Author's reply to review of "Importance of tropospheric volcanic aerosol for indirect radiative forcing of climate" by Reviewer #1

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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 titled 'Importance of tropospheric volcanic aerosol for indirect radiative forcing of climate' by Schmidt et al presents modeling results regarding the importance of tropospheric aerosol in producing CCN particles and its impact on cloud properties. The authors use a modal approach to model aerosol microphysics in a global model. Based on a volcanic sulfur emission inventory they demonstrate tropospheric volcanic aerosols have a great potential to affect our understanding of both natural and anthropogenic aerosol indirect forcing, specifically, aerosol cloud albedo effect. The message is clear and of interest for the community. The manuscript is also well written and generally easy to read. I recommend the publication of this manuscript after the authors address the following comments.

1. The structure of the manuscript may be modified to accommodate a discussion section. This is raised because the manuscript in its current form contains quite a few scattered discussions at several different places and they are related subjects. These discussions are generally interesting and relevant for the manuscript. However, the current arrangement may divert the attention of readers not familiar with technical details of these discussions. It is therefore in my opinion sensible to create a discussion section to go over the discussed points in a concentrated fashion. Another, maybe more important, reason to do so is that the materials presented and the collusions reached in the manuscript do need more discussions on the validity of the assumptions and other technicalities (see later comments).

We agree with the reviewer's suggestion to change the structure of the manuscript. We now have a "Results" section and a "Discussion" section. We moved the paragraphs in question to the Discussion section.

2. It has been increasingly clear that the assumption for a 'pure' cloud albedo effect, i.e. LWP being fixed, is probably not a very good one in reality. As such, latest investigations often use online aerosol-cloud interaction calculations to get a more realistic estimate of cloud albedo effect, including feedbacks. The current study is an offline calculation. Though the aerosol microphysics is quite advanced the way to reach the forcing numbers is a little crude. I am not objecting this kind of analysis. Instead, in my opinion the authors need to clearly state these shortcomings. For example, the authors seem to take the monthly mean grid CDNC and apply it to a cloud climatology to calculate cloud albedo forcing. If it is indeed what is being done it needs to be stated for clarity.

We indeed take monthly mean CDNC grid and apply it to a monthly mean cloud climatology to quantify the cloud albedo effect. We acknowledge that deriving monthly-mean CDNC from monthly-mean size-resolved particle concentrations and monthly-mean particle composition fields ignores temporal variations in aerosol properties. We also acknowledge the fact that our offline study cannot account for all the aerosol-cloud interactions and we clearly state these shortcomings in the text. However, we also doubt whether coupled aerosol-climate models provide any meaningful information on aerosol-cloud couplings given their very low resolution and lack of detailed information on cloud microphysics and mixing processes. However, we are confident that our approach is entirely appropriate to evaluate the relative importance of volcanic effects against a well established (if incomplete) metric used in past IPCC reports. Please see response to suggestion 3 for the changes we made to the manuscript.

3. Regarding equation 1, it is not clear what CDNC is used. PI-vol? PI-no vol? or a fixed number? Or is it changing for every model run? This is a critical issue that needs clarification. And, is the re_control always 10 μm in calculating albedo forcing? Are all the forcing numbers reported here annual and global mean?

The CDNCs used in equation 1 correspond to the particular model experiment considered, e.g. in the model experiment where we calculate the cloud albedo effect between the PD_vol and PD_no_vol, $CDNC^{control}$ corresponds to PD_vol and $CDNC^{perturbed}$ corresponds to PD_no_vol.

We amended the manuscript as follows:

Section 2.4 Radiative transfer code is now moved to follow on from section 2.5 Model experiments. We added additional text to the new section 2.5 Radiative transfer code:

The cloud albedo effect between a control and a perturbed experiment is quantified by modifying the cloud drop effective radius r_e (in μm), for low and mid-level water clouds (up to 600 hPa) only, as follows:

$$r_e^{perturbed} = r_e^{control} \times (CDNC^{control}/CDNC^{perturbed})^{1/3} \quad \textbf{(1)}$$

where CDNC (in cm^{-3}) corresponds to the monthly mean cloud droplet number concentration calculated from a particular GLOMAP-mode simulation, and a fixed value for $r_e^{control}$ = 10 μm is considered in order to ensure consistency with the ISCCP cloud retrievals. For example, when calculating the volcanically induced cloud albedo effect for PD_vol and PD_no_vol, $CDNC^{control}$ corresponds to PD_vol and $CDNC^{perturbed}$ corresponds to PD_no_vol. When calculating the anthropogenic cloud albedo forcing between PI and PD, $CDNC^{control}$ corresponds to PD_vol and $CDNC^{perturbed}$ corresponds to PI_vol.

The $r_e^{control}$ is indeed always considered to be 10 μm in our cloud albedo effect calculations. This is done to ensure consistency with the ISCCP cloud retrievals used to create our cloud climatology, where in the derivation of the cloud liquid water path (LWP), a constant effective radius of 10 μm was assumed. In Spracklen et al. (2011) we have tested both this constant $r_e^{control}$ approach, together with an alternative approach where the effective radius was variable for both the control and the perturbed experiments. Both approaches gave very similar answers, suggesting the methodology is robust.

All cloud albedo forcing values reported are annual global means, except for the values in Table 4 where we also state annual hemispheric and annual tropics means. We now state in all relevant Figure and Table captions that the values correspond to the global (expect where hemispheric means were given) annual mean.

Also, from the model-observation comparison the uncertainty (or bias) with the model is quite significant in light of what is being tracked down in this study. Would be beneficial to include this uncertainty into consideration?

Yes, we discussed in Section 3.4 (page 8027, lines 25 - 29) that further uncertainties originate in the PI and PD baselines (hence in radiative forcing) from missing or poorly defined aerosol sources. We now moved this paragraph into the Discussion section. Model realism is of course a common issue with all global assessments of forcing, although we think we have gone further than previous studies by actually evaluating a key diagnostic in the aerosol microphysics model.

Minor Comments:

1. *Page 8010, line 10: consider adding 'estimated based on our methodology'. The assumptions of the methodology shall be clearly stated as suggested in major comments.*

We now also clearly state the underlying assumptions in the "Method" section as outlined above.

2. *Line 15 page 8012: it is worth to note that the Hawaiian study seems to report volcanic aerosol impact 6000km away from the source.*

This is a very good suggestion. We added the following sentence to the introduction:

Yuan et al. (2011) showed that sulphur emissions emitted from Kilauea's Halema'uma'u Crater on Hawaii affect trade cumulus cloud amount and cause a regional total shortwave radiative forcing of up to −20 W m^{-2} as far downwind of the volcanic source as 6000 km.

3. *Line 4, page 8016: why 2000 dust is used while other fields are from 2004? In the same paragraph, does the model treat aerosol mixing at all?*

We used prescribed daily-varying dust emissions for the year 2000 from datasets provided for the AEROCOM model intercomparison initiative (Dentener et al., 2006). We included these dust emissions to ensure our representation of the present-day aerosol contained all the major particle types (sulphate, sea-salt, carbonaceous aerosol and dust).The meteorological fields used in the 3D offline transport model are from 6-hourly varying ECMWF reanalyses for a different year (2004), but we do not expect this minor discrepancy to have any impact on our simulated cloud albedo effects

4. Equation 1, page 8019: due to many factors effective radius does not respond to CDNC in a theoretical 1/3 power law. It is the upper limit, which may be worth noting here.

Equation 1 was derived from the Bower et al. (1994) effective radius r_e (in µm) parameterisation, namely:

$$r_e = 100 \times [LWC \times 3/(4\pi \times \rho_w \times CDNC)]^{1/3},$$

where *LWC* is the liquid water content in g m^{-3}, ρ_w (in g cm^{-3}) is the density of liquid water, and *CDNC* is the cloud droplet number concentration in cm^{-3}. This parameterisation was used by Bower et al. (1994) for layer clouds and small cumuli. Other studies also used 1/3-power-laws of the *LWC/CDNC* ratio, i.e.

$$r_e = \alpha \times (LWC/CDNC)^{1/3}$$

to parameterise the effective radius. Martin et al. (1994) for example, derived values of α for stratocumulus clouds using in situ measurement data. While Bower et al. (1994) derived α=62.04, Martin et al. (1994) derived α=66.8 for maritime and α=70.91 for continental stratocumulus clouds. More recently, Nair et al. (2012) also used various 1/3-power-laws of the *LWC/CDNC* ratio to compare with aircraft observations during the CAIPEEX campaign.

We acknowledge the fact that in practice the effective radius does not always respond to CDNC in an exact theoretical 1/3 power law, but several sensitivity calculations we performed for different effective radius expressions showed that, since in our case we calculate the radiative effect of a change in r_e, rather than the radiative flux for a particular r_e, the exact expression used for effective radius had virtually no effect on the calculated cloud albedo effect.

5. *Starting at line 20, page 8023: the emission may be quite different between the Hawaiian study and what is used in this manuscript. This may be worth noting.*

The reviewer is correct. We added the following sentence to the Discussion section:

Yuan et al. (2011) used satellite retrievals to show that emissions from Kilauea's summit vent (Hawaii) induced a regional cloud albedo effect of up to −4 W m^{-2}. Using our global aerosol microphysics model, we calculate an annual mean cloud albedo effect around the Islands of Hawaii of −2.32 W m^{-2} (uncertainty range −1.48 to −3.17 W m^{-2}) for PI, and of −1.07 W m^{-2} (uncertainty range −0.64 W m^{-2} to −1.68 W m^{-2}) for PD. Note that our estimate of the radiative effects will be partly due to volcanic sulphur emissions from Hawaii and partly due to long-range transport of aerosol from other volcanoes. In the model, we emit a total of ~2600 t of SO_2 per day in the gridboxes above Hawaii accounting for emissions from both the Kilauea's summit and east rift zone vents. Therefore, the comparison of our model estimate to the Yuan et al. (2011) study is for qualitative purposes only.

6. 3RD paragraph on page 8024 and 2nd on page 8025 are good discussions that may be moved to the discussion section.

OK, moved into Discussion section.

7. Starting at line 20, page 8029: this example is a little out of place. Again, putting this kind of discussion in a right place would make both the manuscript easier to read and the discussed points more outstanding.

OK, moved into Discussion section.

References:

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