The patterns and implications of diurnal variations in d-excess of plant water, shallow soil water and air moisture

Running title: Diurnal variations in d-excess of different water pools

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Abstract

Deuterium excess (d-excess) of air moisture is traditionally considered a conservative tracer of oceanic evaporation conditions. Recent studies challenge this view and emphasize the importance of vegetation activity in controlling the dynamics of air moisture d-excess. However, direct field observations supporting the role of vegetation in d-excess variations are not well documented. In this study, we quantified d-excess of air moisture, shallow soil water (5 cm and 10 cm), and leaf and xylem water of multiple dominant species at hourly interval during three extensive field campaigns at two climatically different locations within the Heihe River Basin, northwestern China. The ecosystems in the two locations range from forest to desert. The results showed that with the increase of temperature (T) and decrease of relative humidity (RH), the δD-$\delta^{18}O$ regression lines of leaf water, xylem water and shallow soil water deviated gradually from their corresponding local meteoric water line. There were significant differences in d-excess values among different water pools at all the study sites. The most positive d-excess values were found in air moisture (9.3‰) and the most negative d-excess values were found in leaf water (-85.6‰). The d-excess values of air moisture ($d_{moisture}$) and leaf water (d_{leaf}) during the sunny days, and shallow soil water (d_{soil}) during the first sunny day after rain event showed strong diurnal patterns. There were significantly positive relationships between d_{leaf} and RH and negative relationships between $d_{moisture}$ and RH. The correlations of d_{leaf} and $d_{moisture}$ with T were opposite to their relationships with RH. In addition, we found opposite diurnal variations for d_{leaf} and $d_{moisture}$ during the sunny days, and for d_{soil} and $d_{moisture}$ during the first sunny day after rain event. The steady state Craig-Gordon model captured the diurnal variations of d_{leaf} with small discrepancies on the magnitude. Overall, this study provides a comprehensive and high-resolution dataset of d-excess of air moisture, leaf, root, xylem and soil water. Our results provide direct evidence that $d_{moisture}$ of the surface air at continental locations can be significantly altered by local processes, especially plant transpiration during the sunny days. The influence of shallow soil water on $d_{moisture}$ is generally much smaller compared with that of plant transpiration but the influence could be large in a sunny day right after rainfall events.

Key words: ecohydrology, deuterium excess, arid regions, the Heihe River Basin, hydrogen isotope, oxygen isotope

1. Introduction

Measurements of water isotopic compositions (e.g., δD, $\delta^{18}O$) provide insights into the study of hydrologic cycle, ecological processes, and palaeoclimate across multiple temporal and spatial scales (e.g., Brunel et al., 1992; Gat, 1996; Dawson et al., 2002; Newman et al., 2010; Wang et al., 2010; Zhang et al., 2011; Good et al., 2012; Wang et al. 2013). Plant uptake does not fractionate source water (White et al., 1985), δD or $\delta^{18}O$ therefore can be used to track plant water source (Ehleringer and Dawson, 1992), to investigate relative rooting depth (Jackson et al., 1999) and to identify hydraulic redistribution (Dawson, 1993). Water isotopes can also be used to trace catchment water movements (Brooks et al., 2010), the geographic origin of water vapor (Clark and Fritz, 1997), basin-level water recycling (Salati et al., 1979), and to reconstruct past environmental parameters such as ambient temperature (T) and relative humidity (RH) (e.g., Helliker and Richter, 2008). The isotopic compositions of water from different areas are affected by specific meteorological processes, which provide a characteristic fingerprint of their origin (Clark and Fritz, 1997). Much work has focused on isotopic compositions of surface water (Zhao et al., 2011b), groundwater (Zhao et al., 2012) and precipitation (Dalai et al., 2002; Karim and Veizer, 2002; Zhao et al., 2011b; Soderberg et al., 2013). However, fewer investigations were conducted to simultaneously measure δD and $\delta^{18}O$ of leaf water, xylem water, shallow soil water and air moisture, especially on the diurnal variations of these pools at ecosystem scale.

Deuterium-excess (d-excess) is defined as d-excess = $\delta D - 8.0 \times \delta^{18}O$ (Dansgaard, 1964). Points that fall on the Global Meteoric Water Line (GMWL) have a constant d-excess of 10.0‰. This is because rainout isotopic fractionation is considered an equilibrium process, which affects the position of the data points on the GMWL, but does not affect the intercept - d-excess. Since the effect of equilibrium Rayleigh condensation processes roughly follows the GMWL slope of 8, variations in d-excess can provide information about the environmental conditions (e.g., RH and T) during non-equilibrium processes in oceanic moisture source regions. In other words, d-excess is considered as a conservative tracer of oceanic evaporation conditions, assuming there are no contributions from surface evapotranspiration as the air mass travels over land (Welp et al., 2012). Therefore d-excess is used to identify the location of moisture source when there are no contributions from surface evapotranspiration (Uemura et al., 2008). Transpiration does not change source water d-excess since transpiration does not fractionate source water. Evaporation, however, usually results in a higher d-excess value (Gat et al., 1994). D-excess has been used to estimate evaporation in previous studies. For example, d-excess was used to quantify sub-cloud evaporation in Alpine regions (Froehlich et al., 2008) and to estimate the contribution of evaporation from the Great Lakes to the continental atmosphere (Gat et al., 1994).

By using meta-analysis approach to synthesize d-excess measurements from multiple sites, Welp et al. (2012) showed that d-excess value of surface atmospheric vapor can be significantly altered by local processes and it is not a conserved tracer of humidity from the marine moisture source region as previously assumed. In addition, modeling simulation also showed that plant transpiration plays an important role in diurnal d-excess variations (Welp et al., 2012), which contradicts with the conventional understanding. Based on isotopic observations from a US Pacific northwest temperate forest and modeling exercise, Lai and Ehleringer (2011) concluded that atmospheric entrainment appears to drive the isotopic variation of water vapor in the early morning when the convective boundary layer develops rapidly, while evapotranspiration becomes more important in mid-afternoon as a primary moisture source of water vapor in the studied forest. These authors therefore also casted some doubts on whether continental water vapor d-excess can be used as a conserved tracer of environmental conditions during evaporation at the moisture source. Despite these new understanding of biological and environmental controls on d-excess variations, field observations of the role of direct vegetation effect on diurnal d-excess variations are not readily seen in literature. In addition, Merlivat and Jouzel (1979), one of the few who theoretically calculated the quantitative relationship between d-excess of evaporating vapor with T and RH, predicted that d-excess is affected by both T and RH, and d-excess of evaporating vapor increases with T (0.35‰/ºC) but decreases with RH (-0.43‰/%) (Merlivat and Jouzel, 1979). Field-testing of such theoretical relationship is lacking. The quantitative relationship will enhance our prediction of climatic and environmental change impact (e.g., changes in T, RH, rainfall and location) on water cycles. Furthermore, it is unclear whether a consistent d-excess-RH relationship, similar to the d-excess-RH relationship of ocean evaporation, exists in evapotranspiration. Evapotranspiration from the earth surface is a key process in the hydrological cycle connecting the earth surface and the atmosphere. Therefore, it is essential to study the evapotranspiration process and its link to the atmospheric circulation in order to better understand the feedbacks between the earth surface and the atmosphere (Aemisegger et al., 2013).

In this study, we quantified the d-excess dynamics of air moisture, shallow soil water (5 cm and 10 cm), and leaf and xylem water of multiple dominant species at hourly interval during three extensive field campaigns at two climatically different locations in the Heihe River Basin, China. We aim to provide a field-based fine-resolution d-excess record and to explore the underlying mechanisms. The questions we addressed in this study are 1) what's the diurnal patterns of d-excess in air moisture, leaf, root, xylem and shallow soil water under different climatic and meteorological conditions? 2) what are the mechanisms of the observed patterns and their controlling factors? and 3) how well do the widely used steady state models capture the leaf d-excess dynamics?

2. Materials and Methods

2.1 Sampling sites

The field sampling took place at two locations (Dayekou and Ejin) with distinct climatic conditions within the Heihe River Basin (HRB), northwestern China (Fig. 1). The temperature is lowest in January, and is highest in July in both Dayekou (Zhao et al., 2011a) and Ejin. Dayekou is located at the upper reaches (Fig. 1). The mean annual temperature of Dayekou is about 0.7°C, with a mean January temperature of -12.9°C and a mean July temperature of 12.2°C. Mean annual precipitation is 369.2 mm, with over 71% of the rainfall occurring between June and September, and the rainfall in July is the highest. Ejin is located at the lower reaches (Fig. 1). The mean annual temperature of Ejin is 8.8°C, with a mean January temperature of -11.3°C and a mean July temperature of 26.8°C. Mean annual precipitation from 1960 to 2007 is 35.0 mm year^{-1}, with 75% of the rainfall occurring between June and September. With a strong potential evapotranspiration of 3700 mm (Gong et al., 2002), Ejin is considered one of the driest regions in China. At Dayekou, three sites were selected with two sites (S1-Sep/S1-Jun and S2-Jun) at the Pailugou valley and the other (S3-Aug) at the Guantan valley. The site names were assigned based on the combination of location and sampling time. The S1 (100°18'E, 38°33'N, 2900 m) was dominated by tree species *Qinghai Spruce* (Q.S.), shrub species *Potentilla fruticosa* (P.F.), and grass species *Polygonum viviparum* (P.V.). The S2 (100°17'E, 38°33'N, 2700 m) was dominated by tree species Q.S., and grass species *Stipa capillata* (S.C). The S3 (100°15′E, 38°32′N, 2800 m) was dominated by tree species Q.S. Two sites were selected at Ejin, one is at the riparian forest (S4-Aug: 101°14'E, 42°01'N, 930 m) with dominant tree species of *Populus euphratica* (P.E.) and shrub species *Sophora alopecuroides* (S.A.), another is at the Gobi (S5-Aug: 101°07'E, 42°16'N, 906 m) with the main shrub species of *Reaumuria soongorica* (R.S.) (Table 1).

Insert Table 1 here

Insert Figure 1 here

2.2 Plant and soil sample collections

Three extensive field samplings were conducted in August 2009 and in June and September 2011 in the upper and lower reaches of the HRB (Table 1). In the upper reaches, at the S1-Jun, samples were taken from 6:00 June 23 to 18:00 June 25 2011 with 1-hour intervals for leaf and stem of Q.S., 5 cm and 10 cm soil as well as atmospheric vapor near the ground (about 20 cm above the ground) and at the canopy. Leaf and stem of P.F. as well as leaf and root of P.V. were taken from the same period with 2-hour intervals. All these samples were refereed as S1-Jun. In S1-Sep, samples were taken from 8:00 September 6 to 17:00 September 8 2011 with 1-hour interval for leaf and stem of Q.S., 5 cm and 10 cm soil and atmospheric vapor near the ground and at the canopy. Leaf and stem of P.F. as well as leaf and root of P.V. were taken from the same period with 2-hour intervals. At the S2-Jun, leaf and stem of Q.S., 5 cm and 10 cm soil and atmospheric vapor near the ground and at the canopy were sampled from 6:00 June 27 to 18:00 June 28 2011 with 1-hour intervals, while leaf and root of S.C. were taken from 6:00 June 27 to 18:00 June 28 2011 with 2-hour intervals. At the S3-Aug, it rained twice during the sampling period (from 17:00 July 31 to 4:00 August 1 and from 10:40 to 20:00 August 2 2009). Leaf and stem of Q.S. as well as 5 cm and 10 cm soil samples were taken from 6:00 August 1 to 18:00 August 2 and from 6:00 to 18:00 August 3 2009 with 2-hour intervals. The atmospheric vapor at canopy was collected from 6:00 August 2 to 18:00 August 3 2009 with 2-hour intervals (Table 1).

In the lower reaches of the HRB, at the S4-Aug, leaf and stem of P.E. and leaf of S.A., 10 cm soil and atmospheric vapor at canopy were taken from 6:00 August 6 to 22:00 August 9 2009 with 2-hour intervals. At S5-Aug, leaf and stem of R.S., 10 cm soil and atmospheric vapor at canopy were taken from 18:00 August 10 to 18:00 August 12 2009 with 2-hour intervals. When samples were taken during rainy day and morning, napkins were used to wipe off water from the leaf and stem surfaces (Table 1).

For the soil, leaf and stem samples, samples from two 8 ml bottles were used to extract water and measure δD and $\delta^{18}O$. All samples were frozen in the Linze and Ejin field stations right after sampling and then transferred back to the laboratory for water extraction. Water samples were extracted from leaves, stems, roots and soil by a cryogenic vacuum distillation line (Zhao et al., 2011b). The extracted water was frozen into a collection tube.

2.3 Air moisture collection

We used a method similar to Wang and Yakir (2000) for short-term sampling of ambient air moisture at different locations such as Qinghai Spruce forest (S1-Sep, S1-Jun, S2-Jun and S3-Aug) in the upper reaches, riparian forest (S4-Aug) and the Gobi (S5-Aug) in the lower reaches. At the S1-Sep, S1-Jun and S2-Jun, the sampling of air moisture was collected within a canopy and near ground (about 20 cm above the ground). At S3-Aug, S4-Aug and S5-Aug, the sampling of air moisture was collected within a canopy (Fig. 1 and Table 1). Air was sucked by a small diaphragm pump through low adsorption plastic tubes and a small cryogenic trap at -80^{o}C at a rate of about 250 ml·min^{-1} for about 50 min. Pump and traps were located on the ground downwind of the sampling site and all the tubing was flushed with sample air before the actual trapping. After sampling, liquid water was transferred from traps to 2 ml glass bottles and transported to the laboratory for δ^{18}O and δD analysis.

2.4 Isotope analysis

The δ^{18}O and δD values of the water samples were measured using Euro EA3000 element analyzer coupled to an Isoprime Isotope Ratio Mass Spectrometer (Isoprime Ltd, UK) at Heihe Key Laboratory of Ecohydrology and River Basin Science, Cold and Arid Regions Environmental and Engineering Research Institute. To avoid the memory effect associated with continuous-flow methods, measurements of each sample were repeated five times, and the first values were discarded. The accuracy was better than ±1.0‰ for δD and ±0.2‰ for δ^{18}O. The δ^{18}O and δD were calibrated using two international standard materials (*V-SMOW* and *GISP* or *SLAP*) and one working standard. The δ^{18}O and δD values are expressed in ‰ on a *V-SMOW- SLAP* scale.

2.5 Meteorological measurements

During each study period, RH, T and photosynthetically available radiation (PAR) were measured due to their significant effects on soil evaporation and transpiration. At S3-Aug, T, RH and PAR were measured every 30 min with a weather station permanently installed at the station (HMP45C for measuring T and RH, LI190SB for measuring PAR) at 2 m, 10 m and 24 m height. At S1-Sep, S1-Jun, S2-Jun, S4-Aug and S5-Aug, RH, T and PAR were measured every 10 min with a portable weather station (Davis Vantage Pro2 portable weather station) at 2 m. Only 2 m height weather data such as T, RH and PAR were used in this study.

2.6 Modeling leaf water $\delta^{18}O$, δD and d-excess

Leaf water isotope enrichment is conventionally described by the steady state Craig and Gordon equation (Craig and Gordon, 1965) and non-steady state forms have also been proposed to account for the less enriched leaf water condition predicted by steady state model (e.g., Cuntz et al. 2007; Farquhar et a. 2007). To test whether we could use the current understanding of leaf water enrichment to reproduce the observed d-excess variations in leaf water, we used steady state Craig and Gordon model to estimate leaf water $\delta^{18}O$ and δD values, then calculate d-excess values using d-excess = $\delta D - 8.0 \times \delta^{18}O$ (Dansgaard, 1964). Only leaf water $\delta^{18}O$, δD and d-excess values of P.E. at S5-Aug were modeled for this study because it was sunny and had the most complete dataset through that entire study period. The leaf water enrichment ($\delta_{l,s}$) is calculated as

$$\delta_{l,s} \approx \delta_x + \varepsilon_{eq} + \varepsilon_k + h(\delta_v - \varepsilon_k - \delta_x),$$

where δ_x represents the $\delta^{18}O$ or δD values of liquid water at the evaporating front, we estimated δ_x using the isotopic composition of xylem water. δ_v is the $\delta^{18}O$ or δD values of the background atmospheric water vapor, $\alpha^*(>1)$ is the temperature-dependent equilibrium fractionation factor between liquid and vapor, $\varepsilon_{eq} = 1000(1-1/\alpha^*)$, α_k is the kinetic fractionation associated with diffusion of water through the soil and $\varepsilon_k = 1000(\alpha_k - 1)$, 1.0189 (~19‰) for oxygen and 1.017 (~17‰) for hydrogen in a turbulent boundary layer (Wang and Yakir, 2000). h is relative humidity normalized to the leaf temperature.

3. Results

3.1 Meteorological conditions at each site during the sampling periods

This study was conducted at the sites with dramatically different climatic conditions. The results showed that T, RH and PAR varied significantly with the meteorological conditions and locations (Fig. 2). Low RH, high T and PAR were found during the sunny days, whereas high RH, and low T and PAR were found during the cloudy days at each site (Fig. 2). The RH decreased and T increased from the upper reaches to the lower reaches, except at S2-Jun with the lowest mean RH (42.2%) (Table 2 and Fig. 2).

Insert Figure 2 here

Insert Table 2 here

3.2 Variations of $\delta^{18}O$ and δD in different water pools

Figure 3 shows measured isotopic compositions of all the water samples in the δD-δ^{18}O plots. In general, the δD and δ^{18}O of xylem and soil water showed the relatively small ranges, compared to those of leaf water and air moisture (Table 3 and Fig. 3). The δD and δ^{18}O in leaf water varied from -37.6‰ to 44.0‰ and from -6.2‰ to 32.4‰, respectively, of all species. The δD and δ^{18}O of air moisture at canopy ranged from -188.9‰ to -25.7‰ and from -24.9‰ and -6.0‰, respectively in all study sites. The δD and δ^{18}O of air moisture near the ground ranged from -133.0‰ and -40.6‰ and from -19.7‰ to -7.9‰, respectively in the upper reaches. The δD and δ^{18}O in xylem water (including stem and root) varied from -72.7‰ to -21.4‰ and from -9.0‰ to 2.9‰, respectively. The δD and δ^{18}O in soil water varied from -67.4‰ to -6.3‰ and from -9.9‰ to 5.1‰, respectively (Table 3).

Insert Figure 3 here

Insert Table 3 here

The air moisture had the lowest average δD and $\delta^{18}O$ in all study sites that increased with rising altitude (Table 3). The δD-$\delta^{18}O$ regression lines followed closely with the local meteoric water lines (LMWL) (Fig. 3). The average δD and $\delta^{18}O$ of air moisture were -101.7‰ and -14.1‰ near the ground and were -99.1‰ and -13.3‰ at the canopy, respectively in the upper reaches. In the lower reaches, the average δD and $\delta^{18}O$ of air moisture were -116.7‰ and -16.2‰ in S4-Aug and -136.3‰ and -17.7‰ in S5-Aug, respectively.

Leaf water had the highest average δD and $\delta^{18}O$ values, and leaf δD-$\delta^{18}O$ regression lines highly deviated from their corresponding LMWL, and leaf water showed the greatest variation in the observed $\delta^{18}O$ values. In addition, leaf water δD and $\delta^{18}O$ values increased with the decrease of altitude and increase of T (Table 2 and Table 3). In the upper reaches, the average δD in leaf water of Q.S., P.F., P.V. and S.C. were 1.9‰, -5.6‰, -2.2‰ and 10.4‰, respectively, and the average $\delta^{18}O$ were 8.3‰, 3.0‰, 1.5‰ and 8.2‰, respectively. In the lower reaches, the average δD in leaf water of P.E., S.A and R.S were 6.2‰, 10.4‰ and 7.5‰, respectively; and the average $\delta^{18}O$ were 14.6‰, 15.6‰ and 27.2‰, respectively.

The average δD and $\delta^{18}O$ values were -34.9‰ and -4.2‰ in 5 cm soil water and -43.2‰ and -5.2‰ in 10 cm soil in the upper reaches, and -34.2‰ and 1.4‰ in 10 cm water in the lower reaches, respectively. With the increase of T and decrease of altitude, the δD-$\delta^{18}O$ regression lines gradually deviated from their corresponding LMWL, and the variations of $\delta^{18}O$ values in xylem and soil water also increased gradually (Table 2 and Table 3). There were significant differences in δD and $\delta^{18}O$ between xylem water of S.C. and 5 cm soil water in the upper reaches. Differences were also seen in

P.E. and R.S. in the lower reaches (Fig. 3 and Table 3).

3.3 Variations of d-excess in each water pool

3.3.1 The diurnal variations of d-excess in leaf and xylem water during the sunny days

Several sunny days were selected based on the meteorological record (Fig. 2). The selected periods included the following: from 6:00 to 18:00 September 7 and 8 at S1-Sep, from 6:00 to 16:00 June 23 at S1-Jun, from 6:00 to 16:00 June 27 at S2-Jun, from 6:00 August 1 to 16:00 August 2 and from 6:00 to 18:00 August 3, 2009 at S3-Aug. At S4-Aug and S5-Aug, all data were selected. The diurnal variations of leaf water d-excess (d_{leaf}) and xylem water d-excess (d_{xylem}) values during the sunny day were shown in Figure 4. During the sunny days, we found clear and robust diurnal variations of d_{leaf} in all the study sites. The maximum values of d_{leaf} occurred from 6:00 to 10:00, gradually decreasing to a minimum value in the mid-afternoon (from 14:00 to 20:00), and increasing again to a maximum value from 4:00 to 8:00 in the next day (Fig. 4). In the upper reaches, the averaged d_{leaf} values of Q.S. were -64.7‰, and varied from 13.4‰ (S3-Aug) to -133.8‰ (S2-Jun). The d_{leaf} values of P.F. (-29.8‰) and P.V. (-14.3‰) were higher than that of S.C. (-55.4‰). In the lower reaches, the mean d_{leaf} value of P.E. (-110.2‰) and S.A. (-114.4‰) in the S5-Aug were higher than those of R.S. (-210.4‰) in the S4-Aug (Table 4).

Insert Figure 4 here

The peak-to-trough amplitudes of d_{leaf} varied greatly. They were 147.2‰ in tree (Q.S.), 122.6‰ in shrub (P.F.) and ranging from 143.1‰ to 52.6‰ in grasses (P.V. and S.C.) in the upper reaches. In the lower reaches, the peak-to-trough amplitudes of d_{leaf} were 124.4‰ in P.E. (tree), 96.9‰ in S.A. (shrub), and 80.6‰ in R.S. (shrub) (Table 4).

Insert Table 4 here

Compared to d_{leaf}, the diurnal variations of d_{xylem} of all species were more stable, and showed no clear diurnal variations (Fig. 4). In the upper reaches, the mean d_{xylem} values of Q.S., P.F., P.V. and S.C.

were 6.2‰, 0.8‰, 7.6‰ and -18.8‰, respectively. The averaged differences between d_{xylem} and d_{leaf} were 70.9‰, 30.6‰, 21.9‰ and 36.6‰ in Q.S., P.F., P.V. and S.C. in the upper reaches. In the lower reaches, the mean d_{xylem} values of P.E. and R.S. were -8.2‰ (S4-Aug) and -44.8‰ (S5-Aug), and the differences between d_{xylem} and d_{leaf} were 102.2‰ and 165.6‰ for S4-Aug and S5-Aug in the lower reaches, respectively (Table 4).

3.3.2 Variations of d-excess in soil water and air moisture during the sunny days

The averaged soil water d-excess values of 5 cm and 10 cm (d_{soil}) were -0.9‰ and -1.2‰, varying from -37.3‰ (S2-Jun) to 14.3‰ (S1-Sep), and from -25.7‰ (S2-Jun) to 16.6‰ (S1-Sep) in the upper reaches, respectively. In the S4-Aug and S5-Aug, the averaged d_{soil} values of 10 cm were -31.0‰ and -59.1‰, ranging from -45.5‰ to -19.8‰ and from -75.3‰ to -48.7‰, respectively. The d_{soil} values decreased with the increase of T and decrease of RH (Table 2 and Table 4). Except at S3-Aug, there were no temporal trends of d_{soil} at 5 cm and 10 cm (Fig. 5). The d_{soil} of the 5 cm and 10 cm were the lowest near 12:00. The highest observed d_{soil} was from 2:00 to 6:00 for site S3-Aug during the first sunny day after rain day (Fig. 5D).

Insert Figure 5 here

Fig. 6 shows the diurnal variations of air moisture d-excess ($d_{moisture}$) from each study site at the sunny days. Although the patterns were similar at all sites, the peak-to-trough amplitudes of $d_{moisture}$ varied greatly. They were 39.9‰ near the ground and 36.7‰ at the canopy in the upper reaches. In the lower reaches, the peak-to-trough amplitudes of $d_{moisture}$ at the canopy were 17.3‰ in the S4-Aug and 30.6‰ in the S5-Aug, respectively (Table 4 and Fig. 6). Except S2-Jun (Fig. 6c), the $d_{moisture}$ values varied diurnally, showing a clear and robust pattern of maximum $d_{moisture}$ during the mid-day (about from 10:00 to 16:00) (Fig. 6).

Insert Figure 6 here

3.3.3 Variations of d-excess in leaf water, xylem water, soil water and air moisture water during the cloudy days

In our study, the cloudy days occurred only at the upper reaches (Table 1). The d_{leaf} values during the cloudy days were significantly higher than those of the sunny days and d_{leaf} values were significantly lower than those of d_{xylem} values (Table 5). During the cloudy days with low PAR at the upper reaches of the HRB, there were no clear diurnal variations for $d_{moisture}$, d_{leaf} and d_{soil} at 5 cm and 10 cm depth, except for $d_{moisture}$ and d_{leaf} at S1-Jun (Fig. 7 and Fig. 8). In addition, at S3-Aug, the d_{leaf} increased gradually from 6:00 to 16:00 and showed the opposite diurnal variations compared to those of the sunny days. The likely reason is the leaf absorption of precipitation with high d-excess (e.g., 11.8‰ as in Zhao et al., 2011b) during the rainy conditions (precipitation occurred from 10:40 to 22:00 on August 2).

During the sunny days, the large difference between d_{xylem} and d_{leaf} was found, and the d_{xylem}-d_{leaf} values varied from 25.9‰ to 116.9‰, with a mean value of 60.0‰. Except at S2-Jun, the mean difference of d_{xylem} and d_{leaf} during the cloudy days was 22.6‰, and this value was lower than that of the sunny days. The large difference of d_{leaf} between the sunny and cloudy days was found with a mean of 32.7‰ (excluding S2-Jun), and the difference varied from 12.0‰ to 55.7‰ (excluding S2-Jun). There was no obvious difference of d_{xylem} between the sunny and cloudy days except at S2-Jun (Table 5).

Insert Table 5 here

Insert Figure 7 here

Insert Figure 8 here

3.4 Controlling factors of the d-excess in different water pools

3.4.1 Relationships between d-excess of various pools and RH

Significantly positive correlations were found between d_{leaf} and RH at all the study sites during the entire study periods (from June to September) (Table 6). Significantly positive correlations were also found between d_{leaf} and RH at all the study sites during the sunny days (Table 7). As RH increased by 1%, the increasing magnitude of d_{leaf} ranged from 0.49‰ to 2.53‰ in the upper reaches. In the lower reaches, as RH increased by 1%, the increasing magnitude of d_{leaf} ranged from 1.21‰ to 1.77‰ (Table 6).

Insert Table 6 here

Except near the ground at S1-Sep, significantly negative correlations were found between $d_{moisture}$ and RH at all the study sites when including both sunny and cloudy days (Table 6). Significantly negative correlation was found between $d_{moisture}$ and RH at S1-Sep when only sunny days were considered (Table 7). The $d_{moisture}$/RH were -0.15‰/% at S1-Jun and -0.27‰/% at S2-Jun for near the ground air moisture. For the canopy air moisture, the $d_{moisture}$/RH were -0.24‰/%, -0.32‰/%, -0.25‰/%, -0.15‰/%, -0.13‰/% and -0.68‰/% at S1-Sep, S1-Jun, S2-Jun, S3-Aug, S4-Aug and S5-Aug, respectively. During the sunny days, the $d_{moisture}$/RH were -0.36‰/% and -0.31‰/%, respectively, for near the ground and at the canopy in the upper reaches, which were larger than the results based on data including both sunny and cloudy days (Table 7). In terms of d_{soil}, the correlations between d_{soil} of 10 cm at S1-Sep, S3-Aug and RH, and between d_{soil} of 5 cm at S1-Jun and RH were significant (Table 6).

Insert Table 7 here

3.4.2 Relationships between d-excess of various pools and T

Significantly negative relationships were found between d_{leaf} and T in both upper reaches and lower reaches except in Q.S. at S1-Jun (Table 8). The decreasing magnitudes of d_{leaf} with T in Q.S. were -3.27‰/ºC, -1.59‰/ºC and -6.25‰/ºC at S1-Sep, S2-Jun and S3-Aug, respectively. The magnitudes were -6.45‰/ºC and -5.10‰/ºC for P.F., -6.74‰/ºC and -5.07‰/ºC for P.V. for S1-Sep and S1-Jun, respectively. The magnitude was -2.21‰/ºC in S.C. at S2-Jun. During the sunny days, there were significantly negative relationships between d_{leaf} and T in both upper and lower reaches (Table 7, Table 8). In the lower reaches, the decreasing magnitudes of d_{leaf} in P.E. and S.A. were -4.40‰/ºC and -2.15‰/ºC at S4-Aug, respectively. It was -1.82‰/ºC for R.S. at S5-Aug (Table 8).

Insert Table 8 here

There were significantly positive relationships between $d_{moisture}$ and T at all sites except S2-Jun (Table 8). The $d_{moisture}$/T values near the ground were 0.54‰/ºC and 0.76‰/ºC at S1-Sep and S1-Jun, respectively. The $d_{moisture}$/T values at canopy were 0.81‰/ºC, 0.91‰/ºC, 0.64‰/ºC, 0.54‰/ºC and 0.83‰/ºC at S1-Sep, S1-Jun, S3-Aug, S4-Aug and S5-Aug, respectively (Table 8). During the sunny days, the $d_{moisture}$/T were 1.18‰/ºC and 1.11‰/ºC, respectively, for near the ground and at the canopy, which were larger than the results based on data including both sunny and cloudy days (Table 7).

At S2-Jun, there were positive relationships between d_{soil} (both 5 cm and 10 cm depth) and T, and the d_{soil}/T values were 0.88‰/ºC (p = 0.021) and 0.34‰/ºC (p = 0.045) for 5 cm and 10 cm depth, respectively. However, at S3-Aug, there were negative relationships between d_{soil} (both 5 cm and 10 cm depth) and T, and the d_{soil}/T values were -0.45‰/ºC (p = 0.009) and -0.54‰/ºC (p = 0.002) for 5 cm and 10 cm depth, respectively. Significantly negative relationship was also found between d_{soil} of 5 cm depth and T at S1-Sep (d_{soil}/T= -0.16‰/ºC, p = 0.002) (Table 8).

3.4.3 Relationships between d-excess of various pools

Insert Figure 9 here

During the sunny days, we found an opposite pattern between the diurnal variations of d_{leaf} and $d_{moisture}$ (Fig. 9). Similar pattern was found between d_{soil} and $d_{moisture}$ during the first sunny day after rain. The d_{leaf} (d_{soil}) became more negative while $d_{moisture}$ became more positive during the afternoon, and opposite patterns were found during the night (Fig. 9 and Fig. 10). There were significantly negative relationships between d_{leaf} and $d_{moisture}$ at three study sites (Table 7). In the upper reaches, d_{leaf} of wood species (Q.S.) were correlated significantly to $d_{moisture}$ both near the ground and at the canopy, and the slopes were -1.47 and -1.40, respectively. The significantly negative relationships were also found between d_{leaf} of shrub/grass and $d_{moisture}$ near the ground, and the slopes were -0.14 and -0.12, respectively. In the lower reaches, the slopes of d_{leaf} and $d_{moisture}$ at canopy were -0.06 in woody species (P.E.), -0.10 and -0.28 in shrub (S.A.) in the S4-Aug and S5-Aug, respectively (Table 7).

Insert Figure 10 here

3.5 Modeling results of leaf water $\delta^{18}O$, δD and d-excess

The stead state Craig-Gordon model captured the diurnal variations of $\delta^{18}O$, δD and d-excess but discrepancy existed between modeled and observed values (Fig. 11). During the day, the observed values of $\delta^{18}O$ and δD were lower, while d-excess values were higher than predicted by the steady state Craig-Gordon model. At night, the observed values of $\delta^{18}O$ and δD were higher, while d-excess values were slightly lower than those of predicted by the model. On average, the modeled leaf water $\delta^{18}O$ and δD values were 1.8‰ and 6.7‰ higher, while d-excess values were 7.5‰ lower than those of observed values. The steady-state predictions explained 79.5% of variations in modeled $\delta^{18}O$, whereas 63.4% and 64.2% of variations in modeled δD and d-excess (Fig. 11).

Insert Figure 11 here

4. Discussion

4.1 Variations of δD and $\delta^{18}O$ in different water pools

Our results show that there are significant differences in δD and $\delta^{18}O$ among leaf water, xylem water, soil water and air moisture, and different δD-$\delta^{18}O$ patterns due to hydrogen and oxygen isotopic discrimination related to soil evaporation, plant transpirations and plant physiology. For example, compared to that of xylem water and shallow soil water, leaf water has the highest average δD and $\delta^{18}O$ values and the largest ranges in all the study sites. In addition, the δD-$\delta^{18}O$ regression lines of leaf water highly deviate from their corresponding LMWL (Table 2 and Fig. 3), suggesting a strong transpiration enrichment effect. With the decrease of RH and increase of T, leaf water δD and $\delta^{18}O$ values increased and the δD-$\delta^{18}O$ regression lines gradually deviate from their corresponding LMWL due to stronger transpiration, suggesting that climatic conditions have significant effect on variations of leaf water δD and $\delta^{18}O$ and their correlations by affecting transpiration (Table 2 and Table 3).

In the upper reaches, at high altitude sites such as S1-Sep and S1-Jun, the patterns of δD-$\delta^{18}O$ regression lines in shallow soil water and xylem water are similar (Fig. 3a and Fig. 3b), suggesting that the water sources of plants are from shallow soil water, and soil water are subject to only mild evaporation. These results are consistent with the fact of the horizontal distributions of Q.S. roots and shallow rooting of herbaceous plants such as P.V. However, at relatively lower altitude such as S2-Jun and S3-Aug, the xylem water δD and $\delta^{18}O$ of Q.S. are lower than that of soil water except the herbaceous plant (S.C.) (Table 3), and the δD-$\delta^{18}O$ regression lines of soil water deviate from the LMWL (Fig. 3). These results may be related to stronger soil evaporation in shallow soil layers. In the lower reaches, the δD and $\delta^{18}O$ of 10 cm soil water are significant higher than those of P.E. and R.S. xylem water, and the δD-$\delta^{18}O$ regression lines obviously deviate from the LMWL of the lower reaches, suggesting that strong soil water evaporation occur in shallow soil in the lower reaches.

As expected, the isotopic results show that the soil water at 5 cm and 10 cm is affected by evaporation which is indicated by a slope less than 8.0 (Dansgaard, 1964). In our study, the slopes of 5 cm and 10 cm soil water evaporation line vary from 2.6 to 7.4 (Table 9). Relative high slopes were found at S1-Sep (7.1) and S3-Aug (7.4) likely due to low temperature during September at S1-Sep and rain event at S3-Aug. The slopes of other sites are lower than 5.0, especially in the lower reaches, and the values in slopes are very small in the S4-Aug (2.6) and the S5-Aug (2.8) (Table 9), revealing strong shallow water evaporation. These slope values are comparable with other studies in vadose zones with evaporation slopes between 2 and 5 (Allison, 1982; Clark and Fritz, 1997; Kendall and McDonnell, 1999; Wenninger et al., 2010; Sutanto et al., 2012). The patterns of δD-δ^{18}O regression lines from shallow soil water gradually deviate from their corresponding LMWL with the decrease of altitude, suggesting a stronger water loss through direct evaporation especially in extremely arid region such as the riparian forest site and the Gobi site in the lower reaches of the HRB.

The air moisture has the most depleted δD and δ^{18}O compared to leaf water, xylem water and shallow soil water (Table 3). The air moisture δD and δ^{18}O data cluster around the corresponding LMWL (Fig. 3). These results are consistent with the isotopic fractionation theory (Gat, 1996) and they are also consistent with previous study in urban settings, agricultural settings, forest and grassland in China, Canada and USA (Welp et al., 2012).

4.2 Variations of d_{leaf}, d_{xylem}, d_{soil} and $d_{moisture}$ under different conditions

4.2.1 Variations of d-excess in leaf water and xylem water and their diurnal patterns

The significant differences of d-excess are found between leaf water and xylem water in both the upper reaches and lower reaches. In order to evaluate the effect of plant transpiration on d_{leaf}, we calculate the difference between d_{leaf} and d_{xylem} assuming d_{xylem} represents the d-excess of source water. The differences of averaged d_{xylem} and d_{leaf} vary from 21.9‰ to 165.6‰, and the differences are 70.9‰ in Q.S., 30.6‰ in P.F., 21.9‰ in P.V. and 36.6‰ in S.C. in the upper reaches, and are 102.0‰ in P.E. and 165.6‰ in R.S. in the lower reaches (Table 4). These differences reach the maximum value in the afternoon (Fig. 4). Since no isotopic fractionation occurs during water uptake and transport from roots to twigs (Washburn and Smith, 1934), the large differences between d_{xylem} and d_{leaf} found in this study indicate that plant transpiration results in lower d_{leaf}, and releasing water vapor with higher d-excess values into atmosphere. These were consistent with those expected from the recycling of surface evapotranspiration (Gat et al., 1994). Therefore, mixing of transpiration moisture into atmosphere will increase $d_{moisture}$, except for the conditions with influence of entrained atmospheric moisture with high d-excess. In addition, during the sunny days, the clear and robust diurnal variations of d_{leaf} with daily maximum in the early-morning and negative peak in the mid-afternoon are found in all the study sites (Fig. 4 and Table 4), while no diurnal variations of d_{leaf} are found at the cloudy days (Fig. 7). These results indicate that d_{leaf} is affected by meteorological conditions through their effect on plant transpiration. At the same time, no diurnal variations of d_{xylem} of all species are found at both the sunny and cloudy days (Fig. 4 and Fig. 7), indicating that d_{xylem} is stable and the effect of meteorological conditions on d_{xylem} is small. These results also suggested that the d-excess of moisture through plant transpiration has an important role on changing the $d_{moisture}$ of local air moisture during the sunny days.

4.2.2 Variations of d-excess in shallow soil water

No clear diurnal trends of d_{soil} are found except at S3-Aug. At S3-Aug, there are clear daily variations of d_{soil}, which reaches the lowest value at around 12:00 and slowly climbs up to the previous level the next day (the first sunny day after rain event) (Fig. 5D). This pattern is similar to d_{leaf}, which is likely due to the strong evaporation during the first sunny day after rain event (rain stopped at about 22:00 and we started make samples at 6:00 the next day) (Fig. 5D). At S3-Aug, during the first day after rain event, we also found the negative relationship between d_{soil} of 5cm, 10cm and T (Table 8), and opposite patterns between the diurnal variations of d_{soil} and $d_{moisture}$ (Fig. 10). These results indicate that d-excess of moisture through soil evaporation also has an important role on changing the $d_{moisture}$ of local air moisture during the sunny day after the rain events. In addition, the effect of soil evaporation on $d_{moisture}$ is similar to plant transpiration effect, and this effect was mainly controlled by temperature indicated by the negative relationship between d_{soil} and T (Table 8).

4.2.3 Variations of d-excess in air moisture near the ground and at the canopy

In our study, the peak-to-trough magnitudes vary greatly, and are 39.9‰ near the ground and 36.7‰ at the canopy in the upper reaches, and are 17.3‰ (S4-Aug) and 30.6‰ (S5-Aug) at the canopy in the lower reaches (Table 4 and Fig. 6). These observed values are higher than that of previous reports that the peak-to-trough magnitudes vary from 3.5‰ to 17.1‰ (Welp et al., 2012)**.** The higher range is likely caused by the large diurnal RH range (up to 80% change) in these environments. The lowest $d_{moisture}$ values are found near the ground (1.5‰) at high altitude during September (S1-Sep). The low values may be related to atmospheric entrainment contribution as atmospheric entrainment has been found to be responsible for the low d-excess values observed in the Pacific Northwest (Lai and Ehleringer, 2011).

In our study, during the sunny days, the $d_{moisture}$ vary diurnally, showing a clear and robust pattern of the highest $d_{moisture}$ at midday, and the lowest $d_{moisture}$ values at night at all the sites (Fig. 6). The same trends also found in urban settings (New Haven and Beijing), agricultural settings (Rosemount and Luancheng), forest (Borden Forest) and grassland (Duolun) (Welp et al., 2012); in one Beijing site (Wen et al., 2010), and in the Pacific Northwest (Lai and Ehleringer, 2011). These results showed that $d_{moisture}$ diurnal variation is not a pattern unique to any particular location or vegetation type, and the diurnal pattern of $d_{moisture}$ may suggest that $d_{moisture}$ is not a conserved tracer of humidity conditions at the marine moisture source region (Welp et al., 2012), and is strongly controlled by local evaporation and transpiration.

No clear diurnal patterns of $d_{moisutre}$ during the cloudy days when plant activity is low, which supports the role plants play in regulating $d_{moisutre}$. Namely, there are no clear diurnal variations for

$d_{moisture}$ except at S1-Jun (Fig. 8). The $d_{moisture}$ at S1-Jun shows diurnal variation (Fig. 8b), which corresponds to patterns of d_{leaf} after 8:00 (Fig. 7b and Fig. 8b).

4.3 The controlling factors of d-excess of various pools

4.3.1 Correlations between d_{leaf} and RH or T

The significantly positive correlations are found between d_{leaf} and RH at all the study sites during our study periods (from June to September) (Table 6). Significantly negative relationships are also found between d_{leaf} and T except in Q.S. at S1-Jun. In addition, during the sunny days, stronger relationships between d_{leaf} and T/RH are found at all study sites (Table 7). These results suggest that meteorological conditions such as RH and T have strong effect on variations of d_{leaf}, likely through the effect on transpiration.

4.3.2 Correlations between d_{soil} and RH or T

There are significant correlations between d_{soil} and RH or T in several cases. For example, the d_{soil} of 10 cm is positively correlated with RH (Table 6), and d_{soil} of 5 cm and 10 cm are negatively correlated with T (Table 8) at S3. The d_{soil} of 10 cm is also positively correlated with RH (Table 6), and d_{soil} of 5 cm is negatively correlated with T at S1-Sep (Table 8). At S3-Aug, during the first day after rain event, the negative relationship between d_{soil} at 5 cm and 10 cm and T (Table 8), the clear diurnal variations of d_{soil} at 5 cm and 10 cm (Fig. 5D) and opposite patterns between the diurnal variations of d_{soil} and $d_{moisture}$ (Fig. 10) are found. These results indicate that d-excess of moisture through soil evaporation also has an important role on changing the $d_{moisture}$ of local air moisture during the sunny days after the rain events, and this role is controlled by meteorological conditions. At the same time, the d_{soil}/RH are 0.08‰/%, and the d_{soil}/T vary from -0.16‰/ºC to -0.54‰/ºC, respectively, which are an order of magnitude lower than those of the d_{leaf}/RH (from 0.49‰/% to 2.53‰/%) and d_{leaf}/T (from -6.74‰/ºC % to -1.59‰/ºC). This means that even at the sunny day, the contribution of shallow soil water evaporation on $d_{moisture}$ is much less than that of plant transpiration (Table 6 and Table 7).

4.3.3 Variations of $d_{moisture}$ and its controlling factors

Main moisture sources of local air moisture come from canopy transpiration, soil evaporation and atmospheric entrainment (Lai and Ehleringer, 2011). If $d_{moisture}$ is a conservative tracer of conditions in the moisture source region, we would not expect it to vary with local relative humidity unless there is a local source of moisture to the atmosphere (Welp et al., 2012). In our study, except near the ground in S1-Sep, significantly negative correlations are found between $d_{moisture}$ and RH at all the study sites. The mean $d_{moisture}$/RH is -0.27‰/%, ranging from -0.68‰/% (S5-Aug) to -0.10‰/% (S4-Aug) (Table 6). Except of S5-Aug, the rate of $d_{moisture}$/RH of all the sites are lower than that of Merlivat and Jouzel's theoretic prediction (-0.43‰/%) (Merlivat and Jouzel, 1979) (Table 6). Aemisegger et al. (2013) reported that the importance of continental moisture recycling. It concluded that the contribution of plant transpiration to the continental evaporation flux can be deduced from the $d_{moisture}$-RH relation at the seasonal timescale and for individual events (Aemisegger et al., 2013). The relationship between $d_{moisture}$ and RH strongly depends on the isotopic composition of the soil moisture and the contribution of transpiration, which can be assumed in first order to be non-fractionating over timescales of > 1 day (Harwood et al., 1999; Farquhar et al., 2007). Welp et al. (2012) also reported that afternoon averages (12:00-18:00 local standard time) of $d_{moisture}$ are correlated with RH at New Haven ($d_{moisture}$/RH = -0.36‰/%) and Borden Forest ($d_{moisture}$/RH = -0.22‰/%) sites during the summer months (June-August). In addition, except at S2-Jun, there are significantly positive relationships between $d_{moisture}$ and T at all the sites. The mean $d_{moisture}$/T are 0.72‰/ºC, varying from 0.52‰/ºC (S4-Aug) to 0.91‰/ºC (S1-Jun) (Table 8). This is higher than that of Merlivat and Jouzel's theoretic prediction (0.35‰/ºC) (Merlivat and Jouzel, 1979). These results suggest that local contributions of moisture on $d_{moisture}$ are high, and local meteorological conditions such as RH and T have an important effect on

$d_{moisture}$. In addition, during the sunny days, the clear diurnal patterns of d_{leaf} (Fig. 4) and $d_{moisture}$ (Fig. 6, except Fig. 6c), the opposite patterns between the diurnal variations of d_{leaf} and $d_{moisture}$ (Fig. 9) and significantly negative relationship between $d_{moisture}$ and d_{leaf} ($p<0.001$) and high significantly relationships between $d_{moisture}$ and RH/T ($p<0.001$) (Table 6, 7 and 8) are found, suggesting that there is a strong linkage between $d_{moisture}$ and d_{leaf}, and the regulation of plant transpiration on the variations of atmospheric vapor isotopic composition is strong.

4.4 Comparison of modeled and observed leaf water $\delta^{18}O$, δD and d-excess

The modeling results reasonably captured the diurnal variations of $\delta^{18}O$, δD and d-excess with some discrepancies (Fig. 11). The discrepancies were larger for δD than for $\delta^{18}O$. The results indicate that a better parameterization of δD or a non-steady state modeling is likely needed to more accurately simulate the d-excess dynamics in leaf water. The d-excess of other components (e.g., soil water, atmospheric vapor) are rarely seen in literature and the current study provide a valuable source to validate modeling work of d-excess of various components.

5. Conclusions

Through extensive characterization of δD, $\delta^{18}O$ and d-excess in different water pools (e.g., leaf water, xylem water, 5 cm and 10 cm soil water and air moisture) in the HRB, we aimed to investigate the effects of local processes (e.g., plant transpiration and evaporation) on d-excess variations of different water pools. We concluded that:

1. There were significant variations of δD and $\delta^{18}O$ in different water pools. The most negative δD and $\delta^{18}O$ were found in air moisture. Average δD and $\delta^{18}O$ of air moisture were -101.8‰ and -14.1‰ in the upper reaches and -124.4‰ and -16.8‰ in the lower reaches, respectively. The most positive δD and $\delta^{18}O$ were found in leaf water. Average δD and $\delta^{18}O$ of leaf water were 0.9‰ and

8.1‰ in the upper reaches and 6.6‰ and 18.2‰ in the lower reaches, respectively. The δD-δ^{18}O regression lines of leaf water, xylem water and shallow soil water deviated gradually from their corresponding LMWL with the increase of T and decrease of RH.

2. Peak-to-trough amplitudes of d_{leaf}, d_{xylem}, d_{soil} and $d_{moisture}$ varied from 52.6‰ to 147.2‰, 10.2‰ to 48.8‰, 25.7‰ to 51.6‰ and 17.3‰ to 39.9‰, respectively, which were an order of magnitude higher than previous observations and predications (e.g., Merlivat and Jouzel, 1979, Welp et al., 2012). The mean $d_{moisture}$ values were the most positive, which were 7.7‰ near the ground and 11.2‰ at the canopy level in the upper reaches, 12.8‰ and 5.6‰ at the canopy level in the riparian forest site and in the Gobi site in the lower reaches. The d_{leaf} values were the most negative, which were -41.1‰ in the upper reaches and -145.0‰ in the lower reaches.

3. Several lines of evidence suggest that $d_{moisture}$ is not a conserved tracer of humidity conditions of the marine moisture source region, and is controlled by local transpiration and evaporation. The evidence includes the clear diurnal patterns of $d_{moisture}$ and d_{leaf} during the sunny days, the strong correlations of d_{leaf} with meteorological conditions (T and RH), the significant correlations of $d_{moisture}$ with d_{leaf}, T and RH, and no diurnal patterns of $d_{moisture}$ and d_{leaf} during the cloudy days when plant activity was low. In addition, large differences between average d_{xylem} and d_{leaf} were observed in our study, indicating the amount of d-excess lost through transpiration into atmosphere was high. Our results indicate plant transpiration strongly regulates $d_{moisture}$, especially during the sunny days. The effect is controlled by local meteorological conditions, such as T, radiation and RH.

4. The influences of shallow soil water evaporation on $d_{moisture}$ variations are generally small. However, the d-excess values of moisture from soil evaporation have strong effect on $d_{moisture}$ at the first sunny day after a rain event. The size of this effect is related to T and RH.

5. The steady state Craig-Gordon model can reasonably capture the diurnal variations of $\delta^{18}O$, δD and d-excess with small discrepancies. Non-steady state models are likely needed to more accurately simulate the d-excess dynamics of leaves and other components.

Our study shows that $d_{moisture}$ of the surface air at continental locations can be significantly altered by local processes at both mountain areas (Qilian Mountains) and extremely dry environments (Ejina), therefore such effect is likely a universal phenomenon across regions with varying climates.

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References

Aemisegger, F., Pfahl, S., Sodemann, H., Lehner, I., Seneviratne, S., and Wernli, H.: Deuterium excess as a proxy for continental moisture recycling and plant transpiration, Atmospheric Chemistry & Physics Discussions, 13, 2013.

Allison, G. B.: The relationship between 18O and deuterium in water in sand columns undergoing evaporation Journal of Hydrology, 55, 163-169, 1982.

Brooks, J. R. e., Barnard, H. R., Coulombe, R., and McDonnell, J. J.: Ecohydrologic separation of water between trees and streams in a Mediterranean climate, Nature Geoscience, 3, 100-104, doi:110.1038/ngeo1722, 2010.

Brunel, J. P., Simpson, H. J., Herczeg, A. L., Whitehead, R., and Walker, G. R.: Stable isotope composition of water vapor as an indicator of transpiration fluxes from rice crops, Water Resour. Res.,

28, 1407-1416, 10.1029/91wr03148, 1992.
Clark, I. D., and Fritz, P.: Environmental isotopes in hydrogeology, CRC press, 1997.
Craig, H., and L. I. Gordon. 1965. Deuterium and oxygen-18 variations in the ocean and marine atmosphere, in Proceedings of the Conference on Stable Isotopes in Oceanographic Studies and Paleotemperatures, edited by E. Tongiogi, pp. 1 – 24, Cons. Naz. delle Ric., Rome.
Cuntz, M., Ogée, J., Farquhar, G., Peylin, P., and Cernusak, L.: Modelling advection and diffusion of water isotopologues in leaves, Plant Cell and Environment, 30, 892-909, 2007.
Dalai, T. K., Bhattacharya, S., and Krishnaswami, S.: Stable isotopes in the source waters of the Yamuna and its tributaries: seasonal and altitudinal variations and relation to major cations, Hydrological Processes, 16, 3345-3364, 2002.
Dansgaard, W.: Stable isotopes in precipitation, Tellus, 16, 436-468, 1964.
Dawson, T. E.: Hydraulic lift and water use by plants: implications for water balance, performance and plant-plant interactions, Oecologia, 95, 565-574, 1993.
Dawson, T. E., Mambelli, S., Plamboeck, A. H., Templer, P. H., and Tu, K. P.: Stable isotope in plant ecology, Annu. Rev. Ecol. Syst., 33, 507-559, 2002.
Ehleringer, J., and Dawson, T.: Water uptake by plants: perspectives from stable isotope composition, Plant Cell Environment, 15, 1073–1082, 1992.
Farquhar, G. D., Cernusak, L. A., and Barnes, B.: Heavy water fractionation during transpiration, Plant Physiology, 143, 11–18, 2007.
Froehlich, K., Kralik, M., Papesch, W., Rank, D., Scheifinger, H., and Stichler, W.: Deuterium excess in precipitation of Alpine regions‚Äìmoisture recycling, Isotopes in Environmental and Health Studies, 44, 61-70, 2008.
Gat, J., Bowser, C., and Kendall, C.: The contribution of evaporation from the Great Lakes to the continental atmosphere : estimate based on stable isotope data, Geophysical Research Letters, 21, 557-560, 1994.
Gat, J.: Oxygen and hydrogen isotopes in the hydrologic cycle, Annual Review of Earth and Planetary Sciences, 24, 225–262, 1996.
Gong, J.D., Cheng G.D., Zhang X.Y., Xiao, H.L., and Li, X.Y. Environmental changes of Ejina region in the lower reaches of Heihe River. Advance in Earth Sciences, 17(4), 491-496, 2002.
Good, S., Soderberg, K., Wang, L., and Caylor, K.: Uncertainties in the assessment of the isotopic composition of surface fluxes: A direct comparison of techniques using laser-based water vapor isotope analyzers, Journal of Geophysical Research, 117, D15301, 10.1029/2011JD017168, 2012.
Harwood, K., Gillon, J., Roberts, A., and Griffiths, H.: Determinants of isotopic coupling of CO2 and water vapour within a Quercus petraea forest canopy, Oecologia, 119, 109–119, 1999.
He, J.Q. The spatial and temporal variations of stable isotopes in precipitation and river water in the Hexi Inland Rivers Basins, A dissertation for the degree of Doctor of Philosophy of Chinese Academy of Sciences. 2011.
Helliker, B. R., and Richter, S. L.: Subtropical to boreal convergence of tree-leaf temperatures, Nature, 454, 511-514, 2008.
Jackson, R., Moore, L., Hoffmann, W., Pockman, W., and Linder, C.: Ecosystem rooting depth determined with caves and DNA, Proceedings of the National Academy of Sciences, USA, 96, 11387-11392, 1999.
Karim, A., and Veizer, J.: Water balance of the Indus River Basin and moisture source in the Karakoram and western Himalayas: Implications from hydrogen and oxygen isotopes in river water, Journal of Geophysical Research: Atmospheres, 107, ACH 9-1-ACH 9-12, 2002.
Kendall, C., and McDonnell, J. J.: Isotope tracers in catchment hydrology, Elsevier, 1999.

Lai, C.-T., and Ehleringer, J. R.: Deuterium excess reveals diurnal sources of water vapor in forest air, Oecologia, 165, 213-223, doi210.1007/s00442-00010-01721-00442, 2011.
Merlivat, L., and Jouzel, J.: Global climatic interpretation of the deuterium-oxygen 18 relationship for precipitation, Journal of Geophysical Research, 84, 5029-5033, 10.1029/JC084iC08p05029, 1979.
Newman, B. D., Breshears, D. D., and Gard, M. O.: Evapotranspiration partitioning in a semiarid woodland: ecohydrologic heterogeneity and connectivity of vegetation patches Vadose Zone Journal, 9, 561-572, doi: 510.2136/vzj2009.0035 2010.
Salati, E., Dall'Olio, A., Matsui, E., and Gat, J. R.: Recycling of water in the Amazon basin: an isotopic study, Water Resources Research, 15, 1250-1258, 1979.
Soderberg, K., Good, S. P., O'Connor, M., Wang, L., Ryan, K., and Caylor, K. K.: Using atmospheric trajectories to model the isotopic composition of rainfall in central Kenya, Ecosphere, 4, art33, 10.1890/es12-00160.1, 2013.
Sutanto, S. J., Wenninger, J., Coenders-Gerrits, A. M. J., and Uhlenbrook, S.: Partitioning of evaporation into transpiration, soil evaporation and interception: a comparison between isotope measurements and a HYDRUS-1D model, Hydrology and Earth System Sciences, 16, 2605-2616, 2012.
Uemura, R., Matsui, Y., Yoshimura, K., Motoyama, H., and Yoshida, N.: Evidence of deuterium excess in water vapor as an indicator of ocean surface conditions, Journal of Geophysical Research: Atmospheres, 113, 2008.
Wang, X.F. and Yakir, D.: Using stable isotopes of water in evapotranspiration studies. Hydrological Processes, 14, 1407-1421, 2000.
Wang, L., Caylor, K. K., Villegas, J. C., Barron-Gafford, G. A., Breshears, D. D., and Huxman, T. E.: Evapotranspiration partitioning with woody plant cover: assessment of a stable isotope technique, Geophysical Research Letters, 37, L09401, 10.1029/2010GL043228, 2010.
Wang, L., Niu, S., Good, S., Soderberg, K., Zhou, X., Xia, J., Sherry, R., Luo, Y., Caylor, K., and McCabe, M.: The effect of warming on grassland evapotranspiration partitioning using laser-based isotope monitoring techniques, Geochimica et Cosmochimica Acta, 111, 28-38, 10.1016/j.gca.2012.12.047, 2013.
Washburn, E. W., and Smith, E. R.: The isotopic fractionation of water by physiological processes, Science, 79, 188-189, 1934.
Welp, L. R., Lee, X., Griffis, T. J., Wen, X.-F., Xiao, W., Li, S., Sun, X., Hu, Z., Val Martin, M., and Huang, J.: A meta-analysis of water vapor deuterium-excess in the midlatitude atmospheric surface layer, Global Biogeochem. Cycles, 26, GB3021, 10.1029/2011gb004246, 2012.
Wen, X., Zhang, S., Su, X., Yu, G., and Lee, X.: Water vapor and precipitation isotope ratios in Beijing, China, Journal of Geophysical Research-Atmospheres, 115, D01103, doi:01110.01029/02009JD012408, 2010.
Wenninger, J., Beza, D. T., and Uhlenbrook, S.: Experimental investigations of water fluxes within the soil-vegetation-atmosphere system: stable isotope mass-balance approach to partition evaporation and transpiration, Physics and Chemistry of the Earth, 35, 565-570, 2010.
White, J., Cook, E., Lawrence, J., and Broecker, W.: The D/H ratios of sap in trees: implications for water sources and tree ring D/H ratios, Geochimica et Cosmochimica, 49, 237–246, 1985.
Zhang, Y., Shen, Y., Sun, H., and Gates, J. B.: Evapotranspiration and its partitioning in an irrigated winter wheat field: A combined isotopic and micrometeorologic approach, Journal of Hydrology, 408, 203-211, 2011.
Zhao, L., Yin, L., Xiao, H., Cheng, G., Zhou, M., Yang, Y., Li, C., and Zhou, J.: Isotopic evidence for the moisture origin and composition of surface runoff in the headwaters of the Heihe River basin.

Chinese Science Bulletin, 56, 406-416, 2011a.
Zhao, L., Xiao, H., Zhou, J., Wang, L., Cheng, G., Zhou, M., Yin, L., and McCabe, M. F.: Detailed assessment of isotope ratio infrared spectroscopy and isotope ratio mass spectrometry for the stable isotope analysis of plant and soil waters, Rapid Communications in Mass Spectrometry, 25, 3071-3082, 2011b.
Zhao, L., Xiao, H., Zhou, M., Cheng, G., Wang, L., Yin, L., and Ren, J.: Factors controlling spatial and seasonal distributions of precipitation $\delta^{18}O$ in China, Hydrological Processes, 26, 143-152, 10.1002/hyp.8118, 2012.

Table 1 The vegetation types, sampling dates and time, and sampling types at the sampling sites in the Heihe River Basin.

Study region	Ecosystem type	Altitude (m)	Location ID	Sampling time and interval	Meteorological conditions	Sampling types
The upper reaches	Forest	2900m	S1-Sep: Pailugou	September 6-8, 2011 1 hour interval September 6-8, 2011 2 hour interval	The cloudy day: September 6, 2011 The sunny day: September 7 and 8, 2011	Qinghai Spruce - leaf and stem 5cm soil water 10cm soil water Atmospheric vapor near ground Atmospheric vapor at canopy Potentilla fruticosa - leaf and stem Polygonum viviparum - leaf and root
	Forest	2900m	S1-Jun: Pailugou	June 23-25, 2011 1 hour interval June 23-25, 2011 2 hour interval	The sunny day: June 23, 2011 The drizzle day: From 9:00 to 20:00 on June 24, 2011 The cloudy day: June 25, 2011	Qinghai Spruce - leaf and stem 5cm soil water 10cm soil water Atmospheric vapor near ground Atmospheric vapor at canopy Potentilla fruticosa - leaf and stem Polygonum viviparum - leaf and root
	Forest	2700m	S2-Jun: Pailugou	June 27-28, 2011 1 hour interval June 27-28, 2011 2 hour interval	The sunny day: June 27, 2011 The cloudy day: June 28, 2011	Qinghai Spruce - leaf and stem 5cm soil water 10cm soil water Atmospheric vapor near ground Atmospheric vapor at canopy Stipa capillata - leaf and root
	Forest	2800m	S3-Aug: Guantan	July 31, August 1-2, 2009 2 hour interval	Rain time: From 17:00 July 31 to 4:00 August 1 From 10:40 to 22:00 August 2, 2009 The sunny day: August 1	Qinghai Spruce - leaf and stem 5cm soil water 10cm soil water Atmospheric vapor at canopy
The lower reaches	Riparian forest	930m	S4-Aug: Qidaoqiao	August 6-9, 2009 2 hour interval	The sunny day	Populus euphratica - leaf and stem Sophora alopecuroides - leaf 10cm soil water Atmospheric vapor at canopy
	Gobi	906m	S5-Aug: Gobi	August 10-12, 2009 2 hour interval	The sunny day	Reaumuria soongorica - leaf and stem 10cm soil water Atmospheric vapor at canopy

Table 2 Meteorological data at each site during the observation periods. Note: The S1-Sep, S1-Jun, S2-Jun and S3-Aug indicate the 2900 m Qinghai spruce forest site in September 2011, 2900 m site in June 2011, 2700 m site in June 2011 and 2800 m site in August 2009 in the upper reaches. S4-Aug and S5-Aug indicate the riparian forest at 930 m and the Gobi site at 906 m in August 2009 in the lower reaches.

	S1-Sep			**S1-Jun**			**S2-Jun**		
	RH (%)	T (°C)	PAR ($\mu mol \cdot m^{-2} \cdot s^{-1}$)	RH (%)	T (°C)	PAR ($\mu mol \cdot m^{-2} \cdot s^{-1}$)	RH (%)	T (°C)	PAR ($\mu mol \cdot m^{-2} \cdot s^{-1}$)
Mean	**74.3**	***6.3***	491.7	**58.9**	***11.5***	576.0	**42.2**	***15.2***	687.1
Minimum	39.8	0.0	0.0	25.0	4.0	0.0	19.4	7.2	0.0
Maximum	91.1	12.4	1886.0	96.5	20.0	2097.0	67.9	22.5	2021.0
Standard diviation	15.5	3.9	637.6	21.9	5.0	625.9	14.3	4.5	713.1

	S3-Aug			**S4-Aug**		**S5-Aug**	
	RH (%)	T (°C)	PAR ($\mu mol \cdot m^{-2} \cdot s^{-1}$)	RH (%)	T (°C)	RH (%)	T (°C)
Mean	**74.8**	***12.0***	541.1	**46.5**	***23.1***	**19.0**	***28.7***
Minimum	38.0	5.2	0.0	17.0	9.1	11.3	17.2
Maximum	95.1	18.8	2036.0	87.5	33.7	34.3	38.0
Standard deviation	19.4	4.0	676.5	21.6	6.5	7.0	7.1

Table 3 Spatial and temporal variations in $\delta^{18}O$ and δD of different water pools in the Heihe River Basin. The numbers in the parenthesis indicate the number of samples. In the upper reaches, Q.S., P.F., P.V. and S.C. refer to *Qinghai Spruce*, *Potentilla fruticosa*, *Polygonum viviparum*, and *Stipa capillata* in the forest ecosystem. In the lower reaches, P.E. and S.A. refer to *Populus euphratica* and *Sophora alopecuroides* in the riparian forest ecosystem. R.S. refers to *Reaumuria soongorica* at the Gobi site.

Study sites	**S1-Sep, S1-Jun, S2-Jun and S3-Aug**								**S4-Aug**				**S5-Aug**	
Plant species	Q.S. (n = 166)		P.F. (n = 51)		P.V. (n = 51)		S.C. (n = 23)		P.E. (n = 36)		S.A. (n = 36)		R.S. (n = 23)	
Leaf water	$\delta^{18}O_{leaf}$	δD_{leaf}	$\delta^{18}O_{leaf}$	δD_{leaf}	$\delta^{18}O_{leaf}$	δD_{leaf}	$\delta^{18}O_{leaf}$	δD_{leaf}	$\delta^{18}O_{leaf}$	δD_{leaf}	$\delta^{18}O_{leaf}$	δD_{leaf}	$\delta^{18}O_{leaf}$	δD_{leaf}
Mean	8.3	1.9	3.0	-5.6	1.5	-2.2	8.2	10.4	14.6	6.2	15.6	10.4	27.2	7.
Minimum	-4.8	-29.4	-5.0	-37.6	-6.2	-35.0	1.7	-6.1	3.4	-9.7	5.8	-3.8	22.2	-5.
Maximum	18.5	22.8	17.7	31.6	20.1	44.0	11.4	22.8	21.3	23.4	20.3	19.1	32.4	23.
SD	6.8	13.3	6.4	17.1	7.0	20.1	2.5	7.9	5.0	8.2	3.9	5.8	2.5	6.
Xylem water	$\delta^{18}O_{xylem}$	δD_{xylem}	$\delta^{18}O_{xylem}$	δD_{xylem}	$\delta^{18}O_{xylem}$	δD_{xylem}	$\delta^{18}O_{xylem}$	δD_{xylem}	$\delta^{18}O_{xylem}$	δD_{xylem}	$\delta^{18}O_{leaf}$	δD_{leaf}	$\delta^{18}O_{xylem}$	δD_{xylem}
Mean	-6.7	-47.2	-5.0	-39.1	-6.5	-44.6	-1.7	-32.7	-5.2	-48.9	/	/	-2.4	-64
Minimum	-9.0	-65.7	-7.5	-60.1	-8.5	-61.5	-5.4	-46.6	-5.6	-51.7	/	/	-4.9	-72
Maximum	-2.1	-21.6	-2.9	-23.6	-4.8	-32.7	1.0	-21.4	-4.2	-43.9	/	/	2.9	-50
SD	1.6	10.1	1.1	8.8	1.0	7.4	1.6	7.3	0.3	1.7	/	/	1.9	6.
δ_{xylem}-δ_{leaf}	**-15.0**	**-49.1**	**-8.0**	**-33.5**	**-8.0**	**-42.4**	**-9.9**	**-43.1**	**-19.8**	**-55.1**	/	/	**-29.6**	**-71.**
Soil water	5cm depth (n = 166)						10cm depth (n = 166)				10cm depth (n = 36)		10cm depth (n = 4)	
	$\delta^{18}O_{soil}$			δD_{soil}		$\delta^{18}O_{soil}$		δD_{soil}		$\delta^{18}O_{soil}$		δD_{soil}	$\delta^{18}O_{soil}$	δD_{soil}
Mean	-4.2			-34.9		-5.2		-43.2		0.0		-31.2	2.7	-37
Minimum	-8.9			-62.0		-9.9		-67.4		-2.0		-36.7	1.1	-46
Maximum	2.5			-6.3		-0.7		-12.0		2.4		-21.0	5.1	-27
SD	2.5			10.7		2.1		10.1		1.1		3.3	1.9	8.
Air moisture	At the canopy (n = 172)						Near the ground (n = 172)				At the canopy (n = 36)		At the canopy (n = 2	
	$\delta^{18}O_{moisture}$			$\delta D_{moisture}$		$\delta^{18}O_{moisture}$		$\delta D_{moisture}$		$\delta^{18}O_{moisture}$		$\delta D_{moisture}$	$\delta^{18}O_{moisture}$	$\delta D_{moisture}$
Mean	-13.3			-99.1		-14.1		-101.7		-16.2		-116.7	-17.7	-136
Minimum	-18.5			-135.4		-19.7		-133.0		-23.1		-167.2	-24.9	-188
Maximum	-6.0			-25.7		-7.9		-40.6		-11.6		-78.0	-11.7	-96
SD	1.8			17.3		2.2		16.6		3.0		22.5	3.3	23.

Table 4 Spatial and temporal variations in d-excess (‰) of each water pools in the Heihe River Basin. The numbers in the parenthesis indicate the number of samples. The location ID and abbreviation of plant Latin name were the same as in Table 2 and 3.

Study sites	**S1-Sep, S1-Jun, S2-Jun and S3-Aug**				**S4-Aug**		**S5-Aug**
Plant species	Q.S. (n = 166)	P.F. (n = 51)	P.V. (n = 51)	S.C. (n = 23)	P.E. (n = 36)	S.A. (n = 36)	R.S. (n = 23)
	Leaf water				Leaf water		Leaf water
Mean	**-64.7**	**-29.8**	**-14.3**	**-55.4**	**-110.2**	**-114.4**	**-210.4**
Minimum	**-133.8**	**-112.9**	**-117.0**	**-72.3**	**-161.2**	**-145.4**	**-245.6**
Maximum	**13.4**	**9.7**	**26.1**	**-19.7**	**-36.8**	**-48.5**	**-165.0**
SD	**43.0**	**35.6**	**37.0**	**15.0**	**34.7**	**26.0**	**17.4**
The peak-to-trough amplitudes	**147.2**	**122.6**	**143.1**	**52.6**	**124.4**	**96.9**	**80.6**
	Xylem water				Xylem water		Xylem water
Mean	**6.2**	**0.8**	**7.6**	**-18.8**	**-8.2**	/	**-44.8**
Minimum	**-7.2**	**-7.5**	**0.3**	**-34.9**	**-14.1**	/	**-73.0**
Maximum	**15.4**	**7.5**	**22.4**	**-3.1**	**-3.9**	/	**-24.2**
SD	**5.0**	**3.8**	**5.2**	**7.2**	**2.1**	/	**12.7**
The peak-to-trough amplitudes	**22.6**	**15.0**	**22.1**	**31.8**	**10.2**	/	**48.8**
Mean d_{xylem} - d_{leaf}	**70.9**	**30.6**	**21.9**	**36.6**	**102.0**	/	**165.6**
Soil water	5 cm soil water (n = 166)		10 cm soil water (n = 166)		10 cm soil water (n = 36)		10 cm soil water (n = 4)
Mean	**-0.9**		**-1.2**		**-31.0**		**-59.1**
Minimum	**-37.3**		**-25.7**		**-45.5**		**-75.3**
Maximum	**14.3**		**16.6**		**-19.8**		**-48.7**
SD	**12.5**		**10.0**		**6.2**		**11.5**
The peak-to-trough amplitudes	**51.6**		**42.3**		**25.7**		**26.6**
Air moisture	At the canopy (n = 172)		Near the ground (n = 172)		At the canopy (n = 36)		At the canopy (n = 23)
Mean	**7.7**		**11.2**		**12.8**		**5.6**
Minimum	**-9.9**		**-7.0**		**2.6**		**-11.4**
Maximum	**26.8**		**32.9**		**19.9**		**19.2**
SD	**8.5**		**9.4**		**4.8**		**9.1**
The peak-to-trough amplitudes	**36.7**		**39.9**		**17.3**		**30.6**
Mean $d_{moisture}$ - d_{soil}	**8.6**		**12.4**		**43.8**		**64.7**

Table 5 Differences between d_{xylem} (‰) and d_{leaf} (‰) in the sunny and the cloudy days. The location ID and abbreviation of plant Latin name were the same as in Table 2 and 3.

Study sites	Plant species	The sunny day			The cloudy day			Difference of d_{leaf}	Difference of d_{xylem}
		d_{leaf}	d_{xylem}	d_{xylem}-d_{leaf}	d_{leaf}	d_{xylem}	d_{xylem}-d_{leaf}	d_{cloudy}-d_{sunny}	d_{cloudy}-d_{sunny}
S1-Sep	Q.S.	-51.9	11.8	**63.7**	-6.8	12.0	**18.8**	***45.1***	***0.1***
	P.F.	-60.6	2.7	**63.3**	-4.9	2.7	**7.6**	***55.7***	***0.0***
	P.V.	-42.0	10.6	**52.7**	11.1	8.3	**-2.8**	***53.2***	***-2.3***
S1-Jun	Q.S.	-72.0	5.0	**77.1**	-47.4	5.2	**52.6**	***24.7***	***0.2***
	P.F.	-37.8	-0.6	**37.2**	-15.5	-1.1	**14.4**	***22.3***	***-0.5***
	P.V.	-20.4	5.5	**25.9**	-4.6	6.4	**11.0**	***15.9***	***1.0***
S2-Jun	Q.S.	-114.0	2.9	**116.9**	-116.9	-0.2	**116.7**	***-2.9***	***-3.1***
	S.C.	-52.9	-15.9	**37.0**	-59.5	-23.7	**35.8**	***-6.6***	***-7.8***
S3-Aug	Q.S.	-64.9	1.4	**66.3**	-52.8	4.0	**56.8**	***12.0***	***2.5***
Mean		-57.4	2.6	**60.0**	-33.0	1.5	**34.5**	***24.4***	***-1.1***

Table 6 Linear least square fits between d-excess of various water bodies and relative humidity (RH) (%) at each site. Here *r* is correlation coefficient, and p is significance level. The $p < 0.001$ indicates statistical significance at the 99.9% significance level, and the $p < 0.05$ indicates statistical significance at the 95% significance level. The location ID and abbreviation of plant Latin name were the same as in Table 2 and 3.

d-excess (‰) versus RH (%)									
	Slope	**Intercept**	***r***	***p***		**Slope**	**Intercept**	***r***	***p***
	S1-Sep					**S1-Jun**			
d_{leaf} of Q.S.	**1.52**	**-146.68**	**0.701**	**<0.001**	d_{leaf} of Q.S.	**0.82**	**-113.36**	**0.589**	**<0.001**
d_{leaf} of P.F.	**2.38**	**-212.45**	**0.846**	**<0.001**	d_{leaf} of P.F.	**1.07**	**-90.47**	**0.825**	**<0.001**
d_{leaf} of P.V.	**2.53**	**-208.14**	**0.879**	**<0.001**	d_{leaf} of P.V.	**0.99**	**-72.05**	**0.723**	**<0.001**
d_{soil} of 5cm	0.03	6.61	0.198	0.122	d_{soil} of 5cm	**-0.10**	**9.42**	**-0.483**	**0.001**
d_{soil} of 10cm	***0.08***	***1.84***	***0.253***	***0.048***	d_{soil} of 10cm	<0.01	1.10	-0.046	0.775
$d_{moisture}$ near the ground	-0.11	9.62	-0.168	0.191	$d_{moisture}$ near the ground	**-0.15**	**18.50**	**-0.477**	**0.001**
$d_{moisture}$ at the canopy	**-0.24**	**20.56**	**-0.457**	**<0.001**	$d_{moisture}$ at the canopy	**-0.32**	**33.83**	**-0.753**	**<0.001**
	S2-Jun					**S3-Aug**			
d_{leaf} of Q.S.	**0.49**	**-135.82**	**0.686**	**<0.001**	d_{leaf} of Q.S.	**1.48**	**-169.36**	**0.716**	**<0.001**
d_{leaf} of S.C.	***0.56***	***-79.08***	***0.523***	***0.022***					
d_{soil} of 5cm	-0.21	-10.54	-0.279	0.094	d_{soil} of 5cm	0.05	-9.86	0.289	0.161
d_{soil} of 10cm	-0.02	-14.72	0.013	0.941	d_{soil} of 10cm	***0.08***	***-12.35***	***0.403***	***0.046***
$d_{moisture}$ near the ground	**-0.27**	**26.82**	**-0.682**	**<0.001**					
$d_{moisture}$ at the canopy	**-0.25**	**27.58**	**-0.689**	**<0.001**	$d_{moisture}$ at the canopy	**-0.15**	**28.37**	**-0.526**	**0.007**
	S4-Aug					**S5-Aug**			
d_{leaf} of P.E.	**1.41**	**-171.76**	**0.844**	**<0.001**	d_{leaf} of R.S.	**1.77**	**-243.96**	**0.716**	**<0.001**
d_{leaf} of S.A.	**1.21**	**-166.99**	**0.947**	**<0.001**					
d_{soil} of 10cm	0.02	-32.08	-0.012	0.939					
$d_{moisture}$ at the canopy	**-0.13**	**17.42**	**-0.602**	**0.003**	$d_{moisture}$ at the canopy	**-0.68**	**18.47**	**-0.526**	**<0.001**

Table 7 Correlations between d-excess of various water bodies and RH (%) and T (^oC), and between $d_{moisture}$ and d_{leaf} during the sunny days at each site. Here *r* is correlation coefficient, p is significance level. The $p < 0.001$ indicates statistical significance at the 99.9% significance level, and the $p < 0.05$ indicates statistical significance at the 95% significance level. The location ID and abbreviation of plant Latin name were the same as in Table 2 and 3. The periods of the sunny days were the same as in Fig. 5.

Study area		**The d-excess values versus RH (%)**				**The d-excess values versus T (^oC)**			
		Slope	**Intercept**	***r***	***p***	**Slope**	**Intercept**	***r***	***p***
S1-Sep S1-Jun S2-Jun S3-Aug	$d_{moisture}$ near the ground	-0.36	27.643	-0.712 (84)	<0.001	1.18	- 4.574	0.771	<0.001
	$d_{moisture}$ at the canopy	-0.31	28.269	-0.617 (101)	<0.001	1.11	0.695	0.716	<0.001
	d_{leaf} of wood	1.26	- 131.626	0.600 (102)	<0.001	-3.84	-19.327	0.630	<0.001
	d_{leaf} of shrub	1.26	-121.121	0.629 (25)	<0.001	-3.66	-15.489	0.547	<0.001
	d_{leaf} of herb	1.21	-99.962	0.635 (37)	<0.001	-3.17	-1.134	0.563	<0.001
	d_{leaf} of wood vs $d_{moisture}$ near the ground	-1.47	- 63.237	-0.360 (84)	<0.001	/	/	/	/
	d_{leaf} of wood vs $d_{moisture}$ at the canopy	-1.40	-52.568	-0.340 (101)	<0.001	/	/	/	/
	d_{leaf} of shrub vs *$d_{moisture}$ near ground*	*-0.14*	*3.69*	*-0.599 (24)*	*0.039*	/	/	/	/
	d_{leaf} of grass vs *$d_{moisture}$ near ground*	*-0.12*	*12.72*	*-0.648 (12)*	*0.023*	/	/	/	/
S4-Aug	d_{leaf} of wood vs $d_{moisture}$ at the canopy	-0.06	7.163	-0.543 (32)	<0.001	/	/	/	/
	d_{leaf} of shrub vs $d_{moisture}$ at the canopy	-0.10	1.827	-0.534 (32)	<0.001	/	/	/	/
S5-Aug	d_{leaf} of shrub vs $d_{moisture}$ at the canopy	-0.28	-57.737	0.540 (25)	<0.001	/	/	/	/

Table 8 Linear least square fits between d-excess of various water bodies and temperature (T) (^{o}C) at each site. Here *r* is correlation coefficient, p is significance level. The $p < 0.001$ indicates statistical significance at the 99.9% significance level, and the $p < 0.05$ indicates statistical significance at the 95% significance level. The location ID and abbreviation of plant Latin name were the same as in Table 2 and 3.

d-excess (‰) versus T (^{o}C)									
	Slope	**Intercept**	***r***	***p***		**Slope**	**Intercept**	***r***	***p***
	S1-Sep					**S1-Jun**			
QS leaf water	**-3.27**	**-13.20**	**-0.419**	**<0.001**	QS leaf water	-1.60	-46.89	-0.202	0.220
PF leaf water	**-6.45**	**5.88**	**-0.612**	**<0.001**	PF leaf water	**-5.10**	**31.80**	**-0.919**	**<0.001**
PV leaf water	**-6.74**	**22.91**	**-0.575**	**<0.001**	PV leaf water	**-5.07**	**45.80**	**-0.942**	**<0.001**
5cm soil water	**-0.16**	**10.17**	**-0.387**	**0.002**	5cm soil water	0.12	2.36	0.075	0.635
10cm soil water	0.06	7.62	0.143	0.268	10cm soil water	-0.09	2.15	-0.087	0.585
Air moisture near ground	**0.54**	**-1.78**	**0.349**	**0.005**	Air moisture near ground	**0.76**	**0.86**	**0.610**	**<0.001**
Air moisture at canopy	**0.81**	**-2.63**	**0.481**	**<0.001**	Air moisture at canopy	**0.91**	**4.12**	**0.494**	**0.003**
	S2-Jun					**S3-Aug**			
QS leaf water (37)	**-1.59**	**-90.88**	**-0.664**	**<0.001**	QS leaf water	**-6.25**	**14.67**	**-0.684**	**0.001**
SC leaf water (19)	**-2.21**	**-22.15**	**-0.646**	**0.003**					
5cm soil water	***0.88***	***-32.89***	***0.379***	***0.021***	5cm soil water	**-0.45**	**-1.15**	**-0.514**	**0.009**
10cm soil water	***0.34***	***-20.73***	***0.332***	***0.045***	10cm soil water	**-0.54**	**-0.16**	**-0.589**	**0.002**
Air moisture near ground	0.42	9.23	0.285	0.087					
Air moisture at canopy	0.31	12.28	0.173	0.305	Air moisture at canopy	**0.64**	**9.39**	**0.491**	**0.005**
	S4-Aug					**S5-Aug**			
PE leaf water	**-4.40**	**2.274**	**-0.642**	**<0.001**	RS leaf water	**-1.82**	**-158.14**	**-0.742**	**<0.001**
SA leaf water	**-2.15**	**-64.28**	**-0.560**	**<0.001**					
10cm soil water	0.08	-32.97	0.050	0.755					
Air moisture at canopy	**0.54**	**0.95**	**0.773**	**<0.001**	Air moisture at canopy	**0.83**	**-18.23**	**0.684**	**0.001**

Table 9 Equations of soil water δD and $\delta^{18}O$ at each site using linear least squares fits method. Here r is correlation coefficient, p is significance level. The $p < 0.001$ indicates statistical significance at the 99.9% significance level, and the $p < 0.05$ indicates statistical significance at the 95% significance level.

Sites	Equation	*r*	*p*	Sites	Equation	*r*	*p*
S1-Sep	$\delta D = 7.114\times\delta^{18}O + 3.030$	0.921	<0.001	S3-Aug	$\delta D = 7.355\times\delta^{18}O - 8.267$	0.914	<0.001
S1-Jun	$\delta D = 4.998\times\delta^{18}O - 16.213$	0.825	<0.001	S4-Aug	$\delta D = 2.615\times\delta^{18}O - 31.128$	0.890	<0.001
S2-Jun	$\delta D = 3.952\times\delta^{18}O - 26.901$	0.888	<0.001	S5-Aug	$\delta D = 2.840\times\delta^{18}O - 44.930$	0.642	0.222

Figures captions

Figure 1. Locations of the sampling sites in the Heihe River Basin. Note: the information of sampling locations, altitude, period of sampling and climatic conditions are listed in Table 1.

Figure 2. Comparison of hourly average relative humidity (RH), air temperature (T) and photosynthetically active radiation (PAR) during the experimental period. The dark cycles and white cycles indicate the RH and T at each site. The grey shadow and blue shadow indicate cloudy days and rainy days. The panels a, b, c and d refer to the Qinghai spruce forest of S1-Sep, S1-Jun, S2-Jun and S3-Aug. The panel e and f refer to S4-Aug and S5-Aug.

Figure 3. Plot of δD and $\delta^{18}O$ of different water pools at each site. The LMWL (cited from He (2011)) is plotted for each site (dark line is the GMWL (the global meteoric water line); blue dotted line and dashed lines are the LMWL of the upper and the lower reaches, respectively). Note: the panels a, b, c, d, e and f refer to the same location as in Figure 2, and the abbreviations of plant Latin names were the same as in Table 3.

Figure 4. Variations in leaf and xylem water d-excess in the sunny days of the upper reaches and lower reaches of the Heihe River Basin. Note: the panels a, b, c, d, e and f refer to the same location as in Figure 2, and the abbreviations of plant Latin names were the same as in Table 3. The following sunny days were selected: S1-Sep: from 6:00 to 18:00 September 7 and 8; S1-Jun: from 6:00 to 16:00 June 23; S2-Jun: from 6:00 to 16:00 June 27; S3-Aug: from 6:00 August 1 to 16:00 August 2 and from 6:00 to 18:00 August 3, 2009. All data at S4-Aug and S5-Aug were selected.

Figure 5. Spatial and temporal variations in soil water d-excess in the Heihe River Basin. Note: the panels a, b, c and d refer to the same locations as in Figure 2, and the panel e refers to S4-Aug and S5-Aug in the lower reaches.

Figure 6. The d-excess of air moisture during the sunny days. Note: the panels a, b, c, d, e and f refer to the same locations as in Figure 2.

Figure 7 Variations in leaf and xylem water d-excess of the upper reaches of the Heihe River Basin during the cloudy days. Note: the panels a, b, c and d refer to the same location as in Figure 2, and the abbreviations of plant Latin names were the same as in Table 3.

Figure 8. Variations in d-excess of shallow soil water and of air moisture during the cloudy days of the upper reaches of the Heihe River Basin. Note: the panels a, b, c and d refer to the same locations as in Figure 2.

Figure 9. Comparison of leaf water and air moisture d-excess values during the sunny days. Note: the panels a, b, c, d, e and f refer to the same locations as in Figure 2, and the abbreviations of plant Latin name were the same as in Table 3. The AC and NG refer to air moisture at the canopy level and near the ground, respectively.

Figure 10. Comparison of soil water and air moisture d-excess values during the sunny days after 4 h rain event on 2 August, 2009 at S3-Aug.

Figure 11. Comparison of leaf water $\delta^{18}O$, δD and d-excess values for *Populus euphratica* between the simulated values (steady state Craig and Gordon model) and observed values at S5-Aug.

Figures

Figure 1

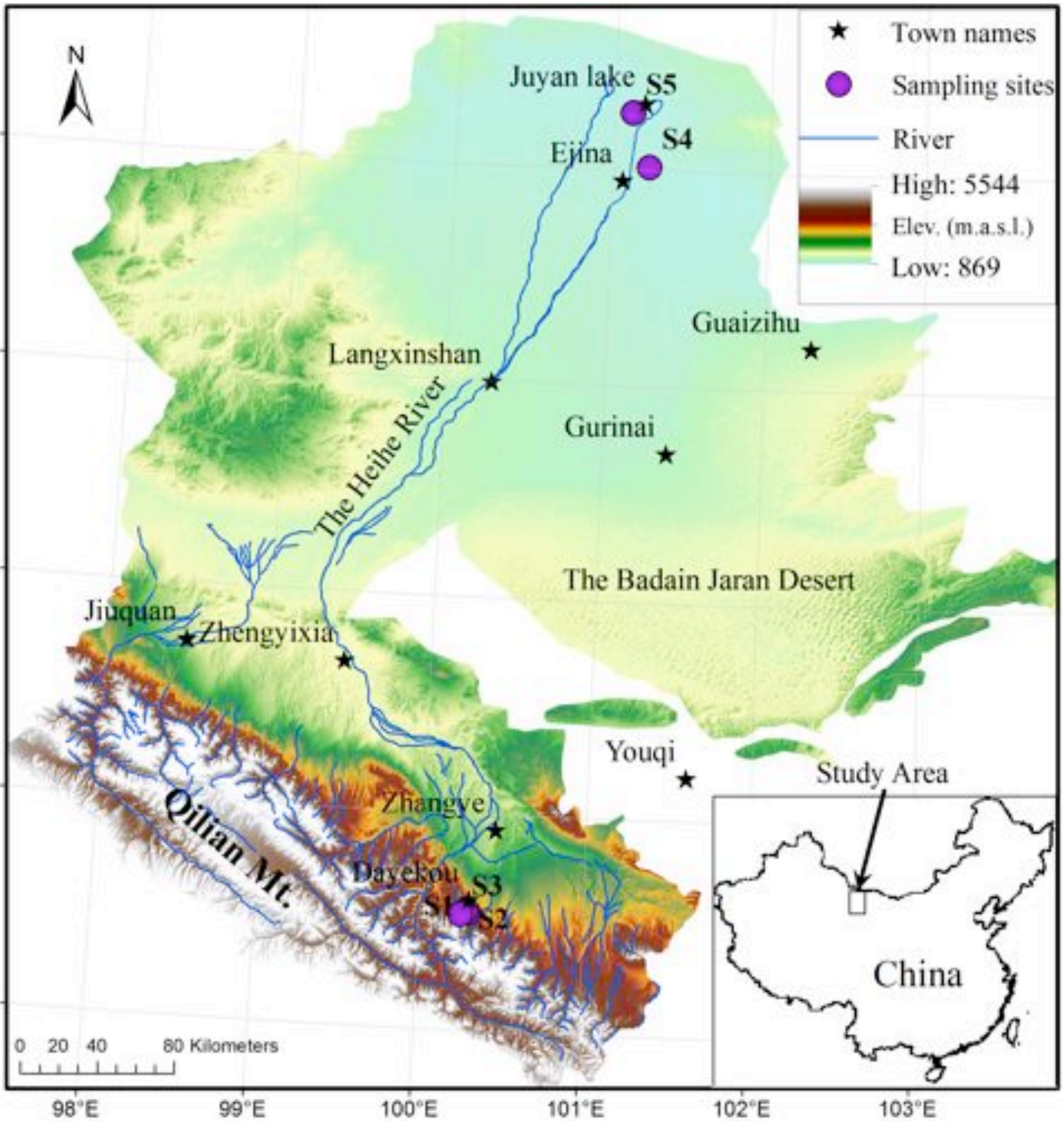


Figure 2

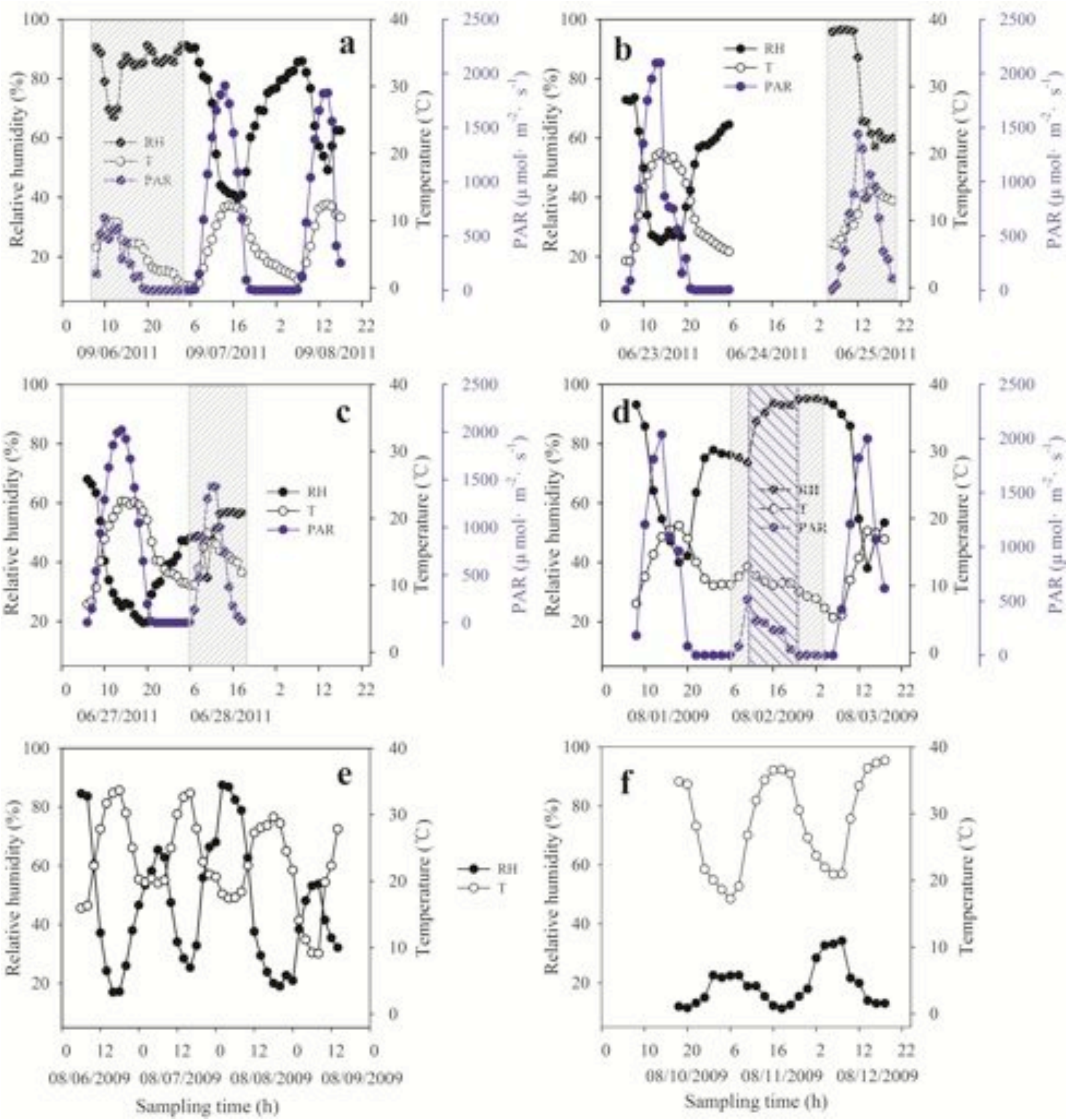
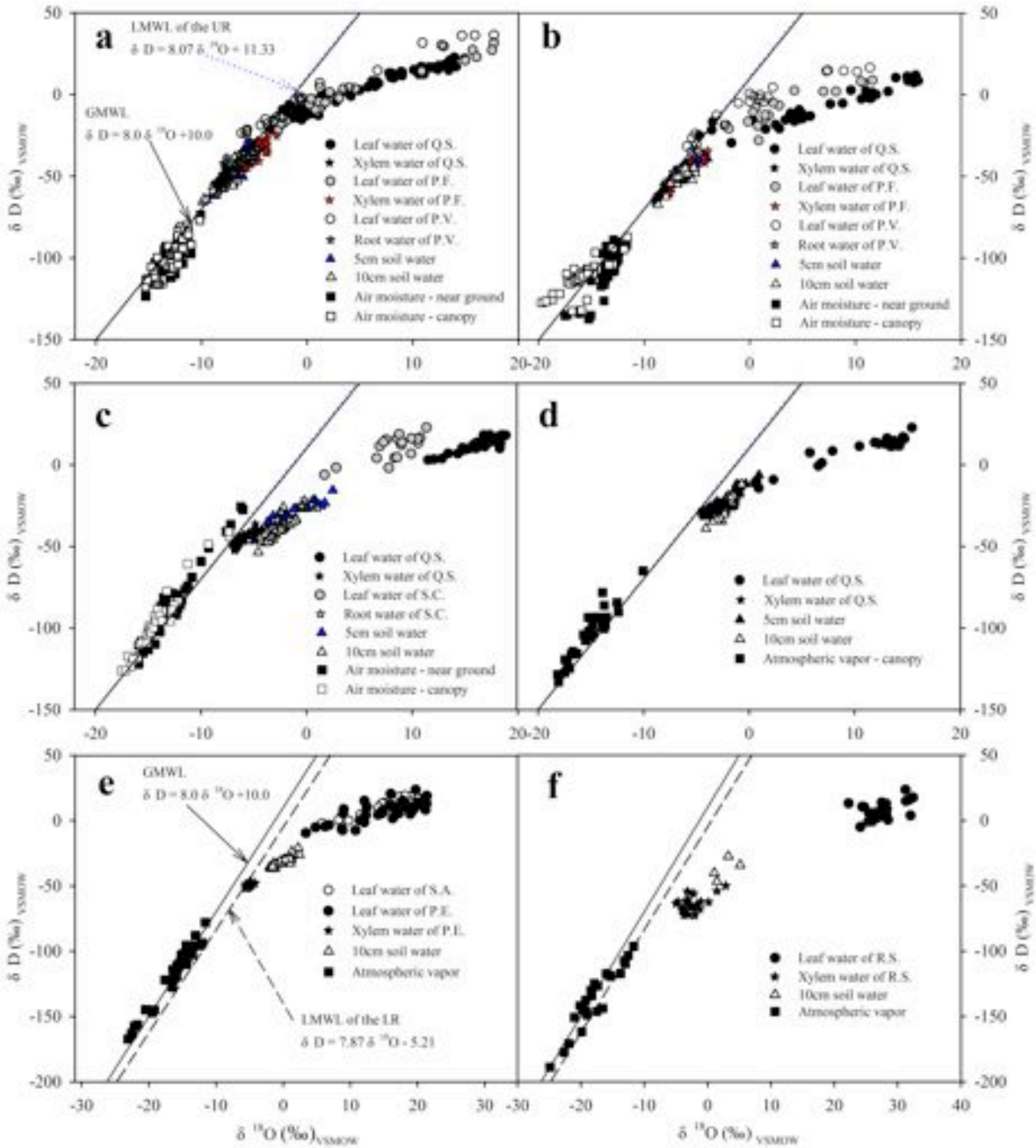

a
b
c
d
e
f
RH
T
PAR
Relative humidity (%)
Temperature (℃)
PAR (μ mol· m⁻²· s⁻¹)
09/06/2011 09/07/2011 09/08/2011
06/23/2011 06/24/2011 06/25/2011
06/27/2011 06/28/2011
08/01/2009 08/02/2009 08/03/2009
08/06/2009 08/07/2009 08/08/2009 08/09/2009
08/10/2009 08/11/2009 08/12/2009
Sampling time (h)

Figure 3



a
LMWL of the UR
δ D = 8.07 δ ¹⁸O + 11.33
GMWL
δ D = 8.0 δ ¹⁸O +10.0
Leaf water of Q.S.
Xylem water of Q.S.
Leaf water of P.F.
Xylem water of P.F.
Leaf water of P.V.
Root water of P.V.
5cm soil water
10cm soil water
Air moisture - near ground
Air moisture - canopy
b
c
Leaf water of S.C.
Root water of S.C.
d
Atmospheric vapor - canopy
e
GMWL
δ D = 8.0 δ ¹⁸O +10.0
Leaf water of S.A.
Leaf water of P.E.
Xylem water of P.E.
Atmospheric vapor
LMWL of the LR
δ D = 7.87 δ ¹⁸O - 5.21
f
Leaf water of R.S.
Xylem water of R.S.
δ D (‰) VSMOW
δ ¹⁸O (‰) VSMOW

Figure 4

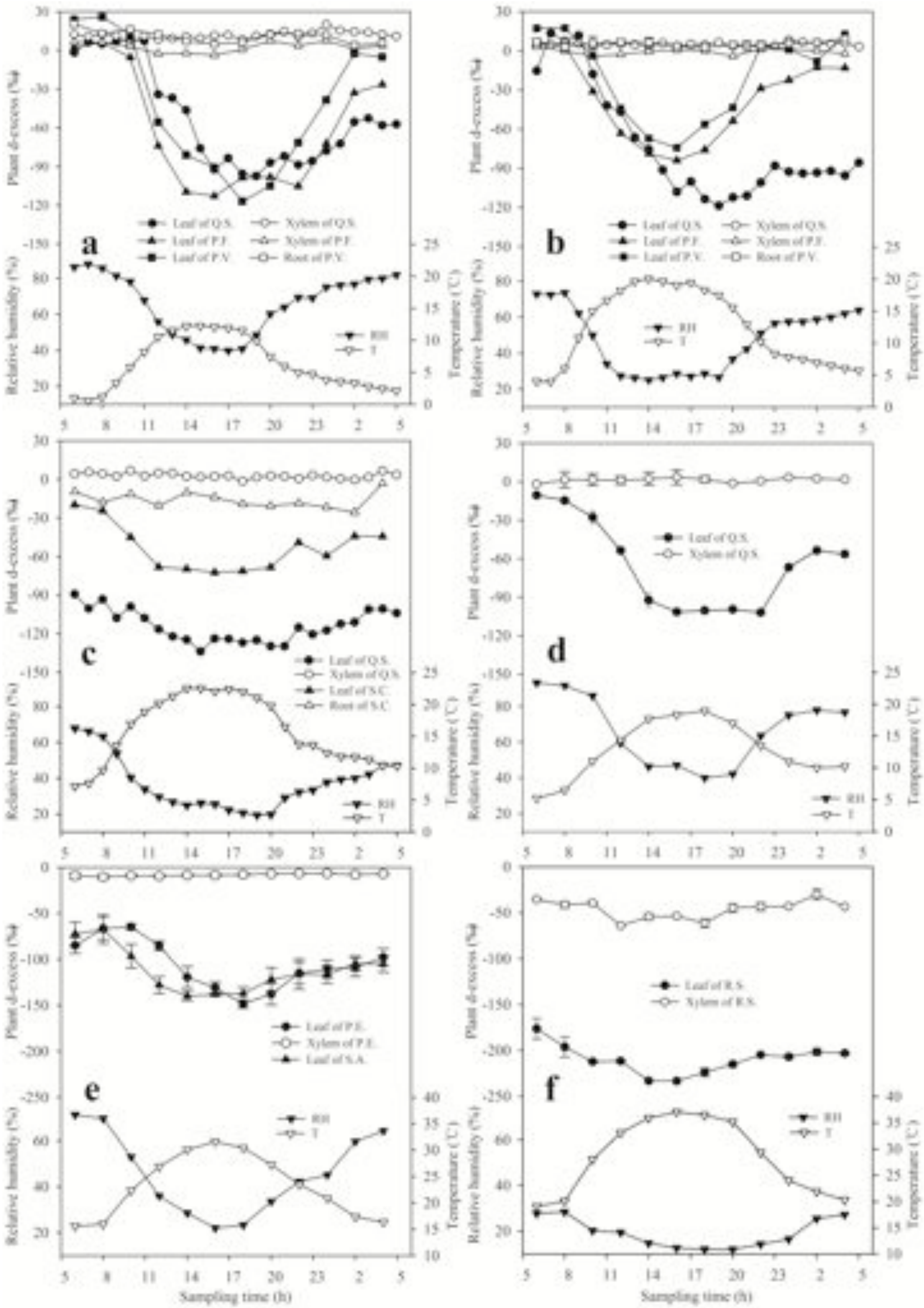
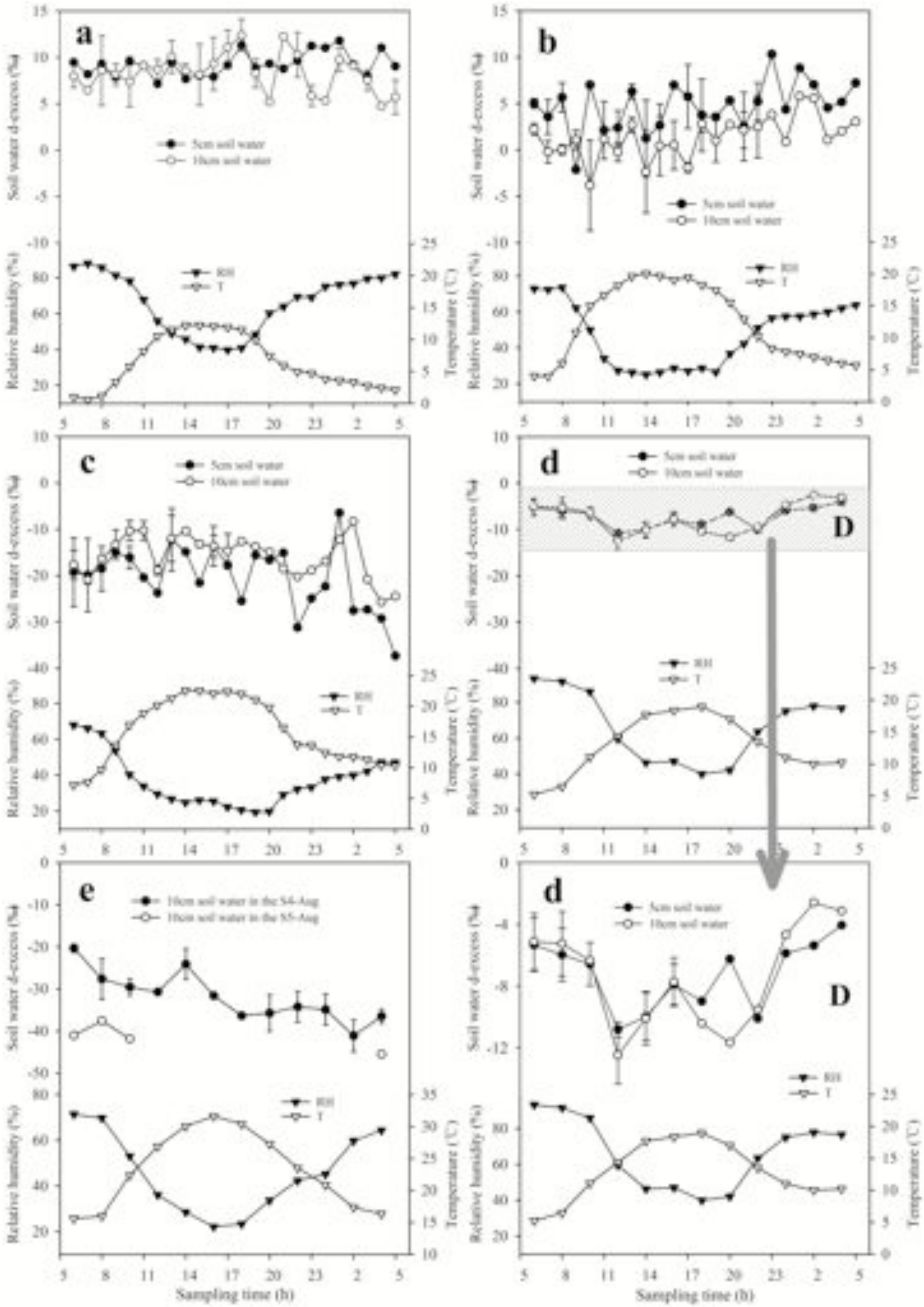
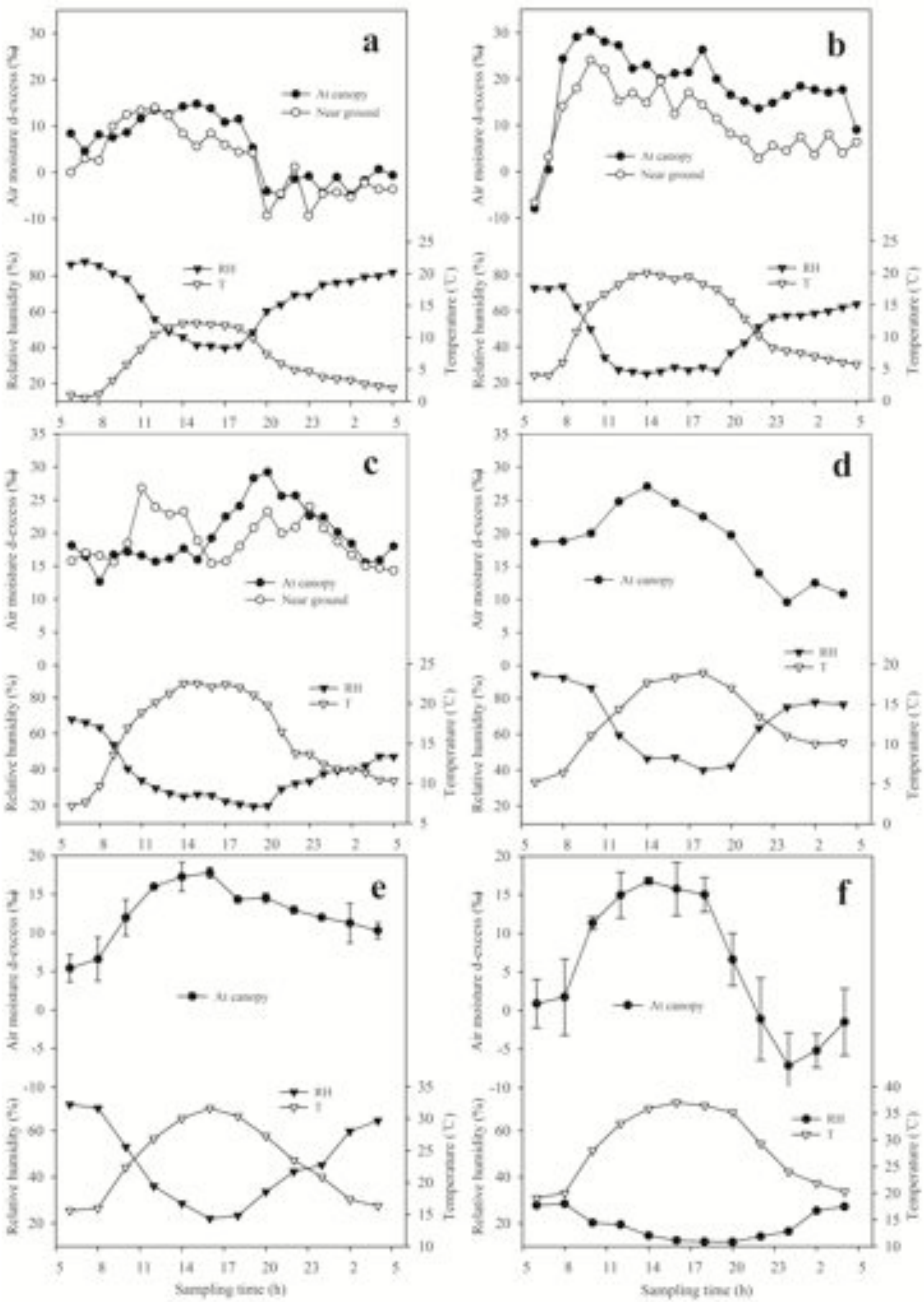

a
b
c
d
e
f
Plant d-excess (‰)
Relative humidity (%)
Temperature (°C)
Sampling time (h)
Leaf of Q.S.
Leaf of P.F.
Leaf of P.V.
Xylem of Q.S.
Xylem of P.F.
Root of P.V.
Leaf of S.C.
Root of S.C.
Leaf of P.E.
Xylem of P.E.
Leaf of S.A.
Leaf of R.S.
Xylem of R.S.
RH
T

Figure 5


a
b
c
d
D
e
5cm soil water
10cm soil water
RH
T
Soil water d-excess (‰)
Relative humidity (%)
Temperature (℃)
Sampling time (h)

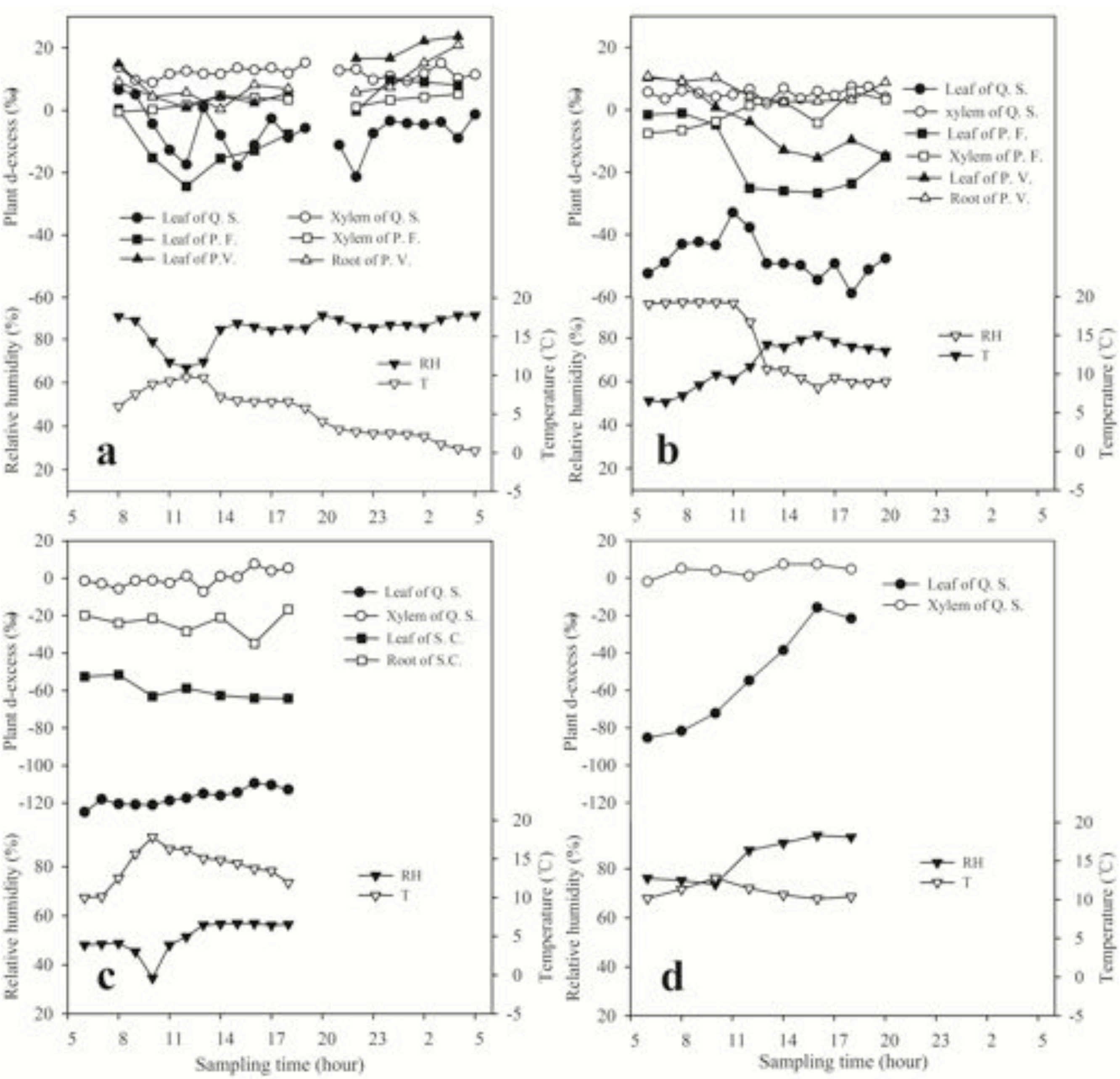
885

Figure 6



886

Figure 7



a
b
c
d
Plant d-excess (‰)
Relative humidity (%)
Temperature (℃)
Sampling time (hour)
Leaf of Q. S.
Xylem of Q. S.
xylem of Q. S.
Leaf of P. F.
Xylem of P. F.
Leaf of P.V.
Leaf of P. V.
Root of P. V.
Leaf of S. C.
Root of S.C.
RH
T

Figure 8

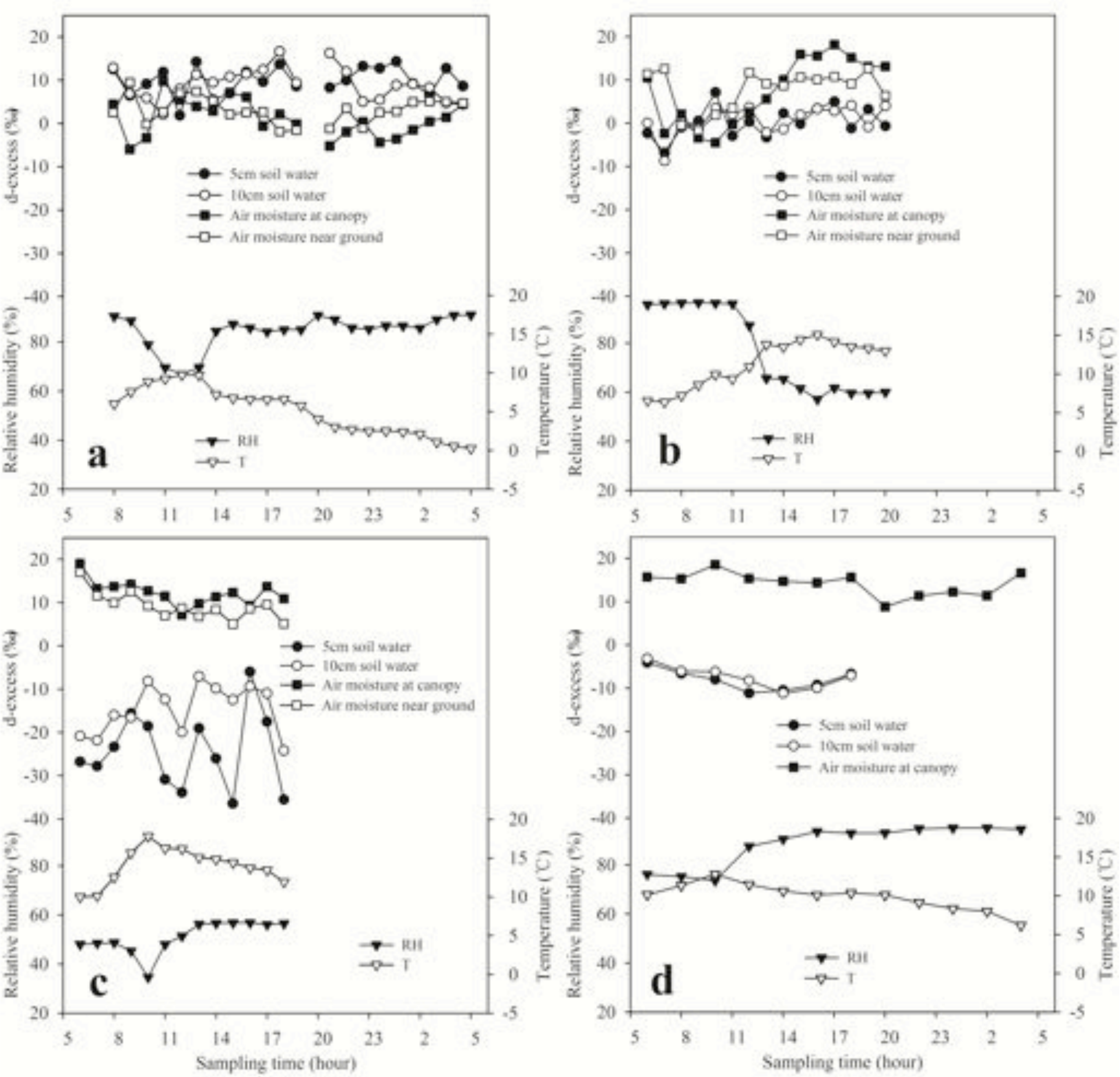

a
b
c
d
5cm soil water
10cm soil water
Air moisture at canopy
Air moisture near ground
RH
T
d-excess (‰)
Relative humidity (%)
Temperature (℃)
Sampling time (hour)

Figure 9

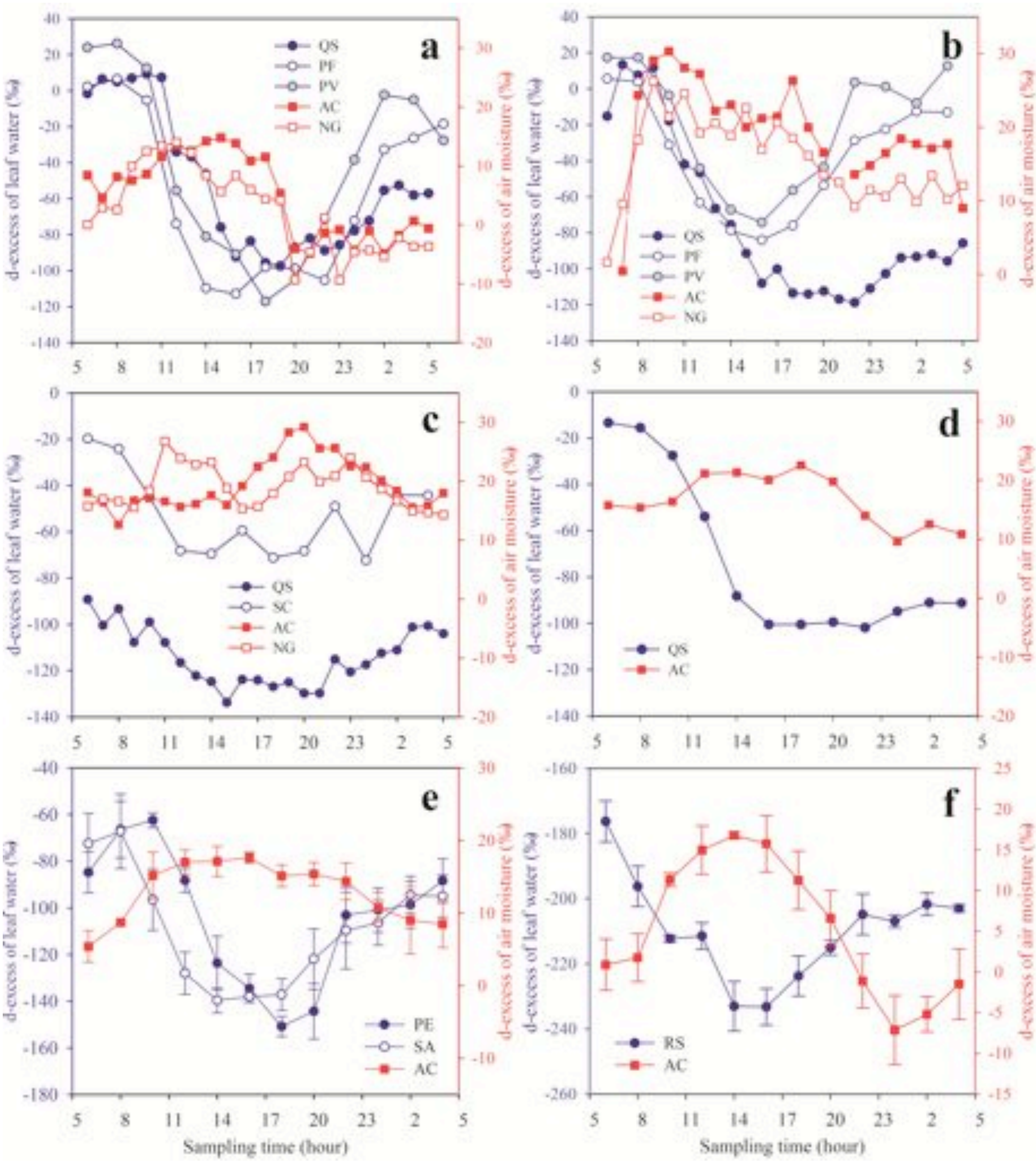
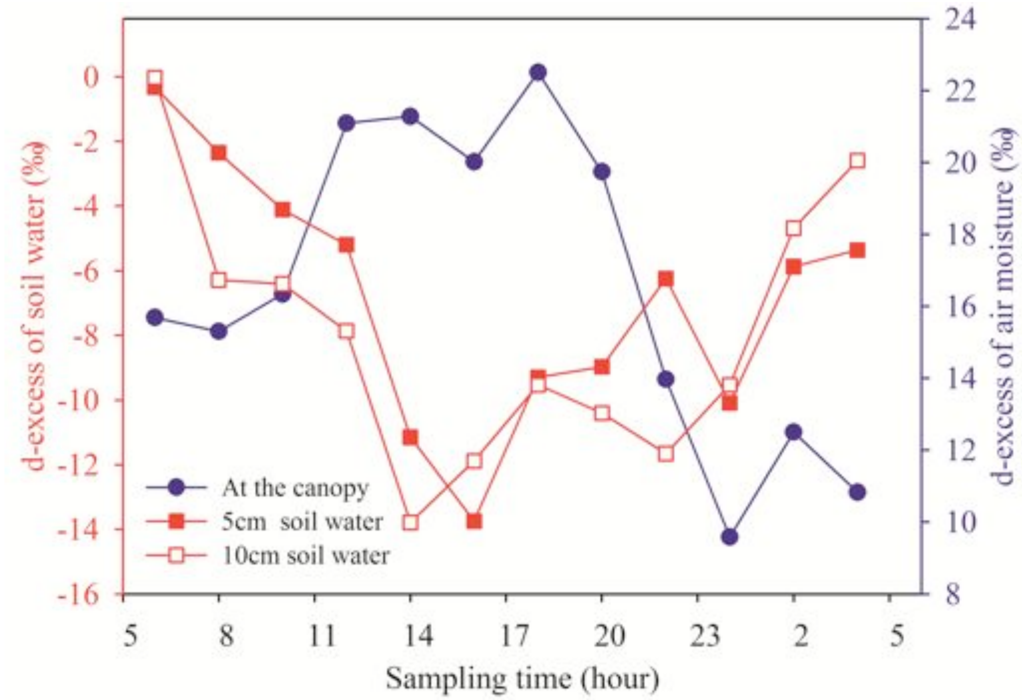
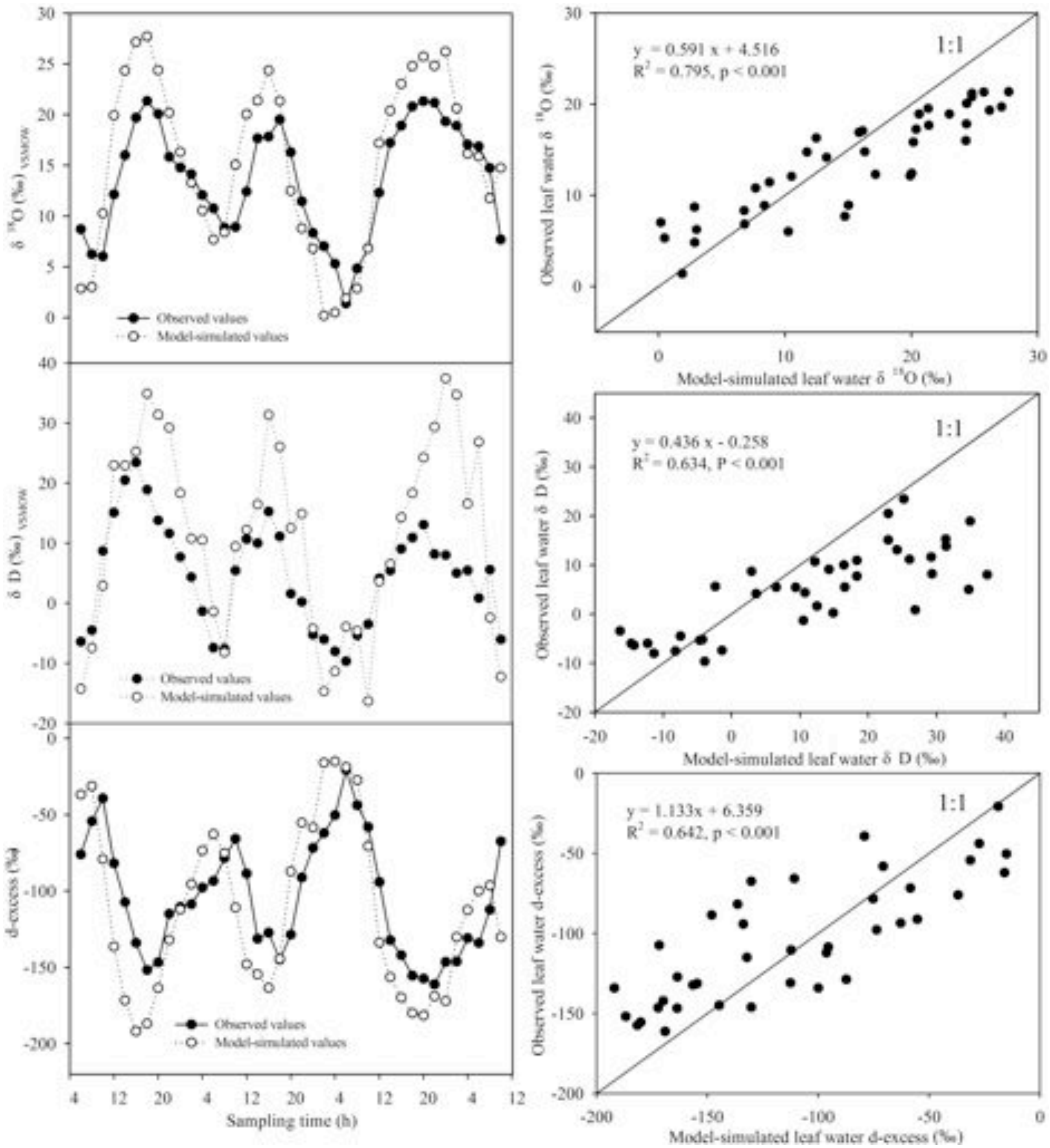

a
QS
PF
PV
AC
NG
b
QS
PF
PV
AC
NG
c
QS
SC
AC
NG
d
QS
AC
e
PE
SA
AC
f
RS
AC
d-excess of leaf water (‰)
d-excess of air moisture (‰)
Sampling time (hour)

Figure 10



d-excess of soil water (‰)
d-excess of air moisture (‰)
Sampling time (hour)
At the canopy
5cm soil water
10cm soil water
0
-2
-4
-6
-8
-10
-12
-14
-16
24
22
20
18
16
14
12
10
8
5
8
11
14
17
20
23
2
5

Figure 11



δ ¹⁸O (‰) VSMOW
Observed values
Model-simulated values
δ D (‰) VSMOW
d-excess (‰)
Sampling time (h)
y = 0.591 x + 4.516
R² = 0.795, p < 0.001
1:1
Observed leaf water δ ¹⁸O (‰)
Model-simulated leaf water δ ¹⁸O (‰)
y = 0.436 x - 0.258
R² = 0.634, P < 0.001
Observed leaf water δ D (‰)
Model-simulated leaf water δ D (‰)
y = 1.133x + 6.359
R² = 0.642, p < 0.001
Observed leaf water d-excess (‰)
Model-simulated leaf water d-excess (‰)