We thank Referee #3 for his or her comments and constructive suggestions on how to improve the contents of the manuscript. The comments as posted are listed below on green font, our responses to them on red font and the specified modifications to the text on blue font.

Review of Manuscript acp-2021-111:

Particle emissions from a modern heavy-duty diesel engine as ice-nuclei in immersion freezing mode: an experimental study on fossil and renewable fuels, by Korhonen et al.

General comments:

Korhonen et al. perform systematic experiments about the ice nucleation ability of diesel engine particulate emissions at high relative humidity and mixed phase cloud relevant conditions, with auxiliary measurements about particle size distribution, hygroscopicity and chemical composition. The ice nucleation activity data and conclusion presented, demonstrating the poor ice formation ability of diesel engine particles and limited effects from photochemical ageing processes on the particle ice nucleation activity, are of interests to the ice nucleation community. However, some part of discussion and data interpretation are not thoroughly or comprehensively presented. I would like to suggest further revisions before recommending acceptance.

The ice nucleation experiments performed cover a variety of diesel engine emissions generated by three kinds of fuels. Different engine exhaust treatment techniques are used to mimic diesel engine particulate emission atmospheric relevant ageing processes. The research interests are of significance of the atmospheric ice nucleating particles and thus climate. An interesting ice nucleation story is clear. Nevertheless, some improvements need to be made, regarding to data interpretation and results discussion. In general, I have five major comments on this manuscript.

The authors should clarify their samples and research focus clearly and construct an unambiguous approach about the research story, illustrating the relations among all the measurements. For instance, the manuscript title and abstract tell me the particulate emission from a diesel engine will be the object for this experimental study, but the authors directly introduce soot particles in the introduction and the following parts. Note, the particulate emissions from diesel engines are not only comprised of soot particles. Differentiating the concept of soot particles from diesel engine particulate emissions is necessary. The authors also need to explain why soot particles emitted by land transportation diesel engines are atmospherically relevant.

We have added the following paragraph to the introduction, and consider that it clarifies the atmospheric relevance of the studied particles:

“Particulate emissions from diesel engines can form a notable fraction of total aerosol burden in urban areas, especially in regions where diesel-powered vehicles outnumber ones using different types of fuel (DeWitt et al., 2014). For instance, such regions can comprise arterial roads near seaports, transport hubs, or other facilities whose function relies on heavy-duty transport. In addition to impairment of air quality near the surface level, convective draft mixes the aerosol within the atmospheric boundary layer and can lift the particles to altitudes up to several kilometers, depending on weather conditions. Studies focusing on urban impacts on climate and weather show evidence of precipitation anomalies downwind from large cities (Han et al., 2012; Han et al., 2013;

Zhong et al., 2015). Hence, understanding the role of urban aerosol particles in cloud processes is essential for further understanding of the origins of the abovementioned anomalies in precipitation due to urban aerosol particles. However, the number of previous studies focusing on the ice nucleating potential of fossil and renewable diesel fuels (Schill et al., 2016; Chou et al., 2013) is limited, to our knowledge."

Second, I highly recommend to harmonise the ice nucleation terminology through the manuscript, see Vali et al. (2015) as a reference. The aim is to make dissemination more uniform and consistent within the community.

We agree on the importance of uniform terminology and explain why we refer to immersion freezing in the methodology section of the revised manuscript. Also reference to Vali et al. (2015) added. For instance, the following statement has been added to Sect. 2.2:

"The high relative humidity on water enabled investigation of the IN activity in immersion freezing mode as it is defined by Vali et al. (2015) because liquid formation on the sample particles is expected before possible ice formation takes place; immersion and condensation freezing modes are indistinguishable in the SPIN."

Third, the ice nucleation pathway, immersion mode freezing, is not well introduced. Only referring to a reference (Korhonen et al. 2020) without a brief introduction about how this can be achieved is not enough, in my opinion. Both the concept and the approach to achieve it should be explained even as a summary and then referring to the previous literature can be of more clarity and convincingness.

We acknowledge that the initial formulation required reading Korhonen et al. (2020) and have revised Sect. 2.2 in a way that it explicitly explains the behavior of droplets and ice crystals inside the SPIN, when the settings used in this study are applied. Besides, the reasoning for using the 4 µm size threshold in size-separation is now explained. The reasoning for the selection of the size threshold has been added to the text:

"Therefore, we used basic size-separation for ice crystals and droplets and consider particles larger than 4 µm ice crystals. Given the high relative humidity, 110% over water and 150-163% over ice (depending on lamina temperature during the T-scan), the formed ice crystals grow fast inside the SPIN to optical sizes much higher than the 4 µm threshold used, typically to a range between 7-15 µm."

Forth, a new approach to present normalized ice activation fraction results should be explained more clearly in Sect. 2 and 3. Also, it should be applied carefully. Note that, the particle samples are polydisperse with different particle size distributions (see Fig. 2) and the proportion of large particles (e.g. > 100 nm ~ 10 %) is still comparable or even higher than the highest ice activation fraction measured. For instance, the highest ice activation fraction for unaged RME fuel engine particles is less than 0.35 % even at the lowest temperature addressed, as shown in Fig. 6. To figure out the contribution of large particles (e.g. 100, 200 or 300 nm) to the ice activation fraction, ice nucleation experiments for size selected (e.g. 100, 200 or 300 nm) large particles are expected to performed. In addition, the data processing or the calculation method needs to be formulated and then the specific equation can help the understanding.

We have re-analyzed all SPIN data, and we have identified an artifact in the data processing. Whenever the SPIN OPC counts per second exceeds a certain threshold, then the SPIN data are stored in a slightly different manner, which is very simple to correct for. That correction was unfortunately not carried out for the ice-activated fractions (AFs) presented in figures 3 to 6 in the previous version of the manuscript. The correct AFs may be up to about 1 order of magnitude higher than what was previously presented for the very lowest temperatures around -41°C, while the differences in the AFs were relatively smaller or insignificant for temperatures above -39°C. Generally, these corrections do not affect the main findings of little to no indications of heterogeneous immersion freezing. Previously, we interpreted the pronounced low AF levels for several diesel samples near T=-41°C relative to the homogeneous freezing reference to be due to small and/or very hydrophobic diesel particles not activating into cloud droplets with the potential for subsequent immersion freezing inside SPIN. For that reason, we found that there was a risk of misinterpreting low AFs as a result of low immersion freezing ice nucleating ability – rather than an effect of very low CCN activity. That was the motivation for the AF scaling carried out. With the correctly analyzed AFs, there are still indications of low CCN activity influencing the AFs but to a significantly less pronounced level. We still think that it is of relevance to consider how high concentrations of nucleation mode particles and/or very hydrophobic seed particles may bias the immersion freezing AF low by contributing to the total particle concentration while not acting as CCN/INPs. However, we no longer consider it a risk that the 'raw' AFs can be misinterpreted in this context, so we have decided not to carry out the AF scaling and the figures 3.b to 6.b do not appear in the revised version of the manuscript. Consequently, the paragraphs in L215-225 and L273-282 have been deleted.

The main foci of the study were to (1) directly link the ice-nucleating ability of the studied particles to their physico-chemical properties, and (2) to investigate to which extent simulated photochemical aging potentially would influence the ice-nucleating ability of the particles. Hence, there were a number of constraints on the particle number concentration and particle mass that could be studied. It would typically not be possible to obtain reasonable number concentrations of 200 or 300 nm mono-disperse particles for the SPIN measurements. It is not uncommon to study poly-disperse particle populations with a CFDC as it is generally done in ambient measurements and a number of laboratory studies (e.g. Petters et al., 2009; Levin et al.,2016). Furthermore, the fact that we do not observe any indications of heterogeneous freezing for temperatures well above the homogeneous freezing reference, indicates a low ice-nucleating ability for any particle size of the studied population. Thus, with the experimental constraints and the scientific aims, it was not meaningful to carry out size-selected CFDC measurements, and it is highly questionable whether such activities would add any substantial additional and essential information regarding the low ice-nucleating ability of the studied particles.

Finally and most importantly, the results discussion is performed not thoroughly and reasonably. For example, comparing the ice nucleation results of polydisperse aerosol dominated by fine particles (< 100 nm) with the homogeneous freezing of larger (350 nm) ammonium sulfate (AS) particles is inappropriate. Instead of 350 nm AS particles, the comparison with the homogeneous freezing results of small (< = 100 or 150 nm) AS particles would be of more relevance to the results of diesel soot particles which has a small size distribution. Especially, the diesel engine particles already exhibit a homogeneous freezing depression event at temperatures lower than the homogeneous freezing temperature at such a high relative humidity (RH_w = 110 %). In addition, the

ice nucleation data is not well linked to the auxiliary measurement results. Similar findings in the literature are also helpful to support the conclusion (see detailed comments in next part).

It is not clear why the reviewer finds that it would make more sense to study e.g. 100 nm particles rather than 350 nm ammonium sulfate particles to obtain a homogeneous freezing reference, since no scientific arguments are provided supporting that point of view. In this context, it is relevant to point out that the critical supersaturation needed to activate a soot particle with a mobility diameter of 100 nm is much higher than the corresponding value for a 100 nm ammonium sulfate particle. So, we do not expect that to be the reason, and we cannot think of another reason to justify the argument.

The aim of the homogeneous freezing reference is simply to detect when dilute aqueous cloud droplets freeze. The chosen ammonium sulfate particle size should ensure that (1) as large a fraction as possible of the particles activate into droplets inside SPIN, and (2) the dissolved ammonium sulfate seeds in the cloud droplets do not depress the freezing point substantially.

We speculate, that for ammonium sulfate particles located in the vicinity of the lamina closest to the cold wall would contribute to droplet formation and freezing, with that contribution potentially increasing with the increasing particle seed size (associated with lower critical supersaturation). The freezing point depression is about 0.03 K for 4 µm droplets and 350 nm ammonium sulfate seeds following Ignatius et al. (2016). Hence, we do not expect the solute effect to substantially lower the freezing point, while a more than reasonable fraction of the particles is likely to contribute to the formation of dilute droplets. Hence, we consider 350 nm particles a reasonable choice in this context. We could of course obtain very similar spectra for e.g. 100 nm seed particles, but the comparability to measurements carried out years ago is questionable due to potential minor differences in the flow control and aspects similar to that.

Specific comments:

Line 17: change ‘continuous-flow diffusion chamber’ to the same as it is in Line 87

Done.

Line 19: change to ‘-43 and -32ºC’. The same for Line 43, 92, 96, 175, 234 and 268. Please check through the mathematical notation and make it satisfy ACP terminology.

Done.

Line 23: change ‘present’ to ‘presented

The sentence has been rephrased from:

“In addition to ice-nucleation experiments, we used supportive instrumentation to characterize the emission particles and present six different physical and chemical properties of them.

to:

"In addition to ice-nucleation experiments, we used supportive instrumentation to characterize the emitted particles for their physicochemical properties and presented six of them.

Line 24: make 'different emission after-treatment systems' specified

The text has been changed from:

"We found that the studied emissions were poor ice-nucleators and substitution of fossil diesel with renewable fuels, using different emission after-treatment systems and photochemical aging of total exhaust had only little effect on their ice-nucleating abilities."

to:

"We found that the studied emissions contained no significant concentrations of ice nucleating particles likely to be of atmospheric relevance. The substitution of fossil diesel with renewable fuels, using different emission after-treatment systems such as a diesel oxidation catalyst, and photochemical aging of total exhaust had only little effect on their ice-nucleating abilities."

Line 27: change to 'the radiative forcing of the Earth and thus climate in different ways'

Following the recommendations by all referees, the text has been changed from:

"Atmospheric aerosols affect the radiative forcing budget of the Earth and thus climate in multiple different ways, directly through absorption/scattering of radiation and indirectly through impacts on cloud properties."

to:

"Atmospheric aerosols affect the energy budget of the Earth and thus climate in different ways: directly through absorption and scattering of heat and light, respectively, and indirectly via affecting cloud formation and lifetime."

Line 39: change to 'homogeneous ice nucleation'

Done.

Line 41: change to 'ice nucleating particles' and specify its abbreviation 'INPs'

According to suggestion by Referee #4, we have rephrased the sentence from:

"but there is evidence that combustion emissions from different hydrocarbon fuels can have potential as active ice-nucleating particles in temperatures higher than that."

to:

"but there is evidence that combustion emissions from hydrocarbon fuels have the potential to nucleate ice at higher temperatures."

In the revised text, we introduce the abbreviation INP later on L44.

Line 39 to 41: Please provide evidence or reference to show combustion emissions are relevant to the lower troposphere ice nucleation activities.

We assume that Referee #1 has also pointed out the same aspect and have added Ikhenazene et al. (2020) and Thomson et al. (2018).

Line 44 and 45: If you write that soot particles are not active INPs, the relative humidity and temperature condition also need to be reported.

We have added mentioning that the reported soot particles were not active INPs at MPC-relevant temperature and humidity conditions.

Line 50: the reference 'Mahrt et al. 2018' should be irrelevant to the atmospheric aging processes for INPs but Mahrt et al. (2020a) and (2020b) can be references.

Removed Mahrt et al. (2018), suggested references added to replace it.

Line 58 and 59: change to 'the climate forcing due to anthropogenic soot particles immersion freezing'

Done.

Line 60: change to 'ice nucleation abilities'

Done.

Line 72 to 74: The environmental pollution caused by diesel engine without DPF or DOC technique is not relevant to this research topic.

Following the suggestion by Referee #1, we have revised the entire paragraph to be more concise and more focusing on 1) the atmospheric relevance of diesel emissions and 2) the motivation for ice nucleation studies, such as this one. Please check the revised chapter where this statement is corrected.

Line 129 to 131: was the Aerosol Instrument Manager (AIM) software used to log the SMPS data? If so, the SMPS scan size upper limit should be much larger than 500 nm with such a high sheath to aerosol sample flow ratio (10:1) and a 180 s scanning time. And if the size scan did not cover the whole range of the aerosol particle size distribution, the multiple charge correction is not finished and then the results are biased by the uncomplete correction calculation.

Yes, we used the AIM software. The parameters describing the SMPS settings in the original manuscript were unfortunately wrong, we deeply apologize for this. The nominal flow rates used in the majority of the campaign was 5 lpm sheath and 1 lpm aerosol flow rate. The size range was 11-500 nm as stated previously. The number fraction of particles above 500 nm in the soot mode was according to the lognormal fits less than 0.5%. This means there were only very small uncertainties from the multiple charge correction. There is some minor noise in the channels 450-500 nm for some of the experiments as seen in figure 2. These are not from multiple charge correction. The signal is extremely low at these channels it is unclear where this noise comes from.

Line 130 to 142: Better to introduce the measurements work flow following the sample flow sequence depicted in Fig. 1.

We have revised the chapter to depict the work flow better.

Line 179 to 181: The authors need to make a more conceivable and clear statement for distinguishing water droplets from ice crystals. I understand that the basic idea is to let the OPC running in different size channels and then to differentiate the particle phase according to their survival abilities through the evaporation section, i.e. water droplet can be evaporated because of the relative humidity condition. The statement about CCN ability and immersion mode freezing make readers confused. In addition, referring to the study performed by Korhonen et al. (2020) as an example does not make sense for me. This is because the samples are different between the current study (i.e. diesel engine particulate emissions) and the previous study (i.e. particulate emissions from solid-biomass-fired cookstoves). The OPC channel size used to discriminate water droplets from ice crystals should be stated from the current study.

The reference to Korhonen et al. (2020) relates to the experimental approach and associated advantages and limitations. The approach can be applied in general for any type of particle studied, so we disagree with the reviewer comment in this context, and it would be bad practice not to let readers know where the relevant information about the instrument operation was introduced.

The statement:

"The main motivation for this operation procedure is 1) to ensure that a significant number of particles with low CCN activity form droplets inside SPIN, and 2) that immersion freezing over a wider temperature range can be investigated."

has been modified to:

"In the present study, the main motivation for this operation procedure was 1) to ensure that particles with low CCN activity form droplets inside SPIN due to a relatively high supersaturation, and 2) that immersion freezing over a wider temperature range can be investigated within a single run."

Using size to discriminate between ice and non-ice is fundamental in CFDC studies, and widely applied. The SPIN OPC is advanced and not limited to discrete channels. The ice size threshold has been changed from 6 to 4 µm, which was clearly stated in the previous as well as the revised manuscript.

Line 190: change to 'exiting the IN chamber' or 'exiting the SPIN'. Or, the authors can decide to use 'SPIN' or 'the SPIN' through the whole manuscript.

Done. We have decided to use form 'the SPIN' throughout the text.

Line 215 to 225: In this paragraph is not well organised. In my point of view, the authors may need to explain how the freezing of a particle immersed in a water droplet could happen when the temperature decreases lower than the homogeneous freezing temperature (HNT), to illustrate the results presentation. A suggestion could be that sample particles might be activated as cloud

droplets at RHw = 110 % for temperature conditions higher than the HNT, thus makes it possible to investigate the particles immersion mode freezing ability at T < HNT in the flowing temperature scan because a droplet would freeze homogeneously when T is lower than HNT. Here again, the ice crystal formation of droplet activated particles at T < HNT should be homogeneous freezing. If the authors claim this is immersion freezing, evidence of this should be presented. But if the freezing occurs at RH conditions above homogeneous RH condition at the same T, then it is unclear how the authors can conclude immersion freezing to be the relevant mechanism.

It seems as if the reviewer is of the impression that there is a single homogeneous freezing temperature (HFT) determining whether a droplet will freeze or not. We do not share that point of view, and in the following we will describe why the observed ice spectra are very much in line with what can be expected from homogeneous freezing.

Homogenous freezing is generally expected to be explained by classical nucleation theory (Ickes et al., 2015). Hence, the freezing probability of a droplet is a function of temperature, time, and droplet size. In addition, solutes may depress the freezing point as described further above.

Inside SPIN, the aerosol sample located in the lamina will be exposed to a range of temperatures, supersaturations and different residence times in the lamina due to the non-uniform flow profile.

It is not straightforward, which droplet size and corresponding residence time of such a droplet in the growth part of the SPIN chamber that should be applied in the calculations. Ignatius et al. (2016) estimated that inside the SPIN, a frozen fraction of 10% could be expected pure water droplets with a temperature between -38.2 and -37.6°C. In the figures 3-6, the average lamina temperature is shown on the ordinate axis. Hence, roughly 50% of the aerosol population can be expected to be exposed to either a lower or alternatively a higher temperature than the average lamina temperature depending on the respective particle trajectories inside SPIN. That, in conjunction with a higher freezing probability for lower droplet temperatures explain why the ice-activated fraction increases with decreasing average lamina temperatures in the temperature range from –41 to -38°C. So qualitatively, the ice spectra are in line with what can be expected for homogenous freezing.

When it comes to the quantitative aspects of the ice-activated fraction, then the AFs are lower than what we could expect from the theoretical considerations by Ignatius et al. (2016). As discussed in the present manuscript, we would expect the AFs to be biased significantly low due to only a fraction of the sample particles being focused in the lamina (Garimella et al., 2017). In addition, a few other effects may bias the ice crystal counts low, such as (i) the potential presence of ice crystals of sizes below 4 µm, and (ii) losses of ice crystals between the evaporation section and the OPC detection volume. Finally, the 'theoretical' calculations related to (i) homogeneous freezing conditions, and (ii) the lamina conditions inside SPIN are associated with some uncertainty. Hence, overall, we find a reasonable agreement between observations and what can be expected from 'theory' considering relevant errors and biases. Since most potential biases are likely to be comparable between samples studied with identical SPIN operation conditions, we consider the homogeneous freezing reference a reasonable reference for comparison within the present study.

Line 223 to 225: A clear definition for the normalization of the ice activation fraction curves for each sample should be made. A formulation for this approach or an example may help.

We have omitted this normalization method from the revised manuscript and present only the 'classical' method for calculating the AF in the text. Hence, the lines mentioned in the comment have been deleted.

However, we have added the INAS density normalization and present its formulation and results in supplementary information.

Line 232 and 233: change 'L/min' to 'L min^{-1}'. Please check the unit through the manuscript.

Corrected to form "L min^{-1}" throughout the revised text.

Line 235: The CCNC calibration curves should be provided in the following section or in an Appendix part.

We have included a figure with the CCNC calibration curves in the supplementary information. We also provide the necessary information regarding the figure.

Line 251: The calibration results should be provided in the following section or in an Appendix part.

We have included a figure with the CCNC calibration curves in the supplementary.

Line 265: What is the 'GMDs'?

The GMD means geometric mean diameter of the particles, we have defined the acronym in the revised text.

Line 285: change to 'Ice activation fraction curves for fossil diesel emissions are presented in Figs. 3 and 4'

Done.

Line 287: change to 'Fig. 3a'. And the similar suggestion to that of Line 298, 310, 314, 324, 326 and 327. Please check the abbreviation for 'Figure' through the manuscript.

Done.

Line 291: change to 'Fig. 3b'; Specify which two samples

Done.

Line 294: Here, what is the size range for the so-called ultrafine particles? It should be 100 nm if the number 90 % refers to the size distribution results mentioned in Sect. 3.1. Please make the ultrafine particle with a quantitative value for clear discussion.

Quantification added.

Line 300 to 307: The discussion in this paragraph can be better. First, the ice formation enhancement by lowering the ice onset temperature values should be clearly connected to the sample to make it

easier for readability. Second, some evidence form auxiliary measurements should be provided to interpret the results. Also, relevant studies in the literature can be referred to for comparison, e.g. Zhang et al. (2020) also investigated the photochemical aging effects on soot particles ice nucleation activities at T < HNT.

The focus of that section is on the potential of heterogeneous immersion freezing. Hence, Zhang et al. (2020) is not a relevant study to consider in this context, since they studied deposition freezing.

The discussion concerning the ice-nucleation results from photochemically treated particles has been changed from:

"Photochemical aging was found to have little effect on ice-activity, as Fig. 4 shows: for the whole sample population, the most ice-active case produced ice onset at approximately 0.5 ^{o}C higher temperature than homogeneous freezing. The normalization test revealed that the highest activity was only slightly higher than in unaged emissions (1.2 ^{o}C vs. 0.6 ^{o}C, relative to the homogeneous freezing reference), which indicates that the simulated photochemical aging through OH and O3 exposure has some, but only little effect on IN activity of fossil diesel emissions. The aged samples showed similar behavior in ice-activity relative to the unaged ones, with maximum activated fraction reaching up to 1.7% from whole sample population, regardless of the equivalent atmospheric aging time. Generally, the ice-activated fractions never reached above 1.7% in any experiment on polydisperse sample emissions from fossil diesel."

to:

"Photochemical aging was found to have little effect on the observed ice-activated fractions. It may be non-ideal to compare the ice-activated fractions directly between the PAM-processed samples versus the fresh emissions. The particle number size distributions may differ significantly between the samples as illustrated in Fig. 2, so the ice-activated fractions have been normalized to particle number concentrations representing different average particle sizes – and different hygroscopic properties. Nevertheless, we observe no indications of heterogeneous freezing more than about 1.0 ^{o}C above the homogeneous freezing in any experiment. Considering the potential errors and biases, we find it unlikely that the type of photochemical aging simulated in these experiments are likely to significantly improve the ice-nucleating ability of the engine emissions studied. However, that does not rule out that other types of atmospheric processing can be of importance for the ice-nucleating ability of diesel engine emissions."

Line 313: change to 'the lowest temperature'

Done.

Line 316 and 317: Arguing that the -36.1 ºC is outside of the instrument uncertainty should refer to the homogeneous freezing temperate detection ability of SPIN.

We agree, reference to that added.

Line 327 and 328: Why use the size distribution results about fossil fuel in Fig. 2a ('left-hand panel of Fig. 2' in text) to interpret the ice activation results of RME emissions?

We apologize the wrong reference to the figure, corrected to “Fig. 2c”.

Line 335 to 347: The discussion in this paragraph is too general and not specific enough. For example, the auxiliary measurement results for each sample should be connected to the sample directly, instead of making a general statement or a conclusion (e.g. Line 336 to 338 about CCNC results) for the overall study. The statements also should be clearly related to the quantitative values obtained from the supportive measurements. In addition, explanation or definition about each measurement result, e.g. OA, C_{11}/C_3, should be made in the main text. Necessary references in the literature also need to be referred.

We acknowledge this shortcoming and have added depth to the discussion. For instance, the analysis regarding the CCNC has been expanded. The statement:

“The CCNC experiments showed no CCN activity in any experiment without the PAM, which is a clear indicator of extreme hydrophobicity of the studied particles that was also observed in the SPIN as weak droplet formation despite high supersaturation RH_{water} =110 % during the T-scans. The PAM experiments where SOA formation on diesel emission particles took place increased the κ_α values to distinguishable levels, yet this increased hygroscopicity had no observed effect on ice-activity.”

Has been replaced with:

“Detection of significant concentrations of CCN for any of the bypass or PAM-TD experiments, even for the highest supersaturation approaching ~2.4%, was impossible. We conclude that the associated particles were hydrophobic. For the PAM experiments, we observed droplet formation, and it was possible to infer the CCN activity. For the PAM experiments related to different fuels, the 58 nm particles had kappa values of 0.08-0.10, the 107 nm particles had kappa values of 0.04-0.08, while they were about 0.01 for the 196 nm particles. The number concentrations for the 296 nm particles were too low for the CCN measurements. An increase in the κα when the aerosol is exposed to photochemical ageing can be due to (i) formation of SOA, (ii) oxidation of the soot surface, and (iii) collapse of fractal soot aggregates (Tritscher et al., 2011). Gren et al. (2021) reported the effective density to increase significantly for the very same particles after sampling through PAM, which is indicative of significant SOA coatings which appeared to be relatively more pronounced for the 58 and the 107 nm particles. In combination with the dramatic increase in κα from undetectably low to levels of up to 0.1, formation of SOA is likely to be the dominant process. The kappa for SOA formed from diesel engine exhaust has been reported in the range 0.09 to 0.14 (Tritscher et al., 2011). Following the approach described by Wittbom et al., (2014) about the κα of aged diesel soot particles, our observations indicate that the aged 58 nm particles were likely to be dominated by SOA by volume, while the 107 nm particles were likely to be soot particles with a significant SOA coating. The more modest kappa values for the 196 nm are indicative of a relatively lower ratios between the SOA and the primary soot particle volumes.”

We would expect the increase in κ_α and associated increase in particle volume due to the PAM processing to increase the fraction of particles activating into cloud droplets inside SPIN. Hence, that would increase the number of particles potentially contributing to immersion freezing. On the other hand, the PAM processing clearly leads to pronounced new particle formation with modes centered near ~20 nm (Fig. 2). It is questionable to which extent such particles (i) act as CCN inside SPIN, and (ii) subsequently may play a role as INPs in immersion freezing mode. Hence, the ice-activated fractions observed with SPIN may get biased high for the PAM-processed aerosol due to a larger

fraction of soot particles being immersed in cloud droplets, or the activated fraction may get biased low due to the pronounced increases in nucleation mode particles, which are unlikely to contribute significantly to immersion freezing as studied with SPIN. These observations illustrate that it is complicated to directly compare the AF between these samples with substantially different κ_α and particle number size distributions.

Line 348: change to 'Summary and conclusion'. Because the discussion is largely presented in previous sections and this part is more about conclusions.

Done.

Line 349 to 368: I disagree with the logicality in this part. On the one hand, the authors conclude that small diesel engine particles have no contribution to ice nucleation activities (Line 361). On the other hand, they are comparing the ice nucleation ability of the particles produced by different fuels. The authors need to firstly demonstrate ice nucleation activity via the immersion mode really occurs then they can make statement about the efficiency of the soot particles as potential ice nucleating particles (INPs). The reference about the study presented by Kanji et al. (2020) in Line 354 is inappropriate, which states that their findings are in complete agreement with Kanji et al. (2020).

It is not possible to discern between condensation freezing and immersion freezing with typical CFDC measurements. However, as described above, the homogeneous freezing reference agrees well with what could be expected from homogeneous immersion freezing considering potential errors and biases. In addition, we have replaced the statement including the reference to Kanji et al. (2020) from:

"Besides, our results are in complete agreement with Kanji et al. (2020) who studied whether hydrocarbon soot from propane combustion and different commercially available black carbon particles can induce immersion freezing and found all of them inefficient ice-nucleators."

to:

"Our results, in conjunction with a lot of previous studies indicate that a wide range of black carbon and soot particle types are inefficient as immersion freezing INPs"

Figures and Tables:

Figure 1: I cannot find where the 'FPA-fast particle analyser' is in the figure. It is not mentioned in the main text, either.

The referee is correct that no FPA data was used in our analysis. We have removed the mentioning from the caption.

Figures 2: The size distribution measurement for 'engine-out + BP' sample presented in Fig. 5 is missed in Fig. 2b and should be provided. And there is no SPIN experiment corresponding to the sample 'DOC + PAM' in Fig. 2b. In addition, it would be helpful if the figure grids are on to guide reader's eyes.

Upon revision of the calculation method, the panel b of Figs. 3-6 has been omitted from the revised manuscript. Therefore, this comment is no longer of relevance.

Figure 3: Is the ice activation curve for ''Engine-out + BP' sample normalized by the sample approach as those of other samples? The highest ice activation fraction should be the unity. It looks in corrected.

Upon revision of the calculation method, the panel b of Figs. 3-6 has been omitted from the revised manuscript. Therefore, this comment is no longer of relevance.

Added References:

Garimella, S., Rothenberg, D. A., Wolf, M. J., David, R. O., Kanji, Z. A., Wang, C., Rösch, M., and Cziczo, D. J.: Uncertainty in counting ice nucleating particles with continuous flow diffusion chambers, Atmos. Chem. Phys., 17, 10855–10864, https://doi.org/10.5194/acp-17-10855-2017, 2017.

Gren, L., Malmborg, V.B., Falk, J., Markkula, L., Novakovic, M., Shamun, S., Eriksson, A., Kristensen, T.B., Svenningsson, B., Tunér, M., Karjalainen, P. and Pagels, J., Effects of renewable fuel and exhaust aftertreatment on primary and secondary emissions from a modern heavy-duty diesel engine, J. Aerosol Sci., 156, 105781, 2021.

Ickes, L., Welti, A., Hoose, C., and Lohmann, U.: Classical nucleation theory of homogeneous freezing of water: thermodynamic and kinetic parameters, Phys. Chem. Chem. Phys., 17, 5514–5537, 2015.

Ignatius et al.: Heterogeneous ice nucleation of viscous secondary organic aerosol produced from ozonolysis of a-pinene, Atmos. Chem. Phys., 16, 6495–6509, https://doi.org/10.5194/acp-16-6495-2016, 2016.

Ikhenazene, R., Pirim, C., Noble, J. A., Irimiea, C., Carpentier, Y., Ortega, I. K., Ouf, F.-X., Focsa, C., and Chazallon, B.: Ice Nucleation Activities of Carbon-Bearing Materials in Deposition Mode: From Graphite to Airplane Soot Surrogates, J. Phys. Chem. C, 124, 489–503, https://doi.org/10.1021/acs.jpcc.9b08715, 2020.

Levin, E. J. T., McMeeking G.R., DeMott, P.J., McCluskey C.S., Carrico C.M., Nakao S., Jayrathne T., Stone E.A., Stockwell C.E., Yokelson R.J. and Kreidenweis S.M., Ice-nucleating particle emissions from biomass combustion and the potential importance of soot aerosol, J. Geophys. Res. Atmos., 121, 5888–5903, doi:10.1002/2016JD024879, 2016.

Thomson, E. S., Weber, D., Bingemer, H. G., Tuomi, J., Ebert, M., and Pettersson, J. B. C.: Intensification of ice nucleation observed in ocean ship emissions, 8, 1111, https://doi.org/10.1038/s41598-018-19297-y, 2018.

Tritscher, T., Jurányi, Z., Martin, M., Chirico, R., Gysel, M., Heringa, M. F., DeCarlo, P. F., Sierau, B., Prévôt, A. S. H., Weingartner, E., and Baltensperger, U.: Changes of hygroscopicity and morphology during ageing of diesel soot, Environ. Res. Lett. 6, 034026, 2011.

Wittbom, C., Eriksson, A. C., Rissler, J., Carlsson, J. E., Roldin, P., Nordin, E. Z., Nilsson, P. T., Swietlicki, E., Pagels, J. H., and Svenningsson, B.: Cloud droplet activity changes of soot aerosol upon smog

chamber ageing, Atmos. Chem. Phys., 14, 9831–9854, https://doi.org/10.5194/acp-14-9831-2014, 2014.

Suggested References:

Kanji, Z. A., Welti, A., Corbin, J. C., and Mensah, A. A.: Black Carbon Particles Do Not Matter for Immersion Mode Ice Nucleation, Geophys Res. Lett., 47, https://10.1029/2019gl086764, 2020.

Korhonen, K., Kristensen, T. B., Falk, J., Lindgren, R., Andersen, C., Carvalho, R. L., Malmborg, V., Eriksson, A., Boman, C., Pagels, J., Svenningsson, B., Komppula, M., Lehtinen, K. E. J., and Virtanen, A.: Ice-nucleating ability of particulate emissions from solid-biomass-fired cookstoves: an experimental study, Atmos. Chem. Phys., 20, 4951-4968, http://10.5194/acp-20-4951-2020, 2020.

Mahrt, F., Alpert, P. A., Dou, J., Gronquist, P., Arroyo, P. C., Ammann, M., Lohmann, U., and Kanji, Z. A.: Aging induced changes in ice nucleation activity of combustion aerosol as determined by near edge X-ray absorption fine structure (NEXAFS) spectroscopy, Environ. Sci.: Processes Impacts, https://10.1039/c9em00525k, 2020b.

Mahrt, F., Kilchhofer, K., Marcolli, C., Grönquist, P., David, R. O., Rösch, M., Lohmann, U., and Kanji, Z. A.: The Impact of Cloud Processing on the Ice Nucleation Abilities of Soot Particles at Cirrus Temperatures, J. Geophys. Res. Atmos., 125, 1-23, https://10.1029/2019jd030922, 2020a.

Vali, G., DeMott, P. J., Möhler, O., and Whale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmos. Chem. Phys., 15, 10263-10270, https://10.5194/acp-15-10263-2015, 2015.

Zhang, C., Zhang, Y., Wolf, M. J., Nichman, L., Shen, C., Onasch, T. B., Chen, L., and Cziczo, D. J.: The effects of morphology, mobility size, and secondary organic aerosol (SOA) material coating on the ice nucleation activity of black carbon in the cirrus regime, Atmos. Chem. Phys., 20, 13957-13984, https://10.5194/acp-20-13957-2020, 2020.

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