



1 **Moisture sources and dynamics over southeastern Tibetan Plateau**  
2 **reflected in dual water vapor isotopes**

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11

12 **Abstract**

13 The Tibetan Plateau (TP) serves as a water tower for major rivers in Asia, and mountain valleys in southeastern  
14 TP are key channels for moisture entering the TP. Water resources on the TP are experiencing spatially opposite  
15 changes due to climate change, and understanding the sources and dynamics of atmospheric moisture is vital. To  
16 investigate the role of ocean surface evaporation, continental air mass intrusion, and rain-vapor interaction, we  
17 present a three-year daily time series of near-surface water vapor isotope compositions ( $\delta^{18}\text{O}$  and  $d$ -excess) from  
18 the South-East TP station. We find that apparent negative correlations between  $d$ -excess and relative humidity over  
19 the Indian Ocean mainly reflect their similar seasonality. When analyzed for different seasons, the correlation is  
20 insignificant or only explains a marginal fraction of variance. Therefore, caution is required when interpreting the  
21  $d$ -excess as a conservative tracer of ocean surface evaporation. Instead, local and upstream specific humidity is the



22 main factor determining non-monsoon season  $d$ -excess variability due to the intrusion of cold and dry air from upper  
23 levels. During the summer monsoon season,  $d$ -excess and  $\delta^{18}\text{O}$  mainly reflect the effect of raindrop evaporation on  
24 humidity during transport which decreases lower vapor  $\delta^{18}\text{O}$  but increases  $d$ -excess values. These findings provide  
25 new insights into the significance of using water isotopes to track moisture sources and dynamics over the TP with  
26 seasonally alternating circulation systems. Particularly, the findings for  $d$ -excess will improve the understanding of  
27 different moisture sources and guide the interpretation of  $d$ -excess derived from other water bodies and ice cores.

28

## 29 **1 Introduction**

30 The Tibetan Plateau (TP) and its surrounding regions, also termed the Third Pole and the Asian Water Tower,  
31 form the highest and largest plateau on Earth that influences climatic and hydrological systems at regional to global  
32 scales, such as the formation of the Asian Summer Monsoon (Wu et al., 2022; Yao et al., 2022). In addition, the TP  
33 stores the largest amount of frozen water outside of polar regions and sustains freshwater supplies of major river  
34 systems in Asia. However, the water balance on the TP has experienced significant changes under the backdrop of  
35 global warming (Yao et al., 2022). For instance, the southeastern TP is experiencing a drying trend while wetting in  
36 the northern TP (Jiang et al., 2023; Zhang et al., 2023). Atmospheric water vapor is the input of the water storage  
37 system and understanding its sources and dynamics is vital for understanding the imbalance of TP's hydrological  
38 system.

39 Water stable isotopes are natural tracers of the water cycle (Bowen et al., 2019; Galewsky et al., 2016) and  
40 have been intensively studied on the TP in precipitation, surface water, and ice cores (Yao et al., 2013; Thompson  
41 et al., 2024; Bershaw, 2018). In general, precipitation isotope ratios ( $\delta^{18}\text{O}$  and  $\delta^2\text{H}$ ) over the southern TP have lower  
42 values during the summer monsoon season and higher values during the non-monsoon season under the influence



43 of the westerlies (He et al., 2015; Tian et al., 2007; Guo et al., 2024; Yang et al., 2017). Recent studies have  
44 confirmed that monsoon convection at upstream along moisture transport pathways, rather than local precipitation  
45 amount, is the key process that controls summer monsoon season precipitation  $\delta^{18}\text{O}$  (Cai et al., 2017; He et al.,  
46 2015). Different processes have been proposed to elucidate the relationship between precipitation  $\delta^{18}\text{O}$  and  
47 convection where the amount effect (Dansgaard, 1964) is present (Bowen et al., 2019; Galewsky et al., 2016). A  
48 relatively classical interpretation is that the continuous rainout associated with stronger convection could cause  
49 depleted precipitation following the Rayleigh distillation (Cai and Tian, 2016; Scholl et al., 2009; Vuille et al., 2003).  
50 Another interpretation emphasized the role of rain-vapor interaction that partial evaporation of raindrops formed at  
51 higher altitudes isotopically depletes lower tropospheric water vapor and then affects subsequent precipitation  
52 isotope compositions (Risi et al., 2008a; Kurita et al., 2011; Cai et al., 2018; Lee and Fung, 2008). Observations of  
53 vapor isotope compositions could help disentangle the different processes involved in the amount effect, especially  
54 the secondary parameter deuterium excess (*d*-excess). The *d*-excess is defined as  $\delta^2\text{H} - 8\delta^{18}\text{O}$  by Dansgaard (1964)  
55 and mainly reflects the effects of kinetic fractionation. The rainout process mostly involves equilibrium fractionation  
56 while raindrop evaporation is associated with kinetic fractionation, and they can therefore have different *d*-excess  
57 signatures in water vapor. Isotopic compositions in the vapor phase have only been observed at a few stations on  
58 the TP and isotope ratios ( $\delta$  values) have been the major focus of previous studies (Tian et al., 2020; Dai et al., 2021;  
59 Chen et al., 2024; Yu et al., 2016; Yu et al., 2015), less is known about vapor *d*-excess.

60 It is less certain regarding what caused higher isotope ratios during the non-monsoon season. Following the  
61 regional amount effect (Galewsky et al., 2016; Bowen et al., 2019), high  $\delta^{18}\text{O}$  values could be explained by  
62 weakened convection during the non-monsoon season. However, precipitation  $\delta^{18}\text{O}$  is lower during late- to post-  
63 monsoon season in regions extending from the southeastern TP to the head of the Bay of Bengal (BOB), which is  
64 not consistent with the weakening of convection (Breitenbach et al., 2010; Cai and Tian, 2020). Shifts of moisture



65 transport pathways between convection active and non-active regions have been invoked to explain this abnormal  
66 seasonal pattern (Cai and Tian, 2020; Lekshmy et al., 2022). On the other hand, higher precipitation  $\delta^{18}\text{O}$   
67 accompanied by higher  $d$ -excess during the non-monsoon season has been interpreted as more intense continental  
68 recycling or moisture from the Mediterranean delivered by the westerlies compared with moisture from the Indian  
69 Ocean during the summer monsoon (Tian et al., 2007; Yao et al., 2013; An et al., 2017; Breitenbach et al., 2010). In  
70 addition, understanding of the atmospheric water cycle for a full seasonal cycle is complicated by the lack of  
71 precipitation during the non-monsoon season which can be compensated by monitoring atmospheric water vapor  
72 isotopes as it is not limited by precipitation events.

73 Both theoretical predictions and observations over ocean surface suggested that  $d$ -excess reflects ocean surface  
74 evaporation conditions, such as sea surface temperature (SST) and relative humidity normalized to SST ( $\text{RH}_{\text{SST}}$ )  
75 (Merlivat and Jouzel, 1979; Bonne et al., 2019; Liu et al., 2014; Craig and Gordon, 1965). Interpretations of  $d$ -  
76 excess over the TP also frequently invoke these relationships with ocean evaporation conditions (Zhao et al., 2012;  
77 Shao et al., 2021; Chen et al., 2024; Liu et al., 2024). However, relationships with either  $\text{RH}_{\text{SST}}$  or SST are much  
78 weaker than those observed over ocean surface. For instance, Shao et al. (2021) showed significant correlations  
79 between an ice core  $d$ -excess record derived from the central TP and  $\text{RH}_{\text{SST}}$  over the northern BOB and Arabian Sea  
80 (AS). However, the correlation coefficient is only -0.44 and the slope between  $d$ -excess and  $\text{RH}_{\text{SST}}$  is as steep as -  
81  $0.99\text{‰} \text{‰}^{-1}$ . The slope over oceanic regions generally ranges from  $-0.3\text{‰} \text{‰}^{-1}$  to  $-0.6\text{‰} \text{‰}^{-1}$  based on in-situ  
82 observations (Bonne et al., 2019; Liu et al., 2014; Benetti et al., 2014; Uemura et al., 2008). Many studies have  
83 suggested that  $d$ -excess at terrestrial sites is not a conservative tracer of evaporation conditions at the oceanic source  
84 regions (Fiorella et al., 2018; Aemisegger et al., 2014; Welp et al., 2012; Wei and Lee, 2019; Samuels - Crow et al.,  
85 2014). Besides temporal variations, ice core  $d$ -excess values are generally higher than that observed in precipitation  
86 at lower altitudes on the TP (Shao et al., 2021; Tian et al., 2001; Zhao et al., 2012; Joswiak et al., 2013; Zhao et al.,



87 2017; Thompson et al., 2000). It is still unclear what caused the abnormally high  $d$ -excess in these high-altitude ice  
88 cores relative to precipitation and river isotope observations at lower altitudes.

89 Mountain valleys in the southeastern TP are believed to be major moisture transport channels delivering water  
90 vapor toward the TP (Araguás-Araguás et al., 1998; Tian et al., 2007; Yao et al., 2013). Therefore, we started a water  
91 vapor sampling campaign at the South-East Tibetan Plateau Station for integrated observation and research of alpine  
92 environment (SETP) in June 2015 to study the moisture sources and dynamics and their influence on water isotope  
93 compositions. Following the previous study (Yao et al., 2013), we define June-September (JJAS) as the summer  
94 monsoon season. In contrast, we define November-April (Nov-Apr) as the non-monsoon season and May and  
95 October as the transition between the two seasons. We will show distinct seasonal moisture sources and dynamics  
96 between the two seasons as reflected in our vapor isotope observations and Lagrangian moisture source diagnostic.  
97 Our results suggest that the apparent correlation between SETP vapor  $d$ -excess and oceanic surface evaporation  
98 conditions is mainly a result of their similar seasonality. Alternatively, we suggest the intrusion of dry and cold air  
99 by the westerlies from high altitudes contributes to high  $d$ -excess. In contrast, vapor  $\delta^{18}\text{O}$  and  $d$ -excess confirm the  
100 significant role of rain-vapor interaction in the amount effect during the summer monsoon season.

## 101 **2 Data and methods**

### 102 **2.1 Atmospheric water vapor sampling**

103 Atmospheric water vapor samples were collected using a cryogenic trapping method at the SETP station  
104 (29°46'N, 94°44'E, 3326 m above sea level, and Fig. S1). The sampling system includes an air pump pumping  
105 ambient air into the cold trap, a linked-ball-shaped glass cold trap, and an electric-powered system that creates and  
106 maintains a cold environment filled by 95% ethanol as cold as below -80 °C. Ambient air was pumped from an inlet  
107 at approximately 8 m above ground level through a Teflon tube to a glass trap immersed in a cold environment with



108 a temperature at  $-70\text{ }^{\circ}\text{C}$ . The airflow rate was adjusted to  $\sim 5\text{ L/min}$  to allow 10-20 ml of water samples throughout  
109 each sampling operation. During summers, the duration of each sampling operation is 24 hours. During dry winters,  
110 however, we increased the sampling duration to 48 hours if a 24-hour sampling period cannot guarantee enough  
111 sample amount. The samples were collected at 20:00 Beijing Standard Time (12:00 UTC). The efficiency of  
112 extracting water vapor from ambient air was tested by connecting an additional cold trap to the outlet of the initial  
113 cold trap, and no visible condensation was noticed in the additional cold trap (Yu et al., 2015). Further comparison  
114 against direct measurements of vapor isotope composition by the Picarro L2130-i Cavity Ring Down Spectroscopy  
115 (CRDS) at Lhasa, southern TP also confirmed the reliability of this method in sampling atmospheric water vapor  
116 over the TP (Tian et al., 2020).

117 The sampling campaign was started on 25 June 2015 and ended on 14 June 2018. In total, 742 samples were  
118 collected, and all the collected samples were kept frozen until transportation to the laboratory for measurements.  
119 Samples collected before 28 June 2016 were measured at the Key Laboratory of Tibetan Plateau Earth System,  
120 Environment and Resources, Institute of Tibetan Plateau Research, Chinese Academy of Sciences by a Picarro  
121 L2130-i analyzer. Samples collected after 28 June 2016 were measured at the Institute of International River and  
122 Eco-security, Yunnan University by a Picarro L2140-i analyzer. The isotopic values are calibrated and expressed  
123 relative to Vienna Standard Mean Ocean Water 2 (VSMOW2). The precisions of measurements at both laboratories  
124 are 0.1‰ for  $\delta^{18}\text{O}$ , 0.4‰ for  $\delta^2\text{H}$ , and 1.2‰ for  $d$ -excess.

## 125 **2.2 Meteorological data**

126 Daily local meteorological data before 2018, including precipitation amount, air temperature, air pressure, and  
127 relative humidity, at the SETP station were provided by the station at the National Tibetan Plateau / Third Pole  
128 Environment Data Center (Luo, 2018). Specific humidity ( $q$ ) at SETP station is calculated from air temperature, air  
129 pressure, and relative humidity at the station.



130 To facilitate analyses on larger spatial scales, we obtained meteorological variables (including 2-meter air  
131 temperature, 2-meter dew point temperature, and SST, etc.) at  $0.25^\circ \times 0.25^\circ$  and hourly resolution from the European  
132 Centre for Medium-Range Weather Forecasts fifth generation reanalysis (ERA5) (Hersbach et al., 2019).  $RH_{SST}$  is  
133 estimated from ERA5 2-meter meteorological data and SST using  $RH_{SST} = e_{air}/e_{sat}$ , where  $e_{air}$  is vapor pressure  
134 of air and  $e_{sat}$  is saturation vapor pressure with respect to SST. We further obtained precipitation data at  $0.1^\circ \times 0.1^\circ$   
135 and half-hourly resolution from the Integrated Multi-satellitE Retrievals for GPM (V07) (Huffman et al., 2023). In  
136 addition, meteorological data at  $1^\circ \times 1^\circ$  and 3-hourly resolution from the Global Data Assimilation System (GDAS)  
137 are used to calculate air mass trajectories (see section 2.4).

### 138 **2.3 Theoretical framework for the understanding of isotope compositions and humidity**

139 Besides complex atmospheric circulation models, the evolution of vapor isotope compositions during different  
140 moistening and dehydration processes can be understood by a compilation of atmospheric processes, such as  
141 condensation, mixing, and raindrop evaporation, that lead to different pathways of isotopic evolution along  
142 atmospheric humidity (Noone, 2012; Worden et al., 2007; Galewsky et al., 2016). The progressive condensation of  
143 water vapor and removal as rain droplets or ice is best described by the canonical Rayleigh distillation model  
144 (Dansgaard, 1964). In the Rayleigh distillation framework, condensate is removed from the air mass as soon as it  
145 forms, and the isotope ratio of remaining vapor is described as  $\delta = (1 + \delta_0)(q/q_0)^{\alpha-1} - 1$ , where  $\delta$  is the  
146 isotope composition expressed as per mil deviation from a standard,  $q$  is the specific humidity, and  $\alpha$  is the  
147 fractionation factor. A subscript of 0 refers to the initial condition of the air mass. The falling raindrop may partially  
148 evaporate or exchange isotopes with ambient vapor. As raindrops are formed at higher altitudes where water vapor  
149 is depleted in heavy isotopes, the partial evaporation of raindrops would preferentially deplete its surrounding water  
150 vapor but increase the atmospheric humidity, which leads to  $\delta$  values lower than that predicted by the Rayleigh  
151 distillation (Risi et al., 2008a; Worden et al., 2007). The evolution of  $\delta$  along with  $q$  under partial evaporation of



152 raindrops can be described in a “super-Rayleigh” trajectory by inflating the effective fractionation factor ( $\alpha_e$ ) as  
153  $\alpha_e = (1 + \phi)\alpha$ , where  $\phi$  quantifies the degree to which  $\alpha$  deviates from equilibrium. We note that Worden et al.  
154 (2007) and Noone (2012) have given different equations to quantify the deviations of  $\alpha_e$  from  $\alpha$  under different  
155 degrees of raindrop evaporation, and the same deviation of  $\alpha_e$  from  $\alpha$  requires very different degrees of raindrop  
156 evaporation. In this study, we follow the formulations by Noone (2012). Finally, the influence of air mass mixing  
157 on humidity and isotopic compositions can be modeled through the mass balance perspective. When considering  
158 mixing a dry air mass with a moist air mass, for instance, the specific humidity of mixed air mass can be described  
159 as  $q = f_{dry}q_{dry} + f_{moist}q_{moist}$ , where  $f$  is the fraction of each air mass with the subscript denoting different air  
160 masses and  $f_{dry} + f_{moist} = 1$ . Isotopic compositions of the mixed air mass can be derived similarly by solving the  
161 mass balance equations for the light and heavy isotopes, respectively. The outcome of the mixing process leads to a  
162 hyperbolic relationship between  $\delta$  and  $q$ . In other words,  $\delta \times q$  and  $q$  should have a linear relationship in the  
163 mixing process (Fiorella et al., 2018). In a framework of the Keeling plots (Keeling, 1958), the intercept of the  
164 regression between  $\delta$  and  $1/q$  or the slope between  $\delta \times q$  and  $q$  gives an estimation of the isotope  
165 composition of the moist end member.

166 Assuming a surface temperature of 25 °C and relative humidity of 85%, following the evaporation model by  
167 Craig and Gordon (1965) we can derive that  $\delta^{18}\text{O}$  of ocean surface evaporation is -11.5%,  $\delta^2\text{H} = -81.4\%$ , and  $d$ -  
168 excess = 10.6%. We use this isotopic signature of evaporated water vapor as a wet end member to model the  
169 moistening process by mixing with ocean surface evaporation. A hypothetical dry end member from the Rayleigh  
170 curve at  $q = 0.5$  g/kg is chosen to represent the dehydrated dry air. The dehydration process by condensation is  
171 modeled by choosing a starting point at the mixing line with a relative humidity of 80%. Similarly, the “super-  
172 Rayleigh” distillation with partial rain evaporation is started from the same starting point. For the cases of “super-  
173 Rayleigh”, we simulated the isotopic evolution under two scenarios (Rain\_evap\_A and Rain\_evap\_B). Following





174 the equations in Noone (2012), Rain\_evap\_A represents that 2% of rain is evaporated and Rain\_evap\_B represents  
175 an evaporated fraction of 5%. Mixing with evapotranspiration over south Asia and the TP is another way that could  
176 modify the atmospheric humidity and vapor isotope compositions over southeastern TP. Accurate quantification of  
177 the isotopic composition of land surface evapotranspiration is challenging. Given that the precipitation  $\delta^{18}\text{O}$  over  
178 south Asia is generally between -1.0‰ and -5.0‰ (Bowen and Wilkinson, 2002; Terzer-Wassmuth et al., 2021) and  
179 transpiration may account two-thirds of evapotranspiration or more (Cao et al., 2022; Han et al., 2022; Good et al.,  
180 2015), we assume the  $\delta^{18}\text{O}$  of land surface evapotranspiration has a value of -5.0‰ as an upper bound. Similarly,  
181 we assume that the  $d$ -excess of the wet end member of land surface evapotranspiration is 15.0‰.

#### 182 **2.4 Air mass trajectory and moisture source diagnostic**

183 Air mass backward trajectories were calculated to investigate the air mass transport and diagnose moisture  
184 sources and transport pathways toward SETP using the Hybrid Single-Particle Lagrangian Integrated Trajectory  
185 model (HYSPLIT) (Stein et al., 2015). Trajectory calculation is driven by the meteorological data of the GDAS. We  
186 released air parcels from 5 locations (the studied site and points displaced  $0.2^\circ$  in each cardinal direction) at 7  
187 different vertical levels at 10, 50, 100, 200, 300, 400, and 500 m above ground level. For each day during the  
188 sampling campaign, trajectories were initiated every 3 hours to calculate 10-day backward trajectories. In this setting,  
189 280 trajectories were derived for each single day. Geographical and meteorological variables, including location,  
190 pressure, temperature, specific humidity, rainfall amount, boundary layer height, and the terrain height along  
191 trajectories, were stored at hourly intervals.

192 Moisture contribution along air mass trajectories to the humidity at SETP is quantified using the Lagrangian  
193 moisture source diagnostic method of Sodemann et al. (2008). The method considers mass balance along the  
194 trajectory and assigns increases in specific humidity (forward in time) as moisture uptake, and decreases in specific  
195 humidity as moisture lost due to precipitation. The method also proportionally considers the decreased contribution



196 of early uptake due to the precipitation en route. We have previously adapted this method to quantify the moisture  
197 sources of precipitation in sub-regions of South Asia and East Asia (Cai et al., 2018; Cai and Tian, 2020). The  
198 diagnostic results suggest that the unattributed fraction of moisture arriving at SETP is ~5%, and therefore 10-day  
199 trajectories are capable of diagnosing most of the moisture sources. Instead of focusing on quantifying the  
200 evaporative moisture source from the Earth surface, in this study, we focus on the contribution of the air parcel itself  
201 to the humidity at SETP. This variable is readily available from the diagnostic method, and the change of air parcel  
202 contributions between time steps within the boundary layer is the moisture uptake from the Earth surface. The  
203 moisture contribution by air parcel to humidity at SETP gives a measure of the importance of upstream air masses.  
204 Using this variable as the weight, mean upstream geographical and meteorological variables are hence calculated  
205 as weighted means. We also applied cluster analysis on the trajectories to visualize the major transport pathways  
206 using the K-means clustering method. When calculating the mean trajectory for each cluster and meteorological  
207 variables along each mean trajectory, the moisture contribution by air parcel is also considered as the weight to  
208 calculate weighted means.

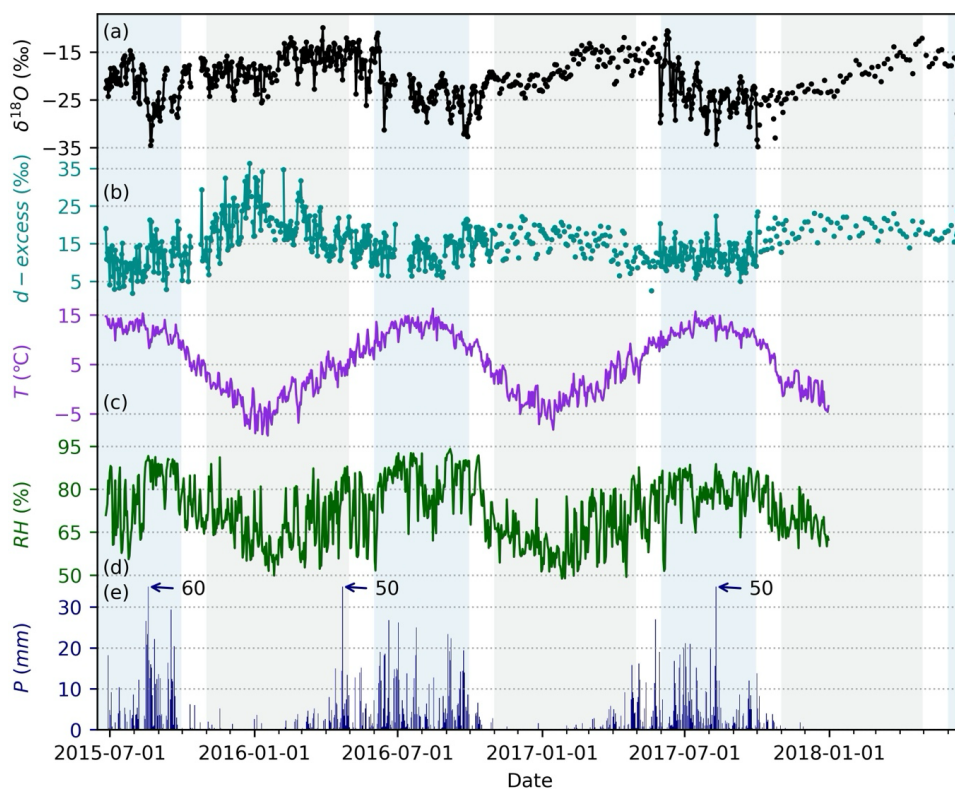
## 209 **3 Results**

### 210 **3.1 General characteristics**

211 In general, water vapor  $\delta^{18}\text{O}$  values are at a lower level during the summer monsoon season and a higher level  
212 during the non-monsoon season (Fig. 1a). Mean vapor  $\delta^{18}\text{O}$  values are -18.4‰ for the non-monsoon season, -23.3‰  
213 for the summer monsoon season, -16.9‰ for May, and -22.8‰ for October. During the onset of the summer  
214 monsoon, the vapor  $\delta^{18}\text{O}$  shows a dramatic decrease to lower values. Without a sharp rebound to values before the  
215 summer monsoon, the  $\delta^{18}\text{O}$  value shows a gradual increase trend from the end of the summer monsoon season  
216 toward the highest values during spring and early summer. Although this region is significantly influenced by the



217 amount effect, the seasonal trend of vapor  $\delta^{18}\text{O}$  does not strictly follow the seasonal variation of local precipitation.  
218 For instance, local precipitation shows clear cessation after the summer monsoon (Fig. 1e) while  $\delta^{18}\text{O}$  does not  
219 rebound to the level before summer monsoon onset. These seasonal characteristics of vapor  $\delta^{18}\text{O}$  is consistent with  
220 precipitation  $\delta^{18}\text{O}$  observed in southeastern TP, northeast India, and Bangladesh (Yao et al., 2013; Cai and Tian,  
221 2020; Yang et al., 2017). Overall, water vapor *d*-excess also has lower values during the summer monsoon season  
222 and higher values during the non-monsoon season (Fig. 1b). Mean vapor *d*-excess values are 18.3‰ for the non-  
223 monsoon season, 11.9‰ for the summer monsoon season, 13.7‰ for May, and 14.9‰ for October. However, the  
224 timing of the seasonal transition of vapor *d*-excess is different from that of vapor  $\delta^{18}\text{O}$ . The highest vapor *d*-excess  
225 values generally occur during winter months when the air temperature and relative humidity (RH) are the lowest  
226 (Fig. 1c and 1d) and the *d*-excess starts to decrease from spring which is earlier than the sharp drop of vapor  $\delta^{18}\text{O}$   
227 during the onset of the summer monsoon.



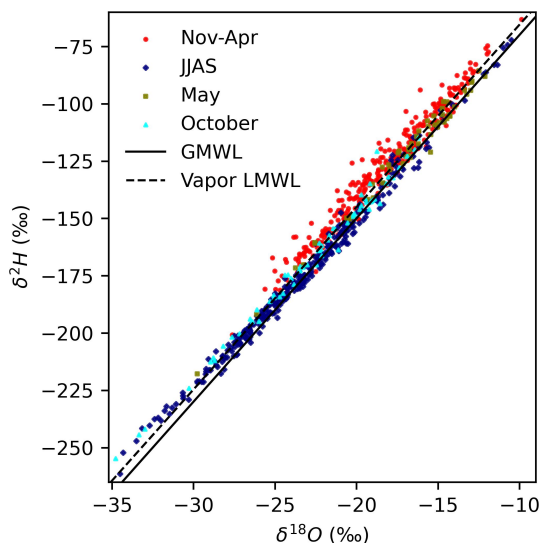
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229 **Figure 1. The general temporal variability of observed water vapor isotope compositions and daily local**  
 230 **meteorological variables from 2015-2018: (a)  $\delta^{18}\text{O}$ , (b)  $d$ -excess, (c) air temperature, (d) relative humidity**  
 231 **(RH), and (e) precipitation amount. The light blue shading highlights the summer monsoon season and the**  
 232 **light steel blue shading highlights the non-monsoon season.**

233 The linear relationship between paired  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  data points and their locations relative to the global  
 234 meteoric water line (GMWL,  $\delta^2\text{H} = 8\delta^{18}\text{O} + 10$ ) (Craig, 1961) generally provide additional insights into isotopic  
 235 fractionation (Putman et al., 2019). The local meteoric water line (LMWL) estimated from all vapor  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$   
 236 data points is  $\delta^2\text{H} = 7.96\delta^{18}\text{O} + 14.04$  ( $R^2 = 0.98$ ) which plots above but approximately parallel with the GMWL.  
 237 This relatively higher intercept of vapor LMWL reflects the continental location of the site and further kinetic  
 238 fractionation after ocean surface evaporation. The  $\delta^2\text{H}$ - $\delta^{18}\text{O}$  relationship also varied seasonally. The vapor LMWL



239 for non-monsoon season is  $\delta^2\text{H} = 7.58\delta^{18}\text{O} + 10.61$  ( $R^2 = 0.96$ ) and for summer monsoon season is  $\delta^2\text{H} = 7.53\delta^{18}\text{O}$   
240  $+ 0.91$  ( $R^2 = 0.99$ ). Non-monsoon season vapor isotope compositions mainly plot above the GMWL and even above  
241 the overall vapor LMWL. While the majority of monsoon season isotope data plot below the overall vapor LMWL,  
242 those data points that have the lowest  $\delta$  values plot above the overall vapor LMWL indicating further kinetic  
243 fractionation such as rain evaporation. Vapor isotope compositions for May are more similar to those during the  
244 non-monsoon season but plot closer to the GMWL and LMWL, while data for October show a more similar behavior  
245 with the monsoon season observations.



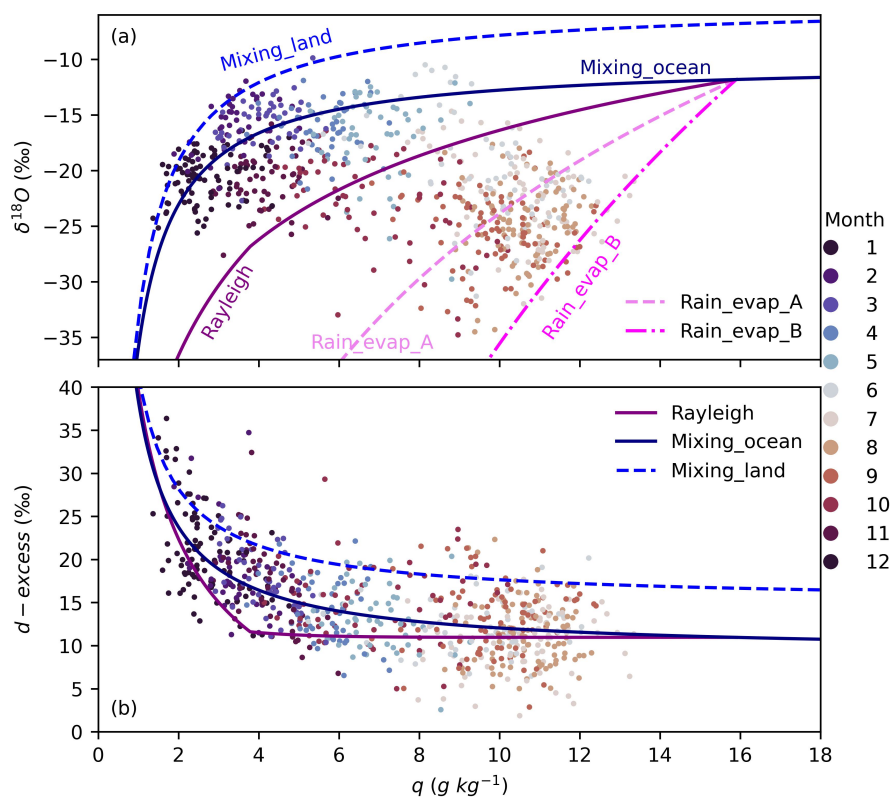
246  
247 **Figure 2. Relationship between vapor  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$ . Data during the non-monsoon season (Nov-Apr),**  
248 **the summer monsoon season (JJAS), May, and October are shown as red dots, navy diamonds, olive squares,**  
249 **and cyan triangles, respectively. The solid line indicates the global meteoric water line (GMWL). The dashed**  
250 **line indicates the local meteoric water line (LMWL) estimated from all vapor  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  data points.**

251 The relationships between vapor  $\delta^{18}\text{O}$  and specific humidity ( $q$ ) further indicate seasonally contrasting  
252 moisture dynamics (Fig. 3a). Note that only data before 2018 are shown as local meteorological data are unavailable  
253 in 2018. In addition, the sampling frequency during 2018 was reduced (Fig. 1) due to logistic issues. Therefore,



254 when analyzing relationships between vapor isotope compositions ( $\delta^{18}\text{O}$  and  $d$ -excess) and meteorological variables  
255 (both locally and regionally) we only focused on data before 2018. For months during the non-monsoon season, the  
256 majority of data points fall above the Rayleigh distillation line but below the mixing line of an upper bound of  
257 hypothetical evapotranspiration over South Asia, especially for the winter months. In contrast, data for the summer  
258 monsoon season months predominately fall below the Rayleigh distillation line and are constrained by “super-  
259 Rayleigh” lines with different degrees of rain evaporation. The relationships between  $\delta \times q$  and  $q$  further indicate  
260 seasonal contrast moisture source signatures (Fig. S3). Distribution of non-monsoon season  $\delta \times q$  and  $q$  suggests  
261 the mixing between a dry end member that has almost totally dehydrated through condensation and a moist end  
262 member of surface evaporation or moisture that has been partially dehydrated through Rayleigh distillation (Fig.  
263 S3). A simple estimation through the linear regression between  $\delta \times q$  and  $q$  suggests  $\delta^{18}\text{O}$  of the moist end  
264 member for the non-monsoon season is  $-13.9\text{‰} \pm 0.6\text{‰}$ . The amount weighted annual mean precipitation  $\delta^{18}\text{O}$  at  
265 this site was about  $-14.5\text{‰}$  (Yao et al., 2013). However, the overall estimation for the summer monsoon season  
266 suggests  $\delta^{18}\text{O}$  of the moist end member is  $-30.9\text{‰} \pm 1.8\text{‰}$  which is much lower than the estimation for the non-  
267 monsoon season. This exceptionally low value requires an additional moisture source of rain evaporation that is  
268 much depleted in heavy isotopes than surface evapotranspiration and is consistent with the distribution of  $\delta^{18}\text{O}$ - $q$   
269 below the Rayleigh distillation line (Fig. 3a).

270 The relationships between vapor  $d$ -excess and  $q$  also suggest seasonal contrasts in moisture dynamics (Fig. 3b).  
271 During non-monsoon season months, vapor  $d$ -excess shows a negative correlation with  $q$ , and the highest  $d$ -excess  
272 values are generally associated with the driest and coldest air (Figs. 1 and 3b). However, vapor  $d$ -excess does not  
273 show a clear relationship with  $q$  and shows a substantial variability ( $\sim 20\text{‰}$  in range) at a given  $q$  during the summer  
274 monsoon season. These relationships suggest that vapor  $d$ -excess is less predictable by  $q$  than  $\delta^{18}\text{O}$ , except for low  
275 humidity levels.



276

277 **Figure 3. Relationships between vapor isotope compositions and specific humidity ( $q$ ) from 2015-2017.**

278 **(a) scatter plot of  $\delta^{18}\text{O}$  against specific humidity ( $q$ ). (b) scatter plot of  $d$ -excess against  $q$ . The months for the**

279 **data points are color-coded. The reference lines are the same as those in Fig. S2, and interpretations of these**

280 **reference lines are referred to Fig. S2 and section 2.3.**

### 281 3.2 Moisture sources and transport pathways for different seasons

282 To understand drivers of the seasonal contrasting moisture dynamics reflected in vapor isotope compositions,

283 we first analyzed the moisture sources and transport pathways during different seasons (Fig. 4). Note again that we

284 focus on the contribution by historical (last 10 days) air mass to humidity at SETP instead of moisture uptake from

285 the Earth surface. During the non-monsoon season, moisture is mainly transported by two branches with one from

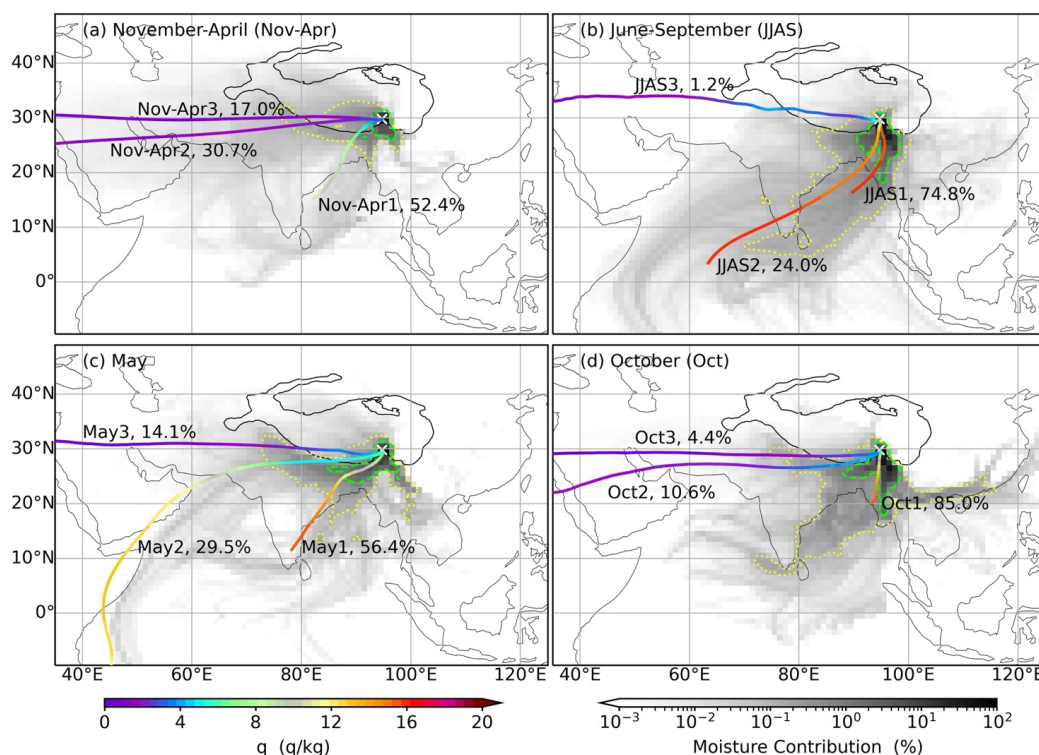
286 the west of SETP by the westerlies (clusters Nov-Apr2 and Nov-Apr3) and the other from the south of SETP such



287 as the BOB (cluster Nov-Apr1). Quantitatively, the fraction of moisture from the south pathway and the sum of the  
288 two west branches is comparable (52.4% vs. 47.7%). We note that if moisture contributions by air parcels are not  
289 considered, trajectories for all three clusters are from the west of SETP and they only reflect the transport of air  
290 masses (Fig. S4). These differences between pure air mass trajectories and considering moisture contribution by air  
291 masses call for caution when interpreting air mass trajectories. During the summer monsoon season, moisture is  
292 predominantly transported from the south of SETP by the summer monsoon. The moisture sources and transport  
293 pathways for May show some similarities with the results for the non-monsoon season. Compared with the second  
294 transport pathway during the non-monsoon (cluster Nov-Apr2), the second transport pathway during May (cluster  
295 May2) shows an overall southward shift toward the AS. Although the air mass transport pattern during October is  
296 also similar to that during the non-monsoon, the moisture sources and transport pathways for October show  
297 similarities with the results for the summer monsoon season with a slight eastward shift (Figs. 4d and S4d).

298 Another emerging feature of moisture source distributions is that humidity at SETP is predominantly  
299 contributed by air masses over proximal terrestrial regions, especially those regions in its south (Fig. 4). In contrast,  
300 air masses over oceanic regions make a much smaller contribution to humidity at SETP. For instance, the 1% contour  
301 of moisture contribution by air parcels over each  $1^{\circ} \times 1^{\circ}$  grid box does not reach oceanic regions during all four  
302 seasons. Therefore, moisture uptake of surface evaporation from oceanic regions, such as from the BOB and AS, is  
303 also very limited as most of the moisture in air masses over these oceanic regions is lost by precipitation and  
304 replenished by evapotranspiration during the transport toward SETP. This result questions whether vapor isotopic  
305 compositions at SETP still preserve the meteorological information at the ocean surface.





306

307 **Figure 4. Moisture sources and transport pathways during different seasons from 2015-2017. (a) spatial**  
 308 **distribution of relative contribution of moisture by all air parcels overall each  $1^\circ \times 1^\circ$  box (shading) to**  
 309 **humidity at the SETP station and specific humidity ( $q$ ) along mean trajectories (weighted by the moisture**  
 310 **contribution of air parcels) for the non-monsoon season of November-April (Nov-Apr). (b-d) are the same as**  
 311 **(a), but for the monsoon season of June-September (JJAS, b), May (c), and October (d), respectively. The**  
 312 **dotted yellow and dashed green contours indicate the moisture contribution at 0.1% and 1%, respectively.**  
 313 **The yellow crosses indicate the location of the SETP station. The black solid lines denote the Tibetan Plateau**  
 314 **with altitude contour at 3000 m.**



## 315 4 Discussion

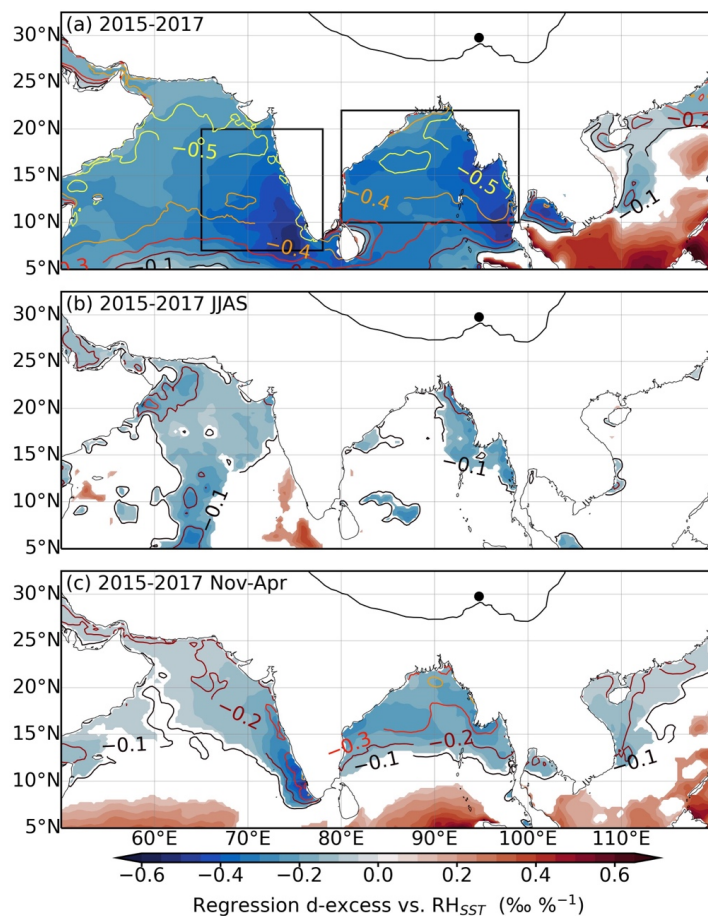
### 316 4.1 Role of ocean surface evaporation conditions

317 Relationships between vapor *d*-excess and ocean surface evaporation conditions of  $RH_{SST}$  and SST are first  
318 tested using all the data from 2015-2017 (Fig. 5a and Fig. S5a). Results indeed show negative correlations between  
319 vapor *d*-excess and  $RH_{SST}$  over northern Indian Ocean, especially the northern part of AS and BOB (Fig. 5a).  
320 Quantitatively, slopes of the regression between *d*-excess and  $RH_{SST}$  over the northern Indian Ocean range from higher  
321 than  $-0.1\% \%^{-1}$  to values below  $-0.6\% \%^{-1}$ . The regression slopes over the northern BOB (10-22°N and 80-99°E)  
322 and the eastern AS (7-20°N and 65-78°E; Fig. 5a) fall within the range reported in previous studies (Uemura et al.,  
323 2008; Benetti et al., 2014; Liu et al., 2014; Bonne et al., 2019). Vapor *d*-excess and regional average  $RH_{SST}$  yields  
324 an overall slope of  $-0.49\% \%^{-1}$  ( $r = -0.52$  and  $p < 0.01$ ) for the eastern AS (Fig. S6) and  $-0.52\% \%^{-1}$  ( $r = -0.55$  and  
325  $p < 0.01$ ) for the northern BOB (Fig. S7). However, the distribution of data in the *d*-excess and  $RH_{SST}$  space suggests  
326 a clustering of data that observations during summer months are mainly located in the lower right with high  $RH_{SST}$   
327 and low *d*-excess, but in the upper left part of the space for winter months. During each season, there is substantial  
328 variability in vapor *d*-excess for a given  $RH_{SST}$ . These results suggest that the apparent negative correlation between  
329 *d*-excess and  $RH_{SST}$  may mainly arise from their opposite seasonality. Similarly, apparent negative correlations  
330 between vapor *d*-excess and SST also emerge over the northern Indian Ocean (Fig. S5a). However, both theoretical  
331 prediction (Merlivat and Jouzel, 1979) and in-situ observations above the ocean surface (Bonne et al., 2019; Liu et  
332 al., 2014) suggest a positive correlation between vapor *d*-excess and SST. Therefore, we argue that the overall  
333 correlations between SETP vapor *d*-excess and surface evaporation conditions over the northern Indian Ocean are  
334 mainly a result of their seasonality and do not hold realistic causal relationships.

335 We further examined the relationship between vapor *d*-excess and  $RH_{SST}$  for the summer monsoon and non-  
336 monsoon seasons, respectively. The negative correlation between vapor *d*-excess and  $RH_{SST}$  almost totally



337 diminishes, especially during the summer monsoon season when absolute values of correlation coefficients drop to  
338 below 0.3 (Fig. 5b). Stronger correlations during the non-monsoon season (Fig. 5c) could be due to the overall  
339 intraseasonal variation that  $d$ -excess is the highest during winter and lower at the beginning and ending stages (Fig.  
340 1b) which could be accompanied with an opposite  $RH_{SST}$  trend. Even so, the correlations during the non-monsoon  
341 still only explain a marginal fraction of variance in  $d$ -excess (10%-16% at maximum over the northern BOB).  
342 Similarly, correlations with SST over the northern Indian Ocean also become trivial when separately considering  
343 the summer monsoon or non-monsoon season (Fig. S5). In summary, vapor  $d$ -excess at SETP is less likely a  
344 conservative tracer of surface evaporation conditions (neither  $RH_{SST}$  nor SST) over the northern Indian Ocean.  
345 Therefore, it should be cautious when interpreting  $d$ -excess in meteoric water or paleo archives from the TP as a  
346 proxy of evaporation conditions over the Indian Ocean.

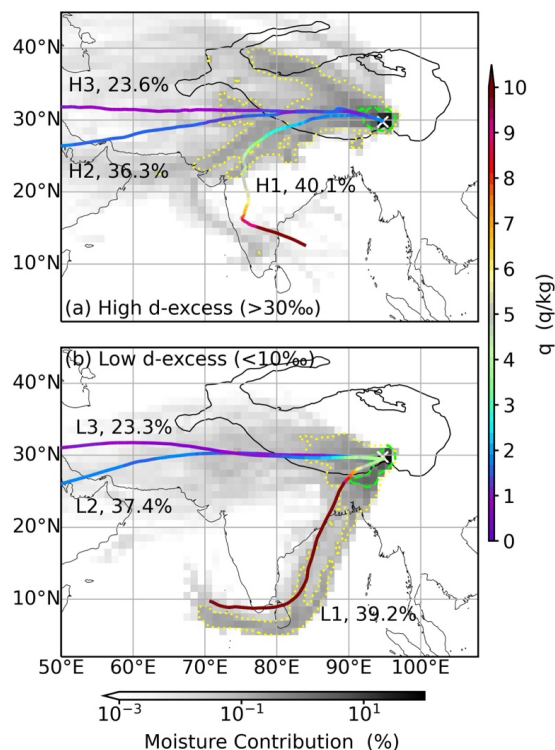


348 **Figure 5. Relationships between water vapor  $d$ -excess and relative humidity scaled to sea surface**  
349 **temperature ( $RH_{SST}$ ). (a) regression of vapor  $d$ -excess against  $RH_{SST}$  (shading and only values significant at**  
350 **the 95% significance level are shown) and correlation coefficients between them (contours at an interval of**  
351 **0.1 and only negative correlations are shown) for all the data from 2015-2017. (b) and (c) are the same as (a)**  
352 **but only for the data within the summer monsoon season (JJAS) or the non-monsoon season (Nov-Apr),**  
353 **respectively. The black dots indicate the location of the SETP station. The black solid lines denote the Tibetan**  
354 **Plateau with altitude contour at 3000 m.**



355 **4.2 Role of dry and cold air intrusion during the non-monsoon season**

356 Both theoretical predictions by the Rayleigh model and observations during the non-monsoon season suggest  
357 an increasing trend of vapor  $d$ -excess when  $q$  goes to extremely low values (Fig. 2). In addition, results for both air  
358 mass transport and moisture transport show the dominant role of the westerlies (Figs. S4a and 4a). Therefore, we  
359 hypothesize that the mixing of cold and dry air transported by the westerlies from higher altitudes with surface vapor  
360 controls vapor isotope compositions during the non-monsoon season. Surface vapor influenced by recycled moisture  
361 from terrestrial evapotranspiration would further elevate vapor  $d$ -excess at a given  $q$  (Fig. 3b). We first did a  
362 composite analysis on moisture sources and transport pathways for the highest (higher than 30‰ and  $n = 10$ ) and  
363 lowest  $d$ -excess observations (lower than 10‰ and  $n = 8$ ) during the non-monsoon season (Fig. 6). For high vapor  
364  $d$ -excess values, moisture is predominantly transported by westerlies from the west of SETP, such as over the TP  
365 and northwestern India. In addition, air masses along backward trajectories are characterized by extremely low  $q$   
366 i.e. as low as below  $2 \text{ g kg}^{-1}$  along the mean trajectories (weighted by the moisture contribution) over the TP (Fig.  
367 6a). For low  $d$ -excess cases, a substantial amount of moisture transport pathways (account for 39.2% by the L1  
368 cluster, Fig. 6b) shift toward more humid areas of northeast India, Bangladesh, and the BOB. This contrasting  
369 moisture transport pattern between high and low  $d$ -excess cases agrees with our hypothesis that the high  $d$ -excess  
370 is associated with dry and cold air transported by the westerlies.



371

372 **Figure 6. Composite of moisture sources and transport pathways for high and low  $d$ -excess days during**  
 373 **the non-monsoon season of November-April. (a) spatial distribution of relative contribution of moisture by**  
 374 **all air parcels overall each  $1^\circ \times 1^\circ$  box (shading) to humidity at the SETP station and specific humidity ( $q$ )**  
 375 **along mean trajectories (weighted by the moisture contribution of air parcels) for  $d$ -excess values higher than**  
 376 **30‰ during the non-monsoon season ( $n = 10$ ). (b) is the same as (a) but for  $d$ -excess lower than 10‰ ( $n = 8$ ).**  
 377 **The yellow crosses indicate the location of the SETP station. The black solid lines denote the Tibetan Plateau**  
 378 **with altitude contour at 3000 m.**

379 The relationship between vapor  $d$ -excess and the intrusion of cold and dry air is further tested by relationships  
 380 among vapor  $d$ -excess, local  $q$ , upstream  $q$ , upstream air temperature, and upstream air altitude (Fig. 7). Upstream  
 381 variables are mean values along the 10-day backward trajectory weighted by the moisture contribution of the air  
 382 parcel at each time step (section 2.4). The non-monsoon season vapor  $d$ -excess shows robust negative correlations



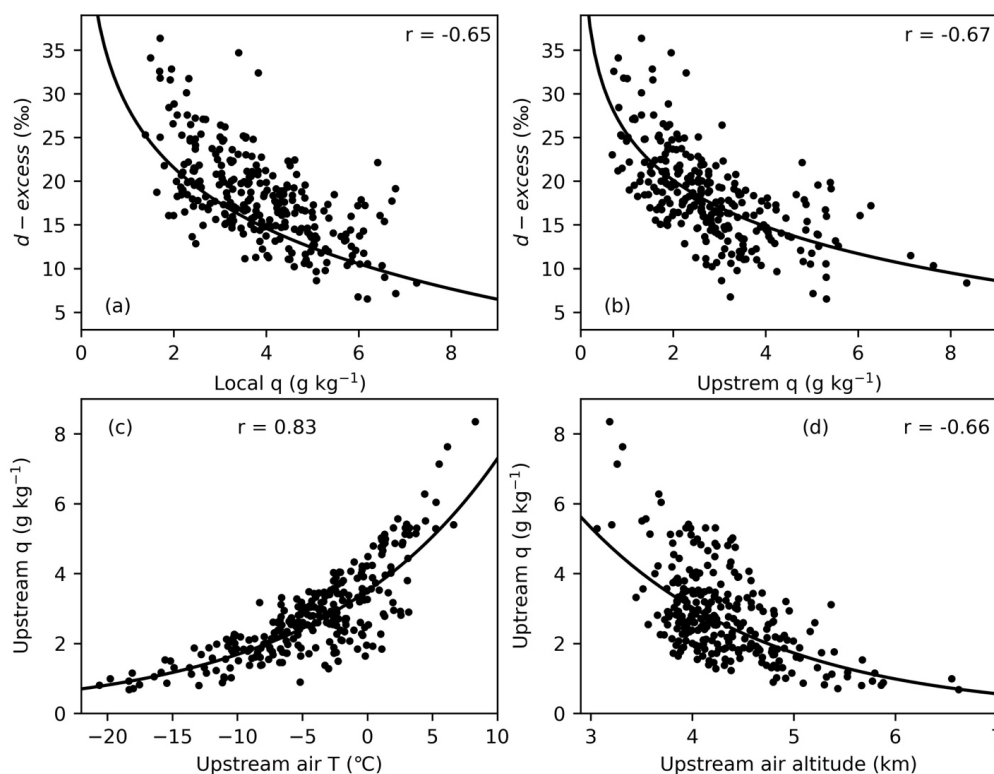
383 with  $q$  both at the local scale as well as at upstream ( $r = -0.65$  and  $-0.67$ , respectively, and  $p < 0.01$  for both). At the  
384 same time, low  $q$  is associated with air masses with low temperature and from high altitudes (Figs. 7c and 7d). This  
385 effect could also have an impact on vapor  $\delta^{18}\text{O}$ .  $\delta^{18}\text{O}$  during the high  $d$ -excess cases is lower than  $\delta^{18}\text{O}$  during the  
386 low  $d$ -excess cases (at a significance level of 95.3%), and the overall correlation coefficient between  $\delta^{18}\text{O}$  and  $d$ -  
387 excess during the non-monsoon season is  $-0.29$  ( $p < 0.01$ ). Correlations between  $\delta^{18}\text{O}$  and local  $q$  ( $r = 0.42$  and  $p <$   
388  $0.01$ ) or upstream  $q$  ( $r = 0.38$  and  $p < 0.01$ ) are weaker than the correlations between  $d$ -excess and  $q$ . The relationship  
389 between non-monsoon season  $\delta^{18}\text{O}$  and humidity is mainly expressed as the relationship between  $\delta \times q$  and  $q$  ( $r$   
390  $= 0.82$  for local  $q$  and  $r = 0.90$  for upstream  $q$ ).

391 We further analyzed the spatial distribution of correlations between SETP vapor isotope compositions ( $\delta^{18}\text{O}$   
392 and  $d$ -excess) and 2-meter air temperature as well as humidity measured by 2-meter dew point temperature during  
393 the non-monsoon season (Fig. 8). Results show significant negative correlations between  $d$ -excess and dew point  
394 temperature at the regional scale over southeastern TP, northeast India, and northern Bangladesh (Fig. 8a).  
395 Correlations with air temperature are generally similar with correlations between  $d$ -excess and dew point  
396 temperature but the most significant correlations are in a smaller region (Fig. 8b). In contrast, correlations between  
397  $\delta^{18}\text{O}$  and dew point temperature is not as strong as that for  $d$ -excess (Fig. 8c). Instead,  $\delta^{18}\text{O}$  shows stronger positive  
398 correlations with air temperature over the India subcontinent and the northwestern part of southeast Asia (Fig. 8d).

399 Extremely high  $d$ -excess values at very low  $q$  levels are predicted in Fig. 3b, and previous study has shown  
400 that as  $q$  approaches zero, vapor  $d$ -excess can approach 7000‰ following the Rayleigh distillation trajectory (Bony  
401 et al., 2008) caused by the definition of the  $d$ -excess (Dütsch et al., 2017). High vapor  $d$ -excess values have also  
402 been observed in low humidity conditions such as in the polar regions (Bonne et al., 2014; Steen-Larsen et al., 2017)  
403 or at high altitudes (Samuels - Crow et al., 2014; Webster and Heymsfield, 2003; Sayres et al., 2010; Sodemann et  
404 al., 2017). Therefore, we infer that the increasing trend of vapor  $d$ -excess along with decreasing local  $q$ , upstream



405  $q$ , and regional dew point temperature is a result of intensified mixing with dry and cold subsiding air transported  
 406 by the westerlies from high altitudes. Relationships between upstream  $q$  and upstream air temperature as well as  
 407 altitude support this inference that low humidity condition is associated with subsiding dry and cold air from high  
 408 altitudes (Figs. 7c and 7d). Therefore, vapor  $d$ -excess during the non-monsoon not only provides information on the  
 409 specific humidity but also indicates the source of humidity.



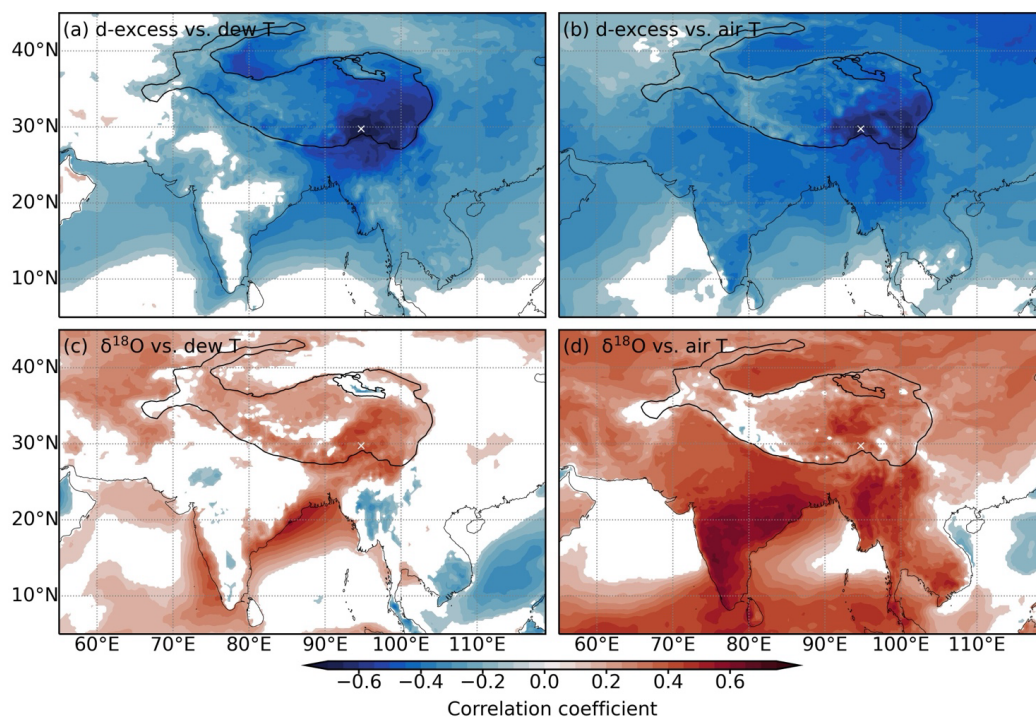
410

411 **Figure 7. Relationships among water vapor  $d$ -excess, local specific humidity ( $q$ ), upstream  $q$ , upstream**  
 412 **air temperature (T), and upstream air altitude during the non-monsoon season of November-April. (a) scatter**  
 413 **plot of  $d$ -excess against local  $q$ . (b) scatter plot of  $d$ -excess against upstream weighted-mean  $q$ . (c) scatter plot**  
 414 **of upstream  $q$  against upstream air T. (d) scatter plot of upstream  $q$  against upstream air altitude. All the**  
 415 **upstream variables are mean values along backward trajectories weighted by the moisture contribution of**  
 416 **air parcels. The solid curves indicate the log regression between the respective variables with the correlation**





417 coefficients indicated by the numbers.



418

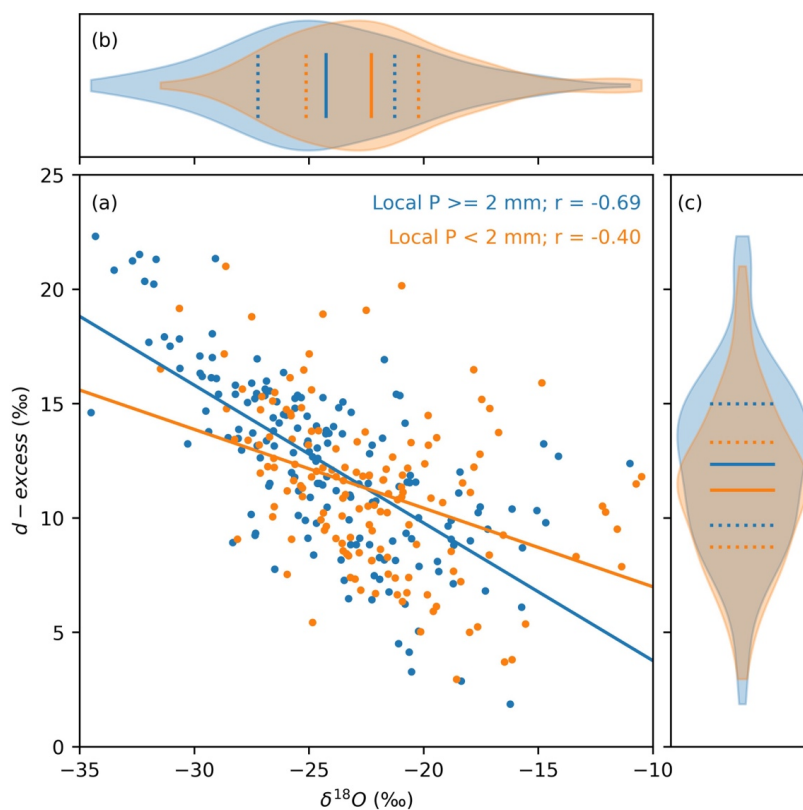
419 **Figure 8.** Spatial distribution of correlation coefficients among water vapor isotope compositions, dew  
 420 point temperature, and air temperature during the non-monsoon season of November-April. (a) spatial  
 421 distribution of correlation coefficients between SETP vapor *d*-excess and 2-meter dew point temperature. (b)  
 422 the same as (a) but with 2-meter air temperature. (c) the same as (a) but between  $\delta^{18}\text{O}$  and 2-meter dew point  
 423 temperature. (d) the same as (c) but with 2-meter air temperature. Only values significant at the 95%  
 424 significance level are shown. The white crosses indicate the location of the SETP station. The black solid lines  
 425 denote the Tibetan Plateau with altitude contour at 3000 m.

426 **4.3 Role of rain-vapor interaction during the summer monsoon season**

427 Different from the significant dependence of vapor *d*-excess on specific humidity during the non-monsoon  
 428 season, vapor *d*-excess is not correlated with specific humidity ( $r = 0.04$  and  $p = 0.51$ ) during the summer monsoon



429 season. The behavior of  $\delta^{18}\text{O}$  is also distinct during the two seasons (Fig. 3). Distribution of summer monsoon  
430 season observations in the  $\delta^{18}\text{O}$ - $q$  space suggests that the vapor has undergone a certain degree of rain-vapor  
431 interaction by rain evaporation (Fig. 3a). On the other hand, partial rain evaporation in an unsaturated atmospheric  
432 environment is associated with kinetic fractionation which decreases the  $d$ -excess values of the raindrop but  
433 increases the  $d$ -excess of surrounding vapor (Risi et al., 2008b). This effect of rain-vapor interaction on vapor isotope  
434 compositions has been suggested as a major process responsible for the amount effect in the tropics (Risi et al.,  
435 2008a; Kurita et al., 2011; Bowen et al., 2019; Galewsky et al., 2016). Therefore, we hypothesize that vapor isotope  
436 compositions during the summer monsoon season at SETP are controlled by the degree of rain-vapor interaction.  
437 The first evidence supporting this hypothesis is that vapor  $\delta^{18}\text{O}$  is significantly correlated with  $d$ -excess during the  
438 summer monsoon season ( $r = -0.55$  and  $p < 0.01$ , Fig. 9a). In addition, there is a trend that vapor  $\delta^{18}\text{O}$  and  $d$ -excess  
439 are less correlated when  $\delta^{18}\text{O}$  is high and the opposite for low  $\delta^{18}\text{O}$  levels (Figs. 9a and S8). If there is no rain, rain-  
440 vapor interaction is not possible. Therefore, we flitted data during days when the daily precipitation amount is not  
441 less than 2 mm as rainy days and days with precipitation less than 2 mm as no rain occurs locally (non-rainy days).  
442 Vapor  $\delta^{18}\text{O}$  during rainy days is significantly higher than during non-rainy days and the opposite trend applies for  
443  $d$ -excess ( $p < 0.01$  for both  $\delta^{18}\text{O}$  and  $d$ -excess) (Figs. 9b and 9c). The correlation between vapor  $\delta^{18}\text{O}$  and  $d$ -excess  
444 during rainy days becomes stronger ( $r = -0.69$  and  $p < 0.01$ ). However, vapor  $\delta^{18}\text{O}$  is still negatively correlated with  
445  $d$ -excess during non-rainy days ( $r = -0.40$  and  $p < 0.01$ ). Even if a stricter threshold of 0 mm for daily precipitation  
446 amount is used for flittering non-rainy days, there is still a significant negative correlation between vapor  $\delta^{18}\text{O}$  and  
447  $d$ -excess ( $r = -0.37$  and  $p < 0.01$ ). In addition, correlations with local precipitation amount are weak both for  $\delta^{18}\text{O}$   
448 ( $r = -0.31$  and  $p < 0.01$ ) and  $d$ -excess ( $r = 0.26$  and  $p < 0.01$ ). Therefore, we further infer that the effect of rain-vapor  
449 interaction is not only from the local scale but also inherits the history of rain-vapor interaction before vapor has  
450 been transported to SETP.



451

452 **Figure 9. Relationships between SETP vapor  $d$ -excess and  $\delta^{18}\text{O}$  during the summer monsoon season. (a)**

453 **scatter plot of  $d$ -excess against  $\delta^{18}\text{O}$  and linear regression lines between them. (b) distribution of  $\delta^{18}\text{O}$  values**

454 **with the dashed lines indicate values at the lower and upper quartiles and the solid lines indicate the mean**

455 **values. (c) is the same as (b) but for  $d$ -excess. Orange colors indicate data observed during daily precipitation**

456 **amount less than 2 mm and blue colors indicate data observed during days with precipitation amount not**

457 **less than 2 mm.**

458 If there is a larger amount of rainfall, the effect of rain-vapor interaction on atmospheric humidity would be

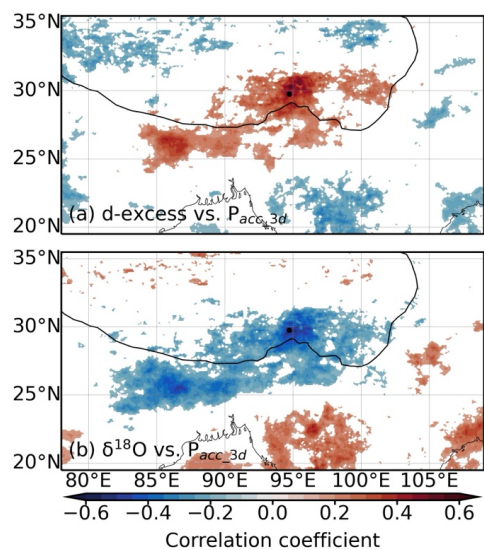
459 stronger. Therefore, we use total precipitation amount ( $P_{acc}$ ) as a measure of rain-vapor interaction. To account for

460 the history during moisture transport, the total precipitation amount during several days before sampling is

461 considered. We have tested the relationships between vapor isotope compositions ( $\delta^{18}\text{O}$  and  $d$ -excess) and  $P_{acc}$  over



462 1-10 days prior to sampling (Figs. S9 and S10). Vapor *d*-excess reaches an optimal correlation with  $P_{acc}$  when the  
463 total precipitation amount during 3 days before sampling ( $P_{acc\_3d}$ ) is considered. Vapor  $\delta^{18}O$  shows a slightly longer  
464 memory and reaches an optimal correlation with  $P_{acc}$  when the total precipitation amount during 5-6 days before  
465 sampling is considered. Fig. 10 shows the spatial distribution of correlations between vapor isotope compositions  
466 ( $\delta^{18}O$  and *d*-excess) and  $P_{acc\_3d}$ . Vapor *d*-excess shows significant positive correlations with  $P_{acc\_3d}$  in the region  
467 surrounding SETP with a spatial scale of  $\sim 5^\circ \times 5^\circ$  and the positive correlation extends southwestward to the foothill  
468 of the Himalayas (Fig. 10a). In contrast, vapor  $\delta^{18}O$  shows significant negative correlations with  $P_{acc\_3d}$  in similar  
469 regions (Fig. 10b). For non-rainy days, vapor  $\delta^{18}O$  and *d*-excess still show significant correlations with  $P_{acc\_3d}$  at  
470 regional scale, albeit with weaker correlation levels and smaller spatial extent (Fig. S11). These significant  
471 correlations among vapor  $\delta^{18}O$ , *d*-excess, and  $P_{acc\_3d}$  provide further evidence for understanding processes that are  
472 responsible for the amount effect. The negative correlation between  $\delta^{18}O$  and  $P_{acc\_3d}$  has also been observed in  
473 precipitation and can be interpreted in terms of either continuous rainout (Cai and Tian, 2016; Scholl et al., 2009;  
474 Vuille et al., 2003) or the effect of rain-vapor interaction (Lawrence et al., 2004; Risi et al., 2008a; Kurita et al.,  
475 2011; Worden et al., 2007). Although continuous rainout with increased rainfall amount can explain the decreasing  
476 trend of  $\delta^{18}O$  by the Rayleigh distillation model, *d*-excess stays at a relatively stable level when specific humidity  
477 is not very low (above  $\sim 4 \text{ g kg}^{-1}$  in Fig. 3b for example). Therefore, the positive correlation between vapor *d*-excess  
478 and  $P_{acc\_3d}$  provides an additional constraint that the amount effect is not simply a result of rainout but rain-vapor  
479 interaction plays an important role in altering lower tropospheric isotope compositions.



480

481 **Figure 10. Relationships between vapor isotope compositions for rainy days (local daily precipitation**  
482 **amount not less than 2 mm) and total precipitation amount at the regional scale during the summer monsoon**  
483 **season. (a) spatial distribution of correlation coefficients between vapor *d*-excess and total precipitation**  
484 **amount during 3 days prior sampling ( $P_{acc\_3d}$ ). (b) is the same as (a) but for  $\delta^{18}\text{O}$ . Only values significant at**  
485 **the 95% significance level are shown. The black dots indicate the location of the SETP station. The black**  
486 **solid lines denote the Tibetan Plateau with altitude contour at 3000 m.**

#### 487 4.4 An alternative interpretation for the high *d*-excess in high-altitude TP ice cores

488 Interpretations of *d*-excess in meteoric water and ice cores on the TP are complicated by evaporation conditions  
489 over the northern Indian Ocean ( $RH_{SST}$  and SST) and continental recycling (Shao et al., 2021; Zhao et al., 2012;  
490 Joswiak et al., 2013; Pang et al., 2012; An et al., 2017). Attempts have been made to establish a relationship between  
491 vapor *d*-excess and  $RH_{SST}$  (Chen et al., 2024; Liu et al., 2024) as well as between ice core *d*-excess and  $RH_{SST}$  (Shao  
492 et al., 2021) or SST (Zhao et al., 2012). Based on relationships between vapor *d*-excess and surface evaporation  
493 conditions discussed above, however, the apparent relationships are mainly a result of similarities in the seasonality



494 of these variables. Furthermore, the direct contribution of water vapor contained in air masses over oceanic regions  
495 to humidity at SETP is very limited (Fig. 4), which implies that the contribution to humidity over the TP is further  
496 decreased than at SETP as it is at the forefront of moisture transport toward TP (Fig. S1). The dominant terrestrial  
497 origin indicates that the moisture has undergone a certain degree of continental recycling. Mixing with terrestrial  
498 sources is also reflected in the relationship between isotope compositions and  $q$  (Fig. 3). The degree of continental  
499 recycling also alters vapor isotope compositions that transpiration introduces isotopically enriched moisture and  
500 evaporation introduces moisture with high  $d$ -excess. Seasonally changing isotope signatures in precipitation and ice  
501 cores as well as variations at longer timescales have been interpreted as moisture source shift between recycled  
502 moisture over terrestrial regions and oceanic moisture sources or their relative contributions (An et al., 2017; Yang  
503 and Yao, 2020). A further inference of this process is that the oceanic moisture is brought by the summer monsoon  
504 while the westerlies bring moisture from continental recycling or even the Mediterranean Sea, and therefore water  
505 isotope signatures reflect the interplay between the summer monsoon and westerlies (Joswiak et al., 2013; Pang et  
506 al., 2012; Tian et al., 2007). Although our results also indicate seasonally shifting moisture sources, continental  
507 recycling prevails throughout the year (Fig. 4). Besides focusing on moisture sources at the Earth surface, we  
508 provide an alternative perspective to explain the high  $d$ -excess induced by the westerlies as dry and cold air  
509 intrusions. In this circumstance, the interpretation of the interplay between the summer monsoon and westerlies is  
510 still valid, but we emphasize changes in air mass property driven by the different circulation systems.

511 The proposed alternative interpretation could also help understand the increasing trend of precipitation and  
512 river water isotope observations toward ice cores at higher altitudes on the TP as specific humidity is very low at  
513 ice core sites and prolonged interaction with cold and dry air may further modify snow isotope compositions (Ma  
514 et al., 2024; Wahl et al., 2022). In addition, intense rain-vapor interaction during the summer monsoon is another  
515 source of higher vapor  $d$ -excess (section 4.3). Higher vapor  $d$ -excess signal could be inherited in subsequent



516 precipitation when it feeds the precipitation (Risi et al., 2008b). However, a clear relationship between TP  
517 precipitation *d*-excess and monsoon convection has not been established yet, partly due to less attention has been  
518 paid to *d*-excess in previous studies. Nevertheless, summer monsoon rainfall *d*-excess observed on the TP is  
519 generally between 0-10‰ (Tian et al., 2001). Raindrop evaporation at upstream increases vapor *d*-excess and  
520 therefore could cause elevated *d*-excess in downstream rainfall. On the contrary, this effect can be compensated by  
521 on-site raindrop evaporation as it lowers raindrop *d*-excess values. The overall positive correlation between  
522 precipitation *d*-excess and altitude in Asia has been sometimes interpreted as stronger evaporation at lower altitudes  
523 (Bershaw, 2018). For snowfall on glaciers, evaporation is less likely for falling snowflakes due to cold temperatures  
524 and the short distance between the cloud base and the glacier surface. Therefore, elevated vapor *d*-excess signal  
525 caused by accumulated rain-vapor interaction at upstream associated with monsoon convection could be another  
526 source for the high *d*-excess in ice cores.

## 527 **5 Conclusions**

528 We present a three-year-long daily near-surface water vapor isotope compositions observed at the South-East  
529 TP station which is at the major channel for moisture entering the TP. Our vapor isotope compositions paired with  
530 specific humidity reflect distinct moisture sources and dynamics between the non-monsoon and summer monsoon  
531 seasons, consistent with Lagrangian moisture diagnostic results. Despite significant negative correlations between  
532 vapor *d*-excess and relative humidity scaled to sea surface temperature existing over the northern Indian Ocean  
533 when data for all seasons are considered, such correlations with oceanic surface evaporation conditions largely  
534 disappear when separately considering each season. This result questions the early interpretation of TP *d*-excess as  
535 oceanic evaporation conditions and guarantees new interpretations in the future.

536 During the non-monsoon season, vapor *d*-excess is mainly influenced by specific humidity both at the local



537 scale and upstream. Highly dehydrated air at the lower end of the Rayleigh distillation is expected to have extremely  
538 high *d*-excess values. Air mass trajectory analyses and moisture source diagnostics suggest that the cold and dry air  
539 intrusion driven by the westerlies during the non-monsoon season leads to the increasing trend of vapor *d*-excess  
540 along with decreasing specific humidity. This process also contributes to a weak negative correlation between vapor  
541 *d*-excess and  $\delta^{18}\text{O}$ . Furthermore, vapor  $\delta^{18}\text{O}$  primarily reflects mixing processes with a relatively enriched moist  
542 end-member compared with the summer monsoon season. The new insight on non-monsoon season vapor *d*-excess  
543 provides an alternative way to interpret the high *d*-excess in high-altitude TP ice cores.

544       During the summer monsoon season, rain evaporation is the dominant process determining water vapor isotope  
545 compositions. First, vapor  $\delta^{18}\text{O}$  systematically shifts below the Rayleigh distillation curve falling in the region  
546 predicted by “super-Rayleigh” distillation driven by partial rain evaporation. Second, vapor  $\delta^{18}\text{O}$  is anti-correlated  
547 with *d*-excess pointing to an origin of depleted vapor by kinetic fractionation which is not likely simply a result of  
548 rainout. Third, vapor  $\delta^{18}\text{O}$  is significantly negatively correlated with total precipitation amount at the regional scale,  
549 but vapor *d*-excess positively correlates with total precipitation amount. These results help us understand the  
550 dynamics of atmospheric humidity and also help disentangle the different effects of rainout and rain-vapor  
551 interaction in the amount effect.

552       Overall, the new findings from the study reveal different moisture sources and dynamics between the non-  
553 monsoon and monsoon seasons over the southeastern TP. The findings will also help the interpretation of ice core  
554  $\delta^{18}\text{O}$  and *d*-excess records derived from glaciers on the TP.

#### 555 **Competing interests**

556       The authors declare that they have no conflict of interest.





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564 **Data availability**

565 The NOAA ARL provided the HYSPLIT model and the GDAS data  
566 (<https://www.ready.noaa.gov/HYSPLIT.php>). The Copernicus Climate Change Service provided the ERA5 data  
567 (<https://doi.org/10.24381/cds.adbb2d47> and <https://doi.org/10.24381/cds.fl7050d7>). The GPM data are available  
568 through GES DISC (<https://doi.org/10.5067/GPM/IMERG/3B-HH/07>). Local meteorological data at the SETP  
569 station are provided by National Tibetan Plateau / Third Pole Environment Data Center  
570 (<https://dx.doi.org/10.11888/AtmosphericPhysics.tpe.68.db>). The observation data at the SETP station have been  
571 uploaded to Figshare and will be made publicly available after publication (10.6084/m9.figshare.27302871).

572 **Author contributions**

573 **Zhongyin Cai**: Conceptualization, methodology, investigation, formal analysis, funding acquisition, writing-  
574 original draft, writing-review & editing; **Rong Li**: Investigation, data curation, writing-review & editing; **Cheng**  
575 **Wang**: Validation; **Qiukai Mao**: Investigation, **Lide Tian**: Resources, project administration, funding acquisition.



576 **References**

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