

Sartelet and co-authors present a comparison of modelled and measured aerosol number concentrations and size distributions at three sites in the greater Paris region. Modelled aerosol concentrations are performed without a nucleation scheme and using three nucleation schemes: binary, ternary, and heteromolecular (including organic components). The paper is clearly written and the analysis of data is mainly about comparing correlation model results and measurements. My expertise is not in aerosol modelling, so I cannot comment on the validity of the modelling framework, however I have a few comments pertaining to the treatment of aerosol emissions that are shown below. In summary, I believe that there is a large number of processes that can influence the aerosol number concentrations (especially the nucleation mode) that are not addressed nor discussed in this study.

It is not clear to me how representative these modelling simulations are compared to measurements given that there are significant uncertainties regarding emissions of gas-phase precursors, especially organic SOA precursors. Have the authors investigated the impacts of changing precursor source emission concentrations and evaluating the impacts on number concentrations and size distributions? How would that affect model results?

Our reply: Emissions and formation of gas-phase precursors of particles are indeed attached to significant uncertainties. To check that our simulation is realistic, the concentrations of PM₁ directly related to these precursors are compared to measurements. Nitrate, ammonium, sulfate and organics compare very well to the measurements, as detailed in section 5.1.1., as well as gas-phase NO₂. For NH₃, the following sentences are added :

« No measurement of NH₃ concentration was available in 2009. However, the simulated concentration range is realistic, considering that measurements of NH₃ performed recently in the Paris city center with a mini-DOAS indicate a mean concentration of 2 µg m⁻³ (Viatte et al. 2021). »

Do the authors consider the emissions of low- and extremely-low organic volatile compounds? if so, how are these treated? if not, I would assume that these LVOCs and ELVOCs can significantly impact modelled nucleation rate, concentrations, and size distributions, hence a discussion of potential impacts would be needed in the manuscript.

Our reply: Emissions of volatile compounds of different volatilities are treated in the model. The following sentences are added in the section 2.2, which deals with emissions :

« Emissions of intermediate, semi and low volatile organic compounds (IVOC, SVOC, LVOC respectively) are estimated from the organic mass of PM_{2.5} as detailed in Sartelet et al. (2018) : the mass of organic vapors is estimated by multiplying by 1.5 the organic mass (Kim et al. 2016). The emitted organics are then divided into volatility classes : 25% is assigned to LVOC, 32% to SVOC and 43% to IVOC (Couvidat et al. 2012). »

The ELVOCs involved in the heteromolecular nucleation are not emitted in the atmosphere but formed from autoxidation of monoterpenes (Ehn et al 2014).

How are organics partitioned to the aerosol phase? Different treatments can affect the evaporation of the nucleation mode (and hence concentrations and size distribution), especially when using the heteromolecular nucleation scheme which consider condensation of organic components. For instance, have the authors explored the impact of absorptive partitioning versus non-adsorptive partitioning? I am bringing this issue since

it will likely affect nucleation mode number concentrations, which is the main focus of this study.

Our reply: Organics partition to the aerosol phase either by absorbing into an organic phase or into an aqueous phase, depending on the properties of the compounds (hydrophilic or hydrophobic). Adsorption is not considered, as we assume that it is negligible in front of absorption because there is always aerosol mass in the atmosphere, and non-volatile compounds condense independently of the particle composition. As detailed in section 2.1, the growth of UFP is strongly impacted by the condensation of low-volatility compounds as well as coagulation. Therefore, numerically, the condensation of non-volatile compounds is solved dynamically with nucleation and coagulation processes. Those non-volatile compounds, such as the ELVOCs involved in the the heteromolecular nucleation, may not evaporate from the nucleation mode, because they are not volatile. Then the condensation/evaporation of semi-volatile compounds is computed by assuming bulk thermodynamic equilibrium between the gas and the particle phases.

Can the author add a temporal comparison of modelled and measured $N_{<10}$, $N_{<100}$ and $N_{>100}$ for the duration of the measurements? It would be informative to understand the temporal evolution of particle concentrations and check if biases occur on given days or are consistent throughout the measurement period.

Our reply: The hourly temporal evolution of N_{10-100} is added and compared to measurements for the different nucleation parameterisations in section 5.2. N_{10-100} concentrations are plotted because they allow us to clearly visualize the strengths and shortcomings of the different nucleation parameterisations for the formation of UFP, and to perform coherent comparisons at all sites.

Added references :

- Cai, R., Chandra, I., Yang, D., Yao, L., Fu, Y., Li, X., Lu, Y., Luo, L., Hao, J., Ma, Y., Wang, L., Zheng, J., Seto, T., and Jiang, J.: Estimating the influence of transport on aerosol size distributions during new particle formation events., *Atmos. Chem. Phys.*, 18, 16 587–16 599, <https://doi.org/10.5194/acp-18-16587-2018>, 2018
- Kim, Y., Sartelet, K., Seigneur, C., Charron, A., Besombes, J.-L., Jaffrezo, J.-L., Marchand, N., and Polo, L.: Effect of measurement protocol on organic aerosol measurements of exhaust emissions from gasoline and diesel vehicles, *Atmos. Environ.*, 140, 176–187, <https://doi.org/10.1016/j.atmosenv.2016.05.045>, 2016.
- Sartelet, K., Zhu, S., Moukhtar, S., André, M., André, J., Gros, V., Favez, O., Brasseur, A., and Redaelli, M.: Emission of intermediate, semi and low volatile organic compounds from traffic and their impact on secondary organic aerosol concentrations over Greater Paris, *Atmos. Environ.*, 180, 126–137, <https://doi.org/10.1016/j.atmosenv.2018.02.031>, 2018.
- Schobesberger, S., Junninen, H., Bianchi, F., Lönn, G., Ehn, M., Lehtipalo, K., Dommen, J., Ehrhart, S., Ortega, I. K., Franchin, A., Nieminen, T., Riccobono, F., Hutterli, M., Duplissy, J., Almeida, J., Amorim, A., Breitenlechner, M., Downard, A. J., Dunne, E. M., Flagan, R. C., Kajos, M., Keskinen, H., Kirkby, J., Kupc, A., Kürten, A., Kurtén, T., Laaksonen, A., Mathot, S., Onnela, A., Praplan, A. P., Rondo, L., Santos, F. D., Schallhart, S., Schnitzhofer, R., Sipilä, M., Tomé, A., Tsagkogeorgas, G., Vehkamäki, H., Wimmer, D., Baltensperger, U., Carslaw, K. S., Curtius, J., Hansel, A., Petäjä, T., Kulmala, M., Donahue, N. M., and Worsnop, D. R.: Molecular

understanding of atmospheric particle formation from sulfuric acid and large oxidized organic molecules, *Proc. Nat. Acad. Sci.*, 110, 17 223–17 228, <https://doi.org/10.1073/pnas.1306973110>, 2013.

- Viatte, C., Petit, J.-E., Yamanouchi, S., Van Damme, M., Doucerain, C., Germain-Piaulenne, E., Gros, V., Favez, O., Clarisse, L., Coheur, P.-F., Strong, K., and Clerbaux, C.: Ammonia and PM_{2.5} Air Pollution in Paris during the 2020 COVID Lockdown, *Atmosphere*, 12, <https://doi.org/10.3390/atmos12020160>, 2021