We have reproduced the reviewers comments in **bold text**, the answers in normal font. We have used *italicised text* from the manuscript and used *blue font* for any new text added and ~~*red strikethrough*~~ for any text removed.

Reviewer 1

General comments

The authors evaluate the Arctic aerosols representation in four CMIP6 models (CESM2, MRI-ESM2, MIROC-ES2H and UKESM1) and a chemical transport model (TM5) using the one-year dataset generated during the MOSAiC expedition. They observe that CMIP6 models underestimate aerosol concentration and fail to reproduce the aerosols seasonal cycle, and then use several TM5 experiments tuning different individual parameters (MSA treatment, DMS emissions, nucleation rate, sea spray aerosols emissions), to investigate the reasons of the discrepancies between models and MOSAiC observations of aerosol number concentration (CN) and cloud condensation nuclei (CCN).

Methanesulphonic acid (MSA) and iodic acid are the main aerosol precursor in spring, summer and autumn during the MOSAiC campaign. Therefore, the authors discover that implementing an MSA dependent nucleation scheme in their TM5 model improves the seasonality of the CN values, and greatly improves the comparison with observations. The implementation of an iodic acid dependent nucleation scheme could also help to improve the aerosols representation in the TM5 model.

I think the authors make good use of the data obtained during the MOSAiC campaign, using the dataset to investigate the current deficiencies in the representation of Arctic aerosols in the studied CMIP6 models. Therefore, I recommend the manuscript to be published after some points are addressed:

We thank the reviewer for their positive assessment of the manuscript and for highlighting the usefulness of the MOSAiC dataset for evaluating Arctic aerosol representation in CMIP6 models and TM5. We agree that iodic-acid-driven nucleation can be a key missing pathway, especially in late summer and autumn, and we have strengthened the discussion of this limitation and its implications. We have also expanded the Methods section and clarified the technical points as suggested by the reviewer. Below, we give our detailed responses to each comment brought forward.

Specific comments

L69: "By using a set of simulations with individually perturbed parameters,…" Please, mention here what individual parameter have been perturbed.

Thank you for the comment. We have included details on parameter perturbations to the end of introduction by adding the following lines

L69 : "By using a set of simulations with individually perturbed parameters, including MSA volatility and nucleation treatment, DMS emissions, boundary layer nucleation rate and sea-spray aerosol emissions, we quantify the sensitivity of Arctic aerosol to uncertainties in

processes and emissions."

L83: What are differences between the CB05 carbon-bond mechanism chemistry scheme and the modified version mCB05? These should be mentioned in the text.

We agree that the original text did not sufficiently identify the chemistry scheme or explain the relationship between CB05 and mCB05. We have revised the manuscript to include the original CB05 reference and the TM5 mCB05 implementation references. We now clarify that mCB05 is the TM5-adapted implementation of the CB05 carbon-bond mechanism for global tropospheric chemistry simulations. Because the detailed reaction-level modifications are described in Williams et al. (2013), we refer readers to those studies rather than reproducing the full mechanism description here.

L83: A modified version of the CB05 carbon-bond mechanism chemistry scheme (Yarwood et al. 2005), mCB05, is implemented in the model (Williams et al. 2013) ~~*(Williams et al. 2017)*~~*. Compared with the earlier modified CBM4 mechanism used in TM5, mCB05 includes a more explicit treatment of lower molecular weight C1–C3 organic species, reducing the number of VOC species lumped into generic tracer species.*

L104 and Eq.2: the oxidation scheme for DMS employed in the TM5 model seems too simple and outdated. There have been several updates in the last years (e.g. Kilgour et al., 2024 (https://doi.org/10.5194/acp-25-1931-2025).

We agree that the DMS oxidation scheme used in TM5 is simplified. In the current model setup, DMS oxidation is represented using a fixed branching ratio to SO_2 and MSA, and therefore it does not include recent developments in DMS oxidation chemistry, such as temperature-dependent MSA production or multi-step pathways involving intermediates such as HPMTF. We have added this limitation to the manuscript and cited Shen et al. (2022)and Kilgour et al. (2025). Implementing a more detailed DMS oxidation mechanism would require substantial changes to the gas phase chemistry scheme and is beyond the scope of the present study, but we now explicitly discuss this as a source of uncertainty in the interpretation of the DMS sensitivity experiments. We have added the following text to clarify these points in the description of the experiment setup

L160: ~~*In TM5, it undergoes oxidation to produce sulphate and MSA (see Eq. 2), both of which contribute to aerosol nucleation and growth processes.*~~ *In the present TM5 setup, DMS oxidation is represented using a simplified reaction with a fixed branching ratio to SO_2 and MSA. This treatment does not include recent developments in DMS oxidation chemistry, including temperature-dependent MSA production and multi-step pathways involving intermediates such as HPMTF (Shen et al., 2022; Kilgour et al., 2025). This simplification is particularly relevant in the Arctic, where low temperatures may affect the relative production of MSA and sulfate. Therefore, the DMS sensitivity experiments should be interpreted as tests of the model response to DMS emission uncertainty rather than as a complete assessment of DMS oxidation chemistry.*

L112-121, section 2.1.2: This section is too short, relying mainly on previous literature. It should at least include a brief description of the implemented emissions, instead of just directing the reader to the literature on which they are based

We thank the reviewer for this comment. We clarify that Section 2.1.2 describes the emission setup used in the control simulation, while the perturbations to DMS and sea-spray emissions are described separately in Section 2.2.4. To make the control setup clearer, we have expanded Section 2.1.2 with a brief description of the baseline anthropogenic, DMS, sea-spray aerosol and biogenic VOC emissions used in TM5.

L112 : *The evolution of the aerosol population in TM5 depends on the modelled and prescribed emissions. For the control simulation, anthropogenic emissions and baseline natural aerosol emissions are prescribed from the CMIP6 emission dataset (van Vuuren et al. 2011; Lamarque et al. 2013; T. P. C. van Noije et al. 2014, 2021), following Shared Socioeconomic Pathway 3-7.0 scenario for 2019–2020 (Gidden et al. 2019). DMS emissions over land are calculated following Spiro et al. (1992), which provides a global sulfur-emission inventory including natural terrestrial sulfur sources, while oceanic DMS emissions are calculated from the Lana et al. (2011) seawater DMS climatology and masked by prescribed 1° sea-ice cover. Wind-driven sea-spray aerosol emissions from breaking waves are calculated using the Gong (2003) source function, with a temperature-dependent size distribution following van Noije et al. (2021). Biogenic NMVOC emissions are based on the MEGAN inventory of Guenther et al. (2012), which estimates emissions from vegetation using land-cover type, emission factors and meteorological drivers such as temperature and radiation. Sensitivity experiments in which DMS and sea-spray aerosol emissions are perturbed are described in Section 2.2.4.* ~~*The anthropogenic emissions and the natural aerosol baseline for the model are from the CMIP6 emission dataset (T. van Noije et al. 2021; T. P. C. van Noije et al. 2014; Lamarque et al. 2013; van Vuuren et al. 2011), following the Shared Socioeconomic Pathway 3-7.0 scenario (Gidden et al. 2019) for the studied period (2019-2020). DMS emissions over land are calculated as described in Spiro et al. (1992), and oceanic DMS emissions are calculated based on Lana et al. (2011), masked by prescribed 1-degree sea ice cover. Wind-driven sea spray emissions from breaking waves (SSA) in TM5 are based on Gong (2003), with a temperature dependent size distribution as described in Van Noije et al. (2021). Biogenic emissions of non-methane volatile organic compounds (NMVOCs) are based on Guenther et al. (2012).*~~*The chemistry setup and other emissions in the model are described in detail in Huijnen et al. (2010) and van Noije et al. (2014).*

L127: 9-month spin-up period. I think that is too short a period of time. Did the authors checked that that time is sufficient? Typically, 1-2 years spin-up is employed for troposphere stabilization.

We thank the reviewer for raising this point. The 9-month spin-up was chosen because the analysis focuses on short-lived tropospheric aerosol number concentrations and precursor-driven aerosol processes along the MOSAiC trajectory, rather than on long-lived chemically active tracers. Most aerosol species considered here have atmospheric lifetimes of days to weeks, so the 9-month spin-up is sufficient for the aerosol fields relevant to this study. Also, 3-6 months is a typical spin up range for global chemistry transport models as can been seen in Zhang et al. (2012), Leon-Marcos et al. (2025) Salzmann et al. (2022) and Wang et al. (2026).

L204-208: at least a brief description of the instruments and methods employed to create the MOSAiC dataset should be included. Simply listing the references is insufficient.

We have expanded Section 2.3.2 to briefly describe the CPC, CCNC, and NO_3-CIMS measurements, including the particle size range, supersaturation used for CCN, and the filtering of pollution-contaminated data.

L201: Aerosol observations onboard RV Polarstern were conducted from the Swiss Container and the ARM Aerosol Observing System using a comprehensive suite of instruments. Aerosol particle number size distributions (10–500 nm) were measured with a Scanning Mobility Particle Sizer (SMPS; TSI 3082 classifier, 3081 DMA and 3772 CPC). Black carbon concentrations were measured with a Magee Scientific AE33 Aethalometer. Aerosol chemical composition of non-refractory PM_1 was obtained using a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS), and supporting measurements included cloud condensation nuclei counters (CCNCs), condensation particle counters (CPCs), an Aerodynamic Particle Sizer (APS). Also, a Wideband Integrated Bioaerosol Sensor (WIBS) measured aerosol microphysical and fluorescence properties. The aerosol instrumentation sampled from dedicated whole-air and interstitial aerosol inlets mounted ~15–18 m above sea level. The instruments used and the processes involved in creating these datasets are discussed in detail in Heutte et al. (2023) *and Boyer et al. (2023)*

L219: "..while TM5 both underestimates or overestimates the observed monthly CN, depending on the experiment." To understand what experiment underestimate o overestimate observations, it will be helpful if figure 2 differentiated the different TM5 experiments, instead of grouping them all with the same symbol (pink cross mark).

We thank the reviewer for raising this point. When plotting Fig. 2 the option to add the names of the experiments were considered. But this reduced the readability of the figure due to the expanded legend and the overlap between the experiments. Also, since Fig. 3 compares the experiments with the observed number concentrations, we thought it would be redundant to include this in Fig. 2. We acknowledge that the text quoted in the comment would confuse the reader and so change the text to make the paper easier to follow as

L129: The variability in the monthly averages differs between the four models. All four analysed CMIP6 models underestimate observed monthly averaged CN concentrations, ~~while TM5 both underestimates or overestimates the observed monthly CN, depending on the experiment~~ while TM5 BASECASE overestimates CN in summer and underestimates CN in winter.

The authors mention and demonstrate (Fig. S4) the importance of iodic acid as a primary driver of nucleation in the Arctic from spring to autumn. Nevertheless, they do not include the formation mechanism in their TM5 model. It should not be difficult to implement the formation mechanism described in the Finkenzeller et al., 2022 (Nat. Chem. 15, 129–135 (2023). https://doi.org/10.1038/s41557-022-01067-z), using lower boundary conditions (LBC) for its precursors (e.g. iodine monoxide) from another model implementing an atmospheric halogen chemistry scheme. I think a sensitivity experiment implementing the formation of iodic acid would help to improve the comparison with observations during that time period, if they also implement an iodic acid dependent nucleation scheme. Although the latter is perhaps more difficult. That could help to give an answer for the question in L440: "Iodic Acid is a main driver of NPF in late summer and early autumn in the Arctic (Price et al. 2023) and could be the reason the model does not capture these events"

We agree that iodine chemistry and iodic-acid-driven nucleation are potentially important for Arctic aerosol formation and that assessing this pathway would be of great interest. However, including iodine nucleation pathways is outside the scope of this paper. A meaningful implementation would require iodine precursor emissions, chemical formation pathways for iodic acid, and an iodic-acid-dependent nucleation scheme. Implementing overly simplified chemical mechanisms (or prescribed precursor concentrations), together with nucleation schemes that have not been extensively evaluated in regional or global models, might lead to an inaccurate assessment of the role of iodine-driven nucleation. Our sensitivity studies are designed to provide a broad assessment of the potential role of secondary and primary aerosol sources in the Arctic. However, omitted precursors and processes, such as iodine chemistry, may still cause temporal and spatial biases that are not captured by the perturbations tested here. We have refined L440 as follows to reflect this

L440 : Iodic acid is thought to play a significant role in NPF in the Arctic in late summer and early autumn (Price et al. 2023) and may contribute to the model's inability to reproduce these events.

Technical corrections

Define CMIP and AMIP projects acronyms. It will be useful for non-modeller readers.

The first occurrence of CMIP and AMIP are changed to CMIP(Community Model Intercomparison Project) and AMIP(Atmosphere only Model Intercomparison Project)

Figure 1: Font size in upper-right label is too small.

Changed.

L127: describe what a "vertical resolution of 34 hybrid sigma pressure levels" is. It will be useful for non-modeller readers.

We have changed the text to the following for added clarity.

L127 : For all simulations, the model was run from October 2019 to September 2020 with a 9-month spin-up period, at a horizontal resolution of 3° longitude by 2° latitude ~~vertical resolution of 34 hybrid sigma-pressure levels~~. The vertical resolution of the run was 34 hybrid sigma-pressure levels, where the lowest level follows the surface elevation and the higher levels are isobaric.

L230-231: "The main difference between the experiments is that the AMIP runs use observed SIC and SST for the plots, …". In this statement the words "for the plots" can be removed.

Done.

L286: replace "$1*10^6$ /cm³" by "1×10^6 /cm³"

Done.

L356: Replace "Figure 5 shows the spatial distribution of CN…" by "Figure 5 shows the modelled spatial distribution of CN".

Done.

Figure 6 and 7: font size of labels and colour bar numbers too small.

The font size has been increased.

Figure 8: include (c) and (d) labels are missing in lower panels. The blank spaces at both the beginning and end of the four panels can be removed.

Changed as described in the comment.

L411-414: that paragraphs is almost the caption of Figure 8, so it should either be removed or rephrased.

L411-414 has been removed.

L440: replace "(Fig. S4)" by (Fig. S4d).

Changed.

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