

Thoughtful comments and suggestions from all three reviewers are appreciated very much. We thank them because by addressing these comments and suggestions the quality of our manuscript will be improved. In the following we provide point-by-point responses to each reviews.

Reviewer 1:

The manuscript by Yuan et al. examines the impact of a long-lasting non-explosive volcanic eruption on the microphysical and macrophysical properties of trade cumuli over the Pacific Ocean downwind of the largest of the Hawaii islands. The idea is brilliant and the manuscript is well written. The results show a significant impact of aerosols, presumably sulphate aerosols from emissions of sulphur dioxide, on the cloud microphysical properties (cloud droplet size and cloud optical depth) and the cloud amount. I think the manuscript has the potential to be an excellent article and it is for that reason only that I am calling for major revisions. I hope that the suggestions below will help to achieve this.

Correlation does not imply causality. So the usual way of proceeding to suggest there is indeed causality between the presence of volcanic aerosols and the cloud modification is to envisage all other possible reasons one can think of to explain the correlations, and try to rule them out one by one. The authors discuss meteorology and the island wake effect as two possible confounding factors. There are a few more, however small and unlikely they might be, that should be discussed as well.

1) The volcano is probably emitting other chemical species like CO₂ and water vapour H₂O. Could these trace species change the heating rates and moisture availability in the region with increased aerosol loading?

2) In a similar vein, the volcano may be a source of heat, could that change the thermodynamic of the clouds in the volcanic plume?

Response:

Here we address these two possibilities. The air volume coming out of the volcanic vent is on the order of parts per million level (or lower). Even though the temperature difference may be several 100s of degrees the tiny volume is not expected to meaningfully change boundary layer properties at such a large scale. The water vapor emitted by the volcano is on par with that of SO₂ (ppm level) and therefore has negligible effect on boundary layer moisture. The volcanic heat source from radiation and conduction would be even smaller considering the small area it covers compared to the region of interest.

3) The presence of aerosols may contaminate the cloud retrievals. Can the authors make a convincing case that any contamination effect is much smaller than the aerosol effect on clouds?

Response:

Most of the aerosol anomaly is composed of sulfates converted from volcanic SO₂. We used CALIPSO to determine that the aerosols stay within about 2km of the sea surface. The presence of sulfate can have some effect on the optical depth retrieval, if the aerosol loading is very high, or if the clouds are very thin. However, the amount of observed AOD (on the order of 0.2) makes this impact negligible compared with the observed change in COD. This is for cloud optical depth. For possible retrieval artifacts in the cloud droplet effective radius (DER), we consulted with the MODIS cloud product team. The DER retrieval is primarily a function of the radiance at 2.1 μm, and no indication of sulfate affecting 2.1 μm is known. In addition, we filtered the cloud data at pixel level and chose only those pixels with COD >10, confidently cloudy (see text for details) and analyzed these clouds. The same results hold.

We therefore conclude the aerosol effect on the retrieval is negligible in our study. We have added a note in the text to address this concern in the data set section: ‘We note that potential retrieval errors in cloud product due to the presence of aerosols and SO₂ is negligible compared to the signal of aerosol effect on clouds as we will show later.’

4) Could it be that the SO₂ or CO₂ emitted by the volcano contaminates the cloud retrievals? For instance SO₂ has absorption bands at 6.7 and 8.3 μm, can the authors make sure that these wavelengths are not used in the retrievals of cloud droplet size and cloud optical depth?

Response:

Cloud retrieval by MODIS uses combination of visible and shortwave IR channels, all below 4 microns. The standard product uses the 2.1 μm channel for droplet size, which is not affected by either gas.

Back-of-the-envelope calculations should be enough to rule out factors 1) and 2) above. Factor 3) might be more tricky to address, while factor 4) can certainly be looked at.

The authors are right to say that the volcanic emissions would not depend on the meteorology, however the atmospheric transport of the plume does, and it would be good to have a feeling for how much variability in the atmospheric circulation and cloudiness there is from year to year.

Response:

The point on transport by the reviewer is valid. The issue is addressed in our analysis of wake effect. In terms of cloudiness the magnitude of cloud fraction change (from minimum to maximum) can be as large as 0.2 depending on the location, which is one important reason to normalize the cloud fraction in one of our analyses.

There are two ways to diagnose the impact of the aerosols on the cumulus clouds: one can either i) compare the cloud properties inside and outside the plume or ii) look at the anomaly in cloud proper ties in 2008 compared to previous years. The authors are doing both: as I understand it, figures 1, 2, 3, 4, 7, 9A and 9C are about the former, while figures 5, 6 and 9B are about the latter. The authors should be more explicit about this. The impact of the island wake on the cloud proper ties can only be ruled out from method ii), not method i).

Response:

The reviewer is right. We added this discussion in the manuscript at the end of the 3rd paragraph of section 2.

I am not convinced by the normalisation procedure that is described on page 6424. How much variability is there from year to year in the cloud properties averaged over the box of Figure 6? Also the region for normalisation (DL to 155W, 10N to 25N) is somewhat arbitrary and it would be good to know the sensitivity of the results to that choice. By normalising the cloud cover or cloud optical depth, the authors eliminate any large-scale impact of aerosols on clouds (because these average to zero by construction). I suspect the areas of negative COD change on Figure 6 are the results of the normalisation procedure. This is something important to establish and discuss because it would be good to know if the aerosols have an impact on the cloud properties in the “far field”.

Response:

We indeed tested quite a few choices of the background region before settling on the area used in the submitted manuscript. The results are all qualitatively similar. The base region of normalization that we used is not the whole area from DL to 155W, 10N-25N. It is the area by 20-25N, 165W-170W. The reviewer’s comments about the zero-sum nature of the normalization may result from this misunderstanding. We ended up using this region to act as the normalization because it is just outside of the aerosol transport route and thus close enough to the region of interest. With this in mind, it is not surprising that there are negative anomalies outside of the volcano track region. However, they are noisy and do not form physical patterns. Most of them are statistically insignificant.

Is the increase in COD downwind of the volcano compensated or not by a decrease in COD further downwind of the volcano when aerosol levels are down but water availability is still high? This might be difficult to prove or disprove but worth investigating. Also it is difficult to figure out the magnitude of the change in cloud optical depth from Figure 6 (I estimate it to be 20% (from -10% to 10%) of 20 or about 4 units, but is this correct?).

Response:

The COD effect does not stop right at DL. As shown in Figure 1, the aerosol anomaly does not stop until it reaches way beyond DL.

As reported in our manuscript, the increase in COD can be as large as 9 or greater. The values in Figure 6 is after normalization. The reviewer is right that the change is about 20%.

It is important to consider the wake effect from the island as a possible confounding factor, however I am not sure why the authors restrict their analysis to an SST effect only. I also wonder if the combination of the wake and aerosol effects could result in a larger sensitivity to the microphysics than the aerosol effect would do if it was alone (pure speculation on my side).

Response:

The direct effect of blocking by islands on clouds is only effective in the proximity (on

the order of 10-100 km) of islands at the lee side as noted in our manuscript. It is the far-field response of 'wake effect' that is more relevant for our study. This response hinges upon the oceanic Rossby wave introduced SST anomaly, which is the reason we use SST anomaly. However, we did not only look at SST. As noted in the text, we do look at fields such as wind, humidity etc.

Figure 2, and page 6422, line 15ff: this is not clear at all from the plots. I'm not sure where to look on Fig. 2d. Are these the best plots to make this point (which is certainly valid if I judge from the other plots)?

Response:

Figure 2d provides aerosol-cloud mask from CALIPSO. It demonstrates that aerosol and cloud in this case are observed to be right next to each other, not aerosol over clouds, clouds over aerosols, or aerosol separated from clouds, for which we cannot be sure aerosol and cloud are interacting.

Minor comments:

page 6416, line 5: what buffering mechanisms?

Response:

On 6417 or second paragraph of the introduction, several examples are provided and references are provided for these buffering mechanisms. For a comprehensive review on the buffering mechanisms we recommend the Stevens and Feingold paper referred in our study.

page 6416, line 7: or should it be the other way around? Modifications in trade cumulus clouds are associated with aerosols.

Response:

Changed.

References should be cited with the year in brackets only when they form part of a sentence, e.g. on page 6417, line 27: "Stevens and Feingold (2009) provide ...". There are many other occurrences in the text where this is not the case. Can the authors go through the text and change this?

Response:

Done.

page 6419, line 14: "... the diffusive nature of the mixing..."

Response:

Changed.

page 6419, lines 25-26 contradict directly lines 17-18. This needs to be phrased better. See also my major comment above.

Response:

We changed the phrasing in line 17-18 to reflect this point by reviewer.

page 6419, line 5: note that the transport of the aerosol from the volcano is not independent from the meteorology.

Response:

Changed.

page 6423, line 15: I don't know what is meant by 3~8 μm .

Response:

Changed to 3 to 8 μm .

page 6425, line 10: "microphysical"

Response:

Done

page 6426, line 19: make it clear that the recent observations are for clouds affected by ships not by a volcano as in this paper.

Response:

Done.

page 6427, line 29: not clear where the 4 Wm^{-2} comes from. Can the authors explain how they come to this number?

Response:

We arrive at this number using the MODIS based susceptibility calculations as referred in the manuscript. We also checked modeling results where only Twomey effect is considered and the magnitude of change in CCN is larger than observed in our study.

page 6438, line 3: "Eeffect" should be "Effect"

Response:

Changed.

Figure 7 presents a longitudinal mean for each latitude, which assumes some latitudinal consistency. However it is clear from the previous figures that the mean aerosol transport and effect is not exactly east-west, but it is a bit slanted. Could values be averaged along such an axis on Figure 7?

Response:

We note first that slant is minimal in the longitudinal band we consider. Indeed, we attempted to fit a straight line to the data (165w-175w) and the angle between the east-west and the line is smaller than 5 degrees for this longitudinal band. Given the data resolution at 1 degree, the sampling that considers this slant is almost identical to the results we are showing now. Nonetheless, we added a note in the figure caption to reflect the reviewer's concern.

Reviewer 2:

A volcano located on the Hawaii Islands injected massive load of sulfur to the atmosphere for a few months during 2008. This provides an opportunity to analyze the impact of CCN on the cloud life cycles. This paper analyzes this particular natural experiment through the use of various spaceborne observations of atmospheric parameters over the Pacific. They demonstrate that, as hypothesized before, aerosols have a large impact on the aerosol life cycle, with a decrease of the droplet radius, a suppression of precipitation, an increase of cloud fraction. Overall, the aerosol load has

a large impact on the albedo, which a dominant contribution by the indirect effect. This is paper is certainly very interesting and convincing. As stated by the authors, there have been several demonstration of the aerosol impact on cloud microphysics, but this one goes a step further as it demonstrates the impact on the cloud fraction and Earth Albedo. There is no doubt that the paper is a very significant contribution to the study of aerosol-cloud interaction, and it should therefore be published.

I have very few suggestions for correction as the paper is very well written and presented. I must admit I have had difficulties following the methods used to discard the Island wake effect impact on clouds, to finally discard it. I was not convinced by the method that uses EOF for various meteorological fields. It seems clear that the wake Island effect is similar from year to year. Is the SST pattern significantly different in JJA 2008 than for the same months of other years? Note that, even if a lower SST is observed in 2008, one may argue that it is a consequence, rather than the cause, of the cloud cover change. I did not feel that the EOF analysis is necessary to discard the wake effect as the cause for cloud parameter changes downwind of the Island. I therefore suggest that the authors reconsider whether such analysis is really relevant.

Response:

The reviewer's point on the causal link between lower SST and cloud cover is well taken. We had similar speculation. To better understand the wake effect we recommend the reviewer to refer to Xie et al (2001). We summarize the effect in a few sentences in the last paragraph of section 2.

We note that the purpose of EOF analysis is two-fold. First, it is to demonstrate the lack of connection between cloud properties and the wake effect, which was originally included upon comments from oceanographers who feel it is absolutely necessary to do so. Second, as importantly, it is also done to check the proposed link between aerosols and cloud properties with a different method. We show in the manuscript that results fulfill both purposes.

The most important comment concerns some of the Figures (3,4,6). I assume the input data are 1 degree resolution monthly means. There is obviously some spatial interpolation/extrapolation on these data that are not needed, and not desired as one does not know its impact. I strongly suggest the authors keep with the original resolution with no spatial interpolation. Figure 1A and b should have the same spatial area. If possible, all figures should have the same area and projection. Figure 12 shows the relative humidity anomaly as given by a re-analysis. However, it is not clear which data have been used to constrain this analysis. It is neither not clear whether the patterns are significant. I do not think this figures add significantly to the demonstration.

Response:

Maybe it is the contouring algorithm in IDL, but we assure the reviewer that we used the original resolution (1degree) data in our plot. We did not do any interpolation for these plots.

Regarding Figure 1a and b, if the reviewer does not think it is critical we would like to keep the configuration. The reason is that in Figure 1a we want to show a broad view of

the aerosol plume with a large context. The SO₂ plume is not reaching far and thus we chose to use the common area used by other plots.

As for the relative humidity fields, it is from the NASA MERRA reanalysis that assimilates quality assured AIRS infrared radiance to constrain the water vapor and temperature profiles. The MERRA analysis has been shown to be one of the best modern era reanalysis products [Rienecker et al., 2011]. We show the humidity field to demonstrate that the large-scale humidity field is not favored for cloud formation. It is also illustrating that no distinct plume of high moisture is due to the volcano, which confirms the back-of-envelope calculation presented above.

Reviewer 3:

This paper deals nicely with one of the most critical problems of cloud-aerosol analysis, namely the separation between the true aerosol effects and coincidental correlations driven by meteorology.

When showing correlations between aerosols and clouds properties one has to separate associations from cause-and-effect. Theoretically, associations between aerosols to clouds can be driven by meteorological states that favor clouds and aerosols with particular properties with no real interaction-driven-effects.

Here, by analyzing clouds inside and out of the volcanic aerosol plume the authors solved much of the decoupling issue. It is fair to say that away from the island the meteorological conditions are expected to be similar. Moreover, the authors make sure that the wake effect is not included in the results.

I think that this paper should be published and have few minor comments:

1) I think that the EOF analysis reduces the clarity of the paper. Without it the paper is clear and the analysis is straightforward. The EOF uses linear mixture of variables (similar to PCA) that does not necessary make a physical sense. I think that the arguments are quite convincing without it.

Response:

The reviewer's point is well taken. This part of analysis resulted from comments we received from oceanographers. They think it is critical to rule the influence of the 'wake effect'. We agreed and did this analysis, which we think is a healthy addition to our other analysis. Please also see our response above on this point.

2) I miss discussion on invigoration. The authors show here clear invigoration of warm clouds. The polluted clouds are taller and thicker. This was shown in Koren et al, (2005) as the warm part of all convective clouds and since then there is a debate if warm clouds are invigorated or actually shrinks due to enhanced evaporation. I see it as a key result of the paper analysis and think it should be discussed and even highlighted. (see for enhanced evaporation: Small, J. D., P. Y. Chuang, G. Feingold, and H. Jiang (2009), Can aerosol decrease cloud lifetime?, Geophys. Res. Lett., 36, L16806, doi:10.1029/2009GL038888).

Response:

There are other possibilities such as entrainment. The invigoration hypothesis by Koren et al (2005) may offer a viable explanation for the observed results in our manuscript. Also, the Small et al. (2009) paper specifically deals with non-precipitating cumulus clouds, different from our case. However, the reviewer's point is well taken and we expand our text to reflect this point following the presentation of result on increase of cloud top.

3) Why Aerosol Index (AI)? Andreae (2009) showed that AOD is a good measure to CCN and for low levels of AOD fine fraction measurements are significantly less accurate. (Ref: Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions. Atmospheric Chemistry and Physics 9, 543-556 (2009)).

Response:

The reviewer's point is well taken. Indeed, when we carried out the analysis we first used the aerosol optical depth and results are qualitatively similar. There is a segment of the community that prefers aerosol index and because the MODIS product is sensitive to particle size with a healthy Angstrom Exponent, we decided to use it. But again, this choice here doesn't affect our main results.

Reference:

Rienecker M., et al., 2011, MERRA - NASA's Modern-Era Retrospective Analysis for Research and Applications, doi: 10.1175/JCLI-D-11-00015.1, J. of climate, in press.