

Interactive comment on “Composition of ice particle residuals in mixed phase clouds at Jungfrauoch (Switzerland): Enrichment and depletion of particle groups relative to total aerosol” by Stine Eriksen Hammer et al.

Anonymous Referee #2

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General Comments

This paper discusses the most recent evaluation of the composition of ice crystal residues as measured by an ice-selective counterflow virtual impactor inlet (CVI) at the Jungfrauoch experiment station. The broader community is surely interested in this work, in general and to understand if all measurement artifacts have been solved to the point that a consistent and informative data set can be collected regarding the source of ice nucleating particles and other information on microphysical processes in winter clouds at this site. The short answer based on this paper is that, while some

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issues have been resolved, this remains a work in progress. While a potentially useful paper, this one could have used stronger editing prior to submission. The overall organization is generally good, but the discussion bounces around such that the various facts are not subjected to a structured discussion. As for specific critical revisions needed, a vital one is to bring forward the fact that a major measurement artefact remains unresolved, that of AI in many particles. I saw no clear discussion of the potential source for this contamination. Otherwise, I appreciated the attempt to categorize electron microscopy microprobe data, although I stumbled on the categorization of “sea salt”, by which was meant an assortment of possibilities. This pointed to a general need to be more descriptive about the categorizations and how artefacts were defined in comparison to specific sources. With revision, this paper will become acceptable for publication, although it remains another step in the direction of attempts to extract information on ice nucleation processes through inspection of ice particles residuals.

Specific questions/comments for potentially addressing are listed below.

Specific Comments

Abstract

A few details here should be clarified. 1) I have no idea what a multi MINI impactor is, but it certainly does not need listing in the abstract. Just the basic technique should be stated. 2) It is not clear how a dilution setup allows for matching a total aerosol sample to the Ice-CVI sample. Can this be explained in plain language? Perhaps, “A dilution system was used to collect total particles at a similar rate to Ice-CVI particle collections”, although I do not know how that is managed, and it seems that it failed about 50%) State temperatures as “local” or “site”. These are not necessarily the cloud activation temperatures. 4) “Approximately 3000 total aerosol particles from five days in clouds were also analysed.” Is this referring to IPRs or to interstitial particles, or to all non-ice particles?

Introduction

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Page 3, line 8 – Spontaneous freezing of supersaturated vapour? In the Earth’s atmosphere? I have not seen such a statement in the literature in some time. Remove unless you can support the feasibility of any process other than homogeneous freezing, not “homogeneous ice nucleation”.

Page 3, lines 12 to 15 – Add “hypothesized” to modes. The last sentence is repetitive with regard to mixed-phase temperatures where heterogeneous nucleation is the source of ice initiation.

Page 3, paragraph 2 – The need for this thesis-type material is questionable. I suggest to revise and reduce or even omit most of this and get straight to the point, which seems to be that information on the relevance and importance of different ice nucleating particle types has come from laboratory measurement, and these emphasize the importance of mineral dust particles except at very modest cloud supercooling. What this paragraph does not seem to mention are specific studies where activated ice nucleating particles have been studied for composition, not simply tested as single collected types in the laboratory.

Page 4, line 4: Ogren et al. (1985) is not in the reference list. There is a substantial amount of literature since in which airborne CVIs have focused on ice clouds, and Cziczo, Froyd and colleagues have emphasized some other constraints on ice cloud sampling of IPRs (e.g., a focus on small ice, as done also in this study – line 6 statement, although no indication is given as to why fresh ice is needed). While an aside of sorts, the utility of sampling in mixed-phase clouds for ice nucleation studies using a non-ice-CVI is not reflected here, since the focus is on IPRs. The fact that one gets both IPRs and liquid cloud residuals when sampling in mixed-phase clouds is not necessarily a detriment, and this makes it suitable for ice nucleation measurements and subsequent collection of the activated INPs for compositional analyses (already mentioned in the preceding point). This is alluded to later in mentioning use of a FINCH for a similar purpose.

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Page 4, lines 8-10: But then this introduction is followed with these lines, which I could not understand - “Knowledge on particle groups acting as ice nuclei in mixed phase clouds is contradictory. IPRs are the residuals ice crystals formed on real INPs after they have been activated in the environment and the measured ice nucleation efficiency of these IPRs is then considered to be the same as for INPs.” I expected the first sentence to be immediately supported. Is this a new paragraph? It is not a good one for sure. Rewrite it to be concise, and get to that point. Is the contradiction mentioned referring only to studies done at Jungfraujoch, or what other studies? Will this study seek to resolve contradictions? What is a “real” INP? I suggest to remove this terminology. I think I understand the last part to mean that the composition of IPRs is considered to be those of INPs that were active at the local temperature of observation.

Experimental

Page 5, lines 5-6: Please explain or omit the statement “. . .original true INPs.” You will simply assume that IPRs represent INPs active at the cloud temperature of observation, correct? Are you trying to infer that other methods will not detect INPs? I think you are trying to say that the residuals reflect INPs that were activated in the cloud. But are you saying that every ice crystal contains an INP? I do not think that can be supported, if for example secondary ice formation processes were active.

Page 5, line 7: typo, “were” not “where”

Page 5, lines 9-10: Can you explain the need for dilution of the total aerosol sample a little better? i.e., there would be too many particles if collected for the entire time period?

Page 6, lines 18-19: Why are pure salt, alumina, Cu-rich and Ni-rich particles considered as contamination? It would be nice to consolidate this information in one place. In the end, no source is identified or even suggested for the alumina particles assumed as contamination, and I find the fresh salt explanation to be questionable. I gather later that the AI is assumed to come from ice crystals striking the walls of the CVI, despite

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coating them with Ni, but it is almost incomprehensible how this contamination exceeds that found in any previous study (page 15).

Page 7, line 5: This seems to require a statement that the cloud sampling temperatures were considered as appropriate as the ice crystal formation temperature. Could satellite data say anything about coldest cloud top temperatures at these times? Or do you also assume that the limited ice crystal size range sampled restricts this condition?

Results

Page 12: A general comment - it might be nice to show both a representative particle image and elemental spectra for each of the different particle composition categories. This could go in the supplement in addition to the single example given.

Page 12: General comment 2 – It is only if one goes immediately to look at Fig. S5 at this point that one realizes that the vast majority of particles were categorized as artifacts. Surely this needs to be mentioned upfront. Greater than 50

Page 12, line 7: I wonder if in the basic analysis performed if a mineral particle could be distinguished as being from desert or from other soils? I assume this would remain unresolved, since the soil particle could have multiple potential actual ice nucleation sources, including trace organics.

Page 12, line 10: When the authors say “sea salt”, what is meant? Is it only NaCl, or does this refer to aerosols of sea spray origin, with a more complex mixing state? There are only two categories, aged and (aged-) mixed, and by mixed are also included mixtures with other aerosols such as minerals. This makes attribution specifically to “sea salt” nebulous, and yet statements are subsequently made in the results about the ice activity of “sea salt”. This is problematic.

Page 12, line 14: I assume that aluminum oxides are omitted from the metal oxide category because of the alumina contamination that is not really discussed?

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Page 15, line 1: Sea salt is similarly ice active as aluminosilicates? Is it the sea salt, the organic content of marine aerosols, or the particles they are mixed with? Hence my earlier question. Perhaps these should be stated to be sea salt-containing particles, and a statement is needed about how this does not identify the “salt” as the ice nucleating component.

Page 15, lines 15-17: A focus on fine particles is mentioned as an explanation for the occurrence of more alumina in this study, apparently from crystals etching this from the CVI walls (nowhere stated clearly). This is the first mention of any different focus in this study. What is meant by a focus on fine particles? Why would there be so much less Ni and so much more Al? Was the coating quickly destroyed? Ineffective? Also on line 15, “but” is misspelled.

Page 15, lines 24-26: If pure or “fresh” salt is an artefactual reflection of secondary ice formation contributions, how is this reliably distinguished from sea spray aerosols? Would aging of sea salt always occur for marine particles reaching the site? Relatively unaged marine aerosols are found at other remote locations.

Page 16, line 12: By “concentration of the total inlet” do you mean the accumulated particle number concentrations sampled from the total particle inlet (after dilution)?

Page 17, lines 8-10: This might well be the third mention of the sample that was exposed to high vacuum in the electron microscope for too long. Please edit.

Page 17, line 23: “section 4.3”

Page 18, lines 14-15: This statement regarding the association of sampling temperatures with actual ice nucleation temperatures should preface measurements in the discussion of methods.

Page 18, line 21: The reason that the authors believe that the current results are correct in regard to the lack of contribution of complex secondary particles and soot as IPRs (and thus INPs), and why the previous studies erred, should be summarized.

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Page 18, lines 24-25: This statement regarding the composition of the secondary particle category also belongs in the methods material, which was painfully short in describing the different categories and their justification.

Page 18, line 28: Are the studies herein and those summarized in Knopf et al. (2018) for cloud activation temperatures in the same range?

Page 20, first paragraph discussion of “sea salt”: This discussion was odd. I could take argument with the authors about the supposedly “controversial” nature of ice nucleation involving marine aerosols overall, but let me focus on lines 6-8. Unless the authors wish to reject clear evidence in the papers mentioned or in papers published since involving specific sampling of sea spray particles (none referenced here), the ice activity is clearly if not definitively associated with contained organics in many instances. It is not really a hypothesis that the salt itself is not the INP, so it is good that the authors will not “exclude” this fact.

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