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QUANTIFYING CLOSED-BASIN LAKE TEMPERATURE AND HYDROLOGY BY INVERSION OF OXYGEN ISOTOPE AND TRACE ELEMENT PALEOCLIMATE RECORDS

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ABSTRACT. Lake systems are important paleoclimate archives that preserve ecosystem and hydrologic responses to critical periods in Earth history, such as carbon cycle perturbations and glacial-interglacial cycles. Geochemical measurements of biogenic carbonate (for example, $\delta^{18}\text{O}$, $\delta^{13}\text{C}$, $^{87}\text{Sr}/^{86}\text{Sr}$, [Li], [U], [Sr], and [Mg]) are indicators of hydrologic variability in lake systems throughout the geologic record. In this study, we present a new closed-basin lake modeling approach, HyBIM (the Hydrologic Balance Inverse Model) that employs a system of total differential equations and uses the measured $\delta^{18}\text{O}$, Sr/Ca, and Mg/Ca of biogenic carbonate to determine changes in temperature, runoff, and lake evaporation. Using equally-spaced time steps, these equations are simultaneously solved to constrain the hydrologic parameters of the lake as recorded in biogenic carbonate. We use a Monte Carlo approach to account for uncertainty in the input parameters, such as $\delta^{18}\text{O}$ temperature relationships, partition coefficient uncertainty, and watershed solute chemistry.

For illustrative purposes, we apply the model to two ostracod valve datasets covering different timescales: (1) the Cretaceous Songliao Basin, northeast China, and (2) Holocene Lake Miragoane, Haiti. Modern water measurements of water isotopes and cation concentrations from each location are required as model inputs. We compare our modeling results with author interpretations and geologic observations. The modeling approach presented in this study can be applied to other closed-basin lake records, can be modified for other calcifying species (for example, gastropods or mollusks) or with calibration to inorganic lacustrine carbonate. In addition, this approach holds promise for extension with additional proxy measurements (that is, δD , U/Ca or Li/Ca) and changing source area on tectonic timescales using proxies that reflect changing source lithology (that is, Sr and Pb isotopes). Future incorporation of age model uncertainty in the Monte Carlo approach will also provide utility by quantifying temporal uncertainty on the hydrologic response recorded by lake sediments.

Keywords: lakes, paleohydrology, inverse model, trace elements, oxygen isotopes

INTRODUCTION

Lake systems provide valuable paleoclimate archives as natural integrators of terrestrial processes. Lake deposits from hydrologically closed basins or terminal lakes record important changes in climate, human activity, hydrology, ecology, and tectonics (Antevs, 1948; Eugster and Jones, 1979; Currey, 1990; Binford and others, 1997; Carroll and Bohacs, 1999; Reheis, 1999; Demko and others, 2005; Horton and Chamberlain, 2006; Davis and others, 2009; Reheis and others, 2014; Hillman and others, 2014; Smith and others, 2014). Additionally, lake sediments provide the most widely available and continuous records of terrestrial climate available to reconstruct

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past climate (for example, Oviatt and McCoy, 1992; Colman and others, 1995; Williams and others, 1997; Bohacs and others, 2003; Melles and others, 2012). Accurate estimates of past precipitation, runoff, and/or temperature are important for studying the evolution of landscapes and the diversification and migration of species, as well as to test climate model predictions of past climate (for example, Rozanski and others, 1997; Partridge and others, 1997; Steinman and others, 2012; Ibarra and others, 2014).

Studies of ancient lake systems have long sought to infer climate conditions leading to the existence and persistence of lakes in terrestrial settings. Inferring the climatic setting and the open versus closed hydrology of a lake system using facies identification has been extensively investigated (for example, Nilsson, 1931; Eugster and Kelts, 1983; Kelts, 1988; Talbot, 1990; Carroll and Bohacs, 1999). Within the Carroll and Bohacs (1999) framework, closed-basin lakes are of two types: balanced-fill and underfilled. Underfilled lake systems are identified by evaporative facies (Jones and others, 1977; Eugster and Jones, 1979; Horita, 1990) and are typical of many Quaternary playa-lake systems found in semi-arid regions of the mid-latitudes, such as the western United States (Mifflin and Wheat, 1979; Reheis and others, 2014). In contrast, balanced-filled lake systems are typical of freshwater to saline conditions with mixed carbonate and siliciclastic sediments (for example, Carrol, 1998; Kempf and others, 2009; Doebbert and others, 2014).

For closed-basin Quaternary lake systems, in which tectonic rearrangement is negligible, climatic conditions have been quantified using measures of lake surface area and tributary size (Snyder and Langbein, 1962; Mifflin and Wheat, 1979; Bowler, 1981; Benson and Paillet, 1989; Bengtsson and Malm, 1997; Reheis, 1999; Sack, 2009; Broecker, 2010; Munroe and Laabs, 2013; Hudson and Quade, 2013; Ibarra and others, 2014; Huth and others, 2015; Hudson and others, 2015). Additionally, recent work modeling stable isotopes in closed-basin balance-filled lakes takes advantage of longer (relative to overfilled/open lakes) residence times to infer climatic conditions by relating the $\delta^{18}\text{O}$ or δD to the hydrologic balance (Horita, 1990; Hostetler and Benson, 1994; Benson and White, 1994; Gibson and others, 2002; Gibson and Edwards, 2002; Benson and Paillet, 2002; Russell and Johnson, 2006; Jones and others, 2007; Jones and Imbers, 2010; Doebbert and others, 2010; Steinman and others, 2010a, 2010b, 2012, 2013; Steinman and Abbott, 2013; Jasechko and others, 2013; Stansell and others, 2013; Ibarra and others, 2014; Jasechko and others, 2014; Gibson and others, 2015). Fundamental to applying an isotopic modeling approach are assumptions concerning the watershed precipitation-runoff relationships (Jones and others, 2007; Ibarra and others, 2014), mixing or stratification of the lake (Imberger and Ivey, 1991), basin-scale and regional vapor recycling (Gat and Matsui, 1991; Gat and others, 1994; Burnett and others, 2003; Winnick and others, 2014), and groundwater influences (Krabbenhof and others, 1990a, 1990b; Shapley and others, 2005; Guay and others, 2004; Steinman and others, 2013). While these models offer powerful methods to reconstruct the hydrology of lake systems they are limited by the numerous input parameters required that are difficult to constrain in ancient lake systems. Another approach, outlined here, is to use additional measurements, such as trace elements, that can be meaningfully related to climatic conditions and will reduce the assumptions necessary for accurate, quantitative climatic reconstructions. Using trace element and oxygen isotope measurements it is then possible to tightly constrain the system using far fewer input parameters.

In this paper, we develop a new modeling framework exploiting variations in $\delta^{18}\text{O}$ and trace elements (Mg and Sr) measured in lacustrine carbonates and utilizing a series of total differential equations. We apply this model to previously published ostracod paleoclimate records for closed basins. Lacustrine ostracod records are used here for illustrative purposes because of their broad paleoclimate applications in historical (for example, Engstrom and Nelson, 1991; Holmes and others, 2007a; Zhang and others, 2009), Quaternary (for example, Edney and others, 1990; Hodell and

others, 1991; Curtis and Hodell, 1993; Lamb and others, 1999; Cohen and others, 2000; Holmes and others, 2007b; Gouramanis and others, 2010), and deep-time (for example, Forester, 1991; Chamberlain and others, 2013) settings. Extensive work has also been carried out on the ecology, taphonomy, and taxonomy of ostracod species (for example, Horne and others, 2002; Frogley and others, 2002; Bennett and others, 2011; Blome and others, 2014), and modern ostracod datasets have been extensively studied and provide the necessary empirical calibrations of trace element distribution coefficients and vital effects (for example, Engstrom and Nelson, 1991; Xia and others, 1997a, 1997b; Wansard and others, 1998; De Deckker and others, 1999; Dettman and others, 2002; Holmes and Chivas, 2002; Dwyer and others, 2002; Ito and Forester, 2009; Gouramanis and others, 2010; Marco-Barba and others, 2012; Börner and others, 2013) necessary to constrain our model. The modeling framework presented in this paper applies, given the existence of relevant and accurate calibrations, to any biogenic carbonate records from a closed-basin lake setting.

MODEL DEVELOPMENT

We present the Hydrologic Balance Inversion Model (HyBIM), an inverse model using a system of total differential equations that are inverted to solve for the extensive variables in lake chemistry. The structure of the model is similar to models that quantify changes in the global carbon cycle and/or other biogeochemical cycles as recorded in marine sediments (for example, Godd eris and Fran ois, 1996; Derry and France-Lanord, 1996; Fran ois and Godd eris, 1998; Li and others, 2009; Li and Elderfield, 2013). We solve for the relative changes in temperature and lake volume, and thus rely only on the first derivative of the input dataset and not the absolute value of the isotopic and trace element measurements. Because of this formulation order of magnitude fluctuations in lake volume cannot be modeled by this approach. Fundamentally, this model quantifies the observations made by previous studies that qualitatively employed end-member mixing (from a fresh source to an evaporative lake) in closed-basin lacustrine deposits based on the covariance of Sr/Ca, Mg/Ca, $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ (for example, M uller and others, 1972; Eugster and Kelts, 1983; Talbot, 1990; Li and Ku, 1997; Garnett and others, 2004; Davis and others, 2009; Horton and Oze, 2012; McGee and others, 2012; Chamberlain and others, 2013; Horton and others, 2015). We do not incorporate variations in $\delta^{13}\text{C}$ in this initial iteration of HyBIM; however, evaporative processes and lake residence times should also control the systematics of the $\delta^{13}\text{C}$ of carbonates from terminal lake systems (see discussion in Horton and others, 2015).

First, we establish the modeling framework using a system of total differential equations. Second, we describe the Monte Carlo routine used to account for parameter and input data uncertainty (for example, Fantle, 2010; Royer and others, 2014). Third, we discuss the input dataset (the time series of measurements) and necessary input parameters. Finally, we describe the quantification of the partial derivatives necessary to constrain the model and necessary input parameters. This model is available as an example R code for modification at <http://earth.geology.yale.edu/~ajs/SupplementaryData/2015/Ibarra/>. For illustrative purposes, we apply HyBIM to two ostracod valve datasets covering different timescales: (1) the Cretaceous Songliao Basin, northeast China, and (2) Holocene Lake Miragoane, Haiti. All model parameters are summarized in table 1 and a schematic of the model structure is presented in figure A1.

Model Framework

Here, we present a modeling framework that produces a time series of temperature changes, effects of evaporation, and relative volume estimates for runoff for carbonate-bearing (here ostracods) paleolake records. Chivas and others (1986) previously suggested a quantitative framework by which a unique solution for paleotemperature and paleosalinity could be calculated using paired Mg/Ca and $\delta^{18}\text{O}$ measurements. Following this observation, we write three total differential equations for lake water that describe the effects of temperature,

TABLE 1
Definition of model parameters for HyBIM as used in the text

Parameter	Description
$dT, \partial T$	Change in basin air temperature (mean annual surface temperature)
$dF_{\text{evap}}, \partial F_{\text{evap}}$	Change in amount of lake water (volumetrically) evaporated, where $F_{\text{evap}} = 1$ is fully evaporated and $F_{\text{evap}} = 0$ is no evaporation
$dF_{\text{input}}, \partial F_{\text{input}}$	Change in volumetric increase in lake water, where $F_{\text{input}} = 1$ is no change
Δt	Time step of model
τ_{res}	Residence time of lake water
α	Combined fractionation factor for Rayleigh equation (kinetic and equilibrium)
$d(\delta^{18}\text{O}), \partial(\delta^{18}\text{O})$	Change in oxygen isotopic composition of lake water
$(\delta^{18}\text{O})_{\text{source}}$	Oxygen isotopic composition of incoming meteoric water
$(\delta^{18}\text{O})_{\text{initial}}$	Oxygen isotopic composition of lake water for a given time step, derived from the measured $\delta^{18}\text{O}$ in biogenic carbonate, corrected for vital effects
Sr/Ca	Strontium to calcium ratio measured in biogenic carbonate
Mg/Ca	Magnesium to calcium ratio measured in biogenic carbonate
$d[\text{Sr}], \partial[\text{Sr}]$	Change in strontium concentration of lake water
$d[\text{Mg}], \partial[\text{Mg}]$	Change in magnesium concentration of lake water
$d[\text{M}], \partial[\text{M}]$	Change in magnesium or strontium concentration of lake water
$[\text{M}]_{\text{source}}$	Magnesium or strontium concentration of incoming meteoric water (weighted streamwater and rainwater)
$[\text{M}]_{\text{initial}}$	Magnesium or strontium concentration of lake water for a given time step, derived from the measured Sr/Ca and Mg/Ca in biogenic carbonate using the appropriate partition coefficient and calcium concentration.
$K_{\text{p}}[\text{M}]$	Partition coefficient for Sr/Ca or Mg/Ca

evaporation, and runoff/precipitation on the oxygen isotopic and trace element concentration of ostracods in a hydrologically closed lake. The three total differential equations (for lake water $\delta^{18}\text{O}$, $[\text{Mg}]$, and $[\text{Sr}]$) are:

$$d(\delta^{18}\text{O}) = \left(\frac{\partial(\delta^{18}\text{O})}{\partial T} \right) dT + \left(\frac{\partial(\delta^{18}\text{O})}{\partial F_{\text{evap}}} \right) dF_{\text{evap}} + \left(\frac{\partial(\delta^{18}\text{O})}{\partial F_{\text{input}}} \right) dF_{\text{input}} \quad (1)$$

$$d[\text{Sr}] = \left(\frac{\partial[\text{Sr}]}{\partial T} \right) dT + \left(\frac{\partial[\text{Sr}]}{\partial F_{\text{evap}}} \right) dF_{\text{evap}} + \left(\frac{\partial[\text{Sr}]}{\partial F_{\text{input}}} \right) dF_{\text{input}} \quad (2)$$

$$d[\text{Mg}] = \left(\frac{\partial[\text{Mg}]}{\partial T} \right) dT + \left(\frac{\partial[\text{Mg}]}{\partial F_{\text{evap}}} \right) dF_{\text{evap}} + \left(\frac{\partial[\text{Mg}]}{\partial F_{\text{input}}} \right) dF_{\text{input}} \quad (3)$$

where $\delta^{18}\text{O}$ is the oxygen isotopic composition of the lake water determined from biogenic carbonate measurements corrected for species-specific vital effects, $[\text{Mg}]$ and

[Sr] are the lake water concentrations of Mg^{2+} and Sr^{2+} determined from the Sr/Ca and Mg/Ca ratios measured in the biogenic carbonate, T is mean annual surface air temperature (K) for the lake basin, F_{evap} is the fraction of the lake evaporation, and F_{input} is the fractional lake volume increase due to changes in runoff and precipitation (see table 1).

We treat equations 1–3 as a system of total differential equations that can be solved for each time step of the first derivative of the [Mg], [Sr], and $\delta^{18}\text{O}$ time series [that is, $\mathbf{x} = \mathbf{A}^{-1}\mathbf{b}$, where matrix \mathbf{A} is composed of the partial derivatives, the \mathbf{b} vector is the first derivative of the input dataset (left side of eqs 1 to 3) and \mathbf{x} is the solution vector of (dT , dF_{evap} and dF_{input})]. We do so by determining the partial derivatives for a specific lake system, calibrated using modern watershed/lake chemistry data and knowledge of the specific ostracod species. Interpolation to an evenly spaced time series is required, which we accomplish using an Epanechnikov kernel smoother applied to the input data. We assume that within each time step, lake evaporation and/or lake volume change does not change more than 20 percent, and we approximate all derivatives according to this assumption (that is at 10%). The model output is a time series of the solution vector (dT , dF_{evap} and dF_{input}). dF_{evap} and dF_{input} are combined to give the fractional net volume increase for each time-step (see fig. A1). We use a Monte Carlo approach to account for uncertainty in the input parameters and in the input dataset, which we derive from kernel smoothing.

Monte Carlo Routine for Uncertainty Quantification

Given the assumptions that underlie the input parameters and data set for this model it is critical to quantitatively assess the errors associated with the calculated runoff input, temperature changes, and evaporative effects. Thus, we use a Monte Carlo routine to account for two types of uncertainty. The first is uncertainty in the input dataset. Since most paleoclimate records display uneven variance and non-uniform sampling density, it is necessary to account for this by producing datasets for each iteration based on the statistics (time series of the mean and residuals) derived from kernel smoothing of the dataset (see below). To do so, for each Monte Carlo iteration we implement a bootstrapping method whereby we account for input dataset uncertainty by re-sampling residuals. The second type of uncertainty accounted for by our model is the uncertainty in the input parameters. The input parameters are best determined by modern watershed and lake chemistry (when available) and knowledge of the species precipitating the biogenic carbonate (described below). Within each Monte Carlo iteration, we derive a time series of partial derivatives based on the input parameters. Input parameter uncertainty distributions are assumed to be normal distributions, requiring that uncertainty is quantified by as many modern measurements of watershed chemistry as possible. The partial derivatives are substituted into the \mathbf{A} matrix at each time step.

Input Dataset Requirements and Interpolation

There are two requirements for the input dataset that need to be met for this model. First, the modeled Mg/Ca, Sr/Ca, and $\delta^{18}\text{O}$ time series must be of sufficient resolution to resolve climatic events of interest and the time step must be greater than the residence time of these elements in a lake. Thus, in order to use the dataset in the system of equations (left side of eqs 1–3), the time series must be interpolated to even time steps, requiring a sufficiently resolved age model for the Mg/Ca, Sr/Ca, and $\delta^{18}\text{O}$ measurements. Smoothing of each time series (Mg/Ca, Sr/Ca, and $\delta^{18}\text{O}$) is performed using an Epanechnikov kernel and interpolated to an even time step following a data driven least squares cross-validation to select the appropriate kernel bandwidth (Hayfield

and Racine, 2008). The Epanechnikov kernel (Epanechnikov, 1969) is used because it has finite boundaries (instead of Gaussian kernel) and is smooth (compared to a triangle or uniform kernel) (see Fan, 1992; Danese and others, 2014). Kernel smoothing of the time series is implemented using the ‘np’ package in R (Hayfield and Racine, 2008). The additional advantage of using a kernel smoother is that uncertainty of the interpolated dataset can be quantified along the time series (mean and residuals) and input into the Monte Carlo routine.

While high-resolution interpolation of the smoothed dataset is desirable to resolve climatic events of interest, linear interpolation of the input dataset to an even time step (Δt) must be such that $\Delta t \gg \tau_{\text{res}}$, where τ_{res} is the residence time (inferred from lake geometry and average input or evaporative fluxes, for example Gibson and others, 2002; Jasechko and others, 2014). For lakes with no modern analogue (see Cretaceous Songliao Basin example), first-order estimates for τ_{res} can be approximated using scaling relationships between outcrop size and basin geometry (Hendriks and others, 2012), which in combination with facies identification can be used to determine estimates of the relationship between potential accommodation and basin average precipitation/evaporation (Carroll and Bohacs, 1999). Thus, due to the formulation and assumptions of the mixing and mass conservation relationships derived below, resolving hydrologic or climatic events over intervals equal to or shorter than τ_{res} is not possible within this modeling framework. This implies that, given the ‘water equivalency rule’ for a region of interest (Hendriks and others, 2012), paleoclimate records from small lake systems, which integrate smaller watersheds and have shorter residence times, provide the highest fidelity temporal records of hydrologic cycle changes. Furthermore, determination of the lake hypsometry and/or outcrop extent (for example, Reheis 1999; Sack, 2009; Steinman and others, 2013; Smith and others, 2014; Doebbert and others, 2014) ensures that the modeling assumptions outlined above are met.

The second requirement for the model is that all of the input data need to be placed in terms of lake composition. Although we use measured values of Mg/Ca, Sr/Ca, and $\delta^{18}\text{O}$ of ostracods, these values need to be corrected so that they represent the values of these elements and isotopes in the lake water. To make this correction one must correct for “vital effects” and temperature. Correcting for vital effects is relatively straightforward as the measured vital effect is simply subtracted from the measured ostracod values. Vital effects associated with the calcification of biogenic carbonate are observed because shell carbonate is not precipitated in isotopic equilibrium with water (Holmes and Chives, 2002). Vital effects observed in ostracod $\delta^{18}\text{O}$ are positive (0.3 – 2.5 ‰) and have been observed to be constant within individual genera (Xia and others, 1997a, 1997b; von Grafenstein and others, 1999; Chivas and others, 2002; Didie and Bauch, 2002). Since vital effects are not temperature-dependent, data from a variety of different modern species fall on slopes close to the inorganic calcite equilibrium slope (-0.250 ‰/K) of Kim and O’Neill (1997) (see compilation by Marco-Barab and others, 2012). Our model ideally should be used with a time series produced by a single ostracod species or genera. Although, if multiple species are measured (for example, Lister and others, 1991) this could be accounted for using vital effect offsets.

In contrast, correcting for temperature effects is less straightforward and could, if done incorrectly, lead to “circular reasoning” since we are attempting to constrain temperature changes in the model. We assume temperature could vary from 10 to 35 °C (uniform distribution). For each Monte Carlo iteration we select a starting temperature and assume that $\delta^{18}\text{O}$ of the lake water is in equilibrium with the measured $\delta^{18}\text{O}$ of ostracods using the temperature-dependent fractionation relationship of Kim and O’Neill (1997) corrected for vital effects. We avoid “circular reasoning” since our

system of linear equations calculates the relative changes in temperature, dT (in the x vector), using the first derivative of the $\delta^{18}\text{O}$ of the lake water (in the b vector), not the absolute temperature.

Distribution coefficients (K_D -values) relating trace element to calcium ratios are commonly determined for biogenic carbonates through modern and/or culturing studies. The distribution (or partition) coefficient is defined as:

$$K_D[M] = \frac{M/\text{Ca}_{\text{water}}}{M/\text{Ca}_{\text{shell}}} \quad (4)$$

where M is the trace metal, such as Sr, Mg, or U, and M/Ca are molar ratios. K_D -values for Mg and Sr are both less than one, meaning that both trace metals are actively excluded during calcification. Additionally, Chivas and others (1986) demonstrated that K_D -values are similar among members of the same or closely related genera.

In many species, K_D has been shown to be temperature-dependent. In most ostracod species, there is little or no temperature effect on Sr/Ca (Chivas and others, 1986; Holmes and Chivas, 2002, Marco-Barab and others, 2012), but the Mg/Ca K_D in ostracods is temperature-dependent (Engstrom and Nelson, 1991; DeDeckker and others, 1999; Holmes and Chivas, 2002). For the example datasets used in this paper, we use species-specific K_D -values (or from related genera when not available), and uncertainties are obtained from regression statistics of the original datasets (table 2). To place the input data $[\text{Mg}]$ and $[\text{Sr}]$ in terms of lake composition (b vector) we calculate the Mg/Ca K_D (and Sr/Ca K_D if necessary) using the temperature and lake water $[\text{Ca}]$ selected for each Monte Carlo iteration.

Determining Partial Derivatives

In the following sections each of the partial derivatives are derived or determined using either mass conservation (for example, $\frac{\partial[\text{Mg}]}{\partial F_{\text{evap}}}$) and mixing (for example, $\frac{\partial[\text{Mg}]}{\partial F_{\text{vol}}}$), or empirical relationships from environmental and/or laboratory measurements (for example, $\frac{\partial(\delta^{18}\text{O})}{\partial T}$).

Temperature Partial Derivatives

To solve for the oxygen isotope partial derivative $\frac{\partial(\delta^{18}\text{O})}{\partial T}$ requires knowing how $\delta^{18}\text{O}$ of meteoric water (precipitation and runoff) varies as a function of temperature. We recognize that there are numerous factors affecting the $\delta^{18}\text{O}$ of precipitation beyond that of temperature, which include seasonality, vapor recycling, moisture source *et cetera*. However, the commonly held assumption that underpins terrestrial paleoclimate studies is the well-known positive correlation between $\delta^{18}\text{O}$ values of precipitation and temperature for temperate and high latitude areas (Dansgaard, 1964). For the mid-latitude site (Songliao Basin) we use the observed relationship between temperature and precipitation Rozanski and others (1993) of 0.58 ‰/K, as has been used in numerous studies to calculate the temperature for paleoclimate archives in mid- to high-latitude sites, such as paleosols (for example, Koch and others, 2003), paleolakes (for example, Anderson and others, 2001), and ice cores (see Jouzel and others, 1997 and references therein). For the low latitude site (Lake Miragoane, Haiti) it is unlikely that the correlations of Rozanski and others (1993) apply as these are for mid- to high-latitudes. Although it is not ideal to use the approach we outline in this paper to low latitudes because temperature is not as well correlated to the $\delta^{18}\text{O}$ of precipitation, we use this site only as an example of how this approach can be used in

TABLE 2
Details of input dataset and input parameters for HyBIM application to Lake Miragoane and Songliao Basin

Record	Age Range	Original References	$\delta^{18}\text{O}$		Mg/Ca		Sr/Ca		$\delta^{18}\text{O}$ Kernel		Mg/Ca Kernel		Sr/Ca Kernel	
			Average Resolution	Average Resolution	Average Resolution	Average Resolution	Bandwidth	Bandwidth	Bandwidth	Bandwidth	Bandwidth	Bandwidth	Bandwidth	Bandwidth
Lake Miragoane, Haiti	Holocene (10.56 - 0.05 ka BP)	Hodell and others (1991); Curtis and Hodell (1993)	16.9 years	176.5 years	173.5 years	38.9 years	422.6 years	440.1 years						
Songliao Basin, China	Cretaceous (88.04-84.02 Ma)	Chamberlain and others (2013)	24.4 kyrs	27.9 kyrs	27.9 kyrs	61.7 kyrs	240.9 kyrs	219.0 kyrs						
Record	Ostracod Genus	Partition Coefficient	Vital Effect		Input Solute Concentrations		Input $\delta^{18}\text{O}$							
Lake Miragoane, Haiti	<i>Candona</i> genus (identified by R. Forester, see details in Curtis and Hodell, 1993)	As determined for <i>Candona rawsoni</i> by Engstrom and Nelson (1991). Temperature effect for Mg/Ca only.	+ 2.5 \pm 0.2 ‰ based on <i>Candona candida</i> . (Keatings, 1999)		Average of watershed [Sr] and [Mg], and lake [Ca] from Holmes and others (1995a,b)		-8 \pm 2 ‰ (OIPC; Bowen and Revenaugh, 2003)							
Songliao Basin, China	Sampled extinct <i>Cypridea</i> genus, closely related to extant genus <i>Cyprideis</i>	As determined for <i>Cyprideis</i> by DeDecker and others (1999). Temperature effect for both Sr/Ca and Mg/Ca.	Assumed to be 1.3 \pm 0.2 ‰ (Average of compilation in Holmes and Chivas, 2002)		Average of watershed [Sr] and [Mg], and lake [Ca] from Han and Huh (2009), Wang and others (2012), Liu and others, (2013), Moon and others (2009)		-18 \pm 2 ‰ (estimated from Poulsen and others, 2007)							

lakes because this record offers the type of data that are necessary for these calculations. Recognizing these limitations we use a slope of 0.26‰/K for the tropical sites (see fig. A2). For both cases we incorporate the error associated with the empirical temperature versus $\delta^{18}\text{O}$ correlation in the Monte Carlo.

In our model, thus, one of the assumptions inherent in our calculations is that temperature is the primary driver of $\delta^{18}\text{O}$ value of water input to the lake. Note that since the slope between temperature and $\delta^{18}\text{O}$ of precipitation can and does vary geographically, more accurate approaches can be tailored for individual sites by measurements of site-specific correlations between temperature and $\delta^{18}\text{O}$ of precipitation.

In addition, to the partial $\frac{\partial(\delta^{18}\text{O})}{\partial T}$ discussed above it is necessary to place the ostracod values as the $\delta^{18}\text{O}$ of lake water for these calculations (b vector of eqs 1 to 3). To do this we need to account for both vital effects and the temperature-dependent isotopic fractionation (see above).

In addition, there are minor temperature effects on the Sr/Ca (typically none) and Mg/Ca (see above discussion). Since the inputs (b vector of eqs 1 to 3) are placed in terms of lake composition, after accounting for the Mg/Ca or Sr/Ca of the lake water and selected temperature for the Monte Carlo iteration the temperature partial derivatives ($\frac{\partial[\text{Mg}]}{\partial T}$ and $\frac{\partial[\text{Sr}]}{\partial T}$) are equal to zero.

Evaporation Partial Derivatives

To account for the evaporative concentration of trace elements, we assume mass conservation of Sr and Mg. Thus, this model is not applicable to brine or saline environments (see Jones and others, 1977; Eugster and Jones, 1979) nor is it applicable to systems that form large deposits of chemical sediments with high Mg and/or Sr concentrations. For biogenic carbonates, we apply the applicable distribution coefficient (K_D) and take the first derivative. In doing so, we assume that the Sr/Ca and Mg/Ca measured in the biogenic carbonates are faithfully recording changes in the Sr and Mg concentrations of the lake water. Assuming mass conservation, the change in solute M concentration (from the initial concentration to an increased concentration at time 2) during fractional lake evaporation (F_{evap} fraction remaining, from 0 to 1, where 1 is fully evaporated) is given by the equation (fig. 1):

$$[\text{M}]_2 = \frac{[\text{M}]_{\text{initial}}}{(1 - F_{\text{evap}})} \quad (5)$$

Solving for the partial derivative of equation 5 has the analytical solution:

$$\frac{\partial[\text{M}]}{\partial F_{\text{evap}}} = \frac{[\text{M}]_{\text{initial}}}{(1 - F_{\text{evap}})^2} \quad (6)$$

This relationship is non-linear; thus, to provide a linear slope for $\frac{\partial[\text{M}]}{\partial F_{\text{evap}}}$ we assume that for any given time step, the lake does not evaporate more than 20 percent by volume. By inspection of equation 6, the average evaporation between 0 to 20 percent, given by $F_{\text{evap}} = 0.1$, yields a slope of $\frac{[\text{M}]_{\text{initial}}}{(0.9)^2}$. Therefore, the initial trace element concentration (that is, the Mg and Sr concentrations), which we assume to be approximated using the observed distribution of trace element measurements, is important for setting $[\text{M}]_{\text{initial}}$ at each time step.

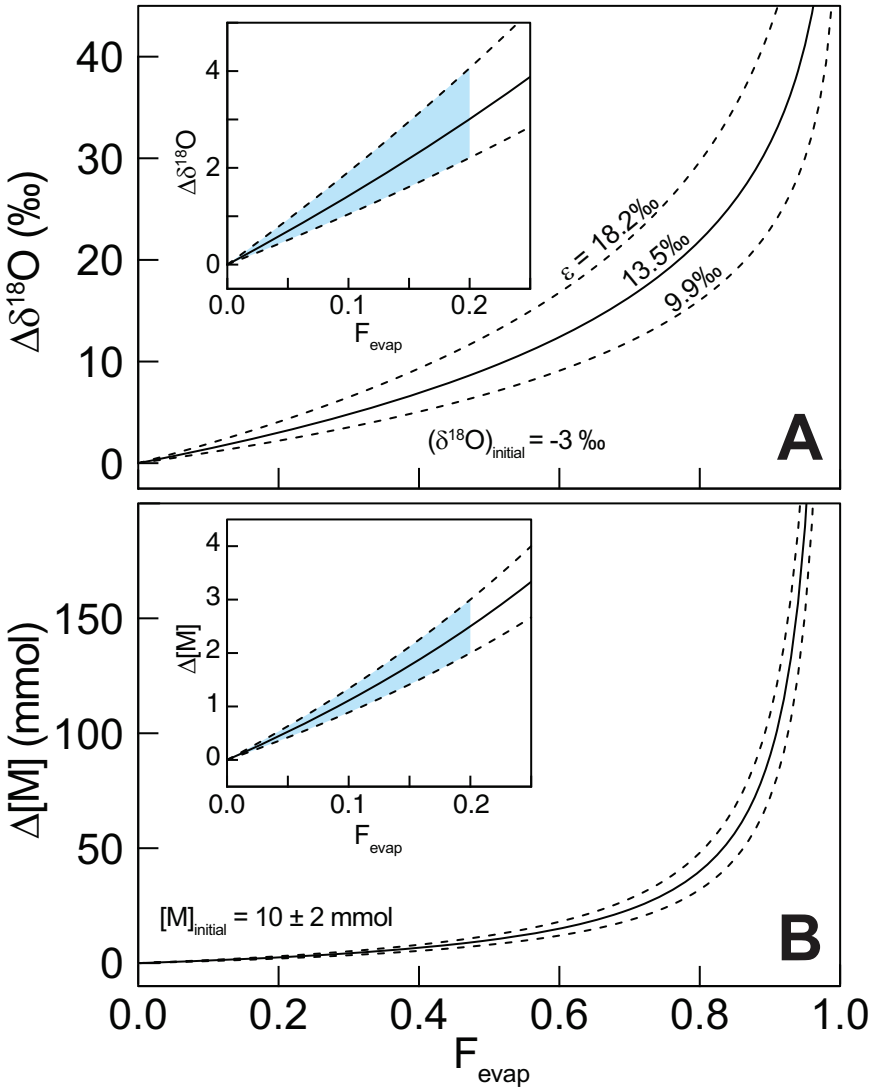


Fig. 1. Illustration of evaporation relationships used to parameterize the partial derivatives. Inset plots show regions of linear approximation assuming $F_{\text{evap}} < 0.2$ within each time step gray shading (blue shading in online versions). Dashed lines are uncertainty. See text for equations describing relationships. (A) Rayleigh evaporation of $\delta^{18}\text{O}$, total (kinetic and equilibrium) fractionation, ϵ , is $13.5 + 4.7 / -3.6\text{‰}$ ($5 - 35\text{ }^{\circ}\text{C}$ and $50 - 90\%$ humidity). (B) Mass conservation of solute, M (that is, Mg or Sr), with an initial lake concentration of 10 ± 2 mmol.

The evaporative enrichment of lake water $\delta^{18}\text{O}$ with progressive evaporation was originally parameterized by Craig and Gordon (1965). The limitation of applying the Craig and Gordon (1965) evaporation model to lacustrine paleoclimate records, as outlined in detail by Gonfiantini (1986), is the large number of input parameters and the difficulty in constraining the isotopic composition of the evaporated water vapor (see discussion in Jones and Imbers, 2010). Recent work has attempted to simplify evaporative assumptions (Benson and White, 1994; Jones and others, 2007; Jones and Imbers, 2010; Placzek and others, 2011), but numerous input parameters are still

required, making the generic application of these lake water evaporation models to paleoclimate records challenging.

Central to the difficulties encountered by previous researchers is the influence of humidity on the evolution of the isotopic composition of an evaporating water body (Gonfiantini, 1986). This is due to changes in the kinetic enrichment factor due to changes in humidity (Gat, 1970; Merlivat, 1978; Merlivat and Jouzel, 1979). However, if within a given time step lake evaporation does not exceed ~25 percent total lake volume and humidity is <90 percent, the isotopic evolution of an evaporating water body can be approximated by a Rayleigh relationship (Gonfiantini, 1986). Thus, we derive the partial derivative for $\delta^{18}\text{O}$ and fractional lake evaporation as the first derivative of the Rayleigh equation (fig. 1):

$$\frac{\partial(\delta^{18}\text{O})}{\partial F_{\text{evap}}} = (\alpha - 1)(\delta^{18}\text{O}_1 + 1000)(1 - F_{\text{evap}})^{(\alpha - 2)} \quad (7)$$

where α is the combined fractionation factor of the temperature-dependent equilibrium fractionation factor (Horita and Weslowski, 1994) and humidity-dependent kinetic fractionation factor (Gonfiantini, 1986), F_{evap} is the fraction of the lake evaporation (as above) and $\delta^{18}\text{O}_{\text{initial}}$ is the initial isotopic composition of the lake from the measured $\delta^{18}\text{O}$ of the biogenic carbonate corrected for vital effects (see below). While a simplification of evaporative processes, assuming a Rayleigh relationship and a combined fractionation factor in this manner has successfully been applied to both surface soil reservoir and lake system modeling (for example, Chamberlain and others, 2014; Caves and others, 2015). We assume that relative humidity for each Monte Carlo iteration can vary from 50 to 90 percent (uniform distribution), and temperature varies from 5 to 35 °C (uniform distribution) as determined for each iteration (as described above). By doing so we calculate the combined fractionation factor, α , using the equations of Horita and Weslowski (1994) and Gonfiantini (1986) (see fig. 1). The humidity and temperature ranges selected here are conservative and broadly apply to both illustrative lake systems used in the examples below. The relevant latitude and geographic setting of the lake system being modeled should inform the humidity and temperature ranges chosen as model input.

Lake Input Partial Derivatives

Determination of the lake input partial derivatives is similar for both trace elements and oxygen isotopes. We rely on binary mixing equations assuming that the isotopic and chemical composition of water entering the lake is sufficiently well characterized and relatively invariant (relative to the lake water). Water in terminal lakes is sufficiently evaporatively enriched relative to the input $\delta^{18}\text{O}$ that in most examples, the input water $\delta^{18}\text{O}$ composition is typically statistically distinguishable from the lake water $\delta^{18}\text{O}$ composition (see for example compilation by Horton and Oze, 2012). This is similar for trace elements in terminal lake systems, such that the input concentration of Sr and Mg in terminal lakes is typically more dilute than the lake water.

Given these principles, we define the partial derivative relating increased lake volume, from the addition of source water (F_{input} , which varies from 1 to infinity, where 1 is the original lake volume) and variations in $\delta^{18}\text{O}$ as the first derivative of a binary mixing relationship (fig. 2):

$$\frac{\partial(\delta^{18}\text{O})}{\partial F_{\text{input}}} = \frac{(\delta^{18}\text{O})_{\text{source}} - (\delta^{18}\text{O})_{\text{initial}}}{(F_{\text{evap}})^2} \quad (8)$$

where $\delta^{18}\text{O}_{\text{source}}$ is the meteoric water composition and $\delta^{18}\text{O}_{\text{initial}}$ is determined as previously described. The meteoric water $\delta^{18}\text{O}$ composition should be determined by

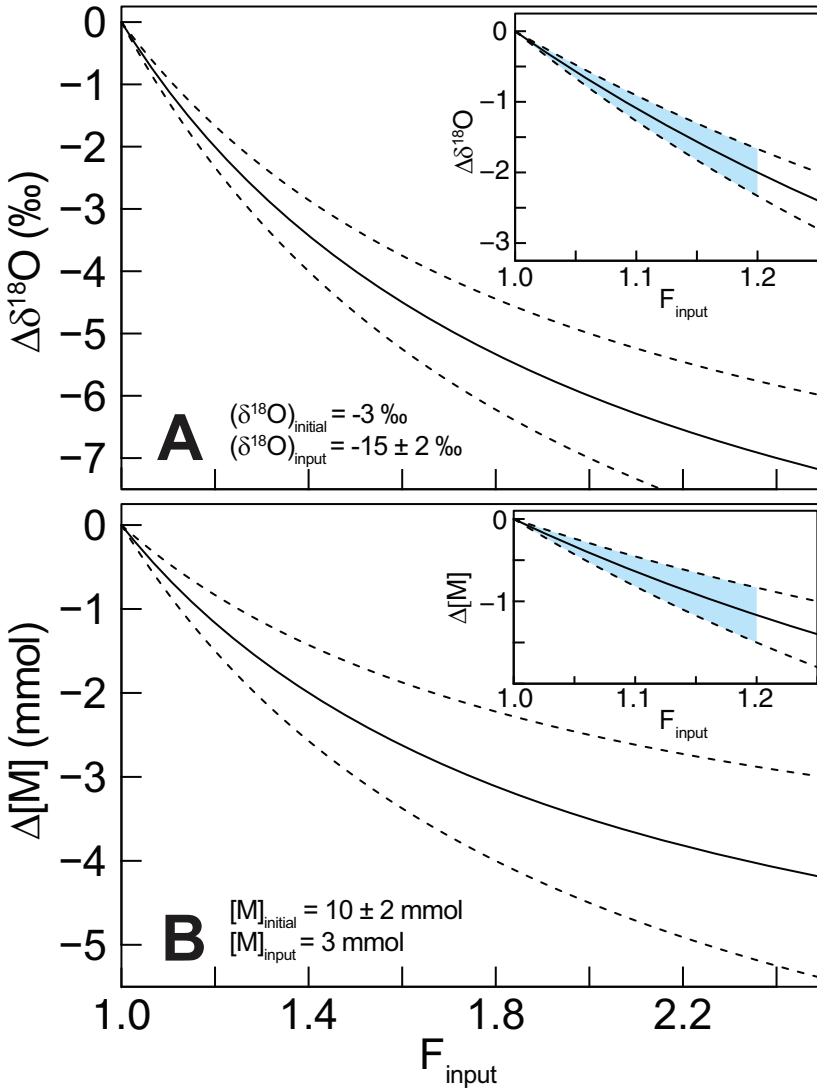


Fig. 2. Illustration of mixing relationships used to parameterize the partial derivatives for volume increase. Inset plots show regions of linear approximation assuming $F_{\text{input}} < 1.2$ within each time step (blue shading in online version). Dashed lines are uncertainty. See text for equations describing relationships. (A) $\delta^{18}\text{O}$ mixing (input water of $-15 \pm 2\text{‰}$, initial lake water of -3‰). (B) Mixing of initial concentration ($10 \pm 2\text{ mmol}$) with a dilute input solute concentration of 3 mmol .

direct observation, by upstream fluvial carbonates (for example Ibarra and others, 2015) or by assuming an empirical relationship related to the amount of annual rainfall (amount effect), as observed in many tropical systems (Risi and others, 2008). Similarly for trace element concentrations:

$$\frac{\partial [M]}{\partial F_{\text{input}}} = \frac{[M]_{\text{source}} - [M]_{\text{initial}}}{(F_{\text{evap}})^2} \quad (9)$$

where M is the Sr or Mg concentration, $[M]_{\text{source}}$ refers to the runoff water concentrations, and $[M]_{\text{initial}}$ is determined as previously described from the measured Mg/Ca and Sr/Ca of the biogenic carbonate.

Input Parameter Requirements

There are several input parameter requirements that need to be satisfied for robust use of this model. First, measurements of solute chemistry ($[\text{Sr}]$, $[\text{Mg}]$, and $[\text{Ca}]$ from lake water and/or runoff) and $\delta^{18}\text{O}$ of meteoric water are necessary to constrain the mixing equation end-members. This can best be done by measuring the elemental and isotopic composition of modern rivers and precipitation in the lake to be studied or the site of the ancient lake as is done here in the Songliao Basin (table 2). There are obvious complications to this approach since the drainage basin may have changed through time as well as the isotopic composition of precipitation – for example by a change in moisture source. In addition, the uncertainty (standard deviation) of the end-members is required. This can be quantified through repeat measurements of stream water/subsurface runoff chemistry.

Second, empirical relationships for temperature effects on the species-specific partition coefficient (for Mg/Ca) are required; such as those fit by Engstrom and Nelson (1991) and DeDeckker and others (1999). As discussed above, species-specific vital effects for $\delta^{18}\text{O}$ offsets are applied uniformly prior to derivation of the partial derivatives from the mixing and mass conservation equations. Input parameters are supplied to HyBIM as the mean and standard deviation, assuming a normal distribution. However, implementation of other probability distributions (such as uniform distributions) is possible. The probable temperature and relative humidity ranges (used for the $\delta^{18}\text{O}$ equilibrium fractionation equations and Mg/Ca K_D values), which are treated as uniform distributions, are specified by the user and should be informed by paleoenvironmental, latitudinal, climate modeling, and/or modern observations.

EXAMPLE APPLICATIONS

We apply HyBIM to two lacustrine paleoclimate records covering differing time-scales, geochemical conditions, ostracod species, and dataset resolutions (figs. 3 and 4). We chose these two data sets because: (1) they had the elemental and isotopic measurements necessary for using this model and; (2) they represent two very different cases that demonstrate the utility and flexibility and limitations of the HyBIM for a variety of biogenic carbonate paleoclimate records. In each case, the original age model is used for all datasets and input parameters are constrained using modern catchment and lake chemistry and species-specific partition coefficient calibrations. Sampling resolution, the least squares cross-validated kernel bandwidths, length of the paleoclimate record, and input parameter sources are listed in table 2.

Lake Miragoane, Haiti (Hodell and others, 1991; Curtis and Hodell, 1993)

The paleoclimate records from Holocene Lake Miragoane in Haiti (Hodell and others, 1991; Curtis and Hodell, 1993; Higuera-Gundy and others, 1999) were one of the first to use detailed, high-resolution stable isotope and trace element measurements on lacustrine ostracods. This hydrologically closed lake provides a complete record of the Holocene at high resolution (see age resolutions for $\delta^{18}\text{O}$, Mg/Ca and Sr/Ca records in table 2). Two cores were recovered and sampled at fine (~ 10 cm) resolution for $\delta^{18}\text{O}$ and coarser (~ 150 cm) resolution for Mg/Ca and Sr/Ca (Curtis and Hodell, 1993). The discrepancy in sample resolution results in the cross-validated kernel bandwidth for $\delta^{18}\text{O}$ (bandwidth = 39 years) to be much shorter than the Mg/Ca and Sr/Ca records (422 and 440 years, respectively) (fig. 3A). Data from both cores were combined and kernel smoothed using the original age model.

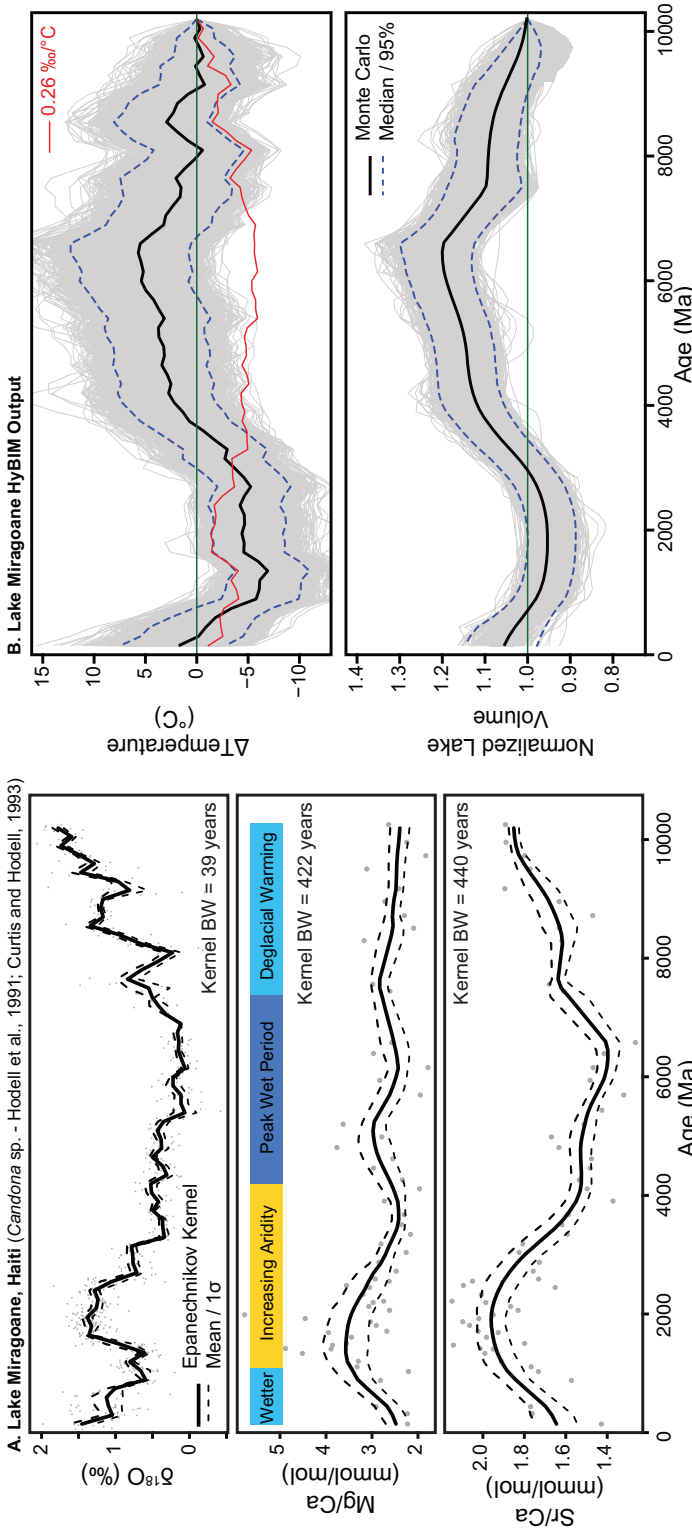


Fig. 3. Illustrative Application of HyBIM to the Lake Miragoane Ostracod (*Candona* sp.) record. (A) Original $\delta^{18}\text{O}$, Mg/Ca and Sr/Ca records (gray dots) from Hodel and others (1991) and Curtis and Hodel (1993). Data are plotted on the original age model. For model input the data are kernel smoothed using an Epanechnikov kernel. The cross validated bandwidth (BW; see text for explanation) is listed for each record ($\delta^{18}\text{O}$, Mg/Ca and Sr/Ca). The solid black line is the mean and the dashed black lines (dashed blue lines in online version) are the 1σ . (B) HyBIM results for the Lake Miragoane records using partition coefficient and vital effect data from Engstrom and Nelson (1991) and Keatings (ms, 1999) derived for *Candona* sp. The thin gray line (thin red line in online version) is the empirical relationship for isotopes in precipitation ($0.26\text{‰}/^\circ\text{C}$) derived from empirical tropical GNIP stations (fig. A2; after Rozanski and others, 1993).

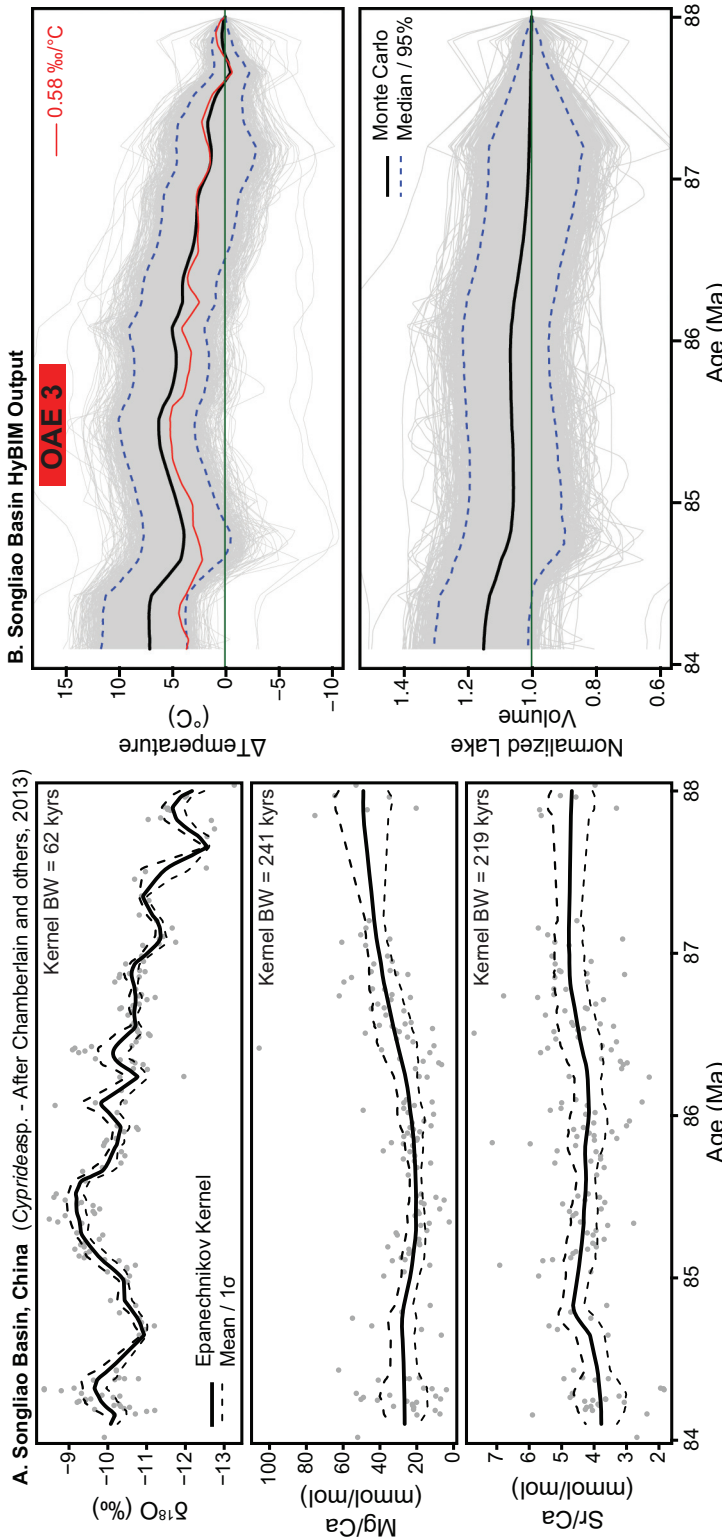


Fig. 4. Illustrative Application of HyBIM to the Songliao Basin Ostracod (*Cypridea* sp.) Record. (A) Original $\delta^{18}\text{O}$, Mg/Ca and Sr/Ca records (gray dots) from Chamberlain and others (2013). Data are plotted on the original age model of Wan and others (2013). For model input the data are kernel smoothed using an Epanechnikov kernel from 84 to 88 Ma (densely sampled portion of the record). The cross validated bandwidth (BW; see text for explanation) is listed for each record ($\delta^{18}\text{O}$, Mg/Ca and Sr/Ca). The solid black line is the mean and the dashed black lines are the 1σ and the 95% confidence interval of the Monte Carlo estimation. The light gray line (red line in online version) is the empirical relationship of coefficient data from DeDecker and others (1999) derived for *Cypridea* sp., the extant, closely related genus of *Cypridea* sp. The solid line is the median and the dashed black lines (dashed blue lines in the online version) are the 95% confidence interval of the Monte Carlo estimation. The approximate position of OAE 3 is denoted by the red bar (in online version), which corresponds to a peak in precipitation (0.58 ‰/°C) derived from mid-latitude Global Network of Isotopes in Precipitation (GNIP) stations. Rozanski and others (1993) for isotopes in precipitation (0.58 ‰/°C) derived from mid-latitude Global Network of Isotopes in Precipitation (GNIP) stations.

The elemental and isotopic data for these studies were from ostracods identified as *Candona* sp., by R. Forester (see discussion in Curtis and Hodell, 1993), which is an ostracod species closely related to *Candona rawsoni*. See table 2 for detailed model input parameters, including the partition coefficients for *Candona rawsoni* as determined by Engstrom and Nelson (1991).

The modeled temperature and lake volume changes for the Lake Miragoane record are largely in accordance with the expected response (see interpretation in Curtis and Hodell, 1993, their fig. 8). These authors suggested based on the isotopic and elemental data that lake levels were highest and climate was “mesic” (wetter and hotter) during the early Holocene, peaking between 7,000 and 4,000 years BP (Hodell and others, 1991; Curtis and Hodell, 1993). In the late Holocene they suggest that between ~4,000 and ~2,500 years BP there was a two-step increase in aridity towards lower lake level and increased salinity, based primarily on the oxygen isotopes (Hodell and others, 1991), followed by wetter conditions beginning at ~1,000 year BP based on additional evidence from the trace element measurements (Curtis and Hodell, 1993).

Our model agrees with this interpretation and shows that modeled temperature and lake volume covary. Lake volume and temperature increase to a peak at ~6300 years BP, decline to a minimum at ~2000 years BP, and increase again nearing present day. The deglacial warming (~10,000 – 7,000 years BP), and increasing aridity (from ~4,000 – 1,000 years BP) are captured by the predicted temperature fluctuations. Driven by the covariation in the input datasets, the modeled temperature is substantially different than that expected from just using the

$\frac{\partial(\delta^{18}\text{O})}{\partial T}$ empirical relationship of 0.26 ‰/°C (thin gray line-thin red line in online version- of fig. 3B; relationship derived in fig. A2). In addition, while the higher resolution $\delta^{18}\text{O}$ record determines the sub-1000-year variations in temperature, the long-term trends largely reflect the trace element records. The sampling resolution discrepancy of the trace elements (low resolution) and $\delta^{18}\text{O}$ records (high resolution) is not necessarily ideal, but our results demonstrate that, by kernel smoothing the records and interpolating to even time-steps, the problem of differing sampling resolution can be resolved.

Songliao Basin, China (Chamberlain and others, 2013)

The Songliao Basin is a Cretaceous terrestrial lake basin that contains up to 10,000 m of fluvial-lacustrine, volcanoclastic and alluvial sediments (Feng and others, 2010; Wang and others, 2013). During the first phase (SK-1) of the International Continental Scientific Drilling Project’s (ICDP) efforts to recover strata covering the entire Cretaceous, a total of 2,486 m of core were recovered that span the late Turonian to the end Cretaceous (Wan and others, 2013; Deng and others, 2013; Wu and others, 2014). Recent paleoclimate reconstruction efforts have used a variety of proxies, including ostracods from lacustrine facies (Chamberlain and others, 2013) and paleosol carbonate nodules (Huang and others, 2013; Gao and others, 2015). We model the Sr/Ca, Mg/Ca and $\delta^{18}\text{O}$ ostracod record originally published by Chamberlain and others (2013) spanning 84 to 88 Ma (fig. 4A). Data is kernel smoothed on the original age model (Wan and others, 2013; Wang and others, 2013; Chamberlain and others, 2013), and this portion of the record was selected because of the high density sampling through this period, relatively stable source area (based on $^{87}\text{Sr}/^{86}\text{Sr}$), and the likely existence of Ocean Anoxic Event 3 (OAE 3 at ~85.5 Ma; Waple, 2012). Because the ostracod genus *Cypridea* sampled by Chamberlain and others (2013) is extinct, we use partition coefficient and vital effect calibrations for the extant genus *Cyprideis*, a closely related genus also of the *Cyprididae* family (table 2). See table 2 for detailed model input parameters.

The relatively invariant (kernel smoothed) Mg/Ca and Sr/Ca records (fig. 4A) result in several interesting observations when HyBIM is applied to the Songliao Basin ostracod record. The Mg/Ca and Sr/Ca records demonstrate much greater spread around the mean kernel-smoothed values and, due to slightly lower sampling density, have a greater cross-validated kernel bandwidth than the $\delta^{18}\text{O}$ record (62 kyrs vs. 241 kyrs and 219 kyrs; fig. 4A). Because of the relatively invariant Mg/Ca and Sr/Ca record, the modeled temperature is similar to the result expected if a $\frac{\partial(\delta^{18}\text{O})}{\partial T}$ empirical relationship of 0.58 ‰/°C is applied to the kernel smoothed mean of the $\delta^{18}\text{O}$ record (red line in online version in fig. 4B), unlike the Lake Miragoane record.

We applied the HyBIM to a portion of the Songliao record given in Chamberlain and others (2013) from 88 to 84 Ma because this time interval captures a portion of the Oceanic Anoxic Event 3 (OAE 3) during a time of global warming. Moreover, we have excellent oxygen and trace element data for this time range in the Songliao Basin. It is recognized that unlike earlier OAEs, OAE 3, may be a series of events distributed over longer time frames and is recorded predominantly in the western hemisphere (Wagreich, 2012). Thus, we are interested in how the temperature record in the Songliao Basin recorded this “event”. Our results show peaks of warming of $\sim 6^\circ\text{C}$ between 86 and 85 Ma and again between 85 and 84 Ma. These two warm periods are consistent with multiple events defining OAE 3, but cannot be correlated with those in marine sections because: 1) the age constraints in the Songliao Basin lack the necessary time resolution; and 2) there are multiple carbon isotope excursions during OAE 3 that vary both temporally and spatially across the globe (Wagreich, 2012). Nevertheless, our results do show multiple warming events during this time interval of OAE 3. In addition, the HyBIM results agree with the interpretation of increasing lake size, based on outcrop extent, during this time interval (Feng and others, 2010), suggesting that over this interval lake volume increased by ~ 10 percent. However, we point out that due to the large spread in the measured Sr/Ca and Mg/Ca, the modeled volume changes are not particularly robust.

LIMITATIONS AND GUIDELINES

The modeling framework presented in this paper represents a new approach to quantitatively constraining the hydrologic balance and temperature fluctuations as recorded by the trace elements and stable isotopes of biogenic lacustrine carbonate. Inherently the modeling of paleoclimate records requires simplifying assumptions. As such, we provide several guidelines for assessing the potential application of HyBIM to a lake system:

- (1) Lake systems with highly variable (inorganic) chemical sedimentation, typically driven by order of magnitude changes in lake level, cannot be modeled using HyBIM. A fundamental assumption of the evaporation partial derivatives is that we assume mass conservation of the trace elements within each time step. In addition, we assume that within each time step lake volume does not change by more than 10 percent (see derivation of partial derivatives). Thus, HyBIM solutions should be limited to “balanced-filled” (Carroll and Bohacs, 1999) lake systems with biogenic carbonate from periods of similar lithologic deposition.
- (2) Many large lakes break up into smaller lakes during desiccation due to complex hypsometric basin geometries and watersheds (for example Quaternary Lake Lahontan; Reheis and others, 2014). While this is not readily assessed from sediment cores alone, we emphasize the need for determination of the lake hypsometry via shoreline and/or outcrop extent (for example, Reheis, 1999; Sack, 2009; Zimmerman and others, 2011; Steinman and

others, 2013; Ibarra and others, 2014; Smith and others, 2014; Doebbert and others, 2014). HyBIM should not be applied to these systems over intervals of breakup, as the river chemistry of the smaller lake basins within a large system may differ due to bedrock heterogeneity and catchment weathering processes.

- (3) Well-constrained partition coefficients are necessary to place the measured Mg/Ca and Sr/Ca (or other trace element measurements) in terms of lake water concentrations. Presently modern studies of terrestrial biogenic carbonates from ostracods (for example, Engstrom and Nelson, 1991; Xia and others, 1997a, 1997b; Wansard and others, 1998; De Deckker and others, 1999; Dettman and others, 2002; Holmes and Chivas, 2002; Dwyer and others, 2002; Ito and Forester, 2009; Gouramanis and others, 2010; Marco-Barba and others, 2012; Börner and others, 2013) represent some of the only calibrated partition coefficients available for use in the HyBIM modeling framework outlined here. Future application of this modeling framework to inorganic carbonate or other types of biogenic carbonate from lake systems will require further measurement of trace elements and calibration of partition coefficients.
- (4) We assume that lake water temperature and the basin average air temperature are directly proportional. HyBIM solves for temperature changes (ΔT), not absolute values of temperature, but selects an absolute temperature for each Monte Carlo iteration used for placing the measured Mg/Ca and $\delta^{18}\text{O}$ into lake water composition (see previous discussion). For the lake systems with mean annual air temperature greater than 0 °C included in a recent global lake temperature compilation the slope between the mean annual water temperature and mean annual surface air temperature is indistinguishable from 1 (slope = 1.02 ± 0.03 (1σ), $r^2 = 0.94$, $n = 81$, mean annual air temperatures from 0 to 27 °C; see dataset from Appendix A of Hren and Sheldon, 2012), suggesting that this assumption of proportionality is valid.

CONCLUSIONS AND FUTURE MODEL REFINEMENT

In this paper, we develop and apply a new modeling framework using variations in $\delta^{18}\text{O}$ and trace elements (Mg and Sr) measured in biogenic lacustrine carbonates for closed basin lacustrine paleoclimate records. By applying this modeling approach to two ostracod records, we demonstrate the utility of HyBIM to quantify paleoenvironmental changes across different timescales. Due to a lack of significant variation in the (kernel smoothed) Mg/Ca and Sr/Ca records from the Songliao Basin, the $\delta^{18}\text{O}$ record primarily controls the predicted temperature fluctuations and the volume changes are minimal. Additionally, the Lake Miragoane results demonstrate how the combination of high-resolution and low-resolution datasets are accounted for by HyBIM via kernel smoothing and interpolation to even time steps. Future application of HyBIM to a high-resolution historical record from a small, constrained terminal lake system will provide the best independent validation of this modeling methodology.

Two additions could provide further extensions to this modeling approach. First, over-determination of the **A** matrix by adding a fourth equation to the system of equations describing additional trace elements (such as U or Li; for example Holmes and others, 1995), δD (for example Tierney and others, 2008; Feakins and others, 2014; Kirby and others, 2014) or $\delta^{13}\text{C}$ (controlled by similar processes described in Li and Ku, 1997; Horton and Oze, 2012; Horton and others, 2015) would provide greater control on the partitioning of salinity changes due to evaporation and dilution by volumetric increases. Second, additional isotope systems, such as Sr or Pb isotopes, could be included as a fourth equation (the **A** matrix would be 4x4) and would extend the function of HyBIM to also de-convolve other possible variables, such multiple

riverine inputs, rearrangement of the drainage basin, *et cetera*. For example, assuming different watershed source regions have different Sr or Pb isotope signatures (and also trace element concentrations and/or meteoric water $\delta^{18}\text{O}$ inputs), a fourth set of partial derivatives can be derived to define the relationship between two distinct end-members. This approach could be applied to the full Songliao Basin record (lower resolution) presented in Chamberlain and others (2013), or to basins with sufficient data and known tectonic changes, such as Cenozoic basins in the western United States (for example, Davis and others, 2009; Smith and others, 2014; Doebbert and others, 2014).

The application of this modeling framework is not limited solely to lake systems. For example, recently published speleothem records have measured $\delta^{18}\text{O}$, Mg/Ca, and Sr/Ca, in addition to other trace elements (such as U/Ca, Ba/Ca) and isotope systems (such as $^{234}\text{U}/^{238}\text{U}$) and $^{87}\text{Sr}/^{86}\text{Sr}$) (for example, Oster and others, 2009; Steponaitis and others, 2015). By modifying the modeling framework presented here the relative influence of temperature and vadose zone residence times (influenced by prior carbonate precipitation) on the stable isotope and trace element measurements could be quantitatively deconvolved. However, modern cave monitoring studies to assess the variability and controls on the stable isotope and trace element variations in cave drip water (for example, Fairchild and others, 2000; Musgrove and Banner, 2004; Wong and others, 2011; Oster and others, 2012a), would be necessary for calibration. Similarly, trace element and stable isotope measurements on pedogenic soil minerals and the associated soil pore water in arid soil systems (for example Amundson and others, 1996; Oster and others, 2012b; Maher and others, 2014) may provide an additional avenue for quantitative paleoclimate reconstruction using a similar modeling framework.

Finally, the Monte Carlo approach presented in this paper only includes uncertainty in the input parameters and input dataset. However, age-models in sedimentary settings, and many terrestrial paleoclimate archives, including lakes, can be highly uncertain due to sedimentation rate changes, hiatuses and a lack of dateable material (for example, Huybers and Wunsch, 2004; Blaauw and Christen, 2011). Further extension of HyBIM to include incorporation of age model uncertainty would provide added utility by including temporal uncertainty quantification of the hydrologic response to climatic events and trends recorded by the lake carbonate geochemistry. Recent work has included this type of Monte Carlo age model uncertainty into paleoclimate reconstructions for individual records and large paleoclimate compilations (for example, Shakun and others, 2012; Anchukaitis and Tierney, 2012; Tierney and others, 2013; Marcott and others, 2013; Steinman and others, 2014). Ultimately paleoclimate proxy modeling efforts, such as those illustrated in this paper, are only valuable if the uncertainty of the timing and magnitude of climatic changes is robustly quantified.

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APPENDIX

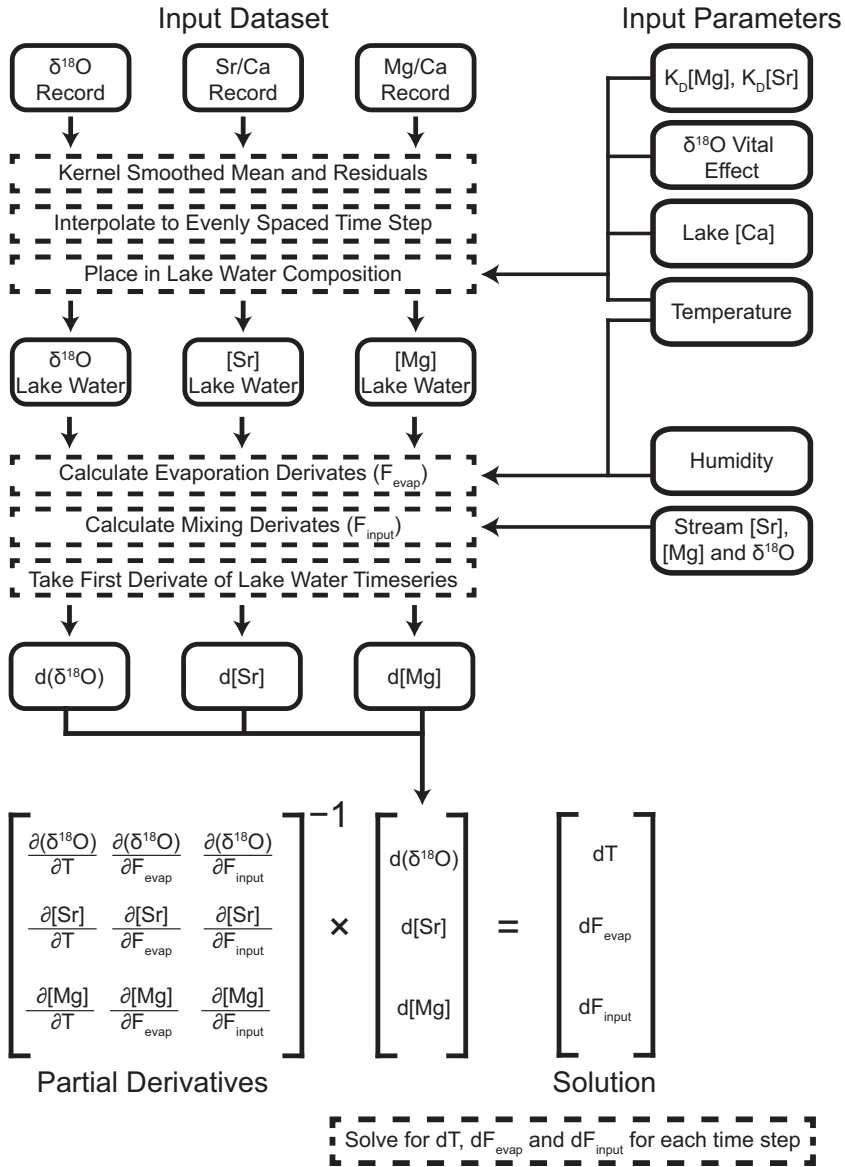


Fig. A1. Schematic of HyBIM model structure. The model structure uses a matrix inversion to solve a system of three equations ($x = A^{-1}b$) implemented at each time step to calculate changes in lake volume (from the combined evaporation and inputs) and basin average air temperature. After smoothing and interpolating to an even time series, the input dataset (time series of δ¹⁸O, Mg/Ca and Sr/Ca from biogenic carbonate), given a set of adjustable input parameters (right side of the fig.; table 1), is first placed in lake water composition (δ¹⁸O, [Mg] and [Sr] Lake Water). The first derivative of the lake water time series is inputted as the b vector at each time step (d(δ¹⁸O), d[Mg] and d[Sr]). The partial derivatives that make up the A matrix are substituted at each time step using mixing equations and evaporation equations (eqs 5 to 9 for F_{evap} and F_{input} derivatives), or estimated for temperature (the T derivatives) based on the geographic location (see text for details). For each Monte Carlo iteration the model produces a time series of the x solution vector. To provide summary statistics after combining the evaporation and lake input variables to calculate changes in volume we calculate the median and 95% range (as in figs. 3 and 4).

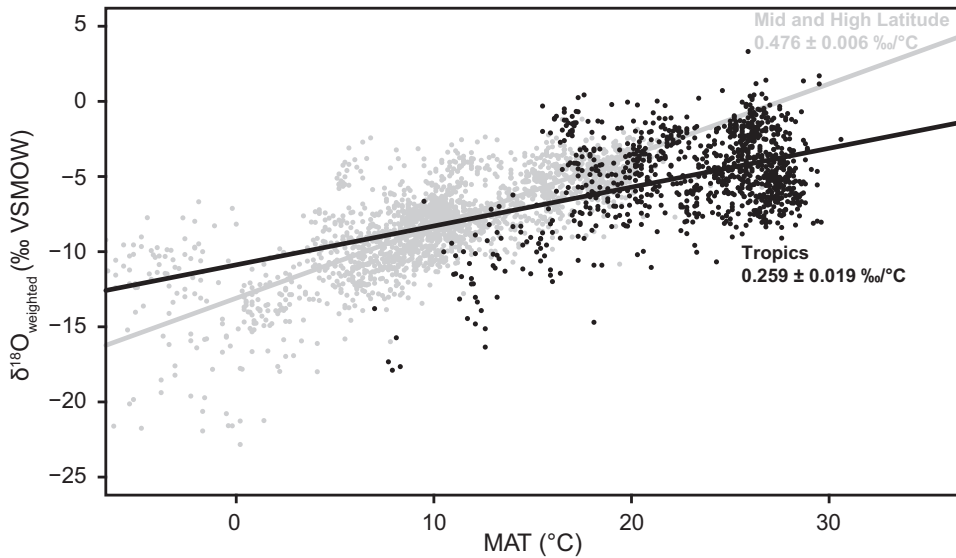


Fig. A2. Annual weighted precipitation $\delta^{18}\text{O}$ and mean annual temperature from the Global Network in Precipitation Data (GNIP) as compiled by the Stable Water Isotope Intercomparison Group (SWING). Following Rozanski and other (1993) we calculate slope of the temperature vs. weighted precipitation $\delta^{18}\text{O}$ for the mid and high latitude ($> 23^\circ$) sites (gray), and tropics (black). We only use the empirical regression (slope and uncertainty) for the tropics in modeling of Lake Miragoane temperature changes. For the Songliao Basin we use the original regression of Rozanski and others (1993) of $0.58\text{‰}/^\circ\text{C}$.

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