Isotopic Characteristics of Salt Lake Carbonates in the Badain Jaran Desert Hinterland and Their Environmental Significance: Postprint

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Abstract

The formation of lake carbonates is closely related to the physicochemical properties of lake water and climatic environment, serving as an important carrier of lake environmental information. Taking two types of carbonates (tufa and lake sediment carbonates) from six lakes in the hinterland of the Badain Jaran Desert as the research object, and based on stable carbon, oxygen, and clumped isotope (Δ47) indicators, this study comparatively analyzed the precipitation processes of the two carbonates and the environmental information they reveal. The research shows: (1) The carbon and oxygen isotopic characteristics of tufa and sediment carbonates from the six lakes in the Badain Jaran Desert are significantly different, with the oxygen isotopes of tufa deposits being significantly more negative than those of lake sediment carbonates. (2) The clumped isotope temperature [T(Δ47)] of lake sediment carbonates is close to the regional summer temperature, whereas the T(Δ47) of tufa carbonates is lower than that of lake sediment carbonates. (3) The $ $18O of carbonate precipitation water estimated based on oxygen isotopes and T(Δ47) shows that the water $ $18O forming tufa carbonates records the signal of mixing between spring water and lake water. The research results preliminarily reveal the environmental indicative significance of salt lake carbonates in the hinterland of the Badain Jaran Desert, providing data support for revealing the hydrological evolution of desert lakes on long timescales.

Full Text

Preamble

The formation of lake carbonates is intimately linked to the physicochemical properties of lake water and the climatic environment, serving as a crucial

archive of lake environmental information. This study investigates two types of carbonates (tufa and lake sediment carbonates) from six lakes in the hinterland of the Badain Jaran Desert, analyzing their precipitation processes and environmental implications based on stable carbon, oxygen, and clumped isotope (Δ47) proxies. The research reveals: (1) The carbon and oxygen isotope characteristics of the two carbonate types differ significantly, with tufa oxygen isotopes being substantially more depleted than those of lake sediment carbonates. (2) The clumped isotope temperatures [T(Δ47)] of lake sediment carbonates are consistent and approximate regional summer temperatures, whereas tufa T(Δ47) values are lower. (3) Based on oxygen isotopes and T(Δ47), the estimated $ $18O composition of the water from which tufas precipitated records a mixed signal of spring water and lake water. These findings provide preliminary insights into the environmental significance of salt lake carbonates in the Badain Jaran Desert hinterland and offer data support for revealing the hydrological evolution of desert lakes over long timescales.

Keywords: lake sediment carbonates; tufa; clumped isotopes; Badain Jaran Desert

The Badain Jaran Desert is China' s second-largest desert, with numerous lakes in its hinterland forming a unique landscape of sand dunes and lake clusters. Understanding the hydrological evolution of this desert is critical for comprehending climate change processes in the transitional zone between monsoonal and westerly wind regimes. The desert hinterland lacks surface runoff, with groundwater accounting for the majority of lake water replenishment, making the sources of groundwater and its recharge mechanisms to lakes long-standing scientific questions. Previous studies on Badain Jaran Desert lakes have shown that abundant carbonates exist in the area, including lake sediment carbonates and tufa carbonates associated with spring outcrops, providing material support for understanding lake evolution and regional environmental change. Research on Spanish tufas has demonstrated that tufa carbon and oxygen isotopes exhibit covariation, which was used to argue for their utility in indicating the oxygen isotope composition of spring water recharging lakes. Rosen et al. systematically compared oxygen isotopes in groundwater, lake water, and tufa from Big Soda Lake, Nevada, further indicating that tufa oxygen isotope values represent a mixture of lake water and groundwater signatures. Subsequently, Hudson et al. combined carbonate carbon, oxygen, and clumped isotope techniques to evaluate spring and lake water mixing information recorded in tufa deposits from Lake Albert in the Chewaucan Basin, North America. Building on this work, our study examines tufa and lake sediment carbonates from Badain Jaran Desert salt lakes, using stable carbon-oxygen and clumped isotope techniques to compare the isotopic characteristics of these two carbonate types and their environmental implications. Our conclusions provide data support for addressing key scientific questions regarding lake environmental evolution and water sources.

1 Study Area Overview

The Badain Jaran Desert is located in the western Alxa Plateau of Inner Mongolia, China, distributed east of Gurnai Lake, south of Guaizi Lake, west of Zongnai Mountain and Yabulai Mountain, and north of Beida Mountain (Fig. 1). Geographically, it lies between 39°04 15 -42°12 23 N and 99°23 18 -104°34 02 E, covering an area of approximately 5.21$×10^{4}$ km^2. The desert is situated in a mid-latitude arid region with a temperate continental climate, receiving about 100 mm of annual precipitation while evaporation exceeds 1500 mm. Multi-year observations indicate a mean annual temperature of 9.96°C, with the coldest month averaging -9.73°C (January) and the warmest month averaging 26.90°C (July). The Badain Jaran Desert consists primarily of mobile dunes with very low surface vegetation coverage, dominated by xerophytic and super-xerophytic shrubs and semi-shrubs.

More than 100 lakes are distributed between the high dunes in the hinterland and southern regions, most with areas smaller than 1 km^2. Due to scarce precipitation and absence of surface runoff, all lakes are groundwater-fed closed systems, predominantly highly mineralized salt lakes with marshy halophytic meadows around their shores. A few lakes contain tufa deposits, with previous surveys indicating that tufa is mainly distributed in the large lake area north of Yikeliaobao. To facilitate comparison with previous monitoring results, this study conducted field investigations and sampling in the large lake area north of Yikeliaobao in July 2023, collecting tufa and lake sediment carbonates from six lakes: Hujilin (HDGJL), Sumujilin (SMJL), Cherigele (CRGL), Bulte (BLT), Bilutu (BLT2), and Hudugejilin (HDGJL2). All six lakes are salt lakes with water chemistry types of Na-Mg-Cl-SO_4 or Mg-Na-Cl-SO_4.

In the field, tufa samples were collected at locations with obvious spring outcrops, while surface sediment samples were collected from nearshore positions without visible springs. Lake sediments are predominantly grayish-brown, consisting mainly of loose aeolian sand. Tufa samples are mostly grayish-white, lack obvious bedding structures, and contain numerous bubble cavities.

2.1 Material Collection and Preprocessing

In the laboratory, collected lake surface sediment samples were washed with ultrapure water to remove soluble salts and freeze-dried using an FD-1A-50 vacuum freeze dryer (Beijing Boyikang). Dried samples were then ground to pass through a 200-mesh sieve for subsequent analysis. Tufa samples were cleaned with ultrapure water to remove loose surface particles, air-dried at room temperature, and then ground to pass through a 200-mesh sieve for subsequent testing.

2.2 Isotope Testing Methods

Clumped isotope measurements were performed at the Stable Isotope Center of Fujian Normal University using a laboratory-built vacuum sample preparation system and a MAT 253 Plus isotope ratio mass spectrometer. The procedure was as follows: Approximately 10 mg of powdered sample (for tufa) or 70 mg (for sediments) was placed in a 15 mL reaction tube and reacted with excess phosphoric acid (1.95 $g \cdot mL^{-1}$) under vacuum at 90°C for 15 minutes. Water produced during the reaction was absorbed using an ethanol-liquid nitrogen trap (approximately -80°C), and the generated CO_2 was collected in a liquid nitrogen cold trap (approximately -196°C). The collected CO_2 was then pushed by high-purity helium through a Porapak Q gas chromatography column to remove trace organic components that could interfere with measurements. Finally, the purified CO_2 was introduced via dual-inlet into the MAT 253 Plus mass spectrometer for clumped isotope analysis, with a signal intensity of 20 V and 8 sample-standard comparison cycles per measurement. To reduce analytical errors and obtain more reliable results, each sample was measured at least 10 times.

During the experiments, ETH-1 ($\Delta47 = 0.293‰$), ETH-2 ($\Delta47 = 0.297‰$), and ETH-3 ($\Delta47 = 0.701‰$) standards were used to establish a laboratory empirical conversion equation, while IAEA-C1 ($\Delta47 = 0.392‰$) and IAEA-C2 ($\Delta47 = 0.732‰$) reference materials monitored data quality. All results were processed using Easotope software and reported relative to the absolute reference frame established by Dennis et al., with Pee Dee Belemnite as the standard. During tufa and lake sediment carbonate analyses, monitoring data for carbonate standards showed long-term analytical precision better than $±$0.010‰ and measurement repeatability better than $±$0.15‰.

3 Results and Analysis

3.1 Carbon and Oxygen Isotope Characteristics of Carbonates

Based on stable isotope results for tufa and lake sediment carbonates from Badain Jaran Desert salt lakes (Fig. 2), lake carbonates show $\delta^{13}C$ values ranging from -0.45‰ to 1.68‰ and $\delta^{18}O$ values from -0.57‰ to 0.47‰. Tufa $\delta^{13}C$ values range from -0.45‰ to 1.68‰, while lake sediment carbonate $\delta^{13}C$ ranges from 0.57‰ to 1.68‰. Tufa $\delta^{18}O$ values are more depleted (ranging from -0.57‰ to -0.45‰) compared to lake sediment carbonates (ranging from 0.47‰ to 1.68‰). The ranges of $\delta^{13}C$ and $\delta^{18}O$ values for tufa are 2.26‰ and 1.13‰, respectively, both larger than those for lake sediment carbonates (1.11‰ and 1.21‰). Overall, tufa $\delta^{13}C$ is more negative than lake sediment carbonates (by approximately 0.47‰), and tufa $\delta^{18}O$ is more depleted (by approximately 1.68‰). These characteristics reveal that the water bodies and precipitation processes forming tufa and lake sediment carbonates are significantly different.

Typically, carbonates in lake sediments precipitate when lake water becomes supersaturated due to biological or physicochemical factors. Regarding oxy-

gen isotopes, carbonate $\delta^{18}O$ is controlled by water temperature and the $\delta^{18}O$ composition of the precipitation fluid. In the Badain Jaran Desert, although sediments and tufa from the same lake deposit within the same system, all six lakes show sediment carbonate $\delta^{18}O$ values more enriched than tufa (by approximately 2.26‰), indicating that the two carbonate types utilize water bodies with different isotopic compositions or precipitate at different temperatures.

Tufa formation is generally associated with spring outcrops, and its isotopic composition is influenced by: (1) degassing, where reduced pressure during groundwater emergence causes dissolved CO_2 to escape, making the water $\delta^{13}C$ significantly more negative than groundwater because $^{12}CO_2$ preferentially escapes during degassing; and (2) mixing, where convergence of different water bodies causes local supersaturation and carbonate precipitation. These processes can lead to covariation of tufa carbon and oxygen isotopes. Our study shows significant positive correlations between carbon and oxygen isotopes in tufa from different lakes, similar to tufa characteristics reported in other regions. These differences reveal that the isotopic signatures of Badain Jaran Desert lake sediment carbonates and tufa carbonates are closely related to their deposition processes.

Comparatively, research on lake tufa carbon and oxygen isotopes is limited. Previous studies of Pyramid Lake tufa and Searles Lake tufa have shown that tufa precipitation results from mixing between relatively fresh, Ca-rich spring water and CO_3^{2-}-rich lake water. The Badain Jaran Desert salt lake water and spring water chemistry follow a similar pattern: lake water is relatively depleted in Ca^{2+} but enriched in CO_3^{2-}, while spring water is relatively enriched in Ca^{2+} but depleted in CO_3^{2-}. When these two water bodies mix, carbonate supersaturation and precipitation occur. Modeling of spring and lake water mixing shows that when lake water proportion exceeds 60%, the carbonate saturation index of the mixed water increases rapidly, promoting carbonate precipitation. Degassing and mixing effects cause differences between tufa and lake sediment carbonates, explaining their isotopic compositional differences.

3.2 Clumped Isotope Characteristics

Extensive observations and modeling studies show that significant disequilibrium of carbonate clumped isotopes in nature mainly includes: (1) rapid carbonate precipitation causing isotopic disequilibrium, and (2) CO_2 degassing causing disequilibrium. For lake sediment carbonates in the Badain Jaran Desert, authigenic carbonate precipitation primarily results from evaporation, with weak rapid degassing effects. First, the desert lake ecosystem' s multi-year cumulative monthly average CO_2 flux is negative year-round, indicating CO_2 uptake from the atmosphere, making rapid CO_2 degassing unlikely in these salt lakes. Second, theoretical calculations show that under these conditions, the time required for CO_2 hydration and isotopic equilibrium is less than one day, while carbonate precipitation in these closed lakes without surface inflow can take years, making CO_2-H_2O isotopic disequilibrium unlikely. Additionally, under high pH condi-

tions, even rapid carbonate precipitation causes minimal kinetic fractionation effects on Δ47 values, with temperature effects not exceeding 10°C.

Since Ghosh et al. established the first Δ47-temperature conversion equation, numerous targeted empirical temperature calibration equations have been developed based on natural samples and laboratory precipitation experiments. Due to significant differences among these equations and lack of consensus, we selected the Kele et al. conversion equation based on the principle of similar materials. This calibration was established using numerous samples and types, with a wide temperature range (6–95°C), and is minimally affected by carbonate $\delta^{18}O$ values, mineral types, and precipitation rates, providing more accurate carbonate precipitation temperatures for this study:

$$\Delta_{47} = (0.044 \pm 0.005) \times 10^6/T^2 + (0.205 \pm 0.047)$$

where T is temperature in Kelvin.

Calculated results show tufa T(Δ47) ranges from 23.8–33.9°C (mean 27.9°C), while lake sediment carbonate T(Δ47) ranges from 17.5–21.2°C (mean 20.1°C). Tufa Δ47 values ($= 0.726‰$) are more positive than lake sediment carbonates ($= 0.717‰$), with a mean difference of 0.009‰, exceeding laboratory analytical error. This difference suggests either different precipitation temperatures or kinetic isotopic fractionation during carbonate precipitation.

If summer is considered the main period for lake sediment carbonate formation, comparison shows that lake sediment carbonate T(Δ47) is essentially consistent with monitored summer lake surface temperatures (25–30°C) in the Badain Jaran Desert hinterland, aligning with conclusions that lacustrine authigenic carbonates primarily record summer temperatures. Tufa formation involves mixing of spring and lake water with different temperatures, making direct T(Δ47) comparison difficult. From a tufa formation perspective, spring water typically has high CO_2 partial pressure, and degassing during emergence can cause kinetic fractionation and Δ47 value decreases. However, theoretical calculations show that when degassing dominates, the relationship between Δ47 and $\delta^{13}C$ should have a slope of approximately -0.0290, while our tufa samples show a slope of only -0.003, indicating that even if degassing affects isotopic composition, the resulting clumped isotope fractionation is insignificant.

From the temperature perspective recorded by tufa, although tufa T(Δ47) is lower than lake sediment carbonates, it is generally higher than annual average spring water temperatures (17–18°C), showing characteristics of spring-lake water mixing. When lake water temperature exceeds spring water temperature, spring emergence and contact with lake water cause mixing temperature elevation, favoring CO_2 escape and carbonate precipitation, making tufa-recorded precipitation temperatures intermediate between spring temperature and summer lake surface temperature. This may relate to the small water volume and intense evaporation in desert lakes causing large $\delta^{18}O$ fluctuations. For example,

Sumu Jilin Lake shows significant differences among reported lake water $\delta^{18}O$ values. In contrast, lake sediment carbonates represent average conditions of lake water over longer periods, so estimated $\delta^{18}O$ values should represent an average level, explaining why water $\delta^{18}O$ values estimated from lake sediment carbonates are relatively stable among different lakes.

3.3 Estimation of Water $\delta^{18}O$ During Carbonate Formation

Under isotopic equilibrium conditions, carbonate $\delta^{18}O$ is a function of temperature:

$$1000 \ln \alpha = 18.03 \times 10^3 / T - 32.42$$

where α is the oxygen isotope fractionation factor between carbonate and its precipitation fluid. Using measured carbonate $\delta^{18}O$ and T(Δ47), we estimated the $\delta^{18}O$ composition of the water from which carbonates precipitated. Comparison with previously reported lake and spring water $\delta^{18}O$ values shows that estimated water $\delta^{18}O$ values fall within the range of observed lake and spring water values, indicating that our estimates represent carbonate precipitation under observed water conditions.

Results show that water $\delta^{18}O$ values estimated from lake sediment carbonates are relatively stable among lakes, partially explaining why these values are consistent. In contrast, tufa samples record water $\delta^{18}O$ values that are generally more depleted than lake sediment carbonates and show large variations, reflecting differences in spring-lake water mixing during tufa formation. For example, Hudugejilin tufa-estimated water $\delta^{18}O$ (-5.5‰) is very close to previously measured spring water values (-5.6‰), indicating great potential for using Badain Jaran tufa to reconstruct spring water $\delta^{18}O$, consistent with conclusions from Andrews et al.

If we consider the estimated water $\delta^{18}O$ values reliable and use lake sediment carbonate T(Δ47) and estimated water $\delta^{18}O$ as references, differences in Δ47 between tufa and sediment samples can indicate the degree of water mixing during tufa formation. Results show that while lake sediment carbonate and tufa T(Δ47) values are relatively close, some tufa values are more depleted than modern lake water $\delta^{18}O$. Tufa-estimated water $\delta^{18}O$ shows a significant positive correlation with Δ(T47) (the difference in T(Δ47) between lake sediment carbonates and tufa), further demonstrating that carbonate isotopic composition can reveal tufa deposition processes and assess spring-lake water mixing degrees.

4 Conclusions

Based on analysis of carbon, oxygen, and clumped isotope characteristics of two types of carbonate deposits from six lakes in the Badain Jaran Desert, this study examined differences in precipitation processes and environmental implications between tufa and lake sediment carbonates. The results show:

1) Differences in carbon and oxygen isotope characteristics between tufa and lake sediment carbonates are closely related to their precipitation processes. Lake sediment carbonate precipitation is primarily influenced by lake water evaporation, while tufa oxygen isotopes are significantly more depleted than lake sediment carbonates. Tufa and lake sediment carbonates have different environmental indications.

2) Lake sediment carbonates precipitate under near-equilibrium conditions, with T(Δ47) primarily recording summer temperatures. Tufa T(Δ47) values are generally higher than spring water temperatures but lower than summer lake temperatures, representing the result of spring-lake water mixing.

3) Based on carbonate isotopes, estimated water $\delta^{18}O$ values during precipitation fall between corresponding lake and spring water values, indicating that tufa precipitation fluids are products of lake-spring water mixing. Carbonate Δ47 can indicate the degree of spring-lake water mixing during tufa formation, providing an important supplement to existing geochemical methods for lake research.

Therefore, we infer that Badain Jaran Desert lake tufa has potential for revealing lake water sources, and future research based on more lakes and expanded water chemistry characterization could further develop applications of carbonate clumped isotopes and oxygen isotopes in desert lakes.

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Note: Figure translations are in progress. See original paper for figures.