

Co-Editor Decision: Reconsider after minor revisions (Editor review) (03 Apr 2017) by James Allan

Comments to the Author:

While the paper looks good on the whole, there were certain 'major' comments from both referees that I do not see as being adequately addressed. I would like to see these tackled properly before the paper goes to publication.

Firstly, regarding referee #1's comment concerning anthropogenic influence, I note that the authors have rebutted this point, however I see that as a sufficiently important issue that this should be addressed in the manuscript itself. I would ask that the authors amend the text to reflect this.

In the summary and conclusions we added the text:

In this analysis the possibility that sporadic anthropogenic emissions were interpreted as NPF events cannot be excluded completely. However, there are a number of facts arguing strongly against this possibility leading to serious misinterpretation of the data:

- a) *Location and operation of the Mt. Zeppelin station exclude local contamination to a very large extent.*
- b) *Manual inspection of the time series by one of the co-authors (PT) further reduced the risk of contaminated data.*
- c) *The temporal evolution of MEV events, i.e. concurrent and sustained concentration increases at several particle sizes below 60 nm does not correspond to a typical passage of stack emissions from a large combustion source, (Ogren and Heintzenberg, 1990). Instead, it looks very much like MEV events observed under even stricter constraints on local or regional sources of contamination on icebreaker Oden in the central pack ice area, (Karl et al., 2013), and also looks similar to nocturnal NPF-events in Australian forests, (Suni et al., 2008; Junninen et al., 2008).*

Second, referee #2's points 1 and 2 about the metrics being reported has not been addressed, which they regard as being very important (see their comments). However, on reading the rebuttal and the paper referenced by the referee, it seems to me that the authors may have misunderstood what the referee was asking for. The metrics in question are defined in the paper Kulmala et al. (2012) doi:10.1038/nprot.2012.091, where 'GR' is defined as 'growth rate' and 'CS' as 'condensation sink'. Furthermore, the term 'formation rate' is applied to the rate of particle number formation at a defined size, not the growth rate referred to in section 3.2 and in the rebuttal.

In light of this clarification, can the authors generate these metrics and comparisons, as requested? I would expect that this would be most appropriate as an extension to the DGR approach. As well as ensuring comparability with other works, this will also allow the dataset to be more accessible for any future meta-analyses. Even if these comparisons turn up null results, this would still be an important observation to report in its own right. If the authors still feel that generating these statistics would be inappropriate, they are still entitled to make that argument, but such an argument should probably be reflected in the text of the paper, so as not to leave it open to accusations of deliberate omission. I should state however that my personal inclination is to err on the side of generating these statistics unless there is a technical reason that prevents this.

In response we modified the discussion of DGR-events and added the following text and Table 3 to the manuscript. Additionally we formulated a supplement to the manuscript, in which we present the graph that ref#2 requested.

In the analysis of atmospheric data and theoretical modeling of NPF-events of type DGR two key parameters are discussed, namely particle formation rate J ($\text{cm}^{-3}\text{s}^{-1}$) and growth

rate GR (nmh^{-1}) of particle diameters. For both parameters the measurement protocol by Kulmala et al. (2012) provides specific calculation procedures, (equations. 2, 7, and 9), which we follow in the present study, albeit with the caveat that the one-hour temporal resolution of our time series is far below the ten-minute time resolution that the protocol of Kulmala et al. (2012) requests in order to be able to follow the rapid development of NPF-events. Furthermore, only the 127 DGR-events identified from 2011 on are based on particle size distributions measured down to a diameter of five nanometers.

The sizes of newly nucleated aerosol particles are of order 1–2 nm, which is below or near the limit of existing measurement techniques. When the nuclei grow in size their number concentration decreases because of various removal mechanisms. Instead of particle formation rates at the initial nucleus size so-called apparent nucleation rates J_x are often reported, i.e. rates at which new particles appear at some larger observable particle diameter dx . For the present study two apparent nucleation rates are calculated: DGR events of the whole time series have been identified with particle size distributions measured at diameters from 10 nm up through the growth of the number median diameter D_{50} in the size range 10 – 50 nm. Thus we calculated J_{22} for these 235 events at the nominal geometric mean diameter of 22 nm. For 127 of these events size distributions reached down to five nanometer diameter, (years 2011 and later). For these events we calculated J_{11} at the geometric mean diameter 11 nm as representative for the diameter range 5 – 25 nm, which is close to the frequently reported apparent formation rate J_{10} at 10 nm diameter. Additionally, the two corresponding grow rates GR_{22} and GR_{11} were calculated in the respective diameter ranges.

Statistics of these four key parameters of the DGR events are collected in Table 3. Depending on the pollution level at the measuring site widely varying values of J_{10} have been reported. For the polluted subtropical environment of Taiwan Young et al. (2013) give values from 4.4 to 30 $\text{cm}^{-3}\text{s}^{-1}$ whereas Pierce et al. (2014) published values between 0.22 and 0.84 $\text{cm}^{-3}\text{s}^{-1}$ from a rural Canadian setting. The latter range is within the range 0.1 – 9.4 $\text{cm}^{-3}\text{s}^{-1}$ with a median value of 1.2 $\text{cm}^{-3}\text{s}^{-1}$ reported by Yli-Juuti et al. (2009) for a station in rural Hungary. The two formation rates of the present study cover the range 0.1 – 1.4 $\text{cm}^{-3}\text{s}^{-1}$ for the 25% to 75% percentiles (see Table 3), which covers the range of 0.05 to 0.13 $\text{cm}^{-3}\text{s}^{-1}$ given by Vencaz et al. (2009) for a remote site in the Himalaya. The environmental conditions at the Siberian station Tiksi at the coast of the Laptev Sea may come closest to our Arctic setting. From this site Asmi et al. (2016) published formation rates of 0.01 to 0.41 at an unspecified particle size.

In terms of 25% to 75% percentiles the particle growth rates of the present study range from 0.4 to 1.4 nmh^{-1} in the range 5 – 25 nm and 1.0 to 1.8 nmh^{-1} in the diameter range 10 - 50 nm, which is near the range of results of 1 – 2 nmh^{-1} derived by Ström et al. (2009) for new particle formation in the lower boundary over Ny-Ålesund, Spitsbergen but considerably lower than the maximum growth rate of 3.6 nmh^{-1} reported by Asmi et al. (2016) for July at the Siberian station Tiksi at the coast of the Laptev Sea. For open ocean new particle formation events over the North Atlantic O’Dowd et al. (2010) report a “typical growth rate” of 0.8 nmh^{-1} whereas Ehn et al. (2010) give an average growth rate of 3 nmh^{-1} . We note that the average length of DGR-events was 10 ± 1 h, (one standard deviation). Further details about the connection between growth rates and the two formation rates can be found in the supplement.

Statistics	J11	GR11	J22	GR22
Minimum	0.1	-1.2	0.1	-0.1
25%	0.4	0.1	0.2	1.0
50%	0.7	0.4	0.3	1.4
75%	1.4	0.6	0.7	1.8

Maximum	19	2.2	22	4
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Table 3 Statistics of particle formation rates of DGR-events J11, and J22, ($\text{cm}^{-3}\text{s}^{-1}$), at the nominal geometric mean diameters 11 nm, and 22 nm and corresponding diameter growth rates GR11, and GR22, (nmh^{-1}) in the two diameter ranges 5 – 25 nm, and 10 – 50 nm.

Finally, I think the point referee #2 had in their point 3 was not how the PCT events had been defined, but that there was no comparison with similar results in the literature. I think whether these the observations defined this way definitely represent NPF is likely to be a moot point, however a paper of this nature should be placed in the context of other works where possible. A quick comparison with other works would be welcome.

We are afraid to admit that we cannot find any reports in the literature that could be used in comparison to our findings concerning PCT-events. Any suggestions by the reviewer would have been highly welcome.

As a separate issue concerning referee #1's point regarding trajectory accuracy, I do not regard the issue of back trajectory validation to be one that is easily solved. Because the arctic region is so data-poor, it is highly likely that the wind field used has assimilated (and placed great weighting on) local weather observations. As a result, good agreement here is not surprising but does not necessarily validate the fidelity of the model field away from the observations. I think it should be sufficient to state that there are inherent uncertainties with this technique.

In the first revision of the manuscript we did address referee #1's point regarding trajectory accuracy by adding "Trajectories extending backwards for ten days are inaccurate at origin due to the trajectory uncertainty of 25-30% of its length, (Stohl, 1998)". Please let us know if you want us to elaborate the point of trajectory accuracy any further.

Literature

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