Author response to comments by Referee #2

We thank Referee #2 for his/her work on the manuscript, and comprehensive comments and suggestions on revisions how to improve the contents. The detailed responses (written on red font) to comments as posted (green font) are listed below.

1. To better understand the implications of these measurements, it would be best convert the data shown in Figures 4 to 6 to active site density (ns) or active fraction kind of metric and compare against other INP data (soot, dust, etc.) from literature. This will help to put the data in the context of other INPs.

We agree that comparable metrics are essential when reporting scientific results. However, as we investigated the conversion options, we found that the active site density metric would very likely include big uncertainties due to assumptions (surface area) that are needed for such a conversion: biomass combustion soot particles have a tendency of being irregularly shaped agglomerates, unlike e.g. mineral dusts and/or biological particles such as bacteria. Therefore, when only their electrical mobility and effective density are known, the total surface area can range significantly from nearly spherical to a complex agglomerate yet maintaining the same electrical mobility and effective density. Hence, we consider results from this conversion speculative.

Instead, we shall present the data in activated fraction metrics and offer two-dimensional uncertainty analysis that contains uncertainties on lamina temperature (CFDC cooling system) and the statistical analysis of detector uncertainty comprising the OPC and the CPC.

2. Figures and Tables.

In Figure 1, do the ejector dilution (ED) also helps to cool the samples? (this is described on line 95).

Yes, because the compressed air we used was at room temperature. Regarding the high dilution ratio of 1:100, it can be expected that flue gas experienced also cooling during ejector dilution.

It is not clear how ice crystals can grow to size up to 11 um (Figure 2) as droplets only grew to 4.5 um only (page 256). Would you please explain this observation? If these droplets freeze, the size of the ice crystal should be equal to the droplet size, correct? From ~20.5 to 20.6 hrs (Figure 2), where ice crystals are observed, there are some particles of size 2±1 um observed. How is this possible? All the droplets should be frozen at this temperature. If these are not droplets, then why such small ice crystals are observed?

There are several reasons why the ice crystals appear significantly larger than the droplets detected with the OPC. In the following, we list two main reasons for the observed differences.

The detected particles had been exposed to an evaporation section where the droplets evaporated to some extent, while less is expected to happen to the ice crystals, so we do not know the exact size of the droplet population in the upper part of the chamber where freezing occurs.

The equilibrium vapour pressure is lower over an ice surface relative to a liquid water surface for temperatures below 0°C, so the growth of ice crystals is faster than the growth of liquid droplets in the chamber.

Please elaborate caption of figure 3. SPIN data is not shown here. How is the contribution from multiple charge particles is corrected for the data shown in figure 3?

The SMPS software includes automatic multiple charge correction, which was applied in the presented results in Fig. 3.

SPIN was operated at RHw =115% and without depol. detector. How was droplet breakthrough artefact addressed? It is not clear if the ice activation threshold is 1e-3, then how data is shown up to 1e-4(see figures 4 to 6). If the data (from 1e-3 to 1e-4) is not trustworthy because of the threshold limit, I would revise the figures to show data from 1e-3 to 1 only.

We addressed the droplet breakthrough artefact via choosing the ice crystal size threshold above the maximum droplet growth size (about 6 µm), which was determined from considering all experiments carried out in this manner during the campaign.

In chamber experiments, the threshold of 10^{-3} activated fraction does not represent the detection limit but the ice onset as we defined in the beginning of Sect. 3 on line 280: the purpose of this was to provide an ice activation threshold that is commonly used in the literature and is independent of measurement technique. The data between 10^{-4}-10^{-3} in ice-active fraction are well above the detection limit and thus trustworthy.

Please explain what X-axis labels in Figure 7 are. What is K500? How these labels are related to figures 4 to 6. I think there is a typo (‘ja’) on line 762. The ice detection limit (figure4 to 6) shows 1e-3, but in Table 1 detection limit is in the range of 1e-06. Please clarify this discrepancy and definition of the detection limit. From a readability perspective, it would be better to spell out the abbreviations (e.g., ND, FD etc.) that appeared in Tables 1 to 3 in the Table caption-text itself.

We have revised Fig.7 and tables in the following way:

- X-labels of Fig.7 have been changed to correspond with ones in Fig.6
- The typo on line 762 corrected
- Added definitions of ND and FD to caption of Table 3.

Please see the response to previous comment about the difference between detection limit and defined ice onset.

3. It is mentioned that the evaporation section is not efficient (line 262). Would you please explain this feature of the SPIN. Why it is not efficient, how it affects the data presented here, and how it is operated differently from other SPIN and CFDC style chambers. A paragraph from Line 273 to 276. Please elaborate on this argument. Does the correction factor was applied? If yes, how this factor was determined. There is some discussion in section 3.3; however, it is not very clear. The factor estimates described on line 458 to 461 are not proved using INP measurements. These are speculations. Please justify

Most of these issues have been addressed in our response to reviewer 1, and associated changes have been made to the manuscript. So here, we will only provide a few short comments.

The evaporation section of SPIN is shorter than for many other CFDCs, which most likely is due to a more advanced OPC in SPIN enabling detection of the depolarisation of the particles. Unfortunately, the depolarisation signal detected with this particular OPC did not suffice for assessing the phase state of single particles.

All shown results were not exposed to any corrections of the ice-activated fractions, since we find the correction too uncertain to do so.

For the estimated correction factors, please see our response to reviewer #1 with respect to L445-461. Our estimates of corrections factors have been described in more detail in the revised version of the manuscript.