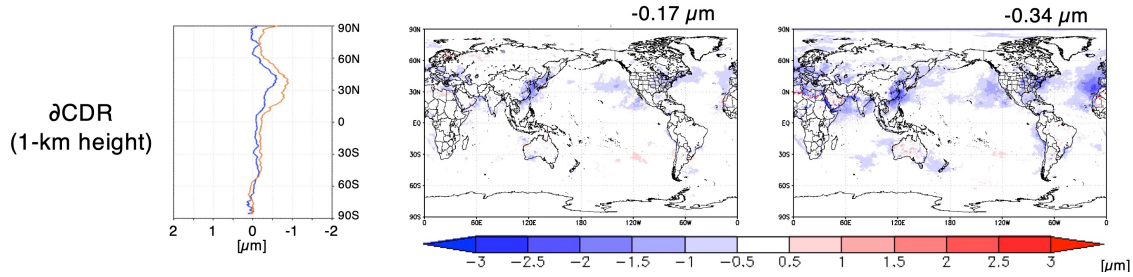
Dear Editor and Reviewers,

We appreciate your continued contributions to improving our manuscript. In this revision, we found our mistake in the calculation of the NDW6-simulated cloud droplet effective radius (CDR) used in Figures 11 and 12(c) in the original manuscript. The NDW6-simulated CDR should be calculated using the NDW6-simulated CDNC. However, the NDW6-simulated CDR was calculated using the NDW6-simulated CCN, as was the case for the NSW6-simulated CDR. Therefore, in the revised version, the NDW6-simulated CDR was newly calculated using the NDW6-simulated CDNC. In the original version, this CDR calculation was performed at each time step in the model, but due to the computer resource limitations, the 6-year integration cannot be performed again in the model. Therefore, we recalculated the CDRs for both NDW6 and NSW6 using monthly averages of cloud mass and number concentrations. Using these results, the global mean difference in the NDW6-simulated CDR between PD and PI, ∂CDR, was changed from -0.17 μm to -0.62 μm, whereas the NSW6-simulated ∂CDR was changed from -0.34 μm to -0.31 μm. The regional averages were also changed (Figure 13(c) in the revised manuscript). These were also shown in this reply. Note that this CDR recalculation does not affect other parameters (like LWP, ERFaci, so on), because this CDR was diagnostic and variable for the output.

Previous version



Revised version

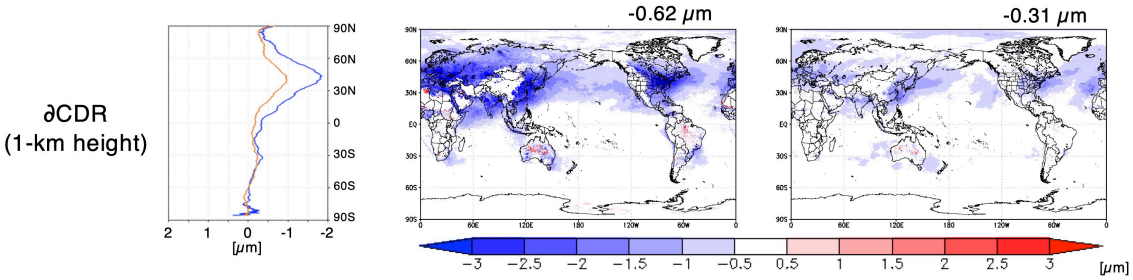


Figure: Modified ∂CDR used in Figure 11 in the previous version (Figure 12 in the revised manuscript).

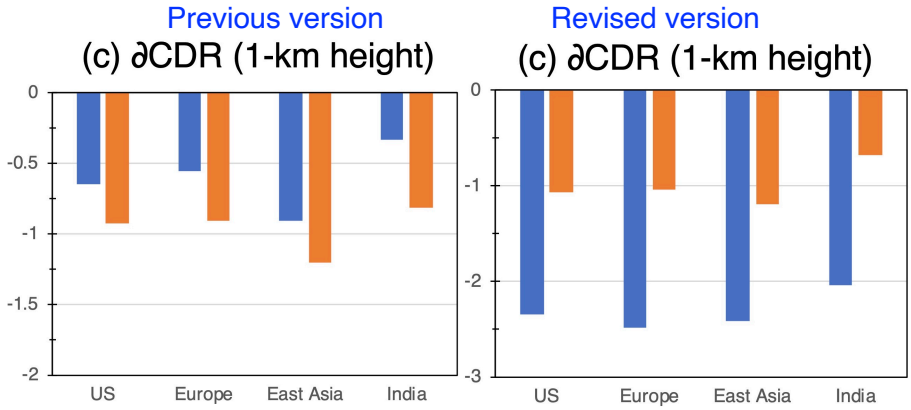


Figure: Modified ∂CDR used in Figure 12 in the previous version (Figure 13 in the revised manuscript).

In this revision, we realized that we did not evaluate CDR in the previous manuscript, so we newly added the evaluation to Table 1 and section 3.1 in the revised manuscript (Lines 134-142, 243, 250, 343-344, 346, and 348). The simulated CDRs were evaluated using CDRs at warm-topped clouds requiring specific conditions of cloud mixing ratio and cloud top temperature. To calculate the simulated CDRs, we newly integrated the model for only 1-year (due to limitations in available computer resources, 6-year integration was too heavy to recalculate). As a result, both the NDW6-simulated and NSW6-simulated CDRs at warm-topped clouds were lower than the MODIS-estimated results, but the NDW6-simulated CDR was generally closer to the observation compared to the NSW6-simulated results.

In the end, one of our main conclusions was slightly modified. In the previous version, we did not address the importance of the Twomey effect in NDW6, but we concluded that both the Twomey and the cloud lifetime effects in NDW6 were larger than in NSW6. This point was reflected to the revised version (Lines 32-33, 587-589, and 645-648). Except for this point, the conclusions remain.

In addition, we corrected some typographical and other errors.

- As noted in [A-2], a new figure (Figure 3) was added to the revised manuscript and the figure numbers were changed.
- IPCC(Forster et al., 2021): -0.25 Wm^{-2} (-0.45 Wm^{-2} to -0.05 Wm^{-2}) to -0.3±0.3 Wm^{-2}
- We added “the lower limit of” to Line 501 of the revised manuscript.
- We corrected the bars used in Thorsen21 in Figure 11(a).
- We corrected a typo in “and” in the caption of Table 2, deleted “shortwave and” from the caption of Figure 11, and corrected “sink processes” in the caption of Figure 12.
- We removed the explanation for the abbreviations, which are explained in the text, from the captions of Figures 12, 13, and 14.

We apologize for our mistake in the calculate of CDR in NDW6 and apologize for the correction made here, which should have been addressed in the first revision.

Sincerely yours,
Daisuke Goto

Reviewer 1

[C-1] 2nd review of Goto et al.
Thanks for making many of the changes suggested by the referees. I am mostly happy with the paper now. There are just a few places where some additional figures would be useful to show what is written in the text and where the text could be improved to make things clearer.

[A-1] We appreciate your great contributions to our manuscript. Your comments and suggestions are very helpful. We would like to answer each point below.

Major points
[C-2] L413 – "Therefore, the overestimation of the NSW6-simulated LWP in the eastern Pacific Ocean and Southern Atlantic Ocean effectively balanced the underestimation of the zonal averages of the simulated LWP and unexpectedly led to zonal LWP values closer to the MAC results."
[C-2-1] I found this difficult to follow. I suggest rewording to: "Therefore, the overestimation of the NSW6-simulated LWP in the eastern Pacific Ocean and Southern Atlantic Ocean effectively balanced the underestimation in the Western Pacific Ocean and Indian Ocean, which led to zonal LWP values that were closer to the MAC results."
[C-2-2] Also, it is still not easy to make this comparison from Fig. 2. Map plots of the bias of the models vs MAC would be helpful. As would averages (or the bias and/or LWP fields) for 0-30S as a function of the longitude.

[A-2] As for [A-2-1], we agreed your suggestion. We used this expression in the revised manuscript. As for [A-2-2], we newly added Fig 3 to the revised manuscript by calculating differences in the LWP among NDW6, NSW6, and MAC. To reflect this, the numbers of Figures are changed in the revised manuscript. We also prepared a new figure for 0-30S averages as a function of the longitude, but we thought that this information is overlapped with new Figure 3, so we didn't insert the specific figure to the revised manuscript (we showed it only in this reply).

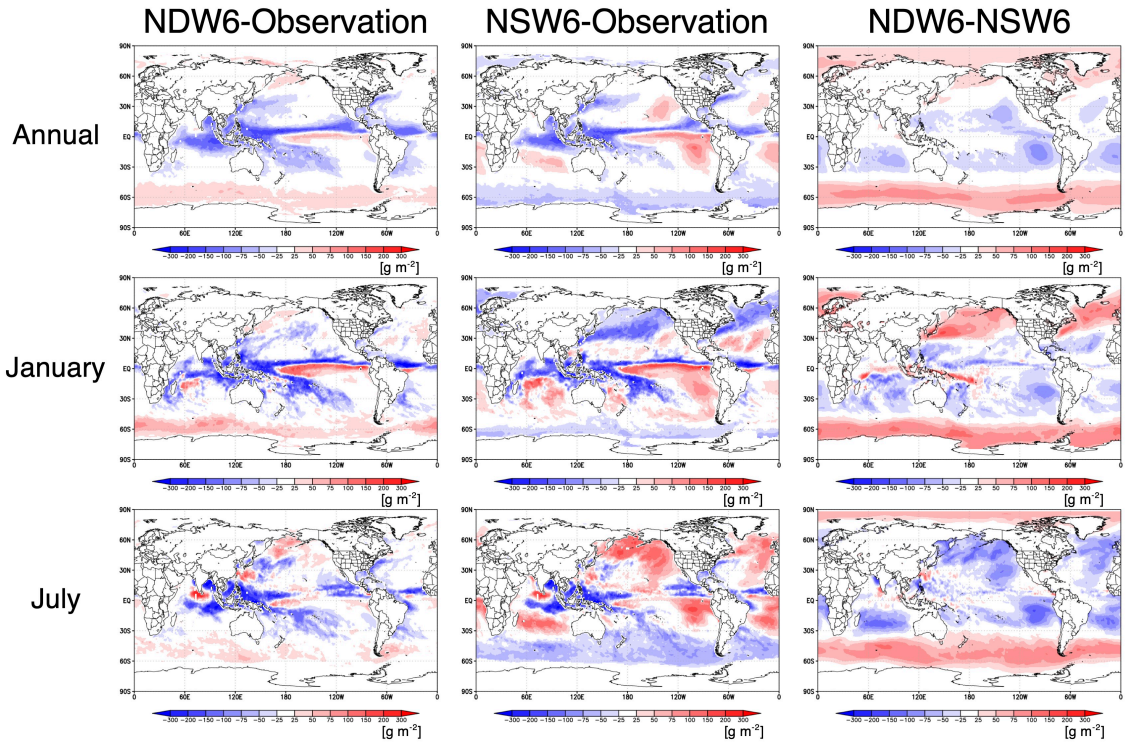


Figure 3 (new figure): Horizontal distributions of differences in LWP among NDW6, NSW6 and MAC over only the ocean as annual, January, and July averages. All units are in g m^{-2}.

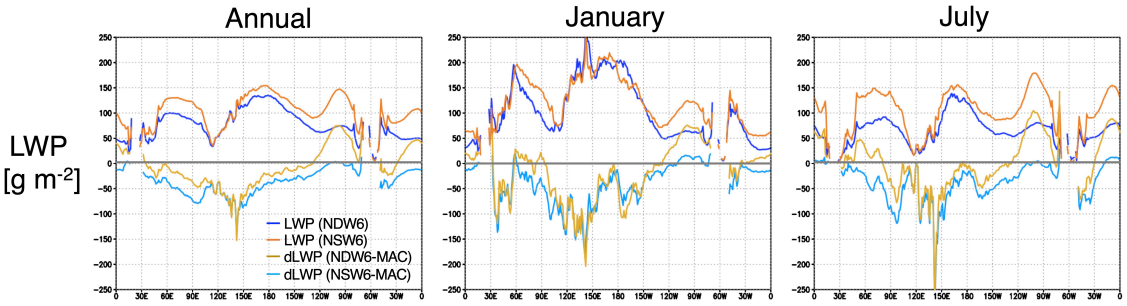


Figure (not used in the revised manuscript): Bias and LWP fields for 0-30S as a function of the longitude, as annual, January, and July averages.

[C-3] L614 – "When the simulated LWP is underestimated, the simulated aerosols are also underestimated, because the simulated precipitation is generally comparable to the observations in this study, as shown in Figure 1. When the simulated CDR is overestimated, the simulated CCN must be underestimated. This is consistent with the underestimation of the simulated aerosol. Therefore, if the negative biases in the simulated SWCRF are eliminated, the simulated aerosols will increase."

- This is quite speculative. It's not clear how well linked the LWP, CDR and precipitation in the clouds in question are. For frontal mid-latitude precipitation the relationships may not be as straightforward as for stratocumulus. Precipitation is likely to be determined by large scale meteorology. Plus, it's not clear how close to the observations the model

needs to be in order to allow a constraint on LWP or CDR. The comparison between models and observations is likely highly uncertain for precipitation. I'm not sure whether you need the extra text here (and it is quite difficult to explain what you are trying to say in a clear way!).

[A-3] Thank you for your comment. As you pointed out, this part is quite speculative. We largely modified this part as follows: "The NDW6-estimated SWCRF values are concluded to be better than the NSW6 results, but the underestimation of the NDW6-simulated SWCRF is mainly caused by the underestimation of the simulated LWP due to the underestimation of the simulated CDR shown in Table 1. The impacts of this negative biases in the simulated SWCRF and LWP on the aerosol simulations are still unclear due to complex interactions between aerosols, clouds, and precipitation."

[C-4] L663 – "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."
[C-4-1] You would need to add dCDR to Fig. 13 to show this.
[C-4-2] Generally it seems strange that such large changes in CDNC in NSW6 (assumed to be the same as dCCN) produce fairly small changes in CDR. E.g., Fig. 12 states that for the US there is around a 60 per cm3 increase in CDNC for NSW6. If we assume a baseline CDNC of 200 per cm3 (which is quite high) then the equates to a 260/200 = 1.3x increase. Since CDR is approximately proportional to CDNC^(-1/3) (assuming equal LWP) then this would be expected to lead to a 1.3^(-1/3) = 0.916x change in CDR. For a PI CDR value of 10um this would lead to a new CDR of 9.16um, or a 0.84um decrease. This is similar to what is seen in Fig. 12 for NSW6. However, for NDW6 Fig. 12 says that there was only a 4 per cm3 increase in CDNC. Using the same numbers, this would lead to only a 0.07um decrease in CDR; but the quoted change is around 0.6um. So, there seems to be an inconsistency there. Using lower PI values for CDNC could allow the NDW6 numbers to work out, but then the NSW6 value would be out. So, something is not quite right here I think – could it be that the radiation scheme (presumably where CDR is calculated?) of NSW6 doesn't actually assume that the change in CCN is equal to the change in CDNC?

[A-4] Thank you for your comment. As you suggested, we added ∂CDR to Figure 13 (Figure 14 in the revised manuscript). As mentioned in the beginning of this reply, the NDW6-simulated ∂CDR is larger than the NSW6-simulated ∂CDR. When plotting the vertical profile of ∂CDR in the global average, we found that we used incorrect weighting factors to estimate the global average for each layer, so we fixed them in Figure 14 in the revised manuscript. We also found typos in units of ∂CDNC, so we corrected them. This correction does not affect other parts in the manuscript. After the correction, the conclusions remained unchanged, except for the difference in the ∂CDR between NDW6 and NSW6. In the revised manuscript, we modified this part as follows: "Specifically, above a height of 3 km, ∂Qc is close to zero, but ∂CDR is not zero because ∂CDNC has a positive value. Even though the magnitude of ∂CDR in NSW6 is lower than that in NDW6, this represents possible overestimation of the Twomey effect in NSW6."

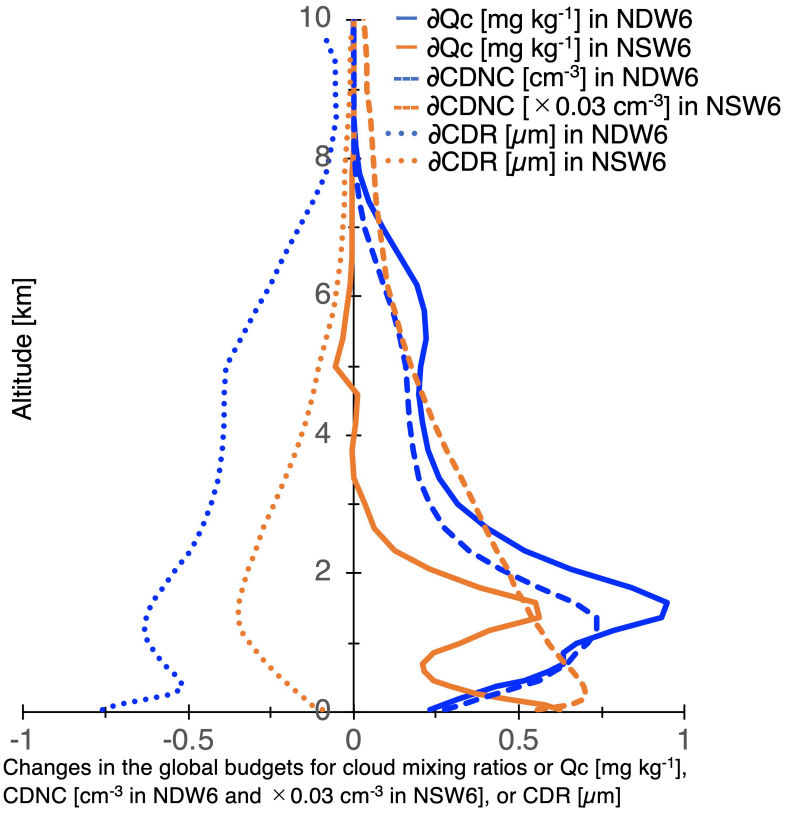


Figure 14 in the revised manuscript: Global budgets of the annual averages of the NDW6- and NSW6-simulated Qc (mixing ratio of cloud droplets), the NDW6-simulated CDNC, the NSW6-simulated CDNC (which is equal to CCN number concentrations), and the NDW6- and NSW6-simulated CDR (cloud droplet effective radius for warm clouds)

Regarding [A-4-2], the main point is the difference in the PI value of CDNC between NDW6 and NSW6. The simulated value of CDNC for PI in NSW6 is approximately 40 per cm^3 in the US, for example. According to your calculations, CDNC increases by a factor of 2.5 and CDR changes by a factor of 0.737. Since the CDR value for PI in the US is 6 μm, this leads to a new CDR of 4.42 μm, or a 1.6 μm decrease. In contrast, the simulated CDNC value for PI in NDW6 is approximately 2 per cm^3 in the US; The CDNC in NDW6 tends to be lower (which we understand is too low, but it is still difficult to resolve this issue within this manuscript). Thus, a 3-fold increase in CDNC results in a 0.693-forld change in CDR. Therefore, we think that the relationship between CDNC and CDR in NDW6 are consistent to the results in NSW6. We added this point to the revised manuscript (Lines 545-550) as follows: "For example, in the United States, the NSW6-simulated ∂CDNC (=∂CCN) is approximately 60 cm^{-3} and the NSW6-simulated ∂CDR is approximately -1.1 μm, whereas the NDW6-simulated ∂CDNC is approximately 4 cm^{-3} and the NDW6-simulated ∂CDR is approximately -2.3 μm. The difference in the ∂CDNC-∂CDR relationship between NDW6 and NSW6 is caused by the difference in the baseline of CDNC and CDR. The NDW6-simulated CDNC under both the PD and PI aerosol conditions is much lower than the NSW6-simulated results, whereas NDW6-simulated CDR under both the PD and PI aerosol conditions is larger than the NSW6-simulated results."

[C-5] L680 - “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 the industrial regions, such as the United States, Europe, and East Asia, increase in magnitude.” Presumably you are referring to NSW6 vs NDW6 here? You need to make that clear. However, dCDR is larger (in magnitude) for NSW6, and dLWP, dCA and dCF are smaller. Maybe you meant CDR is more negative, but better to talk in terms of magnitude I think.

[A-5] Thank you for your comment. This sentence is for both NDW6 and NSW6, so we added “In both NDW6 and NSW6” to this part in the revised manuscript.

[C-6] L685 – “by considering the uncertainty caused by the assumption in the PI conditions.” - It’s not clear what you mean here?

[A-6] Thank you. We deleted this from the revised manuscript.

[C-7] L691 – “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,” - This needs a bit of explanation. Perhaps something like “This, combined with the smaller magnitude of decrease in CDR for NDW6 vs NSW6 and the larger magnitude increase in LWP and CF, suggests that the ERFaci due to the cloud lifetime effect in NDW6 is larger than that in NSW6 due to the Twomey effect,...”

[A-8] Thank you for your suggestion. The correction of CDR in NDW6 suggests that the ERFaci due to both the Twomey and cloud lifetime effects in NDW6 was larger than that in NSW6. However, the comparisons in the vertical profiles of ∂Qc, ∂CDNC, and ∂CDR suggest that the NSW6-simulated ERFaci certainly includes some bias due to the overestimation of the Twomey effect, especially above 3 km height. Therefore, we modified this (Lines 586-588 in the revised manuscript) and sentences in summary (Lines 643-646 in the revised manuscript) as follows: “Therefore, it was suggested that the ERFaci due to both the Twomey and cloud lifetime effects in NDW6 was larger than that in NSW6, although the NSW6-simulated ERFaci certainly includes some bias due to the overestimation of the Twomey effect.”

Typos/grammar

[C-8] L18 – “but the differences between the results of NDW6 and NSW6 experiments were larger for some aerosol species, especially dust and sulfate, compared to those between the experiments with different horizontal resolutions, i.e., 14 km and 56 km grid spacing, as shown in a previous study.” - I suggest that you start a new sentence and slightly rewrite this to make this sentence clearer : “However, for some aerosol species, especially dust and sulfate, the differences between the NDW6 and NSW6 experiments were larger than those between experiments with different horizontal resolutions (14 km and 56 km grid spacing), as shown in a previous study.”

[A-8] Thank you so much for the correction. We used them in the revised manuscript.

[C-9] L34 – “caused by ignorance of sink process in the cloud droplet number concentrations.” -> “caused by the ignorance of sink processes for cloud droplet number concentrations.”

[A-9] Thank you so much for the correction. We reconsidered this and decided to remove this from the abstract since it is not directly related to why the ERFaci in NDW6 is larger than that in NSW6. We also found places in the manuscript that needed correction related to your comment, so we changed from “sink process” to “sink processes” in Line L540 and Figure 12 in Lines 1295 in the revised manuscript.

[C-10] L218 - “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).” - I think this should be “Hoesly et al. (2018) estimated that the globally averaged emissions in 1850 were 2.1% of the 2010 emissions for sulfate, 12.0% for BC and 22.7% for OC.”

[A-10] Thank you so much for the correction. We used them in the revised manuscript.