Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-537

Anonymous Referee #1

Referee comment on "The effect of marine ice nucleating particles on mixed-phase clouds" by Tomi Raatikainen et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-537-RC1, 2021

Review of Raatikainen et al.

This manuscript describes a very interesting experiment carried out using high-resolution large eddy simulations to study the effect of INP concentrations in mixed-phase clouds and particularly the role of marine INPs. The study focuses around different cases with different background INP concentrations, switching on and off the marine INP emissions. The results are relevant and interesting, highlighting the effect of marine INPs under certain conditions (related to the background INP and the meteorological conditions). The study also highlights the importance of INP recycling. However, some important issues need to be addressed. I would recommend the manuscript for publication after these issues have been resolved.

One of the major points to address is the fact that the methods section is difficult to follow. This is because it is a bit unorganised (e.g., the INP description is scattered through the text). Additionally, there are many things that need to be described much more precisely, since they are relevant for the results (particularly the ice-nucleation scheme, the calculations of INPs from aerosol particles and the studied area). The fact that this section is complicated and a lot information is missing might have affected my understanding of other parts of the manuscript. I suggest the authors spend some time improving this section substantially (see specific comments below).

Another important point I see in this manuscript is the fact that some processes appear to be disconnected from existing measurements. This is particularly the case of INPs concentrations. I understand it is difficult to do so, given the fact that model assumes a temperature independent INP concentration, which then might or might not act as an INP depending on its size and temperature. Most measurements of INP use a singular description of the process and therefore produce INP vs temperature spectrums, which might be difficult to compare to the concentrations reported here. However, I still think substantial effort needs to be done in order to address this. Are the simulated background and marine INP concentrations realistic? Without these comparisons, one could argue that maybe the modelled marine INP component is too high compared with the background (or the opposite). Realistic INP concentrations are necessary to support one of the main conclusions of the study (marine INPs have an influence on mixed-phase clouds).

Specific comments

Introduction

- General: As a suggestion, I do not think it is necessary to add e.g. before the references.
- Lines 21-28: The first sentence refers to immersion mode, while the rest of the paragraph defines the ice-nucleation modes, finishing with the immersion one. I suggest altering the order of the paragraph: starting with a description of the modes, which finishes with the immersion mode. Then add the statements in how immersion mode affects shallow mixed-phase clouds.
- Line 33: The mentioned reference only provides INP vs temperature data. I would add other examples of fieldwork where INP vs temperature and INP vs relative humidity are shown.
- Line 35: I suggest not including soot as one of the most important types of INPs. Although this might be the case for deposition mode, there is not much evidence that it is for the immersion mode and these part of the introduction (and the manuscript in general) is referring to the immersion mode.
- Line 38 "In the absence of dust": I suggest mentioning that there some dust sources in cold high-latitude environments (Bullard et al., 2016) and they could be important local INP sources in the absence of dust from deserts too (Tobo et al., 2019, Sanchez-Marroquin et al., 2020).

Methods

- Sect. 2.2: Have Secondary Ice Processes been implemented?
- Line 65: A bit more of information on the ISDAC should be added; where and when it happened, the type of measurements that were conducted that are used in this work.
- Line 70: Is the 4 L^{-1} concentration based in something? If this comes from a measurement of the ISDAC campaign, it should be stated.
- Line 77: I suggest adding a full description of the species used for this work. I would also add a description of the bin range.
- Line 83: It is necessary to add a description of the ice-nucleation scheme and how it relates to dust and sea spray. Is the scheme (and its output) consistent with recent experimental INP measurements?
- Line 91 "adjusting the initial concentrations of INPs": How was this done? Is this validated based on any experimental study?
- Line 92 "ammonium bisulphate" (and in general through the whole manuscript): Is the ammonium bisulphate an immersion mode INP or is it just there to activate the dust as a CCN prior to ice-nucleation? The later does not seem the case, since later the authors state that the dust and ammonium bisulphate are externally mixed. I do not think ammonium bisulphate is a relevant type of INP in the immersion mode, according to the literature (it could be for cirrus clouds but the focus here is mixed-phase clouds). The inclusion of this species to the INP population needs to be much better explained and justified (there are not even mentions to it in the introduction but then it seems to be a major thing for this work) or removed.
- Figure 1. As a suggestion, I would use μm instead of m for consistency with most of the

aerosol studies.

- Line 95 "(set to 0.00015)": Does this mean that the INP concentration is 0.00015 times the dust concentration, regardless of the temperature? In general, this point would be addressed by including a much more detailed description of the ice-nucleation scheme, and if it is linked or justified based on any experimental study.
- Line 108 "Ice nucleation follows the approach used in Ahola et al. (2020)". The description of the ice-nucleation scheme seems to be split between the lines 85 to 97 and 108 to 120. I suggest merging this. Additionally, as previously explained, the description needs to be much more extensive, clear and organised.
- Line 115 "but marine and background INPs become internally mixed": I do not really understand this part, please explain it better. If ammonium bisulphate can be excluded as an immersion mode INP, maybe you can have the background and marine INPs externally mixed?
- Line 117 "internally mixed dust and ammonium": Isn't this externally mixed as stated in line 114?
- Line 121-128: How is the SSA related to INP? Is it based in any experimental parameterization? Is there any temperature dependence? How do the modelled INP concentrations compare with available measurements of INP or concentrations from other models?
- Line 139 "adjusting the fractions of INPs". Explain this and if the adjusted INP concentrations are comparable to measurements.

Results and discussion

- Figure 2 (legend): I suggest using only 5 colours for this graph, each of them corresponding to each INP fraction, and then use the "dashed" "not dashed" to refer to Marine emissions on and off.
- Line 169 "Simulations where marine INP emissions are switched off are excluded for clarity": Why, isn't this comparison interesting too?
- Figures 3 and 6 would benefit from having a title specifying that each column refer to mass and number respectively.
- Lines 182 to 186: From what I understand, this analysis is done on what the authors call "INP" which is not the particles that nucleate ice but a temperature independent fraction of the total aerosol (background and marine). Then, some of these "INPs" will act as an INP based on their size and temperature. Hence, this analysis of INP budget, seems more like and aerosol budget analysis. I think the authors should justify why this analysis is relevant to the particles that will nucleate ice in the model, which are likely the ones that are numerous and carry enough surface area (see next comment). As previously mentioned, a more concise, systematic and expanded description of the INP scheme would help understanding this too.
- Line 186 "we will focus more on the INP mass": What about INP surface area? Your mass INP will be dominated by the upper end of the aerosol range, however, aerosol particles of medium sizes would carry a substantial amount of surface area (if not the majority) and they might not contribute much to the mass or number, biasing the results. This seems important since I guess the stochastic ice-nucleation scheme will be dependent on the surface area of the aerosol particles? I suggest addressing this too, or justifying why surface area has been excluded of the analysis.
- Section 3.2 (general): This comment is related to previous comments about comparison with observations. I think more effort should be done in order to show that

the modelled INP budgets are realistic, showing some consistency with measurements (I am aware there might not be measurements at that specific time and location, but the concentrations could be compared to the closest available observations). I appreciate it is difficult to perform this comparison, since most measurements are carried out using a singular description of ice-nucleation while this work uses a stochastic one. However, I still think some effort should be put into this.

- Line 203 "over simulation time 7-8h": Make clear than you are doing a one hour average starting at the seventh hour of the simulation. It took me a while to realise what the authors mean.
- Line 205 "The other simulations show": Does this mean the same analysis applied in other 1 hour time intervals? Are all the conclusions of this analysis valid for the other time intervals? This should be explained more concisely.
- Line 237 " Simulations where marine INP emissions are switched off are excluded for clarity": Why? Isn't it making any difference?
- Line 265 "ice nucleation rates": Could you describe what this magnitude means in the model? Is it the number of primary ice freezing events per second and per surface area of INPs?
- Figure 8 would benefit from having a title indicating which panel refers to marine missions on an off.
- Line 271 "updraughts have higher INP mass concentrations": This section is interesting. However, I find the magnitude INP mass in a column a bit disconnected. Wouldn't it be better to give this in INP number?
- Line 282: "aerosol freezing": Is this what other studies refer to as deposition (or pore condensation freezing) ice-nucleation? It is not clear. If so, please indicate in the method section how this process is parameterized. If my assumption is right, link it with the existing literature, which suggests the same (this process is not very important for mixed-phase clouds, when comparing with immersion freezing).

Conclusions:

- Line 323 "which has prognostic aerosol, cloud and ice phase INP size distributions". This sentence is describing the methods; it would go better in the first paragraph of the conclusions.

Other

- Ice nucleation: Ice-nucleation
- Line 3 "ice nucleating particles (INPs)": Ice-Nucleating Particles (INPs)
- Line 88 "258 K or -15 ^{o}C": I suggest sticking to one temperature unit through the text.