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 Δ^{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 pyrolysis method was applied for the oxygen isotopic measurements in our study. As demonstrated by our preliminary tests, the yield of O_2 produced from the Ag_2SO_4 (mixed with black/brown particles inspected by eyes) pyrolysis was much lower than expected indicating the Ag_2SO_4 might be contaminated by organic materials. Thus, we did additional removal of organics to improve the yield of O_2 by applying a high-temperature heating method, which would not result in measurable oxygen isotopic exchanges (Xie et al., 2016). In brief, an aliquot of extracted solution was dried in a freeze dryer, and then the dried salts were further heated at 450 °C for 2 h following Xie et al., (2016). After heating, the dried salts were re-dissolved by deionized water prior to separation of sulfate from other anions via ion chromatography. In order to make the description more clear and logic, we made a revision accordingly.

Action: The laboratory procedure for the heating method was re-organized in Lines 162-170: "In this study, oxygen isotopic compositions were determined by an Ag_2SO_4

pyrolysis method as introduced by Savarino et al. (2001) with further modifications described in several later studies (Geng et al., 2013; Schauer et al., 2012). An aliquot of extracted solution (containing ~1.5 μmol of sulfate) was dried in a freeze dryer (T = 45 °C) to concentrate water-soluble ions. Dried salts were further heated at 450 °C for 2 h to remove organics following Xie et al., (2016). This heating procedure, which would not lead to measurable oxygen isotopic exchanges (Xie et al., 2016), is important in our study because organics contaminants may reduce the precision and accuracy of $\Delta^{17}O$ measurements via consuming produced O_2 during the Ag_2SO_4 pyrolysis state. Our preliminary tests showed that the yield of O_2 produced from the Ag_2SO_4 pyrolysis was much lower (i.e., ~50 %) than expected (higher than 90 %) when the Ag_2SO_4 was contaminated by organics (black/brown particles inspected by eyes). After heating, the dried salts were re-dissolved by 10 mL of deionized water".

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 $\Delta^{17}O$ 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 $\Delta^{17}O$ 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 $\Delta^{17}O$ values clearly indicate the importance of S(IV) oxidation by O_3. However, the seasonality between $\Delta^{17}O$ and O_3 levels present opposite trend, which indicates that the O_3 level is not the ultimate determinant of high $\Delta^{17}O$ values as well as its seasonality. Interestingly, the atmospheric humidity shows a good co-variation with $\Delta^{17}O$. Thus, considering the frequently occurred dust storms, we infer that the atmospheric acidity as well as the heterogeneous reactions (e.g., SO_2 oxidation by O_3) on dust surface which favors high humidity conditions might play important roles in promoting the O_3 oxidation pathway. To test our assumptions, we conducted the model simulations (GEOS-Chem) with 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 cloud water pH dose play an important role in the high $\Delta^{17}O$ values, but the contribution of heterogeneous O_3 oxidation on dust surface was estimated to be less than 5 %. 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 consider that the observation data is a major significant of this study and the modeling results are to support the interpretation of our observation.

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 comment. 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 420-425, 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. Meanwhile, since the GEOS-Chem was set to simulate sulfate produced via each formation pathway as different tracers that are transported, the estimated $\Delta^{17}O$ also includes sulfate produced during the transport. Thus, we have considered not only sulfate produced around our sampling site but also sulfate produced during the transport from source regions throughout the manuscript. In order to make it more clearly, we added some related description in lines 425-426 and 431-432.

Action: Lines 425-426: "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 431-433: "Note that the sulfate simulated by the chemical transport model

includes both local and distant sources, thus the estimated $\Delta^{17}O(SO_4^{2-})_{SAS}$ also includes sulfate produced during the transport".

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 conducted this research in the Tibetan Plateau region. This would make this study more logical.

Action: Lines 22-24: "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 attention".

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 71-75: "It has been demonstrated that sulfate plays 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: We added the explanation about the importance of sulfate formation specifically in the TP region.

Action: Lines 77-81: "Thus, atmospheric sulfate not only has an influence on radiative

forcing of the climate, to some extent, it could also affect the snow/glacier melting indirectly and further impact the supplies of fresh water to larger numbers 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 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 $\Delta^{17}O$ of atmospheric sulfate collected in that remote site on the northern slope of Mt. Everest? If there is an alternative source, what different $\Delta^{17}O$-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 120-125: "Considering the specific climate and environment in the Tibetan Plateau region, especially the Mt. Everest region, e.g., low temperature, high elevation, and frequently occurring dust storms, we investigate if the $\Delta^{17}O$ characteristics in sulfate are different from other regions. As a result, we can infer the formation mechanisms of atmospheric sulfate in this unique region. We also examine if the sulfate level in this remote site is influenced by long-range cross-border transport. With these expectations, here, for the first time, we present relatively long term $\Delta^{17}O$ 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. Please refer our reply to first part of your major comment where we clarified the measurement procedure. Please note that we evaporated sample solutions without adding $BaCl_2$, even which recovered sulfate in the obtained precipitates.

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 232-234: "Since BC is mainly produced from incomplete combustion of fossil fuel and biomass, which are also important sources of SO_2, thus in order to investigate the potential contribution of combustion source to 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 might 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 299-307: "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 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 originate 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 discussion or 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. Besides, in order to make the comparison between estimated f_{O3} from observations and modeling results more intuitive, we modified Fig. 7 by plotting $f_{O3, min}$ and $f_{O3, max}$ in Fig. 7(c-d).

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 249, 403: 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 405: The "hypothesize" was changed to "explain" now.

11. 363: Change "hypothesis" to "explanation".

Response: Corrected.

Action: Line 408: 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 459-463 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 an explanation in lines 471-475.

Action: Lines 459-463: "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 increasing the hygroscopicity of atmospheric aerosols, RH can promote SO_2 oxidation on the surface of dust aerosols. Thus, RH should play a crucial role in the heterogeneous oxidation processes".

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) + O₃ 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 471-475: "Also note that, although the seasonal trends in $\Delta^{17}O$ for both nitrate and sulfate result from the relative importance of S(IV) + O₃ pathway, the mechanisms behind it are different. For nitrate, the main factor controlling the $\Delta^{17}O$ seasonality is the O₃ 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 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 O₃. 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 O₃. 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 analyzed the relevance between $\Delta^{17}O$ and meteorological data, and calculated the contribution of O₃ oxidation based on the observations. Based on these analyses, 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 446-448: "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 indicates the importance of

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.

References

Bao, H., Campbell, D. A., Bockheim, J. G., and Thiemens, M. H.: Origins of sulphate in Antarctic dry-valley soils as deduced from anomalous ^{17}O compositions, Nature, 407, 499-502, https://doi.org/10.1038/35035054, 2000a.

Bao, H., Thiemens, M. H., Farquhar, J., Campbell, D. A., Lee, C. C.-W., Heine, K., and Loope, D. B.: Anomalous ^{17}O compositions in massive sulphate deposits on the Earth, Nature, 406, 176-178, https://doi.org/10.1038/35018052, 2000b.

Bauer, S. E., and Koch, D.: Impact of heterogeneous sulfate formation at mineral dust surfaces on aerosol loads and radiative forcing in the Goddard Institute for Space Studies general circulation model, J. Geophys. Res. Atmos., 110, https://doi.org/10.1029/2005jd005870, 2005.

Dominguez, G., Jackson, T., Brothers, L., Barnett, B., Nguyen, B., and Thiemens, M. H.: Discovery and measurement of an isotopically distinct source of sulfate in Earth's atmosphere, Proc. Natl. Acad. Sci. U.S.A., 105, 12769-12773, https://doi.org/10.1073/pnas.0805255105, 2008.

Geng, L., Schauer, A. J., Kunasek, S. A., Sofen, E. D., Erbland, J., Savarino, J., Allman, D. J., Sletten, R. S., and Alexander, B.: Analysis of oxygen-17 excess of nitrate and sulfate at sub-micromole levels using the pyrolysis method, Rapid Commun. Mass Spectrom., 27, 2411-2419, https://doi.org/10.1002/rcm.6703, 2013.

Kang, S., Qin, D., Mayewski, P. A., Sneed, S. B., and Yao, T.: Chemical composition of fresh snow on Xixabangma peak, central Himalaya, during the summer monsoon season, J. Glaciol., 48, 337-339, https://doi.org/10.3189/172756502781831467, 2002.

Kang, S., Zhang, Y., Qian, Y., and Wang, H.: A review of black carbon in snow and ice and its impact on the cryosphere, Earth Sci. Rev., 103346, https://doi.org/10.1016/j.earscirev.2020.103346, 2020.

Kunasek, S., Alexander, B., Steig, E., Sofen, E., Jackson, T., Thiemens, M., McConnell, J., Gleason, D., and Amos, H.: Sulfate sources and oxidation chemistry over the past 230 years from sulfur and oxygen isotopes of sulfate in a West Antarctic ice core, J. Geophys. Res. Atmos., 115, https://doi.org/10.1029/2010jd013846, 2010.

Lee, C. W., and Thiemens, M. H.: The δ^{17}O and δ^{18}O measurements of atmospheric sulfate from a coastal and high alpine region: A mass-independent isotopic anomaly, J. Geophys. Res. Atmos., 106, 17359-17374, https://doi.org/10.1029/2000jd900805, 2001.

Li, L., Chen, Z., Zhang, Y., Zhu, T., Li, J., and Ding, J.: Kinetics and mechanism of heterogeneous oxidation of sulfur dioxide by ozone on surface of calcium carbonate, Atmos. Chem. Phys., 6, 2453-2464, https://doi.org/10.5194/acp-6-2453-2006, 2006.

Lim, S., Lee, M., Kim, S.-W., and Laj, P.: Sulfate alters aerosol absorption properties in East Asian outflow, Sci. Rep., 8, 1-7, https://doi.org/10.1038/s41598-018-23021-1, 2018.

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, 2020.

Patris, N., Delmas, R., Legrand, M., De Angelis, M., Ferron, F. A., Stiévenard, M., and

Jouzel, J.: First sulfur isotope measurements in central Greenland ice cores along the preindustrial and industrial periods, J. Geophys. Res. Atmos., 107, ACH 6-1-ACH 6-11, https://doi.org/10.1029/2001jd000672, 2002.

Prospero, J. M., Ginoux, P., Torres, O., Nicholson, S. E., and Gill, T. E.: Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product, Rev. Geophys., 40, 2-1-2-31, https://doi.org/10.1029/2000rg000095, 2002.

Pye, H. O., Nenes, A., Alexander, B., Ault, A. P., Barth, M. C., Clegg, S. L., Collett Jr, J. L., Fahey, K. M., Hennigan, C. J., and Herrmann, H.: The acidity of atmospheric particles and clouds, Atmos. Chem. Phys., 20, 4809-4888, https://doi.org/10.5194/acp-20-4809-2020, 2020.

Savarino, J., Alexander, B., Darmohusodo, V., and Thiemens, M. H.: Sulfur and oxygen isotope analysis of sulfate at micromole levels using a pyrolysis technique in a continuous flow system, Anal. Chem., 73, 4457-4462, https://doi.org/10.1021/ac010017f, 2001.

Schauer, A. J., Kunasek, S. A., Sofen, E. D., Erbland, J., Savarino, J., Johnson, B. W., Amos, H. M., Shaheen, R., Abaunza, M., and Jackson, T. L.: Oxygen isotope exchange with quartz during pyrolysis of silver sulfate and silver nitrate, Rapid Commun. Mass Spectrom., 26, 2151-2157, https://doi.org/10.1002/rcm.6332, 2012.

Shah, V., Jacob, D. J., Moch, J. M., Wang, X., and Zhai, S.: Global modeling of cloud water acidity, precipitation acidity, and acid inputs to ecosystems, Atmos. Chem. Phys., 20, 12223-12245, https://doi.org/10.5194/acp-20-12223-2020, 2020.

Sofen, E., Alexander, B., and Kunasek, S.: The impact of anthropogenic emissions on atmospheric sulfate production pathways, oxidants, and ice core [delta] 17O (SO42-), Atmos. Chem. Phys., 11, 3565, 2011.

Wang, W., Xu, W., Collett Jr, J. L., Liu, D., Zheng, A., Dore, A. J., and Liu, X.: Chemical compositions of fog and precipitation at Sejila Mountain in the southeast Tibetan Plateau, China, Environ. Pollut., 253, 560-568, https://doi.org/10.1016/j.envpol.2019.07.055, 2019.

Xie, L., Spiro, B., and Wei, G.: Purification of $BaSO_4$ precipitate contaminated with organic matter for oxygen isotope measurements ($\delta^{18}O$ and $\Delta^{17}O$), Rapid Commun. Mass Spectrom., 30, 1727-1733, https://doi.org/10.1002/rcm.7610, 2016.