Interactive comment on "Single-particle characterization of ice-nucleating particles and ice particle residuals sampled by three different techniques" by A. Worringen et al.

Anonymous Referee #1

Worringen et al. paper present the characterization of single-particle ice-nucleating, based on three different techniques. The paper is very interesting and a valuable contribution to the ice nucleation community; however, major corrections are needed before this manuscript can be accepted. Many sections of the paper are not clear and are written with not good flow, forcing reading the sections several times in order to understand it. It seems that different people wrote different sections and the writing is not uniform. That is why I will recommend the authors to rewrite so sections in order to increase their clarity and flow.

Details on the PCVI and CVI that were used in this work are missing, for example what type or model were used, what conditions were used in order to get cutoff sizes?
Details on the used instrumentation is given in the companion paper in AMTD (Schenk et al., 2014), which was just published during the time of the review. This paper is correctly referenced now. Details on the Ice-CVI are given in Mertes et al. 2007, which was referenced in the manuscript.

Detail on comparison between the three methods is missing, the authors mentioned that they were not measure at the same time, but do the authors know if all methods can see the same particles, and detect the same IN.
We are totally aware that in many cases parallel sampling was not performed. This is shown in Fig. 1 and discussed in the details. This is certainly a limitation of the current data set. Still, we believe that this data set is valuable for the scientific community. In addition, we address this point in the recommendation for future work (new chapter in the manuscript).

Where there any comparisons between these methods before the fieldwork to make sure they all see the same thing?
There were comparative previous measurements in a joint field campaign, whose results were used to optimize the instrument performance. However – added in the new recommendation section – we have to conclude that additional comparative measurements are necessary in future (an according laboratory campaign is scheduled for March 2015).

Each method had different artifact that should be mentioned with more details
We do not understand this comment as a major part of the manuscript addresses the artifacts.

It seem that the sampling with the different methos did not had any overlap, where each method used at different time, the author combine all the data to one data,
The author combine the data from all method at some part for the entire time period, does all days had the exact same conditions, is it possible to assume that some days had different IN types and the differences between the instrument come because of that?
See above (comment on non-parallel sampling).

Specific comments
Abstract:
The abstract is not clear I will recommend rewriting it again.
We agree and have completely rewritten the abstract.

Page 23029 lines1-8
The sentence is too long I will recommend to separate it to at least two sentences.
Changed as requested, now three sentences.

Page 23029 line 2
I think using the two tern ice-nucleating particles (INP) and ice particle residuals (IPR) is a bit confusing for the reader, the term ice-nuclei residuals (INR) will be a better term since the analysis is for IN particles. If the author want to use both I will recommend on using ice crystal residuals (ICR) instead of IPR.
We did not change the acronyms, as they were chosen by the INUIT consortium (after long discussions), and are used in all INUIT papers.

Page 23029 lines 11
Use the term January-February and not January/February
Changed.

Page 23029 lines 12-19
Sentences are not clear
We have added the word "instrumental" to the expression "contamination artifacts". In addition, the measurement artifacts are now explained explicitly.

Page 23029 lines 14
I think it will be better to use the term FINCH+ PCVI then the term FINCH+IN-PCVI, it is obvious that the PCVI is used for IN residuals and that that FINCH was used for IN anlysis.
We did not change the acronyms, as they were chosen by the INUIT consortium (after long discussions), and are used in all INUIT papers.

Page 23029 line 24
The choice of word yielded is not recommended I will replace it with something else
Replaced by "obtained".

1 Introduction

The introduction is missing many things as the principle behind a PCVI or CVI method Examples of laboratory and field works that used this technique not just for IN but also for CCN Elementals that should be expected in IN particles
The PCVI and CVI principle were discussed in previous literature, e. g., Boulter et al. (2006), and are thus not explained in the manuscript. Technical details of the used specific instruments are given in the cited referenced (Mertes et al., 2007; Schenk et al., 2014).

Tell the reader about previous IN measurement in the Jungfraujoch station.
We have added an according section to the introduction.

Page 23030 line 17-18
I will recommend not to use the sentence as it is because it seem negative. Instead, I recommend the authors to use a sentence like this: although, in the last decade, large attention has been given to field studies at different parts of the world (e.g., Prenni et al., 2009a,c; Santachiara et al., 2010; Ardon-Dryer et al., 2011; Conen et al., 2012; Ardon-Dryer and Levin, 2014), these measurement covers small fraction of the world and many filed work are still needed.
Changed as requested.

Page 23030 line 20
I think this will be a better way (e.g. Hoose and Möhler, 2012, and references therein)
Changed as requested.

Page 23030 Line 24-26
The sentence is not clear and it should rewriting
We have rephrased the sentence

Page 23030 line 26- page 23030 line1
Rewrite the sentences
Changed as requested

Page 23031 line 4
Replace the word 'reached' with another word
Replaced by "realized".

Page 23031 lines 11-14
The authors should not include the FRIDGE in this part, since it is impossible to separate the activated particles from the non-activated ones in FRIDEG and FRIDGE samples the particles on a substrate.
Indeed they are not separated by FRIDGE, but they can be distinguished. Sentence was rephrased.

Page 23031 lines 20-21
Use the term PCVI instead of pumped counterflow virtual impactor
Changed as requested.

Page 23031 lines 24-28
Rewrite the sentence it is cumbersome
After reading this sentence several times, we came to the conclusions not to change it. We believe that the message is quite clear.

2 Experimental

I suggest to separate this section to three parts sampling site, sampling the ice crystals (all CVI) and last section analyzing of the IN property (as microscope and laser) or combin it with the CVI part.
We agree and have restructured the experimental section

Page 23032 lines 1-10
Rewrite it, the flow of this paragraph is bad the description is bad, give more explanation on the station, why sampling there.
We agree and have rewritten this section.

A figure with experimental set up will be very helpful for the reader; it might be even better then table 1.
We disagree and think that the information in table 1 is sufficient.

2.1 INP/IPR differentiation

The whole section is not clear it will be better to use each section in the explanation of the chambers or setup instead here
We agree and have changed the according section

Page 23032 line 12
Bundke et al., 2008 did not use a PCVI in this paper, rewrite it
Bundke et al. 2008 introduced the basic principles of FINCH. We have added the reference Schenk et al. 2014 who describe the current instrument setup.

Page 23032 lines 1-18
"Two-stage impactor system (see above)", what does the author mean, not clear. The explanation seems to

appear afterword is at line 20.
Section was rewritten (see above)

2.1.1 Coupling of FINCH and IN-PCVI
Page 23033 line 6
Use the word by, counted by an optical
Use the word was and not the word is, "The OPC used in this instrument was able"
Changed as requested.

Page 23033 lines 9-11
Not clear
We still believe that this sentence is clear.

Page 23033 line 13
There is not reference for this paper Schenk et al., 2014, there is one for Schenk, 2014
Corrected

Page 23033 lines 12-15
Rewrite this sentence, say how they were separated with what, this is a suggestion for this sentence: The activated IN grow into an ice crystals. The ice crystals were separated from the non-activated particles and from the small-supercooled droplets by using the PCVI
Page 23033 lines 14-21
These sections are not clear at all
We have rewritten these sections to make them clearer.

What does it mean: "This is realized by a counterflow that meets the FINCH output flow which is at the same time the IN-PCVI input flow".
Does the authors mean that the flow from FINCH was the same as the input value that needed for the PCVI in order to get a cut off size of 4.5-8 micron, is this the 50% cut off?
Rewritten to make it clearer.

What values were used for input and output for the PCVI that gave values of 4.5-8 micron? please write it.
Please refer to the according publication (Schenk et al., 2014).

2.1.2 Ice-CVI
The whole section is not clear, as setup figure would be helpful
This section is only a short summary, as the setup was described in detail in the cited paper by Mertes et al. 2007.

Page 23033 line 24
Delete the words "so-called"
Changed as requested.

2.1.3 Ice Selective Inlet (ISI)
Well written, it is clear.
Thank you!

2.1.4 Laser Ablation Spectrometry (LA-MS)
Page 23035 lines 9-10
What is the meaning of 104h and 32h?
It is hours. This is now explained in the text.

Perhaps this sentence will be better: Total of 1809 particles mass spectra were sampled with the LA-MS. A

1663 mass spectrum were when the Ice-CVI was in use and 146 when the ISI was in use.
Changed as requested.

2.2 Sample analysis by electron microscopy

Were only 46 particles analyzed or each sample contained many particles, please explain. In addition the author does not mention after which equipment this sampling were taken from.
We are talking about samples. We now include the number of samples obtained from each technique (as requested). Thus, the meaning of "samples" should now be clear.

2.3 Particle classification
Is this a combination of all the analysis methods if so say it?
The chapter particle classification is now part of the electron microscopy chapter. Thus, it is now clear that we talk about particles classification in SEM analysis.

Why there is a classification of droplets if the author mentioned that all droplets should evaporated or cut by the CVI? is there a sampling error, if so mention it before, which sampling method gave droplets?
In principle, the particles should have dried until reaching the impactors. However, evidently this was not always the case, and some arrived as droplets. Thus, we included a separate class for them.

Why there is definition of other with many elements? Are the materials are what the CVI built from or material of the grid itself, please explain.
These are not artifact particles. The 'other' group simply contains different particle types which have only singular or very rare occurrence. We have added a comment on their low abundance.

Was there any compression between the different analysis methods? Or the author assume they are all measure the same parameters?
The major objective of the present manuscript is to investigate the differences of three techniques applied to separate and analyze INP/IPR! We discuss the differences of the techniques in detail and it is not assumed that they measure the identical parameters.

Page 23036 line 1
Use the word were and not the word are
Changed as requested.

3 Results

I will recommend to combined the result with the discussion it will be better to understand and will allow the author to make stronger points about their findings
After careful consideration of this comment (we took it very serious) we came to the conclusion not to combine these sections. The main reason is that in the discussion section we go beyond the method comparison, which is central subject of the results section. Combining results and discussion chapter would not give appropriate weight, for the example to the comparison with other field experiments.

3.1 Artifact particles

Information on the amount of artifact particles from the total particles that were sampled could tell the reader how this sampling methods had on the artifacts amounts
We do not understand this comment, as this information is given in Figs. 3 and 7 (formerly 3 and 5).

It will be clearer to mention what each CVI or chamber was built from and to state that some of the artifact could be due to that, the way it is written now is not clear.
We have added a comment on the materials used for manufacturing or as calibration aerosol. We have also rewritten the Pb-artifact discussion regarding the Ice-CVI.

3.1.2 Potential INP/IPR sampling artifacts

Can the author explain how or by with methods these particles were sampled, is it by all methods or just by some?
We do not understand this comment, as this information is given in Fig. 7 (formerly 5).

3.2 Composition of INP/IPR at the Jungfraujoch in winter

The author should mention in the text how many particles were sampled by each methods, and not just show it in figure 5
We give a total number in the text, but we think that this information is more appropriate in the figures.

Why are the artifact part of the figure if the authors say that artifact are not an IN source and will not be shown (page 23037 line 21)
Droplets, sulfate and sea-salt are classified as POTENTIAL sampling artifacts. We came to the conclusion that the current knowledge is not sufficient for unambiguous identification of sampling artifacts. The discussion in the literature is still controversial. We show these groups in the figures and tables in order to allow the reader to come to his/her own conclusion on the basis of the full data set.

The comparison between the instruments (fig 7) is good because finally there are two samples that were taken at the same time, however perhaps some of the difference appear in Fig 7 are due to the different sampling methods, I think that the author should mention it in the text.
We agree and have added an according sentence.

3.3 Size distribution of INP/IPR components
How was the size determined?
We give the sizes as equivalent projected area diameter determined in electron microscopy, given in section 2.2.1

Are the artifact taken into account in this calculations?
Artifacts are excluded from this discussion. An according sentence was added.

More information and comparison with the literature will improve this section
The size distribution can't be compared between the instruments due to different inlet transmission efficiencies and – in part – yet uncharacterized instrument transmission efficiencies. We are not aware of size distribution measurements for individual INP/IPR compounds.

3.4 Composition of total aerosol

Does the author think that such small sampling time represent the total aerosol type in that area for the entire period?
I think it would have been better to start and end every experiment by looking on the total aerosol composition and not just sample it for such small period.
Why only showing on Fig 9 the sample from the 16 and not all the times that were sampled?
We agree that this short sampling time is not representative for the campaign. Therefore, we removed section 3.4 and modified section 4.2 considerably.

4 Discussion
4.1 Which particle classes can be regarded as INP/IPR?

Page 23044 line 13
There is no need to writer Al- but only write Al unless the author wanted to say Al-rich

This is exactly what we want to say (Al-rich). Thus, no change is required here.

In addition, it is not clear if these are particle who only had these elements or perhaps they were parts of particles with other elements for example mineral dust particles contained many elements as Si, Al, Mg, K, Ca and Fe.
Silicates would have been classified separately as such (section before). Therefore, these particles are metal oxides.

Page 23045 lines 12-13
I think this sentence is unnecessary in this part.
We think that this sentence is appropriate at this part of the manuscript.

4.2 Relative ice nucleation ability of the different particle classes

I am not sure if it is correct to do this comparison since the total aerosol composition represent small period of time form this experiments.
This discussion was removed from the manuscript.

Why should the author compare with dust particles as Feldspar when there is no dust event that were sampled and the chances for such particles to reach the station at winter is very low.
Mineral dust particles are still the major fraction of INP/IPR on Jungfraujoch in winter. Thus, a discussion of the silicate composition is meaningful.

I am not sure if what written in page 23047 is relevant under this section, perhaps a better place will be under section 4.1
The comment does not apply anymore, as this section was restructures considerably.

4.3 Comparison between FINCH+IN-PCVI, Ice-CVI and ISI
I think that this section is very important and it should be in an earlier part of the paper.
We believe that the reader will recognize the importance of this chapter independent of its location in the manuscript. In addition, we have added a more extended discussion.

4.4 Comparison with other field experiments

The author have already compared some of these result in previous sections, I will recommend to combine all of it to one section
Combined as suggested into chapter 4.1

Page 23049 lines 12-16
I do not agree with combining all the data from the different methods due to their limitation of sampling at different times, but in any case, such information should have been included in the result section
We are aware of the limitation of the dataset. However, we have shown in Fig. 4 that (with exception of organics/carbonaceous) the relative abundances of the INP/IPR classes between the techniques do not differ extremely. Thus, we find it justified to combine all data to increase the statistical significance.
We kept these sentences as short introduction to the discussion, which would be otherwise difficult to understand

Page 23049 lines 17-16
Perhaps a figure or a table with these comparisons will be clearer
We agree and have compiled a short table (Table 4)

Page 23050 lines 12-19
The author does not say much about the different days that were sampled therefore I am not sure if this paragraph is appropriate.

We have removed this paragraph.

Page 23050 lines 20-28
The author already discuss about this point at an earlier stage I will recommend combining the two.
Section was combined with a previous one.

4.5 Comparison between scanning electron microscopy analysis and laser ablation mass spectrometry

I think this section should be in an early part of the paper, perhaps a better place will be to combine the result and the discussion to one part.
Results and discussion are intentionally not combined (see above comment to chapter 3).
We agree that this section does not fit perfectly into the general manuscript outline, as it discussion a special technical point. However, we feel that it fits better into the discussion section as into the results section.

In addition, I do not think it is a good idea to compare something that was not measure in parallel because there could be many elements that could affect the comparison.
From the flow restriction it was not possible to have both techniques working in parallel. As the measurements were intermittent, there is no reason to assume a systematic difference based on this non-parallelity.

5 Summary and conclusions

I would recommend rewriting this part based on the artifact which should be taken out from all calculation.
In addition, the author should say that although three methods were used they were not used in parallel.
We believe that the information on the artifacts is an essential part of the manuscript. Thus, we would like to keep it into the summary.
We explicitly mention the non-parallel sampling now.

Table 1

The freezing mechanism should not be included for IN-CVI or ISI since they both measure ice crystal after they nucleate
Nevertheless, they were subject to a freezing mechanism (occurring naturally in the atmosphere).

Figure 1
This figure is not clear, what are the gray or the colored marked represent.
Are the values of aerosol concentration, temperature, wind are they daily average, it will be good to mention it here and in the text as well.
We have added an explanation in the caption. Continuous curves are shown for concentration, temperature, wind parameters.

Figure 3

This figure is not so clear and it seem that the effect of the artifact is too big, a different way of presenting this information might be better.
Box plots are a standard way of displaying point and interval estimates of measurement data. The artifacts are shown as they are.

Figure 6-7
I would suggest to delete the artifact from these figures
We have discussed above why we want to keep the potential sampling artifacts in the figure (note that Fig. 7 was removed and the numbering has changed).

Figure 9

This figure is very important and show the reader about the type of particle found in that area, however the author should mention that these particles were sampled on vary narrow time period, only on the February the 16 and for short period of time.
Fig. 9 was removed as request from the other reviewers. Reason was the extremely short sampling interval and the subsequent potential statistical uncertainty.

Figure 10

The author should mention at lease in the text which one is the current project and with one is the other for example “inuit- current project”
Changed as requested.

References used in reply

Boulter, J. E., Cziczo, D. J., Middlebrook, A. M., Thomson, D. S., and Murphy, D. M.: Design and Performance of a Pumped Counterflow Virtual Impactor, Aerosol Sci. Technol., 40, 969-976, 10.1080/02786820600840984, 2006.

Mertes, S., Verheggen, B., Walter, S., Connolly, P., Ebert, M., Schneider, J., Bower, K. N., Cozic, J., Weinbruch, S., Baltensperger, U., and Weingartner, E.: Counterflow Virtual Impactor Based Collection of Small Ice Particles in Mixed-Phase Clouds for the Physico-Chemical Characterization of Tropospheric Ice Nuclei: Sampler Description and First Case Study, Aerosol Sci. Technol., 41, 848-864, 10.1080/02786820701501881, 2007.

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