Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2016-916-AC2, 2017


Atmospheric Chemistry and Physics Discussions
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# *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.

**Elham Baranizadeh et al.**

tuomo.nieminen@uef.fi

Received and published: 12 July 2017

We thank Referee #2 for his/her thorough review of our manuscript. The comments were extremely valuable and we have redone most of the analysis based on them. The main weakness of the previous submitted version was the poor performance in estimating the growth rate with the mode fitting method, which further meant poorly predicted time lag and poor performance in the time resolved formation rate comparison. We have now reanalyzed the growth rates with the so-called maximum concentration method and the results are overall muck better. All figures and table 1 are modified



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accordingly, and we also removed the old Figure 3 comparing time-lags, as we believe that it is unnecessary in the new version. We also removed the standard error color coding, related to the uncertainties when determining GR, from Figure 1.

Below we give our detailed responses to the referee's comments.

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 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.*

The method used in our manuscript for scaling the formation rates is indeed the same as described by Kulmala et al. in their Nature Protocols paper. While the method itself is not new, it has not been tested with atmospheric particle number size distribution data before (to our knowledge), although widely used in e.g. global modelling of aerosol dynamics. In our work, we present comparisons with the scaled formation rates to those calculated directly from the measurement data, and thus evaluate the applicability of this method with real atmospheric data.

*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.*

As the referee notes, the main weakness of the previous submitted version was the poor performance in estimating the growth rate with the mode fitting method, which further meant poorly predicted time lag and poor performance in the time resolved formation rate comparison. We have now reanalyzed the growth rates with the so-called maximum concentration method and the results are overall much better (the updated Fig. 1 included in the revised manuscript is shown as Figure 2 in our replies to Referee #1). All the figures and Table 1 are modified accordingly, and we also removed the old Fig. 3 comparing time-lags, as we believe that it is unnecessary in the new version.

*3. I do wonder how there can be only 65 days with good enough data from 12 or 13 years of Hyytiälä 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.*

The number of days included in our analysis is not limited by data availability, but rather the criteria of the NPF event analysis: the growing nucleation mode needs to be clearly observable for several hours (i.e. no changes in air masses). Typically in Hyytiälä the number of these “well-behaved” NPF events is around 10% of all days (Dal Maso et al., 2005). In the Dal Maso classification the NPF events in Hyytiälä are classified as Ia, Ib, and II. For the analysis of this manuscript, we only chose class Ia NPF events, producing the number of events analyzed here. This strict selection of NPF events was done because we wanted to eliminate the possible effect of e.g. changes in air masses in our results.

CONTENTS

*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.*

We added this reference.

*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 hyytiälä data since the point is testing the approach. but for puijo?*

The referee is correct that nowadays the aerosol instruments are able to measure particles down to 1.5 nm. However, since the vast majority of particle formation rates reported in the literature is at 3 nm, we chose to scale also the Puijo data to this size. That way the Puijo results can be more directly compared to observations at other sites.

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

The method is the same as described in the Nature Protocol paper, however we feel that presenting the method in our manuscript makes it easier for the user to read our paper. One other reason for this choice is the need to use growth rates in the equations. For Hyytiälä data, growth rates are available down to 3 nm while for Puijo only above 7 nm. Thus we wanted to make clear in the equations what size ranges we are using.

*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; hyytiälä isn't exactly under-studied after all.*
The referee has a valid point here. As we recalculated the growth rates using maximum-concentration method, there is indeed a size-dependency in the growth rates, as is shown in Figure 1 below (the red line shows the median, the edges of the box the 25th and 75th percentiles, and the error bars the 10th and 90th percentiles of the GR values of NPF events in Hyytiälä; the red data points are all GR values which are larger than the 90th percentile). However, this does not affect greatly the correlation between J3,obs and J3,est (calculated either using GR3-10 or GR7-20), as can be seen from Fig. 1 in the revised manuscript.

*line 173 "85 %" –> wasn't it 84 in the abstract?*
All the results related to comparison of the estimated and observed formation rates are updated in the revised manuscript according to the new J3,est values, which are calculated using the GR from maximum-concentration method. The correlation coefficient between J3,est and J3,obs is now 0.90 and 91% of the J3,est daily mean values are within factor of 2 from J3,obs.

*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.*
We agree, and therefore we have left the median values out from the revised manuscript.

*line 201ff, line 210ff, and lots of other places –> i won't comment on the GR stuff here, see general comments above.*
As mentioned previously, we chose a better method for the GR analysis, and the results improved a lot.

*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.*
As mentioned before we were very selective with the events analyzes and chose only the so-called 1A events based on the Dal Maso et al. classification. This was done in order to be sure that e.g. small changes in airmasses would not influence the results of the J estimation.

*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.*
So true. As we redid our analysis with the improved growth rate analysis, we also checked each analyzed event visually.

*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 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.*
With our new results, having improved growth rates as well as a much better match between the observed and estimated formation rates, we decided to remove (the old) figure 3 as well as related text.

*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?*
Yes, true. Now they are.

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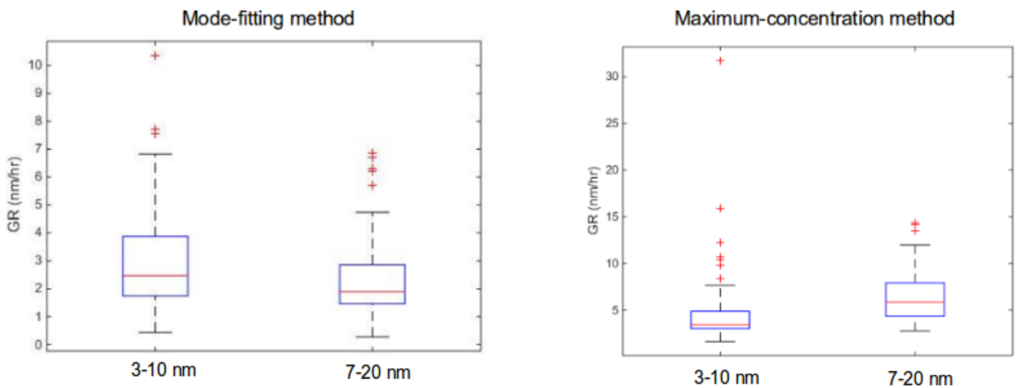


**Fig. 1.** Statistics of the growth rates in 3-10 nm and 7-20 nm size ranges in Hyytiälä, calculated by mode-fitting (left panel) and maximum-concentration (right panel) methods.



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