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Pre-hispanic wetland irrigation and metallurgy in the South Andean Altiplano (Intersalar Region, Bolivia, XIVth and XVth century CE)

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ABSTRACT

The high-altitude Andean Altiplano has been subject to abrupt climate changes during the Holocene. The resulting impact on the hydrological cycle has obliged ancient societies to adapt and develop strategies to face droughts and sustain agropastoral activities. Here, we present the results of archaeological prospections together with the biogeochemical characterization of a well-dated core collected at Saitoco wetland in the arid southern Altiplano (Intersalar Region, Bolivia). Archaeological survey allowed the mapping of a network of channels collecting water from the surrounding mountains to the wetland, and the presence of small copper mines and metallurgical installations. Traces and major element concentrations and accumulation rates, together with elemental and isotopic characterization of organic matter (OM) (i.e., C_{org} and $\delta^{13}C_{org}$) in the wetland core have been used to document the evolution of the landscape in relation to climatic and anthropogenic pressure. While OM, bromine (Br), selenium (Se) and mercury (Hg) data were used to assess the variability of precipitations during the Late Holocene (4.2 ka BP to the present), local and regional mining activities have been reconstructed through the variations of lead (Pb), copper (Cu), and antimony (Sb). Our results show an abrupt change in OM composition during the 14th and 15th century CE, characterized by an abrupt shift in $\delta^{13}C_{org}$ synchronous with a rise in Br, Se and Hg, testifying for the waterlogging of the wetland during a known arid period in the region. This change is attributed to anthropogenic transformation of the landscape through the irrigation of the wetland by channeling streams from the surrounding mountains. At the same time, a mining pollution signal was recorded supporting local Cu mining and metallurgy. The mining signal then reached its maximum values during the Inca and Colonial periods, which matches with reported enhanced mining activities in the region. From the colonial era onward, the wetland progressively dried up, likely resulting from the abandonment of the site. Through the combination of a biogeochemical record and archaeological prospections, this study provides evidences that societies of the arid Intersalar region have transformed their landscape and developed wetland irrigation and mining during the arid 14th and 15th century CE Period.

1. Introduction

Holocene climate in the high-altitude tropical Andes has been

marked by abrupt changes that have impacted hydrological cycles (Baker et al., 2001). Such changes have also affected the development of ancient societies and their occupation of the land (Bruno et al., 2021;

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Guédron et al., 2023; Vella et al., 2024). Following a wet period during the 6th century, Tiwanaku became a major archaic state of the Altiplano until ca. 1000 CE (Janusek, 2004; Marsh et al., 2023). During the following epoch, i.e., from ca. 1000 to 1450 CE, regional paleoclimatic studies suggest a general dry period (Guédron et al., 2023; Thompson et al., 2013), during which major population migrated from the Andean highlands (i.e., shores of Lake Titicaca) to settle in fortified hilltop settlements named Pukaras (Arkush, 2009). While some authors attributed this behavior to a context of widespread conflicts resulting from the collapse of Wari and Tiwanaku polities (Arkush, 2008), other suggest that some reasons are of a symbolic nature (Housse and Mouquet, 2023). In the arid south-central Altiplano region (Salar Uyuni), archaeological studies have shown evidence of intense agricultural activities associated with quinoa cultivation between 1200 and 1450 CE, which has been defined as the Late Regional Development Period [i.e., LRDP (Cruz et al., 2017)]. Many of these sites were then abandoned during the Inca invasion (Bouysse-Cassagne et al., 1992). From the middle of the Inca occupation period (ca. 1450 to ca. 1570 CE) until the end of the Colonial era (ca. 1570–1825 CE), the Altiplano was marked by increased precipitation (Apaéstegui et al., 2018; Delaere and Guédron, 2022; Guédron et al., 2023; Thompson et al., 2013; Weide et al., 2017). Despite this climate improvement, the Intersalar region (i.e., in the southern Altiplano between Salars Coipasa and Uyuni) became considerably depopulated from the Inca domination period onward, perhaps as a result of population transfer to agricultural areas and mining centers (De Galvez, 1977; de Morales, 1977; Espinoza Soriano, 1981; Wachtel, 1980). Before the arrival of the Spaniards (i.e., ca 1535 in the Altiplano), Altiplano's pre-Columbian societies already had developed sophisticated mining and metallurgical knowledge since at least three millennia (Lechtman, 2014; Lechtman and Macfarlane, 2005; Schultze et al., 2009). Although generally performed at small scales, these activities have released significant amounts of metal such as lead (Pb), copper (Cu), mercury (Hg), and antimony (Sb), recorded in numerous environmental archives (i.e., lake and peat sediment, ice cores) of the central Andes (Abbott and Wolfe, 2003; Cooke, 2006; Cooke et al., 2007, 2008b; Eichler et al., 2017; Guédron et al., 2019, 2021). Major regional mining and metallurgical signals have been attributed first to the Tiwanaku period, followed by an increase in intensity culminating during the Spaniards mining period centered on silver (Ag) extraction in the region of Potosi (Cooke et al., 2013; Hylander and Meili, 2003a).

Although reconstructions of paleoclimate and mining activities are relatively numerous in the northern part of the Altiplano, they are scarce in the arid southern part due to restricted presence of reliable environmental archives such as lake sediment. Documenting this region is however essential to understand how populations were able to adapt and develop an economy in hostile territories during an arid era that followed the decline of the great Andean societies (Tiwanaku/Wari). Several archaeological studies have highlighted the construction of hydraulic infrastructure to improve water management in such arid context (Lane et al., 2022; Vella et al., 2024). These studies have, however, emphasized the difficulty of dating these infrastructures, and the value of paleoenvironmental records in overcoming these problems.

In such arid region, Andean wetlands, also called *bofedales* or *humedales* (Monge-Salazar et al., 2022), appear as a useful alternative environmental archives to sediment records to reconstruct historical variations of moisture and human occupation (Domic et al., 2018; Villarreal et al., 2014). As peatlands, their convex shape and organic rich composition allow the trapping of elements originating from the atmosphere, which can be used as proxies of climate change and anthropogenic activities (e.g., mining and agropastoralism). While elements like as silver (Ag), lead (Pb), copper (Cu), antimony (Sb), and mercury (Hg) were extensively used in the northern Altiplano to reconstruct mining activities (see references herein), elements such as bromine (Br), selenium (Se) as well as Hg can be good candidates for paleoclimate reconstructions (Guédron et al., 2018; Jiskra et al., 2022; Martinez-Cortizas et al., 2007). These elements are emitted as gases or

particles to the atmosphere and then deposited on the land mostly via wet deposition (precipitation). In pristine environments, Se, Br, and Hg are mainly released from the ocean through biogeochemical processes (biosynthesis of volatile organic forms, photo or bio-reduction), sea-salt aerosols, and in a lesser extent from volcanoes (Amouroux et al., 2001; Eggenkamp, 2014; Jiskra et al., 2021; Le Bras et al., 2023). While Hg is considered conservative in organic rich soil and sediment (Cossa et al., 2021; Guédron et al., 2013), a study documenting historical reconstructions of halogens have indicated that Br is not always conservative in peatbogs as its concentration is linked to peat decomposition processes (Biester et al., 2004). In parallel, two studies showed evidence of uptake and accumulation of inorganic Br by plants (Xu et al., 2004) followed by biogeochemical transformations to stable organobromine species during plant decomposition leading to Br accumulation in soils (Leri and Myneni, 2012). For Se, there is no study assessing in details if it is a conservative element in wetland archives, but Se is well-known to be largely and strongly bound to OM leading to its accumulation in soils and wetland (Sharma et al., 2015; Tolu et al., 2022; Winkel, 2016).

In this study, archaeological survey was combined with the study of a core collected in the Saitoco wetland, located north of the Salar Uyuni. To reconstruct changes in local and regional moisture as well as mining activities, we combined geochemical analysis of trace (Br, Hg, Se, Pb, Cu, Sb) and major (Si, Ti, Al, Fe) elements together with organic carbon content and isotopic composition (i.e., C_{org} and $\delta^{13}C_{org}$). Archaeological observations of the surrounding area were then confronted to geochemical data to understand, distinguish and reconstruct natural and anthropogenic signals recorded in the area.

2. Materials and methods

2.1. Site of study and environmental context

The sampling site, i.e., the Saitoco wetland (19.78° S, 67.71° W, 3650m asl) is located in the Andean Altiplano, a high altitude (~3700 m asl) endorheic basin which includes Bolivia, Peru, Argentina and Chile (Fig. 1).

Over the Andean altiplano, aridity increases southward from the temperate shores of Lake Titicaca to the cold deserts of the Salar Uyuni (Bolivia) and Atacama (Chile) regions. Precipitation mainly originates from northeasterly (summer monsoonal) airflows that bring convective precipitation from Amazonia, and in a lesser extent, from southerly (westerly) airflows that bring frontal precipitation from extratropical cyclones, during the winter (Garreaud et al., 2003; Houston and Hartley, 2003; Valdivielso et al., 2022; Vuille, 1999).

The sampled Saitoco wetland is located in the Intersalar region, between the two world's largest salt flats (Salar de Uyuni and Salar de Coipasa, Fig. 1). In this area, the climate is cold and dry up to arid (average precipitation ~ 200 mm per year), with a rainy season centered in December to March. This area is composed of a series of composite volcanic centers, flat lava domes, and extensive ignimbrites (Salisbury et al., 2022), and the Saitoco wetland is found at the foot of the Tunupa Volcano.

Three hundred meters north of the Saitoco wetland, lies the pre-Hispanic site of Saitoco (ca. 2.3 ha), which has been dated to 1292–1392 CE (Cruz et al., 2017). In addition, 1.3 km east of the wetland lies Jach'a Pukara, the largest and most populous pre-Hispanic village in the Intersalar region (3.5 ha), dated to 1294–1394 CE (Cruz et al., 2017). Archaeological studies have shown evidence of relatively dense population in this arid region, together with the organization of an agricultural and pastoral landscape for the subsistence and development of the community (Cruz et al., 2017).

2.2. Archaeological context, prospections and mapping

The occupation of the southern Altiplano has been documented in previous studies (Cruz et al., 2017, 2022). Briefly, the period before

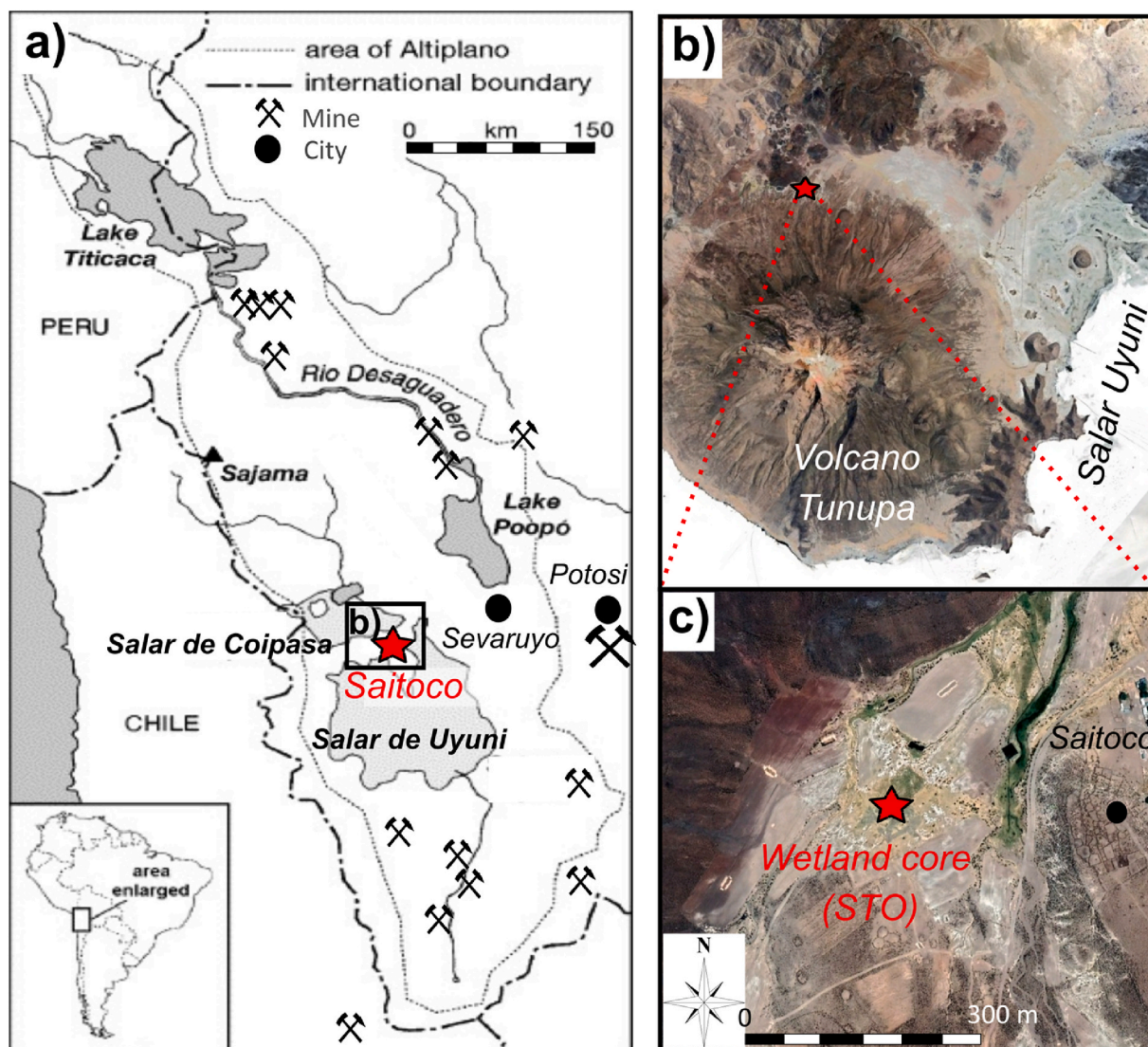


Fig. 1. Location and geographical characteristics of the sampling site. Panel a) shows the location of the Saitoco wetland in a map of the Andean Altiplano that informs on main historical mining and metallurgical centers (adapted from [Chepstow-Lusty et al. \(2005\)](#) and [Macfarlane and Lechtman \(2014\)](#)), and the ancient capital of the Quillacas region (i.e., Tambo de Sevaruyo or Oma Porco) of which the Intersalar was part ([Cruz, 2024](#)). Panel b) is a satellite image locating the Saitoco wetland at the foot of the Tunupa Volcano (google earth image), and Panel c) the location of the core STO in the wetland (Google Earth image).

1200 CE is considered to be a long transition from mobile hunting-gathering and herding communities to more aggregated and proliferating agropastoral settlements. Evidence of pre-Hispanic agrarian societies is mainly related to the Late Regional Development Period (LRDP), i.e. between 1200 and 1450 CE, and the following Inca Period (ca. 1450 to ca. 1570 CE). During the Inca period, precise historical data indicate significant depopulation resulting from displacement of local population by the Incas towards the agricultural areas of Cochabamba ([de Morales, 1977](#); [Espinoza Soriano, 1981](#)). Following the Inca period, the Spanish settlement started around 1570 CE ([Espinoza Soriano, 1981](#)), with the development of small scale mines in the 1580 ([Gil Montero, 2017](#)) that developed in the following decades.

Prospections and surveys of residential, storage, and cropland areas at the Saitoco wetland were conducted from 2007 to 2017. Systematic surveys were carried out by analyzing geographic data from high-resolution satellites, as well as from geomorphology, and topographic maps. A detailed site survey was performed to elaborate i) the typology of archaeological cropland areas and mapping of irrigation canals ([Fig. S2](#)), ii) archaeological surveys and excavation of residential structures, and iii) archaeological surveys of ancient mining and

metallurgical sites. The geographic data used in this study consisted of (i) the land surface topography extracted from the Shuttle Radar Topography Mission (SRTM) v2 Digital Elevation Model (DEM) with 1-arc sec resolution (~30 m) downloaded from the Reverb|ECHO website (<http://reverb.echo.nasa.gov/>), and (ii) high-resolution satellite imagery available on Google Maps [source: GeoEye, DigitalGlobe, CNES (Centre National d'Études Spatiales)/Astrium, and CNES/Airbus]. Archaeological cropland areas were digitized by photo interpretation of satellite images with QGIS editing tools (www.qgis.org/). The detailed data processing for studying the distribution of topographic variables (elevation, slope, and aspect) over cropland areas is described elsewhere ([Cruz et al., 2017](#)).

2.3. Core collection, sampling, pollen and chemical analysis

An 80 cm core (STO) was collected in the downstream part of the Saitoco wetland in 2012 ([Fig. 1](#)). The core was sliced with 1 cm intervals. For the entire core, one sample out of two was dedicated to pollen analysis and the other to geochemical analysis. Pollen was analyzed in sub-samples accounting for a volume of 0.5 cm³ at the ISEM laboratory

in France. A spike of two Lycopodium tablets (36814 *Lycopodium clavatum* spores per tablet) per sample was added to calculate the concentration of pollen. The samples were first treated with 40% HF, 15% HCl and washed in 10% KOH in a hot water bath before density separation with $ZnCl_2$ (Faegri and Iversen, 1989). For each sample dedicated to geochemical analyses, a volume of $\sim 2 \text{ cm}^3$ was weighted before and after freeze-drying to determine dry bulk density (DBD). These sub-samples were then crushed in a planetary mill (Pulverisette 7 premium line model, Fritsch®) to obtain a size smaller than $63 \mu\text{m}$ before chemical (elemental and isotope) analyses.

All details for the sample digestion methods and analytical protocols are provided in Guédron et al. (2021), and quality assurance and quality control (QA/QC) are provided in Table S1. Briefly, total mercury (THg) concentrations were determined by atomic absorption spectrophotometry with an automatic Hg analyzer (Altec, model AMA 254) following a published procedure (Guédron et al., 2009). Concentrations obtained for repeated analyses ($N = 15$) of certified reference materials (CRMs) never exceeded the published range of concentrations for BCR 277 ($128 \pm 17 \text{ ng g}^{-1}$) and ERM-CC-141 ($93 \pm 17 \text{ ng g}^{-1}$). All samples were duplicated providing a residual standard deviation below 5%.

Major (Fe, Al, Ti, Si, S, P) and trace (Se, Pb, Sb, Cu) elements were measured, respectively, by inductively coupled plasma optical spectrometry (ICP-AES, model Varian 720-ES) and inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7900) after an acid digestion (HNO_3 , HF) in Teflon Savillex vessels (Guédron et al., 2006). QA/QC included the digestion and analysis of using the CRM PACS-2 (National Research Council of Canada, Table S1). Bromine (Br) was measured by ICP-MS after acid digestion (HNO_3 , HF) followed by ammonia dilution (He et al., 2018).

Total carbon (TC) and total organic carbon (C_{org}) concentrations as well as their $\delta^{13}C$ signatures were determined using a Cavity Ring-Down Spectrometer (Picarro G-2201-i, Inc.®) coupled with Combustion Module (Costech, Inc.®) (CM-CRDS) using previously reported analytical methods, calibration, and sample preparation before and after decarbonation of sediments to compare total and organic carbon content (Guédron et al., 2019).

Elemental accumulation rates (AR) were calculated by multiplying the elemental concentrations by the sedimentation rate (SR), and the dry bulk density (DBD). Calculations of elemental enrichment factors with respect to a conservative crustal element (Equation (1)) were used to discriminate between the natural and anthropogenic signals of the considered elements.

$$EF(X) = ([X]/[Ti])_{\text{sample}} / ([X]/[Ti])_{\text{background}} \quad (\text{Equation 1})$$

Where, EF corresponds to the enrichment factor of the studied element (X) with respect to a conservative crustal element (Titanium, Ti) that is not supplied by atmospheric deposition. It is calculated by normalizing the concentrations of the studied element X by the concentrations of Ti in the samples and in the local geochemical background taken as the deeper sample of the wetland record (Eichler et al., 2017; Guédron et al., 2021). Hence, the lithogenic contribution for each sample corresponds to the product of the studied element (X) by the ratio of the sample over the background value of the conservative crustal element (Ti) calculated as shown in equation (2).

$$EF(X)_{\text{litho}} = [X] * ([Ti]_{\text{sample}} / [Ti]_{\text{background}}) \quad (\text{Equation 2})$$

Finally, the exogenic contribution (natural and/or anthropogenic contribution) is the difference between the total EF and the lithogenic EF (cf. Equation (3)).

$$(EF(X)_{\text{exo}} = EF(X) - EF(X)_{\text{litho}}) \quad (\text{Equation 3})$$

2.4. X-ray absorption spectroscopy

Br K-edge HERFD-XAS measurements were performed at the beamline BM16 at the European Synchrotron Radiation Facility (ESRF) in Grenoble (France). The beamline was equipped with a Si(220) monochromator, 12 Si(880) crystal analyzers, and a 2D detector Imixpad (Proux et al., 2017). The samples analyzed (i.e., freeze-dried and crushed STO samples) were analyzed together with Br reference compounds diluted in BN to a Br concentration of $2000 \mu\text{g g}^{-1}$. Br reference compounds included an aliphatic bromine compound (Bromoeicosane), an aromatic bromine compound (Bromophenol) as well as sodium bromide (NaBr). All samples were pressed as 5 mm diameter pellet, and introduced in a He cryostat. Spectra were recorded at 15 K to limit radiation damage. Energy calibration was performed with a Pt metallic foil recorded every ten spectra, and setting the inflection point of the edge at 13.273 eV. Br K-edge HERFD-XAS spectroscopy allows a clear distinction of the Br species (aliphatic, aromatic and inorganic Br), as shown in Fig. S1. After energy calibration and normalization of all data, linear combination fits were done to quantify the three Br species (aliphatic, aromatic and inorganic Br), on the energy range $-15; +30 \text{ eV}$ relative to the absorption edge (Table S2).

2.5. Radiocarbon dating and age model

Chronological framework was established with radiocarbon ages obtained from collected organic remains and bulk sediment (Table S3), using Accelerator Mass Spectrometry (AMS) at the Laboratoire de Mesure du Carbone14 (LMC14)-UMS 2572 (CEA/DSM-CNRS-IRD-IRSN-Ministère de la Culture et de la Communication, Saclay, France) and at the radiocarbon facility of the Belgian Royal Institute for Cultural Heritage (RICH). Radiocarbon ages were calibrated to calendar years Before Present (cal. yr BP) using the calibration curve for the Southern Hemisphere SHCal20 (Reimer et al., 2020) and a post-bomb curve (Hua et al., 2013). Age-depth model was established with the clam R package (Blaauw, 2010).

3. Results and discussions

3.1. Archaeological evidences of human occupation at Saitoco

3.1.1. Archaeological evidences of agricultural development at Saitoco

Recent archaeological studies in the region have demonstrated increasing human occupation during the Late Regional Development Period [(LRDP), i.e., 1250–1450 CE (Mendez-Quiros et al., 2023)] and the development of agricultural systems for quinoa cultivation and pastoral activities (Cruz et al., 2017). Such development of agricultural practices in this arid region implied landscape modifications for water-saving practices [i.e., micro-terracing and channeling (Cruz et al., 2017)]. Archaeological prospections at Saitoco have allowed the identification of a large number of ancient agricultural micro-terraces (corals) attesting for pastoral exploitation in the area. It further allowed mapping of a network of anthropogenic canalization in the wetland collecting water from the slopes of Cerro Condor Samaña and volcano Tunupa (Fig. 2 and Fig. S2).

The construction of terracing structures for cultivation on the slopes were developed for slowing down surface water runoff, improving water infiltration, and reducing soil erosion (Cruz et al., 2017). In addition, practices of collecting water to irrigate the wetlands were probably used to improve grazing areas devoted to camelid farming as described in the region (Lane, 2021; Lane et al., 2022; Palacios Ríos, 1977; Verzijl and Quispe, 2013). The set-up of these hydraulic infrastructure is difficult to date, and previous archaeological studies speculated that such channeling has been developed during the dwelling period of Saitoco, Ququisani, and Jach'a Pukara sites (Fig. 6), dated to the 13–15th century CE (Cruz et al., 2017).

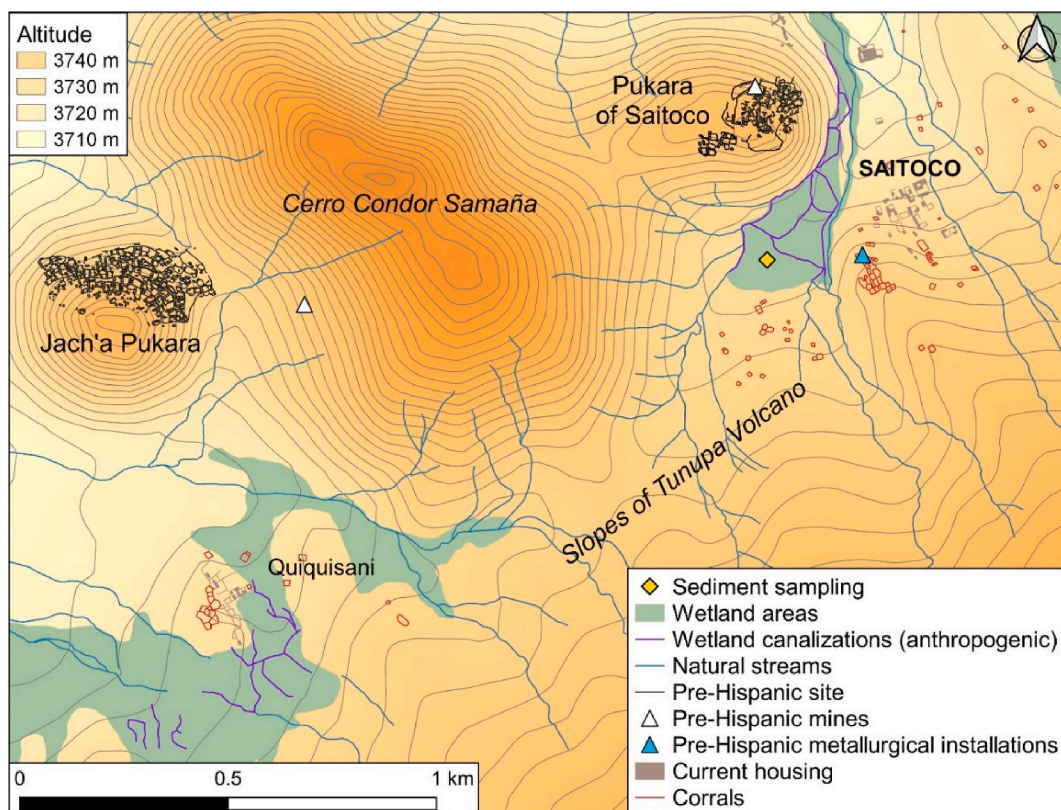


Fig. 2. Archaeological mapping of the pre-Hispanic sites of Saitoco and Jach'a Pukara, indicating natural streams, anthropogenic canals in the wetland area, and identified agricultural micro-terraces (corrals), pre-Hispanic copper mine and metallurgical installations.

3.1.2. Archaeological evidences of early mining and metallurgy at Saitoco

Archaeological prospections at Saitoco have also allowed the identification of ancient mining and metallurgical installations 200 m from the Saitoco wetland site, including a pre-Hispanic copper mine (Fig. 3A), metallurgical installations (Fig. 3C), slag and copper metal droplets (Fig. 3B and D) at Pukara and Jach'a Pukara (Fig. 2).

The copper mine is associated with the Jach'a Pukara site, located less than 200 m away. This latter site has been dated to 1294–1394 CE (95% error range), and the copper metal sample (Fig. 3D) was retrieved in this archaeological context supporting a similar indirect dating for both the mine and the sample (Cruz et al., 2017). Similar pre-Hispanic metallurgical structures have also been recorded in northern Chile during this mining period (Figuera et al., 2018; Mille et al., 2013).

3.2. Saitoco wetland core lithology and chronology

Based on the DBD and the concentrations of minerogenic elements (Si, Ti, Fe and Al) and organic carbon (C_{org}), four main units were identified in the STO core (Fig. 4). From core bottom to top, the three first units (1, 2 and 3) exhibited high DBD ($1.6 \pm 0.4 \text{ g cm}^{-3}$) indicating high content of detrital materials such as quartz and phyllosilicates (Si), as well as Fe, Al and Ti oxides (Fig. 4). A major change occurred at the transition between Units 3 and 4, characterized by a decline of the amount of minerogenic fraction (i.e., drop in DBD, Si, Ti, Fe and Al) to the benefit of an increase of C_{org} , from a baseline of ca. 0.8% up to ca 9.5 % on the core top.

Amongst the 12 radiocarbon ages, 4 were rejected (considered too young or inconsistent with a neighboring age) and 2 were dated post-bomb. Hence, seven radiocarbon ages were kept to elaborate the age depth model (Fig. 4). The resulting chronological framework for the entire core spanned more than four millennia. Sedimentation accumulation rate (SAR) was very low in the 3 bottom units ($0.16 \pm 0.01 \text{ mm. yr}^{-1}$) testifying for low sedimentation between before the first age

(3691–3888 cal BP, 95% error range) and the age at the top of unit 3 (i. e., 706–831 cal BP, 95% error range), followed by a gradual rise from 0.2 to 0.7 mm. yr^{-1} in Unit 4 (i.e., ca 780 cal BP to modern).

3.3. Abrupt change in wetland composition during the 14th and 15th century

From 2000 BCE to ca. 1300 CE, the wetland C_{org} accumulation rates (AR) were low with negative and stable $\delta^{13}C_{org}$ values (ca. -27 ‰ , Fig. 5a), which indicates highly decomposed terrestrial OM in dry conditions (Biester et al., 2004; Guédron et al., 2018). It is worth mentioning that pollen grains were not preserved in the entire core likely due to inappropriate taphonomic condition (i.e., high pH, oxidation) and high ultraviolet rays at this altitude (Andersen, 1986; Torabinejad et al., 1998; Twiddle and Bunting, 2010; Zhang et al., 2014). During the end of the 14th and 15th century [i.e., from calibrated 1393–1486 CE ($\pm 95\%$ error confidence; 2σ) onward], a major ecological and geochemical transition occurred as recorded by an abrupt rise in $\delta^{13}C_{org}$ from -27 to -20 ‰ (Fig. 5a), which indicates a change of the wetland vegetation from terrestrial to aquatic plants (Guédron et al., 2018) and higher water supply and accumulation in the wetland (Alewell et al., 2011). Synchronously to the $\delta^{13}C_{org}$ anomaly, carbonate ($CaCO_3$) content increased from few up to 50 % indicating favorable moisture condition for bacterial $CaCO_3$ precipitation in this alkaline environment (Canakci et al., 2015), whereas carbonates are almost absent from the profile in the previous dry period. FeARs mirrored the profile of $CaCO_3$ supporting the presence of a waterlogged and anoxic wetland that allows iron reduction and thus dissolution and export out of the wetland. The higher $\delta^{13}C_{org}$ lasted until the beginning of the Inca period (ca. 1450 to 1570 CE), and then gradually decreased to reach its baseline in the modern times.

Hence, our biogeochemical results (i.e., decline in $\delta^{13}C_{org}$ and $CaCO_3$) suggest that the wetland became progressively drier during the

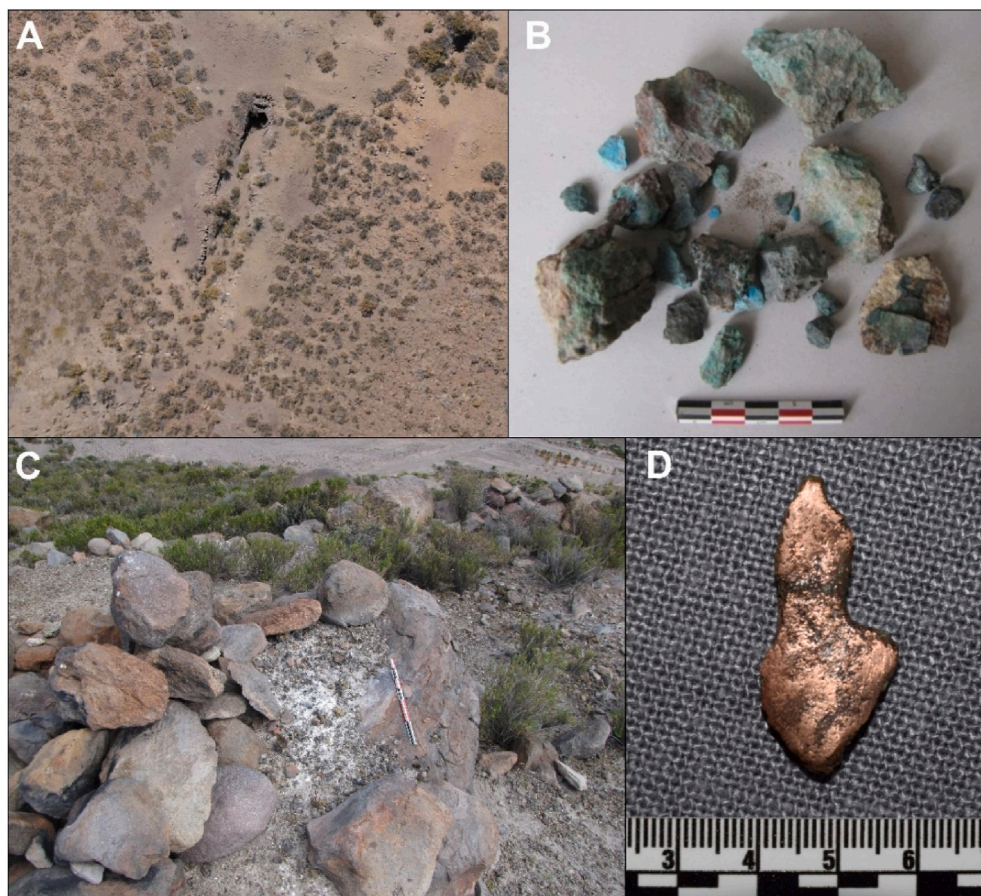


Fig. 3. A) Photograph of an ancient copper mine located 250 m west of the Pre-Hispanic site of Jach'a Pukara. B. Samples of copper ore recovered at the ancient mines of Saitoco. C. pre-Hispanic metallurgical structure (bench) used for copper ores smelting into copper metal located 200 m from the core sampling site. D. Copper metal piece found in a pre-Hispanic metallurgical context at Saitoco.

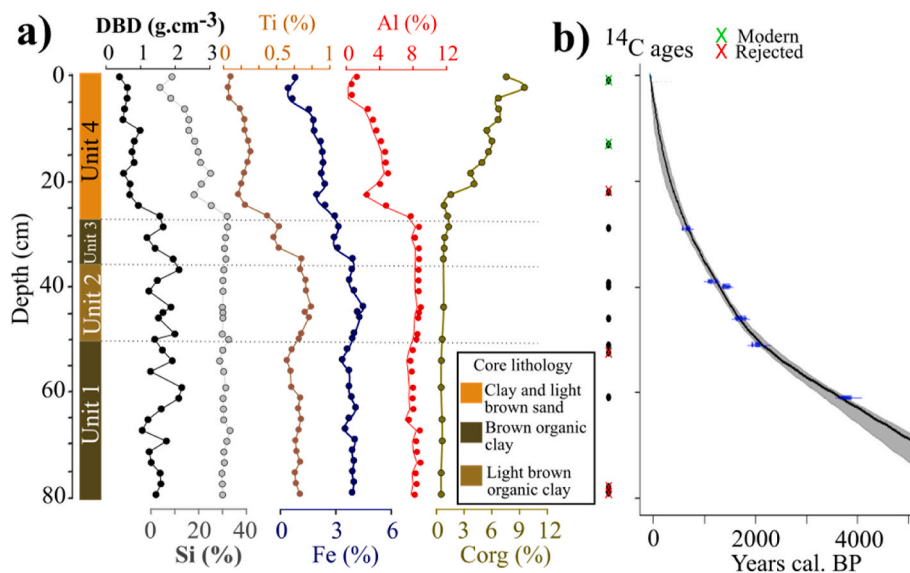


Fig. 4. Core lithology and age depth model. Panel a) shows depth profiles for dry bulk density (DBD), concentrations of minerogenic elements (Si, Ti, Fe and Al), and concentration of organic carbon (Corg), indicating three main unit in sampled Saitoco core. Panel b) gives the obtained age depth model for the sampled core.

Inca and Colonial periods. The last - 2‰ shift of $\delta^{13}\text{C}_{\text{org}}$ after 1850 CE is attributed to the Suess effect, i.e., a decrease of the atmospheric $\delta^{13}\text{C}_{\text{CO}_2}$ values since the beginning of the industrial era due to the combustion of fossil fuels depleted in ^{13}C (Suess, 1953). Finally, within the last past two

centuries, the wetland returns to a likely more productive (i.e., increase in C_{org} AR) and wetter context (increase in CaCO_3 and decline in Fe ARs), but with $\delta^{13}\text{C}_{\text{org}}$ values back to -27 ‰ indicating a predominance of terrestrial plants.

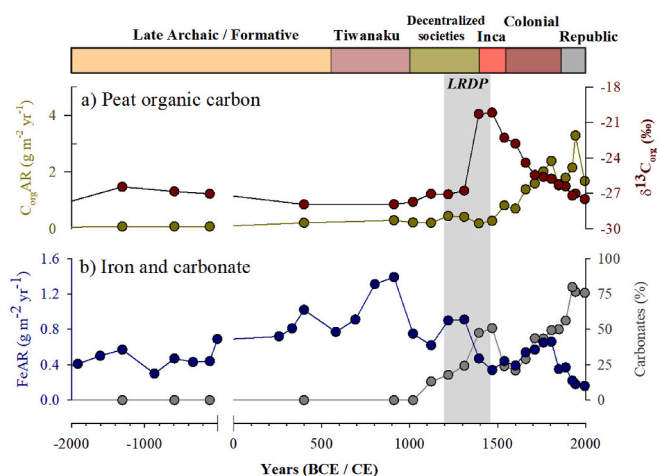


Fig. 5. Major ecological and geochemical transition in the Saitoco wetland. Panel a) shows the organic carbon accumulation rates ($C_{org}AR$) and isotopic composition ($\delta^{13}C_{org}$). Panel b) shows the iron accumulation rates (FeAR) and carbonate content (%). The shaded grey band highlights the Late Regional Development Period (LRDP), which corresponds to human occupation and development of rain fed agriculture in the region (Cruz et al., 2017).

3.4. Evidence of wetland irrigation during the Late Regional Development Period

3.4.1. Bromine, selenium and mercury as proxies of water inputs to the wetland

The accumulation rates (AR) of Br, Se and Hg depicted similar trends across the entire STO record (Fig. 6), and are significantly correlated with r^2 values of 0.43 (BrAR vs SeAR; $p < 0.05$), of 0.68 (BrAR vs HgAR; $p < 0.01$), and of 0.75 (SeAR versus HgAR; $p < 0.01$), supporting similarity in their sources and behavior (accumulation) in the Saitoco wetland. Only Hg showed several differences that can be attributed to anthropogenic contribution.

For Br, we also investigated its speciation in five samples from Unit 4 (i.e., top of the STO core) using K-edge HERFD-XAS to evaluate possible changes in speciation with sediment aging. In the underlying layers (units 1 to 3), Br speciation could not be determined due to Br concentrations being below the detection limits of XAS spectroscopy. The five obtained XAS spectra revealed that Br was mainly found associated with OM as aromatic Br compounds (>80%) with a minor percentage (<15%) of inorganic Br (Fig. 7 and Table S3). Aliphatic Br was not detected in the samples. A similar predominance of aromatic Br was observed in the humus fraction of a forest soil in New Jersey (USA) (Leri and Ravel, 2015).

In the Saitoco record, Br speciation was similar in the five samples from the top unit 4, indicating no change in speciation during OM aging. These results hence suggest that Br accumulates in wetland as aromatic organobromine preserved during OM humification (Leri and Ravel, 2015; Martínez-Cortizas et al., 2016).

Before the LRDP, accumulation rates (AR) and enrichment factors (EF) of Br, Se and Hg remained at their lowest baseline values with the exception of a Hg peak during the Tiwanaku period (i.e., ca. 800 CE) and a very slight increase in SeARs during the Formative and Tiwanaku periods (Fig. 6). This pattern indicates a generally dry Late Holocene before 1200 CE, consistently with regional records from the Salar Coipasa and Uyuni (Baker et al., 2001; Nunnery et al., 2019), from northern Chile Puna (Domic et al., 2018), and from Atacama highlands (Domic et al., 2023; Gayó et al., 2012). To distinguish between natural and anthropogenic sources of Hg, Hg was normalized to Se (Fig. 6e), as this latter is not expected to have any anthropogenic source. Hg:Se ratio shows fine and abrupt peaks during the Tiwanaku, LRDP and colonial/industrial revolution periods, corroborating significant Hg

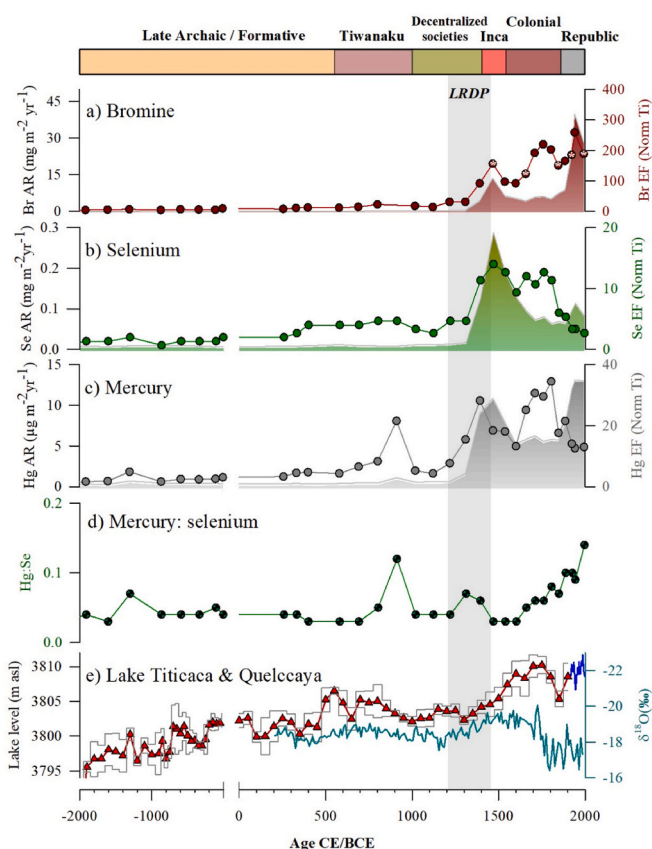


Fig. 6. Reconstruction of paleo-climate trace element proxies in Saitoco and comparison with published reconstructions of precipitations in the Altiplano. Panels a), b) and c) show the accumulation rates (AR) and enrichment factors (EF) of respectively bromine (Br), selenium (Se) and mercury (Hg). Samples used for XAS spectroscopy are indicated with a white star within dots in panel a). Panel d) gives the Hg:Se ratio, which reflects the anthropogenic Hg contribution when the ratio increases above the baseline ratio. Panel e) informs on variations in Lake Titicaca water level (red symbols; Guédron et al., 2023) and $\delta^{18}O$ record (cyan line) of Quelccaya Ice core (Thompson et al., 2013) during the same time period. The vertical shaded grey highlights the Late Regional Development Period (LRDP).

anthropogenic inputs during periods of intense mining activities in the region (Cooke et al., 2008a, 2011; Guédron et al., 2019, 2021). A major increase in Br, Se and Hg appears synchronous with the $\delta^{13}C_{org}$ anomaly during the late LRDP, followed by a decline at the onset of Inca Period. Moderately wetter conditions in the arid Atacama region have been associated with an increase in precipitation during the Medieval Climate Anomaly (i.e., MCA: ca. 850 to 1250 CE) due to the strengthening of the SASM in association with the southward displacement of the ITCZ (Apaéstegui et al., 2018; Gayo et al., 2012; Nester et al., 2007). However, the observed peak in our proxies (Br, Se and Hg) appears later than the MCA event, and is inconsistent with reconstructions performed in the Atacama Desert (Domic et al., 2023; Gayó et al., 2012), NW Argentina (Lupo et al., 2006) as well as regional paleoclimate records (Chepstow-Lusty et al., 2009; Guédron et al., 2023; Jomelli et al., 2009; Morales et al., 2012; Thompson et al., 2013). Finally, the return to elevated values of Br, Se and Hg ARs and EFs around 1700 CE is in line with increased regional precipitation pattern during the last past 300 years which support increased deposition with higher precipitations (Baker and Fritz, 2015; Baker et al., 2009; Guédron et al., 2023; Thompson et al., 2013).

The inconsistency of the 14–15th century anomaly in $\delta^{13}C_{org}$, Br, Se and Hg (which suggests a wet event) with regional records support that it is unlikely to be of climate origin. Hence, our sedimentary record

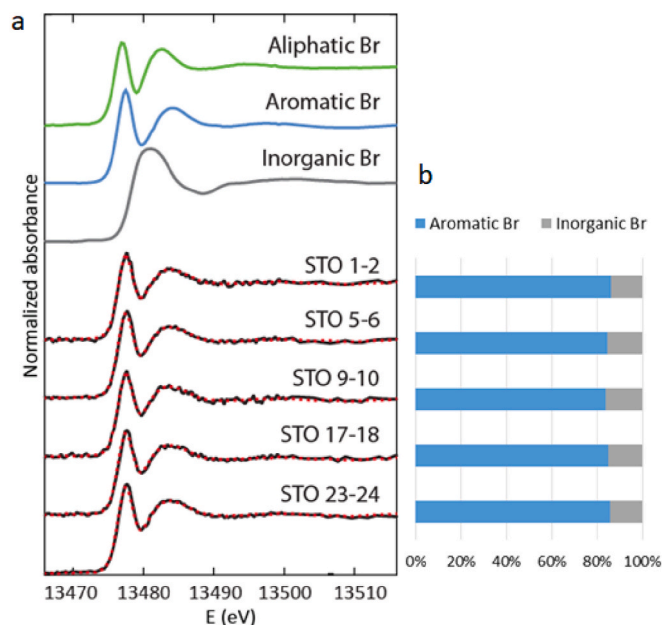


Fig. 7. Bromine (Br) speciation in the Saitoco wetland core. Panel a) shows the Br K-edge HERFD-XAS spectra for selected layers of the core (plain, black spectra, values in cm), and the linear combination fits (LCFs, dashed, red spectra) using Br standards. Three representative Br standards are shown on top, i.e., aliphatic Br (Bromoeicosane), aromatic Br (Bromophenol) and inorganic Br (NaBr). Panel b) gives the proportion of Br species obtained by the LCFs, in % of total Br in each sample. Fits with aliphatic Br instead of aromatic Br were rejected because of poor reconstruction (R factor multiplied by 24).

corroborates the hypothesis of the anthropogenic transformation of the wetland ecosystem through the installation of the channel during the 14th century. Such anthropogenic management of the water to make the natural streams converge to the wetland may have favored the water-logging of the wetland lowlands resulting in vegetation change from a dominance of terrestrial plants to aquatic (i.e., $\delta^{13}\text{C}_{\text{org}}$ anomaly) and in paleoclimate proxies recorded at STO. In addition, the focusing of meltwaters from the slopes of the Tunupa volcano (5321 m asl) into the wetland might be responsible for the observed enrichment in Br, Se and Hg as these elements have been reported in high concentration in snow and ice meltwaters (Alfthan et al., 1995; Jeong et al., 2022; Paudyal et al., 2017; Søndergaard et al., 2015).

From the 15th to the 17th century, the decline in $\delta^{13}\text{C}_{\text{org}}$ values, and in CaCO_3 , Br, Se and Hg contents, which reflect the gradual return of the wetland to its original drier state, suggests the abandonment of irrigation and possibly the site. A regional demographic decline is notably documented in the mid-15th century with the arrival of the Incas and the integration of the territory into their empire (Cruz et al., 2017). During the following Colonial era, historical sources and archaeological studies confirm little occupation of these lands (Cruz et al., 2017). Hence, although higher regional precipitations at that time are reported in the area and over the Altiplano, the probable abandonment of the wetland did not allow sufficient water to maintain it in a waterlogged state.

3.5. Evidences of local and regional mining and metallurgical activities

3.5.1. Geochemical evidences of mining and metallurgical activities

Lead (Pb), copper (Cu), and antimony (Sb) are relevant trace metal proxies to reconstruct historical periods of ore extraction and metal smelting in the Altiplano region (Eichler et al., 2016, 2017; Guédron et al., 2019, 2021).

Because the minerogenic contribution of Pb, Cu and Sb is important in the record (Figs. S3 and S4), results are presented in enrichment factors considering the exogenic contribution of each metal. Exogenic

Pb, Cu and Sb deposition to the wetland are relatively low or moderate (<25%) during the Formative and Tiwanaku Epochs (Fig. 8). The increased anthropogenic Hg deposition identified at Saitoco during the 9th century (i.e., late Tiwanaku epoch, Fig. 6c) can thus be attributed to regional mining activities, such as the Lake Titicaca Basin during a period of increased demand in metals due to the reconstruction of the Tiahuanaco capital (Guédron et al., 2021; Macfarlane and Lechtman, 2014).

The first major significant and synchronous increase of Pb, Cu and Sb EFs occurs after the Tiwanaku period, and is continued by a second major increase during the end of the LRPD, and peaking during the Inca period (Fig. 8). These two epochs have been documented in other parts of the Altiplano as significantly intense metallurgical periods. For the post-Tiwanaku period, Lake Titicaca records showed evidence of intense but discontinuous metallurgical signal (Guédron et al., 2021) attributed to increasing endemic warfare in the Northern Altiplano (Arkush, 2011; Covey, 2012). Although not quantified, archaeological studies report numerous small-scale metallurgical centers over the southern Altiplano (i.e., South Bolivia and Northern Argentina and Chile) including Cu, Ag and Pb mining and smelting (Cruz, 2009; Cruz and Joinville Vacher, 2008; Figueroa et al., 2018; Mille et al., 2013). At Saitoco, the second and concomitant rise in Pb, Cu, Sb (Fig. 8) and Hg:Se (Fig. 6d) during the late LRPD and Inca period overlaps the marked geochemical anomaly attributed to the human occupation of the site. In contrast to the signal recorded for the wetland irrigation period, the metallurgical signal peaks during the Inca period. This suggest that mining and metallurgy reached their maximum activity during the Inca occupation, or that this signal reflect the rise in regional or sub-regional metallurgical activities (Abbott and Wolfe, 2003). Finally, Pb, Cu, and Sb levels remain high during the entire colonial era testifying of the intense regional mining activities reported over the entire Altiplano (Abbott and Wolfe, 2003; Cooke et al., 2008a, 2011; Guédron et al., 2019, 2021).

4. Conclusions

Water management and maintenance of Andean wetlands are well known in modern socio-ecosystem. For past environments, the methodological complexity of dating these ecosystems, which have generally experienced discontinuous functioning, rarely enables reliable

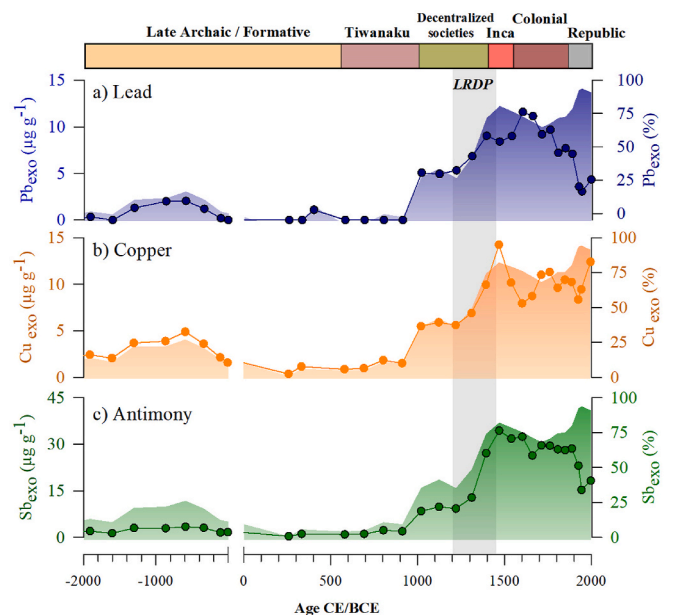


Fig. 8. Calculated exogenic concentrations (circles) and percentage of the total elemental concentration (areas filled) for a) lead (Pb), b) copper (Cu), and c) antimony (Sb).

chronologies to be reconstructed. We here provide solid evidence of water management in Andean wetlands during pre-Hispanic times through the combination of archaeological, paleoenvironmental, and geochemical studies. Our results demonstrate that LRDP societies of the arid Intersalar region have transformed their landscape by creating irrigation channels to supply wetland areas. In parallel, we provide evidence of local copper mining and smelting activities, which probably lasted during the Inca period. A significant outcome of this reconstruction of the anthropogenic signal on the landscape is the assessment of the magnitude and the duration of the human imprint, as well as how the landscape has recovered after the abandonment of the site.

Archaeological studies in the Intersalar region have revealed that the society's economy during the LRDP was based on extensive non-irrigated cultivation of quinoa. Hence, irrigated pastures were probably maintained for the breeding of camelids, which were not only a major source of protein, wool and other materials, but also a fundamental tool for the economy, as it enabled the transport and exchange of surplus production (i.e., quinoa, salt, metals ...). Complementary studies in the intersalar region are thus needed to provide new information on the precise use of these irrigated wetlands, and their sudden abandonment.

CRedit authorship contribution statement

Stéphane Guédron: Conceptualization, Formal analysis, Investigation, Methodology, Resources, Supervision, Validation, Writing – original draft, Writing – review & editing. **Clément Roy:** Formal analysis, Writing – review & editing. **Géraldine Sarret:** Formal analysis, Writing – review & editing. **Julie Tolu:** Resources, Formal analysis, Writing – review & editing. **Marie-Pierre Ledru:** Conceptualization, Formal analysis, Investigation, Methodology, Project administration, Resources, Funding acquisition, Writing – review & editing. **Sylvain Campillo:** Resources, Formal analysis. **Sarah Bureau:** Resources, Formal analysis. **Anne-Lise Develle:** Resources, Formal analysis. **Charline Guiguet-Covex:** Formal analysis. **Eduardo Queiroz Alves:** Formal analysis. **Mathieu Boudin:** Resources, Formal analysis. **Richard Joffre:** Project administration, Formal analysis, Investigation, Writing – review & editing. **Pablo Cruz:** Project administration, Conceptualization, Formal analysis, Investigation, Methodology, Resources, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.quascirev.2024.108826>.

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