

We very much appreciate the comments from the handling editor Dr. Graham Feingold on our paper. The following are our point-by-point responses to these comments (the Editor's comments are displayed first in bold *Italic* font).

***In addition to the reviewers' comments, I have a number of important comments that I would like you to take into account before resubmission:***

***1) To begin with, the quality of the writing is not up to par. I am sympathetic to the fact that the authors are probably not naive English speakers but we cannot underestimate the importance of clearly written and sharp text that communicates the ideas effectively. Unfortunately, much work needs to be done in this regard.***

We agree that the manuscript could be better prepared. The entire manuscript has now been largely rewritten. The revised manuscript should reflect our best effort in making a significant improvement of the paper's readability.

***2) On a scientific note, the authors do not seem to have strong familiarity with the literature on aerosol-cloud-radiation interactions and thus, while the results seem robust, some of the explanations are well-known and could be stated much more simply, with appropriate references. In many places this leaves the incorrect impression that the authors have discovered something new.***

As a general response to the editor's above comment, we have modified discussions in several sections of the paper, primarily to emphasize on the outcomes of our study that are different from previous analyses after introducing aerosol's role into consideration. For example, new discussions (and new figures) regarding LPW and CF response to both aerosol abundance and chemical composition have been added. We appreciate specifically the references suggested by the editor, and we have cited most of them to indicate the consistency as well as difference between results of ours and others.

***Examples:***

***- Liquid water path (LWP) and cloud fraction (CF) adjustments***

***The notion that the Twomey effect is the dominant one has long been shown to be inadequate, especially given the much stronger control of N on LWP (2.5 x more important in a relative sense) and the dominance of CF, about which much less is known regarding aerosol effects. There is a very large body of literature on this topic and in various places, the text comes across as naive (e.g., bottom of page 24, and bottom of page 25).***

We believe that what we expressed in the text, e.g., in the bottom of Page 24 alongside other places, is basically the same message as the above comment, i.e., the Twomey effect is not always the dominant factor in controlling the radiative effect of stratiform clouds particularly regarding the incoming solar radiation at the ground. To better understand the issue, the responses of LWP and cloud fraction to aerosol variation need to be considered. As the editor perhaps also agree that our effort is specifically to examine the role of aerosols on this regard comparing to many other works in the large body of literature, especially by taking both aerosol size distribution and chemical composition into consideration, aside from the key microphysical processes interacting with dynamics that has caused such an outcome. We certainly should express our points more clearly. Thus, we have made modifications in several related discussions. A new Fig. 12 and Fig.A4 have been added to show the timely variations of LWP

and CF in various runs. These newly added analyses indeed brought quite many revisions of Abstract, Conclusion, and main discussions. See the following response in more specific.

**- LWP adjustments are usually negative in stratiform clouds**

[https://doi.org/10.1175/1520-0469\(2003\)060<0262:TCALWT>2.0.CO;2](https://doi.org/10.1175/1520-0469(2003)060<0262:TCALWT>2.0.CO;2)

<https://doi.org/10.5194/acp-19-5331-2019>, doi:10.1029/2006GL027648

***This makes it a central variable for aerosol-cloud-radiation interactions, and yet other than the Table values, it's hard to get a good picture of LWP evolution and how N might be affecting LWP. The same is true for CF: the smaller the cloud fraction the smaller the radiative effect of the clouds, and the less leverage there is for aerosol effects on clouds. Aerosol effects on CF are less well quantified but this might be where some extra work gives you an opportunity to say something new.***

The editor's point is well received. As mentioned in the response to the previous comment, we have added new figures (new Fig. 12 and Fig. A4) to show the time evolutions of LWP and CF in different runs. New and revised discussions can be seen from Abstract, Section 4, and Conclusion, among others. In brief, our result of LWP response (negative) to CDNC is in an agreement with the previous studies (note that our configurations for aerosol-CDNC relation brought in size distribution alongside chemical composition). We agree that the CF-aerosol has rarely been addressed in-depth, and with the unique modeling configurations we should be able to obtain some new knowledge. We now show that CF-CDNC relation varies in different stages, for most of the convection stage before massive cloud break-up occurs, they are inversely correlated. In addition, semi-direct effect causes a lower CF in the same period.

Here is one example of the revisions (newly added in Section) 4.2): "Looking into timely varying metrics of LWP in various run, 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 why such an inverse relation between LWP and CDNC (or aerosol number concentration in works with simple aerosol-cloud model) (e.g., Ackerman *et al.*, 2004; Bretherton *et al.*, 2007). 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 convection stage, in the mid-convection stage (13:00-15:00 UTC), the above relation would reverse, not to mention that the difference in cloud extent among different runs as discussed previously".

In 4.3, a newly added paragraph regarding semi-direct effect reads as: "The impact of the semi-direct effect on other critical macrophysical features such as cloud fraction and LWP can be also seen from the model results. For instance, LWPs are clearly lower in the AODON runs of the two polluted cases (REF and POL) (Fig. 12). In addition, an increase of cloud fraction due to the semi-direct effect can be seen throughout the convection stage until 15:00 UTC when massive cloud break-up occurs (Fig. A4). All these imply a critical role of semi-direct effect in cloud radiation".

**- On page 21, the discussion of the microphysical responses is long and not very informative because much is already anticipated. Is the POLLUTED case even needed given that REF is so polluted already and that at some point updraft/supersaturation production cannot activate any more aerosol?**

The indicated discussion has been rewritten to simplify description of certain expected results while emphasize on some special cases. By the way, in the discussion regarding the role of different evaporation rate due to aerosol size distribution, Wang et al. (2003).

Regarding whether the POL is even needed since REF is already polluted, we believe the answer is yes. This is because that size distribution and chemical compositions of these two observed profiles are different despite that they have the similar peak number concentrations. Such a difference could cause different semi-direct effect aside from activation, as suggested by the results. We have revised 4.1 to emphasize the differences between POL and REF aerosol profiles. In addition, discussions have also been revised to indicate different modeled outcomes associated with the above difference.

One example of newly revised discussions reads as: “At the cloud formation (02:00 UTC), despite having similar liquid water content (LWC) around  $0.35 \text{ g m}^{-3}$  at 250 m in both cases,  $N_c^{POL}$  reaches  $333 \text{ droplets cm}^{-3}$  and  $r_c^{POL}$   $6.45 \text{ }\mu\text{m}$  instead of  $653 \text{ droplets cm}^{-3}$  and  $5.1 \text{ }\mu\text{m}$  for REF case, indicating a result of differences mainly in the Mode 2 aerosol numbers between the two scenarios (at 02:00 UTC the updraft near cloud base is rather weak at less than  $0.30 \text{ m s}^{-1}$  in both cases). This trend is reversed at 06:00 UTC when the CDNC and radius are equal to  $1208 \text{ droplets cm}^{-3}$  and  $6.43 \text{ }\mu\text{m}$  in POL, and  $1305 \text{ droplets cm}^{-3}$  and  $6.12 \text{ }\mu\text{m}$  in REF, respectively. After 08 UTC and until the cloud break up,  $N_c^{POL}$  is superior to  $N_c^{REF}$  by reaching a maximum difference of  $1425 \text{ droplets cm}^{-3}$  at 14:00 UTC. Their respective radii are  $4.42 \text{ }\mu\text{m}$  and  $5.18 \text{ }\mu\text{m}$  while the liquid water content profiles are quite the same as near  $0.47 \text{ g m}^{-3}$  at 750 m. The difference between POL and REF in CDNC after sunrise suggests that the activation favors the POL profile with higher sulfate content when updraft is strengthened”.

**- Effect of drizzle: it has been proposed that weak drizzle can stabilize clouds (by preventing deepening [https://doi.org/10.1175/1520-0469\(1998\)055<3616:LESOSP>2.0.CO;2](https://doi.org/10.1175/1520-0469(1998)055<3616:LESOSP>2.0.CO;2)) and if drizzle evaporates just below cloud base, can strengthen turbulence by destabilizing the BL (doi:10.1029/2001JD001502) When discussing drizzle, please engage in these ideas and see if they are relevant to your analysis (e.g., CLEAN, Fig. 10). In Fig. 10, a TKE profile would help to show whether weak drizzle just below cloud base might be enhancing cloud turbulence/deepening. You could show divergence of the modeled drizzle flux to get a sense of evaporative cooling below cloud base.**

We appreciate the excellent point regarding drizzle. We did not expand our discussion on this aspect is primarily due to the fact that drizzle was believed to be rare for this case based on observations. From our modeling results, it can be clearly seen from the profiles that the cloud droplet size is rather small (sub-10 micron; Fig. 7, 8, 9, & 10), and the formation of drizzle sized particles were rare even for the CLEAN case despite a “drizzle-sensitive” scheme of Khairoutdinov and Kogan was specifically introduced in our modeling. The point is still valid, we have added the following sentence in discussion: “Despite the relatively larger size of the droplets in CLEAN comparing to the cases of POL and REF, there is no clear sign of massive formation of drizzles even in the convection stage (Fig. 10). Nevertheless, sedimentation thus evaporation of larger droplets from cloud base could likely create a thermodynamic perturbation

(e.g., Stevens *et al.*, 1998; Jiang *et al.*, 2002), though the quantity of such a perturbation seems rather small here”.

**- Where does the absorbing aerosol reside? This makes a significant difference to the dynamical response (e.g. doi: 10.1256/qj.03.61, doi:10.1029/2005JD006138, 10.1002/2015GL066544). And please convey the essence of knowledge already known from these papers, rather than simply providing lists of references. The current version of the text is not careful about using those references to provide context.**

Aerosols are relatively well mixed within the PBL (the initial profile and the periodic lateral boundary condition) (e.g., Fig. 7, 8 & 9). We did not apply any specific isolated BC layer as in the referred idealized studies. We have added a few more sentences in the revised manuscript in Introduction and in discussions. For example, the new sentence in Introduction reads as: “Such a semi-direct effect can be positive or negative depending on the relative distribution of the aerosol with respect to clouds (e.g., Johnson *et al.*, 2004; Feingold *et al.*, 2005)”. Also in 4.3, “Note that our modeling configurations are based on 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 (e.g., Johnson *et al.*, 2004; Feingold *et al.*, 2005)”.

### **3) Missing information/other comments:**

**- The cloud radar is mentioned but we aren't told its wavelength, which makes it hard to interpret what it sees. (See drizzle discussion above)**

We have added information of the Ka band mobile, dual-polarization Doppler radar (8.5 mm, 35.5 MHz) in the revised text.

**- You mention supersaturation quite a bit but is it actually prognosed, or diagnosed based on a parametrization that includes updraft? And all diagnostic activation parameterizations depend on  $w$  (line 463). A problem is that it's not just vertical motion that drives supersaturation but the total effects of dynamics.**

The activation is calculated using Abdul-Razzak and Ghan scheme (2004) as described in the text, and thus supersaturation is obtained from diagnostic method with several corrections mainly at cloud top. We have made these clearer in the revised manuscript, in the description of LIMA and ORILAM. We certainly agree that considering 3D air motion could in theory lead to a more accurate estimation, though the difference might not be so significant comparing to the outcome derived by vertical-dominated diagnostics, because the large temperature gradient in vertical direction and the maximum activation near the cloud base. Based on our knowledge, the current effort is still on making a better correction at cloud top.

**- Model radiation: the model top is at 2 km. Does this mean you ignore the influence of the gases above the domain. If so, this is a serious omission. A column of atmosphere should be patched above for radiation calls so that the radiative effect of gases is included.**

Meso-NH model is a community model just like WRF and developed for various applications, not all of them would have a domain to cover the whole atmosphere. The radiation above the model domain, as in any other such modeling application, is calculated based on prescribed (e.g., climatological) profiles. To avoid unnecessary misunderstanding, we have added in the revised

text that: “Note that the radiation module still proceeds calculations above the 2 km using prescribed profiles”.

***- The low domain top might also explain why your modeled cloud deepens too much in the afternoon: If in fact there were upper-level clouds and the model doesn't see them then your cloud top cooling will be too strong and your cloud will deepen more than it should***

Comparing to many other similar modeling efforts, our domain is not necessarily too low considering the vertical extent of the cloud layer. Certainly, a mid-cloud layer with drizzle could lead to a faster break up as hypothesized (cited in the paper), or without this mid-cloud the modeled cloud would actually last longer than observed one did. Nevertheless, this is the reason why we decided to design a configuration to avoid this issue, rather than focusing on the aerosol impacts within lower atmosphere. We have added a statement regarding this in responding to the reviewer #1 comments, as “Nevertheless, our focus of this study is on the diurnal cycle of LLSC as influenced by aerosols alongside planetary boundary layer dynamics rather than examining the above hypothesis, which appeared to be related to a process beyond the local scale. Therefore, our model setting is made to specifically eliminate the influence of mid-cloud layer for the purpose as described later”.

***- Is hygroscopic growth included in the aerosol radiative effects and optical properties?***

The aerosol code ORILAM has a gas-particle equilibrium (EQSAM for inorganics and MPMPO for organics) that allows the model to calculate the water content of the aerosol. The solver will then combine the moment 0 (integrated number) and 3 (integrated new volume which integrates the hygroscopic growth) to calculate the new dimensional distribution (Tulet et al., 2005, 2006; both were cited in the manuscript). The parameterization of aerosol optical properties introduced in Meso-NH (Aouizerats et al., 2010; cited in the manuscript) allows the calculation to be processed at each time step, with a refractive index corresponding to the chemical composition of aerosol particle (including the amount of water) according to the Maxwell-Garnett equation (Maxwell-Garnett, 1904) as defined in Tombette et al. (2008). This approach considers that the aerosol is composed of an inclusion and an extrusion. The inclusion is composed of the primary and solid parts of the aerosol, while the extrusion is composed of the secondary and liquid parts of the aerosol. These are largely described in the manuscript already. We have reorganized the contents with certain added information regarding these aspects in the revised manuscript, in the paragraph of ORILAM description.

***- Typically, radiation is called much more often than 10 minutes (usually order 20 s). What effect is this having on simulations?***

It has been tested with values as short as 30 seconds, the step is selected as an optimized solution based on the modeled outcomes and also overall computation burden.

***- I was surprised that the aerosol model uses the 6<sup>th</sup> moment as one of its moments. That's typically a choice for rain (radar reflectivity = 6<sup>th</sup> moment)***

The ORILAM aerosol model adopts a 3-moment approach to close the log-normal distribution defined by a median radius, an integrated number, and a dispersion. The choice of moment 6 is numerical, it allows one to calculate the coagulation coefficients explicitly and to facilitate the integration of the aerosol solver. All these were described in detail in the two cited Tulet et al.

papers. We have also added a note about the selection of the 6<sup>th</sup> moment in the ORILAM description paragraph.

**- *Cloud void space is the 1-CF. Why not speak in terms of the familiar cloud fraction and make the reader's life easier?***

The point is well received. We have now used the term of cloud fraction as much as possible in the discussions unless when cloud void space appears to be more natural in expression (positive, negative or increase and decrease). By the way, the axis label of “Cloud Presentation Probability” in Fig. 4 and A2 have been replaced by Cloud Fraction with a notation in the caption and associated text to mark the layer-defined nature of this quantity.

**- *As noted by a reviewer, the earlier part of the simulation is probably affected by spin-up. This is worth checking so that your discussion of the 0:00-04:00 UTC period is robust.***

This has been checked through testing runs in the early stage of the modeling. The simulation started at 23 UTC of the previous day, though the cloud formed around 2 UTC based on observation (see 2.2), thus the spin up is not necessarily too short. We have also found that with a cyclic lateral boundary condition without topography, the modeled results converge with profiles rather quickly. Based on the results of testing runs and actual simulations, we believe that the impact of the spin-up time on simulations is quite limited.

**- *Bottom of page 11, other reasons include incorrect surface fluxes, and model weaknesses.***

It has been modified to “The differences between the model and the observation between 13:00 and 16:00 UTC could come from the different representation of simulated result (a domain average) versus that of ceilometer detection (limited to only one vertical direction), the vertical resolution of observed profiles, the limitation of radar in detecting hydrometeors, and in the end, certain model weaknesses likely associated with a lack of hourly radiosondes during the afternoon period to provide sufficient observational constrain”.

**- *Caption Fig. 7h: why mention SWHR (no lines) at night***

There is a mistake in the discussion of Fig.7h in the text, where SWHR (bottom horizontal axis in the Figure) should be LWHR (top horizontal axis). This has been corrected.

**- *Lines 370-371: The increase in cooling with higher  $N$  is only true for clouds with  $LWP < 25$  g/m<sup>2</sup>.***

Solid point, LWPs of many cloud blocks during the stratiform stage meet that criterion (Fig. 5).

**- *Line 595: references?***

Since the according discussion with references has been provided in the main text (e.g., Section 4), thus, we would like to limit the citation as a common practice in Summary and Conclusion. We have added words of “as demonstrated in this study and several previous ones” in the revised manuscript.