Anonymous Referee #4

Some additional comments for the authors:

The authors would like to thank the referee again for thoughtful comments, that are helpful to improve the quality of our manuscript. Below, we provide our point-by-point responses. The referee's comments (RC) are shown from here on in black. The authors' responses (AR) are in blue below each of the referee's statements.

RC: - L137: replace 'RGB1000' with 'brush dispenser'as it tells the reader more than an instrument ID . AR: Replaced.

RC: - Given this is a submission to ACP, thus I would suggest the authors to consider to word section 2.5.1 (Extraction of total DNA and metagenomics analysis of sample microbiomes) in a more accessible way to the atmospheric science community.

AR: We agree. We reword the subsection title to "2.1.5. Analysis of sample microbiomes". We also begin the section with the following phrase, which explains the method to the atmospheric science community:

"The microbiome of our samples was characterized by metagenomics analysis. With this approach, total DNA is extracted from environmental samples; this DNA is a mix from all microorganisms and macroorganisms present in a sample. The qualitative and quantitative identification of microorganisms is carried out by amplifying (by polymerase chain reaction) and sequencing (several methods are in use) specific DNA segments of phylogenetic markers (genes that are used for identifying an organism) from the extracted and purified total DNA. Bioinformatics analysis of sequences obtained determines the nature and abundance of microorganisms in this sample."

RC: - L 269: for the section 2.2.3 title consider adding the article ,The' at the beginning as otherwise it is a bit hard to read as title/instrument name rather than a sentence on the first read of the section heading. And/or add the instrument acronym in brackets to the section heading; this should clarify for the reader.

AR: We added both "The" and the instrument acronym in brackets for Sects. 2.2.3, 2.1.3, and 2.1.4.

RC: - L 278 to 280, something is a bit odd in the sentence structure – the order of information (particular of ,in this study') is confusing.

AR: Our apologies for this confusion. We realized that our detection limit of 0.05 INP per L air is stated and given in the previous sentence. As this sentence (L278-280) contains repetitive information, we simply decided to omit it as the paragraph makes sense without it.

RC: - L282: here you talk about a grayscale to detect freezing onset for the drop freezing setup WT-CRAFT. How is this for the INSEKT instrument you describe earlier, can you specify if grey scale is applicable for INSEKT to?

AR: Yes, it was applied for INSEKT. For this study, we set a threshold value for the greyscale on our LabView-based image analysis tool. Once the optimized threshold value was achieved, we considered the well was frozen. We now clarify this point in L162-163 as:

"If a well froze upon the presence of an INP, a camera detected the associated brightness change based on an optimized greyscale threshold value set on the LabView software for this study." RC: - L329: RH also decreases in case c.ii. Thus I would recommend rewording the sentence to "... in almost all cases RH drops/decreases...".

AR: Ok. We rephrased the sentence as "In almost all cases, the *RH* dropped during some expansions at low temperatures."

RC: -L405 (Table 6): How do the authors account for the different sampling duration for the listed samples? What are/might be the implications for the n(INP)?

AR: Different sampling durations result in different air volumes sampled for individual samples. As can be calculated from Table 2, the average air volume sampled for individual samples (\pm standard deviation) was 955.0 \pm 300.5 L (min – max = 317.8 – 1560.0 L). While the air volumes deviated (mainly limited by on-site feedlot activities), we conducted our sampling activities when cattle were active in the evening hours under relatively stable ambient conditions as seen in Table 7 (except the winter sampling activities). To respond to the reviewer's point, we checked a correlation between the air volume (V_{air} , Table 2) and n_{INP} @ -25 °C (Table 6). Our results show r^2 of 0.069. Thus, our result implies that the different air volumes did not significantly impact to n_{INP} (otherwise, they might linearly correlated).

RC: - Figure 6: The order of the seasons (in panels a,b,c) is not intuitive. I would suggest the authors to change the order e.g. to (a) Winter, (b) Spring, (c) Summer and for easier reading of the graph to include the season names in the respective figure panel.

AR: Reordered as suggested. The figure captions as well as texts in Sect. 3.2.2 are also revised for consistency.



Figure 6. Downwind OLLF n_{INP} spectra from 2017 – 2019 sorted based on meteorological seasons are shown; winter (a), spring (b), and summer (c). The uncertainties in temperature and $n_{\text{s,geo}}$ are $\pm 0.5 \,^{\circ}\text{C}$ and $\pm \text{CI95\%}$, respectively, and error bars are shown at -5, -10, and -15 °C. The shaded area represents minimum – maximum n_{INP} .



Figure 8. The $n_{s,geo}$ spectra of OLLF aerosol particles from field ambient samples collected in 2017 – 2019. All downwind $n_{s,geo}$ spectra from winter (a), spring (b), and summer (c) are shown. Different symbol shapes correspond to individual OLLF sites as indicated in the legend. The uncertainties in temperature and $n_{s,geo}$ are ± 0.5 °C and ± 23.5 %, respectively, and representing error bars are shown at - 5, -10, and -15 °C. The shaded area represents minimum – maximum $n_{s,geo}$.

RC: - SI, Figure S4: There is a typo in the caption.

AR: Corrected. filed \rightarrow field