Response to Dr. Cyril Brunner

We thank Dr. Brunner for his careful reading and review of our paper. Our detailed responses to his comments follow. Reviewer comments are in blue, our responses are in black, and our corresponding revisions in the manuscript are in red.

General comments:

The authors present a very interesting study on the effect of marine organic aerosol (MOA) on mixed-phase clouds. In particular, they present and validate results from three different MOA emissions schemes, quantify the resulting spatial cloud condensation nuclei and ice nucleating particles (INP) number concentrations and compare it to modeled INP concentrations of dust, using state of the art parametrizations. In contrast to previous work by other authors, they present data comparing the INP population of MOA to INPs of dust.

The writing (from an editorial standpoint) is to be commended. The methodology is stringent and valid. The assumptions made are to a majority stated and their impact on the result comprehensibly assessed. The work addresses relevant scientific atmospheric questions with impacts on global climate simulations. The topic of the paper is well suited for ACP. I recommend the manuscript for publication if the following minor comments are addressed:

We thank the reviewer for the encouraging comments. We have revised the manuscript following your comments and clarified the text to improve the paper.

Specific comments:

Page 2 (line 12/13). I do not fully understand why the three regions are stated. Are mixed-phase clouds only observed in the Arctic, Antarctic, and over the Southern Ocean? As the paper is also not focusing on these regions, I would propose to rephrase the sentence. E.g., include all regions or specify what is unique about mixed-phase clouds in the three stated regions.

We thank the reviewer for the suggestion. We have revised the sentence to read:

"Mixed-phase clouds are frequently observed in high-latitude regions, and..."

Page 3 (lines 44-48). Kanji et al, 2017 provide an excellent overview of the different modes of freezing. The stated mechanism, however, was not introduced by Kanji et al., 2017. Please cite the original source or e.g. Vali, G., DeMott, P. J., Möhler, O., andWhale, T. F.: Technical Note: A proposal for ice nucleation terminology, Atmos. Chem.Phys., 15, 10263–10270, https://doi.org/10.5194/acp-15-10263-2015, 2015.

We thank the reviewer for the suggestion. We have added the reference as you suggested, and the revised sentence reads as:

"In mixed-phase clouds in which air temperature is between –38˚ C and 0˚ C, ice is initialized only by heterogeneous nucleation on ice nucleating particles (INPs) (Vali et al., 2015)."

Page 15 (line 399). Is for the NULL approach the annual global MOA or sea salt emission 4.6 Tg yr-1? Please specify.

We thank the reviewer for the suggestion. We meant the annal global MOA emission of 4.6 Tg yr^{-1}. We have revised the sentence as:

"The NULL approach only gives an annual global MOA emission of 4.6 Tg yr^{-1}".

Page 23/24 (lines 652-683). The missing representation of secondary ice formation is nicely formulated. However, the study also does not model other IN species, such as ash, biomass-burning particles, or other land-borne bio particles. Please elaborate on the impact of this (valid) simplification on the study's results.

We thank the reviewer for the excellent comment. We have added some discussion on missing representation of other types of INPs in the model in the newly added third paragraph of section 4. The revised sentences read as:

"In this study, other potential INP species than dust and MOA, such as ash, biomass-burning particles, or other land-borne biological particles (Hoose et al., 2010; Jahn et al., 2020; Schill et al., 2020) are not represented in the model. These INP species can be regionally important at certain temperature regimes of mixed-phase clouds. Accounting for these species may increase the INP concentrations predicted in the model and change the mixed-phase cloud properties, particularly at warmer temperatures > -15ºC. The impacts of these INP species will be quantified in our future studies."

Page 32 (Table 1). Please explain variables in the caption, such as g, to support the reader's quicker understanding.

We thank the reviewer for the suggestion. We have added the explanations for variables in Table 1, and the revised caption for Table 1 reads:

Table 1. Aerosol species in MAM4 modes

	Accumulation	**Aitken**	**Coarse**	**Primary Carbon**
Species[1]	num_a1, so4_a1, pom_a1, soa_a1, bc_a1, dst_a1, ncl_a1, moa_a1	num_a2, so4_a2, soa_a2, ncl_a2, dst_a2, moa_a2	num_a3, dst_a3, ncl_a3, so4_a3	num_a4, pom_a4, bc_a4, (moa_a4 if externally mixed)
Size range	0.08 – 1 μm	0.02 – 0.08 μm	1–10 μm	0.08 – 1 μm
Standard Deviation σg	1.6	1.6	1.2	1.6
Number-median diameter *Dgn*	1.1×10^{-7}	2.6×10^{-8}	2.0×10^{-6}	5.0×10^{-8}
Low bound *Dgn*	5.35×10^{-8}	8.7×10^{-9}	4.0×10^{-7}	1.0×10^{-8}
High bound *Dgn*	4.8×10^{-7}	5.2×10^{-8}	4.0×10^{-5}	1.0×10^{-7}

[1]so4_aX: sulfate mass mixing ratio in mode X; pom_aX: particulate organic matter (POM) mass mixing ratio in mode X; soa_aX: secondary organic aerosol (SOA) mass mixing ratio in mode X; bc_aX: black carbon (BC) mass mixing ratio in mode X; dst_aX: dust mass mixing ratio in mode X; ncl_aX: sea salt mass mixing ratio in mode X; moa_aX: marine organic aerosol (MOA) mass mixing ratio in mode X;

and num_aX: number mixing ratio of mode X. *_a1: accumulation mode; *_a2: Aitken mode; *_a3: coarse mode; and *_a4: coarse mode.

Page 46 (Figure 6). No need to change anything, just a general comment. With INP measurements, we are often divided about how the show the INP concentration most representatively over a long period. If we calculate the mean concentration, the result will be "biased" towards higher INP concentrations if a few events with INP concentrations in the order of 102 to 103 are present. IMHO showing presenting the reader also with the median concentration provides a complete picture.

We thank the reviewer for the suggestion. We agree with the reviewer that mean concentration will be "biased" towards the higher INP. However, in GCMs we are simulating the climatological states of aerosols, not episodical events.

Supplement (Figure S1). The unit of plot d) is not fully visible

Thanks. We have fixed this. The revised Figure looks like: