We thank the referee for the useful comments, all of which we considered carefully. In the following, we respond to them separately, where the original comments are colored, while the answers are given in black.

Review of "The immersion freezing behavior of ash particles from wood and brown coal burning" by Grawe et al.

The authors present the immersion freezing ability and efficiency of combustion ash particles from the laminar flow tube study. The authors provide the immersion freezing dataset in two metrics, f ice and n s , in the temperature (T) range of -38 °C < T < -24 °C. More specifically, this study suggests the followings: (1) the brown coal burning ash particles, which is the proxy of anthropogenic combustion ashes, are more ice nucleation (IN) active/efficient as compared to the wood burning products (the natural proxy), (2) the fly ash particles are more IN active/efficient as compared to the bottom ash particles, (3) two aerosolization processes, namely dry and wet dispersion, result in different f_ice (T) and n_s (T) spectra, (4) the ultrasonic bath application prior to particle generation increases the IN activity/efficiency of a ash sample, presumably due to the presence of fewer agglomerates in the sonicated sample than the non-treated one.

General comments

The topic itself is an important addition to ACP, and the authors' new IN results potentially complement the results from previous study (Umo et al., 2015, ACP), in which the droplet-freezing assay was used to investigate in the immersion freezing ability and efficiency of combustion ashes in the T range of -11 to -36 °C. In general, the authors conducted a careful study, with their dedicated effort to examine a variety of sample preparation methods, e.g. atomization vs. dry dispersion, ultrasonic bath application. Unfortunately, such care was not taken in the preparation of the manuscript, with the manuscript containing a number of ambiguous statements, non-intuitive figures and over-interpreted results (e.g., Sect. 4, P5 L19-30). I have numerous critical revisions as listed below. Major revisions/suggestions are listed first, followed by the section-based specific and technical revisions. I would urge the authors of the manuscript to thoroughly proof read their manuscript as this list is too long. While authors may be able to address these issues, I do believe that the revision of the manuscript could be time consuming and result in a significantly different paper. For these reasons, I encourage the authors to resubmit it with a completely different format.

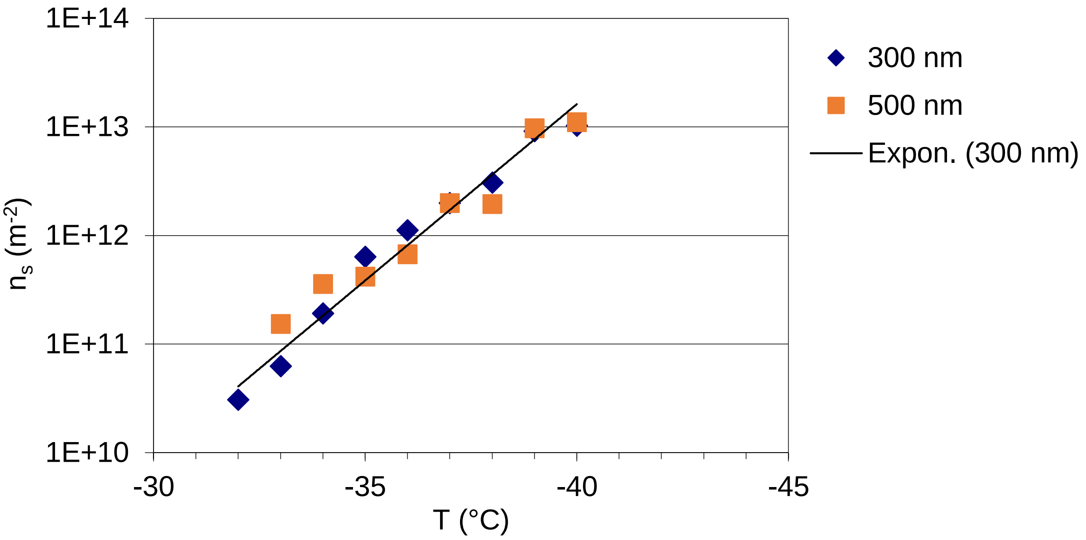
Specific comments

I suggest the following major revisions.

Discuss the representativeness of 300 nm diameter ash particles - Justification of selecting 300 nm diameter (P3 L26-28) is missing. The authors state that the ash samples are a composite material (e.g., P15 L22), but do not discuss why 300 nm diameter is representative for their IN analyses. Different size of particles may possess different composition and/or IN behavior (e.g., Wheeler et al., 2014, J. Phys. Chem. A). I strongly suggest the authors to conduct additional IN measurements and surface characterization with different sizes. Having another set of measurements for e.g. 600 nm (or even with polydispersity aerosol population) would add clarity.

From size distribution measurements during coal combustion, we know that there is a bimodal distribution: one submicron peak below 0.1 µm and one broad supermicron peak in the range from 3 to 50 µm (Damle et al., 1981, Aerosol Science and Technology 1:1). As particle collection techniques are less efficient for submicron particles (Flagan and Seinfeld, 1988, Fundamentals of Air Pollution Engineering), we decided to select particles smaller than 1 µm. 300 nm particles were chosen because we could get sufficiently high and stable number concentrations from the variety of samples when selecting this size (the mode diameters vary from 130 to 200 nm for dry particle generation). While there are a lot of different particle types included in bulk fly ash (i.e., Ramsden and Shibaoka, 1982,

Atmos. Environ. 16, 9), it has also been found from single particle analysis that the chemical composition of fly ash particles with aerodynamic diameters between 0.2 µm and 4.8 µm is remarkably consistent (Kaufherr and Lichtman, 1984, Environ. Sci. Technol., 18). Hence, we would not assume to see a difference in n_s, would we select another particle size.
In case of bottom ash, exemplarily, measurements were performed with 500 nm brown coal bottom ash particles which showed no difference in n_s within the range of our measurement uncertainty compared to the measurements with 300 nm particles (see below, now also added as green triangles in Fig. 4). As LACIS measurements are very time consuming and we do not expect to see a difference in n_s in the size range we are able to select, we would like to avoid performing further experiments with other particle sizes.
In the manuscript, a paragraph explaining why 300 nm were chosen was added (P4 L17-28).



Provide the results (no hype) to support your data interpretations and conclusions - One of the major findings out of this study is that the immersion behavior of brown coal ash particles changes depending on the sample preparation methods. The authors need to **investigate and discuss** in-depth physical reasons of why the observed difference appears. It seems not meaningful to attribute the reason to just the 'sample preparation and particle generation' (P11 L14-16), speculate potential reasons (e.g., P12 L1-3; P13 L10-14) and put it off as future work (e.g., P14 L28-32; P15 L20-25). To date, there have been some recent publications attempting to identify potential reasons of the data diversity due to different experimental methods and sample preparation methods (e.g., Hiranuma et al., 2015, ACP, Emersic et al., 2015, ACP). I suggest the authors to elaborate what can be further clarified on top of given previous findings. Reporting only the IN observations seems not novel enough to complement the previous ash IN study (i.e., Umo et al., 2015).

You are correct that a difference in IN activity between dry particles and particles in suspensions has been observed before. The studies you cite above also did not reach final conclusions on this. And we are the first to examine the difference between using and not using ultrasonification. This is an important new contribution to the topic, while obtaining the reasons for our observations is beyond the scope of this study. However, in our manuscript we now also discuss the literature you suggest, adding their speculations to ours (P18 L5-P19 L3).
Additionally to the discussion about differences between LACIS and cold stage measurements, we included a paragraph about differences between dry dispersion and wet particle generation with the help of an atomizer (P16 L7-P12). Concerning this, there have been observations showing that composition of wet and dry generated mineral dusts differ from one another and causing differences in

hygroscopicity (Herich et al., 2009, Phys. Chem. Chem. Phys. 11; Sullivan et al., 2010, Aerosol Science and Technology, 44).

Consider removing Sect. 4 - The atmospheric implications (P14 L8-17) sound too speculative and ambitious. The abundance data (concentration and size distribution) with some spatio-temporal distributions seem necessary to estimate the ambient ash-derived INPs. The back of envelop calculation presented here seems not novel enough for you to support your sub-conclusion, which appears in P14 L13-14 ("In conclusion,…") and P1 L13-14 ("ash from brown…a regional scale"). If the authors wish to keep the atmospheric implication section, the difference between airborne (fly) ash and surface (bottom) ash with respect to their mixing state, degree of agglomeration and atmospheric lifetime should be somehow discussed. Also, discuss the contribution of natural ashes to ambient INPs vs. that of anthropogenic ones. Otherwise, the authors may consider removing the entire section.

We consider this section an interesting supporting information to the manuscript. Differently from mineral dust particles, which have received much attention, not much is known about ash as INP in the atmosphere, and this section shows that it is worthwhile not omitting this aerosol type. We therefore do not want to remove this section. However, we have added some additional / updated information and more recent references.

Tighten up the writing by removing unnecessary words - Improving the language, structure, presentation seems necessary. The authors should avoid making a review question how careful the research team is.

For example, I suggest minimizing ambiguous (and unnecessary) adverbs and adjectives to make the manuscript more scientifically sound than the current form. Such words should be replaced with specific and explanatory descriptions/values. Otherwise, the authors should reinforce them by adding proper citations. My suggestions include, but not limited to:

P1 L6: more (specify how much in what T range?)
P1 L14: at least
P2 L12: major
P2 L15: rough
P2 L20: for a long time & large
P2 L22: strongly
P2 L24: very similar
P2 L32: lower (than what?)
P3 L4: slight
P3 L23: some
P3 L24: mostly (define which samples)
P3 L30: larger & more (than what?)
P4 L30: exactly
P5 L12: most (amongst what?)
P5 L13: slightly
P5 L14: significantly
P5 L17: most striking
P5 L19: small
P5 L23: perfectly (I do not think so) & most
P5 L24: significantly less (is it fair to say this by inspecting a single snapshot picture?)
P5 L27: similar
P5 L30: small
P5 L31: small
P6 L1: supposedly & obviously
P6 L5: possibly & weakly
P8 L10: almost entirely
Fig. 4 caption: at least
P10 L2: significant & low
P10 L4: most effective & least & rather similar
P10 L6: small & more (than what?)
P10 L7: more (than what?)
P11 L2: certain (specify)
P11 L15: considerably
P11 L16: by several tens of percent (just give a number)
P11 L20: probably exclusively & completely
P12 L1-2: most likely
P12 L24: numerous
P12 L25: large
P12 L26: higher (as compared to what?)
P12 L29: larger (how much?)
P13 L12: likely
P14 L15: significantly low
P14 L24: significantly (how much?)
P15 L3: barely
P15 L4: significant
P15 L6: likely
P15 L 11: eventually
P15 L20: most likely

The text is now more specific where useful. However, for those of the locations you listed below that refer to figures, adding numbers to the text only decreases the readability without adding information to the manuscript. Hence, where numbers can be seen from figures, we did not change anything. Furthermore, it is often not possible to give specific values, as those have not been reported in literature until now. Words like “possibly”, “supposedly”, “probably”, “likely” need to be used in those parts of the manuscript where we discuss hypotheses.

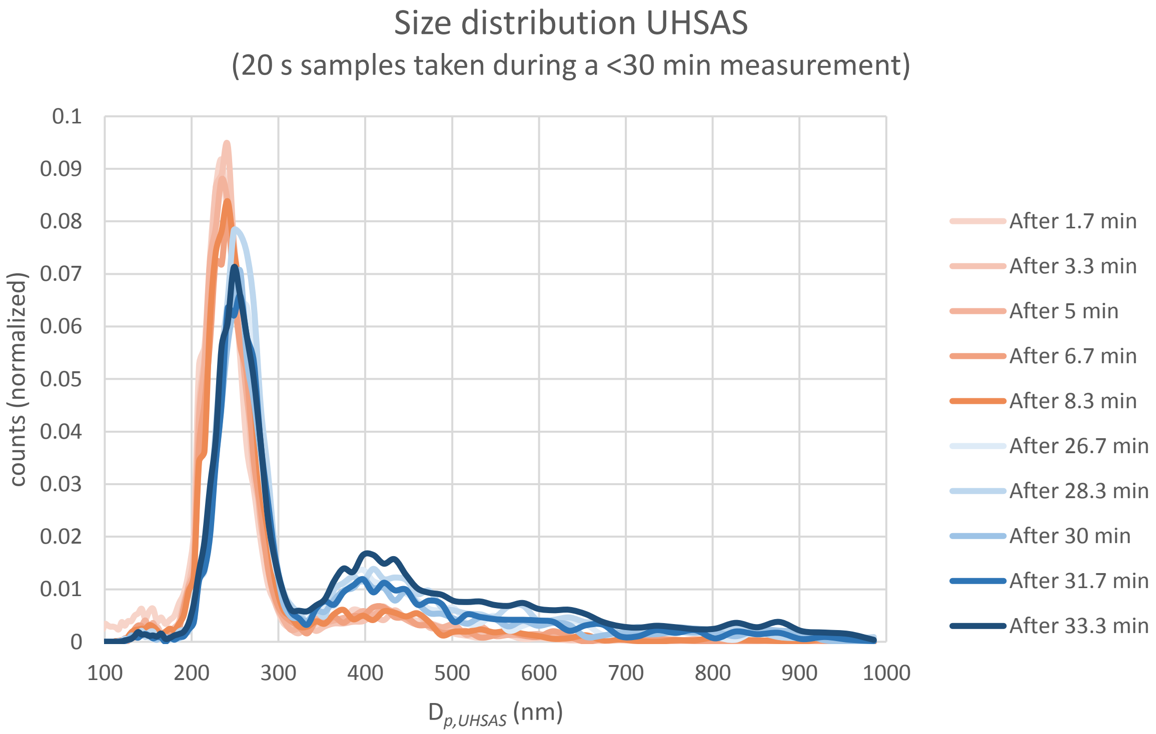
Improve the figure and table presentations - In general, all figures and tables should be self-explanatory. My suggestions include the followings:

Fig. 1: It seems that more than 50% of mass are composed of materials that are not listed in the figure. The authors need to clarify this point to the reader by adding descriptive text or adding another group of 3 bars showing the sum of the other components in the figure. The authors implies such contribution may in part come from ‘amorphous’ material (P11 L1). I suggest the authors to give an idea of what they are (perhaps carbonaceous materials?). I also suggest the authors to give a proper reason of why beech ash composition is not shown (P5 L9). Perhaps, presenting data (± uncertainty) with the table format may be more intuitive to the reader than using the figure format. In addition, the figure caption should read “bottom ashes from spruce, birch…”.

In addition to Fig. 1 we also included a table format (Tab. 3) with the corresponding values. There, the sum of the analyzed components as well as the associated uncertainties are given. We did not want to omit the diagram, as the illustration of the numbers might help to take in the large amount of information. You correctly observed that the given values do not add up to 100 %. This is due to the analysis procedure: It is standard to recalculate the concentrations of the ten major ions into the mass of their most common oxide forms, even though they may have had other counterions in the original sample. In some cases the compounds are indeed oxides, but we do not know the counterions in each case. K for example would be more likely to occur as KCl, K2CO3 etc., than as K2O. Any missing percentage is due to other than these ten elements and the fact that other counterions would have been involved. This has been added to the manuscript (P7 L2-P9 L2).
From previous studies we know that the amorphous material in fly ash is mainly aluminosilicate glass (Ramsden and Shibaoka, 1982, Atmos. Environ., 16, 9; Querol et al., 1996, Atmos. Environ., 30, 21). This was added on P14 L22-24. The low loss on ignition (LOI) value for fly ash (see Tab. 3) indicates that there is barely any carbon left in our sample. Hence it cannot be contained in the amorphous fraction.
Beech ash is not shown in the analysis as it was provided later, after the other samples had already been analyzed in Sweden (added on P6 L24-P7 L1). We decided to include the data, even without the chemical analysis, because we wanted to show that beech ash is comparable to the other wood bottom ashes.
The caption was corrected.

Fig. 2: I see at least six particles that have >1 μm diameter in the panel b. As stated in P5 L24-26, the doubly charged particles of 300 nm cannot be >1 μm. The authors state that “…the number of particles with three or more negative charges was negligible…” (P3 L32-P4 L2), but it seems not negligible and contradicting. In terms of the number, those large particles may have only a small contribution. However, they may substantially contribute to the total surface. If that is the case, they should be accounted for the immersion freezing parameterization. Otherwise, your ice nucleation active surface site density would be overestimated because of overlooking the presence of large particles. Adequate surface estimation is one of the important keys for the n_s parameterization. Ultimately, the authors may want to assess if such correction can explain the discrepancy between dry and wet (or not). Just to start with, you may first estimate the particle size distributions by analyzing SEM images (i.e., estimate the area equivalent diameter for several hundreds of particles; see Hoffmann et al., 2013, AMT). This approach may be better than relying only on the USHAS measurements. In addition, such work will help clarify the vague statement in P5 L24-30.

The multiple charge correction was performed based on size distribution measurements with the UHSAS. For this, 300 nm particles were generated in the same way as for LACIS measurements and sampled for several minutes. For UHSAS and LACIS measurements, particle production only operated for short amounts of time (<30 min), before cleaning the cyclone prior to a next set of measurements. The filter samples, however, were collected over several hours to assure a sufficiently high concentration on the filter surface. New UHSAS measurements (see below) show that the cyclone, which we used to minimize multiply charged particles, is filled after around 30 minutes, so that more larger particles are present than for the beginning of the UHSAS measurement. Additionally, it was also seen in APS measurements that there was an increasing amount of supermicron particles passing the DMA and cyclone as the generation system ran for more than 30 min. As LACIS measurements typically last around 20 minutes only (due to wall glaciation effects), and the cyclone was cleaned after each LACIS measurement, we are certain that the UHSAS determined multiple charge fractions (see below) correspond to the actual fractions in the flow tube. The large particles which can be seen in Fig. 2 b) of the original manuscript have accumulated during the long collection time in which the cyclone was not cleaned often enough. We decided to collect fly ash particles from dry generation on filters once again, now cleaning the cyclone every 30 minutes. The new SEM pictures (see examples below, one was added to the manuscript as Fig 1b) show a majority of particles in the size range of 300 nm. 68 of 84 counted particles were in the size range of around 300 nm, while 16 particles classified as particles larger than 500 nm. This might not be significant from a statistical point of view, but it is similar to the findings of the UHSAS measurements, where 19 % of all particles were identified as doubly charged.
We omitted Fig. 2b) from the original manuscript to avoid confusing the reader with the occurrence of large particles which are only present on these particular filters but not in the LACIS measurements. In light of the new observations, we changed the paragraph on P5 L19-30 of the original manuscript accordingly (P9 L14-24).



SEM images of fly ash particles from dry generation:

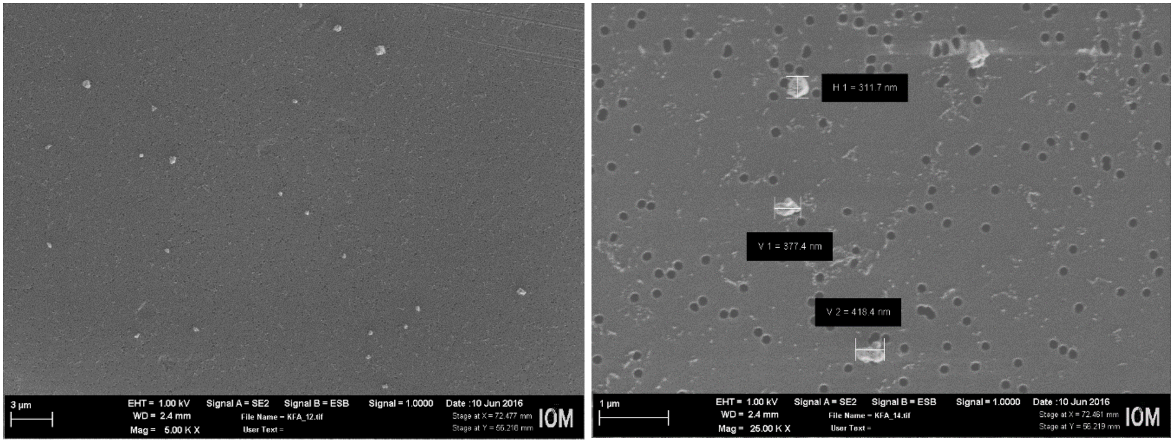


Fig. 4: The authors need to explain the factor of 4.54 in text at the first appearance of Fig. 4. I also suggest clarifying what the 'clay mineral baseline' means. Better presentation may be made with multiple panels. For example, the authors may use individual panels for ash type comparison, dry vs. wet, +US vs. –US. The same may apply to Fig. 5.

Fig. 4 (now Fig. 3) was changed as suggested and the correction factor is now explained in the caption. We added further explanation concerning the clay mineral baseline on P14 L30-P15 L1 as well as in the caption of Tab. 4. The original Fig. 5 (now Fig. 4) was not altered in terms of its layout.

Fig. 5: Specify if your n_s metric is based on the BET specific surface area or the geometric surface. Though I agree with your statement in P13 L8-10 (i.e., the influence/difference is small anyway), it is important to compare the results with a same metric. If this figure contains both BET-based n_s and geometric n_s spectra, I suggest presenting all data and spectra using either one of two metrics. The authors may simply apply a factor of 4 (P13 L9) for the conversion.

This is a fair point. However, instead of showing the BET based n_s values calculated from the specific surface area given by Umo et al. (2015), we decided on introducing a second y-axis to the plot. The left y-axis is for the n_s values from our measurements assuming spherical particles (circles) and the right y-axis is for the BET based n_s values (lines) reported by Umo et al. (2015). We added arrows to indicate which axis belongs to which data set. We decided to present the data in this way, because we cannot provide BET measurements for our samples within this short time frame and we cannot be sure that our samples compare to the samples from the Umo et al. (2015) paper concerning their specific surface area. However, we also recalculated our n_s into a BET based n_s using the specific surface area values given by Umo et al. (2015) and saw a change of a factor 3.5 at most. This and a detailed explanation of Fig. 4 (former Fig. 5) have been added to the manuscript (P17 L10-12 and P17 L23-33).

Add proper references – I suggest the authors to give a careful look at the followings:

P1 18: Add citation after "…and climate models".
We added Koop and Zobrist (2009, Phys. Chem. Chem. Phys. 11) as a reference.
P1 L21: Replace Murray et al., 2012 with Koop et al., 2000 (Nature); Murray et al., 2010 (Phys. Chem. Chem. Phys.); Rosenfeld and Woodley, 2000 (Nature).
Done.
P2 L5: Add citations after "…been conducted".
Done.
P3 L1-2: Whale et al. (2015, AMT) may be a good reference to add here.
Added.
P3 L20: Add reference for your dry dispersion method.

There is no publication mentioning the generator. We now cite a dissertation from our group which is unfortunately only available in German (M. Rösch, 2015).

P4 L8: Add reference for “water contamination”. The authors may also want to reduce the tone and call it as a background contribution or something similar.

We changed water contamination to impurities. As references we added Budke and Koop, (2015, AMT 8) and Whale et al. (2015, AMT, 8) (P5 L9-11).

P7-8: Provide the reference for R1 & R2.

We chose not to include the chemical reactions any more, as they were already described in the text. Steenari et al. (1999, Fuel, 78) was added as a reference (P11 L1).

P10 L17: Any reference for insoluble K to be highly ice active? You should not speculate for all insoluble materials to be IN active.

K is insoluble in K-feldspars which are known to be ice active. Sure, we do not know about the mineralogy of our samples, but we have made it clear that this is an assumption rather than a fact.

P12 L1-3: multiple citations seem missing.

This part was revised. We added two studies as a reference where it was shown that particle generation affects particle surface properties and composition (Herich et al., 2009, Phys. Chem. Chem. Phys. 11; Sullivan et al., 2010, Aerosol Science and Technology, 44).

P12 L8: Look into references given in Zolles et al. (2015). There have been a few other studies of active sites and their influence on IN activities.

We now also cite Hiranuma et al. (2014, ACP) and state that the fragmentation in the ultrasonic bath could have a comparable effect on the ice fraction as the milling procedure.

P12 L16: e.g., Connolly et al., 2009 (ACP), Niemand et al., 2012 (JAS)

Added.

Other specific and technical suggestions sorted out for each section are listed below.

Abstract

P1 L2: …trigger ice nucleation when they interact with water vapor and/or supercooled droplets.

P1 L4: …worldwide, and…

P1 L6-7: I suggest separating this sentence into two sentences.

P1 L6: ice active in the immersion mode

P1 L8: …the effect of various particle generation methods on…

P1 L8: For this → For instance

All done.

P1 L14: heterogeneous ice nucleation → immersion freezing in the T range of XX to YY °C

We changed “heterogeneous ice nucleation” to “immersion freezing”. However, including the temperature range does not help the readability of the sentence.

Sect. 1

P1 L21-23: The authors may focus on immersion freezing by rephrasing L21-23 to, “At water saturation, this temperature limit of droplet freezing … referred to as immersion freezing.”. Otherwise, describe heterogeneous IN and then immersion freezing. Note that the deposition nucleation can occur at temperatures below -38 °C.

Done.

P2 L4: However, up to now only a very few → To date, only a few

Done.

P2 L5: → material, which

Done.

P2 L12: contribute to

Done.

P2 L12: → Furthermore, the ash from natural source is…

Biomass burning is often anthropogenically induced. We'd rather not imply that all biomass burning ash is emitted due to natural causes.

P2 L13-14: Awkward sentence. Rephrase.

We changed the sentence to: "The impact of ash particles as potential INPs must be put into perspective by comparing ash emission rates to those of other INP containing aerosols…". Hopefully, this brings more clarity.

P2 L18: I wonder where the authors find the 7% number in DeMott et al. (2003).

It is not stated directly in the text. However one can calculate that 20% of 33% is equal to 7% of the total ice crystal residue number.

P2 L19-20: I suggest clarifying that the result presented in DeMott et al. (2003) is from a limited time segment of measurements in cirrus clouds.

Done.

P2 L23-26: I do not understand what this sentence mean. Please clarify.

We are not completely sure what is unclear. This part was divided into two sentences to increase readability.

P2 L27-29: I suggest separating this part into two sentences.

Done.

P2 L33: What are water soluble components? Sugars? Any biological materials? How important are they for IN as compared to the insoluble components?

The sentence before the one you refer to here explains your last question ("…particles freed from water soluble components initiated freezing at lower temperatures."). Maybe you were thinking of ice active bacterial macromolecules, which is not what we are talking about here. Hence, the addition (P3 L7-8) that it is dominantly $CaSO_4$ hopefully clarifies your questions well enough.

P2 L35-P3 L2: Provide more information regarding Umo et al. (2015). At least the investigated T ranges for individual ash materials and their IN efficiency/activity with some quantities.

Done.

P3 L2: Reword "however". I do not find smooth transition.

Done.

P3 L5: Reword "assumed". e.g., suggested/postulated

Done.

P3 L11-12: Move this part to the method section.

Another referee requested a more detailed motivation of our study in the introduction. We would hence like to keep the sentence in this position because it underlines the difference in methodology in comparison to the previous study.

Sect. 2.1.1

P3 L21: as → along with

Done.

P3 L21-23: Subdivide the sentence ("Variations caused … transported downwards.") into two.

Done.

P3 L23-24: Clarify if glass beads have any influence on modification of particle surface (e.g., by scratching) and IN behavior.

We do not expect the beads to have any influence on the IN behavior of the particles. Firstly, we used only 20 beads, hence, the probability of a particle which would later be investigated colliding with one of the beads is rather low. Secondly, if collisions with these beads would lead to a particle modification, then collisions with the flask wall would also. Wall collisions are common with other dry dispersion techniques (e.g., fluidized bed generator) and are not expected to modify the particle surface, so why should our beads? Secondly, the beads did not appear milky after several hours of particle generation, indicating that no significant scratching took place. An explanation was added to the manuscript (P4 L11-12).

P3 L24-26: I suggest rephrasing the complete sentence ("In addition … dryer unit."). Just say you used a custom-built atomizer. Is it perhaps the one used in Wex et al., 2015, ACP? If so, add it as a reference.
We prefer not to cite anything here, as this is a standard procedure used frequently by many groups in aerosol research.
P3 L31: efficient → active due to the presence of large surface
Done.

Sect. 2.1.2

P4 L4-5: Awkward sentence. Rephrase. The LACIS reference is repetitive.
We changed the sentence to "The immersion freezing behavior of the previously generated and size selected ash particles was investigated with LACIS." The reference was deleted.
P4 L6: "LACIS offers … nucleation processes."; please provide the reference showing the 'surface' (of the droplet assay plate - I assume this is what you mean) interferes with IN measurements. If not, I suggest rephrasing the sentence.
We omitted the half sentence in question.
P4 L14-15: I suggest briefly clarifying how the phase discrimination can be done in text.
We added that "the approach to determining the phase state of the hydrometers is based on the fact that the former polarization of light is maintained for scattering at spherical hydrometers (supercooled water droplets) while non-spherical hydrometeors (ice particles) cause depolarization" (P5 L18-20).

Sect. 2.2

P4 L18-23: I suggest to summarize your ash samples using a table along with columns of ash type (bottom or fly), source (natural or anthropogenic), particle generation method (dry or wet), and information regarding your IN experiments (e.g., examined T range and IN observed T range).
We included the suggested table in the manuscript (Tab. 2).
P4 L25-26: "It has to be noted that…"; not much adding. I suggest removing this sentence.
The composition of the coal depends on the deposit it was taken from. Consequently, ash from different coal will have different compositions. We think the reader should know that both coal types were taken from different deposits.
P4 L27: If it is commercially available, specify it.
We could only find out the manufacturer of one of the domestic ovens. For two of the ovens it was simply not traceable. It turned out that one was actually home-built. In the light of this, we omitted "commercially available" in the cited sentence.
P4 L27: Explain how you "extracted" it. Provide some information regarding the electrostatic precipitator (or reference). What is the cutsize? What is the collection efficiency? etc.
The ash was simply taken out of the collection tank of the electrostatic precipitator. Our contact at the power plant provided the overall efficiency which is 99.98 %. This information was added in Sec. 4. Furthermore, the flue gas behind the filter must not contain more than 50 mg per m^3. There is no information concerning the cut-size or the size-dependence of the efficiency.
P4 L28: → station, which
Done.
P5 L4: affected → modified
Done.
P5 L5: "one fly ash suspension sample"; specify which one and why the authors choose this one in text.
This simply implies that several identical samples were prepared, one of which was not put into the ultrasonic bath. Nothing was changed.
P5 L9: Provide brief description of atomic adsorption methods along with proper reference(s). The rest of the paragraph (up to L18) may better fit in the results section. Consider reorganizing the sections.

Our partners in Sweden further specified the method to be Inductively Coupled Plasma Sector-Field Mass Spectrometry (ICP-SFMS). This is now stated in the text. We added a reference describing the method (Zheng and Yamada, 2006, Talanta, 69).

We did not move the chemical composition analysis to the results section as none of the other referees raised objections. We think the sample characterization better fits in the methods and materials section.

P5 L11: obtained → estimated

Done.

P5 L19-30: I suggest the authors to minimize over-interpretation from snap shot pictures. The authors may consider providing the reader with the proxy of particles' sphericity. For example, a particle aspect ratio of individual particles can be estimated by the 2D SEM images. Inspecting several hundreds of particles and providing statistically valid data in the result section will strengthen the paper.

We weakened the statement and avoided the use of the word "spherical".

P5 L23: Irregular shape may suggest the predominance of carbonaceous compounds (e.g., Hiranuma et al., 2008, Atmos. Environ.). Do the authors have any information regarding its organic fraction and content?

Yes, there is information concerning the organic fraction which is now included in Tab. 3. Here the loss on ignition (LOI) value corresponds to the organic content, i.e., it tells us about amount of unburnt fuel in the sample:

Spruce: 22.9 %, Coal bottom: 10.5 %, Coal fly: -0.8 %, Birch: 26.7 % (uncertainty +- 5%)

We do not know the exact composition of this organic fraction, but there will be carbonaceous matter included since these particles are produced during incomplete combustion (Kucbel et al., 2016, Perspectives in Science, Vol. 7). We added a paragraph about the relation between carbon content and particle shape including the citation you suggested (P9 L25-32).

P5 L23-24: "…most show significantly less surface defects…"; it is hard to judge the presence of defects by looking at given SEM pictures with low magnification. Can the authors provide the image with high magnification as an example? Better option would be measuring BET surface of both bulk samples and comparing each other.

We weakened the statement and now say that the lowest BET values were found for coal fly ash in the previous study by Umo et al. (P9 L31-32).

P5 L25-26: I wonder why UHSAS was not able to measure them.

Our new SEM images show that the UHSAS was never the problem. The UHSAS has an upper size limit of 1 µm. As already elaborated on in more detail above, we saw the large particles on the filters because particles were collected for several hours during which the cyclone filled up. The UHSAS and LACIS measurements, however, did not last longer than 20 min and the cyclone was cleaned after each measurement. We are now sure that the UHSAS determined double charge fractions correspond to the fractions in the flow tube (see estimation given above).

P5 L27-28: "…similar particle losses should have occurred."; I suggest removing any opinion statements. The authors may provide the results instead if available.

The entire paragraph (P5 L24-30 of the original manuscript) where we try to explain possible reasons for the discrepancy between SEM and UHSAS was removed. As we now know that the filled cyclone was the reason, the discussion about particle losses seems unnecessary at this point. However, we added a short sentence concerning this on P5 L4-5.

P5 L33-35: "It is reasonable to assume … additional fly ash particles."; I do not understand your logic here. Please clarify.

The needle shaped particles are not present in the dry sample and hence are special to the suspension. Insoluble material will not change when put in water, so these needle shaped particles can be assumed to be formed from dissolved material, and hence our statement. As it is not clear to us what was unclear to you. Here, nothing was changed.

P6 L3: → all components are dissolved in droplets

Surely not all, just the water soluble components. Nothing was changed.

P6 L5 to the end of this section: this part seems not belonging to the method section. Should be moved to the discussion section?

The section is called "Methods and materials". As we kept the discussion of the chemical composition of the dry samples in its place, it would not be consistent to move the part about the soluble components to the results section.

P8 L9-10: Awkward sentences. I suggest rewording.

Done.

Sect. 3

P8 L13-16: Subdivide this sentence to two parts.

Done.

P8 L17-19: Why don't you look at only quasi-spherical ash particles on the SEM images? Analyzing the SEM images, you should be able to distinguish ash particles from the recrystallized components, which seem possessing high aspect ratios. This procedure perhaps enables you to estimate the contributions of multiple charge components. The same applies to your statement in P11 L12-13. The correction seems feasible and important.

The correction you suggest here was done. This was described on P11 L26-30 of the original manuscript. As the number of ash particles on the filters, compared to others, was rather small, a number of multiply charged ash particles cannot be derived from these SEM images. With the UHSAS it is not possible because the two populations cause overlapping signals (P8 L17-19 of the original manuscript). However, to estimate the multiple charge fractions in the suspension measurements, we weighted the bipolar charge distribution (Wiedensohler, 1988, Journal of Aerosol Science), i.e., the probability of the particles to receive one, two or three negative charges in the neutralizer, with the measured size distributions of the ash-water suspensions. There is a caveat to this estimate, as we do not know how the size distributions would look like for the insoluble particles only. It turned out, that the highest multiple charge fractions would probably occur for the fly ash suspension (+US), where we calculated 80.5 % singly, 16.8 % doubly, and 2.7 % triply charged particles. The multiple charge fractions were even lower for the other suspension samples. Would we perform the multiple charge correction using these fractions, our measured data would be reduced by a maximum factor of 2 only. A short version of this was added to the manuscript (P15 L10-16).

P8 L21: → in Fig. 4. These calculations are based on…

Done.

Sect. 3.1

P10 L3: Specify the T range for the reader.

Done.

P10 L4-6: Discuss why all the wood burned ash particles have similar IN behavior. There seems difference in SiO2 content for spruce (~10%) and birch (<5%). Based on your statement in P5 L12-13 (i.e., the higher SiO2 , the more IN efficient), could the SiO2 content be responsible for the observed result of n_s spruce > n_s birch in Fig. 5? If this holds true, the bulk beech ash should contain the highest SiO2. Would you consider carrying out the atomic adsorption measurement of beech ash (currently missing without any intuitive explanation) to support your idea?

Yes, SiO2 could play a role for the wood ashes as well. However, the wood ashes are really close together (logarithmic scale) so that we would like to avoid the over-interpretation of these slight tendencies.

P10 L9 - P11 L3: The authors suggest that the fraction of SiO2 , Hg and insoluble K has some contributions to determine the immersion freezing behavior of ashes based on their bulk observation of atomic adsorption technique (P10 L9-22). In the following sentences, the authors

introduce the hypothesis, which infers that the amorphous material content is also important as a determining factor of IN behavior.
Discuss which one is more dominant. Further, what is the mixing state of your ash samples? What is the relative importance of the mixing state as compared to the "bulk" composition?
Our measurement methods are not able to distinguish between the importance of these different possible contributions, and hence we discuss them as suggestions. The same applies for the mixing state: as long as it is not known what it is that makes the difference (or, in other words, what it is that makes the IN activity - which is something the whole IN community is looking for), it can also not be stated how the mixing state of this unknown component is. Still, we had hoped that the bulk composition would reveal something more meaningful. This was, unfortunately, not the case.

P10 L10-12: Rephrase. Not easy to follow.
Done.
P10 L14: → act as INP. This previous observation may be relevant to our study as…
Done.
P10 L14: Is the atomic adsorption technique used in this study sensitive to iodine? If so, please show I in Fig. 1 too.
Iodine was not investigated because, initially, we expected it to be a minor component in the ash samples. Unfortunately, there is no time to rerun the analysis and investigate for the I content. We cannot rule out HgI_2 as a factor, so nothing was changed.
P10 L16: Why soluble? Presumed? Or measured? Please clarify.
We added the following explanation on P14 L15-16: "K in wood ashes is present in the form of soluble salts and oxides, whereas coal ashes contain K in clay minerals with low solubility", citing Steenari et al., 1999, Fuel, 78. In the course of this, we changed the previous citation of Andreae et al., 2004, Science, 303 (original manuscript P5L16) to Steenari et al., 1999, because the reasoning is presented more clearly in the latter.
P10 L18-22: "Apart from the chemical composition, other properties such as ice active sites…"; Discuss the relative importance of what you found to the other potential factors. I suggest rephrasing and clarifying the last sentence. I do not understand what the authors mean here.
This sentence was deleted. We cannot make any statements on how likely these speculations are.

Sect. 3.2

P11 L12-13: I disagree. The correction is possible and feasible, and the authors should account for it. See my comment in Sect. 3.
As stated earlier, we are talking at cross purposes here. The multiple charge correction is not synonymous with the correction for the occurrence of needle shaped crystals (P11 of this document).
P11 L13-14: This sentence seems not needed here. I suggest removing.
The sentence is needed to remind the reader that n_s values might be lower for the wet generation measurements.
P11 L14-16: The authors need to investigate and discuss physical reasons of why the influence of the US application on IN behavior is material dependent rather than just saying 'sample preparation' methods resulted in the difference. The same goes to P13 L3-4.
We now discuss possible reasons for the difference in IN efficiency when changing from dry to wet particle generation in more detail (P16 L6-25 and P17 L35-P19 L9). That we do not see a decrease in f_ice for spruce suspension is hence related to the fact that the discussed possible reasons are apparently not relevant in case of particles from the spruce ash suspension.
P11 L18: meaning → suggesting
Done.
P11 L17-30: Multiple topics seem to be squashed within this short section. Reorganize and rephrase the sentences. Again, the scaling factor of 4.54 should be introduced prior to its first appearance in Fig. 4.

The scaling factor is now mentioned in the caption of Fig. 3 (former Fig. 4). We think the detailed explanation better fits where we discuss the results for wet particle generation. However, we reorganized the paragraph and now start with the discussion of the filtered fly ash suspension and later go on with the non-filtered suspension. We hope that the need for the scaling factor of 4.54 and the paragraph itself are more understandable now.

P12 L1-3: Sounds too speculative. Any experimental evidence of a destruction of former active sites? I do not understand the last sentence. How do water soluble components play a role in what?

This was rephrased. We do not mention the destruction of sites anymore. Instead there is a discussion about how particle properties could be changed during wet particle generation (P16 L6-12).

P12 L8-13: What is the atmospheric implication of your findings here?

The relevance of these findings is, that possibly samples that are treated with an ultrasonic bath might yield different concentrations of ice nucleation active particles than when this treatment is omitted, and that the use of the respective reported concentrations should be done with caution. This follows implicitly from the text, and nothing was changed.

P12 L23-24: Repetitive. Delete.

Done.

P12 L24-30: Rephrase. This part may better fit in the introduction section as part of an example of previous work regarding ash IN characterization.

We think the information is better suited to be stated here, as it belongs to the explanation of Fig. 4 (former Fig. 5).

Sect. 3.3

P12 L15: ice nucleation active surface site density

Done.

P13 L5: nucleation spectrum → ns spectrum

Done.

P13 L10-14: Sounds too speculative. I suggest removing the opinion statements (that is, must be, expect, likely) and rephrasing the sentences.

We added a references for the time-dependence statement (Ervens and Feingold, 2012; Welti et al., 2012; Wex et al., 2014) and a discussion about how the methodology (wet generation vs. cold stage suspension) could affect n_s values citing the suggested papers by Hiranuma et al. (2015, ACP) and Emersic et al. (2015, ACP).

Sect. 4

P14 L2-4: The authors may consider removing this paragraph, which seems not adding much.

P14 L5-8: These sentences better fit in the introduction section (e.g., P2 L15) rather than here.

The section has been revised (see P3 of this document).

Sect. 5

P14 L21: at the

Done.

P14 L23: differences (in what?)

Differences in the immersion freezing behavior. This was added.

P14 L28: "a decisive factor…"; I disagree. The authors need to provide much more comprehensive composition data (e.g., beech ash data, bulk vs. single particle, mixing state, organic speciation) than what are presented in this manuscript to have this statement as your conclusion.

As we cannot provide the requested data for now, we weakened the statement and omitted "decisive" so that the sentence now reads: "… however, a factor could be the presence of insoluble K in the coal ashes.".

P14 L28-32 & P15 L20-21: I agree with the authors that multiple particle properties inherently influence particle's immersion freezing behavior, and it is not easy to understand what the controlling(s) factor is(are). With some extra experiments suggested above, the authors may be able to shed light on the questions raised here.

Additional experiments to round up our results were done with respect to multiply charged particles (see our answers to your remark on P5 of this document), as well as regarding particle hygroscopicity (as requested by another reviewer, now added in P11 L17-21). However, further additional examinations are beyond the scope of the here presented study, so that mentioning these issues here, at the end of the summary and conclusion sections, can be seen rather as a hint towards readers on directions in which additional rewarding research can be made.

P15 L9: nucleation spectrum → f_ice and n_s spectra; the same applies to elsewhere, e.g., P15 L17

Done.

P15 L12-13: What is its atmospheric implication?

We added: "…which could imply that previous results might have overestimated the ice nucleation ability of certain substances." (P21 L18-19).

References cited in our answers:

Andreae et al., Science (2004)
Budke and Koop, AMT (2015) and
Damle et al., Aerosol Science and Technology (1981)
Emersic et al., ACP (2015)
Ervens and Feingold, ACP (2012)
Flagan and Seinfeld, Fundamentals of Air Pollution Engineering (1988)
Herich et al., 2009, Phys. Chem. Chem. Phys.
Hiranuma et al., ACP (2014)
Hiranuma et al., ACP (2015)
Kaufherr and Lichtman, Environ. Sci. Technol. (1984)
Koop and Zobrist, Phys. Chem. Chem. Phys. (2009)
Kucbel et al., Perspectives in Science (2016)
Querol et al., Atmospheric Environment (1996)
Ramsden and Shibaoka, Atmospheric Environment (1984)
Rösch, PhD thesis University of Leipzig (2015)
Steenari et al., Fuel (1999)
Sullivan et al., ACP (2009)
Umo et al., ACP (2015)
Welti et al., ACP (2012)
Wex et al., ACP (2014)
Whale et al. AMT (2015)
Wiedensohler, Journal of Aerosol Science (1988)
Zheng and Yamada, Talanta (2006)