Replay to the review of: "Time dependent, non-monotonic response of warm convective cloud fields to changes in aerosol loading"

We would like to thank the reviewers for their efforts and beneficial comments that helped us improve our paper and present a clearer and more complete study. Before addressing all of the reviewers' comments in a point by point manner (Referee #1 followed by Referee #2), we would like to list a few general revisions that are relevant to both reviewers:

1. We added information in the methodology section for describing better the treatment of aerosol in the model including the aerosol size distribution, chemistry, activation etc.
2. Following both of the reviewers' comments we added a supporting information (SI) file with more technical information. The SI includes information about the mean cloud and rain drop concentration and size for the different simulations, vertical profiles of mean cloud fraction and additional information about the time evolution of the thermodynamic conditions.

A point by point reply to all of the reviewers' comments (answers in blue).

Referee #1

General Comments:

This article investigated the response of thermodynamic properties of cloud fields to changes in aerosol loading, using a Large Eddy Simulations (LES) with bin micro-physics. The results that pollution acts to suppress rain and increase atmospheric in-stability, that is, warming of the lower part of the cloudy layer and cooling of the upper part, are very important and add some new insights into the understanding of aerosol-cloud-radiation interactions. The article is generally well written, concise and should be publishable if the following specific comments and suggestions can be considered in revision.

Answer: we thank the reviewer for finding this work important and for the encouragement.

Specific Comments:

1. Since I did not see the article by Dagan et al. (2016), but from the title and introduction in this manuscript, it seems to me that the results and conclusions of these two paper are similar. What are the main differences between them?

   Answer: In this study we examine the response of cloud fields' mean properties to changes in aerosol loading. This is done both globally (during the entire simulation period, Sec. 3.1) and for different parts of the simulation period (Sec. 3.2). We show that the cloud fields' mean properties response in a non-monotonic way to an increase in aerosol loading. The non-monotonic trends with an optimal aerosol concentration of the mean cloud field properties are explained by contradicting aerosol effects. The time evolution of this response and the increase along time of the optimal aerosol concentration are driven by differences in the evolution of the thermodynamic conditions in the cloud field under different aerosol concentrations. This thermodynamic evolution under different aerosol concentrations is the focus of Dagan et al. (2016) paper. In Dagan et al. (2016) we don't discuss the aerosol effect on the mean cloud field properties and their non-monotonic trend but only the thermodynamic evolution. As the changes in the thermodynamic conditions is the driving force of the evolution of the non-monotonic trend, and for the purpose of writing a "stand-alone" paper this issue is explained here again.

   We added clarification to the revised manuscript: "*All the aerosol effects that were discussed up to this point (condensation-evaporation efficiencies, η and water loading) are applicable both on the single cloud scale as well as on the cloud field scale. However, on the cloud field scale, another aspect needs to be considered, namely the time evolution of the effect of clouds on the field's thermodynamic conditions (which was the focus of a recent study by Dagan et al., 2016).*"

2. Different initial concentrations of aerosol particles are used in the simulation. How the initial aerosols are distributed vertically, uniform or decrease according to a certain function? Whether they change with time? What are the altitudes of these aerosol concentrations referred to? Did you consider aerosol

regeneration after evaporation of cloud particles? This could be a very important source of aerosols, especially in polluted conditions, and could be of important effects to the subsequently developed clouds and precipitation (e.g., Yin et al. 2005).

Answer: We thank the reviewer for rising this important point. The model is initialized using an oceanic aerosol size distribution (Jaenicke, 1988; Altaratz et al., 2008). The concentrations we report are near the surface level and the aerosols are assumed to maintain constant mixing ratio with height. A constant aerosol mixing ratio is expected in a well mixed environment i.e. in the boundary layer. We note that the focus here is on shallow convective clouds and hence this is a reasonable assumption. Under the assumption of constant aerosol mixing ratio, its concentration decreases with height according to the decrease in the air density. Initially at the inversion base height (~1500m) the aerosol concentration is ~12.3% less than at the surface.

The aerosols serve as potential cloud condensation nuclei (CCN) and undergo activation based on Kohler theory (Khain et al., 2000). A prognostic equation for the aerosol is solved, which includes regeneration upon evaporation and removal by surface rain. The aerosols are assumed to be composed of ammonium-sulfate.

We added clarification to the revised manuscript: "*The shape of the aerosol size distribution was based on measurements of marine aerosol distribution (see details in Jaenicke 1988 and Altaratz et al., 2008). Eight different simulations were conducted with a changing aerosol concentration (total concentration of: 5, 25, 50, 100, 250, 500, 2000 and 5000 cm^{-3} near ground level, Dagan et al., 2015a). For reducing the sensitivity of our results to the aerosol size distribution shape and focusing on the aerosol number concentration effect, the different aerosol concentrations are calculated by multiplication of all bins in the smallest concentration size distribution by a constant factor and maintaining a similar shape of the size distribution. The aerosol is assumed to be composed of ammonium-sulfate and maintains constant mixing ratio with height. A prognostic equation is solved for the aerosol mass, including regeneration upon evaporation and removal by surface rain. Regeneration upon evaporation of cloud drops was shown to be a very important source of aerosols, especially in polluted conditions (Yin et*

al., 2005). The aerosol serves as potential cloud condensation nuclei (CCN) and it is activated based on the Kohler theory (Khain et al., 2000). The aerosol (water drop) size distribution is calculated between 5 nm to 2 μm (2 μm-3.2 mm). For both aerosol and drops, successive bins represent doubling of the mass."

3. Whether the reversing point (line 182) change with thermodynamic and dynamic conditions?

   Answer: we thank the reviewer for this comment. The reversing point (in this case for the trend of the maximum water mass as a function of aerosol loading) does change with the thermodynamic conditions (Dagan et al., 2015a). The interplay between the different aerosol effects depends on the environmental conditions. For example, under dryer conditions or smaller clouds the enhanced evaporation at the cloud margin and the consequence mixing with the outside dryer air under polluted conditions will be more significant than under humid conditions and for larger clouds (Dagan et al., 2015a).

   We added clarification to the revised manuscript: *"The total water mass (calculated as mean over time in each domain) as a function of aerosol concentration shows a clear reversal in the trend (Fig. 1A). For the given environmental conditions simulated here, it increases when increasing aerosol loading from 5 to 50 cm^{-3}. Additional increase in the aerosol loading results in a decrease in the total water mass in the domain."*

   In addition, we added clarification about the dependency of the optimal aerosol concentration on the environmental conditions to the introduction of the revised manuscript: *"Aerosol effects on single warm convective clouds were shown to have an optimal value with respect to maximal water mass, cloud depth and rain yield (Dagan et al., 2015a,b), which depends on the environmental conditions."*

   In Sec. 3.2 of the manuscript, and especially in Fig. 6, we show how, in our case, the optimal aerosol concentration changes with the thermodynamic conditions along the simulations: *"These changes in time push the optimum aerosol concentration to higher values along the simulation time."*

4. Some of the results (Line 189-190, 201) for more polluted simulations contradict with the Twomey effects. Is there any observational evidence to support these results?

   Answer: according to Twomey effect (Twomey, 1974; Twomey, 1977), for a given liquid water path (LWP) aerosol act to reduce the droplets size and hence increase the cloud albedo. Figure S1 below presents the mean vertical profiles of the droplet radius and concentration for four different simulations differ by the aerosol concentration. It demonstrates, in agreement with Twomey effect, that the mean cloud droplet radius decreases with aerosol loading (in the cloudy layer H>500m). Below cloud base the trend is reversed and the rain drops size increases with the aerosol loading (Altaratz et al., 2008; Berg et al., 2008). Based on this figure we believe that our results do not contradict the Twomey effect.

   In lines 189-190 of the manuscript we describe the results presented in Fig. 1C of the manuscript of a decrease in cloud fraction (calculated as the area covered by clouds with optical path $\tau>0.3$) with aerosol loading above 25 cm^{-3}. This was shown before based both on observational (Small et al., 2009) and numerical studies (Seigel, 2014; Xue and Feingold, 2006; Xue et al., 2008). In line 201 we mention that additional increase in the aerosol loading above 500 cm^{-3} results in a minor decrease in the cloud top height which is also related to the enhanced evaporation at the cloud margin with aerosol loading (Small et al., 2009) and does not contradict the Twomey effect.

   For clarifying this issue and providing more information about the aerosol effect on the clouds mean properties in our simulations we have added the figure below to the SI (Fig. S1).

   The additions to the revised manuscript: *"The effects of changes in aerosol concentration on the drop concentration and its mean size, for the different simulations can be found in Fig. S1 in the supporting information (SI)."*

   The additions to the SI: "*Figure S1 presents vertical profiles of the mean concentration and mean cloud drop size per height. It demonstrates that at the cloudy layer (H>500m) the mean drop size decreases with aerosol loading, while its concentration increases (Twomey, 1977). Below cloud base the trend*

is reversed – larger rain drops and lower concentration under more polluted conditions (similar to what was shown in Altaratz et al., 2008)."

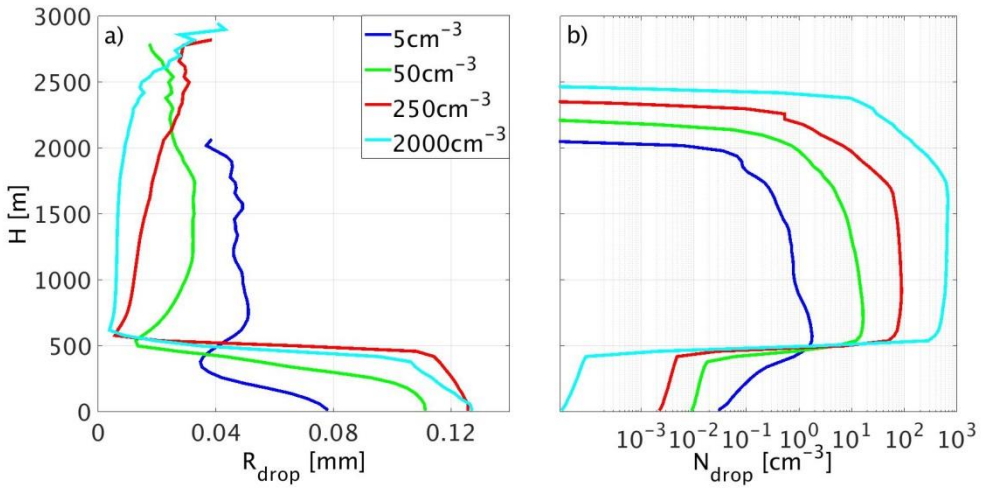

Figure S1. Vertical profiles of a) the mean (over time and domain) drop radius, and b) the mean (over time) of drop maximum concentration. These results include both cloud and raindrops for four simulations (with aerosol loading levels of 5 cm^{-3} – blue, 50 cm^{-3} – green, 250 cm^{-3} – red, and 2000 cm^{-3} – cyan).

5. Line 198-200: Is the invigoration effect limited to aerosol concentration lower than 500 cm^{-3}?

   Answer: For the maximum cloud top height and the case simulated here (BOMEX case study) the reversal in the trend occurs at aerosol concentration of 500 cm^{-3} (we expect this value to be higher for deeper clouds). Above this concertation the negative effects of aerosols, namely the increase in evaporation efficiency at the cloud margin and the water loading take over and dominate over the positive effects of increased condensation efficiency at the cloud core and the decrease in effective terminal velocity. The optimal aerosol concentration for the maximum cloud top height is higher than that of the liquid water mass (Dagan et al., 2015a) because it is affected more by the positive aerosol effects in the cloud core. As we explain it in the text: *"The maximum cloud top height (Fig. 1D), calculated as a mean over time of the*

altitude of the highest grid box in the domain that contains liquid water content (LWC >0.01g/kg) increases significantly when increasing aerosol loading up to 500 cm^{-3} (increase from 1692 m to 2120 m when increasing aerosol loading from 5 to 500 cm^{-3}). Additional increase in the aerosol loading results in a minor decrease in the maximum cloud top height (down to 2030 m for aerosol loading of 5000 cm^{-3}). The minor decrease seen for this range of aerosol concentration (compared with the larger decrease in the mean LWP for example) can be explained by the location of the maximal cloud top height above the cloud core, which is affected mainly by the invigoration processes (enhanced condensation and latent heat release) and less by margin oriented processes (enhanced entrainment and evaporation) that significantly impact the total cloud mass (Dagan et al., 2015a). Another reason is the cloud deepening effect under polluted conditions (Stevens, 2007; Seifert et al., 2015) that will be described later."

6. Line 251-252: Similar trend is also seen for maximum cloud top height. Is the decrease in COG height for larger aerosol concentration related to the inversion layer above cloud which prohibited the further growth of clouds?
   Answer: In the polluted conditions simulations the highest cloud tops penetrate the inversion layer, evaporate there and hence drive an increase in the inversion base height with time. However, as the initial inversion base height is similar in all simulations it cannot explain the differences in cloud top between the simulations at the initial stage. At aerosol concentration above 500 cm^{-3} the mean COG slightly decreases with aerosol loading since the negative effects of aerosol take over. Above this concertation the increase in evaporation efficiency at the cloud margin and the water loading take over and dominate the increase in the condensation efficiency at the cloud core and the decrease in effective terminal velocity. For the COG (as well as for the maximum cloud top height) the optimal aerosol concentration is higher than the one for the liquid water mass because it is more effected by the positive aerosol effects in the cloud core (Dagan et al., 2015a).

7. Line 259-260: The LWP is decreasing with larger aerosol concentration. Is the water loading larger?

Answer: We thank the reviewer for this comment. It is true that the mean LWP decreases in the simulations with aerosol concentration above 100 cm^{-3}. Below 100 cm^{-3} an increase in the aerosol loading results in an increase in the mean LWP (Fig. 1B in the main text). For high aerosol loading (>100 cm^{-3}) both the LWP and the cloud fraction decrease with aerosol loading but for most vertical levels the liquid water content (LWC) increases.
Figure 4 in the text presents vertical profiles of the mean LWC for four simulations differ by the aerosol loading which implies about the water loading effect.

The increase in the water loading effect under polluted conditions is a result of the rain suppression (Fig. 1F) and the decrease in the effective terminal velocity (Fig. 3B) that drive the water mass to be pushed higher in the atmosphere.

Regarding the apparent contradiction between the trend in LWP (decreasing for aerosol concentration above 100 cm^{-3} – Fig. 1B) and the mean LWC (generally increases with aerosol loading – Fig. 4). The values calculated in Fig. 4 are for the cloudy pixels. The LWC generally increases with height but the mean cloud fraction decreases (above the cloud base H~500m - see Fig. S2 below). Considering the two factors together (vertical profiles of CF and LWC) results in a decrease in LWP with aerosol concentrations above 100 cm^{-3}.

We added the figure below to the SI as well as explanations and clarifications regarding this point to the revised manuscript: *"We show that both the height and the magnitude of the maximum LWC increase with the aerosol loading. This is due to both rain suppression (Fig. 1F) and an increased V_{COG} (Fig 3C) with aerosol loading. There is a reduction in the mean LWP (for >100 cm^{-3} - Fig. 1B) although there is an increase in the LWC with aerosol loading due to the differences in cloud fraction (Fig. 1C, Fig. S2). Below the clouds' base (H<~550m) the LWC trend is reversed due to the enhancement of rain in the clean runs (Fig. 1F). The increase in LWC with aerosol loading implies a larger water loading negative component in the clouds' buoyancy."*

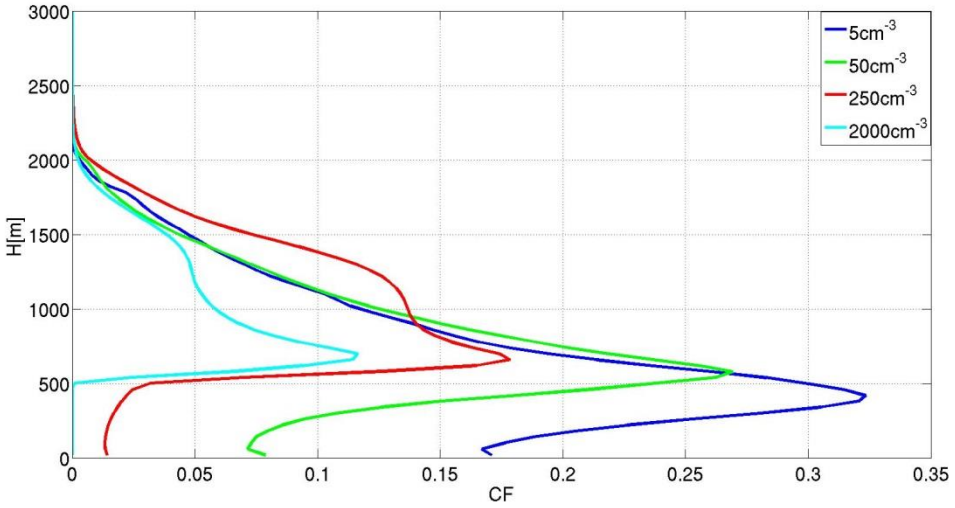

Figure S2. Vertical profiles of the mean (over time and domain) cloud fraction (CF) per height for four simulations (with aerosol loading levels of 5 cm^{-3} – blue, 50 cm^{-3} – green, 250 cm^{-3} – red, and 2000 cm^{-3} – cyan).

8. Line 297-299: Suggest to show 1-2 figures related the time variations of cloud fields to support the statements.

   Answer: we thank the reviewer for the suggestion. Following this comment we have added to the new supporting information (SI) file a figure presenting the temporal evolution of the vertical profiles of the thermodynamic conditions (temperature and water vaper mixing ratio). Figure S3 (see below) presents the time evolution of the mean thermodynamic conditions for two simulations with aerosol loading of 50 cm^{-3} (clean – upper row) and 2000 cm^{-3} (polluted – lower row). This figure demonstrates that under clean conditions the cloudy layer (H~500-1500m) becomes warmer and dryer with time while the sub-cloud layer becomes colder and wetter. On the other hand, under polluted conditions the trend is different and the most significant change occurs at the inversion layer which significantly cools and becomes wetter with time.

   These different changes in the thermodynamic conditions under clean and polluted conditions are the focus of another new paper (Dagan et al., 2016).

   The addition to the revised manuscript: *"Figure 5 presents the changes (final value minus initial one) in the temperature (T) and water vapor content (q_v) vertical profiles as a function of aerosol concentration used in the simulation.*

The initial profiles were identical in all simulations. Figure S3 (in the SI) presents the full temporal evolution of those parameters."

The additions to the SI: "*Figure S3 presents the temporal change in vertical profiles of the temperature (left column) and water vapor mixing ratio (right column) for clean (50 cm^{-3} – upper row) and polluted (2000 cm^{-3} – lower row) conditions. It demonstrates that under clean conditions the cloudy layer (H~500-1500m) becomes warmer and dryer with time while the sub-cloud layer becomes colder and wetter. On the other hand, under polluted conditions the trend is different and the most significant change occurs at the inversion layer which significantly cools and becomes wetter with time".*

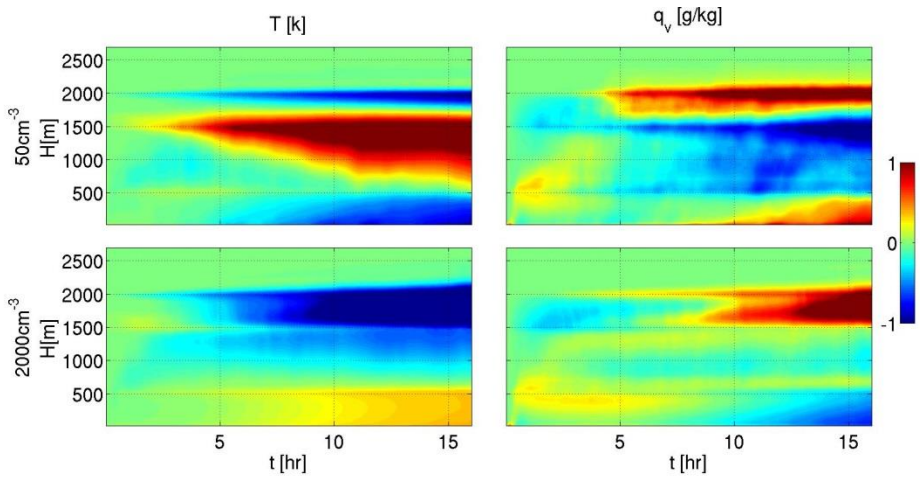

Fig. S3 Temporal changes compared to the initial profiles of mean environmental temperature [K] (left) and mean water vapor mixing ratio [g/kg] (right). For two different simulations with aerosol concentrations of 50cm^{-3} (clean – upper row) and 2000cm^{-3} (polluted – lower row).

9. Line 331-335: Suggest to add more explanations to the results.

Answer: we thank the reviewer for this comment. Detailed explanations for this point are given at the next paragraph. We added clarification to the revised manuscript: "*Trends in the mean rain rate show that in the cleanest simulations (5, 25 and 50 cm^{-3}) it decreases with time (Fig. 1H, 53.3, 32.9 and 40.1%,*

respectively). In the regime of medium to fairly high aerosol loading (100, 250 and 500 cm^{-3}) the rain rate increases (19.6, 598.1 and 841.5%, respectively). And in the most polluted simulations (2000 and 5000 cm^{-3}) the surface rain is negligible throughout the simulation time. These trends are explained below

The time evolution of the thermodynamic conditions (Fig. 5) shows a reduction (enhancement) in the thermodynamic instability with time in the clean (polluted) simulations. Figure 6 and table 1 indicate that under clean conditions the decrease in the thermodynamic instability with time leads to a decrease in the mean cloud field properties such as total mass, cloud top height and rain rate. Under polluted conditions the trends are opposite and the mean cloud field properties increase with time due to the increase in thermodynamic instability (Dagan et al., 2016) and due to the cloud deepening (Stevens and Seifert, 2008; Stevens, 2007; Seifert et al., 2015). These differences between the clean and polluted simulations drive changes in the optimum aerosol concentration with time. For example, for the LWP (Fig. 1B) the optimum aerosol concentration is 50, 100 and 250 cm^{-3} for the first, second and third parts of the simulation, respectively."

Technical corrections:

1. Line 35: add “water vapor and” at the end of this line;

   Answer: Following this comment, we have added the reviewer suggestion to the revised manuscript: "*Warm cloud (containing liquid water only) formation depends on the availability of water vapor and aerosols acting as cloud condensation nuclei (CCN)."*

2. Line 63: remove “,”between the parentheses;

   Answer: Thank you for this comment. We have removed it: *"For aerosol concentrations lower than the optimum, the positive relationship between aerosol concentration and cloud development is a result of two main processes: 1) larger latent heat release driven by the increase in the condensation efficiency causing stronger updrafts, and 2) decrease in the*

effective terminal velocity (η, i.e. mass weighted terminal velocity of the hydrometeors) (Koren et al., 2015) due to initial smaller droplets and the delay in the collision-coalescence process."

3. Line 158: “in (Siebesma et al., 2003)”should be replaced by “by Siebesma et al. (2003)”;

   Answer: We corrected it in the revised manuscript: *"This case was initialized using the setup specified in Siebesma et al. (2003)."*

4. Line 184-185: remove ();

   Answer: We have removed it in the revised manuscript: "*The LWP (Liquid Water Path - Fig. 1B) calculated as a mean over time over all cloudy columns in each domain, which is strongly correlated with the total water mass, also shows the same non-monotonic general trend."*

5. Line 295: change “less” to “minus”;

   Answer: We corrected it in the revised manuscript*: "Figure 5 presents the changes (final value minus initial one) in the temperature (T) and water vapor content (q_v) vertical profiles as a function of aerosol concentration used in the simulation."*

6. Line 625: Add the variable for the abscissa.

   Answer: It was added in the revised manuscript:

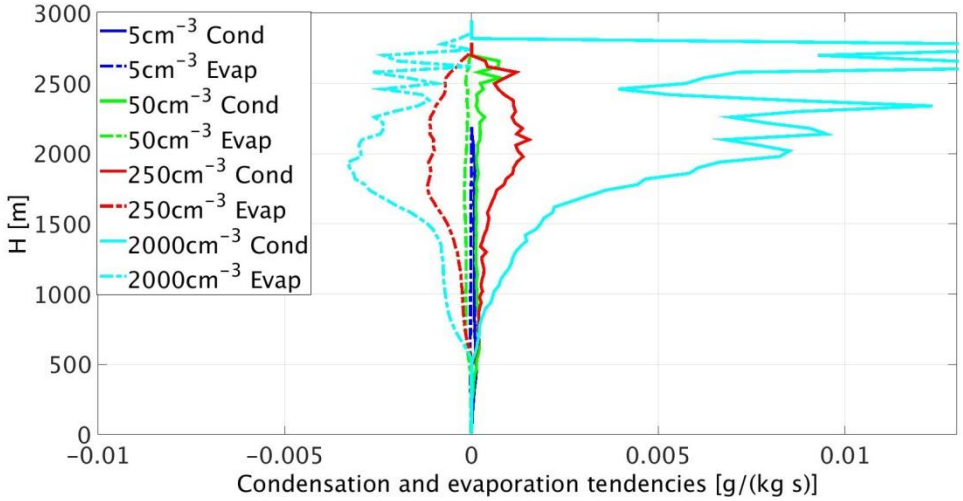

Figure 2. Domain mean condensation (solid lines) and evaporation (dashed lines) tendencies for four different simulations conducted with different aerosol concentration levels (5 cm^{-3} blue, 50 cm^{-3} green, 250 cm^{-3} red and 2000 cm^{-3} cyan).

Anonymous Referee #2

The manuscript investigated cloud-aerosol interaction in warm cloud environment using a Large Eddy Simulations (LES) with bin microphysics. The paper is generally well written, contains some potentially interesting model results, but does not go into sufficient detail in any one of the cases and there is no comparison with observations which is necessary for a reality check. The article can be considered for publication after major revision.

Answer: we thank the reviewer for acknowledging our interesting results. We have changed the manuscript according to all of the reviewer's comments and we believe it presents the work in sufficient details now.

Some specific comments and suggestions follow:

1. The algorithm of cloud-aerosol interaction in the model is not clear. Suggest to elaborate the section 2 (methodology) in details. How the aerosol initiations are taking place? What is the composition of aerosols they have considered in their model?

   Answer: We thank the reviewer for rising this important point. The model is initialized using an oceanic aerosol size distribution (Jaenicke, 1988; Altaratz et al., 2008). The concentrations we report are near the surface and the aerosols are assumed to maintain constant mixing ratio with height.

   The aerosols serve as potential cloud condensation nuclei (CCN) and activated based on Kohler theory (Khain et al., 2000). A prognostic equation for the aerosol is solved, which includes regeneration upon evaporation and removal by surface rain. The aerosols are assumed to be composed of ammonium-sulfate.

We added this information to the revised manuscript: *"The shape of the aerosol size distribution was based on measurements of marine aerosol distribution (see details in Jaenicke 1988 and Altaratz et al., 2008). Eight different simulations were conducted with a changing aerosol concentration (total concentration of: 5, 25, 50, 100, 250, 500, 2000 and 5000 cm^{-3} near ground level, Dagan et al., 2015a). For reducing the sensitivity of our results to the aerosol size distribution shape and focusing on the aerosol number concentration effect, the different aerosol concentrations are calculated by multiplication of all bins in the smallest concentration size distribution by a constant factor and maintaining a similar shape of the size distribution. The aerosol is assumed to be composed of ammonium-sulfate and maintains constant mixing ratio with height. A prognostic equation is solved for the aerosol mass, including regeneration upon evaporation and removal by surface rain. Regeneration upon evaporation of cloud drops was shown to be a very important source of aerosols, especially in polluted conditions (Yin et al., 2005). The aerosol serves as potential cloud condensation nuclei (CCN) and it is activated based on the Kohler theory (Khain et al., 2000). The aerosol (water drop) size distribution is calculated between 5 nm to 2 μm (2 μm-3.2 mm). For both aerosol and drops, successive bins represent doubling of the mass."*

2. Does the model take nucleation scavenging? Need to be cleared.

   Answer: The prognostic equation for aerosol solved by the model does consider removal by surface rain. As was mentioned above, this information was added to the revised manuscript: *"A prognostic equation is solved for the aerosol mass, including regeneration upon evaporation and removal by surface rain."*

3. How does different size distribution of aerosols (CCN) affect the DSD? What is the effect of large CCN on the processes of droplet nucleation?

Answer: We thank the reviewer for this comment. For reducing the sensitivity of our results to the aerosol size distribution and focusing on the aerosol number concentration effect we have kept the shape of the aerosol size distribution constant in all different simulations. We have increased the aerosol number concertation by a constant factor, multiplying all bins. We added this information to the methodology section of the revised manuscript: "*For reducing the sensitivity of our results to the aerosol size distribution shape and focusing on the aerosol number concentration effect, the different aerosol concentrations are calculated by multiplication of all bins in the smallest concentration size distribution by a constant factor and maintaining a similar shape of the size distribution".*

In our case, as will be demonstrate later (answer no. 13), increasing the aerosol number concentration results in more and smaller cloud droplets. This can also be seen in Fig. 1 below which presents examples of aerosol and drop size distributions for four different simulations. The aerosol size distributions represent the initial conditions near the surface while the drop size distributions represent a level slightly above the cloud base (H=660m) after 2 hours of simulations. The drop size distribution is an average over all cloudy pixels (LWC>0.01 g/kg) with vertical velocity above 1 m/s.

Figure 1 below demonstrates that under polluted conditions there are more numerus and smaller droplets compared to clean conditions.

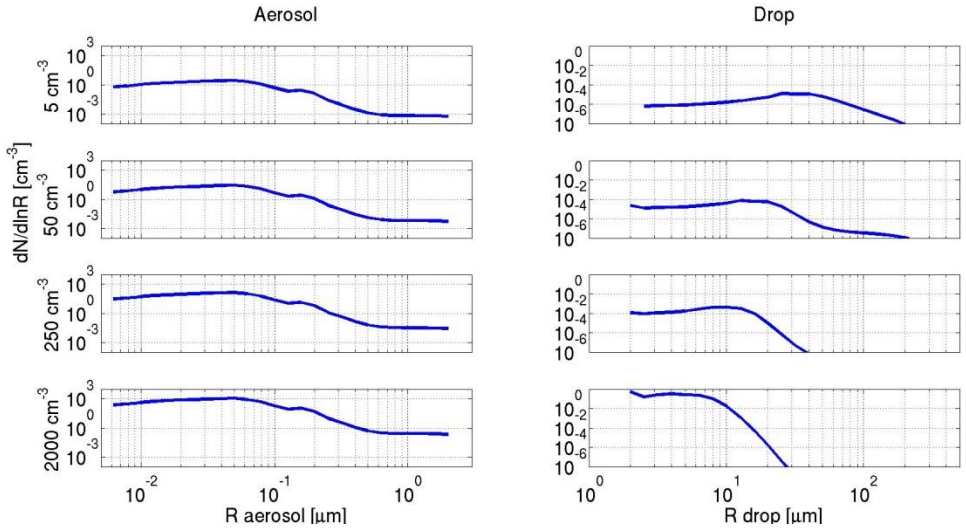

Fig. 1. Mean initial near surface aerosol size distribution (left column) and mean drop size distribution above cloud base (H=660m right column) after two hours of simulation for four different simulations (5cm^{-3} – first row, 50cm^{-3} – second row, 250cm^{-3} – third row, and 2000cm^{-3} – bottom row). The drop size distribution is an average over all cloudy pixels (LWC>0.01 g/kg) with vertical velocity above 1 m/s.

The sensitivity of the results presented here (especially the non-monotonic response of the cloud fields' mean properties) to different aerosol size distributions that do not include GCCN (giant CCN) is small. The largest aerosol radius bin used in this study is 2 μm and hence large GCCN are not considered here.

However, the response of rain amount to aerosol concentration is sensitive to the availability of GCCN (Dagan et al., 2015b; Feingold et al., 1999; Yin et al., 2000). The availability of GCCN can produce rain even under extremely polluted conditions. The existence of GCCN at the cloud base height can be considered as a special case. Khain et al. (2000) claimed that GCCN can affect the initiation of the collection process only if they reach the cloud base height in concentrations that are comparable to rain drop concentrations (10^{-4}-10^{-3} cm^{-3}), which for large aerosol is not always trivial. Thus, for simplicity reasons, in this study we choose to avoid the GCCN effect. Moreover, even if GCCN are available, some reduction in rain amount is expected under high aerosol loading (Dagan et al., 2015b).

4. Authors have discussed on the role of aerosol on collision-coalescence, delayed surface rain in their introduction (line 53- 56) and also about cloud invigoration mechanism in line 80-83. In this context authors should put the references (Hazra et al. 2013a, Journal of Atmospheric Science; Hazra et al. 2013b, Journal of Geophysical Research-Atmosphere) studied cloud-aerosol interactions using 2 moment bulk microphysical scheme (Cheng et al. 2007, QJRMS) in meso-scale model. In this regard, I suggest refer/include all those papers in the manuscript.

   Answer: we thank the reviewer for this comment. We added those references to the revised manuscript:

   "The initiation of collision-coalescence is delayed in polluted clouds (Gunn and Phillips, 1957; Squires, 1958; Albrecht, 1989). This drives a delay in rain formation and can affect the amount of surface rain (Rosenfeld, 1999, 2000; Cheng et al., 2007 ; Khain, 2009; Levin and Cotton, 2009; Koren et al., 2012; Hazra et al., 2013a;b; Dagan et al., 2015b).

   "*The invigoration mechanism, which refers to deeper and larger clouds with larger mass that develop under polluted conditions was studied mainly in deep convective clouds (Andreae et al., 2004; Koren et al., 2005; Rosenfeld et al., 2008; Tao et al., 2012; Fan et al., 2013; Hazra et al., 2013a; Altaratz et al., 2014). Our focus here is on warm cloud fields for which previous observational studies reported on invigoration effect or a non-monotonic response of the clouds to an increase in aerosol loading."*

5. Line 87-91: How aerosol impact on cloud fraction (CF), discussion not clear.

   Answer: in this part of the introduction we discuss previous observational works that examined the relations between aerosol and warm convective clouds' properties. Cloud fraction (CF) was proposed to be affected by aerosol due to rain suppression and prolonging the cloud lifetime (Albrecht, 1989). In addition, warm convective cloud invigoration by aerosol (larger, deeper and stronger precipitating clouds under polluted conditions) also drive increase in the cloud fraction (Koren et al., 2014).

Following this comment we have added clarification to the revised manuscript: *"From convective stability considerations deeper clouds tend to have larger area (larger CF). It was shown that warm convective cloud's area correlates positively with cloud's depth (Benner and Curry, 1998; Koren et al., 2008)."*

6. Line: 169-172: Authors have mentioned the aerosols number concentration. Is it total concentration of all bins? Or which bin size of aerosols number concentrations they are increasing. This should be cleared to the readers.

   Answer: In this sentence we were referring to the total aerosol concentration. For simplicity reasons (as was explained in answer No. 3), the changes in aerosol concentration are done by multiplying all bins by a constant factor and maintaining a similar aerosol size distribution. We added clarification to the revised manuscript: *"Eight different simulations were conducted with a changing aerosol concentration (total concentration of: 5, 25, 50, 100, 250, 500, 2000 and 5000 cm^{-3} near ground level, Dagan et al., 2015a). For reducing the sensitivity of our results to the aerosol size distribution shape and focusing on the aerosol number concentration effect, the different aerosol concentrations are calculated by multiplication of all bins in the smallest concentration size distribution by a constant factor and maintaining a similar shape of the size distribution."*

7. Line 177-179: Authors mentioned "the eight simulated cloud fields are examined first". Are the eight simulated cloud fields are in similar or different. Need to be explained.

   Answer: in this sentence we describe that at this section the aerosol effect on the mean cloud field properties are examined for eight simulations. The eight different simulations describe different aerosol concentrations. To make this point clearer, this sentence was revised: *"The aerosol effects on the mean properties of the eight simulated cloud fields (differ by the aerosol loading) are examined first."*

8. In Figure 1 and Figure 6: Total liquid water mass, LWP, CF and rain rate show a “tipping” point. It will be worthy if authors discuss on the “tipping point” as revealed in Figure 1 and 6. Another important point authors should explain why the “tipping point” for total water mass, LWP are different from the CF and rain rate.

   Answer: We thank the reviewer for this comment. We agree with the reviewer that this “tipping point” is the heart of this paper and should be explained thoroughly.

   We refer to this “tipping point” as a non-monotonic response with an optimal aerosol concentration and try to put this result in the focus of this paper (starting from the title). The non-monotonic trends with an optimal aerosol concentration of the mean cloud field properties are explained by contradicting aerosol effects. Namely, two positive aerosol effects: 1) increase in the condensation efficiency (as shown in Fig. 2 of the main text), and 2) increase in the droplet mobility (as shown in Fig. 3 in the main text). And two negative aerosol effects: 1) increase in the evaporation efficiency (as shown in Fig. 2 of the main text), and 2) increase in the water loading (as shown in Fig. 4 in the main text). All of those effects are discussed and presented in the paper.

   Regarding the different optimal aerosol concentration for the different cloud field properties, in a previous study (Dagan et al., 2015a) it was shown that more core oriented cloud properties have a higher optimal aerosol concentration than periphery based properties. For example, the strongest vertical velocities (W) in the domain are more frequent at the clouds' cores in which the conditions can be considered as closer to adiabatic. As a result W will be more affected by the increase in the condensation efficiency with aerosol loading (more related to the cloud core) then by the increase in the evaporation efficiency (more related to the cloud periphery) and will have high optimal aerosol concentration (Fig. 3A). A different example is the total cloud mass which is determine by both the cloud core and cloud periphery and hence affected by both the positive and negative aerosol effects. As a result the total cloud mass will have lower optimal aerosol concentration. Each of the different cloud field properties presented in figures 1 and 6 is a result of a different blend of core and periphery oriented processes and hence has a different optimal aerosol concentration.

This is being explained in the text: "*The minor decrease seen for this range of aerosol concentration (compared with the larger decrease in the mean LWP for example) can be explained by the location of the maximal cloud top height above the cloud core, which is affected mainly by the invigoration processes (enhanced condensation and latent heat release) and less by margin oriented processes (enhanced entrainment and evaporation) that significantly impact the total cloud mass (Dagan et al., 2015a). Another reason is the cloud deepening effect under polluted conditions (Stevens, 2007; Seifert et al., 2015) that will be described later.*"

9. Can authors consider another aerosol concentration at lower rage?

   Answer: We thank the reviewer for this comment. The lowest aerosol concentration used in this study is 5 cm^{-3} which is already extremely low and is not common in the natural environment. We have used an extremely large range of aerosol concentrations (3 orders of magnitudes, between 5 and 5000 cm^{-3}). Which is much larger then in many of the recent cloud-aerosol interactions studies (e.g. Seifert et al., 2015 who used a range of 35-105 cm^{-3} – factor of three). In addition, we have used relatively high resolution (8 different aerosol concentration) especially at the lower range (5, 25, 50, 100, 250, 500, 2000 and 5000 cm^{-3}).

   Under aerosol concentration lower than 5 cm^{-3} we expect a very limited clouds' development due to the very low aerosol concentration. The total droplet surface area will be extremely low and hence the condensational growth of the drops will be very non-efficient. In addition, the low amount of droplets would result in insignificant competition between the droplets on the available water vapor and hence they would grow fast and start to precipitate early. The early initialization of precipitation will act as a positive feedback and reduce even more the droplet surface area and condensation rate.

10. Authors have showed the response of vertical wind (updraft) to varying CCN concentrations (Figure 3A). It shows updraft is monotonically increasing with aerosol concentrations. Is it correct? Need to be explained.

Answer: Increasing aerosol loading result in an increase in the condensation efficiency at the cloud core (super-saturated volumes) and hence increase in the latent heat release. This results in an increase in the buoyancy and vertical velocity in the polluted clouds. In Fig. 3A the mean weighted by the mass vertical velocity is presented for being consistent with the COG point of view. This mass-weighted mean represent more the cloud core then the cloud margin (which has less mass). As was explained in answer 8, the strongest vertical velocities (W) in the domain are more frequent at the clouds' cores in which the conditions can be considered as closer to adiabatic. As a result W will be more effected by the increase in the condensation efficiency with aerosol loading (more related to the cloud core) then by the increase in the evaporation efficiency (more related to the cloud periphery) and will have high optimal aerosol concentration. At high aerosol concentrations (above 2000 cm^{-3}) this effect saturates and the mean updraft remains similar.

This is being explained in the text: *"The mean updraft (in both space and time, weighted by the liquid water mass in each grid box to be consistent with the COG point of view - Fig. 3A) increases with the increase in aerosol loading, in agreement with previous studies (Saleeby et al., 2015; Seigel, 2014). This indicates an increase in the latent heat contribution to the cloud buoyancy, driven by increase in the condensation efficiency (Dagan et al., 2015a,b; Koren et al., 2014; Pinsky et al., 2013; Seiki and Nakajima, 2014) (Fig. 2)."*

11. Another doubt: Figure 1B and Figure 4: Figure 1B shows clear tipping point of LWP, first it increases then it decreases, whereas when I am looking into the Figure 4, LWC increasing with aerosol number concentrations. Authors should make it unambiguous.

    Answer: The apparent contradiction between the trend in LWP (decreasing for aerosol concentration above 100 cm^{-3} – Fig. 1B) and the mean LWC (generally increase with aerosol loading – Fig. 4) is because of the different cloud fraction in the different heights (see Fig. S2 below). The values calculated in Fig. 4 are for the cloudy pixels. The LWC generally increase with height but the mean cloud fraction decreases (above the cloud base H~500m - Fig. S2). Considering the two factors together (vertical profiles of

CF and LWC) results in a decrease in LWP with aerosol loading at concentrations above 100 cm^{-3}.

The increase in the water loading effect under polluted conditions is a result of the rain suppression (Fig. 1F) and the decrease in the effective terminal velocity (Fig. 3B) that drive the water mass to be pushed higher in the atmosphere.

We added the figure below to the SI as well as explanations and clarifications regarding this point to the revised manuscript: *"We show that both the height and the magnitude of the maximum LWC increase with the aerosol loading. This is due to both rain suppression (Fig. 1F) and an increased V_{COG} (Fig. 3C) with aerosol loading. There is a reduction in the mean LWP (for >100 cm^{-3} - Fig. 1B) although there is an increase in the LWC with aerosol loading due to the differences in cloud fraction (Fig. 1C, Fig. S2). Below the clouds' base (H<~550m) the LWC trend is reversed due to the enhancement of rain in the clean runs (Fig. 1F). The increase in LWC with aerosol loading implies a larger water loading negative component in the clouds' buoyancy."*

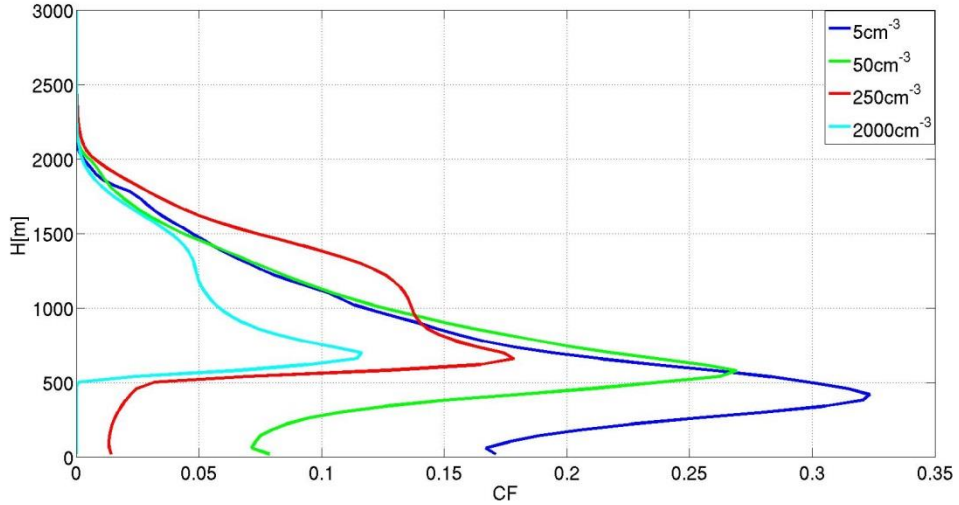

Figure S2. Vertical profiles of the mean (over time and domain) cloud fraction (CF) per height for four simulations (with aerosol loading levels of 5 cm^{-3} – blue, 50 cm^{-3} – green, 250 cm^{-3} – red, and 2000 cm^{-3} – cyan).

12. Time dependent part (Figure 6, line 338-349) is not much convincing. Need to be explained in details.

Answer: we thank the reviewer for this comment. Following this comment we have added additional information to the revised manuscript to improve clarity. Figure S3 (see below) presents the temporal evolution of the vertical profiles of the thermodynamic conditions (temperature and water vaper mixing ratio) for two simulations with aerosol loading of 50 cm^{-3} (clean – upper row) and 2000 cm^{-3} (polluted – lower row). This figure demonstrates that under clean conditions the cloudy layer (H~500-1500m) becomes warmer and dryer with time while the sub-cloud layer becomes colder and wetter. Those changes drive reduction of the thermodynamic instability with time. On the other hand, under polluted conditions the trend is different and the most significant change occurs at the inversion layer which significantly cools and becomes wetter with time. Those changes drive increase in the thermodynamic instability with time.

The differences in the thermodynamic evolution under clean and polluted conditions drive changes in the mean cloud field properties (Fig. 6). Under clean conditions the thermodynamic instability decreases and hence the mean cloud field properties (total mass, CF, mean LWP) also decrease with time. On the other hand, under polluted conditions the thermodynamic instability increases with time and hence also the mean cloud field properties.

These different changes in the thermodynamic conditions under clean and polluted conditions are the focus of another new paper (Dagan et al., 2016).

The addition to the revised manuscript: *"Figure 5 presents the changes (final value minus initial one) in the temperature (T) and water vapor content (q_v) vertical profiles as a function of aerosol concentration used in the simulation. The initial profiles were identical in all simulations. Figure S3 (in the SI) presents the full temporal evolution of those parameters."*

The additions to the SI: "*Figure S3 presents the temporal change in the temperature (left column) and water vapor mixing ratio (right column) vertical profiles for clean (50 cm^{-3} – upper row) and polluted (2000 cm^{-3} – lower row) conditions. It demonstrates that under clean conditions the cloudy layer (H~500-1500m) becomes warmer and dryer with time while the sub-*

cloud layer becomes colder and wetter. On the other hand, under polluted conditions the trend is different and the most significant change occurs at the inversion layer which significantly cools and becomes wetter with time".

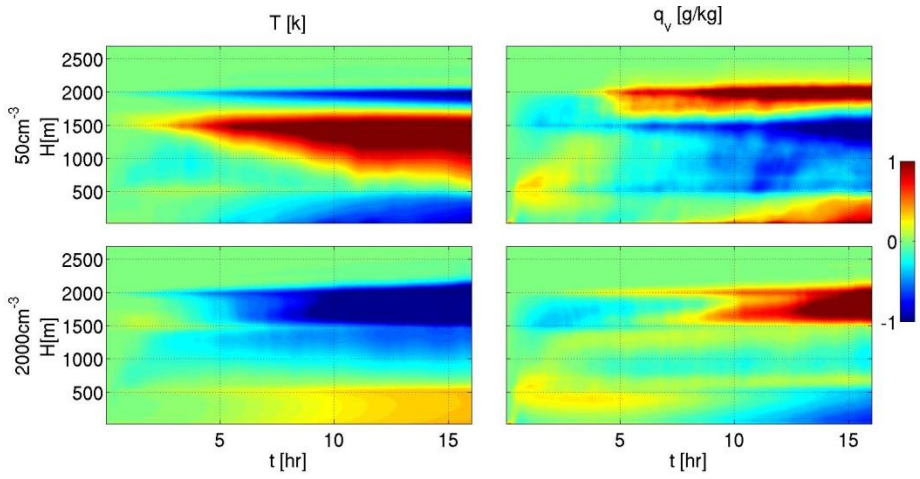

Fig. S3. Temporal changes compared to the initial profiles of mean environmental temperature [K] (left) and mean water vapor mixing ratio [g/kg] (right). For two different simulations with aerosol concentrations of 50cm^{-3} (clean – upper row) and 2000cm^{-3} (polluted – lower row).

13. It will be worthy if authors provide cloud drop number concentration and cloud drop size with increasing aerosol concentration.

    Answer: we thank the reviewer for this comment. Following this comment we have added the manuscript supporting information file (SI) some information about the cloud drop mean size and concentration for the different simulations (differ by the aerosol loading). Information about the aerosol and drop size distribution is presented in answer number 3 above. Figure S1 below presents the vertical profiles of the mean cloud drop concentration and size. It demonstrates that at the cloudy layer (H>500m) the mean drop size decreases with the aerosol loading, while its concentration increases (Twomey, 1977). Below cloud base the trend is reversed – larger concentration and smaller rain drops under clean conditions (Altaratz et al., 2008).

The additions to the revised manuscript: *"The effects of changes in aerosol concentration on the drop concentration and its mean size, for the different simulations can be found in Fig. S1 in the supporting information (SI)."*

The additions to the SI: "*Figure S1 presents vertical profiles of the mean concentration and mean cloud drop size per height. It demonstrates that at the cloudy layer (H>500m) the mean drop size decreases with aerosol loading, while its concentration increases (Twomey, 1977). Below cloud base the trend is reversed – larger rain drops and lower concentration under more polluted conditions (similar to what was shown in Altaratz et al., 2008).*"

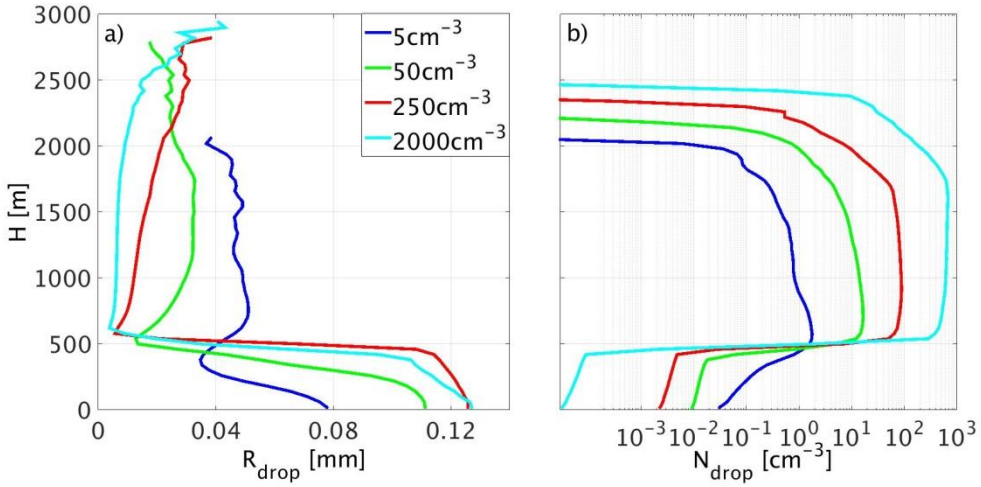

Figure S1. Vertical profiles of a) the mean (over time and domain) drop radius, and b) the mean (over time) of drop maximum concentration. These results include both cloud and raindrops for four simulations (with aerosol loading levels of 5 cm^{-3} – blue, 50 cm^{-3} – green, 250 cm^{-3} – red, and 2000 cm^{-3} – cyan).

14. Can authors put some hints of cloud-aerosol interactions in the mixed-phase clouds in their "Conclusion" section?

Answer: We thank the reviewer for this comment. The results presented in this study are for the aerosol effect of the mean properties of warm convective cloud fields. Warm processes act as the initial and boundary conditions for mixed-phase processes in deep convective clouds. Hence, understanding the

aerosol effect on the warm processes is the first step in understanding aerosol effects on deep convective clouds. We can expect that the aerosol effect on the warm processes (condensation-evaporation, collection etc.) in deep convective clouds would be similar to the ones described here for warm convective clouds (especially during the first stages of the cloud, when there are only warm processes). However, ice processes are much more complicated and much is still unknown about them so taking them into consideration making the aerosol effect much harder for prediction. The additional phase transition and the interactions between phases add many levels of complexity. Great part of the challenges in understanding aerosol effects on deep convective clouds is attributed to the fact that ice nucleation is far from being fully understood (DeMott et al., 2015). Ice nucleation processes are shown to be extremely sensitive to the aerosol surface properties in a way that is not yet understood (Vali, 2014) and many freezing schemes are based on empirical relations that are far from being converged to one comprehensive theory. Therefore we have chosen to gain first a detailed process level understanding of the warm phase.

Nevertheless, we expect the general trend to be similar. Regarding the thermodynamic feedback for example, in deep convective clouds, higher aerosol loading was shown to result in larger water mass being pushed to higher levels (i.e. Hazra et al., 2013a; Koren et al., 2010; Rosenfeld et al., 2008; Storer and van den Heever, 2012, among many others). This is expected to result in more evaporation at higher levels, hence cooling and moistening of those levels and increasing the atmospheric instability. Since the convective clouds that pass the freezing level are usually thicker, we expect the optimal aerosol concentrations to be higher (Dagan et al., 2015a). And indeed, aerosols were reported to increase rain amount from deep convective clouds (Hazra et al., 2013a; Koren et al., 2012; Li et al., 2008). This could lead to the opposite result of warming of the cloudy layer and cooling of the sub-cloud layer by rain evaporation, hence consumption of the atmospheric instability. Determining the end result of those two contradicting effects cannot be done a-priory and should be investigated in details. Because this subject is highly speculative and the fact that we do not show anything regarding aerosol effect on deep convective clouds in this manuscript, we prefer not to include any

speculations about it in the discussion and to keep this subject to future studies.

15. Line 35-36: It is not only depends on CCN concentrations, also depends on availability of water vapor.

    Answer: We thank the reviewer for this comment. We changed this sentence in the revised manuscript: "*Warm cloud (containing liquid water only) formation depends on the availability of water vapor and aerosols acting as cloud condensation nuclei (CCN).*"

16. The authors probably should proof read the manuscript carefully to correct some typos

    Answer: we thank the reviewer for this comment. The manuscript has been gone thought proof reading.

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