Author's reply to review of "Importance of tropospheric volcanic aerosol for indirect radiative forcing of climate" by Reviewer #2

Anja Schmidt, School of Earth and Environment, University of Leeds, LS2 9JT, Leeds, UK

a.schmidt@leeds.ac.uk

We would like to thank the anonymous reviewer for their constructive and detailed comments on our manuscript. To guide the review process, we have inserted the reviewer's comments in italics and our responses in regular font. Text added to the manuscript itself is indicated in bold.

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?

Mann et al. (2010) showed that the modal scheme performs generally well in comparison to a range of observations. Mann et al. (2012) examined differences between the sectional and modal GLOMAP schemes finding that surface level CCN concentrations simulated by the two versions within 25% in most regions on the annual mean although biases were higher in some marine regions due to the necessarily simpler approach to size-resolved growth and cloud processing. Note also that the runs in this paper use the latest GLOMAP-mode version which was improved in the Mann et al. (2012) study to reduce bin-mode biases. Our results in the above papers confirm that the differences between modal and bin schemes are always smaller than between either model and observations, making it difficult to conclude which model is better. Although heuristically one might expect a bin model to perform better, this conclusion has very limited observational evidence.

The study we present is part of a series of studies performed using the modal version of GLOMAP to quantify the contribution of several natural aerosol sources to cloud condensation nuclei number concentration and indirect radiative forcing of climate. Using the modal scheme enabled us to put our results in context with other natural aerosol sources such as DMS by comparing our results with a previous study using GLOMAP-mode (Woodhouse et al., 2010) and to the ECHAM5-modal scheme (Thomas et al., 2010).

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.

We agree with the Reviewer that the choice of nucleation scheme will affect the results. However, as we state in our manuscript so far the precise nucleation mechanism operating in the boundary layer is not understood. Even though Spracklen et al. (2010) showed that the empirical BL mechanism improves the modelled aerosol number concentrations, there is increasing evidence that aside from H_2SO_4, other molecules (likely organics) are involved. Furthermore, Reddington et al. (2011) have shown that using an empirical BL mechanism does not necessarily improve modelled CCN number concentrations compared to observations over polluted regions in Europe. Moreover, most of the volcanic SO_2 is emitted well above the boundary layer where binary homogenous nucleation is the dominant new particle formation mechanism. We feel that we already provide a good indication of the uncertainty that could arise from including (or excluding) BLN by calculating the differences in cloud albedo effect under present-day atmospheric conditions.

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 base cases.

We agree with the reviewer that the fraction of sub-grid sulfate and assumptions about its size distribution will influence particle number concentrations. However, in terms of the volcanic source, there is no evidence of a sub-grid sulfate fraction as high as 9% of the emitted SO_2. Our choice of 2.5% is in agreement with several observations at various continuously degassing volcanoes (e.g., Allen et al., 2002, Mather et al., 2003a,b, 2004, 2006, 2012; Martin et al., 2008), and the majority of other global aerosol modelling studies. We added further discussion to both Section 2.2 (Methods) and to the Discussion section (see reply to Reviewer 1) to clearly state these shortcomings and recommendations for future work. Even a 10% change in CDNC in either direction due to process uncertainties will not affect our main conclusions: uncertainty in the volcanic SO_2 flux does result in a very large spread in the magnitude of the anthropogenic cloud albedo forcing.

We amended the manuscript as follows:

Measurements of near-source sulphate particles at several continuously degassing volcanoes indicate a fraction of 1 % to 5 % of the total emitted SO_2 (e.g., Allen et al., 2002; Mather et al., 2003a,b, 2004, 2006, 2012; Martin et al., 2008). Luo and Yu (2011) have shown that global CCN number concentrations are sensitive to both the sub-grid sulphate mass fraction and the assumptions made about its size-distribution, amongst many other aerosol processes. We do not test the sensitivity of our results to the assumptions made about the sub-grid sulphate fraction because we expect the uncertainty in the volcanic sulphur source strength to have a much larger impact on our results than the chosen sub-grid sulphate fraction.

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?

We amended the Introduction as outlined below to more clearly state the advancements compared to previous studies:

Previous modelling studies have assessed the relative contribution of volcanic degassing to the global sulphur mass budget (Chin and Jacob, 1996; Stevenson et al., 2003a) as well as its radiative effects (Graf et al., 1997, 1998; Langmann et al., 1998) under PD atmospheric conditions. Graf et al. (1997), amongst others, concluded that volcanic sulphur emissions are at least as important as anthropogenic sulphur emissions with regard to the global sulphur cycle and their contribution to the radiative forcing of climate. Langmann et al. (1998) used a regional chemistry transport model and found that natural sulphur sources such as DMS and volcanoes account for ~34% of the total direct sulphate aerosol radiative effect over Europe. However, these earlier studies neither assessed the impact of volcanic degassing under PI conditions (when the aerosol background loading was generally lower) nor did these studies address the role of the uncertainty in the volcanic source strength.

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?

We expect the specific year for the cloud fields used will have only a very minor effect on our results. Our methodology is in line with many studies using the offline version of the Edwards & Slingo (1996) radiative transfer code.

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

We amended that sentence as follows:

Since this study uses a 3D offline chemistry transport model, we do not account for feedbacks between the additional aerosol loading and atmospheric dynamics.

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

We agree that these statements are better placed in the Introduction so we removed them here.

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

In Section 3.1 we discussed that non-linear interactions between nucleation, condensation, gas-phase oxidation of SO_2 and aqueous-phase oxidation of SO_2 drive the production of additional CCN-sized particles depending on both aerosol background concentration and volcanic sulphur flux strength. We amended the sentence in Section 3.3 as follows:

However, absolute changes in global mean CDNC are also less in the PD (174 cm^{-3} versus 158 cm^{-3}, difference 16 cm^{-3}) compared to the PI (107 cm^{-3} versus 76 cm^{-3}, difference 31 cm^{-3}) as a result of non-linear interactions between chemical and microphysical processes, resulting in the production of more particles per unit of volcanic sulphur emission in the PI (see Sect. 3.1 and Sect. 3.2).

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.

Yes, that is correct. We amended the figure caption.

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

Thanks, this has now been corrected.

References (those added in the revised version of the paper are indicated in bold):

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