

# Geochemistry and $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology of lavas from Tunupa volcano, Bolivia: Implications for plateau volcanism in the central Andean Plateau

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## ABSTRACT

Tunupa volcano is a composite cone in the central Andean arc of South America located ~115 km behind the arc front. We present new geochemical data and  $^{40}\text{Ar}/^{39}\text{Ar}$  age determinations from Tunupa volcano and the nearby Huayrana lavas, and we discuss their petrogenesis within the context of the lithospheric dynamics and orogenic volcanism of the southern Altiplano region (~18.5°S–21°S). The Tunupa edifice was constructed between  $1.55 \pm 0.01$  and  $1.40 \pm 0.04$  Ma, and the lavas exhibit typical subduction signatures with positive large ion lithophile element (LILE) and negative high field strength element (HFSE) anomalies. Relative to composite centers of the frontal arc, the Tunupa lavas are enriched in HFSEs, particularly Nb, Ta, and Ti. Nb-Ta-Ti enrichments are also observed in Pliocene and younger monogenetic lavas in the Altiplano Basin to the east of Tunupa, as well as in rear arc lavas elsewhere on the central Andean Plateau. Nb concentrations show very little variation with silica content or other indices of differentiation at Tunupa and most other central Andean composite centers. We propose that this distinct compositional domain reflects an amphibole- and/or phlogopite-rich mantle lithospheric source. Breakdown of these minerals during lithospheric delamination may provide a melting trigger for Tunupa, as has been suggested for other rear arc plateau lavas of the central Andes, and for plateau regions globally. The ca. 11 Ma Huayrana lavas indicate that this process had begun in the central Altiplano Basin by this time. The enriched Nb-Ta-Ti signature of plateau lavas may be an important indicator of hydrous mineral breakdown within the mantle lithosphere, and it can be detected in lavas that have likely experienced crustal contamination.

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## INTRODUCTION

The central Andean Plateau (Altiplano-Puna) of western South America (14°S–28°S) is the only modern geologic setting on Earth where subduction of an oceanic plate beneath a continent has led to the formation of a major continental plateau (Isacks, 1988), surpassed in elevation and extent only by the Tibetan Plateau (continent-continent collision). This apparent plate-tectonic paradox has been the focus of numerous studies seeking to understand the processes of its formation and evolution (e.g., Allmendinger et al., 1997; Lamb and Davis, 2003; Garziona et al., 2006; Oncken et al., 2006; Barnes and Ehlers, 2009; Faccenna et al., 2013). Whereas active subduction has resulted in frontal arc volcanism along the western edge of central South America for much of the past 200 m.y. (James, 1971; Davidson et al., 1991; Haschke et al., 2002), the sources and processes involved in producing the abundant mid- to late Cenozoic rear arc magmatism, hundreds of kilometers east of the frontal arc, remain poorly understood (Davidson and de Silva, 1992; Kay and Kay, 1993; Trumbull et

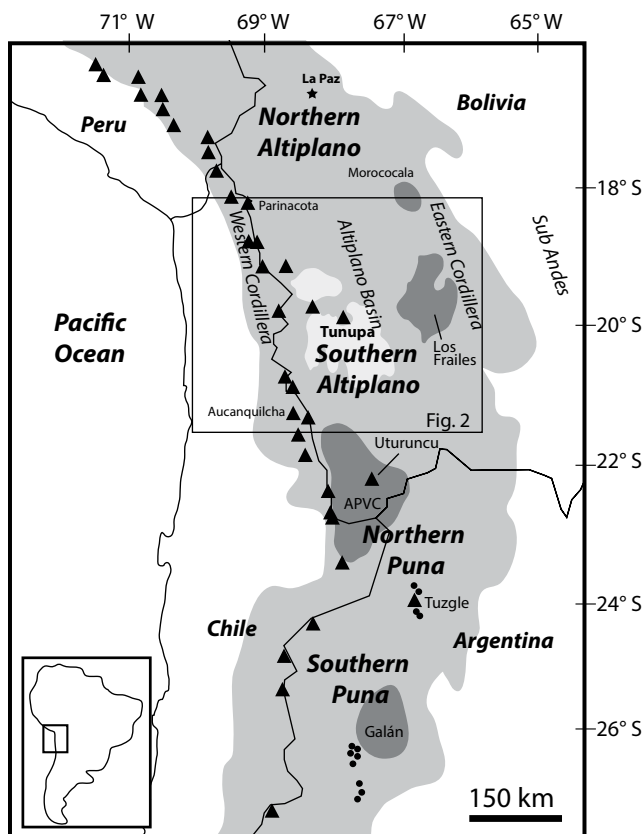
al., 2006; Hoke and Lamb, 2007; Kay and Coira, 2009; Mamani et al., 2010).

Rear arc magmatism in the central Andes may be closely related to the processes of plateau formation, as suggested by the close spatial, and temporal, correlation between crustal thickening and volcanic vent distribution (e.g., Allmendinger et al., 1997; Trumbull et al., 2006). Based largely on seismic studies that suggest a dominantly felsic crustal composition and lower than expected thicknesses of mantle lithosphere in this region of intense crustal shortening (Whitman et al., 1996; Myers et al., 1998; Beck and Zandt, 2002; Yuan et al., 2002; McQuarrie et al., 2005), many researchers have argued for an important role for density-driven removal (delamination) of varying amounts of the mafic lower crust and mantle lithosphere beneath the central Andes. Although a magmatic response to lithospheric removal is generally expected in orogenic zones such as the central Andes (e.g., Kay and Kay, 1991, 1993; Elkins-Tanton, 2005), the petrogenetic sources and melting triggers are a matter of considerable debate (Kay et al., 1994; Davidson and de Silva, 1995; Hoke and Lamb, 2007; Jiménez

and López-Velásquez, 2008; Drew et al., 2009; Kay and Coira, 2009; Ducea et al., 2013). In this study, we present new  $^{40}\text{Ar}/^{39}\text{Ar}$  and whole-rock elemental data for the composite rear-arc Tunupa volcano of the Bolivian Altiplano, and we evaluate these data within the context of regional plateau volcanism. We find that Tunupa and regional rear arc lavas are enigmatically enriched in Nb-Ta-Ti in relation to lavas of the arc front, and we propose that this enrichment is ultimately derived from the breakdown of hydrous minerals (amphibole, phlogopite) within the delaminating mantle lithosphere. Nb-Ta-Ti-rich lavas are also found in the Tibetan Plateau, and we suggest that this geochemical signature may be an important reflection of magmatism associated with plateau formation globally.

## TUNUPA VOLCANO

The Pleistocene Tunupa volcano (19.8°S, 67.6°W) is centrally located within the latitude-defined, southern Altiplano transect (17°S–21°S; Kay and Coira, 2009) of the central Andean Plateau (Figs. 1 and 2). Tunupa is situated near the



**Figure 1. Location map of the central Andes of South America. Quaternary composite volcanoes are shown as black triangles and major mid-Miocene to recent ignimbrite fields are darkly shaded. Dark circles in Argentina are locations of rear arc lavas discussed in the text (Déruelle, 1991; Schreiber and Schwab, 1991; Drew et al., 2009). The >3 km elevations of the central Andean Plateau (Altiplano-Puna) are lightly shaded. The southern Altiplano segment (box, Fig. 2) is defined here as between ~18°S and 21.5°S. APVC—Altiplano-Puna volcanic complex.**

center of the Altiplano Basin of Bolivia, ~115 km east of the Pleistocene frontal arc of the Western Cordillera, ~115 km west of the fold-and-thrust belt of the Eastern Cordillera, and ~175 km above the subducting Nazca plate. Despite its rear arc location, Tunupa shares broad geomorphological similarities with the composite centers of the frontal arc, including edifice height (1.8 km), summit elevation (5.4 km), cone diameter (15 km), and eruptive volume (~84 km<sup>3</sup>). Tunupa is deeply eroded on its east-southeastern flank, where glacial and volcanoclastic sediments form a depositional apron around much of the edifice. Its central peak is extensively hydrothermally altered with abundant sulfur visible in aerial photographs. A series of morphologically distinct domes located on the eastern flanks overlie flows of the main Tunupa edifice (Fig. 3). Small-volume pyroclastic deposits of unknown depth crop out within the drainages of the northern flank (Fig. 3). Two K-Ar ages ( $1.8 \pm 0.2$  Ma [biotite] and  $2.5 \pm 0.5$  Ma [plagioclase]) were reported in a regional study by Baker and Francis (1978), who also reported an  $11.1 \pm 0.4$  Ma whole-rock K-Ar age for the Huayrana lavas located less than 3 km to the east of Tunupa (Figs. 2 and 3). No other geochemical or geochronologic data were available for Tunupa or Huayrana prior to this study.

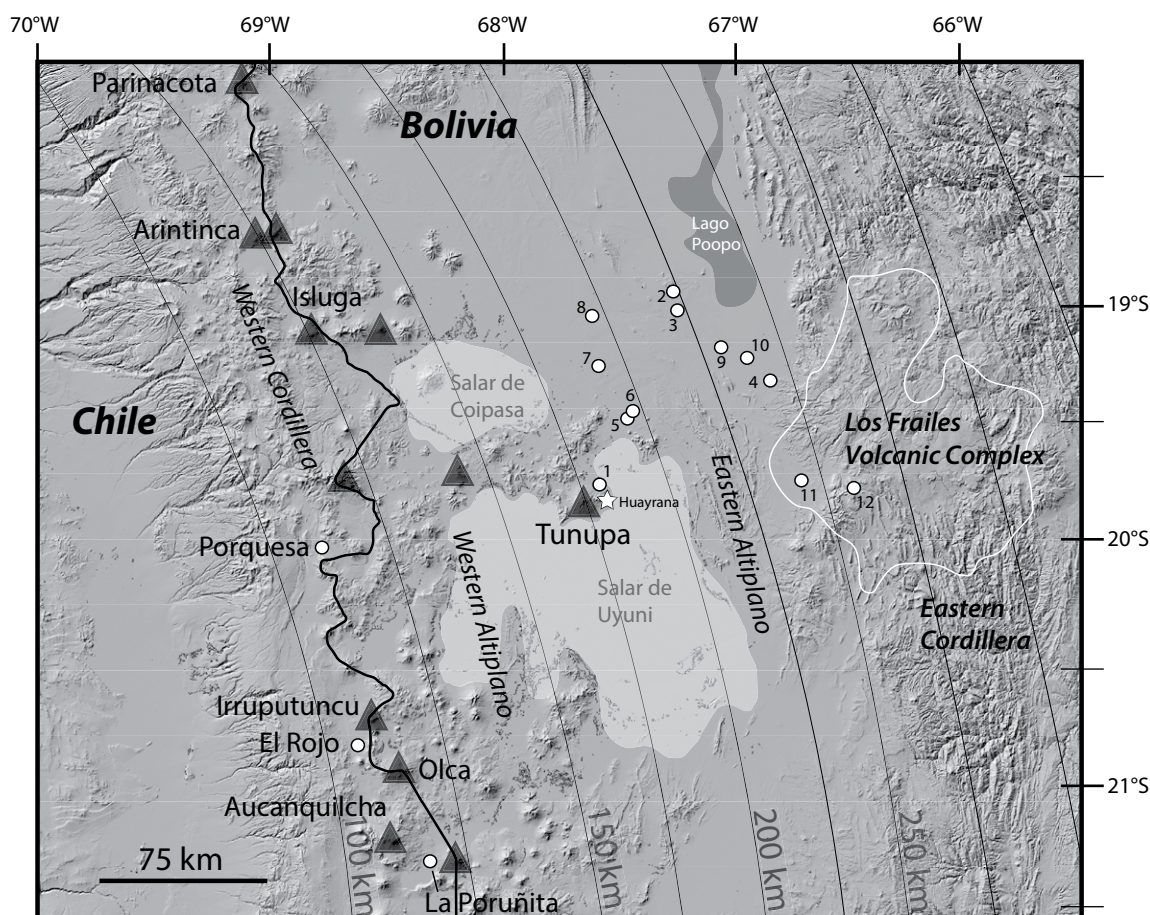
## REGIONAL CENOZOIC REAR ARC MAGMATISM

Widespread rear arc volcanism began in the central Andes between ca. 30 and 25 Ma, following ~10 m.y of intense crustal shortening and relative volcanic quiescence (James and Sacks, 1999; Trumbull et al., 2006). Miocene and younger ignimbrites, stratovolcanoes, monogenetic structures, and intrusive units are widely distributed in the central Andean rear arc region of Peru, Bolivia, and Argentina (for reviews, see Jiménez and López-Velásquez, 2008; Barnes and Ehlers, 2009; Mamani et al., 2010; Kay and Coira, 2009). Miocene and younger, rear arc, large-volume, silicic ignimbrite fields such as Morococala, Los Frailes, Altiplano-Puna, and Cerro Gálán (Fig. 1) are restricted to the thick crustal regions of the central Andes. These fields likely represent large-scale crustal melting, and suggest a petrogenetic origin associated with plateau construction (Coira et al., 1993; de Silva, 1989; Francis et al., 1989; Kay et al., 2010; Salisbury et al., 2011). Rear arc composite cones such as Tunupa, Uturuncu, and Tuzgle are located between 70 and 120 km from the rear arc and are generally composed of intermediate compositions similar to volcanoes of the frontal arc (e.g., Davidson et al., 1991), with subtle differences in major- and trace-element

geochemistry (Sparks et al., 2008; Coira and Kay, 1993; Kay et al., 1994; Michelfelder et al., 2013). The most mafic rear arc lavas are generally associated with smaller-volume, monogenetic, calc-alkaline lavas, shoshonites, and other alkaline lavas (Déruelle, 1991; Davidson and de Silva, 1992, 1995; Redwood and Rice, 1997; Hoke and Lamb, 2007; Carlier et al., 2005).

In the southern Altiplano transect, a similar range of eruption styles and compositions as the entire plateau is represented. The eastern region is dominated by the early Miocene to Quaternary Los Frailes volcanic plateau, covering a surface area of ~8000 km<sup>2</sup>, with an eruptive volume of ~2000 km<sup>3</sup> (Jiménez and López-Velásquez, 2008). West and north of Los Frailes, latest Oligocene to early Miocene basaltic and shoshonitic lava flows and sills crop out with mid-Miocene, Pliocene, and Quaternary monogenetic lavas (Davidson and de Silva, 1992, 1995; Redwood and Rice, 1997; Hoke and Lamb, 2007). Tunupa is located in the central Altiplano, forming the eastern part of an E-W-trending, late Oligocene to Quaternary volcanic field, known as the Serranía Intersalar (Leytón and Jurado, 1995). The western edge of the Serranía Intersalar is marked by the Quaternary Sillajhuayo composite volcano (Salisbury et al., 2013), located within the region of diminished late Pleistocene frontal arc activity known as the Pica Gap (Wörner et al., 1992, 1994, 2000a), between Isluga and Irruputuncu volcanoes (Fig. 2).

Compositions, and proposed petrogenetic sources and melting mechanisms, of the Cenozoic rear arc magmatism in the central Andes vary widely. Mantle-normalized trace-element patterns, including those from the more mafic samples, generally fall along a spectrum between rare intraplate (ocean-island basalt [OIB]-like) signatures and the more common, arc-like signatures (e.g., Kay et al., 1994; Redwood and Rice, 1997; Hoke and Lamb, 2007; Jiménez and López-Velásquez, 2008). As clearly demonstrated in the postcollisional, potassic lavas from the Tibetan Plateau, melting to produce arc-like trace-element patterns in orogenic plateau settings does occur in the absence of active subduction, and the mantle lithosphere is thought to be a major source of these melts (e.g., Turner et al., 1996; Williams et al., 2004). Derivation of melts within hydrous, mica-bearing mantle lithosphere has been argued in the central Andes for potassic magmas in both the rear arc regions of the northern Puna (Déruelle, 1991) and the northern Altiplano (Carlier et al., 2005). Although the proposed, plateau-wide, catastrophic delamination event between ca. 10 and 6 Ma (Garzzone et al., 2006) is controversial (e.g., Barnes and Ehlers, 2009), the removal of large amounts of mantle lithosphere has been implicated in the generation of the voluminous



**Figure 2.** Southern Altiplano region of the central Andes. Dark triangles show locations of Quaternary composite volcanoes; open circles are select Pliocene and younger monogenetic centers described in the literature. Data for our regional Quaternary geochemical comparison are compiled from the labeled arc-front centers of Parinacota (Hora et al., 2009); Aucanquilcha and La Poruñita (Grunder et al., 2008; Walker, 2011); Arintinca, El Rojo (Wörner et al., 1992); Irruputuncu, Isluga, and Olca (Wörner et al., 1992; Mamani et al., 2010). Data from monogenetic rear arc centers are labeled with numbers 1–4 (Davidson and de Silva, 1995); 5–8 (Hoke and Lamb, 2007); 9–10 (McLeod et al., 2012); 11–12 (Jiménez and López-Velásquez, 2008). Depths to the subducting Nazca plate are from Cahill and Isacks (1992).

ignimbrites beneath the volcanic centers of Los Frailes and Gálan (Kay et al., 1994; Myers et al., 1998). Melting accompanying such large-scale foundering events may be dominated by asthenospheric upwelling, whereas the removal of smaller (50–1 km) blocks may result in the heating, dehydration, and melting of the mantle lithosphere, producing the smaller-volume, discrete episodes of central Andean Plateau magmatism (Drew et al., 2009; Ducea et al., 2013).

Much of the uncertainty of possible petrogenetic sources (asthenosphere, mantle lithosphere, continental crust) in the central Andes is due to a lack of detailed knowledge of potential end-member compositions. Further complicating the spatiotemporal and geochemical patterns of central Andean Plateau lavas are variations of slab dip (e.g., Coira et al., 1993; Ramos et al., 2002; Mamani et al., 2010), variable degrees of partial melting (e.g., Kay and Kay, 1993;

Kay et al., 1994), and assimilation of heterogeneous crustal material (e.g., Davidson et al., 1991; Davidson and de Silva, 1995; McLeod et al., 2012), as well as the possible influence of subducting aseismic oceanic ridges and subduction erosion of the forearc (Kay and Mpodozis, 2002). It is within this uncertain context that we examine the rear arc composite Tunupa volcano and regional rear arc magmatism. A better understanding of the temporal, spatial, and geochemical history of the plateau is a key factor in resolving the petrogenetic-tectonic relationships in orogenic plateaus such as the central Andes.

## METHODS

### Sample Collection

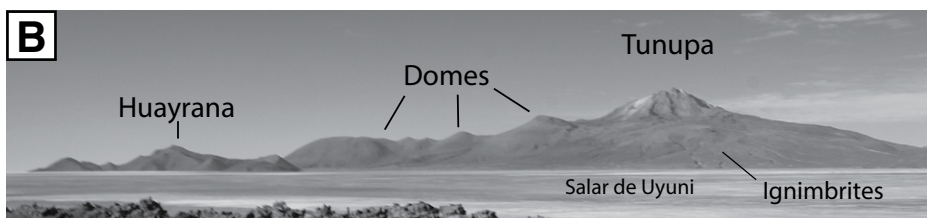
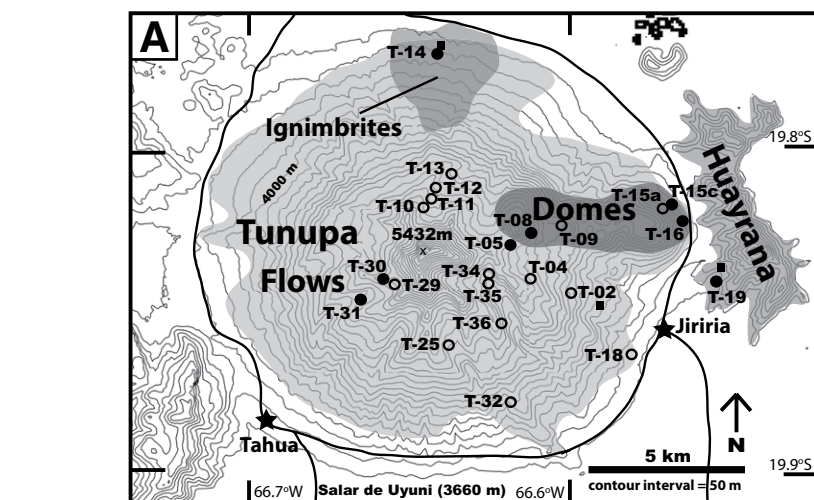
Samples of individual lavas were collected at the lowest and highest accessible stratigraphic

locations on the Tunupa edifice (Fig. 3). Care was taken to collect the freshest samples and to remove weathering rinds, although many samples are slightly altered, with oxidized mafic phenocrysts common. A single lava sample from the highly eroded edifice of Huayrana was also collected and analyzed.

### Geochronology and Geochemistry

The  $^{40}\text{Ar}/^{39}\text{Ar}$  analyses of groundmass glass separates were performed at the University of Wisconsin Rare Gas Geochronology Laboratory. In the absence of sanidine, groundmass separates were preferred for this study over biotite and plagioclase separates, as recent geochronologic studies of the central Andes have demonstrated that these phases often result in imprecise and inaccurate ages (e.g., Hora et al., 2010; Salisbury et al., 2011). Furnace experiments were





**Figure 3.** (A) Annotated topography of Tunupa and Huayrana with sample localities of analyzed rocks. Open circles denote locations of samples with whole-rock analyses, and closed circles denote samples with whole-rock and  $^{40}\text{Ar}/^{39}\text{Ar}$  analyses. (B) Photograph looking toward the north-east side of Tunupa. Field of view is approximately 15 km.

performed following the methods of Jicha et al. (2012). Ages are reported with  $2\sigma$  analytical uncertainties and were calculated relative to a Fish Canyon standard age of  $28.201 \pm 0.046$  Ma (Kuiper et al., 2008) and a value for  $\lambda^{40}\text{K}$  of  $5.463 \pm 0.107 \times 10^{-10} \text{ yr}^{-1}$  (Min et al., 2000).

Whole-rock major- and trace-element analyses (Table 1) were performed at the Geo-Analytical Laboratory at Washington State University–Pullman. Whole-rock major-element concentrations were analyzed by X-ray fluorescence (XRF) using lithium tetraborate fused beads and a Rigaku 3370 spectrometer with a Rh target. Trace-element concentrations were measured by XRF and inductively coupled plasma–mass spectrometry (ICP-MS). In cases where the same element was measured by both methods, ICP-MS results are reported. See Johnson et al. (1999) and Knaack et al. (1994) for complete analytical details.

## Results

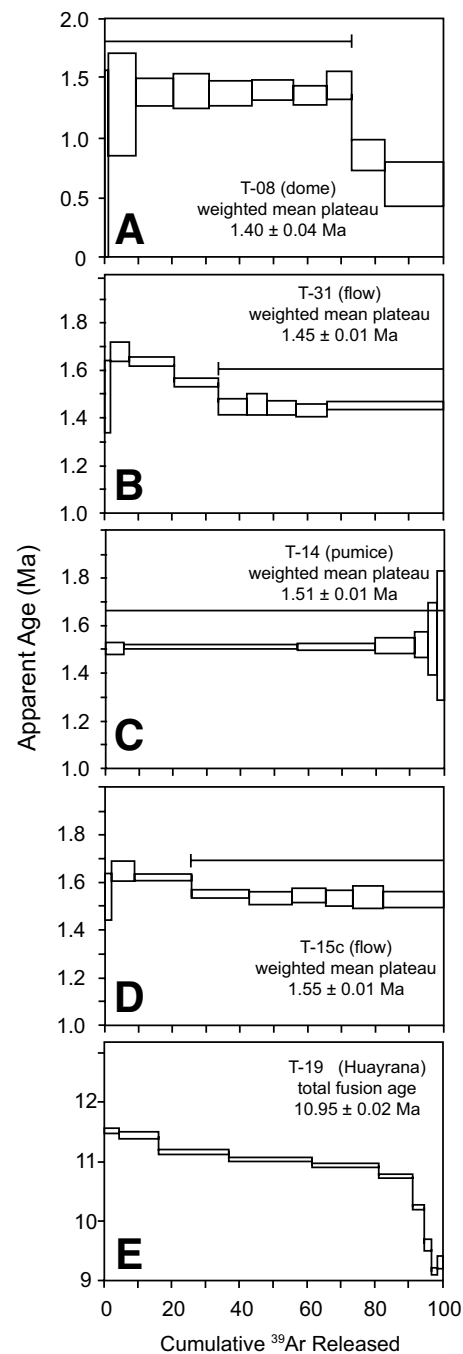
Phenocryst modal percentages of Tunupa lavas range from 15% to 30% and consist of plagioclase, clinopyroxene, amphibole, biotite, oxides, and rare orthopyroxene and olivine. Apa-

tite and zircon are observed as accessory phases. The groundmass varies from glassy to crystalline with microlites of plagioclase comprising much of the matrix. Disequilibrium textures of plagioclase, including sieved, or mottled, cores with clear, euhedral overgrowth rims (e.g., Tsuchiyama and Takahashi, 1983), are common.

## $^{40}\text{Ar}/^{39}\text{Ar}$ Age Determinations

Stratigraphically consistent weighted mean plateau ages are reported for two Tunupa flows, one pumice sample and one lava dome (Table 1; Fig. 4). Plateaus consist of at least three contiguous steps containing  $>50\%$  of the total  $^{39}\text{Ar}$  released and having ages that agree within 95% confidence limits (Renne, 2000). These ages indicate edifice construction between  $1.55 \pm 0.01$  and  $1.40 \pm 0.04$  Ma, and they are significantly younger than the K-Ar ages reported by Baker and Francis (1978).

Downward-trending age spectra without obvious plateaus are also reported for two Tunupa flows, one Tunupa dome and the Huayrana sample (Fig. 4E). These spectra may indicate  $^{39}\text{Ar}$  recoil or may be related to heterogeneous alteration of the groundmass. In



**Figure 4.**  $^{40}\text{Ar}/^{39}\text{Ar}$  age spectra from incremental heating experiments. (A–D) Concordant age spectra and weighted mean plateau ages from Tunupa lavas and pumice. (E) Discordant spectra; total fusion age is shown.

these cases, we use the total fusion ages with caution, three of which fall within the preferred age of Tunupa construction (Table 1). The total fusion age of  $10.95 \pm 0.02$  Ma for Huayrana is within uncertainty of the  $11.1 \pm 0.4$  Ma K-Ar whole-rock age of Baker and Francis (1978).

TABLE 1. WHOLE-ROCK MAJOR- AND TRACE-ELEMENT DATA FOR TUNUPA VOLCANO AND HUAYRANA LAVAS

Sample:	T-02	T-04	T-05	T-10	T-11	T-12	T-13	T-15c	T-18	T-25	T-29	T-30	T-31	T-32	T-35	T-36	T-14	T-08	T-09	T-15a	T-16	T-19
	flow	flow	flow	flow	flow	flow	flow	flow	flow	flow	flow	flow	flow	flow	flow	flow	pumice	dome	dome	dome	dome	Huayrana
<b>XRF<sup>†</sup> (wt%)</b>																						
SiO <sub>2</sub>	63.2	63.0	63.2	62.4	61.8	62.0	62.0	61.3	63.6	61.1	61.8	62.5	62.8	63.4	60.6	63.2	61.3	65.8	65.7	63.5	65.7	65.8
TiO <sub>2</sub>	1.1	1.1	1.1	1.2	1.2	1.2	1.2	1.2	1.1	1.3	1.2	1.1	1.2	1.2	1.4	1.1	1.2	0.8	0.9	0.9	0.9	0.7
Al <sub>2</sub> O <sub>3</sub>	15.9	15.9	15.9	15.7	15.9	15.9	16.1	15.8	16.0	15.7	16.0	15.7	15.6	15.6	16.7	15.7	16.7	15.6	15.6	17.1	15.8	15.7
FeO <sup>*</sup>	5.2	5.0	4.9	5.6	5.7	5.6	5.4	5.7	4.9	5.8	5.6	5.3	5.1	5.0	6.2	4.9	5.3	4.0	4.2	4.4	4.1	4.2
MnO	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
MgO	1.87	2.19	2.03	2.56	2.46	2.47	2.60	2.95	1.86	3.03	2.80	2.56	2.49	2.12	1.96	2.46	2.63	1.68	1.46	1.81	1.33	1.23
CaO	4.08	4.13	4.06	4.38	4.68	4.58	4.39	5.08	3.94	4.91	4.73	4.63	4.35	4.24	5.16	4.39	4.45	3.36	3.32	3.68	3.20	3.27
Na <sub>2</sub> O	4.49	4.55	4.62	4.29	4.29	4.31	4.34	4.16	4.58	4.32	4.03	4.25	4.34	4.37	4.52	4.12	4.42	4.36	4.36	4.10	4.42	4.23
K <sub>2</sub> O	3.7	3.6	3.6	3.5	3.4	3.4	3.5	3.3	3.7	3.3	3.3	3.4	3.6	3.6	2.9	3.6	3.5	4.0	4.1	4.1	4.1	4.5
P <sub>2</sub> O <sub>5</sub>	0.38	0.37	0.39	0.38	0.41	0.39	0.39	0.38	0.38	0.46	0.45	0.41	0.40	0.41	0.52	0.37	0.39	0.28	0.32	0.31	0.34	0.32
Sum <sup>†</sup>	99.04	99.45	99.08	100.11	99.10	98.95	98.90	98.26	98.82	98.89	96.91	98.87	99.10	99.10	98.35	97.06	97.47	99.55	98.74	97.15	98.61	98.47
<b>ICP-MS<sup>§</sup> (ppm)</b>																						
Rb	84	83	81	83	77	78	65	82	82	74	73	80	88	86	56	86	71	99	104	82	98	174
Sr	814	852	881	763	790	770	777	771	879	881	872	865	817	860	980	809	827	747	733	817	736	777
Ba	1188	1204	1252	1087	1069	1066	1130	1051	1262	1015	1075	1156	1074	1076	1208	1079	1155	1134	1207	1313	1209	1128
Cs	1.2	1.2	1.1	1.8	1.0	1.1	0.9	1.4	1.1	1.8	1.6	0.9	2.0	1.2	0.5	1.8	1.4	1.8	1.4	1.8	1.2	2.4
Pb	12.3	11.2	13.8	15.5	13.1	13.5	12.8	12.6	14.1	13.8	13.9	14.2	14.7	13.1	9.3	16.4	16.6	18.3	16.3	19.6	13.7	13.7
Y	16	13	13	15	16	16	15	17	15	15	16	14	14	14	25	12	16	11	13	13	13	23
Zr	267	268	276	267	259	257	272	217	283	242	205	224	247	251	263	215	286	233	239	257	240	332
Hf	6.8	6.7	6.8	6.8	6.6	6.5	6.8	5.6	6.9	6.3	5.4	5.8	6.4	6.5	6.4	5.7	7.2	6.2	6.2	6.9	6.3	8.8
Nb	21.2	20.7	20.9	21.9	21.5	21.2	22.9	19.9	21.1	22.0	20.8	21.2	21.4	21.0	22.1	20.1	23.1	19.8	20.5	21.5	20.5	39.8
Ta	1.6	1.5	1.5	1.6	1.5	1.5	1.6	1.4	1.5	1.6	1.5	1.5	1.6	1.6	1.4	1.5	1.7	1.6	1.6	1.7	1.6	3.0
Th	10.7	10.6	10.4	11.1	10.5	10.1	11.4	9.3	10.4	10.3	8.3	8.7	11.5	11.6	5.3	9.9	11.9	12.2	12.1	13.2	11.9	32.5
U	2.5	2.4	2.4	2.6	2.5	2.5	2.5	2.3	2.5	2.5	2.1	2.1	2.7	2.8	1.2	2.3	2.6	3.0	3.0	2.8	2.9	8.0
La	54.5	53.0	54.9	51.6	50.0	48.5	51.8	55.0	67.0	49.7	48.2	51.7	49.8	52.0	45.8	48.9	55.5	51.7	53.0	74.0	53.3	94.8
Ce	97.7	99.2	102.6	97.8	95.2	95.8	98.9	91.1	102.4	95.4	91.7	97.0	94.1	98.0	94.4	91.2	101.3	95.0	97.0	114.1	96.9	152.4
Pr	11.4	11.2	11.8	11.3	11.0	11.0	11.3	11.7	14.0	11.3	11.2	11.3	10.8	11.4	12.1	10.5	12.1	10.7	11.0	15.3	11.0	18.3
Nd	41.9	40.7	42.7	41.6	40.9	41.0	41.8	44.5	50.6	43.0	42.7	42.7	40.2	42.9	50.5	38.8	44.7	38.3	39.3	54.3	39.9	63.0
Sm	7.3	7.0	7.2	7.1	7.3	7.4	7.2	7.9	8.2	7.7	7.8	7.5	7.1	7.4	10.8	6.8	7.8	6.5	6.8	8.6	6.8	10.1
Eu	1.9	1.9	1.9	1.9	1.9	2.0	1.9	2.1	2.1	2.1	2.1	2.0	1.9	1.9	3.1	1.8	2.0	1.7	1.7	2.0	1.7	2.1
Gd	5.3	4.8	4.9	5.2	5.4	5.5	5.3	6.1	5.7	5.5	5.8	5.2	5.0	5.2	9.1	4.7	5.7	4.3	4.6	5.7	4.7	7.0
Tb	0.7	0.6	0.6	0.7	0.7	0.7	0.7	0.8	0.7	0.7	0.7	0.7	0.7	0.7	1.2	0.6	0.7	0.6	0.6	0.7	0.6	0.9
Dy	3.4	3.1	3.1	3.5	3.6	3.7	3.4	4.0	3.5	3.5	3.7	3.1	3.2	3.3	6.2	2.9	3.6	2.7	2.9	3.4	2.9	4.8
Ho	0.6	0.5	0.5	0.6	0.6	0.6	0.6	0.7	0.6	0.6	0.6	0.5	0.5	0.5	1.0	0.5	0.6	0.4	0.5	0.5	0.5	0.9
Er	1.4	1.2	1.2	1.4	1.4	1.5	1.4	1.6	1.4	1.4	1.5	1.2	1.3	1.2	2.2	1.1	1.5	1.0	1.1	1.1	1.1	2.2
Tm	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.1	0.2	0.1	0.1	0.2	0.2	0.3
Yb	1.1	1.0	0.9	1.1	1.2	1.2	1.1	1.2	1.1	1.1	1.2	0.9	1.0	1.0	1.5	0.8	1.2	0.8	0.9	0.9	0.9	1.9
Lu	0.2	0.1	0.1	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.1	0.2	0.2	0.2	0.1	0.2	0.1	0.1	0.1	0.1	0.3
Ni	14	13	13	12	13	12	14.3	24.2	12.8	22.6	22.3	15.4	17.6	14.3	19.3	15.7	14.1	9.4	10.3	10.6	11.9	10.5
Cr	38	36.6	35.3	41.3	41.7	37.4	39.7	92.2	33.5	76.9	62.9	50.7	54.7	42.7	57.6	49	48	26.8	29	28	27.6	20.1
Sc	8.0	7.9	7.6	9.5	9.5	10.6	9.9	11.9	7.8	10.6	9.4	9.0	9.0	8.4	10.2	8.0	9.4	6.0	6.3	7.2	6.4	7.9
V	106	99	110	124	126	129	117	137	101	135	127	124	119	114	141	109	115	75	85	84	73	79
Ga	22.2	23.4	26	21.9	22.8	21.6	23.6	22.0	22.0	22.0	21.9	22.4	23.5	22.5	24.0	22.6	23.0	21.6	23.4	24.8	23.1	22.6
Cu	18.7	18.6	11.7	18.3	16.3	18.2	18.6	21.3	15.7	34.1	21.7	15.2	16.8	20.9	35.9	12.7	21.1	14.8	17.9	11.3	16.8	17.0
Zn	95	88	91	99	95	100	107	91	117	105	97	90	96	94	125	89	101	94	99	93	94	66
Lat. (°S)	19.841	19.836	19.833	19.818	19.814	19.809	19.799	19.812	19.846	19.859	19.842	19.838	19.848	19.873	19.841	19.848	19.787	19.827	19.822	19.814	19.812	19.825
Long. (°W)	67.602	67.618	67.620	67.649	67.646	67.643	67.643	67.591	67.574	67.641	67.661	67.661	67.670	67.620	67.629	67.627	67.648	67.614	67.603	67.591	67.591	67.553

<sup>†</sup>X-ray fluorescence (XRF) analyses are normalized to 100%. Sum is prenormalized total. All analyses are from Tunupa except T-19.

<sup>§</sup>ICP-MS—inductively coupled plasma—mass spectrometry.

## Major and Trace Elements

Tunupa whole-rock compositions (Table 2) comprise a high-K, calc-alkaline suite that ranges from 60 to 66 wt% SiO<sub>2</sub>, with MgO < 3% and K<sub>2</sub>O > 2.9%, typical of differentiated magmas of the central Andean arc and rear arc. Major-element trends are generally well defined, with increasing SiO<sub>2</sub> accompanied by decreases in CaO, MgO, FeO\*, and TiO<sub>2</sub>, and increases in K<sub>2</sub>O (Fig. 5). Na<sub>2</sub>O concentrations are generally higher than K<sub>2</sub>O and are not defined by a linear trend. Dome samples plot at the silicic end of the Tunupa major-element trends, with an ~2 wt% gap in SiO<sub>2</sub> between the flows and domes. The Huayrana sample is transitionally shoshonitic and similar to the Tunupa domes with respect to major elements.

Mantle-normalized trace-element patterns of Tunupa and Huayrana lavas are characterized by enrichments of large ion lithophile elements (LILEs) relative to high field strength elements (HFSEs) (Fig. 6). Tunupa and Huayrana rare earth element (REE) patterns are steep (Fig. 6; La/Yb<sub>n</sub> = 20–55, Dy/Yb<sub>n</sub> = 2.0–2.7). With increasing SiO<sub>2</sub>, most HFSEs show only moderate variation, whereas the middle to heavy REEs decrease, and Rb and Ba increase (Fig. 7).

## DISCUSSION

### Comparison to Regional Lavas of the Central Andean Plateau

The Pleistocene Tunupa volcano shares morphological, temporal, mineralogical, textural, and compositional features characteristic of previously studied composite centers of the central Andes (Salisbury, 2011), consistent with an overall similar pattern of differentiation during ascent through, and interaction with, the thick Andean crust (e.g., Wörner et al., 1988; Davidson et al., 1991; Feeley and Davidson, 1994; Grunder et al., 2008). A minimum volume of

84 km<sup>3</sup> for the Tunupa edifice is estimated using a simple cone with a height of 1.64 km and a diameter of 14 km. Assuming that the youngest and oldest <sup>40</sup>Ar/<sup>39</sup>Ar ages represent the interval of Tunupa eruptions, we calculate extrusion rates between 0.43 and 0.93 km<sup>3</sup>/k.y., similar to those calculated for Parinacota (0.75–1.0 km<sup>3</sup>/k.y.; Hora et al., 2007) and Lascar (0.70–0.93 km<sup>3</sup>/k.y.; Gardeweg et al., 1998), although considerably higher than lavas from the ≤1 Ma Aucanquilcha (0.04 km<sup>3</sup>/k.y.; Grunder et al., 2008) and Uturuncu (0.14–0.27 km<sup>3</sup>/k.y.; Sparks et al., 2008) edifices.

Tunupa lavas are typically high-K, calc-alkaline, plagioclase-dominated trachyandesites to trachydacites, with evidence for magma mixing including disequilibrium plagioclase textures and rimward increases in An content of many plagioclase phenocrysts (e.g., Tsuchiyama and Takahashi, 1983; Salisbury, 2011). Mixing is a common process in many composite cones of the central Andes (Davidson et al., 1991; Giniere and Wörner, 2007) and may be necessary to facilitate eruption in the central Andes, as well as arc volcanoes worldwide (Kent et al., 2010; Kent, 2013). The strongly fractionated REE ratios of Tunupa and most other central Andean Plateau lavas imply a significant role for garnet, commonly attributed to magma differentiation at the base of the thick Andean crust (Hildreth and Moor bath, 1988; Davidson et al., 1991).

Major-element concentrations generally fall within the range for Parinacota and Aucanquilcha lavas, with the exception of TiO<sub>2</sub> values, which are higher at Tunupa for a given wt% SiO<sub>2</sub> (Fig. 5). Despite overall arc-like patterns on mantle-normalized diagrams (Fig. 7), HFSE compositions are enriched within Tunupa lavas, with the most pronounced enrichment in Nb and Ta (Figs. 5 and 6). The predominance of Nb and Ta (fewer Ta analyses are available in the literature) is apparent in nearly every trace-element ratio involving these elements, including those relative to the less mobile HFSEs such as Nb/Zr (Fig. 8).

Available data from the southern Altiplano rear arc show that the observed Nb-Ta-Ti enrichment at Tunupa is ubiquitous in lavas from the Quaternary centers of the eastern Altiplano and Eastern Cordillera (Figs. 2 and 5–8). This enrichment is apparent in both ratios and concentrations, and it suggests a distinct, rear arc, high-Nb (>~18 ppm) compositional domain. In contrast, available data for Nb concentrations of Quaternary lavas from the southern Altiplano frontal arc average less than 12 ppm (Mamani et al., 2010). The Nb-Ta-Ti rear arc enrichment appears to be plateau-wide and is apparent in rear arc lavas including from composite cones such as Uturuncu (Sparks et al., 2008; Michelfelder et al., 2013) and Tuzgle (Coira and Kay, 1993) in the northern Puna and the <7 Ma mafic lavas of the southern Puna (Drew et al., 2009), as well as the shoshonitic and alkalic lavas from across the plateau (Déruelle, 1991; Carlier et al., 2005).

### Crustal versus Mantle Sources for Nb-Ta-Ti Enrichment

A long-standing debate of central Andean frontal arc magmatism concerns the relative involvements, and influence in trace-element systematics, of the asthenosphere, mantle lithosphere, and continental crust (Hildreth and Moor bath, 1988; Rogers and Hawkesworth, 1989; Davidson et al., 1991). Rear arc magmatism is similarly controversial, with the added complication of poorly constrained models for melt generation. In the southern Altiplano rear arc, Davidson and de Silva (1992, 1995) attributed the higher Nb/Zr ratios relative to the frontal arc as likely due to lower degrees of partial melting related to a diminished slab flux. Lower degrees of partial melting of mantle peridotite, however, would promote higher abundances of the more incompatible trace elements, and lower values of ratios of Nb relative to more incompatible elements such as Th and U. Although there is overlap in the data (Fig. 8), such trends are gen-

TABLE 2. <sup>40</sup>Ar/<sup>39</sup>Ar SUMMARY OF GROUNDMASS SEPARATE LASER INCREMENTAL HEATING EXPERIMENTS

Sample number	Sample type	Location		K/Ca total	Total fusion age (Ma ± 2σ)	<sup>40</sup> Ar/ <sup>39</sup> Ar <sub>i</sub> (±2σ)	Isochron age (Ma ± 2σ)	N	% <sup>39</sup> Ar	MSWD	Plateau age (Ma ± 2σ)
		Lat (°S)	Long (°W)								
T-08	Tunupa dome	19.827	67.614	3.63	1.20 ± 0.06	294.8 ± 0.8	1.48 ± 0.11	8 of 10	72.8	0.74	1.40 ± 0.04
T-31	Tunupa flow	19.848	67.670	1.04	1.50 ± 0.01	295.7 ± 3.7	1.45 ± 0.03	5 of 9	66.3	0.40	1.45 ± 0.01
T-14	Tunupa pumice	19.787	67.648	8.26	1.51 ± 0.01	295.8 ± 1.1	1.51 ± 0.01	7 of 7	100.0	0.15	1.51 ± 0.01
T-15c	Tunupa flow	19.812	67.591	0.93	1.56 ± 0.01	293.4 ± 3.8	1.55 ± 0.02	6 of 9	74.6	0.52	1.55 ± 0.01
T-05	Tunupa dome	19.833	67.620		1.52 ± 0.01						No plateau
T-16	Tunupa flow	19.812	67.591		1.52 ± 0.01						No plateau
T-30	Tunupa flow	19.838	67.661		1.59 ± 0.01						No plateau
T-19	Huayrana flow	19.825	67.553		10.95 ± 0.02						No plateau

Note: Ages were calculated relative to 28.201 Ma for the Fish Canyon sanidine standard. Preferred ages are in bold. MSWD—mean square of weighted deviates.

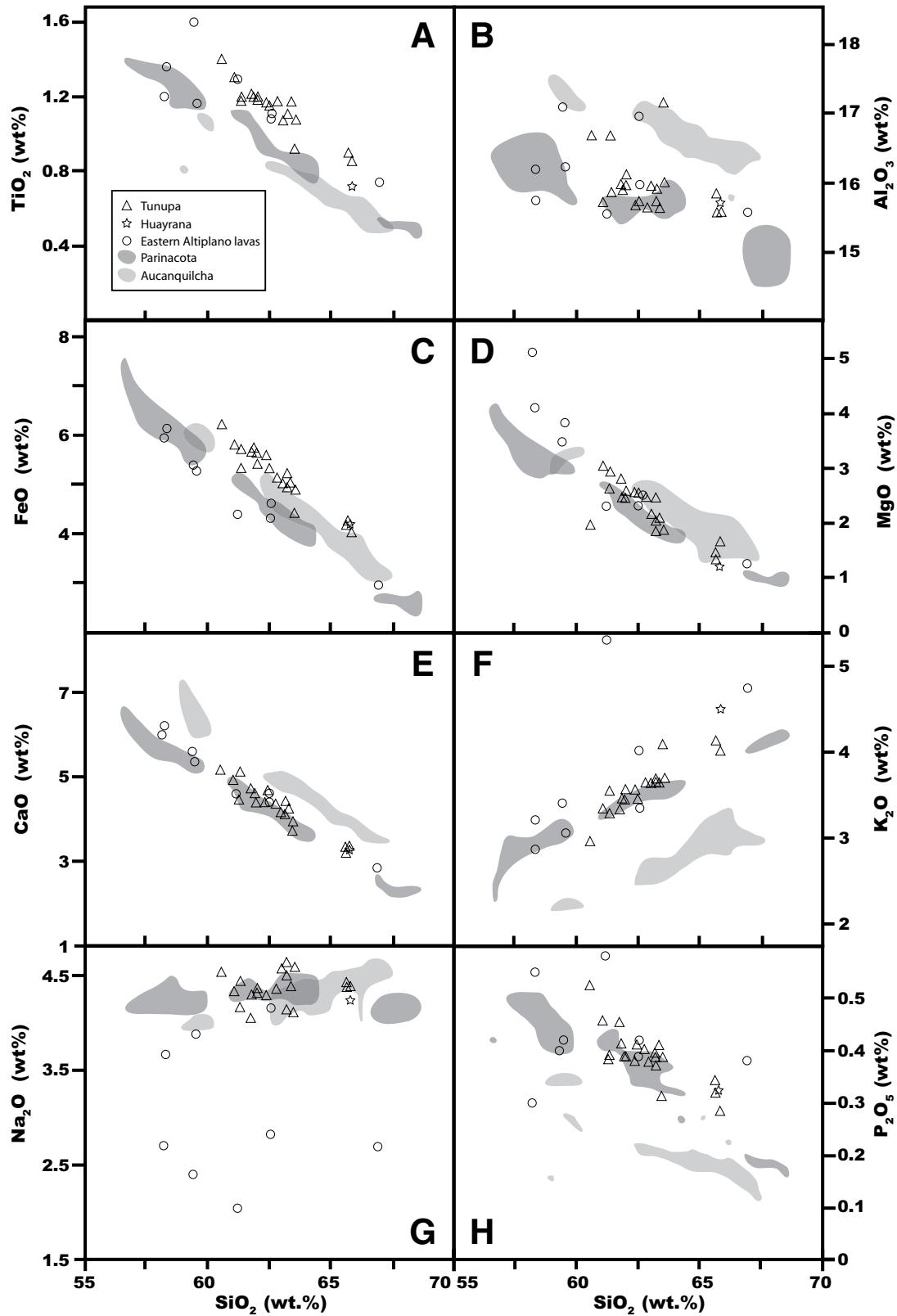
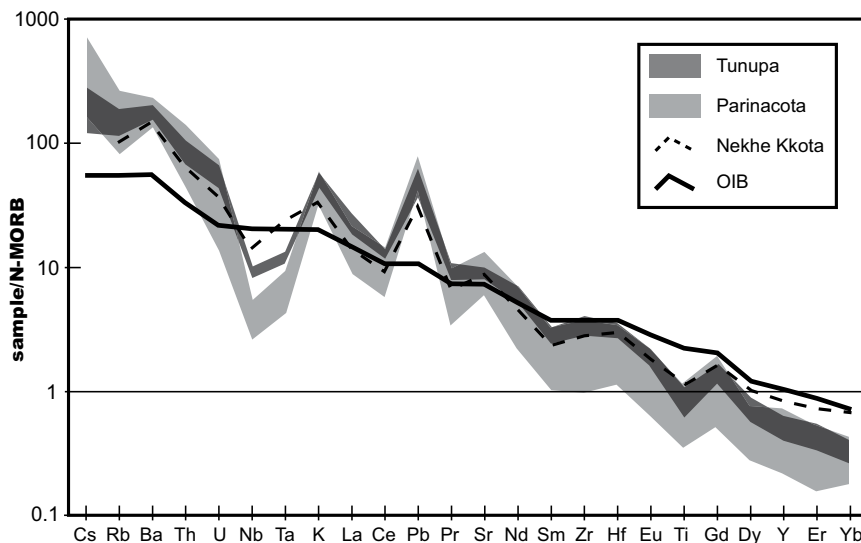


Figure 5. Major-element data for Tunupa and Huayrana samples with regional southern Altiplano centers for comparison. Open triangles are Tunupa; star is Huayrana. Shaded regions define the distribution of compositions for Quaternary composite centers Parinacota and Aucanquilcha for comparison to the arc front. Open circles are monogenetic rear arc centers restricted to  $>55$  wt%  $\text{SiO}_2$  for comparison to the more-evolved composite lavas of Tunupa. Locations and data references are listed in Figure 2.



**Figure 6.** Normalized trace-element data showing range of compositions of Tunupa and Parinacota lavas (Hora et al., 2009). The monogenetic eastern Altiplano center of Nekhe Kkota (<1 Ma, 7 wt% MgO, #6 in Fig. 2; Hoke and Lamb, 2007) and ocean-island basalt (OIB) from Sun and McDonough (1989) are shown for comparison. Data are normalized to normal mid-ocean-ridge basalt (N-MORB; Sun and McDonough, 1989). Note the high variation in normalized Nb and Ta concentrations.

erally not observed in Tunupa and eastern Altiplano rear arc lavas, particularly with respect to the well-studied Quaternary lavas from Aucanquilcha and Parinacota. Furthermore, geodynamic modeling of subduction zones with geometry comparable to the central Andes generally does not support significant amounts of melting by slab dehydration at large (>100 km) distances behind the frontal arc (Grove et al., 2009). Thus, although lower degrees of partial melting related to a diminished slab flux in the rear arc cannot be ruled out definitively, it is unlikely to explain the high-Nb, rear arc geochemistry.

In a seminal study of arc volcanoes along strike in the southern volcanic zone of the central Andes, Hildreth and Moorbath (1988) concluded that northerly increases in such mobile elements as K, Rb, Th, and U are best explained by increased involvement of thick, continental crust. Many of these same elements were also implicated by Michelfelder et al. (2013), who argued for increased involvement of older, more felsic continental crust in the rear arc at Uturuncu volcano compared to the frontal arc. Although crustal contamination is likely in most, if not all, lavas of the central Andean Plateau, we believe it is unlikely to explain the observed rear arc enrichment in Nb-Ta-Ti. Whereas K, Rb, Th, U, and La all increase with increasing  $\text{SiO}_2$  at many individual central Andean centers, including Tunupa, Aucanquilcha, Ollagüe, Uturuncu, and Parinacota, Nb and Ta concen-

trations show very little variation with  $\text{SiO}_2$ , suggesting that Nb and Ta concentrations are not strongly affected by crystal fractionation or crustal assimilation in the central Andes. Furthermore, Nb concentrations are relatively low in average continental crust (~12 ppm; Hawkesworth and Kemp, 2006) as well as in the limited exposures of pre-Cenozoic Andean basement rocks (average = 12.5 ppm,  $n = 76$ ; Damm et al., 1994; Wörner et al., 2000b; Lucassen et al., 1999a) and crustal xenoliths (average = 14.4 ppm,  $n = 34$ ; Lucassen et al., 1999b; McLeod et al., 2013).

We consider the most likely source for Nb-Ta-Ti enrichment to be the subcrustal lithospheric mantle. The association of Nb with Ta and Ti, and their enrichment in alkaline magmas that are generally assumed to derive from metasomatized mantle lithosphere (e.g., Best and Christiansen, 2001), has long been recognized (e.g., Parker and Fleischer, 1968). The most likely hosts for Nb and Ta (as well as an important host for Ti) in metasomatized mantle lithosphere are the volatile-bearing phases of amphibole and phlogopite (Ionov and Hoffman, 1995; Kepezhinskis et al., 1996; Grégoire et al., 2000). Analyses of mantle xenoliths show that amphiboles and micas from metasomatic veins are highly enriched (50–200-fold) in Nb and Ta compared to primitive mantle and are characterized by high (Nb,Ta)/(Th, U, LREE) values (Ionov and Hofmann, 1995). Ti-oxides (rutile, ilmenite) can contain much higher concentrations of Nb and Ta but are not common

mantle minerals (Ionov et al., 1997; Kalfoun et al., 2002). Long-term subduction along the western edge of South America, including periodic episodes of low-angle subduction, could directly hydrate the mantle lithosphere (James and Sacks, 1999; Haschke et al., 2002), resulting in a metasomatized mantle rich in Nb-Ta-Ti-bearing, hydrous mineral phases beneath the pre-Andean Plateau region.

Cenozoic arc and rear arc, mantle-derived xenoliths and primary alkaline magmas are absent in the central Andes (Lucassen et al., 2005), and as a result, no direct estimates of mantle compositions are available for this time frame. However, studies of Nb-rich alkaline lavas and metasomatized mantle xenoliths from eruptions that predate Cenozoic plateau development provide evidence for the presence of ancient, Nb-Ta-rich, metasomatized lithospheric mantle beneath what is now the central Andes (Lucassen et al., 2005; Lucassen et al., 2007). Viramonte et al. (1999) described amphibole + Ti-rich phlogopite + apatite veins in preplateau mantle xenoliths and suggested that such veins served to enrich the Cretaceous lithospheric mantle. Such material would also be a viable source for Nb-Ta-Ti-enriched components in younger plateau magmas.

#### Delamination of the Mantle Lithosphere and Plateau Melt Generation

As the metasomatized lithosphere provides a viable source for the Nb-Ta-Ti-rich central Andean rear arc magmas, lithospheric delamination is a likely mechanism to trigger mantle melting. Although the details of the timing, scale, and physical mechanisms for delamination in the central Andes remain a matter of focused research, geophysical and structural evidence is consistent with the removal of large amounts of the mantle lithosphere and mafic lower crust beneath the central Andean Plateau during Cenozoic shortening of the Andean orogeny (Kay and Kay, 1993; Whitman et al., 1996; Myers et al., 1998; Beck and Zandt, 2002; Yuan et al., 2002; McQuarrie et al., 2005; Barnes and Ehlers, 2009; Kay and Coira, 2009). Displacement of mantle lithosphere to greater depths and higher pressures is expected to result in the breakdown of hydrous phases, leading to dehydration melting (e.g., Elkins-Tanton, 2005) of the surrounding material (Fig. 9). If, as detailed herein, these phases also serve as a significant host for Nb-Ta-Ti, melts generated from this process would also be expected to be enriched in these elements. Delamination may also trigger dry, decompression melting of upwelling asthenosphere, and it is likely that multiple source components are involved in rear arc, plateau magmatism (e.g., Redwood and Rice, 1997), resulting in a wide



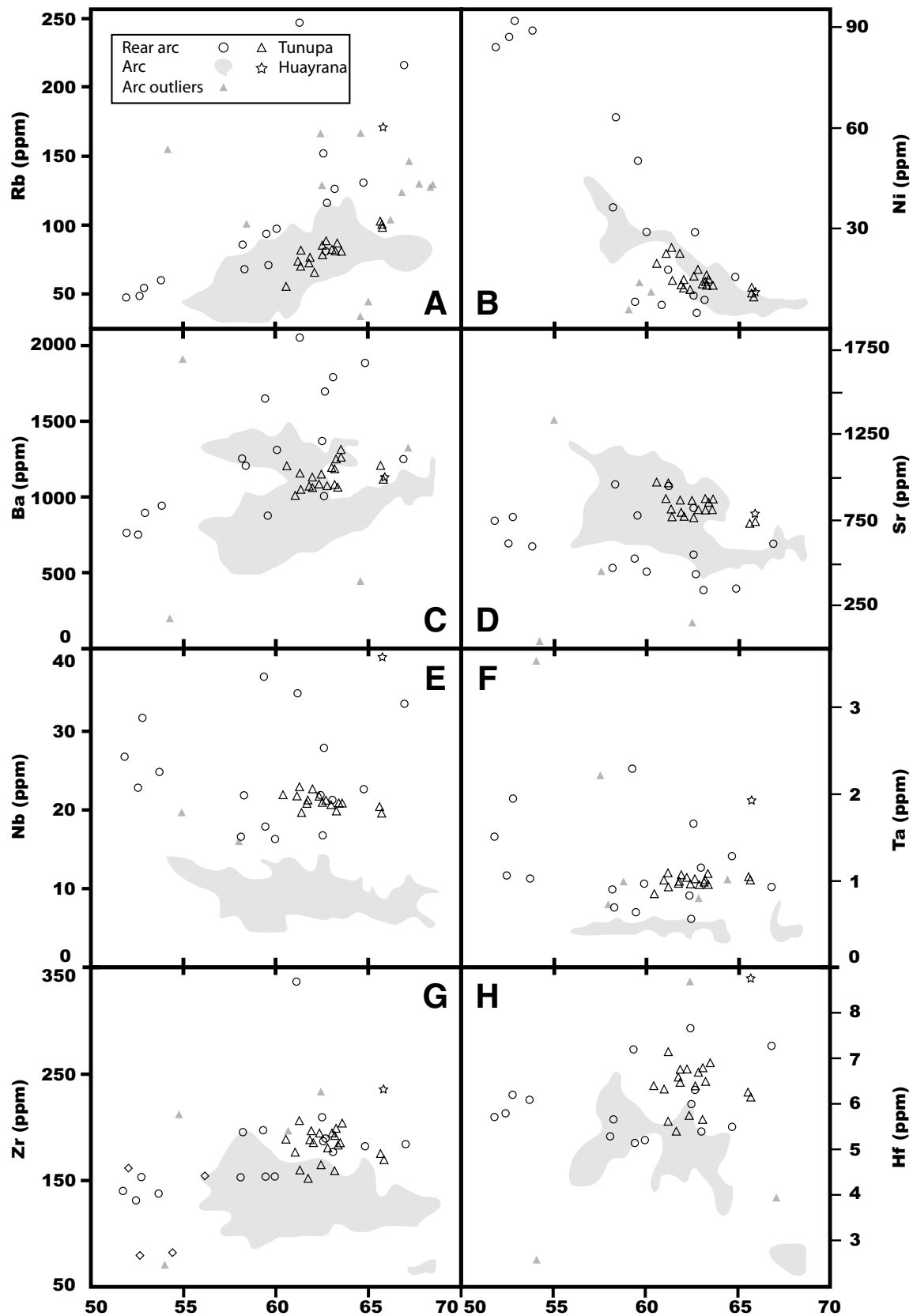
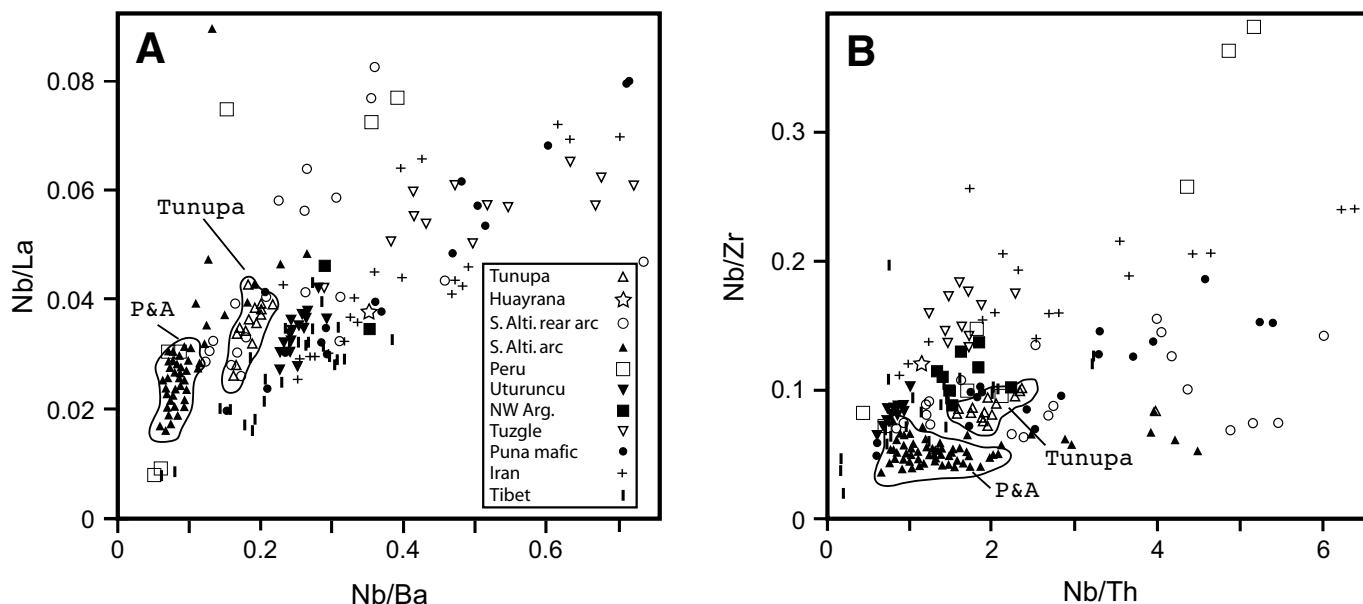
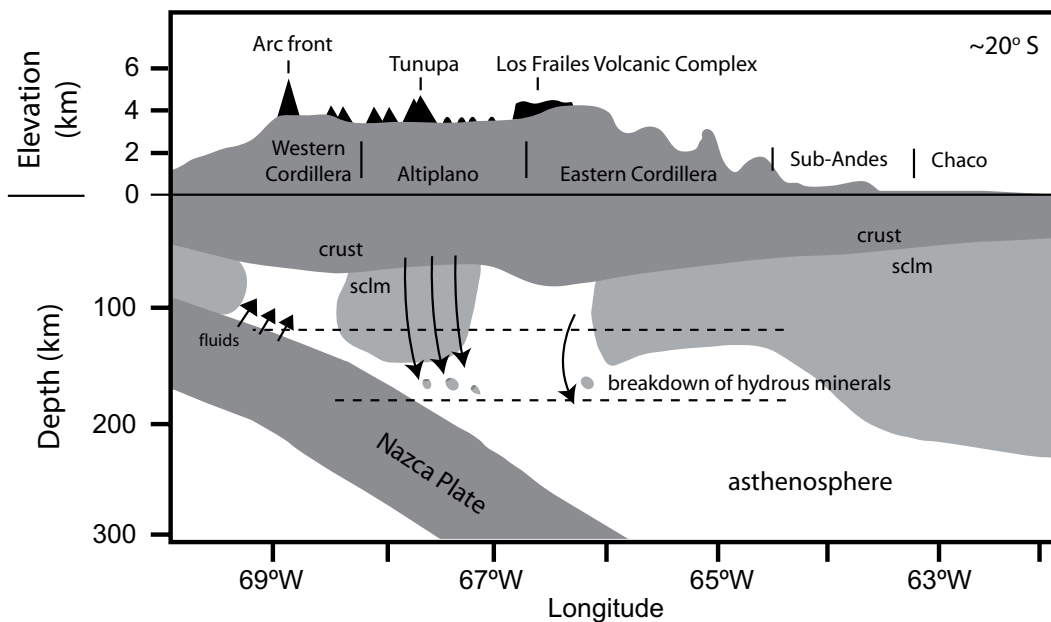


Figure 7. Trace-element data for Tunupa lavas. Huayrana and southern Altiplano Quaternary lavas shown for comparison. Note the prominent Nb-Ta rear arc enrichments. Arc outliers are data points that plot outside of the concentrated data of the shaded distribution. See Figure 2 for Quaternary arc and rear arc locations and data references.



**Figure 8.** Nb/element ratios showing the rear arc enrichment of Nb at Tunupa and plateau lavas from the central Andes, Iran-Turkey, and Tibet compared to the frontal arc of the southern Altiplano. P&A refers to the frontal arc composite volcanoes of Parinacota and Aucanquilcha (Hora et al., 2009; Grunder et al., 2008; Walker, 2011), whereas the remaining symbols are for rear arc, Pliocene and younger monogenetic rear arc lavas of the southern Altiplano (Davidson and de Silva, 1995; Hoke and Lamb, 2007; McLeod et al., 2012; Jiménez and López-Velásquez, 2008); Peru—Quaternary potassic lavas from the northern Altiplano rear arc (Carlier et al., 2005); Uturuncu volcano (Sparks et al., 2008); NW Argentina—potassic lavas from the northern Puna (Déruelle, 1991; Schreiber and Schwab, 1991); Tuzgle volcano (Coira and Kay, 1993); Puna mafic—late Miocene and younger potassic lavas from the southern Puna rear arc (Drew et al., 2009); Iran—Quaternary lavas from the Turkish-Iranian Plateau (Allen et al., 2013); Tibet—Miocene and younger lavas from the northern Tibetan Plateau (Williams et al., 2004). Note that the Cretaceous, mantle lithosphere-derived, alkaline lavas that erupted in what is now the Eastern Cordillera of Bolivia between 19°S and 21°S are too high in Nb/La, Nb/Ba, Nb/Zr, and Nb/Th to plot on the diagrams (Nb = 90–144 ppm; Lucassen et al., 2007).



**Figure 9.** Simplified model showing small-scale removal of lower crust and mantle lithosphere. Breakdown of hydrous minerals during delamination (foundering, removal) within the subcontinental lithospheric mantle (sclm) is proposed to trigger melting and produce the Nb-Ta-Ti-rich magmas. Recent geodynamic models of lithospheric removal suggest much more geodynamic complexity than illustrated here, with possible ductile dripping and partial, piecemeal removal of lower crust and lithosphere (e.g., Göğüş and Pysklywec, 2008; Krystowicz and Currie, 2013). Depths of hydrous mineral breakdown are not well constrained and likely involve a number of intermediate reactions over variable pressures and depths (e.g., Trønnes, 2002). Crustal and lithospheric depths from Beck and Zandt (2002).

range of magma compositions. We argue here that the Nb-Ta-Ti enrichment may be an important indicator of increased involvement of the mantle lithosphere, whereas magmas with lower relative Nb enrichments may reflect a higher proportion of the convecting mantle.

In the rear arc of the southern Altiplano segment, the discrete episodes of mid-Miocene and younger plateau magmatism may each be related to small-scale delamination events (Ducea et al., 2013). If this is the case, the ca. 11 Ma, Nb-Ta-rich Huayrana lavas indicate that this process had begun in the region by this time. It remains unclear if older magmatism (late Oligocene–early Miocene) of the region was related to small-scale delamination, or to a much larger episode of lithospheric removal and slab steepening (Hoke and Lamb, 2007). Small-scale delamination and accompanying Nb-Ta-Ti-rich magmatism may have then continued across the southern Altiplano rear arc throughout the late Miocene to recent time, resulting in the monogenetic magmatism of the eastern Altiplano and the composite Tunupa lavas at ca. 1.55 Ma. The relative proximity to the frontal arc and general similarity in composition and morphology may also indicate a subduction influence in the Tunupa lavas. Nb-Ta-Ti enrichments observed in rear arc, orogenic lavas that erupted across the central Andean Plateau (Déruelle, 1991; Schreiber and Schwab, 1991; Carlier et al., 2005; Drew et al., 2009; Michelfelder et al., 2013; Coira and Kay, 1993; Hoke and Lamb, 2007; Jiménez and López-Velásquez, 2008) suggest that a plateau-wide relationship exists among piecemeal delamination, mineral dehydration within the mantle lithosphere, and the generation of Nb-Ta-Ti-rich magmatism in restricted centers (Figs. 8 and 9). Therefore, further refinement of the time scales, volumes, and compositions of central Andean Plateau volcanism may help to further constrain the spatiotemporal dynamics of plateau delamination.

On a global scale, we also note that similar Nb-Ta-Ti enrichments are observed in the mantle lithosphere-derived magmas from the Tibetan and Turkish-Iranian Plateaus, implying that this relationship may be a global phenomenon (Fig. 8; Nomade et al., 2004; Williams et al., 2004; Zhang et al., 2008; Allen et al., 2013). Many of the magmas from the southern Puna, Tibet, and Turkey-Iran are considerably more mafic (<55 wt% SiO<sub>2</sub>) and are argued to contain little to no contributions from the continental crust (Williams et al., 2004; Drew et al., 2009; Allen et al., 2013). A key implication of this study is that the Nb-Ta-Ti enrichment remains apparent in lavas that have received significant additions from the continental crust, such as those from Tunupa volcano. We, thus, suggest

that the Nb-Ta-Ti signature may be useful in identifying a hydrous-mineral-bearing mantle lithosphere component in regions where concentrations of K, Rb, and Ba may be strongly influenced by crustal contamination.

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### Geochemistry and $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology of lavas from Tunupa volcano, Bolivia: Implications for plateau volcanism in the central Andean Plateau

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