

1 **Spatial Distribution and Temporal Variability of Stable Water Isotopes in a Large and**
2 **Shallow Lake**

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16
17 **Abstract**

18 Stable isotopic compositions of lake water provide additional information on hydrological,
19 meteorological and paleoclimate processes. In this study, lake water isotopic compositions
20 were measured for more than three years in Lake Taihu, a large and shallow lake in southern
21 China, to investigate the isotopic spatial and seasonal variations. The results indicated that (1)
22 the whole-lake mean H²HO and H₂¹⁸O composition of the lake water varied seasonally from
23 48.4‰ ± 5.8‰ to -25.1‰ ± 3.2‰ and from -6.5‰ ± 0.9‰ to -3.5‰ ± 0.8‰, respectively,
24 (2) the spatial pattern of the lake water isotopic compositions was controlled by the direction
25 of water flow and not by local evaporation rate, and (3) using a one-site isotopic measurement
26 to represent the whole-lake mean may result in unreasonable estimates of the isotopic
27 composition of lake evaporation and the lake water residence time in poorly mixed lakes. The
28 original data, documented here as an online supplement, provides a good reference for testing
29 sensitivity of lake water budget to various isotopic sampling strategies. We propose that

30 detailed spatial measurement of lake water isotopic compositions provides a good proxy for
31 water movement and pollutant and alga transports, especially over big lakes.

32

33 **Keywords:** lake water; H²HO; H₂¹⁸O; spatial distribution; seasonal variability

34

35 **1. Introduction**

36 Compositions of stable isotopes of lake water provide unique information on hydrological,
37 meteorological and climatic processes. In the process of lake evaporation, the heavier isotopic
38 molecules of water (H²HO and H₂¹⁸O) tend to accumulate in the liquid water, causing
39 depletion in the vapor phase. Isotopic measurements of the water can be used to estimate
40 annual evaporation of surface water bodies (e.g. [1-6]), which is an important component of
41 the local hydrological cycle, but is difficult to measure with in-situ instruments, especially in
42 remote areas due to cost, time and infrastructure considerations [7]. Lake isotopic
43 measurements are also widely used in research on lake water yield [8,9], water residence time
44 [10,11], water mixing [12,13], and connection with groundwater [14,15]. Furthermore, data
45 on stable isotopes in lakes with long residence times can be used to study the effect of past
46 climate on lake evolution [10,16].

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

56 Here, we investigate the spatial distribution and temporal variability of stable isotopes in
57 Lake Taihu, a large and shallow freshwater lake in the Yangtze River Delta, China. We chose
58 Lake Taihu for four reasons. First, Lake Taihu is a large and shallow lake; therefore, we

59 expect measureable spatial variability. Lake Taihu is the third largest freshwater lake in China,
60 with mean water depth of 1.9 m, area of 2400 km², lake length from north to south of 69 km
61 and width from east to west of 56 km [20]. Second, the lake has distinct inflow and outflow
62 regions throughout the year (Figure 1). Among the 172 rivers and canals connected with Lake
63 Taihu [21], most act as either inflows or outflows all the time. The main exceptions are three
64 waterways whose water flow may be reversed for the purpose of water quality management.
65 Isotopic enrichment along the direction of water flow in the lake can be expected. Third, the
66 lake is located in a subtropical monsoon climate, where 45% of annual precipitation fall in the
67 months of June to August. The spatial isotopic patterns may reveal different roles of
68 precipitation and river input in the rainy season and in the dry season. Fourth, the isotopic
69 study was supported by concurrent measurements at five eddy covariance sites on the lake
70 [22]. Variables measured by the eddy flux network, such as lake evaporation and lake
71 temperature, can help us interpret temporal and spatial patterns seen in the isotopic data.

72 A number of factors may contribute to spatial variability of the lake water isotopic
73 composition. In particular, we are interested in two competing hypotheses: (1) spatial
74 variations of the lake isotopic composition are indicative of spatial variations in the fraction of
75 the water parcel that has previously evaporated. To put it differently, high H²HO or H₂¹⁸O
76 composition may indicate high local evaporation rate; and (2) spatial variations in the H²HO
77 and H₂¹⁸O compositions are controlled by “water age” or time spent in the lake since the
78 water first entered. If this hypothesis holds, the lake isotopic composition may be a good
79 tracer for water movement across this vast lake. These hypotheses are relevant to ongoing
80 efforts to restore the water quality of Lake Taihu. The lake provides the drinking water supply
81 for five million people in cities such as Shanghai, Wuxi and Suzhou [23]. Located in one of
82 the most developed regions in the country, Lake Taihu has been suffering from severe water
83 pollution problems [20]. Algal blooms are common occurrence. A better knowledge of the
84 water flux and movement across the lake may improve predictions of the timing and location
85 of algal growth and the subsequent spread of algal biomass. If our second hypothesis is valid,
86 the isotopic tracers may be a more viable method for tracking horizontal water movement
87 than model calculations based on hydrodynamic principles because of the large uncertainties
88 in specifying the inflow and outflow boundary conditions associated with so many waterways

89 for models.

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

94

95 **2. Materials and Methods**

96 *2.1 Measurement of lake and river water isotopes*

97 Isotopic measurements consisted of daily lake water sampling at one location and seasonal
98 lake and river surveys. Since 11 May 2012, one lake water sample was collected daily at
99 13:00 at a distance of 200 m from the shore. The sampling site is the Taihu Lake Ecosystem
100 Research Station of Chinese Academy of Sciences, located in Meiliang Bay, at the north shore
101 of the lake (the MLW site, 31°24'N, 120°13'E, Figure 1). The first seasonal lake survey was
102 conducted at 29 sites spread over the whole lake on August 15-16, 2011. One water sample
103 was collected at each location. Subsequent surveys were made every three months in
104 February, May/June, August and November, each survey lasting 1-2 days and covering the
105 same 29 locations in the lake. The first river water survey was conducted in May 2013 on the
106 same days when the lake survey took place. Subsequent river surveys were also timed with
107 the lake surveys. One water sample was collected at ~100 m from each entry or exit point of
108 the lake from 51 rivers (Figure 1). These rivers contributed more than 80% of the total water
109 inflow and outflow of the lake. Lake and river samples were collected from 20 cm below the
110 surface.

111 The water samples were collected in 300-mL glass bottles. The bottles were sealed and
112 placed in a refrigerator with temperature of 4 °C before analysis. The samples were analyzed
113 for H²HO and H₂¹⁸O compositions using a liquid water isotope analyzer (Model DLT-100;
114 Los Gatos Research, Mountain View, CA, USA) in the Key Laboratory of Ecosystem
115 Network Observation and Modeling, Chinese Academy of Sciences. The typical measurement
116 precision was ±0.3‰ for H²HO and ±0.1‰ for H₂¹⁸O. Data collected before the end of 2014
117 were used in this study. Delta notation $\delta^2\text{H}_L$ and $\delta^{18}\text{O}_L$ were used to represent H²HO and
118 H₂¹⁸O isotopic enrichment in reference to the Vienna Standard Mean Ocean Water.

119 From August 2011 to May 2012, we also measured vertical profiles of the H²HO and
120 H₂¹⁸O compositions at the 20-, 80- and 150-cm depth. Differences among the three depths
121 were less than 0.2‰ for whole-lake mean $\delta^2\text{H}_L$ and less than 0.1‰ for $\delta^{18}\text{O}_L$. The vertical
122 gradients were small because convective overturning occurs regularly at night [22], and the
123 profile measurement was discontinued.

124

125 **3. Results and Discussion**

126 ***3.1 Spatial patterns of the lake water isotopes***

127 Figure 2 shows the spatial patterns of the lake isotopic composition averaged over a three-
128 year period (August 2011 to June 2014). A gradual isotopic gradient from the northwest river
129 inflow zone to the southeast river outflow zone is clearly evident. The lowest $\delta^2\text{H}_L$ and $\delta^{18}\text{O}_L$
130 occurred in the northern part of the lake, with minimum values of $-45.8\text{‰} \pm 9.7\text{‰}$ and $-6.3\text{‰} \pm 1.5\text{‰}$,
131 respectively. The southeast zone was most enriched in H²HO and H₂¹⁸O, with
132 maximum values of $-28.1\text{‰} \pm 7.5\text{‰}$ and $-3.7\text{‰} \pm 0.9\text{‰}$. The spatial pattern of deuterium-
133 excess (d_L) was reversed, with higher values in the northwest zone (with a maximum of $5.1\text{‰} \pm 3.2\text{‰}$)
134 and lower values in southeast zone (with a minimum of $1.1\text{‰} \pm 3.5\text{‰}$). For
135 comparison, the annual mean $\delta^2\text{H}$ (± 1 standard deviation) of inflow and outflow rivers was -
136 $38.8\text{‰} \pm 6.3\text{‰}$ and $-30.0\text{‰} \pm 4.8\text{‰}$, respectively (Figure 5).

137 Isotopic data from individual seasonal surveys are shown in Figures 3 and S1. With the
138 exception of August, the seasonal spatial patterns are similar to the annual patterns. In August,
139 the northeast portion of the lake was equally enriched as or more enriched in H²HO and H₂¹⁸O
140 than the southeast portion. The range of spatial variability also changed through the year. The
141 minimum spatial variability occurred in June 2014 (with a range of 11.0‰ for H²HO), while
142 the maximum occurred in November 2012 (with a range of 37.1‰ for H²HO).

143 Changes in local evaporation rate were not a contributor to these spatial patterns because
144 eddy covariance data show little spatial variations in water surface temperature and in local
145 evaporation rate [24]. The annual mean $\delta^2\text{H}_L$ (± 1 standard deviation) at location 27 in the
146 southeastern region was $-30.9\text{‰} \pm 5.3\text{‰}$, which was 6.6‰ greater than at location 10 near the
147 western shore (Supplementary Figure 1). At these two locations, lake evaporation rate
148 measured by eddy covariance differed by only 1%.

149 Instead, the progressive enrichment in H^2HO and H_2^{18}O from northeast to southwest was
150 associated with the water flow pattern. As water moves from the inflow to the outflow region,
151 it undergoes evaporation for a longer time and becomes more isotopically enriched. In other
152 words, the spatial variability in the isotopic composition of the lake was associated with the
153 direction of water flow. Not surprisingly, when some of the rivers reversed flow direction, the
154 spatial patterns also shifted. Water was diverted, at times of low water levels, from Yangtze
155 River through Wangyuhe River, a reversible waterway at the northeast corner of the lake
156 (Figure 1) in order to raise the lake water level for navigation and for the purpose of pollution
157 dilution [25]. In our study period, water diversion was conducted from November 2011 to
158 February 2012, November 2012 to January 2013, July to October 2013, December 2013 to
159 March 2014, and November 2014. During these times, lake water in the north part near the
160 mouth of Wangyuhe River was less enriched in H^2HO and H_2^{18}O than the rest of the lake. In
161 August, with Wangyuhe River becoming an outflow river, the localized lake water became
162 more enriched in H_2^{18}O and H^2HO . Similarly, isotopic seasonal fluctuations in the southwest
163 portion of the lake were associated with reversible flow patterns of the two rivers there
164 (Figure 1).

165 To further quantify the spatial patterns, we divided the lake into three zones: inflow zone
166 (12 sites), transition zone (9 sites) and outflow zone (8 sites). The isotopic differences
167 between the outflow and inflow zone are shown in Figure 4. The outflow zone was always
168 more enriched in H^2HO and H_2^{18}O than the inflow zone, with the mean $\delta^2\text{H}_L$ difference
169 ranging from 1.7‰ to 12.5‰ and the $\delta^{18}\text{O}_L$ difference ranging from 0.1‰ to 1.8‰.
170 Deuterium excess in the outflow region was lower than that in the inflow region except for
171 June 2014. In general, the spatial gradients were high in cold seasons and low in warm
172 seasons. For example, the spatial difference of $\delta^2\text{H}_L$ was 3.8‰, 2.8‰ and 1.7‰ in May 2012,
173 August 2013 and June 2014 respectively, and increased to 11.0‰, 7.3‰ and 10.6‰ in the
174 following November; the corresponding $\delta^{18}\text{O}_L$ changed from 0.8‰, 0.5‰ and 0.1‰ to 1.8‰,
175 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‰.

176

177 ***3.2 Environmental controls on seasonal and interannual variabilities***

178 Seasonal and interannual variations of meteorological variables and river and lake isotopic

179 compositions are shown in Figure 5. The outflow rivers were always more enriched in H²HO
180 and H₂¹⁸O than the inflow rivers. The difference between the outflow and inflow rivers ranged
181 from 3.4‰ to 11.5‰ for δ²H and 0.7‰ to 1.6‰ for δ¹⁸O. The seasonality of the deuterium
182 excess difference between the two sets of rivers was not as clear as for H²HO and H₂¹⁸O.
183 Daily δ²H_L and δ¹⁸O_L at the MLW site and the whole lake mean values showed similar
184 seasonal variations, with higher δ²H_L and δ¹⁸O_L in warm seasons than in cold seasons.

185 Linear correlation analysis (Table S1) indicates that evaporation (E), the difference
186 between precipitation and evaporation ($P - E$) and throughflow index (x) are three key
187 environmental factors influencing the isotopic compositions and their temporal variations.
188 Here E is monthly lake evaporation calculated using the Priestley-Taylor equation [26]
189 calibrated against the eddy covariance measurement, P is monthly precipitation, and x is
190 throughflow index, defined as the ratio of monthly evaporation to the sum of precipitation and
191 river input. Consistent with other lake studies [27–29], throughflow index was the most
192 important factor controlling temporal variations in the whole-lake mean isotopic composition
193 as well as in the difference in deuterium-excess between the inflow and the outflow rivers
194 (Supplementary Figure S2). In months of higher x , the whole-lake mean δ²H_L and δ¹⁸O_L
195 decreased, d_L increased, and the difference in the deuterium-excess between the outflow and
196 the inflow rivers became smaller. The whole-lake mean d_L was positively correlated with $P -$
197 E . With regard to the spatial variations in δ²H_L and δ¹⁸O_L, E was the only factor having
198 significant correlation with temporal variations of the strength of the spatial gradient: When
199 evaporation increased, the difference between the inflow and outflow zone became smaller.

200

201 **3.3 Isotopic composition of lake evaporation**

202 The H²HO and H₂¹⁸O compositions of lake evaporation (δ²H_E and δ¹⁸O_E) were calculated
203 using the isotopic mass balance method (Table 1; Supplemental Materials). Traditionally,
204 isotopic measurements are used in connection with mass conservation to estimate lake
205 evaporation. In the present study, lake evaporation was measured directly with eddy
206 covariance, so the mass balance principle was used instead to constrain the isotopic
207 composition of lake evaporation. In this calculation, all components except δ²H_E and δ¹⁸O_E
208 were provided by measurements, and δ²H_E and δ¹⁸O_E were backed out from the isotopic mass

209 balance equations. The study period was from June 2013 to May 2014 when the
210 measurements were available. Three sets of calculations were performed, one based on the
211 daily isotopic measurement at the MLW site, one on the whole-lake surveys, and the third on
212 the seasonal measurement at site 16 (the ZSW site; Figure 1), but keeping the same values for
213 all other input parameters. The goal was to determine uncertainties associated with the use of
214 isotopic data obtained at a single location as a substitution for the whole-lake mean. Based on
215 the MLW site measurement, the annual $\delta^2\text{H}_E$ and $\delta^{18}\text{O}_E$ were -81.9‰ and -11.6‰,
216 respectively. The annual $\delta^2\text{H}_E$ and $\delta^{18}\text{O}_E$ were -83.7‰ and -11.5‰ based on the whole-lake
217 survey. It turns out that the isotopic composition at the MLW site were not too far from the
218 whole lake mean values (Table 1). The lowest annual mean $\delta^2\text{H}_L$ and $\delta^{18}\text{O}_L$ were observed at
219 the ZSW site, in northwest position in the lake (Figure 1). If these observations were used in
220 the mass balance calculation, $\delta^2\text{H}_E$ and $\delta^{18}\text{O}_E$ would change to -85.2‰ and -15.2‰, which
221 were much more negative than the estimates based on the whole-lake mean.

222 Linear regression of the whole-lake water isotopic composition yielded a local
223 evaporation line of $\delta^2\text{H}_L = 6.76\delta^{18}\text{O}_L - 2.59$ (Figure S3). Mass conservation requires that $\delta^2\text{H}_E$
224 and $\delta^{18}\text{O}_E$ should fall on the evaporation line. The $\delta^2\text{H}_E$ and $\delta^{18}\text{O}_E$ estimates based on the
225 whole lake and the MLW isotopic data are indeed closer to the line than those based on the
226 ZSW data (Figure S3).

227

228 **3.4 Lake water residence time**

229 The lake water H^2HO and H_2^{18}O data were used as tracers to calculate the residence time of
230 water in Lake Taihu. A steady-state model [29] was employed, assuming Lake Taihu is a
231 throughflow lake with inflow (inflow plus precipitation) balanced by evaporation plus liquid
232 outflow (Supplemental Materials). Also, the seasonality of evaporation rate was considered. If
233 the H^2HO data was used, the residence time was 75 ± 12 days. If H_2^{18}O data was used, the
234 residence time was 36 ± 8 days. For comparison, if the residence time (τ) was computed from
235 the water budget as $\tau = V/(P + I)$ (where V represents lake water volume, P is precipitation, I
236 is inflow), it should be around 143 ± 14 days, where V is $49.1 (\pm 4.9) \times 10^8 \text{ m}^3$, P is $27.5 (\pm$
237 $2.8) \times 10^8 \text{ m}^3$, and I is $98.8 (\pm 9.9) \times 10^8 \text{ m}^3$ (Supplementary Materials). To calculate
238 residence time, a steady-state model was used under the assumption that the inflow is

239 continuous and balanced by outflow (evaporation and liquid outflow) (Gibson et al. 2002).
240 For Lake Taihu, the assumption may not be fulfilled since the mean depth was small and there
241 was short-term perturbation such as water diversion from the Yangtze River. It appears that
242 the unsteady state conditions affected the isotopic and the water mass balance calculations
243 differently. Based on a numerical model of water transport across the lake, Li et al. [25] found
244 that the lake residence time or water age is 130 days in the summer and around 230 days in
245 other seasons, with a maximum value of 254 days in 3% of the lake. The relatively short
246 residence time of Lake Taihu is comparable with those of large and shallow lakes. Of the 73
247 lakes surveyed by Jasechko et al. [6], nine lakes have water area greater than 2000 km² and
248 depth less than 4.6 m (the threshold for shallow lakes according to the Minnesota Pollution
249 Control Agency (MPCA) [30], seven of these shallow and large lakes have residence time less
250 than 3 years, and two have a 6-year residence time. According to the research of Brooks et al.
251 [11], half of the 1028 lakes across the conterminous USA have residence time less than 190
252 days, and the median residence time of the lakes shallower than 2 m was 124 days. They also
253 found that the isotope-derived residence time was longer than the residence time from the
254 water balance model, but their lake water sampling was limited in summer and may not
255 represent a whole-year condition.

256 If our residence time calculations were based on the isotopic data collected at the single
257 site (MLW), the residence time was reduced to 42 ± 7 and 24 ± 7 days using H²HO and H₂¹⁸O
258 as tracer respectively. Using the data from site ZSW, the results were unreasonably negative
259 since the isotopic content of lake water was even lower than the amount-weighted isotopic
260 value of inflow water.

261

262 **4. Conclusions**

263 In this study, the spatial patterns and temporal variability of lake water isotopes over Lake
264 Taihu, a large and shallow lake, were investigated over more than three years. The results
265 indicated (1) that spatial patterns of the lake isotopic composition were characterized with a
266 gradual isotopic gradient from the northwest river inflow zone to the southeast river outflow
267 zone, and (2) that the spatial was controlled by the water flow pattern, not by the changes in
268 local evaporation rate. With regard to temporal variability, we found that the key environmental

269 factors influencing the temporal variability of the isotopic composition were evaporation rate,
270 the difference between precipitation and evaporation, and throughflow index. The lake water
271 residence time calculated with the isotopic method was shorter than those computed from the
272 lake water budget or numerical modeling.

273

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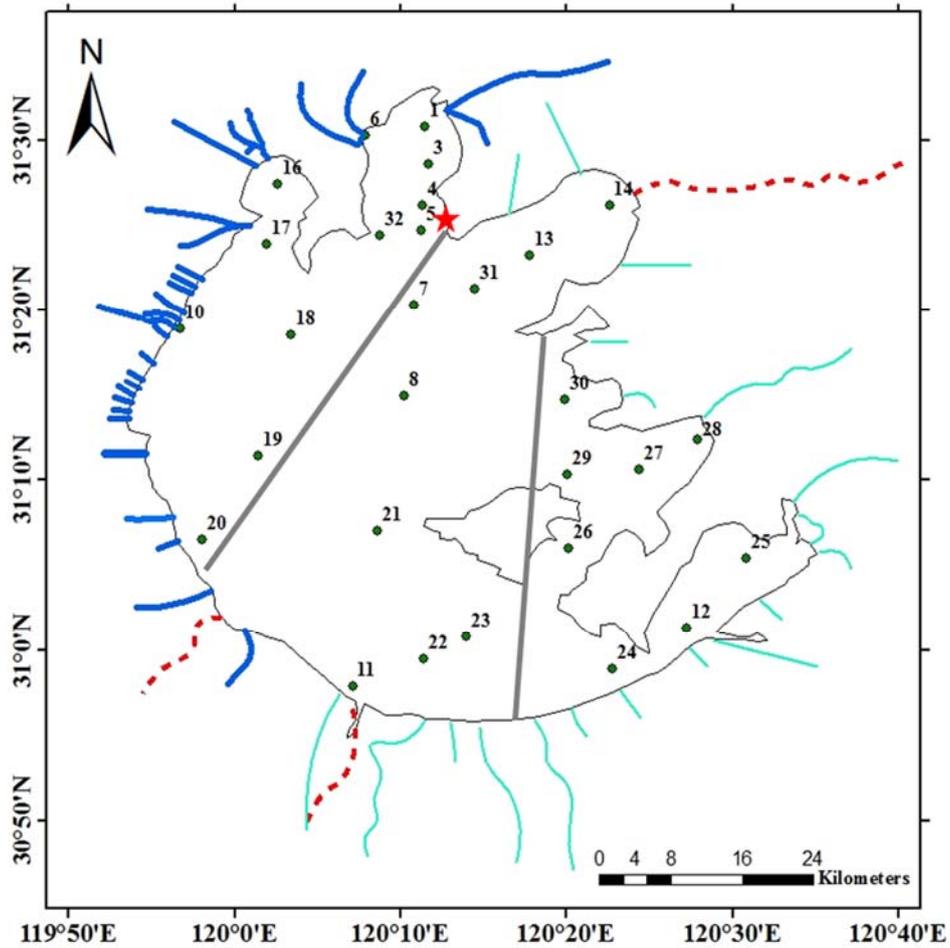
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362 **Table 1.** Results of isotopic mass balance method for a complete annual cycle (June 2013 to May 2014). The mean $\delta^2\text{H}$ and $\delta^{18}\text{O}$ values of water
 363 vapor were -108.4‰ and -15.7‰.

| | Precipitation | Inflow | Outflow | Lake water | Change in lake water | Evaporation |
|--------------------------------------|---------------|--------|---------|-------------------------|----------------------|-------------------------|
| Amount ($\times 10^8 \text{ m}^3$) | 27.5 | 98.8 | 105.3 | 49.1 | -0.67 | 21.7 |
| $\delta^2\text{H}$ (‰) | -38.8 | -38.6 | -28.7 | -34.4*/-31.5**/-41.6*** | -2.0*/-1.2**/-0.6*** | -81.9*/-83.7**/-85.2*** |
| $\delta^{18}\text{O}$ (‰) | -6.0 | -4.9 | -3.7 | -4.5*/-4.2**/-5.5*** | -0.2*/-0.2**/1.4*** | -11.6*/-11.5**/-15.2*** |

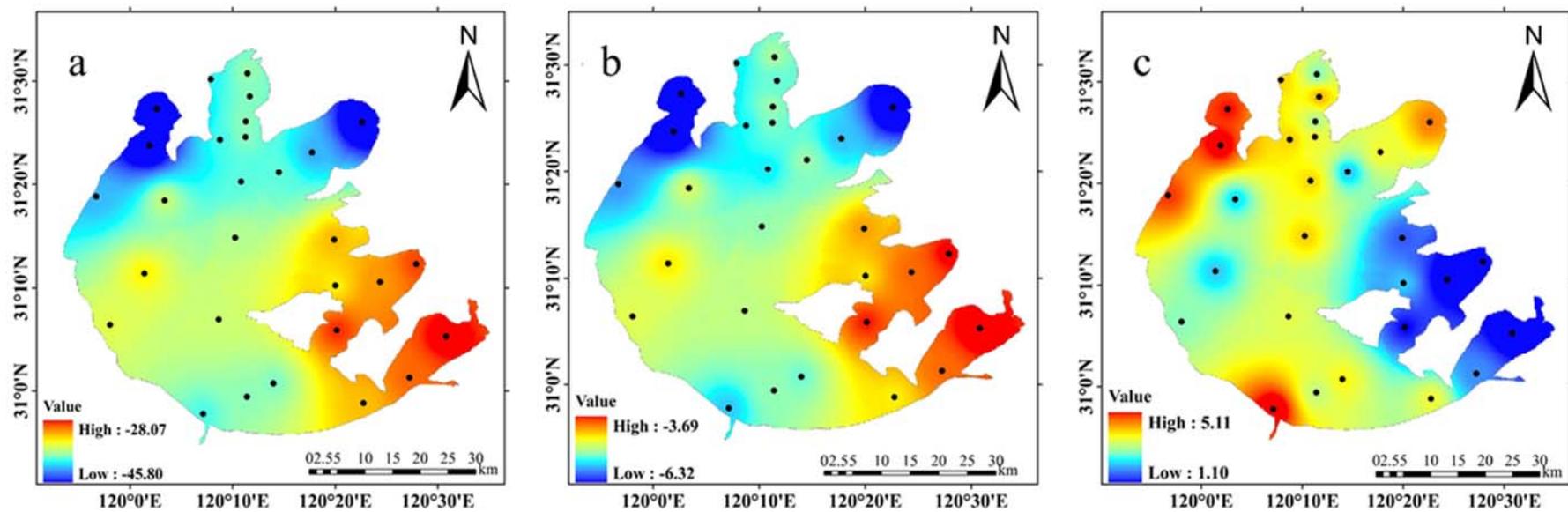
364 Note: Change in lake water represents difference in the amount, $\delta^2\text{H}$ or $\delta^{18}\text{O}$ between 1 June 2013 and 31 May 2014. The data marked with *
 365 represent results based on one-site daily measurement at the MLW site; those marked with ** represent results based on whole-lake surveys;
 366 and those marked with *** represent results based on the seasonal measurement at the ZSW site.

367 **Figure 1.** Map of Lake Taihu and the sites of lake and river surveys. Star: the MLW site; dots:
 368 lake survey sites; thick blue lines: inflow rivers; thin green lines: outflow rivers; red dashed
 369 lines: inflow/outflow rivers (the river at the northeast corner was Wangyuhe River); Grey
 370 lines mark approximate boundary of the inflow and outflow regions.



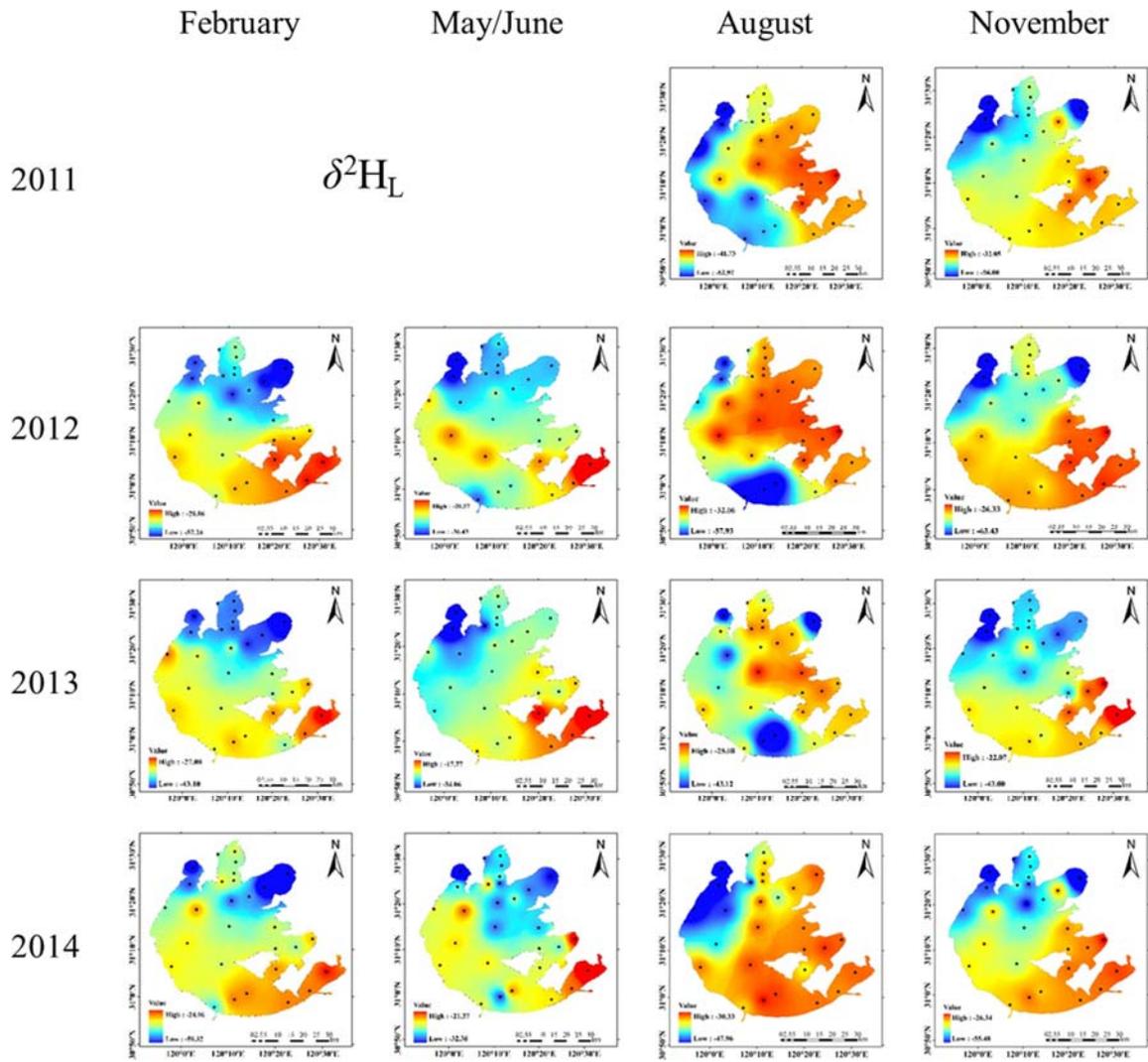
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372 **Figure 2.** Spatial patterns of the annual mean lake water isotope composition: a: H^2HO ; b: H_2^{18}O ; c: d-excess.

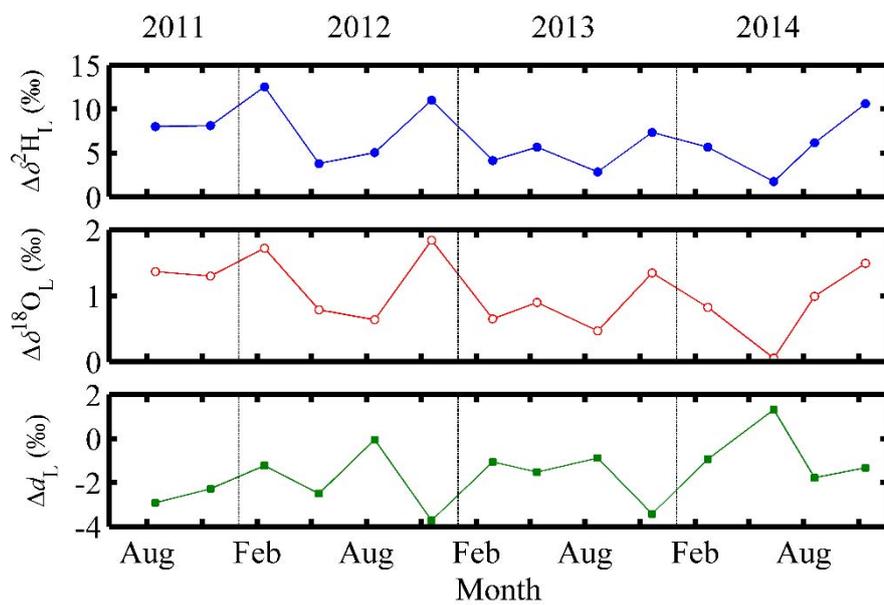


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374 **Figure 3.** Spatial distribution of H^2HO at each lake survey.

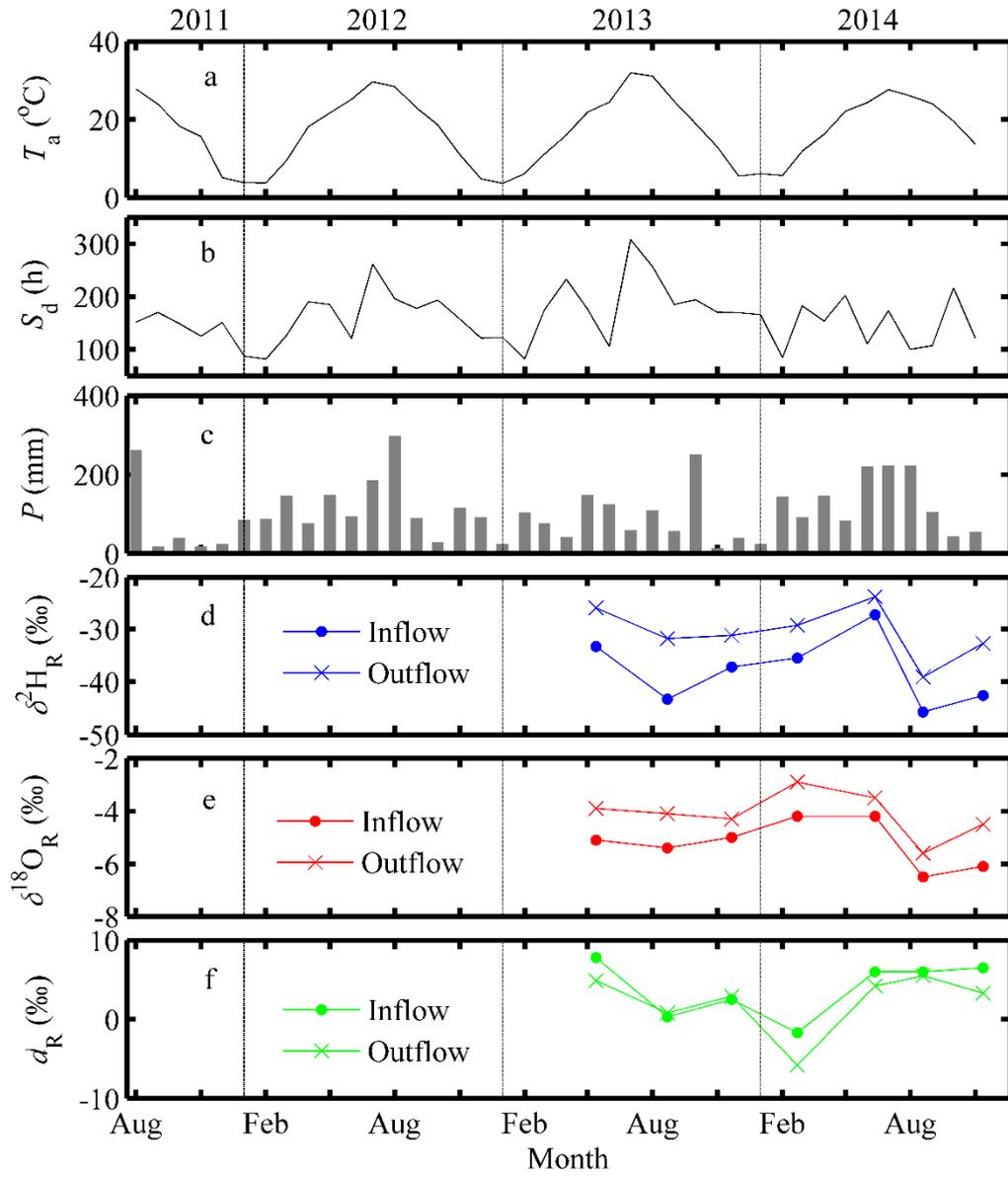


376 **Figure 4.** Difference of $\delta^2\text{H}_L$, $\delta^{18}\text{O}_L$ and d_L between outflow and inflow regions of the lake
 377 (outflow– inflow).

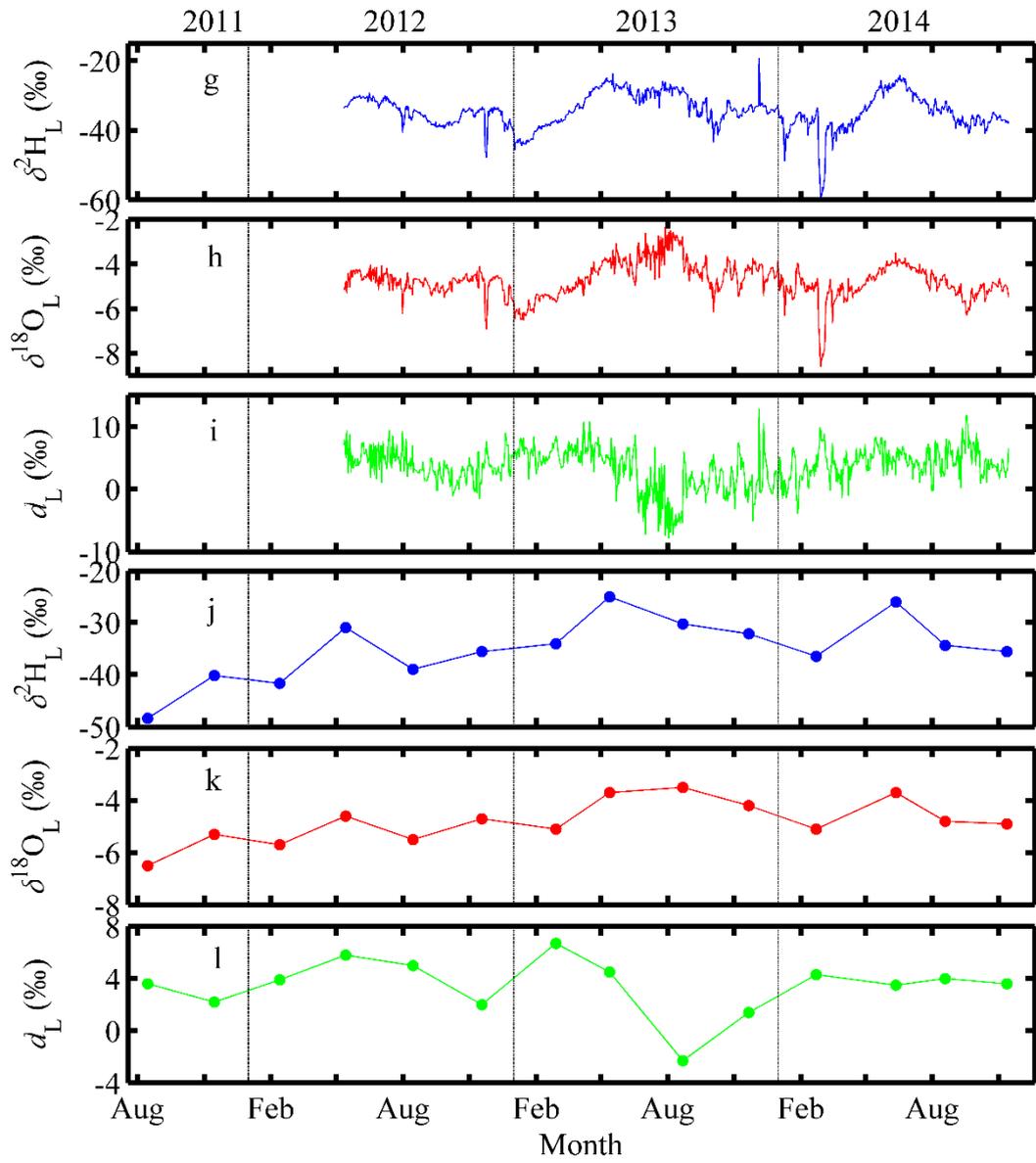


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379 **Figure 5.** Seasonality of meteorological variables, lake and river water isotopes: a: air temperature;
 380 b: solar radiation; c: precipitation; d-f: $\delta^2\text{H}$, $\delta^{18}\text{O}$ and d of inflow (dots) and outflow (crosses) rivers;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



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