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Response to Anonymous Referee #1

To facilitate reading we use different fonts for (1)comments from Referee, (2) our response, *(3) the changes we made to the manuscript.*

The authors show the results of monitoring of ice nucleating particles (INPs) active at a relatively warm temperature (-8° C) in PM10 and PM2.5 at a coastal site in southern Norway. They also discuss possible linkages of INPs and local meteorological events. Despite low time resolution and limitation of available temperature (weekly INP data at -8° C), this work provides rare and very valuable datasets, since there are few studies that have reported seasonal variations of atmospheric INPs based on continuous longterm monitoring. However, I thought that more detailed discussion is necessary to suggest that INPs active at -8 °C may consist largely of fungal spores. I would therefore recommend this manuscript be published after several revisions.

We thank Referee #1 for having taken the time to read our manuscript and for providing valuable feedback on how to improve it.

The claim that fungal spores make up a majority of INP_{-8} at Birkenes seems too weak to stand the discussion. It was also questioned by Alex Huffman, the second Referee. Additional reading related to this issue led us to a more differentiated view and according changes to the manuscript.

Specific comments:

1) The wording "maritime climate (e.g., title)" may not be appropriate. This work is based on the measurements at a coastal site, but I didn't think that the measurements have been conducted in remote marine boundary layer. For example, the authors described that "probably, INP-8 were aerosolised locally by the impact of raindrops on plant, litter and soil surfaces (Page 1, Lines 10-11)". This obviously indicates the influence of terrestrial origin and not marine origin. To avoid misleading, I would strongly suggest avoiding the use of the wording "maritime climate" in the main text and considering a more appropriate title.

With "maritime climate" we meant to indicate that the climate at the observatory is shaped by the proximity of the sea. "Coastal climate" is equally valid and avoids misunderstanding.

We replaced "maritime climate" with "coastal climate" throughout the manuscript (incl. title).

2) It is a little difficult to understand the motivation of measurements of arabitol and mannitol. Although there is the description of "fungal markers arabitol and mannitol (Page 1, Line 15)" in the abstract, the authors should clearly explain why arabitol and mannitol can be regarded as fungal biomarkers in Section 2.4. In addition, in the first paragraph of Section 3.4, the authors may need

to briefly describe the reason why they decided to use arabitol and mannitol data.

Sorry for the operational blindness. We fully agree that more explanation is necessary.

We added a short sentence to the beginning of section 2.4: "Arabitol and mannitol have previously been identified as amenable tracers of fungal spores (Bauer et al., 2008)." (page 3, line 20).
Greater detail seemed more appropriate under 'Results and Discussion', at the beginning of Section 3.4. There, we added: "Arabitol and mannitol serve as carbohydrate stores in fungal spores. Their ambient air concentration has been found to correlate well with number concentrations of airborne fungal spores (Bauer et al., 2008), but not necessarily with ergosterol (Burshtein et al., 2011), a dominant sterol in most fungi (Weete et al., 2010). It seems that arabitol and mannitol are specifically associated with spores released under moist conditions, as occur in forests during nighttime (Zhu et al., 2016)." (page 7, lines 6-9).

3) The results of measurements of INPs after heating to 90 °C should be presented more clearly. The authors conclude that "sensitivity to heat treatment (90 °C) suggests bacterial or fungal sources, not pollen (Page 7, Lines 22-23)". For this results, however, there is only a very short description that "exposure to 90 ° C deactivated on average >93% of INP-8 in our assays (Page 6, Line 16)". If "the punches from 10 filters in each size fraction were tested a second time after they had been immersed for 10 min in a water bath at 90 °C (Page 3, Line 10-11)", it is probably important to show the results in a figure and/or table (and seasonal variations of INP-8 after heat treatment if possible).

For nine of the 10 samples tested a second time after heat treatment we can only provide an upper estimate for heat resistant INP because their number was below detection limit. Therefore, there is no seasonal structure we could show.

We now explain this issue in more detail in the 3rd paragraph of section 3.3: "However, we can exclude a major contribution of pollen-derived INP, because exposure to 90 °C deactivated all INP_{-8} in nine of the 10 samples treated that way and 92% of INP_{-8} in the remaining sample (26.3.-02.4.2014). Still, a few heat resistant INP_{-8} might have escaped observation at concentrations below the limit of detection in our approach (0.08 m^{-3}). If so, they would on average not have constituted more than 7% of all INP_{-8} found prior treatment." (page 6, lines 26-29).

4) It is hard to understand why the authors consider that "INP-8 may consist largely of fungal spores during the warm part of the year (Page 1, Line 15)" and "bacterial contributions may be more important than fungal sources during the colder part of the year (Page 7, Line 25, 26)". I assume that this is only based on the results that "from mid-May to mid-September, INP-8 correlated positively with the fungal markers arabitol and mannitol (Page 1, Line 15; Page 6, Lines 26-27)". This result may suggest that fungal spores are a potential important source of INPs from mid-May to mid-September, but I think that it is still impossible to rule out the possibility that the contribution of bacteria and soil organic particles were also significant. Did you try any other approaches to support the authors' hypothesis? For example, did you evaluate the relationship of INP-8 with other markers (e.g., bacteria, soil organic particles) from mid-May to mid-September?

Your interpretation of our views in the initial manuscript are correct. Reflecting on your comment and reading new papers we agree that the evidence we have at hand to support that view is not as strong as we initially thought it was. We do not have data for other parameters. When searching for answers to your comment we came across an interesting paper showing how bacterial and fungal communities on decaying leaves evolve in a highly dynamical and interacting succession (Purahong et al., 2016), which makes a strong point for the INP-8 sources probably also being highly dynamic throughout the seasons.

Accordingly, we added to the end of section 3.4 the two sentences: "Overall, the relative contribution of INP_{-8} from any type of microorganism might have changed by the end of September as a result of leaves starting to be shed by deciduous trees. Decaying leaves provide the substrate for a highly dynamical succession of interacting fungal and bacterial populations (Purahong et al., 2016)." (page 8, lines 6-8).
We also changed the statement cited in your comment, from "INP_{-8} may consist largely of fungal spores during the warm part of the year (Page 1, Line 15)" to "some fraction of INP_{-8} during that period may consist of fungal spores" (page 1, line 16); and changed "bacterial contributions may be more important than fungal sources during the colder part of the year" to "it might be that the fungal composition had changed or that bacteria had become more important sources of INP_{-8}." (page 8, line 3).

5) It is a little hard to understand that "from mid-May to mid-September, INP-8 correlated positively with the fungal markers arabitol and mannitol (Page 1, Line 15)" only based on Figure 5. For example, could you show the results (additional figures like Figure 4 or tables) comparing INP-8 with arabitol and mannitol measured in different seasons (e.g., the period of mid-May to mid-September vs. other periods; spring vs. summer vs. autumn vs. winter)?

We have added the requested Figure (Figure 6).

Technical corrections:
6) Page 2, Line 5: [f]ollow-up => follow-up?

Changed as suggested.

7) Please explain the definition of INP-8 more clearly. Is it ice nucleating particles "active at -8 °C or warmer (Page 1, Line 25)" or "active at -8 °C"?

Done.

The sentence now reads: "ice nucleating particles active at -8 °C or at warmer temperatures (from here on collectively denominated as INP_{-8})" (page 1, lines 26-27).

8) It is a little difficult to see the data on precipitation and/or snow depth in Figures 2 and 5. I would like to suggest that the values would increase from bottom to top if there are no special reasons.

We have tried the suggested change and found the Figure looks too crowded to properly capture its essence: the dynamics of INP mirror those of rainfall. By "mirroring" the precipitation and snow depth values on the horizontal axis (i.e. increasing from top to bottom) we find this feature is easier to capture.

We added to the legend of Figure 2 the brief explanation "inverse scale to mirror precipitation values on the horizontal axis, disentangling an otherwise crowded display of the data" (page 13, lines 8-9).

9) What is the value of 0.47 in Figure 4 (r or r2)? In addition, I would like to suggest that the authors would indicate the equation of the regression line, since they noted that "in PM2.5 we found consistently about half as many INP-8 as in PM10 (Page 1, Line 14)".

The value 0.47 denotes the slope of a regression line fitted to the data (i.e. there were on average 0.47 times as many INP_{-8} in $PM_{2.5}$ as there were in PM_{10}). The regression has an r value of 0.90.

Changed Figure and its legend accordingly. (page 14).

10) Is Figure 3 FLEXPART output? If so, please describe it in Section 3.2 and/or the caption of Figure 3. In addition, I would like to suggest including the explanation of why the unit of potential emission sensitivity is seconds (Page 4, Lines 3-4).

Yes, Figure 3 is FLEXPART output. The Figure legend is now more informative.

Changed Figure legend to: "Source receptor sensitivity (SRS) fields for situations with > 4 INP_{-8} m^{-3} (left) and when INP_{-8} were < 4 m^{-3} (right) as derived from FLEXPART. The SRS unit is seconds, which would result in a mass concentration (kg m^{-3}) at the receptor when multiplied with an emission flux (kg m^{-3} s^{-1}) into the model grid cells. Since emission fluxes are not known for INP_{-8}, SRS values can be considered as a measure of relative impact that INP emissions from a particular area would have had on INP concentrations at Birkenes. The potential influence was strongest from areas shown in red colour and weakest from those in white and purple colours." (page 14, lines 3-8).

<u>References</u>

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