The paper addresses an interesting topic related to the physical and chemical processes that determine the atmospheric levels of the secondary inorganic aerosols (SIA) in urban scale. The manuscript is very well written. The scientific tools used (i.e. Eulerian modeling and PM chemical composition measurements) and methodological approach are robust, while the conclusions are clear. The paper can be accepted for publication. The main minor comment is that the authors should provide maps with the chemical speciated total emission fields (precursors of SIA and SIA), so as to be easier to associate the impact of emission sources on the spatial distribution of SIA concentrations. For the same reason, the relative contribution of emission sectors on the precursors of SIA and SIA total emissions should be presented.

Replies to RC1

We thank the reviewer for the careful reading of the manuscript and the insightful comments. Please find below our point-by-point replies:

1. The main minor comment is that the authors should provide maps with the chemical speciated total emission fields (precursors of SIA and SIA), so as to be easier to associate the impact of emission sources on the spatial distribution of SIA concentrations.

Reply: A supplementary material document has been created for the manuscript, where a new figure (Fig. S1) has been added, depicting the spatial distribution of total emission fields of the relevant species indicated by the reviewer. A relative discussion has been added to the main text.





Figure S1: Maps of total annual emission fields of: a) total NO_x emissions, b) total NH_3 emissions, c) total SO_2 emissions, d) total $PM_{2.5}$ emissions, e) fine-mode sea salt emissions (SSf), and f) potassium (K^+) emissions from domestic burning (Kbb). The database used is CAMS regional anthropogenic emissions, which are spatially disaggregated (to 1 km²) by the UrbEm approach (Ramacher et al., 2021); sea salt emissions are calculated online based on Vignati et al. (2010) parameterizations along with available updates (see text); K^+ emissions are derived based on non-sea salt $K^+/PM_{2.5}$ concentration ratios are

derived from filter-based $PM_{2.5}$ measurements in the center of Athens by Paraskevopoulou et al. (2014) (see text).

2. For the same reason, the relative contribution of emission sectors on the precursors of SIA and SIA total emissions should be presented.

Reply: A new figure has been added to the supplement, depicting the contribution of emission sectors to the yearly values of SIA and their precursors as a total value for the domain of interest. A relative discussion has been added to the main text.



Figure S2: Spatio-temporal totals of SIA-related tracer emissions for SO₂, NH₃, PM_{2.5}, Sea-Salt (expressed in NaCl), Potassium (K^+), SO₄⁻², and NOx (in tonnes per year per simulation domain), and the contribution of each source sector (GNFR category A: power plants; B: industrial sources; C: other stationary combustion; D: fugitives; E: solvents; F: road transport; G: shipping; H: aviation; I: off-road; J: waste; K: agriculture)

References

Paraskevopoulou, D., Liakakou, E., Gerasopoulos, E., Theodosi, C., and Mihalopoulos, N.: Long-term characterization of organic and elemental carbon in the PM2.5 fraction: the case of Athens, Greece, Atmos. Chem. Phys., 14, 13313–13325, https://doi.org/10.5194/acp-14-13313-2014, 2014.

Ramacher, M. O. P., Kakouri, A., Speyer, O., Feldner, J., Karl, M., Timmermans, R., Denier van der Gon, H., Kuenen, J., Gerasopoulos, E., and Athanasopoulou, E.: The UrbEm Hybrid Method to Derive High-Resolution Emissions for City-Scale Air Quality Modeling, Atmosphere, 12, 1404,

https://doi.org/10.3390/atmos12111404, 2021.

Vignati, E., Facchini, M. C., Rinaldi, M., Scannell, C., Ceburnis, D., Sciare, J., Kanakidou, M., Myriokefalitakis, S., Dentener, F., and ODowd, C. D.: Global scale emission and distribution of sea-spray aerosol: Sea-salt and organic enrichment, Atmospheric Environment, 44, 670–677, https://doi.org/10.1016/j.atmosenv.2009.11.013, 2010.