

Interactive comment on “Estimation of atmospheric particle formation rates through an analytical formula: Validation and application in Hyytiälä and Puijo, Finland” by Elham Baranizadeh et al.

Anonymous Referee #2

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GENERAL

The manuscript is basically suitable for ACP but a few important points need to be addressed first. I'll rate this "major revision" for now but remain skeptical that all concerns can be addressed to my satisfaction.

1. The authors present a method to estimate formation rates for smaller particles based on measurements of the formation rate for larger particles. This is no doubt necessary to compare measurements with different instruments and to gain information on the actual nucleation rate which happens at sizes which are often outside the measurement

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range. However, a few years back, Kulmala et al. have published a sort of how-to guide on nucleation measurements in *Nature Protocols*. And that looks very, VERY similar to what is presented in this manuscript while the text reads as if something new is shown. So my question would be: What is actually new in the approach that the authors present? Or is this just another application of the same formula that has been used in lots and lots of papers for quite a number of years? If this were the case, the manuscript's content would be very slim indeed (and all the description of methods used obsolete) and one would have to ask if just running an old formula on a new set of data would justify a scientific publication.

2. The method to determine GR obviously doesn't work as the authors themselves point out. That is not a surprise since mode-fitting at the edge of a size distribution is always a bad idea. However, there are other methods. How can you justify using a method that clearly produces bad results while there are alternatives available? Sure, other methods typically are much more labour-intensive but given the current state of affairs it seems quite clear to me that other approaches MUST be employed. At least a preliminary test on a smaller subset of the data is absolutely and totally necessary.

3. I do wonder how there can be only 65 days with good enough data from 12 or 13 years of Hyttiälä observations. Assuming 12 full years à 365 days and an NPF frequency of 23% (Nieminen et al., 2014), that's a tiny 6.5% of all nucleation events observed during that time (about 1000). How can that be? Are we supposed to believe that one of the longest and probably the most published data set of aerosol size distributions is actually total crap? I mean, I have worked with DMPS and SMPS data quite a bit but never ever has the data been so terrible that a proper analysis was possible for less than 10% of events. And even if I was to accept this low percentage (which I can't and won't) then the question arises if this kind of cherry-picking doesn't introduce a bias into the analysis that would make all and any results highly questionable.

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line 31f "(e.g. Almeida et al., 2013; Berndt et al., 2014; Kirkby et al., 2016)" → Bianchi et al., 2016 should probably be added there.

line 51 "we aim to estimate 3 nm particle formation rates" → why? the 3 nm limit has no physical meaning, it's just a tradition born out of instrumental limitations from two decades back. i understand that you do that for the hyttiälä data since the point is testing the approach. but for puijo?

line 88ff → this whole section is useless if this is the same approach as in the Nature Protocol

line 164 "the size dependence of the growth rate in the range 3-20 nm is typically weak" → really? or is this just an artefact of the GR approach not working (which we know is true). certainly you could cite some previous studies that have found this; hyttiälä isn't exactly under-studied after all.

line 173 "85 %" → wasn't it 84 in the abstract?

line 181ff → the whole median thing seems a bit silly. i mean, you take the median over 12 hours during most of which there is no formation of 3 nm particles. Of course the result will be close to 0 (as it is). a pointless exercise which tells us nothing.

line 201ff, line 210ff, and lots of other places → i won't comment on the GR stuff here, see general comments above.

line 216f "the days during which a clear peak in each of the [different] time evolution curves could be observed (39 days out of 65 days)" → 39 out of 65 sounds quite good. but really it is 39 out of 1000, and that is not acceptable.

line 224 "It can be also concluded that visual inspection of the data is still valuable" → that's sound advise that you might want to follow with regard to the GR business.

line 227 "There are 15 NPF days for which the estimated time-lag is within 1.5 hours of the observed time-lag." → please take a moment and think what you have written

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there. with an average GR of roughly 4 nm/h, the average time-lag should be around 1 hour, right? that 15 cases lie within 1.5 hours is no proof that the method works sometimes but rather that the method does not work AT ALL.

line 227ff "Overall these results from analyzing Hyytiälä data show that Eq. (4) can be used to estimate the mean formation rates of 3-nm particles with reasonably good accuracy." → but maybe things could be much better with an improved determination of GR?

TECHNICAL

not necessary at this point

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