Response to referee comments #1

In this study, the authors conducted numerical simulations targeting the eruption of Holuharaun volcano by the NWP mode of ICON, and investigated the impacts of aerosol emitted from the volcano on N_d, LWP, cloud fraction, and cloud albedo. Through their analyses the authors clarified that the impact of the aerosols on the LWP and cloud fraction was mainly explained by the difference of the meteorological condition between inside and outside plume, although the increase of N_d was clearly seen as the impact of the aerosols. In my understanding, the response of the LWP and cloud fraction to the aerosol variation are featured topics in the scientific community and the results of this study are interesting. So, I encourage the authors to conduct this study. Most part of the manuscript is well written, but there are several issues to be addressed. Based on the descriptions outlined above, my decision is "major revision", and I encourage the authors to revise the manuscript.

We thank the reviewer for their assessment of our manuscript. The review helped to improve the manuscript significantly.

Major comments

1. The authors discussed the effects of the emitted aerosol on the radiative forcing based on the results of cloud albedo. However, the goal of this study is to understand how LWP and cloud fraction respond to the aerosol variation, as the authors indicate in the body of the manuscript. So, the discussion about the radiative forcing and cloud albedo will make readers to confuse the main topic of this study. Of course, I understand that the radiative forcing and cloud albedo are really important for the climate study, but focusing on the LWP and cloud fraction makes the main message of this study clear. The discussion about the cloud albedo and radiative forcing in discussion section, which can be created in the revised manuscript is better, or the discussion of them in supplementary information is another option.

We appreciate the reviewer's suggestion. A new section "Implications for the radiative impact" is now added in the revised manuscript before the "conclusion" section and all the information regarding radiative forcing is moved to this section to tone down this aspect of the analysis, in light of the reviewer's concern.

2. In the section 3, the authors suggest that the decrease of the light rain and increase of the heavy rain in volcano simulation based on Fig. 6a. In addition, the authors suggest that the enhancement of the RWP is decreased by 15 % and no difference in RWP over outside of plume based on Table 1. I agree these suggestions, but it is not clear about why the decrease of the light *rain and the increase of the heavy rain can result in the decrease of the enhancement of RWP and as consequence, increase of LWP. The manuscript is not so long at current version, and therefore, the author can add detailed descriptions about the reason. Such descriptions will help readers to understand the authors' suggestions more clearly.*

We appreciate the reviewer's suggestion. We conclude in the manuscript that the decrease of light rain and the increase of heavy rain results in the enhancement of LWP inside of the volcano plume. The reason for this conclusion is that when light rain is depressed, cloud droplets must grow larger in order to reach the size that they can start to precipitate which leads to a shift in the LWP distribution through the higher values, and if cloud droplets grow larger, they produce heavier precipitation. Therefore there is an enhancement in the probability of heavy rain compared to no-volcano simulation inside of volcano plume, even if the average RWP (Table 1) does not change much. These informations are added in the revised manuscript.

3. *First of all, I appreciate the authors' effort to conduct the numerical simulation by using ICON-NWP and to develop the method for implementing aerosol effects. The effects of aerosols on the cloud microphysical properties can be calculated by the coupling method used in this study. However, the method cannot implement feedback of cloud process to aerosol field. If the authors conducted the ICON-NWP coupled with aerosol transport model or chemical transport model online, the results about the LWP adjustment and cloud fraction adjustment would be changed. I understand that the simulation by ICON-NWP coupled with aerosol transport model or chemical transport model online is one of the future study of authors' group, however, discussions about the limitation about the method used in this study and discussions about the difference of the results from the online coupled model and this study should be added.*

We appreciate the reviewer's suggestion in pointing out the limitation and potential difference of the method used in this study compared to using an interactive aerosol model. The approach used in this study to implement aerosol effects in the ICON-NWP does not consider transport and transformation of aerosols in the model domain and indeed an atmospheric model with interactive aerosol physics would be able to represent the evolution of the aerosol field in more detail. However, an important buffering mechanism, namely the consumption of CCN on activation, is considered in the method used in this study. So CCN are lost when they are activated but will eventually relax back to their initial profile . This was indeed not made clear in the first version and is now clarified in the revised manuscript. It should also be noted that there is an important advantage of our method compared to a fully interactive aerosol scheme, which is that the location of the plume is derived from observations and so is at the same region as in satellite retrievals. This allowed us to analyze inside and outside of the plume in simulations and satellite products with confidence.

Specific Comments:

1. *Figure 1: What does the color of Fig. 1 mean? Elevation from sea surface? The caption about the color and the color bar should be added.*

This Figure indicates the domain of the simulation with orography color-coded. Blue color indicates the ocean surface and other colors indicate the elevation of land above sea surface. A color bar is added, and the caption is revised to include information about the color coding.

2. *Line 59: The information of the layer thickness is required.*

We use 75 vertical levels spanning from the surface to 30 km altitude with a vertical resolution of 20 m at the lowest model level that gradually gets coarser towards the model top; the coarsest vertical resolution is 400 m. This information is added to the manuscript.

3. *Line 65-66: What physical variables were used for the initial and boundary condition? Such information is important for other scientists to trace the simulations by other models.*

The physical variables that are used are temperature, horizontal wind components, surface pressure, surface geopotential, geopotential, specific humidity, cloud liquid water content, cloud ice content, rainwater content, snow water content, snow temperature, water content of snow, density of snow, snow albedo, skin temperature, sea surface temperature, soil temperature level 1,2,3,4 (level 1 to 4 are located at 3.5cm, 17.5cm, 49.5cm and 64cm respectively), sea-ice cover, water content of interception storage, surface roughness, Land/sea mask, soil moisture index layer 1,2,3,4. These information are added in the manuscript in a more summarized form.

4. *Line 68: In this part, the authors indicate that analyses period is from 1 to 7 September, 2014. Is the period corresponding to the period of the calculation? If so, did the author check the effects of spin-up was sufficiently small? In such regional scale simulation, we do not analyze first several hours to avoid the artificial wave generated during initial shock.*

The period of the analysis is from 1 to 7 September 2014. In order to consider the spin-up effect the first 9 hours of simulation were not considered in the analysis.

5. *Line 79: In this part, the authors indicate that the input variables of COSP are temperature, pressure, cloud fraction, and cloud water content. For simulating MODIS's signal, the information of size distribution function is required. The information about the size distribution of hydrometeor should be added in the method.*

Apart from the variables mentioned in the submitted version of manuscript as an example of COSP's inputs, in the MODIS simulator setup which has been used in this study, the effective radius of cloud droplets and ice crystals are used as an input to the simulator. The approach for calculating the effective radius of cloud liquid droplet and ice crystal is to use the actual parameters from the hydrometeor size distribution from the two-moment microphysics scheme in the model. This approach had already been implemented and tested in ICON-NWP by Kretzschmar et al. (2020) and Costa-Surros (2020) for improving cloud radiative properties. Equation B8 in Kretzschmar et al. (2020) indicates how effective radius of liquid cloud droplets and ice crystals are calculated from their respective size distributions. This additional information about the way of using size distribution function as input to the COSP MODIS simulator indeed was lacking and is now added in the revised manuscript.

6. Section 2.2.: In this section, the authors describe the method for implementing aerosol effects on the ICON-NWP, and the authors shows distribution of column-mean CCN as shown in Fig. 3. I think the distribution of CCN is reasonable. However, there are no information about the vertical distribution of CCN. Based on the body of the manuscript, the data for SO_2 was originated from OMPS product. I think that the product is vertical column amount of SO_2. Which layer did the authors add the SO_2? Based on my experiences, the layer that aerosols are input is really sensitive to the simulated impact of aerosol on cloud microphysical properties. In addition, did the authors assume SO_2 gas is as sulfate aerosol particle?

As well as the SO_2, water vapor is also emitted by the eruption, and the emitted water vapor can affect the meteorological field and cloud properties. Did the author only consider the emission of SO_2?

The activated CCN concentration was computed from aerosol components (including sulfate aerosol) mass mixing ratio in a box model, and this box model requires the sulfate mass mixing ratio at each vertical level. In order to scale the sulfate aerosol from CAMS reanalyses inside the volcanic plume, we scaled the sulfate aerosol mass in each vertical level based on the SO_2 retrieved OMPS for the lower troposphere. It is thus assumed that in each level, sulfate aerosols in the troposphere are enhanced in the plume by the same ratio of enhancement as SO_2 in the lower troposphere (up to 3 km) in the OMPS satellite retrievals. We now clarified in the revised manuscript that it is not total-column that is used to scale the cloud-active aerosol, but the one in the boundary layer beneath the clouds.

7. Line 193-195: In this part, the authors suggest that the decrease of the probability of clouds with low LWP and the increase of the probability of clouds with high LWP. I agree the suggestion, but I cannot agree "thicker clouds (with high LWP)" and "shallower clouds (with low LWP)" from the results shown the manuscript. Does the word "thick" and "shallow" mean "geometrically thick" and "geometrically thin"? If so, the author should show the cloud geometrical thickness. If the authors just want to discuss the probability of clouds with low and high LWP, the words "thick" and "shallow" are not necessary.

As well as the terminology, if the thick clouds increase in the simulation with volcano emission, such difference can result in the change of the circulation. Did the author check the cloud distribution (geographical distribution, vertical structure of clouds and so on)?

The discussion about why the decrease of the probability of clouds with low LWP and the increase of the probability of clouds with high LWP occurred is also useful for readers.

In light of this reviewer's concern, the phrases “thick” and “shallow” were removed from the manuscript because we indeed did not analyze the geometrical thickness of clouds. Rather, by the terms thick and shallow clouds, we meant clouds with higher LWP and lower LWP, respectively. We revised the manuscript to have a discussion and conclusion part and we added an explanation about the reason for the shift in LWP distribution toward the higher LWP.

8. In addition, grid line in each frequency distribution (Figs. 4, 5, 6, and 7) will be helpful for readers to distinguish the shift of peak and find the decrease of low LWP and the increase of high LWP.

The grid lines was added to the figures.

9. Line 218-238: As I mentioned in the general comment, this part is not the main topic of this study. So, I recommend the authors to create new section “discussion” after conclusion or just before the conclusion and move this part to the new section. Again, I understand that the radiative effect is important, but this is not the main topic of this study. Alternatively, I ask the authors to add more descriptions about the enhancement of RWP and precipitation as I mentioned in the general comment.

The section ‘Implications for the radiative impact’ is created before the conclusion part and the results about cloud albedo were moved to this section. In addition, in the conclusion, more discussion about the reason for the enhancement of RWP and precipitation as mentioned in the response to the general comment was added.

Minor Comment:

-Line 47: “(Toll et al., 2017)” should be “Toll et al. (2017)”

Changed.

-Table 1: The unit of LWP and RWP is g m^{-2}

Changed.

-Line 157: I think that “(factual and counterfactual)” is not necessary.

Changed.

-Figure 4. How did the author define the "Inside" and "Outside" plume?

The method to define the inside and outside of the plume as is those marine pixels that correspond to the concentration of SO2 more than 1 Dobson in figure 2 are assumed to be located inside the volcano plume and the rest of the pixels are considered as outside of the volcano plume.

-Line 275: The URL of CAMS reanalysis data is not correct. The URL has been moved to "https://confluence.ecmwf.int/display/COPSRV/Copernicus+Atmosphere+Monitoring+Service+-+CAMS"

Corrected.

Reference

Kretzschmar, et al. "Employing airborne radiation and cloud microphysics observations to improve cloud representation in ICON at kilometer-scale resolution in the Arctic." *Atmospheric Chemistry and Physics* 20.21 (2020): 13145-13165.

Costa-Surós, M., Sourdeval, O., Acquistapace, C., Baars, H., Carbajal Henken, C., Genz, C., Hesemann, J., Jimenez, C., König, M., Kretzschmar, J., Madenach, N., Meyer, C. I., Schrödner, R., Seifert, P., Senf, F., Brueck, M., Cioni, G., Frederik Engels, J., Fieg, K., Gorges,K., Heinze, R., Kumar Siligam, P., Burkhardt, U., Crewell, S., Hoose, C., Seifert, A., Tegen, I., and Quaas, J.: Detection and attribution of aerosol-cloud interactions in large-domain large-eddy simulations with the ICOsahedral Non-hydrostatic model, Atmospheric Chemistry and Physics, 20, 5657–5678, https://doi.org/10.5194/acp-20-5657-2020, 2020.