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Comment on acp-2022-22

Anonymous Referee #1

Referee comment on "Influence of emission size distribution and nucleation on number concentrations over Greater Paris" by Karine Sartelet et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-22-RC1, 2022

This paper presents three-dimensional simulations with a chemistry-transport model coupled to a sectional aerosol module to assess the influence of three different nucleation parameterizations on particle number concentrations and size distributions in the Greater Paris region. The authors developed an innovative methodology for distributing particulate matter emissions to the size distribution of emitted particles in the sectional representation that ensures conservation of mass and number. This method for particle emissions is successfully applied in the simulations without nucleation and with three nucleation schemes. Model results are evaluated against observations of total particle number and qualitatively also to observed size distributions at three observation sites. Overall, the manuscript is written in good style and interpretation of the results for different nucleation scheme is convincingly presented.

However, when comparing the modelled size distributions to measured size distributions, especially for the heteromolecular nucleation mechanism that involves biogenic oxidation products, it becomes obvious that with the current approach, the discrepancy can be attributed neither to inaccuracies of the formation rate alone nor to inaccuracies of the growth rate alone. I strongly suggest to perform a new particle formation event analysis including all nucleation-event days. The formation rate J10-50 (or J10-100) of particles can be derived from the observations (change of total number concentration plus losses through coagulation and growth) and compared to the formation rate in the model for different nucleation parametrizations. I think the paper will greatly benefit from the additional nucleation-event analysis.

Specific Comments:

1.) P. 3 line 5: Another uncertainty when dealing with emissions of particulate matter is the emission of low-volatile organic vapors. Depending on the distance from the source these may be in gas phase (at high temperature) or in the condensed phase (after cooling to ambient temperature). The difficulty in accounting for the organic vapors in the emission inventory arises from the fact that they might already be partly included in $PM_{2.5}$ (as organic carbon).

2.) Section 2.1: Related to the previous point: how is the primary emission of semi-volatile and low-volatile organic vapors from road traffic estimated in the model?

3.) Cai et al. (2018) show different influence of long-range (or short-range) transported aerosols and gaseous precursors on new particle formation events in clean and polluted environments. How is transport of particles through the regional boundaries estimated?

4.) P. 4, lines 17-19: The proxy of biogenic oxidation products (BioOxOrg) in the Riccobono parameterization is strictly speaking not the same as ELVOC from the auto-oxidation of monoterpenes. BioOxOrg represents later-generation oxidation products of biogenic monoterpenes. The authors should discuss what this means for the time scale of formation.

5.) P. 4 line 23: Guo et al. (2020) find that aromatic VOC from vehicular exhaust are important precursors for nucleation and growth. While monoterpene emissions of forests may be important for regional new particle formation, I would assume that the urban local particle formation events are more likely caused by vehicle-emitted precursors.

6.) Section 2.2: The distribution of PM2.5 emissions over the size bins of the model is an innovative approach. It would be interesting to know if the distribution of PM2.5 emissions to size sections of emitted particles is done online in the CTM or prior to the simulation in an emission model. Further, I suggest adding a table that shows the distribution procedure for different PM size ranges to the 25 model size sections. The diameter bounds for the sections of the aerosol model could also be included in that table to avoid listing them in the text (P. 4, lines 28-29).

7.) P. 11, effect of the ternary nucleation: To better understand the influence of ternary nucleation in the region, it would be helpful to show the ammonia (NH3) concentration field as well and discuss the uncertainties of NH3 emissions.

8.) P. 12 lines 5-7: The comparison of the size distribution from heteromolecular nucleation with observations indicates that the <20 nm particles do not grow sufficiently, either because they are too numerous or because concentrations of condensable vapor are too low. As stated in my general comments, a nucleation-event analysis would greatly help to evaluate the formation rate in the model.

Technical Corrections:

P. 2 line 6: should be “organs”.

Figures 4 and 5: annotate longitude and latitude on the x- and y-axes of the maps.

Figure 5: better denote the simulation name at the plots, for example in the header of the maps.

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, 16587-16599, https://doi.org/10.5194/acp-18-16587-2018, 2018.

Guo, S., Hu, M., Peng, J., Wu, Z., Zamora, M. L., Shang, D., Du, Z., Zheng, J., Fang, X., Tang, R., Wu, Y., Zeng, L., Shuai, S., Zhang, W., Wang, Y., Ji, Y., Li, Y., Zhang, A. L., Wang, W., Zhang, F., Zhao, J., Gong, X., Wang, C., Molina, M. J., and Zhang, R.: Remarkable nucleation and growth of ultrafine particles from vehicular exhaust, Proc. Nat. Acad. Sci., 117, 3427-3432, https://doi.org/10.1073/pnas.1916366117, 2020.