Author comment in response to the comments provided by Referee #1

We thank Referee #1 for their effort in reading and commenting our manuscript and giving helpful feedback. In the following, we repeat the **referee's comments in bold typeface**, and give point-by-point answers in normal typeface; extracts from the *original manuscript are presented in red italic*, and from the *revised manuscript in blue italic*. Line numbers are referring to the updated manuscript version.

General Comments:

Due to the low and large variability of the concentrations in the atmosphere, our understanding of the chemical composition and sources of ice nucleating particles is still quite limited. This article investigated the potential chemical characteristics and sources of ice nucleating particles (INPs) at the high-altitude research station Jungfraujoch (JFJ) in winter 2017 based on measurements from a suite of sophisticated instruments and the FLEXPART source emission model calculations. Their results show that mineral dust and particles of marine origin are the most important contributors to ice nucleation. These are supported by analysis of airmass back trajectories and the chemical composition of ice particle residuals. The results are very important and add some new insights into the understanding of possible chemical composition and sources of INPs. The article is generally well written, concise and should be publishable if the following specific comments and suggestions can be considered in revision.

We appreciate these general remarks regarding our manuscript and believe that we have addressed the comments and suggestions provided by Referee #1 in the revised manuscript as indicated below.

The authors used the INPs concentration at -31 °C to represent ice formation in mixed-phase clouds. I think this temperature is too low for typical mixed-phase clouds. Actually, the authors have also indicated in Line 367, that temperature ranged from -5 °C to -18 °C to be a relevant range in which MPCs can form.

The reviewer addresses two points here. First, mixed-phase clouds can form within the temperature range from 0 °C to approximately -38 °C (e.g., Korolev et al., 2017). Thus, our chosen nucleation temperature of -31 °C albeit toward the lower end, is still very relevant for mixed-phase cloud formation, but perhaps for a higher altitude than where we measured at the JFJ.

Second, indeed, the mixed-phase clouds present at Jungfraujoch are warmer than -31 °C. Field INP concentration measurements are typically performed at temperatures colder than the ambient conditions, because they investigate the *potential* of the aerosol population to nucleate ice if the air mass containing those particles would experience this temperature and supersaturated conditions. The great advantage of our sampling location and sampling time in the winter months is that the measurements were taken in the free troposphere, which is not impacted by local emissions but of long-range air masses, and thus is better representative of

aerosol particles of a global burden (Lacher et al., 2018a). Thus we believe that our INP concentration measurements at -31 °C are relevant for cloud microphysical properties in the free troposphere and not only locally at the JFJ. When measuring at the ambient temperature or warmer, it is further possible that the INPs are depleted due to a pre-activation during the transport from INP source regions to the measurement site (Conen et al., 2015). At a measurement site as Jungfraujoch, which is far away from INP source regions, it is especially likely that this process takes place.

Last but not least, the ambient INP concentrations at Jungfraujoch are typically very low, and by choosing a nucleation temperature as low as -31 °C, we ensure that most of our measurements are still above the detection limit of HINC (Lacher et al., 2017).

We clarify this aspect by mentioning in lines 140 - 141:

The RH_w of 103% ensures that the entire aerosol layer which experiences a varying RH between 101 – 103% is above water saturation such that the particles can activate into droplets in the given residence time of HINC.

A reference by Jiang et al. (2016) should be added in Line 70, it provided another evidence that the INP concentration increased from about 10 per liter to more than a hundred per liter.

Thank you for this suggestion. We include now the study by Jiang et al. (2016) to reference the importance of mineral dust for ice nucleation (see lines 63 - 64 in revised manuscript version).

In Line 95, after Murphy et al., I suggest to add Chen et al. (2021), who used a single particle mass spectrometer together with a wide-range aerosol particle spectrometer to determine the possible chemical components and sources of INPs.

Thank you for pointing at this interesting study. We include Chen et al. (2021) now in the statement in line 85 of the revised manuscript.

Line 149: "-31 °C (±0.4 °C; [INP]-31) and at a relative humidity with respect to water of 103% (±2%), representing condensation/immersion freezing, relevant for the formation of MPCs": As has pointed out above, -31°C might be lower than typical temperatures in mixed-phase clouds, and RH of 103% is possibly much higher than general winter orographic clouds.

Please see our comment above regarding the temperature range for mixed-phase clouds. Indeed, a relative humidity with respect to water (RH_w) of 103% is higher than ambient supersaturation conditions in mixed-phase clouds. There are three reasons for this. First, HINC has an uncertainty in RH_w of 2%, and by setting a RH_w of 103% we ensure that the entire aerosol layer in the chamber is exposed to conditions of or above water saturation. Some of this uncertainty arises from the fact that the part of the aerosol layer closer to the warm wall will experience RH_w below 103%. By maintaining the nominal RH_w at 103%, we ensure that even this part of the aerosol layer is still at or above RH_w = 100%. Second, the residence time of particles in continuous flow diffusion chambers is on the order of a few seconds, which limits

the activation and growth time for cloud droplets and ice crystals; by creating such a high supersaturation, we are counteracting this time effect. Moreover, a high supersaturation also favors the activation of all CCN (and INPs) into cloud droplets, even in the case of hydrophobic particles and high CCN number concentrations (see also the discussion in Rogers et al. (2001)).

Lines 174-175: "which requires the surface area distribution concentration to be calculated from the number size distribution.": How the surface areas of aerosol particles were calculated? The assumption of aerosol particle shape could significantly impact on the calculated surface area.

The reviewer is right that the aerosol particle shape can impact the calculated aerosol particle surface area. For our calculation we assumed a spherical particle shape, as the true particle shape was not measured. We add a statement that our assumption might lead to increased uncertainties in lines 164 - 167:

Measurements from the GAW SMPS and OPS are used to calculate the available surface area of the aerosol population by assuming a spherical particle shape and that the refractive index of the ambient aerosol population is represented by the calibrated value of the OPS. We acknowledge that this can lead to higher uncertainties, which are not quantified here.

Lines 211-212: "The overall detection efficiency of the LAAPTOF is between 0.01 (±0.01) % to 4.2 (±2.4) %": Is it so low?

Yes, that is correct. We acknowledge that due to this low detection efficiency in LAAPTOF the large majority of the particles is not analyzed; however, we still consider that the measured particles are representative for the characteristics of the overall aerosol properties and for their change over time, which is used in this study to investigate parallel changes in the INP population.

During the review process we realized that the values stated from ALABAMA and LAAPTOF were not referring to the same detection efficiencies; while the values given for LAAPTOF are the *overall detection efficiencies*, which we define as detection efficiency combined with the hit rate, the ALABAMA stated only the detection efficiency. Moreover, we added a graph on the size-dependent detection efficiencies from the two single particle mass spectrometers and HINC, with and without the PFPC. As our aim is to investigate size dependence of detection efficiency in relative terms, we normalized those measurements to the maximum value in each instrument.

We updated the manuscript accordingly in lines 207 - 225, and include Fig. S1 in the appendix:

The overall detection efficiency (combining the detection efficiency and the hit rate) of the LAAPTOF is between 0.01 (±0.01) % to 4.2 (±2.4) %, in the size range of 0.2 to 2 µm based on polystyrene latex particles (PSL). The highest overall detection efficiency is for 1µm and lowest for 2 µm (Shen et al., 2018). Note that such efficiency is also particle type dependent (Shen et al., 2018, 2019). More details on the LAAPTOF can be found in Gemayel et al. (2016), Reitz et al. (2016), and Shen et al. (2018, 2019). Details on the ALABAMA have been presented in Brands et al. (2011), Roth et al. (2016), Schmidt et al. (2017), and Clemen et al. (2020). The detection efficiency of the ALABAMA during this campaign was between 40% and

60% in the size range of 0.3 to 1.0 µm based on PSL particles. Up to a particle size of about 1.3 µm, the detection efficiency of the ALABAMA decreased to less than 30% and is estimated to be about 5 (±5) % for 2 µm. At the same time, the hit rate during those tests using PSL particles was lower, such that the overall detection efficiency for the ALABAMA was only between 1% and 16% in the size range from 0.3 to 1 µm. As those values are based on measurements using PSL particles, they can vary considerably during field applications; e.g., the ALABAMA hit rates were significantly higher than those of the PSL test measurements (which is attributed to particle charge effects during the nebulization of the PSL particles). In the light of our research objectives, focusing on the general trend of the aerosol particle composition, we therefore provide an overview of the size dependent overall detection efficiency from the LAAPTOF and the ALABAMA normalized to the maximum value measured, together with the normalized transmission efficiency from HINC (Fig. S1). From those normalized values it is visible that both SPMSs detect aerosol particles in the same size range, with a maximum between 0.5 and 1 µm, and therefore yield comparable information on the particle composition in this size range. HINC measures particles below 2 µm with a high efficiency which can have an impact on the comparison between the INP measurements from HINC and the results obtained from the ALABAMA and the LAAPTOF.

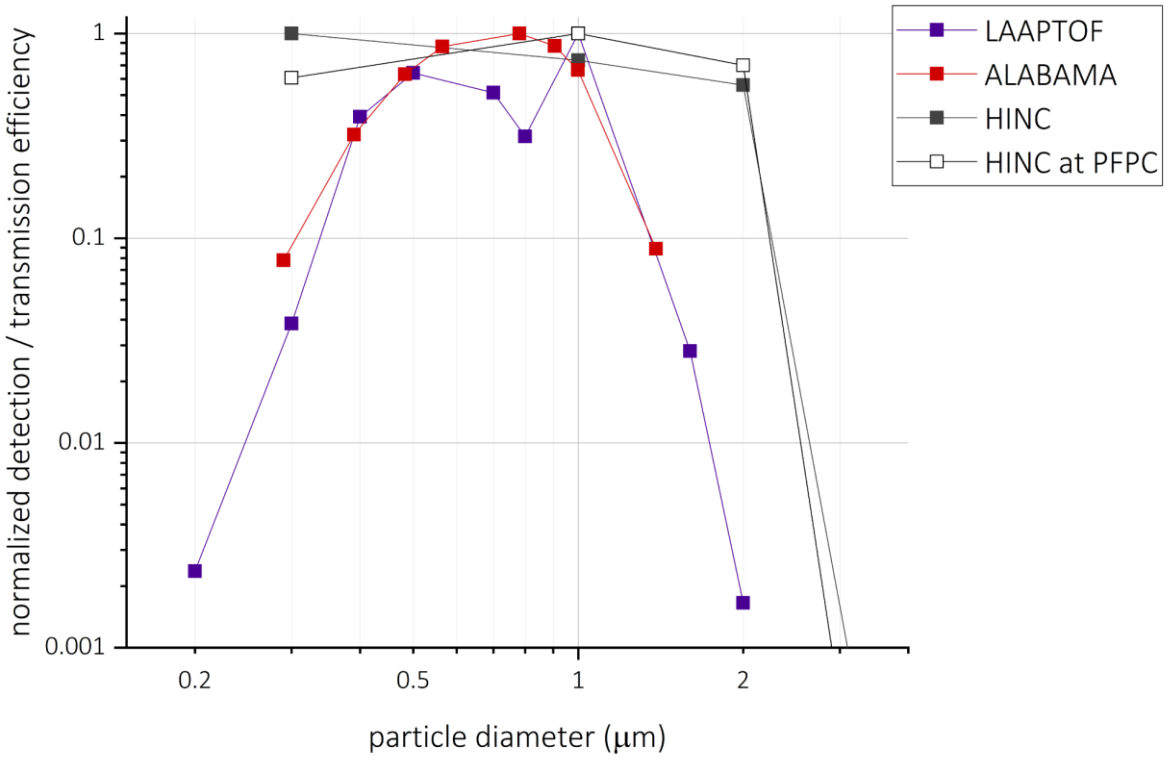

Figure S1: Normalized size-dependent detection efficiencies for aerosol particles in ALABAMA and LAAPTOF), and normalized transmission efficiency for HINC, and for HINC at the PFPC (based on calculations using the measured transmission efficiency with size-dependent enrichment from the PFPC (Gute et al., 2019)); the measurements were normalized to the maximum detection efficiencies of 4.3% (LAAPTOF), 15.6% (ALABAMA), 100% (HINC), and 1480% (HINC at PFPC).

Line 366-367: "The ambient temperature ranged from -5 °C to -18 °C (Fig. S3, panel b), which is a relevant range in which MPCs can form": If the ambient temperature ranged from -5°C to -18°C, why choose -31°C as the nucleation temperature for INP analysis?

Please see our answer to the first comment. In addition, we would expect that any aerosol particles active as INP in the range -5 to -18 °C will also be active at -31 °C thus we should capture these particles in our reported INP concentrations.

Line 396-397: "particle types "secondary inorganics", "K, organic sulfate", and "more mixed/aged" measured by the LAAPTOF…": "secondary inorganics" and "organic sulfate" were not shown in the figure. Please keep consistent with the chemicals shown in the figure.

Secondary inorganics are referring to NH_4, NO_3, SO_4; we include "secondary inorganics" now in the legend of Fig. 2, panel c. Indeed, it should read "K, organics, sulfate", thank you for pointing this out. We updated the sentence in line 391:

...particle types "secondary inorganics (NH_4, NO_3, SO_4)", „K, organics, sulfate"...

Please note that Fig. 2 uses particle type clusters based on fuzzy c-means, in order to give a general overview of the time series of particle composition during the campaign. Figure 5, on the other hand, assigns the best correlating marker ions to a particle type.

Lines 477-479: "Such compounds are typically water soluble and are therefore not expected to contribute to immersion freezing INPs at conditions assessed in HINC": Negative correlations means the present of those chemicals would inhibit ice formation.

Indeed, this finding can be interpreted that those chemicals inhibit ice formation, which is, however, unlikely as any potential coating does not impact the ice nucleation ability in the immersion freezing mode (e.g., Kulkarni et al., 2014; Kanji et al., 2019) but can inhibit ice nucleation in the deposition or pore condensation freezing mode. Thus, we did not interpret this finding to be an effect of ice nucleation inhibition, but of an air mass containing a higher fraction of such water soluble aerosol particles which are all together not ice-active. We add this discussion to the manuscript in lines 471 - 472:

Moreover, we do not expect that those components reduced the ice nucleation ability of the ambient aerosols by a coating effect in the immersion freezing regime (e.g., Kulkarni et al., 2014; Kanji et al., 2019).

Lines 576-578: "Thus, the timeseries of the ALABAMA and AMS sulfate measurements are anticorrelated to the [INP]-31 time series which is not surprising given soluble aerosol particles are not expected to contribute to heterogeneous ice nucleation above -38 °C": As has been pointed out above, negative correlation means that the present of sulfate would suppress those particles to act as INPs.

Please see our answer to the comment above.

Line 637: mineral dust particles can also be incorporated into cloud particles when coated dust particles serves as CCN or collected by cloud particles, not necessarily as INPs, especially for cloud formed at warmer temperatures (>-17°C as indicated in the text).

We agree that the IPR analysis can be impacted by mineral dust particles when sampling secondary formed ice crystals, which originally were cloud droplets and might contain mineral dust. However, we see a strong mineral dust particle signal sampling different cloud events, and do not observe this highly variable fraction as we did for sea spray indicating ions.

Line 739: remove "immersion freezing", since some of the instruments, such as FRIDGE, only measure INPs of deposition or condensation-freezing mode.

Thank you for this attentive correction, agreed and changed.

Technical corrections

Line 55: remove "the" after "by";

We have modified the statement to read (lines 48 - 49) "*... they can modulate the microphysical properties of cirrus and mixed-phase clouds (MPCs) by initiating ice crystal formation.*"

Line 81 and Line 103: change "as e.g." to "such as that";

Agreed and changed.

References, such as Eriksen Hammer et al. (2018) in Line 113, Collaud Coen et al. (2004) in Line 125 and 127, 2013; Pandey Deolal et al. (2014) in Line 330, should be cited with the last name of the first author;

These are the last names of the first authors, please see the respective publication details.

Line 189: remove "for analysis";

Corrected.

Line 383: remove "then";

We removed "then".

Line 387: Should "FRIDGE and INSEKT" be "FRIDGE and HINC"?

Thank you, this is correct, we changed the statement accordingly.

Line 639: add "by" after "caused".

Agreed and changed.

References

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