April 2nd, 2021

Dear Editor Eliza Harris for *Atmospheric Chemistry and Physics*

We are pleased to submit our revised manuscript for our manuscript abd4610 entitled "*Isotopic constraints on atmospheric sulfate formation pathways in the Mt. Everest region, southern Tibetan Plateau*" for your consideration for publication in *ACP*.

We have carefully reviewed and considered the two reviewer's comments, and we are grateful for the valuable comments, and we mainly agree with the reviews. The relevant changes except mentioned in the response letters are summarized as follows.

1. Change the font of delta and DELTA to Italic throughout the manuscript.
2. Adding a space between value and %(‰) throughout the manuscript.
2. Change the D17O value of O3 + S(IV) to 6.4±0.3‰ according to the recent our study surveys (see Ishino et al 2021 and Hattori et al 2021), also the related values throughout the manuscript and the supplementary tables were corrected accordingly.
3. "10% of [Fe]_dust and 1% of [Fe]_anthro" in section 2.3 was changed to "1% of [Fe]_dust and 10% of [Fe]_anthro". This is just a mistake.
4. Additional three references are added.
(1) Hattori, S., Iizuka, Y., Alexander, B., Ishino, S., Fujita, K., Zhai, S., Sherwen, T., Oshima, N., Uemura, R., Yamada, A., Suzuki, N., Matoba, S., Tsuruta, A., Savarino, J., and Yoshida, N.: Isotopic evidence for acidity-driven enhancement of sulfate formation after SO2 emission control, Sci. Adv., 7, http://doi.org/10.1126/sciadv.abd4610, In Press, 2021.
(2) Ishino, S., Hattori, S., Legrand, M., Chen, Q., Alexander, B., Shao, J., Huang, J., Jaegle, L., Jourdain, B., and Preunkert, S.: Regional characteristics of atmospheric sulfate formation in East Antarctica imprinted on 17O-excess signature, J. Geophys. Res. Atmos., 126, e2020JD033583, https://doi.org/10.1029/2020JD033583, 2021.
(3) Lin, M., Wang, K., Kang, S., Li, Y., Fan, Z., and Thiemens, M. H.: Isotopic signatures of stratospheric air at the himalayas and beyond, Sci. Bull., 66, 323-326, https://doi.org/10.1016/j.scib.2020.11.005, 2021.
5. Add two items into the Acknowledgement, i.e., "M.L. acknowledges financial supports from the JSPS Postdoctoral Fellowship and Guangdong Pearl River Talents Program (2019QN01L150)." And "We acknowledge two anonymous referees for their constructive comments."
6. The date error and references in Fig. 5 were corrected.

We look forward to hearing from you at your earliest convenience.

Yours sincerely,

Kun Wang and Shohei Hattori on behalf of all co-authors

Black: Reviewers' comments

Red: Response

Blue: Action

Response to Reviewer #1

This study presents two-season's triple oxygen isotope data for atmospheric secondary sulfate (SAS) from the Mt. Everest region or Tibet Plateau (TP), a geographic-climatically unique area. The data reveal an unexpected seasonal trend with the monsoon season having higher $\Delta^{17}O$-SAS value than the pre-monsoon season. The data are consistent with the observed neutral to alkaline rainwater in the region, indicating an enhanced O_3 pathway in S(IV) oxidation. I have some comments on lab procedures and the explanations based on simple correlations vs. GEOS-CHEM modeling.

Response: We thank the reviewer for his/her time and the professional review work on our manuscript. These comments and suggestions are very valuable and helpful for revising and improving our manuscript. We have considered these comments and suggestions very carefully, and the following are the detailed point-by-point responses.

Three major issues:

1. The laboratory procedure has a discrepancy. I could not connect the heating of solid precipitates to remove organics and the IC-column separation and precipitation of Ag_2SO_4 (See details in my specific comments below).

Response: We thank the reviewer for this comment and sorry that we did not present this part in a clear way, which would lead to a misunderstanding. An Ag_2SO_4 hydrolysis method was applied to do the isotopic measurements in our study. After dried, the Ag_2SO_4 was found to be mixed with some black/brown materials, and the yield of O_2 is also much lower than normal. We think this might be due to the larger amounts of organics in environmental samples. Thus, we used a heating method to remove the organics (Xie et al., 2016). Firstly, we evaporated the samples to get the solid precipitates, and then heated the precipitates in an oven at the temperature of 450℃ for 2 h. That is, the sulfate was not in Ba form before and after heating. After that, the samples in solid form were ultrasonic dissolved prior to using ion chromatography to separate sulfate from other ions and trace organics. Also, it has been demonstrated that the effect of oxygen isotopic exchange during the heating (T = 450℃) is negligible (Xie et al., 2016).

Action: The laboratory procedure for the heating method was re-organized in Lines 161-173: "In this study, a method of Ag_2SO_4 pyrolysis was applied to do the oxygen isotopic measurements. However, after dried in a vacuum centrifuge, the Ag_2SO_4 was found to be mixed with some black/brown particles, and also the yield of O_2 after Ag_2SO_4 pyrolysis is much lower than normal. Given all of this, we suggested that the environmental samples might contain much organics, which would reduce the precision and accuracy of measurements. Thus, we did additional removal of organics to improve the yield of O_2 by applying a high-temperature heating method (Xie et al., 2016). Briefly, before heating in the oven, the samples were evaporated to get solid precipitates, and then the precipitates were heated at 450 °C for 2 h prior to isotopic measurements. After heating, the organic materials were largely removed as detected by the ion chromatography, and the yield of O_2 was improved significantly. As demonstrated by Xie et al. (2016), the effect of oxygen isotopic exchange during the heating is negligible. After the removal of organic materials, the oxygen isotopic compositions of sulfate were measured using a pyrolysis technique in a continuous flow system as introduced by Savarino et al. (2001) with further modifications described in several later studies (Geng et al., 2013; Schauer et al., 2012)". which can be also seen in the response for specific comments below.

2. Many ad hoc explanations of the observed data seem to be unnecessary if you already have an isotopically enhanced GEOS-CHEM model to account for the $\Delta^{17}O$-SAS. Of course, after the modeling results, you can highlight the major factors. But any discussion on the controlling factors should not be isolated from the modeling—which seems to me is the case in the manuscript.

Response: Thanks a lot for the comment. One of the important purposes of this work is to report a new dataset of $\varDelta^{17}O$ in atmospheric sulfate observed from the Mt. Everest region, a climatically important region in the world. The comparisons with other regions (i.e., other mid-latitude sites and polar regions) as well as the related discussion revealed that the $\varDelta^{17}O$ data observed from the modern atmospheric sulfate in the Mt. Everest region is relatively high and show unique seasonal characteristics. Based on these comparisons and discussions, we tried to find the related mechanisms behind them. The high $\varDelta^{17}O$ values clearly indicated the importance of S(IV) oxidation by O_3. However, the seasonality between $\varDelta^{17}O$ and O_3 levels present opposite trend, which indicated that the O_3 level is not the ultimate determinant of high $\varDelta^{17}O$ values as well as its seasonality. Interestingly, the atmospheric humidity shows a good trend with $\varDelta^{17}O$. Thus, considering the frequently occurred dust storms, we infer that the atmospheric acidity as well the heterogeneous reactions (e.g., SO_2 oxidation by O_3) on dust surface (which can be favored by the atmospheric humidity) might play important roles in promoting the O_3 oxidation pathway. In this case, we conducted the model simulations

(GEOS-Chem) to test these assumptions by comparing three model set ups with/without consideration of cloud water pH and heterogeneous oxidation of SO_2 by O_3. The results show that the pH dose play an important role in the high $\Delta^{17}O$ values, but the heterogeneous O_3 oxidation seems not so important. Based on the analysis above, we concluded that the atmospheric acidity (i.e., cloud water pH) should play an important role in controlling sulfate formation pathways particularly for dust-rich environments. Although the model predicted minor relative contribution of heterogeneous oxidation of SO_2 by O_3 (which might be due to the model's uncertainties), we suggest it a potential important pathway of sulfate formation, which might explain the seasonality of $\Delta^{17}O$ values.

Overall, we treat the model results as necessary addition to this work, which would be based on the analysis and discussions as listed above.

3. The data have NOT ruled out or considered the possibility that the SAS collected in TP was already formed in the atmosphere in South Asia. That is to say that a portion or maybe a dominant fraction of the SAS collected in the field station in TP is not formed locally but transported there in long-distance. If true, local rainwater pH should not have played a role in the elevated $\Delta^{17}O$-SAS value. This requires the GEOS-CHEM modeling results to clarify.

Response: We agree with the reviewer for this insight comments. Yes, based on current data, we cannot exclude the fraction of SAS formed during the long-range transport which would/should contribute to the atmospheric sulfate level in the Mt. Everest region. The description in lines 419-424, i.e., "Indeed, the pH in fog, rain and snow in the TP region were reported to be high, respectively, pH = 6.4, 6.2, and 5.96 ± 0.54 (Wang et al., 2019; Kang et al., 2002), and the modeled cloud water pH in the Mt. Everest region and the South Asia showed approximately pH = 6 (Shah et al., 2020; Pye et al., 2020). Such high pH condition favors S(IV) oxidation by O_3. We suggest that the frequently occurring dust storms, not only in the Mt. Everest region but also in South Asia (Fig. 3 & Prospero et al., 2002), are likely to play an important role for promoting the S(IV) oxidation by O_3 through the high cloud water pH conditions" indicated the possibility of sulfate formation during the long-range transport. Besides, the drive data sets of GEOS-Chem also include transportation, thus we have considered both source and transportation's contributions to the sulfate level at our sampling site throughout the manuscript. In order to make it more clearly, we added some related description in lines 424-425 and 430-431.

Action: Lines 424-425: "That is, the sulfate produced in South Asia and during the long-range transport should contribute significantly to the observed sulfate in the Mt.

Everest region".

Lines 430-431: "Note that the drive data sets of the model also include transportation, thus both source and transportation's contributions to the observed sulfate are considered by the model".

Some of the specifics while I was reading over the manuscript.

1. Abstract: The scientific problem is not specifically expressed. Something "poorly characterized" is not a reason for research because almost everything is "poorly characterized" to an unspecified standard.

Response: We agree with the reviewer and thank him/her for this valuable comment. At least, in the Abstract section, we should emphasize the reason why we will conduct this research in the Tibetan Plateau region. This would make this study more logical.

Action: Lines 21-23: "as an important component of atmospheric particulates, sulfate has been thought to exert a profound impact on the environment and climate, while the environmental and climatic change in the TP region has attracted wide attentions".

2. 58-59: "It contains the largest land ice masses outside the poles and supplies water for more than one billion people (Immerzeel et al., 2010)." Please state the relevance of this fact to your study.

Response: We are sorry that we did not present the relevance between the snow/ice melting and sulfate level in the Tibetan Plateau in a clear way. Actually, one recent study has pointed out that the sulfate play an important role in both enhancing single scattering albedo and altering the absorption properties of carbonaceous aerosols (e.g., black carbon and brown carbon) (Lim et al., 2018), while black carbon deposited on the glacier can accelerate snow/ice melting (Kang et al., 2020).

Action: Lines 70-74: "It has been demonstrated that the sulfate play an important role in both enhancing single scattering albedo and altering the absorption properties of carbonaceous aerosols (e.g., black carbon (BC) and brown carbon) (Lim et al., 2018). As a product of incomplete combustion of fossil fuel and biomass, BC deposited in the TP region has attracted much attention, since it can accelerate snow/glacier melting, although the magnitude of this effect is uncertain (Kang et al., 2020 and references therein)".

3. 66: "a deep understanding" is vague in its meaning. Are you trying to link the secondary sulfate to the recent weakening of temperature seasonality in TP? A testable hypothesis that is related to sulfate formation pathway in TP would benefit the presentation of your research.

Response: Corrected accordingly.

Action: Lines 76-80: “In sum, the atmospheric sulfate not only have an influence on the radiative forcing via its climatic role, to some extent, it could also affect the snow/glacier melting indirectly and further concern the supplies of fresh water to larger amount of people in Asia. Thus, it is meaningful to investigate the formation mechanisms of sulfate in the Tibetan Plateau region for an accurate assessment of the regional climatic change as well as cryospheric extent variation in this region”.

4. 100-105: The need to do the SO4 collection and isotope analysis is tenuous. “No observational studies” itself is not a good reason. What I am expecting to be shown is the link of sulfate formation pathways to specific climate, environmental, or meteorological issues in TP. For example, if indeed Indian subcontinent is supplying atmospheric pollutants to TP, what should we expect to see in the Δ^{17}O of atmospheric sulfate collected in that remote site on the northern slope of Mt. Everest? If there is an alternative source, what different Δ^{17}O-SAS are we expecting?

Response: We thank the reviewer for this valuable comment and suggestion. Yes, we agree that a clear statement of novelty is really important for this work. In order to further elucidate the motivation of this work, we added some hypotheses and expectations in the Introduction section.

Action: Lines 119-124: “Considering the specific climate and environment in the Tibetan Plateau region, especially the Mt. Everest region, e.g., low temperature, high elevation, and frequently occurred dust storms, we were wondering if the $\mathit{\Delta}^{17}$O characteristics in sulfate would differ from other regions. As a result, we can infer the formation mechanisms of atmospheric sulfate in this unique region. We also want to know if the sulfate level in this remote site is influenced by the long-range cross-border transport. With these expectations, here, for the first time, we present relatively long term $\mathit{\Delta}^{17}$O observations in atmospheric sulfate as well as its concentrations in this region”.

5. 140-142: Before briefing on the heat treatment of “sample precipitates”, you omitted the precipitation step (e.g., did you evaporate and acidify the solution before adding $BaCl_2$ solution?). If this method is used, why was the method of Savarino et al (2001) and Geng et al (2013) developed specifically for smaller samples also used? If precipitates were heated at 450°C, how did you check organic matter by ion chromatography? Did you re-dissolve the precipitates? It is not mentioned in text. If the SO4 is separated and purified by IC, why is the heating solid precipitates necessary?

Response: We thank for this comment and sorry for the vague expressions. A method of Ag_2SO_4 pyrolysis was applied to measure the oxygen isotopes in our study. However, after drying Ag_2SO_4 samples, we found some black/brown particles mixed with Ag_2SO_4, and also the yield of O_2 after Ag_2SO_4 hydrolysis was found to be much lower than normal. Given these factors, we suggested that the environmental samples might contain much organics, which would reduce the precision and accuracy of oxygen isotopic measurements. Thus, we did additional removal of these organics to improve the yield of O_2 from the samples. Here, we applied a heating method as indicated by (Xie et al., 2016) to do the removal organics. Before heating in the oven, the samples was firstly evaporated to get the solid precipitates, and then subjected to be heated at 450℃ for 2 h. After that, the samples were ultrasonic dissolved again prior to separate sulfate from other ions and trace organics by using ion chromatography. It turned out that the yield of O_2 has indeed increased significantly after the heating method. In order to make the expression more clear, we made a revision accordingly.

Action: We made a correction in Lines 161-173: "In this study, a method of Ag_2SO_4 pyrolysis was applied to do the oxygen isotopic measurements. However, after dried in a vacuum centrifuge, the Ag_2SO_4 were found to be mixed with some black/brown particles, and also the yield of O_2 after Ag_2SO_4 pyrolysis is much lower than normal. Given all of this, we suggested that the environmental samples might contain much organics, which would reduce the precision and accuracy of measurements. Thus, we did additional removal of organics to improve the yield of O_2 by applying a high-temperature heating method (Xie et al., 2016). Briefly, before heating in the oven, the samples were evaporated to get solid precipitates, and then the precipitates were heated at 450 °C for 2 h prior to isotopic measurements. After heating, the organic materials were largely removed as detected by the ion chromatography, and the yield of O_2 was improved significantly. As demonstrated by Xie et al. (2016), the effect of oxygen isotopic exchange during the heating is negligible. After the removal of organic materials, the oxygen isotopic compositions of sulfate were measured using a pyrolysis technique in a continuous flow system as introduced by Savarino et al. (2001) with further modifications described in several later studies (Geng et al., 2013; Schauer et al., 2012)".

6. Section 2.4.1: Please add the relevance of black carbon data to the SO4 story in TP. It is not obvious to me.

Response: Thank you for your suggestion. We revised accordingly.

Action: Lines 231-233: "Since the BC is mainly produced from the incomplete combustion of fossil fuel and biomass, which are also important sources of SO2, thus in order to investigate the potential sources of sulfate, the BC data is also presented in this study".

7. Line 247-261: There is a distinct possibility that the $CaSO_4$ (i.e., terrigenous sulfate has a positive $\Delta^{17}O$ value) because a large portion of the sulfate in the arid surface salts could come from atmospheric deposition. Ignoring this possibility would increase the estimated $\Delta^{17}O$ of SAS, although quantitatively it is not a big deal (i.e., ~6%).

Response: We agree with the reviewer that a larger portion of sulfate contained in the arid surface soil could come from the atmospheric deposition. Thus it is very important to choose a proper *k* value to do the calculation. The $k = 0.18$ applied in this study has been widely used to do such estimation in previous studies (Kunasek et al., 2010; Lin et al., 2020; Patris et al., 2002), although the uncertainty remains. As suggested by the reviewer, the increase of $\Delta^{17}O$ values should be very limited due to the minor portion of terrigenous sulfate in our study (~ 6.3%), and would not change our discussions and conclusions. But for the sake of preciseness, we added the related description, and gave some analyses in a quantitative way.

Action: Lines 298-306: “Note that, although the terrigenous sulfate has been generally considered to possess zero $\Delta^{17}O$ signal (Dominguez et al., 2008; Lee and Thiemens, 2001), some previous studies observed positive $\Delta^{17}O$ values in sulfate mineral deposits formed in arid surface environments (e.g., central Namib Desert and Antarctic dry-valley soils) due to the atmospheric deposition (Bao et al., 2000b; Bao et al., 2000a). If this is also true for the Mt. Everest region, the estimated $\Delta^{17}O(SO_4^{2-})_{SAS}$ would be overestimated. However, the overestimation should be very limited due to the small portion of terrigenous sulfate (6.3 ± 3.0 %) in our study. As described in section 3.2, the average of $\Delta^{17}O(SO_4^{2-})_{SAS}$ in the Mt. Everest region is 1.7 ± 0.5 ‰. If we assume that half of the terrigenous sulfate originates from the oxidation processes in the atmosphere, the estimated $\Delta^{17}O(SO_4^{2-})_{SAS}$ would be ~1.6 ‰, which is slightly lower than the reported data in our study. Thus, the potential overestimation would not change our discussions and conclusions”.

8. 341-352: Do you really need this simple estimation of minimum and maximum O_3 path fraction involved in SAS formation? I am afraid that such a discussion does not add to the story. I thought you have already run modeling that incorporates meteorological data and atmospheric chemistry (isotope-enabled) and transport, i.e., GEOS-CHEM. The model should give you more accurate prediction of the respective fractions of O_3 pathway during pre-monsoon and monsoon reasons. The simple minimum and maximum estimations ignore other pathways such as Mn-Fe catalyzed cloud-water O_2 oxidation and mineral-surface heterogeneous oxidation.

Response: We thank the reviewer for this kind suggestion. The main purpose of calculating minimum and maximum contribution of oxidation by O_3 is to examine the relative importance of S(IV) + O_3 oxidation pathway based on our measured $\Delta^{17}O$ data, which can be used to compare with the modeled results. We think combination of the measurement and modeling results would make the analyses more convincible.

9. 359: Please change “SR” to “solar radiation” because you only used the abbreviation twice (another in line 216) in the entire paper.

Response: Thanks for the reasonable comments. Now it was corrected.

Action: Lines 248, 402: The abbreviation of “solar radiation” (i.e., SR) was deleted in line 248, and the “SR” was changed to “solar radiation” in line 402.

10. 360: Change “hypothesize” to “explain”.

Response: Corrected.

Action: Line 404: The “hypothesize” was changed to “explain” now.

11. 363: Change “hypothesis” to “explanation”.

Response: Corrected.

Action: Line 407: The “hypothesis” was changed to “explanation” now.

12. 375-381: Now, my understanding here is that the GEOS-CHEM atmospheric chemistry-transport model (isotopically enabled) could not offer a prediction on the SAS $\Delta^{17}O$ value for the pre-monsoon and monsoon reasons. Correct? If true, what’s the point of doing the modeling?

Response: As shown in Fig. 7 in the manuscript, the model could offer a prediction on the $\Delta^{17}O(SO_4^{2-})_{SAS}$ values for different seasons, but the seasonal variation in the $\Delta^{17}O(SO_4^{2-})_{SAS}$ values is not very clear. Also, the modeling results tell us that the pH play an important role in the oxidation of S(IV) by O_3 and predicted comparable $\Delta^{17}O(SO_4^{2-})_{SAS}$ values with our measurements within the entire sampling period. Although we gave some discussions on the potential relevance between $\Delta^{17}O(SO_4^{2-})_{SAS}$ seasonality and RH, a more convincing conclusion need further investigation.

13. 398-406: There could be no major problem with an acidic cloud-water in these cited models. The problem could be the poor parameterization of heterogeneous oxidation of SO_2 or aqueous S(IV) on dust surface.

Response: In section 3.3.3, we mainly focus on the impact of atmospheric acidity on $\Delta^{17}O$, and found that the pH plays an important role in determining the relatively high $\Delta^{17}O(SO_4^{2-})$ in the Mt. Everest region. Based on our observation and modeling results, we suggest that the lack of full consideration of pH in previous models should be one of the factors influencing the accuracy of prediction. One example is that, when the pH is fixed to 4.5 for Northern Hemisphere in the model, the modeled $\Delta^{17}O$ is too low to explain the observed high $\Delta^{17}O$ values for some specific regions (Sofen et al., 2011), which potentially indicated the importance of atmospheric acidity in such kind of modeling work. Additionally, our modeling results do not support the importance of heterogeneous oxidation on dust surface, which might be due to the poor parameterization in the model, so we give additional discussions about heterogeneous oxidation independent of modeling results in section 3.3.4. That is why we pointed out the pH, but not heterogeneous oxidation when we mentioned these cited models.

14. Section 3.3.4.: Your explanation of the higher $\Delta^{17}O$ during monsoon season is due to the enhanced moisture on alkaline dust surface which increases the overall fraction of the S(IV) + O_3 reaction pathway than during the pre-monsoon when the air is relatively drier. If so, it is worth of another repeat here. Please also link and explain the opposite trends in the $\Delta^{17}O$ between NO3- and SO4 in this framework (it can be explained).

Response: We thank the reviewer for the reasonable comments. Now we added the related description in lines 458-463 and 465-468 to explain the seasonal trend by considering the effect of RH on heterogeneous SO_2 oxidation by O_3 on the alkaline dust surface, while for the opposite seasonal trend in $\Delta^{17}O$ between sulfate and nitrate, we gave some explanations in lines 471-474.

Action: Lines 458-463: "The adsorption of SO_2 is thought to be one of the important rate-determining steps for SO_2 oxidation on dust surface suggesting that the heterogeneous reactions on dust surface are very dependent on the SO_2 uptake coefficient, which is further determined by RH (Bauer and Koch, 2005; Li et al., 2006). By improving the hygroscopic property of atmospheric aerosols and result in increasing water content and surface areas, RH can promote SO_2 oxidation to occur on the surface of these aerosols. Thus, RH should play a crucial role in the heterogeneous oxidation process".

Lines 465-468: "Thus, the relatively high RH during the monsoon season in the Mt. Everest region would facilitate the heterogeneous SO_2 oxidation by O_3 on the alkaline dust surface, which increases the overall fraction of the S(IV) + O_3 reaction and results in relatively higher $\Delta^{17}O(SO_4^{2-})_{SAS}$ values in monsoon season than that in the pre-monsoon season".

Lines 471-474: "One should also note that, although the seasonal trends in $\Delta^{17}O$ for both nitrate and sulfate are result from the relative importance of S(IV) + O_3 pathway, the mechanism behind it is different. For nitrate, the main factor controlling the $\Delta^{17}O$ seasonality is the O_3 concentrations, while for sulfate, RH might play a more important role in determining the $\Delta^{17}O$ seasonality as discussed above".

15. Importantly, if none of these surface heterogeneous reaction, nature and flux of dust particles, or the enhancing effect of moisture was incorporated in the GEOS-CHEM model, I wonder why you need to do the GEOS-CHEM modeling at all. On the other hand, if the model can do the job in explaining the observed $\Delta^{17}O$ data, why would you need to single out some of the factors and do the simple minimum/maximum estimates and interpretation? That is something that confuses me.

Response: We appreciate the reviewer for this comment. By considering the cloud water pH, the model predicted comparable $\Delta^{17}O$ values with our observations. Thus, the most significant point that revealed by our modeling work is the importance of atmospheric acidity in the formation of atmospheric sulfate as observed in the Mt. Everest region. So, we think the modeling work is necessary in this study. As described in section 2.3, the GEOS-Chem simulations in our work incorporate the heterogeneous oxidation on dust aerosols by O_3. Although the model has predicted minor relative contribution of heterogeneous reaction, considering the poor parameterization of heterogeneous oxidation in the model, we gave a further discussion on the potential importance of heterogeneous oxidation of S(IV) by O_3. Since the main content of this paper is to investigate the possible formation mechanisms of atmospheric sulfate in the Mt. Everest region based on our measurements, thus we firstly did some analyses and discussions including the relevance between $\Delta^{17}O$ and meteorological data, and simple estimations. Based on these analyses and discussions, we induced that the atmospheric acidity and/or heterogeneous reactions might play an important role in the sulfate formation. Thus, finally we used the modeling results to verify the potential importance of pH and heterogeneous reactions. So, the analyses based on the observed $\Delta^{17}O$ prior to the modeling part are necessary. To make it more logical between contexts, we add one sentence in section 3.3.3.

Action: Lines 445-447: "The above results show that, the consideration of variable cloud water pH significantly improved the model's ability to simulate the $\Delta^{17}O(SO_4^{2-})_{SAS}$ in the Mt. Everest region, which indicate the importance of atmospheric acidity in favouring the oxidation of S(IV) by O_3 in this region".

Summary: I think the $\Delta^{17}O$-SO4 data are solid, unique, interesting, and can be explained by known reaction mechanisms. With proper cleaning-up and reorganization of the flow of the manuscript, the data may reveal important unknown parameters we need to calibrate and therefore quantitatively improve the isotope prediction of the GEOS-CHEM model.

Response: We thank the reviewer for his/her positive attitude towards our work and also for the valuable comments. According to these comments, we have revised the manuscript carefully.

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