Supplemental Materials for

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

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S1. Isotopic mass balance method

Isotopic mass balance method (IMBM) was employed to calculate the isotopic compositions of evaporation (δ_E). The water balance equation is given by,

$$I + P = E + \Delta V + O \quad (1)$$

where I represents inflow, P is precipitation, E is evaporation, V is lake water volume, ΔV is the variation of water volume, and O is outflow. In this calculation, time step was one year. Monthly I, V and ΔV data were from the report on the website of Taihu Basin Authority of Ministry of Water Resources (http://www.tba.gov.cn). Precipitation was the mean value of monthly precipitation observed at Wuxi, Huzhou and Dongshan meteorological station located in north, south-west and south-east of the lake, respectively. Evaporation was calculated using the Priestley-Taylor evaporation model [1] validated against the evaporation measured by the Taihu Eddy Flux Mesonet [2], with input variables of radiation, air pressure, air and water temperature measured at the MLW eddy covariance site. Outflow O was calculated as a residual using equation 1 to ensure perfect water balance.

The isotopic mass balance equation is given by

$$\delta_I I + \delta_P P = \delta_E E + \Delta(\delta_L V) + \delta_O O \quad (2)$$

where δ represents the HDO or $H_2{}^{18}O$ composition. Here δ_I and δ_O are the isotopic compositions of inflow and outflow rivers, δ_P and δ_E are the isotopic composition of precipitation and lake evaporation. For the isotopic composition of lake water (δ_L), three datasets were used, i.e. the one-site data at the MLW site, the ZSW site, and the seasonal whole-lake mean data. We did not measure the isotopic compositions of rainwater in the study period. To calculate monthly δ_P over Lake Taihu, the regression equations derived from the measurement in Changshu Agricultural Experiment Station (31°33'N, 120°42'E) were employed [3]. This site is part of the Chinese Network of Isotopes in Precipitation, and is located in the Lake Taihu catchment. Monthly δ_P was calculated from monthly mean air temperature (T_a) and monthly total sunshine duration (S) using the local precipitation lines $\delta^{18}O_P = -7.564 - 0.006T^2 + 0.023S$ and local MLW $\delta D_P = 8.77\delta^{18}O_P + 13.96$, where air temperature T and sunshine duration S were mean values observed at the three weather stations (Wuxi, Huzhou and Dongshan). Readers should be reminded that regression equation for $\delta^{18}O_P$ is based on statistical analysis and does not mean that the isotopic composition of precipitation was controlled by sunshine duration.

Based on equation 2, δ_E can be calculated as

$$\delta_E = \frac{\delta_I I + \delta_P P - \Delta(\delta_L V) - \delta_O O}{E} \quad (3)$$

S2. Throughflow index and Residence Time

The steady-state models of Gibson et al. [4] was employed to calculate the throughflow index and the residence time of Lake Taihu. Lake Taihu is a throughflow lakes with continuous inflow balanced by a combination of evaporation and outflow. Under the assumption of constant hydrologic fluxes and minor volume variation, the lake can be viewed as in approximate hydrologic steady state.

The throughflow index (x) is the ratio of evaporation to the sum of water incomes (precipitation and inflow), i.e. $x = E/(P + I)$, and can be calculated using the isotopic method as

$$x = \frac{(\delta_L - \delta_{IN})}{m(\delta^* - \delta_L)} \quad (4)$$

where $\delta_{IN} = (I\delta_I + P\delta_P)/(I+P)$, δ^* is the limiting isotopic enrichment [5,6] calculated as

$$\delta^* = (h\delta_v + \varepsilon)/(h - 10^{-3}\varepsilon) \quad (5)$$

and m is the enrichment slope [7,8] given by

$$m = (h - 10^{-3}\varepsilon)/(1 - h + 10^{-3}\varepsilon_k) \quad (6)$$

where h is relative humidity referenced to the lake surface temperature, δ_v is isotopic composition of atmospheric water vapor, ε is the total fractionation factor comprised of equilibrium fractionation factor (ε^*) and kinetic fractionation factor (ε_k). The equilibrium factor ε^* was calculated from lake surface temperature using the function of Majoube (1971) [9], and the kinetic factor was given as $\varepsilon_k = C_k(1 - h)$, where C_k is 14.3‰ and 12.5‰ for $H_2{}^{18}O$ and H^2HO, respectively [10].

The residence time of the lake water is calculated as

$$\tau = xV/E \quad (7)$$

The annual mean value of δ_A, δ^*, h and m were calculated as evaporation flux-weighted means from their respective monthly values. The annual isotopic compositions δ_L and δ_{IN}

were amount-weighted mean values. In this study, data from one complete year (May 2013 to April 2014) when both the lake and the river isotopes measurements were available.

S3. Uncertainty analysis

Monte Carlo simulations were carried out to determine the uncertainty in the residence time calculated from the water budget and the isotopic method. The uncertainty range of the liquid water isotope measurement was 0.3‰ for H^2HO and 0.1‰ for $H_2{}^{18}O$. Uncertainties of the water budget components (*P*, *E*, *V* and *I*) were assumed to be 10% of the measured values. The input variables were assumed to vary in their respective uncertainty ranges according to the normal distribution. The uncertainty of the residence time was calculated as one standard deviation of a total of 20,000 Monte Carlo samples.

References

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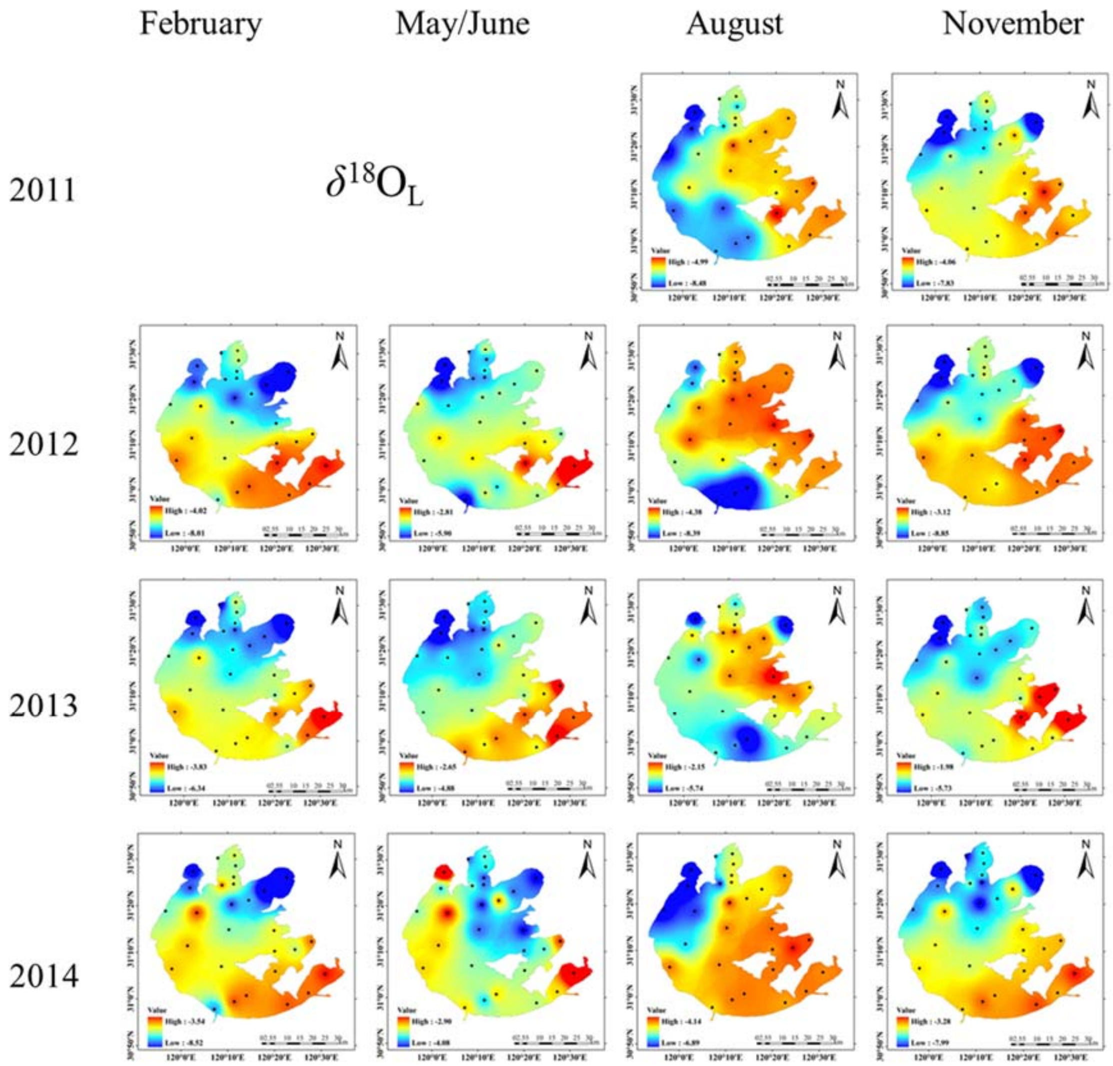
Figure S1. Spatial patterns of $\delta^{18}O_L$ and d_L at each lake survey. The minimum and maximum ranges were 1.2‰ and 5.7‰ for $\delta^{18}O_L$, 7.0‰ and 14.6‰ for d_L.



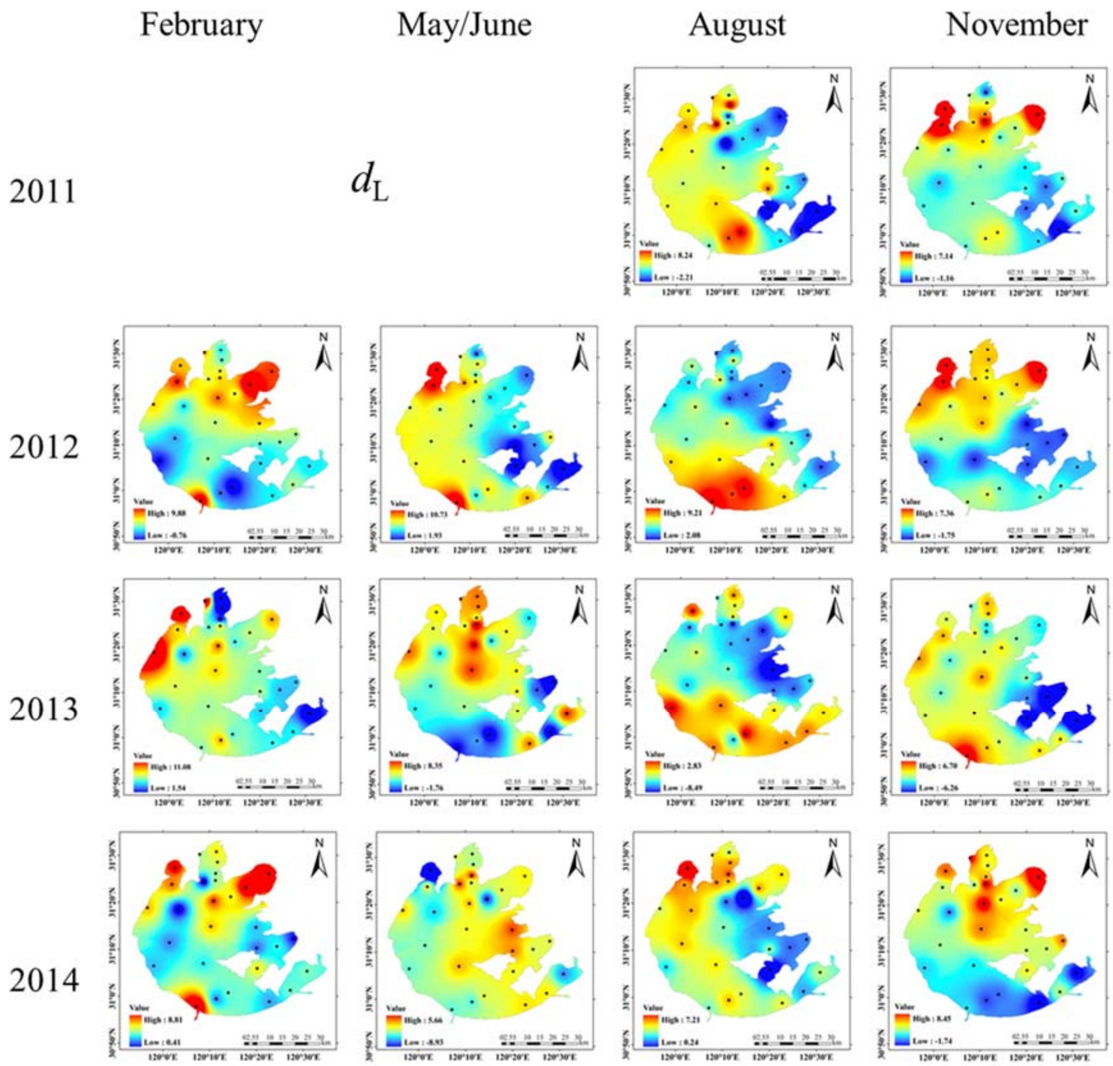

February
May/June
August
November
2011
d_L
2012
2013
2014

Figure S2. Whole-lake mean isotopic composition and outflow/inflow differences of river water isotopes versus $E/(P + I)$.

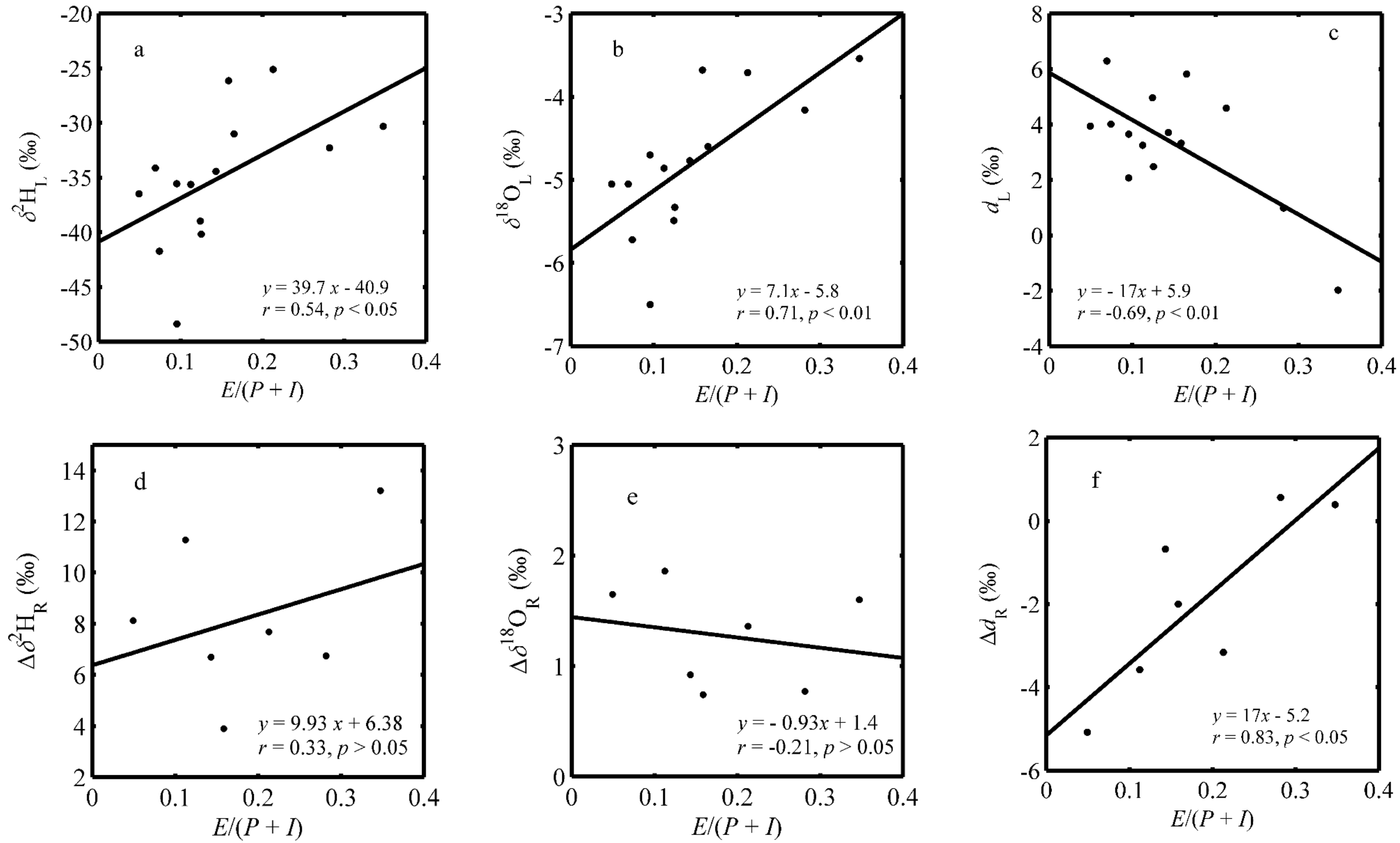


Figure S3. δ^2H versus δ^{18}O values of lake water, river water and lake evaporation. Closed circles: δ_L in inflow zone; open circles: δ_L in transition zone; triangles: δ_L in outflow zone; crosses: δ_R of inflow rivers; pluses: δ_R of outflow rivers; diamond, open square and closed square: δ_E calculated from mass balance using the water isotopic measurement at the ZSW site (location 16), the MLW site and the whole-lake survey; thick solid line: Global Meteoric Water Line (GMWL); thin dashed line: Local Evaporation Line (LEL). Each data point for δ_L and δ_R represents the mean value in one survey, and the data point for δ_E represents a whole-year value.

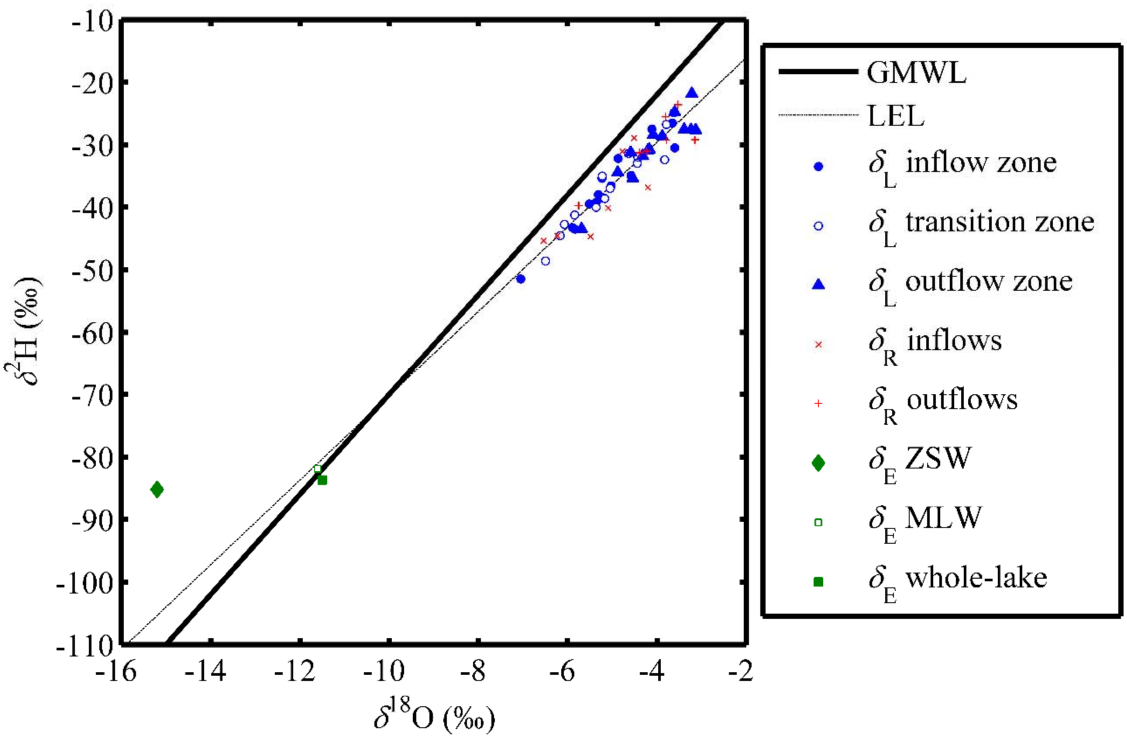


Table S1 Linear correlation of lake and river water isotopic composition with environmental variables.

	Lake isotopes (whole-lake mean value)			Lake isotopes (outflow zone - inflow zone)			River isotopes (outflows-inflows)		
	δ^2H_L	$\delta^{18}O_L$	d_L	$\Delta\delta^2H_L$	$\Delta\delta^{18}O_L$	Δd_L	$\Delta\delta^2H_R$	$\Delta\delta^{18}O_R$	Δd_R
E	0.27	0.36	-0.36	-0.58*	-0.55*	0.31	0.35	-0.05	0.59
P	-0.12	-0.22	0.35	-0.39	-0.43	0.38	-0.48	-0.31	-0.17
P-E	-0.32	-0.50	0.65*	-0.08	-0.15	0.25	-0.69	-0.27	-0.54
x	0.54*	0.71**	-0.69**	-0.40	-0.30	-0.05	0.33	-0.21	0.83*
T_a	0.15	0.22	-0.25	-0.52	-0.47	0.19	0.12	-0.25	0.60
Water depth	-0.40	-0.42	0.16	0.02	0.06	-0.12	-0.22	-0.47	0.48
S_d	0.35	0.43	-0.36	-0.51	-0.46	0.17	0.20	-0.20	0.62
S	0.22	0.33	-0.41	-0.42	-0.28	-0.16	0.22	-0.19	0.64

Note: *, $p < 0.05$; **, $p < 0.01$