# Hydrologic implications of the isotopic kinetic fractionation of open-water evaporation

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**Abstract**    The kinetic fractionation of open-water evaporation against the stable water isotope $H_2^{18}O$ is an important mechanism underlying many hydrologic studies that use $^{18}O$ as an isotopic tracer. A recent *in-situ* measurement of the isotopic water vapor flux over a lake indicates that the kinetic effect is much weaker (kinetic factor 6.2‰) than assumed previously (kinetic factor 14.2‰) by lake isotopic budget studies. This study investigates the implications of the weak kinetic effect for studies of deuterium excess-humidity relationships, regional moisture recycling, and global evapotranspiration partitioning. The results indicate that the low kinetic factor is consistent with the deuterium excess-humidity relationships observed over open oceans. The moisture recycling rate in the Great Lakes region derived from the isotopic tracer method with the low kinetic factor is a much better agreement with those from atmospheric modeling studies than if the default kinetic factor of 14.2‰ is used. The ratio of transpiration to evapotranspiration at global scale decreases from 84±9% (with the default kinetic factor) to 76±19% (with the low kinetic factor), the latter of which is in slightly better agreement with other non-isotopic partitioning results.

**Keywords**    Kinetic fractionation factor, Craig-Gordon model, Moisture recycling, Evapotranspiration partitioning, Deuterium excess

## 1.    Introduction

Evaporation from water bodies is important in hydrological and meteorological studies for a number of reasons. Precise quantification of evaporation is a prerequisite for studies on water budgets of lake catchments and also on evapo-transpiration partitioning at the regional and global scale

(Coenders-Gerrits et al., 2014; Gibson et al., 2016). Because lake evaporation provides moisture to the local atmosphere, accurate estimation of the moisture recycling rate helps to improve the ability to calculate the atmospheric moisture budget and local weather prediction (Bryan et al., 2015). Furthermore, knowledge on evaporation also provide a reference state for paleoclimate reconstruction using isotopic tracers (Jouzel et al., 2007).

Because the stable isotopes of water provide unique constraints on evaporation and the water cycle, they have been

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widely used in studies of lake evaporation and the associated hydrological and meteorological processes. These isotope-based studies take advantage of discrimination against the heavier isotopoluges ($H_2^{18}O$ and $H^2HO$) than the lighter one ($H_2^{16}O$), or fractionation effects, during the phase change from liquid water to water vapor. Because the saturation water vapor pressure and molecular diffusivities of $H_2^{18}O$ and $H^2HO$ are lower than those of $H_2^{16}O$, it is easier for the lighter water molecules to escape from liquid water and the heavier molecules tend to accumulate in the liquid phase (Merlivat, 1978; Merlivat and Jouzel, 1979; Cappa et al., 2003). The isotopic fractionation associated with the saturation vapor pressure difference is called the equilibrium effect and that associated with the diffusivity difference is called the kinetic effect. The imprints on the vapor and on the liquid caused by these effects are the fundamental basis for the applications of the isotopic method in water cycling studies. These studies span a range of spatial scales, ranging from quantification of evaporation and water mixing of individual lakes (e.g., Kabeya et al., 2008; Yi et al., 2008; Gibson and Reid, 2010; Wassenaar et al., 2011; Jasechko et al., 2014; Gibson et al., 2016, 2017), to regional moisture recycling (Gat et al., 1994; Machavaram and Krishnamurthy, 1995; Bowen et al., 2012), and regional and global evapotranspiration partitioning (e.g., Jasechko et al., 2013; Kool et al., 2014). Furthermore, the isotopic method is also used in general circulation models to provide additional constraints on the water cycle (e.g., Risi et al., 2010; Dee et al., 2015; Werner et al., 2011, 2016) and in paleoclimate reconstruction from ice cores (Jouzel et al., 2007; Steffensen et al., 2008).

The applications cited above all require accurate quantification on the isotopic composition of evaporation ($\delta_E$). Generally, $\delta_E$ is calculated with the model developed by Craig and Gordon (1965, referred to as Craig-Gordon model hereafter). According to the Craig-Gordon model, $\delta_E$ is determined by the isotopic compositions of atmospheric water vapor ($\delta_V$) and liquid water ($\delta_L$), atmospheric humidity referenced to water surface temperature ($h$), and factors describing the isotopic equilibrium and kinetic fractionation effects. Except for the kinetic fractionation factor ($\varepsilon_k$), all the inputs can be measured directly or approximated by robust empirical relationships.

The precise value of $\varepsilon_k$ in the natural environment is a subject of debate. In the hydrological literature, values of 14.2‰ for $H_2^{18}O$ and 12.5‰ for $H^2HO$, referred to as the LK values hereafter, have been widely used to describe the kinetic effect of lake evaporation (Gonfiantini, 1986). However, for ocean surface (OS) evaporation, the kinetic effect is expressed as a function of wind speed according to the physical model developed by Merlivat and Jouzel (1979), with a typical value of about 6.2‰ for $H_2^{18}O$ and a value of 5.5‰ for $H^2HO$ under the smooth regime. These OS values suggest that the kinetic effect is much weaker than assumed

in most lake evaporation studies.

This study aims to quantify the uncertainty the traditional LK $\varepsilon_k$ introduces to studies of the atmospheric water cycle. Specifically, we are interested in the impact of the kinetic factor on moisture recycling rate calculations and on global partitioning of terrestrial evapotranspiration (*ET*) into component contributions. This study complements a recent study we published (Xiao et al., 2017). In that study, we showed that the annual evaporation of Lake Taihu based on the lake isotopic mass balance is very sensitive to $\varepsilon_k$, increasing by 72% if the OS $\varepsilon_k$ is used for the $^{18}O$ kinetic effect as compared to the evaporation estimate using the LK $\varepsilon_k$ for $^{18}O$. We also showed that the evaporation estimate using the OS $\varepsilon_k$ is in good agreement with the evaporation rate measured independently by eddy covariance. That study is concerned with the water cycle at a local site. The present study extends the analysis to the regional (moisture recycling) and the global scale (*ET* partitioning).

## 2. Theory on isotopic evaporation

The isotopic compositions of lake evaporation ($\delta_E$) is given by Craig and Gordon (1965) as

$$\delta_E = \frac{\alpha_{eq}^{-1}\delta_L - h\delta_V - \varepsilon_{eq} - (1-h)\varepsilon_k}{1 - h + 10^{-3}(1-h)\varepsilon_k},\quad (1)$$

where $\delta_V$ and $\delta_L$ are the isotopic compositions of atmospheric water vapor and liquid water, respectively, $h$ is relative humidity in fraction in reference to water surface temperature $T_s$, and $\varepsilon_{eq}$ and $\varepsilon_k$ are equilibrium and kinetic fractionation factors expressed in per mil, respectively, with the former related to the equilibrium factor $\alpha_{eq}$ as $\varepsilon_{eq}=(1-1/\alpha_{eq})\times10^3$. Here $\alpha_{eq}$ is a function of $T_s$ (Majoube, 1971).

While the function described by Majoube (1971) for $\alpha_{eq}$ is widely accepted, there is no consensus on how $\varepsilon_k$ should be calculated. A general parameterization relates $\varepsilon_k$ to the diffusivity ratio, as (Gonfiantini, 1986; Cappa et al., 2003; Lee et al., 2009),

$$\varepsilon_k = n\left(1 - \frac{D_i}{D}\right) \times 1000\,‰,\quad (2)$$

where $D_i$ and $D$ are molecular diffusivity of the heavy isotopologue ($H_2^{18}O$ or $H^2HO$) and that of $H_2^{16}O$, respectively, and $n$ is an aerodynamic parameter. According to Craig and Gordon (1965), $n$ falls between the value of zero and unity. If the thin interfacial air layer (several mm in thickness) above the water surface is fully turbulent, $n$ is equal to zero, and no kinetic fractionation occurs. If diffusion in the interfacial layer is completely molecular, $n$ is equal to one. Under natural conditions, both molecular and turbulent diffusion are expected. For diffusion through the laminar boundary layer of plant leaves, $n$ is 2/3 (Farquhar and Lloyd, 1993). In most lake studies, a value of $n=1/2$ is used, which is equivalent to

having $\varepsilon_k$ of 14.2‰ for $H_2^{18}O$ and 12.5‰ for $H^2HO$ (Gonfiantini, 1986; Jasechko et al., 2013; Skrzypek et al., 2015; Gibson et al., 2016).

In the studies of ocean evaporation, the kinetic factor is parameterized as a function of wind speed (Merlivat and Jouzel, 1979). This parameterization recognizes that because input variables of the Craig-Gordon model are measured at some height above the interfacial layer, the diffusion pathway consists of both the molecular part in the interfacial layer and the turbulent part in the atmospheric surface layer. In the study of Merlivat and Jouzel (1979), $\varepsilon_k$ is an implicit function of wind speed at the height of 10 m above the surface. Here for convenience of computation, this function in the smooth regime is approximated by a fourth-order polynomial as (Appendix 1, http://earth.scichina.com),

$$\varepsilon_{k,18} = 4.6351u^4 \times 10^{-4} - 0.01465u^3 \\ + 0.1696u^2 - 0.9261u + 7.9230, \quad (3)$$

for $H_2^{18}O$, and

$$\varepsilon_{k,D} = 4.0917u^4 \times 10^{-4} - 0.01293u^3 \\ + 0.1497u^2 - 0.8174u + 6.9882, \quad (4)$$

for $H^2HO$, where $u$ is wind speed in m at the height of 10 m above the surface. Under typical wind conditions over a lake, $\varepsilon_{k,18}$ is about 6.2‰ and $\varepsilon_{k,D}$ is about 5.5‰. These OS values are less than half of the LK values, suggesting that the kinetic effect of open-water evaporation may be much weaker than assumed by the LK values.

The Craig-Gordon model can be rearranged to isolate the kinetic effect on the vapor isotopic compositions. Deuterium excess ($d$), a measure of the $H^2HO$ abundance relative to the $H_2^{18}O$ abundance, is given as

$$d = \delta^2 H - 8\delta^{18}O. \quad (5)$$

Because the equilibrium effect of $H^2HO$ is about 8 times that of $H_2^{18}O$, $d$ is a tracer that preserves the kinetic effect on evaporation. Combining eq. (5) with the Craig-Gordon expressions for $H^2HO$ and $H_2^{18}O$, we obtain the deuterium excess of evaporation ($d_E$), as

$$d_E = \frac{d_L - h d_V - (\varepsilon_{eq,D} - 8\varepsilon_{eq,18}) - (1-h)(\varepsilon_{k,D} - 8\varepsilon_{k,18})}{1-h}, \quad (6)$$

where $d_V$ and $d_L$ are deuterium excess of atmospheric water vapor and lake water, respectively. Since $\left(\varepsilon_{eq}^D - 8\varepsilon_{eq}^{18}\right)$ is close to zero, the equation can be simplified to:

$$d_E = \frac{d_L - h d_V}{1-h} - \left(\varepsilon_{k,D} - 8\varepsilon_{k,18}\right). \quad (7)$$

On the global scale, an isotopic closure assumption generally holds, in that the isotopic compositions of water vapor in the atmospheric surface layer are equal to those of the evaporation flux, that is (Merlivat and Jouzel, 1979; Jouzel and Koster, 1996)

$$d_E = d_V. \quad (8)$$

Manipulation of eqs. (7) and (8) yields the following linear relationship between $d_E$ (and $d_V$) and relative humidity,

$$d_V = d_E = d_L + (8\varepsilon_{k,18} - \varepsilon_{k,D}) - (8\varepsilon_{k,18} - \varepsilon_{k,D})h. \quad (9)$$

So the deuterium excess of evaporation (and water vapor) should increase linearly with decreasing relative humidity and the linear line slope of $d_V$ versus $h$ is given by $-\left(8\varepsilon_{k,18} - \varepsilon_{k,D}\right)$.

This isotopic closure assumption, originally made for the whole water cycle, has been questioned at the regional scale by a GCM modeling study (Jouzel and Koster, 1996). However, recent observations of atmospheric vapor in the surface layer over the open oceans offer support for eq. (9) (Uemura et al., 2008; Benetti et al., 2014; Steen-Larsen et al., 2014, 2015).

In the following, we will use eq. (9) to check the validity of the two sets of kinetic factor values using the published data on the $d_v$ versus $h$ relationship.

## 3.  Data and methods

### 3.1  Data on deuterium excess versus humidity

To test the effect of kinetic fractionation factor on the relationship between deuterium excess and humidity described by eq. (9), we used published data on water vapor isotope compositions measured over the Mediterranean Sea, the Southern Ocean and North Atlantic Ocean. (1) Gat et al. (2003) conducted a shipboard sampling campaign over the Mediterranean Sea (32.498°–41.460°N, 2.606°–32.843°E). They collected water vapor samples from the mast height (27.9 m) using a cold trap at a total of 25 locations. (2) In the eastern Mediterranean, measurement of isotopic water vapor was conducted on a rooftop (about 6 m above the ground) at the Weizmann Institute of Science in Rehovot, Israel (31.9°N, 34.8°E, 76 m above sea level) from October 2000 to 2006. Time interval was around 8 h and 265 measurements were used (Pfahl and Wernli, 2008). (3) Over the Southern Ocean, shipboard measurement was conducted from 4 to 30 Jan 2006, in a latitude range from 33.47° to 66.83°S and a longitude range from 20.11° to 114.35°E. The atmospheric water samples were collected 2 to 3 times per day, and a total of 60 vapor samples was measured (Uemura et al., 2008). (4) *In-situ* and continuous measurements on isotopic water vapor using laser isotope instruments was conducted at the Bermuda Islands (32.26°N, 64.88°W) in the North Atlantic from November 2011 to June 2013 (Steen-Larsen et al., 2014). (5) Continuous measurement was made on the top of a lighthouse at Selvogsviti on the south corner of Iceland (63.83°N, 21.47°W) between November 2011 and April 2013 (Steen-Larsen et al., 2015), representing the North Atlantic. (6) Continuous measurement at the 20 m height above the sea surface was conducted on a ship over the

subtropical Eastern North Atlantic Ocean (26°N, 35°W) from 16 August to 10 September 2012 (Benetti et al., 2014).

In the following analysis, air humidity ($h$) was normalized to water surface temperature.

### 3.2 Moisture recycling over the Great lakes

#### 3.2.1 Basic principle

Because the deuterium excess of atmospheric vapor is a conserved quantity during atmospheric transport, the water recycling ratio *f*, or the relative contribution of evaporation of a lake to the local atmospheric moisture, can be expressed as (Gat et al., 1994)

$$f = \frac{d_{V,downwind} - d_{V,upwind}}{d_E - d_{V,upwind}}, \tag{10}$$

where $d_{V,\,upwind}$ and $d_{V,\,downwind}$ represent deuterium excess of atmospheric water vapor upwind and downwind of the lake, $d_E$ is the deuterium excess of lake evaporation.

The denominator ($d_E - d_{V,\,upwind}$) can be derived from the Craig-Gordon model. If the isotopic composition of water vapor and humidity is observed in the upwind region of lakes (referred to as $\delta_{V,\,upwind}$ and $h'$), the Craig-Gordon model can be written as

$$\delta_E = \frac{\alpha_{eq}^{-1}\delta_L - h'\delta_{V,upwind} - \varepsilon_{eq} - (1 - h')\theta\varepsilon_k}{1 - h' + 10^{-3}(1 - h')\varepsilon_k}, \tag{11}$$

where $\theta$ is a weighting factor regarding the relative humidity at water surface ($h$) and that in the turbulently mixed sublayer ($h'$) as

$$\theta = \frac{1 - h}{1 - h'}. \tag{12}$$

Then an equation similar to eq. (7) can derived as

$$d_E = \frac{d_L - h'd_{V,upwind}}{1 - h'} + \theta(8\varepsilon_{k,18} - \varepsilon_{k,D}). \tag{13}$$

Rearranging eq. (13), we have:

$$d_E - d_{V,upwind} \approx \frac{(d_L - d_{V,upwind})}{(1 - h')} + a \times \theta, \tag{14}$$

where $d_L$ was the deuterium excess of the lake water body, and $a = 8\varepsilon_{k,\,18} - \varepsilon_{k,\,D}$. In the study of Gat et al. (1994), the value of $\theta$ is 0.88, and *a* is equal to 107 since the values of $\varepsilon_{k,D}$ and $\varepsilon_{k,18}$ are set to 13‰ and 15‰, respectively. In this study, *a* equals 44.1 since the OS factors were used. A schematic diagram indicating heights and locations of the variables in the above equation was shown in Figure 1.

The reader is reminded that the weighting factor $\theta$ is used to account for the fact that $h$ and $d_V$ are not measured over the lake surface.

In this study, we evaluate the sensitivity of regional moisture recycling for the Great Lakes region in North America. We chose this region because all the input data required by the isotopic method are readily available. The moisture recycling ratio was estimated using the improved
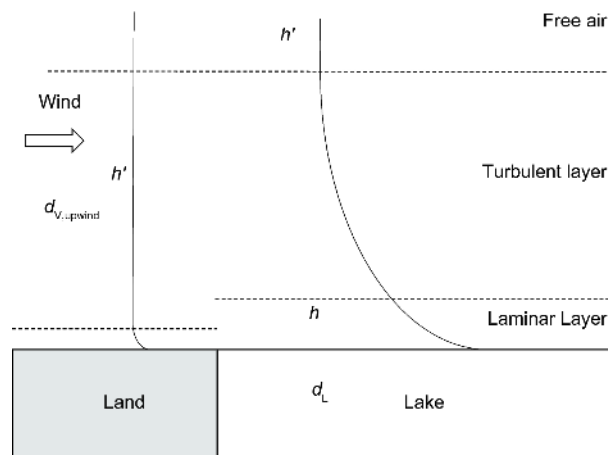


**Figure 1** Schematic diagram indicating heights and locations of the variables in eq. (14) for the moisture recycling ratio estimation. Solid line represent humidity profiles.

isotopic and meteorological data given in the study of Jasechko et al. (2014) and the same methodology described by Gat et al. (1994). Although the analysis was done for one region, we expect the OS $\varepsilon_k$ to give equally robust assessment of moisture recycling for other regions of the world.

#### 3.2.2 Data sources and uncertainty analysis

Here, the relative humidity referenced to lake temperature was calculated from the specific humidity and lake temperature for each lake in the Great Lakes region of North America (Jasechko et al., 2014). The mean value is $h$=64.3 ±9.3% (mean±1 standard deviation). The d-excess of atmospheric water vapor was calculated from the measured d-excess of precipitation according to the precipitation-equilibrium assumption. The annual mean d-excess of the incoming vapor ($d_{V,\,upwind}$), determined with the precipitation data collected at Atikokan, Trout Lake, Lake Geneva and Chicago, is 12.2±3.8‰. The annual mean d-excess of water vapor downwind of the Great Lake region ($d_{V,\,downwind}$), determined from the precipitation data collected at downwind locations (Otawa, Chutauqua, Ste. Agathe and Aurora) is 16.4±3.2‰. Thus, the increase of deuterium-excess of atmospheric vapor, $d_{V,\,downwind} - d_{V,\,upwind}$, is 4.2‰.

Uncertainty estimates for the moisture recycling calculation were made with a Monte Carlo procedure employing a Gaussian distribution for errors in input variables and an ensemble of 800000 realizations. The top and bottom 0.5% outliers were removed before computing the mean values and standard deviations. The error ranges of the input variables are given in the paragraph above.

### 3.3 Global evapotranspiration partitioning

#### 3.3.1 Theory of isotopic mass balance and watershed ET partitioning

The ratio of transpiration to evapotranspiration on the global

scale can also be calculated based on the isotopic mass balance (Jasechko et al., 2013). For a lake catchment, the hydrologic steady state can be expressed as the balance between inputs and outputs of the water budget

$$I = xP + E + T + Q, \tag{15}$$

where $I$ is water input (including precipitation and inflows from upstream lakes), $xP$ is precipitation interception, $E$ is open-water and soil evaporation, $T$ is transpiration, $Q$ is river runoff or ground water discharge.

For the lake catchment, the stable isotope mass balance can be written as

$$\delta_I I = \delta_P xP + \delta_E E + \delta_T T + \delta_Q Q, \tag{16}$$

where $\delta_I$, $\delta_P$, $\delta_E$, $\delta_T$ and $\delta_Q$ represent the isotopic compositions of water input, precipitation, open-water and soil evaporation, transpiration and river runoff or ground water discharge, respectively.

Combining the two equations above, we obtain the catchment transpiration as

$$T = \frac{I(\delta_I - \delta_E) - Q(\delta_Q - \delta_E) - xP(\delta_P - \delta_E)}{\delta_T - \delta_E}. \tag{17}$$

To estimate the global transpiration, the deuterium excess of the water cycle components was used instead of the $\delta$ value as

$$T = \frac{P(d_P - d_E) - Q(d_Q - d_E) - xP(d_P - d_E)}{d_T - d_E}, \tag{18}$$

where $d$ is deuterium excess, the subscripts P, E, Q and T represent precipitation, evaporation, water lost through river discharges and transpiration. In the original paper of Jasechko et al. (2013), the LK values were used for kinetic fractionation factor and the isotopic compositions of open-water and soil evaporation were grouped into $\delta_E$, which was calculated from eq. (1). Here, the OS factors were used as default.

Once $T$ is known, the transpiration fraction is computed from eq. (15), as

$$\frac{T}{xP + E + T} = \frac{\dfrac{P(d_P - d_E) - Q(d_Q - d_E) - xP(d_P - d_E)}{d_T - d_E}}{P - Q}. \tag{19}$$

### 3.3.2   Input data and uncertainty range

To calculate global terrestrial transpiration, each individual variable in the combined water budget and isotopic budget equation (eq. (18)) was integrated to a global value according to Jasechko et al. (2013). Briefly, the terrestrial precipitation was obtained from a surface climate data set (New et al., 2002; $P$=110000±10000 km$^3$ yr$^{-1}$). The global canopy interception ratio ($x$) was based on a satellite study (Miralles et al., 2010), giving an interception amount of $xP$= 7500±1500 km$^3$ yr$^{-1}$. River discharges to the oceans was based on a river-based estimate from atmospheric moisture

budget reanalysis conducted by Dai and Trenberth (2002) ($Q$=37300±700 km$^3$ yr$^{-1}$). The deuterium excess of discharge was calculated based on the isotopic measurement of 31 of the world's 200 largest rivers and was weighted by mean annual discharge of each river ($d_Q$=6.8±3.8‰) (Jasechko et al., 2013). The deuterium excess of precipitation ($d_P$) was based on the data from the IAEA/WMO network stations (Rozanski et al., 1993) ($d_P$=9.5±1‰). The deuterium excess of transpiration was estimated as the spatially weighted mean value of $d_P$ by mean long-term normalized difference vegetation indices (NDVI) ($d_T$=8±3‰). The deuterium excess of terrestrial evaporation was calculated by averaging the deuterium excess of evaporation for 73 lakes globally, giving a mean value of $d_E$=75±30‰ if the LK $\varepsilon_k$ values were used (Jasechko et al., 2013) and $d_E$=18±30‰ if the OS values were used.

Uncertainty estimates of the transpiration fraction were also made with the Monte Carlo procedure described in Section 3.2.2. The error ranges of the input variables are given in the paragraph above.

## 4.   Results and discussion

### 4.1   Relationship between deuterium excess and humidity

The LK and OS kinetic factors are theoretical values based on different assumptions about the role of turbulence in the diffusion process near the water surface. Direct measurement of $\delta_E$ at Lake Taihu indicates that the OS $\varepsilon_k$ is more appropriate than the LK $\varepsilon_k$ for describing the kinetic effect of open-water evaporation. However, it is not clear if the results obtained at Lake Taihu can be extended to other water bodies where turbulence regimes may be different. Here we offer additional evidence for the OS $\varepsilon_k$ by examining the relationship between deuterium excess and humidity. If the kinetic fractionation parameterization is accurate, the predicted slope in eq. (9) should agree with the actual slope measured under conditions of isotopic closure state, such as over the open ocean.

To investigate the sensitivity of deuterium excess-humidity relationship to humidity, we have compiled several recently published data sets on the vapor isotope ratios observed in the marine atmospheric surface layer as described in Section 2.4.1. Because these measurements were made on ships or very close to an oceanic vapor source, the assumption of isotopic closure should hold, and the theoretical relationship between deuterium excess versus relative humidity (eq. (9)) can be evaluated against the observations.

The observational results are presented in Figure 2, along with the theoretical relationships based on the OS and the LK kinetic parameters. The OS parameters yield a slope value of −0.44 per mil per percent relative humidity change (‰ %$^{-1}$),
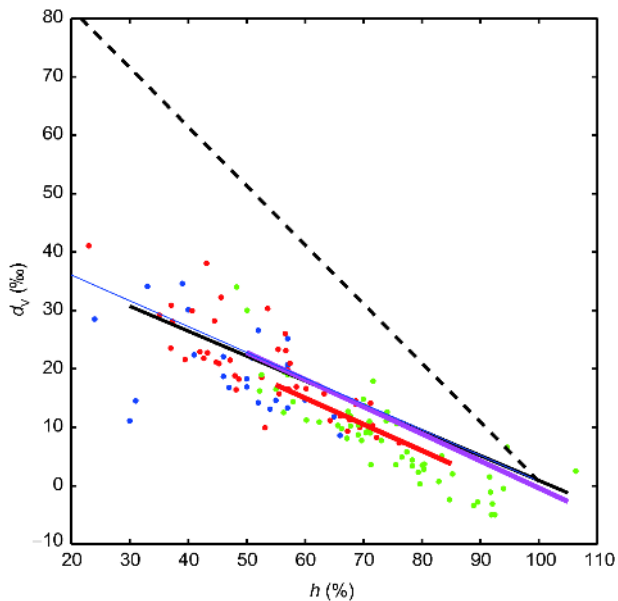
**Figure 2** Deuterium excess of atmospheric vapor versus relative humidity referenced to water surface temperature. Red dots, Eastern Mediterranean Sea (Pfahl and Wernli, 2008); blue dots, Mediterranean Sea (Gat et al., 2003); green dots, the Southern Ocean (Uemura et al., 2008); solid black line, North Atlantic (Steen-Larsen et al., 2014); red line, the subtropical Eastern North Atlantic Ocean (Benetti et al., 2014); pink line, the south coast of Iceland (Steen-Larsen et al., 2015); solid blue line, theoretical line with the OS kinetic factors; dashed black line, theoretical line with the LK factors. Error bars are ±1 standard deviation.

which is in broad agreement with the published observations, offering further evidence of a weak kinetic effect. We note that the original parameterization of Merlivat and Jouzel (1979) consists of a smooth region and a rough regime. The observations presented by Steen-Larsen et al. (2014) (and the other cited studies) are an indirect validation of the OS parameterization for smooth conditions (the $^{18}O$ $\varepsilon_k$ value 6.2‰; wind speed less than 6 m s$^{-1}$), but not validation of the values of 2‰ to 4‰ predicted for rough conditions (wind speed greater than 7 m s$^{-1}$). In comparison, the slope resulting from the LK kinetic parameters is too steep (−1.01‰ per % relative humidity change).

The results presented in Figure 2 confirms the conclusion reached by Xiao et al. (2017) that the OS $\varepsilon_k$ as given by eqs. (3) and (4) should be used in conjunction with the Craig-Gordon model for calculating the isotopic compositions of evaporation.

## 4.2 Moisture recycling over the Great Lakes region

Figure 3 compares the recycle rate calculated with the isotopic method with that obtained by reanalysis and regional climate models (Bryan et al., 2015). The reanalysis data sets included the North American Regional Reanalysis (NARR) (Mesinger et al., 2006) and the European Centre for Medium-Range Weather Forecasts Reanalysis (ERA) Interim reanalysis data set (Dee et al., 2011). The regional climate

model (RegCM; Giorgi et al., 2012) was driven by the EAR interim reanalysis data set, the National Oceanic and Atmospheric Administration (NOAA) Geophysical Fluid Dynamics Laboratory Earth System Model using the Modular Ocean Model version 4.1 (GFDL-ESM2M) (Dunne et al., 2012, 2013) and the Met Office Hadley Centre Hadley Global Environment Model 2 Earth System model (HadGEM2-ES) (Collins et al., 2011; Martin et al., 2011). Using the lake $\varepsilon_k$, the evaporated water is too depleted of $H_2^{18}O$, resulting in high bias in $d_E$. In order to maintain the $H_2^{18}O$ mass balance in the atmosphere, we needed an exceedingly low estimate of evaporative contribution or water recycling rate (5.7% to 9.5%) to the atmospheric water pool. The mean recycling rate is 7.4±0.5% (mean ±1 standard deviation), which is similar to the original estimate given by Gat et al. (1994) but is still too low in comparison with the results obtained with atmospheric transport models. These modeling studies indicate a much more intensive moisture recycling, with a recycling rate of 20.7±2.6% (Bryan et al., 2015). This conflict largely vanishes in our reassessment using the OS $\varepsilon_k$ (Figure 3). Using the OS kinetic factors, we obtained a mean recycling rate of 16.8±2.8%.

Our study clearly shows the importance of the kinetic factors in the isotope-based recycling calculation. Other studies have also highlighted other parameters in need of attention (Machavaram and Krishnamurthy, 1995; Bowen et al., 2012). First, the value of $\theta$ may vary from month to month (Bowen et al., 2012). Second, the assumption of isotopic equilibrium between atmospheric vapor and precipitation may be not justified in some regions (Machavaram and Krishnamurthy, 1995). Third, if wind direction is variable between seasons, such as in monsoon climates, the downwind effect will be weakened, and make the estimation difficult because of high noise to signal ratio.

## 4.3 Global Evapotranspiration partitioning

The results of *ET* partitioning in this study are compared with those published in the literature using water isotopes, land surface models, remote sensing and site measurements (Figure 4). Using the LK kinetic factors, we obtained a transpiration fraction of 84±9%. If we used the OS kinetic factors, the transpiration fraction was lowered slightly to 76±19%.

These isotope based estimates are generally higher than other published results. A compilation of ecosystem-scale observations suggests that the global *T/ET* ratio is 61±15% (Schlesinger and Jasechko, 2014). Zhou et al. (2016) estimated that the *T/ET* ratio is bounded between 41% and 68% based on eddy covariance measurements and the correlation between carbon and water flux. Wang et al. (2014) synthesized published experimental studies and concluded that the ratio is equal to 38–77%. Zhang et al. (2016) obtained a ratio
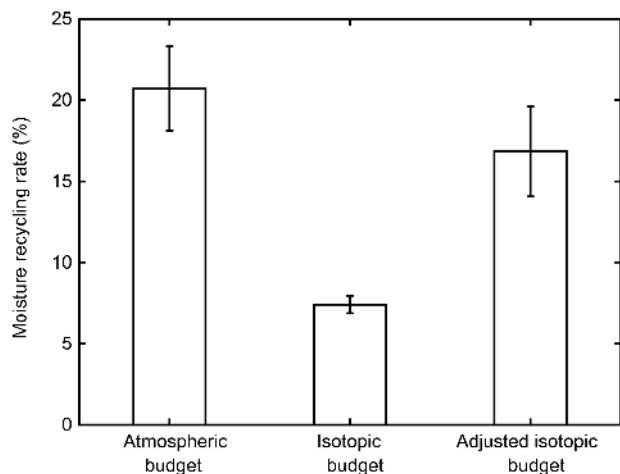
**Figure 3**   Moisture recycling rate for the Great Lakes region. The atmospheric budget value is the mean of two reanalysis and three regional climate model estimates (Bryan et al., 2015). The isotopic budget value is calculated with an isotopic budget method (Gat et al., 1994) using the data for the Great Lakes (Jasechko et al., 2014) and the LK $\varepsilon_k$. The adjusted value is obtained with the same isotopic budget method but using the OS $\varepsilon_k$. Error bars are ±1 standard deviation.

of about 65% using an observation-driven Penman-Monteith-Leuning (PML) modeling study. Miralles et al. (2011) arrived at an estimate of 80% using a process-based methodology and satellite data. Miralles et al. (2016) obtained an estimate of 76%, 24% and 56%, using a remote sensing-based *ET* product (Global Land Surface Evaporation: The Amsterdam Model, GLEAM), the Penman-Monteith algorithm behind the official Moderate Resolution Imaging Spectroradiometer (MODIS) evaporation product (PM-MOD), and the Priestley-Taylor Jet Propulsion Laboratory model (PT-JPL), respectively. Wei et al. (2017) combined global evapotranspiration estimates from remote sensing and land surface model together with the relationship between LAI and *T/ET* for each vegetation class and got an estimate

of 57.2±6.8%. Wang and Dickinson (2012) gave an average value of 58% according to global land surface modeling studies. According to 9 climate models including GSWP-2, CMIP5, STEAM, IsoMatsiro, CLM3, CLM3.5, CLM4CN, CLM4CNE and CLM4SP, the global transpiration ratio is about 43% (Dirmeyer et al., 2006; Yoshimura et al., 2006; Lawrence et al., 2007, 2011; Wang-Erlandsson et al., 2014). Maxwell and Condon (2016) obtained an estimate of 62 ±12% based on groundwater-based partitioning coupled with vegetation and land-energy processes. Fatichi and Pappas (2017) obtained a value of 70±9% using a mechanistic ecohydrological model.

The isotope methodology of Jasechko et al. (2013) was challenged by several studies. Coenders-Gerrits et al. (2014) argued that if a larger and more realistic values for runoff and interception are used, the transpiration ratio will be reduced to 35–80%. Another isotopic-based global assessment using a much higher rainfall interception estimate yields a low transpiration ratio of 64±13% (Good et al., 2015). Schlaepfer et al. (2014) argued that the relative importance of transpiration is overestimated by Jasechko et al. (2013) due to two invalid assumptions: (1) that lake water integrates all processes (e.g. soil evaporation, snow sublimation and isotopic exchange effects in upland ecosystems) and areas (e.g. arid soil) over the entire catchment, and (2) that the mean deuterium excess of 73 catchments can represent global water fluxes.

In this study, we showed that another implicit assumption of this isotopic method, that the LK kinetic fractionation factors can be used to describe isotopic effects of lake evaporation, is invalid. However, despite that adoption of the alternative, OS-based kinetic factors rendered the estimate of *T/ET* to be in a closer agreement with other global estimates (Figure 4), the estimate is still biased too high. In other words, an improved global *ET* partitioning using the isotopic
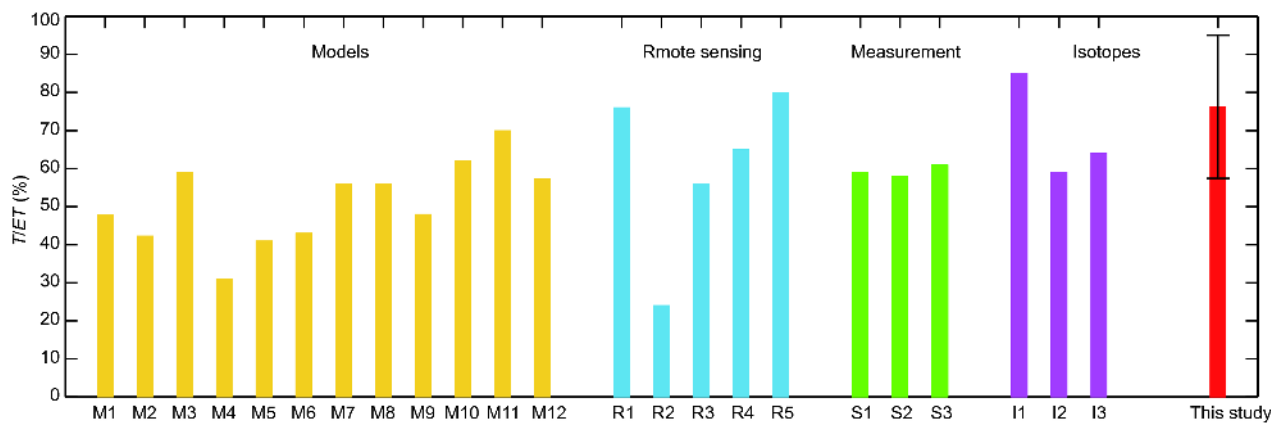


**Figure 4**   Ratio of transpiration to total evaporation on global scale. Models include M1: GSWP-2 (Dirmeyer et al., 2006), M2: CMIP5 (Wei et al., 2017), M3: STEAM (Wang-Erlandsson et al., 2014), M4: IsoMatsiro (Yoshimura et al., 2006), M5: CLM3 (Lawrence et al., 2007), M6–M9: CLM3.5, CLM4CN, CLM4CN3, CLM4SP (Lawrence et al., 2011), M10: ground-water based model (Maxwell and Condon, 2016), M11: (Fatichi and Pappas, 2017), M12: (Wei et al., 2017). Results based on remote sensing include R1–R3: GLEAM, PMMOD and PTJPL (Miralles et al., 2016), R4: PML (Zhang et al., 2016), R5 (Miralles et al., 2011). Site measurements include S1–S3 (Wang and Dickinson, 2012; Schlesinger and Jaschko, 2014; Zhou et al., 2016). Isotopic method include I1–I3 (Jasechko et al., 2013; Coenders-Gerrits et al., 2014; Good et al., 2015) and this study.

method requires not only accurate parameterization of the kinetic effect but also improved estimates of other terms of the water budget.

In comparison to the global $T/ET$ ratio, lake evaporation based on the isotopic mass balance is much more sensitive to the kinetic factor values. For instance, evaporation rate over Lake Taihu calculated using the OS $\varepsilon_k$ for $^{18}$O is 72% higher than if the LK value is used (Xiao et al., 2017).

## 5. Conclusions

The deuterium-humidity relationships observed over the ocean confirm that the OS kinetic factor (~6.2‰ for $^{18}$O) is a more appropriate description of the kinetic fractionation of open-water evaporation than the LK kinetic factor (14.2‰).

The isotopic method for hydrological budget analysis shows different sensitivity to the kinetic factor $\varepsilon_k$ at different spatial scales. Previously, we have demonstrated that the estimate of evaporation flux using the $^{18}$O tracer at a single lake is very sensitive to the choice of the kinetic factor. Here we show that the moisture recycling rate at the regional scale is also sensitive to $\varepsilon_k$. Using the OS and the LK kinetic factors, the recycling rate of the Great Lakes region is 16.8 ±2.8% and 7.4±0.5%, respectively, with the former in much better agreement with the recycling rates derived from atmospheric reanalysis. The high sensitivity is largely related to the fact that lake evaporation dominates the surface water vapor flux in the Great Lakes region. In contrast, the estimate of the global transpiration ($T$) to evapotranspiration ($ET$) ratio $T/ET$ is less sensitive to $\varepsilon_k$, decreasing slightly from 84 ±9% if the LK $\varepsilon_k$ is used to 76±19% if the more appropriate OS $\varepsilon_k$ is used. Even with the OS $\varepsilon_k$, the $T/ET$ ratio is still biased high in comparison to other non-isotopic estimates, suggesting that other terms of the global water budget must be improved first before a successful partitioning of the global $ET$ can be achieved via the isotopic method.

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