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 2

General Comments

This manuscript by Chandrasekharan et al., titled "Understanding uncertainties in Arctic aerosol representation in climate models", evaluates the representation of Arctic aerosols in four CMIP6 models and the TM5 model using the full annual cycle of observations from the MOSAiC expedition. The topic is timely and of clear importance to the Arctic climate modelling community. The ensemble of sensitivity experiments of parameter purturbation is quite comprehensive, and the study sorted out the relative contributions of several key processes, including MSA-driven nucleation, DMS emissions, sea spray aerosol, and nucleation rate uncertainties, to Arctic aerosol number and CCN concentrations across seasons. These results provide valuable and actionable insight into the structural gaps in current climate models. However, several findings would benefit from more thorough physical interpretation before the manuscript is ready for publication (see detailed comments below). Overall, the writing can use some improvement since the manuscript is a bit hard to follow. I would recommend minor revision.

We thank the reviewer for their thorough assessment of the paper and for highlighting the importance of ensemble studies in furthering the understanding of aerosol modelling in the Arctic. We have expanded the description and discussion of CCN guided by the suggestions from the reviewer. We have also taken the suggestion regarding improving the writing and flow of the manuscript into account and made changes to improve the manuscript.

Specific Comments

Line 91 Please provide a brief description of the ELVOC treatment, including how their gas-phase concentrations are calculated and how they partition to the aerosol phase.

We thank the reviewer for bringing this to our attention. We have changed the text as follows to explain the ELVOC treatment in detail.

L91: ~~*SOA in the model consists of extremely low volatility organic compounds (ELVOCs) and semi-volatile organic compounds (SVOCs). ELVOCs condense on particles depending on the particle surface area of each mode, whereas SVOCs condense on particles depending on the total mass of organic aerosols (OA) existing in each mode. As a result of their treatment in the model, SVOCs mainly contribute to the growth of accumulation mode and coarse mode particles, and ELVOCs contribute to nucleation and early growth of aerosols, as well as the growth of larger aerosol particles (Bergman et al. 2022).*~~ *The formation of SOA in TM5 is based on Jokinen et al. (2015), where products of BVOC oxidation are categorised simply as extremely low volatility organic compounds (ELVOCs) and semi-volatile organic compounds (SVOCs), and formation is based on prescribed yields for each oxidant. Our simulations only consider total monoterpene emission instead of detailed speciation, and we use the ELVOC/SVOC yields corresponding to α-pinene for all monoterpene. A total SOA yield of*

15% and 5% is assumed for monoterpene and isoprene, respectively. Both SVOC and ELVOC partition to aerosol phase during the timestep. ELVOCs condense on particles depending on the particle surface area of each mode, whereas SVOCs condense on particles depending on the total mass of organic aerosols (OA) existing in each mode. As a result of their treatment in the model, SVOCs mainly contribute to the growth of accumulation mode and coarse mode particles, and ELVOCs contribute to nucleation and early growth of aerosols, as well as the growth of larger aerosol particles (Bergman et al. 2022) .

Line 108 The CCN diagnostic based on Petters and Kreidenweis (2007) is introduced without sufficient methodological detail. Please give a brief description. Also, please clarify (1) how the scheme treats coarse-mode particles (e.g., sea spray aerosol) as potential CCN, and (2) whether and how competition effects between particles of different sizes during activation are accounted for. This is relevant in the Arctic context where both freshly nucleated particles and large primary particles coexist.

We thank the reviewer for this clarification request. In TM5, all particles are treated similarly depending on their size and Kappa. Sea salt resides in accumulation and coarse modes, but it is mixed with other compounds. As with all modes, a volume-weighted kappa value is calculated for these particles as well. The competition for water vapour is not accounted for, as no real activation is calculated. This formulation is for diagnostic cloud condensation nuclei only, meaning it only provides an estimate of the potential number of cloud droplets. We have also revised the section describing CCN as follows

L 108 : The calculation of CCN is based on the Eq. 10 from Petters and Kreidenweis (2007). ~~*Cloud Condensation Nuclei (CCN) calculation in TM5 is based on equation 10 from Petters and Kreidenweis (2007). CCN is a climate-relevant variable; it is a fraction of ambient aerosols that can participate in cloud droplet formation. The ability of an aerosol particle to act as a CCN depends on its size and chemical composition, as well as on the ambient supersaturation. A fraction of accumulation and coarse mode particles can act as CCN.*~~ *Over the simulated aerosol population it follows th procedure in Mann et al. (2010) and Block et al (2024). In practice, the CCN threshold size is calculated for soluble Aitken, accumulation and coarse modes based on the prescribed supersaturation(s) and modally determined volume-weighted Kappa. The total CCN for a given supersaturation is then the sum over the particle concentration of individual modes exceeding the threshold size. The Kappa values of different aerosol components in TM5 can be found in Table 1 in Fanourgakis et al. (2019) .*

Table 2 (BULKMSA) Consider replacing "bulk tracer" with "passive tracer" to more precisely convey that MSA in this experiment undergoes no further chemical processing or size-resolved partitioning.

We thank the reviewer for asking this clarification. According to Saltzmann et al. (1983) and Pzenny et al (1992), the MSA aerosol is formed through condensation onto existing particles in the accumulation and coarse modes. However, in TM5, MSA is assumed to condense only on the particles in the accumulation mode to keep the implementation simple (aan de Brugh et al. 2011). Therefore, it affects the size in the accumulation mode as well as the refractive index of the mode. It is true that, compared to the other species in the M7 context, MSA is rather passive. However, we would rather call it a mass-only tracer or bulk tracer than a passive tracer. Also, we have used the same nomenclature as van Noije (2014) regarding TM5 aerosol description: "The other aerosol components included in the model are nitrate, ammonium, and methane sulfonic acid (MSA), which is produced in the oxidation of DMS. These are represented by their total mass, i.e. using a bulk aerosol approach." With this in

mind, in the BULKMSA experiments, methylsulfonic acid is treated as a mass-only tracer which is not influenced by the aerosol growth processes. This means that the mass is added at every time step to the accumulation mode after the calculation of aerosol microphysics. As such, it will affect the calculation of refractive indices and sedimentation of the accumulation mode. To clarify the handling, we will rephrase the section on the MSA handling in the main text.

Table 2 : *BULKMSA – Nucleation scheme is the same as BASECASE, but MSA is treated using a bulk aerosol approach (Sect. 2.1.1)* ~~*as a bulk tracer and*~~ *does not participate in chemistry.*

Line 206 The authors state that aerosol precursor gases (H_2SO_4, HIO_3, and MSA) were measured during MOSAiC and used in the analysis, yet no model–observation comparison for these species is presented in either the main text or the supplement. It is unclear to reader how well the model reproduces precursor concentrations and therefore difficult to assess how model parameters should be adjusted relative to the BASECASE to improve CN and CCN simulations. Please include these comparisons in the main text (or did I overlook?).

We thank the reviewer for bringing this to our attention. As pointed out by the reviewer, in the current version of the manuscript, there is no direct comparison of modelled and observed aerosol precursors. In Fig, S4, we use aerosol precursor values to try to find possible reasons for discrepancies between observed and modelled CN. To make the comparison more explicit, we have added a new figure,Fig. S5, which compares the monthly average values of the observed aerosol precursors to the available modelled aerosol precursors(MSA and H2SO4). We also add the following analysis to the main text

L271: The modelled seasonal cycle of sulphuric acid is different from the observations. We find that the model predicts lower concentration in winter and higher concentrations than the observations in summer. Despite the higher concentration of sulphuric acid in summer, we find that the values are still below the cutoff of $1x10^{-6}$ cm^{-3} and so seems to have a negligible effect on CN in summer. With MSA, we see that the modelled values are one degree of magnitude lower than the observations. Moreover, the modelled values have a steep decline from July 2020 when compared with the observations.

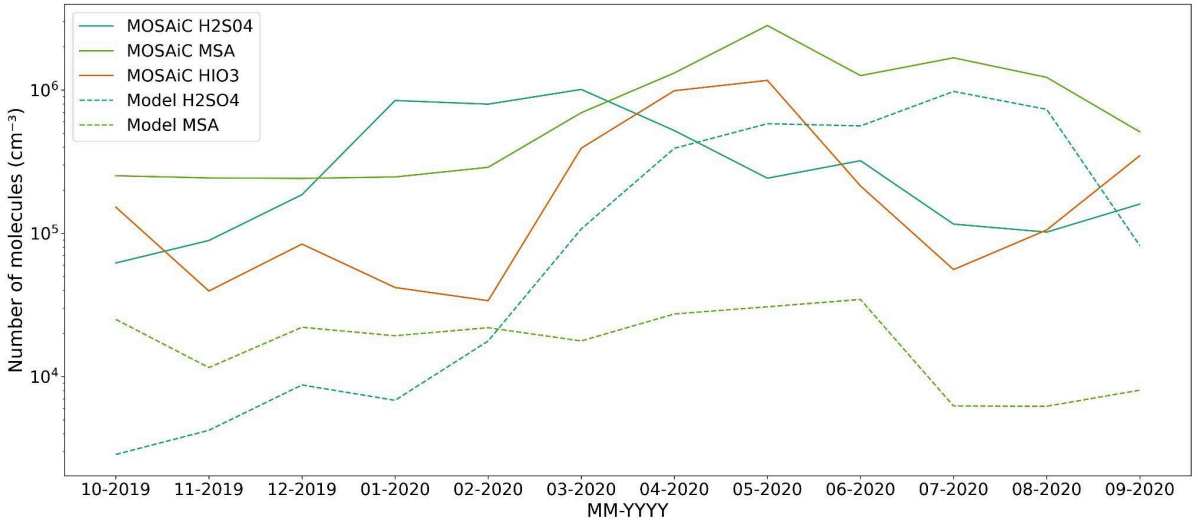

Fig S3: Comparison between modelled and observed monthly average aerosol precursor concentration from October 2019 to September 2020. The solid lines are the observed sulphuric acid, MSA and Iodic acid concentrations measured during the MOSAiC campaign. The dashed lines show the modelled monthly averages of sulphuric acid and MSA. Iodic acid emissions are not present in TM5.

Lines 220–221 It is worth noting that models such as CESM2 do not include MSA as an aerosol component or nucleation precursor. This omission is likely a primary reason for CESM2's severe underestimation of Arctic CN and CCN.

We agree with the reviewer about the importance of missing MSA in CESM. However, none of the other CMIP6 models included have MSA participating in nucleation either. Also, when looking at the comparison between AMIP and historical runs of CESM, we find a significant difference in CN. This points to an inaccurate representation of sea ice concentration being the primary cause of the underestimation of CN in the historical run. But we thank the reviewer for bringing up the importance of Arctic-relevant aerosol represented in the models. We have added the presence of mineral dust and DMS emissions as important reasons for CN modelled in MIROC and UKESM to be closer to MOSAiC values in Section 3.1 .

Line 262 Most CMIP6 models employ highly simplified chemistry schemes that are known to poorly represent long-range transport and ageing of anthropogenic aerosols even at mid-latitudes. Therefore, I wonder how Arctic haze is modeled. The authors should compare simulations of anthropogenic aerosols (e.g., sulfate) against MOSAiC, and discuss how these limitations in the CMIP6 models' chemical schemes may contribute to the wintertime and springtime CCN underestimation.

We thank the reviewer for bringing up this point. We agree that CMIP6 models poorly represent long-range transport of aerosols. This is partly due to the simplified chemistry schemes employed and partly due to incorrect modelling of meteorology in climate models. Here, since TM5-MP is run with the ERA5 reanalysis data, we have ignored incorrect modelling of transport as a reason for deficiencies in the model during the winter and Arctic haze period.

We have added Fig. S3 to the supplementary material comparing the aerosol precursor concentration between MOSAiC and the model and also added discussions regarding this as mentioned in the answer to the review comment of L206.

Line 269 The discussion of factors contributing to overestimated summer CN in TM5 should acknowledge the potential role of missing Arctic dust. Mineral dust can act as a condensation sink for H_2SO_4, MSA, and SOA; its absence in the model would reduce the condensation sink, leading to higher gas-phase precursor concentrations and consequently elevated nucleation rates and CN.

We thank the reviewer for bringing this to our attention. The text on underestimation of CN in the summer has been edited to account for this.

L269 : While the simulations including BASECASE overestimate the CN values in summer, it is also important to note that the model is still missing important sources of aerosols. The version of TM5 used here does not include mineral dust, which acts as a sink for aerosol precursors. Including this could bring down CN during summer.

Lines 272–274 It looks like that the SS_HIGH run produces CN values closer to the MOSAiC observations in summer than the BASECASE (Fig. 3d), which may be good.

We agree with the reviewer that the monthly averages are close to the observations. But we also see that the monthly average values remain the same from July 2020 through September 2020. So, while we see that the average values are closer to the observations, we can also see that there is very little difference between the average values for the months. This experiment does not seem to capture nucleation events that occur in summer. This can be seen in Fig. 8(b) and 8(c), which show that the SS_HIGH experiment underestimated CN during such events.

Line 281 (BULKMSA figure reference) There appears to be an error in the figure reference: BULKMSA is shown in Fig. 3a, not Fig. 3b as cited. Please verify and correct. Additionally, it would be helpful to clarify whether the BULKMSA line overlaps substantially with MSA_SVOClike in Fig. 3a, as the two lines are difficult to distinguish visually.

Corrected

Line 281 (MSA_SVOClike figure reference) Similarly, MSA_SVOClike is plotted in Fig. 3a, not Fig. 3c as stated in the text. Please correct the figure reference throughout.

Corrected

Line 283 The authors could make the interpretation of Fig. 3c more explicit. The difference between BASECASE and SO4NUCL represents the isolated contribution of MSA to nucleation, since SO4NUCL uses sulphuric acid as the only nucleation precursor while all other settings remain identical. As shown in Fig. 3c, this difference is substantial in summer, indicating that MSA contributes more to Arctic NPF than sulphuric acid during this season, consistent with the observational findings of Boyer et al. (2024). This point can be stated more explicitly.

L288 : The results from the SO4_NUCL experiment are in agreement with these observations: the negligible contribution of sulphuric acid to nucleation results in low CN during the summer period.

Line 290 The DMS oxidation reaction producing SO_2 and MSA is given as Eq. 2 in the methods section, but is referenced here as Eq. 3. Please verify and correct.

Corrected.

Line 295 The authors state that modelled MSA concentrations are lower than observations by approximately one order of magnitude, which is a critical result bearing directly on the interpretation of the nucleation experiments. However, there is no figure of this model–observation MSA comparison in the main text or supplementary material. Please present this comparison explicitly.

We have added Fig. S3, as referenced in the answer in the comment to L206, to back this up and the text has been changed to reference the figure here as follows

L295: even though the modelled MSA concentration is lower than the measurements by about one order of magnitude (Fig. S3).

Line 307 The comparison between DMS_LOW, NUCLRATE_LOW, and MOSAiC observations in Fig. S2 is difficult to assess because the lines for these experiments are too thin and closely spaced to distinguish clearly, particularly during May–June 2020 where the authors report the largest deviations. Please increase line thickness and use more contrasting colours or line styles in this figure to improve readability.

We agree with the reviewer, this was an issue that we encountered when working with this plot. We tried replotting with thicker lines, but the reason for being unable to distinguish between the lines seems to be that the lines overlap. We have added text in the figure description to ensure that readers understand this as well.

Fig S2: Violin plot of the daily average CN from the MOSAiC expedition and the simulation, BASECASE, along the MOSAiC track. MOSAiC and BASECASE values are plotted as the left and right half violin, respectively, with the shaded area showing the probability density function of the CN values. The horizontal lines at the top and bottom of the violins represent the maximum and minimum, and the line in the middle is the median. The 11 simulations run for this study are shown as line plots, with the point corresponding to each month representing the median value for the month. We see significant overlap between the experiments DMS_LOW and NUCLRATE_LOW from late spring to early autumn, especially from July 2020 to September 2020.

Line 349 The discussion of CCN underestimation focuses primarily on particle size and hygroscopicity biases, but several other potentially important contributors are not considered. These include: (1) the absence of anthropogenically influenced aerosols associated with Arctic haze, which are likely important CCN in winter and spring; (2) missing iodic acid-driven nucleation, which, while primarily a summer process, can produce particles that grow to CCN sizes; (3) Arctic mineral dust, which, once chemically aged through condensational growth of H_2SO_4 or organics, can become hygroscopic and CCN-active; and (4) marine-derived organic compounds such as methanethiol, which may contribute to secondary aerosol formation and CCN activity over the open Arctic Ocean. Please discuss these missing processes in the context of the CCN underestimation.

We thank the reviewer for this comment and for highlighting several potentially important processes that may contribute to the simulated CCN biases. We have added relevant discussion on the above-mentioned processes in both 3.2.2, where relevant for CCN, and in the conclusion section discussing omitted or potentially poorly represented processes and the importance of future research.

Line 350 Please briefly state what particle size and hygroscopicity (κ) thresholds correspond to the transition between sub-CCN and CCN sizes in the Petters and Kreidenweis (2007) framework as implemented in TM5 at the 0.2% supersaturation used for comparison with MOSAiC observations.

We thank the reviewer for asking for this clarification. In order to provide a comprehensive answer regarding the treatment of CCN in TM5, we have answered this in an earlier question, from L108.

Line 371 The spatial maps in Fig. 6 show that NUCLRATE_HIGH leads to a decrease in CCN in some Arctic regions (Fig. 6l). Please provide a physical explanation for this result.

Here, an increased nucleation rate means that there is more competition for aerosol precursors with the same vapours being required for particle growth as well as nucleation. We have included the following to the text to explain this

L371: ...regions with higher CN also have higher CCN concentration. The only exception being the NUCLRATE_HIGH(Fig 6l) experiment in February 2020. Here we see a small decrease in Ccn concentration over North America even though the CN values increase over the area. This is due to the increased nucleation rate resulting in more competition for the aerosol precursors to grow and resulting in a smaller number of particles growing to bigger size(Fig. S3(j)).

Line 421 The authors attribute the model's failure to capture the second blowing snow event to the low wind speed observed during that period, but do not speculate on what physical mechanism produced the CN spike in the observations. Was this event potentially associated with Arctic haze transport rather than local blowing snow? Or could it reflect a local emission mechanism not captured by wind-speed-dependent parameterisations, such as sublimation of blowing snow particles at low wind speeds? Please provide a more explicit discussion of the possible causes.

We thank the reviewer for bringing this to our attention. The blowing snow event in Fig. 8 (a) has been described in detail in Gong et al. (2023). In the paper they mention that for the sources for both aerosol events. For the aerosol event that started on 09 December 2019 and lasted till the end of the plotted period, the initial peak was caused by a blowing snow event and the higher concentration in the latter period is caused by aerosols due to biomass burning. The text will be changed to

L421 : Fig. 8a is a blowing snow event that takes place along the MOSAiC track, ~~which has been discussed in detail in Gong et al. (2023).~~ Gong et al. (2023) conducted source attribution for the event and identified two aerosol ~~There are two blowing snow~~ events in the time period, one from 2 December 2019 to 6 December 2019 and another ~~shorter one~~ from 7 December 2019 to ~~8~~ 10 December 2019. The first event is primarily driven by blowing snow. For the second event, the initial peak, till 8 December 2019, is driven by blowing snow and the later part of the event is primarily driven by biomass burning. The model seems to behave differently for the two events. We can see that the model captures the first event to some extent, especially in the SS_HIGH. The first period has higher wind speed, which could be the reason for better agreement between the model and observations. However, for the second event, the wind speed is much lower, but a bigger spike in CN is observed. This could be the reason why the model is unable to capture this event. Simple parameterisations which do not include factors other than wind speed and SIC will also not be able to capture this event.

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