Atmospheric Measurement Techniques Discussions

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Comment on amt-2021-26

Anonymous Referee #1

Referee comment on "A phase separation inlet for droplets, ice residuals, and interstitial aerosol particles" by Libby Koolik et al., Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2021-26-RC1, 2021

The manuscript by Libby Koolik and co-authors provides a thorough description of the novel SPIDER inlet, which can discriminate interstitial aerosol particles, cloud droplet and ice crystal residuals in ambient mixed-phase clouds. SPIDER uses a combination of a large pumped-counterflow-virtual-impactor (rejection of interstitial aerosol particles) and a droplet evaporation section and a PCVI (rejection of droplet residuals), which allows that only ice crystal are transmitted in the system. A set of laboratory experiments is conducted to verify the transmission of ice particles in the main sample flow of SPIDER, and the rejection of interstitial aerosol particles and cloud droplets. The rejected unactivated aerosol particles and the cloud droplet residuals can thereby still be measured in the pump flows of the PCVIs. The SPIDER inlet was also successfully tested in a field campaign during in-cloud conditions, measuring the size distribution ice crystal residuals.

Only a few of such ice-selective inlets exist worldwide, therefore the novel SPIDER inlet is an enrichment for the field of mixed-phase cloud reserach.

The manuscript is generally well written, and I only have the following minor comments.

General comments:

- The citation in the introduction needs to be improved. Often references are repeated and sometimes not specific enough, or not appropriate. See specific comments below.
- Transmission efficiencies: As you claim that the SPIDER inlet is able to sample simultaneously interstitial aerosol particle, droplet residuals, and ice crystal residuals, it would be needed to adress the transmission efficiencies for the different channels. To my understanding this can be retrieved from the existing measurements.

- No particles smaller than the lower size limits of the OPC/OPS (~0.3 µm) were measured. Hiranuma et al. (2016) used a condensation particle counter to adress the question of transmission of small particles in the different channels. In my opinion, such measurements can help to verify that e.g. no small aerosol particles or small evaporated cloud droplets are able to be transmitted in the droplet or ice channel, respectively. Further, such measurements can also be used for transmission efficiency measurements at the interstitial aerosol channel. This is rather a recommendation for future work and does not imply that new measurements need to be presented in this manuscript.
- Ice crystal residuals and cloud droplet residuals are not necessarily only INPs and CCNs, respectively, as cloud droplets can also contain scavenged particles and ice residuals can also contain droplet residuals due impact from secondary ice crystal formation (e.g. see discussion in Kamphus et al., 2010). Based on your statement in the introduction (lines 67 – 70) and in the conclusions (lines 290 – 291), you should be more specific about what ice residuals and cloud droplet residuals are when you sample them with SPIDER. Are you truly only measuring INPs and CCNs?

Specific comments

- Abstract: I suggest to give the size range of ice particles which can be analyzed with SPIDER.
- Lines 26 – 28: I assume that the most important criteria about the Storm Peak Laboratory campaign was that you were able to sample ambient supercooled clouds, which I would mention here.
- Lines 29 – 30: „Possible design improvements of SPIDER are also suggested", are you refering here to using more robust OPCs or OPCs with a higher resolution? It is not clear to me what those design improvements would be.
- Lines 33 – 34: „Mixed-phase clouds are important factors in aviation and climate (Shupe et al., 2008)", please add more and also more recent literature, as e.g. Lohmann et al. (2017), McCoy et al. (2016).
- Lines 38 – 40: „Mixed-phase clouds are particularly complicated because the partitioning of phases is critical in assessing these effects (Atkinson et al., 2013; Hirst et al., 2001; Korolev et al., 2003; Shupe et al., 2006)." Atkinson et al. (2013) was not investigating this specific research question; also, there is more and also more recent literature about this, e.g. Korolev et al. (2017), Tan and Storelvmo et al. (2019), just to name a few.
- Lines 41 – 43: „This has resulted in a global effort to study these clouds (Abel et al., 2014; Davis et al., 2007a; Hiranuma et al., 2016; Kupiszewski et al., 2015; Mertes et al., 2007; Patade et al., 2016)." Also here, include more recent studies, e.g. Lohmann et al. (2017), Lowenthal et al. (2019), Schmidt et al. (2017), Ramelli et al. (2021), Ruiz-Donoso et al. (2020).
- Lines 45 – 46: „At this saturation aqueous droplets are the favored state and particles that activate are termed cloud condensation nuclei (CCN) (Lohmann and Hoose, 2009; Wang et al., 2012)." Those references are not specifically investigating warm cloud actication. I recommend to change the references to e.g. Pruppacher and Klett (1997).
- Lines 42 – 49: „Ice can form homogeneously, via spontaneous nucleation of ice in a solution droplet, at temperatures below -40°C (Atkinson et al., 2013; Kamphus et al., 2010; Korolev et al., 2003; Storelvmo et al., 2008; Verheggen et al., 2007; Wang et al., 2012)." None of those publications focus on homogeneous freezing of solution droplets, I recommend to reference Heymsfield et al. (2017) or Koop et al. (2000).
- Lines 49 – 51: „At higher temperatures, ice forms heterogeneously through different

pathways promoted by ice nucleating particles (INPs) (Atkinson et al., 2013; Kamphus et al., 2010; Lohmann and Hoose, 2009; Storelvmo et al., 2008; Tsushima et al., 2006; Verheggen et al., 2007; Wang et al., 2012)." I recommend to reference rather review papers specifically on INPs, e.g. Hoose and Möhler (2012), Kanji et al. (2017).

- Lines 51 – 52: „The specific properties that determine an effective INP remain poorly understood (Shupe et al., 2008)." Shupe et al. (2008) did not investigate INP properties. I suggest to reference Kanji et al. (2017).
- Lines 62 – 63: „Motivated by climate change, estimated to be warming approximately twice as fast as the global average (Verlinde et al., 2007)…" please reference more recent literature here.
- Line 94: Please introduce the abbreviation for IS-PCVI.
- Lines 102 – 103: I suggest to include the expected D50 for those flow settings.
- Line 103: I suggest to move „the PCVI PF, AF, and SF at 8.0, 2.5, and 1.0 L min-1, respectively." to below when you introduce the PCVI, e.g. to lines 111 – 112.
- Lines 103 – 105: I suggest to give the Weber Number here (0.3) in comparison to a value of 10 and larger when droplet breakup is expected.
- Section L-PCVI: Based on the experiments presented in Fig. 3 and 4 you could determine the transmission efficiency of interstitial particles in the PF, taking into account the dilution ratio.
- Section Droplet Evaporation Chamber: What is the residence time of cloud droplets and ice crystals in the droplet evaporation section, and does this impact the partial evaporation?
- Section Sustaining Ice Crystals: It is not clear to me which „chamber" is meant here. Was the droplet evaporation chamber used to induce homogeneous freezing and form ice crystals? If so, how could you determine if ice crystals survived in the droplet evaporation chamber? As this section belongs to 4.2, I understand that the intention is to test if ice crystals are sustaining in the droplet evaporation chamber, which, in theory, is not needed, as the droplet evaporation chamber is maintained at ice-coated walls (saturated with respect to ice). Maybe you should consider to move this section to 4.3.
- Lines 198 - 199: What is the size of the formed ice crystals? And how were they validated visually?
- Line 206: Which particle sizes are generated with this AS solution?
- Lines 234 – 235: I recommend to not include the dicsussion about the different OPCs used at the interstitial aerosol channel and cloud residual channel, as you don´t show those results.
- Line 241: I suggest to give a number for „low aerosol particle conditions".
- Lines 249 – 250: This is a repetition from your statement in line 246, I would delete one of the sentences.
- Lines 253 – 254: Where is this „inadvertent transmission" coming from?
- Lines 290 – 291: „Ultimately, information on cloud nucleation capabilities of various aerosol particles could be compared to laboratory work and integrated into climate models (Shupe et al. 2008)" I recommend to cite also more recent literature here.

- The author contributions is missing
- Figure 4: There is no panel (c)
- Figure 7: Please indicate that this is the transmission efficiency from the PCVI
- Figure 8 (and related discussion in the text): Another important parameter for the description of these timeseries would be the ambient temperature, which one could relate to the nucleation temperature of ice crystals in the cloud. More, on 2019-01-21 at ~ 18:30, ice crystal concentrations are as high as 0.03 cm-3, which is a relative high INP concentration at temperatuers < -20°C (the lower limit of ambient temperature, as I understand from line 240). Thus, is this an indication for an impact of sampling ice

crystals formed by secondary processes?

- Figure 9 (panels c, d): Also here, are your measurements impacted by secondary ice crystal production in the smaller size bins? It is quite surprising that the concentration of ice crystal residuals increase towards smaller sizes.

Editorial comments:

- Line 92: Please introduce the abbreviation „SPIDER", since it is the first time using it in the main text.
- Line 93: Please introduce the abbreviation „L-PCVI".
- Line 222: The abbreviation for INP was introduced earlier.
- Lines 304 – 305: Remove those test citations.
- I suggest to either use supersaturation or relative humidity with respect to water (especially in the droplet evaporation chamber section).
- As the abbreviation for ammonium sulfate (AS) is only used a few times in the manuscript I suggest to use the full name.
- The resolution of Figures 3, 4, 8, and 9 can be improved.

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