

Interactive comment on “Potential of polarization lidar to provide profiles of CCN- and INP-relevant aerosol parameters” by R. E. Mamouri and A. Ansmann

Anonymous Referee #3

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General Comment

This manuscript will ultimately be a good contribution to the literature, continuing the efforts that this group is leading in attempting to utilize remote sensing to retrieve profiles of cloud active aerosols. I have no issues with the methodology development itself. However, I list a number of minor comments, and a couple major ones. The first major point is that the authors have used a parameterization to represent pollution INPs that was developed based on data that appears not at all to have included strong anthropogenic pollution influences. In fact, it appears to have effectively isolated such influence by restricting relation only to aerosols larger than 500 nm diameter in regions

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away from urban areas where those number concentrations never exceeded about 10 per cc, and then appear likely the consequence of sampling mineral dust aerosols. This application of the DeMott et al. (2010) parameterization for pollution for the first time is a misstep, one that could then be erroneously referenced as indicating that pollution particles are efficient INPs, perhaps as efficient as mineral dust under some conditions. This has never been shown using actual data to my knowledge, and if such data did exist, then surely evidence would be widespread throughout the ice nucleation literature. It is not. I suggest that, alternately, this parameterization could be used as a contrast to others that are purely for mineral dusts, but should not be ascribed in any manner to pollution particles. I do not know if that is possible, or if it meshes with the lidar profiling that has been done. I will guess that this is problematic for the authors because they are using the lidar response to anthropogenic haze and biomass smoke specifically for a category termed “continental pollution” aerosols. The solution to this problem is not clear, unless specific parameterizations are proposed for smoke and/or pollution on the basis of data in the literature.

A few other key things needing attention in my opinion are:

- 1) It would be useful to introduce the fact that aerosol numbers are not corrected for RH growth that may be present in the ambient observations. That is, dry aerosol distributions are not used.
- 2) Terminology for relevant quantities should be unified throughout the paper (see specific comments)
- 3) The parameterization of Steinke et al. (2015) is applied for deposition nucleation, but with little concern for the fact that that study exclusively focused on Arizona Test Dust as the basis for the parameterization. It is widely known in the ice nucleation community that ATD is not a good atmospheric dust surrogate. Consequences seem apparent in the results given here, yet no qualifications are given.

Specific Comments

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1. Introduction

Page 34152, line 1: After introducing the shorthand APC for aerosols and CCN, why not use it for INP, for example APC_{INP} instead of creating an alternate form (INPC)? Furthermore, a more standard notation (n_{INP}) is used in section 3. Are there reasons not to use a single notation, preferably the latter form, from the beginning of the paper?

Page 34152, line 5: The same issue is present for CCNC versus (APC_{CCN} already defined, and $n_{CCN,xx}$)

Page 34152, lines 11-12: Please explain what is meant by mineral dust particles being fully activated below -20°C . Many studies, including Murray et al. (2012) indicate that nucleation is not fully stochastic for all INP and that the active fraction and site density for many mineral dusts are strongly temperature dependent even below -20°C . Hence, I believe this statement to be false.

Page 34152, lines 14-17: Herein starts a problem in including a variety of aerosol types, indistinguishably with regard to ice nucleation, as “continental particle mixtures” that always contain efficient INPs. While it is certainly true that all of these particle types are emitted from continents, they are distinct types that need distinct definition as INPs. This problem becomes a flaw when it is later assumed that the DeMott et al. (2010) parameterization can encapsulate these specific sets of sources, and pollution (“anthropogenic haze”) INPs in particular. I will elaborate on this below.

Page 34153, line 16: Where do soil dust particles fit (soil and road dust mentioned on the last page)? Are these assumed the same as mineral dusts? Reducing this category now to anthropogenic haze and smoke mixtures is unfortunate, as smoke particles have been identified (though not parameterized) as INPs in published studies (e.g., McCluskey et al., 2014), while little or no data exists for anthropogenic particles contributing as INPs at mixed phase cloud temperatures in the free troposphere (unless the authors can prove so). There is stronger evidence for sources of organic and biogenic INPs from soils and plants that are present in air over continents at sizes

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above 500 nm. Page 34154, line 9: Can you explain what is meant by a simulation study? Does that mean using a global aerosol model as ground truth? If so, please state so.

2. Instrumentation

Page 34158, lines 10-13: These statements bring to mind for the first time that these numbers could drastically differ depending on RH, and to this point in the paper, nothing is said about how this factor is dealt with.

3. Methodology

Page 34162: Is some typical hygroscopicity value assumed in (4) to (6)? And doesn't one require dry sizes first, prior to computation of CCN number?

Page 34163: Absent a mention of assumed composition, it is not clear where the supersaturation associated with APC_{40} comes from.

Page 34164, line 8: There is no justification given (and none possible in my opinion) for using the parameterization of DeMott et al. (2010) for continental pollution for the reasons already stated. Projects are listed in the DeMott et al. supplemental section that include mineral dust influences, but none for which pollution was a key type, and the creation of an INP surrogate via particles larger than 500 nm appears to have been done specifically to avoid pollution if possible (considering the impacts of pollution on such INP-size relationships evident in Richardson et al, 2007, referenced by DeMott et al., 2010). Figure S1 in DeMott et al. (2010) shows a range of aerosol concentrations that does not seem to reflect what might occur for heavily polluted air. It is likely that data and sampling scenarios from that figure are available, and could be used to assess if any pollution influences were included in that “global” parameterization. That it may not be the case always that pollution particles stay at diameters below 500 nm creates a dilemma for generally applying the D10 parameterization, but one that has to be stated as a dilemma nonetheless, not solved by assuming that the parameterization is

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valid for pollution. The parameterization can be used as a contrast for a more globally-averaged INP, but cases where it is falsely enhanced by pollution should be specially noted and probably omitted from consideration (problematic for this paper that creates a category for pollution, but with no data on INP to ascribe toward it).

Page 34165, line 1: Wex et al. (2014) do not discuss ice activation by purely anthropogenic particles, just coated ones. Please correct. In fact, this surrogate coating for anthropogenic organic particles has no active impact (positive) on ice activation in the temperature regime examined.

Page 34165: It is necessary to note that the parameterization of Steinke et al. is for Arizona Test Dust, an atmospheric surrogate that exceeds the INP activation properties of Asian or Saharan dust (see, e.g., Niemand et al. 2012). This probably explains the overactive deposition process in figures shown in this paper in comparison to immersion freezing by mineral dust.

Page 34166, lines 1-4: Justification for the statements made here is not given, and these statements are problematic. Why assume a constant S_{ICE} , when this is a variable? How is Steinke et al. (2015) applied for pollution? These particle types were not examined by those authors. The lack of detail and discussion here is unacceptable.

4. AERONET observations of the relationships of APC and ASC with AEC

Page 34168: Were RH effects removed somehow? The title of Shinozuka et al. (2015) implies use of dry particle size, but that is not the case here. I could not resolve your discussion of any implications for the present study.

5. Lidar estimates. . .

Page 34175: Please explain better the reason for shifting the temperature profile by 15K. Is it meant to mimic the presence of similar aerosols at lower temperature? D10 is not for pollution though, so omit or apply only the dust number to it. That would be justified.

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Page 34175 and Figures 9 and 10: The higher values in N12 versus D15 could result from substantial ASC existing below a radius of 250 nm, as stated, but could also relate to the failure to convert to dry size distributions before comparing a parameterization based on size with one based on surface area.

Page 34175 and Figure 10: Why is the S15 parameterization shown for temperatures warmer than it is specifically valid. This is an issue because deposition does not typically occur for these conditions, at least on the basis of laboratory observations (i.e., it is a more typical behavior in the cirrus temperature regime). Then one wonders why S15 exceeds N12. This is partly a difference in INP type and partly because it is probably invalid to plot S15 at these conditions.

6. Conclusions

This section now requires revision for the discussion about pollution. Although the lidar may detect it, one cannot escape the fact that INP parameterizations have not been developed specifically for pollution. More likely, they have been developed to avoid it, because it does not represent a very efficient INP source and so is extremely poorly characterized. Similarly, the lidar may detect biomass burning layers, but the authors should then perhaps work with others who have collected data for such particles in order to apply a specific parameterization. These are clear current weaknesses in this paper, and clear future needs that should be discussed.

Technical editorial comments

Page 34151, line 13: "can be used" for "is requested"

Page 34152, line 10: "efficacy"

Page 34156, lines 15-16: please note that this sentence repeats the first sentence of this section.

Page 34158, lines 6-9: Suggest rewriting for awkward language as, "ASC from AERONET is almost 98

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Page 34159, lines 6-7: “profiles” repeats. Remove one.

Page 34161, line 26-27: remove “to introduce”

Page 34162: Begin sentence “Determination of the specific. . .”

Page 34162: Again, use of multiple terms to define the same quantities inside and outside of equations here is very confusing and unnecessary. Section 3.3 even amends the APC term to include supersaturation now, but this could have been done at the beginning of the paper. Nevertheless, it is again redefined starting in Eq. 4.

Page 34163, line 10: missing “our” before “own”.

Page 34165, line 11: replace “leave out to” with “do not”.

Page 34172, line 12: use “disturb” for “disturbed”

Page 34175, line 7: replace “after” with “of”

References

McCluskey, C. S., P. J. DeMott, A. J. Prenni, E. J. T. Levin, G. R. McMeeking, A. P. Sullivan, T. C. J. Hill, S. Nakao, C. M. Carrico, and S. M. Kreidenweis, 2014: Characteristics of atmospheric ice nucleating particles associated with biomass burning in the US: prescribed burns and wildfires, *J. Geophys. Res. Atmos.*, 119, doi:10.1002/2014JD021980.

Richardson, M. S., P. J. DeMott, S. M. Kreidenweis, D. J. Cziczo, E. Dunlea, J. L. Jimenez, D. S. Thompson, L. L. Ashbaugh, R. D. Borys, D. S. Westphal, G. S. Cassuccio and T. L. Lersch, 2007: Measurements of heterogeneous ice nuclei in the Western U.S. in springtime and their relation to aerosol characteristics. *J. Geophys. Res.*, 112, D02209, doi:10.1029/2006JD007500.

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