

Interactive comment on “Resolving nanoparticle growth mechanisms from size- and time-dependent growth rate analysis” by Lukas Pichelstorfer et al.

Anonymous Referee #2

Received and published: 5 October 2017

Review of Pichelsdorfer et al: "Resolving nanoparticle growth mechanisms from size- and time-dependent growth rate analysis".

The authors present two novel methods to derive size-dependent particle growth rate from successive particle size distribution measurements. The methods are based on analyzing changes in the particle number concentration changes as time advances. The methods are novel and interesting to the community as determining the growth rate accurately is a key question in resolving particle formation mechanisms. The methods are sound and the testing of the methods has been done rigorously and carefully. I have a few, fairly minor questions and requests for clarification regarding the manuscript; if

C1

these can be answered, I suggest that the manuscript should be published in ACP.

Questions/requests for clarification:

* The authors compare the new methods to the appearance time (maximum concentration) method which is described as the standard protocol for ddp/dt determination in Kulmala et al., (2012). This method is not suited to produce time-dependent growth rates, although it gives a size dependence. However, the other method that has been described in the same protocol, namely the log-normal distribution function method (Kulmala et al., 2012, p.1659) can give the time-dependent growth rate of a log-normal particle population as a function of time. The authors present no comparison of their methods to this other, quite widely used growth rate determination method. I think that such a comparison could be beneficial, and should not be too laborious as simulated PSDs are already available. Otherwise, it would be good to mention why such a comparison is not considered.

* In section 2.1, it might be helpful to keep the time indexes in the derivation at all times. If I'm reading the method correctly, the experimental size distribution at t_{j-1} , $n^{exp}(t_{j-1})$ is taken as the starting point for producing the simulated distribution $n^{sim}(t_j)$, and this simulated distribution is then compared piecewise to the experimental distribution $n^{exp}(t_j)$. Differences are attributed to growth. From this, I would consider that the time period for which the growth is calculated is $[t_{j-1}, t_j]$. However, In equation (4), the time points used are t_j and t_{j+1} , which is confusing, especially as the times are not given for the (time-dependent) count median diameters. Using the time indexes consistently in Eqs. (2), (3) and (4) would make this much easier to read for a reader trying to implement the method. Also, in the appendix A the methods is described as 'forward-looking', i.e. the interval $[t_j, t_{j+1}]$ is considered.

* Regarding the times, Eq. (4) should include the time stamp similarly to Eq (9).

* Is there a reason why the TREND method looks backward from t_j while INSIDE looks forward?

C2

* Section 2.2. line 17: to me it seems that Eq (5) still gives the number concentration but in volume space (change '...in integrated volume concentration...' to 'in integrated number concentration')

p. 4, line 25: Dilution losses should also be mentioned as they are explicitly in Eq. (5)

p. 5: I do not fully understand why the INSIDE methods needs to use the number volume distribution instead of the number size distribution. It seems unnecessarily confusing especially as it is later converted to size anyway. Could this be explained?

p.5 line 27: '...the more pronounced the differences between the models become.' Which models? Only one model was used?

p. 6:, line 13: '...sensitive to scatter...' what is meant by scatter here? Experimental error in the size distribution or something else? Please clarify.

p10, line 7: 'This trend suggests...' This is confusing, as it reads like lower growth rates in the sub-3nm range indicate lower-volatility vapours. I think the trend meant here is the 'somewhat less significant' part of the sentence; the manner of expressing this is confusing and could be re-structured.

p. 11 line 7: '...sufficient time-resolution and appropriate counting statistics'. I did not find any criteria for this statement. The error was not analysed as a function of time resolution or counting statistics. Please give some more information on what is 'sufficient / appropriate', otherwise reword.

References

Kulmala et al., 2012, Nature Protocols 7, 1651–1667 (2012)
doi:10.1038/nprot.2012.091

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-658>, 2017.