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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Importance of tropospheric volcanic aerosol for indirect radiative forcing of climate" *by* A. Schmidt et al.

Anonymous Referee #2

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The manuscript by Schmidt et al. investigates the effect of volcanic emissions on cloud condensation nuclei (CCN) and cloud droplet number (CDN) concentrations. The global chemistry transport model GLOMAP is run with several emission conditions, to test the sensitivity of aerosol concentrations in present-day and pre-industrial. The CDN concentrations are used to estimate the climate effect of volcanoes via the cloud albedo effect. The manuscript is well written and concise, and the analysis contains all the necessary information for the reader. The results are also put into context, by comparing to other natural (DMS) aerosol sources. The main problems with the manuscript are related to certain methods chosen, when taking into account the purpose of the manuscript: indirect forcing of volcanoes. The specific comments can be found numbered below.

1) There are two versions available of GLOMAP, sectional and modal. In this study, the modal version was chosen. However, it is likely that the modal version does not perform as well as the sectional model, when considering nucleation and growth. Also, cloud processing might make a difference between the sectional and modal versions. Since there are not that many model simulations, the computational benefits should not be significant. Is there a specific reason to choose modal version over the sectional one?

2) The model includes binary homogeneous sulphuric acid-water nucleation. The paper by Stevens et al. (2012) studied nucleation and growth in power plant plumes, with several important implications regarding the current manuscript. For example, they found that the binary nucleation did not predict any nucleation in the plume, and that there is an order of magnitude uncertainty in the formation of >30nm particles due to the chosen nucleation scheme. Certainly, the situation is different in a LES model (Stevens et al.) and a global model grid box of 2.8x2.8 degrees. However, the choice of nucleation scheme should not be overlooked.

The effect of nucleation scheme is discussed on page 8026. Additional sensitivity simulations were done using boundary layer nucleation (BLN), leading to decreased cloud albedo effect. It is concluded in Spracklen et al. (2010) that including BLN improves the modeled number concentrations, compared to simulations without BLN. As is shown in this manuscript, the BLN affects the baseline CDNC and would affect the final conclusions. While it is true that, "we do not fully understand the precise nucleation mechanism operating", I would strongly suggest to perform the model simulations with the nucleation mechanism that fits the observations best.

3) 2.5% of emitted SO2 is assumed to nucleate in subgrid-scale and is directly partitioned to accumulation and coarse modes, increasing number and mass concentrations. A similar assumption is used in many global model studies. However, this is one of the topics that should be focused more in the manuscript. The issue of sub-grid scale nucleation is investigated for example in Stevens et al. (2012), indicating that

the "primary sulfate" fraction could be much higher, even up to 9%. Also, Luo and Yu (2011) show that in addition to the "primary sulfate" fraction, aerosol number concentrations are highly sensitive to assumed size distribution of the "primary sulfate". The issue of "primary sulfate" should be discussed in the manuscript. Also, I suggest a few sensitivity runs testing either the "primary sulfate" mass fraction or it's size distribution, for example for PD and PI basecases.

Smaller remarks:

4) Introduction: maybe indicate more clearly, what is new compared to earlier studies? Is it mainly the comparison of PD and PI conditions?

5) The meteorological fields are for year 2004 for all simulations. However, the radiative calculations use average cloud fields from years 1983-2005. For the sake of consistency, could the radiative code apply fields from year 2004?

6) P.8017, l.28: What are the dynamics-induced changes in CCN, that are not accounted for?

7) P.8020, l.12: Do the first two sentences belong here, or more in the introduction?

8) Why is the absolute change in CCN and CDN concentrations higher in PI than in PD?

9) Fig.2. Indicate the definition of CCN used in the figure. I assume it varies from location to another, so mention this in the figure caption.

10) Fig.A1. Typo: should be "cloud condensation nuclei".

References:

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 8009, 2012.