

# Study on Atmospheric and Terrestrial Water Circulation Processes Using Stable Water Isotopes

(水の安定同位体比情報を利用した大気と陸面の  
水循環過程の解明に関する研究)

WEI ZHONGWANG

魏 忠旺

Department of Natural Environmental studies

Graduate School of Frontier Sciences

The University of Tokyo

自然環境学専攻 新領域創成科学研究科 東京大学大学院

February 2016

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# Abstract

We observed stable isotopes in precipitation and atmospheric water vapor over a humid subtropical rice paddy field in Tsukuba, Japan, from June 2013 to May 2014. We used observed isotope ratios, in combination with an isotope-enabled general circulation model (GCM; Isotopes-incorporated Global Spectral Model: *IsoGSM*) to improve our understanding of the impacts of moisture sources and transport on the variability of water vapor isotopes. The isotopic measurements of water vapor and precipitation suggested that vapor isotopes in the study area were controlled by not only air-rain isotopic exchange, but also other kinetic effects associated with land evapotranspiration and large scale atmospheric circulation at the seasonal time scale. The contribution of land evapotranspiration to local water vapor content ( $F_{ET}$ ) was approximately  $16.0 \pm 12.3\%$  as an annual average, with a summer maximum of  $20.5 \pm 12.9\%$ . Our results show that large-scale atmospheric circulation is the primary control on the variability of near surface water vapor isotope. An *IsoGSM* tagging simulation experiment demonstrated that the large temporal variation of surface water vapor isotopes can primarily be attributed to advection and mixing of moisture from different oceanic source regions.

By using high-frequency isotope measurements and continuous surface water measurements, we investigated the isotope ratios in soil-vegetation-atmosphere transfer and the physical mechanisms involved over a paddy field for a full growing season. The isotopic signals ( $\delta$ ) of evapotranspiration ( $ET$ ) transpiration ( $T$ ) and evaporation ( $E$ ) were determined by the Keeling plot method, surface water isotopic measurements, and the Craig-Gordon model, respectively. The fraction of transpiration in evapotranspiration ( $T/ET$ ) ranged from 0.2 to 1, with an almost continuous increase in the early growing season and a relatively constant value close to 1 later in the year. The result was supported by  $T/ET$  derived from simulated  $T$  and eddy correlation measured  $ET$ . The seasonal change in the transpiration fraction could be described quite well as a function of the leaf area index ( $LAI$ ) ( $T/ET = 0.67LAI^{0.25}$ ,  $R^2 = 0.80$ ), implying that transpiration plays a dominant role in the soil-vegetation-atmosphere continuum during the growing season. The two end-member uncertainty analysis suggested that further improvement in the estimation of  $\delta_T$  and  $\delta_{ET}$  is necessary for partitioning evapotranspiration using the iso-

topic method. In the estimation of  $\delta_{ET}$ , the assumptions underlying Keeling plot method were rarely met and the uncertainty was quite large. A high frequency of precise isotopic measurements in surface water was also necessary for  $\delta_T$  estimation. Furthermore, special care must be taken concerning the kinetic fractionation parameter in the Craig and Gordon Equation for  $\delta_E$  estimation under low- $LAI$  conditions. The results demonstrated the robustness of using isotope measurements for partitioning evapotranspiration.

Recently isotopic method provides an unprecedented opportunity for  $ET$  partitioning across a variety of spatial and temporal scale which is unattainable by conventional technique. However, most of isotope-based studies show that  $T$  generally contributes more than 70% to the  $ET$ , while non-isotopic measurement generally leads to considerably smaller transpiration fractions. As a summary, quantitatively understanding of  $ET$  partitioning is somewhat arbitrary and remains the necessary of a theoretical and technical change, in both in-situ measurement and global scale. Considering the above tasks and background, we developed an  $ET$  partitioning algorithm, combining  $ET$  estimated by remote sensing, land surface models and  $LAI$  regression documented by 64 previous studies. The algorithm was used to understand spatial patterns and global average  $T/ET$ . Our global mean transpiration ratio (50%-60%) is smaller than the literature complication studies [Wang *et al.*, 2014; Schlesinger and Jasechko, 2014] but in good agreement with the value reported in GCMs (47%). On the other hand, it was significantly smaller than that reported in isotopic approaches [e.g., Jasechko *et al.*, 2013; Good *et al.*, 2015a]. Although further validation is required, our measured provided a new inspiration for partitioning global evapotranspiration and suggested vegetation plays a major role in driving the contribution of  $T/ET$ . Moreover, a further study about canopy interception is also required because canopy interception loss at various regions of the globe was few reported in the literature.

# Acknowledgements

I would like to thank to my supervisor Prof. Kei Yoshimura for his considerable support throughout my PHD studies. He has provided me with a lot of help, support and encouragement. Thanks to his patience, kindness, encouragement and good advice, I can finish writting my PHD disertation. Thanks also to Prof. Taikan Oki and Prof Kazuo Oki, who have contributed with his expertise during seminar and lecture. Thanks to Prof Wang, Prof.Yamanaka, Prof. Zhongfang Liu, Prof. Chun-Ta Lai, Prof. Toshihiko Sugai and Prof. Ryoichi Imasu for their valuable suggestion and encouragement for my research.

Thanks are also extended to my friends and colleagues in Oki Lab and Yoshimura Lab, Kim-Sensei, Noda-san, Utsumi-san, Yano-san, Nishijima-san, Nitta-san, Satoh-san, Okazaki-san, Yoshikane-san, Nomoto-san, Fukubayashi-san, Kanazawa-san, Kakehashi-san, Cherry-san, Mehwish-san, Okaneya-san, Saya-san, Ronald-san, Hatono-san, Takahashi-san, Suzuki-san, Hamada-san, Yoshida-san, Fukuda-san, Makino-san, Watanabe-san and so on. I appreciate them a lot not only for their help on my research, but also for their kindness to make me feel welcome. Special thanks are given to Okazaki-san. I am so glad to know you and I will never forget your continuous quick, friendly and good help. In addition, I really appreciate the guidance from Dr. Eun-Chul Chang and Dr. Suryun Ham, who are so patient to give me help to settle the problem regarding modeling studies.

I am very grateful to researchers in National Institute for Agro-Environmental Sciences, Tsukuba, Ibaraki, Japan. Kim-san and Ono-san, thank you provided us abundant high quality data. Thanks to Masaharu Yokoi from Sanyo Trading Co., Ltd., Tokyo, Japan. With their help in field work and care in many practical matters such as instruments, measurements and data analysis. I would like to thank my Chinese friends, Xiaogang He, Yaping Liu, Pei Wang, who always lends me courage and we fought together shoulder by shoulder. They make my daily life full of joy.

At last, thanks also to all my other friends who gave me unforgettable experience in my life. Thanks to my brother, without his help, I could not imagine how I can live through the last there years. At the same time, I would like to thank my parents. They gave me life, and spent countless love on me.

# List of symbols Variables

*CBL* Convective Boundary Layer, m

*CLM* Community Land Model

*d – excess* Deuterium excess , ‰

*E* Evaporation, mm day<sup>-1</sup>

*E<sub>c</sub>* Canopy interception, mm day<sup>-1</sup>

*E<sub>s</sub>* Soil evaporation, mm day<sup>-1</sup>

*ET* Evapotranspiration, mm day<sup>-1</sup>

*F<sub>ET</sub>* Fraction of evapotranspiration to near surface water vapor content

*F<sub>v</sub>* Vegetation Type %

*ISI – MIP1* Inter-sectoral impacts model inter-comparison project 1

*IsoGSM* Isotopes-incorporated Global Spectral Model

*GLEAM* Global Land surface Evaporation: the Amsterdam Methodology

*GRACE* NASA’s Gravity Recovery and Climate Experiment

*GPCC* Global Precipitation Climatology Centre

*GSWP – 2* Global Soil Wetness Project 2

*H* Sensible heat flux, W m<sup>-2</sup>

*IGBP* International Geosphere-Biosphere Programme

*JPL* Jet Propulsion Laboratory

*LAI* Leaf area index, m<sup>-2</sup> m<sup>2</sup>

*LE* Latent heat flux, W m<sup>-2</sup>

*MODIS* Moderate Resolution Imaging Spectroradiometer

|                  |   |
|------------------|---|
| $NSS$            | isotopic non-steady state   |
| $P$              | Precipitation amount  |
| $Q_v$            | water vapor mixing ratio at 2-m   |
| $Q_{bg}$         | background air vapor mixing ratio   |
| $R$              | Runoff, mm day <sup>-1</sup>  |
| $RH$             | Relative humidity   |
| $S_i$            | The evaporative stress and is computed combining observed vegetation water content and estimates of root-zone soil moisture from a multilayer soil module driven by observed precipitation and surface soil moisture. |
| $SSA$            | isotopic steady-state   |
| $STEAM$          | Simple Terrestrial Evaporation to Atmosphere Model  |
| $T$              | Transpiration, mm day <sup>-1</sup>   |
| $T_a$            | Air temperature   |
| $T_d$            | dew temperature   |
| $T/ET$           | Fraction of transpiration to evapotranspiration   |
| $TWSC$           | Terrestrial Water Storage Changes   |
| $vt$             | Vegetation type   |
| $\epsilon_K$     | The Kinetic fractionation factor  |
| $\alpha_{eq}$    | Temperature dependent equilibrium fractionation factor.   |
| $\sigma$         | Std of isotopic ratios  |
| $\beta$          | A constant to account for declines in transpiration when the canopy is wet.   |
| $\delta$         | Isotope composition of sample water relative to a standard, ‰   |
| $\delta D_{v,e}$ | $\delta D$ of precipitation predicted water vapor, ‰  |
| $\delta D_v$     | $\delta D$ of water vapor, ‰  |
| $\delta D$       | Stable isotope composition of hydrogen in water, ‰  |
| $\delta^{18}O$   | Stable isotope composition of oxygen in water, ‰  |
| $\delta_E$       | $\delta$ of soil evaporation flux, ‰  |
| $\delta_{ET}$    | $\delta$ of evapotranspiration flux, ‰  |

|               |                                   |
|---------------|-----------------------------------|
| $\delta_T$    | $\delta$ of transpiration flux, ‰ |
| $\delta_{sw}$ | $\delta$ of surface water, ‰      |
| $\delta_v$    | $\delta$ of water vapor, ‰        |
| $\delta_P$    | $\delta$ of precipitation, ‰      |
| $\delta_{bg}$ | $\delta$ of background air, ‰     |

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# Chapter 1

## Introduction

### 1.1 Stable isotopes approach: theory and measurement

The stable isotopologues  $^2\text{H}^1\text{H}^{16}\text{O}$  and  $^1\text{H}_2^{18}\text{O}$  are powerful tools to trace hydrological processes. Compare to the most abundant water  $^1\text{H}_2^{16}\text{O}$ , their slightly different masses affect their thermodynamic (e.g., differences in latent heat as a result of different hydrogen bonds between molecules) and kinetic properties (e.g., difference in the molecular diffusivity of water vapor when water evaporates into unsaturated vapor or when condensation occurs from supersaturated vapor) [Yoshimura, 2015; Rothfuss *et al.*, 2013; Jouzel and Merlivat, 1984]. As a result, differences in the rates of evaporation and condensation between heavy and light isotopologues of water is able to create spatial and temporal variability in the isotopic composition of water in the air and also on the land surface such as lakes, rivers, soils, and plants [Welp *et al.*, 2012]. These variability can help to address the origin and movement of atmospheric water [Craig, 1961; Dansgaard, 1964], partition water flux in terrestrial water cycle, reconstruct the past climate [Liu *et al.*, 2014] and understand groundwater dynamic [Liu and Yamanaka, 2012].

Since Dansgaard [1964], global isotopic datasets (such as Global Network of Isotopes in Precipitation (*GNIP*)) developed over the past 60 years have enabled continental-scale

assessments of transpiration/evaporation ratios, oceanic water vapor source tracing and the recycling of rainfall back into the atmosphere [Evaristo *et al.*, 2015]. Recently, Global Network of Isotopes in Rivers (*GNIR*) database contained around 21,000 stable water isotope records from 750 sites in 35 countries was developed for better understanding of the resilience of our major global river systems to environmental and human perturbations [Halder *et al.*, 2015]. However, few water vapor study was conducted because traditionally the water vapor have to be collected using cryogenic method, which is less practical and less accurate [Yoshimura, 2015]. Before ten years ago, mass spectrometry is often used for measuring the composition of stable water isotopes such as ice core and precipitation, or isotopes storage in speleothems, coral skeletons and tree cellulose. Recent development in high frequency laser spectrometer in-situ technology and remote sensing retrieval has made it possible to measure the vapor isotopic ratios at high temporal resolution and on a continuous basis [e.g., Lee *et al.*, 2006; Frankenberg *et al.*, 2013]. Such measurements offer unique opportunities to examine the temporal variation in water vapor isotopic ratios and gains insights into mechanisms involved. For example, it is possible to use in-situ stable isotope measurement to partition long-term evapotranspiration [Wei *et al.*, 2014, 2015]. It is also used to study vertical mixing process [Benetti *et al.*, 2014; Wei *et al.*, 2016] or rain-precipitation interaction [Laskar *et al.*, 2014]. Not long ago, the in-situ leaf water and continuous in-situ liquid water measurement were also developed (Picarro L2140-i and L2130-i Analyzer). These provide powerful tools towards a detailed quantitative understanding of air-precipitation exchange processes, ground water-underground water interaction and vegetation activity dynamic. Moreover, new remote sensing techniques allows us to measure water vapor isotope in global scale. For example, Using the Tropospheric Emission Spectrometer (*TES*), Worden *et al.* [2007] investigated evaporation and continental convection in the tropical water cycle. After that, several improved sensors (such as Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (*SCIAMACHY*) [Frankenberg *et al.*, 2009], Greenhouse gases Observing SATellite (*GOSAT*) [Frankenberg *et al.*, 2013; Risi *et al.*, 2013] and Infrared Atmospheric Sounding

Interferometer (*IASI*) [*Lacour et al.*, 2015]) were also used for to investigate aspects of the atmospheric hydrological cycle that are not well constrained by observations of precipitation or atmospheric vapor content. Stable isotope method has been become one of the most useful tools for quantifying water-cycle components and the linkages between plant ecology, physical hydrological and atmospheric dynamic.

## 1.2 Variability of near-surface water vapor isotopes

Water vapor isotopic measurement have been used in many studies to trace the origin and movement of atmospheric water [e.g., *Craig*, 1961; *Dansgaard*, 1964; *Strong et al.*, 2007; *Gat*, 1996; *Samuels-Crow et al.*, 2014a,b; *Lai and Ehleringer*, 2011; *Steen-Larsen et al.*, 2013, 2014a; *Tremoy et al.*, 2012, 2014; *Worden et al.*, 2007]. Most of these studies have focused on condensed phases (i.e., precipitation), with fewer studies considering the vapor phase. Despite a wide range of applications, water isotope in the condensed phase is too coarse to determine the dynamical and microphysical processes involved in atmospheric moisture evolution. Water vapor observation is not limited to rainy days, but can be done continuously over synoptic events and seasons [*Angert et al.*, 2008]. Water vapor isotopic composition is also a useful tool to investigate land vegetation activity and to understand vegetation-atmosphere exchange processes [e.g., *Good et al.*, 2014; *Wei et al.*, 2015].

Recently, water vapor isotopic measurements were shown to be useful for constraining state variable simulations in general circulation models (GCMs) [e.g., *Bonne et al.*, 2014; *Steen-Larsen et al.*, 2014b]. *Yoshimura et al.* [2014] conducted the first ideal observation system simulation experiment, through the combination of a local transform ensemble Kalman filter and an isotope-enabled GCM simulation. The authors showed that by incorporating a synthetic global dataset of Satellite-based water vapor isotope measurements and near surface water vapor isotopic measurements, model simulations were significantly improved in both the vapor isotopic and meteorological fields. Satellite-based water vapor isotope measurements have been used for validating GCMs and partitioning the global

hydrologic water flux [e.g., *Frankenberg et al.*, 2009; *Good et al.*, 2015b,a]. However, their inherent uncertainties are likely too large to quantify sub-grid scale hydrological processes.

Previous studies have identified the factors that control the isotopic signatures of water vapor over oceans [e.g., *Benetti et al.*, 2014, 2015; *Steen-Larsen et al.*, 2014a, 2015], ice sheets [*Steen-Larsen et al.*, 2013], plateaus [e.g., *Galewsky and Samuels-Crow*, 2014a,b; *Samuels-Crow et al.*, 2014a,b], and land surfaces under various climatic conditions [e.g., *Welp et al.*, 2012; *Berckelhammer et al.*, 2013; *Tremoy et al.*, 2012, 2014; *Lee et al.*, 2006; *Noone et al.*, 2013; *Wen et al.*, 2010; *Huang and Wen*, 2014; *Laskar et al.*, 2014]. Common findings of these studies are large variability of the observed atmospheric water vapor isotopic composition from daily to seasonal time scales, irrespective of geographical conditions. Such variation is related to specific hydrological processes. For example, during precipitation events, water vapor isotopes are significantly affected by air-precipitation isotopic exchange [e.g., *Huang and Wen*, 2014; *Lawrence et al.*, 2004; *Wen et al.*, 2010; *Tremoy et al.*, 2014]. Thus, some studies have suggested that it is possible to estimate the monthly water vapor isotope ratio from the precipitation isotope ratio [e.g., *Jacob and Sonntag*, 1991; *Lee et al.*, 2006; *Wen et al.*, 2010]. This method is fascinating because information regarding the liquid phase (such as surface water and precipitation isotope ratios) is readily available [*Lee et al.*, 2006]. A number of studies have concluded that land evapotranspiration play a important role on the isotopic variability of near surface water vapor in well-vegetated areas [e.g., *Welp et al.*, 2008; *Noone et al.*, 2013] or the sea surface [e.g., *Benetti et al.*, 2014].

Some studies have also confrimed the significance of other factors, such as the advection of large-scale moisture sources. *Strong et al.* [2007] used water vapor isotopes to diagnose moisture paths transported from three different oceans. They reported a low deuterium isotope ratio when moisture advection originated from the Gulf of Mexico (warmer than the Pacific Ocean) and a high ratio if water vapor is originated from the Pacific Ocean. By investigating the relationship between meteorological variables and water vapor isotope ratios, *Lee et al.* [2006], *Wen et al.* [2010] and *Huang and Wen*

[2014] suggested the dominant effect of Rayleigh distillation during large scale moisture transport.

The use of high frequency water vapor isotopes has a great potential to improve our understanding of the mechanisms that control stable isotope variability in hydrological processes. Despite these advances, there are several problems when using high frequency water vapor isotope measurements: 1) continuous measurements that cover multiple seasons are scarce [e.g., *Farlin et al.*, 2013; *Noone et al.*, 2013]; 2) very few study has yet been conducted at sites with changing vegetation cover over multiple seasons [*Bastrikov et al.*, 2014]; and 3) it is difficult to understand how large scale moisture transport controls the isotopic variability of water vapor observed at a single tower. Studies investigating the exact cause(s) of what controls long-term isotopic variability are rare.

### 1.3 Partitioning $ET$ using stable isotopes

To elucidate eco-hydrological processes and their underlying mechanisms, detailed knowledge of the relative contribution of different sources of water, such as soil evaporation and vegetation transpiration over land surfaces, is required. Although the total contribution of the land surface  $ET$  has been widely measured (e.g., *FLUXNET*), its partitioning into plant transpiration ( $T$ ) and surface evaporation ( $E$ ) is still subject to debate [e.g., *Coenders-Gerrits et al.*, 2014; *Jasechko et al.*, 2013; *Schlaepfer et al.*, 2014; *Schlesinger and Jasechko*, 2014; *Sutanto et al.*, 2014]. A quantitative relationship between  $ET$  partitioning and vegetation cover index (e.g.,  $LAI$ ) for different systems (i.e., agricultural versus natural systems) in global scale was established by *Wang et al.* [2014]. However, large variations in the fraction of transpiration in evapotranspiration ( $T/ET$ ) occurred across all  $LAI$  ranges with wider variability at lower  $LAI$ . It is still not clear to what extent that  $T/ET$  is controlled by vegetation and what are the additional factors that could further explain the  $T/ET$  variations at a global scale. The partitioning of ecosystem  $ET$  and its controlling factors are therefore crucial to better understand hydrological

processes at the landscape scale.

Various methods have been developed for *ET* partitioning which have been well documented in several review studies [e.g., *Kool et al.*, 2014; *Sutanto et al.*, 2014]. The most widely used among these methods is the combination of the eddy correlation method to measure *ET* with other measurements (e.g., lysimeters [*Agam et al.*, 2012], sapflow [*Williams et al.*, 2004], chambers [*Daikoku et al.*, 2008], or model simulations based on aerodynamic and biophysiological theories (e.g., the Penman Equation [*Penman*, 1948])). A complementary method of partitioning *ET* using isotopic biogeochemistry has been successfully applied in different vegetation types [e.g., *Dubbert et al.*, 2013, 2014a; *Good et al.*, 2014; *Moreira et al.*, 1997; *Rothfuss et al.*, 2010; *Wang and Yakir*, 2000; *Wang et al.*, 2010, 2013; *Williams et al.*, 2004; *Yopez et al.*, 2003] and at the global scale [e.g., *Coenders-Gerrits et al.*, 2014; *Jasechko et al.*, 2013; *Schlaepfer et al.*, 2014; *Schlesinger and Jasechko*, 2014]. To apply this method, detailed knowledge of isotopes in the different phases of water in soil, vegetation, and the atmosphere is required. Historically, because of the practical experimental difficulties in collecting and quantifying ground-based water vapor isotope samples, very few studies have reported successful implementation of the isotopic methodology [*Scanlon and Kustas*, 2012]. Recent development in high-frequency laser spectrometer in-situ technology has made it possible to measure water vapor isotopic ratios at a high temporal resolution and on a continuous basis [*Lee et al.*, 2006]. Such measurements provide a unique opportunity to examine temporal variation in water vapor isotopic ratios and to determine the mechanisms involved [*Lee et al.*, 2006]. The number of isotope-based studies therefore has been increasing dramatically in recent years [e.g., *Rothfuss et al.*, 2010; *Wang et al.*, 2012, 2013; *Dubbert et al.*, 2013, 2014b,a; *Good et al.*, 2014; *Hu et al.*, 2014].

Partitioning *ET* is specifically important for high water consuming agricultural products such as rice, which is widespread in the monsoon Asia. Although some other model-based studies have been conducted [e.g., *Kondo and Watanabe*, 1992; *Maruyama and Kuwagata*, 2010], no direct long term experiment has been reported in a rice paddy field

yet, mainly because of the experimental difficulties in the measurement of sapflow or leaf transpiration. The isotopic method offers an easy way to investigate the fraction of transpiration in evapotranspiration and its controlling factors in rice paddy fields. The long-term water vapor isotope-based partition technique has been used in a series of studies as mentioned above, however, no study has been performed over a wetland such as rice paddy. To the best of our knowledge, this study is the first report of long-term evapotranspiration partitioning using high-frequency continuous water vapor isotope observations over a paddy field. Due to significantly different surface condition (flooded) from that of other studies, the isotope characteristic of each composition in rice paddy field  $ET$  partitioning is not clear.

Although it has been addressed by many studies [*Braud et al.*, 2005, 2009a,b; *Dubbert et al.*, 2013; *Good et al.*, 2012; *Rothfuss et al.*, 2010, 2012], the uncertainties of the isotope-based method is still not well known. At the global scale, the isotope-based method shows a higher  $T/ET$  than the hydrometric method [*Sutanto et al.*, 2014]. To partition  $ET$  using the isotopic method, the isotopic signals of  $ET$ ,  $T$ , and  $E$  must be quantified first. However, estimation of these three terms has a degree of uncertainty [*Good et al.*, 2012; *Hu et al.*, 2014]. The isotopic composition of  $ET$  ( $\delta_{ET}$ ) is traditionally measured by a Keeling plot approach [*Keeling*, 1958], underlying which the assumptions used to calculate  $\delta_{ET}$ , as described in sect. 2.2.3, are rarely met perfectly in nature. On time scales from hours to days, much of the variation in the levels of water vapor isotopes  $\delta_v$  is caused by the advection of air masses in varying stages of rainout and has little to do with  $ET$ . Therefore, the intercept parameter in a Keeling plot depends on which part of the weather cycle is measured and not on  $ET$  [*Lee et al.*, 2006]. Usually,  $\delta_T$  is assumed to be equal to that of root uptake, maintaining a condition referred to as the isotopic steady-state assumption (*SSA*) [e.g., *Flanagan et al.*, 1991; *Yakir and Sternberg*, 2000]. However, many previous studies showed that the steady state is approached only at midday [e.g., *Lai et al.*, 2006; *Lee et al.*, 2007; *Peters and Yakir*, 2010], mainly because of the non-steady state (*NSS*) induced by changes in humidity and leaf energy balance [e.g.,

*Farquhar and Cernusak*, 2005; *Lai et al.*, 2006]. Furthermore, the isotopic composition of soil evaporation (surface water in our case) is commonly estimated from the Craig-Gordon model [*Craig and Gordon*, 1965]. However, the kinetic effect in the model depends on the relative roles of molecular and turbulent diffusion [e.g., *Dubbert et al.*, 2013; *Cappa et al.*, 2003; *Craig and Gordon*, 1965; *Gat*, 1996; *Kim and Lee*, 2011]. Recently, the methods coupling the laser spectrometer and the gas-exchange chamber have been used for directly estimating these three isotopic components [e.g., *Dubbert et al.*, 2013, 2014b,a; *Good et al.*, 2014; *Rothfuss et al.*, 2013; *Wang et al.*, 2012, 2013]. However, well-trained operators are needed and the installment and maintenance of the chamber are difficult.

## 1.4 Global *ET* estimation and its partitioning

Quantifying the global terrestrial water budget and energy cycles is essential for improving our understanding of the availability of water resources, the potential for hydrologic extremes, and the interactions of the land surface with the atmosphere and climate, and thus improving our predictions of large scale weather and climate, helping us make a robust water resource managements, estimating the risk of floods and droughts, and understanding of the land-air water exchange [e.g., *Sheffield et al.*, 2009; *Sahoo et al.*, 2011]. Therefore, it becomes one of main object of recent and ongoing projects such as the Coupled Model Intercomparison Project (*CMIP*), Global Energy and Water Experiment (*GEWEX*), Moderate Resolution Imaging Spectroradiometer project (*MODIS*), Global Earth Observing System of Systems (*GEOSS*). The terrestrial water budget can be defined as the balance between the change in water storage and the difference between the incoming water fluxes of precipitation and outgoing fluxes of evapotranspiration and discharge at the Earth's surface [*Sahoo et al.*, 2011]. Among these components, land evapotranspiration is the second largest component (after precipitation) of the terrestrial water flux and returns more than 60% of precipitation at global scale [*Mu et al.*, 2011]. *ET* uses up more than half of the total solar energy absorbed by the land surface

[Jasechko *et al.*, 2013]. Especially,  $ET$  typically accounts for over 90% of water losses in semi-arid and arid areas [Zhang *et al.*, 2001]. Accurate evapotranspiration estimates therefore are critical for understanding the mechanism of water cycling and evaluating the impacts of climate change, providing an important linkage between the land surface and the atmosphere.

To quantify global land surface evapotranspiration, many datasets including remote sensing-based estimations, reanalysis, land surface model driven with observations-based forcing [Miralles *et al.*, 2011a,b], vegetation coupled land surface model ([Lawrence *et al.*, 2011]), water balance based estimation [Oki and Kanae, 2006], have been derived in recent years. Although having being calibrated by local scale evapotranspiration measurements such as *FLUXNET* eddy correlation dataset (<http://fluxnet.ornl.gov>), the analyzed reference datasets displayed large uncertainty in spatial and temporal variability [Mueller *et al.*, 2011, 2013]. These datasets are somewhat arbitrary, because they are all based on some degree of modeling assumption but not the direct measurement. Although some datasets showed similar results, they may share common biases related to common calibration or forcing dataset, and/or common model assumption. These leave an open question about whether it could be verified by independent methods or experiments.

It is more important that the understanding of mechanism of  $T/ET$  has been a fundamental challenge since more over 70 years ago. The contribution of  $T$  to  $ET$  is a prerequisite for understanding changes in carbon assimilation and water cycling in a changing environment. On local scale, evapotranspiration partitioning can be achieved by a combination of in-situ measurements, such as eddy covariance systems, Bowen-ratio systems, weighing lysimeter, sap flow meters, leaf conductance upscaling, chamber measurement and isotopic measurements. On global scale, dozens of datasets including satellite-based estimations, reanalysis, land surface model driven by observations-based forcing, upscaling of point observation and water balance based estimation can be used for quantifying the contribution of each component in evapotranspiration (Table 1.1). For example, by combining a wide range of remotely-sensed observations, Miralles *et al.*

[2011b] found 80% annual land evapotranspiration corresponds to transpiration. Recently, isotope approach has been used for partitioning  $ET$  in both global and local scale. Based on isotopic mass and water balance, some quantities difficult to be measured such as transpiration can be easily obtained. Basing on isotopic analysis of a global data set of large lakes and rivers, *Jasechko et al.* [2013] used the lake isotopic information to show that transpiration represents 80 to 90% of the terrestrial evapotranspiration. *Good et al.* [2015b] suggested the transpired fraction of evapotranspiration is estimated a mean of 64% based on satellite isotopic measurements, which is much lower than previous studies. Although these results suggested that plant transpiration is a majority of terrestrial water fluxes, the uncertainties were significant. Potential uncertainties derives from:

1. The concept of total evapotranspiration sometimes is confusing. In general, the total evapotranspiration is the combination of soil evaporation ( $E_s$ ), canopy transpiration ( $T$ ) and canopy interception evaporation ( $E_c$ ). However, due to measurement difficulty,  $E_c$  was rarely measured in field based partitioning studies. Thus in a lot of field studies, the total evapotranspiration was the combination of  $E_s$  and  $T$ . On the other hand, except very few study (e.g. *Yang et al.* [2015]),  $E_c$  is generally included in global or regional studies. Due to lack of observation in  $ET$  partitioning studies,  $E_c$  remains large uncertainty in global simulation.
2. Quantification of contribution of transpiration in large scale have long been associated with large uncertainties due to no reliable, benchmarking or good-quality forcing data sets for the evaluating the various products. The local scale  $ET$  partitioning techniques remain difficult to be implemented at large scale (such as regional or continental scale). Some studies directly compared several site measurement results with global simulation [*Wang et al.*, 2014; *Schlesinger and Jasechko*, 2014]. However, field based observation is still rare and only for point scale (also see Table 1.1). Global coverage by such point measurements is not feasible and the representativeness of such local-scale observation remains challenging. *Wang et al.* [2014] reviewed the published studies (48 individual publications) that measured at least

two of the three components ( $E$ ,  $T$ , and  $ET$ ) and  $LAI$  simultaneously in an attempt to establish a quantitative relationship between  $ET$  partitioning and  $LAI$  and extend the in-situ observation to global scale. However, the relationship between  $T/ET$  and  $LAI$  was not well constrained in his study.

3. The different methods generally yield different results. Isotopic method provides an unprecedented opportunity for  $ET$  partitioning across a variety of spatial and temporal scale which is unattainable by conventional technique. However, as cautioned by several studies, isotope-based  $T/ET$  partitioning studies show that  $T$  generally contributes higher than 70% to the  $ET$ . On the other hand, non-isotopic measurement generally leads to considerably smaller transpiration fractions. Thus they argued that the isotopic measurement may overestimate the contribution of  $T$ . Moreover, global isotopic partitioning is sensitive to bulk flux estimates (such as precipitation and interception amount).

As a summary, quantitative understanding of  $ET$  and its partitioning are somewhat arbitrary and remains the necessary of a theoretical and technical change, in both in-situ measurement and global scale.

## 1.5 Objective and outline of this dissertation

The research presented in this dissertation focuses on the development and application of a stable isotope method to investigate the key factors affecting the water cycling including raindrops-air isotopic interaction, land-atmosphere water exchange and large scale moisture circulation. Specifically, based on our observation, we develop a new approach to quantitatively understanding of  $ET$  and its partitioning in global scale. The dissertation is organized into papers destined for publication in peer-reviewed journals and paper in preparation [Wei *et al.*, 2014, 2015, 2016; Wei and Yoshimura, 2016]. The flow chart of this study is presented in Figure 1.1. The introduction and methods are presented in Chapter 1 and 2. Each chapter from 3-5 within the main body is organized to investigate

a specific objective as:

1. In Chapter 3, we conduct a long-term water vapor isotopic study that uses in-situ isotopes and meteorological measurements along with an isotope-enabled GCM, to explore the impact of oceanic water sources on water vapor isotopic variability from daily to seasonal time scales over a rice paddy field in Tsukuba, Japan. We use isotopic information to partition water vapor content into contributions from local and remote-source components. Tagging experiments using the Isotopes-incorporated Global Spectral Model (*IsoGSM*) are conducted to understand the role of large scale moisture transport in the observed isotopic variability.
2. In Chapter 4, we use high-frequency water vapor isotope measurements and surface water measurements to investigate isotopic variation in soil-vegetation-atmosphere transfer and to determine the physical mechanisms involved in a paddy field throughout a full growing season, which is the first observation-based long term  $ET$  partitioning study over wetland surface. The main objectives are: 1) to identify the temporal characteristics of isotopic variation in soil, vegetation and evapotranspiration over the paddy field; 2) to partition evapotranspiration into evaporation and transpiration and to investigate its relation with the vegetation growth; and 3) to explore the potential uncertainties in  $\delta_{ET}$ ,  $\delta_T$ ,  $\delta_E$  and their impact on  $ET$  partitioning. We examine the capability of the Keeling plot method to estimate the isotopic signals of the  $ET$  flux and the steady state assumption for transpiration. We also estimate  $T/ET$  and compare it to water flux based partitioning results.
3. In Chapter 5, by combining our isotopic  $T/ET$  observation and the published observation based  $T/ET$  studies (64 individual publications), we investigated global synthesis of  $LAI$  control on  $ET$  partitioning.  $T/ET$  is estimated as different functions of  $LAI$  with different vegetation types. We estimated the  $ET$  from remote sensing (*GLEAM*), land surface model (*CLM4.5SP*) and water balance approach based on *GRACE* satellite. We develop a new approach to quantitative estimation

of  $T$  in global scale and further compare the global  $T/ET$  results with previous studies.

The conclusion and outlook in the papers are appended to this dissertation (Chapter 6) as a summary of the methods we used, the most significant results, the limitation and its future orientation in each papers.

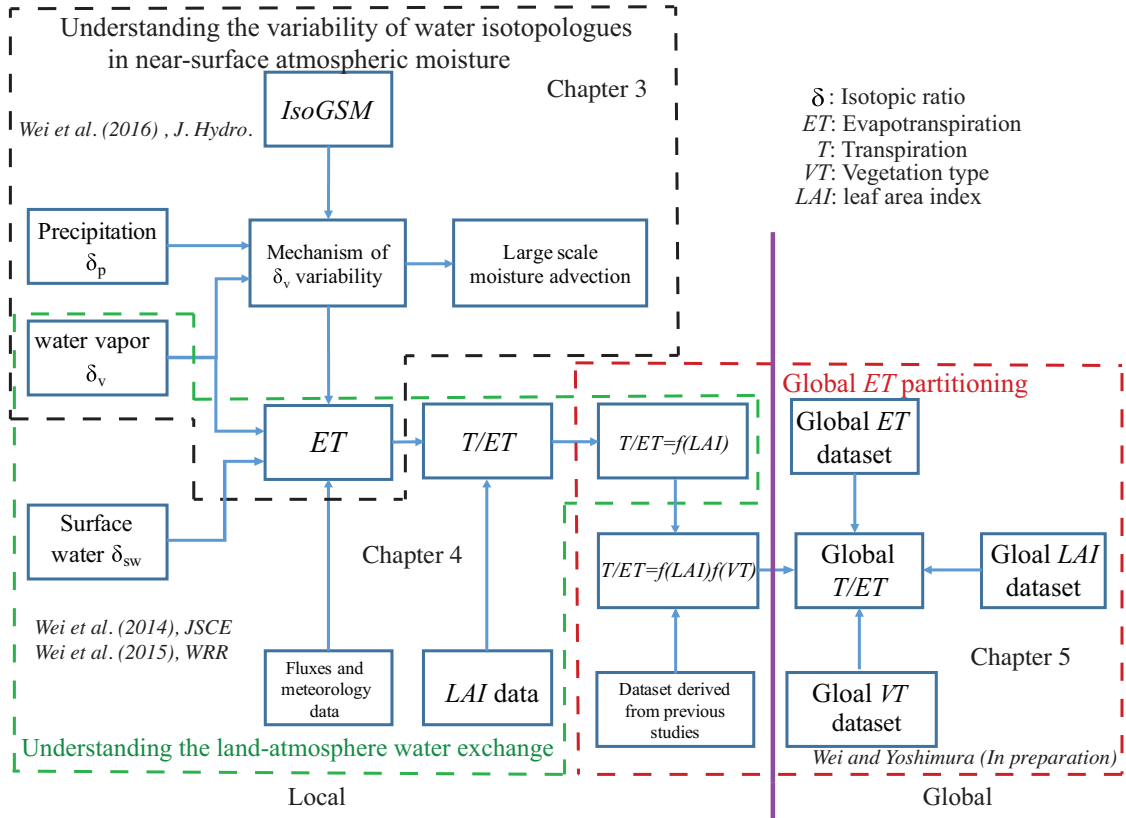


Figure 1.1: Objective and outline of this dissertation.

Table 1.1: Previous studies of *ET* partitioning.

| Methods                           | $E_c/ET$         | $T/ET$ | $T/(ET - E_c)$ | Scale         | Source                                 |
|-----------------------------------|------------------|--------|----------------|---------------|--|
| <b><i>Land surface models</i></b> |                  |        |                |               |  |
| <i>GSWP - 2</i>                   | 16%              | 48%    | 57%            | Global        | <i>Dirmeyer et al.</i> [2006]          |
| <i>ISI - MIP1</i>                 | 27%              | 36%    | 49%            | Global        | Calculated from ISI-MIP1 models        |
| <i>STEAM</i>                      | 21%              | 59%    | 74%            | Global        | <i>Wang-Erlandsson et al.</i> [2014]   |
| <i>IsoMatsiro</i>                 | 36%              | 31%    | 49%            | Global        | <i>Yoshimura et al.</i> [2006]         |
| <i>LCM3</i>                       | 17%              | 41%    | 49%            | Global        | <i>Lawrence et al.</i> [2007]          |
| <i>LCM3.5</i>                     | 18%              | 43%    | 52%            | Global        | <i>Lawrence et al.</i> [2011]          |
| <i>LCM4CNb</i>                    | 21%              | 56%    | 70%            | Global        | <i>Lawrence et al.</i> [2011]          |
| <i>LCM4SP</i>                     | 20%              | 48%    | 60%            | Global        | <i>Lawrence et al.</i> [2011]          |
| <i>LCM4CNEc</i>                   | 22%              | 56%    | 71%            | Global        | <i>Lawrence et al.</i> [2011]          |
| <b><i>Isotopes</i></b>            |                  |        |                |               |  |
| Lakes isotope                     | 10% <sup>d</sup> | 85%    | 94%            | Catchment     | <i>Jasechko et al.</i> [2013]          |
| Lakes isotope                     | 29% <sup>e</sup> | 67%    | 94%            | Catchment     | <i>Coenders-Gerrits et al.</i> [2014]  |
| Global isotope budget             | 27% <sup>e</sup> | 64%    | 88%            | Global        | <i>Good et al.</i> [2015b]             |
| <b><i>Satellite</i></b>           |                  |        |                |               |  |
| <i>GLEAM</i>                      | 11%              | 80%    | 89%            | Global        | <i>Miralles et al.</i> [2011b]         |
| <b><i>Site measurements</i></b>   |                  |        |                |               |  |
| Isotope+hydrometric+model         |                  | 61%    |                | Site averaged | <i>Schlesinger and Jasechko</i> [2014] |
| Isotope+hydrometric               |                  | 60%    |                | Site averaged | <i>Wang et al.</i> [2014]              |
| <b><i>Other</i></b>               |                  |        |                |               |  |
| <i>LAI</i> regression             |                  | 55%    | 70%            | Global        | This study                             |

# Chapter 2

## Methods

### 2.1 Site description

The study site ( $36^{\circ} 03' 14.3''$  N,  $140^{\circ} 01' 36.9''$  E, a.s.l. 12 m) was located in a rectangular 100 m $\times$ 50 m cultivated plot near the center of a 150-ha rice paddy field in Tsukuba (Figure 2.1). The location of the measuring instrument provided a sufficient fetch of uniform land cover for flux and isotope measurements. Cultivation activity influenced the land cover in different seasons. The field was sparsely covered by weeds in spring (March–April), and by a canopy of rice seedlings from May until harvesting at the end of August. The field was flooded during the growing season and allowed to drain in the rest of the year. The maximum canopy height was about 1.1 m and the maximum leaf area index was  $6.2\pm 1.4$ . The site has participated in AsiaFlux activities since 1999. Detailed information regarding meteorological and flux measurements can be found on the AsiaFlux website ([http://asiaflux.net/index.php?page\\_id=83](http://asiaflux.net/index.php?page_id=83)).

During our study period, irrigation started on April 23, 2013 and April 24, 2014. Rice was sown on May 2, in both 2013 and 2014, at a density of about 16 hills/m<sup>2</sup> (each hill contains a group of transplanted rice). The crop was harvested at the end of each August. Harvest residuals remained on site, and ratoon (re-sprouting young rice plants) continued to grow. Field management of the study plot in 2013 is summarized in Figure

2.2. During the full growing season, the field was flooded to a mean water depth of 3.0 cm [Ono *et al.*, 2013]. Periodic measurements of the  $LAI$  and vegetation height  $z_v$  undertaken in 2013 are plotted in Figure 2.2. Our study area is located near the western Pacific Ocean, which is under the influence of a tropical to subtropical climate, with a monsoon period in both summer and winter. In different seasons, this region potentially receives moisture from different oceans. Furthermore, our study was conducted in a large paddy field. Cultivation and irrigation activities influenced the surface cover; thus, we expected significant differences in the evapotranspiration isotopic pattern among seasons.

## 2.2 In-situ observation

The study site is part of the AsiaFlux Network. As Figure 2.3 showed, data in this site is available from (i) regular meteorology measurements, (ii) eddy covariance measurements, and (iii) isotopic measurements (also see photos in Figure 2.1). For (i) and (ii), data have been collected since 1999 by the National Institute of Agro-Environmental Sciences (*NIAES*). For (iii), new measurements began in June 2013, a month after the rice was planted. (i) and (ii) are briefly summarized here with a detailed description given in *Saito et al.* [2005]. The eddy covariance instruments, consisting of an open path infrared gas analyzer (LI-7500, Li-cor) and a sonic anemometer thermometer (DA-600, Kaijo), were mounted at 3.0 m above the ground. Air temperature ( $T_a$ ) and relative humidity ( $RH$ ) were measured at two heights, 3.8 and 1.1 m, with the temperature-humidity sensors (HMP-45A, Vaisala Helsinki, Finland). For (iii), water vapor and precipitation data measured from June 2013 were collected. All isotope measurements are presented as the ratio of heavy to light isotopologues and are relative to the normalized delta notation in per mil (‰),

$$\delta = \left( \frac{R_{sample}}{R_{standard}} - 1 \right) \times 1000 \quad (2.1)$$

where  $R_{sample}$  and  $R_{standard}$  are the oxygen mass 18 to mass 16 ( $^{18}\text{O}/^{16}\text{O}$ ) or deuterium to hydrogen ( $D/H$ ) ratios of a sample and the Vienna standard mean ocean water ( $V - SMOW$ ), respectively.  $d - excess$ , defined as  $\delta D - 8\delta^{18}\text{O}$  [Dansgaard, 1964], was also calculated to provide further insight into the kinetic effects associated with evaporation processes.

### 2.2.1 Isotopic measurement system

The 1 Hz water vapor isotopic measurement system consisted of a Cavity Ring-down Spectrometer (L2120-i), Standard Delivery Module (A0101), High-Precision Vaporizer (A0211) and 16-port Distribution Manifold (A0311) (Picarro, Sunnyvale, CA, USA). Measurements were made every 2 seconds and the results were averaged into 1-minute. Air sampling was undertaken using a distribution manifold. A shielded air intake was installed at a height of 2.0 m and was connected to the distribution manifold with a 14-m, 1/8-inch Teflon inlet tube (Du Pont-Mitsui Fluorochemicals, Tokyo, Japan) at a rate of about  $40 \text{ mL min}^{-1}$ . The flushing time was about 2 min. Because the observed  $Ta$  was higher than the dew point temperature (Figure 2.4), it is not necessary to heat the tubing.

Precipitation samples were collected every six hours using an Eigenbrodt NSA181 rain sampler, which is a wet-only automatic sampling system. The closure system is designed to prevent evaporation of the sample water or potential exchange with the atmosphere. The equipment guarantees automatic all year operation, possible through the use of heating at temperatures of  $0^\circ\text{C}$  and below. This collector contains a permanent cooling and sample-changing system, allow samples to be changed every 6-hours. Daily and monthly averages of  $\delta D$  and  $\delta^{18}\text{O}$  were weighted by the precipitation amount and calculated as follows:

$$\delta_p = \frac{\sum_{k=1}^n \delta_{p,k} \times P_k}{\sum_{k=1}^n P_k} \quad (2.2)$$

where  $n$  is the number of observations for each day, and  $\delta_{p,k}$  and  $P_k$  are the  $\delta$  value and

the total amount of the  $k$ th precipitation, respectively. Assuming that water vapor and precipitation are in isotopic dynamic equilibrium, the equilibrated water vapor isotopic ratio  $\delta_{v,e}$  can be calculated from  $\delta_p$  by incorporating a temperature-dependent equilibrium fractionation factor  $\alpha_{eq}$  [Majoube, 1971]:

$$\delta_{v,e} = \left( \frac{1 + \delta_p/1000}{\alpha_{eq}} \right) \times 1000 \quad (2.3)$$

Water vapor isotopic measurements are briefly summarized here and a detailed measurement information was conducted in Wei *et al.* [2015]. Unfortunately, there was a data gap from 22 October to 23 December 2013, due to a pump failure. Surface water was sampled 1.5 days per week in the growing season of 2013 and increased to 3 days per week on average for 2014. Seven sampling locations were chosen to provide a general distribution of surface water isotopes.

### 2.2.2 Calibration protocol

The collected liquid water samples were transferred into high-density glass storage bottles and stored in a refrigerator at 15 °C in the laboratory until analysis. The laboratory room temperature was kept at 25 °C to prevent condensation during the analysis process. The samplers were analyzed using Cavity Ring-down Spectrometer (L2120-i). A new technique proposed by van Geldern and Barth [2012] was performed as a correction procedure for liquid water isotopic measurement. This technique required modifications to the hardware for liquid water injection, an optimized sequence layout, and a simple post-run correction procedure to remove memory effects. Three standard waters which were referenced against Vienna standard (i.e., KW: -0.3‰, -1.2‰; AW: -26.0‰, 197.9‰; SW: -9.6‰, -64.8‰ for  $\delta^{18}O$  and  $\delta D$ , respectively) were included in each analysis batch to correct raw data for memory effects caused by the influence of the previous sample. The correction results showed that the final mean values agreed very well with the true values (Figure 2.5a). To check the correction performance, we also measured the same

sample several times (after each 8 unknown samples) in one measurement event (Figure 2.6). The carry-over from the different previous samples were virtually removed. Our method reduced the number of injections per sample to four (compared to six in the manufacturer’s recommended method) with similar precision and accuracy (std  $\pm 0.2\text{‰}$  for  $\delta^{18}\text{O}$  and  $\pm 0.5\text{‰}$  for  $\delta D$ ). After memory correction, four injections of each sample were averaged and normalized to the Vienna Standard Mean Ocean Water-Standard Light Antarctic Precipitation (*VSMOW* – *SLAP*) scale using a two-point linear calibration generated from reference waters.

The air samples were analyzed with a cavity ring-down spectrometer and were carefully calibrated with a standard delivery module. The calibration was performed using a self-calibration program provided by Picarro CO. LTD. Two in-house water standards (SW and AW) were vaporized by the standard delivery module, mixed with dry room air and transferred to the analyzer. Each standard sample was measured continuously for 10 min with a dry air cleanup of about 15 min every 9 hours during the whole measurement period. The results showed a high precision with std  $\pm 0.3\text{‰}$  for  $\delta^{18}\text{O}$  and  $\pm 1.3\text{‰}$  for  $\delta D$ . The standard water was changed every 6 months. We also evaluated the concentration dependence of our instrument by injecting different amounts of standard water into the vaporizer. We did not correct the concentration dependence effect because the measurements generally worked well within the range of water vapor concentration encountered during our study period (Figure 2.5b). However, the large measurement uncertainty under low water vapor concentration suggested that the correction of the raw measurements using the humidity-isotope response functions [*Steen-Larsen et al.*, 2014a] might be required. Unfortunately, there was a data gap from 22 October to 23 December 2013, due to a pump failure.

### 2.2.3 $\delta_E$ , $\delta_T$ and $\delta_{ET}$ estimation

Considering water vapor as a mixture of *ET* (including *E* and *T*) from local surfaces and that in the background air, water vapor isotopic variations can be influenced by isotopic

variations in either of the components [Yakir and Sternberg, 2000]. Based on water and isotope mass balance, the isotopic composition of water can be directly used to partition  $ET$ . It should be noted that only  $\delta D$ -based results are presented here, as variation in  $\delta D$  largely mimics that in  $\delta^{18}O$ .

The Keeling plot method, in combination with water vapor isotope ratios, has been successfully used to investigate the variation in  $\delta_{ET}$  [e.g., Moreira *et al.*, 1997; Wang and Yakir, 2000; Yopez *et al.*, 2003; Williams *et al.*, 2004; Lee *et al.*, 2006; Good *et al.*, 2012, 2014]. Based on the mass conservation of water vapor and isotopes, the isotopic composition of water vapor can be determined by

$$\delta_v = a \times \frac{1}{Q_v} + \delta_{ET} \quad (2.4)$$

where  $a = (\delta_{bg} - \delta_{ET}) Q_{bg}$ ,  $Q$  is the water vapor mixing ratio. The subscripts  $bg$  and  $v$  represent the background atmosphere and atmospheric water vapor at the measurement height, respectively. Our Keeling plot was based on 1-hr  $Q_v$  and  $\delta_v$  measurement. Two assumptions must be satisfied [e.g., Lee *et al.*, 2006; Wen *et al.*, 2010] here: (i)  $\delta_{bg}$ ,  $Q_{bg}$  and  $\delta_{ET}$  are constant over the observation period, which means that  $a$  should be constant; (ii) temporal variations in  $\delta_v$  (in our case, water vapor  $\delta D$ ) are caused by evapotranspiration only. An example is given in Figure 2.7.

Several experiments have been conducted in controlled experiments [e.g., Cappa *et al.*, 2003; Kim and Lee, 2011; Rothfuss *et al.*, 2010] or over bare soil [e.g., Dubbert *et al.*, 2013, 2014a] to understand isotopic compositions of soil evaporation, although it is still rare because of the technology limitations. Generally, the Craig and Gordon Equation [Craig and Gordon, 1965] is used to quantify the evaporative enrichment

$$\delta_E = \frac{\alpha_{eq}^{-1} \delta_w - h \delta_v - \varepsilon_{eq} - (1 - h) \varepsilon_K}{(1 - h) + 10^{-3} (1 - h) \varepsilon_K} \quad (2.5)$$

where  $\alpha_{eq} = (24840/T^2 - 76.25/T + 0.0526)$  is the equilibrium fractionation factor [Majoube, 1971], and  $\varepsilon_{eq} = (1 - 1/\alpha_{eq}) \times 1000$ ,  $\delta_w$  is the isotopic composition of surface water,

$h$  is  $RH$  normalized by the saturation vapor pressure at the water surface. The Kinetic fractionation factor  $\varepsilon_K$  is defined by

$$\varepsilon_K = n(1 - D_i/D) \times 10^3 \quad (2.6)$$

where the ratio of the molecular diffusion coefficients of water vapor in dry air  $D_i/D$  is taken as 0.9839 from *Cappa et al.* [2003];  $n = 0.67$  is taken from *Kim and Lee* [2011] and is used to understand the isotopic enrichment of liquid water during evaporation from a water surface.

The chamber-based direct measurements of transpired water vapor isotopic composition ( $\delta_T$ ) have been developed recently [e.g., *Wang et al.*, 2010, 2012, 2013; *Dubbert et al.*, 2014a]. Such chamber measurement is a challenge for our study. It is difficult to ensure good air tightness of air chamber because of hollow stems of rice. Moreover, it requires changing the size of chambers frequently since rice is a fast growing crop. The flooded condition also increases the difficulty of the installation and maintainance of chamber. Practically,  $\delta_T$  can be assumed the same as pond water  $\delta_{sw}$ , although some bias might be produced by non-steady state isotopic enrichment especially during early morning and late afternoon [e.g., *Dongmann et al.*, 1974; *Lai et al.*, 2006, 2008; *Dubbert et al.*, 2014a,b]. Based on the water balance ( $ET = T + E$ ) and isotope mass balance ( $ET\delta_{ET} = T\delta_T + E\delta_E$ ), without considering rainfall interception evaporation,  $T/ET$  is then estimated as

$$\frac{T}{ET} = \frac{\delta_{ET} - \delta_E}{\delta_T - \delta_E} \quad (2.7)$$

The key point in this study is to precisely determine the isotope ratio of  $ET$  ( $\delta_{ET}$ ),  $T$  ( $\delta_T$ ), and  $E$  ( $\delta_E$ ) using isotopic measurements. To identify the principal uncertainties, we quantify the  $T/ET$  uncertainties through a first-order Taylor series expansion of the uncertainty in the final partitioned value [*Good et al.*, 2014; *Phillips and Gregg*, 2001]

$$\sigma_{T/ET}^2 = \frac{\sigma_{\delta_{ET}}^2}{(\delta_T - \delta_E)^2} + T/ET \frac{\sigma_{\delta_T}^2}{(\delta_T - \delta_E)^2} + (1 - T/ET) \frac{\sigma_{\delta_E}^2}{(\delta_T - \delta_E)^2} \quad (2.8)$$

The three terms on the right-hand side of the equation represent the contribution of uncertainties in  $\delta_{ET}$ ,  $\delta_T$ , and  $\delta_E$  to the total variance.

## 2.3 *IsoGSM* simulation and water tagging experiments

### 2.3.1 *IsoGSM* simulation

To investigate the impact of large-scale moisture transfer on the isotopic variability of near surface water vapor, we used *IsoGSM*, a global spectral model that incorporates water isotope tracers [Yoshimura and Kanamitsu, 2009]. *IsoGSM* simulations have been validated by ground-based isotope observation [e.g., Farlin et al., 2013; Uemura et al., 2008] and satellite observation [e.g., Frankenberg et al., 2009]. These studies showed that *IsoGSM* is capable of reproducing the daily averaged variability in  $\delta D$  of near-surface water vapor associated with synoptic weather cycles. The model in this study had a horizontal scale of about 200 km (T62) and 28 vertical levels (L28), with six-hour intervals. *IsoGSM* was nudged by the spectral nudging technique [Yoshimura et al., 2008] to include the actual atmospheric thermodynamic situation. The default land surface scheme in *IsoGSM* is the Noah model [Ek et al., 2003]. In *IsoGSM*, isotopic fractionation takes place whenever phase transition occurs. *IsoGSM* assumes a constant isotopic value (0‰) for the ocean water and no isotopic fractionation occurred in land surface evapotranspiration (i.e., 100%  $T/ET$  is assumed). Readers are directed to Yoshimura et al. [2008], Yoshimura et al. [2014] and He et al. [2014] for a detailed description of the model setup. Over a long time period (such as monthly time scale), the planetary boundary layer can be effectively assumed as a one dimension model, with input (evapotranspiration) and output (air from the free atmosphere) only from the top and bottom. Near surface hor-

horizontal advection impact is small and can be cancelled out by the temporal averaging [Helliker *et al.*, 2004; Angert *et al.*, 2008]. Therefore, the water vapor source over this site was considered to originate from: 1) air from the background atmosphere (convective boundary layer (hereafter *CBL*) water vapor, represented by *IsoGSM* simulated water vapor and its associated  $\delta D_v$  output ( $\delta_{bg}$ ) at *CBL* height); and 2) water vapor supplied from local land evapotranspiration (estimated by a Keeling plot). The relative contribution of evapotranspired water to surface water vapor was evaluated as follows [e.g., Yakir and Sternberg, 2000]:

$$F_{ET} = \frac{\delta_v - \delta_{bg}}{\delta_{ET} - \delta_{bg}} \quad (2.9)$$

where  $F_{ET}$  is the contribution of *ET* to the atmospheric boundary layer (*ABL*) water vapor. We further quantified uncertainties through a first-order Taylor series expansion of all the uncertainty in the final partitioned value [Phillips and Gregg, 2001].

### 2.3.2 Water-tagging experiments using *IsoGSM*

In addition to local evapotranspiration, water vapor isotopes are also affected by original oceanic sources. To understand the origin of near-surface water vapor and its impact on the isotopic variability, we conducted a tagging simulation using *IsoGSM*. The mechanism of our water-tagging experiments was very similar to that of Yoshimura *et al.* [2004], Frankenberg *et al.* [2009] and Risi *et al.* [2010]. By running *IsoGSM* in tracer mode, it is possible to track a given moisture mass back to its last time at sea level, within a certain time span. This was achieved by setting the surface evaporative fractionation factor to 1 for the target region and 0 for the other regions in the model, and by turning off all the other isotopic fractionation in the atmosphere. For each of these simulation, the evaporated water is marked by its origin with a tag. Once this tagged vapor is added to the atmosphere, it undergoes exactly the same processes as normal vapor (It is allowed to advect, mix, condense and rainout in the atmosphere. Therefore, it has influence neither on the hydrological cycle nor on isotopic processes.), except it is not factored into

energy budget calculations. The method used here allows a direct quantification of the amount of water advected from tagging region. Similar experiments were conducted to understand the cause of the seasonal precipitation isotopic variation along the western U.S. coast [Buening *et al.*, 2012]. In our study, vapor was tagged within a  $10^\circ \times 10^\circ$  boxed sub-region. The atmospheric column perceptible water derived from tagged boxes was used to identify the source region of the water vapor.

## 2.4 Global $ET$ estimation and its partitioning

### 2.4.1 Global synthesis of $LAI$ control on dry canopy $ET$ partitioning

In order to understand global synthesis of  $LAI$  control on  $ET$  partitioning, we collected article searches in Google scholar and Web of Science and evaluate the relationship between  $LAI$  and  $T/ET$ . To select datasets of high quality, the following data screening criteria were used: (1) under dry canopy condition or soil evaporation is known. (2) at least two out of the three members ( $E$ ,  $T$ , and  $ET$  for non-isotopic methods or  $\delta_E$ ,  $\delta_T$  and  $\delta_{ET}$  for isotopic method) were measured independently; (3)  $LAI$  was quantified during the measurement period; and (4) Vegetation types were recorded.

As a result, 64 individual publications (Among these studies, 31 individual publications shown in Wang *et al.* [2014] were adopted.) before 30 November 2015 were included in our analysis (Appendix A). Similar with Wang *et al.* [2014], the end members in  $ET$  partitioning and  $LAI$  values directly from tables or text in original papers are summarized. Some indirect data from figures was extracted using GraphClick software (Arizona software, USA). Based on vegetation types, we classified all the studies into 6 species: Broad leaf forests, Needle leaf forests, Mixed forests, Shrubs and grasses, crops and wetlands. To reduce systematic error, the data were averaged into 0.5-bin average of  $LAI$ . The relationship between  $LAI$  and  $T/ET$  for each piece of vegetation was investigated using power regression ( $T/ET = aLAI^b$ , where  $a$  and  $b$  depend on vegetation

type).

### 2.4.2 *ET estimated from LSM, RS and GRACE*

Three different *ET* products were used in this study (Table 2.1), i.e., (1) land surface modelling (*LSM*) estimated *ET* (Community Land Model 4.5, *Lawrence et al.* [2011]), (2) remote sensing (*RS*) based *ET* (Global Land surface Evaporation: the Amsterdam Methodology (*GLEAM*), *Miralles et al.* [2011a,b]) and (3) *GRACE*-inferred *ET*. *GRACE* satellite-derived total water storage change (*TWSC*) can be used to estimate *ET* as a residual in the water balance equation:

$$ET = P - R - TWSC \quad (2.10)$$

where *P* is precipitation and *R* is runoff [*Ramillien et al.*, 2006; *Long et al.*, 2014]. *GRACE* satellite provides information on changes in the gravity field which are controlled primarily by variations in water distribution and are used to derive *TWSC* at a spatial resolution of 1 degree. To estimate *TWSC*, we used GRCTellus land data based on the Level-1 *GRACE* observations, processed at JPL [*Watkins et al.*, 2015]. The spatial resolution is 1× degree. Because of the sampling and post-processing of *GRACE* observations, *TWSC* at small spatial scales tends to be attenuated. Therefore, *TWSC* was corrected by multiply the GRCTellus land data by the scaling coefficient provided by Community Land Model 4 [*Landerer and Swenson*, 2012; *Wiese*, 2015; *Watkins et al.*, 2015]. Detailed information can be addressed in <http://grace.jpl.nasa.gov/data/get-data/monthly-mass-grids-land/>. *TWSC* includes mass changes in groundwater storage, surface water and soil moisture and [*Long et al.*, 2014]. For runoff evaluation, monthly mean of composite runoff from Global Runoff Data Centre (*GRDC*) were used (Figure 2.9). *GRDC* composite runoff was corrected by observed inter-station discharge [*Fekete et al.*, 2002]. Moreover, Global Precipitation Climatology Centre (*GPCC*) monthly precipitation dataset was also used (Figure 2.8). A detail data description can be found in

Table 2.2.

*GLEAM* is designed to maximize the use of satellite-based observations to create a spatially coherent estimate of the evaporative flux over land [Miralles *et al.*, 2011a]. Global maps of evapotranspiration from land-surface are available with a daily resolution on a  $0.25 \times 0.25$ -degree grid. *GLEAM* relies on a Priestley-Taylor ( $P - T$ ) approach [Priestley and Taylor, 1972] to derive daily potential *ET* based on satellite observed surface net radiation ( $R_n$ ),  $T_a$  and a Gash's analytical model based on observed precipitation ( $P$ ) and different vegetation's characteristic was used to derived  $E_c$ . As a result estimates of *ET* for day  $j$  are derived from

$$ET_j = E_{P,i}S_i + (1 - \beta)E_{c,i} \quad (2.11)$$

where  $E_p$  is derived through  $P - T$  approach.  $S_i$  denotes the evaporative stress and is computed combining observed vegetation water content and estimates of root-zone soil moisture from a multilayer soil module driven by observed precipitation and surface soil moisture.  $\beta$  is a constant to account for declines in transpiration when the canopy is wet. A full description of *GLEAM* was conducted in Miralles *et al.* [2011a,b] and the data can be download from <http://foofoo.ugent.be/satex/GLEAM/>.

The Community Land Model versions 4.5 (hereafter *CLM*) are the latest in a series of land models developed through the Community Earth System Model (*CESM*) project. *CLM* is a physically based model having a more complex *ET* computation for turbulent heat transfer based on Monin-Obukhov similarity theory, which is solved numerically to account for atmospheric stability [Choi and Liang, 2010]. In the case of a vegetated surface, *ET* is partitioned into vegetation and ground fluxes that depend on canopy temperature ( $T_c$ ) and ground temperature  $T_s$  in addition to air temperature  $T_a$  and specific humidity  $q_a$ . Newton-Raphson iteration is used to solve for  $T_c$ ,  $H$  and *ET* from vegetation simultaneously using the ground temperature from the previous time step.  $E_c$  in *CLM* is derived through a simple model based on observed  $P$ ,  $LAI$  and stem area index

(*SAI*). Detailed information can be found in Technical Description of version 4.5 of the Community Land Model [Oleson *et al.*, 2013].

### 2.4.3 Global *ET* partitioning

A global *LAI* product (<http://globalchange.bnu.edu.cn/research/lai/#usage>), released by the Land-Atmosphere Interaction Research Group at Beijing Normal University, was used in this study (Table 2.1). This *LAI* product was generated by reprocessing the *MODIS LAI* products which is available from 2000 to 2014 is generated in 8-days intervals. In this dataset, *MODIS LAI* products were improved by using a two-step integrated method. This improved *MODIS LAI* data are more accurate, spatiotemporally continuous and consistent than current *MODIS LAI* data, and can be considered as input for *LSM* and *GCM* modelling [Yuan *et al.*, 2011].

The International-Geosphere-Biosphere-Program (*IGBP*) land cover dataset over the period 2000-2001 with  $1 \times 1$  degree resolution [Friedl *et al.*, 2010; Hall *et al.*, 2006] was also used in our study (Table 2.1). This products describe the geographic distribution of the 17 land cover classification scheme proposed by *IGBP* (Table 2.3). In each grid cell, Fraction of each land cover class ( $F_v$ ) was also involved. In this study, these 17 land cover classification scheme is re-classified into 6 land cover classifications, to match global synthesis of *LAI* control on *ET* partitioning analysis.

Based on estimated *ET*, fraction of each land cover class ( $F_{v_j}$ , a value of 0 to 1 for each vegetation class and 0 for non-vegetation type) and *LAI* regression for each class of vegetation ( $aLAI^b$ ),  $T$  in each grid can be calculated as:

$$T = \sum_{j=1}^n [(ET - E_c) \times F_{v_j} \times aLAI^b] \quad (2.12)$$

where canopy interception  $E_c$  is taken from *GLEAM* dataset or *CLM* dataset. As a

result,  $T/ET$  in each grid or in global is given by:

$$\frac{T}{ET} = \frac{1}{ET} \sum_{i=1}^n [(ET - E_c) \times F_{v_j} \times aLAI^b] \quad (2.13)$$

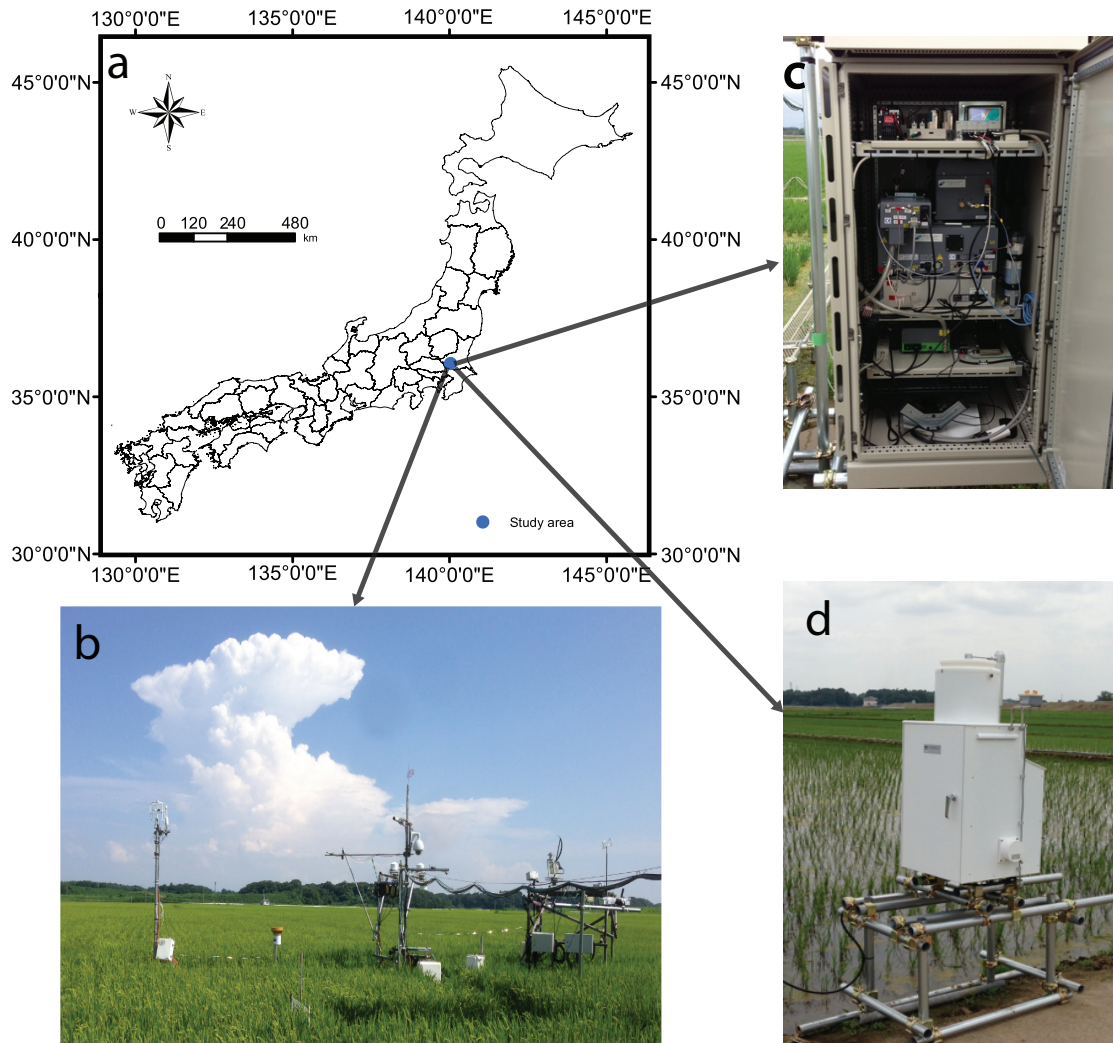


Figure 2.1: (a). Location of the study site in Mase, Japan (closed blue circle). (b). A photo about measurement site. (c). A photo about water vapor isotopic measurement system (Picarro L2120-i). (d) A photo about Automatic rain sampler (Eigenbrodt NSA181).

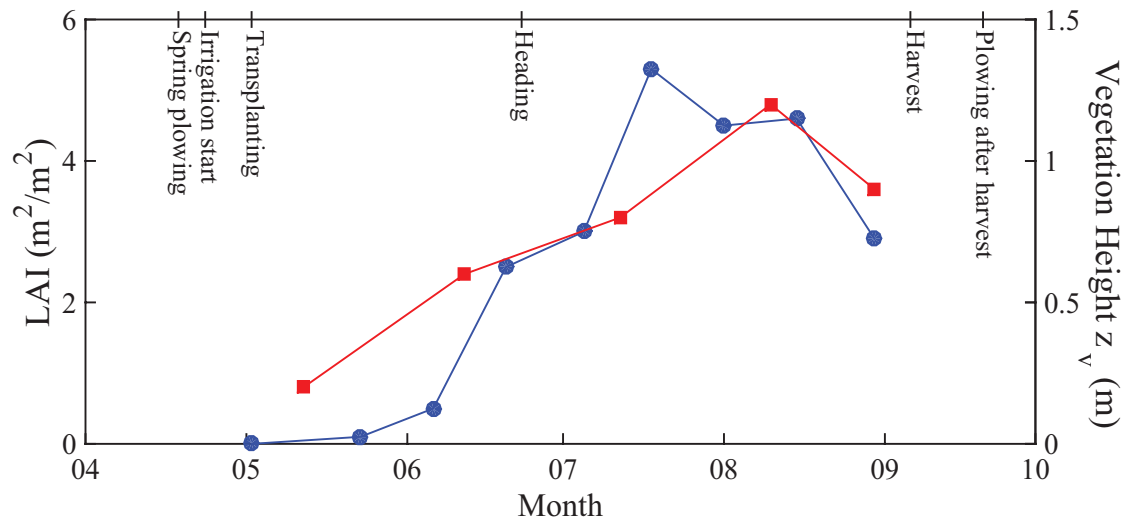


Figure 2.2: Linearly interpolated measurements of leaf area index ( $LAI$ , blue filled circle) and Vegetation height ( $z_v$ , red filled square) throughout the whole growing season in 2013.  $LAI$  and  $z_v$  was measured by the National Institute for Agro–Environmental Sciences (*NIAES*). Field management of the study site in 2013 is also summarized.

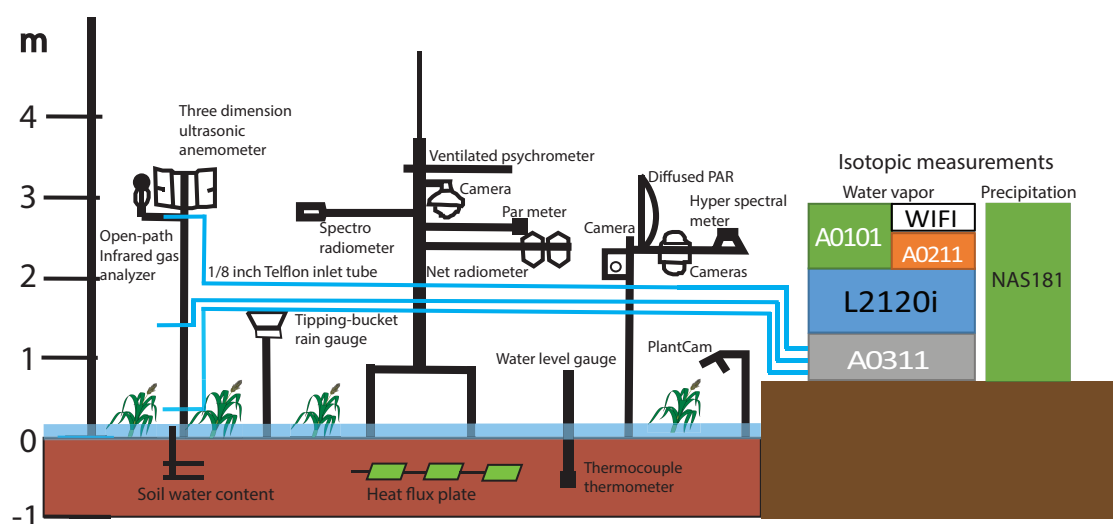


Figure 2.3: The measurement content schematic in this study.

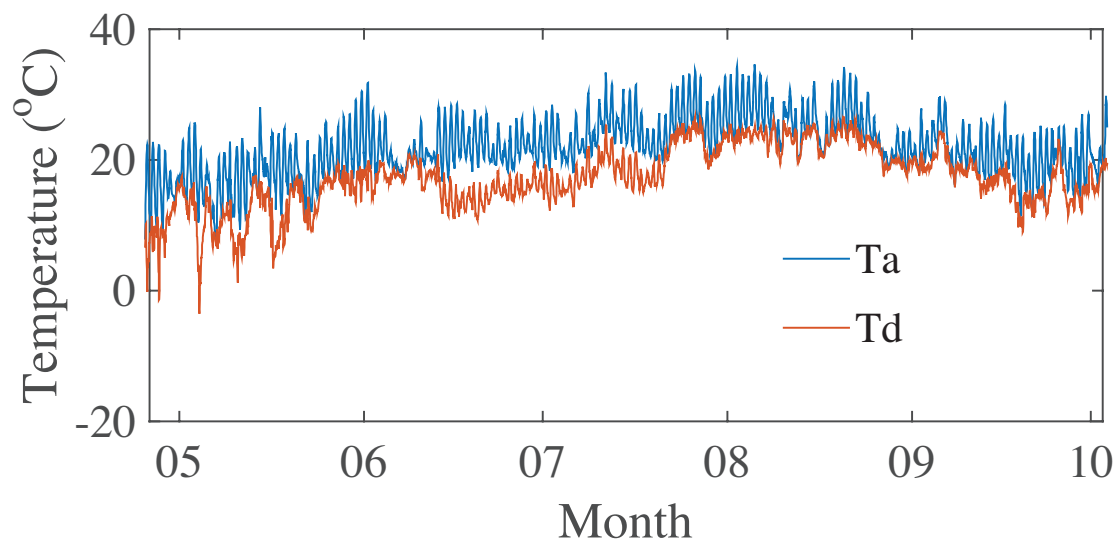


Figure 2.4: Comparison of dew temperature ( $T_d$ ) and air temperature ( $T_a$ ).

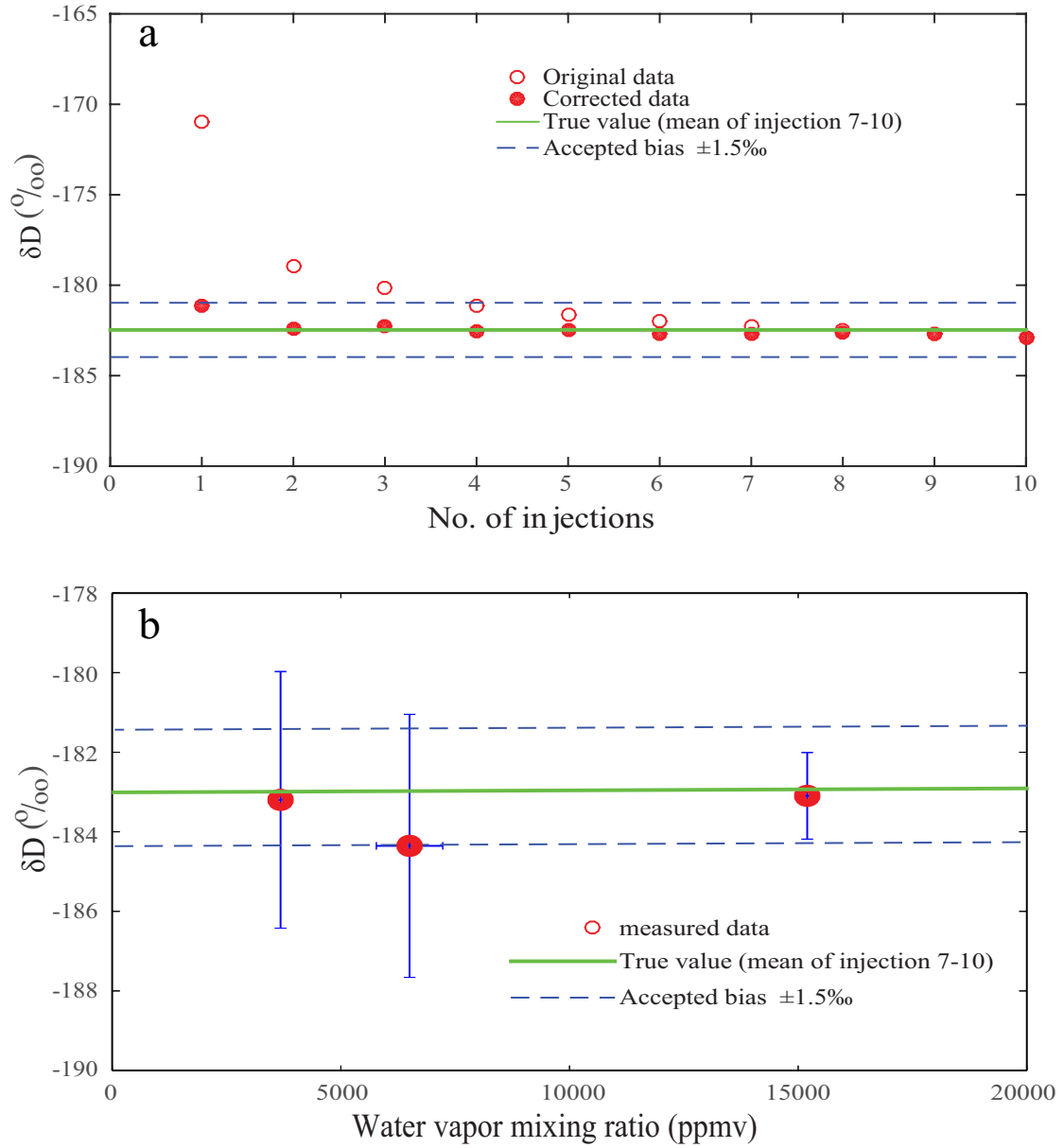


Figure 2.5: Calibration of water vapor and precipitation data (a). An example of correction procedure [van Geldern and Barth, 2012] to reduce memory effect in precipitation isotopic analysis. All the injections of a sample can be used after correction and the number of rejections per unknown can be reduced to 4. Four injections of each sample were averaged and normalized to the *VSMOW/SLAP* scale by using a two-point linear calibration. (b). Evaluation of the dependency of the measured isotopic value on water vapor concentration by injecting different amounts of standard water into the vaporizer. Three tests have been done in field conditions, during 2013/09/01–2013/09/02 ( $Q = 15200 \pm 200$  ppmv,  $\delta D = 183.1 \pm 1.1\text{‰}$ , Total number,  $n = 1516$ ), 2014/09/16–2014/09/17 ( $Q = 6500 \pm 700$  ppmv,  $\delta D = 184.3 \pm 3.3\text{‰}$ ,  $n = 2109$ ) and 2015/03/31–2015/04/01 ( $Q = 3700 \pm 100$  ppmv,  $\delta D = 183.2 \pm 3.2\text{‰}$ ,  $n = 1136$ ).

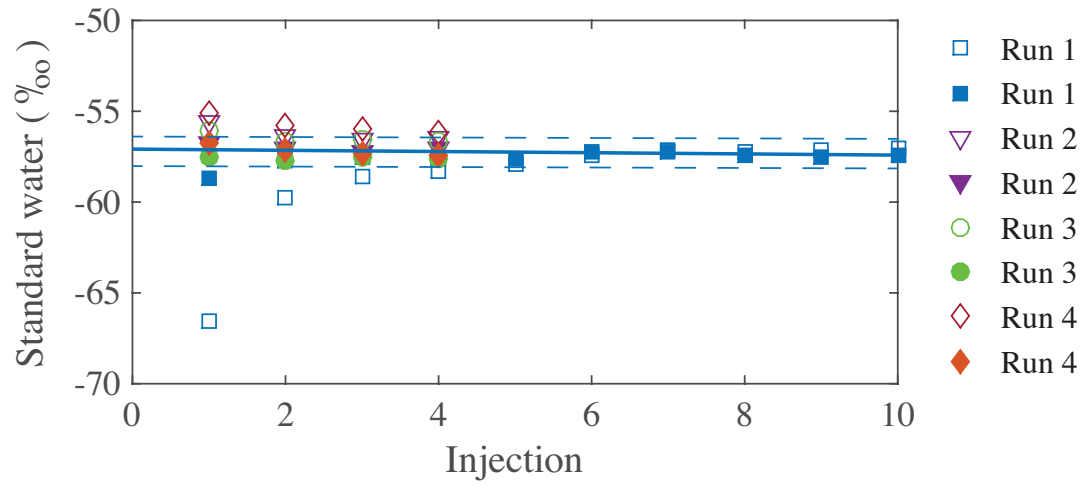


Figure 2.6: Comparison of dew temperature ( $T_d$ ) and air temperature ( $T_a$ ).

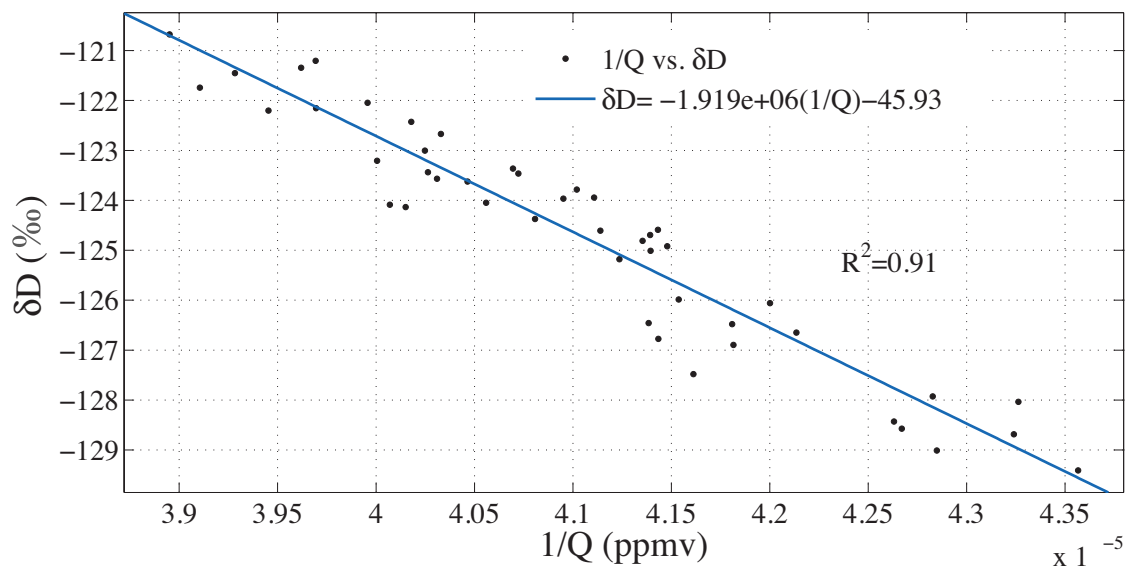


Figure 2.7: An example of  $\delta_{ET}$  estimation using Keeling plot.

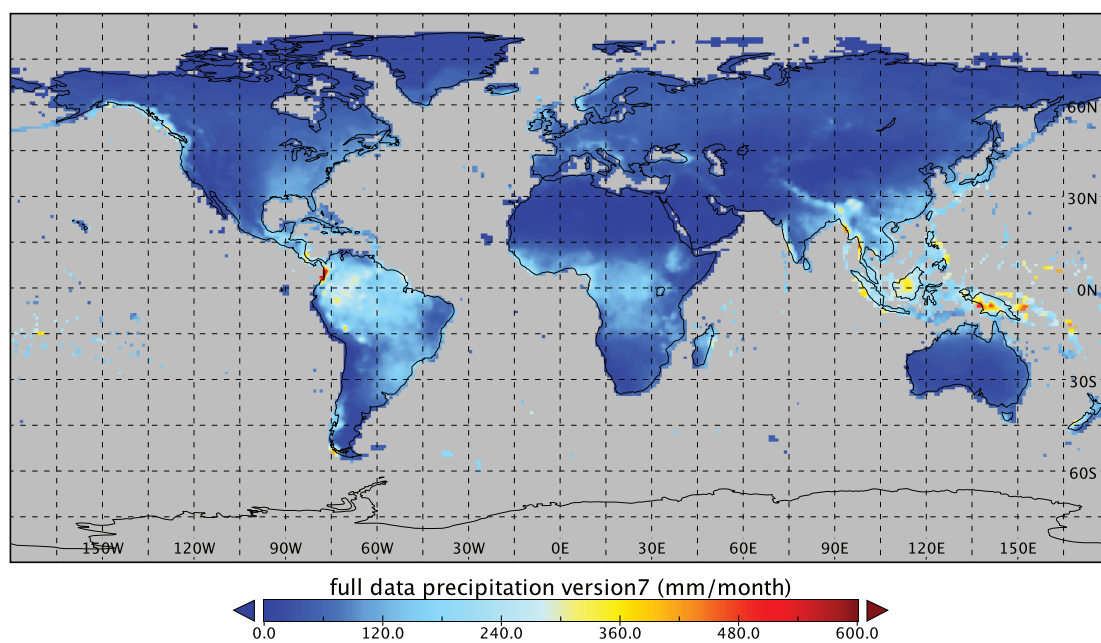


Figure 2.8: Mean precipitation in mm/month (2004–2010), 1.0 degree, obtained from *GPCP* full data product version 7.

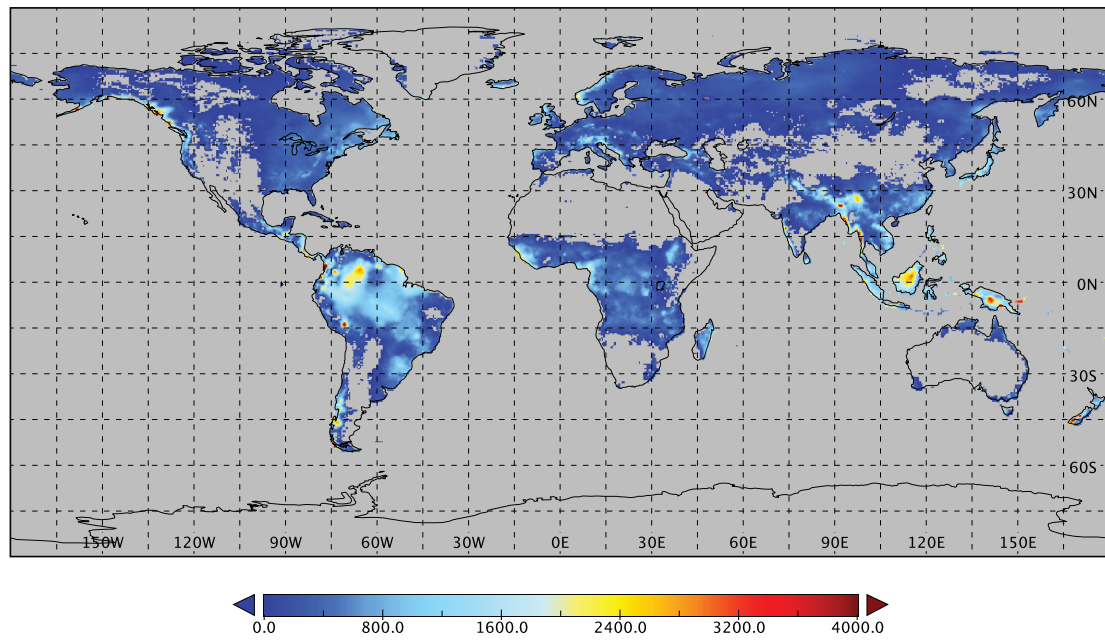


Figure 2.9: *GRDC* composite annual runoff (mm/year).

Table 2.1: The dataset used in global  $T/ET$  estimation.

| Variables         | Datasets                     | Spatial extent | Spatial Resolution | Temporal Resolution | Time Span | References  |
|-------------------|------------------------------|----------------|--------------------|---------------------|-----------|---|
| $ET$              | <i>CLM</i>                   | Global         | 1°                 | Monthly             | 1850–2010 | <i>Lawrence et al.</i> [2011]                                     |
|                   | <i>GLEAM</i>                 | Global         | 0.25°              | Daily               | 2000–2011 | <i>Miralles et al.</i> [2011a];<br><i>Miralles et al.</i> [2011b] |
|                   | <i>GRACE</i> de-rived        | Global         | 1°                 | Monthly             | 2002–2015 | See Table 2.2   |
| $LAI$             | Improved <i>MODIS</i> data   | Global         | 1 km               | 8 days              | 2000–2013 | <i>Yuan et al.</i> [2011]   |
| Land cover        | <i>ISLSCP II</i>             | Global         | 1°                 | annual              | 2000–2001 | <i>Friedl et al.</i> [2010]                                       |
|                   | <i>MODIS IGBP</i> land cover |                |                    |                     |           |   |
| Site measurements | Previous studies             | site           | -                  | -                   | -         | See Appendix A  |

Table 2.2: Data used in estimation of *GRACE*-inferred *ET*.

| Variable | Spatial<br>Resolution | Temporal<br>Resolution | Time<br>Span | Reference                      |
|----------|-----------------------|------------------------|--------------|--------------------------------|
| $P$      | 1°C                   | Monthly                | 1901–2013    | <i>Schneider et al.</i> [2014] |
| $R$      | 0.5°C                 | Monthly                | –            | <i>Fekete et al.</i> [2002]    |
| $TWSC$   | 1°C                   | Monthly                | 2002–2015    | <i>Watkins et al.</i> [2015]   |

Table 2.3: Land cover type codes and definitions and *IGBP* and this study.

| Codes In IGBP | Definitions                        | New classification        |
|---------------|------------------------------------|---------------------------|
| 1             | Evergreen Needleleaf Forests       | Needleleaf Forests        |
| 3             | Deciduous Needleleaf Forests       |                           |
| 2             | Evergreen Broadleaf Forests        | Broadleaf Forests         |
| 4             | Deciduous Broadleaf Forests        |                           |
| 5             | Mixed Forests                      | Mixed Forests             |
| 6             | Closed Shrublands                  | Scrublands and Grasslands |
| 7             | Open Shrublands                    |                           |
| 8             | Woody Savannas                     |                           |
| 9             | Savannas                           |                           |
| 10            | Grasslands                         |                           |
| 11            | Permanent Wetlands                 | Wetlands                  |
| 12            | Croplands                          | Croplands                 |
| 14            | Cropland/Natural Vegetation Mosaic |                           |
| 13            | Urban and Built-Up                 | Others                    |
| 15            | Permanent Snow and Ice             |                           |
| 16            | Barren or Sparsely Vegetated       |                           |
| 17            | Unclassified                       |                           |
| 0             | Water surface                      |                           |

# Chapter 3

## Understanding the variability of $\delta_v$

### 3.1 Characteristics of observation and simulation

Figure 3.1 shows the daily precipitation ( $P$ ), daily averaged water mixture ratio ( $Q$ ),  $\delta^{18}O$ ,  $\delta D$  and  $d - excess$  of water vapor (with subscript  $v$ ), and the weighted precipitation amount of  $\delta^{18}O$ ,  $\delta D$  and  $d - excess$  (with subscript  $p$ ).  $Q$  varied from 3000 to 35000 ppmv.  $\delta^{18}O_v$  fluctuated from -28.4‰ to -11.0‰,  $\delta D_v$  fluctuated from -206.8‰ to -78.6‰, and  $d_v - excess$  fluctuated from -1.8‰ to 31.9‰. There was considerably less day-to-day variation in  $\delta^{18}O_v$ ,  $\delta D_v$ , and  $d_v - excess$  in the warm season than the cold season. Similar results were observed in Beijing (40° 00' N, 116° 23' E [Lee *et al.*, 2006]). The monthly isotopic variation can be explained by strong summer monsoon activity in East Asia, including Japan and southeastern China [Wen *et al.*, 2010; Laskar *et al.*, 2014]. The observed  $\delta_p$  varied widely, ranging from -15.7‰ to 2.9‰ for  $\delta^{18}O_p$ , from -114.9‰ to 34.5‰ for  $\delta D_p$ , and from -21.6‰ to 27.7‰ for  $d_p - excess$ . Positive  $\delta^{18}O_p$  and  $\delta D_p$  values tended to come up in light rain, which were subject to sub-cloud evaporation [Wen *et al.*, 2010; Huang and Wen, 2014]. Water vapor appeared to be isotopically in equilibrium with surrounding raindrops during a precipitation event. Because the water vapor mixing ratio increased with precipitation, the vapor isotope ratios became progressively lower during rainy periods.  $\delta^{18}O_v$  and  $\delta D_v$  were consistently lower than but varied together

with precipitation isotopes. Similarly,  $d_v - excess$  consistently varied with  $d_p - excess$ . As noted in previous studies [e.g., *Lee et al.*, 2006; *Welp et al.*, 2008; *Wen et al.*, 2010], near surface vapor isotope ratios approached the equilibrium state with the ratios in precipitation. These interactions between raindrops and the surrounding water vapor during precipitation events have been quantitatively estimated by *Lee and Fung* [2008].

Monthly isotope and weather variables are shown in Table 3.2. Seasonal changes of relative humidity ( $RH$ ),  $\delta D$ ,  $d - excess$  and air temperature ( $T_a$ ) are plotted in Figure 3.2.  $RH$  and  $T_a$  displayed a clear seasonal signal. There was no clear seasonal cycle in  $\delta_v$  or  $\delta_p$ , although slightly lower values were apparent in winter. In contrast,  $d_v - excess$  and  $Q$  displayed a clear seasonal cycle, which was different from that observed in those areas influenced by the typical arid continental climate at similar latitudes (e.g., Zhangye, China ( $38^\circ 51' N$ ,  $100^\circ 22' E$ ) [*Huang and Wen*, 2014]). The highest and lowest monthly mean values of  $d_v - excess$  were found in January (27.3‰) and July (11.4‰), respectively.  $d_p - excess$  was more variable with the highest value found in January (16.3‰) and the lowest in November (1.5‰). Different from those ocean surface studies [e.g., *Benetti et al.*, 2014, 2015; *Uemura et al.*, 2008, 2012; *Steen-Larsen et al.*, 2014b, 2015], the overall relationships between isotopes and weather variables were found weak and controversial (Table 3.1). Seasonal changes of diurnal averaged  $Q$ , isotope ratio  $\delta^{18}O$ ,  $\delta D$ ,  $d - excess$  of water vapor were shown in Figure 3.3.  $\delta^{18}O$  and  $\delta D$  diurnal variations was likely in phase with the variations in  $Q$ . The diurnal variability of  $d - excess$  was in opposite phase with those of the vapor  $\delta^{18}O$ ,  $\delta D$ . The vertical mixing of air between  $CBL$  and  $ET$  may have played a role in d-excess diurnal variations [*Wen et al.*, 2010].

Figure 3.1 compares an *IsoGSM* isotopic simulation at 2 m with surface observations. *IsoGSM* reproduced the variability of  $Q$  very well ( $R^2 = 0.95$ ,  $RMSE = 832$  ppmv). This implies that our site can reasonably capture  $\delta_v$  variation under the influence of regional-scale advection of atmospheric moisture ( $R^2 = 0.64$ ,  $RMSE = 10.8$ ‰ for  $\delta D_v$ ,  $R^2 = 0.70$ ,  $RMSE = 1.2$ ‰ for  $\delta^{18}O_v$ ). Variability in  $d - excess$  was difficult to simulate [e.g., *Bonne et al.*, 2014]. *IsoGSM* predicted a relatively constant  $d_v - excess$  value of 10‰ and

generally underestimated observed  $d_v - excess$  values throughout the observation period ( $R^2 = 0.20$ ,  $RMSE = 5.5\text{‰}$ ). Model discrepancies indicated that water vapor isotopes were an additional constraint that highlighted the misrepresentation of hydrological processes in the simulation. We did not further explore the possible causes of model bias. Rather, we focused on identifying the controls of near surface  $\delta_v$ . Despite the discrepancy in modeled isotopic variability, *IsoGSM* provided important information regarding how large-scale moisture transfer influenced  $\delta D_v$ . The strong fit at our site suggested that the variability of  $\delta_v$  is primarily influenced by large-scale advection, whereas local evapotranspiration likely played an insignificant role (because *IsoGSM* reproduces large-scale advection rather than local evapotranspiration). However, more evidence is required to determine the driving forces behind the variability of  $\delta D_v$ . Hereafter, we only present results based on  $\delta D$  and  $d - excess$ . Similar results obtained from  $\delta^{18}O$  are not shown here.

## 3.2 Effect of precipitation on the $\delta D_v$ variability

The observed and simulated isotopic ratios (Figure 3.1) indicated that during non-rainy days, there was no significant isotopic variation of water vapor. On rainy days,  $\delta D_v$  changed suddenly and considerably in relation to the isotopic composition of rain (Figure 3.1). For example, in association with a precipitation event that occurred on October 15 (19 mm),  $Q$  increased from 14520 ppmv to 18180 ppm, whereas  $\delta^{18}O_v$  decreased from -99.2‰ to -180‰. This variation indicated a significant isotopic impact of raindrops on near surface water vapor. To verify this quantitatively, we calculated  $\delta D_{v,e}$  based on Equation 2.3 and compared it with  $\delta D_v$  in Figure 3.4.  $\delta D_v$  agreed reasonably well with  $\delta D_{v,e}$  ( $R = 0.74$ ) at the daily time scale. This robust relationship indicated that atmospheric vapor approached an isotopic equilibrium with precipitation at daily time scale. At a monthly time scale, our equilibrium predictions differed greatly from the monthly  $\delta D_v$  (Figure 3.2,  $R = 0.22$ ). These results contradicted previous findings, which demonstrated a reasonable agreement between  $\delta D_v$  and  $\delta D_{v,e}$  on a monthly time scale [Jacob and

*Sonntag*, 1991; *Lee et al.*, 2006; *Wen et al.*, 2010; *Huang and Wen*, 2014]. Furthermore, we found no obvious relationship between monthly  $d_v - excess$  and predicted equilibrium values. Predicted  $d_{v,e} - excess$  was generally similar to that of precipitation. A possible reason for this is that condensation affected  $\delta$  without changing  $d - excess$  [*Dansgaard*, 1964; *Bonne et al.*, 2015].

### 3.3 Effect of local $ET$ on the the $\delta D_v$ variability

During non-rainy days, surface  $ET$  can significantly contribute to the temporal variation in  $\delta_v$ . We investigated this effect using Equation 2.9 with three parameters,  $\delta_{ET}$ ,  $\delta D_v$  and  $\delta_{bg}$  (the isotopic ratio of the background air). Hourly  $\delta_{ET}$  values were calculated using the Keeling plot approach [*Keeling*, 1958], whenever data points were available ( $N > 40$ ) in a given hour and with a high coefficient of determination ( $R^2 > 0.8$ ) from each linear regression. *Wei et al.* [2015] demonstrated that at this site, the assumptions underlying the Keeling plot were rarely met and the uncertainty was quite large, especially under nighttime or weakly turbulent conditions. We assumed no evapotranspiration at night (19:00 p.m. - 7:00 a.m.) and used daily average  $\delta_{ET}$  values (sensible heat weighted).  $\delta D_v$  at simulated  $CBL$  height ( $\delta_{bg}$ ) was derived from linear interpolation of water vapor isotopic ratio from 925 hpa (about 750 m), 850 hpa (about 1500 m) and 700 hpa level (about 3000 m) for three reasons: 1) *IsoGSM* simulated the convective boundary layer extended with a height of 0.8 to 2 km during our observation periods (Figure 3.5); 2) *IsoGSM* is nudged by United States National Centers for Environmental Prediction (*NCEP*) reanalysis data, and thus reproduces the day-to-day variability induced by large-scale convection; and 3) the model produces a satisfactory qualitative agreement for the isotopic composition of atmospheric moisture at 2-m level (Figure 3.1).

The daily variability of  $\delta_{ET}$ ,  $\delta_{bg}$ , and  $\delta D_v$  is shown in Figure 3.6.  $\delta_{ET}$  ranged from -112.2‰ to 37.1‰. A significant difference in  $\delta_{ET}$  was found between the growing and after haverst seasons. During the summer season (June-July-August, JJA),  $\delta_{ET}$  generally

depended on transpiration.  $\delta_{ET}$  was closely coupled with irrigation water, indicating small isotopic fractionations during this period [Wei *et al.*, 2015]. In the non-growing season,  $\delta_{ET}$  was considerably more variable than in JJA. Without irrigation and consequently with negligible transpiration,  $\delta_{ET}$  was controlled by other meteorological parameters, such as precipitation and radiation. For example, a rapid decrease of  $\delta_{ET}$  corresponded to evaporation after precipitation on 18 October (from  $-51.8\text{‰}$  on 18 October to  $-97.81\text{‰}$  on 20 October). Then  $\delta_{ET}$  values gradually increased due to soil evaporation ( $-69.2\text{‰}$  on 21 October). Compared to  $\delta_{ET}$ ,  $\delta D_v$  and  $\delta_{bg}$  clearly displayed seasonal variation and were highly correlated with each other. These results suggest that  $\delta D_v$  was mainly controlled by the air from the free atmosphere. The largest difference between  $\delta D_v$  and  $\delta_{bg}$  was found in JJA, whereas the smallest difference was found in winter (December-January-February, DJF). In JJA, relatively high  $\delta D_v$  values were likely due to enrichment by transpiration, whereas a low  $d_v - excess$  is a result of the negligible kinetic effect associated with transpiration by short vegetation [Lai *et al.*, 2008]. Our results showed a large  $ET$  impact during the rice growing season (JJA). The monthly averaged  $\delta D_v$ ,  $\delta_{ET}$  and  $\delta_{bg}$  used to estimate monthly  $F_{ET}$  are listed in Table 3.3. On average,  $F_{ET}$  was estimated as  $16.0 \pm 12.3\%$ , with the highest value in JJA ( $20.5 \pm 12.9\%$ ) and lowest in DJF ( $7.1 \pm 13.2\%$ ). These estimates are consistent with simulated seasonal averaged latent heat fluxes (Figure 3.7). Thus, we suggest that the primary control on the variability of near surface  $\delta_v$  and humidity during non-rainy days is large scale moisture transfer, while local  $ET$  plays only the secondary role, especially in DJF.

Nevertheless, large uncertainties were still present in these estimates. The ability of *IsoGSM* to reproduce water vapor and its isotopic variability observed at a single location was unexpected because the *IsoGSM* simulation was performed at a coarse resolution ( $200 \text{ km} \times 200 \text{ km}$ ), which is far greater than the footprint of ground-based measurements [Farlin *et al.*, 2013]. Furthermore, feedback from the local land contribution was not carefully addressed. *IsoGSM* neglects possible fractionation during transpiration, which is a common problem in most isotope-enabled GCMs. Bias also arises from neglecting

horizontal advection, which is likely to alter the near surface  $\delta D_v$ . Very few measurements of  $\delta_{bg}$  are currently available [Tsujimura *et al.*, 2007]. Here, we used isotopic ratio of water vapor linear interpolated from water vapor isotopic ratio at different levels as background. However, *IsoGSM* simulated *CBL* height is rough, and the vertical profile of water vapor isotopes is general non-linear. To reduce the uncertainty in evaluating  $F_{ET}$ , a further study based on measured middle and low troposphere isotopes of water vapor isotopes (such as satellite measurement) should be conducted. Nevertheless, our results suggested that near surface water vapor isotope ratio is not significantly affected by *ET*. As explained previously, the high correlation between measured  $\delta D_v$  at 2-m and simulated  $\delta_{bg}$  at *CBL* height ( $R = 0.74$ ) with the insignificant correlation between measured  $\delta D_v$  at 2-m and Keeling plot estimated  $\delta_{ET}$  ( $R = 0.18$ ) also suggested the dominant role of air from the free atmosphere.

### 3.4 Remote controls on the $\delta D_v$ variability

To understand the remote controls on water vapor isotopic variation, we conducted a water tagging experiment to identify the origin of the water vapor. As an example, the model simulation indicated different sources of water vapor in JJA (Figure 3.8, Results of SON, DJF and MAM were shown in Appendix B). There was a significant contribution from the surrounding ocean (both the Japan Sea and Pacific Ocean; Areas c, d, e, and f). Assuming all water vapor originated from the tagged boxes, we were able to calculate the fractional relative contribution for each box. As Figure 3.9 shows, Area e had the most significant impact on moisture sources in JJA, corresponding to about 32% of the total contribution. In SON, flow from either the Japan Sea (Area d, 24%) or the high-latitude Pacific Ocean (Area f, 34%) had the dominant role in controlling the observed isotopic variability at our site. Water vapor generally originated from the Japan Sea in DJF (Area d, 40%). Likewise, variable sources of water vapor were found in MAM (March-April-May), with the largest contribution from Area d (29%). We documented

the 850 hpa isotope ratios based on the wind direction ( $0^\circ$  to  $90^\circ$ , Area f;  $90^\circ$  to  $180^\circ$ , Area e;  $180^\circ$  to  $270^\circ$ , Area a+c;  $270^\circ$  to  $360^\circ$ , Area b+d). The results showed that the original source had a significant impact on water vapor isotopes (Figure 3.10). Seasonal and regional differences of  $\delta D_v$  at the 850 hpa level were related to  $RH$  relative sea surface temperature (SST) ( $RH_s$ ) differences (not shown). More water vapor evaporated from areas with a low  $RH_s$  and a higher isotopic composition than from relatively high  $RH_s$  water bodies region. Through vertical mixing, surface water vapor isotopes can be significantly affected by different oceanic sources. Based on the tagging experiments, the observed seasonal cycle of  $d_v - excess$  (Table 3.2) can also be explained by the seasonal change in the primary marine moisture sources. As shown in Figure 3.1, the highest  $d_v - excess$  occurred in winter, while the lowest occurred in summer. Because moisture was generated under conditions with a high relative humidity in summer (not shown), evaporated water tended to have a lower  $d_v - excess$ . On the other hand, the high  $d_v - excess$  resulting from strong kinetic effects corresponded to low relative humidity and strong evaporation (Figure 3.7) above the ocean in DJF. These results were consistent with the negative correlation between  $d_v - excess$  and relative humidity observed above the ocean [Uemura *et al.*, 2008; Dansgaard, 1964; Benetti *et al.*, 2015, 2014].

The *IsoGSM* simulation suggested that the mixing of air from multiple sources was likely a major factor controlling the water vapor isotopic variation at our study site on non-rainy days. To identify the source of atmospheric moisture and its transport pattern, we briefly describe the theoretical framework for interpreting paired isotope ratios and specific humidity established by Noone [2012]. The framework, composed of a set of simple processes, enables the use of isotopic data to identify the hydrological process under idealized atmospheric conditions. We used two processes in the framework: 1) Rayleigh distillation curves (case b in Noone [2012], hereafter *RC*), representing a pseudoadiabatic process based on oceanic temperatures, with an initial  $\delta D$  value of -80‰ (approximating the  $\delta D_v$  value in equilibrium with that evaporated from the ocean) in an open system, and 2) a mixing line (hereafter *ML*), which represents a special case when the source water

is the infinite reservoir of vapor in thermodynamic equilibrium (i.e., saturated) with the ocean surface (case e in [Noone, 2012]). We plotted daily mean of  $\delta D_v$  as a function of specific humidity ( $q$ ) for each season. As shown in Figure 3.11, the moistening process (increasing  $q$ ) acted to enrich water vapor isotope ratios. During the winter season (DJF),  $q$  generally varied from 2 to 10 g/kg.  $\delta D_v$  was clustered tightly around the mixing line, suggesting a mixing of marine moist air and a dry air mass. This supports the idea that in general, cold northerly winds transport water vapor that originates from the Japan Sea to our site (Figure 3.9). Outside of the winter season,  $q$  varied over a wide range ( $2 < q < 25$  g/kg), while  $\delta D_v$  fell in the range between the  $RC$  and  $ML$ .  $ET$  might be one of the factors pulling data points away from the evaporation line. However, most data points were more depleted than -80‰, suggesting a negligible impact of  $ET$  on  $\delta D_v$  variation (most of the time,  $ET$  tends to increase  $\delta D_v$  [Risi et al., 2010]). Local moisture at our site was likely a result of combined marine sources in these seasons. The large variability of  $q$  and  $\delta D_v$  was related to the mixing of different oceanic sources (Figure 3.9) of distinct isotopic ratios (Figure 3.10). Mixing can occur between several different air masses that deviate from a single curve. Hence, the entire area between  $RC$  and  $ML$  can be considered a result of mixing [Galewsky and Samuels-Crow, 2014a]. The tagging experiment and  $q$ - $\delta D_v$  framework provide consistent evidence that the mixing of multiple moisture sources plays an important role in controlling  $\delta D_v$  variability of this region. In contrast to previous studies those conducted in inland regions [e.g., Lee et al., 2006; Huang and Wen, 2014; Welp et al., 2008; Wen et al., 2010], our measurements showed that  $q$  was a poor predictor of  $\delta D_v$  because of the mixing processes. These results suggest that the common assumption in a Rayleigh distillation model which assumed that water vapor is contributed from a single source, is unable to reproduce the water vapor isotopic variability over a subtropical area. The isotopic variability of water vapor also depends on its air trajectory. In DJF, a lot of vapor is supplied from the Japan Sea, which creates large amounts of snow along the coast of the Japan Sea, resulting in isotopic depleted water vapor and a high  $d$  - excess. By contrast, in JJA, water vapor was derived from

several adjacent oceans, with high humidity and a low evaporation rate, which resulted in an enriched isotopic signature and low  $d_v - excess$ . Being different from those inland area, water vapor source area is very close to our site, the removal of heavy isotopes from water vapor by Rayleigh distillation processes (moisture recycling) is insignificant. On the other hand, large differences of  $\delta D_v$  at 850 hpa level from different areas were clearly observed in each season (Figure 3.12). Thus, we believe source origin of water vapor, rather than physical processes along an air mass moving trajectory, is the primary control of water vapor isotopic variation. We also noted that some of the  $\delta D_v$  values in the wet season were lower than those predicted by *RC*. These data can be explained by shallow dry convection. Strong shallow dry convective activity generates water vapor  $\delta D_v$  below the Rayleigh curve, in a process known as the isotope amount effect [Galewsky and Samuels-Crow, 2014a,b; Bony et al., 2008]. However, more evidence is required to confirm this. Moreover, daily measured  $d - excess$  vs. *IsoGSM* estimated  $RH_s$  from the source regions ( $10 \times 10$  degree averaged) was plotted (Figure 3.13). A negative correlation similar with Steen-Larsen et al. [2015] was observed but with an insignificant correlation. The insignificant correlation may be resulted from 1): very rough resolution  $RH_s$  ( $10 \times 10$  degree averaged); 2) in this  $d - excess$  v.s.  $RH_s$  analysis, we assumed only one source in each day. It would raise significant error if the mixing of multiple moisture sources within a day and 3) We also assume that no time delay during water vapor transport (the evaporated oceanic water was transported to our site immediately) because we used  $RH_s$  at same day as measured  $d - excess$ . The slope (-22.77) and intercept (36.0) are also different from Steen-Larsen et al. [2015]. Nevertheless, further study should be conducted to understand the relationship between  $d - excess$  and water vapor source.

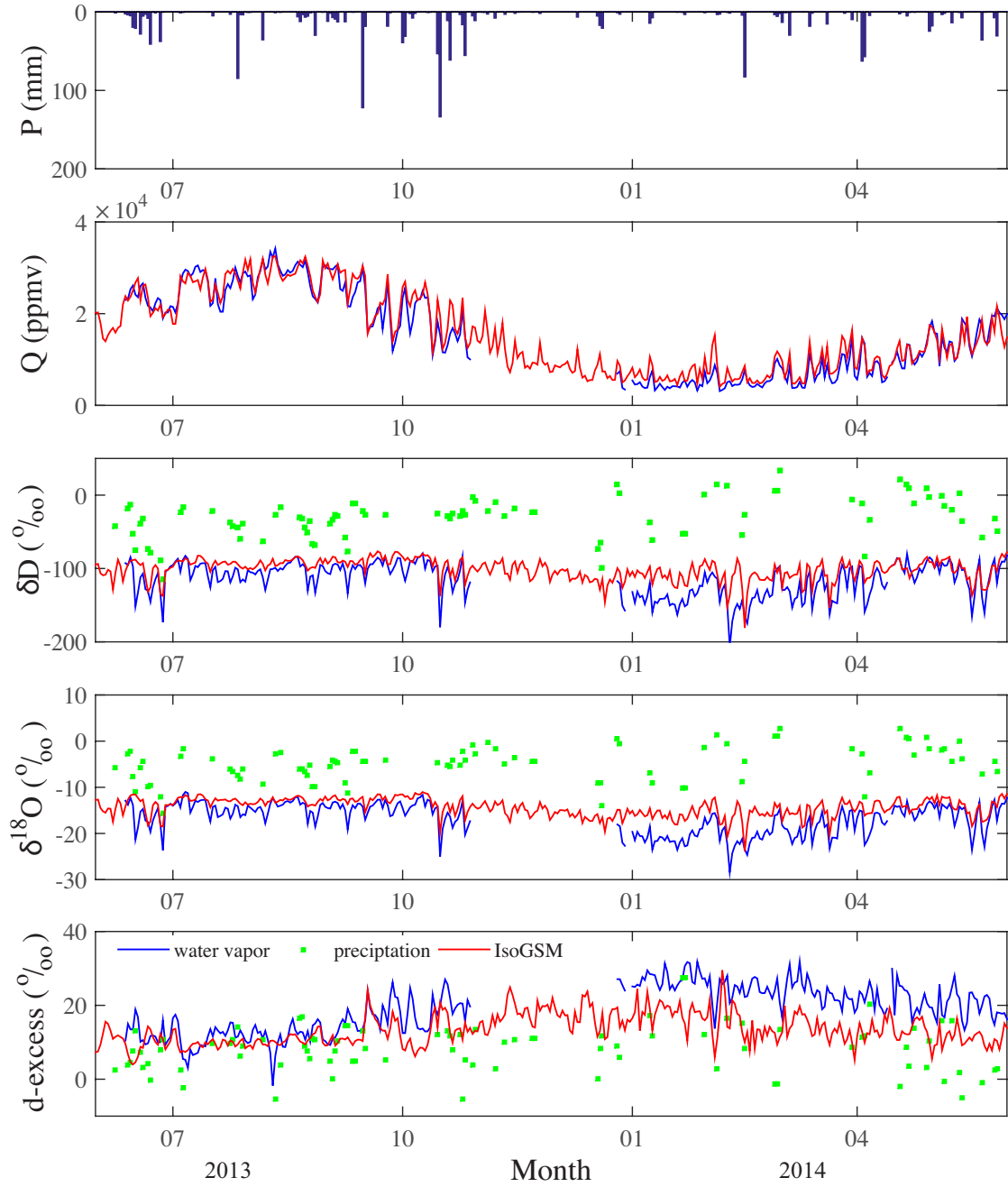


Figure 3.1: Time series of daily observation in precipitation amount ( $P$ ), vapor mixing ratios ( $Q$ ), water vapor and precipitation  $\delta^{18}O$ ,  $\delta D$  and  $d - excess$  (blue line for water vapor, green square for precipitation). The red lines represent the simulated  $Q$  and  $\delta$  by *IsoGSM*.

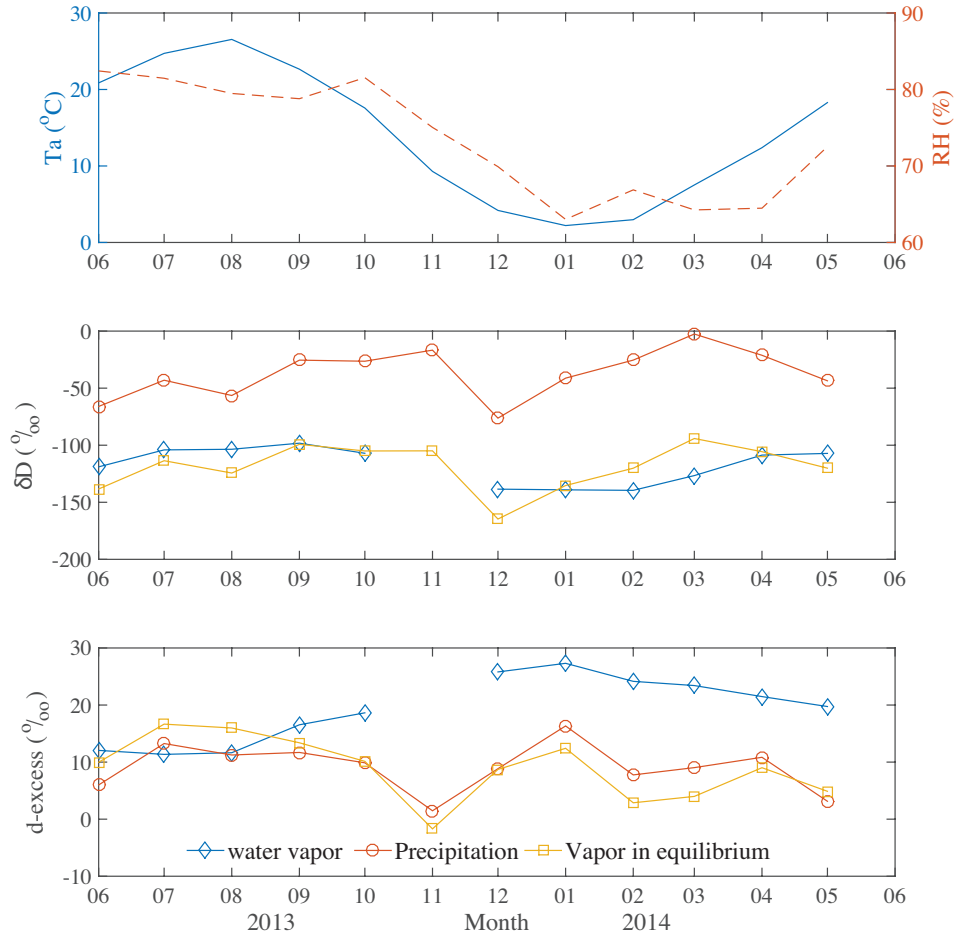


Figure 3.2: Monthly averaged relative humidity ( $RH$ , dash), Temperature ( $T_a$ , solid), isotope ratio  $\delta D$ ,  $d$  – excess of water vapor (diamond) and precipitation (circle). Water vapor isotope  $\delta D_{v,e}$  and  $d_{v,e}$  – excess predicted from precipitation (square) are also plotted.

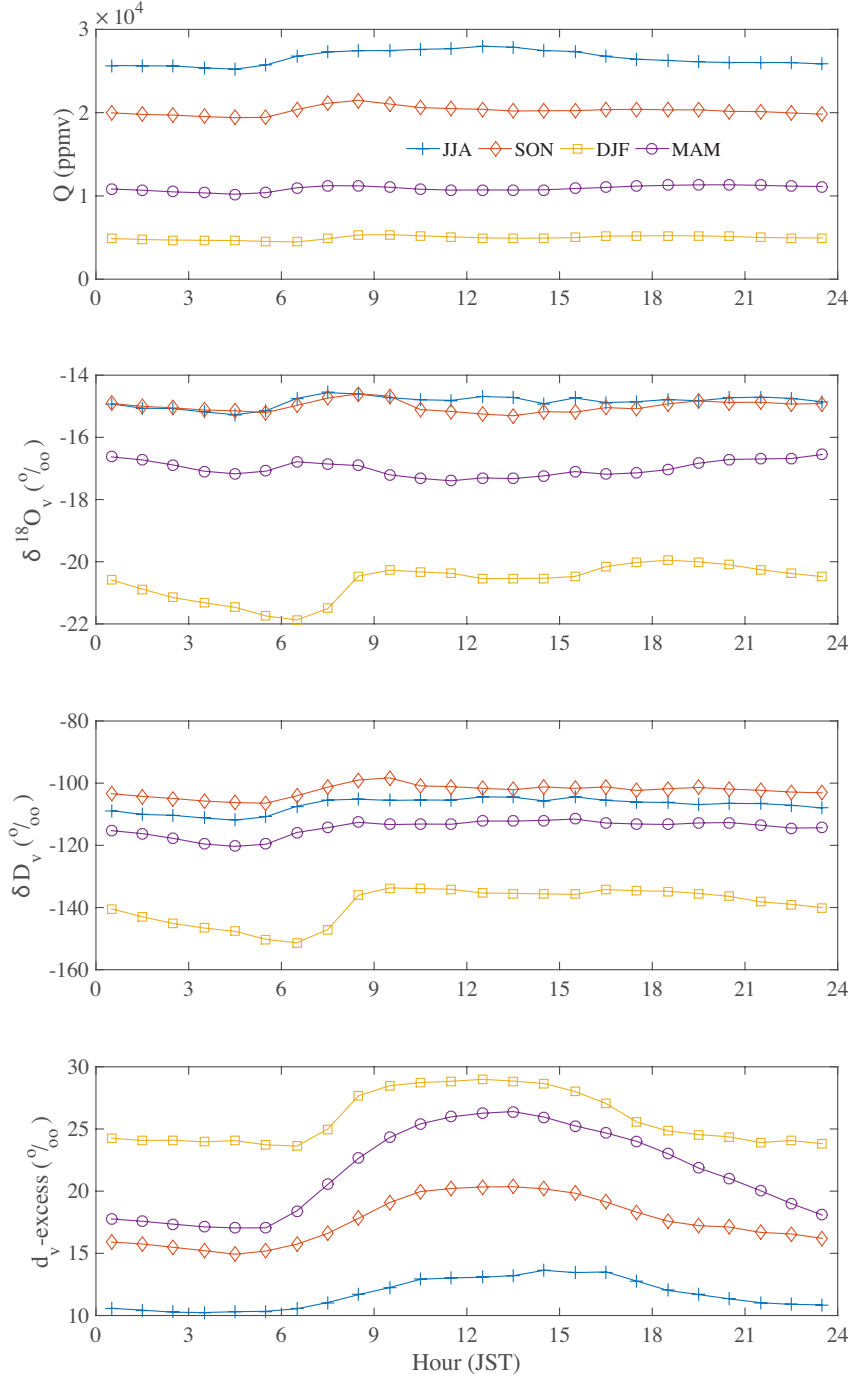


Figure 3.3: Diurnal averaged water vapor mixing ratio, isotope ratio  $\delta^{18}O$ ,  $\delta D$ ,  $d-excess$  of water vapor.

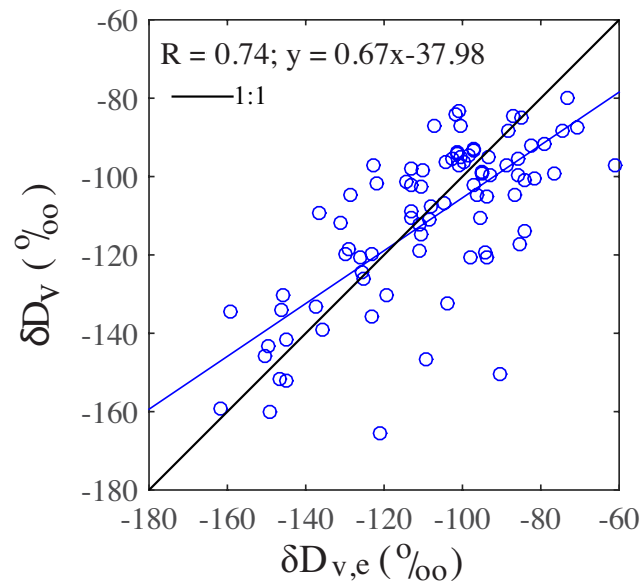


Figure 3.4: Comparison between the observed water vapor  $\delta D_v$  and precipitation predicted  $\delta D_{v,e}$ . Each point represents a rainy day.

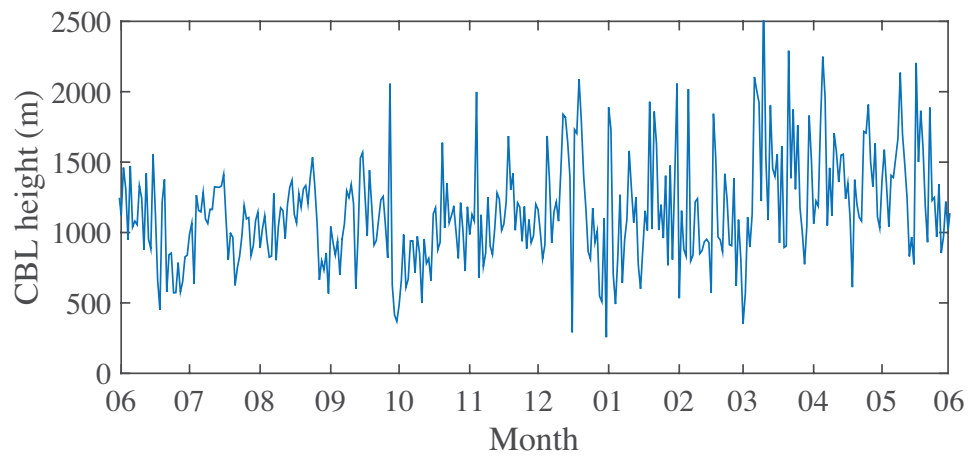


Figure 3.5: Day time *CBL* height simulated using *IsoGSM*.

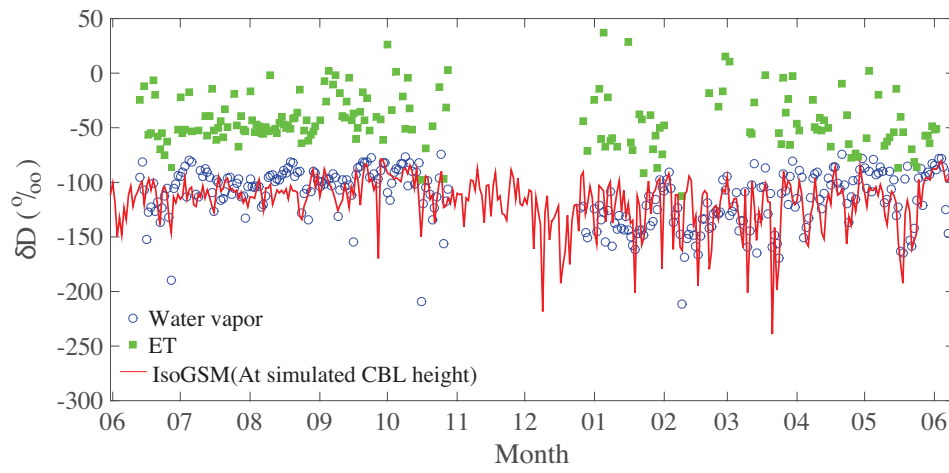


Figure 3.6: Daily variation of isotope ratios of evapotranspiration (green filled square, excluded rainy days), water vapor (blue unfilled circle, excluded rainy days) and background derived from *IsoGSM* simulation.

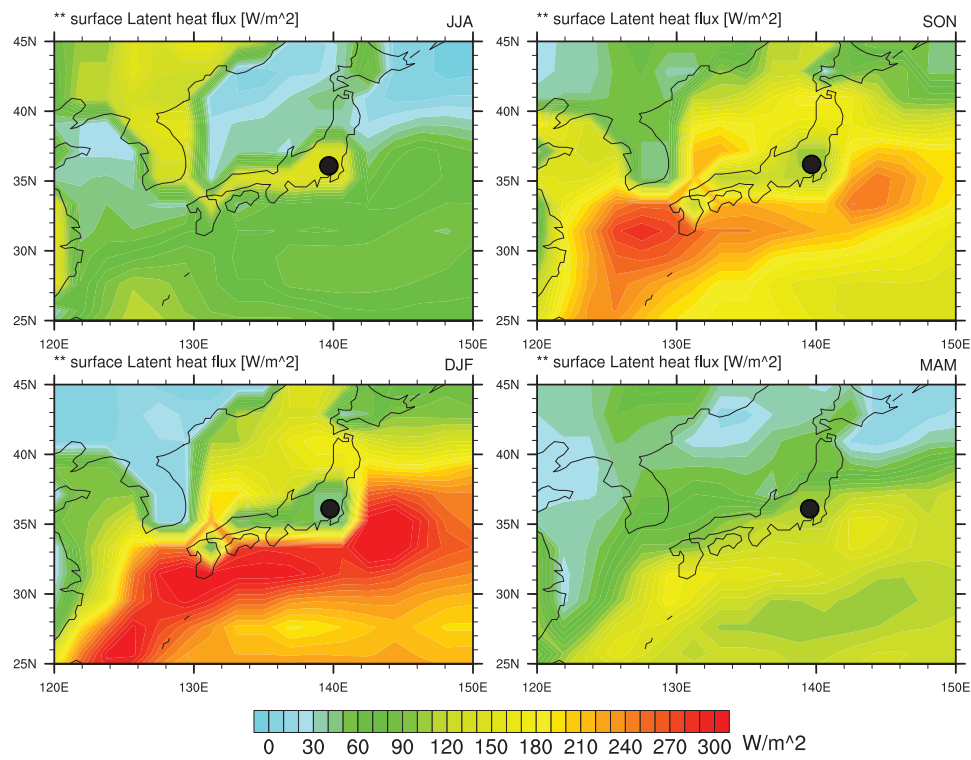


Figure 3.7: *IsoGSM* simulated seasonal mean latent heat fluxes. Black dot shows the study site.

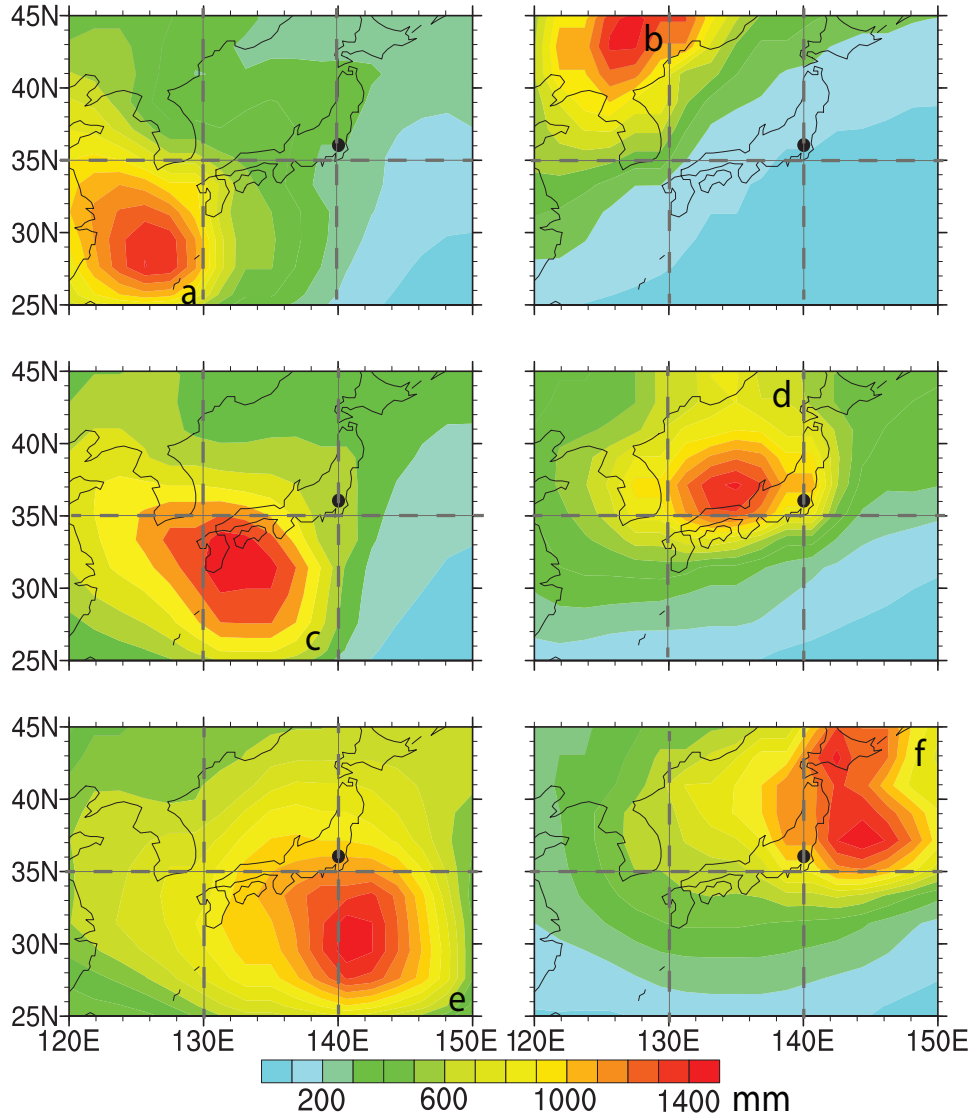


Figure 3.8: The atmospheric column precipitable water contributed from tagged boxes in JJA. Source regions for tagging experiments of water vapor tagged within  $10 \times 10$  degree boxes (contour): a,  $25^\circ\text{N} - 35^\circ\text{N}$ ,  $120^\circ\text{E} - 130^\circ\text{E}$ ; b,  $35^\circ\text{N} - 45^\circ\text{N}$ ,  $120^\circ\text{E} - 130^\circ\text{E}$ ; c,  $25^\circ\text{N} - 35^\circ\text{N}$ ,  $130^\circ\text{E} - 140^\circ\text{E}$ ; d,  $35^\circ\text{N} - 45^\circ\text{N}$ ,  $130^\circ\text{E} - 140^\circ\text{E}$ ; e,  $25^\circ\text{N} - 35^\circ\text{N}$ ,  $140^\circ\text{E} - 150^\circ\text{E}$ ; f,  $35^\circ\text{N} - 45^\circ\text{N}$ ,  $140^\circ\text{E} - 150^\circ\text{E}$ .

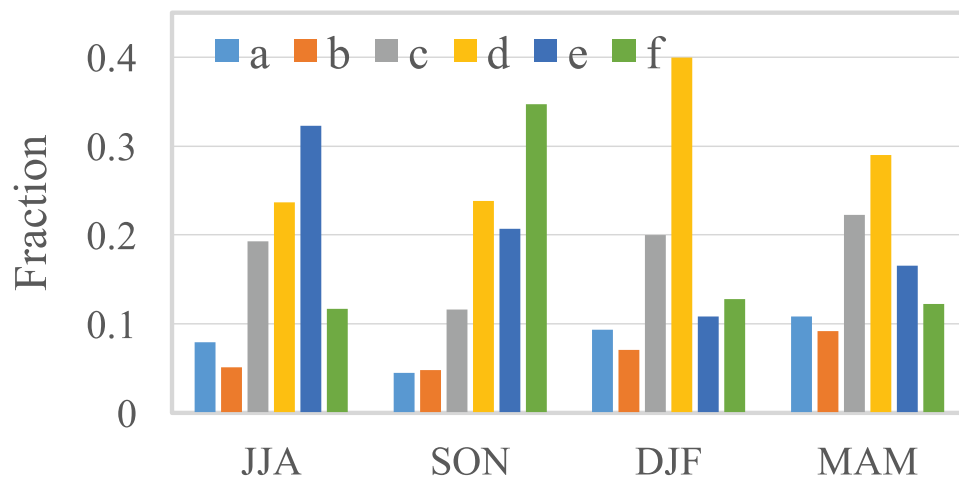


Figure 3.9: The contribution fraction from tagged boxes in JJA, SON, DJF and MAM.

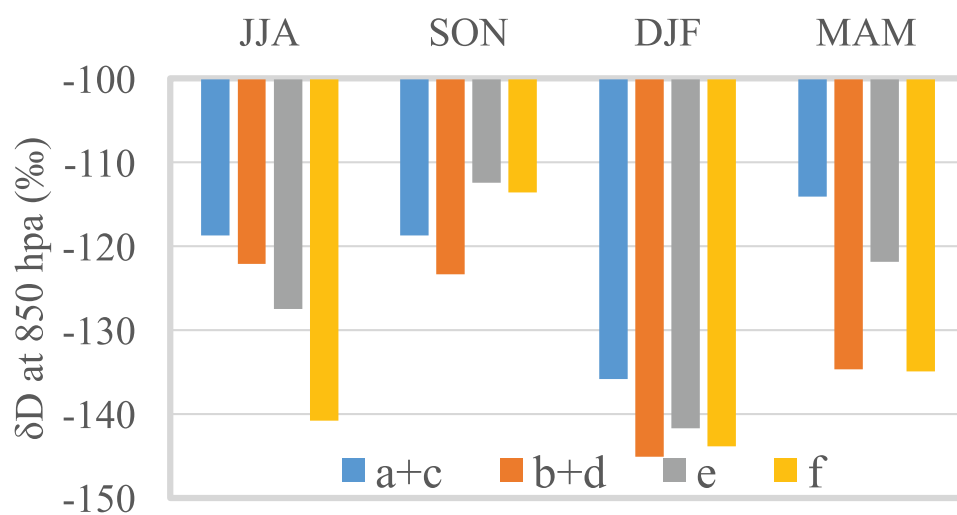


Figure 3.10: 850 hpa isotope ratios documented based on the wind direction ( $0^\circ$  to  $90^\circ$ , Area f;  $90^\circ$  to  $180^\circ$ , Area e;  $180^\circ$  to  $270^\circ$ , Area a+c;  $270^\circ$  to  $360^\circ$ , Area b+d).

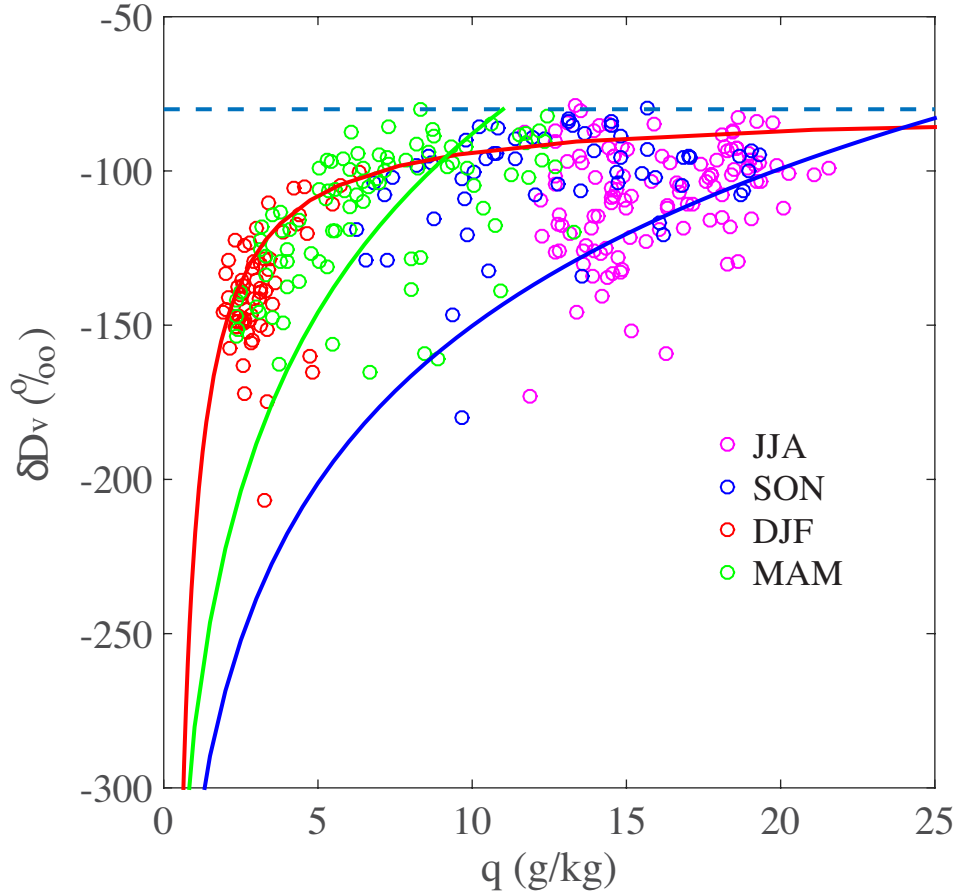


Figure 3.11: The variation of hourly water vapor  $\delta D$  as a function of  $q$  for JJA, SON, DJF and MAM. Rayleigh distillation curves representing a pseudoadiabatic process based on oceanic temperatures with an initial  $\delta D$  values of  $-80‰$  (approximate value of vapor in equilibrium with ocean, shown as blue dash line) are shown as green solid line ( $15^\circ\text{C}$ ) and blue solid line ( $25^\circ\text{C}$ ). The red line called Evaporation line represents a special case where the source water is the infinite reservoir of vapor that is in thermodynamic equilibrium (i.e., saturated) with the ocean surface.

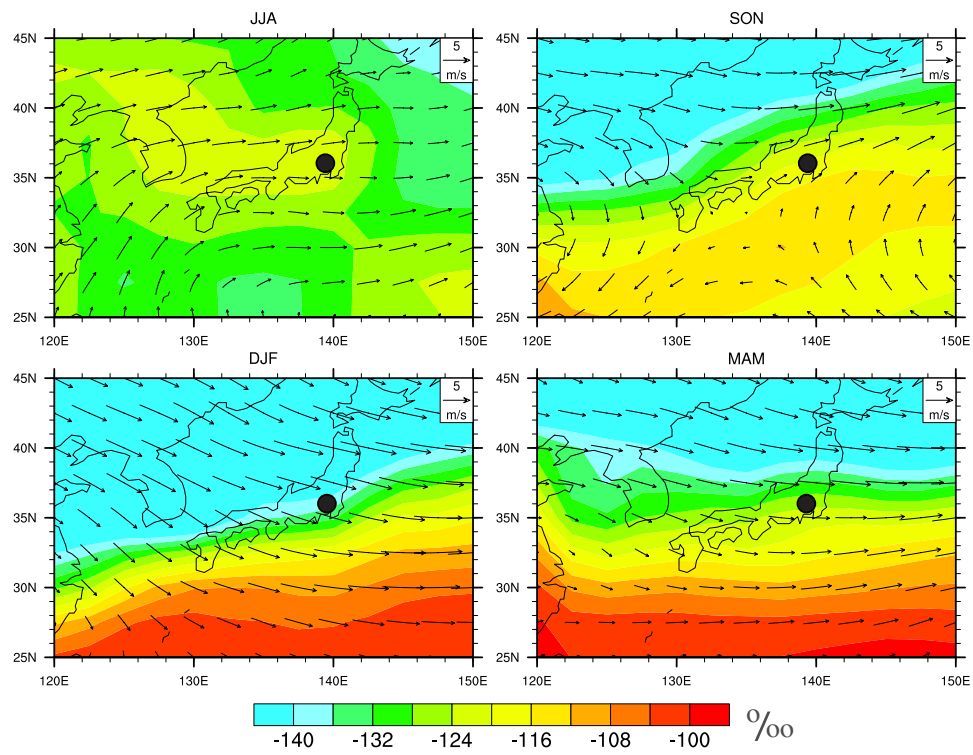


Figure 3.12: *IsoGSM* simulated the wind field and water vapor  $\delta D$  at 850 hpa.

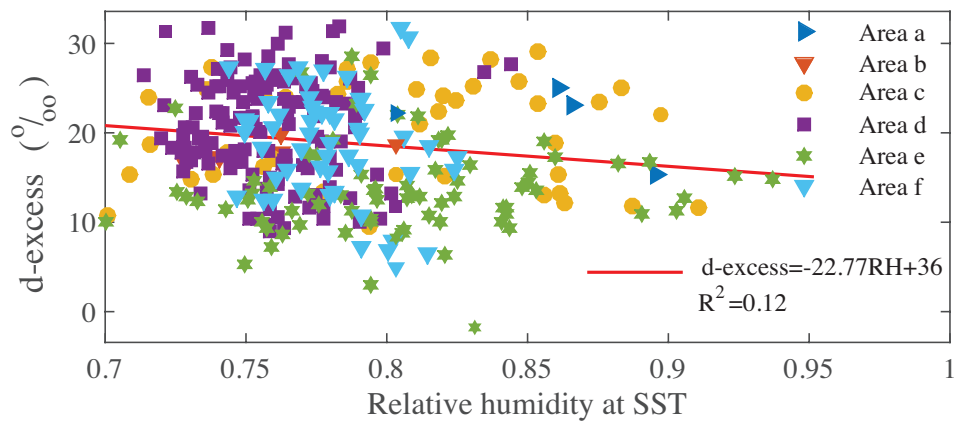


Figure 3.13: Relationship between daily  $d - excess$  and  $RH$  at  $SST$  from the source regions ( $10 \times 10$  degree). Different symbols present water vapor from different source shown in Figure 3.8.

Table 3.1: The linear correlation coefficient R and p-value (a: p-value < 0.001; b: 0.001 < p-value < 0.05 and none: p-value > 0.05) of hourly isotope ratios ( $\delta D$ ,  $\delta^{18}O$ , and  $d - excess$ ) with observed weather variables (water vapor mixing ratio  $Q$ , Relative humidity  $RH$ , surface air temperature  $T_a$ ) for all observed data in each month from June 2013 to May 2014.

| Year | Month | $\ln(Q)$          |                   |                    | $RH$               |                    |                    | $T_a$             |                   |                    |
|------|-------|-------------------|-------------------|--------------------|--------------------|--------------------|--------------------|-------------------|-------------------|--------------------|
|      |       | $\delta D$        | $\delta^{18}O$    | d-excess           | $\delta D$         | $\delta^{18}O$     | d-excess           | $\delta D$        | $\delta^{18}O$    | d - excess         |
| 2013 | 6     | 0.36 <sup>a</sup> | 0.33 <sup>a</sup> | 0.05               | -0.03              | 0.01               | -0.27 <sup>a</sup> | 0.28 <sup>a</sup> | 0.23 <sup>a</sup> | 0.25 <sup>a</sup>  |
| 2013 | 7     | 0.33 <sup>a</sup> | 0.42 <sup>a</sup> | -0.47 <sup>a</sup> | -0.14 <sup>a</sup> | -0.03              | -0.36 <sup>a</sup> | 0.35 <sup>a</sup> | 0.34 <sup>a</sup> | -0.12 <sup>b</sup> |
| 2013 | 8     | 0.41 <sup>a</sup> | 0.48 <sup>a</sup> | -0.38 <sup>a</sup> | -0.29 <sup>a</sup> | -0.19 <sup>a</sup> | -0.27 <sup>a</sup> | 0.46 <sup>a</sup> | 0.45 <sup>a</sup> | -0.08 <sup>b</sup> |
| 2013 | 9     | 0.02              | 0.24 <sup>a</sup> | -0.72 <sup>a</sup> | -0.15 <sup>a</sup> | 0.08 <sup>b</sup>  | -0.70 <sup>a</sup> | 0.10 <sup>b</sup> | 0.17 <sup>a</sup> | -0.25 <sup>a</sup> |
| 2013 | 10    | 0.44 <sup>a</sup> | 0.55 <sup>a</sup> | -0.59 <sup>a</sup> | 0.06               | 0.15 <sup>a</sup>  | -0.46 <sup>a</sup> | 0.40 <sup>a</sup> | 0.45 <sup>a</sup> | -0.29 <sup>a</sup> |
| 2013 | 12    | 0.89 <sup>a</sup> | 0.86 a            | 0.38 <sup>a</sup>  | 0.70 <sup>a</sup>  | 0.68 <sup>a</sup>  | 0.32 <sup>b</sup>  | 0.52 <sup>a</sup> | 0.51 <sup>a</sup> | 0.19               |
| 2014 | 1     | 0.49 <sup>a</sup> | 0.50 <sup>a</sup> | 0.01               | -0.04              | 0.06               | -0.42 <sup>a</sup> | 0.36 <sup>a</sup> | 0.28 <sup>a</sup> | 0.36 <sup>a</sup>  |
| 2014 | 2     | 0.43 <sup>a</sup> | 0.47 <sup>a</sup> | -0.19 <sup>a</sup> | 0.02               | 0.08 <sup>b</sup>  | -0.37 <sup>a</sup> | 0.40 <sup>a</sup> | 0.40 <sup>a</sup> | 0.05               |
| 2014 | 3     | 0.65 <sup>a</sup> | 0.68 <sup>a</sup> | -0.07              | 0.27 <sup>a</sup>  | 0.32 <sup>a</sup>  | -0.20 <sup>a</sup> | 0.41 <sup>a</sup> | 0.41 <sup>a</sup> | 0.04               |
| 2014 | 4     | 0.59 <sup>a</sup> | 0.59 <sup>a</sup> | 0.06               | 0.33 <sup>a</sup>  | 0.43 <sup>a</sup>  | -0.32 <sup>a</sup> | 0.28 <sup>a</sup> | 0.17 <sup>a</sup> | 0.43 <sup>a</sup>  |
| 2014 | 5     | 0.39 <sup>a</sup> | 0.46 <sup>a</sup> | -0.44 <sup>a</sup> | 0.21 <sup>a</sup>  | 0.38 <sup>a</sup>  | -0.79 <sup>a</sup> | 0.13 <sup>a</sup> | 0.02              | 0.41 <sup>a</sup>  |

Table 3.2: Monthly mean values of observed  $\delta D$ ,  $\delta^{18}O$ , and  $d - excess$  for atmospheric water vapor, precipitation, and the vapor in Equilibrium with precipitation in Mase, Japan. Meteorological variables (Temperature  $T_a$ , Humidity  $RH$ , Precipitation amount  $P$  and water mixing ratio  $Q$ ) are also listed. All isotope parameters were presented in ‰.

| Date    | Vapor  |                   |          | Precipitation |                   |          | Vapor in equilibrium |                   |          | RH (%) | T <sub>a</sub> (°C) | P (mm) |
|---------|--------|-------------------|----------|---------------|-------------------|----------|----------------------|-------------------|----------|--------|---------------------|--------|
|         | δD     | δ <sup>18</sup> O | d-excess | δD            | δ <sup>18</sup> O | d-excess | δD                   | δ <sup>18</sup> O | d-excess |        |                     |        |
| 2013 6  | -119.0 | -16.4             | 12.1     | -66.0         | -9.0              | 6.0      | -138.4               | -18.5             | 10.0     | 82.4   | 20.9                | 176.5  |
| 2013 7  | -104.1 | -14.4             | 11.4     | -43.0         | -7.0              | 13.3     | -113.6               | -16.3             | 16.7     | 81.5   | 24.7                | 101.9  |
| 2013 8  | -103.6 | -14.4             | 11.6     | -56.4         | -8.5              | 11.3     | -124.4               | -17.5             | 16.0     | 79.5   | 26.6                | 102.0  |
| 2013 9  | -98.2  | -14.3             | 16.5     | -25.5         | -4.6              | 11.7     | -99.2                | -14.1             | 13.4     | 78.8   | 22.7                | 214.9  |
| 2013 10 | -107.1 | -15.7             | 18.6     | -26.4         | -4.5              | 9.9      | -105.0               | -14.4             | 10.1     | 81.5   | 17.6                | 432.9  |
| 2013 11 | -      | -                 | -        | -16.69        | -2.27             | 1.47     | -104.92              | -12.90            | -1.70    | 75.0   | 9.3                 | 18.6   |
| 2013 12 | -138.5 | -20.5             | 25.8     | -76.3         | -10.6             | 8.9      | -164.7               | -21.7             | 8.7      | 69.9   | 4.2                 | 53.0   |
| 2014 1  | -139.1 | -20.8             | 27.3     | -41.2         | -7.2              | 16.3     | -135.3               | -18.5             | 12.4     | 63.0   | 2.2                 | 26.6   |
| 2014 2  | -139.6 | -20.5             | 24.2     | -25.4         | -4.2              | 7.8      | -120.2               | -15.4             | 2.9      | 66.9   | 3.0                 | 103.7  |
| 2014 3  | -126.6 | -18.8             | 23.4     | -2.6          | -1.5              | 9.0      | -94.1                | -12.3             | 4.0      | 64.3   | 7.5                 | 93.0   |
| 2014 4  | -108.7 | -16.3             | 21.5     | -21.2         | -4.0              | 10.9     | -105.6               | -14.3             | 9.0      | 64.5   | 12.4                | 157.3  |
| 2014 5  | -107.2 | -15.9             | 19.7     | -43.6         | -5.8              | 3.1      | -120.1               | -15.6             | 4.9      | 72.5   | 18.3                | 118.3  |

Table 3.3: The contribution of evapotranspiration ( $F_{ET}$ ) to that of the *ABL* water vapor estimated from monthly water vapor  $\delta D$ , background isotope ratio  $\delta_{bg}$  and evapotranspiration isotope ratio  $\delta_{ET}$ . Data from rainy days, data of latent heat  $LE < 50 \text{ W/m}^2$  and daily  $F_{ET} < 0$  data were excluded. All isotope parameters presented in ‰.

| Year | Month | $\delta D_v$ | $\delta_{bg}$ | $\delta_{ET}$ | $F_{ET}$          |
|------|-------|--------------|---------------|---------------|-------------------|
| 2013 | 6     | -99.6        | -109.7        | -56.9         | $19.2 \pm 14.0\%$ |
| 2013 | 7     | -96.3        | -111.1        | -45.7         | $22.7 \pm 13.6\%$ |
| 2013 | 8     | -97.8        | -110.3        | -46.8         | $19.7 \pm 11.1\%$ |
| 2013 | 9     | -92.8        | -107.4        | -31.0         | $19.1 \pm 12.5\%$ |
| 2013 | 10    | -89.7        | -94.6         | -36.3         | $8.4 \pm 10.6\%$  |
| 2013 | 11    | -            | -             | -             | -                 |
| 2013 | 12    | -            | -             | -             | -                 |
| 2014 | 1     | -148.7       | -149.7        | -91.7         | $0.2 \pm 13.1\%$  |
| 2014 | 2     | -124.3       | -143.6        | 9.7           | $12.6 \pm 13.4\%$ |
| 2014 | 3     | -115.2       | -123.8        | -49.5         | $11.6 \pm 8.3\%$  |
| 2014 | 4     | -106.4       | -124.5        | -51.8         | $24.9 \pm 12.7\%$ |
| 2014 | 5     | -117.2       | -131.1        | -62.5         | $20.2 \pm 14.2\%$ |

# Chapter 4

## *ET* partitioning over a paddy field

### 4.1 Estimation of $\delta_{ET}$ , $\delta_T$ , and $\delta_E$

Hourly variations in  $\delta_{ET}$  were estimated using the Keeling plot method. We selected hourly data whenever there were sufficient data points ( $N > 40$ ) with a high coefficient of determination ( $R^2 > 0.8$ ) for each linear regression, as shown in Figure 4.1. Several difficulties with the Keeling plot approach were found. Some of the calculation results did not satisfy the threshold of  $R^2 > 0.8$ . As a result, only 24% of the observed period could be used in our study. The results were divided into daytime (8:00–17:00) and nighttime (18:00–7:00). Daytime observations in the whole growing season were further divided into two periods, namely early (May to June) and late growing seasons (July to August). In the early growing season,  $\delta_{ET}$  gradually increased with time because the dominance of evaporation resulted in significant heavy isotope enrichment of residual waters. In the late growing season,  $\delta_{ET}$  mainly consisted of transpiration and was closely coupled with surface water. A possible reason was that isotopic fractionation through transpiration was much smaller than evaporation. It can be seen from Figure 4.1 that ambiguous changes in  $\delta_{ET}$  during nighttime were frequently observed. Due to the weak nighttime turbulence conditions, the intercept parameter of the Keeling plot (even  $R^2 > 0.8$ ) was generally an ambiguous quantity when the change in specific humidity was insufficient [Lee *et al.*, 2006].

This suggests that improving the estimation of  $\delta_{ET}$  is an important objective in future studies, especially under low-evapotranspiration conditions, such as those experienced in semi-arid and arid areas. Due to the unreliable  $\delta_{ET}$  at nighttime, we only considered daytime data in this study.

The water used for irrigation in the field was derived primarily from large storage reservoirs. The irrigation water used in the field had a stable isotope composition ( $\delta D$ ) of approximately  $-47.46 \pm 6.50\text{‰}$  and was relatively constant throughout the whole growing season. We used an average value for all sampling points due to the uncertainties induced by the uneven distribution of surface water isotope ratios. Figure 4.2 demonstrates that the spatially averaged pond water  $\delta D$  showed a general decreasing trend from May to June with considerable variability and then remained constant after June. The surface water was evaporating constantly in May and June, but then, there was a gradual shift to a transpiration loss. The  $\delta D$  of surface water decreased from  $-21.75\text{‰}$  (May) to  $-33.22\text{‰}$  (June) due to a change in the source of the water loss (shift from  $E$  to  $T$ ) with large variability during this period. As the rice crop emerged and dense stands of plants were established after June, water in the ponded fields became less enriched in  $\delta D$  ( $-45.30\text{‰}$ ). Large standard deviation in each measurement day in the early growing season indicated a significantly uneven spatial distribution of surface water isotope ratios (more enriched in outflow site because water evaporated for a longer time period during its flow from the inflow to outflow sites). In the late growing season, however, the dominance of transpiration might result in isotope ratios that were similar to those in the irrigation water (small deviation).

It was assumed that the  $\delta D$  value of transpiration was the same as that of collected surface water in this study. However, the  $NSS$  might increase the uncertainty in our estimation. We therefore selected data of mid-day (11:00–15:00) to guarantee the  $SSA$  satisfied. We assumed the rice plant can reach a  $SSA$  state in this time period because

1) We compared diurnal variations in  $\delta_{ET}$  in the late growing season (July and August in 2013) and the non-growing season (September and October in 2013) that were shown

in Figure 4.3. During the late growing season, daytime  $\delta_{ET}$  (8:00–17:00) did not vary significantly and was similar to that of the surface water ( $-43.15\text{‰}$  for  $\delta_{ET}$  and  $-45.30\text{‰}$  for surface water on average), implying little isotopic fractionation during the water-exchange process, which supported the *SSA*. Compared with that in the late growing season, the variations in the diurnal  $\delta_{ET}$  had larger amplitude due to the lack of transpiration after harvest. This demonstrated that the assumption of an isotopic steady state might be satisfied in the daytime. 2) Many studies found the *SSA* is satisfied in midday time [Lai *et al.*, 2006, 2008] because the magnitude of transpiration related to the volume of leaf water is large during mid-day. 3) The *SSA* is satisfied when the ratio of transpiration to leaf water volume is small (short turnover time of leaf water), which has been shown in shorter canopies [Yakir and Wang, 1996; Lai *et al.*, 2006, 2008]. Furthermore, Lai *et al.* [2008] showed that turnover time of leaf water is shorter under wet condition. On the other hand, based on the measured surface water and water vapor isotopes, the hourly  $\delta_E$  could be estimated using the Craig and Gordon Equation (see sect. 2.2.3).

## 4.2 *ET* partitioning and its environmental control

We partitioned evapotranspiration on a daily averaged basis for a full growing season because the surface water measurements were temporally too coarse to provide a realistic description at a smaller scale, i.e., a diurnal time scale. Those sub-data sets that did not satisfy the criteria were removed from the near-continuous data set for a full growing season. To provide an accurate and representative result, the criteria for the data removal was (i) data measured on a rainy day were rejected, (ii) data measured outside the mid-day (11:00–15:00) period were rejected to avoid possible uncertainty due to *NSS*, and (iii) data acquired with a low coefficient of determination for each linear regression in the Keeling plot method ( $R^2 < 0.8$ ) were excluded to maintain a high-quality  $\delta_{ET}$  estimation. Since 72% water evapotranspired during the selected time period, it would not make significant bias in long-duration *ET* partitioning. The selected data was averaged on

a daily timescale, and was plotted in Figure 4.4. Isotopic ratios varied from -83.48 to -14.64‰ for  $\delta_{ET}$ , -48.58 to -4.63‰ for  $\delta_T$ , and -422.81 to -49.33‰ for  $\delta_E$ .  $\delta_E$ ,  $\delta_T$ , and  $\delta_{ET}$  were similar during the early growing season;  $\delta_{ET}$  and  $\delta_T$  were less negative than  $\delta_E$  and very similar during the late growing season; and  $\delta_E$  displayed a decreasing trend during the full growing season. The mean contribution of transpiration to evapotranspiration estimated by Equation 2.5 was shown in Figure 4.5 as a function of time. The  $T/ET$  ranged from 0.2 to 1 and increased almost continuously in May and June, but then became relatively constant, with a value close to 1. The total uncertainty estimated by Equation 6 (also see sect. 4.1) and shown as a vertical bar in Figure 4.5, ranged from 0 to 0.8. Unexpectedly,  $T/ET$  on some days was over 1 or less than 0. This can be explained by the coarse resolution of surface water sampling, neglect of the leaf interception evaporation, and the measurement errors in  $\delta_{ET}$  (see sect. 4.1). We excluded unreliable data ( $T/ET > 1.2$  and  $T/ET < -0.2$ ; about 7% of the data used in Figure 4.5 were excluded) because they were clearly inaccurate and differed from the surrounding  $T/ET$  results. Because both underestimation and overestimation occurred, some of the errors tended to cancel out through averaging, and we therefore did not exclude data with an acceptable error ( $1 < T/ET < 1.2$  or  $-0.2 < T/ET < 0$ ). The monthly  $T/ET$  values during the growing season were 0.47, 0.78, 0.98 and 0.94 from May to August (Figure 4.5), respectively. The monthly  $T/ET$  variation can be seen in Figure 4.5 to be in the same phase as the variation of  $LAI$  in Figure 2.2. The averaged daily change in  $T/ET$  was represented quite well as a function of a 0.5-bin averaged  $LAI$  ( $FT = 0.67LAI^{0.25}$ ,  $R^2 = 0.80$ ), implying that vegetation plays a major role in driving the contribution of  $E$  and  $T$ .  $E$  played a large role in the early growing season while  $T$  dominated the later stage (Figure 4.7). Furthermore, it is suggested that a crop parameter such as  $LAI$  can be an effective alternative for the estimation of  $T/ET$  over a paddy field.

### 4.3 The error analysis

The results demonstrated the robustness of using isotope measurements for partitioning evapotranspiration. However, there were some uncertainties in our estimation. Potential uncertainty derived from neglecting the leaf interception evaporation in Equation 2.7 should have been taken into account, because the rainfall events occurred frequently during the observation period (Figure 4.2). The neglect of interception could be the cause that  $T/ET$  is higher than 1 during the late growing season if the interception evaporation isotope ratio is lighter than  $\delta_E$ . Unfortunately, the interception losses are difficult to estimate in general. As shown in Figure 4.5, the  $T/ET$  uncertainty is large in some data used in our estimation. This uncertainty in the results of this single-isotope two-source mixing model is directly related to the uncertainty in all three of the terms:  $\delta_{ET}$ ,  $\delta_T$ , and  $\delta_E$ . The uncertainty therefore was estimated by Equation 2.8. It was assumed that the values of  $\sigma_{\delta_{ET}}$  and  $\sigma_{\delta_E}$  are the standard deviations of  $\delta_{ET}$  and  $\delta_E$  for each day, and the value of  $\sigma_{\delta_T}$  is the standard deviation of  $\delta_T$  of the spatial distribution on each measurement day. In addition to excluding the days where  $T/ET$  was higher than 1.2 or less than -0.2, we further excluded days where only one measurement of  $\delta_T$  and less than 3 hours' highly accurate  $\delta_{ET}$  estimation were available for each day. The average uncertainty in  $\delta_{ET}$  was about 9.06‰. This uncertainty was smaller than the average uncertainty in  $\delta_E$  (14.27‰), but larger than  $\delta_T$  (5.79‰). The average uncertainty in  $T/ET$  was estimated as  $14 \pm 10\%$ . As shown in Figure 4.6, the uncertainty of  $\delta_E$  was less important than that of the other two parameters throughout the study period, although a high average uncertainty value was obtained. The  $ET$  partitioning uncertainty was derived from the uneven spatial distribution of surface water isotopes ( $\delta_T$ ), with levels generally decreasing with increased  $T/ET$ . As  $T/ET$  increased, the impacts of the Keeling plot  $\delta_{ET}$  uncertainty become much larger than that of  $\delta_T$  and  $\delta_E$ . This result is consistent with  $\delta_T$  measurements. During the early growing season,  $\delta_T$  was quite variable, whereas  $\delta_{ET}$  was more poorly quantified than  $\delta_T$  during the late growing season (Figure 4.4). This suggests that further improvement in the estimation of  $\delta_T$  and  $\delta_{ET}$  is necessary for partitioning evapotranspiration using the

isotopic method. It is noted that about 80% of the data have  $T/ET$  values above 0.75 because rice is a fast-growing plant (Figure 2.2). To partition evapotranspiration under low  $LAI$  conditions, more observations are necessary.

The uncertainty analysis mentioned above is a statistical analysis based on the assumption that source and end members are independently measured. The inherent errors in the estimation methods of  $\delta_E$ ,  $\delta_T$ , and  $\delta_{ET}$  should also be considered. In the estimation of  $\delta_{ET}$ , the Keeling plot approach is robust when combined with high-temporal-resolution data [Good *et al.*, 2012]. However, that only 24% data ( $R^2 > 0.8$ ) can be used in our study suggested the assumptions underlying the Keeling plot method are rarely met perfectly in nature. Even a high  $R^2$  value can be obtained, the error in  $\delta_{ET}$  estimation in some cases can still be quite large (e.g., under weak latent heat conditions). For example, Lee *et al.* [2006] reported that if the change in the range of the water mixing ratios was inconspicuous, the error would increase dramatically. To understand the uncertainty associated with the measured water vapor mixing ratio and isotopic composition, uncertainty assessments following Good *et al.* [2012] (Equation 16) were performed on 1-hr estimation of  $\delta_{ET}$ . A significant uncertainty of  $\delta_{ET}$  was found ( $22.05 \pm 12.14\%$ , excluding the data (41%) with uncertainty larger than 50%). If we consider this uncertainty in  $\sigma_{\delta_{ET}}$ , the average uncertainty in  $T/ET$  would increase to  $24 \pm 15\%$  and 88% of this uncertainty is attributed from  $\sigma_{\delta_{ET}}$ . However, we believe the bias in  $\delta_{ET}$  may be much smaller and may be cancelled out through averaging because we did not find a large variability of daily averaged  $\delta_{ET}$  in the late growing season (Figure 4.4). We also found the uncertainty of  $\delta_{ET}$  showed a similar variation with  $stdQ$  (Figure 4.8). The highest uncertainty can be found in mid-night and the lowest uncertainty of  $\delta_{ET}$  can be found in mid-day. The result is that the turbulent is weakest at midnight and resulted a high uncertainty. We can found the standard deviation of water vapor mixing ratios in lower in nighttime. We confirmed that if a change in the range of the water mixing ratios were inconspicuous, the error would increase dramatically, which is suggested by Lee *et al.* [2006]. Therefore the reason of the lowest  $\sigma_{\delta_{ET}}$  values during the 10-15 h window is that the uncertainty is

lowest during this time scale. On the other hand, we find when changes in the range of the water mixing ratios were inconspicuous, most of time  $\sigma_{\delta_{ET}}$  tended to have a positive value. A similar trend has been observed by *Welp et al.* [2008]. The reason is still not clear. A further study thus should be conducted.

In addition to the uneven distribution of surface water isotopes mentioned in section 4.3, measurements of the time resolution of surface water isotopes should also be considered. Our measurements of the isotopic composition of transpiration were obtained through direct measurement of surface water and were assumed to be constant until the next measurement. Although the surface water isotope levels were consistent throughout the late growing season (since transpiration does not enrich the residual water), they changed significantly in the early growing season (Figure 4.2). For example, surface water  $\delta_D$  in June 2 was much higher than that measured in May 29, 2014. During the early growing season, the residual water isotope was sensitive not only to plant cover but also to water amount (data is not available) and evaporation rate in the field. Assume the same  $T/ET$  in these two days, a higher daily averaged  $ET$  in June 2, 2014 ( $430 \text{ W/m}^2$ ) than in May 29, 2014 ( $298 \text{ W/m}^2$ ) might result in a more enrich surface water  $\delta D$  in June 2, 2014. To reduce the uncertainty, we therefore increased the surface water observation frequency in May and June 2014 (from 1.5 days per week in 2013 to 3 days per week in 2014). Furthermore, although the input water seemed to have a stable isotope ratio, the surface water sometimes received precipitation between two surface water measurements, resulting in a significant bias in the estimation of  $\delta_T$ . For example, input from precipitation may play a significant role in determining surface water isotope during May and June 2013 (Figure 4.2). An additional observation has been done to understand the impact of precipitation, in which we observed the surface water  $\delta D$  in daily basis from August 11 to August 16, 2014. During the observation period, two precipitation events were observed (9.7 mm, 16:00–23:00 in August 12 and 3.9 mm, 16:00–17:00 in August 14). We did not find a significant change of surface water  $\delta D$  in the day after precipitation ( $-48.18 \pm 0.97\%$  for the whole observation period). The possible reason was that

the pond water was refreshed by abundant input water and then showed a similar  $\delta D$  value with input water  $\delta D$ . Although this result suggests that rainfall does not have a significant influence on surface water isotope, it should be noted that this test was under a short period where only 2 small rainfall events occurred. The cause of changes in isotopic values in surface water may be confounded by the storm size, precipitation isotope value, surface water amount and so on. Nevertheless, frequent precise isotopic measurements of surface water are necessary especially during the early growing season. Further uncertainty derived from *SSA* assumption should also be considered. We did not measure the  $\delta_T$  directly but assumed  $\delta_T$  equal to surface water  $\delta D$ . The surface water might be enriched by evaporative enrichment compared to the root absorbed water from soil. However, we believe it would not induce a large error because of the well mixing of surface water and soil water under the flooded conditions. The soil water (Soil Water Sampler, DIK-8392, Japan) and surface water sampled on April 9, 2014 (the second day after a heavy rain, the field was flooded) showed a similar  $\delta D$  (-33.22‰ for soil water and -31.55‰ for surface water, without vegetation and irrigation). The *NSS* was neglectable in our long-duration  $T/ET$  estimation. Even given a high uncertainty raised by *NSS* ( $\pm 10\%$ ) in  $\delta_T$ , it would not make a significant bias in our long-duration  $T/ET$  estimation (averaged  $T/ET$  uncertainty was estimated as  $16 \pm 8.41\%$ ). For most of the time,  $T/ET$  was sensitive to  $\delta_E$  rather than  $\delta_T$  and  $\delta_{ET}$  in Equation 2.7, because  $\delta_E$  was significantly lower than  $\delta_T$  and  $\delta_{ET}$  (Figure 4.4). On the other hand, under low  $T/ET$  condition or on diurnal time scale, the *NSS* should be considered seriously. Nevertheless, a further study should be conducted on *NSS* impact on  $T/ET$  estimation. As shown in Equation 2.5,  $\delta_E$  is controlled by several parameters. The uncertainty can either come from measurement errors or parameter selections. Measurement errors such as the uncertainty in surface water isotopes can significantly affect the accuracy of  $\delta_E$  estimation. In addition to measurement errors, the choice of the kinetic fractionation factor  $\varepsilon_K$  is also important. In this study,  $D_i/D$  was taken as 0.9839 from *Cappa et al.* [2003], and  $n = 0.67$  was taken from *Kim and Lee* [2011] in Equation 2.6. Furthermore, values of  $D_i/D = 0.9755$ , and  $n$

$= 1$  are also widely used [Gat, 1996]. We tested the influence of these values on the  $T/ET$ , and the results were shown in Figure 4.9. Although there is no obvious difference in the high- $T/ET$  period, the influence of this selection cannot be neglected in the low- $T/ET$  period. Unfortunately, there is still no agreement on the selection of  $\varepsilon_K$ . Different values of  $\varepsilon_K$  are still in use for the purpose of  $\delta_E$  estimation [e.g., Braud *et al.*, 2005, 2009a,b; Dubbert *et al.*, 2013, 2014b; Mathieu and Bariac, 1996; Rothfuss *et al.*, 2010], although this value may significantly affect  $ET$  partitioning from a vegetative surface with a low  $LAI$ . Generally, it was less important than  $\delta_T$  and  $\delta_{ET}$  in our study because the  $LAI$  was relatively high most of the time ( $LAI > 1$ ).

In summary, to reduce the uncertainty in evaluating  $T/ET$ , it was necessary in our study to concentrate on the estimation of two parameters:  $\delta_T$  and  $\delta_{ET}$ . 1) With regard to  $\delta_T$ , recent improvements have occurred in the temporal resolution at which surface/soil water samples can be collected and analyzed [Pangle *et al.*, 2013]. Incorporating these with a high frequency of water vapor isotope measurements will offer an extremely precise and quantitative insight into the partitioning of evapotranspiration. Moreover, the  $NSS$  should also be conducted in further study especially at the diurnal time scale. Recent studies showed that  $T/ET$  at diurnal time scale is significantly affected by non-steady state of transpiration [e.g., Dubbert *et al.*, 2013, 2014a,b]. 2) With respect to  $\delta_{ET}$ , we suggest that dual-level measurements of isotopes should be made at the same time. Estimating  $\delta_{ET}$  using the flux gradient relationship [e.g., He and Smith, 1999] has been found to decrease the uncertainty of this estimation [Good *et al.*, 2012].

## 4.4 Comparison with other studies

Several studies based on indirect methods such as the water isotope balance, or model simulations have been done because no direct measurements are available for partitioning evapotranspiration over a paddy field [e.g., Kondo and Watanabe, 1992; Maruyama and Kuwagata, 2010; Simpson *et al.*, 1992]. For example, Simpson *et al.* [1992] found that tran-

spiration accounted for about 60% of total losses to the atmosphere using a stable isotope mass balance based on a water budget (i.e. irrigation inflow + precipitation = evaporation + transpiration + infiltration). However, it was based on coarse measurements and without the consideration of the outflow and groundwater recharge in the water balance. This may result in a significant error in the water budget. By coupling land-surface and crop growth models, *Maruyama and Kuwagata* [2010] suggested that  $T/ET$  was about 45% during the rice growth period. The average  $T/ET$  from our results (80%) was higher than that reported in these studies. To compare with our  $ET$  partitioning result, a crop growth model [*Maruyama and Kuwagata*, 2010] coupled two-source model was developed (Detail can be found in Appendix.). The monthly averaged  $T/ET$  estimated from simulated  $T$  and measured  $ET$  during the growing season was similar to isotope-based estimation (0.24, 0.86, 0.93 and 0.84 from May to August (Figure 4.5), respectively). In daily time scale, high variability could be found in later growing season. This high variability might be attributable to the insufficient consideration and large uncertainty of parameters such as canopy resistance, which were not measured and validated. It highlights the simplicity and versatility of isotope-based method in long-duration  $T/ET$  partitioning. The parameters in this method can be easily determined by water vapor and soil isotope measurements.  $\delta_E$  was estimated from the Craig and Gordon Equation and  $\delta_{ET}$  can be determined by the Keeling plot. Over days to weeks timescales,  $\delta_T$  can be assumed as soil water isotope ratios, because mass balance requires the isotope composition of the source water be equal to that transpired back to the atmosphere [*Griffis et al.*, 2011]. Compared to isotope-based methods, the simulation is difficult to apply to different vegetation cover since it is highly affected many factors such as radiation, air temperature, air humidity, wind, soil water content, crop characteristic, etc (also see Appendix). For cultivated land, the crop development stage, environment, irrigation practice, and crop management also strongly influence the transpiration rate [*Sutanto et al.*, 2014]. Compared to direct measurements of  $E$  and  $T$ , the problem of scaling up from leaf-level to canopy-scale in hydrometric methods diminishes when isotope-based is applied. On the

other hand, as mentioned above, it is still a great challenge in isotope-based study to determine the isotope ratios of  $ET$ ,  $T$  and  $E$ .

In practice, it is difficult to partition  $ET$  at regional or global scale because of the lack of detailed data [Hu *et al.*, 2009]. However, we demonstrated that  $T/ET$  could be well described by a function of  $LAI$ . This suggests that  $LAI$  could be used to partition  $ET$  in spatial studies, as  $LAI$  can be easily obtained through both in situ observations and remote sensing techniques. Many previous studies identified a relationship between  $T/ET$  and  $LAI$  [e.g., Hu *et al.*, 2009; Wang *et al.*, 2010]. Recently, Wang *et al.* [2014] reviewed the published studies (48 individual publications) that measured at least two of the three components ( $E$ ,  $T$ , and  $ET$ ) and  $LAI$  simultaneously in an attempt to establish a quantitative relationship between  $ET$  partitioning and  $LAI$  for different systems (i.e., agricultural versus natural systems) and to explain the variations in observed  $T/ET$  at the global scale. Nonlinear relationships between  $T/ET$  and  $LAI$  were identified for both agricultural ( $T/ET = 0.91LAI^{0.07}$ ) and natural ( $T/ET = 0.77LAI^{0.10}$ ) data subsets, suggesting the feasibility of using  $LAI$  information to generate a global-scale  $T/ET$  data set. Our results generally agree with the global scale nonlinear relationships in Wang *et al.* [2014], but tend to have a slightly lower proportion of transpiration under low- $LAI$  conditions (Figure 4.7). Estimations at both the global scale and local scale (this study) show that even under low- $LAI$  conditions (e.g.,  $LAI = 0.5$ ),  $T/ET$  could have a very high value (0.72 and 0.90 for natural and agricultural systems in global estimations, respectively, and 0.58 in our study). Wang *et al.* [2014] suggested the possibility of a high proportion of water use by vegetation even under low- $LAI$  conditions. However, as mentioned in section 3.1, the error under low- $LAI$  conditions may also lead to a significant uncertainty with respect to  $T/ET$ . Nevertheless,  $LAI$  could be a useful tool to understand the contribution of transpiration water fluxes to continental evapotranspiration and to reduce the simulation uncertainty in water recycling circulation, which has been demonstrated in previous studies [e.g., Hu *et al.*, 2009; Wang *et al.*, 2010; Wang and Yamanaka, 2014].

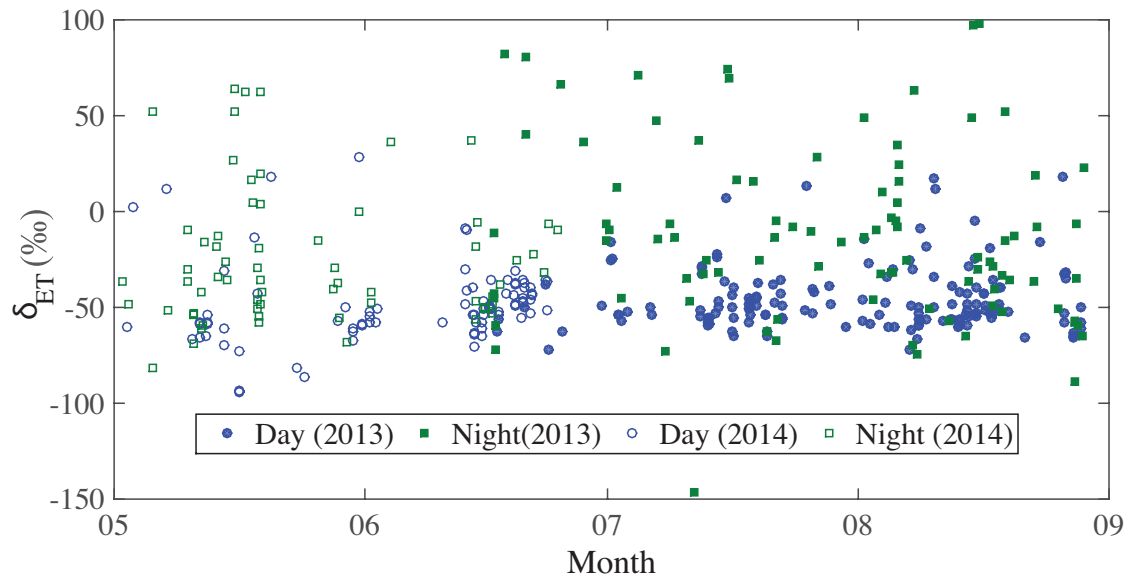


Figure 4.1: Hourly variation in the isotope ratios of  $ET$  from June to August 2013 and May to June 2014. Blue circles (filled, 2013; un-filled, 2014) indicate daytime data (8:00–17:00), and green squares (filled, 2013; un-filled, 2014). indicate nighttime data.

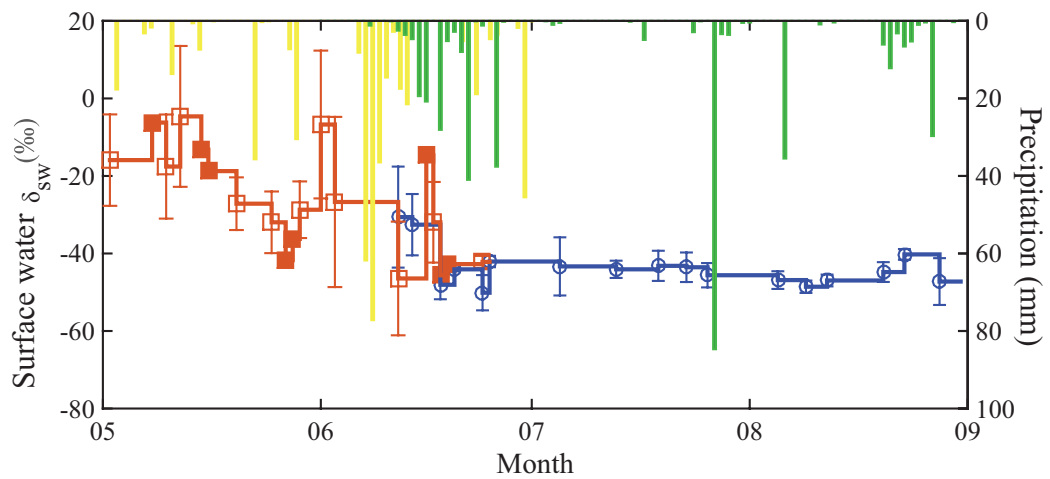


Figure 4.2: Observed  $\delta D$  in surface water from June to August 2013 (blue line) and May to June 2014 (red line). Filled squares with error bars indicate the spatial average values and their standard deviations. Unfilled squares represent the measurement of only one point in 2014. Vertical bars denote daily precipitation amount from June to August 2013 (green bar) and May to June 2014 (yellow bar).

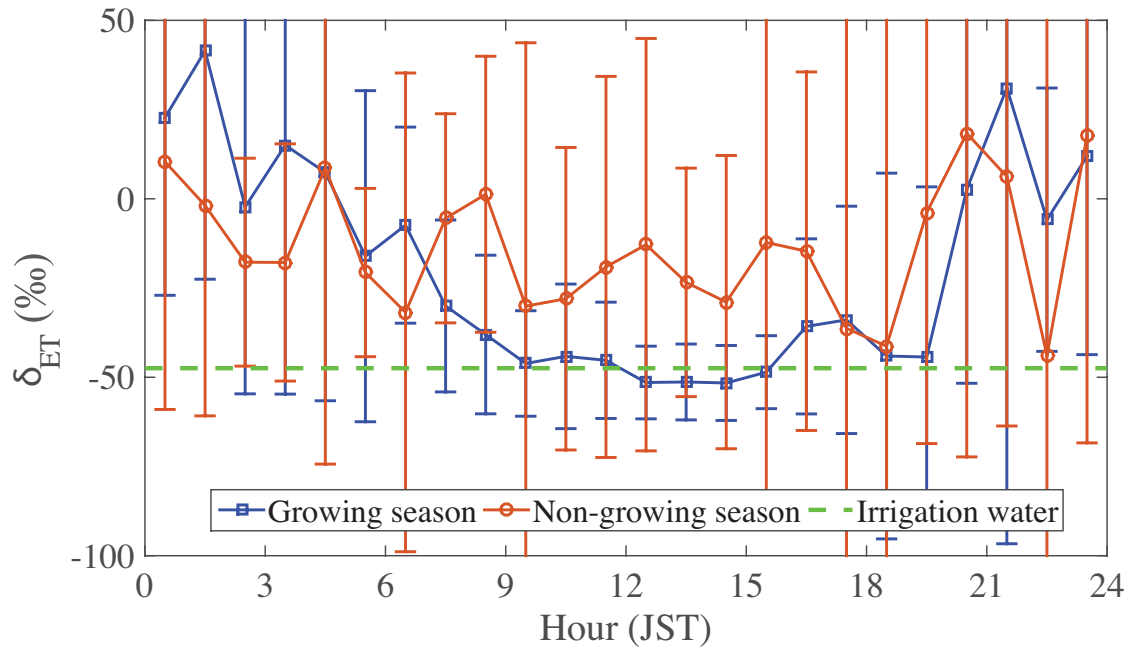


Figure 4.3: Comparison of the ensemble mean diurnal variation in the evapotranspiration  $\delta D$  signal during the growing season (July and August 2013, blue squares) and non-growing season (September and October 2013, red circles). Vertical bars denote the standard deviation for each hour. Green dash line indicates the mean irrigation water  $\delta D$  ( $-47.46\text{‰}$ ).

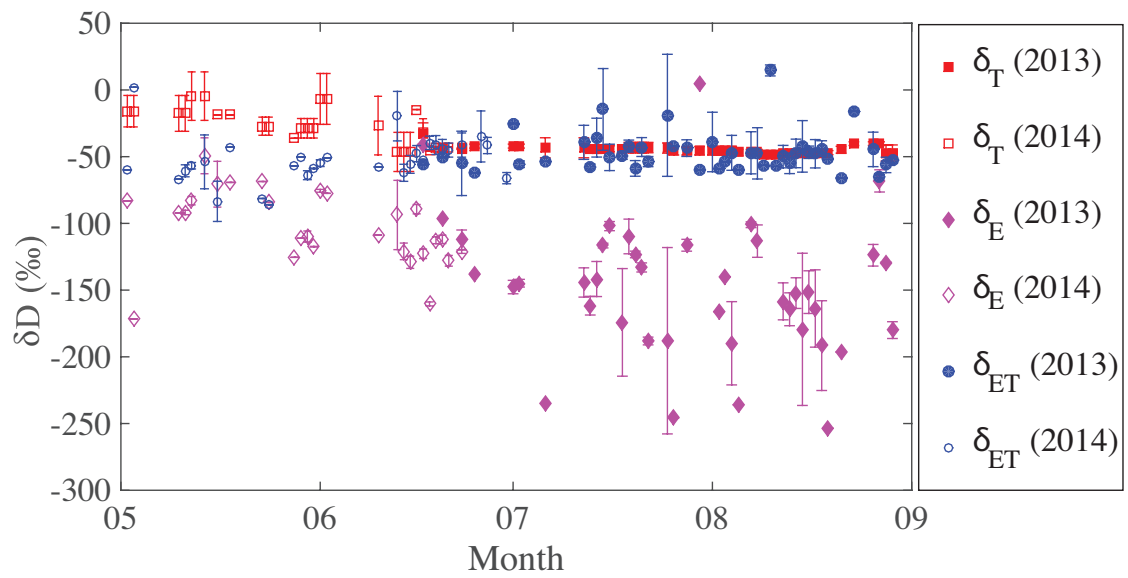


Figure 4.4: Daily averaged variation in isotope ratios of  $ET$  (blue circles),  $T$  (red squares), and  $E$  (pink diamonds). Two points of  $\delta_E$ , which were less than  $-300\text{‰}$ , are not shown in this figure.

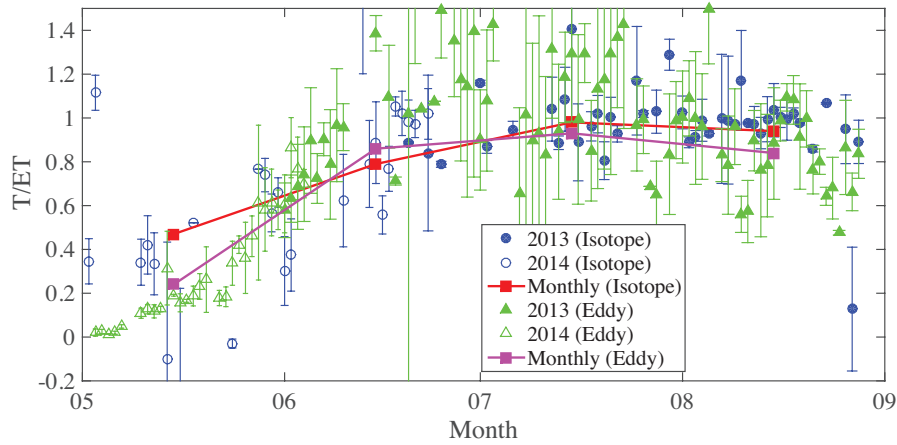


Figure 4.5: Transpiration fraction determined by the isotope-based method and eddy correlation based method. Blue circles represent isotope-based daily averaged  $T/ET$ . Green triangles represent the simulated  $T$  and measured  $ET$  based daily averaged  $T/ET$ . Vertical bars denote the standard deviation for each day. The red line with filled squares represents the isotope based monthly average  $T/ET$  excluding unreliable values ( $T/ET > 1.2$  or  $T/ET < -0.2$ ). Pink line with filled squares represents the simulated  $T$  and measured  $ET$  based monthly averaged  $T/ET$ .

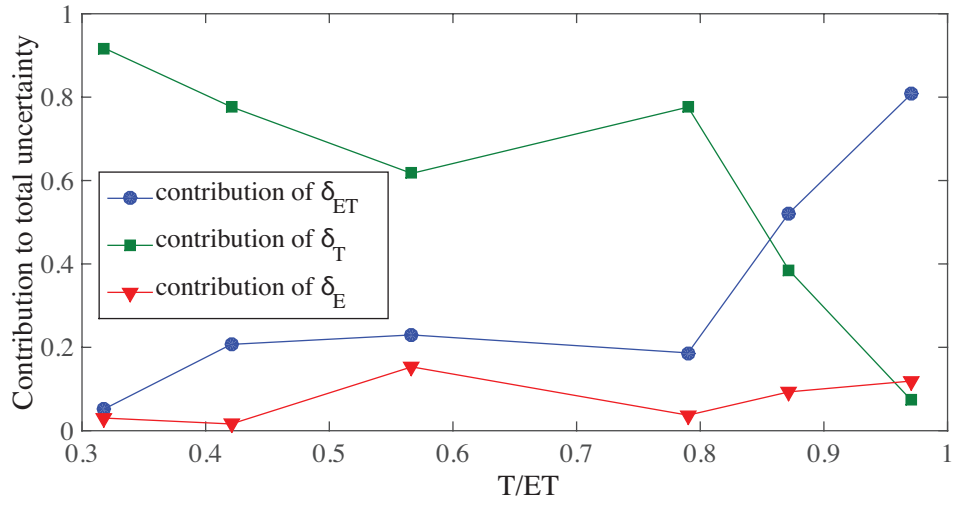


Figure 4.6: The fractional contribution to the partitioning uncertainty of uncertainty  $\delta_{ET}$  (blue circle) and end-member uncertainty ( $\delta_T$  (green square) and  $\delta_E$  (red diamond)) as a function of the transpired fraction of evapotranspiration,  $T/ET$ . The fractional contribution was calculated from Equation 2.8. Data where  $T/ET > 1.2$  or  $T/ET < -0.2$ , with only one point measurement of  $\delta_T$  and with less than 3 hours of highly accurate  $\delta_{ET}$  estimation in each day excluded.

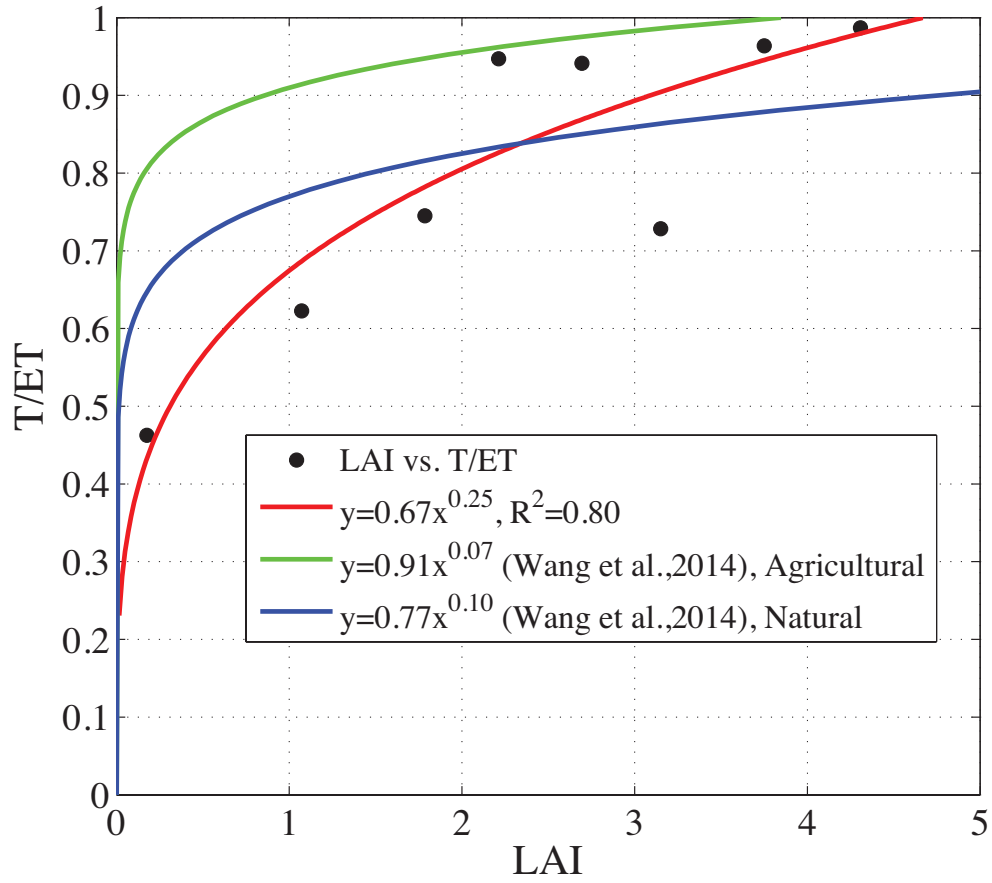


Figure 4.7: Relationship between the 0.5–bin averaged  $LAI$  and the daily averaged transpiration fraction ( $T/ET$ ).  $T/ET > 1.2$  or  $T/ET < -0.2$  data are excluded. Two regression lines (green, agricultural system; blue, natural system) derived from a global observation data set Wang *et al.* [2014] are also plotted.

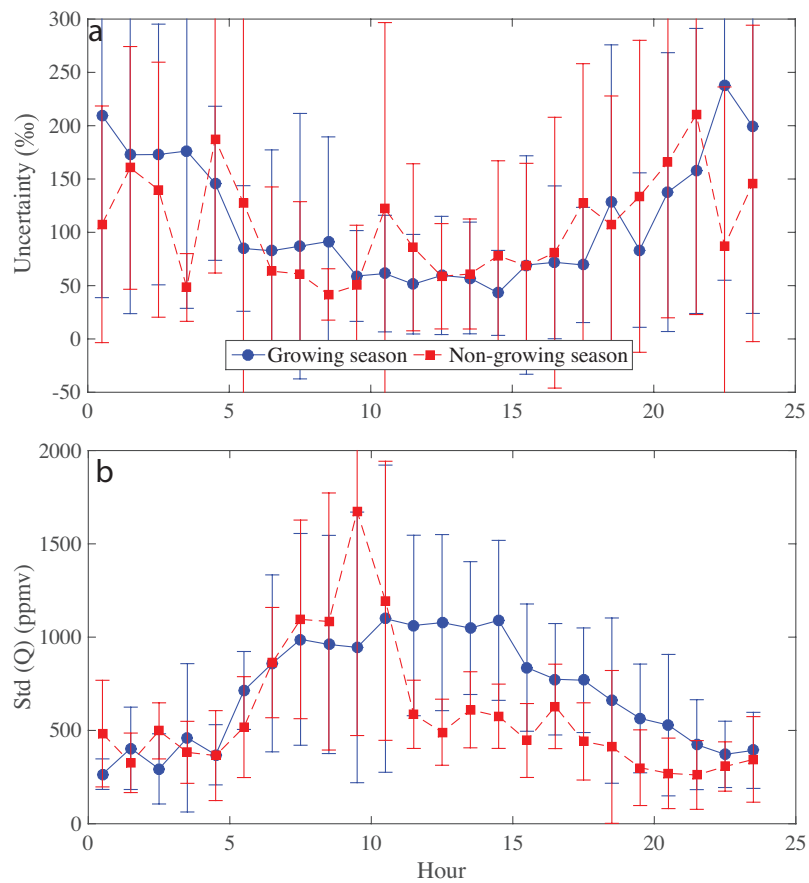


Figure 4.8: Comparison of the ensemble mean diurnal variation in uncertainty of  $\delta_{ET}$  (a) and standard deviation of water vapor of mixing ratio (b) during the growing season (July and August 2013, blue circle) and non-growing season (September and October 2013, red square). Vertical bars denote the standard deviation for each hour.

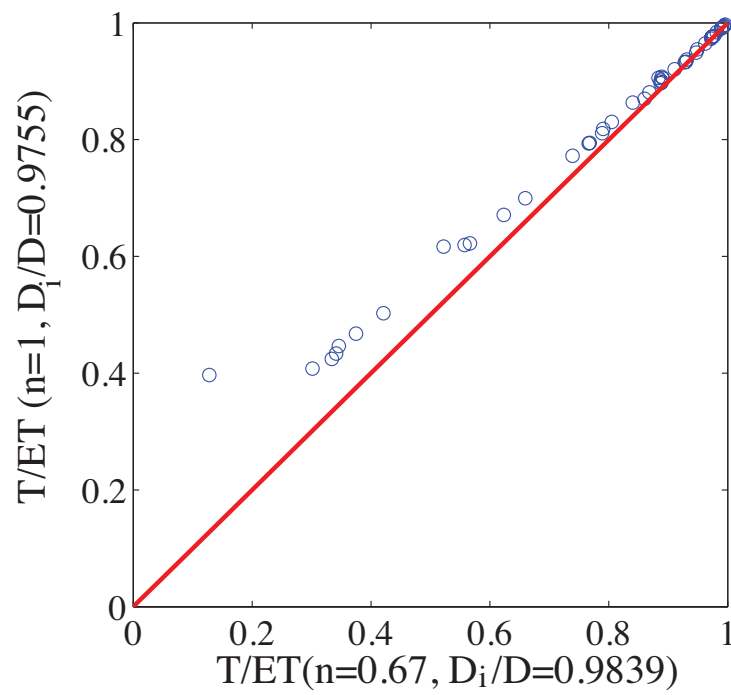


Figure 4.9: Impact of using different parameters for  $\delta_E$  estimation in the  $T/ET$  estimation. Data with  $T/ET > 1$  or  $T/ET < 0$  are excluded.

# Chapter 5

## Global $ET$ partitioning

### 5.1 Global synthesis of $LAI$ control on dry canopy $ET$ partitioning

$T/ET$  ratio has been shown highly relative to  $LAI$  [e.g., *Raz-Yaseef et al.*, 2010; *Yepez et al.*, 2005; *Lawrence et al.*, 2007; *Mitchell et al.*, 2009], but varying among ecosystems [e.g., *Raz-Yaseef et al.*, 2012; *Cavanaugh et al.*, 2011]. Additional suggesting causes including (1) water stress, (2)  $ET$  amount related to  $T/ET$  partitioning and (3) climate condition, are not considered as the major factor of  $T/ET$  variations (see [*Wang et al.*, 2014]). A plant growing stage function was included in *Wang et al.* [2014] to explain the variations, with a correlation of  $R^2 = 0.43$  for the global  $T/ET$  and  $LAI$  dataset. However, as *Wei et al.* [2015] suggested, the growing stages is generally related to  $LAI$  variation (growing stages is also a function of  $LAI$ ). Here we classified the whole vegetation into six classes and document previous studies related to each class in Sec 2. The majority of prior  $ET$  partitioning studies focused on crops (30/64) and grasses (13/64), and most of which were conducted in South America. The global synthesis of  $LAI$  controls on  $ET$  partitioning for each class is shown in Figure 5.1. In general,  $T/ET$  was represented quite well as a function of a 0.5-bin averaged  $LAI$  for different vegetation types, implying that vegetation plays a major role in driving the contribution of  $T/ET$ .

However, different vegetation types showed different exponential relationships and correlations (Table 5.1). The best regression was  $T/ET = 0.69LAI^{0.31}$  for grasses ( $R^2=0.95$ ) and the worst one was  $T/ET = 0.52LAI^{0.25}$  for crops ( $R^2=0.51$ ). The crops showed large variation in  $T/ET$  under different  $LAI$  conditions before bin-averaged process. Variable vegetations can be found in crops class, including corn [Sauer *et al.*, 2007], wheat [Zhang *et al.*, 2002; Herbst *et al.*, 1996], rice [Wei *et al.*, 2015], cowpeas [Sepaskhah and Ilampour, 1995], grapevines [Yunusa *et al.*, 2004] and so on. Although we further divided crops class into C3 and C4 group, no improved correlation was found. A further sub-classification may be required for the large difference of stomatal characteristic among these crops, although a well correlation was found after 0.5-bin averaged of  $LAI$ .

The  $T/ET$  estimated from  $LAI$  regression is described in Table 5.1. Overall, the crops systems tended to have high  $T/ET$ , mainly because agricultural vegetations are typically less constrained by environmental stress [Wang *et al.*, 2014].  $T/ET$  ranged from 0.43 to 0.69 under low  $LAI$  conditions (i.e.  $LAI=1$ ), which is significant higher than we thought. Although Wang *et al.* [2014] suggested a possibility of high proportion of vegetation water use under low  $LAI$  conditions, measurement errors at the same time, should also be noted [Wei *et al.*, 2015]. Forests dataset tended to have a low  $T/ET$  at the same  $LAI$  value ( $T/ET=0.47$ ,  $LAI=1$ ), whereas a high  $T/ET$  value can be found in crops, shrubs and grasses datasets ( $T/ET=0.63$ ,  $LAI=1$ ). For all the classes  $T/ET$  were positively related to  $LAI$ . When  $LAI=3$ ,  $ET$  of shrubs and grasses class increased dramatically ( $T/ET=0.97$ ), which shows a dominate role of  $T$ . Due to canopy structure, no  $LAI>3$  study was found in this class. For high  $LAI$  condition, a consistent value of  $T/ET$  can be found in forests class (about 0.79). The wetlands class (Only one study [Wei *et al.*, 2015] based on isotope method was used in this class.) showed the highest value ( $T/ET=0.88$ ,  $LAI=3$ ;  $T/ET=1.0$ ,  $LAI=6$ ), perhaps because of the consideration of only high  $T/ET$  timescale (mid-day) (see below). Nevertheless, well correlation between  $LAI$  and  $T/ET$  obtained from different datasets suggested that vegetation plays a major role in driving the contribution of  $T/ET$ .

## 5.2 *ET* and $E_c$ estimation from *CLM*, *GLEAM* and *GRACE*

Global patterns of *ET* estimated from *GLEAM*, *CLM* and *GRACE* based water balance are displayed in Figure 5.2. The mean values of the three products reveal high congruence. For example, high *ET* can be found in equator area while low *ET* can be found in high latitude area. On the other hand, the uncertainties also increase with increasing *ET*. *GRACE* derived *ET* appears to be the noisiest among these *ET* products. *GRACE* *ET* overestimates *ET* gradients in warm area (e.g. Amazon, South Asia). Mean annual *GRACE* inferred *ET* from 2004 to 2010 is 11% and 31% higher than *CLM* and *GLEAM*, respectively. *GLEAM* estimated *ET* is very similar to *CLM* estimated but lower than that of *CLM* in most of cases, except in Amazon basin. These differences could be related to energy balance or water balance constraints [Long *et al.*, 2014]. *GRACE*-inferred *ET* is easy to calculate but highly sensitive to the input data (*TWSC*, *R* and *P*) and their related uncertainties. Non-closure errors ranging from 5% to 25% of *P* are primarily attributed to errors in satellite-derived *TWSC* and *P*. Long *et al.* [2014] thus suggested uncertainties in *GRACE* inferred *ET* is higher than that in land surface model and remote sensing *ET* products. Moreover, large uncertainty of runoff estimation is a remained issue in this approach. On the other hand, *CLM* is based on *SVAT* schemes, in which actual *ET* is constrained directly by simulated soil moisture as well as meteorological forcing. In contrast, *ET* from *GLEAM* is determined largely by radiation, without explicit soil moisture constraints based on water balance. Considering that each product has its own uncertainties, it is necessary to investigate  $T/ET$  based on different *ET* products.  $E_c$  is derived from *GLEAM* and *CLM*. Though *CLM* and *GLEAM* *ET* generally consist with each other, significantly lower  $E_c$  can be found in *GLEAM* (about 14% of  $E_c/ET$  in global scale).

### 5.3 Global $ET$ partitioning

Based on global synthesis of  $LAI$  control on  $ET$  partitioning,  $LAI$  dataset, land cover, different  $ET$  and  $E_c$  products, the  $T/ET$  ratio can be estimated by our developed algorithm (Equation 2.13). The *GRACE* based results are shown in Figure 5.5 and the others were shown in Table 5.3. It is shown that  $T$  dominates in the densely vegetated areas of the tropic region. Moreover,  $T/ET$  is also very high over the grasses and crops areas.  $T/ET$  obtained from different  $E_c$  datasets show about 8% uncertainty (Figure 5.5). Interestingly, the Amazon area showed much lower  $T/ET$  (about 60% on average) than we thought, compare to grasses regions. There can be two reasons for this phenomenon: 1) A high canopy  $E_c$  can be found in Amazon region (Broadleaved, evergreen forest) because rainfall interception varies from 14 to 21% of annual rainfall [Kelliher *et al.*, 1993]. It means that much of the atmospheric demand for evaporation is satisfied by  $E_c$ , thereby limiting transpiration. 2) Compare to grasses, even with a high  $LAI$  (e.g.  $LAI=4$ ), the dry leaf condition  $ET$  is not dominated by  $T$  (Figure 5.1). Because  $T/ET$  in each grid cell is spatiotemporally weighted by  $ET$ , our results are highly sensitive to those latitudes and regions that contribute most of the  $ET$  in global hydrologic cycle (i.e. tropical and mid-latitude region). As shown in Figure 5.6, we note that our global mean of  $T/ET$  (50%-60%) is smaller than those of the literature complication studies [Wang *et al.*, 2014; Schlesinger and Jasechko, 2014] but in good agreement with the value reported in GCMs (47%) (also see Table 1.1). On the other hand, it is significantly smaller than that reported in isotopic approaches [e.g., Jasechko *et al.*, 2013; Good *et al.*, 2015b].

### 5.4 The uncertainties in our partitioning

The results demonstrated the robustness of our algorithm for  $ET$  partitioning. However, there were some uncertainties in our estimation. Multi-monthly averaged  $LAI$  and vegetation types were used in Equation 2.13. The day to day variation of these parameters may bias our  $LAI$  regression results. Because the runoff data on which our analysis is

based is not valid at daily timescale, we cannot account for this possible effect in current analysis. However, because our global analysis is mostly sensitive to tropical and mid-latitude regions ( $LAI$  and vegetation type has not significantly changed through the years), the time scale induced bias in the majority of the globe hydrologic cycle is likely small. The uncertainties derived from  $LAI$  regression ( $aLAI^b$  in Equation 2.13) should also be considered. Vegetation type, through its effect on the proportion of the shaded surface fraction, also influences  $T/ET$ , which generally increases from grasses to forests [Raz-Yaseef *et al.*, 2012].  $T/ET$  also varies among ecosystems according to the depth of water uptake. Tree responds primarily to deep soil moisture availability while grasses responds to water in the uppermost soil layer [Holdo and Nippert, 2015]. Some previous studies suggested that  $T/ET$  is larger for deep-rooted trees than for grasses, as well as for soil with better infiltration regimes than more impermeable soils [e.g., Raz-Yaseef *et al.*, 2012; Cavanaugh *et al.*, 2011]. Different from these views, our results found under the same  $LAI$  condition, transpiration fraction shows a larger values for grasses. One possibility is grasses transpiration predominantly reduces shallow soil moisture content which is the dominant factor in  $E_s$  estimation. Another possible reason is that the maximum stomatal conductance difference for different vegetation (transpiration efficiency). Canopy stomatal control of gas exchange is quantified by the stomatal conductance,  $g_s$ , which is a function of maximum stomatal conductance ( $g_{smax}$ ). Under similar meteorology, soil and  $LAI$  condition, a higher  $g_{smax}$  would give a higher  $T$  value. Generally, high  $g_{smax}$  can be found in crops and grasses, as concluded in Kelliher *et al.* [1995]: A highest  $g_{smax}$  can be found in crop ( $12.2 \text{ mm s}^{-1}$ ) while the lowest in forest ( $5.7 \text{ mm s}^{-1}$ ). Moreover, there is no significant different regression results among different forest classes, which was also confirmed in Kelliher *et al.* [1995] (no significant  $g_{smax}$  variation for different forest pieces). Furthermore, another uncertainty is  $T$  transpired by understory vegetation. Some studies suggested understory vegetation ranges from 10% to 50% of total  $ET$  and tends to increase with decreasing  $LAI$  [Kelliher *et al.*, 1997; Wilson *et al.*, 2000]. Nevertheless, the regression between  $T/ET$  and  $LAI$  suggested vegetation

generally controlled  $ET$  partitioning, but with some variability among vegetation types.

Another uncertainty is conducted from different measurement methods. Both isotopic and non-isotopic measurement have been included in our  $LAI$  regression estimation. Isotopic method provides an unprecedented opportunity for  $ET$  partitioning across a variety of spatial and temporal scale which is unattainable by conventional technique. However, most of isotope-based studies showed that  $T$  generally contributes more than 70% to the  $ET$ , while non-isotopic measurement generally leads to considerably smaller transpiration fractions. Thus some studies argued that the isotopic measurement may overestimate the contribution of  $T$ . However, *Wei et al.* [2015] and *Sutanto et al.* [2012] found that no systematic internal difference between isotope and non-isotope methods. Moreover, global simulation shows large discrepancy (Table 1.1). To understand the discrepancy among different methods, we divided the whole dataset into isotope and non-isotope groups. We further analyzed two satellite-based global  $ET$  datasets, (*GLEAM* and *CLM4.5SP*), and compared them with in-situ measurements. Without considering interception ( $ET = E_s + T$  here), relationships between the 0.5 bin averaged  $LAI$  and  $T/ET$  are shown in Figure 5.4. The  $T/ET$  is represented quite well as a function of a 0.5 bin averaged  $LAI$ . However, different datasets showed different nonlinear relationships. *GLEAM* dataset tended to have the highest  $T/ET$  under the same  $LAI$  value. Even under low  $LAI$  conditions ( $LAI < 1$ ),  $T/ET$  in *GLEAM* data was about 80%. Isotopic measurement showed a similar result under high  $LAI$  condition. However,  $T/ET$  derived by isotopic method was much smaller than that derived from *GLEAM* (60% at  $LAI = 1$ ). *CLM* and non-isotope method showed similar results, tending to have lower  $T/ET$  under high  $LAI$  conditions ( $< 80\%$  after  $LAI > 2$ ). The discrepancy between isotope and non-isotope measurement may result from mismatch of measurement time scale. The limitation of isotope method is that it is only applicable under strong latent heat condition. Under weak turbulence condition such as in the morning or evening, the uncertainties of Keeling plot was quit large. As a result, the isotope method estimated  $T/ET$  is only for mid-day (strong  $T$  condition), whereas non-isotope may also account for other time scale

(night-time, weak  $T$  but  $E$  still occurs). For example, from our paddy field observation, we found about 20% of total  $ET$  occurred at night-time (Figure 5.8) because the surface water temperature was higher than air temperature. This night-time evaporation was not accounted into  $LAI$  regression estimation. Thus, those isotopic based studies included in  $LAI$  regression estimation might resulted a overestimation of  $T/ET$  at the same  $LAI$  condition. The different  $LAI$  regression between  $CLM$  and  $GLEAM$  is resulted from significantly different interception fractions in evapotranspiration (also see below).  $E_c/ET$  in  $GLEAM$  is low (8%), resulting in high transpiration ratios.

Clearly, the most egregious bias in  $ET$  partitioning is the excessively large contribution from  $E_c$ . Not only in our study, *Jasechko et al.* [2013] showed a high  $T/ET$  because of the low  $E_c/ET$  in their study. If we change the interception data,  $T/ET$  would reduces to about 60%. The same problem can also be found in *Good et al.* [2015b]. The two  $E_c$  datasets used in our study was not verified. A further study about interception is required because canopy interception loss at various regions of the globe was few reported in the literature. Here we compared these datasets with observations (Figure 5.7 and comparison of measured value and simulated result of each study was shown in Table 5.2). These data were based on the published studies we reviewed (14 individual long term  $ET$  partitioning measurement studies) that measured at least two of the three components in an attempt to compare them with global simulation.  $E_c$  is significantly underestimated for both datasets. If a higher  $E_c$  was confirmed,  $T/ET$  in our analysis would be smaller. Moreover, the omission of snow interception in global simulation clearly needs to be addressed. Until now, there has been very little research on snow and there is no obvious candidate model to employ [*Miralles et al.*, 2010]. In summary, to reduce the uncertainty in evaluating global  $T/ET$ , the improvement of interception evaporation is a prerequisite in our futher study. Although further validation is required, our measurements provided a new inspiration for partitioning global evapotranspiration and suggested vegetation plays a major role in driving the contribution of  $T/ET$ .

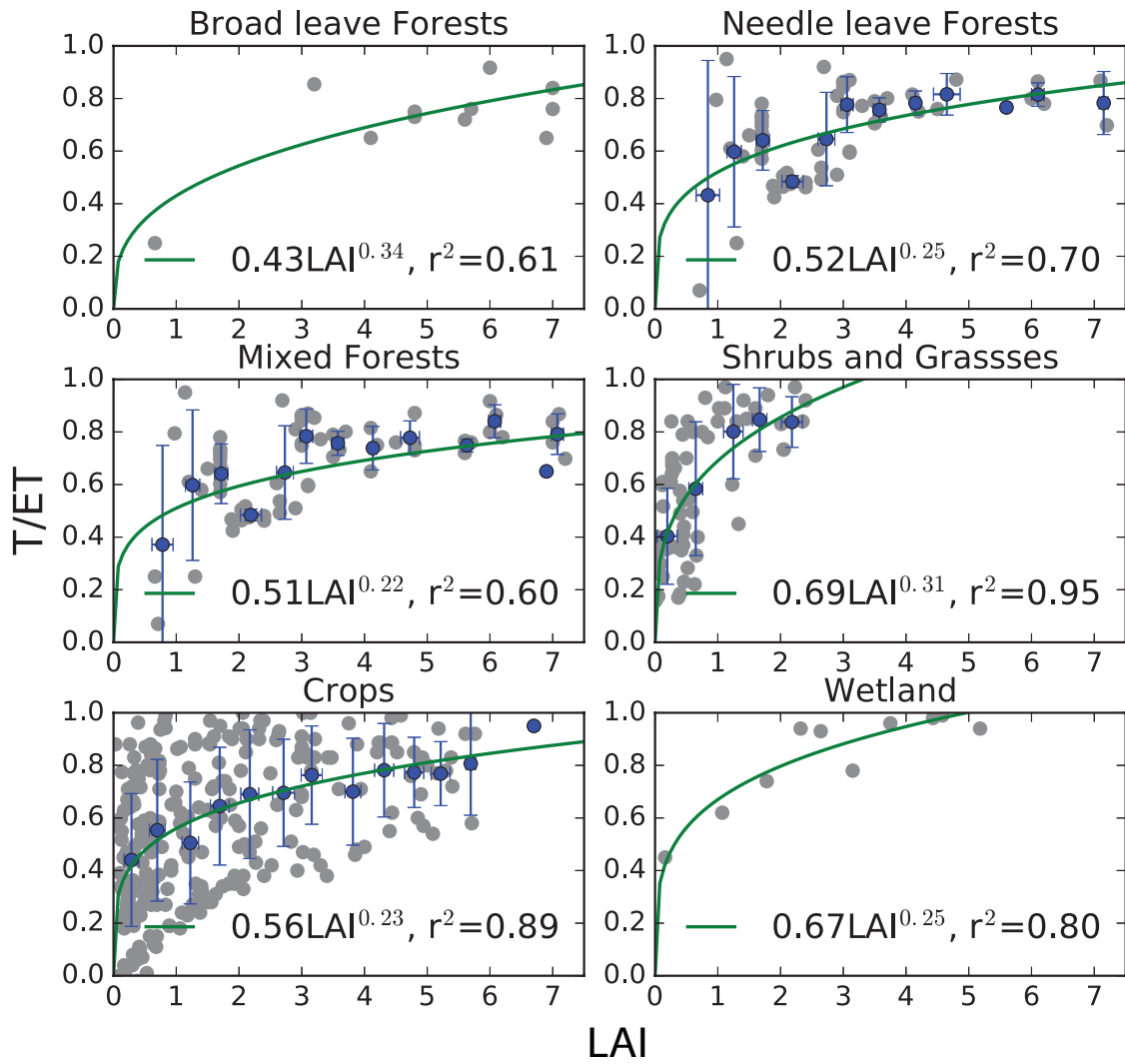


Figure 5.1: Global synthesis of  $LAI$  control on  $ET$  partitioning.

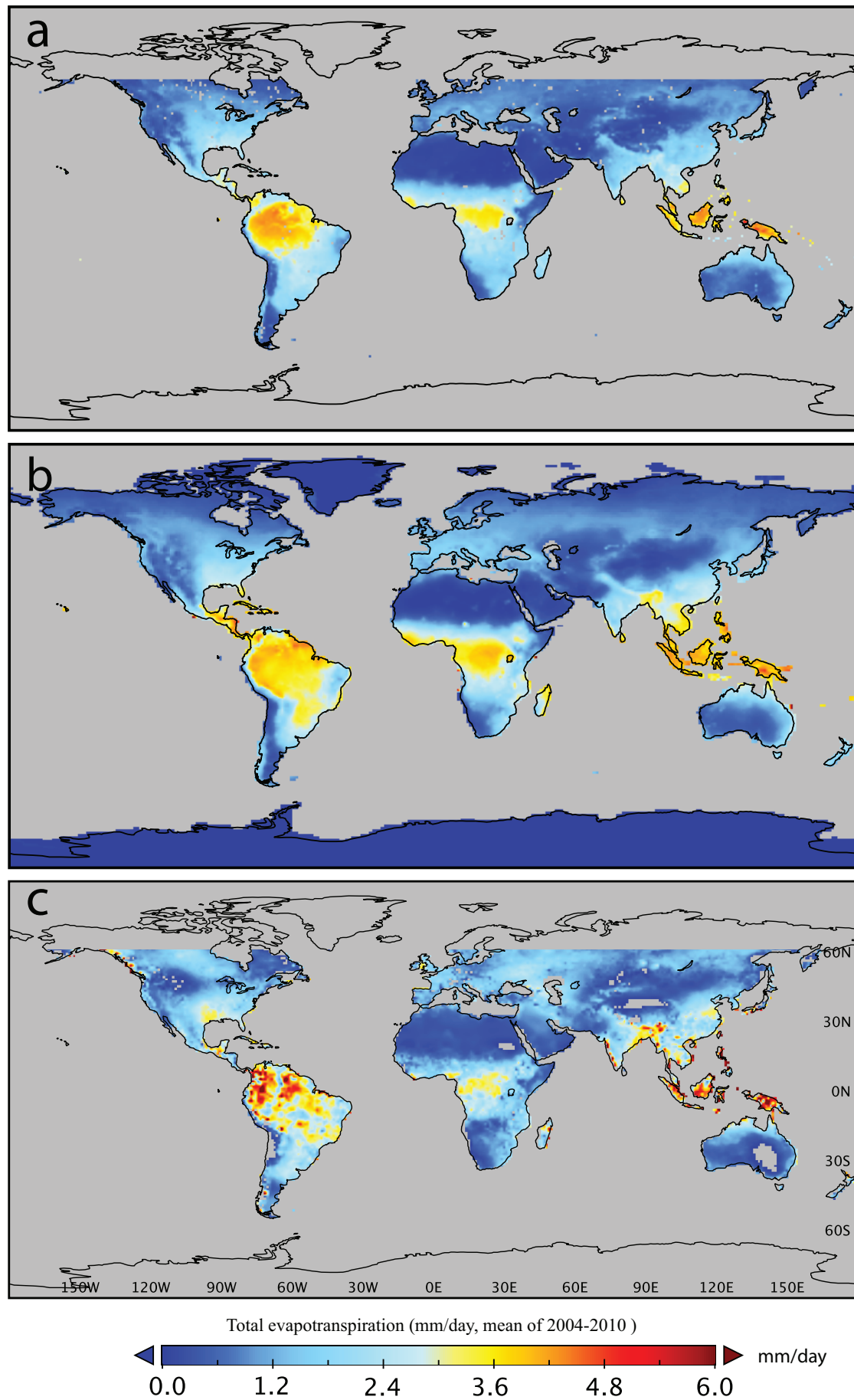


Figure 5.2: Comparison of *ET* derived from *GLEAM* (a), *CLM* (b) and *GRACE* (c).

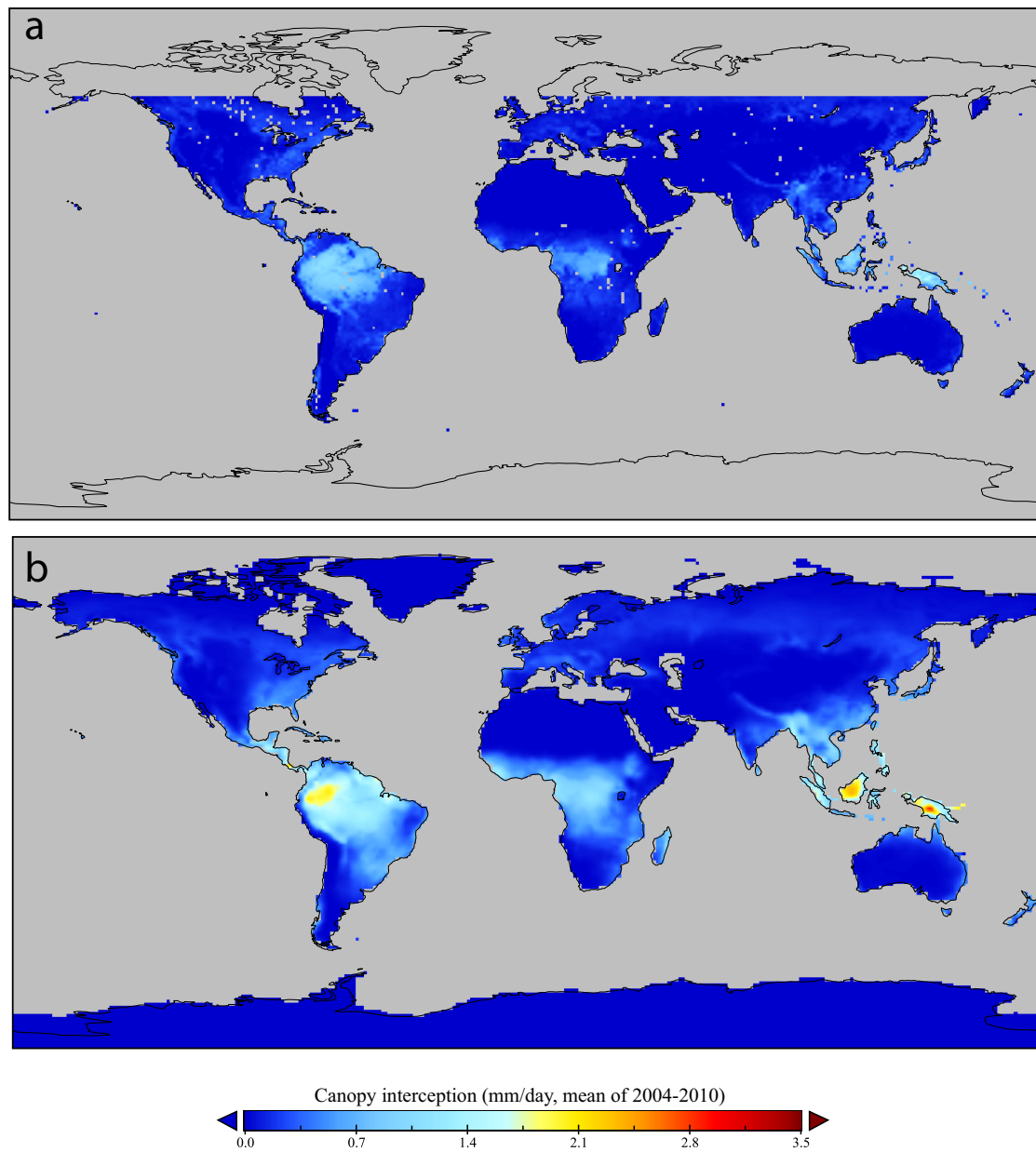


Figure 5.3: Comparison of canopy interception derived from *GLEAM* (a) and *CLM* (b).

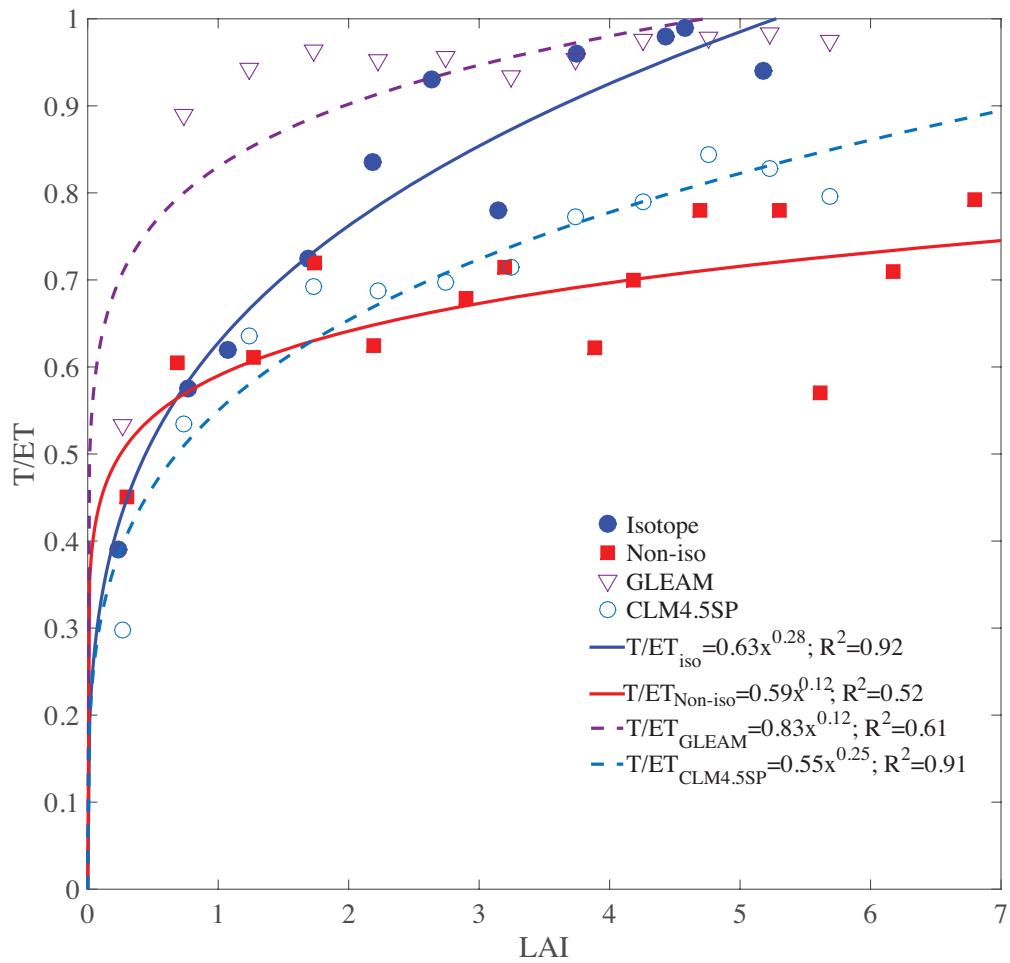


Figure 5.4: *LAI* control on *ET* partitioning based on different methods.

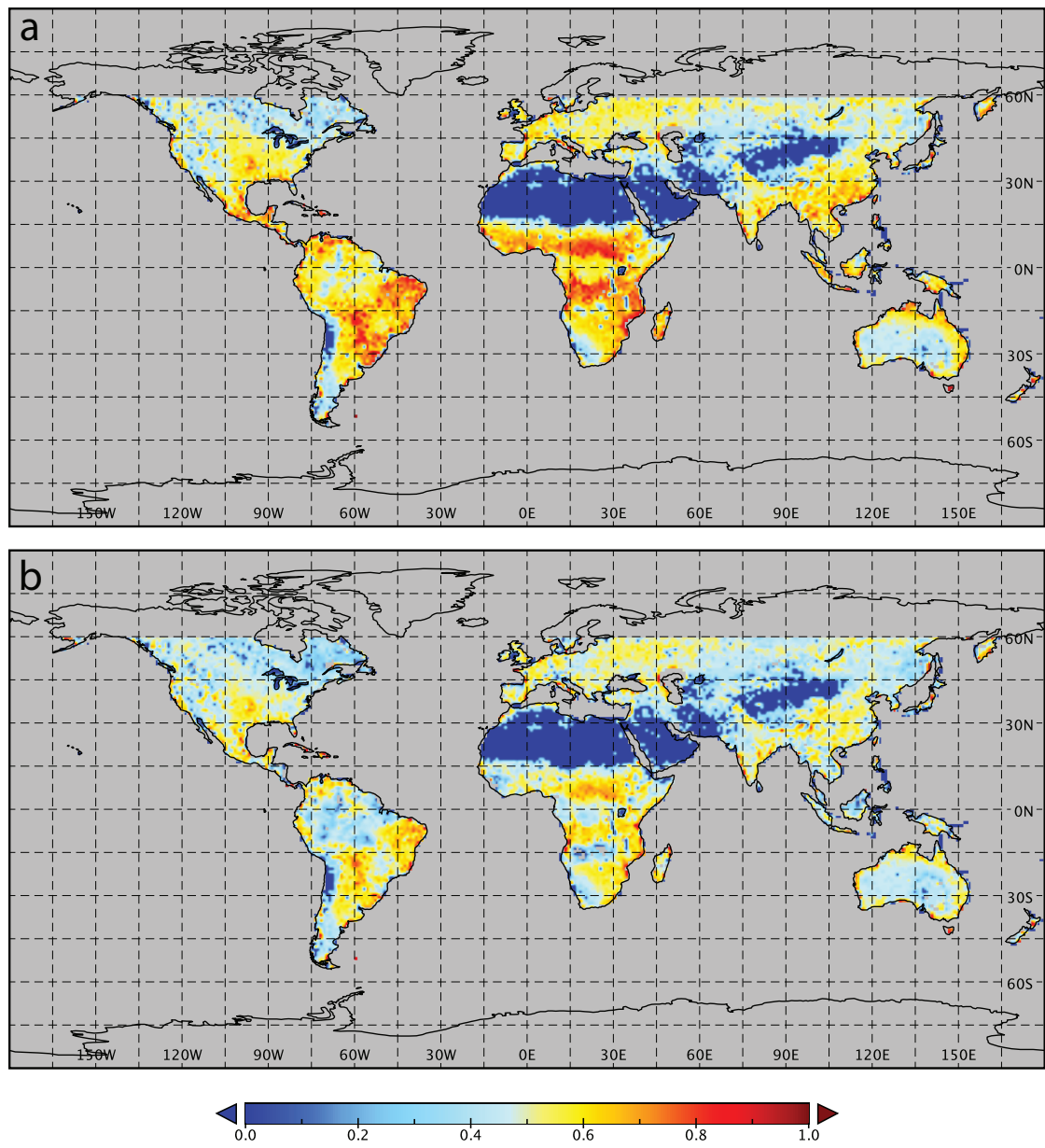


Figure 5.5: Global  $T/ET$  derived from  $LAI$  regression: (a) *GRACE* derived  $ET$  with GLEAM  $E_c$  (b) *GRACE* derived  $ET$  with CLM  $E_c$ ).

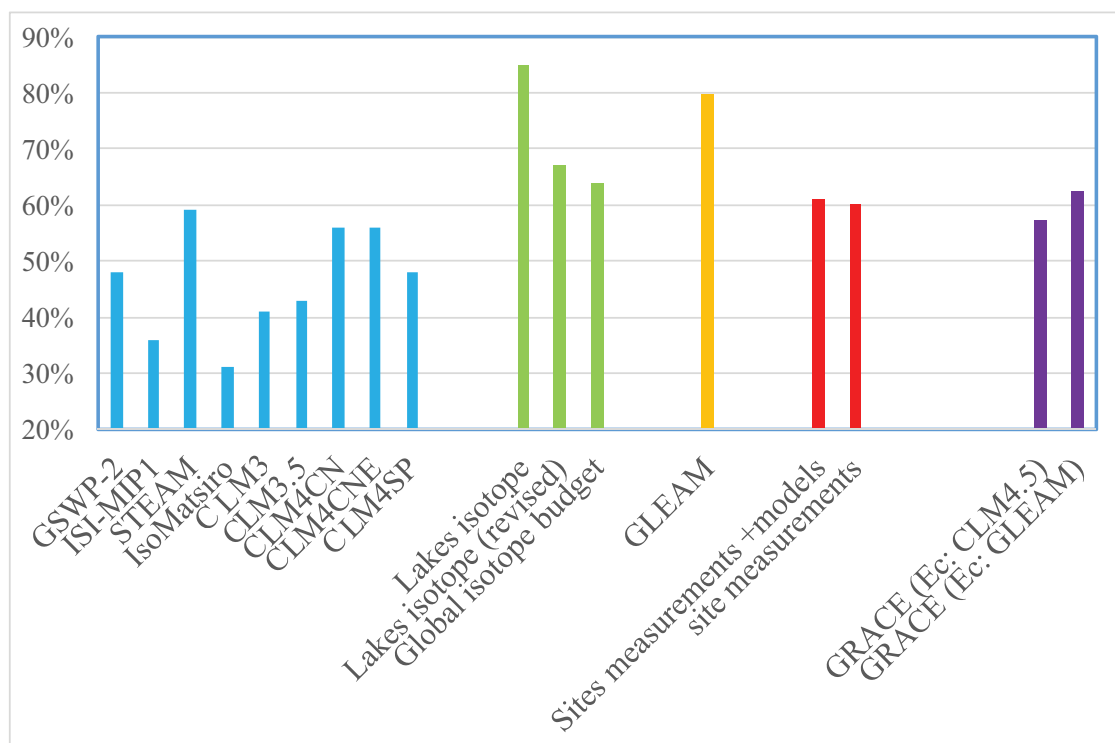


Figure 5.6: Global  $T/ET$  estimated from land surface models, isotopes, satellites, site measurements and  $LAI$  regression.

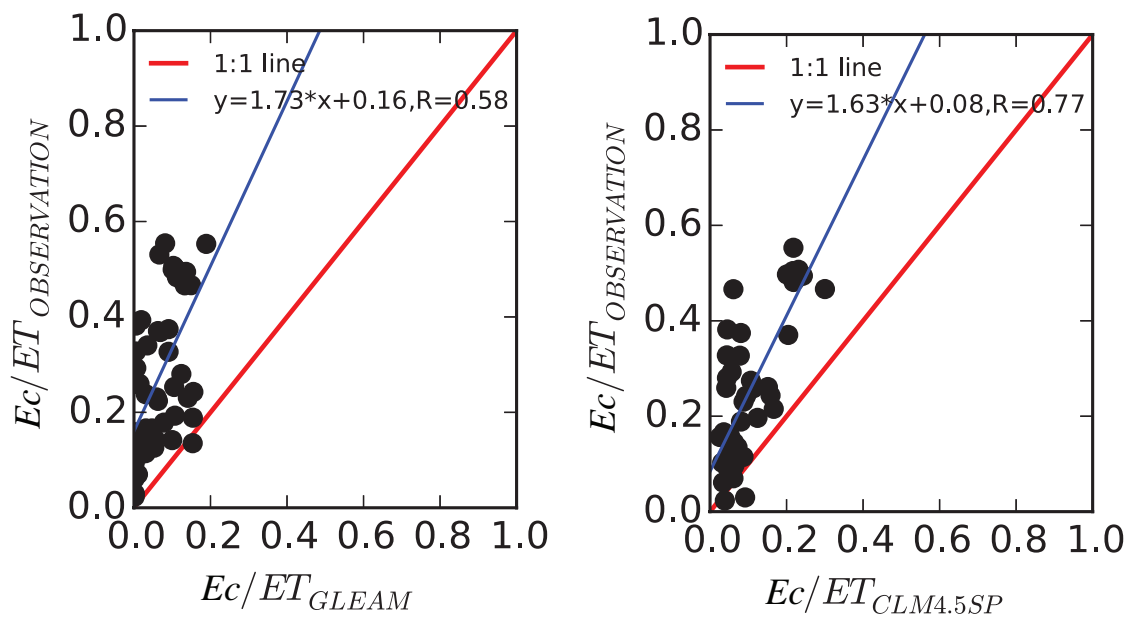


Figure 5.7: Comparison of simulated  $E_c/ET$  and measured  $E_c/ET$ . The measured data was obtained from *Mitchell et al.* [2009], *Mitchell et al.* [2012], *Lee et al.* [2010], *Oishi et al.* [2008] and *Jian et al.* [2015]

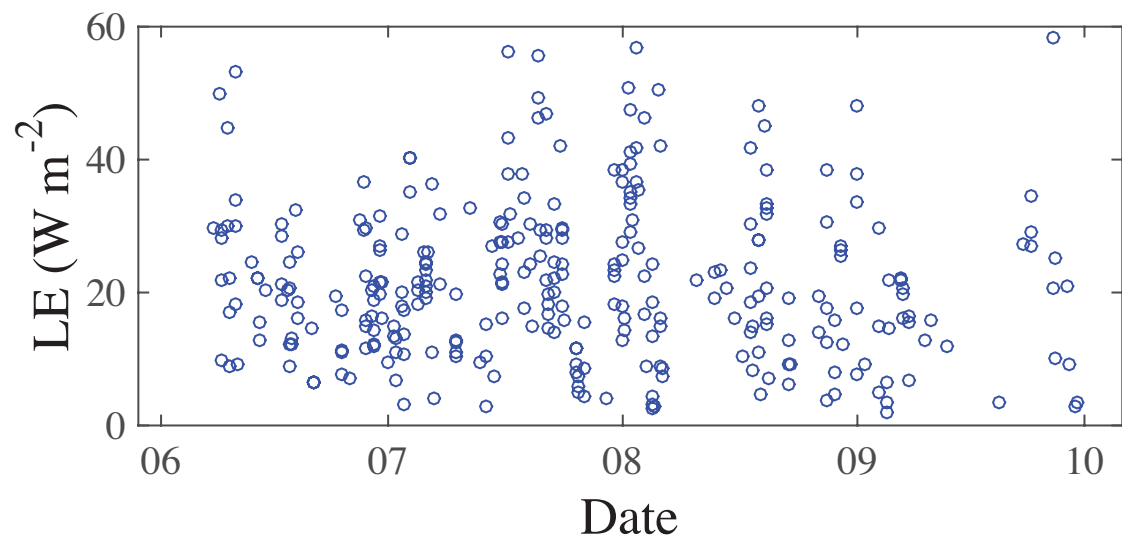


Figure 5.8: Hourly averaged latent heat observed at night time.

Table 5.1: Global systhesis of  $LAI$  control on  $ET$  partitioning.

| Vegetation Class     | $T/ET$ Regression | Correlation ( $R^2$ ) | $T/ET$ ( $LAI=1$ ) | $T/ET$ ( $LAI=3$ ) | $T/ET$ ( $LAI=6$ ) |
|----------------------|-------------------|-----------------------|--------------------|--------------------|--------------------|
| Broad leave forests  | $0.43LAI^{0.34}$  | 0.61                  | 0.43               | 0.62               | 0.79               |
| Needle leave forests | $0.52LAI^{0.25}$  | 0.70                  | 0.52               | 0.68               | 0.81               |
| Mixed Forests        | $0.51LAI^{0.22}$  | 0.60                  | 0.51               | 0.65               | 0.77               |
| Shrubs and Grasses   | $0.69LAI^{0.31}$  | 0.95                  | 0.69               | 0.97               | -                  |
| Crops                | $0.56LAI^{0.23}$  | 0.89                  | 0.56               | 0.72               | 0.84               |
| Wetlands             | $0.67LAI^{0.25}$  | 0.80                  | 0.67               | 0.88               | 1                  |

Table 5.2: Comparison of measured and simulated  $E_c/ET$ 

| Studies                          | $VT$            | Lat   | Lon   | $E_c/ET$ ( <i>obs</i> ) | $E_c/ET$ ( <i>CLM</i> ) | $E_c/ET$ ( <i>GLEAM</i> ) |
|----------------------------------|-----------------|-------|-------|-------------------------|-------------------------|---------------------------|
| <i>Verstraeten et al.</i> [2005] | Forest          | 51.3  | 4.5   | 0.24                    | 0.11                    | 0.06                      |
| <i>Mitchell et al.</i> [2009]    | Forest          | -32.3 | 117.9 | 0.15                    | 0.08                    | 0.15                      |
| <i>Mitchell et al.</i> [2012]    | Forest          | -36.7 | 146.7 | 0.06                    | 0.01                    | 0.01                      |
| <i>Lee et al.</i> [2010]         | Forest and crop | 37.7  | 127.1 | 0.49                    | 0.22                    | 0.11                      |
| <i>Oishi et al.</i> [2008]       | Forest          | 37.0  | -79.1 | 0.01                    | 0.04                    | 0.03                      |
| <i>Jian et al.</i> [2015]        | Forest          | 35.6  | 104.7 | 0.09                    | 0.05                    | 0.09                      |
| <i>Yimam et al.</i> [2015]       | Grass           | 36.2  | -97.0 | 0.25                    | 0.19                    | -                         |
| <i>Ringgaard et al.</i> [2014]   | Forest          | 56.0  | 9.3   | 0.39                    | 0.15                    | 0.11                      |
| <i>Kumagai et al.</i> [2014]     | Forest          | 33.1  | 130.7 | 0.46                    | 0.69                    | 0.15                      |
| <i>Herbst et al.</i> [2008]      | Shurb           | 51.4  | -1.3  | 0.20                    | 0.30                    | 0.30                      |
| <i>Sun et al.</i> [2013]         | Forest          | 36.4  | 139.6 | 0.55                    | -                       | 0.08                      |
| <i>Raz-Yaseef et al.</i> [2012]  | Forest          | 32.35 | 35.0  | 0.11                    | 0.09                    | 0.02                      |

Table 5.3:  $T/ET$  estimated based on Equation 2.13 with different  $ET$  and  $E_c$ .

| $ET$         | $E_c$        | $T/ET$ |
|--------------|--------------|--------|
| <i>GRACE</i> | CLM          | 57%    |
| <i>GRACE</i> | <i>GLEAM</i> | 62%    |
| <i>CLM</i>   | <i>CLM</i>   | 52%    |
| <i>CLM</i>   | <i>GLEAM</i> | 61%    |
| <i>GLEAM</i> | CLM          | 53%    |
| <i>GLEAM</i> | <i>GLEAM</i> | 60%    |

# Chapter 6

## Conclusions and Future Plan

We collected long-term observations of stable isotopes for oxygen and hydrogen in precipitation and water vapor from June 2013 to May 2014 in Tsukuba, Japan. We also investigated the isotopes of precipitation, water vapor, evapotranspiration, and large-scale moisture derived by IsoGSM to understand the variability of water isotopologues in near-surface atmospheric moisture. We conducted water tagging experiments, together with the observed  $d_v - excess$  and  $q - \delta D_v$  framework, to discuss how remote controls could affect the isotopic variability of water vapor. The following conclusions were drawn:

- (1)  $d_v - excess$  and  $Q$  displayed clear seasonal cycles, while the seasonal variations of  $\delta D_v$  and  $\delta^{18}O_v$  were insignificant.
- (2) We successfully combined the GCM simulation (*IsoGSM*) with high-frequency water vapor isotopic measurements to disentangle the water vapor source and its hydrological processes. The model successfully replicated observed isotopic variability, suggesting its ability to reproduce moisture transport.
- (3) The isotopic measurements of near surface water vapor and precipitation suggested that water vapor appeared to be isotopically in equilibrium with surrounding raindrops during a precipitation event. However, the equilibrium predictions from monthly  $\delta D_v$  were heavily biased.

- (4) The contribution of land evapotranspiration to local water vapor content ( $F_{ET}$ ) was approximately  $16.0 \pm 12.3\%$  on average. This revealed a significant seasonal cycle, where  $F_{ET}$  was highest in JJA ( $20.5 \pm 12.9\%$ ) due to the increased transpiration while lowest in DJF ( $7.1 \pm 13.2\%$ ). Our results indicated that the primary control on near surface water vapor  $\delta D$  variability is atmospheric circulation, with local evapotranspiration playing a secondary role.
- (5) The tagging experiments together with the  $d_v - excess$  and  $q - \delta D_v$  framework provided consistent evidence that the large temporal variation of  $\delta D_v$  could primarily be attributed to the advection of moisture from different source regions.

In this study, we adopted a recently developed partitioning technique using high-frequency in situ water vapor isotopic measurements and surface/soil water sampling over a paddy field near Tokyo, Japan, throughout a full growing season. We partitioned  $ET$  by estimating the isotopic signals of  $ET$ ,  $T$  and  $E$ , i.e.,  $\delta_{ET}$ ,  $\delta_T$ , and  $\delta_E$ , respectively.  $\delta_{ET}$  was determined using the Keeling plot method. Estimates of  $\delta_T$  were obtained directly from surface/soil isotopic measurements, assuming a steady state of transpiration.  $\delta_E$  was estimated using the Craig-Gordon model in combination with surface and vapor water isotopic measurements. The fraction of transpiration in evapotranspiration ( $T/ET$ ) ranged from approximately 0.2 to 1, with an almost continuous increase during the early growing season before finally reaching a relatively constant value close to 1, which was confirmed by a two source model simulated  $T$  and eddy correlation  $ET$  measurement. We also investigated the measurement uncertainties when estimating these three terms ( $\delta_{ET}$ ,  $\delta_T$ , and  $\delta_E$ ) and their effect on  $ET$  partitioning. The average uncertainty of  $T/ET$  estimation was  $14 \pm 10\%$ , and the average uncertainty in  $\delta_{ET}$ ,  $\delta_T$ , and  $\delta_E$  were about 9.06, 5.79, and  $14.27\text{‰}$ , respectively. It was apparent that the uncertainties in the estimation of  $\delta_T$  and  $\delta_{ET}$  significantly affected  $ET$  partitioning. The uncertainty in  $ET$  partitioning resulted from the uneven spatial distribution of decreases in the levels of surface water isotopes with an increase in  $T/ET$ , whereas as  $T/ET$  increased, the impact of uncertainty in  $\delta_{ET}$  became much larger than that for the other parameters ( $\delta_T$  and  $\delta_E$ ).  $FT$  was strongly

controlled by the leaf area index ( $LAI$ ) on a daily time scale ( $T/ET = 0.67LAI^{0.25}$ ,  $R^2 = 0.80$ ), suggesting the possibility of using  $LAI$  data to partition evapotranspiration at the global scale. The results demonstrated the robustness of using isotope measurements for partitioning evapotranspiration.

In order to understand global synthesis of  $LAI$  control on  $ET$  partitioning, we collected relevant article from Google scholar and Web of Science and analyze the relationship between  $LAI$  and  $T/ET$ . Based on vegetation types, we classified all the studies into 6 species: Broad leave forests, Needle leave forests, Mixing forests (combination of broad and needle forests), Shrubs-Grasses, Crops and wetlands. The  $T/ET$  was represented quite well as a function of a 0.5-bin averaged  $LAI$ , implying that vegetation plays a major role in driving the contribution of  $T/ET$ . However, different vegetation showed different nonlinear relationships. Forest dataset tended to have a low transpiration proportion under the same  $LAI$  value, whereas a high  $T/ET$  value can be found in Crops and Shrubs-Grasses datasets. This discrepancy may result from vegetation differences in height and bulk density. The  $ET$  estimated from  $RS$ ,  $LSM$  and  $GRACE$  based water balance showed a similar large-scale pattern. Based on global synthesis of  $LAI$  control on  $ET$  partitioning and different  $ET$  products, the  $T/ET$  ratio was reported to be 50% at the global scale, supporting the results reported in GCM models (47%). However, it was significantly smaller than that reported in isotopic approaches [e.g., Jasechko *et al.*, 2013; Good *et al.*, 2015a]. Although further validation is required, our measurements provided a new inspiration for partitioning global evapotranspiration and suggested vegetation plays a major role in driving the contribution of  $T/ET$ . Moreover, a further study about interception is also required because canopy interception loss at various regions of the globe has been scarcely reported in the literature.

## Appendix A

Previous studies used in *LAI*  
regerssion estimation.

Table A.1: Previous studies used in  $LAI$  regression estimation.

| Study                           | Latitude | Longitude | Method  |
|---------------------------------|----------|-----------|---------|
| Broad leave forests             |          |           |         |
| <i>Wilson et al.</i> [2000]     | 36.0     | -84.3     | Non-iso |
| <i>Wilson et al.</i> [2001]     | 36.0     | -84.3     | Non-iso |
| <i>Kelliher et al.</i> [1992]   | -42.1    | 172.2     | Non-iso |
| <i>Mitchell et al.</i> [2009]   | -32.3    | 117.9     | Non-iso |
| <i>Granier et al.</i> [2000]    | 48.7     | 7.1       | Non-iso |
| <i>Unsworth et al.</i> [2004]   | 45.8     | -122.0    | Non-iso |
| <i>Nizinski et al.</i> [2011]   | -4.6     | 11.9      | Non-iso |
| <i>Oishi et al.</i> [2008]      | 37.0     | -79.1     | Non-iso |
| <i>Tang et al.</i> [2006]       | 46.2     | 89.4      | Non-iso |
| <i>Raz-Yaseef et al.</i> [2012] | 31.4     | 35.0      | Non-iso |
| Needle leave forests            |          |           |         |
| <i>Diawara et al.</i> [1991]    | 44.7     | -0.8      | Non-iso |
| <i>Lee et al.</i> [2010]        | 37.8     | 127.1     | Non-iso |
| <i>Jian et al.</i> [2015]       | 35.6     | 104.7     | Non-iso |
| <i>Benyon and Doody</i> [2015]  | -37.8    | 140.8     | Non-iso |
| <i>Barbour et al.</i> [2005]    | -43.2    | 170.3     | Non-iso |
| <i>Oren et al.</i> [1998]       | 32.9     | 80.0      | Non-iso |
| <i>Raz-Yaseef et al.</i> [2012] | 31.4     | 35.0      | Non-iso |
| <i>Simonin et al.</i> [2007]    | 35.3     | -111.6    | Non-iso |
| <i>Domec et al.</i> [2012]      | 35.1     | -76.11    | Non-iso |
| <i>Lin et al.</i> [2012]        | 29.3     | 101.5     | Non-iso |
| <i>Sun et al.</i> [2013]        | 36.4     | 139.6     | Non-iso |
| <i>Jansson et al.</i> [1999]    | 60.5     | 17.3      | Non-iso |

Table A.2: Previous studies used in *LAI* regression estimation (continue).

| Study                                | Latitude | Longitude | Method  |
|--------------------------------------|----------|-----------|---------|
| Shrubs and grasses                   |          |           |         |
| <i>Allen and Grime</i> [1995]        | 13.3     | 2.2       | Non-iso |
| <i>Gibbens et al.</i> [1996]         | 32.6     | -106.8    | Non-iso |
| <i>Dugas et al.</i> [1996]           | 32.8     | -106.8    | Non-iso |
| <i>Stannard and Weltz</i> [2006]     | 29.8     | 52.8      | Non-iso |
| <i>Schlesinger et al.</i> [1987]     | 33.6     | -101.9    | Non-iso |
| <i>Cavanaugh et al.</i> [2011]       | 31.9     | -110.8    | Non-iso |
| <i>Li et al.</i> [2015]              | 43.5     | 116.5     | Non-Iso |
| <i>Yepez et al.</i> [2003]           | 31.7     | -110.2    | Iso     |
| <i>Xu et al.</i> [2008]              | 30.9     | 103.0     | Iso     |
| <i>Yepez et al.</i> [2005]           | 31.8     | -110.9    | Iso     |
| <i>Good et al.</i> [2014]            | 0.3      | 36.9      | Iso     |
| <i>Wang et al.</i> [2010]            | -        | -         | Iso     |
| <i>Wang et al.</i> [2015]            | 36.1     | 140.1     | Iso     |
| Crops                                |          |           |         |
| <i>Ashktorab H. and Paw U</i> [1994] | 38.5     | -121.7    | Non-iso |
| <i>Soegaard and Boegh</i> [1995]     | 13.5     | 2.5       | Non-iso |
| <i>Allen</i> [1990]                  | 35.9     | 37.1      | Non-iso |
| <i>Ham et al.</i> [1990]             | 33.6     | -101.8    | Non-iso |
| <i>Wallace et al.</i> [1993]         | 13.2     | 2.3       | Non-iso |
| <i>Massman and Ham</i> [1994]        | 33.6     | -101.8    | Non-iso |
| <i>Gutiérrez and Meinzer</i> [1994]  | 21.9     | -154.5    | Non-iso |
| <i>Sepaskhah and Ilampour</i> [1995] | 29.8     | 52.8      | Non-iso |
| <i>Cooper et al.</i> [1983]          | 35.9     | 37.1      | Non-iso |
| <i>Sadras et al.</i> [1991]          | -36.4    | 145.2     | Non-iso |
| <i>Yunusa et al.</i> [2004]          | -34.2    | 142.0     | Non-iso |
| <i>Sauer et al.</i> [2007]           | 41.9     | -93.6     | Non-iso |
| <i>Jara et al.</i> [1998]            | 46.2     | -119.7    | Non-iso |
| <i>Eastham et al.</i> [1999]         | -32.1    | 117.2     | Non-iso |
| <i>Sakuratani</i> [1987]             | 36.0     | 140.1     | Non-iso |
| <i>Zhang et al.</i> [2002]           | 37.9     | 114.7     | Non-iso |
| <i>Eberbach and Pala</i> [2005]      | 35.6     | 37.1      | Non-iso |
| <i>Yunusa et al.</i> [1997]          | -34.2    | 142.03    | Non-iso |
| <i>Herbst et al.</i> [1996]          | 54.1     | 10.25     | Non-iso |
| <i>Roupsard et al.</i> [2006]        | -15.4    | 167.2     | Non-iso |
| <i>Al-Khafaf et al.</i> [1978]       | 32.3     | -106.8    | Non-iso |
| <i>Harrold et al.</i> [1959]         | -        | -         | Non-iso |
| <i>Lascano et al.</i> [1987]         | 32.6     | -106.8    | Non-iso |
| <i>Villegas et al.</i> [2015]        | -        | -         | Non-iso |
| <i>Ding et al.</i> [2015]            | 37.9     | 102.5     | Non-iso |
| <i>Wei et al.</i> [2015]             | 36.0     | 140.1     | Iso     |
| <i>Wen et al.</i> [2016]             | 38.9     | 100.3     | Iso     |
| Wetland                              |          |           |         |
| <i>Wei et al.</i> [2015]             | 36.0     | 140.1     | Iso     |

## Appendix B

Water tagging experiments for SON,  
DJF and MAM.

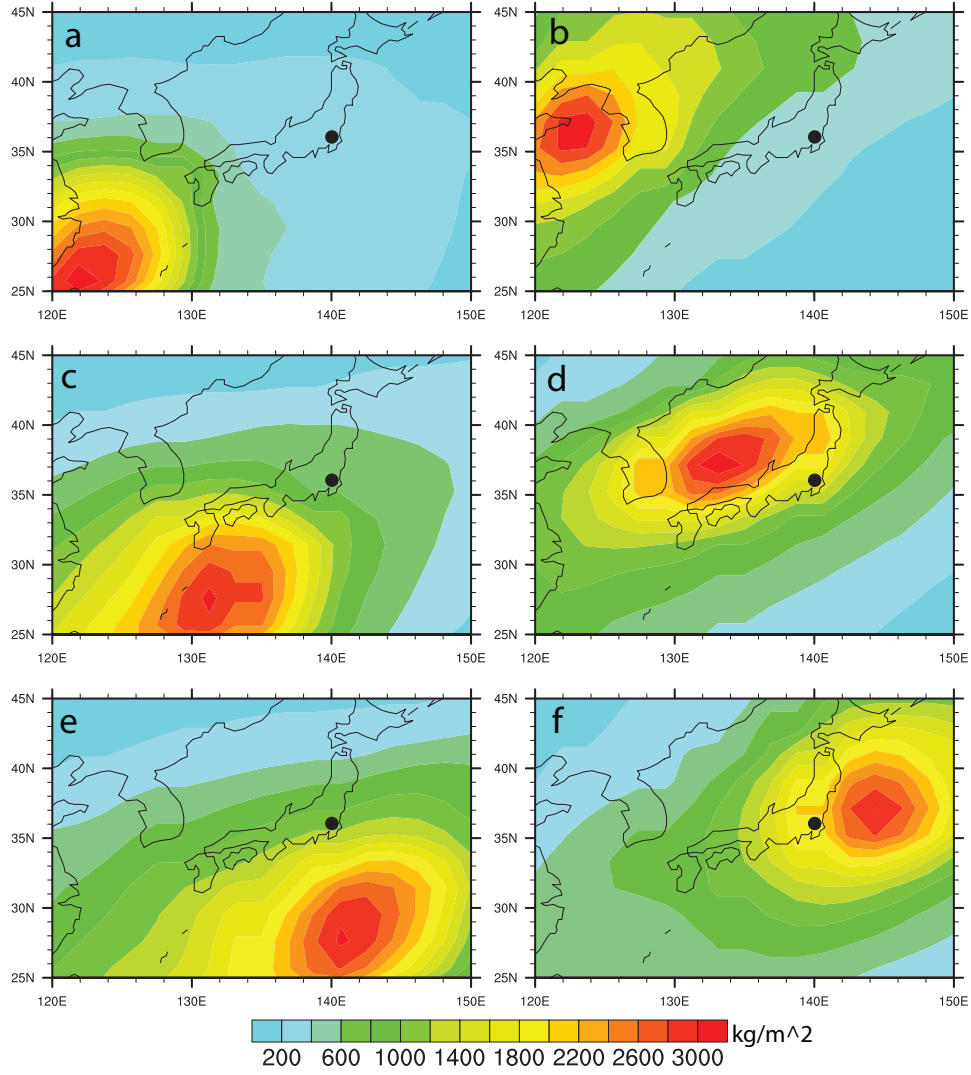


Figure B.1: The atmospheric column precipitable water contributed from tagged boxes in SON. Source regions for tagging experiments of water vapor tagged within  $10 \times 10$  degree boxes (contour): a,  $25^\circ\text{N} - 35^\circ\text{N}$ ,  $120^\circ\text{E} - 130^\circ\text{E}$ ; b,  $35^\circ\text{N} - 45^\circ\text{N}$ ,  $120^\circ\text{E} - 130^\circ\text{E}$ ; c,  $25^\circ\text{N} - 35^\circ\text{N}$ ,  $130^\circ\text{E} - 140^\circ\text{E}$ ; d,  $35^\circ\text{N} - 45^\circ\text{N}$ ,  $130^\circ\text{E} - 140^\circ\text{E}$ ; e,  $25^\circ\text{N} - 35^\circ\text{N}$ ,  $140^\circ\text{E} - 150^\circ\text{E}$ ; f,  $35^\circ\text{N} - 45^\circ\text{N}$ ,  $140^\circ\text{E} - 150^\circ\text{E}$ .

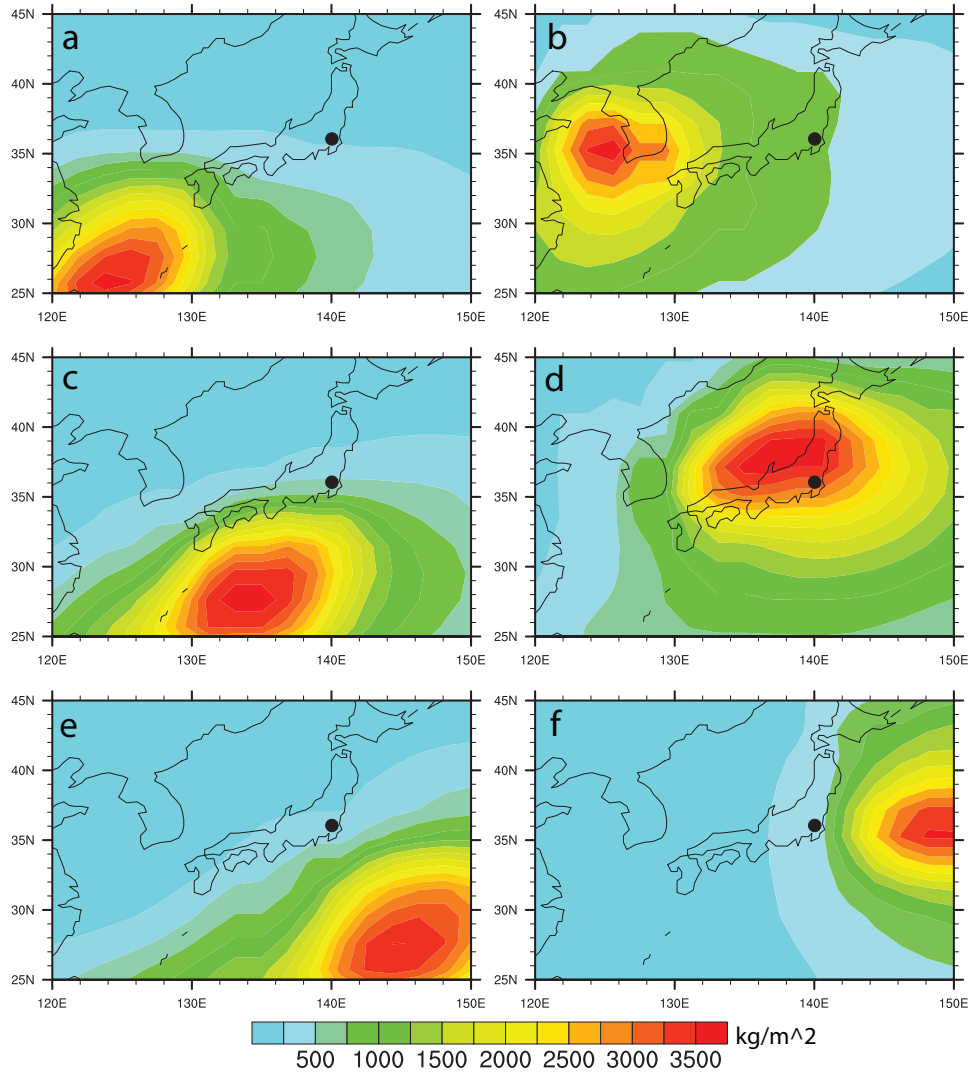


Figure B.2: The atmospheric column precipitable water contributed from tagged boxes in DJF. Source regions for tagging experiments of water vapor tagged within  $10 \times 10$  degree boxes (contour): a,  $25^\circ\text{N}$ – $35^\circ\text{N}$ ,  $120^\circ\text{E}$ – $130^\circ\text{E}$ ; b,  $35^\circ\text{N}$ – $45^\circ\text{N}$ ,  $120^\circ\text{E}$ – $130^\circ\text{E}$ ; c,  $25^\circ\text{N}$ – $35^\circ\text{N}$ ,  $130^\circ\text{E}$ – $140^\circ\text{E}$ ; d,  $35^\circ\text{N}$ – $45^\circ\text{N}$ ,  $130^\circ\text{E}$ – $140^\circ\text{E}$ ; e,  $25^\circ\text{N}$ – $35^\circ\text{N}$ ,  $140^\circ\text{E}$ – $150^\circ\text{E}$ ; f,  $35^\circ\text{N}$ – $45^\circ\text{N}$ ,  $140^\circ\text{E}$ – $150^\circ\text{E}$ .

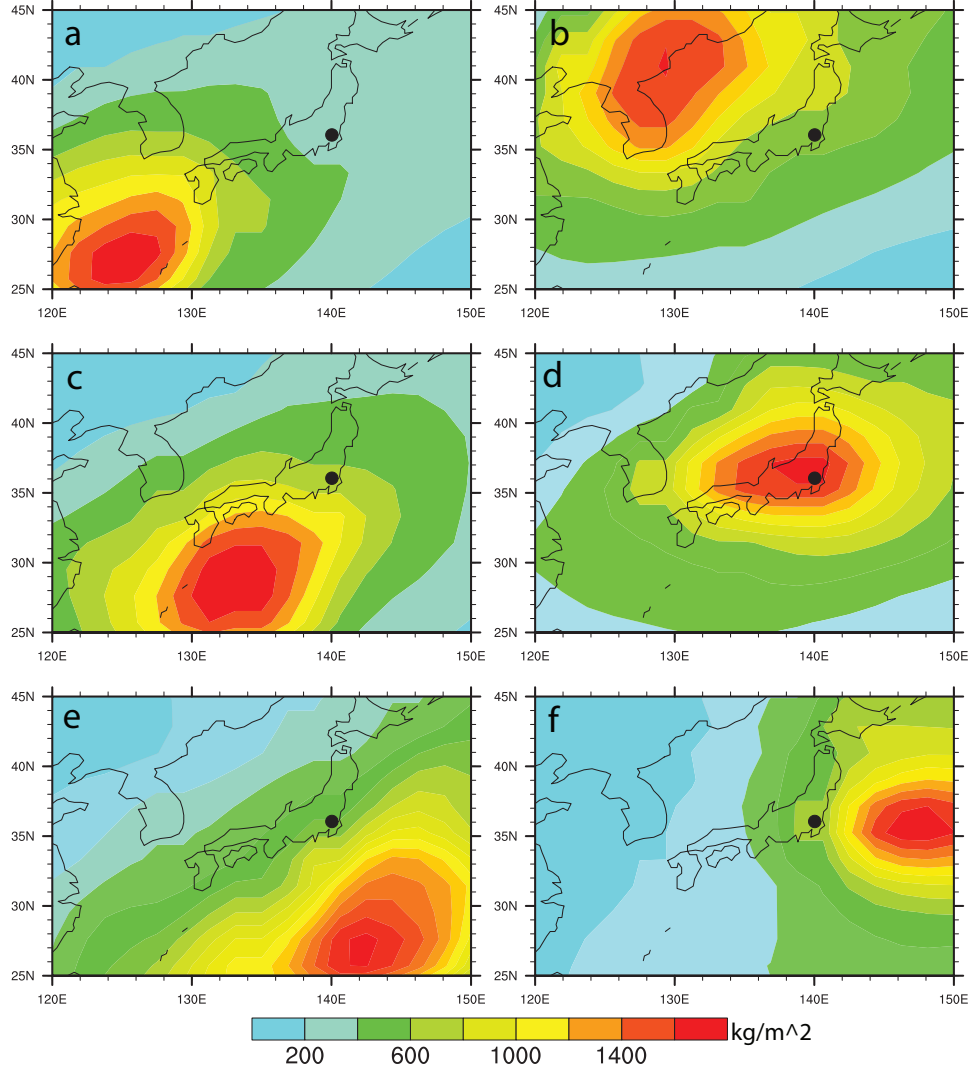


Figure B.3: The atmospheric column precipitable water contributed from tagged boxes in MAM. Source regions for tagging experiments of water vapor tagged within  $10 \times 10$  degree boxes (contour): a,  $25^\circ\text{N}$ – $35^\circ\text{N}$ ,  $120^\circ\text{E}$ – $130^\circ\text{E}$ ; b,  $35^\circ\text{N}$ – $45^\circ\text{N}$ ,  $120^\circ\text{E}$ – $130^\circ\text{E}$ ; c,  $25^\circ\text{N}$ – $35^\circ\text{N}$ ,  $130^\circ\text{E}$ – $140^\circ\text{E}$ ; d,  $35^\circ\text{N}$ – $45^\circ\text{N}$ ,  $130^\circ\text{E}$ – $140^\circ\text{E}$ ; e,  $25^\circ\text{N}$ – $35^\circ\text{N}$ ,  $140^\circ\text{E}$ – $150^\circ\text{E}$ ; f,  $35^\circ\text{N}$ – $45^\circ\text{N}$ ,  $140^\circ\text{E}$ – $150^\circ\text{E}$ .

# Appendix C

## *ET* partitioning based on Two layer method.

The partitioning of water fluxes into canopy and soil components could also be performed on the basis of eddy correlation measured *ET* and simulated *T*. Based on two-source model, *T* can be given by the following bulk equation:

$$T = \frac{\rho_a [q_{sat}(T_c) - q_a]}{r_{av} + r_c} \quad (C.1)$$

where  $\rho_a$  is the density of air ( $\text{kg m}^{-3}$ ),  $q_{sat}(T_c)$  is saturated specific humidity for the canopy temperature ( $\text{kg kg}^{-1}$ ),  $q_a$  is the air specific humidity ( $\text{kg kg}^{-1}$ ), the canopy temperature  $T_c$  ( $^{\circ}\text{C}$ ) which can be estimated according to the aerodynamic principle [Lee *et al.*, 2007]:

$$T_c = T_a + \frac{\overline{w't'}u}{u_*^2} \quad (C.2)$$

where  $u_*$  is friction velocity and  $\overline{w't'}$  is kinematic sensible heat. The aerodynamic resistance for water surface  $r_{av}$  ( $\text{s m}^{-1}$ ) can be determined as follow [Brutsaert, 1982]:

$$r_{av} = \ln \left( \frac{z_m - d_0}{z_{0mv}} \right) \ln \left( \frac{z_h - d_0}{z_{0hv}} \right) / k^2 u \quad (C.3)$$

where  $z_m$  is the height of wind speed measurement (m),  $d_0 = 0.666z_v$  ( $z_v$  is vegetation height in m) is the zero-plane displacement height (m),  $z_{0mv} (= 0.123z_v)$  is the roughness length governing momentum transfer above vegetation canopy (m),  $z_h$  is the height of temperature and humidity measurements (m),  $z_{0hv} (= 0.1z_{0mv})$  is the roughness length governing latent heat transfer above vegetation canopy (m) [Wang and Yamanaka, 2014],  $k = 0.40$  is the von Karman's constant and  $u$  is the wind speed at measurement height ( $\text{m s}^{-1}$ ). The canopy resistance  $r_c$  ( $\text{sm}^{-1}$ ) is estimated from:

$$r_c = r_{st}/LAI \quad (\text{C.4})$$

the leaf stomatal resistance  $r_c$  is derived from rice crop growth model [Maruyama and Kuwagata, 2010]:

$$r_{st} = (1 + S_{absH}/S_{abs})/g_{Smax} \quad (\text{C.5})$$

$$S_{abs} = [(1 - r) - \tau_s(1 - r_g)]Rs/L$$

where  $S_{abs}$  is the mean absorbed solar radiation per unit leaf area ( $\text{Wm}^{-2}$ );  $\tau_s$  is the transmissivities of the canopy for short-wave;  $g_{Smax}$  is the maximum bulk stomatal conductance ( $\text{m s}^{-1}$ ) for saturated  $S_{abs}$ ; and  $S_{absH} = 66 \text{ W m}^{-2}$  is the absorbed solar radiation ( $\text{Wm}^{-2}$ ) when  $g_s$  is equal to half of  $g_{Smax}$ .  $g_{Smax}$  for rice canopy decreases with rice growth due to change in plant phenology is expressed using following empirical function of developmental stage ( $P_s$ ):

$$g_{Smax} = g_{Smax1} + (g_{Smax0} - g_{Smax1} \exp(-(\theta_1 P_s)^{\mu_1}) + (g_{Smax2} - g_{Smax1}) \exp[-(\theta_2(2 - P_s))^{\mu_2}]) \quad (\text{C.6})$$

where  $g_{Smax0} = 0.060$ ,  $g_{Smax1} = 0.023$  and  $g_{Smax2} = 0.009$  are the values of  $g_{Smax}$  at transplanting ( $P_s = 0$ ), heading ( $P_s = 1$ ) and maturity ( $P_s = 2$ ), respectively;  $\theta_1 = 1.65$  and  $\theta_2 = 1.08$  are parameters affecting the timing of the decline in  $g_{Smax}$  after transplanting and heading, respectively; and  $\mu_1 = 8$  and  $\mu_2 = 8$  are parameters affecting the gradient of the

decline in  $g_{Smax}$  after transplanting and heading, respectively [Maruyama and Kuwagata, 2008, 2010]. The developmental stage  $P_s$  is determined by:

$$\begin{aligned} P_s &= N_{DAT}/N_{DTH} (0 < P_s \leq 1) \\ &= 1 + N_{DAH}/N_{DHM} (1 < P_s \leq 2) \end{aligned} \tag{C.7}$$

where  $N_{DAT}$  is days after transplanting,  $N_{DTH}$  is days from transplanting to heading,  $N_{DAH}$  is days after heading, and  $N_{DHM}$  is days from heading to maturation [Maruyama and Kuwagata, 2008]. Using these parameters and Equations C(1)-(7), the transpiration can be calculated at any given growth stage. As same as isotope-based method, data measured on a rainy day and outside the mid-day (11:00–15:00) period were rejected. Moreover, the weak turbulent cases ( $u_* < 0.05$ ) were excluded to maintain a high-quality  $LE$  estimation.

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