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Comment on acp-2021-111

Anonymous Referee #4

Referee comment on "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 Kimmo Korhonen et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-111-RC3, 2021

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 â□°C and -32 â□°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.

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).

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).

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.

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

Specific comments:

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

Ln18: "and" change to "while the…"

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.

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

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

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

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

LN29: "instruments" – provide examples and/or references

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

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

LN38: "than those required"

LN39: "Particles that are active ice nuclei…"

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

LN41: "than that" – specify what "that" refers to

LN43: you haven't defined "INP"

LN44: "on the contrary" – on the other hand?

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

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".

LN53: abilities change to activity

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

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

LN55: " (IPCC) have identified gaps in our knowledge of ice nucleation activity of soot in their…"

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

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.

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

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

LN69-71: reword the sentence

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

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

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

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.

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

LN78: diesel combustion emissions

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

LN84: "as well as they impact" – reword

LN91: "temperatures of -35 â□°C and -30 â□°C. No immersion freezing…"

LN93: remove ice-nucleating potential

LN104: alternatives in the near future

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

LN106: “We investigate…” – reword this sentence

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?

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).

LN115: “approach” – perhaps setup or configuration?

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

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

LN124: PAM – reference for the instrument?

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

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

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.

LN131: CCNc-100 ? one column?

LN138: remove Moreover

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

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?

LN171: remove in the orders of

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

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?

LN201: 15 experiments – did you do any repetitions?

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

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

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?

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?

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 6um threshold for ice in SPIN, which was set mostly because of droplets?

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

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

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.

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

LN272: what is reasonable detection sensitivity?

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?

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?

LN274: remove "calculating"

LN284: In addition, we estimate…

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.

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?

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?

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?

LN545-547: switch places

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

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)?

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

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