Review of "Impact of Holuhraun volcano aerosols on clouds in cloud-system resolving simulations" by Haghighatnasab et al., submitted to Atmospheric Chemistry and Physics (ACP)

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This report contains overall, major and minor comments from this reviewer to the manuscript.

A summary of the manuscript and overall assessment:

Recommendation: Major revision

This study performed sensitivity experiments for a region around the North Atlantic to investigate the effects of volcanic smoke aerosols as cloud condensation nuclei (CCN) associated to the eruption in Holuhraun on the cloud properties in the volcanic smoke trails. A scientific goal is to investigate how liquid water path (LWP) and cloud fraction change in response to increase in CCN and subsequently cloud droplet number (Nd) in the volcanic plumes. Regional cloud-resolving simulations with approximately 2.5 km grid spacings were conducted for a week when emission of SO2 from the eruption was clearly identified in satellite observations. The simulation results were compared to satellite observations for cloud to check how the observed difference in cloud properties between in and outside the volcanic plumes was reproduced in the simulations. The simulation considering effects of volcanic smokes replicated the observed increase in Nd in the plumes. However, the same simulation overpredicted increase in significantly LWP and slightly in cloud fraction.

I think the current investigation and discussion on the relationship between LWP and Nd or Na (aerosol or CCN number concentration) is insufficient. As described in the current manuscript and in reports from model intercomparison projects (e.g., Quaas et al. 2009), conventional global aerosol transport models tend to overpredict increase in LWP in response to increase in Na or Nd, compared to global-scale satellite observations. However, several recent modeling studies, particularly using high-resolution (cloud- or large-eddy-resolving-scale) models at a regional or global scale, reported little change or even decrease in LWP in the response, according to condition. Their results may be more consistent with the finding in Malavelle et al. (2017), which is the case in this study. First, the manuscript should include careful literature reviews about the advances in recent modeling studies on the sensitivity of LWP to variation in aerosol, CCN, or cloud drop number concentration. Then, more discussion and investigation are needed to examine why the results of the cloud-resolving model simulation in this study contradict findings in some of those recent modeling studies as well as the observation results for the volcanic smoke case. I think at least this effort has to be done toward being acceptable. I have several other major comments listed in the following section. The authors are encouraged to revise the manuscript to improve the quality and readability.

Major comments:

1. LWP-Nd

As an example of limited-area large-eddy simulation for aerosol-cloud interaction, Seifert et al. (2015) conducted an extensive series of sensitivity simulations. They reported a negative lifetime effect (unchanged LWP and decrease in cloud cover with increasing Nd) in addition to positive one which has been seen in other previous LES studies, depending on the meteorological condition and the stage of cloud life cycle. Similar dependency of the sensitivity on meteorological condition and cloud regime was found in other LES studies (e.g., Lebo and Feingold 2014). On the other hand, Sato et al. (2018) conducted one-year global cloud-resolving simulation to examine the sensitivity. They successfully reproduced negative λc (the definition can be found in the paper) seen in satellite observations, mostly over regions where cumulus was dominant. They suggested that evaporation process of cloud droplets around cloud top was important to resulting in negative values. More details of the discussion can be found in the paper. As I wrote in the overall comment, since some of other modeling studies could reproduce near-zero and even negative sensitivity, the authors should make efforts to examine and explain why the current simulation could not do it in discussion together with findings in previous studies not limited to those shown above. I understand models have various uncertainty and hence often cannot reproduce observations. But the manuscript should show some advances toward the next step.

Seifert, A., Heus, T., Pincus, R., & Stevens, B. (2015). Large-eddy simulation of the transient and near-equilibrium behavior of precipitating shallow convection. Journal of Advances in Modeling Earth Systems, 7(4), 1918-1937.

Lebo, Z. J., & Feingold, G. (2014). On the relationship between responses in cloud water and precipitation to changes in aerosol. Atmospheric Chemistry and Physics, 14(21), 11817-11831.

Sato, Y., Goto, D., Michibata, T., Suzuki, K., Takemura, T., Tomita, H., & Nakajima, T. (2018). Aerosol effects on cloud water amounts were successfully simulated by a global cloud-system resolving model. Nature Communications, 9(1), 1-7.

2. Meteorological and cloud information of the target case

The manuscript should show what meteorological condition and what types of cloud were dominant in the period and the domain for the simulations. These information is quite important in the discussion because previous studies, e.g., in comment #1, showed some dependency of the aerosol-cloud interaction on those factors. Some MODIS true-color images may help it. And another question, is only warm-topped cloud with cloud top temperature over 273.15 K analyzed and is the other cold-topped cloud excluded?

3. Vertical distribution of the volcanic aerosol plume

The OMPS satellite retrieval products were used to identify the column total SO2, and then sulfate aerosol mass mixing ratio was calculated based on the difference in column total SO2 between in and outside the volcanic (around Ln. 143). But I think the vertical profiles of SO2 and sulfate aerosol concentrations might differ between, because they might be contaminated in limited vertical layers into which smoke was injected. How did the authors consider the vertical injection or vertical distribution of the volcanic aerosol plume? Or, maybe I am confused, does the model not need the information of vertical distribution of aerosol but just use column-integrated value to calculate activated CCN concentration at each vertical level?

4. Definitions of LWP in MODIS product and the simulation

It is clearly written that Nd in the simulations were calculated using a satellite simulator through same pathway as for the MODIS products. But what about LWP? The definition of LWP has large uncertainty between the satellite products and the model simulation even using a simulator because bulk cloud microphysics has a category gap between cloud water and rain. This is problem in the radiative transfer calculation in simulator to determine LWP that is consistent with that in satellite products. This problem may affect the calculation of other variables such as Nd also.

5. Discussion on cloud fraction

I think 2.5 km model grid spacing may be still coarse for comparative discussion of cloud fraction over ocean with the Level-2 MODIS-Aqua cloud product (swath 1km). The model simulation might miss parts of scattered shallow cumulus over ocean and overemphasize extent of deeper cloud. This might contribute the overprediction of positive cloud lifetime effects on cloud fraction in the plumes in the simulation too. The shallow convection parameterization of

Tiedtke (1989) has no effects on the calculation of the cloud fraction, correct?

6. CERES 20 km resolution

Is the 20 km resolution of the CERES products enough to distinguish in and outside the smoke plume? The spatial scales of the smoke trails are unclear to me. And what algorithm was used for remapping the model results from the native model grid structure to those with 20 km grid spacing? The selection of the algorithm may strongly affect the results because it was from fine to very coarse grid structures.

Minor comments:

Ln. 39: “cloud” => “could”

Ln. 84: Same question as in major comment #6, what algorithm was used for remapping?

Around Ln. 110: Can you summarize the variables in the look-up table and the value ranges into a table?

Table 1: Could you add comparison of τc and re into Table 1 too?

Figs. 2 and 3: Please add lines of latitude and longitude to the maps.

Ln. 253-255: These sentences are a bit awkward. Please rephase and improve the readability.

Ln. 256-257: The sentence is confusing. The vertical axis of the plots in Fig. 6 is at a log-scale. The frequency of high RWP over 200 gm-2 in the volcano simulation is quite or neglectably small, and the difference in mean RWP in the plumes is due to the difference in the frequency of lower RWP values.