Response to referee comments #2

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.

We thank the reviewer for their assessment of our manuscript. The review helped to improve the manuscript significantly. In particular, the suggestions led to a clearer formulation of the main messages of the study.

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.

L143.

We appreciate the reviewer's suggestion; indeed it is important to provide a balanced and detailed report about the state of the art. It is of course difficult to be exhaustive, but we made an effort to discuss the key results of large-eddy simulations or kilometer-resolution simulations of the effect of enhanced aerosol on LWP for various cloud regimes in the revised manuscript. Among these, the papers the reviewer suggested are now discussed. We made an effort to include a number of further relevant studies, such as the ones by Ackerman et al. (2004) and Bretherton et al. (2007) that also described the relevant processes in stratocumulus, where reduced sedimentation of smaller droplets may lead to a decrease of the cloud-top entrainment rate and thus a decrease of LWP. The reasons for discrepancies in different cloud-resolving modeling studies are now also discussed in the revised manuscript.

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?

The reviewer is right that this is key information and indeed the broad idea of synoptically driven clouds is insufficient. We now include a new Figure 1 which shows the MODIS visible images from 1 to 7 September 2014 over the simulation domain. A synoptic frontal system is dominant over the North Atlantic ocean. The cloud regime consists of both ice and liquid phase clouds and remains approximately the similar regime during the simulation period. Malavelle et al. (2017) analyzed the cloud regime derived from satellite measurement and showed that the region surrounding the Holuhraun volcano contains the whole spectrum of liquid-dominated cloud regimes. In our analysis, we exclude clouds over land both in satellite observation and simulations. The method to select liquid phase clouds in the MODIS product was to use cloud phase optical properties. This product contains a flag dedicated to liquid phase clouds, therefore, all the grid points with liquid phase clouds were chosen for our analysis. For simulations, the COSP simulator computes the microphysical properties (effective radius, liquid water path, and cloud optical depth) for the liquid and ice phase clouds separately and we used the simulator output dedicated to the liquid phase.

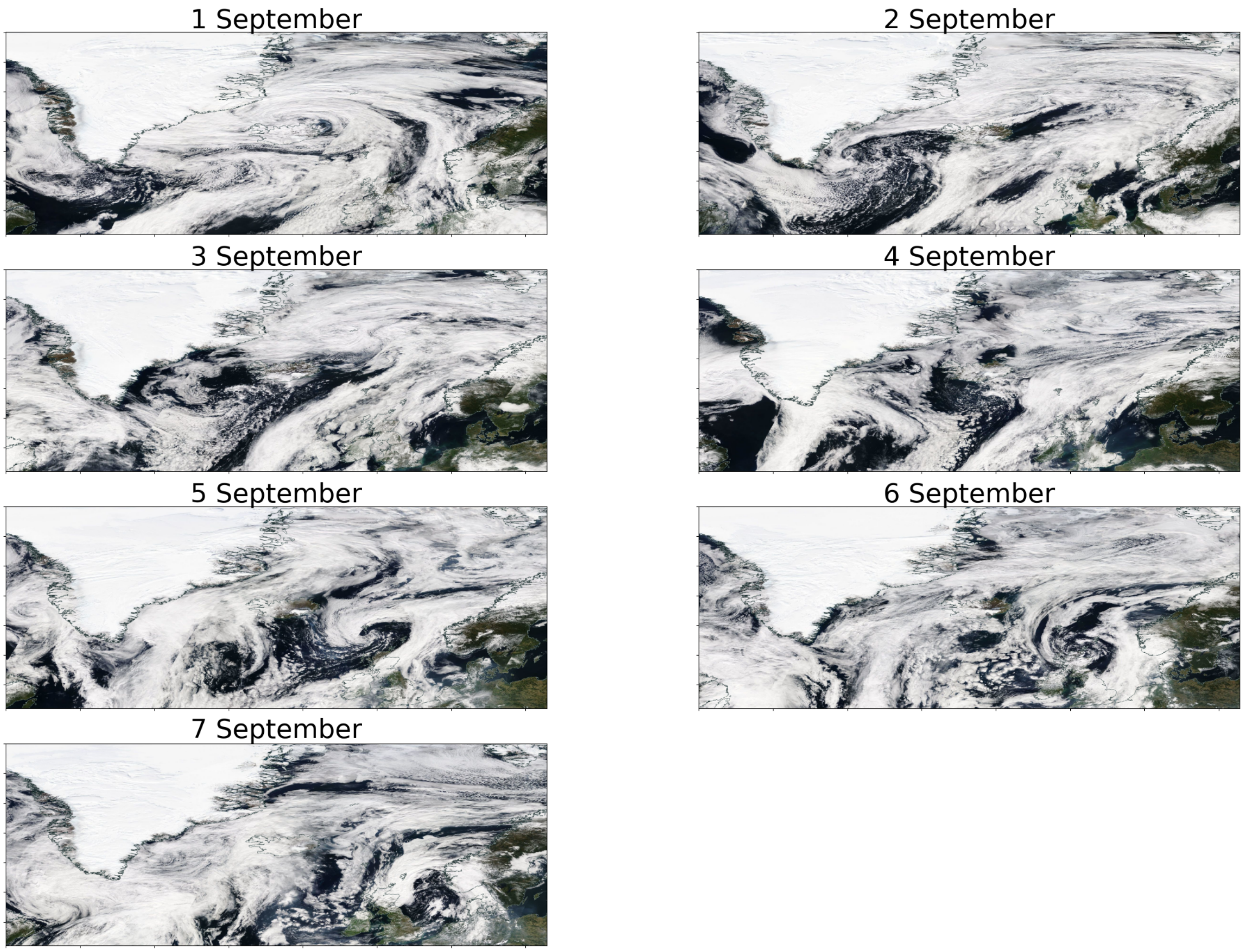

Figure 1. Visible image of MODIS-AQUA from 1 to 7 September 2014.

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?

It seems that we did not explain very clearly what we did to define the plume and in light of the reviewer's comment we now clarified and enhanced the corresponding section in the revised manuscript. The concentration of potentially activated CCN was computed from aerosol components (including sulfate aerosol) mass mixing ratio using a box model, and this box model requires the mass mixing ratio at each level. The potentially activated CCN profile, produced to be used as input in ICON is vertically resolved. In order to define the volcanic plume on the basis of the distribution of sulfate aerosol from the CAMS reanalysis, we scaled each vertical level of sulfate aerosol in CAMS based on the lower troposphere column amount of SO_2 in OMPS data. So we assume that in each level sulfate aerosols in the troposphere are enhanced in the plume as the same ratio of column amount of SO_2 in the lower troposphere (up to 3 km) in OMPS data. In consequence, the vertical distribution within the plume follows the one generated by the reanalysis without the plume, but the scaling makes use of the vertical information from the satellite retrievals such that only the boundary-layer enhancement is used, i.e. the aerosol that is relevant for the formation of the liquid-water clouds investigated in our study.

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.

We appreciate the reviewer's concern about the category gap that exists between cloud water and rain in bulk microphysics schemes. We add this point that the category gap between cloud droplets and rain in size distribution because of using a bulk microphysics scheme in the simulation could cause some uncertainty in LWP between simulations and MODIS and this issue could also affect the computation of cloud droplet number concentration. However, the issue is in our opinion less important when analyzing the differences between the clouds within and outside the volcanic plume.

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 the 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?

In the MODIS-Aqua level 2 cloud fraction product is available at 5x5 km pixels . While of course finer resolution can improve the results in comparing the resolution of simulation to MODIS, it can be seen that this resolution is sufficient for the purpose of discussing the results in this study. We used a grid-scale cloud cover scheme in our simulation. This scheme works in a way that if the sum of specific cloud water content and specific cloud ice content is larger than a threshold (1e-8 kg/kg), cloud fraction is set to 1 and else set to 0. The Tiedtke (1989) shallow convection implicitly contributes to the computation of specific cloud water and ice content but doesn't have an explicit effect in the computation of cloud fraction. These explanations are now added to the revised manuscript.

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.

Indeed it is evident that more detail is needed in this regard. Grid-points with SO_2 concentrations in the lower troposphere exceeding 1DU are considered to constitute the plume, and SO_2 concentration was obtained from OMPS satellite retrievals which are in 50km×50km footprint data in Level 2. We remapped the Level 2 data into the 50km (0.5° degree) resolution, and due to the fact that CERES products are on 20km resolution, it has sufficient resolution to identify the plume. We used the CDO (climate data operators) tools to remap the original unstructured ICON grid to a regular latitude-longitude grid. The same algorithm was used to remap the model grid with 2.5km resolution to 20km resolution to be compared with the CERES product. This CDO module contains operators for an inverse distance weighted average remapping of the four nearest neighbor values of fields between grids in spherical coordinates. Figure 2 shows a comparison between net-shortwave-flux at the top of atmosphere from the simulation which is remapped by the mentioned operator to both 2.5km and 20km resolution. The overall pattern and values are very similar even though there are some small differences due to the remapping from fine to coarse resolution. The relevant additional information is added to the revised manuscript.

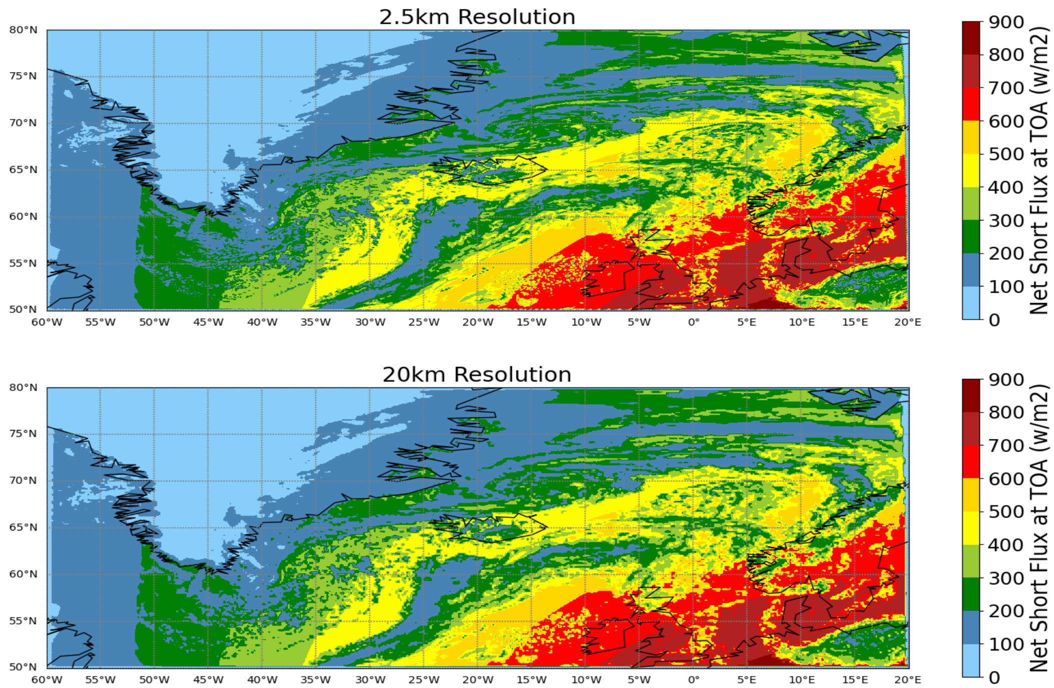

Figure 2 . A comparison between the net shortwave flux of different resolution of 2.5 km and 20km.

Minor comments:

Ln. 39: "cloud" => "could"

Corrected.

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

The MODIS Level 2 data at 1km resolution was remapped to latitude-longitude grids with 2.5 km resolution. The MODIS swaths were used and for each specific point, the mean value of different amounts of a specific variable in each swath were computed.

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

The look-up table consists of potentially activated CCN number concentrations for 10 specific vertical velocities and height for each hybrid-sigma-pressure level (60 levels) and 3 hour interval. This lookup table was calculated offline for 1 to 7 September 2014 (the period of simulation). Here, to show the range of values, we choose 2 September and compute a daily mean. The model level corresponding to approximately (850 hPa) was chosen. The table below summarizes each specific vertical velocity that has been used in the box model for computations of potentially activated CCN concentration. The value range is shown as the mean value for the whole domain and the first and the third quartile of grid point values.

2 September 2014			
Variables	First Quartile(cm^{-3})	Mean value(cm^{-3})	Third Quartile(cm^{-3})
CCN_act (w = 0.01 m/s)	5	7	8
CCN_act (w = 0.0278 m/s)	20	26	32
CCN_act (w = 0.0774 m/s)	54	73	87
CCN_act (w = 0.215 m/s)	117	166	204
CCN_act (w = 0.599 m/s)	230	334	414
CCN_act (w = 1.67 m/s)	406	605	753
CCN_act (w = 4.64 m/s)	639	994	1235
CCN_act (w = 12.9 m/s)	918	1492	1842
CCN_act (w = 35.9 m/s)	1219	2070	2534
CCN_act (w = 100 m/s)	1528	2691	3271

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

The variables effective radius and cloud optical thickness are added to Table 1. It can be seen that effective radius decreased inside the plume by 7% compared to outside the plume. In the MODIS data, the effective radius decreased by 8% inside the plume compared to outside the plume. Cloud optical thickness increased by 33% inside the plume compared to outside the plume in MODIS. This enhancement is about 24% in the “volcano” simulation while in the “no-volcano” simulation, cloud optical thickness decreased by 3% inside the plume compared to outside the plume. This discussion is now added to the manuscript.

Figs.2 and 3: please add lines of latitude and longitude

The grid lines were added to the figures

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

The sentence is rephrased.

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.

The sentence is rephrased.

References:

Ackerman, A. S., Kirkpatrick, M. P., Stevens, D. E., and Toon, O. B.: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, Nature, 432, 1014–1017, https://doi.org/10.1038/nature03174, 2004.

Bretherton, C. S., Blossey, P. N., and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and subtropical stratocumulus albedo, Geophys. Res. Lett., 34, L03813, https://doi.org/10.1029/2006GL027648, 2007.