

We thank Referee #4 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**.

The authors have submitted an article titled: Particle emissions from a modern heavy-duty diesel engine as ice-nuclei in immersion freezing mode: an experimental study on fossil and renewable fuels. The article describes an ice nucleation study with combustion exhaust aerosol ice nuclei. The authors used 3 types of fuel and a diesel engine for aerosol generation. The polydisperse aerosol was introduced to a continuous-flow diffusion chamber at a constant RH while ramping the temperature between $-43\text{ }^{\circ}\text{C}$ and $-32\text{ }^{\circ}\text{C}$. Reference aerosol and 2 processing steps of the engine exhaust after-treatment system were intercompared. In addition, the impacts of different atmospheric processing and ageing steps on ice nucleation activity were evaluated. A range of complementary measurements were taken to characterize the aerosol and explain the IN observations. The authors report overall a poor ice-nucleation performance for all the different aerosol types, after-treatment systems, and photochemical aging with minor differences between the 15 experiments. Overall, the article fits within the scope of the journal and has the potential to have environmental relevance. The authors used state of the art instrumentation and decent experimental planning however, there are few points that would need to be addressed before this manuscript is published.

General comments

The connection between the selected diesel engine and the atmosphere is not established, not in the introduction, nor in the discussions of the results. The relevance of the specific diesel engine and these fuel types globally is further diminished in the paper. This undermines the justification of the selected MPC conditions for the experiments and the global relevance of this study. Consequently causing vagueness in the conclusions.

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. 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. Understanding the potential changes in atmospheric INP budget due to altering anthropogenic emissions, such as ones reported by Klimont et al. (2017) in this context, is essential for a further understanding of processes that may affect the climate change.”

The manuscript lacking substantial information, some of which is split between several other publications e.g. setup details, characterization of the aerosol, size distributions, laminar flow uncertainty etc. (more details in minor comments).

We acknowledge this structural shortcoming of the initial submission and have largely revised the text. Please see our responses to the specific comments.

The selected threshold of 6 micron for ice classification and calculation of activated fraction needs further clarification. This selection and the new approach for calculations of activated fraction are not sufficiently discussed (see minor comments).

The following revisions to address this shortcoming are done:

- 1) As we investigated the data again and found that the diesel emission particles exhibit no ice activity at sample temperatures greater than $-35\text{ }^{\circ}\text{C}$ even with lower threshold sizes for separating the ice crystals from droplets. After this discovery, we have revised re-analyzed the ice-nucleation data.
- 2) We have revised Sect 2.2 (methodology: ice nucleation experiments) to explicitly explain the reasoning behind the choice of $4\text{ }\mu\text{m}$ size threshold I the revised text: please see our responses to the minor comments.

To address the new approach as named in the comment, the following procedure has been performed to address this: We have re-analyzed all SPIN data, and we have identified an 'artifact' in 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 $-41\text{ }^{\circ}\text{C}$, while the differences in the AFs were relatively smaller or insignificant for temperatures above $-39\text{ }^{\circ}\text{C}$. 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 $T=-41\text{ }^{\circ}\text{C}$ 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.

Given the comprehensive list of instruments used, one would expect to see a deeper analysis. For example, shape factor, effective density, composition accompanied with figures or histograms, intercomparing aerosol properties in this study with other studies or IN activity of similar particles

in previous studies that were mentioned here. This additional information would help to expand the discussion on the observations.

The main focus of the current study is the potential contribution of diesel exhaust emissions to immersion freezing. Gren et al. (2021) report a lot of the suggested physico-chemical particle properties of the very same samples. The presentation and discussion of the CCN results have been expanded in the revised manuscript.

Grammar: in particular sentence structure, fluency, and connection between sentences require significant revisions.

We have revised the text following the comments by all referees and consider the contribution to this comment completed that way.

Specific comments:

LN2: "an experimental study" I suggest to change to a laboratory study for clarity

Done.

Ln18: "and" change to "while the..."

Done.

LN19: The tested fuels i.e., EN 590 compliant low-sulfur fossil diesel, hydrotreated vegetable oil (HVO), and rapeseed methyl ester (RME), all were used without blending.

Done.

LN23: "...emitted particles for their physicochemical properties".

Done.

LN23: "We found that the studied particles were poor ice nuclei. The substitution...."

Done.

LN27: First sentence: reword, don't use "/", what are "impacts on cloud properties" - the sentence is not clear

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.”

LN28: change to “Direct effects can be monitored....”, also provide some examples of direct effects

Added aerosol optical thickness and light scattering and absorption as examples.

LN29: “instruments” – provide examples and/or references

Done. We have added sun photometer and lidar as examples of instruments which monitor direct aerosol effects. References to instruments also provided.

LN30: “due to complexity of the processes that contribute to their final effects” – sentence not clear, reword.

The text has been changed from:

“due to complexity of the processes that contribute to their final effects”

to:

“due to complexity of the processes within the clouds that contribute to the total effect”

LN33: “Furthermore, most precipitation events...” - I couldnt find support for this definitive statement in the reference you provided.

We have deleted the statement and the reference from the revised text.

LN38: “than those required”

Done.

LN39: “Particles that are active ice nuclei...”

Done.

LN41: “hydrocarbon fuels have the potential to nucleate ice at temperatures higher..”

Done.

LN41: “than that” – specify what “that” refers to

The two comments above: 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.”

LN43: you haven't defined “INP”

Corrected.

LN44: “on the contrary” – on the other hand?

Corrected.

LN50: add Zhang et al., 2020 to the reference list

Done.

LN50: “these studies taken altogether...” – sentence needs rewording e.g. “The studies mentioned above demonstrate the challenge in associating soot INP properties to the ambient soot particle population”.

Sentence corrected as suggested.

LN53: abilities change to activity

Done.

LN54: “Multiple studies such as” – you listed only one study

The text has been changed from:

“The inconsistency in reported observations on soot IN activity implies that it is unclear which types of soot are efficient INPs, and it has been addressed by multiple studies such as Bond et al. (2013).”

To:

“The inconsistency in reported observations on soot IN activity implies that it is unclear which types of soot are efficient INPs, and it has been addressed by multiple studies such as Hoose and Möhler (2012), and Bond et al. (2013).”

We consider that Hoose and Möhler (2012) present an adequate review of soot-IN studies and their results, both positive and negative.

LN54: “In addition to that” - this sentence doesn't add to the previous sentence. Wrong conjunction.

Conjunction changed to “besides”.

LN55: “ (IPCC) have identified gaps in our knowledge of ice nucleation activity of soot in their...”

Corrected.

LN56: This uncertainty? You haven't mentioned uncertainty, specify what uncertainty you mean

Please see our response to the comment on LN57.

LN56: “..uncertainty reflects to available parameterizations estimating the IN ability of the soot, causing them to range several orders of magnitude” – the context of this sentence is not clear, reword.

Please see our response to the comment below.

LN57: “Consequently, it leads” – what “it” refers to? clarify in the text

We have revised the two sentences mentioned. The text has been changed from:

“This uncertainty reflects to available parameterizations estimating the IN ability of the soot, causing them to range several orders of magnitude (Vergara-Temprado et al., 2018). Consequently, it leads to...”

to:

“The wide range in the reported immersion freezing ability of soot particles is reflected by the available parameterisations spanning several orders of magnitude (Vergara-Temprado et al., 2018).” Consequently, the variation leads to...”

LN58: “challenges when the potential...” – rephrase e.g. “high uncertainty in estimation of the radiative forcing via modeling”

Rephrased as suggested.

LN69-71: reword the sentence

The text has been changed from:

“The introduction of the advanced after-treatment systems, such as the DPF and diesel oxidation catalyst (DOC), have led to a decline of 20-65% in black carbon (BC) emissions from heavy-duty road diesel engines in developed markets between 1990-2010 (Klimont et al., 2017). “

to:

“Klimont et al. (2017) reported a decline of 20-65% in black carbon (BC) emissions from heavy-duty road diesel engines in developed markets between 1990-2010, during the period when advanced emission after-treatment systems, such as the DPF and diesel oxidation catalyst (DOC), became mandatory in new diesel-powered road vehicles.”

LN72: globally still widely used – reference? Can you provide global estimates that will support the relevance of your study?

Please see our response to the comment on LN74.

LN73: “long time” - please support this qualitative projection with a reference

Please see our response to the comment below.

LN74: “still about to maintain its global popularity for decades” – reference for this forecast?

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 the commented aspects are either removed or corrected.

LN75: I think there is a missing paragraph here that connects between diesel combustion emissions at ground level and how they reach and interact with atmospheric humidity and temperature to form clouds, what's their known fraction at different altitudes etc. In what clouds they are most predominant to support your choice of temperature range. Perhaps connection to airborne measurements of combustion emissions and their ability to nucleate ice at altitude detected in flight e.g. Brown, 2018.

The following paragraph has been added to the paragraph describing the effects of diesel emissions:

“This diesel vehicle exhaust is emitted into the planetary boundary layer (PBL) close to the ground level. From mid to high latitudes, low-level mixed-phase clouds may be present within the PBL (e.g. Gierens et al., 2020) and can potentially get influenced by diesel vehicle emissions. Alternatively, soot particles can be transported to higher altitudes and over long distances (Storelvmo, 2012), so they can potentially influence clouds far from the source region. Soot particles have been identified in ice crystal residues in mixed-phase clouds (Cozic et al., 2008; Ebert et al., 2011), and thus it is of relevance to study their potential to facilitate immersion freezing.”

LN76: the sentence is not clear and is it relevant to this study?

Please check the description of the entirely revised paragraph commented in the comment on LN74.

LN78: diesel combustion emissions

Changed.

LN79: “among other factors, remains less studied” – what other factors and why it's less studied?

We have revised the paragraph as a whole (see our response to comment on LN74): in this revised text, the mentioning of other factors has been removed and the following sentences explain why there are fewer studies on immersion-freezing INP potential of diesel emissions.

LN84: “as well as they impact” – reword

We have moved the description of HVO and RME fuels to Sect 2.1, following the recommendations to improve the structure of the manuscript.

LN91: “temperatures of -35 °C and -30 °C. No immersion freezing...”

Done.

LN93: remove ice-nucleating potential

Done.

LN104: alternatives in the near future

Corrected.

LN104: “it is likely that heavy-duty diesel engines will be in use further than that” – here you say likely in LN73 you were much more decisive

We have removed the statement on LN73 due to more comprehensive revision of the entire chapter, please see our response to comments regarding lines 72-74.

LN106: “We investigate...” – reword this sentence

The text has been changed from:

“We investigate the immersion freezing ability at temperatures down to where homogenous freezing dominates, to detect potentially low ice-nucleating abilities.”

to:

“Our ice nucleation experiments aimed in detection of potentially low ice nucleating abilities in immersion freezing mode, down to temperatures where homogeneous freezing starts to dominate.”

LN111: did you monitor the temperature and humidity in the sampling line, if so, where? Please add to figure 1. Did you monitor pressure and airflow in the different sections of your setup? Would the high concentration cause sedimentation and narrowing of inner tubing diameter? Did that affect your measurements during the experiments?

It is very expectable that the sample temperature has been equal to room temperature after PTD and ED dilution by a factor of 1:600-1200 in total, prior to entering any instruments. Moreover, we used ¼” stainless steel tubing in our set-up between the last ED and the SPIN which should further assure that the sample temperature has been equal to room temperature in the SPIN experiments. Given the total sample flow rate of several liters per minute, we expect no gravitational sedimentation for submicron particles inside the tubing.

LN112: “The test engine used was a six-cylinder inline Scania D13 heavy-duty diesel engine” - why this engine was selected? How representative it is of diesel engines in the world? You should provide few more details to establish how this experiment will provide conclusions relevant to real world (outside the laboratory).

The Scania D13 series comprises commercial and industrial diesel engines ranging 270-390 kW in power output, which is a typical size for modern diesel engines used in heavy road vehicles and we consider it a representative option for testing diesel engine emissions.

LN115: “approach” – perhaps setup or configuration?

Changed to “configuration”.

LN116: what is one full temperature scan? How many repetitions did you do to test repeatability?

With one full temperature scan we mean scanning over chosen freezing conditions at constant RH on water (in this study $RH_w = 110\%$), over a temperature range that was -43 to -32 °C in all experiments – this is later quantified on lines 180-181 in the revised text. Terms ‘temperature scan’ or ‘T-scan’ are commonly used in the CFDC community for this scan-type experimentation.

LN122: “were cooled down” – did you monitor the temperature? Where? How?

We used PTD+ED cooling with a total dilution ratio of 1:600-1200, using dry compressed air as diluter. Therefore, we consider that such a high dilution ratio ensures a significant cooling of the sample aerosol at any rate.

LN124: PAM – reference for the instrument?

The following details about the PAM reactor used in this study have been added to the text:

“The PAM reactor used in this study consisted of a 13 L steel chamber containing two Hg lamps with peak intensities at 185 and 254 nm. The UV light generates ozone and hydroxyl radicals (OH) that oxidize the aerosol as it moves through the chamber. The flow rate through the PAM was controlled to 5–7 L min⁻¹. The same UV light intensity was used in all experiments. The aging corresponded to 4.8 ± 2.6 days assuming an average OH concentration of 1.5×10^6 molecules cm⁻³ (Mao et al., 2009). Extensive SOA formation occurred in the PAM for all engine out experiment, while less SOA was formed in measurements after the DOC. The thermodenuder (Aerodyne Inc.) was held at 250 °C in all experiments where it was used.”

LN127: $RH < 10$, where and how it was measured?

Given the high dilution ratio of 1:600-1200 with dry (dew point -40 °C) compressed air, it can be expected that the dew point of the sample flow downstream of the last ED has a RH_{water} way below 10%.

LN129: “The latter scanned continuously” - you mentioned only SMPS so not clear what is the “latter”.

We have replaced “The latter” with “The SMPS”.

LN130: “The size range of SMPS...” - The mobility diameter sampling range of the SMPS was set between 11 and 500 nm with automatic multiple charge correction in the software.

Corrected as suggested.

LN131: CCNc-100 ? one column?

Yes, that was the instrument we used. the following change has been made from:

“(CCNC, Droplet Measurement Technologies)”

to:

“(CCNC, Droplet Measurement Technologies, CCN-100)”

LN138: remove Moreover

Done.

LN142: RH<5, in line127 you said RH<10, was it measured?

The *RH* value on line 127 means the situation downstream of the second ejector dilution. There was an additional molecular sieve dryer upstream of the CFDC, which dried the sample aerosol further to <5% *RH*. This statement is based on the calibration of 60cm mol sieve dryer (marked as ‘dryer’ in Fig. 1), according to which a freshly regenerated desiccant material (13x mol sieves) is capable of drying room air ($T = 20\text{ }^{\circ}\text{C}$, $\text{RH} = 40\text{-}50\%$) down to dew point of $-40\text{ }^{\circ}\text{C}$ with air flow rate of 2.05 SLPM (the SPIN 1 + CPC 1.05).

LN148: “A low level of exhaust gas recirculation (EGR) setting was used, 18% oxygen on intake air to the combustion cylinder” - why these settings were used? Are they common settings?

To limit the formation of NO_x emissions, different levels of EGR are applied depending on engine operating conditions. 18% oxygen is representative for the operating conditions used during the experiments.

LN171: remove in the orders of

Done.

LN184: “Thus, we consider observed particles larger than $6\text{ }\mu\text{m}$ ice crystals” - what about ice smaller than $6\text{ }\mu\text{m}$? For example, if you have pore filling happening as described. in Mahrt et al. 2018, where they chose $1\text{ }\mu\text{m}$ size threshold to detect ice crystals in their chamber. How would such shift in this threshold affect your results?

Following the comment by Referee #2 about the same issue, we 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 $6\text{ }\mu\text{m}$ size threshold in size-separation is now explained.

LN199: “such as particle losses at the laminar flow are discussed in detail by Korhonen et al. (2020)” - are discussed in Korhonen (2020) but how do you address these deviations from laminarity in your

study? or any of the other issues discussed in the conclusions of Garimella et al 2017. If you dont, how much uncertainty it introduces to your results? are they still valid despite those known issues of SPIN?

Korhonen et al. (2020) discusses the operation of the very same SPIN unit in a manner similar to this study (T scans), thus it can be expected that the systematic errors are unchanged between these two sets of ice-nucleation measurements. In the SPIN, the particle losses due to lamina spreading are a well-acknowledged, yet not well-constrained issue as it is concluded in both Korhonen et al. (2020) and Garimella et al. (2017). Therefore, we prefer not to speculate and introduce arbitrary correction factors to the ice-nucleation data of this study. Given the high experiment reproducibility as reported by Korhonen et al. (2020), we consider the results of this study valid.

LN201: 15 experiments – did you do any repetitions?

Yes, we did whenever the pre-planned operation cycle of the engine allowed to repeat the upward T scan without changing any sampling parameters. For instance, the reported fossil diesel + DOC + bypass and HVO + DOC + bypass runs, two of each, are experiment repetitions.

LN202: “SPIN, and polydisperse” – split into 2 sentences

Done.

LN220: switching back and forth between CFDC and SPIN, stick with one naming for the instrument

We have revised the naming as follows: before introduction of the ice nucleation measurements we refer to CFDCs in more general sense, after that the SPIN is always mentioned as the name of the unit.

LN220: “We expect this particle fraction to be dominated by the larger particles, since they are more likely to act as CCN” – what about the doubly charged particles selected with DMA, how they affect this measurement?

The comment only relates to the poly-disperse aerosol samples studied with SPIN. Hence, there was no selection with a DMA prior to those SPIN measurements.

LN223: “The normalization was calculated via averaging the five highest observed ice-activation...” – do you expect to measure only one nucleation mechanism in these temperatures e.g. Marcolli 2014?

Yes. We expect homogeneous freezing of formed droplets at lamina temperatures significantly below homogeneous freezing temperature that was empirically observed to take place at -38.9 °C in the SPIN.

LN237: “In many cases, the supersaturation did not suffice to activate the studied particles, and results are presented only when a full CCN spectrum could be identified” - do these "many cases" have impact on the setting of 6µm threshold for ice in SPIN, which was set mostly because of droplets?

The ice size threshold is determined from the SPIN data analysis and has in the revised manuscript been changed from 6 to 4 μm . We apply the same ice size threshold for the homogeneous freezing reference and the other samples, and the measurements with the CCN counter did not play a role in this context.

LN243: “to elemental carbon (EC) concentrations” – how was EC measured?

The following text has been added:

“The EC was assessed by sampling of undiluted exhaust on quartz filters (Pallflex Tissuquartz, 47 mm), thermal-optically measured according to EUSAAR_2 protocol (Cavalli et al., 2010).”

LN253: “was calculated from SP-AMS and aethalometer data” – what is the propagated uncertainty combining the measurements for calculation of this ratio?

With respect to the accuracy of this ratio, we estimate approximately 50% (95% CI). However, the precision, which is more important for our study as we seek correlations between ice nucleation ability and other physicochemical parameters, is much better. Yet, upon revision, we find that the third digit in the OA/eBC ratio is indeed influenced by noise and hence, we have removed it. It is our assessment that noise did not influence the ordering of these samples and that the lack of correlation with ice nucleation abilities is not due to noise in AE33 and SP-AMS data.”

LN262: “Gren et al. (2021) present a more comprehensive description of the particle size distributions” - There are references to at least 3 other papers that contain substantial information about this study and the reviewer/reader needs to read these to understand this paper e.g. Gren et al. 2021, Kristensen et al. 2020 Korhonen et al. 2020. Some of the information specific to this experiment is missing and needs to be added either as main text, as appendix, or supplementary material.

We acknowledge this issue and have decreased the dependency on other papers using the following actions: First, the ice nucleation pathway inside the SPIN is explained in the Sect 2.2 of the revised manuscript, in a way that it is independent from Korhonen et al. (2020). Second, we have added a deeper discussion on the results supporting the ones from ice nucleation, in a way that understanding their relevance to ice-nucleating abilities of the particles studied does not require reading the abovementioned papers.

LN268: “significant fraction” – how would you quantify this statement?

It is not straightforward to quantify that effect, since it is particle size dependent. For clarification, we have extended the description from:

“Thermodenuder treatment (250°C) removed a significant fraction of the SOA emissions.”

to:

“Thermodenuder treatment (250°C) removed a significant fraction of the OA emissions for both fresh and aged aerosol, as indicated by the differences between the respective particle number size

distributions presented in Fig. 2, and further discussed in relation to the presentation of CCN results below.”

LN272: what is reasonable detection sensitivity?

When the instrument background signal is close to zero, a number concentration of 1200 cm^{-3} would yield a minimum detection sensitivity in the order of $AF = 10^{-5}$. This is mentioned in the revised text.

LN273: “negligible, in the order of 10^{-4} from total sampled concentration” - how this number compares to activated fraction, could all INP be larger than 250nm?

It is very likely that all ice crystal formation was induced by the largest particles in each sample.

LN273: “Due to this” - Here you claim that most particles were small thus this approach for activated fraction is valid. If the IN are all large, would this approach still hold?

Due to the correct re-analysis of the ice-activated fractions, as described in more detail above, our interpretation of the potential effects of small and/or hydrophobic particles has changed. Consequently, L.273-281 have been deleted in the revised manuscript.

LN274: remove “calculating”

Done.

LN284: In addition, we estimate...

Corrected.

LN334: This short section should include deeper analysis, providing more than 1-2 sentences per instrument. For example you describe APM and effective density in the experimental section but I dont see effective density mention in the results, tables, or in the discussions.

The discussion of the CCN results has been extended significantly. Also, the particle effective density is discussed in the revised manuscript.

LN354: “Besides, our results are in complete agreement with Kanji et al. (2020) who studied...” - how propane combustion aerosol and commercial black carbon particles relevant to this study?

We have replaced the statement 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”

LN357: “was found on the other fuels, HVO and RME” - where is the discussion part of the results? what is the possible explanation for this difference?

We have changed L357 from:

“We also found that photochemical aging increased the IN activity on fossil diesel, but no distinguishable effect was found on the other fuels, HVO and RME.”

to:

“We found that photochemical aging had minor if any effects on the ice-activated particle fractions.”

LN370-376: “It is worth mentioning that all experiments in this study were conducted under well-controlled laboratory...” – The authors start the article by suggesting there is an importance for such study in MPC conditions. From this paragraph, if I understand correctly, the authors conclude that their results can't contribute much to our understanding of interactions of diesel combustion emissions in MPC and real atmospheric environment?

Diesel engine emissions comprise a significant fraction of anthropogenic particles in many environments. Hence, it is very important and relevant to investigate their potential impact on clouds and climate. However, it is not possible to capture all potential aspects and complex properties of atmospheric diesel engine emissions within a single study. Therefore, this study can be considered an important contribution to the understanding of the potential effects of diesel emissions on mixed-phase clouds. However, further experimental studies are needed before overall conclusions can be made. That is the reasoning behind the formulation.

LN545-547: switch places

The reference to Mülmenstadt et al. (2015) has been deleted during the revision process.

LN595: Fossil diesel has twice more 400nm particles, would you expect it to affect the comparison to HVO and RME?

The particle number size distributions presented in Fig. 2 represent the undiluted conditions, so the absolute concentrations are not of direct relevance to the SPIN measurements. However, it does appear as if the relative number fraction of 400 nm particles was slightly higher for the diesel emissions relative to the other fuels. In the revised manuscript, we provide estimates of the ice nucleation active surface site (INAS) densities in order to account for differences in surface areas between the samples.

LN598: “see Gren et al. (2021) for a more detailed analysis on emission particle properties of this study” - this sentence is not clear to me. Why more detailed analysis of this study is in another paper? is this an accompanying paper (part1)?

This study, as well as Gren et al. (2021), was a part of an extensive measurement campaign that produced several studies with different foci. We explain this in the introduction of the revised manuscript – the following text has been added:

“This study was a part of an extensive measurement campaign where the particulate emissions from a modern commercial heavy-duty diesel engine that was modified for single-cylinder operation were studied with multiple different foci and more information about the characteristics of the studied particles are presented in separate studies. For instance, Gren et al. (2021) studied the properties of particulate and gas-phase emissions of the test fuels, and how different emission after-treatment systems such as DOC and DPF affected them. This study focuses on immersion freezing ice nucleating abilities of the particulate emissions studied in this experiment campaign.”

LN651 Table2: For the values presented with accuracy to 3 decimal places, what is the estimated error on these?

We acknowledge that using 3 digits was excessive and present the values with two digits in the revised text. According to Gren et al. (2021), the repeatability of BC emission factors between experiments was within 25%.

Suggested references:

- Brown, A.P. "Contrail Flight Data for a Variety of Jet Fuels," AIAA 2018-3188. 2018 Atmospheric and Space Environments Conference. June 2018.
- 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, doi:10.5194/acp-17-10855-2017, 2017.
- Mahrt, F., Marcolli, C., David, R. O., Grönquist, P., Barthazy Meier, E. J., Lohmann, U., and Kanji, Z. A.: Ice nucleation abilities of soot particles determined with the Horizontal Ice Nucleation Chamber, *Atmos. Chem. Phys.*, 18, 13363–13392, <https://doi.org/10.5194/acp-18-13363-2018>, 2018.
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