

Answer to the editor's review on "Modelling of atmospheric variability of gas and aerosols during the ACROSS campaign 2022 in the greater Paris area: evaluation of the meteorology, dynamics and chemistry" by L. Di Antonio et al.

First, we would like to thank the editor for carefully reading the answer to the reviewers and the new version of the manuscript and providing valuable comments that helped to improve the quality of the manuscript. We have taken into account all the comments made by the editor, and have changed the paper accordingly. The details of our changes are highlighted in the text. The point-by-point answers are provided below.

With regard to the comment of the reviewer on the PBLH simulations, the authors do not address in their revised version the importance of errors in the chemical concentrations raised by reviewer #1, who also provides a relevant reference. Please add a comment on this in the manuscript.

We thank the editor for this suggestion. We have added the following comment in section 3.2:

"Daily maxima are captured (± 200 m) for the majority of days, however, for about a third of cases the simulated PBLH remains below the observed MLH by more than 200 m. An inaccurate simulation of PBLH can lead to a poor representation of surface aerosol concentrations (Du et al., 2020). This could be the case of the two very hot days with T_{max} higher than 33°C (June 18 and July 13), the observed MLH at SIRTa nearly reached 3 km, while simulated PBLH are only 1600 and 1900 m, respectively. ... For this day, the underestimated simulated PBLH could contribute to the overestimated OA peak (Fig. 9a)."

Line 36: replace 'will be' by 'are'

Corrected, thanks.

Lines 118- 120: What means 'generally small differences' please provide statistical indicators. You can add those in Figure S1. Also add in this place part of your reply to the comment on the use of a smaller number of vertical layers in the 2km domain.

Thanks for this comment. Section 2.1 has been modified to take into account the comment as follows:

"The 30 and 6 km domains of the CHIMERE model have 15 vertical layers between the surface and 300 hPa, while 10 levels are used for the 2 km domain up to 500 hPa. Although the total number of vertical layers is lower compared to the larger domains, they are still denser near the surface where detailed resolution is most critical. Indeed, differences between the two configurations are generally small at the three campaign sites (mean bias between the 6km and 2km simulations at the three campaign sites ranging between -0.4 and $0.07 \mu\text{g m}^{-3}$ for organics, between -0.04 and $0.02 \mu\text{g m}^{-3}$ for sulfate, between -0.01 and 0.03 for ammonium, between 0.02 and 0.08 for nitrate and -0.002 and $0.001 \mu\text{g m}^{-3}$ for chloride, see Fig. S1). While the 6 km resolution simulations are used for comparisons with meteorological or pollutant observations over France, the finer scale 2 km resolution simulation is used for comparisons with campaign observations, especially in section 4.4."

Line 169: 'partitioning' not 'portioning'

Corrected thanks.

Line 208: please provide link to the documentation of the CHIMERE model.

Thanks, the link to the documentation has been inserted within the text.

Line 209: replace ‘is’ by ‘are’

Done, thanks.

Lines 352-365: add somewhere in this discussion your comment on the nighttime product of the STRATfinder algorithm.

We thank the editor for this suggestion. The description of the mixing layer height (MLH) product in Section 2.2 has been updated to include the discussion of the nighttime reliability of the STRATfinder algorithm as follows:

“Mixed layer height (MLH) derived automatically from profile observations obtained by a network of automatic lidars and ceilometers (ALC) operated in synergy with the PANAME initiative (Kotthaus et al., 2023). At SIRT ALC profile data (Lufft CHM15k) are processed using the STRATfinder algorithm (Kotthaus et al., 2020). However, this algorithm has limited sensitivity below 230 m, making nighttime comparisons less reliable. The data also undergo additional quality control measures, developed in the context of RI-URBANS and the ABL testbed program (<https://ablh.aeris-data.fr/>, last access: 3 July 2024).”

Lines 521-524: NH₃ rich conditions mean also that your aerosol is probably not very acidic and this could explain why HNO₃ partitions to the aerosol phase as nitrate ion. Did you check the simulated acidity of your aerosol?

Thank you for your suggestion. We have indeed checked the acidity of our aerosol in the simulation and added a Figure for the PRG urban site in the supplementary material (see Fig. R1, Fig. S12 in the supplementary material) and observed that aerosols are in general well neutralized, in particular for the overestimated nitrate peak on June 16, 2022 and on some occasions slightly acidic. Furthermore, we found that this overestimated nitrate peak is largely driven by the transport of nitrate from a strong emission source in Northern France, rather than by local nitrate formation pathways.

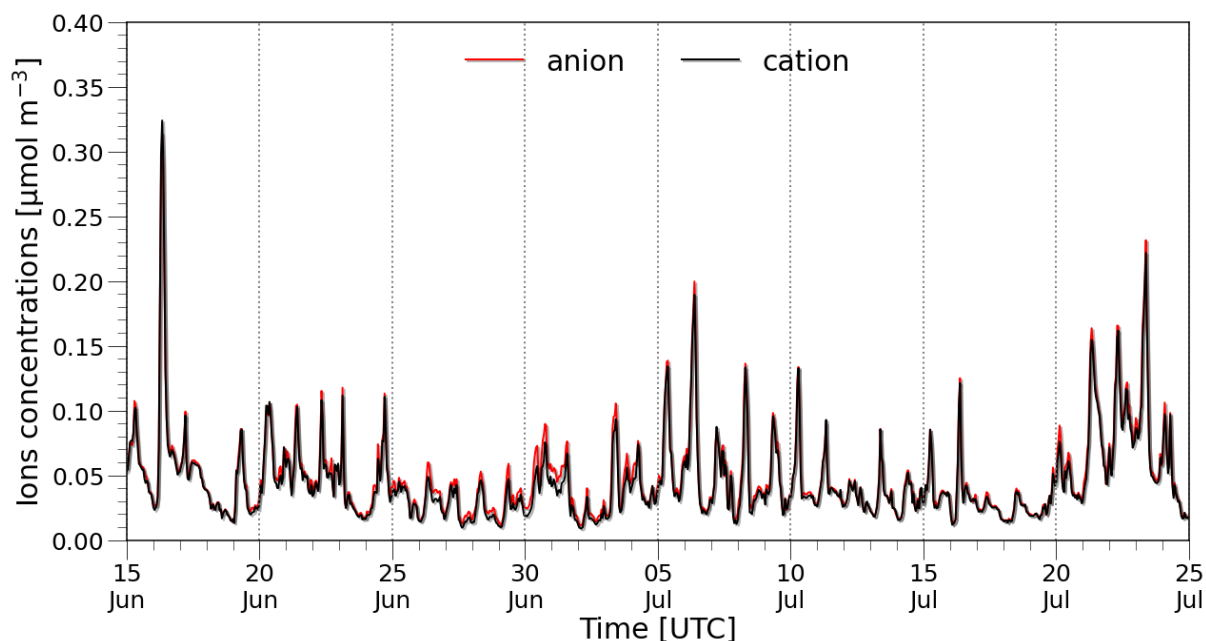


Figure R1: Time series of cation ($\Sigma \text{NH}_4^+ + \text{Na}^+$) and anion ($\Sigma 2 \text{SO}_4^{2-} + \text{NO}_3^- + \text{Cl}^-$) ions concentrations at the urban PRG site during the ACROSS period.

In the new section: Discussion

Please add also your comment/reply on the less pronounced sensitivity of nitrate simulations to the model resolution.

We thank the editor for the suggestion. Since the new Discussion Section 6 addresses uncertainties in the formation of biogenic secondary organic aerosol (BSOA), we have incorporated this aspect into Section 4.4 as follows:

“For these days, aerosol is well neutralized to slightly acidic (see Fig. S12). Although the differences in the nitrate concentrations due to the spatial resolution of the model simulation are small (with a mean bias between the 6km and 2km spatial resolution simulation ranging from 0.02 to 0.08 $\mu\text{g m}^{-3}$ for nitrate at the three campaign sites), when discrepancies with observations occur, the nitrate peaks are typically more overestimated in the 2km simulation compared to the 6km simulation (see Fig. S1).”

Line 601 remove ‘will’

Removed, thanks.

Line 609 replace ‘report’ by ‘reported’

Line 611 replace ‘leads’ by ‘led’

Line 612 replace ‘occur in’ by ‘are computed for’

Lines 614 and 616 , 619 replace ‘find’ by ‘found’

Line 625 ‘comes from THE uncertainty’

Thanks, the above corrections have been taken into account.