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Key Points:

- A new model is developed to estimate δ_T
- Fifteen centimeter is a reasonable depth for soil water sampling for estimating δ_E
- The δ_{ET} is the most critical member for partitioning

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Partitioning of evapotranspiration through oxygen isotopic measurements of water pools and fluxes in a temperate grassland

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Abstract Stable isotopic measurements of water provide a promising tool for partitioning of ecosystem evapotranspiration (ET). This approach, however, is still facing some challenges due to the uncertainties in estimating the isotopic compositions of ET and its components. In this study, a tunable diode laser analyzer was deployed for in situ measurements of the oxygen isotopic compositions of water vapor. Using these measurements together with samples of water in plant and soil pools, we partitioned ET via estimating the oxygen isotopic compositions of ET (δ_{ET}) and that of its two components, i.e., plant transpiration (δ_{T}) and soil water evaporation (δ_{E}). A new δ_{T} model was developed in this study, which illustrated consistent estimations with the traditional model. Most of the variables and parameters in the new model can be measured directly with high accuracy, making its potential to be used at other sites high. Our results indicate that the ratio of plant transpiration to evapotranspiration (T/ET) illustrates a "U" shape diurnal pattern. Mean T/ET at 0630–1830 during the sampling days was 83%. Soil depth of 15 cm is a reasonable depth for soil water sampling for estimating δ_{E} at this site. We also investigated the uncertainties in estimating these three terms and their effects on partitioning. Overall, in terms of partitioning, the uncertainties are relatively small from δ_{T} and δ_{E} but quite large from δ_{ET}. Quantifying and improving the precision of δ_{ET} should be a priority in future endeavors of ET partitioning via the stable isotopic approach.

1. Introduction

Evapotranspiration (ET) is an important process of ecosystem water budget and energy balance and is closely linked to ecosystem productivity [*Law et al.*, 2002; *Scott et al.*, 2006]. Vegetation transpiration (*T*) and soil evaporation (*E*) are the major components of ET, which are controlled by different biotic and physical processes. Partitioning of ET is therefore critical to predict the climatic responses of ecosystem functions and processes [*Williams et al.*, 2004; *Lauenroth and Bradford*, 2006; *Hu et al.*, 2009]. Being theoretically sound and practically nondestructive, the measurements of the ^{18}O or ^{2}H isotope composition of water vapor fluxes from *E*, *T*, and their combination ET are a promising method of ET partitioning. Several attempts have been made to partition ET using this method in recent years [*Moreira et al.*, 1997; *Yepez et al.*, 2003; *Lee et al.*, 2007; *Xu et al.*, 2008; *Wang et al.*, 2010; *Rothfuss et al.*, 2010].

By using the isotopic method to partition ET, we must first quantify the isotopic composition of *T*, *E*, and ET, respectively. Unfortunately, the estimation of these three terms currently has some degree of uncertainty. The isotopic composition of soil evaporation, δ_E, is generally estimated with the Craig-Gordon model [*Craig and Gordon*, 1965], in which the isotopic composition of ambient water vapor above the canopy (δ_V) is one key parameter. Owing to a technical limitation for continuous measurements, δ_V is commonly assumed a constant over an hourly time scale. Recent studies have found that δ_V is very variable at hourly, diurnal, and seasonal time scales. It is also influenced by hydrological events such as precipitation, dew formation, and dew evaporation [*Lee et al.*, 2006; *Welp et al.*, 2008; *Wen et al.*, 2012]. Therefore, to accurately estimate δ_E, we need to measure δ_V with high accuracy and at fine time scale resolutions. The isotopic composition of water at the soil evaporating front ($\delta_{S,e}$) is another important parameter in the Craig-Gorden model. However, sampling the soil water at a certain depth to estimate $\delta_{S,e}$ is usually quite arbitrary. The effect of sampling soil water at different depths on δ_E estimation remains unclear.

The isotopic composition of plant transpiration, δ_T, can also be estimated with the Craig-Gordon model [*Craig and Gordon*, 1965]. In addition to δ_V, estimating the water isotopic composition at the leaf evaporating front ($\delta_{L,e}$) is critical to resolve δ_T. Conventionally, the estimates of $\delta_{L,e}$ assume that the isotopic composition of transpiration is equal to that of the stem water, i.e., the isotopic steady state assumption [*Wang and Yakir*, 2000]. Recent studies have indicated that the assumption is satisfied only during very limited midday hours [*Farquhar and Cernusak*, 2005; *Lai et al.*, 2006; *Lee et al.*, 2006; *Welp et al.*, 2008], suggesting that estimating daily δ_T under the steady state assumption may be not feasible. *Farquhar and Cernusak* [2005] developed a new model to estimate $\delta_{L,e}$., which resolves the problems of the steady state assumption and performs satisfactorily [*Welp et al.*, 2008]. However, two shortcomings may hinder the wide application of this model to estimate $\delta_{L,e}$. First, there are many parameters in the model, and the estimates of these parameters need labor-intensive sampling [*Farquhar and Cernusak*, 2005]. Second, some parameters such as stomatal conductance, a key parameter in the Farquhar-Cernusak (FC) model, are difficult to measure or estimate at canopy level.

The isotopic composition of evapotranspiration (δ_{ET}), is conventionally inferred by fitting δ_V to the inverse of water vapor mixing ratio, w_a, via the Keeling plot approach [*Keeling*, 1958]. The Keeling plot approach however holds an implicit assumption that the temporal variations of δ_V are attributable only to ET. Recent studies have indicated that, in many cases, the contribution of ET to δ_V variations at the diurnal time scale is small, implying that the Keeling plot-derived δ_{ET} is probably not accurate [*Lee et al.*, 2007]. Therefore, in order to use the Keeling plot approach, one needs substantial changes in δ_V within very short time period (<1 h) with an assumption that ET dominates the variations in δ_V during this short-time period. However, this requirement is hardly met at sites with low ET, such as grassland ecosystems in arid area. Direct measurements of δ_{ET} would be a promising solution of this problem.

Although a number of studies on partitioning ET with the isotopic method have been conducted, the sampling of most previous work (i.e., flasks) is discrete and labor intensive [*Moreira et al.*, 1997; *Wang and Yakir*, 2000; *Yepez et al.*, 2003; *Lee et al.*, 2007; *Xu et al.*, 2008; *Wang et al.*, 2010]. Advances in in situ technology, e.g., tunable diode laser (TDL) absorption spectroscopy, have made refining the isotopic method of partitioning ET quite feasible. With the TDL technique, the measurements of δ_V at hourly to seasonal time scales have been reported [*Lee et al.*, 2007; *Wen et al.*, 2008]. In addition, it also provides a good opportunity for in situ measurements of the isotopic composition of ET using the micrometeorological flux-gradient method, which estimates constituent fluxes based on vertical gradients of scalar concentrations in the atmospheric surface layer [*Yakir and Wang*, 1996; *He et al.*, 2001; *Riley et al.*, 2003; *Lee et al.*, 2007].

In this study, by employing the TDL technique, we continuously measured δ_V and δ_{ET} in a temperate grassland in the growing season (May to September) of 2009 in Inner Mongolia, China. Together with sampling of water in plant leaf laminas, stems, and the soil, we estimated δ_E and δ_T with the Craig and Gordon model for partitioning ET with the isotopic method. In addition, a new model was developed to estimate δ_T with the principles of leaf water isotopic mass balance, avoiding the key problems of the Farquhar-Cernusak model mentioned above. Our objectives in this study are to (1) compare the δ_T estimated with the Farquhar-Cernusak model (referred as FC model in this paper) and the model developed in this study; (2) diagnose the effect of soil water sampling at different depths (5, 15, and 25 cm, respectively) on estimating δ_E and ET partitioning; and (3)explore the potential uncertainties of δ_T, δ_E, and δ_{ET} in terms of partitioning.

2. Theory

Water added to the ecosystem air from ET carries unique isotopic signals from plant transpiration and soil evaporation separately. By measuring the isotopic compositions of *T*, *E*, and ET, the relative contributions of *E* and *T* to the total ET can be determined [*Wang and Yakir*, 2000]. The fractional contribution of plant transpiration f_T to ET can be calculated by [*Yakir and Sternberg*, 2000]:

$$\mathrm{ET} = E + T \tag{1}$$

$$\delta_{\mathrm{ET}}\,\mathrm{ET} = \delta_E E + \delta_T T \tag{2}$$

$$f_T = T/ET = \frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E} \tag{3}$$

where $\delta = R_{\mathrm{sample}}/R_{\mathrm{standard}} - 1$, and the standard is the Vienna standard mean ocean water; *R* is the isotopic ratio $^{18}\mathrm{O}/^{16}\mathrm{O}$).

The parameter δ_E is calculated based on the Craig-Gordon model, i.e., CG model [*Craig and Gordon*, 1965] which accounts for equilibrium and kinetic fractionation during phase change and diffusion of water vapor to the mixed boundary layer near the soil surface [*Wang and Yakir*, 2000]:

$$\delta_E = \frac{\alpha^*\delta_{S,e} - h_S\delta_v - \varepsilon^* - (1-h_S)\varepsilon_k}{(1-h_S) \;+\; (1-h_S)(\varepsilon_k/1000)} \tag{4}$$

where $\delta_{S,e}$ is the isotopic composition of liquid water at the soil evaporating front (we sampled the soil water at 5, 15, and 25 cm, respectively, to estimate this parameter); α^* (<1) is the temperature-dependent equilibrium fractionation factor from liquid to vapor calculated with soil temperature [*Majoube*, 1971]; $\varepsilon^* = 1 - a^*$; ε_k is the kinetic fractionation factor for oxygen (31.9‰; [*Cappa et al.*, 2003], a value of 28.6‰ is used in some cases [*Merlivat*, 1978]); and h_s is the relative humidity normalized to the temperature of the evaporation front.

The parameter δ_T can also be calculated with the CG model:

$$\delta_T = \frac{\alpha^*\delta_{L,e} - h_L\delta_v - \varepsilon^* - (1-h_L)\varepsilon_k}{(1-h_L) + (1-h_L)(\varepsilon_k/1000)} \tag{5}$$

where $\delta_{L,e}$ is the isotopic composition of leaf water at the site of evaporation. The parameter h_L is the relative humidity normalized to the leaf temperature, which was measured by the sensor in the leaf chamber of Li-6400 (LI-COR cor., Lincoln, Nebraska, USA). Owing to the difficulty in measuring or calculating, $\delta_{L,e}$ is conventionally estimated with the steady state assumption [*Wang and Yakir*, 2000]:

$$\delta_{L,es} = \delta_S + \varepsilon^* + \varepsilon_k + h_L(\delta_v - \varepsilon_k - \delta_S), \tag{6}$$

where δ_S is the isotopic composition of stem water. *Farquhar and Cernusak* [2005] proposed a model to estimate $\delta_{L,e}$ which solves the problems caused by the steady state assumption:

$$\Delta_{L,e} = \Delta_{L,es} - \frac{\alpha_k\alpha^+}{gw_i}\frac{d(W\Delta L)}{dt} \tag{7}$$

$$\delta_{L,e} = \Delta_{L,e}(\delta_S + 1000) + \delta_S \tag{8}$$

$$\Delta_{L,es} = \frac{R_{L,es}}{R_S} = \frac{\delta_{L,es} - \delta_S}{\delta_S + 1000}, \tag{9}$$

where g is the leaf stomatal conductance (mol m^{-2} s^{-1}). The w_i is the saturated water vapor at the temperature of the water at the sites of evaporation (mol mol^{-1}). W is the water storage in the leaf lamina (mol m^{-2} leaf). ΔL is the isotopic composition of bulk leaf relative to that of the stem. R_S, $R_{L,es}$, and $R_{L,e}$ are the isotopic ratio (heavy to light) (^{18}O/^{16}O) of the stem water and the water at the leaf evaporating front with and without the steady state assumption.

In this study, we developed a new model of δ_T using the mass conservation principle at the ecosystem level. We consider the changes of the water storage in the leaf lamina (ΔW mol m^{-2} leaf) during a time interval (3600 s in this study, i.e., 1 h) to be equal to the accumulated flux into the leaf lamina from the stems ($\sum J$ mol m^{-2} leaf) and lost from the leaf by transpiration ($\sum T$ mol m^{-2} leaf):

$$\Delta W = \sum J - \sum T. \tag{10}$$

Assuming no changes in isotopic composition of influx and outflux during the time interval, the change of the amount of the heavy isotope $\Delta(W\delta_L)$ are correspondingly linked by:

$$\Delta(W\delta_L) = \delta_S\Sigma J - \delta_T\Sigma T \tag{11}$$

where δ_L is the isotopic ratio of the water in leaf lamina. Eliminating $\sum J$ from equation (10) and equation (11), we obtain

$$\Delta(W\delta_L) = \delta_S(\Delta W + \Sigma T) - \Sigma T\delta_T. \tag{12}$$

We solve for total T during the interval:

$$\mathrm{LAI}\sum T = f_T \sum \mathrm{ET} = \frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E}\sum \mathrm{ET}. \tag{13}$$

where LAI is leaf area index. Taking equation (13) into equation (12), we get the solution of δ_T:

$$\delta_T = \frac{\delta_S \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) + \mathrm{LAI}\delta_E[\Delta(\delta_L W) - \delta_S \Delta W]}{\sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) + \mathrm{LAI}[\Delta(\delta_L W) - \delta_S \Delta W]}. \tag{14}$$

Details on deriving equation (14) are available in the Appendix A. Notably, when use this model for estimating δ_T, care must be taken that the sampling interval is short to guarantee δ_L and δ_S remain nearly constant (see equation (11)). In this study, we linearly interpolated δ_L and δ_S to 1 h resolution, assuming δ_L and δ_S remain constant within 1 h.

Both the FC model and our new model need W, δ_L, and δ_S as input variables, which can be measured by sampling leaf laminae and plant stems. In the FC model, in addition to the variables needed to estimate $\delta_{L,e}$ (equation (6), e.g., δ_V, relative humidity, leaf temperature, ε_k, and ε^*), g, and w_{i} must also be measured (or estimated) for inclusion (equation (7)). In our model, ET, δ_E, and δ_{ET} must also be measured or estimated (equation (14)). ET can be measured with eddy covariance technique. The parameter δ_{ET} can be measured with TDL technique. The parameter δ_{E} can be estimated with CG model or measured directly with the chamber method developed in recent years [e.g., *Yepez et al.*, 2005; *Rothfuss et al.*, 2010; *Wang et al.*, 2010].

3. Materials and Methods

3.1. Site Description

The experiment was conducted in Duolun County (42°02′N, 116°17′E, 1324 m above sea level), a semiarid area located in Inner Mongolia, China. The mean annual precipitation during 1953–2007 is 383 mm, of which more than 80% falls in the growing season (May to September). The mean annual temperature is 2.1°C with the monthly mean temperature ranging from 18.9°C in July to −17.5°C in January. The sandy soil of the study site is classified as chestnut soil according to the China's soil classification system, or Haplic Calcisols according to the Food and Agriculture Organization classification, with 63% sand, 20% silt, and 17% clay [*Niu et al.*, 2010]. The site has been fenced since 2001 as a long-term study plot and is dominated by C_3 species, including *Stipa kryroii*, *Agropyron cristatum*, and *Artemisia frigida* [*Zhang et al.*, 2007; *Xia et al.*, 2009]. According to our measurements, the three dominant species account for approximately 75% of the total aboveground biomass of the community in the peak growing season. The vegetation height is 0.3–0.4 m during the peak of the growing season.

3.2. Isotope Measurements

The system for in situ measurements of water vapor $^{18}\mathrm{O}/^{16}\mathrm{O}$ isotope ratio consisted of a tunable diode laser analyzer (model TGA-100A, Campbell Scientific Inc., Logan, UT) and a dripper calibration system [*Wen et al.*, 2008]. The principle of our operation in this study was described by *Lee et al.* [2006] and *Wen et al.* [2008]. Briefly, a dripper calibration system produced moist air with an isotopic composition identical to a liquid source reservoir that dripped into a heated evaporation flask, ensuring immediate, complete evaporation of the water. The calibration stream was split into two, and the humidity of one of the moist airstreams was reduced by 10% by mixing in dry air to give a calibration range of humidity. The humidity of the two moist airstreams was controlled by the rate of water dripping, and the flow of dry air through the dripper system such that the ambient humidity was bracketed by the calibration standards.

Six air species were measured in sequence: one zero gas (i.e., dry air); two span gases produced by the dripper system; and three ambient intake gases at the heights of 0.2, 0.7, and 1.7 m (the ambient air measured at 0.2 m was not used in this study). The sampling line is approximately 20 m in length and insulated with a heated cable to avoid water vapor condensation. Flows through the calibration intakes were controlled at about 0.3 L min^{-1} by stainless steel precision needle valves, and flows in the air intakes were controlled at 0.6–2 L min^{-1} by stainless steel critical orifices. A subsample of one of the six intakes was drawn into the analyzer's sample cell at a rate of 0.2 L min^{-1}. Each intake was measured for 20 s at a frequency of 1 Hz, and

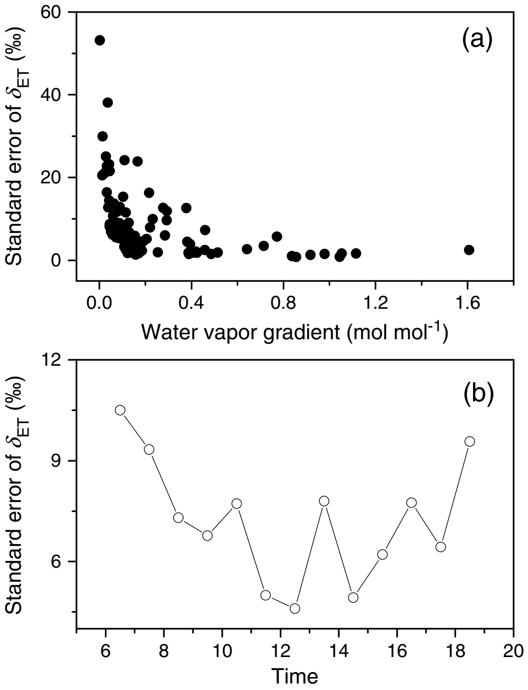

Figure 1. (a) Effect of the water vapor gradient between the two sampling intakes on δ_{ET} uncertainty. (b) Mean diurnal variations in the uncertainty of δ_{ET}.

the two ambient intakes (0.7 and 1.7 m) were switched in every 2 min to exclude systematic bias (i.e., one measuring cycle was 4 min). At the intake of the analyzer, a bypass was configured to ensure that each air species could arrive at the analyzer within 1 s. After the data of first 10 s was discarded, all measurements during the second 10 s period were averaged. The δ_V was calibrated in every cycle using the zero gas and the two span gases [see *Wen et al.*, 2008, equations (2)–(6)]. The system nonlinearity was checked hourly to ensure that the difference between the two calibration streams was within the tolerable threshold of 0.5‰ [*Wen et al.*, 2008]. The measurements were excluded for analysis in the cases where the difference exceeded the tolerable threshold.

Hourly δ_{ET} was estimated with the flux gradient approach [*Yakir and Wang*, 1996] as described and tested by *Griffis et al*. [2005] and *Lee et al*. [2007]. The vertical gradient in water vapor concentration and isotopic composition was measured by the TDL via two intakes above the canopy (0.7 m and 1.7 m). R_{ET} was calculated by

$$R_{ET} = R_d \frac{x^{16}_{s,2} - x^{16}_{s,1}}{x^{18}_{s,2} - x^{18}_{s,1}} \frac{x^{18}_{a,1} - x^{18}_{a,2}}{x^{16}_{a,1} - x^{16}_{a,2}}, \qquad (15)$$

where R_d is the isotopic ratio of the dripper calibration water, superscripts 16 and 18 denote the ^{16}O and ^{18}O molecules in water; s_1 and s_2 indicate span calibration vapor streams 1 and 2, and a_1 and a_2 represent the ambient air sampled at two heights. R_{ET} of each measuring cycle was calculated with equation (15), and the hourly R_{ET} was calculated as the average of these 4 min R_{ET} within each hour. Ideally, 15 R_{ET} would be available for estimating the hourly values. However, due to the data quality control (if the measured values of the isotopic composition of the calibration gas deviated from the real value beyond 0.5‰, the data would be excluded), the number of R_{ET} was different for each hour. The standard errors of those δ_{ET} in each hour were calculated to quantify the uncertainty of hourly δ_{ET} [*Zhang et al*., 2006]. Results indicate that the δ_{ET} uncertainty varied from 0.8‰ to 53‰, with an average of 7.9‰. As expected, the uncertainty was higher when the water vapor gradient between the two sampling intakes was smaller (Figure 1a). The diurnal pattern illustrated that the δ_{ET} uncertainty was higher in early morning and late afternoon, and lower in the middle day hours (Figure 1b). In order to minimize the effects of the high uncertainty from δ_{ET} on ET partitioning, an uncertainty threshold of 15.8‰ (i.e., twice the average) was set to exclude the low-quality δ_{ET} data.

3.3. Ancillary Measurements

In addition to TDL measurements of the ^{18}O of ambient water vapor and δ_{ET}, we also made ^{18}O measurements of water in leaf laminae, stems, and the soil at 5 cm, 15 cm, and 25 cm depths. We collected the leaves, stems, and soil samples every 3 h (mostly from 06:30 to 18:30) on six sunny days (day of year (DOY) 178, 203, 207, 219, 223, and 224) in 2009. Each fresh plant sample was approximately 10 g, and soil sample was 30–50 g. The plant and soil samples were sealed in glass vials and frozen until cryogenic water extraction using a vacuum line. Extracted waters were analyzed for ^{18}O composition using a liquid isotope water analyzer (LGR DLT-100, Los Gatos Research, Inc., USA) and corrected for organic contaminants with the method of *Xiao et al*. [2012]. The average correction was 8.1‰ for leaf water samples, 2.5‰ for stem water samples, and 0.4‰ for soil water samples. The mean difference between the LGR and the isotope ratio mass spectrometer measurements was 0.3‰ [*Xiao et al*., 2012].

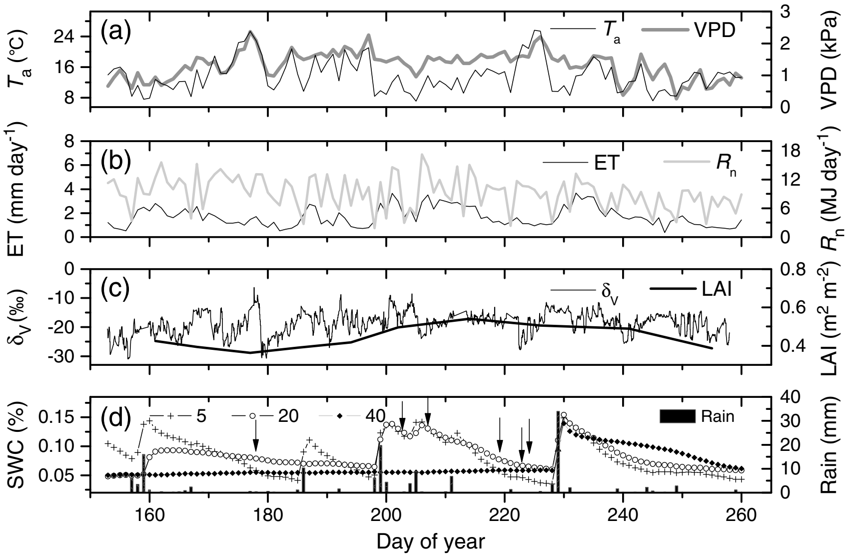

Figure 2. Seasonal variations in (a) daily air temperature and water vapor pressure deficit, (b) evapotranspiration and net radiation (R_n), (c) the hourly isotopic composition of water vapor in the air (δ_V) and leaf area index, and (d) rainfall and soil water content at the depth of 5, 20, and 40 mm The arrows in Figure 2d indicate the 6 days on which leaf water, stem water, and soil water were sampled for ^{18}O measurements.

Leaf stomatal conductance (Li-6400, LI-COR cor., Lincoln, Nebraska, USA) and water content of the three dominant species (*Stipa kryroii*, *Agropyron cristatum*, and *Artemisia frigida*) were measured every 3 h. We weighed fresh leaf biomass immediately after sampling the leaves from the field (the samples were sealed in plastic-filmed bags) and then again weighed the dry leaf biomass after drying in a 75°C oven for 48 h. The leaf area was measured with a portable leaf area meter LI-3000A (Licor Inc., Lincoln, Nebraska) to calculate the specific leaf area (used to express leaf water content on the basis of leaf area, mol m^{-2}). LAI was estimated through harvesting aboveground biomass and measuring the leaf area in 2006 at the study site by *Zhang et al.* [2007]. We established a linear function between normalized difference vegetation index (NDVI, 250 m, from http://fluxnet.ornl.gov) and LAI (LAI = 0.2 NDVI−0.36, R^2 = 0.96). LAI was estimated with the linear function and NDVI in the study year. Values of δ_L, δ_S, g, and W for the whole canopy were weight averaged based on the aboveground biomass of the three dominant species. To get more detailed information on the diurnal dynamics, we linearly interpolated the values based on the 3 h samplings or measurements to hourly values.

3.4. Meteorological Measurements

An open-path eddy covariance (EC) system and a meteorological system were installed at a height of 1.7 m to monitor H_2O fluxes and the meteorological variables. The EC system consists of an open-path CO_2/H_2O gas analyzer (LI-7500, Licor Inc., Lincoln, Nebraska) and a 3-D sonic anemometer/thermometer (model CSAT3, Campbell Scientific Inc., Logan, Utah). Detailed information of the sensors and the flux data processing was described by *Hu et al.* [2009]. The raw fluxes of water vapor were calculated online at 30 min intervals and postprocessed to calculate ET. The meteorological variables, including air temperature (T_a); canopy temperature; soil volumetric water content and soil temperature at 5, 20, and 40 cm; and water vapor pressure were measured simultaneously with the eddy flux system and calculated at 30 min intervals.

4. Results and Discussion

4.1. Seasonal Variations in Main Environmental and Biotic Variables

Mean daily air temperature during the growing season (DOY 150–260) varied between 7 and 25°C (Figure 2). As expected, vapor pressure deficit (VPD) illustrated similar seasonal fluctuations as air temperature, with a range of 0.4 to 2.5 kPa and an average of 1.6 kPa. Evapotranspiration varied in the range of 0.3–3.4 mm d^{-1}, with four distinct peaks during the growing season, which were likely driven by soil water content rather than

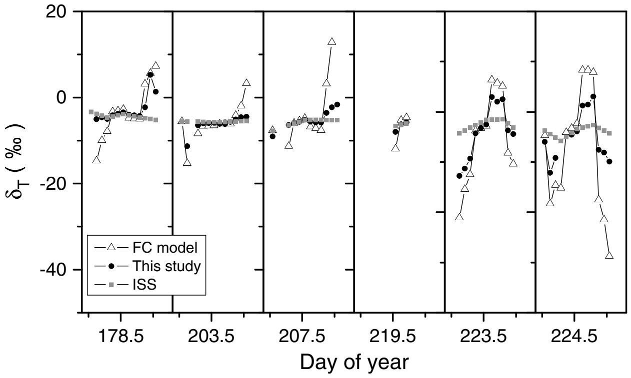

Figure 3. Comparison of the isotopic composition of plant transpiration (δ_T) developed in this study and the Farquhar-Cernusak model. ISS is δ_T calculated with the isotopic steady state assumption.

net radiation. Leaf area index at the study site was quite low, with a maximum of 0.5 $m^2 m^{-2}$ at the beginning of August in 2009. The isotopic composition of water vapor in the air, δ_V, varied from −32‰ to −7‰ with a mean of −18.8‰, which was a bit lower than the values observed at the site in Beijing, China (−15.5‰) [*Wen et al*., 2010] and in Great Mountain, USA (−16.8‰) [*Lee et al*., 2006]. Similar to the findings of previous reports [*Lee et al*., 2006; *Wen et al*., 2010], the seasonal cycle included several weather cycles lasting 5–10 days, which produced isotopic anomalies on the order of 10–25‰ (Figure 2). Soil moisture at the depth of 5 cm was sensitive to the rainfall events, but that at the depth of 40 cm remains constant during most of the growing season (Figure 2d). In general, the whole growing season consisted of four distinct periods of intensive rainfall events, which resulted in four peaks of soil water content.

4.2. Comparisons of δ_T Estimated With the FC Model and the New Model

Figure 3 illustrates the diurnal variations in δ_T estimated with the two models in the six sampling days. The results indicate that δ_T values estimated with the two methods were in good agreement both in terms of absolute magnitude and diurnal variability. Especially in late morning and early afternoon (1030–1530), the determinant coefficient of the linear regression between the estimates of the two methods was 0.94 ($y = 1.3x + 0.6$). The bulk mean of δ_T in midday to early afternoon was −3.0‰ (±5.1) when estimated with FC model and −3.7‰ (±3.1) when estimated with our model. In most cases, δ_T ranged from −30‰ to 10‰ during the study period at our site. In general, δ_T steadily increased from the early morning (0630) until middle afternoon (1530) and then gently decreased (Figure 4). In this study, we found δ_T progressively increased from early morning to afternoon. Similarly, *Lai et al*. [2006] and *Griffis et al*. [2010, 2011] reported the same pattern at a coniferous forest in southern Washington and a cropland in Minnesota. However, *Lee et al*. [2007] found a contrary diurnal pattern of δ_T in a mixed forest in Connecticut. We speculate that $\delta_{L,e}$ determines the diurnal changes in δ_T in the ecosystems, which show an increase in δ_T from early morning to late afternoon. However, we believe that relative humidity controls the variations in δ_T in the ecosystems, which display the contrary pattern. Our results illustrate that δ_T at this site met the isotopic steady state (ISS) assumption during the midday hours (Figure 4), which is consistent with previous studies [*Lai et al*., 2006; *Welp et al*., 2008].

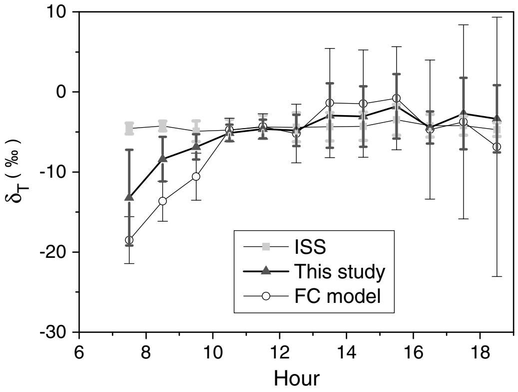

Figure 4. Mean diurnal variations in the isotopic composition of plant transpiration during the six sampling days estimated with three methods: the isotopic steady state assumption, the FC model, and the model developed in this study.

Notably, in early morning and late afternoon, δ_T estimated with the FC model was usually smaller than that estimated with our model when δ_T was negative, but bigger when δ_T was positive, causing a higher diurnal variability for the FC model (Figure 3). We investigated potential sources of uncertainty to identify the cause of the divergence of the two models.

The first cause may be from our model due to the uncertainties of ET and δ_{ET} measurements. Previous studies illustrated no obvious diurnal pattern of ET uncertainties measured with eddy covariance systems [*He et al*., 2010]. However, δ_{ET} measurements in the early morning and late afternoon may have large uncertainties due to the small water vapor gradient between the two sampling intakes [*Lee et al*., 2007]. Assuming an uncertainty as big as 20% in ET, 15.8‰

in δ_{ET} (the threshold for excluding low-quality δ_{ET}), the corresponding uncertainties of δ_T were less than 0.5‰ and 1.4‰, respectively. This implies that ET and δ_{ET} measurements should not be the main sources of the different estimates of the two models in the early morning and late afternoon.

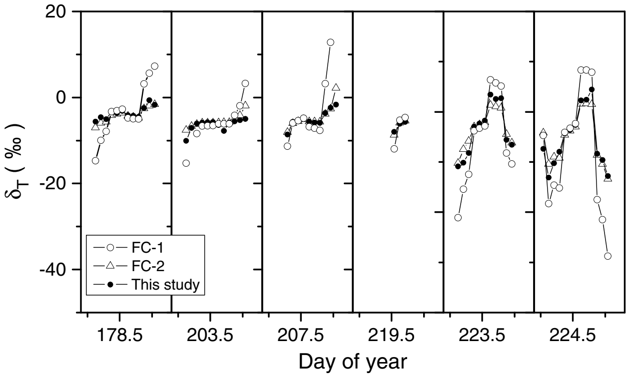

Figure 5. Diurnal variations in the isotopic composition of plant transpiration estimated with the FC model and the model developed in this study. FC-1: canopy level stomatal conductance was estimated with aboveground biomass-weighted average of the stomatal conductance of the three dominant species (the same as Figure 2). FC-2: canopy level stomatal conductance was "tuned" based on the measurements of one dominant species (see details in the text).

The second cause may be from the FC model due to the uncertainties of stomatal conductance measurements. Canopy level stomatal conductance was estimated using the biomass-weight average method in this study. However, the leaf shape of the two dominant species *Stipa kryroii* and *Artemisia frigida* was irregular, causing potentially large uncertainty in the measurements. We tried to "tune" canopy level *g* with different schemes, e.g., simple bulk average, leaf-weight average, and aboveground biomass-weight average. The results indicated that different schemes yield quite different magnitudes of diurnal variability in δ_T. For example, if we use twice of the *g* of *Agropyron cristatum*, whose measurements were reliable, the divergence disappeared, and the two models yield nearly identical δ_T throughout the study period (Figure 5). This result suggests that stomatal conductance is the key source of uncertainty in the FC model and the main cause of the divergence between the FC model and the model developed in this study. Much attention should be paid to *g* calculation when using the FC model at the ecosystem level in future.

Note that the leaf water content and the isotopic compositions of leaf and stem water are needed in both the FC model and our model. Differences among species may result in biased estimate of the canopy level values and introduce uncertainty in the prediction of δ_T and ET partitioning. We estimated the canopy level values with different average schemes. The results indicated that different methods yielded nearly identical δ_T and *T*/ET (data not shown). This implies that the differences in leaf water content and the isotopic compositions of leaf and stem water among plant species will introduce a minor uncertainty in estimating δ_T and *T*/ET.

With δ_{ET} measured using the flux-gradient method and δ_E estimated with CG model (soil water at the depth of 15 cm), we further compared the partitioning performances between the two δ_T models. The results indicate that the two methods yielded nearly the same *T*/ET during most periods throughout each day (Figure 6). The consistent estimation of δ_T with the FC model lends credit to the model developed in this study. In our model, most of the variables and parameters can be measured directly with high accuracy. This increases the potential for it to be used at other sites, especially where direct measurements of leaf stomatal conductance are difficult. Notably, both δ_{ET} and δ_E are input variables, which may be the main uncertainties in

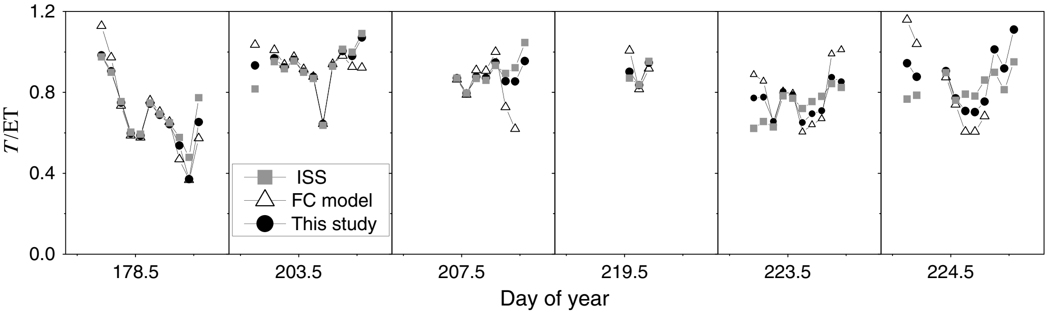

Figure 6. Diurnal variations in the ratio of plant transpiration to evapotranspiration during the six sampling days. *T*/ET was calculated with δ_T estimated with ISS assumption, FC model, or the model developed in this study.

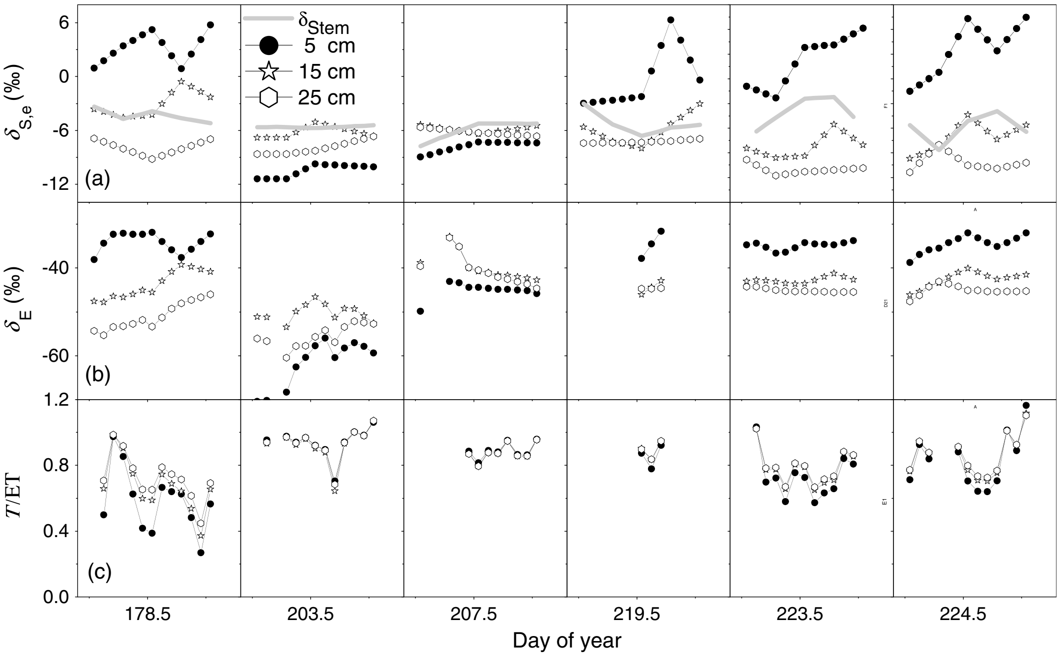

Figure 7. (a) Differences in the isotopic composition of soil water at different depths, comparing with the isotopic compositions of stem, and (b) their effects on the estimate of the isotopic composition of soil water evaporation, δ_E, and (c) on corresponding ET partitioning. The δ_T estimated with the FC model and δ_{ET} estimated with flux-gradient method were used for partitioning.

estimating δ_T using our model. Especially, δ_{ET} measurements tend to have large uncertainties in the conditions with low ET. However, our sensitivity analysis indicated that a change of 10% in δ_{ET} or δ_E will result in a corresponding change of 2% or 3% in δ_T. This means that the uncertainties from δ_{ET} and δ_E could be largely attenuated in terms of estimating δ_T. In addition, some novel methods developed for measuring δ_{ET} and δ_E directly with a chamber would make our model more accurate and conveniently used [*Yepez et al.*, 2005; *Rothfuss et al.*, 2010].

4.3. Effects of Sampling Soil Water at Different Depths on Estimating δ_E

The isotopic compositions of soil water at different depths (i.e., δ_{S_5}, δ_{S_15}, and δ_{S_25}) illustrated certain of differences. The differences were relatively big at days with low soil water content but small at days with high soil water content (e.g., DOY 203 and DOY 207) (Figure 7a). On average, the difference in midday to early afternoon (11:30–14:30) was 2.8‰ between δ_{S_5} and δ_{S_15} and 1.4‰ between δ_{S_15} and δ_{S_25} in the two wet days, and 7.7‰ and 2.9‰ in the other dry days. Comparing with δ_S, the differences of δ_E diminished among soil depths (Figure 7b). For example, the difference between δ_{E_5} and δ_{E_15} was 6.9‰ in 11:30–14:30 of the two wet days, which was 13.5% of the mean of δ_{E_5}, but the corresponding difference in δ_S was as big as 33.3%. In general, the range of variations in δ_E at the three soil depths was approximately -70‰ to -30‰, which is consistent with the results of previous studies illustrating the variations from -50‰ to -30‰ [*Wang and Yakir*, 2000; *Yakir and Sternberg*, 2000; *Yepez et al.*, 2003; *Xu et al.*, 2008].

T/ET calculated with different depths of $\delta_{S,e}$ was quite consistent in most cases (Figure 7c). The partitioning results were nearly identical in the two wet days (DOY 203 and DOY 207). However, a big discrepancy in *T*/ET was observed in the midday of DOY 178, when the difference in soil volume water content at the depth of 5 cm and 20 cm was the largest throughout the six sampling days (Figure 2d). *T*/ET calculated with δ_S at 5 cm was 0.35, whereas it was 0.56 at 15 cm and 0.62 at 25 cm. It is widely acknowledged that plant transpiration accounts for most of ET in the condition with low soil moisture [*Scanlon and Albertson*, 2004; *Lauenroth and Bradford*, 2006; *Raz-Yaseef et al.*, 2012]. It seems that sampling soil water at the depth of 15 and 25 cm produced more reasonable results than that at 5 cm. In addition to the days near DOY 178, three more periods with large differences of soil water content existed among different depths: DOY 160–170, 186–190, and 240–254

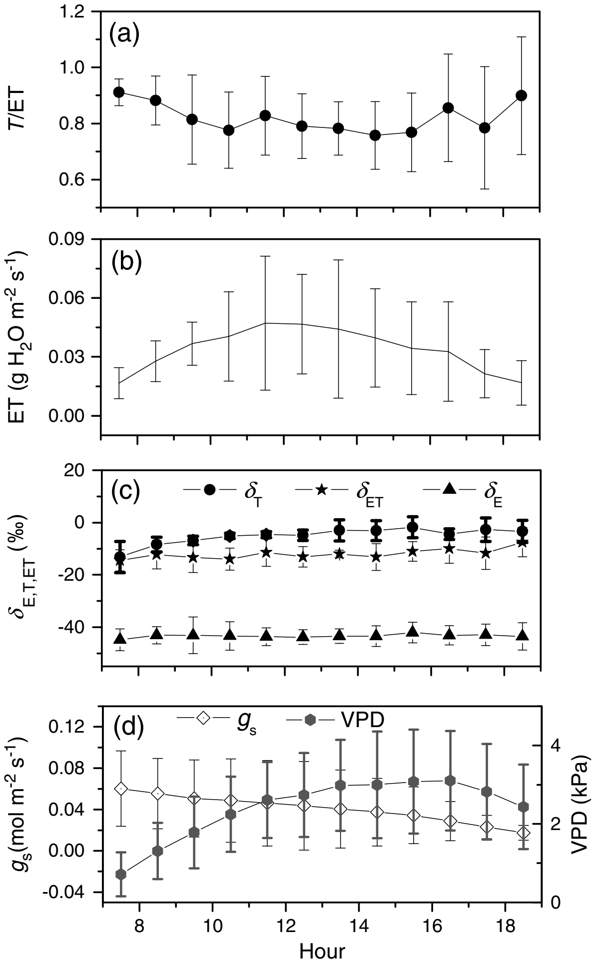

Figure 8. Mean diurnal variations in the (a and b) partitioning results (*T*/ET) and (c and d) relevant variables (δ_E, δ_T, δ_{ET}, stomatal conductance (g_s), VPD, and ET) during the six sampling days.

(Figure 2d). During these periods, sampling soil water at a depth which is too shallow may bring in bias in estimates of δ_E. Considering that more than 80% root biomass is distributed in the first 30 cm soil layer at this grassland, we assume that the soil water, where both *E* and *T* originate, is at the same depth and varies with soil water content. As Figure 7a illustrates, the isotopic composition of stem water was close to that of soil water at the depth of 15 cm. This implies that soil sampling at the depth of 15 cm seems to be reasonable in most cases at our study site.

4.4. Mean Diurnal Pattern of ET Partitioning

Bulk average *T*/ET indicates that plant transpiration accounted for 83% of ET at 0630–1830 during the six sampling days. Ensemble *T*/ET illustrates a U shape diurnal pattern; i.e., a progressive decrease in *T*/ET was found from 0630 to 1030. *T*/ET remained relatively constant during 1030 to 1530 (approximately 75%) and gently increased again in the late afternoon (Figure 8a). In contrary, ET flux increased from early morning until 11:30 and then progressively decreased (Figure 8b). In general, the diurnal changes of *T*/ET were mainly contributed by δ_{ET} and δ_T, and δ_E remained relatively constant (Figure 8c). The diurnal variations in *T*/ET seem to be controlled by the interaction of canopy stomatal conductance and VPD. In the mornings, relatively high *g* and low VPD favor plant transpiration, but not soil water evaporation, resulting in high T/ET (Figure 8d). In the afternoons, however, as *g* declined and VPD increased, increases in soil evaporation are greater than plant transpiration, which lowers *T*/ET. The gentle increase of *T*/ET in the late afternoon seems to be the result of the decrease of VPD, which may cause substantial reduction of soil water evaporation.

The estimated *T*/ET at this site is, in general, consistent with previous studies using the stable isotopic method. For example, *T*/ET was approximately 80% in the midday at our grassland site. *Wang and Yakir* [2000] reports a value of 96–98% in a mature wheat field with dense canopy. *Wang et al.* [2010] found *T*/ET range from 61% to 83% in the Biosphere 2 greenhouse, where the wood cover ranges from 25% to 100%. Our study illustrates that *T*/ET was lower at the days with high soil water content (DOY 203 and DOY 207) than that at the days with low soil water content. This is consistent with the work of *Yepez et al.* [2005], who found an increase of *T*/ET after a 39 mm rain event. Although many attempts have been made to partition ET using a stable isotopic approach, seldom such studies address factors controlling the temporal variations in *T*/ET, due to lack of continuous measurements,. Using a two-source model, *Zhu et al.* [2013] found that diurnal *T*/ET pattern was controlled by stomatal conductance at a wet alpine meadow site. At the arid site in our study, we found that in addition to stomatal conductance, VPD was also of major influence in the afternoon. In addition, most model studies illustrate that *T*/ET is mainly determined by canopy stomatal conductance or LAI at seasonal scale [*Lauenroth and Bradford*, 2006; *Hu et al.*, 2009; *Huang et al.*, 2010; *Zhu et al.*, 2013]. It is quite necessary to compare these findings with the model results through the stable isotopic approach. Although the estimated T/ET in this study was, in general, agreement with previous reports, it failed to be validated with an independent approach. Integrating of some other partitioning methods, such as chamber method, micro lysemeter, sapflow, as well as modeling should be necessary in future work.

4.5. Uncertainties in δ_E, δ_T, and δ_{ET} and Effects on ET Partitioning

All of the three terms, δ_E, δ_T, and δ_{ET}, may introduce uncertainties in terms of ET partitioning. To identify the principal uncertainties, we quantified the sensitivity of T/ET to changes in the three terms and explored the sources of uncertainties for each term via error propagation analysis. There may be two main sources of uncertainty for estimating δ_E with the Craig-Gordon equation. The first one is the use of the kinetic fractionation factor ε_k. A popular value for ε_k is 31.9‰ [*Cappa et al.*, 2003] or 28.6‰ [*Merlivat*, 1978]. The use of different values of ε_k will result in approximately 3‰ difference in δ_E. Second, δ_V is an important factor affecting δ_E. We have fully tested the accuracy of TDL measurements of δ_V, finding that the precision is 0.07‰ for ^{18}O/^{16}O at the dew point temperature of 8°C [*Wen et al.*, 2008]. Obviously, the precision of δ_V will introduce minor uncertainty in estimating δ_E. With all the uncertainties from other sources, e.g., relative humidity and soil temperature being taken into account, the total uncertainty of δ_E might be less than 5‰, which could introduce a partitioning (i.e., T/ET) uncertainty of 2.8%.

In terms of estimating δ_T with the new model, according to equation (14), potential uncertainties might be from leaf water content, ET, δ_S, δ_E, and δ_{ET}. Being directly sampled or measured, the leaf water content, ET and δ_S, would introduce very subtle uncertainties on δ_T. However, δ_E and δ_{ET} may introduce some uncertainties on δ_T. Assuming an uncertainty of 5‰ for δ_E or 7.9‰ for δ_{ET} (i.e., the average δ_{ET} uncertainty during the six sampling days), the corresponding uncertainty of δ_T estimated with our new model would be 0.1‰ (for δ_E) or 0.3‰ (for δ_{ET}). Obviously, the uncertainties from δ_E and δ_{ET} have been largely diminished in terms of estimating δ_T. Therefore, the total uncertainty of δ_T estimated with our model seems to be less than 1‰.

The parameter δ_{ET} was measured with the flux-gradient method by the TDL system in this study. Potential uncertainties in δ_{ET} measurements include instrument precision, variability in atmospheric conditions, different footprints at the two intake heights, and the averaging method to calculate hourly δ_{ET} [*Good et al.*, 2012]. Unfortunately, we failed to fully quantify the uncertainties of δ_{ET} measurements. We provided an imperfect estimation of the uncertainty by calculating the standard errors of δ_{ET} within each hours. The results illustrated an average uncertainty of 7.9‰ in δ_{ET}, which is obviously much larger than that of δ_T and δ_E. Similarly, in terms of partitioning net ecosystem carbon exchange (NEE) with the stable isotopic method, previous work also found that the uncertainty from δ_{NEE} was much larger than that of its components, i.e., gross primary productivity and ecosystem respiration [*Zhang et al.*, 2006]. With the same version of the instrument as the one in this study, *Lee et al.* [2007] investigated the accuracy of TDL in measuring δ_{ET} via a series of laboratory tests. Their study illustrated that the TDL instrument would yield a precision of 1.4‰ for δ_{ET} under the conditions that the vertical vapor gradient was 0.3–0.5 mmol mol^{-1}. Our approach yielded an uncertainty of 3.1‰ within the similar water vapor gradient. It seems like the uncertainty from the field is much larger than that in laboratory. Noteworthy, we sampled plant and soil water to estimate δ_E and δ_T in the nights of DOY 219 and DOY 224. The result indicated that most of the T/ET calculated was out of the rational range, i.e., 0–1 (data not shown). Also, the estimated T/ET was occasionally over 1.0 in the early morning and late afternoon in the sampling days (Figure 6). We speculate that the unrealistic estimate is probably due to the inaccurate measurements of δ_{ET} when the gradient was very small (Figure 1). Suppose an uncertainty of 7.9‰ in δ_{ET}, the sensitivity analysis indicated that this would introduce an uncertainty of 21.4% in T/ET. Obviously, the uncertainty of ET partitioning arise from δ_{ET} was much larger than the other two terms. Thus, quantifying and improving the precision of δ_{ET} should be the priority in future studies.

5. Conclusions

In this study, by employing the TDL technique, together with intensive sampling water in plant and soil pools, we made an attempt to partition ET via estimating δ_E, δ_T, and δ_{ET}. Our main conclusions are as follows: (1) The δ_T model developed in this study illustrated consistent estimations with the traditional model, making its potential to be used at other sites significant. (2) Fifteen centimeter is a reasonable depth for soil water sampling for estimating δ_E at this site. Sampling water at a too shallow depth may bring in biased δ_E estimation when soil moisture is very low. (3) The δ_{ET} has higher sensitivity to the partitioning and larger uncertainties in measurements than δ_T and δ_E. Quantifying and improving the precision of δ_{ET} measurement should be a priority in future endeavors of ET partitioning via stable isotopic approach.

Appendix A: Derivation of the Isotopic Composition of Plant Transpiration (δ_T) Proposed in This Study

Considering the changes of water storage in the leaf lamina (ΔW mol m^{-2} leaf) during a time interval (e.g., 3600 s) is the outcome of the accumulated flux input from the stems ($\sum J$ mol m^{-2} leaf) and that output by transpiration ($\sum T$ mol m^{-2} leaf):

$$\Delta W = \sum J - \sum T. \tag{A1}$$

Assuming no changes in isotopic composition of influx and outflux during the time interval, the change of the amount of the heavy isotope $\Delta(W\delta_L)$, is correspondingly linked by

$$\Delta(W\delta_L) = \delta_S \sum J - \delta_T \sum T, \tag{A2}$$

where δ_L is the isotopic ratio of the water in leaf lamina. Eliminating $\sum J$ from equation (A1) and equation (A2), we obtain

$$\Delta(W\delta_L) = \delta_S(\Delta W + \sum T) - \sum T\delta_T. \tag{A3}$$

As we know, the total T during the interval:

$$\mathrm{LAI}\sum T = f_T \sum \mathrm{ET} = \frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E}\sum \mathrm{ET}. \tag{A4}$$

Incorporating equation (A4) into equation (A3), we get

$$\begin{aligned}\Delta(W\delta_L) &= \delta_S\left(\Delta W + \frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E}\frac{\sum \mathrm{ET}}{\mathrm{LAI}}\right) - \frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E}\frac{\sum \mathrm{ET}}{\mathrm{LAI}}\delta_T \\ &= \delta_S \Delta W + \delta_S \frac{\sum \mathrm{ET}}{\mathrm{LAI}}\frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E} - \delta_T \frac{\sum \mathrm{ET}}{\mathrm{LAI}}\frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E}\end{aligned} \tag{A5}$$

and hence

$$\Delta(W\delta_L) - \delta_S \Delta W = \delta_S \frac{\sum \mathrm{ET}}{\mathrm{LAI}}\frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E} - \delta_T \frac{\sum \mathrm{ET}}{\mathrm{LAI}}\frac{\delta_{\mathrm{ET}} - \delta_E}{\delta_T - \delta_E} \tag{A6}$$

$$(\Delta(W\delta_L) - \delta_S \Delta W) \times (\delta_T - \delta_E)LAI = \delta_S \sum ET(\delta_{ET} - \delta_E) - \delta_T \sum ET(\delta_{ET} - \delta_E). \tag{A7}$$

For simplicity, let

$$Y = \Delta(W\delta_L) - \delta_S \Delta W \tag{A8}$$

and equation (A7) can be expressed as

$$Y(\delta_T - \delta_E)\mathrm{LAI} = \delta_S \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) - \delta_T \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E), \tag{A9}$$

i.e.,

$$Y\delta_T\mathrm{LAI} - Y\delta_E\mathrm{LAI} = \delta_S \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) - \delta_T \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E). \tag{A10}$$

Move the terms with and without δ_T in the different sides of the equation, we can get

$$Y\delta_T\mathrm{LAI} + \delta_T \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) = \delta_S \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) + Y\delta_E\mathrm{LAI}. \tag{A11}$$

The δ_T is arrived as

$$\delta_T = \frac{\delta_S \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) + Y\delta_E\mathrm{LAI}}{\sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) + Y \times \mathrm{LAI}}, \tag{A12}$$

i.e.,

$$\delta_T = \frac{\delta_S \sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) + \mathrm{LAI}\delta_E[\Delta(\delta_L W) - \delta_S \Delta W]}{\sum \mathrm{ET}(\delta_{\mathrm{ET}} - \delta_E) + \mathrm{LAI}[\Delta(\delta_L W) - \delta_S \Delta W]}. \tag{A13}$$

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