Response to Anonymous Referee #2

Pistone et al. present data and analysis of aerosol, cloud and meteorological data in a trade wind regime over the Indian Ocean. Its subject and content is surely of interest to ACP. Here are some comments for the authors and the editor to consider.

We thank this reviewer for their careful reading of our manuscript. Responses to comments (reproduced as normal face text) are given below in bold, with edits to passages within the text provided in *italics*.

Major comments:

1. The abstract needs to be rewritten. I think it can be more specific and to the point. In particular, they could highlight the uncertainties involved in the interpretation of the results they have.

Thank you for the comment, we have revised the abstract as follows:

> There are many contributing factors which determine the micro- and macrophysical properties of clouds, including atmospheric *vertical* structure, dominant meteorological conditions, and aerosol concentration, all of which may be coupled to one another. In the quest to determine aerosol effects on clouds, these potential relationships must be understood. Here we describe several observed correlations between aerosol conditions and cloud and atmospheric properties in the Indian Ocean winter monsoon season.
>
> In the CARDEX (Cloud, Aerosol, Radiative forcing, Dynamics EXperiment) field campaign conducted in February and March 2012 in the northern Indian Ocean, continuous measurements *were made* of atmospheric precipitable water vapor and the liquid water path (LWP) of trade cumulus clouds, concurrent with measurements of water vapor flux, cloud and aerosol vertical profiles, meteorological data, and surface and total-column aerosol. *We present observations indicating* a positive correlation between aerosol and cloud LWP which becomes clear only after the data are filtered to control for the natural meteorological variability in the region.
>
> We then use the aircraft and ground observatory measurements to explore *possible* mechanisms behind the observed aerosol–LWP correlation. *High pollution is found to correlate with higher temperatures and higher humidity measured throughout the boundary layer. The increase in cloud liquid water is found to coincide with a lowering of the cloud base resulting from the increased boundary-layer humidity. Large-scale analysis corroborates these co-variations: high pollution cases are shown to originate as a highly-polluted boundary layer air mass approaching the observatory from a northwesterly direction. This polluted mass exhibits higher temperatures–potentially attributable to aerosol absorption of solar radiation over the subcontinent–and higher humidity than the cleaner cases. While high aerosol conditions are observed to disperse with air mass evolution, along with a weakening of the high-temperature*

anomaly, the high humidity condition was observed to instead strengthen in magnitude coincident with the polluted air mass. Potential causal mechanisms of the observed correlations are then explored, though future research will be needed for a more complete and quantitative understanding of the aerosol–humidity relationship.

2. In the introduction, the material is almost exclusively about previous studies done in the Indian Ocean region, which is perfectly fine since that is the place where this experiment took place. However, since the authors also emphasize a view on the general aerosol-cloud interaction problem within the trade cumulus regime, references and discussions about other papers may be needed to put current study in a proper context.

Following this comment and the comment of Reviewer #1, we have added additional background text to the introduction:

> As nations in southeast Asia have increased bio- and fossil fuel combustion in recent decades, corresponding increases in atmospheric aerosol pollution have been seen over the region (e.g. Ramanathan et al, 2001). The high levels of anthropogenic emissions combine with the seasonal monsoon cycle (Lawrence and Lelieveld, 2010) to cause frequent episodes of heavy air pollution over the northern Indian Ocean, especially in the so-called winter monsoon season (November through March) when the low-level atmospheric flow is northerly/northeasterly, following the temperature gradient from the colder subcontinent to the warmer ocean (Figure 1).
>
> In addition to their direct effects on the climate (i.e. heating or cooling), aerosols are also known to affect clouds by three primary mechanisms: cloud brightening (e.g. Twomey, 1974, the first indirect effect); precipitation suppression (e.g. Albrecht, 1989, the second indirect effect); and radiative (the so-called semi-direct) effects, which may either enhance or diminish cloud cover based on the cloud type and relative position of the aerosol layer (e.g. Koch and Del Genio, 2010). It is important to note that in addition to the often opposing signs of each of these effects, aerosol-cloud interactions have been shown to be highly dependent on the regime (i.e. the typical meteorological conditions, cloud types, location) in which they are found (Stevens and Feingold, 2009). That is, the expression of any or multiple aerosol-cloud effects will be dependent on the conditions under which they are expressed, and thus may vary from one region to another even when considering physically similar clouds. In-situ observations of all types of clouds are thus critical to understanding the full range of indirect effects influencing the Earth's atmosphere.

3. Frankly, the organization of the manuscript is a bit loose. I would encourage the authors to better organize the material if they can.

Following this and the comments of Reviewer #1, we have re-organized and clarified the paper structure. In addition to revising the references to the Appendix throughout the paper as detailed in the Response to Reviewer #1, we have consolidated some of the discussion of Section 3.3 with that in Section 3.1, and have improved transition sentences between each section to better clarify the intended structure of the paper and to highlight the linkages between successive sections.

Minor comments:

1. Abstract Line 3: what is ‘atmosphere structure’ ?

This was meant to refer to atmospheric vertical structure, i.e. how atmospheric properties vary with altitude. The text has been revised to clarify this.

> …including atmospheric *vertical* structure, dominant meteorological conditions, and aerosol concentration…

2. Sentence beginning at Line 25 of Abstract: rewrite this sentence. I had a hard time understanding it.

This part of the abstract has been revised as follows:

> *While high aerosol conditions are observed to disperse with air mass evolution, along with a weakening of the high-temperature anomaly,* the high humidity condition was observed to *strengthen in magnitude as the polluted air mass moves over the ocean toward the site of the CARDEX observations*.

3. 2nd paragraph in Introduction: flux measurement is more relevant to thermodynamics instead of dynamics.

This sentence has been clarified:

> CARDEX follows from these previous studies using UAVs and ground measurements, and for the first time incorporates *measurements of turbulent kinetic energy and latent heat fluxes* for a greater focus on *how thermodynamic factors and atmospheric dynamics* may *influence aerosol effects on clouds*.

4. P29355 Line 8: using linear correlation statistics on log of data is weird. The usual assumption involved in the correlation could be violated in this case and the statistical significance test is meaningless. How about a regular linear correlation? What are the statistics for that?

Thank you for this comment. The observed LWP follows a roughly lognormal distribution, rather than normal; thus taking the log(LWP) values should result in a more normal distribution. The y-axis is illustrated as a logarithmic scale for clarity to the reader. We have also explored other metrics of statistical significance with and without the transform, and added this discussion in the text as follows:

> *However, when the data are filtered to take into account meteorology, there is a positive correlation between LWP and aerosol which is significantly greater than zero (Spearman ρ = 0.48; Pearson R = 0.42, both at the 95% confidence level) for the “dry” (PWV < 40 kg/m^2) cases only (Fig. 6b). Note that for the Pearson correlation analysis we have taken the logarithmic transform of the LWP as these data exhibit a lognormal rather than normal distribution; the non-parametric Spearman coefficient is insensitive to the logarithmic transform.*

5. Last paragraph on P29358: there is no reason to expect this island is representative of the large scale.

We agree, and believe our findings in fact indicate that the island is not representative; however, we find conditions at MCOH to be influenced by variations in large-scale conditions, and we consider it important to compare the observations made here to the larger context in which they were made. This sentence (p. 29358, lines 22-24 in the original text) reads "the pollution level classifications as determined by the conditions over MCOH are not necessarily representative of the region as a whole."

6. Sentence beginning at line 13 of P 29359: rewrite it. I could not understand it.

This sentence has been rewritten for clarity as follows:

> *Although this divergence may act to dilute the polluted air mass, the MODIS AOD shown here suggests that dilution is not the dominant factor distinguishing the two cases. Rather, polluted air is prevented from arriving at MCOH during the low pollution cases due to differences in advection patterns.*

7. Line 26 of the same page: This indicates more of an advection process.

Thank you for this comment. With Section 3.2 as a whole, it was our intention to establish that this coincidently high-aerosol, high-temperature air mass was in fact advected from the Indian subcontinent (and subsequently dispersed through the region), with aerosol heating beginning over the subcontinent (e.g. original draft, p. 29359, lines 21-24). This has been clarified in this section of the text.

> *Rather, polluted air is prevented from arriving at MCOH during the low pollution cases due to differences in advection patterns.* […] Similar to the patterns in the MODIS AOD, the high temperatures in Case H are seen to be concentrated in a region which approaches MCOH from the north, and then slightly dissipates over the four days in question *as the polluted air mass is advected southward*. The remarkable spatial coincidence of temperature with the maximum AOD *over all three days* is strongly suggestive of heating of the air mass due to absorbing aerosol, likely occurring since before the air mass leaves the subcontinent.

8. Line 7 of P 29360: how this correlation is done exactly? It needs to be clearly described. It is very important.

Thank you for the comment. We have clarified these correlations, their analysis, and discussion of their significance.

> For both pollution cases, Fig. 12 shows a region of statistically-significant correlation (95% level, indicated by hatching) between AOD and T. *These correlation coefficients (and those in Fig. 13 [original Figure 14]) were determined by calculating the Pearson correlation R between AOD and T for all days in question (i.e. all H days, or all L days), for each individual 1x1-degree*

latitude/longitude point. Finally, points were only classified as ``significant'' if there were no more than 10% of MODIS retrievals missing. While both Case L and Case H are shown for comparison, it should be noted that due to availability of fewer Case L days being observed (Table 1), the correlations for Case H (left panel) are considered more robust. Analysis for all days indicates a similar pattern to Case H, although weaker in magnitude.

The region of high positive and significant correlation *for Case H* is present over *a broad extent of* the Arabian Sea (the low-level source region to MCOH). The correlation weakens in both magnitude and area of significance between Day H-2 and Day H, which further suggests a dispersion of the polluted air mass *with time, consistent* with the above interpretation of Fig. 11. Case L *shows* a *smaller region of positive correlation concentrated to the north in* the Arabian Sea, *suggesting that while high pollution and temperature are again coincident, the polluted air mass simply is not advected in the direction of MCOH in these cases*.

9. Section 3.2.2: there are many places where correlation is negative. Given the authors' hypothesis there should be no negative correlations anywhere.

With regard to the interpretation of Figure 12, this is addressed in (original manuscript) p. 29360, lines 22-27. Our hypotheses concern vertically-coincident aerosol and meteorological conditions, whereas the region of negative correlation to the east of the subcontinent was likely a result of elevated plumes producing surface cooling. Other regions of negative correlation were not found to be statistically significant, and are likely the result of noise. We have expanded and clarified this discussion.

The smaller region of significant negative correlation to the east of the subcontinent *(particularly evident in Case H)* may be explained by low atmosphere/surface dimming due to *high AOD here indicating* an elevated aerosol plume *rather than the high boundary-layer aerosol responsible for the positive correlation to the northwest*; at higher altitudes, for example at 875 hPa (z~1250m), the AOD and temperature T_{875} show a strong positive correlation in this region. Elevated aerosol plumes are generally seen to approach MCOH from this direction, following the upper-level wind field, consistent with the findings of Höpner et al (2016).

10. Line 7 of P 29363: there are so many meteorological factors to be examined. For example, advection is not considered and it is quite relevant.

We apologize that this was not clear. The intent of this section was not to completely rule out the role of meteorology (including advection), but rather to indicate that with the present analysis, we were not able to determine that this was a dominant factor. For example, the passage immediately preceding this comment presents data which strongly suggests direct advection of high-humidity air is not primarily responsible for the observed patterns. This section has been reworded make this point more clear.

This leaves large-scale factors *(e.g. advection of warm, humid, and polluted air masses)*, local top-of-boundary-layer fluxes, or possible aerosol-induced effects as potential contributing factors to the observed higher relative humidity.

To assess the possible influence of large-scale meteorological conditions on humidity, we *examine* HYSPLIT back trajectories *for any systematic differences in the origin or evolution of the air masses for each case. These* show the upper-level flow *approaching from the northeast* over the subcontinent, consistent with the results shown in Section 2 *(Fig. 5b).* The near-surface flow originates generally from the north/northwest *for both cases; although* low pollution conditions exhibit less extended back trajectories (i.e. lower wind speed above the boundary layer), they come from generally the same direction.

Thus we found no clear meteorological distinction *(in terms of humidity level or origin)* between the two cases which might explain the difference between their boundary-layer conditions. While meteorological conditions may be a potential causal factor of the observed correlation between aerosol and cloud properties (e.g. Mauger and Norris, 2007, 2010), the present observations are not sufficient to definitively establish or discard this hypothesis. Further study of the large-scale context is necessary to more fully explore the potential meteorological influences on the low/high pollution distinction and on the aerosol–humidity relationship.

11. Line 7 of P 29364: increasing T will strongly decrease RH.

We apologize that this was not clear. While we agree that increased T would decrease local RH, this line refers to the possibility that temperature-induced changes in dynamics could potentially change large-scale transport patterns leading to advection of high-humidity air from elsewhere. This section is intended to offer potential explanations for the observed correlations—observations which show increased T to be coincident with increased RH. We have revised this section to make this point more clear.

Another possible mechanism to explain the high humidity relates to the temperature/aerosol relationship. While the *observed* development of the AOD-T relationship *(Figs. 11 and 12)* is consistent with that of aerosol heating of the air mass (Ramanathan et al, 2007), there are two possible interpretations of *how this may relate to the development of high humidity conditions.* First, the humidification of the boundary layer may *be a result of* the meteorological history of the air mass coincident with aerosol conditions (e.g. Mauger and Norris, 2007); second, aerosol conditions may be directly or indirectly increasing the boundary layer humidity. *As shown above, the first interpretation is not supported by the present study, though a more complete analysis is necessary. Regarding the second possibility, aerosol heating may suppress turbulent mixing and stabilize the boundary layer, lowering BL height and inducing higher relative humidity as the polluted plume ages. Alternately, the presence of aerosol heating within the more polluted air mass may be altering the mesoscale circulation to bring more moist air to the region.* Again, further study is needed to establish the plausibility of these potential causal mechanisms, *and to determine* whether meteorological or aerosol mechanisms may be primarily responsible for the observed correlations. *Regardless of their mechanism, these correlations must* be considered in such studies of aerosol–cloud interactions, as secondary changes in atmospheric properties – either directly by aerosol effects or coincident with high-pollution conditions – may alter the effective magnitude of indirect effects.

12. Conclusion: it's overall too general and should be more specific.

Thank you for this comment. We have revised the conclusion as follows:

Here we have presented new results on the characterization of trade cumulus clouds and the dry season cloud climatology in the northern Indian Ocean *using combined ground station observations, vertical atmospheric profiles from UAVs, and large-scale satellite data and meteorological reanalysis*. We describe the general characteristics of the atmosphere in the region and illustrate the existence of two separate climatologies based on the water vapor conditions in the atmospheric column, which result in different populations of clouds forming: *"dry" conditions result in clouds which tend to be constrained by a well-defined boundary layer topped by an inversion, whereas the clouds forming under "wet" conditions exhibit more unconstrained and varied development fed by the availability of more humid upper-layer air.* When the data are *analyzed* according to this climatological separation to *filter out* the large natural variability of high-vapor conditions, we observe a distinct positive correlation between aerosol concentration and cloud liquid water. Highly polluted conditions *(with a high concentration of absorbing aerosol) are found* to be systematically warmer and more humid, *as seen by the ground, aircraft, and large-scale analyses*. From the in-situ aircraft and remote-sensed ground observations, we observe a lower boundary layer height under polluted cases, resulting in a lower cloud base which is responsible for the greater cloud liquid water. *The observed increase in RH was the only potential factor which could account for the magnitude of the observed greater polluted cloud LWP which results from this lower cloud base*. The large-scale analysis indicates that highly polluted air masses exiting the subcontinent are also warmer *initially*, while high-humidity conditions develop along with the air mass as it ages.

While *the strong correlation between aerosol and temperature is likely* attributable to aerosol heating of the air mass (e.g. Ramanathan et al, 2007), with the given observations we are unable to definitively determine a causal mechanism responsible for the observed *correlation between aerosol and humidity. Possible mechanisms which may be producing these correlations include meteorological or aerosol-driven factors, though we were not able to attribute the observed differences to differences in large-scale advection patterns. There remains the possibility that aerosol effects may be driving the observed lagged humidification of the boundary layer, either by influencing the mesoscale circulation or stabilizing the boundary layer locally; this is an intriguing avenue for further study.*

Understanding the consequences of aerosol—cloud interactions in this region requires an understanding of how variations in atmospheric conditions such as temperature and humidity may impact cloud dynamics and water content. Additionally, future research aiming at understanding aerosol--cloud interactions as a whole, and effects of aerosols influencing atmospheric dynamics specifically, should incorporate both local observations of the instantaneous vertical structure and motion of the atmosphere, as well as large-scale observations to understand the air mass history and the potential influence of meteorology on these effects.