



Isotopic tracers of paleohydrologic change in large lakes of the Bolivian Altiplano

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ABSTRACT

We have developed an $^{87}\text{Sr}/^{86}\text{Sr}$, $^{234}\text{U}/^{238}\text{U}$, and $\delta^{18}\text{O}$ data set from carbonates associated with late Quaternary paleolake cycles on the southern Bolivian Altiplano as a tool for tracking and understanding the causes of lake-level fluctuations. Distinctive groupings of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are observed. Ratios are highest for the Ouki lake cycle (120–95 ka) at 0.70932, lowest for Coipasa lake cycle (12.8–11.4 ka) at 0.70853, and intermediate at 0.70881 to 0.70884 for the Salinas (95–80 ka), Inca Huasi (~45 ka), Sajsi (24–20.5 ka), and Taucu (18.1–14.1 ka) lake cycles. These Sr ratios reflect variable contributions from the eastern and western Cordilleras. The Laca hydrologic divide exerts a primary influence on modern and paleolake $^{87}\text{Sr}/^{86}\text{Sr}$ ratios; waters show higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios north of this divide. Most lake cycles were sustained by slightly more rainfall north of this divide but with minimal input from Lake Titicaca. The Coipasa lake cycle appears to have been sustained mainly by rainfall south of this divide. In contrast, the Ouki lake cycle was an expansive lake, deepest in the northern (Poópo) basin, and spilling southward. These results indicate that regional variability in central Andean wet events can be reconstructed using geochemical patterns from this lake system.

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Introduction

Numerous paleolakes once occupied the Poópo, Coipasa, and Uyuni basins on the southern Bolivian Altiplano and provide dramatic evidence of major changes in low-latitude moisture. These were among the largest late Quaternary paleolakes in the Americas (30,000–60,000 km²), and span almost 3° of latitude. The deposits associated with these lakes have been the focus of multiple investigations, including studies of cores (e.g. Sylvestre et al., 1999; Fornari et al., 2001; Baker et al., 2001a; Fritz et al., 2004) and shoreline deposits (e.g. Rondeau, 1990; Sylvestre et al., 1999; Placzek et al., 2006a) from a variety of perspectives, biological, geochemical, and geochronological. Interpretation of these archives has and will continue to make a significant contribution to our understanding of tropical climate change (e.g. Seltzer et al., 2003).

Strontium (Sr), uranium (U), and especially oxygen (O) isotopes are widely used geochemical tools in lake studies for reconstructing paleohydrologic and hence paleoclimate changes. The uses and limitations of oxygen isotopes are well known, since important variables such as temperature, rainfall, and evaporation all influence, often in complex ways, the oxygen isotopic composition of lakes. Strontium and uranium isotopes are less widely employed in paleolake studies and yet can be extremely useful as stratigraphic tools (e.g. Hart et al., 2004; Ku et al., 1998), for constraining lake levels (Benson and

Peterman, 1995), and for reconstructing the history of lake overflow between sub-basins in large complex systems like the one under study here (Hart et al., 2004; Benson and Peterman, 1995).

For this study, our main focus is on the Sr system because individual lake cycles often have unique Sr isotopic ratios. As such, we are able to utilize these values as stratigraphic markers. Furthermore, we measure and then model the modern fluxes of Sr isotopes to contextualize results from ancient carbonates associated with the various paleolakes. Our U and O results add useful constraints on the Sr system. Both the Sr (Coudrain et al., 2002; Grove et al., 2003) and O (Cross et al., 2001) isotopic composition of this lake system have been studied to some degree already. We synthesize these sometimes contradictory results and add to them a large body of new results on both the modern and ancient lake systems. Among other things, this perspective allows us to speculate on possible changes in moisture sources to the lakes.

It is now apparent that two major lake expansions on the southern Bolivian Altiplano during the late Quaternary are part of a regional wet interval (18–8 ka) known as the Central Andean Pluvial Event (CAPE) (Latorre et al., 2006; Quade et al., 2008). The region impacted by CAPE appears to extend from 10 to 26°S (Latorre et al., 2006; Smith et al., 2005). Changes in North Atlantic sea-surface temperature (SST) gradients are implied as the principal driver of the CAPE (Zech et al., 2007; Blard et al., 2009). However, significant modern climate variability is observed across the broad region impacted by CAPE. For example, there are two modes of summer rainfall variability on the southern Bolivian Altiplano, one sourced to the north–northeast and one derived from the southeast (Vuille and Keimig, 2004). Changes in the relative importance of these two modes of modern precipitation variability are

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proposed as the driver of regional asynchrony within the CAPE (Quade et al., 2008). One outcome of our present and ongoing studies is to better differentiate the relative contribution of these two rainfall modes under ancient conditions and ultimately to establish the connections of these rainfall modes to tropical and extratropical climatic changes.

Study area

The Altiplano is a high-elevation (~3800 m), internally drained plateau. Four large lake basins (from North to South, respectively, Titicaca, Poópo, Coipasa, and Uyuni) occupy the region between the Western Cordillera (WC) and the Eastern Cordillera (EC), and are part of a watershed spanning 14° to 22°S (Fig. 1). In this paper we consider the “southern Altiplano” to be the area encompassing all watersheds south of Lake Titicaca. The “northern Altiplano” is covered by Lake Titicaca (3806 m; 8560 km²), a deep (>285 m), fresh-water lake with minor (<10%) outflow to the south along the Río Desaguadero into the oligosaline Lake Poópo (3685 m; 2500 km²) (Argollo and Mourguiart,

2000). By the time the Río Desaguadero reaches Lake Poópo it has more than quadrupled in size, and its water chemistry is notably different from that of Lake Titicaca (Grove et al., 2003). Lake Poópo is separated by the Laca hydrologic divide (3700 m) from the Salars (salt pans) of Coipasa (3656 m, 2530 km²) and Uyuni (3653 m, 12,000 km²) (Fig. 1). During exceptionally wet years (e.g. 1986) water flows over this hydrologic divide and into the Coipasa basin (Zolá and Bengtsson, 2007). The Río Laca Jahuirá also originates here and flows west into the Coipasa basin (Fig. 1). The Río Lauca is the primary source of water for a small ephemeral lake in the Coipasa basin, and has a minimum estimated discharge of $0.2 \times 10^9 \text{ m}^3 \text{ yr}^{-1}$ (Grove et al., 2003). The broad divide between the two salars in the Coipasa and Uyuni basins is only 1 m high and during flood events these salars form a single connected water body. For this reason, we consider the Coipasa and Uyuni basins to be a single basin during lake expansion, the Coipasa/Uyuni basin. The Río Grande is the only major watercourse entering the Uyuni basin and has an estimated annual discharge of $0.4 \times 10^9 \text{ m}^3 \text{ yr}^{-1}$ (Montes de Oca, 1997).

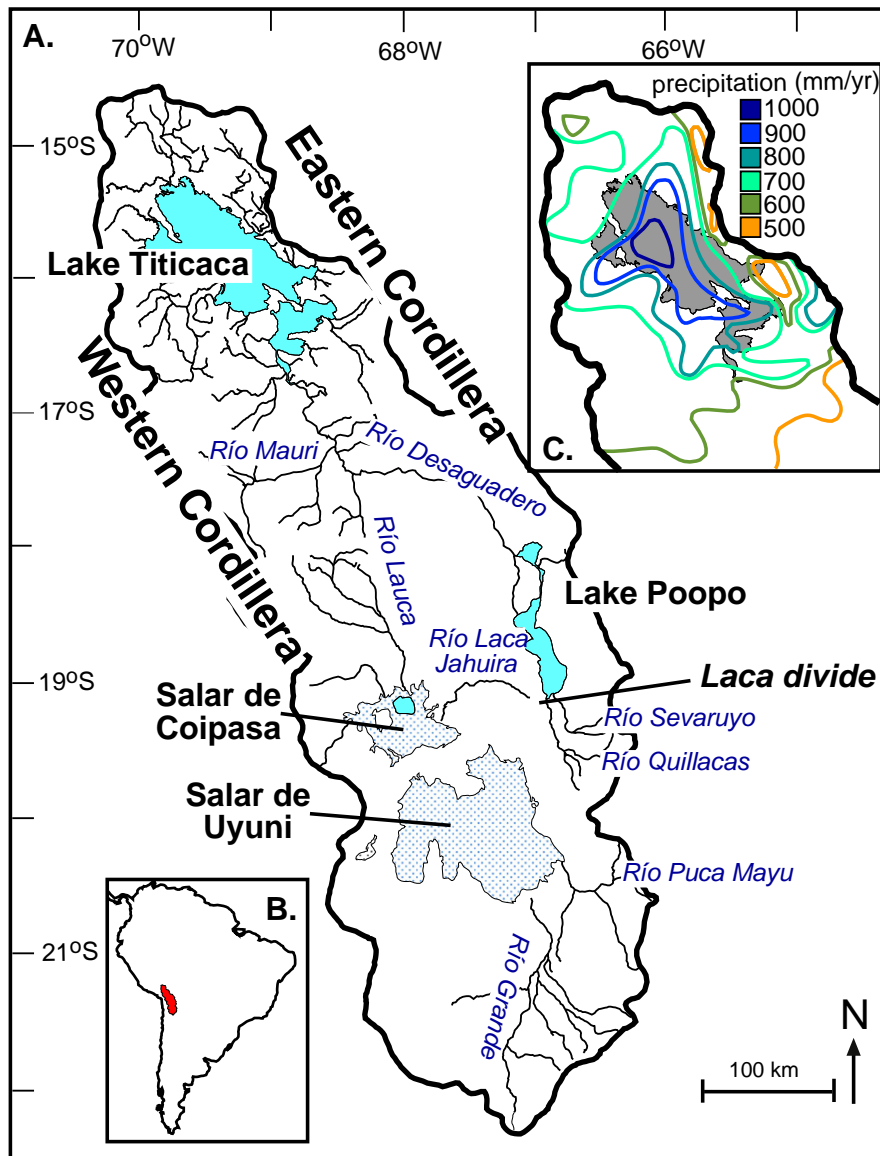


Figure 1. A) Map of the Altiplano study area. Modern lakes are in gray and salars are dotted. Bold line encompasses the hydrographic basin. B) Map of south America with the Altiplano drainage basin indicated in black. C) Precipitation in the Titicaca drainage basin (after Roche et al., 1992). Lake Titicaca is shown in gray. Contour lines of equal precipitation are shown in shades of black (1000 mm/yr) to light gray (500 mm/yr).

Modern climate

The Bolivian Altiplano is located at the western end of the Amazon rainfall belt, and greater than 80% of rainfall occurs in the austral summer (e.g. Vuille, 1999) in what is described by many as the South American Summer Monsoon (SASM) (e.g. Zhou and Lau, 1998). A pronounced north–south precipitation gradient spans the Titicaca–Poópo–Coipasa–Uyuni catchment, with Lake Titicaca receiving ~800 mm of precipitation per year (mm/yr) and the dry southern basins receiving less than 200 mm/yr (Fig. 2A). Two distinct modes of variability in modern summer rainfall occur over the central Andes, and these modes overlap each other on the southern Bolivian Altiplano (Vuille and Keimig, 2004). Rain falling on the northern Altiplano originates from the east and falls predominantly in the summer months when the Intertropical Convergence Zone is displaced southward and convection is most intense in the Amazon Basin (Lenters and Cook, 1997). This rainfall regime is modulated by several factors, including North Atlantic sea-surface temperature (Enfield and Mayer, 1997; Vuille et al., 2000), and El Niño/Southern Oscillation (ENSO), with La Niña years tending to correlate with stronger easterly winds and more rainfall (Aceituno, 1988; Vuille et al., 1998, Vuille, 1999; Garreaud and Aceituno, 2001; Vuille and Keimig, 2004). In contrast, summer rainfall on the southern Altiplano has interannual variability that is most closely tied to precipitation anomalies and humidity levels over the Chaco region of Argentina (Vuille and Keimig, 2004). Interplay between these two modes of modern climate variability may explain the spatial and temporal variability in a regional late Pleistocene wet event, the Central Andean Pluvial Event (CAPE) (Quade et al., 2008).

Lake nomenclature and chronology

Shoreline sediments and >200 radiometric ages place constraints on two high elevation, one mid-elevation, and three shallow lake

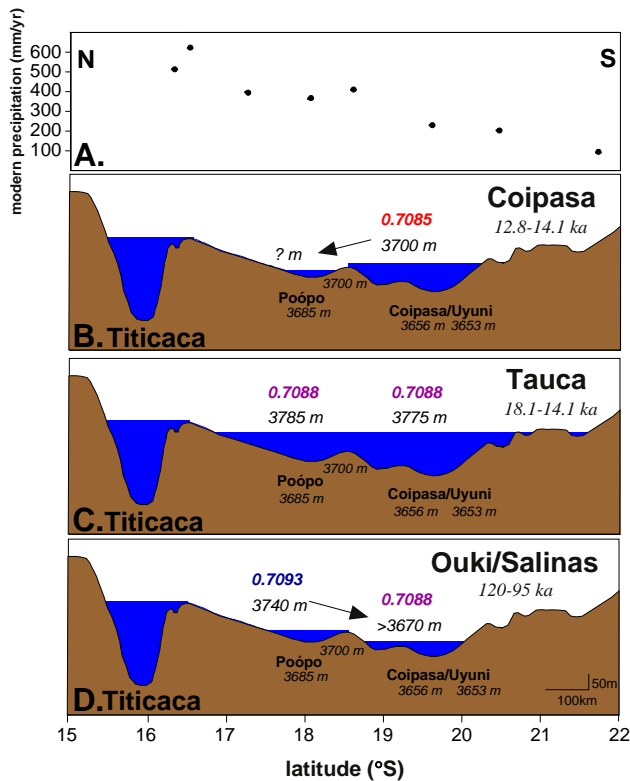


Figure 2. A) The modern precipitation gradient in the Titicaca, Poópo, and Coipasa/Uyuni basins. Schematic cross section of the Titicaca/Poópo/Coipasa/Uyuni hydrographic basins and proposed flow between basins during the Coipasa (B), Tauca (C), and Ouki (D) lake cycles. Ratios of $^{87}\text{Sr}/^{86}\text{Sr}$ and lake elevations for each lake cycle are shown.

cycles over the last 130 ka in the Poópo, Coipasa, and Uyuni basins (Servant and Fontes, 1978; Rondeau, 1990; Bills et al., 1994; Sylvestre et al., 1999; Placzek et al., 2006a,b). These pluvial episodes have been of interest for over a century (e.g. Minchin, 1882), resulting in several, sometimes contradictory, names and chronologies for various lake cycles. In this paper we continue to use the nomenclature for these lake phases employed in our previous research (Fig. 3A; Placzek et al., 2006a,b). The Ouki lake cycle (120–95 ka) is dated at numerous localities in the Poópo basin and reached its maximum elevation (3740 m) between 110 and 100 ka. A much shallower lake, the Salinas lake cycle (<3670 m), was present in the Uyuni Basin between ~95 and 80 ka. No single outcrop contains evidence of a continuous lake over the entire Ouki and Salinas lake intervals (120–95 ka), but Placzek et al. (2006a) differentiate between lacustrine deposits with ages between 95–80 ka and 120–95 ka because they have different elevations in different basins. Examination of individual outcrops suggest that individual sections of the Ouki lake cycle may indeed record different phases of this lake; eventual correlation of shoreline and sediment core stratigraphies could lead to subdivision of the Ouki lake cycle into different sub-phases. Shallow lakes (<3670 m) were also present in the Uyuni basin around 46 ka (Inca Huasi lake cycle), and between 24 and 20.5 ka (the Sajsi lake cycle). The Tauca lake cycle (18.1–14.1 ka) was the deepest lake on the Bolivian Altiplano in the last 120 ka and reached its maximum (~3785 m) between 16.4 and 14.1 ka. The Coipasa lake cycle occurred between 12.8 and 11.4 ka (Placzek et al., 2006a).

Methods

Over 30 exposures of lacustrine sediments were described and sampled as part of a comprehensive effort to obtain and replicate records of lake change from multiple localities in all three major basins (Placzek et al., 2006a); sample sites from this study directly correspond to sites and samples described in these previous efforts. Particular effort was directed towards the various visible paleoshorelines, as this approach to reconstructing lake-level history allows for direct determination of lake level, replication of stratigraphy, and dating by two geochronologic (^{14}C , U–Th) methods. A firm U–Th chronology for the Tauca lake cycle (18.1–14.1 ka) indicates minimal hard-water effects on ^{14}C dates. For older samples (>25 ka), U–Th ages are significantly (>50 ka) older than associated ^{14}C dates, as a result of contamination by as much as 3% modern carbon. The cause for this is likely related to the surface exchange of atmospheric CO_2 with carbonate radicals, a process observed when carbonates gradually take up CO_2 after grinding (Samos, 1949). This contamination of the ^{14}C system is, therefore, not indicative of significant alteration of other isotopic systems; carbonate powders selected for this study are from tufas or shells that were free from secondary cement and low in non-carbonate detritus. Tufas from the southern Altiplano are ubiquitous features in the paleolake basins and have readily identified biologic textures (Placzek et al., 2006b) indicating formation within the photic zone. Sample locations in proximity to springs and/or the rare tufa with a form indicative of formation under the influence of groundwater input (e.g. chimney structures) were avoided in this sampling effort.

Waters and carbonate powders were sampled for Sr and O analyses. We collected water samples during June 2001 and again in November–December 2001; 50 ml of water was passed through disposable 0.2 μm glass syringe filters and stored in sealed acid-washed polyethylene bottles. Samples of water and salt were also collected from the Salars of Coipasa and Uyuni in June 2001. Carbonate samples were soaked in 2% NaOCl for about one hour to remove any organic material and dissolved in doubly distilled 0.1 M acetic acid (Asahara et al., 1995) by placement in an ultrasonic bath for 0.5 h. Samples were then allowed to stand for ~12 h before canting the acid. An ^{84}Sr spike was added to all water and some carbonate samples for the purpose of obtaining

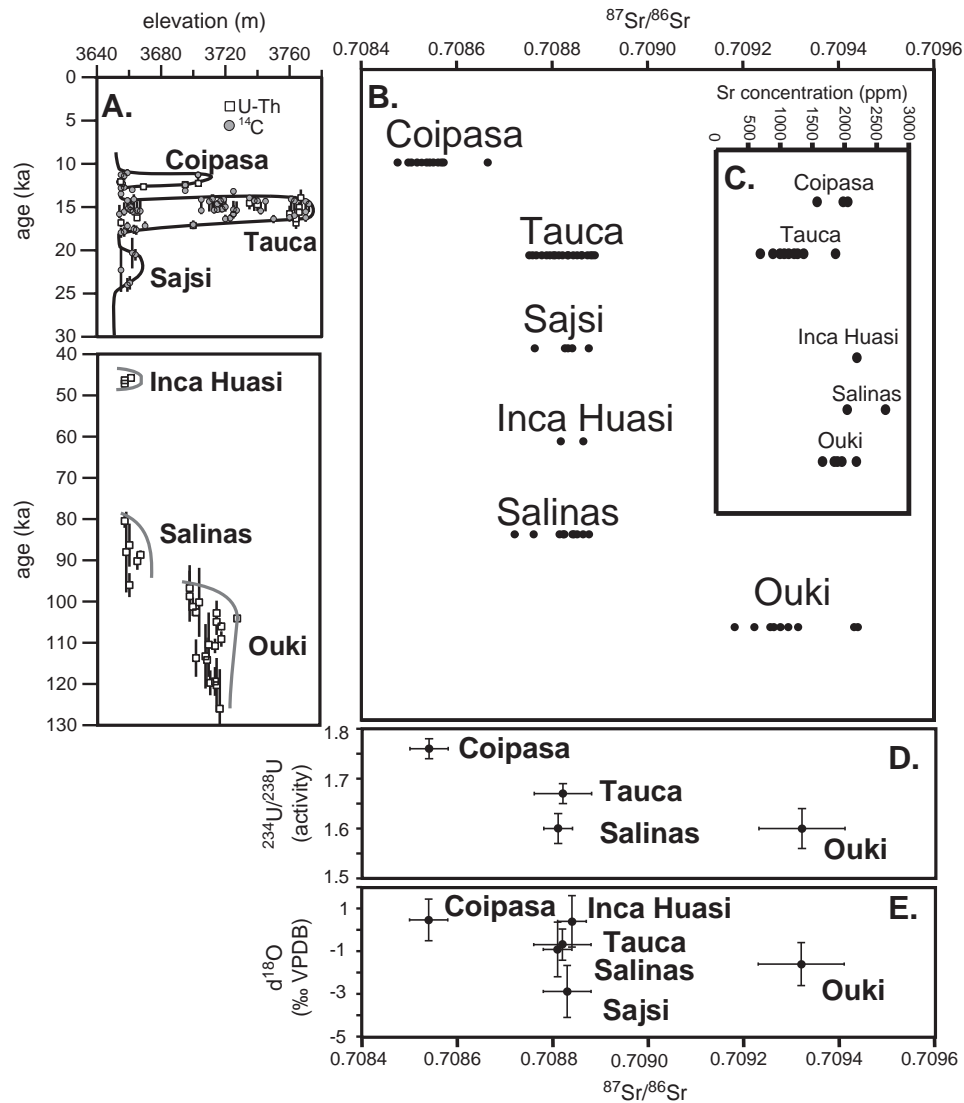


Figure 3. A) Reconstructed lake-level history from shoreline sediments (Placzek et al., 2006a). Radiocarbon (circles) and U–Th (squares) ages are shown, error bars represent 2σ analytical uncertainty and also reflect uncertainty due to initial thorium or radiocarbon calibration. The Ouki lake cycle may represent one lake event or several oscillations. B) The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and concentrations (C) of carbonates from various lake cycles. D) Average $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{234}\text{U}/^{238}\text{U}$ (activity) ratios for paleolake carbonates. E) Average $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and $\delta^{18}\text{O}$ values for paleolake carbonates.

strontium concentration data, and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of spiked samples are corrected for spike composition. Strontium was separated with Eichrom strontium-specific resin, and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were measured on a Micromass Sector 54 thermal-ionization mass spectrometer. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio was normalized to 0.1194 and analyses of the NBS-987 standard run on each 20-sample turret yielded a mean ratio of 0.71026 ± 0.00002 ($n = 12$).

Water isotopic results are reported using standard δ -per mil notation and were processed at the Laboratory of Isotope Geochemistry, University of Arizona. Water samples were prepared using the CO_2 equilibration method on a Kiel III attached to a Finnigan Delta Plus mass spectrometer. The values were corrected based on internal lab standards, which are calibrated through VSMOW and SLAP. The analytical precision is 0.08‰ and 0.1‰, respectively (1σ). Carbonate samples were treated with 2% H_2O_2 for 2 h to remove organic matter and converted to gas using 100% phosphoric acid on a Keil III attached to a Finnigan Delta S mass spectrometer. These values were corrected based on internal lab standards, which are calibrated through VPDB. The analytical precision is 0.05‰ (1σ).

The initial $^{234}\text{U}/^{238}\text{U}$ ratio (expressed throughout this paper as activity ratios) of paleolake carbonates is calculated from the data set

of Placzek et al. (2006a). These initial $^{234}\text{U}/^{238}\text{U}$ ratios are from U– ^{230}Th dates with small corrections for initial ^{230}Th ; reported uncertainties include propagated 2σ envelopes on isotope ratios, decay constants, as well as error on the assumed initial $^{230}\text{Th}/^{232}\text{Th}$ ratio. Carbonates were dissolved in 2 M HNO_3 and spiked with mixed ^{233}U – ^{229}Th . Uranium and Th were co-precipitated with FeOH_3 , separated by anion exchange, and measured on a Micromass Sector 54 thermal ionization mass spectrometer in the Department of Geosciences at the University of Arizona. Details of our analytical procedures are fully discussed in Placzek et al. (2006b). Decay constants are after Cheng et al. (2000).

Results

Modern waters

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of modern waters on the Bolivian Altiplano range from 0.70548 to 0.72553 and strontium concentrations range between 0.03 and 13.8 ppm (Table 1; Fig. 4). Catchments that drain the WC generally have ratios less than 0.708. In contrast, only catchments that drain the EC have $^{87}\text{Sr}/^{86}\text{Sr}$ ratios above 0.710 (Table 1; Fig. 4).

Table 1
Isotopic data from Altiplano waters.

| Catchment #, basin | River | $^{87}\text{Sr}/^{86}\text{Sr}$ (± 1 SE) | Sr ppm (± 1 SE) | Area (km ²) | Rain shadow | $\delta^{18}\text{O}^a$ | $\delta^2\text{H}^a$ | Geology |
|----------------------------|--------------------------|---|--------------------------|-------------------------|-------------|-------------------------|---------------------------|---|
| 1 ^b , Titicaca | Río Ramis | 0.70870 \pm 1 | 0.675 | 4210 | No | 0.0 | 0.0 | Pliocene through Quaternary volcanics with minor Jurassic through Tertiary sediments |
| 2 ^b , Titicaca | Río Huancané | 0.71028 \pm 1 | 0.709 | 4240 | Yes | | | Cretaceous through Devonian sediments |
| 3 ^b , Titicaca | Río Suches | 0.71200 \pm 05 | 0.135 | 3370 | Yes | | | Cretaceous through Devonian sediments |
| 4 ^b , Titicaca | Río Huaycho ^c | 0.70973 \pm 1 | 0.301 | 1020 | Yes | | | Cretaceous through Devonian sediments |
| 5 ^b , Titicaca | Río Coata | 0.70718 \pm 1 | 1.051 | 1550 | No | | | Pliocene through Quaternary volcanics with minor Jurassic through Tertiary sediments |
| 6 ^b , Titicaca | Río lipa ^c | 0.70746 \pm 1 | 0.777 | 1280 | No | | | Pliocene through Quaternary volcanics with minor Jurassic through Tertiary sediments |
| 7 ^b , Titicaca | Río Ilave | 0.70721 \pm 1 | 0.301 | 14,410 | No | | | Pliocene through Quaternary volcanics with minor Jurassic through Tertiary sediments |
| 8 ^b , Titicaca | Río Keka | 0.72196 \pm 1 | 0.029 | 1680 | Yes | | | Devonian sediments and Neogene plutons and sediments |
| 9, Titicaca | Río Catari | Río Keka values | Río Keka values | 4700 | Yes | | | Devonian plutons and Neogene sediments |
| 10, Titicaca | Río Tiahuanacu | Río lipa values | Río lipa values | 840 | No | | | Neogene plutons and sediments |
| 11, Poópo | Río Jacha Jahuira | 0.70948 \pm 1 | 0.946 | 6370 | No | -4.0 | -65.5 | Paleogene to Neogene sediments with minor Proterozoic crystalline rock |
| 12 ^b , Titicaca | Río Tujso Johuiro | Río Jacha Jahuira values | Río Jacha Jahuira values | 3920 | No | | | Paleogene to Neogene sediments and volcanics |
| 13 ^b , Poópo | Río Mauri | 0.70735 \pm 1 | 0.199 | 12,110 | No | | | Neogene volcanics and plutons |
| 14 ^b , Poópo | Well water | 0.70968 \pm 1 | 0.199 | 16,320 | No | | | Paleogene to Neogene sediments |
| 15, Poópo | n/a | Catchment 14 values | Catchment 14 values | 10,400 | Yes | | | Silurian sediments and Neogene tuffs |
| 16 ^b , Poópo | Río Caranguila | 0.70976 \pm 1 | 0.769 | 7400 | No | | | Silurian sediments and Neogene tuffs |
| 17, Coipasa | Río Lauca | 0.70715 \pm 1 | 0.684 \pm 0.005 | 10,440 | No | -8.8 | -91.1 | Neogene through Quaternary volcanics and tuffs |
| 18 ^b , Poópo | Río Poópo | 0.72412 \pm 1 | 1.113 | 8500 | Yes | | | Neogene through Quaternary volcanics and tuffs |
| 19, Coipasa | Río Barras | 0.72412 \pm 2 | 0.380 \pm 0.003 | 5530 | No | -9.4 | -94.8 | Silurian sediments and Neogene tuffs |
| 20, Coipasa | Seep | 0.70679 \pm 2 | 1.193 \pm 0.008 | 8880 | No | -8.5 | -92.0 | Paleogene to Neogene sediments and Cretaceous sediments |
| 21, Coipasa/Uyuni | Unnamed river | 0.70963 \pm 1 | 13.79 \pm 0.18 | 7470 | Yes | -5.4 | -78.6 | Neogene through Quaternary volcanics and tuffs |
| 22, Poópo | Río Sevaruyo | 0.71637 \pm 2 | 0.097 | 3170 | No | | | Neogene tuffs and Cretaceous volcanics |
| 23, Poópo | Río Quillacas | 0.71591 \pm 1 | 0.321 \pm 0.002 | 1410 | Yes | 3.0 | -48.8 | Neogene tuff |
| 24, Uyuni | Río Sajsi | 0.70946 \pm 1 | 2.40 \pm 0.06 | 3820 | No | | | Neogene tuff and Cretaceous sediments |
| 25, Uyuni | Río Kolcha "K" | 0.70678 \pm 7 | 1.23 \pm 0.02 | 6790 | No | -7.6 | -86.1 | Cretaceous sediments; Paleogene through Neogene sediments, plutonics and volcanics |
| 26, Uyuni | Río Pucá Mayu | 0.71176 \pm 2 | 13.8 \pm 0.6 | 10,490 | Yes | 0.2 | -18.0 | Silurian sediments; Paleogene to Neogene sediments and tuffs |
| 27, Uyuni | Río Grande | 0.70952 \pm 1 | 2.76 \pm 0.02 | 30,584 | No | -4.1; -1.4 ^d | -63.5; -53.1 ^d | Neogene tuffs and volcanics; Paleogene to Neogene sediments, with minor Ordovician and Silurian sediments |

| Catchment #, basin | Sample type | $^{87}\text{Sr}/^{86}\text{Sr}$ (± 1 SE) | Sr ppm (± 1 SE) | Latitude; longitude | $\delta^{18}\text{O}$ | $\delta^2\text{H}$ |
|-----------------------|----------------------|---|----------------------|---------------------|-----------------------|--------------------|
| Uyuni | Uyuni water | 0.70894 \pm 1 | 30.7 \pm 0.5 | 20°07'S; 68°14'W | -3.3 | -36.0 |
| Uyuni | Uyuni salt | 0.70885 \pm 0.01 | 2637 \pm 0.93 | 20°18'S; 67°22'W | | |
| Uyuni ^b | Uyuni water | 0.70878 \pm 1 | 9.90 | 20°34'S; 67°33'W | 4.1 | -4.0 |
| Uyuni | Spring in Uyuni | 0.70874 \pm 1 | 4.5 \pm 0.1 | 20°18'S; 67°22'W | | |
| Coipasa | Coipasa water | 0.70915 \pm 1 | 49.8 \pm 1.5 | 19°32'S; 67°55'W | 2.0 | -30.0 |
| Coipasa | Coipasa salt | 0.70827 \pm 1 | 67.8 \pm 0.7 | 19°32'S; 67°55'W | | |
| Poópo | Poópo water | 0.70938 \pm 1 | 9.45 \pm 0.05 | 18°30'S; 67°15'W | 22.5 | 36.4 |
| Poópo ^b | Poópo water | 0.71404 \pm 1 | 1.32 | 18°23'S; 66°57'W | | |
| Titicaca ^b | Modern Lago Grande | 0.70822 \pm 0.6 | 1.15 | 18°30'S; 67°15'W | | |
| Titicaca ^b | Pleistocene overflow | 0.70830 | | | | |
| Poópo | Río Desaguadero 1 | 0.70885 \pm 1 | 1.90 \pm 0.01 | 18°13'S; 67°09'W | 6.4 | -30.5 |
| Poópo | Río Desaguadero 2 | 0.70874 \pm 1 | | 18°05'S; 67°09'W | -0.2 | -49.1 |
| Poópo | Río Desaguadero 3 | 0.70916 \pm 1 | 1.40 \pm 0.02 | 18°01'S; 67°09'W | 3.0 | -48.2 |
| 24 | Río Sajsi well | 0.70792 \pm 1 | 10.5 \pm 0.1 | 19°50'S; 67°27'W | -11.5 | -102.7 |
| 25 | Empexa spring | 0.70548 \pm 1 | 1.96 \pm 0.02 | 20°32'S; 68°26'W | -10.2 | -90.2 |
| 21 | Spring | 0.70620 \pm 1 | 2.58 \pm 0.02 | 19°19'S; 68°22'W | -8.9 | -96.4 |
| 18 | Pazña hot springs | 0.72553 \pm 3 | 2.87 \pm 0.5 | 18°35'S; 66°56'W | -8.7 | -89.6 |

^a Typical standard error is 0.1‰.

^b Data from Grove et al. (2003).

^c Wet season data, all other data is from the dry season.

^d Two stable isotope samples collected (different seasons).

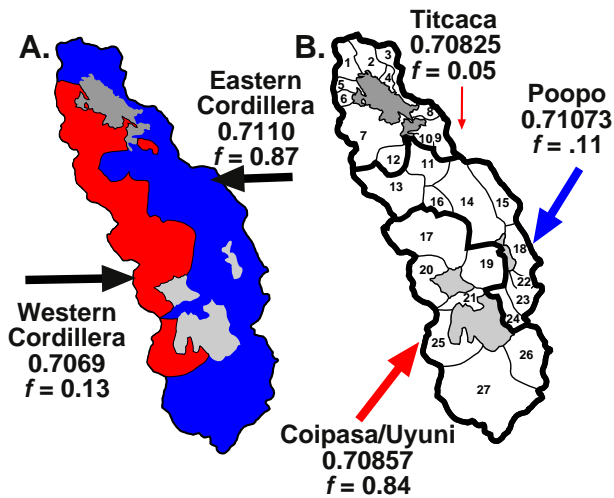


Figure 4. A) The distribution of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of modern catchments on the Bolivian Altiplano based on river and creek sampling. Gray denotes modern lakes and salars. Black represents catchments with ratios <0.709 and white represents ratios >0.709 . Ratios of $^{87}\text{Sr}/^{86}\text{Sr}$ and fractional Sr contributions (f) for the EC and WC are given (see text for computation of f). B) Catchments and sub-basins of the Bolivian Altiplano; individual catchment numbers correspond to Table 1. Ratios of $^{87}\text{Sr}/^{86}\text{Sr}$ and fractional Sr contributions are given for Lake Titicaca, the Poópo basin, and the Coipasa/Uyuni basin.

Some groundwater $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were measured, and ratios for groundwater are similar to those of local surface waters (Table 1).

The $\delta^{18}\text{O}$ values of modern water on the Altiplano vary between -11.5 and -22.0‰ , with $\delta^2\text{H}$ values between -102.7 and 36.4‰ . The $\delta^{18}\text{O}$ value of modern lake Titicaca is -3.8 to -4.6‰ (Cross et al., 2001). Measured values for modern waters from the Poópo, Coipasa, and Uyuni basins are between -11.5 and -22.0‰ (Table 1; Fig. 5). Modern rainfall values average about -17.5‰ (Cross et al., 2001).

Paleolake carbonates

Paleolake carbonates from the Bolivian Altiplano have variable $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{234}\text{U}/^{238}\text{U}$ ratios, but $\delta^{18}\text{O}$ values for all lake cycles are similar (Fig. 3; Table 3). The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios range between 0.70848 and 0.70944 and strontium concentrations are between 690 and 2640 ppm (Fig. 3; Table 2). Shoreline carbonates from the southern Altiplano also have variable initial $^{234}\text{U}/^{238}\text{U}$ ratios between 1.6 and 1.76 (Fig. 3D). The average $\delta^{18}\text{O}$ values for all lake cycles are similar, ranging between -2.9 and 0.5‰ ; the scatter within individual lake cycles produces overlapping $\delta^{18}\text{O}$ values for all lake cycles (Fig. 3E; Supplementary data).

Carbonates from the Coipasa lake cycle have the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.70854 ± 0.00004) and the highest initial $^{234}\text{U}/^{238}\text{U}$ ($1.79 \pm$

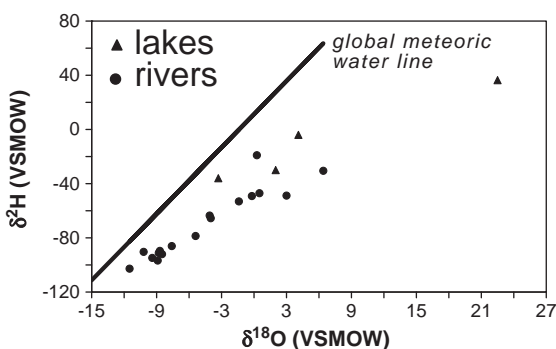


Figure 5. Values of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ from modern rivers (circles) and lakes (triangles) from the southern Altiplano (Table 2). The global meteoric water line (GMWL) is shown; samples show variable degrees of evaporative enrichment.

0.06) (Table 2); the average $\delta^{18}\text{O}$ for the Coipasa lake cycle is 0.4 ± 1.0 . The population of carbonates with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios below 0.7087 includes six undated samples with field associations indicative of formation during the Coipasa lake cycle. One carbonate, whose age and depositional context indicate that it formed during the regression of the Coipasa lake cycle, has a somewhat higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio (0.70867) and was collected along a perennial wash where mixing with local river water may have occurred at low lake levels.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of carbonates of the Tauca, Salinas, Inca Huasi, and Sajsi lake cycles are between 0.70872 and 0.70895, and $^{234}\text{U}/^{238}\text{U}$ ratios range between 1.61 and 1.69 (Table 2); the average $\delta^{18}\text{O}$ for the Tauca, Sajsi, Inca Huasi, and Salinas lake cycles are: $-0.95\text{‰} \pm 1.3$; $-2.9\text{‰} \pm 1.2$; $+0.4\text{‰} \pm 1.2$; $-0.7\text{‰} \pm 0.7$, respectively. Ratios of $^{87}\text{Sr}/^{86}\text{Sr}$ are ~ 0.7088 for all samples from the Tauca highstand at diverse locations around the basins (Table 2). Carbonates that mark the transgression and regression of the Tauca lake cycle to elevations lower than the Laca hydrologic divide (3700 m) also have $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of ~ 0.7088 (Table 2). Carbonates from the Salinas lake cycle (95 to 80 ka) have $^{87}\text{Sr}/^{86}\text{Sr}$ ratios between 0.70872 and 0.70885. Carbonates attributed to the Inca Huasi (~ 46 ka) and Sajsi (24–20.5 ka) lake cycles yielded $^{87}\text{Sr}/^{86}\text{Sr}$ ratios between 0.70876 and 0.70888, but are measured at only one (Sajsi) or two (Inca Huasi) locations.

Carbonates from the Ouki deep lake cycle (120–95 ka) have $^{87}\text{Sr}/^{86}\text{Sr}$ ratios ranging from 0.70918 to 0.70944 and $^{234}\text{U}/^{238}\text{U}$ ratios of 1.60 ± 0.04 . The average $\delta^{18}\text{O}$ value for the Ouki lake cycle is $-1.6\text{‰} \pm 1.0$. These are the highest $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for carbonates of any lake cycle. Strontium concentrations for this lake cycle are also relatively high, between 1657 and 1958 ppm (Fig. 3C).

Modeling the modern lake system

$$^{87}\text{Sr}/^{86}\text{Sr}$$

We constructed a mass balance model of Sr inputs to the modern lake that assists in understanding the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the paleolakes. Much of this model has been developed in previous studies (Grove et al., 2003; Coudrain et al., 2002). In this study we add a substantial body of new evidence from the southern basins. Modeling of the lake $^{87}\text{Sr}/^{86}\text{Sr}_{\text{lake}}$ ratio was estimated by:

$$^{87}\text{Sr}/^{86}\text{Sr}_{\text{lake}} = f_{\text{Sr River1}}(^{87}\text{Sr}/^{86}\text{Sr}_{\text{River1}}) + f_{\text{Sr River2}}(^{87}\text{Sr}/^{86}\text{Sr}_{\text{River2}}) \dots + f_{\text{Sr groundwater}}(^{87}\text{Sr}/^{86}\text{Sr}_{\text{groundwater}}) + f_{\text{Sr Titicaca}}(^{87}\text{Sr}/^{86}\text{Sr}_{\text{Titicaca}}). \quad (1)$$

Here $f_{\text{Sr River 1, River 2, etc.}}$ = the fraction of Sr from each of the major rivers entering the southern Altiplano, and $^{87}\text{Sr}/^{86}\text{Sr}_{\text{River 1, River 2, etc.}}$ = the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for each of the major rivers. Here, we assume that the Sr contribution from direct rainfall onto the lake surface is negligible. In contrast, the groundwater term can make significant contributions to Sr budgets (e.g. Hart et al., 2004) and can be difficult to establish. In most cases, groundwater contributes primarily to streams rather than flowing into lakes, allowing accurate Sr budget models without direct consideration of this term (e.g. Pretti and Stewart, 2002). This assumption was proven valid on the Altiplano by Grove et al. (2003), who successfully reconstructed the modern $^{87}\text{Sr}/^{86}\text{Sr}$ for Lake Titicaca without the groundwater term, and we adopt this simplification in our modern and paleolake models.

Mass balance models typically describe the Sr fractional contribution (f) of individual watershed (River 1) as:

$$f_{\text{river1}} = \frac{(\text{concSr}_{\text{river1}}) \times (\text{discharge}_{\text{river1}})}{\sum_{\text{river1}} (\text{concSr}_{\text{river1,2,3,...x}}) \times (\text{discharge}_{\text{river1,2,3,...x}})} \quad (2)$$

Table 2
Isotopic data from carbonates.

| Sample/lake cycle | $^{87}\text{Sr}/^{86}\text{Sr}$ (± 1 SE) | Sr ppm (± 1 SE) | Sample material | Sample location (latitude; longitude; site # ^a) | $\delta^{18}\text{O}^b$ (‰) |
|----------------------------|---|----------------------|-----------------|---|-----------------------------|
| <i>Coipasa</i> | | | | | |
| S-8-2 | 0.70856 \pm 1 | 1978 \pm 2 | Tufa | 19°50'S; 67°09'W;14 | −0.5 |
| S-10-3 | 0.70856 \pm 1 | | Tufa | 19°50'S; 67°09'W;14 | 0.0 |
| U-31-3 | 0.70852 \pm 1 | | Tufa | 20°14'S; 67°38'W;23 | 1.7; 0.4 |
| U-31-4 | 0.70852 \pm 1 | | Tufa | 20°14'S; 67°38'W;23 | 2.1; 1.4 |
| U-23a-3 | 0.70852 \pm 1 | | Tufa | 20°57'S; 67°02'W | 0.5 |
| U-23a-4 | 0.70850 \pm 1 | | Tufa | 20°57'S; 67°02'W | 0.1 |
| C-14-1 | 0.70853 \pm 1 | | Tufa | 19°07'S; 67°47'W | |
| U-32-3 | 0.70854 \pm 1 | | Tufa | 20°14'S; 67°37'W | |
| U-9-6 | 0.70855 \pm 1 | 1578 \pm 0.5 | Tufa | 20°36'S; 67°37'W;25 | 1.1; 0.1; −1.2 |
| B-17 | 0.70854 \pm 1 | 1572 \pm 1 | Tufa | 19°51'S; 67°33'W;15 | 0.0; 0.3 |
| U-27b-6 | 0.70855 \pm 1 | 2050 \pm 2 | Tufa | 20°09'S; 67°49'W;19 | |
| U-27b-8 | 0.70853 \pm 1 | | Tufa | 20°09'S; 67°49'W;19 | |
| U-27a-2 | 0.70853 \pm 1 | | Tufa | 20°49'S; 67°16'W | |
| U-27a-3ex | 0.70857 \pm 1 | | Tufa | 20°49'S; 67°16'W | |
| U-27a-6 | 0.70851 \pm 1 | | Tufa | 20°49'S; 67°16'W | 1.6; 0.3 |
| U-10 | 0.70850 \pm 1 | 2300 \pm 0.5 | Tufa | 20°49'S; 67°09'W;30 | −0.3; 1.8 |
| S-6-3 ^c | 0.70867 \pm 1 | | Tufa | 19°50'S; 67°09'W;14 | −1.2 |
| Average | 0.70854 \pm 4 | 1990 \pm 320 | | | 0.4 \pm 1.00 |
| <i>Tauca transgression</i> | | | | | |
| U-29a-1 | 0.70887 \pm 1 | 1130 \pm 1 | Ooids | 20°07'S; 67°02'W;18 | 1.3 |
| U-27b-2 | 0.70881 \pm 1 | 1859 \pm 2 | Tufa | 0.7 | 0.7 |
| S-17-4 | 0.70884 \pm 2 | | Tufa | 19°50'S; 67°09'W;14 | |
| S-6-1 | 0.70887 \pm 1 | | Tufa | 19°50'S; 67°09'W;14 | |
| Average | 0.70885 \pm 3 | | | | 1.0 \pm 0.3 |
| <i>Tauca</i> | | | | | |
| S-17-5 | 0.70884 \pm 2 | | Tufa | 19°50'S; 67°09'W;14 | −1.2 |
| S-17-6 | 0.70883 \pm 1 | | Tufa | 19°50'S; 67°09'W;14 | −0.5 |
| RG2-1 | 0.70889 \pm 1 | | Tufa | 20°47'S; 67°06'W | −1.1 |
| RG2-3 | | | Tufa | 20°47'S; 67°06'W | 0.6 |
| CL-2 | 0.70877 \pm 1 | | Tufa | Ascotan, Chile | 0.2 |
| U-23a-2 | 0.70885 \pm 1 | | Tufa | 19°41'S; 67°39'W | |
| U-31-2 | 0.70881 \pm 1 | | Tufa | 20°14'S; 67°38'W;23 | |
| U-26-5 | 0.70882 \pm 1 | | Tufa | 20°48'S; 67°39'W;24 | |
| U-23a-1 | 0.70885 \pm 1 | | Tufa | 20°57'S; 67°02'W | |
| C-1-1 | 0.70880 \pm 1 | 1139 \pm 0.5 | Tufa | 19°37'S; 68°06'W | −2.5 |
| C-1-2 | 0.70880 \pm 6 | 1035 \pm 0.5 | Tufa | 19°37'S; 68°06'W | −2.2 |
| C-1-3 | 0.70880 \pm 1 | 1268 \pm 0.5 | Tufa | 19°37'S; 68°06'W | −1.5 |
| C-1-4 | 0.70885 \pm 1 | 1205 \pm 0.5 | Tufa | 19°37'S; 68°06'W | −0.8 |
| U-5-1 | 0.70881 \pm 1 | 1045 \pm 0.5 | Tufa | 20°36'S; 67°35'W;25 | −1.6; −2.0 |
| U-5-2 | 0.70877 \pm 1 | 1120 \pm 0.5 | Tufa | 20°36'S; 67°35'W;25 | −1.9; −2.4 |
| U-5-3 | 0.70885 \pm 1 | 995 \pm 0.5 | Tufa | 20°36'S; 67°35'W;25 | |
| U-9-2 | 0.70879 \pm 1 | 1161 \pm 0.5 | Tufa | 19°50'S; 67°33'W;15 | −0.5; −0.9 |
| U-9-1 | | | Tufa | 19°50'S; 67°33'W;15 | −2.4 |
| U-9-3 | | | Tufa | 19°50'S; 67°33'W;15 | −1.8 |
| U-9-4 | | | Tufa | 19°50'S; 67°33'W;15 | −1.4 |
| U-9-5 | | | Tufa | 19°50'S; 67°33'W;15 | −1.0 |
| U-7-1 | 0.70881 \pm 1 | 1215 \pm 0.5 | Tufa | 20°43'S; 67°59'W;29 | −1.5 |
| U-7-2 | 0.70884 \pm 1 | 1363 \pm 0.5 | Tufa | 20°43'S; 67°59'W;29 | |
| U-11-2 | 0.70883 \pm 1 | 1060 \pm 0.5 | Tufa | 20°49'S; 67°08'W | −1.9 |
| P-3-1 | 0.70888 \pm 1 | 890 \pm 0.5 | Tufa | 18°34'S; 66°57'W | −2.1; −2.9 |
| P-3-11 | 0.70884 \pm 2 | 883 \pm 1 | Tufa | 19°21'S; 67°13'W | −2.3 |
| B-8 | 0.70881 \pm 1 | 1154 \pm 0.5 | Tufa | 19°51'S; 67°33'W;15 | −1.1 |
| U-27a-3ex | 0.70879 \pm 1 | | Tufa | 20°09'S; 67°49'W;19 | |
| U-27a-5 | 0.70875 \pm 3 | | Tufa | 20°09'S; 67°49'W;19 | 1.6 |
| U-27a-4 | 0.70881 \pm 2 | | Tufa | 20°09'S; 67°49'W;19 | |
| U-27a-3in | 0.70883 \pm 1 | | Tufa | 20°09'S; 67°49'W;19 | 0.4 |
| P-6-1 | 0.70880 \pm 1 | | Tufa | 17°59'S; 67°03'W | |
| C-8-1 | 0.70876 \pm 2 | | Tufa | 19°19'S; 68°22'W | |
| C-8-2 | 0.70878 \pm 2 | | Tufa | 19°19'S; 68°22'W | |
| E-2-1 | 0.70877 \pm 1 | | Tufa | 20°14'S; 68°28'W | |
| C-2-2 | | | Tufa | 19°40'S; 68°06'W;13 | −2.0 |
| Chita-2 | | | Tufa | 20°06'S; 66°58'W;20B | −0.8 |
| U-7-1 | | | Tufa | 20°43'S; 67°59'W;29 | −0.4 |
| U-4-1 | | | Tufa | 20°05'S; 68°13'W;17 | 3.4 |
| S-6-1 | | | Tufa | 19°50'S; 67°09'W;14 | −1.5 |
| S-3-1 | | | Tufa | 19°50'S; 67°09'W;14 | 0.6 |
| S-20-1 | | | Tufa | 19°50'S; 67°09'W;14 | −2.8 |
| Average | 0.70881 \pm 3 | 1150 \pm 215 | | | −1.1 \pm 1.3 |

(continued on next page)

Table 2 (continued)

| Sample/lake cycle | $^{87}\text{Sr}/^{86}\text{Sr}$ (± 1 SE) | Sr ppm (± 1 SE) | Sample material | Sample location (latitude; longitude; site # ^a) | $\delta^{18}\text{O}^b$ (‰) |
|-------------------------|---|-----------------------------|-----------------|---|-----------------------------|
| <i>Tauca regression</i> | | | | | |
| B-12 | 0.70878 \pm 1 | 1067 \pm 0.05 | Tufa | 19°51'S; 67°33'W;15 | 0.1; -0.1 |
| B-14 | 0.70880 \pm 1 | 1099 \pm 0.05 | Tufa | 19°51'S; 67°33'W;15 | -0.1; -0.71 |
| S-20-1 | 0.70878 \pm 1 | | Cemented gravel | 19°50'S; 67°09'W;14 | -2.8 |
| Average | 0.70879 \pm 1 | | | | -0.7 \pm 1.2 |
| <i>Sajsi</i> | | | | | |
| S-17-7 | 0.70883 \pm 1 | | Tufa | 19°50'S; 67°09'W;14 | -1.1 |
| S-17-4 | | | Tufa | 19°50'S; 67°09'W;14 | -3.1 |
| S-17-3 | 0.70888 \pm 1 | | Tufa | 19°50'S; 67°09'W;14 | |
| S-17-1 | 0.70884 \pm 1 | 688 \pm 1 | Ooids | 19°50'S; 67°09'W;14 | -3.8 |
| S-17-2 | 0.70876 \pm 1 | | Ooids | 19°50'S; 67°09'W;14 | -3.5 |
| Average | 0.70883 \pm 5 | | | | -2.9 \pm 1.2 |
| <i>Inca Huasi</i> | | | | | |
| U-28-1 | 0.70882 \pm 1 | | Tufa | 19°57'S; 68°15'W;16 | -0.5 |
| U-27a-1 | 0.70887 \pm 1 | 2191 \pm 3 | Tufa | 20°09'S; 67°49'W;19 | 1.2 |
| Average | 0.70884 \pm 3 | | | | 0.4 \pm 1.2 |
| <i>Salinas</i> | | | | | |
| U-23b-1b | 0.70882 \pm 1 | | Tufa | 19°41'S; 67°39'W | |
| U-23b-2 | 0.70882 \pm 1 | | Tufa | 19°41'S; 67°39'W | |
| U-22-1 | 0.70872 \pm 1 | | Tufa | 19°41'S; 67°39'W;11 | -1.3 |
| U-22-2 | 0.70884 \pm 1 | | Tufa | 19°41'S; 67°39'W;11 | -0.5 |
| U-22-3 | 0.70887 \pm 1 | | Tufa | 19°41'S; 67°39'W;11 | -2.2 |
| U-22-4 | 0.70888 \pm 1 | | Tufa | 19°41'S; 67°39'W;11 | -0.5 |
| U-22-5 | | | Tufa | 19°41'S; 67°39'W;11 | 0.4 |
| U-26-1 | 0.70872 \pm 1 | | Tufa | 20°48'S; 67°39'W;24 | -0.6 |
| U-26-2 | 0.70885 \pm 1 | | Tufa | 20°48'S; 67°39'W;24 | -0.4 |
| U-26-3 | 0.70883 \pm 1 | 2041 \pm 3 | Tufa | 20°48'S; 67°39'W;24 | -0.2 |
| U-26-4 | 0.70876 \pm 1 | | Tufa | 20°48'S; 67°39'W;24 | |
| U-31-1 | 0.70885 \pm 1 | 2637 \pm 4 | Tufa | 20°14'S; 67°38'W;23 | -0.9 |
| Average | 0.70882 \pm 6 | 2340 \pm 420 | | | -0.7 \pm 0.7 |
| <i>Ouki</i> | | | | | |
| T-3 | 0.70926 \pm 1 | 1958 \pm 0.5 | Tufa | 18°17'S; 67°32'W;2 | -1.2 |
| T-9 | 0.70943 \pm 1 | 1851 \pm 0.5 | Tufa | 18°17'S; 67°32'W;2 | -1.7 |
| P-11 | 0.70928 \pm 1 | 1839 \pm 1 | Tufa | 18°35'S; 66°56'W;4 | -2.2 |
| P-17 | 0.70926 \pm 1 | 1657 \pm 0.5 | Tufa | 18°35'S; 66°56'W;4 | -3.3 |
| P-5-3 | 0.70929 \pm 1 | 1874 \pm 1 | Tufa | 18°43'S; 66°52'W;5 | -1.2; -0.9 |
| T-13 | 0.70944 \pm 1 | 1888 \pm 1 | Tufa | 18°17'S; 67°32'W;2 | -2.2 |
| P-7-2 | 0.70928 \pm 1 | 2184 \pm 3 | Tufa | 18°00'S; 67°03'W;1 | |
| C-19-1 | 0.70944 \pm 3 | | Shells | 19°19'S; 67°09'W;8 | |
| C-22-1 | 0.70918 \pm 1 | | Shells | 19°20'S; 67°11'W;8 | |
| C-21-1 | 0.70932 \pm 1 | 1368 \pm 4 | Shells | 19°20'S; 67°11'W;8 | |
| T-24 | | | Tufa | 18°17'S; 67°32'W;2 | -2.5 |
| P-8-2 | | | Tufa | 18°00'S; 67°02'W;1B | 0.4 |
| B-3 | | | Tufa | 18°17'S; 67°32'W;2 | -1.6 |
| Average | 0.70932 \pm 9 | 1830 \pm 240 ^d | | | -1.6 \pm 1.0 |

^a Site numbers correspond to site described by Placzek et al. (2006a).

^b Analytical error (1 σ) on $\delta^{18}\text{O}$ is typically 0.08‰.

^c Excluded from average value.

^d Average concentration values only include data from tufas.

Reliable discharge data is unavailable for most of our basin rivers, and hence we create a simple model that assumes that catchment area is proportional to discharge. Our final model improves on this assumption by weighting catchments by the modern west to east decrease in rainfall seen across the Altiplano (Fig. 1B).

Several useful generalizations can be drawn from the output of this simple model. It is apparent that there are two basic regional inputs that

ultimately determine the isotopic composition of basin lakes past and present (Fig. 1B). The first region is watersheds draining the relatively radiogenic (higher $^{87}\text{Sr}/^{86}\text{Sr}$) rocks of the EC ($^{87}\text{Sr}/^{86}\text{Sr}_{\text{ec}} = 0.71096$, $f_{\text{Sr}} = 0.87$). The second region consists of watersheds draining the relatively non-radiogenic (lower $^{87}\text{Sr}/^{86}\text{Sr}$) rocks of the WC with an $^{87}\text{Sr}/^{86}\text{Sr}_{\text{wc}} = 0.70687$ and $f_{\text{Sr}} = 0.13$; this is dominated by the Río Lauca which flows into the Coipasa Basin. Higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios therefore must mean greater input from the EC and lower ratios greater input from the WC.

$\delta^{18}\text{O}$

We also constructed a $\delta^{18}\text{O}$ evolution model that can be compared to $\delta^{18}\text{O}$ values of modern and ancient lakes. This model is based on that of Craig and Gordon (1965) and predicts changing $\delta^{18}\text{O}$ of lake water with continued evaporation:

$$\delta_L = (\delta_0 - A/B)f^B + A/B \quad (3)$$

Table 3

Average isotopic values for paleolake carbonates.

| Lake cycle | $\delta^{234}\text{U}/^{238}\text{U}$ (activity) | $\delta^{18}\text{O}$ (‰) | $^{87}\text{Sr}/^{86}\text{Sr}$ |
|------------|--|---------------------------|---------------------------------|
| Coipasa | 1.76 \pm 0.02 | 0.4 \pm 1.00 | 0.70854 \pm 4 |
| Tauca | 1.60 \pm 0.03 | -0.95 \pm 1.3 | 0.70881 \pm 3 |
| Sajsi | | -2.9 \pm 1.2 | 0.70883 \pm 5 |
| Inca Huasi | | 0.4 \pm 1.2 | 0.70884 \pm 3 |
| Salinas | 1.67 \pm 0.02 | -0.7 \pm 0.7 | 0.70882 \pm 6 |
| Ouki | 1.60 \pm 0.04 | -1.6 \pm 1.0 | 0.70932 \pm 9 |

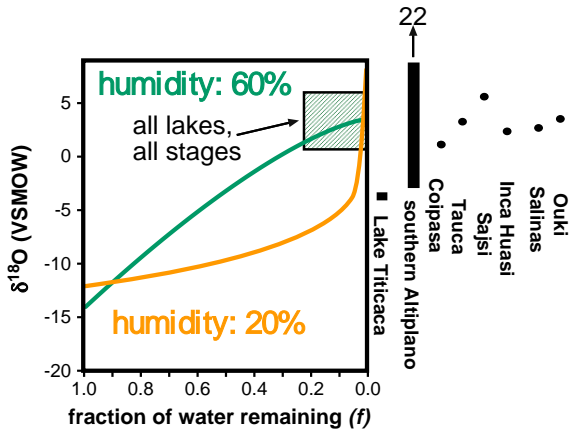


Figure 6. Evaporative model for southern Altiplano lakes. For highly evaporated lakes, changes in $\delta^{18}\text{O}$ due to evaporation are much less for high (>50%) relative humidity (black line) than low humidity (gray line; <50%). Initial $\delta^{18}\text{O}$ (VSMOW) values of -14‰ are assumed for the 60% humidity model, consistent with modern rivers flowing into Titicaca; a value of -12‰ is assumed for the 20% humidity model (after modeled Tauca-phase values from Cross et al., 2001). Both models assume an $\epsilon_{\text{water-vapor}} = 18\text{‰}$ and temperature = 8°C . Boxes indicate values of modern waters. Reconstructed paleolake water values (VSMOW) are shown as circles assuming modern temperature (10.8°C) minus the average temperature change at Vostock (Petit et al., 1999) for each lake cycle.

where,

$$A = \frac{h\delta_A + \Delta\epsilon + \epsilon/\alpha}{1-h + \Delta\epsilon}; \tag{4}$$

$$B = \frac{h-\Delta\epsilon-\epsilon/\alpha}{1-h-\Delta\epsilon}; \tag{5}$$

and

$$\Delta\epsilon = 14.2(1-h) \tag{6}$$

where δ_L is the oxygen isotope composition of the lake water, f is the fraction of water remaining, δ_A is the oxygen isotope composition of the atmosphere, h is the relative humidity, and ϵ is the equilibrium enrichment factor, and α is the equilibrium fractionation factor.

As visible in Eqs. (3)–(6), relative humidity is critical in determining the shape of the $\delta^{18}\text{O}$ evolution curve (Fig. 6). Modern lake values that vary over 30‰ are associated with both low humidity and extreme evaporative enrichment of modern rainfall inputs of about -17.5‰ (Cross et al., 2001).

Discussion

Paleohydrologic implications

The range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios displayed by the lake carbonates must ultimately be tied to the shifting balance of inputs from the WC (0.70687) and the EC (0.71096). Today, an increased flux from the EC (largely the Poópo Basin) would raise composite lake ratios, whereas an increased flux of WC waters (principally the Coipasa/Uyuni basin) would decrease it. Using the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of paleolake carbonates, this would mean that the relative WC/EC Sr contribution to modern Lake Titicaca is 0.67, to the Coipasa lake cycle was 0.60, to the Ouki lake cycle was 0.41, and to all other lakes was about 0.53. Applying this simple model to the modern Titicaca fails to reproduce the modern $^{87}\text{Sr}/^{86}\text{Sr}$ value for Lake Titicaca. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of Lake Titicaca produced by this simple model is 0.7106, much higher than the observed ratio for the lake today of 0.7082–0.7084.

We can bring observed and modeled $^{87}\text{Sr}/^{86}\text{Sr}$ ratios into better agreement for modern Lake Titicaca by recognizing that catchments in the EC with peaks between 4500 and 6400 m have relative discharges that are much smaller than predicted by their catchment area. Indeed, the average discharge of such catchments (Table 1, #2, 3, 4, 8, and 9) in the Lake Titicaca basin is $\sim 40\%$ that of the value predicted by the catchment area (calculated from discharge data in Grove et al., 2003). This likely arises from both orographic effects related to humid air flowing around high peaks of the EC and lake effect precipitation, which is also greater to the west (Roche et al., 1992; Fig. 1B). Our final model uses a weighting factor for catchments in the EC with peaks between 4500 and 6400 m (Table 1). Adding the assumption that Sr contributions from Lake Titicaca are consistent with the modern situation ($\sim 5\%$ input from Titicaca), our model yields $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and fractional contributions for the Poópo basin of 0.7107 and $f=0.11$, and $^{87}\text{Sr}/^{86}\text{Sr}=0.7082$ and $f=0.84$ for the Uyuni/Coipasa basin (Table 4). This model also produces an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70823 for Lake Titicaca, which closely matches observed ratios of 0.708215 to 0.708467 (Table 3; from Grove et al. (2003)). This suggests to us that our estimates of discharge using modified watershed area are valid. Unlike Titicaca waters, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the much smaller southern lakes are seasonally variable and probably influenced by rare flood events and salt dissolution (Table 1), making comparisons of modeled and observed ratios more problematic.

Our model reveals that the relatively low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of waters from the Coipasa/Uyuni basins are balanced by waters with higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from the Poópo basin, but for the past we have to consider the possibility that the fractional Titicaca contribution was larger than 5%, introducing another way, in addition to the WC source, to lever down bulk $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. The treatment of the Titicaca term for the past is the fundamental difference between two previous models of the Sr budget for the southern Altiplano (Coudrain et al.,

Table 4
Lake budgets.

| Basin | Poópo | Coipasa/Uyuni | Titicaca | Poópo | Coipasa/Uyuni | Titicaca |
|-------------------------------------|-------------------|-------------------|------------------|-----------|---------------|-----------------|
| Modern values (Sr model) | 0.71073; $f=11\%$ | 0.70847; $f=84\%$ | 0.70825; $f=5\%$ | | | |
| Modern values (water ^a) | | | | 96% | | 4% |
| Area (southern Altiplano) | | | | 38% | 62% | |
| Area | | | | 28% | 47% | 22% |
| Relative Sr concentration | 0.22 | 1.35 | 0.29 | | | |
| Lake cycle | Sr (%) | Sr (%) | Sr (%) | Water (%) | Water (%) | Water (%) |
| Coipasa | 2.5 | 97.5 | | 10 | 90 | |
| Tauca/Salinas/Inca Huasi/Sajsí | 16 | 80 | <1 | 41 | 54 | 4 ^a |
| Ouki A | 39 | 57 | <1 | 69 | 27 | 4 ^a |
| Ouki B ^b | | 44 | 56 ^b | 31 | | 69 ^b |

^a Assumed values from Coudrain et al. (2002).

^b Assuming no contribution from Coipasa/Uyuni, we suggest that actual values are somewhere between the Ouki A and B models, probably closer to the Ouki A model.

2002; Grove et al., 2003). For the Tauca lake cycle, the Grove et al. (2003) strontium budget model balances modern $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for Lake Titicaca (0.7083) with a composite value of 0.7105 for the entire southern Altiplano determined from 45% of the watersheds, mostly in the Poópo basin. The Grove et al. (2003) model predicts that 70–83% of the water for the Tauca lake cycle originated from Lake Titicaca (Grove et al., 2003), a significant contrast to the modern situation where Lake Titicaca accounts for only 5% of the flux to Lake Poópo (Coudrain et al., 2002).

In our view, the evidence more strongly supports minimal contribution from lake Titicaca, similar to the modern situation. In addition to the evidence we have presented here for an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the southern Altiplano that is considerably less than 0.7105, there are four additional lines of evidence in support of minimal input from Lake Titicaca during the Tauca lake cycle; (1) values of $\delta^{18}\text{O}$ from paleolake carbonates imply significant rainfall in the southern basins for all lake cycles; (2) hydrologic budget considerations point to minimal input from Lake Titicaca during the Tauca lake cycle; (3) $^{234}\text{U}/^{238}\text{U}$ ratios indicate greater input from the southern basin during the Coipasa lake cycle; and (4) the absence of Coipasa-age shoreline tufas in the Poópo basin,

suggests that recharge may have dominantly come from the south. We discuss these lines of evidence as follows.

(1) Average values of $\delta^{18}\text{O}$ (PDB) of carbonates for all paleolake cycles and for all stages within various lake cycles range between -1.6 and $+0.5\%$. Assuming lake paleotemperatures in the $2\text{--}10^\circ\text{C}$ range (see caption of Figure 6 for further explanation), $\delta^{18}\text{O}$ (VSMOW) values of paleolake water can be reconstructed and fall between 0 and 5‰. We can compare the reconstructed $\delta^{18}\text{O}$ values of paleowaters to values modeled for evaporation under differing humidity conditions. We observe that modeled $\delta^{18}\text{O}$ values of highly evaporated waters converge towards the relatively narrow range of reconstructed paleowater values only under humid conditions (Fig. 6; 60% humidity). Under less humid conditions, as with today's southern Altiplano where humidity is $<20\%$, modeled lake water $\delta^{18}\text{O}$ values are higher and more variable than those observed in paleolake carbonates (Fig. 6).

In light of this modeling comparison, two important conclusions can be drawn from the low variance and very positive $\delta^{18}\text{O}$ values of paleolake waters. The very positive values point to highly evaporated lake water, with fraction of water remaining <0.5 at all lake stages. The low variance is most consistent with evaporation under much

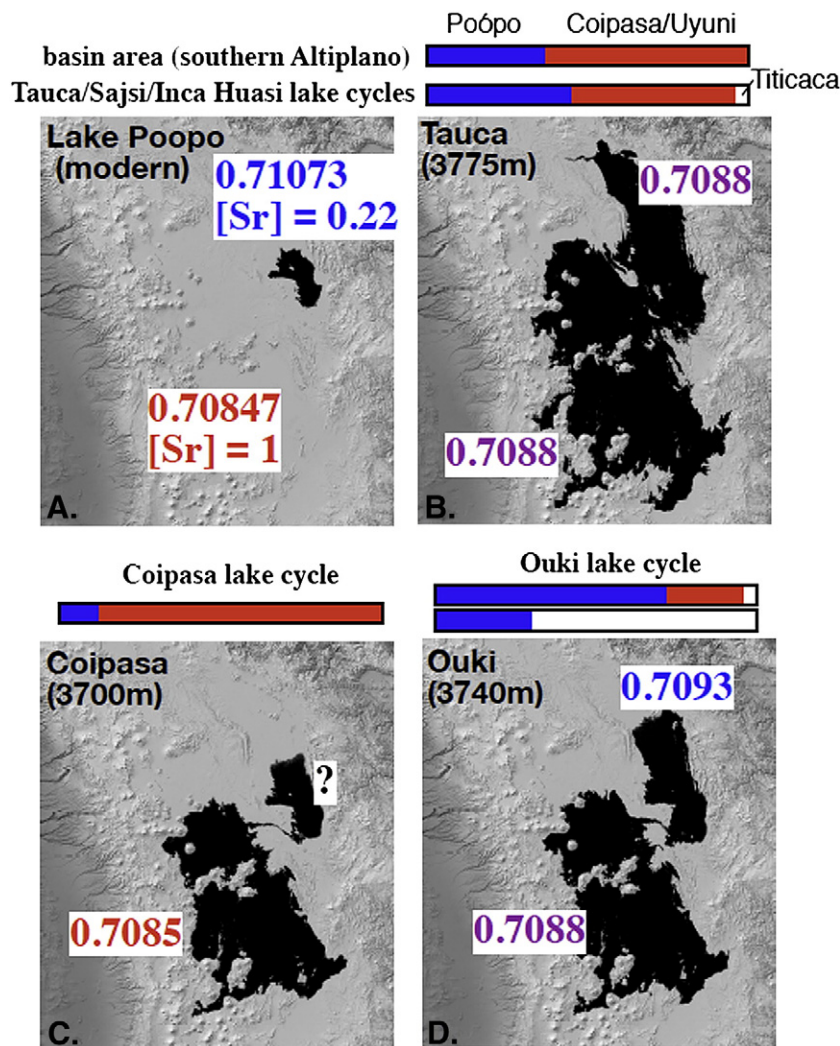


Figure 7. Modern topography (A) showing the possible extent of paleolakes Tauca (B), Coipasa (C), and Ouki (D) at the elevations described in text. The Laca hydrologic divide is broad and open only during the Tauca lake cycle. Ratios of $^{87}\text{Sr}/^{86}\text{Sr}$ from paleolake carbonates are shown for each lake cycle. For comparison, the relative basin area of the southern Altiplano is shown (top). Relative input from Lake Titicaca is assumed to be 4% for the Tauca lake cycle, consistent with the modern situation for Lake Titicaca. The Tauca lake cycle received ~45% of its input from the Poópo basin, which constitutes 39% of southern Altiplano. For the Coipasa lake cycle, only ~13% of the lake water came from the Poópo basin, assuming input from Lake Titicaca is negligible. We model the Ouki lake cycle with both maximum and minimal (4%) input from Lake Titicaca. If this input is minimal (4%), then 73% of the water from the Ouki lake cycle came from the Poópo basin and 22% came from the Coipasa/Uyuni basin. Assuming no input from the Coipasa/Uyuni basin requires that input from the Poópo basin (31%) be balanced by input from Lake Titicaca (69%).

higher humidity (~60%) levels than today (20%), as is graphically visible in Figure 6. The higher humidity in the past implies that more rain than today fell on the southern Altiplano during these lake expansions. These lakes, therefore, were not simply the arid receiving basins of water overflowing from the Titicaca basin.

(2) A variety of hydrologic budget models have been used to reconstruct climate conditions during the Taucas lake cycle (Hastenrath and Kutzbach, 1985; Blodgett et al., 1997; Cross et al., 2001; Condom et al., 2004; Blard et al., 2009). Although these models differ in details, all indicate that evaporation over the considerable surface area of the Taucas lake cycle (~60,000 km²) was huge (>70 × 10⁹ m³/yr), comparable to the discharge of major modern rivers such as the Nile or Rhine Rivers. Since maximum paleodischarge along the Río Desaguadero is estimated to be <20 × 10⁹ m³/yr (Coudrain et al., 2002), it is unrealistic that overflow from Lake Titicaca was sufficient to produce the largest paleolakes.

(3 and 4) Shoreline evidence and ²³⁴U/²³⁸U ratios indicate that the lower ⁸⁷Sr/⁸⁶Sr ratios for the Coipasa lake cycle result from relatively greater contributions from the EC (the Coipasa/Uyuni basin) and not from Lake Titicaca. The Coipasa lake cycle (12.8–11.4 ka) is the youngest lake cycle, and therefore shorelines should be preserved if filling came from Lake Titicaca overflow. Neither Coipasa-age shoreline tufas nor strandlines have been discovered so far in the Poópo basin, suggesting that filling may have been from the south, that is, by northward spilling of the Coipasa/Uyuni Basin. The correspondence between the Coipasa lake cycle highstand and the elevation of the Laca hydrologic divide is also consistent with the Coipasa/Uyuni basin lakes spilling northward into the Poópo basin (Placzek et al., 2009), the reverse of the usual situation today (Fig. 2).

Northward spilling of the Coipasa/Uyuni basins during the Coipasa lake cycle is supported by the very high ²³⁴U/²³⁸U ratios of Coipasa-

age tufas. Changes in the basin averaged ²³⁴U/²³⁸U value of surface waters should reflect changes in the ratio of chemical to physical weathering (Andersen et al., 2009). The ²³⁴U/²³⁸U ratio of surface waters reflects the preferential release of ²³⁴U during aqueous weathering. The ²³⁴U/²³⁸U ratio of surface waters and is strongly tied to soil development; all else being equal, soils formed in dry conditions should be less weathered and retain higher ²³⁴U/²³⁸U ratios (Andersen et al., 2009). Thus, the high ²³⁴U/²³⁸U ratios and shoreline evidence indicate that the source of low ⁸⁷Sr/⁸⁶Sr ratios for the Coipasa lake cycles is the Coipasa/Uyuni basin, not Lake Titicaca.

The relative contribution of water from the Poópo basin and across the Laca hydrologic divide is the principal factor influencing ⁸⁷Sr/⁸⁶Sr ratios of southern Altiplano paleolakes. The Taucas lake cycle, with an ⁸⁷Sr/⁸⁶Sr value of ~0.7088, resulted in a deep (>140 m) lake that integrated the Poópo and Coipasa/Uyuni basin. At highstand, a broad (>70 m), deep (>50 m) section of the lake covered the Laca hydrologic divide (Figs. 2C, 7). Additionally, ⁸⁷Sr/⁸⁶Sr ratios of ~0.7088 are observed not only for all stages of the Taucas lake cycle, but also from carbonates of the Sajsi, Inca Huasi, and Salinas lake cycles. Our models suggest that this ratio of ~0.7088 reflects a marginally greater contribution from the Poópo basin (Fig. 7B), a situation consistent with a very modest north–south precipitation gradient.

Geochemical and field evidence suggests that the Ouki lake cycle filled the Poópo basin to an elevation >3720 m, and overflowed southward over the Laca hydrologic divide (3700 m) into the Coipasa/Uyuni basin, where at times much shallower lakes were apparently present (Fig. 2D). Lakes existed in both the Poópo and Coipasa/Uyuni basin around ~95 ka. Sediments in the Poópo basin (Ouki) are at higher elevation (~3720 m) and have a higher ⁸⁷Sr/⁸⁶Sr ratio (~0.7093) at this time; tufa encrustations in the Uyuni basin (Salinas)

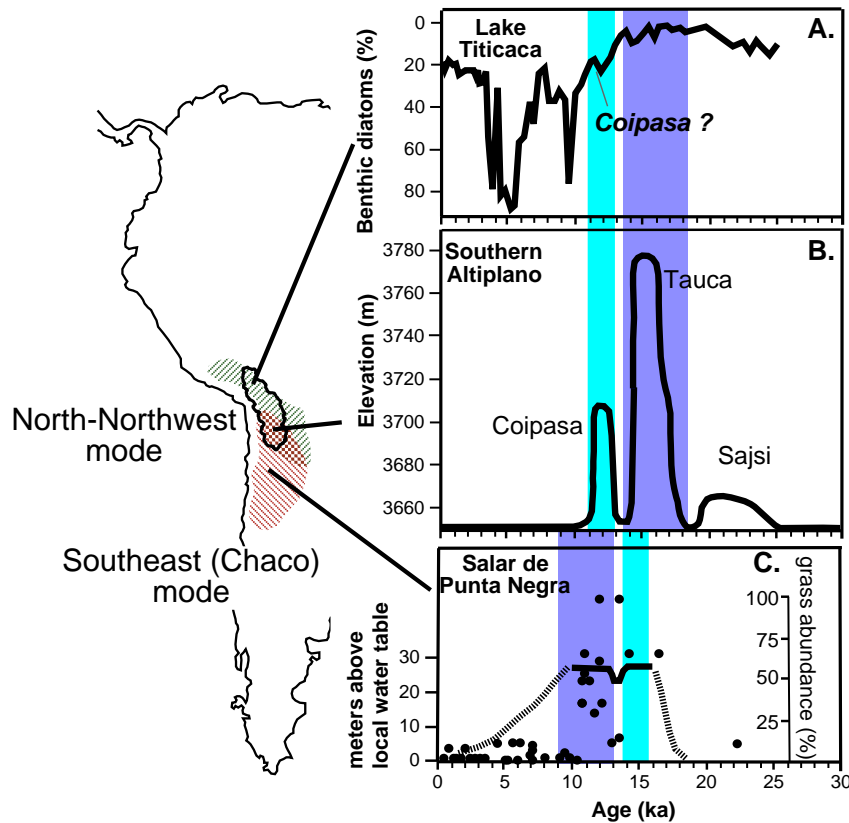


Figure 8. Comparison of paleohydrologic and climate proxies during CAPE and modes of modern precipitation variability (Vuille and Keimig, 2004) represented by overlapping shaded zones. A) Percent benthic diatoms from core 1 PC in Lake Titicaca (Baker et al., 2001b). B) Lake-level curve for the southern Altiplano (Placzek et al., 2006a). C) Reconstructed water table height (black line) (Quade et al., 2008) and grass abundance (circles) in rodent middens from the Punta Negra area (Latorre et al., 2002).

are at much lower elevation (<3675) and have lower (~ 0.7088) $^{87}\text{Sr}/^{86}\text{Sr}$ ratios over the same interval. This implies that although the Poópo basin (Ouki lake cycle) made significant contributions to the Coipasa/Uyuni basin (Salinas lake cycle) during this time, the Coipasa/Uyuni basin (Salinas lake cycle) made little (<22%) or no contribution to the lake (Ouki) in the Poópo basin. Sediments from the Ouki lake cycle consist of thick (generally >1 m) exposures of nearshore carbonates, either sands or massive conical tufa heads. Extensive shoreline sediments indicate prolonged stabilization of the lake by overflow of the Laca hydrologic divide.

Implications for regional climate variability

Recent work suggests regional variability in the timing and relative magnitude of the two phases of CAPE (Quade et al., 2008). Our new data supports this assertion. The first phase of CAPE (18.1–14.1 ka) produced the largest lake (the Taucá lake cycle) documented on the southern Altiplano and coincides not only with cold conditions in the North Atlantic (Heinrich event 1), but with intense and prolonged La Niña like conditions in the Pacific—both patterns that today bring wet years on the Altiplano. The second phase of CAPE corresponds to the Coipasa lake cycle (12.8–11.4 ka), and on the southern Altiplano is exactly coincidental with the Younger Dryas climate interval, another period of cooling in the North Atlantic. Evidence from rodent middens and wetlands in the Atacama region southeast of Uyuni, however, suggests that to the south the second phase of CAPE (12.8–8 ka) was not only longer-lived, but also wetter than the first phase (18.1–14.1 ka) (Latorre et al., 2002; Quade et al., 2008). In contrast, the second (Coipasa) phase of CAPE is less pronounced in a lake record from Lake Titicaca (Fig. 8A). One possible explanation for this pattern is that the first phase of CAPE was dominated by the north–northeast mode of rainfall, modulated by both cool North Atlantic temperatures and La Niña-like ocean and atmospheric conditions (Placzek et al., 2006a; Quade et al., 2008), and the second phase of CAPE may be dominated by the southeast (Chaco) mode of rainfall (Quade et al., 2008). Our strontium budget model for the Coipasa lake cycle indicates enhanced precipitation in the southernmost portion of the Altiplano which is consistent with paleo-precipitation that originated from the Chaco region of Argentina instead of from the Amazon. Variability in the Coipasa phase of CAPE must also be tied to cold

events in the North Atlantic, and coincidental warming of the southern Atlantic Ocean.

Use of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios as stratigraphic markers

The unique $^{87}\text{Sr}/^{86}\text{Sr}$ ratios associated with some of the lake cycles make $^{87}\text{Sr}/^{86}\text{Sr}$ ratios very useful stratigraphic tracers. This requires the assumption that lakes are isotopically uniform during any given lake cycle; an assumption that is validated for the Taucá lake cycle by replicated $^{87}\text{Sr}/^{86}\text{Sr}$ of ~ 0.7088 at diverse locations around the basins (Table 2). For example, ages from the youngest part of the Taucá lake cycle (18.1–14.1 ka) overlap at 2σ with Coipasa-age material (12.8–11.4 ka). In the absence of stratigraphic evidence, how do we distinguish them? Deposits from the Coipasa lake cycle consist of fresh, thin (<0.5 m) tufa crusts at low and moderate elevations (<3680 m) around the Salars of Coipasa and Uyuni. This carbonate is often superimposed on encrustations from the Taucá lake cycle in a manner that is not obviously unconformable (Fig. 9). The differences in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios between carbonates from the Taucá and Coipasa lake cycles can be used to tie carbonates at the lowest (e.g. U-31-4; Fig. 9) and highest elevations (e.g. Table 2; U-9-6, 3703 m) to the Coipasa lake cycle. Thus, both field and geochemical evidence now suggest that the Coipasa lake cycle occurred between 12.8 and 11.4 ka and had a highstand at ~ 3700 m.

Our models predict that the unique ratios associated with various lake cycles should be recorded in all lacustrine carbonates on a given side of the Laca hydrologic divide. Sub-basins divided by the hydrologic divide will have the same $^{87}\text{Sr}/^{86}\text{Sr}$ ratios only when the Laca hydrologic divide is a broad, deep hydrologic connection, as was the situation during the Taucá lake cycle (Figs. 2, 7). In contrast, different values are observed in the Uyuni (Salinas lake cycle) and Poópo basins (Ouki lake cycle) at ~ 95 ka. To what extent the Ouki-age (120–95 ka) lake reached the Coipasa/Uyuni basin is the key remaining unknown. Stratigraphic evidence suggests that this extent was possibly limited: no high elevation tufas of Ouki-age have been dated outside the Poópo basin. A test of this would be the analysis of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of carbonates or salts from cores in the Uyuni basin. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of about 0.7093 would demonstrate that overflow across the Laca hydrologic divide was substantial enough to dominate southern basin waters during Ouki time. Similarly, during Coipasa

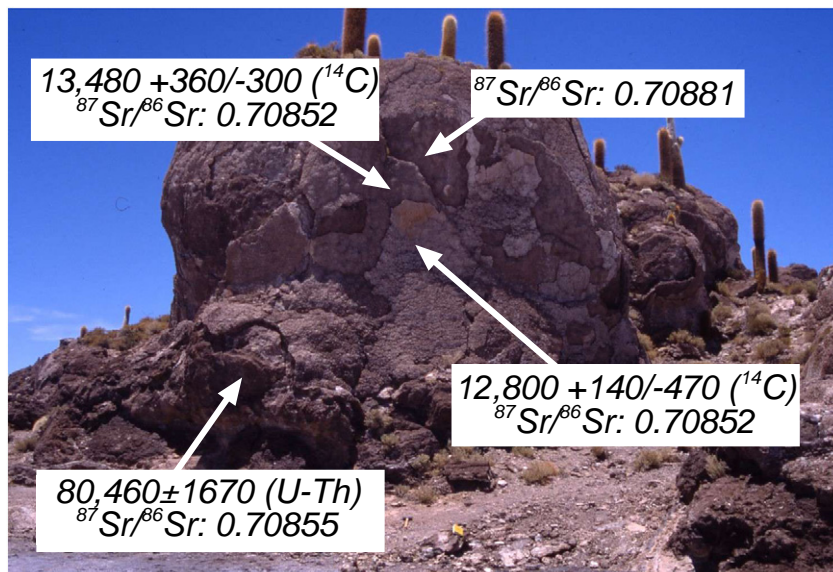


Figure 9. Example of multiple layers of tufa encrustation in the Uyuni basin. This site is only ~ 3 m above the elevation of the modern Uyuni salt pan. Both ages and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the youngest crusts are consistent with the transgression of the Coipasa lake cycle. Older crusts represent the Taucá and Salinas lake cycles and have $^{87}\text{Sr}/^{86}\text{Sr} = 0.7088$.

time (12.8–11.4 ka), $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in Poópo basin carbonates of ~0.7085 would point to the presence of a lake dominated by waters from the Coipasa/Uyuni basin. Intermediate values point to coincidental lakes in both sub-basins with more limited hydrologic transfer across the Laca hydrologic divide.

Conclusions

Strontium, uranium, and oxygen isotopic evidence provide key constraints on the late Quaternary lake history of the southern Bolivian Altiplano. The $^{87}\text{Sr}/^{86}\text{Sr}$ system in the southern Altiplano today is controlled by the interplay of high $^{87}\text{Sr}/^{86}\text{Sr}$ ratios found in the Poópo basin and waters with low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the Coipasa/Uyuni basin, plus or minus modest input from Lake Titicaca. We propose that the Ouki lake cycle (120–95 ka) was an expansive lake, deepest in the northern (Poópo) basin of the system, and spilling southward. Shoreline deposits from this lake cycle have elevated $^{87}\text{Sr}/^{86}\text{Sr}$ and point to recharge mainly from the Eastern Cordillera bordering the Poópo Basin. During the Tauca lake cycle (18.1–14.1 ka), all the major basins south of Titicaca were integrated into one deep lake (3770 m), and inflow to the lake came from local recharge in both the Poópo and Coipasa/Uyuni basins combined with a modest contribution from Lake Titicaca overflow. We attribute this pluvial event to intensification of the tropical summer monsoon, related to cooling in the North Atlantic, perhaps coupled with stronger La Niña conditions in the Pacific. We speculate that the Coipasa lake cycle (12.8–11.4 ka), with a transgression coincidental with the Younger Dryas event was sustained mostly from rainfall in the Coipasa/Uyuni basin, causing this southern lake to rise to the elevation of the Laca hydrologic divide and spill northward into the Poópo basin. This points to intensification of the SE (Chaco) mode of interannual variability in the summer monsoon, probably connected to both tropical and extratropical circulation changes brought about by North Atlantic cooling.

Supplementary materials related to this article can be found online at doi:10.1016/j.yqres.2010.08.004.

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