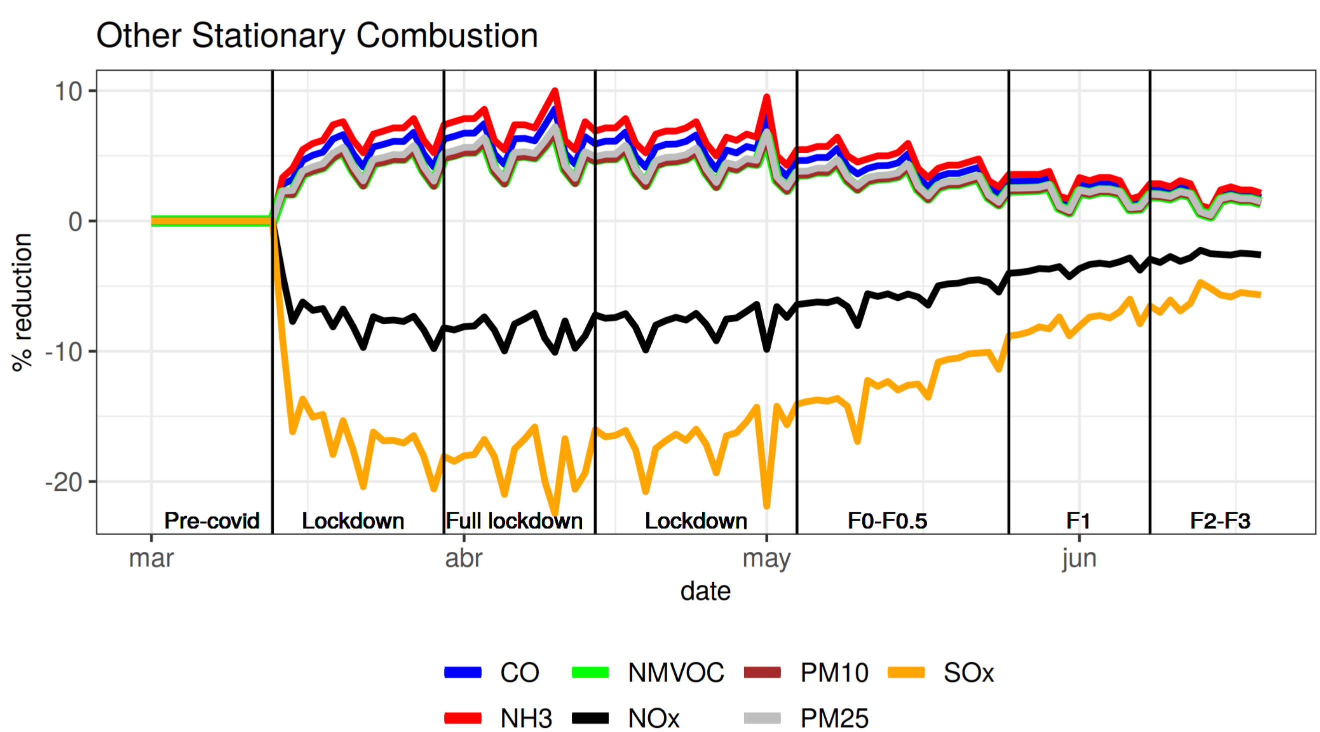

Barcelona
Montseny
Tona
Pardines
Footprint layer 0-300 m agl)
Inter-layer (300-2000 m agl)
22/05/2020
26/05/2020
22/05/2020
26/05/2020
43°N
42.5°N
42°N
41.5°N
41°N
0.5°E 1°E 1.5°E 2°E 2.5°E 3°E 3.5°E
-1 0 1 2 3
Residence time (log10 seconds)

Figure 5 Sectors A and G, B and H, as well as the pollutants CO and NOx and NH3 and PM10 have similar colors and it is difficult to distinguish between different lines.

Response: Amended. We have changed the colors and in the updated manuscript Figure 5 is:





Figures 6-8 As before, we can’t really distinguish the colors. I would suggest using a discrete color scale.

Response: We use a discrete color to display the land-use for each grid (bottom panels). However, we think the ozone concentrations can not be represented in discrete color.

Figures 12-14 There is no reference to these Figures in the text.

Response: Amended. These figures are referenced in the text in Section 5.3.

Table 1 define F0, F1, F2, F3

Response: Amended. The explanation for the acronyms F0, F1, F2 and F3 have been added in Table 1 caption.
Table 1 caption: “F0, F1, F2, F3 are the different phases of the de-escalation period being F0 the first phase after lockdown and F3 being the last phase before all restrictions were eliminated”.

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Mar, K. A., Ojha, N., Pozzer, A., and Butler, T. M.: Ozone air quality simulations with WRF-Chem (v3.5.1) over Europe: model evaluation and chemical mechanism comparison, Geosci. Model Dev., 9, 3699–3728, https://doi.org/10.5194/gmd-9-3699-2016, 2016.

Im, U., Bianconi, R., Solazzo, E., Kioutsioukis, I., Badia, A., Balzarini, A., Baro, R., Bellasio, R., Brunner, D., Chemel, C., Curci, G., Flemming, J., Forkel, R., Giordano, L., Jimenez-Guerrero, P., Hirtl, M., Hodzic, A., Honzak, L., Jorba, O., Knote, C., Kuenen, J.J.P., Makar, P.A., Manders-Groot, A., Neal, L., Perez, J.L., Pirovano, G., Pouliot, G., San Jose, R., Savage, N., Schroder, W., Sokhi, R.S., Syrakov, D., Torian, A., Tuccella, P., Werhahn, K., Wolke, R., Yahya, K., Zabkar, R., Zhang, Y., Zhang, J., Hogrefe, C., Galmarini, S., 2015. Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part I: Ozone. Atmos. Environ. 115, 404e420.