

We thank the reviewer for a thorough review and constructive comments to improve this manuscript. Item-by-item replies are provided below; text in italics shows reviewer's comments.

Review #2:

***Comment.** Page 26442:, Line 8: How accurate is the MODIS cloud liquid water retrieval? A discussion about the data quality would be beneficial here. For example, Min et al. (2012, ACPD) reported that over Southeast Pacific the agreement between MODIS retrieval and in-situ measurement depends on the adiabatic status of the cloud.*

**Reply:** The reviewer is right. The MODIS liquid path is estimated from the retrievals of cloud effective radius and cloud optical depth from the visible and near infrared spectrum. Since the retrieval is most sensitive to the properties of cloud top, the assumption of the constant effective radius throughout the cloud layer generally result in overestimation of liquid water path in MODIS product at the single pixel or small scale level. Of course, many other factors (including 3D effects, with or without drizzle, vertical structure of clouds, cloud type, and geographical location of cloud) can also complicate the analysis of uncertainties in MODIS retrieved liquid water path.

In the main text that describe the MODIS cloud products, we added:  
“Past analysis showed that the biases in the liquid water path data retrieved from MODIS depend on the cloud type, 3-dimensional structure of the clouds (e.g., broken vs. overcast, adiabatic vs. non-adiabatic, etc.), satellite-Sun-Earth geometry, and whether or not having drizzle in the clouds or absorbing aerosols above the clouds [Wilcox et al., 2009; Seethala and Horváth, 2010; Min et al., 2012]. In the middle-to-high latitude oceanic region of our interest, Seethala and Horváth [2010] found that the MODIS liquid water path data overall overestimates the counterpart retrieved from space-borne microwave (AMSR-E) instrument, although significant underestimation can also occur especially for broken clouds. While quantifying the uncertainties in MODIS liquid water path product in our study region and time period is challenging, all past studies support that summation or averaging of MODIS liquid water path over a large spatial domain often reduce the uncertainty [Seethala and Horváth, 2010; Min et al., 2012], which is also the strategy used in this study during the intercomparison of MODIS and GEOS-5 liquid water path (section 4.1).”

In the discussion of GEOS and MODIS liquid water path comparison, we added:  
“This is especially likely after further consideration that MODIS liquid water path may also has a positive bias (~10% in global averages over ocean when compared to the AMSR measurements) [Seethala and Horváth, 2010]. Indeed, our sensitivity experiment shows that a reduction of liquid water path by 15% in the first two days in GEOS-5 field results in a 5% increase in SO<sub>2</sub> total amount (figures not shown).”

***Comment.** Page 26443, Line 20: What's the time step used in the model? Is it the same as in the meteorological/re-analysis forcing data (3 hours)?*

**Reply:** The transport times step is 15 minutes, the convective time step is 15 minutes, emiss time step is 30 minutes, and chemistry time step is 30 minutes. To avoid clutter, we add in the revision that the time step is 15 minutes in the model.

*Comment. Page 26444, Line 27: "A good agreement with no bias was found . . ." I think the performance of the model is overstated here. Compared to the observation, models always have some bias in certain aspects (e.g., in specific regions or seasons).*

**Reply:** We reword the sentence as the following. "A good agreement with no systematic bias was found at the continental scale for comparison of the GEOS-Chem simulated distribution of sulfate-ammonium particles and their extent of neutralization with those from ground-based observations [Park et al., 2004; Martin et al., 2004]."

*Comment. Page 26445, Line 20: Why one day duration was assumed here? Is it a reasonable assumption?*

**Reply:** This assumption is based upon Waythomas et al. (2010) in which they describe Kasatochi eruption as a 'daylong eruption', with near-continuous emissions from ~2200 UTC on August 7 until ~2100 UTC on August 8. The following text is added in the revision: "Based upon Waythomas et al. (2010), the eruption duration is assumed to be 24 hours (with a starting time of 2200 UTC on 7 August 2008) in the model." Waythomas, C. F., W. E. Scott, S. G. Prejean, D. J. Schneider, P. Izbekov, and C. J. Nye (2010), The 7–8 August 2008 eruption of Kasatochi Volcano, central Aleutian Islands, Alaska, *J. Geophys. Res.*, 115, B00B06, doi:10.1029/2010JB007437.

*Comment. Page 26446, Line 3-5: Based on the statement here, it seems that the hygroscopic growth is only considered for optical property calculation. However, (wet) particle size is important for sedimentation and turbulent dry deposition calculations. Using dry size for these calculation will underestimate the particle sedimentation rate. A discussion about this would be helpful.*

**Reply.** The hygroscopic growth effect on the dry deposition and sedimentation of the particle is considered in GEOS-Chem. The revised text now reads as: "Dry deposition is based on the resistance-in-series scheme [Wesely, 1989], with the consideration of the hygroscopic growth of aerosol particles [Park et al., 2004]."

*Comment. Page 26446, Line 5-8: It seems to me the externally mixing was assumed in the calculation. Whether it is true or not, it is necessary to mention the mixing assumption in the calculation.*

**Reply.** We added in the text that "In the RTM calculations, all aerosol and cloud particles are assumed to be externally mixed [Wang et al., 2008]."

**Comment.** Page 26446, Line 11: "all-sky" or "total-sky" is more commonly used than "full-sky" in literature.

**Reply.** "All-sky" is now used through out the text.

**Comment.** Page 26446, Line 16-22: Is the geometric radius (0.07 $\mu\text{m}$ ) for dry aerosols or wet aerosols? If it is the dry radius (as I assume), using a fixed geometric radius of 0.19 $\mu\text{m}$  to consider the enhancement of hygroscopic growth by sulfuric acid would not be appropriate. What's reasoning here?

**Reply.** We now revised the text as follows. "While this set of optical parameters is typical for tropospheric sulfate aerosols that often occur in the neutralized form of ammonium sulfate [Wang et al., 2008], stratospheric volcanic sulfate aerosols may less neutralized (more acid) and thus have greater hygroscopicity than tropospheric sulfate aerosols [Russell et al., 1996]. Indeed, within 3-6 months after Pinatubo eruption, the effective radius (or equivalently, geometric mean radius assuming no change in geometric standard deviation) of stratospheric aerosols was shown an increase by a factor of 2-3 [Russell et al., 1996]. Wang et al. [2008] estimated that for the same amount of sulfate mass with the same size distribution at RH = 5%, ammonium sulfate, ammonium bisulfate, and sulfate acid particles can have 20-30% difference among the radiative forcing efficiencies (normalized to sulfate mass) at RH = 80%; this difference is primarily due to their different hygroscopic growth. To consider the uncertainty due to hygroscopicity and other factors (such as particle coagulation that are not included in the current GEOS-Chem simulation) in the estimate of particle size, we conducted sensitivity experiments to compute the forcing with different sets of sulfate optical properties with increasing particle geometric radius from 0.07  $\mu\text{m}$  to 0.19  $\mu\text{m}$  (Section 3.3)."

**Comment.** Page 26448, Line 3: Fig. 2c, i, o Page 26448, Line 6: C is not in Fig2m. Do you mean A? Page 26448, Line 20: How about the variance?

**Reply:** C is in the top-right corner of Figure 2m, but is surrounded by color contours. We have added "top-right corner of Figure 2m" in the revision. The normalized root-mean square deviation (with respect to OMI retrievals) is 1.25 for the simulation that uses OMI SO<sub>2</sub> data as initial condition, and 1.58 for the simulation that doesn't use OMI data. We added these information in the revision.

**Comment.** Page 26451, Line 23-24: It would be helpful to plot the modeled tropopause height in the figure. Also, how do we know the backscattering is because of SULFATE aerosol loading? Would it be possible that the signal we see in Fig. 6a (A,B) is caused by cirrus cloud and other types of aerosols? How well can CALIOP lidar distinguish aerosol particles from ice particles in cirrus?

**Reply:** The tropopause is now added in the figure 6. The following text is added: “On 14 August 2008, CALIOP data indicate a layer (marked as L1 and L2 in Figure 6a) with high loading of particles above the tropopause (black line in Figure 6b) over North America. Within this layer, the CALIOP measurements of depolarization ratio at 530 nm and backscattering attenuation at 1062 nm both show nearly zero values, and CALIOP layer classification algorithm indicate that this layer are dominated by aerosols with small fraction of cirrus (figures now shown).”

**Comment.** Page 26452, Line 2-5: C, D and G should be in Fig. 6ab. and Fig. 6cd. Also, I can't agree to the statement here. The observed vertical-integrated backscattering and modeled extinction in region C are much larger than those in regions A and B. How do we know the scattering/extinction was caused by deposition? As the authors mentioned later (Line 12, same page), the influence of non-volcanic aerosols (including aerosols other than sulfate) might dominate the signal below 10km in the CALIOP data. More evidence is needed to support this.

**Reply:** Yes, C, D, and G should be in Fig. 6, and the changes were made. We revised the relevant text as follows: “In addition, CALIOP images in Figure 6 a and b also indicate the likely deposition of volcanic sulfate aerosols in the middle-to-lower atmosphere such as over the south central US (marked as C and D in Figure 6a and 6b) and over northeast China (marked as G in Figure 6c and 6d); these “touch down” features are also seen in the similar contour plots showing the difference in GEOS-Chem simulation with and without considering volcanic aerosols (figures now shown), although non-volcanic aerosols from local source also contribute to the high loading of particles in regions C and G.”

**Comment.** Page 26454, Line 3-8: *A discussion about the differences of injection heights and circulation patterns (that is related to poleward transport) between the Pinatubo eruption and Kasatochi eruption would be helpful.*

**Reply:** we added more discussions on the poleward transport, especially regarding the Pinatubo eruptions, as follows. “The significant larger effect of Pinatubo eruption on climate is in part due to the following factors: (a) it ejected ~30 Tg of SO<sub>2</sub> up to 30 km above the sea level, most of which concentrated in 20 – 27 km altitude [McCormick et al., 1995]; (b) at this altitude range over the subtropics, intensity of planetary wave activity and on the phase of the quasi-biennial oscillation regulates the poleward transport, but were shown to be not effective in the N-hemisphere in June-July 1992; (c) the spread of SO<sub>2</sub> to the subtropics in S-hemisphere is found to be unexpectedly faster in June-July 1991, which is attributed to the abnormality of the planetary wave activates over the equator and southern subtropics (Trepte et al., 1993); (d) consequently, SO<sub>2</sub> amount was mainly located in the 30°N – 30°S in the first several months after Kasatochi eruption, and the larger and longer solar illumination in tropic and subtropics enhance the shortwave forcing of volcanic sulfate particles. In comparison, while the volcanic sulfate

aerosols from the Kasatochi eruption (52.1°N, 175.5°W) can reach the tropics within 1-2 month, most of them are concentrated in the mid- and high- latitudes (due to the westerly waves) during N-hemisphere fall-winter season (Fig. 8), and their amount (1-2 Tg) and their e-folding time (9 days) are all much less counterparts (30 Tg and ~1 year) of Pinatubo eruption [McCormick et al., 1995]. Consequently, Kasatochi eruption may have only affected the global radiative energy budget for about 100 days, and has much less impact on climate.”

**Comment.** *Page 26454, Line 18-21: The TOA forcing in GEOS-CHEM and surface forcing in Kravitz et al. (2012) are incomparable. Different models have different relationships between surface and TOA forcings. Is there surface forcing output in your model?*

**Reply:** we revised the text as the follows. “Nevertheless, because of sulfate particle is highly scattering (with the single scattering albedo value close to 1) in visible and other shortwave spectrum, our estimate of global forcing at the TOA and surface is very similar, with a global averages of  $-1.3 \text{ Wm}^{-2}$  in August and  $-0.7 \text{ Wm}^{-2}$  forcing in September, which appear consistent with results in Kravitz et al. (2012) showing a  $-2 \text{ Wm}^{-2}$  of zonal averages of forcing at the surface over the northern hemisphere in August and September.”

Minor comments:

*Page 26437, Line 11: an e-folding time . . . Page 26437, Line 20,22, and many more: Please check the special character. I can't see them in Preview (Mac). Page 26445, Line 21: set to 10km*

**Reply:** These changes were made, and special character is also checked.

*Page 26451, Line 20: The blue line in Fig.5 is hard to see. Please change it to black is possible.*

**Reply:** Changes were made.