Supplement for

“Global climate models are unable to reproduce cloud cover response to aerosol”

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Supplementary Discussion

Section S1 Spatial distribution of cloud property changes in observations and models

The spatial distributions of key cloud properties and the influence of Holuhraun-2014 volcanic plumes are shown in Figures S1-S3, with observations[1] provided in the top row and simulations from six models in the following rows.

Fig. S1 shows the baseline of the control case, i.e. without volcanic eruption. A large divergence is found among six models. For example: regarding cloud droplet number concentration (Nd), CESM2.1.0 underestimates by about 70%, but CNRM-ESM2-1 overestimates by about 40%; regarding droplet effective radius (Re), ECHAM6.3-SALSA2.0 underestimates by about 65%, and other models slightly underestimate; regarding liquid water path (LWP), UKESM1 and CNRM-ESM2-1 underestimate by 50-70% and other models slightly underestimate; regarding liquid cloud cover (LCC), UKESM1 and CNRM-ESM2-1 provide a reasonable estimate, ECHAM6.3-HAM2.3, ECHAM6.3-SALSA2.0 and CESM2.1.0 underestimate by 35-65%, while CAM5.3_Oslo overestimates by about 50%. Fig. S2 shows the volcanic case, in which both large overestimation and underestimation of all cloud variables are observed. This large variation of baseline and experimental simulations is expected and is in line with previous studies, showing great diversity of the chosen models and also justifying that the importance of looking at the relative change of clouds caused by aerosol (rather than absolute change)[2], which is the cloud susceptibility in logarithm scale that we discuss in the main text.

Fig. S3 provides the difference between the volcanic case and baseline, i.e. Fig. S2 minus Fig. S1, which most intuitively shows the region influenced by the volcanic plume. Generally speaking, over the whole North Atlantic, observation shows a clear increase of Nd, a decrease of Re, no clear change of LWP (only -0.03 g m^{-2} for domain-average), and a large increase of LCC; models well capture the spatial patterns of ΔNd and ΔRe (Twomey effect). Four out of six models largely overestimate the increase of LWP, with only UKESM1 and CNRM-ESM2-1 showing a reasonable regional response of LWP, in line with Malavelle et al. (2017)[3]. However, all models show negligible increases in LCC and fail to reproduce the observed strong increase in cloud cover.

These model-observation intercomparison highlights the model's incapability to reproduce the key cloud properties and aerosol-induced cloud responses as observed, especially the LWP and LCC. Although Re and LWP are determined mainly by cloud microphysics diagnostically, while LCC is diagnosed by grid-mean relative humidity, these key cloud variables are interlinked through the partitioning of water in the vapour, liquid, and ice phases in models. To further understand model bias and its capability to represent the observation, we applied different cloud microphysics and cloud cover schemes and conducted sensitivity studies of cloud microphysical processes, see detailed discussion in the main text.

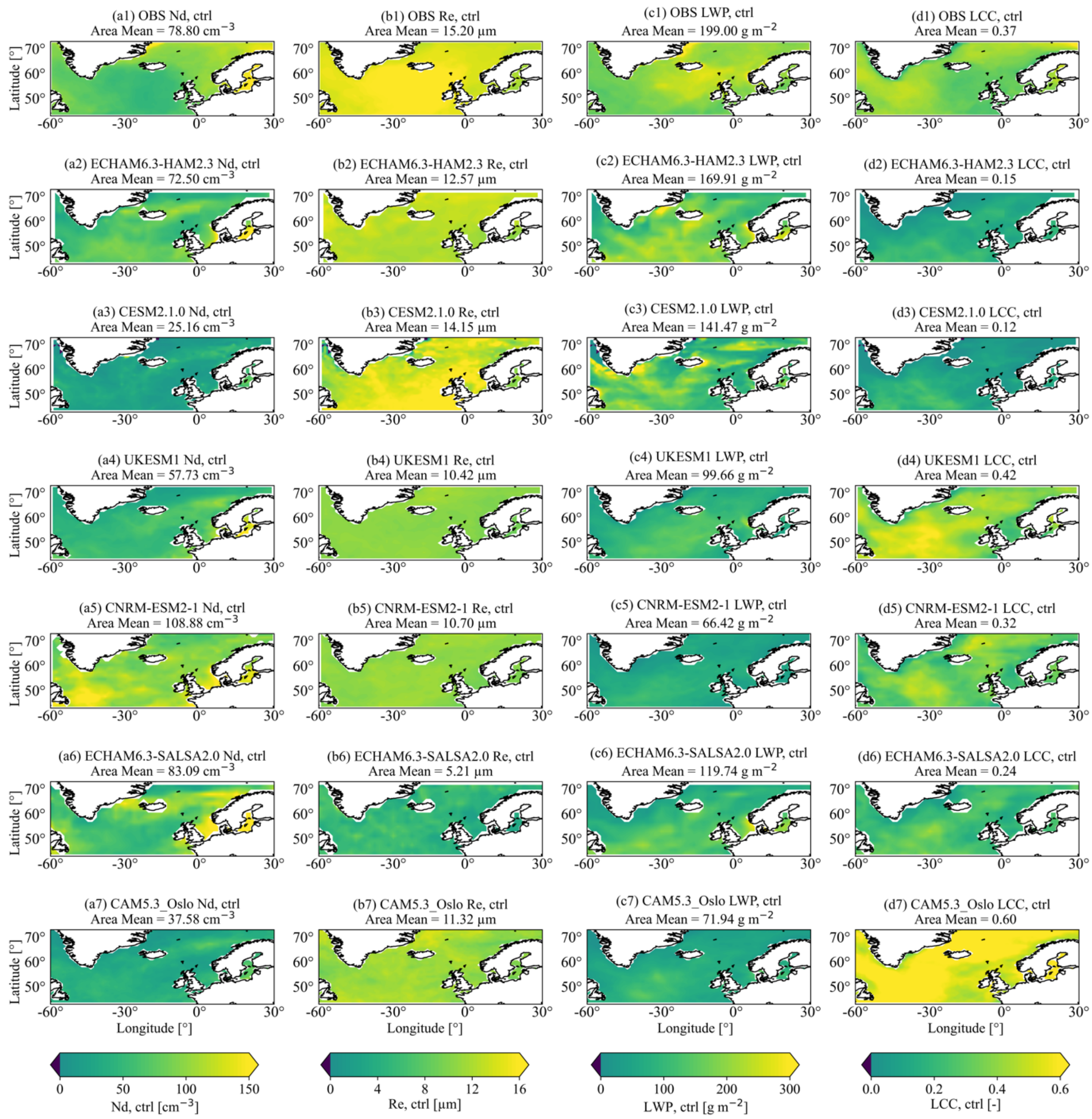

Fig. S1. The **spatial distribution** of Nd, Re, LWP, and LCC in Oct. 2014 (corresponding to columns from left to right), **for the control case**, i.e. without volcanic eruption. Rows from top to bottom are: (**a1-d1**) observations derived from a combination of satellite and machine learning[1]; (**a2-d2**) ECHAM6.3-HAM2.3; (**a3-d3**) CESM2.1.0; (**a4-d4**) UKESM1; (**a5-d5**) CNRM-ESM2-1; (**a6-d6**) ECHAM6.3-SALSA2.0; (**a7-d7**) CAM5.3_Oslo.

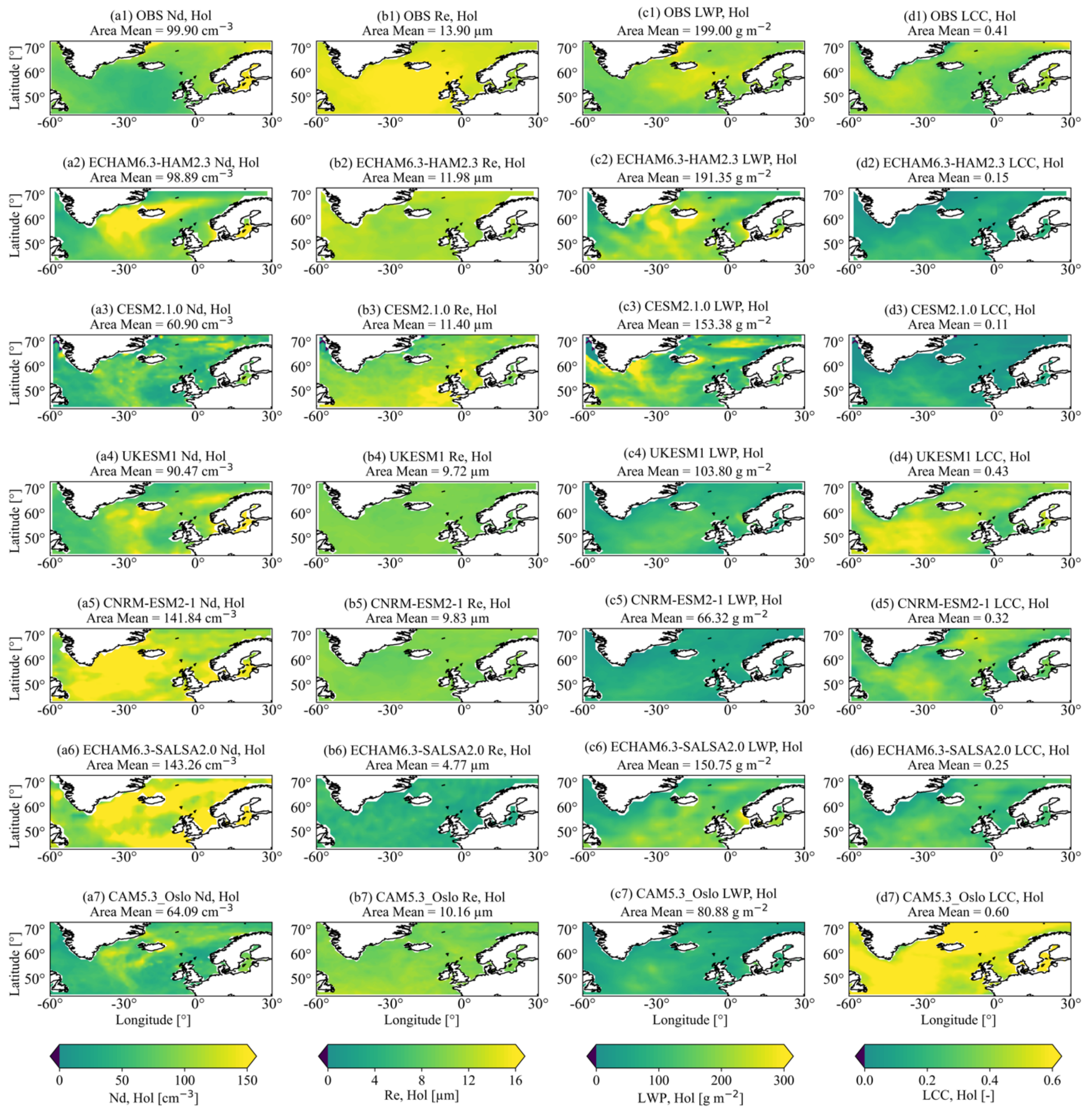

Fig. S2. The **spatial distribution** of Nd, Re, LWP, and LCC in Oct. 2014 (corresponding to columns from left to right), **for the volcano case**. Rows from top to bottom are: (**a1-d1**) satellite observations[1]; (**a2-d2**) ECHAM6.3-HAM2.3; (**a3-d3**) CESM2.1.0; (**a4-d4**) UKESM1; (**a5-d5**) CNRM-ESM2-1; (**a6-d6**) ECHAM6.3-SALSA2.0; (**a7-d7**) CAM5.3_Oslo.

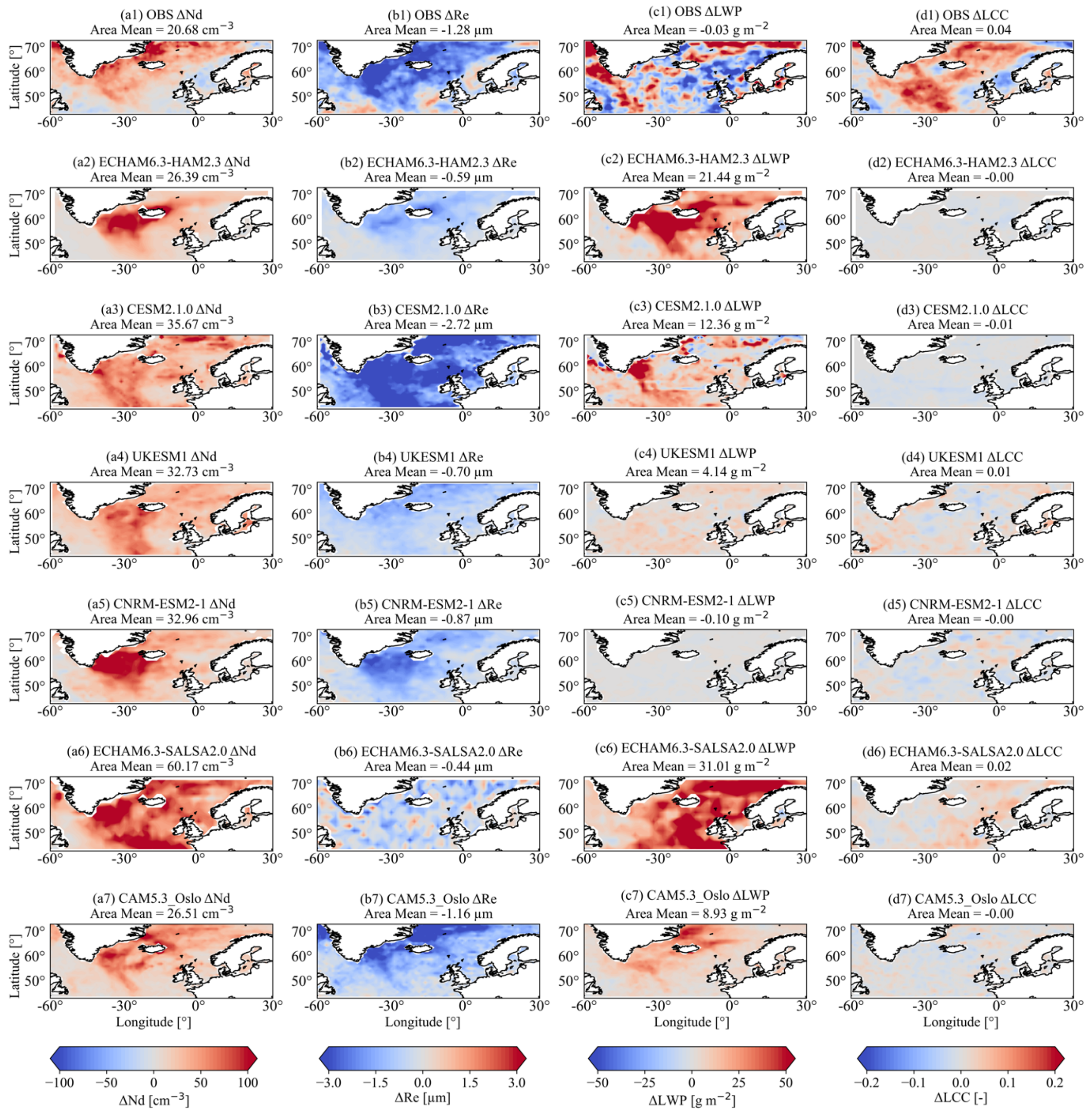

Fig. S3. The **spatial distribution** of Nd, Re, LWP, and LCC in Oct. 2014 (corresponding to columns from left to right), for the **differences between the volcano and control cases**. Rows from top to bottom are: (**a1-d1**) satellite observations minus machine learning derived observations of control case[1]; (**a2-d2**) ECHAM6.3-HAM2.3; (**a3-d3**) CESM2.1.0; (**a4-d4**) UKESM1; (**a5-d5**) CNRM-ESM2-1; (**a6-d6**) ECHAM6.3-SALSA2.0; (**a7-d7**) CAM5.3_Oslo.

References for Supplementary:

1. Chen Y, Haywood J, Wang Y, Malavelle F, Jordan G, Partridge D*, et al.* Machine learning reveals climate forcing from aerosols is dominated by increased cloud cover. *Nature Geoscience* 2022, **15**(8)**:** 609-614.

2. Ghan S, Wang M, Zhang S, Ferrachat S, Gettelman A, Griesfeller J*, et al.* Challenges in constraining anthropogenic aerosol effects on cloud radiative forcing using present-day spatiotemporal variability. *Proceedings of the National Academy of Sciences* 2016, **113**(21)**:** 5804-5811.

3. Malavelle FF, Haywood JM, Jones A, Gettelman A, Clarisse L, Bauduin S*, et al.* Strong constraints on aerosol–cloud interactions from volcanic eruptions. *Nature* 2017, **546**(7659)**:** 485-491.