Review of Cai et al: Impacts of coagulation on the appearance time method for sub-3 nm particle growth rate evaluation and their corrections (V4)

The paper describes a correction for the consideration of coagulation in nucleation aerosol formation modelling. This correction is changing the timing for nucleation mode particles, the time within the model until when particles have grown for example to a certain size. The title restricts this investigation to particle sizes below 3 nm where it might be especially important, however within the manuscript the appearance time is given also for other particles sizes. To illustrate the results a case study for Beijing is given for a late winter nanoparticle event.

General comments

The authors miss to clarify under which environmental conditions their theory is applicable and relevant. Page 3, line 23 they promise to investigate the limitations. However, these limitations are never specified in the text. Page 15, line 18 to 20 states: uncertainties come from instrumental biases, atmospheric turbulence and omitted contributions from transport, mixing and emission to the measured aerosol size distribution.

Summarizing the limits behind these statements mean that the theory presented is valid only for a homogeneous air-mass without any variability during the period between initialization of the model run (e.g. 00:00 until at least noon, the time given for the appearance of 4.9 nm particles (12:00), page 15, line 1). These conditions might be valid in a smog chamber (Stolzenburg et al, 2020) but are unlikely to find in the real atmosphere. Here a coupled model setup is necessary (Baklanov et al, 2008). Whether the model used in the manuscript is applicable at all for stationary ground based measurements under the conditions in the North China Plain is highly questionable.

The authors suggest by plotting model results into a graph with experimental results that the observation on February 24, 2018 can be described with their theory. However, a detailed supporting data set is missing. All available supporting data suggest that model and experiment cannot be compared as the experimental data set is not in agreement with the limitations of the model.

A three dimensional transport model including meteorological variables is available from either Flexpart of HYSPLIT. A local high resolution data set is available for example from the 320 m Beijing meteorological tower, about 7 km away from the particle observation site, close enough for a regional investigation. The data from this tower for February 24, 2018 (private communication, Prof. Fei Hu, IAP Beijing) show, that an intense vertical exchange of air masses began already a few hours before sunrise and continuing the next hours, advecting dry air from the residual layer all the way towards the ground (lowest level 8 m). This advection is reflected for example in the water vapor measurements. Starting at 2.3 g/m^3 the water vapor concentration declined to < 1.1 g/m^3 at 08:00. This is a clear indication for a massive advection of dry air from aloft (~ 0.9 - 1 g/m^3 in the residual layer between 800 and 1500 m above Beijing, HYSPLIT) and a replacement of the humid air in the planetary boundary layer. This air mass transport coincides with a concurrent doubling of the sulphur dioxide concentration between midnight and 08:00 LT. This polluted air mass contained also excess H_2SO_4, roughly doubling from nighttime levels to about $4* 10^6$ $/cm^3$ at the time of sunrise (06:56 LT). Such an increase, even before any uv-radiation would be available for photochemistry, is not at all in agreement with a photochemical production of H_2SO_4 as claimed in the manuscript (Hearth et al, 2004).

The residual layer often contains large quantities of nanoparticles as shown by Quan et al, (2017) for Beijing, by Lampilahti et al, (2020) for Hyytiälä, Finland (also Hao et al, 2018) and by Junkermann and Hacker, (2018) for Germany (see there also for further examples from China and Australia).

Finally: The authors present an analysis how particles might be produced in the atmosphere in a homogeneous air parcel. An admixture of air from the outside either by a replacement of the air mass by horizontal transport or a convective mixing process with air from the residual layer (Lampilahti et al, 2020) is not taken into account. Such an approach is not in agreement with highly variable ambient conditions, typical winds and diurnal cycles in the planetary boundary layer and thus not applicable for the Beijing case study.

The case study however, properly analyzed, would be a good example for a transport and convection driven nanoparticle advection, including gas to particle conversion (Gillani et al, 1979). Such a nanoparticle advection can also superpose the physical constrains of nano-particle GTP under heavily polluted conditions (Kulmala et al, 2017).

References

Baklanov A., Mahura A., Sokhi R. (Eds) (2008): Integrated Systems of Meso-Meteorological and Chemical Transport Models. Report of the COST Action 728 on "Enhancing mesoscale meteorological modelling capabilities for air pollution and dispersion applications". COST Publication, 178p, Feb 2008

Heard, D.E. et al, Geophysical Research Letters, 2004, High levels of the hydroxyl radical in the winter urban troposphere, Volume 31, 18, https://doi.org/10.1029/2004GL020544

Gillani, N.V., Husar, R.B., Husar, J.D., Patterson D.E., and Wilson, W.E., 1978, Project MISTT, Kinetics of particulate sulphur formation in a power plant plume out to 300 km, Atmospheric Environment, 12, 589-598

Hao, L., et al, 2018, Combined effects of boundary layer dynamics and atmospheric chemistry on aerosol composition during new particle formation periods, Atmos. Chem. Phys., 18, 17705–17716, https://doi.org/10.5194/acp-18-17705-2018.

Junkermann W., and Hacker J., 2018, Ultrafine particles in the lower troposphere: major sources, invisible plumes and meteorological transport processes, BAMS, 99, 2587-2622, Dec. 2018, DOI:10.1175/BAMS-D-18-0075.1

Kulmala, M. et al, 2017, Atmospheric gas-to-particle conversion: why NPF events are observed in megacities? Faraday Discuss., 200, 271-288

Lampilahti, J., et al, Aerosol particle formation in the upper residual layer, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-923, in review, 2020.

Quan, J., et al, 2017: Anthropogenic pollution elevates the peak height of new particle formation from planetary boundary layer to lower free troposphere, Geophys. Res. Lett., 44(14), 7537–7543, doi:10.1002/2017GL074553

Stolzenburg, D. et al, 2020, Enhanced growth rate of atmospheric particles from sulfuric acid, Atmos. Chem. Phys., 20, 7359–7372, https://doi.org/10.5194/acp-20-7359-2020