Responses to referee 2

Review

This paper reports on aerosol-cloud interactions observed from satellite measurements and simulated with a high-resolution regional model (Unified Model) using the Holuhraun eruption as an opportunistic experiment. The authors find strong aerosol and "total" effects on cloud droplet number concentrations and effective radius due to the volcanic eruption with minimal and statistically insignificant responses to cloud fraction and liquid water path. The high-resolution of the UM allowed for sufficient realization of the cloud responses to the eruption as compared to more coarse model comparisons that were not as successful. However, uncertainties in the simulation of background aerosols in the UM may complicate these results. This is a fairly straightforward and clearly written paper that carefully walks through the differences in observed and simulated effects and the limitations of each data source in evaluating these differences. I believe this paper is suitable for this journal as it uses innovative approaches for quantifying aerosol-cloud interactions from an opportunistic experiment and provides recommendations for improving future efforts. The authors should consider the following minor comments, questions, and recommendations to improve the work before it should be published.

• Are the authors able to add titles to the sets of columns in Figure 1? Left: Simulated, Right: Satellite? This would allow for a more accessible direct comparison between the plots.

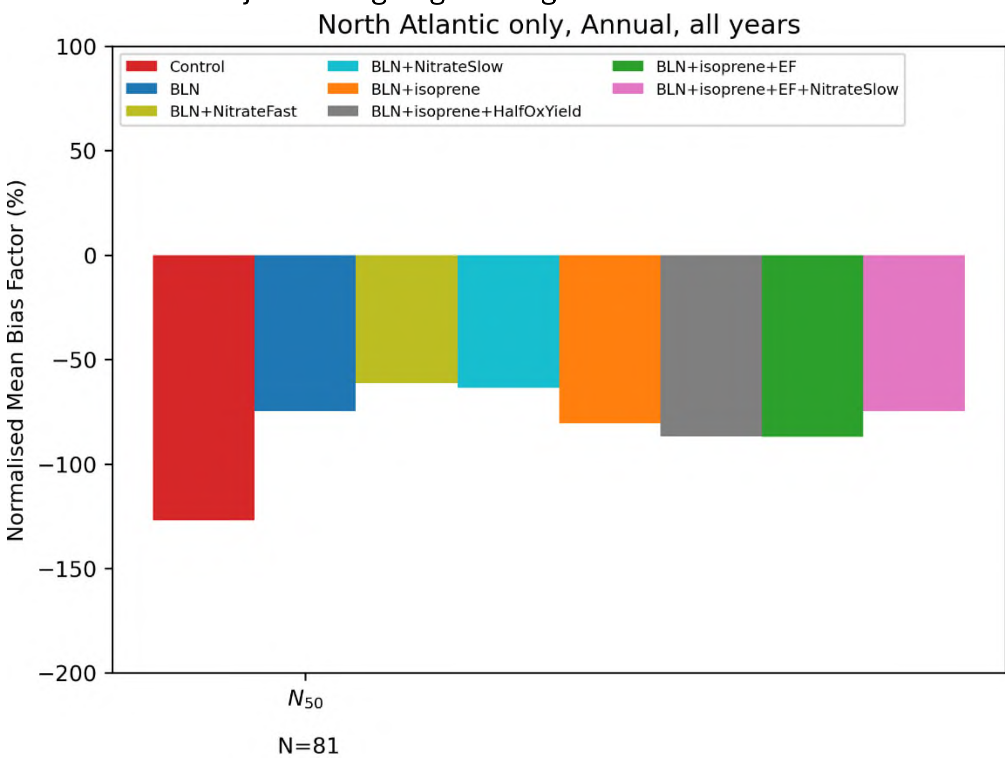
We thank the referee's suggestion. We added 'Simulated' and 'Satellite' at the top of the columns in Fig 1.

• The amount of underprediction in CDNC in the model seems rather notable. Is the magnitude of the CDNC underprediction similar to previous work using this and other models? Does the claim that background aerosols are likely to blame for this discrepancy consistent with underpredictions in other work or a known issue with this model? How does the out-of-plume model AOD compare to the satellite AOD to support this claim?

We agree that the aerosol concentrations are underestimated in our simulations. An underestimation of CDNC over northern North Atlantic has been reported in Grosvenor and Carslaw (2020) for the UKESM global model simulation. Our recent analysis with newer version of model (UKESM1.1) indicates a low bias of N50 by about 40% (NMBF~-75%) in North Atlantic (see the blue bar (BLN) in the figure below). Even stronger low bias in particle number concentration has been seen in UM_UKCA regional simulations off the coast of Portugal (Yoshioka et al., personal communication).

The low bias in our original simulation can also be seen in AOD, although we note that AOD may not be a good predictor of CDNC because it is dominated by second moment of the size distribution ($\int n*r^2$). Table below shows area-averaged AOD in the simulations used in this study compared to MODIS AOD, both at 550 nm, together with relative (%) differences (simulation/MODIS * 100) for the first 6 days. The selected region (50-62N; 40-15W) covers the northern North Atlantic SW of Iceland, mostly outside the volcanic plume (containing regions out of plume and out of bounds). This indicates that the original simulation has a low bias except for the first two days, and that this bias has been somewhat reduced in the enhanced simulation.

However, we note that increasing the background aerosol concentration by changing model settings and perturbing the model parameters did not affect the results of comparing volcano on and off simulations.
Nevertheless, we are fully aware of the pressing need to improve the background aerosol and CDNC in the UM-UKCA model. We added the following sentences in the second paragraph of section 3.1:
General underestimation is considered likely due to biases in background aerosols or an underestimate in the treatment of the activation of cloud droplets than a bias in the Holuhraun volcanic emission implemented in the model. The use of alternate activation schemes is a subject of ongoing investigation.



date	original	%diff	enhanced	%diff	MODIS
20140901	0.148	21%	0.152	24%	0.122
20140902	0.096	-4%	0.102	2%	0.100
20140903	0.087	-23%	0.098	-14%	0.113
20140904	0.077	-25%	0.084	-18%	0.103
20140905	0.088	-26%	0.092	-23%	0.119
20140906	0.087	-46%	0.089	-44%	0.159

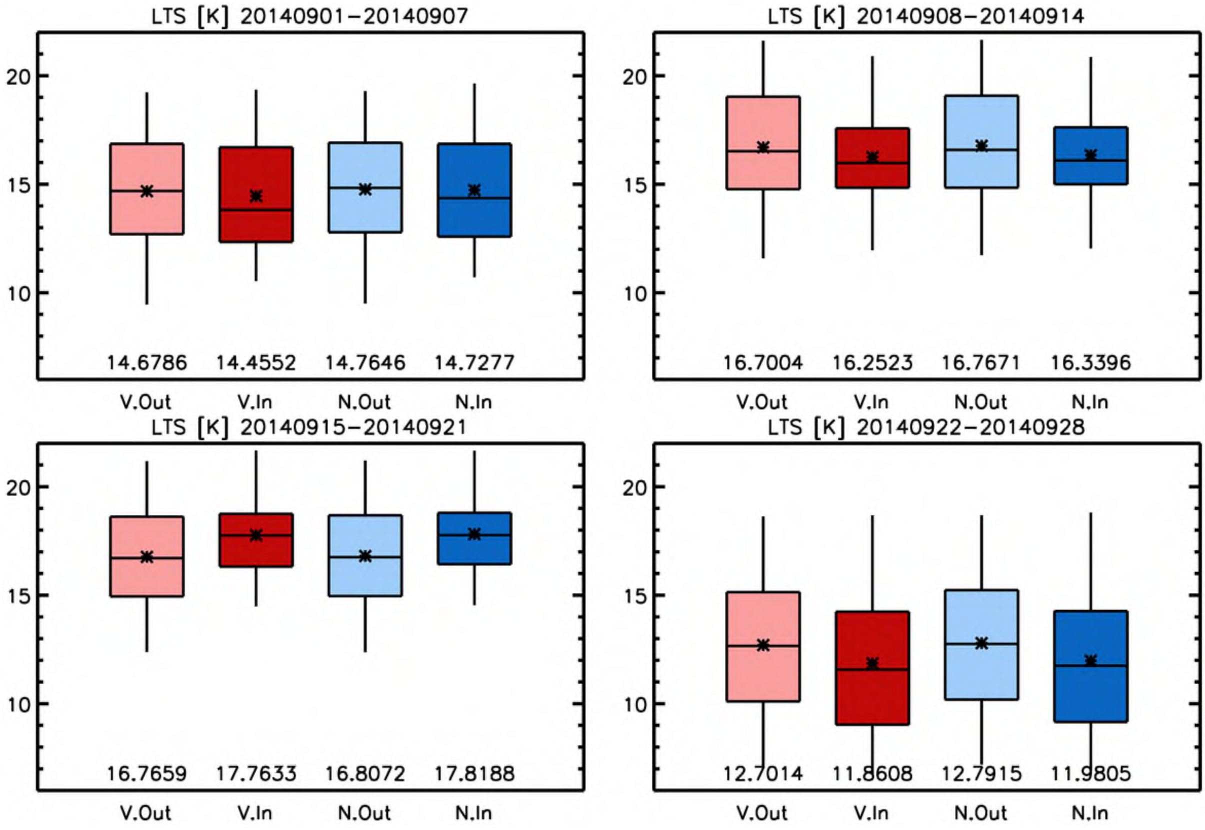
• In the discussion and conclusions, can the authors posit on the potential meteorological covariabilities that my lead to a reduction in the TOTAL and LOCATION effects for CDNC and Reff in week 3? Why did the authors not consider these effects and the other mentioned effects using cloud-controlling factors for this purpose?

We thank the referee for raising this important point. We calculated the lower tropospheric stability (LTS) as the difference between the potential temperatures at the 700 hPa level and the surface. The figure below shows box-and-whisker plots of LTS within and outside the plume regions for both the Volc and NoVolc simulations over four weeks.

The results indicate that LTS was higher inside the plume than outside in week 3, in contrast to the other weeks. They also show that this pattern is insensitive to whether the volcano is on or off in the simulations. Therefore, mesoscale variations in stability are likely to have contributed to the masking effect observed in week 3.

We added these plots in figure 9 and the following sentences in the fourth paragraph of section 4:

Examination of the lower tropospheric stability (LTS), for the high resolution UM, in and out of the plume and for volcano on and off is provided in figure 9. This indicates that LTS is greater inside the plume than outside in week 3, in contrast to the other weeks, and that this result is insensitive to whether the volcano is on or off in the simulations. Therefore, the mesoscale stability variations are likely to have been the source of the masking effect seen in week 3.



• Can the authors briefly speak to some (if any) of the microphysical parameterization scheme differences that could lead to differences in effects between model datasets?

Section 2 describes the CASIM cloud microphysics scheme used in this study.
The UKESM simulation analyzed by Peace et al. (2024) uses a single-moment bulk scheme (Wilson and Ballard, 1999; Sellar et al., 2019) in which droplet number concentration is diagnosed rather than prognosed. In contrast, CASIM explicitly predicts droplet number and mass, allowing a more direct representation of aerosol–cloud interactions (Grosvenor et al., 2017; Field et al., 2023). This difference may contribute, in addition to the resolution difference discussed in Section 4, to the weaker CDNC and Reff responses in UKESM compared to our simulations.
ICON also uses a two-moment bulk scheme (Seifert and Beheng, 2006) similar to CASIM. However, since their results cover only the first week and there are also differences in the atmospheric models and experimental designs used, it is difficult to attribute the simulated differences in cloud properties to differences in the cloud microphysics schemes.

• Lines 468-469: has evidence of this semi-direct effect been suggested or shown in similar previous work? If so, the authors should provide citation here. If not, I still feel it appropriate for the authors to provide some citation to support this point of discussion.

We understand the reviewer's concern about the impact of volcanic aerosols on the cloud field outside the plume. There is indeed precedent in the literature for semi-direct or circulation-mediated effects from aerosol plumes. For example, Diamond et al. (2022) reported that smoke over the southeastern Atlantic not only caused local semi-direct effects but also altered the large-scale atmospheric thermal structure, thereby affecting cloud properties over a broader region.
We have added the following sentence: "This is similar to the effect of smoke over the southeastern Atlantic altering the large-scale atmospheric thermal structure and thereby cloud properties, as reported by Diamond et al. (2022)."

• Lines 473-474 (answer to intro question 1): can the authors please provide a quantification of the CDNC and Reff increases/reductions?

We modified the text and included the ranges of changes in CDNC and Reff as follows; "The modelling results show an increase in droplet number by a factor of 1.6 to 2.6 and a reduction in effective radius by 1.6 to 2.7 μm, except during the third week. These findings are consistent with the direction of changes seen in satellite observations and previous modelling studies, though the exact magnitudes vary among datasets."

References

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