Responses to the Remarks of the Handling Editor Dr. Graham Feingold

Dear Graham,
We very much appreciate your effort in handling our manuscript.

We have carefully addressed each of the new comments from both reviewers. In the meantime, the manuscript has also been revised accordingly, with an emphasis on adding more discussions about relevant previous works, mostly in Section 4. In responding to the recommendation from both reviewers, we have also reduced the content of Section 3 that discusses the results of REF run. Additionally, we have further improved the readability of the paper.

We would also like to respond to your specific note of: "Finally, I do not see any responses to my comments, or an attempt to address them in the manuscript. I reattach those comments here".

Our recollection is that: we received your remarks sometime after our submission of the responses to the two reviewers' previous comments alongside a revised manuscript to the ACP in early April. We then made an extensive effort to carefully address each of your comments and to conduct a massive revision of the manuscript. Specifically, new figures and associated discussions of time variations of LWP and CF in each of the simulations, and the discussions of certain previous works were added. We successfully submitted our responses to your comments to the ACP site on April 25. As ACP has made it available to the public ever since in the interactive discussion stie of our paper, under EC1 pile, we thought that you should have already read it. On the other hand, we could not, however, upload the heavily revised manuscript in responding to your comments (for convenience we noted it as the RE version here), this is due to the ACP uploading rules. Thus, we decided to wait the new comments from the reviewers to merge additional revisions, then upload a combined new version of the manuscript. As a special note, we have now described this matter to both reviewers and informed them that the new revision in responding to their additional comments has been made based on the RE version, which had already addressed many of their new concerns along with yours. We understand the difficulty to track every paper you are handling, and we again appreciate your effort.

Best regards!

Responses to the Additional Comments of the Reviewers

We very much appreciate the additional comments from both reviewers. These comments have led to a further improvement of our manuscript.

We would like to specifically indicate that, soon after we submitted the responses to reviewers' first round comments alongside the revised manuscript, we received the comments from the handling editor, Dr. Graham Feingold. Along with the effort in making our responses to the editor's comments, we also significantly revised the manuscript (hereafter referred as the *RE version* for convenience), where many sections were massively rewritten, and numerous typos or grammatical errors corrected. Most importantly, additional figures and associated analyses of e.g., time evolutions of LWP and CF (we used cloud fraction instead of other diagnostics in this version) in different model runs. More discussions of certain previous studies were added as well. While the responses to the editor were successfully submitted in the ACP website and soon became available to the public (under the EC1 pile), the upload of correspondingly revised manuscript, the RE version, however, was not possible due to the procedure of ACP. Therefore, we decided to merge any additional revisions in responding to the reviewers' new comments into the RE version. The modifications made in the RE version can be easily found in the tracked changes from the currently submitted version of the manuscript. In fact, many of the new comments from the two reviewers have already been addressed in the RE version as would be indicated in our following responses.

The following are our point-by-point responses to the reviewers' comments (here the reviewers' comments are displayed first in bold *Italic* font).

1. **Responses to the Additional Comments of Dr. Mónica Zamora Zapata**

Summary
The authors have included more comparisons, relevant technical details, as well as cleaned up confusing statements, improving the manuscript from its previous version; however, I find that some questions were not fully addressed. From my side, some of them were aimed to ensure that the work is reproducible, so I'm including them in minor comments. Regarding the grammar and style of the manuscript, there is still room for improvement. Nevertheless, their work is still highly relevant and the scope of the paper is now more clear.

We truly appreciate these positive remarks made by Dr. Mónica Zamora Zapata.

Minor comments
• The work is structured in a first part where the REF case is validated, and a second part where the aerosol experiments are performed. I suggest minimizing the description of the REF case as much as needed. For example, I don't see any use of reporting how much is the temperature at cloud top in every hour of the simulation. The relative differences between model results and observations are key but too much description is overwhelming for the reader, what is really useful from there?
The point is well received. We have shortened the REF analysis and made the discussions focus mostly on comparisons with observations rather than providing a hourly report of, e.g., temperature change.

• One of the interesting findings of this work is pointing towards how a polluted scenario can increase spatial cloud variability, which was reported as a research gap in the Introduction. I don't think this is clearly stated in the abstract right now. It motivates it around L30 and then it does not mention spatial variability again. The conclusions summarized it better. Was this process observed/reported in other studies?

Thank you for this excellent point. It has been one of the emphasized enhancements of the RE and the current revision with a specific discussing point of cloud fraction. The Abstract has been rewritten to provide more precise highlights of our findings including the aspect indicated by the reviewer here. In addition to the aerosol concentration factor, the impact from the semi-direct effect by BC has also been highlighted in both Abstract and Conclusion.

Regarding previous works, for example, we have cited Wang et al., 2003 who suggested that cloud under lower aerosol concentration could form thinner cloud layer more easily than in polluted cases, though the model capacity alongside aerosol configuration in that study is differ from ours. We have also discussed Dearden et al. (2018) who conducted a LES simulation of another DACCIWA case though with a passive aerosol configuration, where they run a paired simulations with a binarily configured sedimentation of droplets (i.e., inclusion vs. exclusion). If one simply interprets the sedimentation as a function of droplet size (thus negatively correlated to aerosol concentration), then the derived variation of LWP and CF in that work qualitatively agrees with our finding of LWP and CF changes in responding to aerosol concentration. These are just two examples among those we have added and discussed in the revised manuscript particularly in 4.2 and 4.3.

• Following this topic, it may be worth exploring how different are the trajectories of the POL, REF, and CLEAN cases in a space defined by cloud fraction and reflectivity. This could support the description around L695.

This has been greatly enhanced in the RE and the current version using cloud fraction as the discussing point, aided by additional figures showing the time evolution of both LPW and CF, most importantly, as functions of aerosol concentration as well as chemical composition. Besides the revision in this paragraph, an additional paragraph has been added to enhance the discussion.

• Are there observation based reports finding that solar variability is also greater for more polluted cases?

We have not been able to identify such reports other than estimates based on satellite retrieval.

• Some of my previous comments were aimed towards documenting details of the setup so that the work can be reproduced rather than questioning if they were correct. I'm bringing back some of those topics so that you can evaluate if they can be included:
– What are the initial conditions of this run like?

The model was initialized from sounding at 23:00UTC of July 2 as described in the manuscript, with a one-time perturbation to get the turbulent mixing to start. The thermodynamic and dynamical profiles in the early hours are shown in Figure 7, these data can be obtained from DACCIWA website as indicated in the Code and Data Availability section.

Also, as a general response, we would like to indicate that the entire Model and Data description section has been largely rewritten. Several components of the model such as those regarding aerosol microphysics and chemistry have been added in the RE version, along with certain

details of model configurations (including those in Section 4.1 for providing additional information and explanation for the sensitivity simulations).
– How are the vertical profiles created? Just applying the nearest value to the corresponding grid height? Or is there any averaging performed?
Directly since the vertical resolution is reasonably high.
– I now see that the subsidence velocity is quite high, is this typical of this region? Or was that needed in order to control PBL growth to match observations?
– The way to find the inversion is specific for this case, right?
Answers to both are yes, these can be done rather straightforwardly based on the soundings.

• There is still room for improvement in the readability of the document. I suggest asking for professional help or using one of the many tools available for checking grammar and style. Below are included some suggestions but I didn't have time for a more thorough check.
One of our "senior" co-authors has made an extensive effort to polish the readability.

• Fig. 5 is still stretched
We have resized the figure.

• What is this increase of the vertical wind speed referring to in L525?
The sentence has been modified to "…, implying an increase of surface solar heating".

Typos / writing suggestions
We have generally adopted the reviewer's suggestion in performing the according revision hereafter. Therefore, here we only list the responses where additional answers would need.

• General/style: using more articles like "the" and "a" could help.
Done with a best effort.

• L35 lower?
Here lower is used as a verb.

• L324 you can include how much it varies: 400 to 1200 m
We have revised the sentence to:
"However, the nocturnal-diurnal life cycle in our case involves a dynamically evolving cloud top from 400 to 1200 m, particularly in the daytime, making it a difficult task to prescribe a highlighted zone for finer resolution. Our fast-testing results, on the other hand, did not suggest an alarming difference between the run with 10 m and 5 m vertical resolution (not shown)".

• L404 Midcloud, CPP decreases...
• L405-406 less inhomogeneous or less homogeneous?
We have modified the sentence to:
"Between 10:00 and 13:00 UTC, CF of the layers between domain mean CBH and CTH starts to decrease from near 100% to 90%, while CF at CBH and CTH decreases more substantially to reach near 60% and 80%, respectively. This leads to a less homogeneous cloud deck…".

• L554 Based on the observations, do these upper clouds yield a lower observed solar irradiance?
We do not have direct observational evidence for this.

• L562 I don't know if this is cloud break-up time, as clouds were indeed already broken then
Yes, stratus broke up while separate convective cloud blocks still existed.

• L564 I don't understand this well, is it related to the forcing tendencies?
Primarily surface solar radiation.

All the other issues have been resolved either in the RE version already or this the current revision by adopting the reviewer's suggestions.

2. Responses to the Additional Comments of the Reviewer #2

I find that some of the major and minor issues I raised have not been fully addressed.
Major issue #1:
-Place results in context with previous studies, both when describing the reference case as well as the aerosol sensitivity studies.

I appreciate that a new paragraph has been added in the beginning of Section 4. However, I am afraid such paragraph is too general and does not provide any relevant information into how the presented study relates, or differs, from those mentioned by the author therein. I would appreciate more references and comparisons to other studies both in Section 3 and 4. In general, I find that there is discussion missing in this article. It could be done along the result description, as I suggested first, or in a separate discussion section.

Additional discussions on the similarity and/or differences between our results and those of others have been added in the revised manuscript. Please also note that we have significantly revised the Introduction to highlight the major difference between our modeled case and most others (e.g., land vs. ocean), along with certain different controlling factors as well as expected feedbacks (e.g., surface heat flux responses etc.). New discussions in Sector 4 have been added particularly regarding the aerosol impacts in a context of different aerosol model configuration and profiles.

Major issue #2:
-Language-related issues, like grammatical errors and long and complex sentences make following the manuscript a difficult task. Please correct all grammatical mistakes (only some examples given along the line by line comments) and keep sentences short for the shake of clarity.

I can see that a number of errors, misspellings and confusing sentences have been reworded. I think enough has been done in this respect.

We appreciate the positive remarks made by the reviewer.

Minnor issue #2:
-Section 3 goes through the results of the REF simulation in high detail. Readers would find it easier to understand, however, if instead of a description of each result, a more concise section with the most relevant results is presented. This would also allow, as suggested in the fist Major issue, some room to link the relevant results to previous studies.

I do not think much action has been taken in this respect. I agree that, to a certain extent, this is matter of a style. However, I believe that the current text makes it challenging for a not very expert reader to understand what the new findings are, and what is common knowledge from previous research from such a detailed and indiscriminate description of the results.

The same point from both reviewers is well received. As a response, we have greatly reduced the discussions in REF section, particularly of 3.2. In addition, we have rewritten the summary paragraph in the end of 3.1 to provide a highlight of the overall comparison of our modeled results with observations. In the meanwhile, we decide to keep certain contents of 3.2 to benefit potential reproduction works in near future, besides some later analyses in the paper on the model-observation inconsistency. Here are three examples among others of the newly added discussions in Section 4.2 and 4.3 (note also the new figures shown time variations of LWP and CF):

"The difference between CLEAN and REF in cloud macrophysical features such as CBH and CTH is visible though largely limited to a few tens of meters. However, their differences in cloud fraction and microphysical features are rather significant. As expected, from formation to break-up of the clouds, N_c^{CLEAN} is lower than N_c^{REF} and r_c^{CLEAN} is larger than r_c^{REF}. At 02:00 UTC, N_c^{CLEAN} has a maximum value of 181 *droplets* cm^{-3} and r_c^{CLEAN} of 7.58 μm, in comparison to 653 *droplets* cm^{-3} and 5.1 μm for N_c^{REF} and r_c^{REF} respectively with the same liquid water content value (0.35 $g\ m^{-3}$). r_c^{CLEAN} further increases to 12.55 μm at 08:00 UTC, then decreases slowly to a maximum value of 10.97 μm at 14:00 UTC with LWC^{CLEAN} reaches near 0.45 $g\ m^{-3}$ instead of 0.49 $g\ m^{-3}$ for LWC^{REF}, likely due to an increased activation ratio of aerosols after sunrise. Despite a relatively larger droplet size in CLEAN than POL and REF case, there is no clear sign of massive formation of drizzles even during the convection stage (Fig. 10). Nevertheless, sedimentation thus evaporation of larger droplets from entrainment zone and cloud base could likely create a thermodynamic perturbation (*e.g.*, Stevens *et al.*, 1998; Jiang *et al.*, 2002). In a LES simulations using passive aerosol profile for July 4-5 DACCIWA case, Dearden *et al.* (2018) found that the sedimentation would remove droplets from the entrainment zone thus, through a feedback, lead to a cloud deck with higher LWP while smaller CF than the case where sedimentation is completely excluded. This could imply a similar contrast between CLEAN and the two polluted cases in our simulations, by simply assuming the total sedimentation amount is proportional to the droplet size (*i.e.*, inversely to the CDNC), though the quantity of such a perturbation seems rather small here, not to mention the more sophisticated feedback involved in our case introduced by the dynamic aerosol-cloud interaction in our model".

"Looking into various timely varying metrics of LWP in different model runs, we find that in general, LWP is inversely promotional to CDNC, as LWP in POL < LWP in REF < LWP in CLEAN, and this is applied to different metrics of LWP (Fig. 12, Table 3). However, in comparison, the peak LWP varies less significantly in CLEAN case, while peak LWPs in two other runs decrease with domain averaged quantities in convection stage. There were different opinions regarding the mechanisms behind such an inverse relation between LWP and CDNC (*e.g*., Ackerman *et al*., 2004; Bretherton *et al*., 2007), not to mention that most such hypotheses were proposed based on the cases of marine low clouds that might not be directly applied to the cases over land. In our analysis, the difference in turbulent mixing driven by the surface radiative heating, as influenced by different microphysical features in various cases, seems having played a critical role. The situation of cloud fraction (CF) is somewhat more complicated. As shown in Table 3 and Fig. A4, CF relation with CDNC varies in different stages. An inverse relation between CF and CDNC generally stands in the earlier and later period of the convection stage, in the middle of the convection stage (13:00-15:00 UTC), the above relation, however, would reverse, alongside the vertical cloud extent as discussed previously".

"The above results have demonstrated the important role of solar absorption by aerosols in determining the life cycle of LLSCs. The atmospheric heating by light absorbing BC would limit the elevation of cloud top, especially during the break-up stage (Koch and Del Genio, 2010b; Zhang and Zuidema, 2019). Such a heating can also increase cloud fraction then delay break-up until late afternoon, especially for clouds with higher cloud droplet number concentration in polluted environment such as in POL and REF runs (opposite to the outcome by considering aerosol number concentration only), and thus affect the indirect effect of aerosols. Note that our modeling configurations are based on the aerosol profiles that are relatively well-mixed throughout the PBL then with concentration gradually decreasing along altitude above PBL. Certain previous sensitivity experiments suggested that the location of BC layer within or above PBL could have different impacts on the development of convection, entrainment, and thus life cycle of the low clouds within PBL. For instance, Johnson *et al*. (2004) suggested that without considering the indirect effect of aerosols, BC existing within boundary layer would lower LWP by nearly 20% in a marine low stratocumulus case, where the cloud response is less sensitive to the surface shortwave heating change comparing to the situation in our case. Feingold *et al*. (2005) found that smoke plumes containing BC near the surface would reduce the cloudiness through both the atmospheric heating and weakening effect on surface heat fluxes by BC. These results though obtained with somewhat different model configurations than ours (e.g., coarser vertical resolution, different surface, etc.) are in a qualitative agreement with our findings. Nevertheless, the unique configuration of our model allows us to quantitatively examine the semi-direct effect with varying aerosol chemical compositions and thus extent of aerosol absorption. This has led us to reveal further insights of the complicated interplays among various aerosol effects besides their individual impacts on the life cycle of LLSCs".