Review of: Constraining the aerosol influence on cloud liquid water path

By Gryspeerdt et. al.

This paper examines the response of LWP to changes in droplet number concentration (Nd) using long term (3 years), global observations. The results demonstrate a non-monotonic response with an increase in LWP under low Nd conditions followed by a negative trend under high Nd conditions. The trend flips approximately when precipitation is expected to be suppressed, so the authors conclude that the positive trend is due to rain suppression while the negative one is due to increase in entrainment. They also show that the results are sensitive to the RH of the environment and less sensitive to the LTS. Next, the authors use "natural experiments" of volcanic eruptions and ship track to better understand the causality of the trend. The radiative forcing due to the changes in LWP are calculated and shows that the reduction is LWP can, at most, cancel about half of the cooling due to changes in CF and cloud albedo.

In my opinion, the paper presents an innovative and impressive analysis of the data. A special effort was carried out to avoid artefacts and measurements errors. In addition, the topic is of high importance and hence I strongly recommend it for publication in ACP.

I do have suggestions and comments that the authors may want to consider:

<u>**General comments**</u>

- Another possible explanation for the positive correlation between Nd and LWP under low Nd conditions, that was proposed in the past (beside the rain suppuration that is mentioned in this paper) is warm cloud invigoration by aerosols. One way to separate the different causes using observations is to examining the effect of Nd on the cloud top height (CTH): The rain suppuration argument is expected to result in an increase in LWP without a significant change in CTH. However, the invigoration argument is expected to result in an increase in cloud vertical velocity, CTH and LWP at the same time. I think it could be interesting to examine it.

- For identifying the entrainment feedbacks, you use RH. However, what really determine evaporation and entrainment is the different in water vapor content between saturation (the cloud) and the environment. For a give RH this difference increases with increasing temperatures and hence may cause stronger evaporation and entrainment. For a small range of temperatures RH can serve as a good measure for the water vapor differences, but as here the analysis is conducted globally the range of temperatures are large and I think that this effect can interduce some errors and biases and can't be ignored. I think you should at least check its possible effect on the results and mention it in the text.

- I think that the argument that the "natural experiments" are the ground truth is not supported enough. It is true that it makes sense that the variations in Nd in the volcanic plume ,for example, were created by the aerosol concentration increase (and not by an artefact or any other reason) but you do not provide evidences that the meteorological conditions are really fixed. Can't the volcanic plum change other parameters that effect clouds (such as temperature and humidity) beside the aerosol concentration?

- All figures end at Nd~300 cm^{-3}. It doesn't sound very high. There aren't any cases with higher Nd?

- The abstract could be written in a clearer way (see specific comments below).

- The paper misses a few relevant previous papers.

Specific comments

- The first two sentences in the abstract are a bit confusing.

  "The **impact of aerosols on cloud properties** is one of the largest uncertainties in the anthropogenic radiative forcing of the climate. In recent years, significant progress has been made in constraining this forcing using observations, but uncertainty still remains, *particularly* in the **adjustments of cloud properties to aerosol perturbations**.".

  Both part discus the aerosol effect on cloud properties so the use of "*particularly"* here is not clear to me.

- P1, L11: "…suggesting that aerosol induced LWP reductions could offset a significant fraction of the radiative forcing from aerosol-cloud interactions (RFaci). "

  This sentence wasn't clear to me at the first time I read it. At this point you didn't define yet RFaci as an "instantaneous radiative forcing" so it is not clear why you do not consider the aerosol effect on LWP in "aerosol-cloud interactions"?

- The last sentence of the abstract is not clear to a reader that didn't read the paper yet.

- P1, L21: the change in CF or LWP can be caused not only by delay in precipitation but also by other reasons such as increase in evaporation and entrainment and warm cloud invigoration by aerosol.

- P2, L10: there were also previous studies that found a non-monotonic response of cloud properties (including LWP) to changes in aerosol concentration. The optimal aerosol concentration was shown to depends on the meteorological conditions.

- P2 L12: beside the meteorological conditions, the singe of the effect of aerosol on LWP may be determine by the range of changes in aerosol concentration that is examine in each case.

- P3 L2: see if you can write this part in a clearer way.

- P3: I thought that E1 b and c are more relevant in marine Sc and cumulus clouds, respectively. Is it correct? If yes, it is probably worth mentioning.

- P3: another pathway by which an increase in Nd may affect the LWP is by warm cloud invigoration. The increase in total droplet surface area under polluted conditions would lead to faster condensation (in the super saturated parts of the cloud,) more latent heat release, increase in cloud buoyancy and hence increase in LWP. In addition, under polluted conditions the smaller droplets would be pushed higher in the atmosphere (even under the same air vertical velocity). This could also lead to an increase in LWP with aerosol loading, as the clouds may reach higher in the atmosphere.

- Fig. 2: can you add maps presenting ml and mh for the different regions? I think it could be very interesting to see if the slopes (or even more interestingly the Nd that mark the change in trend) change in the different regions/meteorological conditions and whether you can identify that regions that support the development of more develop clouds are more effected by the increase in Nd than other regions.

- Figures: it will be interesting to add to the figures the Nd that differ between the two different slops. It is interesting to see if it changes for the different cases (presented in the different figures).

- P8. L12: it is consistent with **at list** two aerosol effects in liquid clouds. Under extremely clean conditions the clouds could be “aerosol limited” and so cloud invigoration was suggested to take place.

- P8. L32: another (simpler) way to overcome this difficulty would be to plot the Nd marking the change in trend or the slopes of the different trends on a map.

- Fig. 3: Do you think that it is possible that you don’t see any significant trends for cluster 2 because it mixes many different regions with different meteorological conditions (i.e. tropics and extra-tropics)?

- P12 last line: is it possible that the volcano adds water vapor to the atmosphere as well as aerosols and hence you see a cancelation of effects? In other words, are you sure that all other meteorological conditions are on average the same between the years?

- P13 L13: I am not sure that the statement that the volcanic case has a: “reduced impact of other processes (E2-4)” is supported well enough. For example, are the meteorological condition really fixed? I can imagine that a volcanic plum has other effects rather than increasing the aerosol loading (such as changes in the temperature or humidity vertical profile).

- P13 L29: are they distinguishable from 0?

- P14 L2-4: again, they are not reduced completely. I think you make this argument too strong.

- P16 L14: it doesn’t have to be because of cloud top entrainment. It could also be due to increase in lateral entrainment.