We gratefully acknowledge the comments from the anonymous referee 1 and include them in the manuscript. We believe that the changes help to improve the quality of the paper, especially in terms of clarifying our particle groups making the results better readable for a broader community.

Reviewer comments and questions are printed in this font type.

Our replies are written in Times New Roman.

Changes in the manuscript text is written in italic, sometimes underlining particular word changes.

Interactive comment on "Composition of ice particle residuals in mixed phase clouds at Jungfraujoch (Switzerland): Enrichment and depletion of particle groups relative to total aerosol" by Stine Eriksen Hammer et al. Anonymous Referee #1 Received and published: 17 June 2018

Hammer et al. consider the properties of particles within ice crystals, aka ice particle residuals (IPRs), at the Jungfraujoch, a high altitude mountain site in Europe. For this work they use a published ice selective inlet aka 'Ice-CVI', and compare this to the total aerosol via a simultaneous heated inlet sample to provide all, aka 'total', particles. In total, they consider ~4k ICRs over 7 days and in the ~-10 to -20 deg C range (i.e., mixed phase clouds). These particles were deposited on substrates for off-line analysis with electron microscopy (EM). There are several papers over the last decade, many by the groups represented here describing such measurements at the Jungfraujoch so this is not altogether novel. It does represent important data in an area of atmospheric science that is currently in need of more information. My major concern, however, is that the way ice residuals are described seems very simplistic and not of use to the broader atmospheric science community. For example, what is a C-rich particle? How would someone using AMS or a modeler compare this to their understanding of the atmosphere? Since studies of this type have been published, again, many by this group, I believe they here have a responsibility to make the results more comparable, and therefore more useful, to the broader community. I therefore suggest the authors consider the following suggestions and, pending another review, that this paper could be published in ACP.

1. The literature seems to predominantly use the term 'ice residual' (IR) as opposed to IPR. Is there a reason the authors have suggested a new term? Is it different than IR? If so a comprehensive description – and difference from IR - needs to be made. As it reads it seems they are the same so, if there is no difference, could you please remain consistent with the literature term IR so as not to confuse the reader.
    - We now use the term ice residual (IR) in the paper.

2. Page 3 Line 8: The authors seem to suggest water vapour homogeneously nucleates ice at -38 deg C and 140% RH; but that is not correct. One must first have droplets. Please remove 'vapour or' from this sentence.
    - Removed as suggested

3. Page 5 Line 5 & 6: From the intro, the authors ‘assume’ the IPRs are INPs, not ‘consider’. This is a very important distinction. The authors discuss artifacts; thus they themselves show they cannot make the direct association implied by ‘consider’.
    - We changed the sentence according to reviewer II: *“The campaign lasted for five weeks with the aim to investigate IRs from mixed phase clouds which may reflect* <u>*the initial INPs active*</u> *in the cloud.”*

4. Table 1: Most aerosol composition measurements show the presence of sulfate and SOA particles as a dominant, if not the dominant, aerosol, at least by number. Is the ‘complex secondary particles’ this class? This seems to be implied later in the Discussion but is never clearly stated. If so, even if it is an assumption, this needs to be stated for clarity and comparability to the literature. (note: this seems to be suggested on Page 10 but warrants more than 1 line of text).
    - We agree. A new column is added to table 1 to make it easier to understand our classification and particle groups.

Table 1: Classification criteria and possible sources/ explanation for particle groups for both, total aerosol and ice particle residuals.

Group	**Major elements**	**Morphology/ beam stability**	***Source/ particle explanation***
Soot	C	Chain-like or more compact agglomerates of primary particles	*Combustion, black carbon*
C-rich	C	No soot morphology	*Organic aerosol, biomass burning***, *biological***
Complex secondary particles	No X-ray spectra or S-peak	Most particles evaporating, some relatively stable	*Sulphur rich secondary organic aerosol, might also contain a substantial fraction of nitrates and other organics*
Aged – sea salt	Na, S (sometimes small amount of Cl and Mg)	Relatively stable	*Marine aerosol, sea spray, might contain organics*
Mixed –sea salt	Na, S (sometimes small amount of Cl and Mg) + mineral composition		*Marine aerosol mixed with mineral particles. Might contain organics.*
Ca-rich	Ca, C, O		*Mineral particles, calcium carbonates e.g. calcite*
Ca-sulphate	Ca, S,O		*Mineral particles, e.g. gypsum and anhydrite*
Silica	Si, O		*Mineral particles, e.g. quartz*
Alumosilicate	Al, Si, O		*Mineral particles, e.g. kaolinite*
Fe- alumosilicate	Al, Si, Fe, O		*Mineral particles, e.g. almandine*
Other-alumosilicates	Variable amounts of Na, K, Ca, Si, Al, O, Ti and Fe		*Mineral particles, e.g. feldspars, illite and smectite (montmorillonite)*
Metal/ metal oxides	Fe, O or Ti, O or Fe, Cr, Mn	Fly ash was detected as spherical particles	*Mineral particles like hematite, magnetite and rutile, or steel particles (alloys)*
Pb-rich	Pb, or Pb, Cl	Single particle or inclusions within particle	*Helicopters and small aircrafts, previously reported at Jungfraujoch*
Other	Particles which do not meet the classification criteria above		
Alumina*	Al, O		*Artefact, Ice-CVI*
Ni-rich*	Ni		*Artefact, Ice-CVI*
Cu-rich*	Cu		*Artefact, particle substrate*
Pure salt*	Na, Cl		*Artefact, hypothesised from secondary ice processes e.g. crystal break-up, marine origin***

**Most likely contamination. **Uncertain origin because the chemical characterisation and/or morphology was not the typical for this particle group.*

5. Similarly, biomass burning particles are often noted as being of abundance just below sulfates and SOA. Is this the C-rich class here? If so please state this. I stress here: while the authors are reporting what they observe with their EM technique, they also need to make it clear their classes relate to common particles types if they wish to publish in a journal such as ACP. Please make these comparisons.
   - See new table 1 (same as comment 4)

6. Similarly, what are Ca-rich particles? The discussion seems to suggest they are mineral dust? How are these associated with any common aerosol type?
   - See new table 1 (same as comment 4)

7. The authors discuss observations of biological IRs in the introduction. These don't appear on Table 1 or Figures 4 or 5. Are they not observed or is there an instrumental reason they can't be detected? Are they a subset of the C-rich category? This needs to be stated clearly in the paper, especially in Figure 5, as it goes directly to comparison to the literature on ice residual composition and in the location where Table 1 is described.
   - Primary biological particles are normally classified based on chemistry (C, O and tracer elements i.e., N, P, K) and morphology. We do not see the normal identification specifics in the C-rich group, but still, we cannot exclude that particles in this group might be of biological origin. We therefore added biological particles as a possible explanation of the C-rich group in the new table 1.

8. Page 7, Line 6. Please remove 'Unfortunately, only'. Data are what was collected.
   - Removed as suggested.

9. Sample S-2b looks rather like a local combustion event. This seems to be implied later in the discussion. If it is please state at the location of the Figure for clarity to the reader.
   - We state now that this is a combustion event in the figure caption.
     We changed figure caption to: *"Relative number abundance of the different particle groups within total aerosol samples. Sample S-2b shows a combustion event with air mass history form the Po Valley, and sample S-5b is influenced by an analytical artefact from particle loss of volatile particles."*

10. Given this, could you please present figure S2 at this location – move it into the main paper from the supplement - for the samples to give the origin some context? It is mentioned in the text in the discussion but really needs to be given at this location.
    - We moved Figure S2 to the main paper as suggested.

11. Sample S-5b looks rather like a mixed mineral and perhaps combustion event. Is this so? It again does not look like a clean troposphere. Is this so? If it is please state at the location of the Figure for clarity to the reader. Please also see last comment re: back-trajectories.
    - This sample cannot be explained by back-trajectories, but an analytical artefact from particle loss. We changed the figure caption to (same as comment 9): *Relative number abundance of the different particle groups within total aerosol samples. <u>Sample S-2b shows a combustion event with air mass history form the Po Valley, and sample S-5b is influenced by an analytical artefact from particle loss of volatile particles.</u>*

12. I am confused with Section 4.1, 'Methodological Problems'. I believe this all needs to do in the Experimental section (2), not held until after the data is presented. First, Figure S5 seems to indicate that most of the collected particles are artefact. Is this the case? If so please state the percentage in the Artefact section. Second the reader goes through the results but only after they are presented learns there are issues with the inlet and perhaps the EM data which appear to outnumber the real data by several factors. This is not a logical order. This all needs to be clearly stated and placed in Section 2, not held to Section 4.
    - We moved this section to methodology and added the following sentences to the start of the paragraph to make the contribution of artefacts clearer: "*<u>The IR samples are heavily loaded with artefacts (40-78% of the particles – alumina, Ni-rich and pure salt) easily characterised and removed in further analysis. The Cu-rich particles are a part of the substrates and can in principle be found in both IR samples and total aerosol samples.</u>*"