Black: Reviewers' comments
Red: Response
Blue: Action

Response to Reviewer #2

This study presents triple oxygen isotope data on sulfate aerosols sampled in the Tibetan Plateau. The data reveal an interesting $\Delta^{17}O$-seasonal trend with the monsoon season which would be explained by an enhanced O_3 oxidation pathway in SO_2 oxidation. The data obtained are valuable, the quality of the measurement seems solid to me. The importance of the alkalinity on the sulfate aerosols emphasized in this study is relevant as there are still many uncertainties on the sulfate formation, highlighting the exploration of new perspectives.

However, I have some comments and some suggestions related on how the results are discussed as listed above:

Response： We thank the reviewer for finding our work interesting and important. We also feel great thanks for the reviewer's helpful comments and suggestions on our manuscript. We have considered these comments and suggestions very carefully. Please find our detailed point-to-point responses to the reviewer's concern below.

1. I am not sure to understand why the Tibetan Plateau is an important region and why it is called "Third Pole". Please describe the importance of this area.

Response： We appreciate the reviewer for this valuable comment which would further highlight the importance of our work. We added some sentences to further highlight the importance of the Tibetan Plateau in the section of Abstract.

Action： Lines 62-66: **"**The Tibetan Plateau (hereafter referred to as TP), covering an area of ~2.5×10^6 km^2 and having an average elevation of over 4000 m above sea level (a.s.l.), is the largest plateau in China and the highest plateau in the world. It contains the largest land ice masses outside the poles and supplies water for more than one billion people (Immerzeel et al., 2010). Due to its large area and high altitude, the TP plays an important role in the evolution of Asian monsoon system, and even the Earth's climatic system. Thus, the TP has been known as the 'Third Pole' (Kang et al., 2010; Yao et al., 2012)".

2. In the Method l.155. It is unclear how the authors reported the $\Delta^{17}O$. Is it the raw or the corrected values? In the latter case, how did the authors proceed? A reference is at least required.

Response： We are sorry for the vague expressions which might lead to misunderstanding. The reported $\Delta^{17}O$ values in this study were corrected values. The correction methods have been applied in several studies, e.g., Gautier et al. (2019), Ishino et al. (2021), and Ishino et al. (2017), Hattori et al. (2021). In order to make it in a more clear way, we modified the related description, and add a reference to explain the correction principle.

Action: Lines 186-189: "the raw $\Delta^{17}O$ values were corrected by estimating the magnitude of the oxygen isotope exchange using inter-laboratory calibrated standards as described in Schauer et al. (2012). The correction method has also been applied by several our previous studies (Gautier et al., 2019; Ishino et al., 2021; Ishino et al., 2017; Hattori et al., 2021)".

3. I expected to see a calculation based on Na to exclude the contribution of sea-salt, which would be confirmed by the study of Cong et al. Using only Cong study seems insufficient to me.

Response: We thank the reviewer for the suggestion. Yes, we agree that a calculation based on the ratio of $[SO_4^{2-}]/[Na^+]$ in sea salt would further confirm a negligible amount of sea-salt sulfate in the Mt. Everest region. Due to the high amount of Na^+ in blank filter and/or low Na^+ concentration in samples, the Na^+ concentrations in most of samples are unavailable, thus the Na^+ concentrations are not reported in this study. Actually, Cong et al. (2015) also reported that the Cl^- and Na^+ only consisted of a very minor portion of total ions, and they suggested that the influence of sea salt at our sampling site (QOMS) is negligible. In conclusion, the Mt. Everest region, which is far away from the ocean (~ 750 km), is considered to possess a negligible amount of sea-salt sulfate in its air masses. To make the description more clear, we made a revision here.

Action: Lines 274-279: Due to the relatively high amounts of Na^+ in blank filter papers and/or low Na^+ concentrations ($[Na^+]$) in environmental samples, the $[Na^+]$ data in most of the samples are unavailable and thus the $[Na^+]$ are not reported in this study. Indeed, a previous study has indicated that the Cl^- and Na^+ only consisted of a very minor portion of total ions in TSP, and suggested a negligible influence of sea salt at QOMS (Cong et al., 2015a). In conclusion, the Mt. Everest region, which is located at high elevation and ~750 km away from the ocean, is considered to possess a negligible amount of sea-salt sulfate in its air masses.

4. Knowing that the Ca/Al ratio vary in Asian dust from 0.1 to 35%, how would this affect your result? Ratio from anthropogenic dust is also expected to vary. I also didn't find the values cited by the authors in the literature.

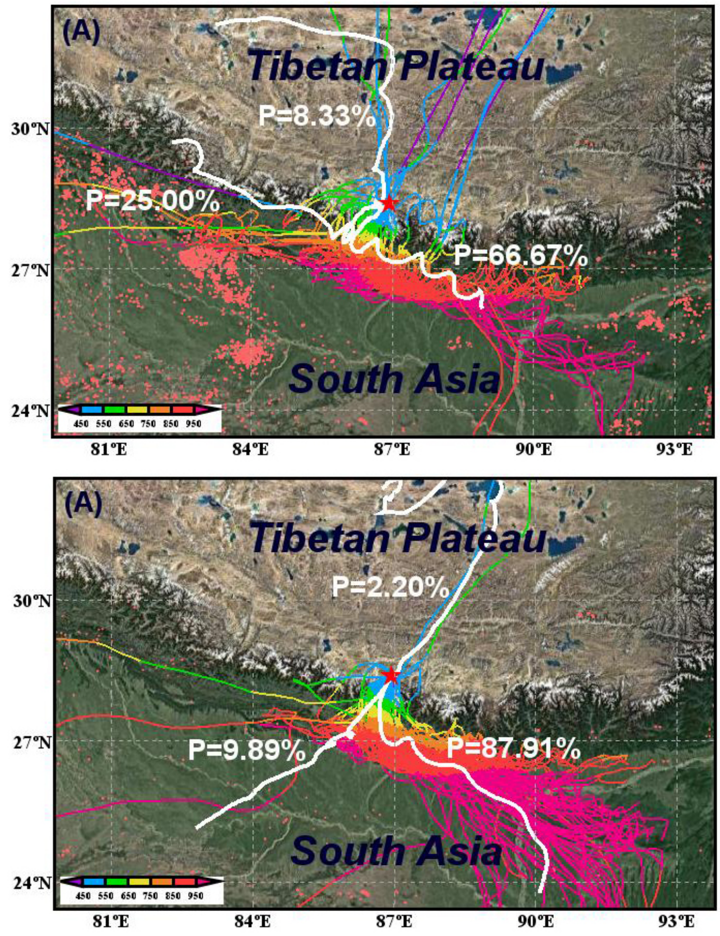
Response: Since Ca^{2+} is a typical tracer of crustal materials (dust) (Ram et al., 2010), a ratio (k) of $[SO_4^{2-}]/[Ca^{2+}]$ in crustal materials is usually used to identify terrigenous sulfate. Either for natural dust or for the anthropogenic dust, the ratio of $[SO_4^{2-}]/[Ca^{2+}]$ indicated a terrigenous source. Thus we think the Ca/Al ratio varying from 0.1 % to 35 % would not affect our results. In our study, the k value of 0.18 (mass ratio) was applied for the estimation, which has been widely used for the estimation of terrigeneous sulfate in different regions including Tibetan Plateau (Kunasek et al., 2010; Patris et al., 2002). In Lin et al. (2020), the k value of 0.18 can be found in the Supplementary Materials, and in Kunasek et al. (2010), the k values can be found in section 3 (Method), while the k value used in Patris et al. (2002) can be found in section 3.3 (Preindustrial Background).

5. It is unclear if the SO_2 concentration has been measured, but adding a discussion using the SO_2 concentration in particular with the use of SOR could help convince me about the predominance of secondary sulfate.

Response: We thank the reviewer for this suggestion. Given the difficulty of in-situ measurements in the Mt. Everest region, we did not measure the SO_2 concentration in this study. We agree that the additional SO_2 concentration data in particular with the use of SOR would make the investigation more comprehensive, and we will consider this in the future studies if the data available.

6. l. 320 Are the HYSPLIT results consistent with your conclusion?

Response: Yes, the HYSPLIT result is consistent with our conclusion. As shown in the Figure below, we can see the pressure along with almost all the trajectories are lower than 450 hPa, thus most of the air masses arriving at our sampling sites are within troposphere.



Five-Day backward trajectories of air masses (colored lines, color represents the elevation in hPa) arriving at QOMS for each day during (A) pre-monsoon (April and May) and (B) monsoon (June, July and August) in 2018. White lines in the map denote different clusters with respective percentage (P) of trajectories.

7. l. 341 A comparison and discussion with the $\Delta^{17}O$ modeled using GEOS-Chem would have been a great addition to the paper. Considering the O_3 concentration, and the kinetic reaction, what $\Delta^{17}O$ values are expected and how does it compare to both min and max fraction you deduced? I am confused on the fact that GEOS-Chem model is only used in cloud pH model

Response： We thank the reviewer for this comment. The changes in concentrations of various oxidants (e.g., O_3, H_2O_2, and OH) would certainly affect the fractional contribution of different formation pathways of atmospheric sulfate. Based on the analyses including the relevance between $\mathit{\Delta}^{17}O$ and oxidants concentrations as well as meteorological data, and the calculated contribution of O_3 oxidation based on the observations, we concluded that the atmospheric acidity and/or the heterogeneous reactions might be responsible for the relatively high $\mathit{\Delta}^{17}O$ signal and its seasonality in the atmospheric sulfate observed in the Mt. Everest region. In order to further verify our thoughts, we conducted the modeling work, and found that the cloud water pH does play an important role in the formation pathways in our study area, and this is also the main conclusion of this study.

Thus, our modeling work was mainly focused on the consideration of cloud water pH and heterogeneous S(IV) oxidation by O_3 on dust surface. 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(b-d).

Action：Line 864 Figure 7, we added estimated f_{O3} from observations in b-d.

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