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# Impact of Bolivian paleolake evaporation on the $\delta^{18}$ O of the Andean glaciers during the last deglaciation (18.5–11.7 ka): diatom-inferred $\delta^{18}$ O values and hydro-isotopic modeling



Benjamin Quesada <sup>a, b, \*</sup>, Florence Sylvestre <sup>a, \*\*</sup>, Françoise Vimeux <sup>b, c</sup>, Jessica Black <sup>a</sup>, Christine Paillès <sup>a</sup>, Corinne Sonzogni <sup>a</sup>, Anne Alexandre <sup>a</sup>, Pierre-Henri Blard <sup>d</sup>, Alain Tonetto <sup>e</sup>, Jean-Charles Mazur <sup>a</sup>, Hélène Bruneton <sup>a</sup>

<sup>a</sup> Aix-Marseille Université, CNRS, IRD, CEREGE, UM 34, Europôle de l'Arbois, BP 80, 13545 Aix en Provence Cedex 04, France

<sup>b</sup> Institut Pierre Simon Laplace (IPSL), Laboratoire des Sciences de Climat et de l'Environnement (LSCE), UMR 8212 (CEA-CNRS-UVSQ), CEA Saclay,

Orme des Merisiers, Bât. 701, 91191 Gif-sur-Yvette, France

<sup>c</sup> Laboratoire HydroSciences Montpellier (HSM), UMR 5569 (CNRS-IRD-UM1-UM2), 34095 Montpellier, France

<sup>d</sup> Centre de Recherches Pétrographiques et Géochimiques (CRPG), UMR 7358, CNRS, Nancy-Université, Vandœuvre-lès-Nancy, France

<sup>e</sup> Aix-Marseille Université, PRATIM, Fédération de Chimie, 3 Place Victor Hugo, 13331 Marseille Cedex 3, France

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## ABSTRACT

During the last deglaciation, the Bolivian Altiplano (15–23°S, 66–70°W) was occupied by paleolake Tauca covering, at least, ~51,000 km<sup>2</sup> at its maximum highstand between 16.5 and 15 ka. Twenty-five hundred years later, after a massive regression, a new transgressive phase, produced paleolake Coipasa, smaller than Tauca and restricted to the southern part of the basin. These paleolakes were overlooked at the west by the Sajama ice cap. The latter provides a continuous record of the oxygen isotopic composition of paleo-precipitation for the last 25 ka. Contemporaneously to the end of paleolake Tauca, around 14.3 ka, the Sajama ice cap recorded a significant increase in ice oxygen isotopic composition  $(\delta^{18}O_{ice})$ . This paper examines to what extent the disappearance of Lake Tauca contributed to precipitation on the Sajama summit and this specific isotopic variation. The water  $\delta^{18}$ O values of paleolakes Tauca and Coipasa ( $\delta^{18}O_{lake}$ ) were quantitatively reconstructed from 18.5 to 11.7 ka based on diatom isotopic composition ( $\delta^{18}O_{diatoms}$ ) and ostracod isotopic composition ( $\delta^{18}O_{carbonates}$ ) retrieved in lacustrine sediments. At a centennial time scale, a strong trend appears: abrupt decreases of  $\delta^{18}O_{lake}$  during lake fillings are immediately followed by abrupt increases of  $\delta^{18}O_{lake}$  during lake level stable phases. The highest variation occurred at ~15.8 ka with a  $\delta^{18}O_{lake}$  decrease of about ~10‰, concomitant with the Lake Tauca highstand, followed ~400 years later by a 7% increase in  $\delta^{18}O_{lake}$ . A simple hydro-isotopic modeling approach reproduces consistently this rapid "decrease-increase" feature. Moreover, it suggests that this unexpected re-increase in  $\delta^{18}O_{lake}$  after filling phases can be partly explained by an equilibration of isotopic fluxes during the lake steady-state. Based on isotopic calculations during lake evaporation and a simple water stable isotopes balance between potential moisture sources at Sajama (advection versus lake evaporation), we show that total or partial evaporation (from 5 to 60%) of paleolake Tauca during its major regression phase at 14.3 ka could explain the pronounced isotopic excursion at Sajama ice cap. These results suggest that perturbations of the local hydrological cycle in lacustrine areas may substantially affect the paleoclimatic interpretation of the near-by isotopic signals (e.g. ice core or speleothems).

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#### 1. Introduction

The isotopic composition of continental sedimentary and ice archives has long been used to infer past climate variability. To reconstruct reliable paleoclimate information, it is necessary to

<sup>\*</sup> Corresponding author. Aix-Marseille Université, CNRS, IRD, CEREGE, UM 34, Europôle de l'Arbois, BP 80, 13545 Aix en Provence Cedex 04, France. \*\* Corresponding author.

*E-mail addresses:* benjamin.quesada@kit.edu (B. Quesada), sylvestre@cerege.fr (F. Sylvestre).

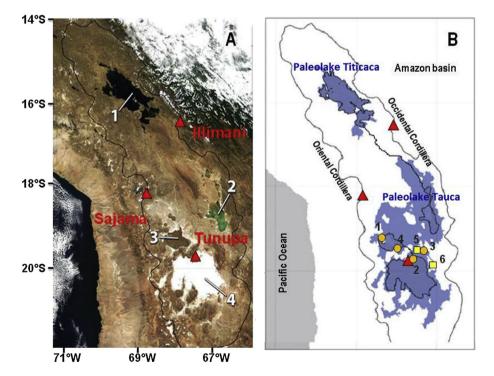
translate geochemical signals into quantified climate parameters. Within this framework, any environmental or climate process affecting the isotopic composition of climate archives must be tracked. In particular, continental water recycling involves fractionating and non-fractionating processes whose magnitudes have to be defined at the regional scale. In this paper, we examine the impact of such processes over the Bolivian Altiplano, which offers a unique opportunity to study the recycling moisture from a local source.

During the Quaternary, the Bolivian Altiplano  $(15-23^{\circ}S, 66-70^{\circ}W)$  has experienced large and abrupt lake-level oscillations recorded in both Lake Titicaca (Baker et al., 2001a) and Salar of Uyuni (Baker et al., 2001b). During the last deglaciation, two lacustrine phases were identified, namely the Lake Tauca and the lake Coipasa, whose highstands are coeval with Heinrich 1 event and the Younger Dryas, respectively (Figs. 1 and 2). Using previous geochronological data (Servant and Fontes, 1978; Sylvestre et al., 1999) and new U–Th series isochrons and <sup>14</sup>C ages, Blard et al. (2011) recently refined the chronology of the Lake Tauca phase between 18 and 14.2 ka. During the highstand at 3770 m, which lasted from 15.7 to 15 ka, the Lake Tauca covered more than 51,000 km<sup>2</sup>. It is recognized as one of the major lacustrine events that occurred in South America during the Late Quaternary. The lake Coipasa phase occurred later, between 12.5 and 11.9 ka, and was of lower amplitude (~3710 m).

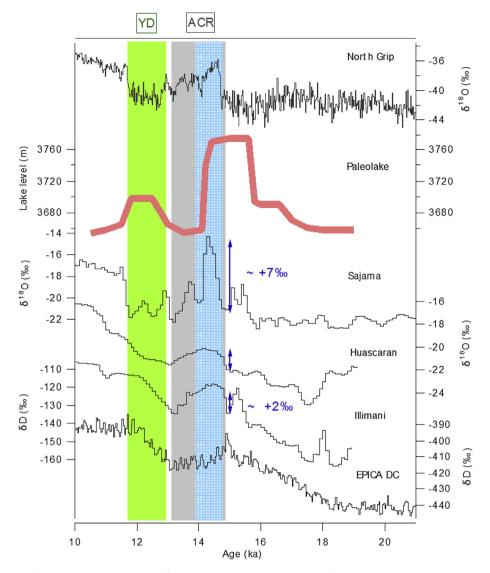
Interestingly, an ice core was extracted from an Andean summit overlooking these Altiplano lakes and covers those periods. It was drilled on the occidental Cordillera (18°06′S, 68°53′W, 6542 m; Fig. 1), at the Sajama ice cap, and displays a 25,000 year-long climate history (Thompson et al., 1998). The Sajama ice isotopic composition is rather constant, between 25 and 15.8 ka. Then, at 14.3 ka, an abrupt  $\delta^{18}O_{ice}$  increase up to +7‰ (hereafter named "isotopic excursion") occurs ( $\delta^{18}O_{ice}$  attains ~-14‰), followed by a return to LGM values (<-20‰) between 14 and 12 ka (Fig. 2). It is worth noting that the deglacial progression at Sajama is highly different from the one recorded in the Huascaran ice core. However, the latter is located far northward in Peru (9°06′S, 77°36′W, 6048 m) on the oriental Cordillera (Thompson et al., 1995) and different climate changes can explain different deglacial histories. Comparing Sajama and nearby Illimani isotopic records, a 5 per mil difference is observed during Tauca phase. The Nevado Illimani is located on the oriental Cordillera (16°37′S, 67°46′W, 6300 m; Fig. 1). However, a recent dating effort put the age of Illimani on a much younger scale (Sigl et al., 2009) then caution should be required regarding the eventual differences between Sajama and Illimani around ~15–18 ka.

To this day, no sound explanation has been put forth to explain this abrupt  $\delta^{18}O_{ice}$  increase at 14.3 ka. According to Blard et al. (2011), an explanation might be related to the evaporation and disappearance of Lake Tauca between 14.5 and 14 ka (Fig. 2). Indeed, recent studies (Lee et al., 2009; Vimeux et al., 2005, 2009, 2011; Vuille and Werner, 2005) combining observations and modeling have showed that the isotopic composition of Andean precipitation is essentially related to regional convective processes and then to regional precipitation rather than temperature changes as suggested by Thompson et al. (1998). Thus, the positive isotopic excursion at Sajama might signal a drier atmosphere, contemporaneous to the end of the Tauca phase (Fig. 2). This latter interpretation could however be in line with Thompson et al. (1998) who find high concentrations of marine and terrestrial anions (Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup> and NO<sup>3-</sup>) and dust in the Sajama core.

In this paper, we explore how the Sajama ice core isotopic excursion at 14.3 ka could be influenced by lake level changes and consecutive evaporation rate changes.



**Fig. 1.** (A) The Bolivian Altiplano today (MODIS image adapted from SALLJEX project: http://www.nssl.noaa.gov/projects/pacs/web/ALTIPLANO/motivation.html) with the location of (1) Lake Titicaca, a deep freshwater lake at 3810 m that extends over ~8750 km<sup>2</sup>; (2) Lake Poopó, a hypersaline shallow lake with an area of ~2500 km<sup>2</sup> at 3686 m; (3) Salars of Coipasa and (4) Uyuni covered by wide salt crusts of ~2500 km<sup>2</sup> and ~10,000 km<sup>2</sup> and located at ~3657 m and ~3653 m respectively. (B) Map of the Altiplano showing the extension of the paleolake Tauca and Titicaca in blue. The current lakes and dry salt flats are also indicated with contours and shaded with lines. The edges of the basin are indicated with a continuous black line. Location of studied sites are represented by orange circles for (1) Estancia Willa Khollu [EWK samples] (2) Churacari Bajo CB [C samples] (3) Pakollo Jahuira PJ [J0 samples] (4) Tauca [BT samples] (same as those of Blard et al., 2011) and by yellow squares for sites (5) Jahuila, (6) Nueva Esperanza from Blard et al. (2011). For both maps, red triangles indicate the Sajama and Illimani ice cores and the Tunupa volcano locations.



**Fig. 2.** Lake level (m) along with the oxygen isotopic composition of North Grip (NorthGrip community members 2004), Sajama (Thompson et al., 1998), Huascaran (Thompson et al., 1995), Illimani (Ramirez et al., 2003) and EPICA DC (EPICA community members 2004) ice cores (in ‰ vs SMOW) over the last 22,000 years. Younger Dryas (YD, green bar) and Antarctic Cold Reversal (ACR, gray bar) are defined with the North Grip and EPICA DC isotopic records respectively. The blue bar points to the end of Tauca phase.

In a first step, we reconstruct the oxygen isotopic composition of lakes Tauca and Coipasa (hereafter,  $\delta^{18}O_{lake}$ ) from 18.5 to 11.7 ka using the oxygen isotopic composition of fossil diatoms (hereafter,  $\delta^{18}O_{diatoms}$ ). This new record is constrained by oxygen isotopic composition of carbonates and of ostracod valves. In a second step, we assess whether basic features observed in the  $\delta^{18}O_{lake}$  signal can be explained by a hydro-isotopic modeling approach. We address to what extent Lake Tauca drying could explain the Sajama  $\delta^{18}O_{ice}$  records during deglaciation. Was Lake Tauca a non-negligible moisture source for precipitation at Sajama? If so, to what extent has lake evaporation contributed to an enrichment of the isotopic composition of ambient water vapor?

#### 2. Modern and past settings

The Bolivian Altiplano, located in tropical Central Andes, is an endorheic basin extending from 15°S to 22°S and 65°W to 69°W, between the oriental and occidental Cordilleras (Fig. 1). With an average elevation of ~3800 m a.s.l, this plateau covers a total area of ~197,000 km<sup>2</sup>. It consists of four main lacustrine basins from north

to south, (i) Lake Titicaca, a deep freshwater lake at 3810 m a.s.l that extends over ~8750 km<sup>2</sup>; (ii) Lake Poopó, a hypersaline shallow lake with an area of ~2500 km<sup>2</sup> at 3686 m a.s.l; (iii) Salars of Coipasa and (iv) Uyuni covered by wide salt crusts of ~2500 km<sup>2</sup> and ~10,000 km<sup>2</sup> and located at ~3657 m and ~3653 m a.s.l respectively. Lake Titicaca overflows into Lake Poopó *via* the Desaguadero River, its main outlet. During wet periods, Lake Poopó can fill Salars of Coipasa and Uyuni, establishing a hydrological link between the north and south of the Altiplano. During the rainy season (November to March), moisture, which mainly originates from the Atlantic Ocean, is advected to the Altiplano (Garreaud et al., 2003; Vimeux et al., 2011).

The lacustrine optimum Tauca most probably results from an increase in precipitation (Kessler, 1984; Servant-Vildary and Mello e Souza, 1993; Baker et al., 2001a,b; Condom, 2002) estimated to be between 1.6 and 3 times more important than nowadays (Blard et al., 2009). Several studies concluded that la Niña years (ENSO cold phase) are often associated with wet conditions on the Altiplano, enhancing easterly flow aloft from Tropical Atlantic ocean (e.g. Garreaud and Aceituno, 2001; Garreaud et al.,

2003). Actually, Tauca phase coincide with higher-than-average La Niña events (Placzek et al., 2006) which is consistent with prevailing easterly winds advecting moisture towards Sajama summit. However, a recent modeling study using the LMDz Atmosphere General Circulation Model shows a different mechanism (Mariotti et al., 2014). According to this study, the increase of moisture advection from Easterly/South-Easterly winds during the Tauca phase is more probably triggered by the inter-hemispheric sea surface temperature contrast in the Atlantic Ocean.

#### 3. Materials

#### 3.1. Diatom and Ostracod samples

Sixteen bulk samples rich in diatoms were selected from sediment outcrops around the Salars of Coipasa and Uyuni. These samples were described in a pioneering study using fossil diatom assemblages for paleoenvironmental reconstruction (Sylvestre, 1997; Sylvestre et al., 1999). Samples EWK 2, 3, 6, 7, 9, 12 and 14 were collected from an outcrop located in the northwest part of Salar of Coipasa in Estancia Willa Khollu (EWK site; 19°16'S–68°20'W, 3685 m) (Fig. 1). Samples C69, C83, C245, C248 and C272 were collected from Churacari Bajo outcrop (CB site; 19°72'S, 67°31'W; 3685 m) located on the northern part of Salar of Uyuni. Samples J038 and J047 came from Pakollo Jahuira outcrop (PJ site; 19°32'S–67°31'W, 3657 m), on the north edge of Salar of Uyuni, and samples BT08 and BT12 were collected on the southern edge of the Salar of Coipasa at Tauca site (19°30'S, 67°98'W, 3657 m) (Fig. 1).

From the aforementioned 16 diatom samples, ostracods were found in 5 samples EWK1, 2, 3, 10 and 11, where from 2 of them we could measure  $\delta^{18}O_{diatoms}$  as well as  $\delta^{18}O_{carbonates}$ .

#### 3.2. Chronology

Ages of the samples were constrained thanks to a chronostratigraphy of the Altiplano lacustrine deposits based on 44<sup>14</sup>C and 6 U/Th ages (Sylvestre et al., 1999) completed by new <sup>14</sup>C and U-Th data by Placzek et al. (2006) and Blard et al. (2011). Although Lake Tauca highstand is now well-dated, discrepancies still remain about the timing of its transgression. A two-step transgression based on diatom assemblages was first evidenced (Sylvestre et al., 1999): a slow first phase between 18.5 ka and 17 ka, and a second, very rapid at ~15.7 ka that reached the highest lake level at 3770 m a.s.l. Between 17 and 15.7 ka, the dominance of benthic diatoms suggested a stable water level at 3690 m. Conversely, dating of shoreline deposits does not evidence a stagnation phase but rather a rapid and continuous transgression after 17.5 ka (Placzek et al., 2006). These two scenarios called "Bioherm transgression scenario" and "Diatom transgression scenario" respectively were recently discussed (see Figure 8 in Blard et al., 2011), but the issue was not settled. The present study, based on the same sediment sections from which fossil diatom assemblages were analyzed, we thus consider here the "Diatom transgression scenario".

The studied sedimentary sections cover the Lake Tauca cycle between ~18.5 ka and 14.1 ka and the lake Coipasa phase between ~12.6 ka and 11.7 ka.

#### 4. Methods

## 4.1. The isotopic composition of diatoms ( $\delta^{18}O_{diatoms}$ )

The complete purification of diatom samples is a key step as any contaminants containing oxygen may change the measured  $\delta^{18}O_{diatoms}$  values. Here, the mineral phase of the sediment was dominated by clays (illite) and plagioclase feldspar (andesine). The

diatoms amorphous silica was cleaned from organic and sedimentary components using the protocol previously set up by Crespin et al. (2010). Seven steps were included: (1) dissolution of carbonates and oxidation of organic matter with an equimolar mixture of 20 ml of  $HClO_4$  (70%) and 20 ml of  $HNO_3$  (65%) heated at 50 °C during ~30 min; (2) oxidation of organic matter using 20 ml of  $H_2O_2$  (33%). Steps (1) and (2) were repeated five times, always followed by (3) a rinsing with distilled water; (4) clay removal by decanting in settling columns, repeated until clear suspension was reached. The final purification, depending on the amount and the size of diatoms, was achieved by physical extraction using (5) a laminar flux separation with SPLITT cell system and/or (6) a densimetric separation using ZnBr<sub>2</sub> at a density of 2.3; (7) samples were washed at least 8 times to remove acids and ZnBr<sub>2</sub> solution, and were dried at 50 °C at least during 24 h.

Purity of diatoms was checked by mineralogical (XRD) and elemental (micro-XRF) semi-quantitative analyses. The lowest diatoms SiO<sub>2</sub> concentration represented ca 98.3% of the concentrates (Table 1). The remaining ca 1% was mainly Al<sub>2</sub>O<sub>3</sub>, K<sub>2</sub>O, CaO under the form of andesine ((Ca, Na)(Al, Si)<sub>4</sub>O<sub>8</sub>), a plagioclase feldspar. Assuming for andesine a  $\delta^{18}$ O value close to the  $\delta^{18}$ O averaged value of continental basalt (6.4‰; Harmon and Hoefs, 1995), 1% of andesine remains may have decreased the  $\delta^{18}$ O values of the concentrates by less than 0.3‰, which is close to the  $\delta^{18}$ O<sub>silica</sub> reproducibility of 0.2‰ (cf below). Additionally, optical and SEM microscopies were used to check that the diatom frustules were devoid of organic remains and were not subject to diagenesis (Fig. 3a–c).

 $\delta^{18}$ O analyses of diatoms started by an inert Gas Flow Dehydration (iGFD; Chapligin et al., 2011): samples were dehydrated by ramp degassing (2 h heating to 1020 °C, 1.5 h held constant at 1020 °C, 2 h cooling down to 400 °C) under a continuous N<sub>2</sub> flow. Oxygen extractions were then performed using the IR Laser-Heating Fluorination Technique (Alexandre et al., 2006; Crespin et al., 2008). No ejection occurred during the analysis. The oxygen gas samples were directly sent to and analyzed by a dual-inlet mass spectrometer ThermoQuest Finnigan Delta Plus. Measured  $\delta^{18}O$ values were corrected on a daily basis using a quartz lab standard  $(\delta^{18}O_{\text{Boulangé 50-100 }\mu\text{m}})$  calibrated on NBS28 (9.6 ± 0.2‰; n = 6) (Alexandre et al., 2006; Crespin et al., 2008). The values were expressed in the standard  $\delta$ -notation relative to V-SMOW. The long term precision of the quartz lab standard was  $\pm 0.2\%$  (1 $\sigma$ ). Amorphous silica working standards used in the frame of the 2011 interlaboratory calibration (Chapligin et al., 2011) were analyzed. BFC (NERC, England), PS1772-8 (AWI, Germany) and MSG60 (CEREGE, France) gave  $\delta^{18}$ O values of 28.3  $\pm 0.6\%$  (n = 7), 43.0  $\pm 0.3\%$  (n = 6) and 37.5  $\pm$  0.3‰ (n = 2) respectively. These values are in the



Semi-quantitative analysis based on micro-XRF for major detectable elements of samples with high biogenic silica contents with (a) sample names and (b) elements detected.

a	b							
Samples	SiO <sub>2</sub>	$Al_2O_3$	K <sub>2</sub> O	CaO	TiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	$MnO_2$	SO <sub>3</sub>
BT08 BT12	98.5 98.5	0.3 0.3	0.4 0.4	0.2 0.2	0.1 0.1	0.4 0.4	0.0 0.0	0.1 0.0
EWK6 EWK7 EWK14 J038 J047 C245	99.2 99.0 99.2 99.5 99.3 98.3	0.2 0.0 0.0 0.0 0.0 0.2	0.1 0.2 0.3 0.2 0.3 0.7	0.2 0.3 0.2 0.1 0.1 0.3	0.1 0.1 0.1 0.1 0.0	0.2 0.2 0.2 0.1 0.1 0.3	0.0 0.0 0.0 0.0 0.0	0.0 0.0 0.0 0.0 0.1
Mean Std	98.3 99.0 0.5	0.2 0.1 0.1	0.7 0.3 0.2	0.3 0.2 0.1	0.1 0.1 0.0	0.3 0.2 0.1	- 0.0 0.0	0.1 0.0 0.0

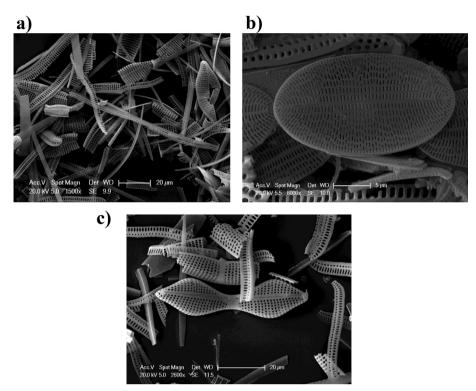


Fig. 3. SEM pictures of purified samples (a) sample EWK7, (b) sample EWK12, (c) sample EWK3 showing that the diatom frustules are devoid of organic remains and were not subject to diagenesis (e.g. clay formation).

range of average values retained for the inter-laboratory calibration (BFC = 29.0  $\pm$  0.3%; PS1772-8 = 42.8  $\pm$  0.8%; MSG60 = 37.0  $\pm$  0.8%). Replicate analysis of the diatom samples gave a reproducibility better than  $\pm$ 0.2% (1 $\sigma$ ).

## 4.2. The isotopic composition of ostracods ( $\delta^{18}O_{carbonates}$ )

About sixty ostracod valves were hand-picked from each sample, using a stereo microscope. For each sample, 1–4 replicates were available for isotopic analyses. Each ostracod valve was cleaned with distilled water and briefly agitated in an ultra-sonic bath to remove detrital particles. The  $\delta^{18}$ O determination was performed by analyzing the CO<sub>2</sub> resulting from the valve carbonate reaction with the H<sub>3</sub>PO<sub>4</sub> acid at 70 °C, with a Finnigan Delta Advantage mass spectrometer. Stable isotopic ratios were expressed in standard  $\delta$ -notation relative to the V-PDB standard. Analytical precision of  $\delta^{18}O_{carbonates}$  was better than ±0.05‰ based on repeated analyses of NBS-19 limestone standard.

Most of the specimen observed were *Lymnocythere* and we kept a single species *L. inopinata* from which vital effect is estimated at  $\delta_{ev} = 0.78 \pm 0.20\%$ , calculated with calcite–water equation of Friedman and O'Neil (1977) (von Grafenstein et al., 1999).

It is worth noting that our temporal resolution (~300 years, only with diatoms) is comparable with other  $\delta^{18}O_{diatoms}$  paleoreconstruction studies. The added value of our work is to go further by proposing a  $\delta^{18}O_{lake}$  profile based on  $\delta^{18}O_{diatoms}$  constrained by  $\delta^{18}O_{carbonates}$ .

#### 5. Results

## 5.1. $\delta^{18}O_{diatoms}$ and $\delta^{18}O_{carbonates}$ values

Mean and reproducibility (SD) of  $\delta^{18}O_{\text{diatoms}}$  values are presented in Table 2 for the 16 diatom samples.  $\delta^{18}O_{\text{diatoms}}$  values range from 23.29‰ to 34.57‰. Mean and reproducibility (SD) are

presented for the 5 ostracod samples in Table 3.  $\delta^{18}O_{carbonates}$  values range from -1.76 to -0.84% vs PDB, and,  $\delta^{18}O_{lake}$  values, reconstructed with ostracods, range from -3.63 to -4.54% vs V-SMOW. For comparison,  $\delta^{18}O_{carbonates}$  measured from tufas and ostracods from Blard et al. (2011) are added in Table 3.

## 5.2. Estimation of the isotopic composition of Lake Tauca ( $\delta^{18}O_{lake}$ )

Calculation of  $\delta^{18}O_{lake}$  from  $\delta^{18}O_{diatoms}$  values requires the selection of (i) a lake water temperature (T<sub>lake</sub>) and (ii) an equation of isotopic fractionation between water and diatoms.

#### 5.2.1. Temperature of Lake, *T*<sub>lake</sub> (18.5–14.5 ka)

Blard et al. (2011) used a gradient of ~+5 °C between the mean temperature of Lake Titicaca (Northern Altiplano) and the cooler local atmosphere, as measured by Richerson et al. (1977) on present-day period. We assume here a similar and constant temperature gradient between the lake and the atmosphere from 18.5 to 11.7 ka although this gradient was probably greater due to lower atmospheric temperature inducing less evaporation (Clausius--Clapeyron law). Note that the atmospheric mean annual temperatures measured on Southern Altiplano on present-day period is ~9 °C (Condom, 2002; Blard et al., 2009). Moreover, glacier equilibrium reconstructions indicate that the atmospheric temperatures were lower by about ~6.5 °C during the Tauca highstands (Blard et al., 2009). Then, local annual mean atmosphere temperature is fixed at ~2.5 °C (i.e 9-6.5 °C) and the Lake Tauca temperature is fixed in this study at 7.5  $^{\circ}$ C (~5  $^{\circ}$ C warmer than atmospheric temperature). We performed Tauca paleolake temperature sensitivity tests (in Fig. 4 and Table 2, a ±2.5 °C uncertainty in lake temperature is considered) showing that the main  $\delta^{18}O_{lake}$  variations cannot be due to the lake temperature variations: differences are less than 1.5% when lake temperature varies between 5 and 10 °C.

#### Table 2

Oxygen isotopic composition of diatom samples ( $\delta^{18}O_{diatoms}$ ): (a) sample names; (b) altitude of the studied site; (c) calibrated ages (ka) using IntCal09; (d) Mean, standard deviation (SD) and number of replicates (*n*) of  $\delta^{18}O_{diatoms}$  values; (e)  $\delta^{18}O_{lake}$  calculated using diatoms-water fractionation from Brandriss et al. (1998); (f)  $\delta^{18}O_{lake}$  calculated using diatoms-water fractionation from Brandriss et al. (1998); (f)  $\delta^{18}O_{lake}$  calculated using corrected  $\Delta_{diatoms-lake water}$  vs Temperature calibration modified from Crespin et al. (2010) and Alexandre et al. (2012); (h)  $\delta^{18}O_{lake}$  calculated using diatoms-water fractionation from Dodd and Sharp (2010); (i)  $\delta^{18}O_{lake}$  calculated using quartz–water fractionation from Matsuhisa et al. (1979).

$\frac{a}{Samples}  \frac{b}{Altitude}  \frac{c}{Calibrated} \\ (m a.s.l)  age ka (\pm 2\sigma)$	c	d	d e		f	g	h	i		
	$\delta^{18}O_{diatoms}$ (‰ vs V-SMOW)			$\delta^{18}O_{lake}$ (‰ vs	$\delta^{18}O_{lake}$ (‰ vs	$\delta^{18}O_{lake}$ (‰ vs	$\delta^{18}O_{lake}$ (‰ vs	$\delta^{18}O_{lake}$ (‰ vs		
	(m a.s.l)	age ka (±2σ)	Mean	SD	n	V-SMOW) after Brandriss et al. (1998)	V-SMOW) after Moschen et al. (2005).	V-SMOW) after Crespin et al. (2010)	V-SMOW) after Dodd and Sharp (2010)	V-SMOW) after Matsuhisa et al. (1979)
BT08 BT12	3657 3657	$11.7 \pm 0.1$ $12.6 \pm 0.1$	33.90 30.51	0.10 0.27	2 2	-1.87 -5.25	-1.01 -4.39	0.66 -2.73	-1.13 -4.51	-5.40 -8.78
EWK2	3685	14.2 ± 0.4	33.14	0.21	3	-2.62	-1.76	-0.10	-1.88	-6.15
EWK3	3685	$14.7 \pm 0.5$	31.80	0.01	2	-3.96	-3.11	-1.44	-3.23	-7.49
C69	3685	$15.2 \pm 0.5$	30.59	0.51	2	-5.17	-4.31	-2.65	-4.43	-8.70
C83	3685	$15.4 \pm 0.5$	23.29	0.05	2	-12.48	-11.62	-9.95	-11.74	-16.01
EWK6	3685	$15.6 \pm 0.8$	23.34	0.20	2	-12.42	-11.56	-9.90	-11.68	-15.95
EWK7	3685	$15.8 \pm 0.8$	33.22	0.27	3	-2.55	-1.69	-0.02	-1.81	-6.08
EWK9	3685	$16.3 \pm 0.6$	33.66	0.43	3	-2.10	-1.24	0.42	-1.36	-5.63
EWK12	3685	$16.8 \pm 0.2$	29.07	0.03	2	-6.69	-5.83	-4.17	-5.95	-10.22
EWK14	3685	$16.9 \pm 0.2$	33.91	0.02	2	-1.85	-0.99	0.67	-1.11	-5.38
J038	3657	$17.2 \pm 0.4$	34.57	0.31	4	-1.20	-0.34	1.33	-0.46	-4.73
C245	3685	$17.7 \pm 0.3$	31.53	0.17	2	-4.23	-3.38	-1.71	-3.50	-7.76
J047	3657	$18.1 \pm 0.3$	33.69	0.15	2	-2.07	-1.21	0.45	-1.33	-5.60
C248	3685	$18.2 \pm 0.4$	27.24	0.19	2	-8.52	-7.67	-6.00	-7.79	-12.05
C272	3685	$18.5 \pm 0.1$	24.80	0.19	2	-10.96	-10.11	-8.44	-10.23	-14.50

5.2.2. Equation of isotopic fractionation between diatoms and lake water ( $\delta^{18}O_{diatoms} - \delta^{18}O_{lake} = \mathbf{a} \cdot T_{lake} + \mathbf{b}$ )

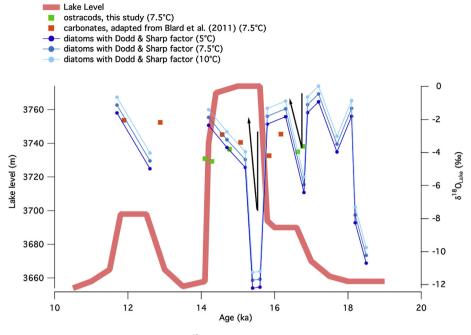
As diatom silica precipitates in isotopic equilibrium with lake water (Leng and Barker, 2006), a thermo-dependent relationship can be expressed as  $[\delta^{18}O_{diatoms} - \delta^{18}O_{lake}]$  (‰ vs V-SMOW) =  $\mathbf{a} \cdot T_{lake}$  (°C) +  $\mathbf{b}$ . According to previous calibration studies (Brandriss et al., 1998; Moschen et al., 2005; Crespin et al., 2010; Dodd and Sharp, 2010; Alexandre et al., 2012; Dodd et al., 2012),  $\mathbf{a}$  is comprised between -0.2 and -0.3%/°C, when  $\mathbf{b}$  strongly differs from one study to another, probably due to discrepancies in methodological approaches (Crespin et al., 2010; Alexandre et al., 2012) or to early post-mortem diagenesis at the sediment–water interface (Dodd and Sharp, 2010; Dodd et al., 2012). For this reason, Dodd and Sharp (2010) and Dodd et al. (2012) suggested the use of thermo-dependent relationships measured for quartz rather than the ones measured for modern diatoms, for interpreting fossil diatom  $\delta^{18}$ O records.

When estimated using the quartz-water relationship (Matsuhisa et al., 1979),  $\delta^{18}O_{lake}$  values were more <sup>18</sup>O-depleted by 4‰ than when estimated using the diatom-water relationship (Brandriss et al., 1998; Moschen et al., 2005; Crespin et al., 2010 corrected in Alexandre et al., 2012; Dodd and Sharp, 2010) (Table 2). Indeed, the results show highly depleted values estimated from quartz-water fractionation of about -8.94‰ at 7.5 °C compared with those calculated with diatoms-water fractionation (average = -4.39%) (Table 2). We used carbonate-based  $\delta^{18}O_{lake}$ reconstruction between 16.8 and 14.2 ka BP (n = 10, see Table 3) representing independent constraint on our diatom-based  $\delta^{18}O_{lake}$ estimates. When compared with diatom-based  $\delta^{18}O_{lake}$  estimates, carbonate-based values (Table 3) differed on average by 0.49% only. On the basis of this comparison in  $\delta^{18}O_{lake}$  reconstruction, the diatom-based calibration proposed by Moschen et al. (2005) and Dodd and Sharp (2010) give similar results with low bias and RMSE compared with the ostracods (respectively, +0.31% and +0.19%

## Table 3

Oxygen isotopic composition of ostracod samples ( $\delta^{18}O_{ostracods}$ ): (a) sample names; (b) calibrated ages (ka) using IntCalO9; (c) Mean, standard deviation (SD) and number of replicates (*n*) of  $\delta^{18}O_{ostracods}$ ; (d)  $\delta^{18}O_{lake}$  calculated using the carbonate–water equation of Friedman and O'Neil (1977) and (e) nature of carbonates. Data from Blard et al. (2011) measured on 5 tufas and 1 ostracod (unknown vital effect) are included and recalculated with the carbonate–water equation of Friedman and O'Neil (1977) for consistency with our measurements. The carbonate–water fractionation is calculated considering a temperature of 7.5 ± 2.5 °C.

a	b	с			$\frac{d}{\delta^{18}O_{lake}}(\text{\% vs V-SMOW})$	e Nature of carbonates	
Samples	Calibrated age ka $(\pm 2\sigma)$	$\delta^{18}O_{carbonates}$	(‰ vs-PDB)				
		Mean	SD	n			
This study							
EWK1	$14.1 \pm 0.2$	-1.6	0.04	3	-4.38	Ostracods	
EWK2	$14.2 \pm 0.4$	-1.76	0.10	2	-4.54	Ostracods	
EWK3	$14.7 \pm 0.5$	-1.01	0.39	4	-3.80	Ostracods	
EWK10	$16.6 \pm 0.4$	-1.17	0.13	4	-3.96	Ostracods	
EWK11	$16.77 \pm 0.4$	-0.84	-	1	-3.63	Ostracods	
Data from Blar	rd et al. (2011)						
Jah-14b	$11.9 \pm 0.2$	-0.04	0.00	3	-2.05	Tufas	
Bo-93.26	$12.8 \pm 0.2$	-0.17	0.01	7	-2.18	Tufas	
Jah-14a	$14.6 \pm 0.4$	-0.91	0.01	4	-2.92	Tufas	
Hua-44	15.1 ± 0.5	-1.38	0.02	6	-3.39	Tufas	
Piv-Ostra	15.9 ± 0.6	-2.19	0.01	1	-4.20	Ostracods	
Jah-12b	$16.2 \pm 0.5$	-0.88	0.01	2	-2.89	Tufas	



**Fig. 4.** Lake level (m) along with the oxygen isotopic composition of the lake  $\delta^{18}O_{\text{lake}}$  (% vs V-SMOW) derived (1) from diatoms (blue curves) using Dodd and Sharp (2010) diatom-water thermo-dependent relationship for T = 5 °C, T = 7.5 °C and T = 10 °C, and (2) from carbonates for T = 7.5 °C (green squares for this study and orange squares for Blard et al., 2011).

for mean bias; 2.66‰ and 2.60‰ for RMSE). Then, the reconstruction proposed in this paper is based on the calibration of Dodd and Sharp (2010).

#### 5.3. Potential caveats

Before discussing the reconstruction of  $\delta^{18}O_{lake}$  presented in the next section, several potential caveats have to be mentioned:

- (1) We implicitly assume a spatial isotopic homogeneity m-deep, throughout the lake (~100 maximal extension > 400 km), which is improbable. We do not know whether Lake Tauca was stratified or mixed. Moreover, we could suppose that during the early stage of the lake transgression, small and disconnected basins may have existed. Nevertheless, our reconstruction relies on a sample series located at different depths and distances from the lake center, suggesting that a potential spatial heterogeneity is partly captured (Fig. 1). For instance, the same planktonic diatoms dominated samples EWK3 and C83 (Sylvestre, 1997), evidencing that the same species could record a large range of isotopic values at different time periods and sites. Finally, our diatom samples are located along a North-South extent and a North-South gradient in amount of precipitation could induce a small isotopic effect (although our samples are located in a less than 1°-latitude band).
- (2) We assume a constant lake temperature during Tauca and Coipasa phases that is obviously erroneous but has only a small influence on results (see Section 5.2.1). Thus, the isotopic shift of ~10‰ between 15.4 and 15 ka is not an artifact due to water lake temperature variation during this period. For the same reasons, a seasonality or temperature stratification of the lake water is conceivable but has probably little effect on interpretations of isotopic variations.
- (3) Here we use the corrected diatoms-water equations deduced from modern calibrations conducted in fresh-water

temperate lakes (Dodd et al., 2012). Although Dodd and Sharp (2010) suggest that  $\delta^{18}O_{\text{fossil diatoms}}$  values are closer to the quartz-water fractionation at (or below) the sediment interface, so far no experimental calibration has been proposed to test this hypothesis. However, even if a postmortem maturation affects the silica skeleton of diatoms, it has been observed that this maturation occurred in less than 0.5 yrs (Dodd et al., 2012), suggesting that diatoms imprint the bottom lake water conditions. This implies that fossil diatom  $\delta^{18}$ O values should directly record the isotopic composition of lake water, at constant temperature, especially in the case of deep lakes. This is potentially a good explanation of our best fit between our  $\delta^{18}O_{lake}$  values calculated from diatoms and those deduced independently from ostracods, because these organisms live in the bottom sediments of the lake. Moreover, contrary to  $\delta^{18}O_{lake}$  reconstruction using diatoms, the ostracods paleotemperature equations are barely dependent on possible bias, even to vital effects (Leng and Barker, 2006). Then diatoms could record the past lake bottom conditions which are usually more stable than the surface, especially in the case of a deep lake, as was Lake Tauca (~100 m deep).

## 6. Evolution of $\delta^{18}O_{lake}$ during the last termination

#### 6.1. Response of Lake Tauca to climate variability

 $\delta^{18}O_{lake}$  reconstruction from 18.5 to 11.7 ka is shown in Fig. 4. It expands on previous results from Placzek et al. (2006) and Blard et al. (2011). The  $\delta^{18}O_{lake}$  exhibits large and rapid variations from ~0 to  $-12\infty$ .  $\delta^{18}O_{lake}$  is ~-10% at the beginning of the sequence at 18.5 ka corresponding with the lake transgression characterized by tychoplanktonic taxa indicators of variable hydrological conditions. This transgression is followed by an abrupt increase by ~9‰ that last less than 200 years until 18.1 ka. Then  $\delta^{18}O_{lake}$  oscillates between -6% and 0% until 15.8 ka. The more <sup>18</sup>O-enriched

composition (ca 0‰) during this phase is recorded during the initial rise of the lake level at around 17.2 ka. At 16.9 ka, a  $\delta^{18}O_{lake}$  depletion of 5‰ is followed by a  $\delta^{18}O_{lake}$  enrichment of 4‰ until 15.8 ka. Then, an exceptional  $\delta^{18}O_{lake}$  drop of 10‰ occurs probably in less than 200 years (see Table 2 column c, for EWK6 and EWK7 samples) during the main lacustrine transgression around 15.8 ka. This abrupt drop is followed by a sudden increase of ~7‰ from 15.4 ka to 14.8 ka (see black arrows on Fig. 4). This particular 2-step  $\delta^{18}O_{lake}$  variation ( $\delta^{18}O_{lake}$  depletion followed by a  $\delta^{18}O_{lake}$  reenrichment) is the so-called "remarkable isotopic feature" studied in the following sections. Note also that the two abrupt isotopic drops at 16.9 ka and 15.8 ka are concomitant with the two successive fillings of the lake during its transgression.

The abrupt  $\delta^{18}O_{lake}$  decrease during lake filling can easily be explained by a regional increase in precipitation (Dansgaard, 1964; Ramirez et al., 2003; Vimeux et al., 2005; Risi et al., 2010). Moreover, this feature appears particularly robust as diatom samples exhibiting this  $\delta^{18}O_{lake}$  drop are of high purity (>99% of silica – see Table 1 for EWK6 and EWK7 samples). However,  $\delta^{18}O_{lake}$  increases measured during stable lake level phases are more surprising observations. This  $\delta^{18}O_{lake}$  re-enrichment has been also recorded at similar timescales in pluvial paleo-lake Bonneville during filling phases (see Figure 7 of McGee et al., 2012). One explanation is related to the lake evaporation and the associated isotopic fractionation leading to an enrichment of the isotopic composition of the lake. According to climate archives in the Andes such as ice cores (at Illimani and Huascaran, Ramirez et al., 2003; Thompson et al., 1995) or speleothems (Kanner et al., 2012; Mosblech et al., 2012), the isotopic composition of precipitation in this region was depleted during the last termination compared to present-day values (estimated around a -14‰ annual average on the Altiplano (IAEA, 2004)). Thus, according to our reconstruction, the lake may have undergone strong evaporation over most of the studied period (Fig. 4).

In order to test the potential impact of evaporation on  $\delta^{18}O_{lake}$ , a simple hydro-isotopic model is developed in the next section. This model is applied to the Tauca phase onset (around 15.8 ka) for which a number of parameters have been constrained in previous studies (Thompson et al., 1998; Condom, 2002; Coudrain et al., 2002; Condom et al., 2004; Vimeux et al., 2005; Blard et al., 2009; this study).

# 6.2. Impact of lake evaporation on $\delta^{18}O_{lake}$ during the Tauca phase?

#### 6.2.1. The hydro-isotopic model

An iterative (annual step) hydro-isotopic model is developed accounting for the Tauca watershed system. It does not take into account external inputs such as groundwater and runoff (see Section 6.3 for further discussions). The model is based on the Tauca watershed hydrological (equation (1)) and isotopic (equation (2)) balances as:

$$\frac{dV_{Lake}}{dt} = (P(t) - E(t)) \cdot S \tag{1}$$

$$\frac{d(\delta_{Lake} \cdot V_{Lake})}{dt} = \left(\delta_p \cdot P(t) - \delta_e \cdot E(t)\right) \cdot S \tag{2}$$

where S is the total surface of the Tauca watershed (lake plus soil): 138.875 km<sup>2</sup> (Blard et al., 2011), P(t) and E(t) are the evolution of precipitation and the total evaporation over the Tauca watershed respectively, dV<sub>Lake</sub>/dt is the volume variation of the lake as defined by Condom et al. (2004) as a function of the lake level,  $\delta_{lake}(t)$  is the isotopic composition of the lake,  $\delta_p(t)$  and  $\delta_e(t)$  are the isotopic composition of incoming precipitation and evaporative flux respectively. The latter is calculated using the (Craig and Gordon, 1965) equation:

$$\delta_{e}(t) = \frac{\delta_{lake}(t) - h(t) \cdot \delta_{atm}(t) + \varepsilon_{v-l}(t) + \varepsilon_{diff}(t)}{1 - h(t)}$$
(3)

where h(t) is the relative humidity normalized to temperature at the lake water surface,  $\delta_{atm}(t)$  is the isotopic composition of the atmosphere above the lake (which is supposed to be in isotopic equilibrium with  $\delta_p(t)$  in our study);  $\epsilon_{v-l}$  (<0) and  $\epsilon_{diff}$  (<0) are the equilibrium and diffusion (or kinetic) fractionation parameters defined as:

$$\varepsilon_{\nu-l} = \alpha_{\nu-l} - 1 = \frac{1}{\alpha_{l-\nu}} - 1 \tag{4}$$

with 
$$\alpha_{l-\nu} = \exp\left(\frac{1.137}{T^2} \cdot 1000 - \frac{0.4156}{T} - 0.0020667\right)$$
  
=  $\frac{\delta_l + 1000}{\delta_{\nu} + 1000}$  (5)

where  $\alpha_{l-\nu}$  is the oxygen isotopic equilibrium fractionation factor between liquid and vapor for oxygen 18 (>1) (Majoube, 1971) (T in °K). We consider atmospheric temperature constant at 2.5 °C (see Section 5.2.1).

$$\varepsilon_{diff} = (1 - h(t)) \cdot \left(1 - \left(\frac{D}{D_i}\right)^n\right) \tag{6}$$

where D and D<sub>i</sub> are the molecular diffusivities of  $H_2^{16}O$  and  $H_2^{18}O$  respectively with a ratio of D/D<sub>i</sub> = 1/0.9723 (Merlivat, 1978), n is a turbulence parameter linked to the surface roughness which is estimated to be 0.5 for mean turbulent flow for a large lake (Gonfiantini, 1986).

At each step, lake isotopic variations are computed and  $\delta_{lake}(t)$  is deduced from Equation (2) as:

$$\delta_{lake}(t) = \delta_{lake}(t-1) + \left(\frac{d\delta_{lake}}{dt}\right)_{t-1} \cdot \Delta t \tag{7}$$

Finally, we can write  $\delta_{lake}(t)$  from equations (1)–(3) and (7) as:

$$\frac{d\delta_{lake}}{dt} + \frac{\delta_{lake}}{\tau} = \frac{A+B}{\tau}$$
(8)

with 
$$\tau = \frac{V_{lake}(t)}{\frac{dV_{lake}}{dt} + \frac{E(t)}{1-h(t)}}; A = \frac{\delta_p(t) \cdot P(t) \cdot S}{\frac{dV_{lake}}{dt} + \frac{E(t)}{1-h(t)}};$$
  
$$B = \frac{h(t) \cdot \delta_{atm}(t) - \varepsilon_{v-l} - \varepsilon_{diff}(t)}{\frac{dV_{lake}}{dt} + \frac{E(t)}{1-h(t)}} \cdot \frac{E(t)}{1-h(t)}$$

During the stable phase (dV<sub>lake</sub>/dt = 0, dS<sub>lake</sub>/dt = 0), the second term (A + B)/ $\tau$  of equation (8) is constant and  $\delta_{lake}$  takes an exponential shape with a time constant  $\tau = ((1 - h)V_{lake}/E)$ . The  $\delta_{lake}$  could be considered invariant after about  $5\tau$ .

Our model is applied during the Tauca phase onset ( $t_0 = 0$  year, around 15.8 ka) until the lake level stabilization at  $t_{Tauca} = 100$  years ( $dV_{lake}/dt$  and  $dS_{lake}/dt = 0$ ). As a first exploration, we use a stabilization time of 100 years which is the lowest estimate given by Condom (2002), and less than the age difference between EWK6 and EWK7 samples (~200 years) which defines the abrupt  $\delta^{18}O_{lake}$  fall. h(t) is supposed to increase from 0.4 to 0.6 during the lake filling. A linear increase in lake altitude from 3650 to 3770 m is defined. P(t) increases linearly from P<sub>0</sub> = 400 mm/year (probably higher than present-day value (300 mm/year) due to wetter conditions) to

 $P_{Tauca} = 600 \text{ mm/year}$  as suggested by Blard et al. (2011). We deduce the  $\delta_p(t)$  variation from P(t) variation according to the isotopicprecipitation gradient of -0.02%/(mm/year) (Rozanski et al., 1993) and considering that  $\delta_p$  at  $t_0$  is -20% (see section 7).

#### 6.2.2. Model results

Model results are shown in Fig. 5 as a function of time. We clearly see that our model is able to reproduce the 2-step  $\delta^{18}O_{lake}$  variation (so-called "remarkable isotopic feature") exhibited in our observations:

- (1) as the lake level increases, the simulated  $\delta^{18}O_{lake}$  decreases, in line with the expected influence of precipitation increase as aforementioned. Moreover, quantitatively, the  $\delta^{18}O_{lake}$ drop is ~17‰ which is consistent with our observations at 15.8 ka, albeit overestimated (~10‰ in data).
- (2) at  $t_{Tauca}$ , a rebound of  $\delta^{18}O_{lake}$  is simulated. The temporal constant  $\tau$  is of ~25 years, leading to a constant  $\delta^{18}O_{lake}$  after 100 years, consistent with the timing of the abrupt increases observed in our reconstruction at ~15.4 ka. This isotopic lake re-enrichment is also simulated by our model (~4‰) albeit slightly lower than our observations (~7‰).

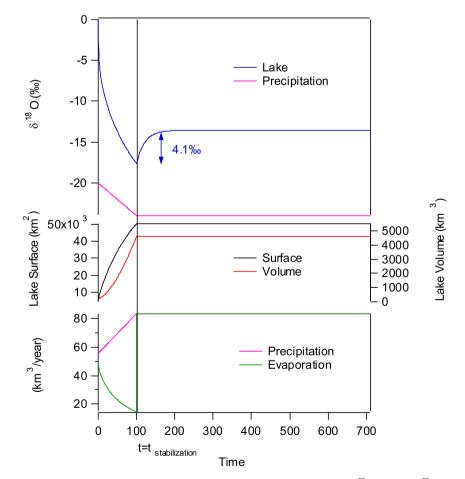
To test the validity of the model results, we performed sensitivity tests with tunable h(t) and P(t) evolutions,  $\delta_p$  at  $t_0$  and  $t_{Tauca}$  (Table 4). The most sensitive parameter is  $t_{Tauca}$ : the more  $t_{Tauca}$  decreases, the more  $\delta^{18}O_{lake}$  is enriched, up to 16‰ when  $t_{Tauca} = 50$  years.

An important assumption in our model deals with the isotopic equilibrium between  $\delta_{atm}(t)$  and  $\delta_p(t)$ . To check the validity of the model results, we reduce the  $\delta_{atm}(t) - \delta_p(t)$  difference by enriching  $\delta_{atm}(t)$  (this mimics a possible realistic change of  $\delta_{atm}(t)$  when lake evaporates). This leads to a more pronounced enrichment in  $\delta^{18}O_{lake}$  more than doubling when  $\delta_{atm}(t)$  tends to  $\delta_p(t)$ .

Thus, we are able to reproduce the  $\delta^{18}O_{lake}$  variations during Lake Tauca filling phase and the following stable phase, considering only the hydrological and isotopic balances of the Tauca watershed and the associated isotopic fractionation. The main reason for this remarkable isotopic feature lies in the competition between evaporation and precipitation: while precipitation increases, the reduction, albeit the inhibition, of evaporation before lake stabilization leads to the depletion of the lake's isotopic composition. When precipitation becomes constant, evaporation is no longer restricted and tends to enrich the lake's isotopic composition. Our model is able to reproduce the  $\delta^{18}O_{lake}$  amplitude increase at 15.4 ka only when Tauca filling time is reduced to around 50 years, compared to previous estimates (see Table 4). The next section explores whether a fraction of the  $\delta^{18}O_{lake}$  enrichment could be also explained by the Titicaca inputs.

# 6.3. Potential role of the Desaguadero River in lake isotopic balance during the Tauca episode

Hydrologic sources from Lake Titicaca during the Tauca episode are controversial: among related studies, some suggest a flow rate of the Desaguadero River between 3 and 30 times higher than its



**Fig. 5.** Hydro-isotopic model results with the parameters mentioned in Table 4 and in Section 6.2.1. From top to bottom:  $\delta^{18}$ O of the lake,  $\delta^{18}$ O<sub>lake</sub> (blue) and the precipitation (pink) in %; Surface (km<sup>2</sup>) and Volume (km<sup>3</sup>) of the lake; Precipitation P(t) and Evaporation E(t) in the Tauca watershed (km<sup>3</sup>/year).

#### Table 4

Sensitivity tests performed on the main parameters of the simple hydro-isotopical model. Each parameter (a, b) is modified around its default value (c) by  $\star \pm 20\%$  (d) to evaluate the  $\delta^{18}O_{lake}$  increase during stable level phase (e). Only one parameter is modified for each sensitivity test.  $\delta^{18}O_{lake}$  increase is ~4.1‰ with the aforementioned default parameterization.  $t_{Tauca}$  is 100 years with tested range coherent with (Condom, 2002). Relative humidity at T = 0 is fixed at 0.4 (and increases to 0.6 at  $t_{Tauca}$ ) but has only a minor influence on  $\delta^{18}O_{lake}$ .

Parameter a	Characterization b	Default value c	Range of change d	Literature constraints e	<sup>18</sup> O increase during stable level phase f
h <sub>Tauca</sub>	Humidity after t <sub>Tauca</sub>	0.6	[0.4; 0.8]	_	[0.2; 8.4] ‰
P(0)	Precipitation before Tauca phase	400 mm/yr	[300; 500]	-	[4.0; 4.3] ‰
P <sub>Tauca</sub>	Precipitation after Tauca phase	600 mm/yr	[400; 800]	(Blard et al., 2009)	[1.8; 8.7] ‰
δ <sub>PO</sub>	<sup>18</sup> O composition before Tauca phase	-20‰	[-15; -25]	(Thompson et al., 1998)	4.1‰
α <sub>P</sub>	Local amount effect	-0.02‰/mm.yr	[-0.06; 0]	(Vimeux et al., 2005)	[1.8; 5.2] ‰
S <sub>TOT</sub>	Tauca catchment area	138,875 km <sup>2</sup>	[100,000; 170,000]	(Blard et al., 2009); (Condom, 2002)	[2.8; 7.6] ‰
t <sub>Tauca</sub>	Time of Tauca filling phase	100 years	[50; 300]	This study; (Condom, 2002)	[0.7; 15.7] ‰

actual flow (Cross et al., 2001; Coudrain et al., 2002; Grove et al., 2003) although a recent study suggests minimal input from Lake Titicaca (Placzek et al., 2011). Moreover, the available water amount provided by surrounding glaciers would have been insufficient to substantially fill Lake Tauca (Blodgett et al., 1997; Condom, 2002) since their maximal extension was reached during the Tauca phase (Blard et al., 2009; Zech et al., 2011). Thus, the inputs from Lake Titicaca and from surrounding glaciers for which no isotopic constraints are known were not taken into account in the first step of our model. We tend nonetheless in this section to explore the possible impact of overflowing via the Desaguadero River. We question the role of the Desaguadero River in enriching the paleolake Tauca and in increasing the simulated isotopic re-enrichment (i.e "extra re-enrichment") during the Tauca stable phase. For this, we add a term for input from Desaguadero Q(t) to our model with its corresponding isotopic composition  $\delta^{18}O_Q$ , without changing the total input budget I(t) (600 mm/year, see Table 4):

$$\frac{dV_{Lake}}{dt} = (I(t) - E(t)) \cdot S \tag{9}$$

$$\frac{d(\delta_{Lake} \cdot V_{Lake})}{dt} = \left(\delta_p \cdot P(t) + \delta_Q \cdot Q(t) - \delta_e \cdot E(t)\right) \cdot S \tag{10}$$

$$I(t) = P(t) + Q(t) \tag{11}$$

Two extreme scenarios of Desaguadero flow evolution during the Tauca phase are considered:

- (1) An "Abrupt scenario": Q(t) is negligible *versus* precipitation input, P(t), during Tauca filling phase (t < t<sub>Tauca</sub>) but equal to  $Q_{Tauca}$  (km<sup>3</sup>/yr) when t = t<sub>Tauca</sub>.
- (2) A "Progressive scenario": Q(t) linearly increases from  $t_0$  to  $t_{Tauca}$  from 0 to  $Q_{Tauca}$  (km<sup>3</sup>/yr) then is equal to  $Q_{Tauca}$  after  $t_{Tauca}$ .

We consider (1) a range of Desaguadero contribution varying from 0% to 70% of the total input budget – which roughly corresponds to interval given by above-cited literature – and (2) a constant Desaguadero isotopic value during the stabilization phase. We test  $\delta^{18}O_0$  values from –20 to 0 ‰.

While scenario 2 exhibits an extra re-enrichment varying from a maximum value of 0‰ (with  $Q_{Tauca} = 0\% \cdot I_{Tauca}$ ) to minimum one of -1.9% (with  $Q_{Tauca} = 70\% \cdot I_{Tauca}$  and  $\delta^{18}O_Q = 0\%$ ), scenario 1 displays an extra re-enrichment from 0‰ (with  $Q_{Tauca} = 0\% \cdot I_{Tauca}$ ) to +6.7% (with  $Q_{Tauca} = 70\% \cdot I_{Tauca}$  and  $\delta^{18}O_Q = 0\%$ ).

In conclusion, the progressive scenario is unable to explain an extra re-enrichment: it contributes to slightly lower the lake reenrichment. On the contrary, with the abrupt scenario, the Desaguadero River could have substantially contributed to the isotopic lake re-increase from 0 to 6.7‰. In this case, the Desaguadero input could complete the enrichment simulated by our simple hydro-isotopic model (with  $t_{stabilization} = 100$  years) and then explain the  $\delta^{18}O_{lake}$  7‰-re-enrichment in our observations at 15.4 ka. Thus, in this case and assuming highest Titicaca outflow estimates, the Desaguadero input could complete the enrichment simulated by our simple hydro-isotopic model (with  $t_{stabilization} = 100$  years) and then explain the  $\delta^{18}O_{lake}$  7‰-re-enrichment in our observations at 15.4 ka. In all the other cases, Desaguadero impact on Tauca isotopic balance is almost negligible.

# 7. Role of Lake Tauca on the isotopic composition of precipitation on Andean summits: case of the Sajama

In this section, we explore to what extent the disappearance of Lake Tauca played a role in the isotopic records from Sajama. Indeed, several studies pointed out the intense convection (e.g. Blodgett et al., 1997) and the water recycling (e.g. Blard et al., 2009) above the lake during Tauca phase that could have provided moisture, eventually further advected towards Sajama summit. These processes are common among pluvial lakes and are likely to contribute substantially to maintain these lakes (Hostetler et al., 1994; Laabs et al., 2009; Mcgee et al., 2012).

Based on the new chronological constraints on Lake Tauca regression (Blard et al., 2011) and accounting for dating uncertainties on ice cores, the abrupt  $\delta^{18}$ O isotopic excursion of +7% recorded in the Sajama ice core between 15 and 14 ka now appears synchronous with the regression of Lake Tauca (see Fig. 2). We question the potential contribution of the lake evaporation when it disappears as a moisture source for precipitation at Sajama. We examine whether the enrichment at Sajama could be explained by moisture supply from paleolake Tauca.

Our discussion will only be based on the isotopic balance. We assume that the signal of the isotopic composition of incoming precipitation on the Altiplano at ~14 ka is  $\delta^{18}O_p=-20\%$  (contrary to  $\delta^{18}O_p = -14\%$  at Sajama) (see Fig. 2). This estimate corresponds to a  $\delta^{18}O_p$  depletion of about 4‰ compared with the modern  $\delta^{18}O_p$ at Illimani. The latter can be estimated by the average value of  $\delta^{18}$ O in Illimani ice core over the last 100 years (Vimeux, 2009). This depletion of 4‰ between 14 ka and present-day is of the same order than the one we observe at Huascaran (this is also the value of  $\delta^{18}$ O in Illimani ice core at ~14 ka if we consider that the dating from Ramirez et al., 2003 is correct). Assuming condensation at isotopic equilibrium at T =  $2.5 \degree C$  (estimated atmospheric mean temperature, see section 5.2.1), the isotopic composition of water vapor from which precipitation forms is -31% (*versus* -25% at Sajama) and will be considered as the isotopic composition of the background atmosphere.

We first consider an extreme situation (although unrealistic) corresponding to a complete and instantaneous lake evaporation. In this case, no isotopic fractionation occurs between the lake water and the atmosphere and, considering that  $\delta^{18}O_{lake}$  is about 5‰ at the lake highest level (Fig. 4), the water vapor input in the atmosphere would have an isotopic composition of  $\delta^{18}O_v = -5\%$ . Consequently, based solely on the isotopic balance, the relative contribution of advected vapor versus vapor originating from the lake (x) to precipitation at Sajama can be estimated as -25% = (1 - x)(-31%) + (x)(-5%). Thus, the lake contribution to Sajama precipitation versus advected water vapor would be 23%. It is worth noting that this calculation is sensitive to  $\delta^{18}O_{atm}$  and varies of about +3%/per mil when  $\delta^{18}O_{atm}$  varies between -25and -35‰.

We now consider a progressive evaporation of Lake Tauca. We use the evaporative model from Gonfiantini (1986) derived from Craig and Gordon (1965). The evolution of the lake's isotopic composition, according to the fraction of remaining water in the lake (f, the lake evaporates as f tends to 0), is given by:

$$\frac{d\delta_{lake}}{d\ln f} = \frac{h \cdot (\delta_{lake} - \delta_{atm}) - (\delta_{lake} + 1) \cdot \left(-\varepsilon_{v-l} - \varepsilon_{diff}\right)}{1 - h - \varepsilon_{diff}}$$
(12)

As an initial exploration, we consider constant conditions for lake evaporation (h = 0.6,  $\delta^{18}O_{atm} = -31$ ‰, T = 2.5 °C). A simple integration of equation (12) leads to:

$$\delta_{lake}(f) = \left(\delta_0 - \frac{A}{B}\right) \cdot f^B + \frac{A}{B}$$
(13)

where:  $A = \frac{h \cdot \delta_{atm} - \varepsilon_{diff} - \varepsilon_{v-l}}{1 - h - \varepsilon_{diff}}; \quad B = \frac{h + \varepsilon_{diff} + \varepsilon_{v-l}}{1 - h - \varepsilon_{diff}};$  and the initial  $\delta^{18}O_{lake}$  is  $\delta_0 = -5\%$ .

Using equation (13), we can deduce the evolution of  $\delta^{18}O_{lake}$  as a function of f, as well as the evolution of the isotopic composition of evaporated water (using equation (3)). In that case,  $\delta_e$  varies from -8.75% (f = 1) to -3.32% (f = 0) and the lake contribution to Sajama precipitation decreases from 27 (f = 1) to 22% (f = 0). It is worth noting that this result is highly sensitive to h and to a lesser extent to  $\delta^{18}O_{atm}$ . We run the model with varying h and  $\delta^{18}O_{atm}$ . Simulations are shown on Fig. 6a and b. Considering all simulations, the extra enrichment of 5‰ at Sajama can be explained with a water vapor input from lake evaporation varying from 10 to 60% (see lake contribution on Fig. 6a and b when f = 0).

We now consider varying conditions during lake evaporation for h and  $\delta^{18}\text{O}_{atm}$  . Indeed, the continuous evaporation of the lake modifies those two parameters as the evaporation occurs. We numerically solve equation (12) by successive iterations (we keep a constant temperature of 2.5 °C and the initial  $\delta^{18}O_{atm}$  is of -31‰.). We consider that h decreases from 0.6 to 0.4. In equation (3),  $\delta^{18}O_{atm}$  is the isotopic composition of the atmosphere in contact with the lake during the lake transgression. We account for the influence of the lake evaporation on  $\delta^{18}O_{atm}$  at each iteration (j) so that  $\delta^{18}O_{atm}$  at the step (j) is calculated as a mixed value of  $\delta_e$  and  $\delta^{18}O_{atm}$  at the previous step (j - 1):  $\delta^{18}O_{atm}(j) = n^*\delta_e(j - 1) + 1$  $(1 - n)\delta^{18}O_{atm}(j - 1)$ . We decide that n is related to f as  $n = (1 - x_f)$ \*f: we account here for the fact that when f = 1 there is no lake contribution and when f = 0 there is a maximal mixing between  $\delta_e$ and  $\delta^{18}O_{atm}$  which is represented by  $x_{f}$ . Fig. 6c shows the lake contribution for different x<sub>f</sub>. We clearly see that the isotopic excursion at Sajama can be explained by a significant water vapor input from the lake evaporation at the beginning of lake disappearance (from almost 50%) which tends to decrease to 0 when the lake disappears as expected.

To conclude, we find in each simulation that around 70% of the abrupt  $\delta^{18}$ O isotopic excursion of +7% recorded in the Sajama ice core between 15 and 14 ka can be explained by the disappearance of Lake Tauca, with a lake contribution varying from a few percent to 60%.

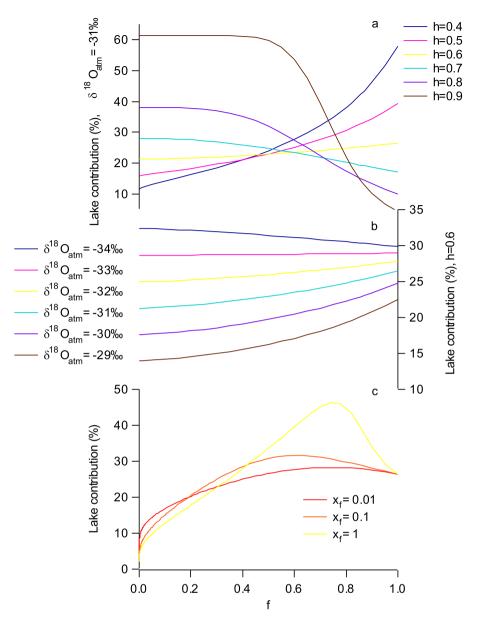
As aforementioned, our calculations are only based on the isotopic balance and do not account for the water availability between advected moisture and lake evaporation. We try here to estimate whether these contributions are realistic. A lake volume of 4600 km<sup>3</sup> corresponds to  $P_{Tauca} = 83.3 \text{ km}^3/\text{year} (0.6 \text{ m/year}^* 138,875 \text{ km}^2) (\text{see})$ Fig. 5). The full disappearance of the lake in 300 years gives a lake water volume availability of 15.3 km<sup>3</sup>/year (4600 km<sup>3</sup>/300 years). We now have to estimate the reduction of PTauca during lake disappearance. For a reduction of 2 (resp. 3) the evaporated moisture versus advected precipitation in volume, and thus the maximum lake contribution to Sajama precipitation, is 37% (= 15.3/(83.3/2)) (resp. 55% = 15.3/(83.3/3)). Considering the range of P<sub>Tauca</sub> given in Table 3 and a reduction of P<sub>Tauca</sub> that can attain a factor 3, we estimate that the maximum lake contribution in terms of water volume versus advected water could vary between 27 and 82%, which is higher than our aforementioned estimates. Consequently, we estimate that the water volume evaporated above the lake can be enough to substantially contribute to the isotopic excursion in the Sajama ice core signal.

#### 8. Conclusion

The Bolivian Altiplano is a unique case for exploring the impact of lakes on surrounding continental climate archives due to the proximity of paleolakes and ice caps. In this paper, we propose for the first time a quantitative reconstruction of the oxygen 18 isotopic variations ( $\delta^{18}$ O) of the Bolivian Altiplano paleolakes from 18.5 to 11.7 ka based on the isotopic composition of fossil diatoms and ostracods. Considering some assumptions (constant lake temperature, choice of the diatoms-water calibration, spatial homogeneity of the lake isotopic composition), a robust feature is evidenced in the evolution of  $\delta^{18}O_{lake}$ . An abrupt decrease of  $\delta^{18}O_{lake}$  during lake fillings is immediately followed by an increase of  $\delta^{18} O_{lake}$  during stable lake level phases. The highest variation occurs at 15.8 ka with a  $\delta^{18}O_{lake}$  decrease of about 10% concomitant with the Lake Tauca transgression, followed by a  $\delta^{18}O_{lake}$  increase of similar amplitude four centuries later, in pace with the lake highstand stabilization.

With a simple hydro-isotopic modeling approach, it is possible to satisfactorily reproduce the 2-step ("decrease-increase")  $\delta^{18}O_{lake}$  variation exhibited in our data. Moreover, this study reveals that the renewed increases of  $\delta^{18}O_{lake}$  after filling phases can be partly explained by the prior reduction of evaporative flux due to high rainfall and a consecutive re-equilibration of the isotopic fluxes in lake steady-state. This explanation assumes a rapid (less than a few centuries) set-up of high rainfall regime. Our model integrates consistent and available constraints on humidity, precipitation, isotopic compositions and geomorphological parameters from the literature and our sensitivity tests show its robustness. However, it remains a simple modeling approach that does not take into account, for example, the seasonality of precipitation, runoff and groundwater, and assumes linear variations of main variables.

The  $\delta^{18}O_{lake}$  reconstruction enables us to explore whether the 'paleolakes' presence have influenced the isotopic composition of precipitation deposited at Sajama. We conclude that the total or partial evaporation of the lake during the Tauca phase regression could explain the distinct isotopic excursion at Sajama. We show that a contribution of the lake evaporation to precipitation at Sajama from a few percent to 60% during the lake disappearance is sufficient to explain the 5‰ extra enrichment and is consistent with available water volume. Consequently, perturbation of local hydrological cycles by lacustrine basins could, at certain periods,



**Fig. 6.** Evolution of the contribution of lake evaporation (in %) to Sajama precipitation as a function of the remaining fraction (f) of water in the lake (when lake has disappeared, f = 0) for (a) varying relative humidity normalized to temperature at the lake water surface, h, with isotopic composition of the atmosphere above the lake  $\delta^{18}O_{atm} = -31\%$  (isotopic equilibrium at  $T = 2.5 \degree C$  with  $\delta^{18}O_p = -20\%$ ); (b) varying  $\delta^{18}O_{atm}$  with h = 0.6 and (c) accounting for the influence of lake evaporation on  $\delta^{18}O_{atm}$  and h, and the mixing (reflected by  $x_f$ , fraction describing mixing between evaporation flux above the lake and water vapor above the lake, see text) between water vapor above the lake and water vapor from evaporation.

substantially affect the interpretation of signals from surrounding records of isotopic composition of precipitation (e.g. ice core or speleothems). Moreover, according to the new geochronological constraints and recent studies on the calibration of the water stable isotopes proxies, these elements could reconcile here the presence of a positive isotopic excursion in the Sajama isotopic record, interpreted as a dry period, consistent with the disappearance of Lake Tauca.

To prove a potential causal link between lake evaporation and this isotopic excursion, a regional modeling approach using a highresolution climate model with water stable isotopes and a campaign for isotopic atmospheric measurements above tropical lakes would be useful for future discussions.

Finally, we neglect here the potential influence of postdepositional effects. Actually, drier conditions over the Altiplano might have led to modify the isotopic composition of snow after deposition (Hardy et al., 1998, 2003; Vuille et al., 1998). Drier conditions could enhance sublimation processes. If sublimation alone is not expected to change the isotopic composition of the ice, condensation of water vapor formed into the firn can lead to an isotopic enrichment.

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