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Interactive Comment

Interactive comment on "Characterization and first results of an ice nucleating particle measurement system based on counterflow virtual impactor technique" *by* L. P. Schenk et al.

Anonymous Referee #2

Received and published: 20 November 2014

Schenk et al. (2014) present a description of a system designed to measure the composition of ice-nucleating atmospheric aerosol particles (INP). The system consists of a combination of three previously-developed systems, an ice nucleation chamber (FINCH), a pumped counterflow virtual impactor (PCVI) and a single particle aerosol mass spectrometer (ALABAMA). A characterization of the lower cut off diameter of the PCVI system was performed, and some limited results on the particles passing through the FINCH-PCVI, interpreted as INP, are presented. These results consist of (i) a discussion of a major sampling artifact (both water droplets and ice crystals were sampled by the PCVI during their atmospheric study), (ii) a comparison of the IN concentrations

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determined as the particle concentration behind the PCVI with the IN concentrations determined by FINCH light polarization measurements, and (iii) a discussion of a very limited set of atmospheric measurements, which is very brief because IN concentrations from the study are discussed elsewhere (cited as Frank et al. 2014) and because the ALABAMA instrument collected only seven mass spectra due to experimental difficulties.

While these studies were well-motivated, I find it difficult to recommend this manuscript for publication in AMT. It is my understanding that AMT aims to publish new experimental or analysis techniques and the present manuscript does not provide an adequate characterization of this specific PCVI design nor represent a new technique. The previous PCVI publications cited by the authors already provide an adequate description of the PCVI technique and the minor differences between the ice nucleation chamber and mass spectrometer used in this study are not significant to the use of a PCVI. The PCVI characterization is similar to that performed by Boulter et al. (2006) and adds a few measurement points to those used in that study. These measurement conditions appear to have caused leakage in the PCVI (see below). The IN results from the PCVI suffer from major limitations due to either extremely poor sampling statistics, the possibility of major instrumental artifacts, or both. A paper about the performance of the commercial Brechtel PCVI would be a useful reference for future studies, but such a paper would need to perform significantly more tests and preferably use more than one pf the commercial PCVIs.

My understanding of the aims of AMT may be incorrect, in which case I would suggest that the paper should be revised considerably before publication for the following reasons:

1. The characterization of the PCVI is too insubstantial. Only a few points were tested, and the influence of PCVI inlet/outlet pressure was not examined (only flow rates). These pressures may change when FINCH or other instruments are connected, or when the temperature of the PCVI input air is lowered by FINCH.

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In addition, the PCVI should not be called an IN-PCVI, as it is not selecting IN but only large particles. As pointed out by the authors, the PCVI technique is well established, and renaming the PCVI as "IN-PCVI" implies that the authors have somehow changed the technique to make it somehow selective towards IN.

2. The major issue of small particles leaking through the PCVI should be discussed. Fig. 4 shows that about 2% of the small particles that were sampled leaked through the PCVI. After reading the Corbin et al. (2012) and Cziczo et al. (2003) studies cited in the introduction, it seems that such leakage should be expected at the low flow rates that were used in the present study. The Corbin et al. (2012) study corrected for leakage using background measurements taken at low saturations and the Cziczo et al. (2003) study avoided leakage by operating their PCVI at higher flows. Why were the current flows chosen as they were, if the PCVI leakage increased significantly as a result and the FINCH was designed to operate at higher flows (Bundke, 2008)?

The reported data were probably affected by this leakage, for example on lines 268-277 the conclusion "only 13 % of the sampled INP are larger than 0.55 μm" could also be interpreted as meaning that small leaked particles represented a major fraction of the measured particles. This alternative interpretation would be fully consistent with the fact that smaller particles are far more numerous in the atmosphere, so that the their transmission (transmission efficiency multiplied by number concentration) would be higher.

The issue of leakage affects the interpretation of the results. For example, Fig. 4 shows an increased transmission efficiency for particles 0.5 to 3 microns in size. Then Fig. 9 suggests that particles 0.6 to 4 microns were observed at higher concentrations behind the PCVI (noting the size comparison issue described below). These particles could therefore be due to enhanced PCVI transmission or in order words reduced PCVI selectivity.

Furthermore, extrapolating the curve of Fig. 4 implies that the PCVI would do a very bad job of removing small particles. This extrapolation is probably wrong, but it would be important to perform additional measurements to show that. In my opinion, the manuscript would be very much improved if the transmission efficiency of particles below the D50 was characterized and minimized.

3. Fig. 4 and Eq. 3 should not be labelled "transmission efficiency" if the data have been arbitrarily scaled. The current labelling implies that 100% of 10 um particles would be sampled downstream of the PCVI, which has not been shown. The true transmission efficiency would affect the calculated IN concentrations.

4. Inferring the influence of droplet contamination by repeatedly fitting the data after removing arbitrarily defined outliers (i.e., Swat < 1.01, 1.02, etc.) is not a robust way to determine the impact of droplets on the measurements. If IN transmission in the PCVI was < 100%, then a 1:1 fit would not be meaningful. For example if large particles were transmitted with only 80% efficiency in Fig. 4, then a 1:1 fit would correspond to a 20% contamination. Anyway, there is no physical reason to calibrate the PCVI performance using the performance of the FINCH optics. The two approaches would have from different strengths and limitations. Ideally, the system would be characterized directly using a known aerosol.

5. Two example mass spectra are shown, but the possibility of leaked particles and of these mass spectra representing droplets rather than IN (was the FINCH water saturation below 100% when these particles were measured?) together with the difficulty of interpreting seven mass spectra in the context of the entire INUIT-JFJ campaign makes it very difficult to assign any meaning to these spectra. Why were these 2 spectra chosen? And are the authors really able to confidently assign the elemental compositions shown in the figure? If yes, how?

Other comments:

- The comparison of linear fits in Fig. 5 is difficult to physically justify. It seems to me that these plots would represent the partial differential of the PCVI transmission

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function, which I would not expect to be linear. Especially Fig. 5b needs more than 3 data points to justify a linear fit.

- The comparison of the APS and OPS in Fig. 9 needs to consider the potential for differences between the instruments. In addition to the expected differences between any two instruments, differences should be expected since the measurements represent different physical properties (aerodynamic and optical sizes). This fundamental difference would not be adequately corrected for by normalizing the OPS to the APS measurements at 0.55 um as was done.

- Section 2.1, please give a short address for Brechtel Manufacturing.

- Page 10591 line 9, "the number size distribution measured with the counterflow switched off was taken as reference", was this reference taken between all measurements? How reproducible was it? I imagine that manual dispersion with a balloon would be highly variable?

- Line 9 page 10592 please remove the word "extended" before characterization.

- Line 1 page 10595 please briefly define the "FINCH closed loop".

- Paragraph 1 page 10596 the feasibility of single particle mass spectrometry coupling to a PCVI has already been verified.

- The 1:1 line in Fig. 7 was invisible on my printout and the authors might consider making it thicker.

- Line 16 page 10597, "calculated" or "fitted"? Was the uncertainty in both variables taken into account during fitting?

- Line 16 page 10598, "consistent with the findings in Section 3.1" - are these not exact the same findings?

- Line 28 page 10598, what reduced counting efficiency? Please explain this counting efficiency and the tests that were carried out.

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- Line 5 page 10599, what does the acronym "PSI" mean?

- Line 1 page 10600, the word "ambitious" is used inappropriately.

- Figure 8, please include error bars to reflect the uncertainty and confidence level discussed in section 3.1. Please include water saturation and not just ice saturation, since Swat is the more important variable and is discussed extensively in the text.

Interactive comment on Atmos. Meas. Tech. Discuss., 7, 10585, 2014.