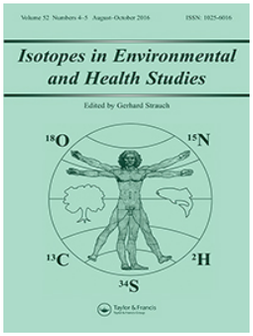

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Spatial distribution and temporal variability of stable water isotopes in a large and shallow lake†

Wei Xiao[a], Xuefa Wen[b], Wei Wang[a], Qitao Xiao[a], Jingzheng Xu[a], Chang Cao[a], Jiaping Xu[a], Cheng Hu[a], Jing Shen[a], Shoudong Liu[a] and Xuhui Lee[a,c]

[a]Yale-NUIST Center on Atmospheric Environment & Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, Nanjing University of Information Science & Technology, Nanjing, People's Republic of China; [b]Key Laboratory of Ecosystem Network Observation and Modeling, Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing, People's Republic of China; [c]School of Forestry and Environmental Studies, Yale University, New Haven, CT, USA

ABSTRACT

Stable isotopic compositions of lake water provide additional information on hydrological, meteorological and paleoclimate processes. In this study, lake water isotopic compositions were measured for more than three years in Lake Taihu, a large and shallow lake in southern China, to investigate the isotopic spatial and seasonal variations. The results indicated that (1) the whole-lake mean δ^2H and δ^{18}O values of the lake water varied seasonally from -48.4 ± 5.8 to -25.1 ± 3.2 ‰ and from -6.5 ± 0.9 to -3.5 ± 0.8 ‰, respectively, (2) the spatial pattern of the lake water isotopic compositions was controlled by the direction of water flow and not by local evaporation rate, and (3) using a one-site isotopic measurement to represent the whole-lake mean may result in unreasonable estimates of the isotopic composition of lake evaporation and the lake water residence time in poorly mixed lakes. The original data, documented here as an online supplement, provides a good reference for testing sensitivity of lake water budget to various isotopic sampling strategies. We propose that detailed spatial measurement of lake water isotopic compositions provides a good proxy for water movement and pollutant and alga transports, especially over big lakes.

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Evaporation; hydrogen-2; isotope hydrology; lake water; oxygen-18; residence time; spatial distribution; seasonal variability; Taihu lake

1. Introduction

Compositions of stable isotopes of lake water provide unique information on hydrological, meteorological and climatic processes. In the process of lake evaporation, the heavier isotopic molecules of water (H^2HO and $H_2^{18}O$) tend to accumulate in the liquid water, causing depletion in the vapour phase. Isotopic measurements of the water can be used to estimate annual evaporation of surface water bodies (e.g. [1–6]), which is an important component of the local hydrological cycle, but is difficult to measure with *in situ* instruments,

CONTACT Wei Xiao wei.xiao@nuist.edu.cn
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especially in remote areas due to cost, time and infrastructure considerations [7]. Lake isotopic measurements are also widely used in research on lake water yield [8,9], water residence time [10,11], water mixing [12,13], and connection with groundwater [14,15]. Furthermore, data on stable isotopes in lakes with long residence times can be used to study the effect of past climate on lake evolution [10,16].

The isotopic composition of lake water is a critical parameter in the applications mentioned above. The literature is rich on data regarding the seasonal isotopic patterns for lake water, driven mostly by precipitation seasonality, and lake ice and snow phenology. Most published studies rely on single-location measurements (e.g. [11,17,18]) or on measurements at a small number of sampling locations (<5; e.g. [4,5]) to represent the whole-lake mean. However, there is evidence of isotopic spatial variability in some lakes [19]. To reduce the uncertainty in lake water balance studies, it is necessary to measure the isotopic composition of lake water at multiple locations and times and to investigate the controlling factors, especially for large lakes.

Here, we investigate the spatial distribution and temporal variability of stable isotopes in Lake Taihu, a large and shallow freshwater lake in the Yangtze River Delta, China. We chose Lake Taihu for four reasons. First, Lake Taihu is a large and shallow lake; therefore, we expect measureable spatial variability. Lake Taihu is the third largest freshwater lake in China, with a mean water depth of 1.9 m, an area of 2400 km^2, a lake length from north to south of 69 km and a width from east to west of 56 km [20]. Second, the lake has distinct inflow and outflow regions throughout the year (Figure 1). Among the 172 rivers and canals connected with Lake Taihu [21], most act as either inflows or outflows all the time. The main exceptions are three waterways whose water flow may be reversed for the purpose of water quality management. Isotope enrichment along the direction of water flow in the lake can be expected. Third, the lake is located in a subtropical monsoon climate, where 45 % of annual precipitation falls in the months of June to August. The spatial isotopic patterns may reveal different roles of precipitation and river input in the rainy season and in the dry season. Fourth, the isotopic study was supported by concurrent measurements at five eddy covariance sites on the lake [22]. Variables measured by the eddy flux network, such as lake evaporation and lake temperature, can help us interpret temporal and spatial patterns seen in the isotope data.

A number of factors may contribute to the spatial variability of the lake water isotopic composition. In particular, we are interested in two competing hypotheses: (1) spatial variations of the lake isotopic composition are indicative of spatial variations in the fraction of the water parcel that has previously evaporated. To put it differently, high H^2HO or $H_2^{18}O$ concentration may indicate high local evaporation rate; and (2) spatial variations in the H^2HO and $H_2^{18}O$ compositions are controlled by 'water age' or time spent in the lake since the water first entered. If this hypothesis holds, the lake isotopic composition may be a good tracer for water movement across this vast lake. These hypotheses are relevant to ongoing efforts to restore the water quality of Lake Taihu. The lake provides the drinking water supply for five million people in cities such as Shanghai, Wuxi and Suzhou [23]. Located in one of the most developed regions in the country, Lake Taihu has been suffering from severe water pollution problems [20]. Algal blooms are common occurrence. A better knowledge of the water flux and movement across the lake may improve predictions of the timing and location of algal growth and the subsequent spread of algal biomass. If our second hypothesis is valid, the isotopic tracers may be a more viable

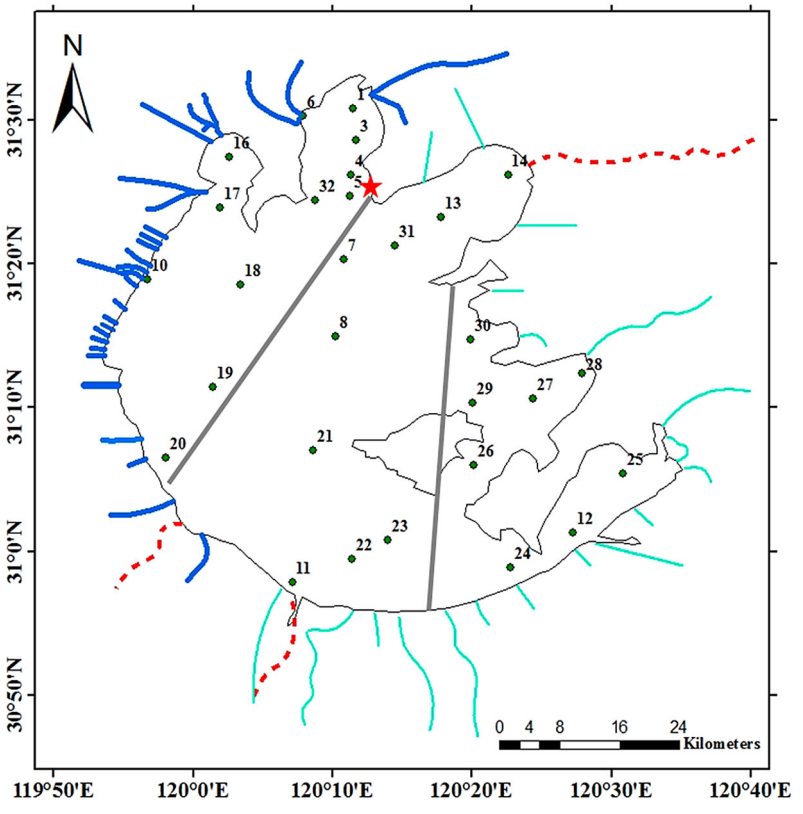

Figure 1. Map of Lake Taihu and the sites of lake and river surveys. Star: the MLW site; dots: lake survey sites; thick blue lines: inflow rivers; thin green lines: outflow rivers; red dashed lines: inflow/outflow rivers (the river at the northeast corner was the Wangyuhe River); grey lines mark approximate boundary of the inflow and outflow regions; created by ArcGIS software (Environmental Systems Research Institute, Inc., Redlands, CA, USA).

method for tracking horizontal water movement than model calculations based on hydrodynamic principles because of the large uncertainties in specifying the inflow and outflow boundary conditions associated with so many waterways for models.

The objectives of this study are (1) to characterize the spatial and temporal variability of the water isotopic composition across Lake Taihu and analyze factors controlling the isotope enrichment patterns; and (2) to evaluate the sensitivity of lake isotopic mass balance and residence time calculations using one-site measurement versus whole-lake sampling.

2. Materials and methods

2.1. Measurement of lake and river water isotopes

Isotope measurements consisted of daily lake water sampling at one location and seasonal lake and river surveys. Since 11 May 2012, one lake water sample was collected daily at 13:00 at a distance of 200 m from the shore. The sampling site is the Taihu Lake Ecosystem Research Station of Chinese Academy of Sciences, located in Meiliang Bay, at the north

shore of the lake (the MLW site, 31°24′N, 120°13′E, Figure 1). The first seasonal lake survey was conducted at 29 sites spread over the whole lake on 15–16 August 2011. One water sample was collected at each location. Subsequent surveys were made every three months in February, May/June, August and November, each survey lasting 1–2 days and covering the same 29 locations in the lake. The first river water survey was conducted in May 2013 on the same days when the lake survey took place. Subsequent river surveys were also timed with the lake surveys. One water sample was collected at ~100 m from each entry or exit point of the lake from 51 rivers (Figure 1). These rivers contributed more than 80 % of the total water inflow and outflow of the lake. Lake and river samples were collected from 20 cm below the surface.

The water samples were collected in 300-mL glass bottles. The bottles were sealed and placed in a refrigerator with temperature of 4 °C before analysis. The samples were analyzed for H^2HO and $H_2^{18}O$ compositions using a liquid water isotope analyzer (Model DLT-100; Los Gatos Research, Mountain View, CA, USA) in the Key Laboratory of Ecosystem Network Observation and Modeling, Chinese Academy of Sciences. Delta notation δ^2H_L and $\delta^{18}O_L$ were used to represent the H^2HO and $H_2^{18}O$ composition in reference to the Vienna Standard Mean Ocean Water (VSMOW). The typical measurement precision was ±0.3 ‰ for δ^2H and ±0.1 ‰ for $\delta^{18}O$. Data collected before the end of 2014 were used in this study.

From August 2011 to May 2012, we also measured vertical profiles of the H^2HO and $H_2^{18}O$ compositions at depths of 20, 80 and 150 cm. Differences among the three depths were less than 0.2 ‰ for whole-lake mean δ^2H_L and less than 0.1 ‰ for $\delta^{18}O_L$. The vertical gradients were small because convective overturning occurs regularly at night [22], and the profile measurement was discontinued.

3. Results and discussion

3.1. Spatial patterns of the lake water isotopes

Figure 2 shows the spatial patterns of the lake isotopic composition averaged over a three-year period (August 2011 to June 2014). A gradual isotopic gradient from the northwest river inflow zone to the southeast river outflow zone is clearly evident. The lowest δ^2H_L and $\delta^{18}O_L$ occurred in the northern part of the lake, with minimum values of -45.8 ± 9.7 and -6.3 ± 1.5 ‰, respectively. The southeast zone was most enriched in H^2HO and $H_2^{18}O$, with maximum values of -28.1 ± 7.5 and -3.7 ± 0.9 ‰. The spatial pattern of deuterium-excess (d_L) was reversed, with higher values in the northwest zone (with a maximum of 5.1 ± 3.2 ‰) and lower values in southeast zone (with a minimum of 1.1 ± 3.5 ‰). For comparison, the annual mean δ^2H (±1 standard deviation) of inflow and outflow rivers was -38.8 ± 6.3 and -30.0 ± 4.8 ‰, respectively (Figure 5).

Isotopic data from individual seasonal surveys are shown in Figure 3 and in the Supplementary Material, Figure S1. With the exception of August, the seasonal spatial patterns are similar to the annual patterns. In August, the northeast portion of the lake was equally enriched as or more enriched in H^2HO and $H_2^{18}O$ than the southeast portion. The range of spatial variability also changed through the year. The minimum spatial variability occurred in June 2014 (with a range of 11.0 ‰ for δ^2H), while the maximum occurred in November 2012 (with a range of 37.1 ‰ for δ^2H).

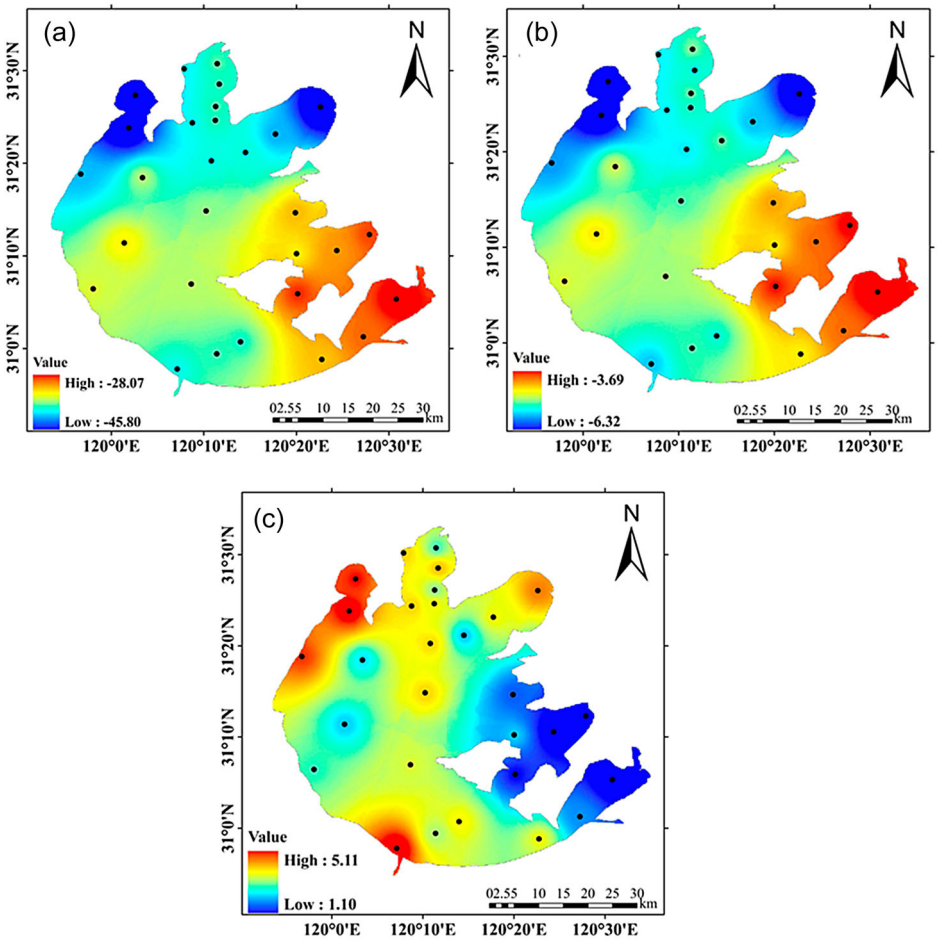

Figure 2. Spatial patterns of the annual mean lake water isotope composition: (a) δ^2H_L; (b) $\delta^{18}O_L$; (c) d_L.

Changes in local evaporation rate were not a contributor to these spatial patterns because eddy covariance data show little spatial variations in water surface temperature and in local evaporation rate [24]. The annual mean δ^2H_L (±1 standard deviation) at location 27 in the southeastern region was -30.9 ± 5.3 ‰, which was 6.6‰ larger than at location 10 near the western shore (Supplementary Material, Figure S1). At these two locations, lake evaporation rate measured by eddy covariance differed by only 1%.

Instead, the progressive enrichment in H^2HO and $H_2^{18}O$ from northeast to southwest was associated with the water flow pattern. As water moves from the inflow to the outflow region, it undergoes evaporation for a longer time and becomes more isotopically enriched. In other words, the spatial variability in the isotopic composition of the lake was associated with the direction of water flow. Not surprisingly, when some of the rivers reversed flow direction, the spatial patterns also shifted. Water was diverted, at times of low water levels, from the Yangtze River through the Wangyuhe River, a reversible waterway at the northeast corner of the lake (Figure 1) in order to raise the lake water level for navigation and for the purpose of pollution dilution [25]. In our study period, water diversion was conducted from November 2011 to February 2012, November 2012 to January

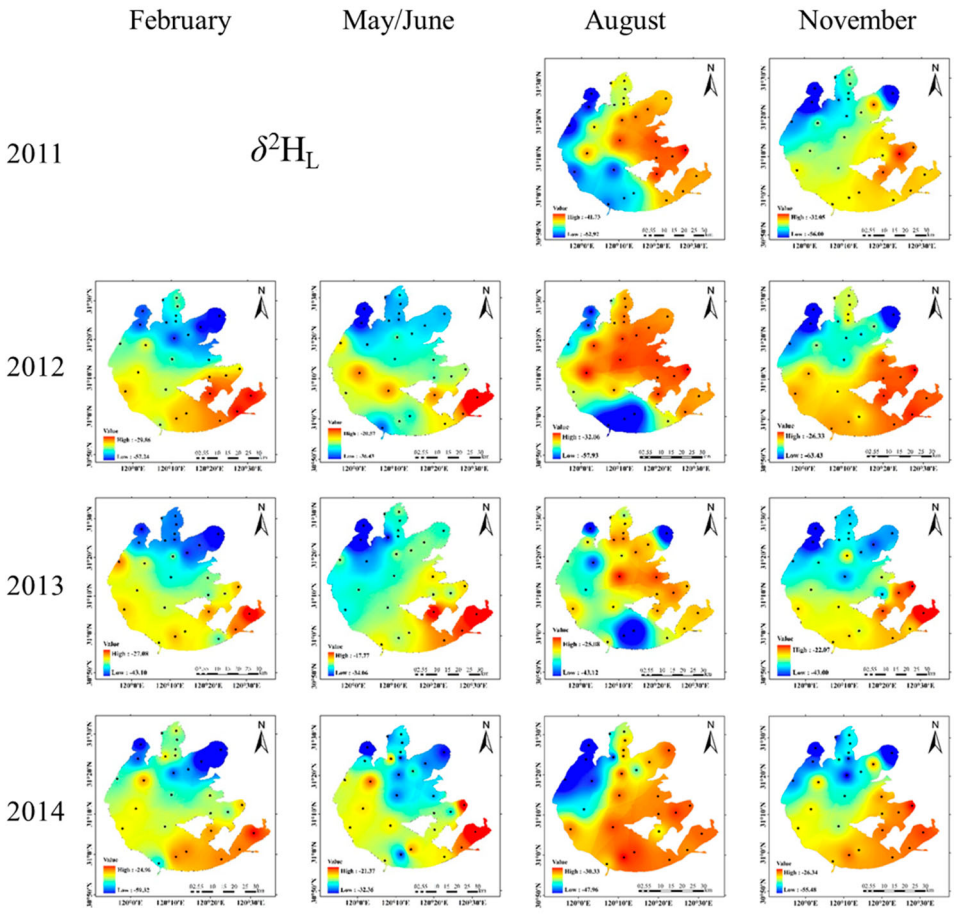

Figure 3. Spatial distribution of δ^2H_L at each lake survey.

2013, July to October 2013, December 2013 to March 2014, and November 2014. During these times, lake water in the north part near the mouth of the Wangyuhe River was less enriched in H^2HO and $H_2^{18}O$ than in the rest of the lake. In August, with the Wangyuhe River becoming an outflow river, the localized lake water became more enriched in $H_2^{18}O$ and H^2HO. Similarly, isotopic seasonal fluctuations in the southwest portion of the lake were associated with reversible flow patterns of the two rivers there (Figure 1).

To further quantify the spatial patterns, we divided the lake into three zones: inflow zone (12 sites), transition zone (9 sites) and outflow zone (8 sites). The isotopic differences between the outflow and inflow zone are shown in Figure 4. The outflow zone was always more enriched in H^2HO and $H_2^{18}O$ than the inflow zone, with the mean δ^2H_L difference ranging from 1.7 to 12.5 ‰ and the $\delta^{18}O_L$ difference ranging from 0.1 to 1.8 ‰. The deuterium excess in the outflow region was lower than that in the inflow region except for June 2014. In general, the spatial gradients were high in cold seasons and low in warm seasons. For example, the spatial difference of δ^2H_L was 3.8, 2.8 and 1.7 ‰ in May 2012, August 2013 and June 2014, respectively, and increased to 11.0, 7.3 and 10.6 ‰ in the following November; the corresponding $\delta^{18}O_L$ changed from 0.8, 0.5 and 0.1 to 1.8, 1.3 and 1.5 ‰; and d_L changed from −2.5, −0.9 and 1.3 to −3.7, −3.4 and −1.3 ‰.

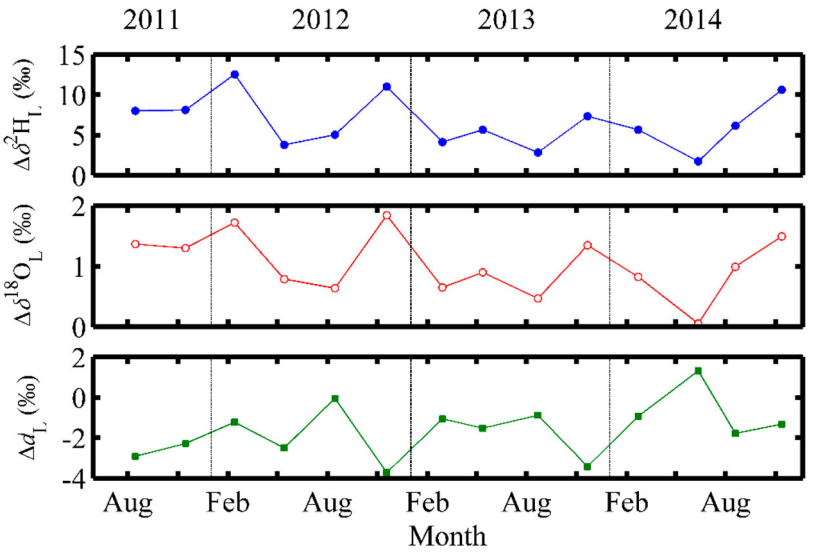

Figure 4. Difference of δ^2H_L, $\delta^{18}O_L$ and d_L between outflow and inflow regions of the lake (outflow–inflow).

3.2. Environmental controls on seasonal and interannual variabilities

Seasonal and interannual variations of meteorological variables and river and lake isotopic compositions are shown in Figure 5. The outflow rivers were always more enriched in H^2HO and $H_2^{18}O$ than the inflow rivers. The difference between the outflow and inflow rivers ranged from 3.4 to 11.5 ‰ for δ^2H and 0.7 to 1.6 ‰ for $\delta^{18}O$. The seasonality of the deuterium-excess difference between the two sets of rivers was not as clear as for H^2HO and $H_2^{18}O$. Daily δ^2H_L and $\delta^{18}O_L$ at the MLW site and the whole-lake mean values showed similar seasonal variations, with higher δ^2H_L and $\delta^{18}O_L$ in warm seasons than in cold seasons.

Linear correlation analysis (Table S1) indicates that evaporation (E), the difference between precipitation and evaporation ($P - E$) and throughflow index (x) are three key environmental factors influencing the isotopic compositions and their temporal variations. Here E is monthly lake evaporation calculated using the Priestley–Taylor equation [26] calibrated against the eddy covariance measurement, P is monthly precipitation, and x is throughflow index, defined as the ratio of monthly evaporation to the sum of precipitation and river input. Consistent with other lake studies [27–29], throughflow index was the most important factor controlling temporal variations in the whole-lake mean isotopic composition as well as in the difference in deuterium-excess between the inflow and the outflow rivers (Supplementary Material, Figure S2). In months of higher x, the whole-lake mean δ^2H_L and $\delta^{18}O_L$ decreased, d_L increased, and the difference in the deuterium-excess between the outflow and the inflow rivers became smaller. The whole-lake mean d_L was positively correlated with $P - E$. With regard to the spatial variations in δ^2H_L and $\delta^{18}O_L$, E was the only factor having significant correlation with temporal variations of the strength of the spatial gradient: When evaporation increased, the difference between the inflow and outflow zone became smaller.

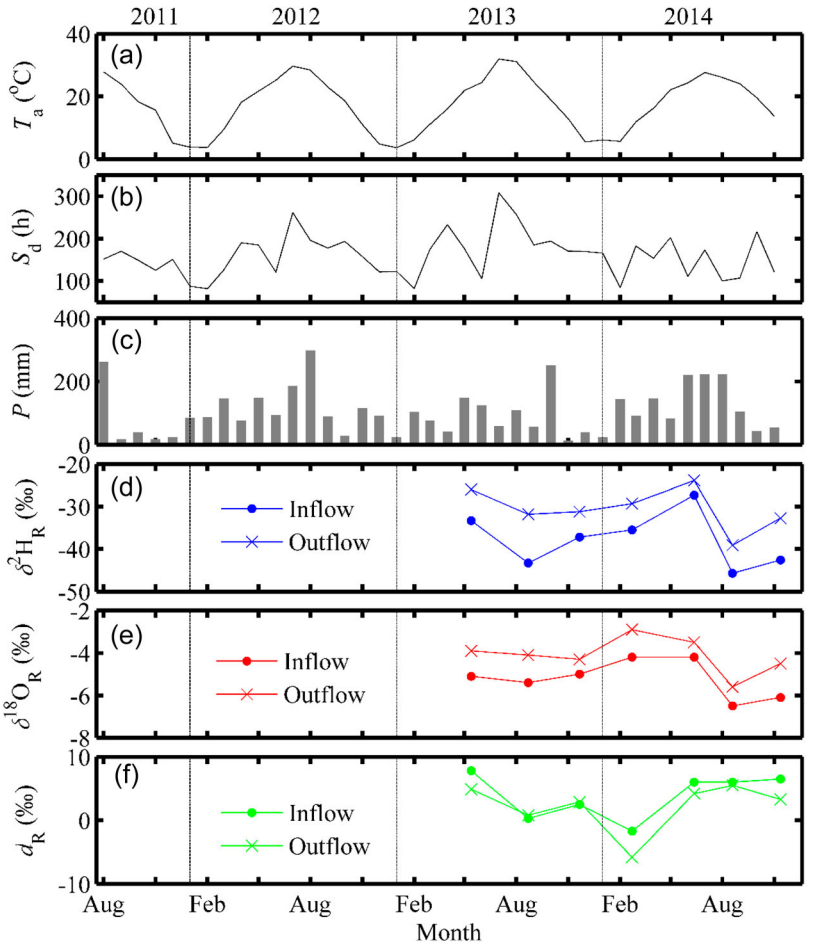

Figure 5. Seasonality of meteorological variables, lake and river water isotopes: (a) air temperature; (b) solar radiation; (c) precipitation; (d–f) δ^2H, $\delta^{18}O$ and d of inflow (dots) and outflow (crosses) rivers; (g–i) δ^2H, $\delta^{18}O$ and d of lake water at the MLW site; (j–l): whole-lake mean δ^2H, $\delta^{18}O$ and d.

3.3. Isotopic composition of lake evaporation

The H^2HO and $H_2^{18}O$ compositions of lake evaporation (δ^2H_E and $\delta^{18}O_E$) were calculated using the isotope mass balance method (Supplementary Material, Table 1). Traditionally, isotope measurements are used in connection with mass conservation to estimate lake evaporation. In the present study, lake evaporation was measured directly with eddy covariance, so the mass balance principle was used instead to constrain the isotopic composition of lake evaporation. In this calculation, all components except δ^2H_E and $\delta^{18}O_E$ were provided by measurements, and δ^2H_E and $\delta^{18}O_E$ were backed out from the isotope mass balance equations. The study period was from June 2013 to May 2014 when the measurements were available. Three sets of calculations were performed, one based on the daily isotope measurement at the MLW site, one on the whole-lake surveys, and the third on the seasonal measurement at site 16 (the ZSW site; Figure 1), but keeping the same values for all other input parameters. The goal was to determine

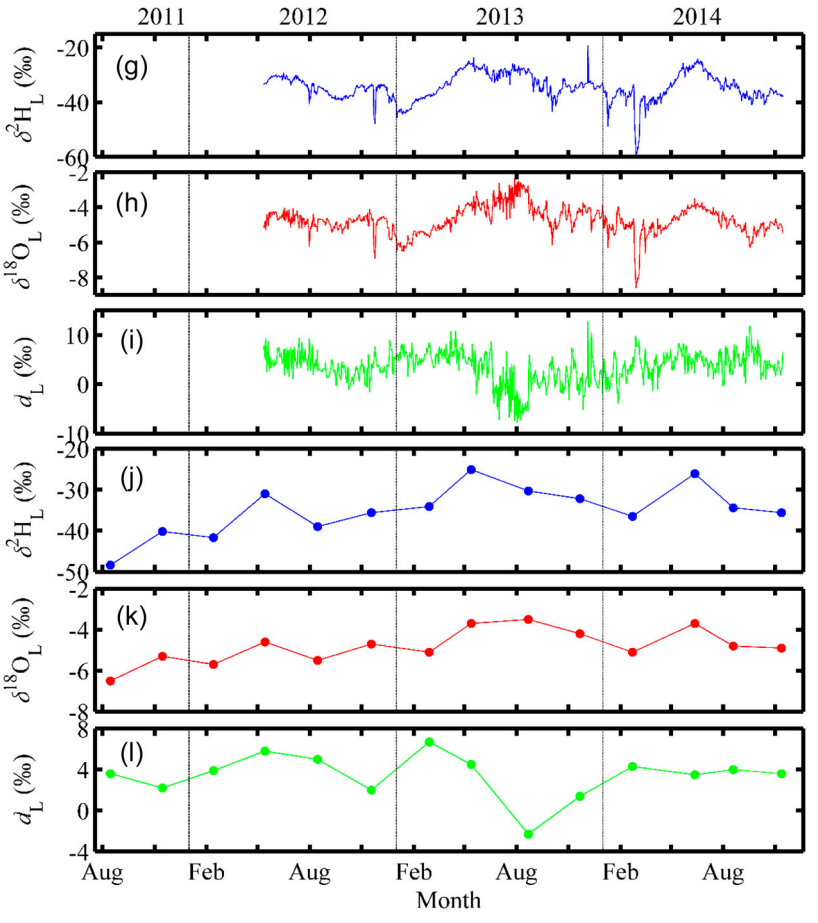

Figure 5. Continued.

uncertainties associated with the use of isotope data obtained at a single location as a substitution for the whole-lake mean. Based on the MLW site measurement, the annual δ^2H_E and $\delta^{18}O_E$ were −81.9 and −11.6 ‰, respectively. The annual δ^2H_E and $\delta^{18}O_E$ were −83.7 and −11.5 ‰ based on the whole-lake survey. It turns out that the isotopic composition at the MLW site were not too far from the whole-lake mean values (Table 1). The lowest

Table 1. Results of isotope mass balance method for a complete annual cycle (June 2013 to May 2014). The mean δ^2H and $\delta^{18}O$ values of water vapour were −108.4 and −15.7 ‰.

	Precipitation	Inflow	Outflow	Lake water	Change in lake water	Evaporation
Amount ($\times10^8$ m^3)	27.5	98.8	105.3	49.1	−0.67	21.7
δ^2H (‰)	−38.8	−38.6	−28.7	−34.4[a]/−31.5[b]/−41.6[c]	−2.0[a]/−1.2[b]/−0.6[c]	−81.9[a]/−83.7[b]/−85.2[c]
$\delta^{18}O$ (‰)	−6.0	−4.9	−3.7	−4.5[a]/−4.2[b]/−5.5[c]	−0.2[a]/−0.2[b]/1.4[c]	−11.6[a]/−11.5[b]/−15.2[c]

Note: Change in lake water represents difference in the amount, δ^2H or $\delta^{18}O$ between 1 June 2013 and 31 May 2014.
[a]Results based on one-site daily measurement at the MLW site.
[b]Results based on whole-lake surveys.
[c]Results based on the seasonal measurement at the ZSW site.

annual mean δ^2H_L and $\delta^{18}O_L$ were observed at the ZSW site, in northwest position in the lake (Figure 1). If these observations were used in the mass balance calculation, δ^2H_E and $\delta^{18}O_E$ would change to −85.2 and −15.2 ‰, which were much more negative than the estimates based on the whole-lake mean.

Linear regression of the whole-lake water isotopic composition yielded a local evaporation line of $\delta^2H_L = 6.76\ \delta^{18}O_L - 2.59$ (Supplementary Material, Figure S3). Mass conservation requires that δ^2H_E and $\delta^{18}O_E$ should fall on the evaporation line. The δ^2H_E and $\delta^{18}O_E$ estimates based on the whole lake and the MLW isotopic data are indeed closer to the line than those based on the ZSW data (Figure S3).

3.4. Lake water residence time

The lake water δ^2H and $\delta^{18}O$ data were used as tracers to calculate the residence time of water in Lake Taihu. A steady-state model [29] was employed, assuming that Lake Taihu is a throughflow lake with inflow (inflow plus precipitation) balanced by evaporation plus liquid outflow (Supplementary Material). Also, the seasonality of evaporation rate was considered. If the δ^2H data were used, the residence time was 75 ± 12 days, and for $\delta^{18}O$ data, the residence time of 36 ± 8 days was calculated. For comparison, if the residence time (τ) was computed from the water budget as $\tau = V/(P + I)$ (where V represents lake water volume, P is precipitation, I is inflow), it should be around 143 ± 14 days, where V is 49.1 $(\pm 4.9) \times 10^8$ m^3, P is 27.5 $(\pm 2.8) \times 10^8$ m^3, and I is 98.8 $(\pm 9.9) \times 10^8$ m^3 (Supplementary Material). To calculate the residence time, a steady-state model was used under the assumption that the inflow is continuous and balanced by outflow (evaporation and liquid outflow) [29]. For Lake Taihu, the assumption may not be fulfilled since the mean depth was small, and there was short-term perturbation such as water diversion from the Yangtze River. It appears that the unsteady state conditions affected the isotopic and the water mass balance calculations differently. Based on a numerical model of water transport across the lake, Li et al. [25] found that the lake residence time or water age is 130 days in the summer and around 230 days in other seasons, with a maximum value of 254 days in 3 % of the lake. The relatively short residence time of Lake Taihu is comparable with those of large and shallow lakes. Of the 73 lakes surveyed by Jasechko et al. [6], nine lakes have water area greater than 2000 km^2 and depth less than 4.6 m (the threshold for shallow lakes according to the Minnesota Pollution Control Agency [30]), seven of these shallow and large lakes have residence time less than 3 years, and two have a 6-year residence time. According to the research of Brooks et al. [11], half of the 1028 lakes across the conterminous USA have residence time less than 190 days, and the median residence time of the lakes shallower than 2 m was 124 days. They also found that the isotope-derived residence time was longer than the residence time from the water balance model, but their lake water sampling was limited in summer and may not represent a whole-year condition.

If our residence time calculations were based on the isotope data collected at the single site (MLW), the residence time was reduced to 42 ± 7 and 24 ± 7 days based on H^2HO and $H_2^{18}O$ as tracers, respectively. Using the data from site ZSW, the results were unreasonably negative since the isotopic composition of lake water was even lower than the amount-weighted isotope value of inflow water.

4. Conclusions

In this study, the spatial patterns and temporal variability of lake water isotopes from Lake Taihu, a large and shallow lake, were investigated for more than three years. The results indicated (1) that spatial patterns of the lake isotopic composition were characterized with a gradual isotopic gradient from the northwest river inflow zone to the southeast river outflow zone, and (2) that the spatial patterns were controlled by the water flow pattern, not by the changes in local evaporation rate. With regard to temporal variability, we found that the key environmental factors influencing the temporal variability of the isotopic composition were the evaporation rate, the difference between precipitation and evaporation, and throughflow index. The lake water residence time calculated with the isotope method was shorter than those computed from the lake water budget or numerical modelling.

Disclosure statement

No potential conflict of interest was reported by the authors.

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