Response to the Referee #1:

This paper described a straightforward sensitivity study of volcanic SO2 emission using the hemispheric CMAQ. It conducted two runs, with and without the volcanic SO2 emissions, and the results were mainly compared to surface sulfate measurements for year 2010. This surface sulfate-only verification is not sufficient for volcanic SO2 emissions since that sulfate concentration can be affected by other processes, such as wet scavenging. You may need to compare the modeled SO2 concentrations to surface/aircraft measurements and satellite retrievals. This manuscript did not mention the temporal variations of volcanic SO2 emission used here, and it likely used static emission rates. If so, the corresponding discussions are needed to justify the treatment since the volcanos unlikely erupted at constant rates for whole year of 2010.

Reply:

We appreciate the reviewer's constructive comments. To address the reviewer's concerns on model evaluation for SO_2, we have significantly expanded the model evaluation discussion to now also include (1) comparisons of column SO_2 predictions for both model cases – with and without volcanic degassing emissions, with OMI inferred SO_2 column distributions across our Northern Hemisphere domain (in Figure S1 in the Supplement), and (2) comparison of ambient SO_2 predictions with observations at CASTNET monitors in the U.S. (Table S1), and (3) comparison of model predicted SO_4^{2-} concentrations in rainwater, precipitation amounts, and SO_4^{2-} wet deposition amounts with observations from the NADP in the US and EANET in Asia (Table S2). Incorporation of volcanic degassing emissions in the model generally helps improve the performance metrics (slightly in some cases) in these model-observation comparisons and are detailed further in the revised manuscript discussion.

In terms of the temporal variation of SO_2 emission, due to lack of other information, we used a time-invariant emission rate. For clarity, we have revised Section 2.1 as follows with underlined for additional discussion.

"In this study, annual estimates of degassing volcanic SO_2 emissions by Carn et al. (2017) are incorporated into H-CMAQ. The estimated emission of SO_2 from the 50 volcanos within our northern hemisphere modeling domain (Figure 1b) is 12.7 Tg/yr. Considering the characteristics of degassing process from volcanoes and the lack of any other information to accurately specify its temporal variations, we use a constant SO_2 emission rate in H-CMAQ based on the annual SO_2 emission estimates by Carn et al. (2017)."

Please also see our replies to your specific comments.

Specified comments:

Page 4, line 24: "In this study, the entire year of 2010 was simulated". Why choose 2010 as the studied year, or is there any specific reason related to the 2010 volcano eruptions?

Reply:

As already stated in the introduction section, we aimed to understand the impact of SO_2 emissions from degassing volcanoes in this study and did not focus on any specific volcanic eruption event.

P2, L32-33: "The volcanic SO_2 emissions from degassing are relatively stable at 23.0±2.3 Tg-SO_2/yr during 2005-2015, and the highest amount was approximately 26 Tg-SO_2/yr in 2010."

P3, L5-7: "According to the comparison between the degassing and eruptive emissions, Carn et al. (2017) estimated that during 2005-2015, volcanic activity contributed about 23 Tg/yr of SO_2 due from degassing while eruptive SO_2 emissions ranged from 0.2 to 10 Tg/yr of SO_2. Therefore, understanding the behavior of persistent degassing SO_2 emissions and its contributions to airborne SO_4^{2-} levels is important."

Based on these reasons, we choose 2010 as the studied year. To address the reviewer's comment, we stated the reason for selection of 2010 explicitly in page 4, lines 23-26:

"In this study, the entire year of 2010 was simulated to analyze the volcanic emission impacts over the Northern Hemisphere, as the SO_2 emissions from degassing during the 2005-2015 period were highest in 2010. Also note that emission estimates presented in Carn et al. (2017) suggest that degassing dominate over eruptive SO_2 emissions during the same time period."

Page 5, line 7. So the volcano emissions have no plume rise, right? If so, why?

Reply:

Yes, plume rise for volcanic degassing emissions were not treated in this study, and this was because the characteristics of the degassing process. As noted earlier, we aimed to analyze the impact of SO_2 emissions from persistent degassing volcanoes and did not focus on any specific volcanic eruption. As such, a plume rise calculation would require characterization of buoyancy of the degassing plume and thus need information (either observations or estimates) on heat content of the degassing plume. Since these are not readily available, volcanic degassing SO_2 emissions were allocated into the model vertical layer corresponding to the volcano's altitude. In this configuration, the 108 km grid spacing used in CMAQ does not allow the model to adequately resolve localized terrain peaks such as volcanoes and assigning their emissions to the first model layer would not account for the fact that in reality these emissions typically occur above the mixed layer. Therefore, the vertical layer to which volcanic degassing emissions were assigned was determined by first calculating the difference between the altitude of a given volcano (this was listed in Table 1) and the CMAQ terrain height for the cell in which it is located, and then determining the vertical CMAQ layer corresponding to this difference.

Page 6, section 3.1. As commented above, the verification with only surface sulfate is insufficient. Even with the coarse resolution, the SO2 comparisons are still preferred. Or, you can use a high-resolution regional CMAQ to study certain region for a certain period.

Reply:

We agree that comparisons with SO_2 measurements would enable more direct assessments of the impacts of including volcanic SO_2 emissions.

CASTNET has been routinely monitoring SO_2 concentration. To address the reviewer's comment, we evaluated SO_2 predictions against all available observations from CASTNET. The evaluation results are now newly presented in Table S1 in the Supplement along with brief discussions of these results in the main text.

Table S1. Statistical analysis of modeled SO_2 concentration against CASTNET observations.

	N	Mean		R	NMB	NME
		Obs.	Model			
CASTNET						
−original H-CMAQ	**4216**	**1.69**	**2.81**	**0.57*****	**+66.1%**	**94.7%**
−incorporation of volcanic emissions			**2.83**	**0.58*****	**+67.4%**	**94.9%**

Note: The unit of mean for observations and simulations is ppbv. Significance levels by Students' t-test for correlation coefficients between observations and simulations are remarked as $^{*}p < 0.05$, $^{**}p < 0.01$, and $^{***}p < 0.001$, and lack of a mark indicates no significance.

Because SO_2 concentration did not change in the case of the incorporation of volcanic emissions, this result showed that SO_2 from volcanic sources was fully oxidized to SO_4^{2-} and there was no direct transport of SO_2 itself. To indicate this point, we have explicitly stated within the modeling evaluation at specific site of Florida in Section 3.3, and it was revised as follows.

"CASTNET also provides measurements of weekly-average SO_2 mixing ratios, which are used to evaluate the model's performance for SO_2 prediction, as shown in Table S1. In addition to domain-mean evaluation, we evaluated SO_2 at the EVE419 site in Florida. At this site, the observed annual mean was 0.61 ppbv whereas base H-CMAQ and H-CMAQ with volcanic SO_2 emissions both showed 0.77 ppbv."

Page 8-9, section 3.3. The volcanic SO2 impact is only shown at two surface sites and for sulfate only, which is insufficient.

Reply:

We appreciate this suggestion. It should be noted that we have evaluated model performance for airborne SO_4^{2-} concentration relative to all locations available from the EANET, CASTNET, and IMPROVE networks as listed in Table 2. Within this comparison, total of 1167, 4216, and 18844 observation-prediction data pairs have been evaluated. In addition, according to your comment, we have also evaluated model performance for ambient SO_2 concentration for CASTNET observation as listed in Table S1.

Taking into account this comment, we have re-examined the analysis in Figure 6, and have revised it to include analysis at an additional observation site in the Virgin Islands. We have revised Figure 6 to show detailed analysis over three specific sites that were most affected by the incorporation of persistent volcanic degassing emissions. The updated discussion in Section 3.3 of the revised manuscript is as follows.

"A second location-specific analysis were conducted at sites located in the Virgin Island and in the state of Florida. As seen in the monthly-average spatial distribution patterns (Fig. 5), the influence of volcanic activity of Soufriere Hills located in Montserrat are more prominent during winter. The nearest observational sites from Soufriere Hills are the IMPROVE site VIIS1, and the southernmost measurement location in Florida (see, Fig. 4) is the CASTNET site

EVE419 and comparison of modeled values with observations at these locations are shown in Fig. 6 (b) and (c). At VIIS1 (Fig. 6 (b)), the base H-CMAQ simulation showed invariant concentrations throughout the year with an annual average value of 0.86 μg/m^3. The statistical scores of R showed scattered correspondence between model and observation. NMB showed negative bias because the base H-CMAQ did not capture the episodical peak concentration. By including the degassing volcanic SO_2 emissions, the model exhibited improved skill in representing the episodic peaks. The statistical scores showed that the average concentration was 1.75 μg/m^3, and NMB showed +44.6%, and R was dramatically improved to 0.72. The deterioration in the value of NME was found, because of the continuous model overestimation. As also suggested the case in Hawaii, further refinement on emission treatments is required. At EVE419 (Fig. 6 (c)), the seasonal pattern with summer minima was captured by H-CMAQ but underestimated throughout the year. Increased concentrations through the incorporation of volcanic emissions are noted during January, late April to early May and December. The increases during late April to early May were not seen from the monthly-averaged spatial distribution (Fig. 5); hence these could be the episodic long-range transport by southeasterly winds. Because of these increased concentrations, all statistical scores of R, NMB, NME showed improvement compared to the base H-CMAQ."