Understanding the variability of water isotopologues in near-surface atmospheric moisture over a humid subtropical rice paddy in Tsukuba, Japan

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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

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of near surface water vapor δD . 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.

Keywords: Water vapor isotope variability, d-excess, kinetic effects, water vapor source

1. Introduction

Measurements of water isotopic composition 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; 5 Lai and Ehleringer, 2011; Tremoy et al., 2012, 2014; Steen-Larsen et al., 2013, 2014b; 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 isotopic composition in the condensed phase is too coarse to determine the dynamical and microphysical processes 10 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 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).

15 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., 2014a). 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 20 GCM simulation. The authors showed that by incorporating a synthetic global

data set of surface vapor isotope 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; 25 Good et al., 2015a,b). 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., 2014b, 2015), ice sheets (Steen-Larsen et al., 2013), plateaus (e.g., 30 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; Berkelhammer 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 variations in the atmospheric water vapor isotopic 35 composition over 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 40 suggested that it is possible to derive a 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 approach is attractive because information regarding the precipitation isotope ratio (such as the Global Network of Isotopes in Precipitation [GNIP] dataset) is readily available (Lee et al., 2006). A number of studies 45 have concluded that local evapotranspiration (ET) is a major contributor to the stable isotope composition 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 identified the importance of other factors, such as the
50 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 origi-
nated from the Gulf of Mexico (warmer than the Pacific Ocean) and a high
ratio if moisture originated from the Pacific Ocean. By investigating the rela-
55 tionship between meteorological variables and water vapor isotope ratios, Lee
et al. (2006), Wen et al. (2010) and Huang and Wen (2014) suggested the dom-
inant effect of Rayleigh distillation during large scale moisture transport.

The use of high frequency water vapor isotopes has a great potential to im-
prove our understanding of the mechanisms that control stable isotope variabil-
60 ity 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)
65 it is difficult to understand how large scale moisture transport controls the iso-
topic variability of water vapor observed at a single tower. Studies investigating
the exact cause(s) of what controls long-term isotopic variability are rare.

In this study, we conducted a long-term water vapor isotopic study that
uses in-situ isotopes and meteorological measurements along with an isotope-
70 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 used isotopic information to partition water vapor
content into contributions from local and remote-source components. Tagging
experiments conducted using the Isotopes-incorporated Global Spectral Model

75 (IsoGSM) were used to understand the role of large scale moisture transport in
the observed isotopic variability.

2. Materials and methods

2.1. Site description

The study site (36° 03' 14.3" N, 140° 01' 36.9" E, a.s.l. 12 m) was located in
80 a rectangular 100 m×50 m cultivated plot near the center of a 150-ha rice paddy
field in Tsukuba. The location of the measuring instrument provided a suffi-
cient 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
85 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 ± 1.4
in July 2014. The site has a warm and humid climate, with a mean annual air
temperature of 13.5°C and mean annual precipitation of 1235 mm (Saito et al.,
90 2005). 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). Our
study area is located near the western Pacific Ocean, which is under the influ-
ence of a tropical to subtropical climate, with a monsoon period in both summer
95 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 ex-
pected significant differences in the evapotranspiration isotopic pattern among
seasons.

100 *2.2. Observation system*

We collected water vapor and precipitation samples. All isotope measurements are presented in parts per thousand using δ notation:

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

where R_{sample} and $R_{standard}$ are the ratios of oxygen mass 18 to mass 16 ($^{18}O/^{16}O$) or deuterium to hydrogen (D/H) of a sample (water vapor or precipitation), and the Vienna Standard Mean Ocean Water (VSMOW), respectively. 105 $d - excess$, defined as $\delta D - 8\delta^{18}O$ (Dansgaard, 1964), was also calculated to provide further insight into the kinetic effects associated with evaporation processes.

Precipitation samples were collected every six hours using an Eigenbrodt 110 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 liquid water samples were sealed into high-density glass storage bottles and stored in a refrigerator in the laboratory until analysis. The air temperature was kept at 25 °C to prevent condensation during 115 sample preparation and analysis. An optimized sequence layout and a simple post-run correction procedure (van Geldern and Barth, 2012) were performed to reduce the memory effect. Three standard waters were included in each analysis batch to correct the memory effect between samples. The correction results showed that the final mean values agreed very well with the true values 120 (Figure 1a). To check the correction performance, we also measured the same sample several times (after each 8 unknown samples) in one measurement event (see supplementary Figure S1). 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)

125 with similar precision and accuracy (std ± 0.2 ‰ for $\delta^{18}O$ and ± 0.5 ‰ for δD).
 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. Daily and monthly averages of precipitation δD
 130 and $\delta^{18}O$ were weighted by the precipitation amount and calculated as follows:

$$\delta_p = \frac{\sum_{i=1}^n \delta_{p,i} \times P_i}{\sum_{i=1}^n P_i} \quad (2)$$

where n is the number of observations for each day, and $\delta_{p,i}$ and P_i are the δ value and the total amount of the i th precipitation, respectively. Assuming that water vapor and precipitation are in isotopic equilibrium, the isotopic composition of water vapor $\delta_{v,e}$ can be calculated from δ_p by incorporating a temperature-dependent equilibrium fractionation factor α_{eq} (Majoube, 1971):
 135

$$\delta_{v,e} = \left(\frac{1 + \delta_p/1000}{\alpha_{eq}} \right) \times 1000 \quad (3)$$

Water vapor isotopic measurements are briefly summarized here and a detailed description can be found in Wei et al. (2015). The measurement system consisted of a high frequency Picarro cavity ring-down spectrometer (L2120-i), standard delivery module (A0101), high precision vaporizer (A0211), and 16-port
 140 distribution manifold (A0311). Measurements were made every 2 seconds, and the results were averaged into 1-minute interval. A shielded air intake installed at a height of 2-m was connected to an A0311 module using a 14-m Teflon inlet tube. The flushing time was about 2 min. We did not heat the tubing because the air temperature was higher than the dew point temperature
 145 (not shown). Two in-house water standards (-0.3 ‰ and -26.0 ‰ for $\delta^{18}O$ and

-1.2‰ and -197.9‰ for δD) were vaporized by the standard delivery module, mixed with dry room air and transferred to the analyzer. Each calibration cycle lasted 30 min for each standard. The results showed a high precision with std $\pm 0.3\text{‰}$ for $\delta^{18}O$ and $\pm 1.3\text{‰}$ for δD . The standard water was changed every
150 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 1b). However, the large measurement uncer-
155 tainty under low water vapor concentration suggested that the correction of the raw measurements using the humidity-isotope response functions (Steen-Larsen et al., 2014b) might be required. Unfortunately, there was a data gap from 22 October to 23 December 2013, due to a pump failure.

To identify the source of atmospheric moisture and its transport pattern, we
160 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)),
165 representing a pseudoadiabatic process based on oceanic temperatures, with an initial δD value of -80‰ (approximating the value of vapor in equilibrium with the ocean) in an open system, and 2) a mixing line, 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)).

170 2.3. *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 δD 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 uses 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. Most isotopic fractionation is assumed to occur at thermodynamic equilibrium, except for three particular cases: surface evaporation from open water, condensation from vapor to ice in supersaturation conditions under -20°C , and evaporation and isotopic exchange from liquid raindrop to unsaturated air. IsoGSM assumes a constant isotopic value (0‰) for the ocean water and no fractionation when water evapotranspires over land surface (i.e., 100% transpiration is assumed). The isotopic ratio of land surface storage (one snow layer and four soil layers) is calculated based on the isotopic mass balance equations, assuming evapotranspiration and surface runoff occur only from the first layer of the soil and that base runoff occurs from the deepest (fourth) layer of the soil without fractionation. 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 treated as a 1-box model, with input (evapotranspiration) and output (air from the free atmosphere) only from the top and bottom. Near surface horizontal advection can be assumed to be small and is smoothed 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 free atmosphere (convective boundary layer (hereafter CBL) water vapor, represented by IsoGSM simulated water vapor and its associated δD_v output (δ_{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):

$$F_{ET} = \frac{\delta_v - \delta_{bg}}{\delta_{ET} - \delta_{bg}} \quad (4)$$

where F_{ET} is the contribution of evapotranspiration 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).

2.4. Water-tagging experiments using IsoGSM

In addition to local evapotranspiration, water vapor isotopes are also affected by original oceanic sources. To determine the origin of water vapor and its impact on the isotopic variability of near-surface water vapor, we conducted a tagging simulation using IsoGSM. The principle 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 calcu-
lations. The method used here allows a direct quantification of the amount of
water advected from tagging region. Similar experiments were conducted to un-
derstand the cause of the seasonal variation in the oxygen isotopic composition
of precipitation along the western U.S. coast (Buenning et al., 2012). In our
study, vapor was tagged within a $10^\circ \times 10^\circ$ boxed sub-region (see Sec.3.4). The
atmospheric column precipitable water derived from tagged boxes was used to
identify the source region of the water vapor.

3. Results and discussion

3.1. Characteristics of observation and simulation

Figure 2 shows the daily precipitation (P), daily averaged water mixture
ratio (Q), $\delta^{18}O$, δD and $d - excess$ of water vapor (with subscript v), and the
weighted precipitation amount of $\delta^{18}O$, δD and $d - excess$ (with subscript p).
 Q varied from 3000 to 35000 ppmv. $\delta^{18}O_v$ fluctuated from -28.4‰ to -11.0‰,
 δD_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 $\delta^{18}O_v$, δD_v , and
 $d_v - excess$ in the warm season than the cold season. Similar results were
observed in Beijing ($40^\circ 00' N$, $116^\circ 23' E$ (Lee et al., 2006)). The seasonal
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 δ_p varied widely, ranging from -15.7‰ to 2.9‰ for $\delta^{18}O_p$, from
-114.9‰ to 34.5‰ for δD_p , and from -21.6‰ to 27.7‰ for $d_p - excess$. Positive
 $\delta^{18}O_p$ and δD_p values tended to occur in small showers, 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 δD_v were consistently lower than but varied together with precipitation isotopes. Similarly, $d_v - excess$ consistently varied with $d_p - excess$.
255 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).

260 Monthly isotope and weather variables are shown in Figure 3. Seasonal changes of relative humidity RH , air temperature T_a , and δD and $d - excess$ are plotted in Figure 3. RH and T_a displayed a clear seasonal signal. There was no clear seasonal cycle in δ_v or δ_p , although slightly lower values were apparent in winter. In contrast, $d_v - excess$ and Q displayed a clear seasonal cycle, which
265 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‰)
270 and the lowest in November (1.5‰).

Figure 2 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 δ_v variation under the influence of regional-scale advection of atmospheric moisture ($r^2 =$
275 0.64, RMSE = 10.8‰ for δD_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; Steen-Larsen et al., 2014a). 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
280 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 δ_v . Despite the discrepancy in modeled isotopic variability, IsoGSM provided important information regarding how large-scale
285 moisture transfer influenced the isotopic variation of surface water vapor. The strong fit at our site suggested that the variability in near-surface water vapor 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
290 required to determine the driving forces behind the variability of δD_v . Hereafter, we only present results based on δD and $d - excess$. Similar results obtained from $\delta^{18}O$ are not shown here.

3.2. *Effect of precipitation on the isotopic variability of water vapor*

The observed and simulated isotopic ratios (Figure 2) indicated that dur-
295 ing non-rainy days, there was no significant isotopic variation of water vapor. On rainy days, the isotopic composition of water vapor changed suddenly and considerably in relation to the isotopic composition of rain (Figure 2). 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
300 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 (3) and compared it with δD_v in Figure 4. δD_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

305 equilibrium with precipitation at daily time scale. At a monthly time scale,
 our equilibrium predictions differed greatly from the monthly δD_v (Figure 3,
 $r = 0.22$). These results contradicted previous findings, which demonstrated a
 reasonable agreement between δD_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).
 310 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
 δ without changing $d - excess$ (Dansgaard, 1964; Bonne et al., 2015).

3.3. *Effect of local evapotranspiration on the isotopic variability of water vapor*

315 During non-rainy days, surface ET can significantly contribute to the tem-
 poral variation in δ_v . We investigated this effect using Equation (5) with three
 parameters, δ_{ET} , δD_v and δ_{bg} (the isotopic ratio of the background air). Hourly
 δ_{ET} values were estimated by the Keeling plot approach (Keeling, 1958), when-
 ever data points were available ($N > 40$) in a given hour and with a high coefficient
 320 of determination ($r^2 > 0.8$) from each linear regression. Wei et al. [2015] demon-
 strated 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 δ_{ET} values (sensible heat weighted). δD_v at
 325 simulated CBL height (δ_{bg}) was derived from linear interpolation of water va-
 por isotopic ratio from 925 hpa (about 750m), 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 (see supplementary Figure S2); 2) IsoGSM is nudged by United States
 330 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 2).

The variability of δ_{ET} , δ_{bg} , and δD_v at the daily time scale is shown in Figure 5. δ_{ET} ranged from -112.2‰ to 37.1‰. A significant difference in δ_{ET} was found between the growing and non-growing seasons. In the summer season (June-July-August, JJA), δ_{ET} generally depended on transpiration. δ_{ET} was closely coupled with irrigation water, indicating small isotopic fractionations during this period (Wei et al., 2015). In the non-growing season, δ_{ET} was considerably more variable than in JJA. Without irrigation and consequently with negligible transpiration, δ_{ET} was controlled by other meteorological parameters, such as precipitation and radiation. For example, a rapid decrease of δ_{ET} corresponded to evaporation after precipitation on 18 October (from -51.8‰ on 18 October to -97.81‰ on 20 October). Then δ_{ET} values gradually increased due to soil evaporation (-69.2‰ on 21 October). Compared to δ_{ET} , δD_v and δ_{bg} clearly displayed seasonal variation and were highly correlated with each other. These results suggest that δD_v was mainly controlled by the air from the free atmosphere. The largest difference between δD_v and δ_{bg} was found in JJA, whereas the smallest difference was found in winter (December-January-February, DJF). In JJA, relatively high δD_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 peak of the rice growing season in JJA. The monthly averaged δD_v , δ_{ET} and δ_{bg} used to estimate monthly F_{ET} are listed in Table 2. 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 6). Thus, we suggest that the primary control on the variability of near surface

δ_v and humidity during non-rainy days is atmospheric circulation, with local
360 ET playing a 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 per-
formed at a coarse resolution ($200 \text{ km} \times 200 \text{ km}$), which is far greater than
365 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 prob-
lem in most isotope-enabled GCMs. Bias also arises from neglecting horizontal
advection, which is likely to alter the near surface δD_v . Very few measure-
370 ments of δ_{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
375 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 δD_v at 2-m and
simulated δ_{bg} at CBL height ($r = 0.74$) with the insignificant correlation be-
380 tween measured δD_v at 2-m and Keeling plot estimated δ_{ET} ($r = 0.18$) also
suggested the dominant role of air from the free atmosphere.

3.4. Remote controls on the isotopic variability of water vapor

To understand the remote controls on water vapor isotopic variation, we
conducted a water tagging experiment to identify the origin of the water vapor.
385 As an example, the model simulation indicated different sources of water vapor

in JJA (Figure 7). 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 8 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° to 90° , Area f; 90° to 180° , Area e; 180° to 270° , Area a+c; 270° to 360° , Area b+d). The results showed that the original source had a significant impact on water vapor isotopes (Figure 9). Seasonal and regional differences of δD_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 1) can also be explained by the seasonal change in the primary marine moisture sources. As shown in Figure 2, 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 6) above the ocean in DJF. These results were con-

sistent with the negative correlation between $d_v - excess$ and relative humidity
 observed above the ocean (Dansgaard, 1964; Uemura et al., 2008; Benetti et al.,
 415 2014, 2015).

The IsoGSM simulation suggested that the mixing of air from multiple
 sources was likely a major factor controlling the water vapor isotopic varia-
 tion at our study site on non-rainy days. To further explore the mechanisms
 responsible for the observed isotopic variation and to validate the simulation
 420 results, we plotted daily mean δD_v as a function of specific humidity q for each
 season. As shown in Figure 10, 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. δD_v was clustered tightly around the mixing line, sug-
 gesting a mixing of marine moist air and a dry air mass. This supports the idea
 425 that in general, cold northerly winds transport water vapor that originates from
 the Japan Sea to our site (Figure 8). Outside of the winter season, q varied over
 a wide range ($2 < q < 25$ g/kg), while δD_v fell in the range between the Rayleigh
 curve and the mixing line. ET might be one of the factors pulling data points
 away from the evaporation line. However, most data points were more depleted
 430 than -80‰, suggesting a negligible impact of ET on δD_v variation (most of
 the time, ET tends to increase δD_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 δD_v was related to the mixing of different oceanic sources
 (Figure 8) of distinct isotopic ratios (Figure 9). Mixing can occur between any
 435 number of air masses that deviate from a single curve. Hence, the entire area
 between the Rayleigh curve and the mixing line can be considered a result of
 mixing (Galewsky and Samuels-Crow, 2014a). The tagging experiment and q -
 δD_v framework provide consistent evidence that the mixing of multiple moisture
 sources plays an important role in controlling δD_v variability in this region. In

440 contrast to previous studies in inland regions (e.g., Lee et al., 2006; Welp et al.,
 2008; Wen et al., 2010; Huang and Wen, 2014), our measurements showed that
 q was a poor predictor of δD_v because of the mixing processes. These results
 suggest that the common assumption in a Rayleigh distillation model that water
 vapor originates from a single source, is unable to reproduce the water vapor
 445 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 humid-
 450 ity 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 δD_v at 850 hpa level from different areas were
 455 clearly observed in each season (see supplementary Figure S3). 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 δD_v values in the wet season were more depleted
 than those predicted by Rayleigh distillation. These data can be explained by
 460 shallow dry convection. Strong shallow dry convective activity generates water
 vapor δD_v below the Rayleigh curve, in a process known as the isotope amount
 effect (Bony et al., 2008; Galewsky and Samuels-Crow, 2014a,b). However, more
 evidence is required to confirm this.

4. Conclusions

465 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
470 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 δD_v and $\delta^{18}O_v$ were insignificant.
- 475 (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
480 suggested that water vapor appeared to be isotopically in equilibrium with surrounding raindrops during a precipitation event. However, the equilibrium predictions from monthly δD_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
485 increased transpiration and lowest in DJF ($7.1 \pm 13.2\%$). Our results indicate that the primary control on near surface water vapor δD variability is atmospheric circulation, with local evapotranspiration playing a secondary role.

490 (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 δD_v could primarily be attributed to the advection of moisture from different source regions.

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Figure 1: 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. After correction, 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, $\delta D=183.1 \pm 1.1$ ‰, Total number $n=1516$), 2014/09/16-2014/09/17 ($Q=6500 \pm 700$ ppmv, $\delta D=184.3 \pm 3.3$ ‰, $n=2109$) and 2015/03/31-2015/04/01 ($Q=3700 \pm 100$ ppmv, $\delta D=183.2 \pm 3.2$ ‰, $n=1136$).

Figure 2: Time series of daily observation in precipitation amount (P), vapor mixing ratios (Q), water vapor and precipitation $\delta^{18}\text{O}$, δD and $d - excess$ (blue line for tower observations, green square for precipitation). The red lines represent the simulated water vapor mixing ratio and water vapor isotope ratios at 2-m level by IsoGSM.

Figure 3: Monthly averaged relative humidity (RH , dash), Temperature (T_a , solid), isotope ratio δD , 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 4: Comparison between the observed water vapor δD_v and precipitation predicted $\delta D_{v,e}$. Each point represents a rainy day.

Figure 5: 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 6: IsoGSM simulated seasonal mean latent heat fluxes. Black dot shows the study site

Figure 7: The atmospheric column precipitable water contributed from tagged boxes in JJA. Source regions for tagging experiments of water vapor tagged within 10×10 degree boxes (contour): a, 25N-35N, 120E-130E; b, 35N-45N, 120E-130E; c, 25N-35N, 130E-140E; d, 35N-45N, 130E-140E; e, 25N-35N, 140E-150E; f, 35N-45N, 140E-150E.

Figure 8: The contribution fraction from tagged boxes in JJA, SON, DJF and MAM.

Figure 9: 850 hpa isotope ratios documented based on the wind direction (0° to 90°, Area f; 90° to 180°, Area e; 180° to 270°, Area a+c; 270° to 360°, Area b+d).

Figure 10: The variation of hourly water vapor δD as a function of specific humidity q for JJA, SON, DJF and MAM. Rayleigh distillation curves representing a pseudoadiabatic process based on oceanic temperatures with an initial δD values of -80 ‰ (approximate value of vapor in equilibrium with ocean, shown as blue dash line) are shown as green solid line (15 °C) and blue solid line (25 °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.

Table 1: One year monthly mean values of observed isotope ratios of atmospheric water vapor, precipitation, and the vapor in Equilibrium with precipitation (δD , $\delta^{18}O$, and $d - excess$) 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 (per mil)			Precipitation			Vapor in equilibrium			RH (%)	T_a (°C)	P (mm)	Q (ppmv)
	δD	$\delta^{18}O$	$d-excess$	δD	$\delta^{18}O$	$d-excess$	δD	$\delta^{18}O$	$d-excess$				
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	22796
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	26261
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	28825
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	22766
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	17494
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	5253
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	4581
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	5385
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	7182
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	9774
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	15602

Table 2: The contribution of evapotranspiration (F_{ET}) to that of the ABL water vapor estimated from monthly water vapor δD , background isotope ratio δ_{bg} and evapotranspiration isotope ratio δ_{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	δD_v	δ_{bg}	δ_{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	$0.11 \pm 8.3\%$
2014	4	-106.4	-124.5	-51.8	$24.9 \pm 12.7\%$
2014	5	-117.2	-131.1	-62.5	$20.2 \pm 14.2\%$