Reviewer 2

[C2-1] This study investigates the impact of newly implemented 2-moment cloud microphysics scheme on the simulated aerosols and their interactions with radiation and clouds in a global model at 14-km resolution. The authors find that with the new scheme the simulated aerosol burden is overall decreased, which is (said) mainly due to a faster cloud to precipitation conversion (as suggested by the increased RPCW ratio). Consequently, the direct effects of all aerosols (natural + anthropogenic) are reduced. On the other hand, the indirect effect (forcing caused by aerosol-cloud interactions) of anthropogenic aerosols is greatly increased (about doubled). The authors state that there are two possible reasons: 1) the cloud water adjustment changes; 2) the lower background aerosol AOT (burden, CCN).
Evaluating the impact of cloud microphysics change on the aerosol lifecycle and aerosol-cloud interactions is important for global aerosol-climate model development, especially for high-resolution applications. Results from this study will serve as a reference for model development and help the modeling community to better understand the behavior of this model. Therefore, I think this study fits the scope of GMD well and it could be a useful reference. However, I think the current manuscript needs to be significantly improved, especially in evaluating the simulated cloud microphysics responses to aerosol perturbations and in explaining the differences in the simulated aerosol indirect effects.

[A2-1] We appreciate your great contributions to improve our manuscript. Your comments and suggestions are very helpful and motivate us to investigate the results more deeply. The evaluation about the simulated cloud microphysics responses to aerosol perturbations and explanation about the differences in the simulated AIE were greatly modified to the revised manuscript.
Through the revision, we modified figures and tables as follows:

- Figures 1, 2, 5, E1 and E2: We changed the color of the zonal averages (NDW6 blue, NSW6 orange, as in the other figures).
- Figures 2 and 5: We replotted the model results in white for grids with missing satellite data. We replotted the zonal averages of the model results by eliminating the grids with missing satellite data.
- Figure 4: We changed the caption named “references” to “AeroCom”.
- Figure 7: We changed the subtitle named “AOD” to “AOT”.
- Figure 9: We provided IRFari of all and each aerosol for shortwave & longwave at the TOA & the surface under all & clear sky conditions. We changed the caption named “references” to “Kinne19” and “Thorsen21”.
- Figure 10: We removed the results of IRFari because they were shown in Figure 9. Instead, we newly added the results of ERFari and sum of ERFari and ERFaci. We also modified the ERFaci for shortwave to net ERFaci (for both shortwave and longwave).
- Figure 11: We modified ∂AOT to ∂CCN. We added new parameters such as ∂CDNC, ∂CDR, ∂CA, ∂CF, and net ERFaci to further explore ACI.
- Figure 12: We replaced Table 3 in the original manuscript to Figure 12 in the revised manuscript by adding relevant parameters such as ∂CCN, ∂CDNC, ∂CDR, ∂CA, ∂CF, and net ERFaci.
- Figure 13: To explain possible overestimations of the NSW6-simulated Twomey effect, we newly plotted global budgets of the annual averages of the NDW6- and NSW6-

simulated Qc (mixing ratio of cloud droplets) and CDNC (cloud droplet number concentrations).

- Table 1 in the original manuscript: We removed it and added a paragraph to explain the HRM and LRM as references in section 2.5 in the revised manuscript.
- Table 1 in the revised manuscript: We simply moved Table 2 in the original manuscript to Table 1 in the revised manuscript.
- Table 2 in the revised manuscript: We showed global and annual mean values of ERFari, ERFaci, and the sum of ERFari and ERFaci for shortwave, longwave, and net radiation under both all-sky and clear-sky conditions.
- Table A1: We newly added the statistical metric to compare results in this study with the references by Goto et al. (2020).
- Table A2: We simply moved Table A1 in the original manuscript to Table A2 in the revised manuscript, with two exceptions. One, we changed "References" to "References from model results". Second, we changed the SO_2 production value from 67.5 to 67.7.

Please note that some English was corrected in the revised manuscript.

Major comments:

[C2-2] 1. Since the focus of this study is on the impact of cloud microphysics on aerosol simulation. It's vital to show the cloud microphysics property changes in the simulation. I would recommend the authors to check the cloud water mass and number budgets in the simulations and evaluate the impact of aerosol perturbation on the budget changes. A good example is shown in Salzmann et al. (2010) in ACP.

[A2-2] Thank you for your comments and providing a good example for comprehensive analysis of the budget changes. But unfortunately, we didn't output the budgets of cloud water mass and number like Salzmann et al. (2010), and due to our computer resource limitations, we are unable to recalculate them. We would like to calculate these budgets and analyze them carefully in the future study. Instead, we calculated differences in the cloud water mass (mixing ratio) and number budgets between the PD and PI aerosol conditions to see the impacts of aerosol perturbation on cloud budget changes. Their global averages in the model heights were plotted in Figure 13 in the revised manuscript. We think this figure clearly indicates a possible overestimation of the cloud water number concentrations (CDNC) in NSW6 above 3 km, because the CDNC sink processes such as accretion, auto-conversion, and evaporation are not considered in NSW6. The following discussions were added to the revised manuscript (Lines 543-556):

"To evaluate the Twomey effect in NDW6 and NSW6, the global averages of differences in the mixing ratios and number concentrations for clouds between the PD and PI aerosol conditions are plotted in Figure 13. The changes in ∂Qc in both NDW6 and NSW6 are positive at most heights, so Qc increases as aerosols increase. This is consistent with the results of ∂LWP shown in Figures 11 and 12(e). The largest value of ∂Qc in both NDW6 and NSW6 occurs at a height of approximately 1.5 km, but the largest values in NDW6 are distributed up to a height of 2 km. Above a height of 3 km, ∂Qc in NDW6 is positive, whereas ∂Qc in NSQ6 is close to zero or negative. This difference in ∂Qc between NDW6 and NSW6 is possibly caused by the differences in the simulated supercooled liquid water in mixed-phase clouds, as mentioned in section 3.1. For ∂CDNC, the largest values in NDW6 occur at a height of 1.2 km, which is slightly lower than the height where the largest value of ∂Qc occurs. This reflects the vertical structure of typical

clouds in NDW6. In contrast, the vertical profile of ∂CDNC in NSW6 is different from that of ∂Qc because NSW6 cannot predict CDNC and adopts ∂CCN. This implies that ∂CDR is anti-proportional to ∂Qc from the surface to the 4-km height and has a low value below the 1.5-km height and the largest value at a height of approximately 1.5 km. Specifically, above a height of 3 km, where ∂Qc is close to zero and ∂CDNC has a positive value, ∂CDR should be small. The possible overestimation of ∂CDR in NSW6 represents possible overestimation of the Twomey effect in NSW6."

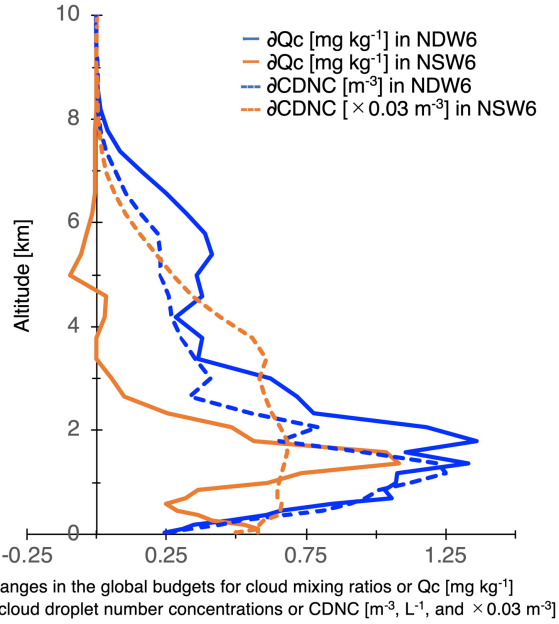

Figure (Figure 13 in the revised manuscript): Global budgets of the annual averages of the NDW6- and NSW6-simulated Qc (mixing ratio of cloud droplets), the NDW6-simulated CDNC (cloud droplet number concentration), and the NSW6-simulated CDNC (cloud droplet number concentration, which is equal to CCN number concentrations)

[C2-3] 2. The authors emphasized the liquid water adjustment (2nd indirect effect) in the abstract and summary. What is the impact of the Twomey effect in the model? The cloud droplet number changes (PD vs PD and PD-PI vs. PD-PI between the simulations) and the impact on effective radius and cloud albedo should be evaluated.

[A2-3] Thank you very much for your comments on the difference in the ERFaci between NDW6 and NSW6. According to many suggestions of you and other reviewers, we checked other parameters, such as CCN, CDNC, CDR (effective radius of clouds), CF (cloud fraction), CA (cloud albedo), and net ERFaci and largely modified our analysis in the revised manuscript (Lines 526-542) as follows:
"Given the verification of the NICAM-simulated CRF above, the simulated ACI due to anthropogenic aerosols is discussed by comparing the results between NDW6 and NSW6 for simulations with aerosol and precursor gas emissions for the preindustrial (PI), mentioned in section 2.3, and the present day (PD). Figure 11 shows the global maps of changes in the simulated CCN at 1-km heights, cloud droplet number concentrations (CDNC) at 1-km heights only for NDW6, cloud droplet effective radius (CDR) at 1-km heights, LWP, cloud albedo (CA), cloud fraction (CF) at 1-km height and net ERFaci between PD and PI. Figure 12 also shows the average values of the selected regions. These figures show that the global average of the NDW6-calculated ∂CCN at a 1-km

height is estimated to be 16.70 cm^{-3} (∂CCN), whereas that in NSW6 is estimated to be 19.59 cm^{-3} (∂CCN). The NDW6-calculated ∂CCN values are lower than the NDW6 results. In ∂CDNC, the NDW6-estimated values are +0.70 cm^{-3} (global), +4.22 cm^{-3} (the United States), +4.58 cm^{-3} (Europe), +3.57 cm^{-3} (East Asia), and +0.34 cm^{-3} (India). However, the CDNC used in NSW6 is equal to the CCN concentrations due to the ignorance of sink process in the CDNC in NSW6, as mentioned in section 2.1, so the difference in ∂CDNC between NDW6 and NSW6 is very large. As a result, the NSW6-simulated ∂CDR values at the 1-km height are much larger than the NDW6 results. The NDW6-estimated ∂CDR is -0.17 μm (global), -0.64 μm (the United States), -0.55 μm (Europe), -0.91 μm (East Asia), and -0.33 μm (India), whereas the NSW6-estimated ∂CDR is -0.34 μm (global), -0.93 μm (the United States), -0.91 μm (Europe), -1.20 μm (East Asia), and -0.81 μm (India). As shown in Figure 11, the NDW6- and NSW6-estimated ∂CDR values are negative near the industrial regions where the ∂CCN is large. Therefore, the approximately 15% difference in ∂CCN between NDW6 and NSW6 causes the approximately 50% difference in ∂CDR. This indicates that the Twomey effect, i.e., the response of ∂CDR to ∂CCN, in NSW6 is larger than that in NDW6."

In addition to the above discussion about ∂CCN, ∂CDNC, and ∂CDR, we discussed ∂LWP, ∂CA, ∂CF, and ERFaci in the revised manuscript (Lines 557-582) as follows:

"As mentioned above, the NDW6-calculated ∂LWP values are higher than the NSW6 results by three times in global averages. The NDW6-estimated values are +2.12 g m^{-2} (global), +7.52 g m^{-2} (the United States), +15.45 g m^{-2} (Europe), +8.77 g m^{-2} (East Asia), and +3.36 g m^{-2} (India), whereas the NSW6-estimated values are +0.65 g m^{-2} (global), +4.96 g m^{-2} (the United States), +2.52 g m^{-2} (Europe), +2.62 g m^{-2} (East Asia), and -0.44 g m^{-2} (India). The positive values in ∂LWP in both NDW6 and NSW6 could be caused by a decrease in auto-conversion due to the increase in CDNC. However, magnitudes of ∂LWP differ between NDW6 and NSW6, which is the largest in Europe among others, whereas the NDW6- and NSW6-simulated ∂CCN are close to each other in most regions. This appears to indicate that the cloud water susceptibility, defined as the difference in ∂LWP against ∂CCN from PD to PI conditions, is larger in NDW6 than in NSW6. Such a different susceptibility could be interpreted in terms of different complexities of hydrometeors interactions between NSW6 and NDW6, particularly whether or not the CDNC and rain drop number concentration (RDNC) are predicted. This generates different variabilities of CDNC and RDNC between the two schemes, possibly leading to the different susceptibilities. Nevertheless, more detailed analysis will be required in future studies to explore microphysical processes responsible for these different behaviors between the two schemes.

The horizontal distribution of changes in the simulated ERFaci is generally consistent with changes in the simulated ∂LWP (Figure 11). By decreasing the simulated ∂CDR, increasing the simulated ∂LWP from PI to PD, and increasing the simulated ∂CA and ∂CF at 1-km height, the negative values of the simulated ERFaci in industrial regions, such as the United States, Europe, and East Asia, increase in magnitude. The global annual averages of the net ERFaci value are estimated to be -1.28 Wm^{-2} (NDW6) and -0.73 Wm^{-2} (NSW6). Both NDW6- and NSW6-estimated ERFaci values range within the results in IPCC-AR6 (Forster et al., 2021), i.e., -0.84 Wm^{-2} (-1.45 Wm^{-2} to -0.25 Wm^{-2}), and the Radiative Forcing Model Intercomparison Project (RFMIP) (Smith et al., 2020), i.e., -0.81±0.30 Wm^{-2}, by considering the uncertainty caused by the assumption in the PI conditions. The magnitude of the ERFaci value in NDW6 is larger than that in NSW6 by 0.55 Wm^{-2} (approximately 43% of the ERFaci value in NDW6), whereas the NDW6-simulated aerosol loadings are smaller than the NSW6 results, as shown in the previous

sections. Figure 12 shows that the negative NDW6-estimated ERFaci values are larger than the NSW6-estimated ERFaci values by 2.33 Wm^{-2} (US), 3.22 Wm^{-2} (Europe), 1.10 Wm^{-2} (East Asia), and 0.89 Wm^{-2} (India). Therefore, it was suggested that the ERFaci due to the cloud lifetime effect in NDW6 was larger than that in NSW6 due to the Twomey effect, although the NSW6-simulated ERFaci certainly includes some bias due to the overestimation of the Twomey effect."

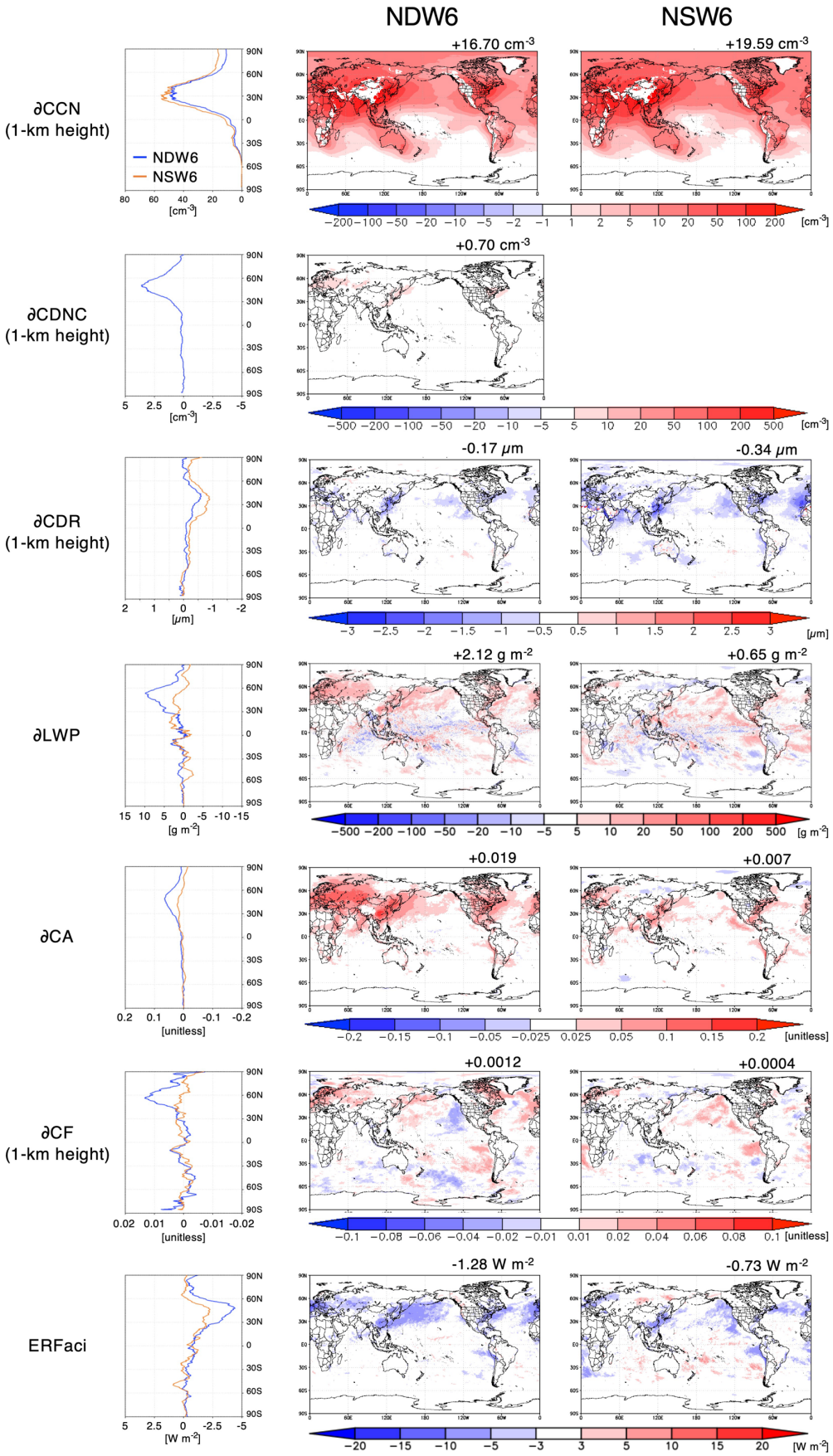
NDW6
NSW6
∂CCN
(1-km height)
NDW6
NSW6
[cm-3]
+16.70 cm-3
+19.59 cm-3
−200 −100 −50 −20 −10 −5 −2 −1 1 2 5 10 20 50 100 200 [cm-3]
∂CDNC
(1-km height)
[cm-3]
+0.70 cm-3
−500 −200 −100 −50 −20 −10 −5 5 10 20 50 100 200 500 [cm-3]
∂CDR
(1-km height)
[μm]
-0.17 μm
-0.34 μm
−3 −2.5 −2 −1.5 −1 −0.5 0.5 1 1.5 2 2.5 3 [μm]
∂LWP
[g m-2]
+2.12 g m-2
+0.65 g m-2
−500 −200 −100 −50 −20 −10 −5 5 10 20 50 100 200 500 [g m-2]
∂CA
[unitless]
+0.019
+0.007
−0.2 −0.15 −0.1 −0.05 −0.025 0.025 0.05 0.1 0.15 0.2 [unitless]
∂CF
(1-km height)
[unitless]
+0.0012
+0.0004
−0.1 −0.08 −0.06 −0.04 −0.02 −0.01 0.01 0.02 0.04 0.06 0.08 0.1 [unitless]
ERFaci
[W m-2]
-1.28 W m-2
-0.73 W m-2
−20 −15 −10 −5 −3 3 5 10 15 20 [W m-2]

Figure 11: Global distributions of the annual averages of the NDW6- and NSW6-simulated CCN change at 1-km height (∂CCN), CDNC (cloud droplet number concentrations, which in NSW6 is equal to the CCN concentrations in NSW6 due to the ignorance of sink process in the CDNC in NSW6) change at 1-km height (∂CDNC), CDR (cloud droplet effective radius for warm clouds) change at 1-km height (∂CDR), LWP change (∂LWP), CA (cloud albedo) change (∂CA), CF (cloud fraction) change at 1-km height (∂CF), and net ERFaci by comparing the results between NDW6 and NSW6 for simulations with aerosol and precursor gas emissions for the present and the preindustrial era. The number located in the upper right in each panel represents the global and annual mean value. The results at 1-km height also include areas with elevations higher than 1-km height in white.

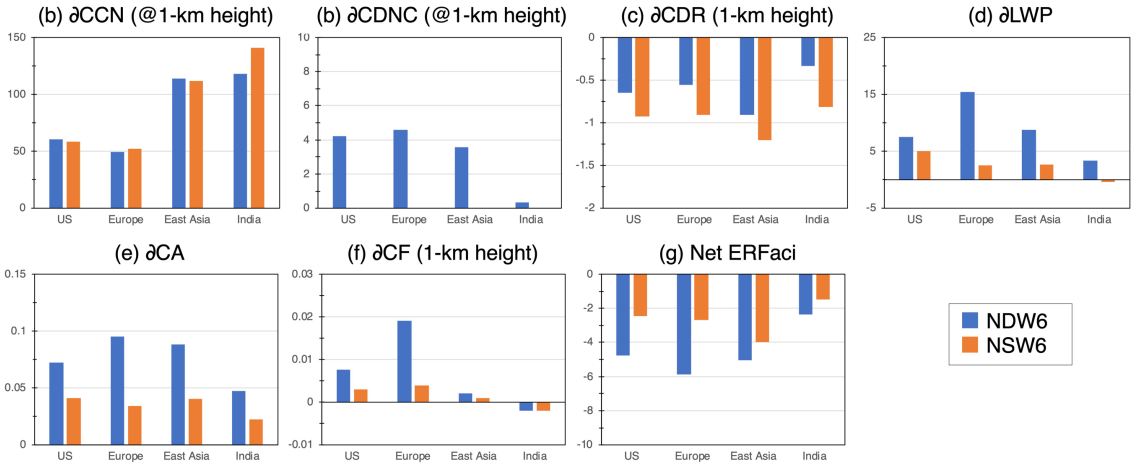

Figure 12: Regional averages of the differences in CCN at 1-km height, CDNC (cloud droplet number concentration only in NDW6), CDR (cloud droplet effective radius at 1-km height), LWP, CA (cloud albedo), CF (cloud fraction at a 1-km height), and net ERFaci between the preindustrial and the present days. The regions are defined as US (90°W-60°W, 30°N-50°N), Europe (0°E-30°E, 40°N-60°N), East Asia (110°E-140°E, 20°N-50°N), and India (70°E-90°E, 10°N-35°N).

[C2-4] 3. It is unclear to me why the authors include the comparison against HRM and LRM (from the other study) and why the discussions are only for some of the fields, but not the others. If the authors want to include this part, the title should be revised (to reflect the impact of resolution and time stepping changes).

[A2-4] Thank you for your comment on the use of HRM and LRM in this study. We treated these results (HRM and LRM) as references to evaluate the model results of NDW6 and NSW6 in this study. In fact, the main target in this study is comparisons between NDW6 and NSW6 in the text, and as a supporting information, comparisons between NSW6, HRM and LRM were mentioned in Appendix. To clarify this, we added a section 2.5 “Reference datasets” in the revised manuscript (Lines 281-302) to explain the information about HRM and LRM as references.
“Our previous model results provided in Goto et al. (2020) using NICAM.16 at a global 14-km high resolution (hereafter referred to as the HRM) and a global 56-km low

resolution (hereafter referred to as the LRM) are used as references to compare the NICAM results. As mentioned in section 2.1, the number of vertical layers is set at 38, and the timestep is 1 minute in both the HRM and LRM. The integration periods in both the HRM and LRM are 3 years as climatological runs. The emission inventories, i.e., 2010 for anthropogenic sources, climatological average in 2005-2014 for biomass burning, and natural sources in the present era, and the nudged SST and sea ice in this study are identical to those in both the HRM and LRM, but the initial conditions in this study are different from those in both the HRM and LRM, which use the model results at the end of December after a 1.5-month spin-up. The initial conditions for the model spin-up are prepared by the reanalysis datasets of the National Centers for Environmental Prediction (NCEP) Final (FNL) (Kalnay et al., 1996) in November 2011. In the cloud microphysics and autoconversion modules, NDW6 coupled to Seifert and Beheng (2006) and NSW6 coupled to Khairoutdinov and Kogan (2000) are used in this study, whereas NSW6 coupled to Berry (1967) is used in both the HRM and LRM. The improvement in the aerosol module described in section 2.2 is also different from that in the HRM and LRM. The results of the HRM and LRM are useful for evaluating the current model results because the observations are limited in some parameters, such as aerosol global budgets and radiative forcings."
To evaluate our results in this study, the results of HRM and LRM are very useful, because the observation and other morel references are limited in some parameters. Therefore, we don't think we need to change the title by reflecting the impact of resolution and time stepping changes.

Detailed comments:
[C2-5] Page 1, Line 21: It would be useful to report the net effective aerosol forcing and ERFari (Ghan's method) as well.

[A2-5] Thank you for your comment on the ERF. According to your suggestions, we added extra forcings (ERFari, ERFaci, and ERFari plus ERFaci) in Figure 10 in the revised manuscript. We showed net ERFari values in abstract (Lines 21-24) and net ERFaci values (Lines 29-31) in the revised manuscript.

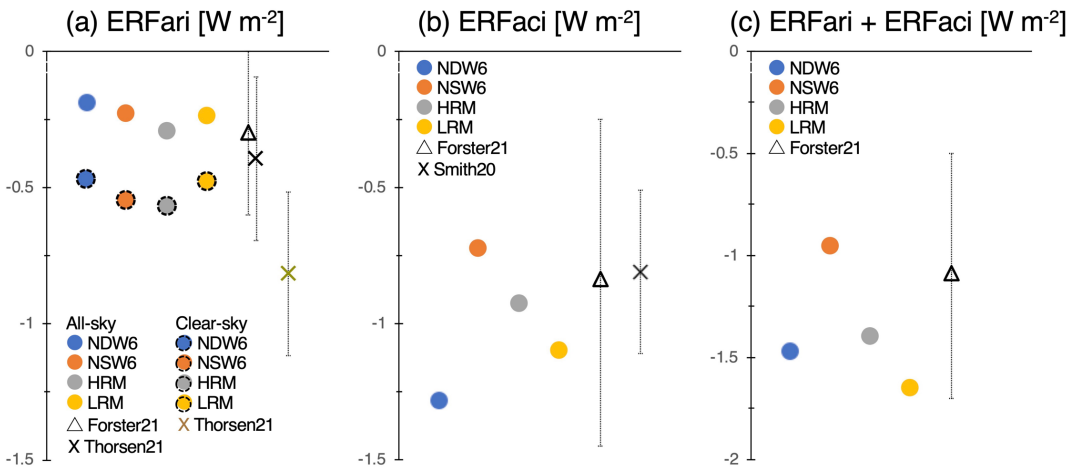

Figure 10 in the revised manuscript: Global and annual mean values of (a) effective radiative forcing for anthropogenic aerosol-radiation interaction (ERFari) for shortwave

and net (sum of shortwave and longwave) radiation, (b) ERFaci for anthropogenic aerosol-cloud interaction, and (c) the net ERF (sum of ERFari and ERFaci). All units are in W m^{-2}. In ERFari, the reference of Forster21 is estimated in the net radiation by IPCC-AR6 or Forster et al. (2021), whereas the reference of Thortsen21 is estimated in the shortwave radiation by Thorsen et al. (2021). The reference for Smith20 is Smith et al. (2020). The values are also listed in Table 2.

[C2-6] Page 1, Line 23: e.g., -> i.e.,

[A2-6] Corrected.

[C2-7] Page 1, Line 31: Why is the 2nd indirect effect (LWP adjustment) so important? How about the Twomey effect in this model?

[A2-7] The details are mentioned in A2-3, so here our modification in Abstract is shown. We modified this in the revised manuscript (Lines 31-34) as follows:
"The magnitude of the ERFaci value in the NDW6 experiment is larger than that in the NSW6 result due to the differences in the susceptibility of the simulated cloud water to the simulated aerosols between NDW6 and NSW6 and the overestimation of the Twomey effect in NSW6 caused by ignorance of sink process in the cloud droplet number concentrations."

[C2-8] Page 2, Line 32: It would be better to look at the ERFaci vs. CCN relationship, rather than ERFaci vs. AOT.

[A2-8] Thanks for your suggestion. We replaced AOD to CCN in the analysis of ACI in the revised Figures 11 and 12.

[C2-9] Page 2, Line 37: better change "aerosol nucleation" to "aerosol activation" to avoid confusion.

[A2-9] Thanks for your correction. We agree.

[C2-10] Page 3, Line 68-69: "difference in the simulated aerosol mass concentrations" Do you mean surface concentrations or mass burden?

[A2-10] Surface concentrations. We added "at the surface" to the revised manuscript (L72).

[C2-11] Page 3, Line 72: Please provide the reference.

[A2-11] Thanks for your comment. This past research is Goto et al. (2020) in the revised manuscript (Line 75).

[C2-12] Page 3, Line 93: Are they climatological runs, or AMIP-style simulations with transient prescribed SST? How is the model initialized?

[A2-12] Thank you for the comments. The experimental conditions were a bit unclear in the original manuscript. The simulations were climatological fields. All the experiments with both NDW6 and NSW6 were carried out for 6-years after the 1-month spinup calculation. The initial conditions for the model spinup were obtained from the end of the 1-year aerosol simulations coupled to NSW6 without nudging the meteorological fields under the present era. We added Section 2.3 named “Experimental conditions” to the revised manuscript (Lines 200-227).

[C2-13] Page 4, Line 109-110: Water vapor is not a hydrometeor.

[A2-13] Thank you for your correction. Yes, it is. We changed this term “hydrometeor” to “water substances” in the revised manuscript of two parts (Lines 112 and 119). We also changed “hydrometeors, except for water vapor” to “hydrometeors” by removing “except for water vapor” in the revised manuscript (Line 156).

[C2-14] Page 4, Line 112: How is the updraft velocity calculated in the model?

[A2-14] Even in 14-km simulations, the updraft velocity is needed to be parameterized using Lohmann et al. (1999). In the revised manuscript (Lines 115-116), we added the following sentence:
“This parameterization is a function of the parameterized updraft velocity with turbulent kinetic energy (Lohmann et al., 1999), aerosol sizes, and aerosol chemical composition”.

Reference:
Lohmann, U., Feichter, J., Chuang, C. C., and Penner, J. E.: Prediction of the number of cloud droplets in the ECHAM GCM, J. Geophys. Res., 104 (D8), 9169-9198, 1999.

[C2-15] Page 4, Line 116-117: What is the background CCN value? Why is it needed? How is the updraft velocity calculated?

[A2-15] Sorry for confusing you. This background CCN was set at NDW6-SN14 and NDW6-S15 (NOT NDW6-G23), because their CCN is not calculated from the aerosol physical module. We added this information to the revised manuscript (Line 121). The calculation method of the updraft velocity is answered in [A2-14].

[C2-16] Page 4, Line 126-127: "although rain does not directly change cloud water” Doesn’t auto-conversion in NSW6 affect cloud water? Also, could you elaborate why the impact of aerosol on cloud water is overestimated?

[A2-16] Thank you for your comment. These parts confuse the reviewers (we got comments on this point from another reviewer). We removed this from the revised manuscript.

[C2-17] Page 4, Line 134: Better present this (cloud/precipitation observational data) in a separate section.

[A2-17] Thank you for your suggestion. Yes, we move this part to new section 2.4 named "Observations" in the revised manuscript.

[C2-18] Page 5, Line 161-162: How is the aqueous chemistry production handled in the original model?

[A2-18] Thank you for your comment on the sulfate modeling. In the original model, the timestep (more than 1 minute) was longer than that in this study, so in this process we assumed the sulfate formed in the clouds by aqueous-phase oxidation at the current timestep is scavenged by the rainout process at the same timestep. However, when the vertical layer is finer and the timestep is shorter than those in the previous studies, this assumption may not be applicable. The critical value of the timestep is unclear, but we think the new assumption (is not scavenged by the rainout process at the same timestep) can be applicable in this study. We added the following sentence to the revised manuscript (Lines 160-162):
"Because the model timestep was more than 1 minute in previous studies (Goto et al., 2020), the original model assumes that the sulfate formed in clouds by aqueous-phase oxidation is scavenged by the rainout process at the same timestep."

[C2-19] Page 6, Line 173: Which year of emission is used?

[A2-19] Thanks for your comment. After moving the emission part to section 2.3 named "Experimental conditions" in the revised manuscript, we added the information about the emission year as follows:
"The emission fluxes used in this study are the Hemispheric Transport of Air Pollution (HTAP)-v2.2 (Janssen-Maenhout et al., 2015) for BC, organic carbon (OC) and SO_2 from anthropogenic sources in 2010 and the Global Fire Emission Database (GFED) version 4 (van der Werf et al., 2017) for BC, OC and SO_2 from biomass burning in climatological average from 2005 to 2014."
These emissions are also used in Goto et al. (2020).

[C2-20] Page 7, Line 207: To better compare with other models, it would better to use 1850 aerosol emissions, rather than using zero emissions.

[A2-20] Thank you for your comment on this assumption. Yes, as you said, a use of the 1850 aerosol emissions is informative for other studies. This point is important but not discussed in the original manuscript. We wanted to recalculate the simulations using the 1850 aerosol emissions, but unfortunately cannot do them due to the limitation of our

computer resources. So, we would like to speculate the uncertainty of the ERF caused by the different assumption of the preindustrial era. As a reference, the results in Hoesly et al. (2018) are shown in Table. We added the following discussions to the revised manuscript (Lines 217-227):

"In the preindustrial experiments, the anthropogenic emission fluxes of BC, OC and SO_2 are assumed to be zero in this study. Hoesly et al. (2018) estimated global averages of the differences in the emission amounts of anthropogenic sources between 1850 and 2010 to be 2.1% (sulfate), 12.0% (BC), and 22.7% (OC). The residential sector has the largest contribution to the total anthropogenic emissions in the preindustrial era. Takemura (2020) calculated the IRFari due to anthropogenic sulfate under the conditions of 0% and 30% of the present emissions and found that the difference in the IRFari was within 0.03 Wm^{-2}. Therefore, differences in the assumptions for the preindustrial era between this study and other studies, such as IPCC-AR6 (Szopa et al., 2021), will result in a difference in the IRFari due to anthropogenic sources of at most 0.05 Wm^{-2}. Takemura (2020) also calculated ERFari and ERFaci due to anthropogenic sulfate under the conditions of 0% and 30% of the present emissions and found that the difference in ERFari plus ERFaci was within 0.2 Wm^{-2}. These are possible uncertainties in the estimated radiative forcings due to anthropogenic sources in this study, but these magnitudes are smaller than the difference between NDW6 and NSW6 in this study, as shown in section 4."

We also added the following sentence to the summary (Lines 651-657) in the revised manuscript:

"As mentioned in section 2.3, the assumption of the preindustrial conditions for aerosols can cause possible differences in the aerosol radiative forcing due to the anthropogenic sources between this study and other studies, such as IPCC-AR6 (Szopa et al., 2021). This study assumes that the anthropogenic emission fluxes of BC, OC and SO_2 are zero in the preindustrial conditions, whereas other studies often use them in 1750 or 1850 provided by Hoesly et al. (2018). Using the results of MIROC by Takemura (2020), the possible difference in the aerosol radiative forcing due to the anthropogenic source will be at most 0.05 Wm^{-2} (IRFari) and 0.2 Wm^{-2} (ERFari plus ERFaci)."

Table Emission amounts in 1850 and 2010 years by Hoesly et al. (2018)

Anthropogenic Source	1850	2010	2010-1850 [%]
SO_2 [$ktSO_2$]	2,481	115,487	2.1
BC [ktC]	934	7,755	12.0
OC [ktC]	4,262	18,755	22.7

Reference:

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750-2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geosci. Model Dev., 11, 369-408, doi:10.5194/gmd-11-369-2018, 2018.

Takemura, T.: Return to different climate states by reducing sulphate aerosols under future CO_2 concentrations, Sci. Rep., 10, 21748, doi: 10.1038/s41598-020-78805-1, 2020.

[C2-21] Page 7, Line 210: Better present this (aerosol observational data) in a separate section.

[A2-21] Thank you for your suggestion. Yes, we moved this part to new section 2.4 named “Observations” in the revised manuscript.

[C2-22] Page 10, Line 301-302: “An increase in the RPCW leads to an increase in the aerosols that are dissolved into clouds,” RPCW increase can only enhance the turnover time of cloud liquid. Does the model consider cloud processing of aerosols?

[A2-22] Thank you for your comment. Yes, the model considers the sulfate production in clouds, so this expression is not accurate. To express this more simply, we modified this in the revised manuscript (Lines 369-370) as follow:
“An increase in the RPCW leads to an increase in the aerosols that are dissolved into raindrops and are removed from the atmosphere”.

[C2-23] Page 10, Line 322: Please define HRM.

[A2-23] Thank you for your suggestion. As we answered in [A2-4], we added a new section 2.5 named “Reference datasets” to the revised manuscript (Lines 281-302).

[C2-24] Page 12, section 4: the ARI and ACI represent different aerosol forcing/effects. Better report both the effective aerosol forcing (ERF_aer_total, ERF_aer_ACI, ERF_aer_ARI) of anthropogenic aerosols and the direct effect (IRF_ARI) of total aerosols. It would be also useful to report the surface forcings.

[A2-24] Thank you for your comment on the ERF. This information of these forcings become very useful for other studies. As you suggested, in the revised Figure 9 we provided IRFari for all and each aerosol for shortwave & longwave at the TOA & surface under all & clear sky conditions. In the revised Figure 10, we showed ERFari, ERFaci, and ERF_total (ERFari+ERIaci). We also added Table 2 (Global and annual mean values of ERFari for anthropogenic aerosol, ERFaci for anthropogenic aerosol-, and the net ERF (sum of ERFari and ERFaci) for shortwave, longwave, and net (sum of shortwave and longwave) radiation under the all-sky and clear-sky conditions) to the revised manuscript.

[C2-25] Page 14, Line 451: The discussion here is a bit vague. Some further analysis is needed to explain the difference.

[A2-25] Thank you for your comment on this statement about a possible reason for the difference in the ERFaci between NDW6 and NSW6. As mentioned in the original manuscript, Carslaw et al. (2013) (and Wilcox et al. (2015) as well) pointed out that the different baseline of AOT fields can provide the difference in the ERFaci between two experiments, even if the difference in the AOT between two experiments is small. In this study, the different baseline of AOT between NDW6 and NSW6 under the present days

can be found up to 20%, so we thought this can be a possible reason of the difference in the ERFaci between NDW6 and NSW6 in the original manuscript. However, when we looked at CCN, which is more sensitive to the ERFaci, the different baseline of CCN at 1-km height between NDW6 and NSW6 under the preindustrial days was smaller even in Europe where the difference in the ERFaci between NDW6 and NSW6 was the largest among the regions. Therefore, in the revised manuscript we concluded that the difference in the baseline of aerosols between NDW6 and NSW6 did not cause the difference in the ERFaci between NDW6 and NSW6. We modified this in the revised manuscript (Lines 583-588) as follows:
"Carslaw et al. (2013) and Wilcox et al. (2015) pointed out that the different baselines of aerosol fields can provide small differences in ERFaci between two simulations. As mentioned in the previous sections for aerosols, the NDW6-simulated aerosols are generally lower than the NSW6 results, for example IRFari is approximately 15% lower. However, the baseline of CCN at 1-km height between NDW6 and NSW6 under the PI conditions is not very different, so the difference in the baseline of aerosols between NDW6 and NSW6 does not cause the difference in ERFaci between the two simulations."
We also added the following sentence to the summary (Lines 645-647) in the revised manuscript:
"Other possible reason for the differences in the ERFaci between NDW6 and NSW6 is the different baselines of aerosol fields, as suggested by Carslaw et al. (2013) and Wilcox et al. (2015), but this is minor because the baseline of CCN at 1-km height between NDW6 and NSW6 under the PI conditions is not very different."
Based on the above discussion, we removed the related statements from the abstract from the original manuscript.

Reference:
Wilcox, L. J., Highwood, E. J., Booth, B. B. B., and Carslaw, K. S.: Quantifying sources of inter-model diversity in the cloud albedo effect, Geophys. Res. Lett., 42, doi:10.1002/2015GL063301, 2015.

[C2-26] Page 15, Line 483: What do you mean "performance improvement" here? Computational performance or better represented physical processes in the model?

[A2-26] Thank you for your comment. The term "performance" may confuse you, so we eliminated this. We simply expressed "these improvements", which include better model reproducibility in the simulated aerosol mass concentrations and AOT.

Reference:
Salzmann, M., Ming, Y., Golaz, J.-C., Ginoux, P. A., Morrison, H., Gettelman, A., Krämer, M., and Donner, L. J.: Two-moment bulk stratiform cloud microphysics in the GFDL AM3 GCM: description, evaluation, and sensitivity tests, Atmos. Chem. Phys., 10, 8037–8064, https://doi.org/10.5194/acp-10-8037-2010, 2010.