Andean uplift and climate evolution in the southern Atacama Desert deduced from geomorphology and supergene alunite-group minerals

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Abstract
Supergene alunite group minerals from the Late Eocene El Salvador porphyry Cu district, the El Hueso epithermal gold deposit and the Coya porphyry Au prospect located in the Precordillera of Northern Chile (~26 to 26°30´ Lat. S) have been dated by the ⁴⁰Ar/³⁹Ar method and analyzed for stable isotopes. These data support published geomorphologic and sedimentologic evidence suggesting that the Precordillera in the Southern Atacama Desert had been uplifted as early as the Late Eocene and, thus, significantly prior to the Altiplano which attained its high elevation only in the late Miocene. The oldest supergene alunite from the Damiana exotic deposit at El Salvador was dated at 35.8±1 Ma and yielded a δD (VSMOW) value of −74‰, which indicates elevations of the Precordillera near El Salvador of at least 3000 m in the Late Eocene. In contrast, Miocene supergene alunite from El Salvador, El Hueso, and Coya have less negative δD signatures reaching values as high as −23 to −25‰ at El Hueso and El Salvador between about 8.2 and 14 Ma. Late Miocene to Holocene supergene alunite, jarosite and natroalunite ages are restricted to El Hueso and Coya located near 4000 m above sea level in the Precordillera, roughly 1000 m higher than the present elevation of El Salvador. The δD values of samples younger than −5 Ma vary between −57 and −97‰. The complex evolution of the δD signatures suggests that meteoric waters recorded in supergene alunite group minerals were variably affected by evaporation and provides evidence for climate desiccation and onset of hyper arid conditions in the Central Depression of the southern Atacama Desert after 15 Ma, which agrees well with published constraints from the Atacama Desert at 23–24° Lat. S. Our data also suggest that wetter climatic conditions than at present prevailed in the latest Miocene and early Pliocene in the Precordillera.

The new and previously published age constraints for El Salvador indicate that supergene mineralization at the Damiana exotic Cu deposit occurred periodically over 23 Ma in a locally exceptionally stable paleohydrologic and geomorphologic configuration.

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1. Introduction
The Andean uplift history, its causes and effects on the climate have been subject of significant research in recent years (e.g., Garzione et al., 2008; Lamb and Davis, 2003; Schlunegger et al., 2006). Much of this work has been concentrated in the northern Chile and Altiplano transects (~18–20° Lat. S, Fig. 1). Farías et al. (2005) and Victor et al. (2004) suggest that up to 2600 m of uplift occurred in the late middle Miocene and was accommodated by high-angle west verging faults in the western Cordillera. Geomorphologic (García and Héral, 2005; Hoke et al. 2007; Schlunegger et al., 2006; Thouret et al. 2007) and stable isotope evidence (Garzione et al., 2008) places the major uplift which gave rise to the present day high elevations of the Altiplano in the late Miocene. The southern Atacama Desert (~26–27° Lat. S, Fig. 1) has received comparatively less recent attention, but available evidence indicates that the uplift history was fundamentally distinct, irrespective of the controversies on the exact timing of Altiplano uplift. For example, no significant high angle west verging faults active during the Miocene have been documented for the southern Atacama Desert. In addition, geomorphologic, apatite fission track and sedimentologic evidence (Nalpas et al., 2005; Riquelme et al., 2007) suggest that in the southern Atacama Desert the Precordillera had attained considerable elevations in the Oligocene or earlier, which greatly precedes the Altiplano uplift. We herein assess the uplift and climate evolution in an oblique transect across the Precordillera at 26–26°30´ Lat. S (Figs. 1, 2) on the basis of the well established geomorphologic framework (Bissig and Riquelme, 2009; Nalpas et al. 2008; Riquelme et al., 2003, 2007, 2008) and eleven new ⁴⁰Ar/³⁹Ar ages and corresponding stable isotope data for supergene alunite group minerals from the El Salvador porphyry Cu district (e.g., Gustafson et al., 2001), the El Hueso epithermal Au deposit (Marsh et al., 1997; Thompson et al., 2004) and the Coya porphyry Au prospect (Rivera et al., 2004), all located in the southern Atacama

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Desert of Chile (Fig. 2). Our new age constraints complement (Fig. 2). Dotted lines indicate physiographic boundaries from Riquelme et al. (2007).

**Fig. 1.** Map of the western Andean slope of northern Chile. The study region is outlined (Fig. 2). Dotted lines indicate physiographic boundaries from Riquelme et al. (2007). Abbreviations: CC: Coastal Cordillera; CD: Central Depression; PC: Precordillera; PD: Preandean Depression; SP: Salar de Pedernales; WC: Western Cordillera; CB: Calama basin; CCG: Cordillera Claudio Gay. Ore deposits and prospects relevant for this paper are 1: Chuquicamata, 2: Escondida; 3: El Salvador; 3: Potrerillos/El Hueso/Coya; 5: La Coipa.

2. Tectonic history and landscape evolution of the southern Atacama Desert

The present day geomorphologic configuration of the fore-arc region of northern Chile (18–28°S) is dominated by extensive pediplain surfaces which are the result of interaction between climatic and tectonic evolution during the late Cenozoic (e.g. Lamb and Davis, 2003; Mortimer, 1973; Rieu, 1975; Riquelme et al., 2003). These relict pediplains have resisted significant modification through erosion for exceptionally long periods of time in some areas (e.g., Dunai et al., 2005). The tectonic and geomorphologic evolution of the studied transect at 26–26°30’ S Lat is summarized in the following.
analyzed for stable oxygen and hydrogen isotopes to potentially constrain the paleo meteoric water at the time of its formation (Arehart et al., 1992; Rye et al., 1992). Since the isotopic composition of meteoric water depends on elevation (e.g., Poage and Chamberlain, 2001), supergene alunite has the potential to record uplift histories (Taylor et al., 1997). However, the isotopic composition of meteoric waters in arid climates may also be influenced by evaporation (e.g., Godfrey et al., 2003) and the relative importance of the latter may be assessed if the tectonic and geomorphologic framework of a region is independently constrained.

4. Samples and analytical methods

Supergene alunite, natroalunite and jarosite, ranging from powdery to porcellaneous, white to slightly greenish to yellowish veins,
specimen, where possible using a micro-drill tool. In these cases the sample for geochronology was extracted first, followed by the sample for stable isotope analysis. Sample material for XRD was extracted last due to the larger amount required. Most ⁴⁰Ar/³⁹Ar analyses were performed at the Noble Gas Laboratory, Pacific Centre for Isotopic and Geochemical Research (PCIGR), University of British Columbia, Vancouver, BC, Canada, but samples CTB43, CTB46, CTB48 and CTB49 were dated at the ⁴⁰Ar/³⁹Ar facility at the Geophysical Institute at the University of Alaska at Fairbanks (UAF). At PCIGR, the samples were step-heated at increasing laser powers in the defocused beam of a 10-W CO₂ laser. The flux monitor used was Fish Canyon Tuff sandine, 28.02 Ma (Renne et al., 1998). For further details on analytical methods refer to Bissig et al. (2008). At the UAF, an 8 W Ar laser was used and the flux monitor was TCR-2 with an age of 27.87 Ma (Lanphere and Dalrymple, 2000); the analytical methods are described in Layer (2000). All ages are reported with the analytical errors at the 2σ level and represent statistically relevant plateau ages unless indicated otherwise. The reported plateau ages are all within error of the corresponding inverse isochron ages. All ⁴⁰Ar/³⁹Ar data are included in digital appendices.

The δ³⁴S, δ¹⁸O₅O₄, δD values for alunite were determined at the Queen’s University facility for Isotope Research using a method modified from Arehart et al. (1992) and Wasserman et al. (1992). Sulfur was extracted online with continuous-flow technology, using a Finnigan MAT 252 isotope-ratio mass spectrometer. Sulfate oxygen was extracted using the technique of Clayton and Mayeda (1963) and hydrogen was extracted from alunite by pyrolysis. All values are reported in units of per mil (%), and were corrected using NIST standards 8556 for sulfur, and 8557 for sulfur and oxygen and NIST 8535 for hydrogen. Sulfur is reported relative to Canyon Diablo Troilite (CDT), oxygen and hydrogen relative to Vienna Standard Mean Ocean Water (V-SMOW). Analytical precision for both δ³⁴S and δ¹⁸O₅O₄ values is 0.3‰ and for δD 5‰.

5. Episodes of supergene mineralization

5.1. El Salvador

Supergene mineralization at El Salvador is principally represented by two exotic deposits, Damiana and Quebrada Turquesa (Figs. 2, 3). Mote et al. (2001) established an overall age range of 35.4 to 11.1 Ma for supergene activity mostly on the basis of Mn-oxide ages in the Damiana exotic deposit. In this study we obtained 6 additional supergene alunite ages (Fig. 4, Table 1) which confirm the overall age range at El Salvador. However, at the outcrop scale, the published ages were not reproducible. At Quebrada Riolita, upstream form the Damiana exotic deposit (Fig. 3, see also Fig. 6 in Mote et al., 2001) two alunite samples extracted from a horizontal vein were dated (Figs. 3, and 4, Table 1): sample STB12A-1 represents homogeneous porcellaneous alunite from the central part of the vein and yielded an ⁴⁰Ar/³⁹Ar age of 14.22±0.16 Ma. Sample STB12A-2 represents alunite completely replacing the feldspars and groundmass from a rhyolitic wall rock clast within the porcellaneous vein and was dated at 35.82±0.95 Ma. Both of our new ages are considerably older than the 12.89±0.06 to 13.02±0.06 Ma age range obtained by Mote et al. (2001) from a subhorizontal vein from the same outcrop. Two additional samples were dated from the brecciated infill of a steeply dipping fault exposed in the Quebrada Riolita outcrop. The alunite is porcellaneous and occurs as white to pale yellowish subangular breccia clasts of less than 1 cm in diameter (Sample STB12B-1), as well as white to pale greenish alunite groundmass (Sample STB12B-2), which suggests that alunite was emplaced in at least two stages separated by fault movement. Alunite extracted from a clast was dated at 15.31±0.63 Ma whereas alunite form the groundmass yielded an age of 13.83±0.23 Ma. Mote et al. (2001) obtained younger ages ranging from 13.22±0.12 to 13.61±0.06 Ma from a sub vertical vein in the same outcrop.
In the El Salvador district, supergene alunites outcropping upstream from the Quebrada Turquesa exotic deposit were collected. Sample STB026 from a powdery white alunite vein yielded a plateau age of 16.31±0.12 Ma (Figs. 3, 4); an additional sample (STB022) yielded, in two separate analytical runs, reproducible age spectra with stepwise increasing ages from ~9 to 14 Ma albeit without attaining a plateau. This sample is interpreted as a mixture of two or more generations of fine grained alunite. Mote et al. (2001) reported one alunite age of 14.8±0.16 Ma as well as supergene Mn oxide ages from 22.9 to 14.4 Ma for Quebrada Turquesa. The geochronological results suggest that exotic mineralization processes at Damiana apparently outlasted those at Quebrada Turquesa.

5.2. El Hueso/Potrerillos

Late Miocene supergene activity at El Hueso led to the precipitation of powdery white alunite within a fracture outcropping on the uppermost bench of the open pit at 3940 m a.s.l. near the pre-mining
The alunites dated in this study have all been analysed for $^{40}$Ar/$^{39}$Ar and XRD analysis, consists of natroalunite mixed with minamiite (Na,Ca,K)Al$_3$O$_9$(OH)$_6$. This sample is also from Coya Maya (3690 m elevation) and yielded an isochron age of 4.83 ± 0.56 Ma on the basis of two aliquots, but similar to sample CTB-46, the age spectra may be affected by $^{39}$Ar recoil loss (Fig. 5). Thus, neither of the aliquots provides a statistically significant age spectrum, but run 1 yielded a pseudo-plateau age of 5.8 ± 0.8 Ma when the errors are increased to two sigma on the individual heating steps. Due to the evidence for recoil effects we prefer the inverse isochron age. An additional sample of natroalunite (CTB-48) was dated from Coya Maya. Scanning Electron Microscope energy dispersive analysis determined the presence of sufficient K for $^{40}$Ar/$^{39}$Ar dating. This sample, like the other samples from Coya Maya, exhibits evidence for recoil effects but two analytical runs yielded an age not significantly different from zero (Fig. 5, Table 1).

6. Stable isotope constraints

The alunites dated in this study have all been analysed for $^{34}$S, $^{34}$S, $^{34}$S and $\delta^{34}$S values of hydroxyl groups in the alunite directly reflect the meteoric water compositions at the time of supergene processes, because the hydrogen isotopic fractionation between water and alunite or natroalunite is minimal at surface temperatures (Bird et al., 1989; Rye et al., 1992) and the $\delta^{18}$O of water in equilibrium with alunite is within the analytical uncertainty from the latter. $\delta^{18}$O values on the sulphate oxygen in the supergene alunite occupy a wide range due to the incorporation of oxygen both from the water as well as the atmosphere (Rye et al., 1992).

The late Eocene alunite from Quebrada Riolita yielded a $\delta^{18}$O value of −73‰ whereas the other alunites from the same location exhibit a marked increase in $\delta^{18}$O from −61‰ at 15.4 Ma to −50‰ at 13.8 Ma (Fig. 7). Alunites from the headwaters of Quebrada Turquesa exhibit significantly higher $\delta^{18}$O values of −34 to −23‰ at ages younger than 16.3 Ma. The $\delta^{18}$O composition of the 8.2 Ma alunite sample from El Hueso is at −25‰, similar to those from Quebrada Turquesa.

At Coya, the early Miocene alunite has a $\delta^{18}$O value of −53‰, whereas the early Pliocene natroalunite and jarosite yielded strongly negative $\delta^{18}$O values of −88‰ and −97‰ respectively. The most recent supergene natroalunite has at −57‰ a less negative $\delta^{18}$O composition.

7. Discussion

7.1. Chronology of supergene oxidation

As documented for an outcrop near the Damiana exotic deposit, ages of supergene alunite vary widely within a single outcrop or vein, indicating that fluids from which these supergene minerals precipitate exploit the same permeability network periodically over extended periods of time. Although this has been known on a porphyry district scale (Sillitoe, 2005), our data, combined with published data (Mote et al., 2001) suggest that this is also the case at a local scale at the Damiana exotic deposit. Here, both within the exotic deposit as well as at the corresponding paleo spring setting ages range from about 36 to 13 Ma, indicating that exotic mineralization processes operated periodically over 23 Ma in an individual ore forming system. Thus, the permeability network exploited by supergene fluids remained active over an extended period of time and implies that the local geomorphologic configuration has not changed substantially. Although the pediment hosting Damiana has likely experienced modifications and was shaped most recently during the formation of the multi-stage Atacama pediplain, erosion was never substantial enough to strip the gravels down to the paleosurface.

The timing of the cessation of supergene activity in the Central Depression and western Precordillera, proposed at ca. 13 Ma (Mote et al., 2001), has been roughly confirmed. The respective youngest supergene ages of Damiana and Quebrada Turquesa correspond to the inferred relative ages of the pediment surfaces hosting these two exotic deposits (Figs. 3, 8), indicating a potential link between local pediment formation and exotic mineralization. The cessation of
Fig. 5. $^{40}$Ar/$^{39}$Ar age spectra and inverse isochron diagrams for supergene alunite group minerals from El Hueso and Coya dated in this study. Sample HTB04 is from El Hueso, the remainder of samples are from Coya.
supergene alunite precipitation at El Salvador occurred at a similar time as in porphyry Cu and epithermal districts farther North (e.g., Arancibia et al., 2006; Bouzari and Clark, 2002; Hartley and Rice, 2005; Sillitoe and McKee, 1996), which, together with other paleoclimatic evidence (Alpers and Brimhall, 1988; Rech et al., 2006) indicates climate desiccation in the middle Miocene (Fig. 8).

The periods of most intense supergene activity in the late Oligocene and Middle Miocene originally defined for northern Chile and southern Peru (Sillitoe and McKee, 1996) become more blurry as more geochronological data become available (Hartley and Rice, 2005) and recent studies suggest a continuous period of intense supergene processes lasting from the late Eocene to the early late Miocene in Northern Chile (Arancibia et al., 2006). Our results are consistent with a prolonged history of supergene mineralization for the El Salvador district.

In the eastern Precordillera at El Hueso and Coya, at elevations approximately 1000–1200 m higher than at El Salvador, 40Ar/39Ar constraints, admittedly still limited, indicate that supergene processes occurred in the late Oligocene and early Miocene as well as from the late Miocene to early Pliocene and may still be occurring at the present day (Fig. 8). Contrasting with El Salvador, supergene oxidation in the eastern Precordillera appears to have been limited throughout the middle Miocene. While the late Oligocene and early Miocene ages roughly coincide with the incision of the Sierra Chocos del Cobre and Asientos pediplains (Fig. 8) and supergene oxidation may have been related to these erosive processes, we interpret the Late Miocene and younger oxidation to be controlled by uplift to elevations sufficient to capture increased precipitation combined with the incision of deep canyons into the previous planar landscape (Bissig and Riquelme, 2009). This would lead to depression of the water table, but increased availability of meteoric water in the vadose zone, generating conditions favorable for sulfide oxidation.

7.2. δD through time

The Late Eocene meteoric water at El Salvador was at δD = −73‰, similar to the present day precipitation at ~3500 m a.s.l. when calculated using the empirical relationship for South America from Poage and Chamberlain (2001). The estimated elevation for the Late Eocene would be no more than 500 m lower if the long term oxygen isotopic variations in seawater (Zachos et al., 2001) are considered. Miocene meteoric waters are considerably less deuterium depleted and the least negative δD values of −23 to −34‰ were obtained for samples between 8.2 and 16.3 Ma from both El Hueso and El Salvador. Early Pliocene waters at Coya were at δD = −88 to −97‰, similar to present day precipitation around the 3800–4000 m elevation at which Coya is presently situated (Poage and Chamberlain, 2001). The most recent sample yielded a less negative δD value of −57‰. Our data starkly contrast earlier work (Taylor et al., 1997) which suggests sharply decreasing δD values from −15‰ in the Late Oligocene to as much as −60‰ in the middle to late Miocene which they interpret as evidence for a marked uplift pulse in the Middle Miocene. The discrepancy between the two datasets can probably be explained by the different scales of the two studies. Taylor et al. (1997) analyzed alunite samples from 20 to 27° S Lat S (see also Sillitoe and McKee, 1996) which likely reflect significant along strike variations in geomorphology, uplift history and climate. North of about 23° Lat S, there is no Preandean Depression (Fig. 1) and independent evidence suggests that much of the uplift of the Altiplano has occurred in the middle or late Miocene (e.g., Gregory-Wodzicki, 2000; Hoke et al., 2007). In the southern Atacama Desert, the Precordillera attained elevations of at least 2000 m in the early Oligocene (Riquelme et al., 2007) and our stable isotope data suggest a Late Eocene elevation of 3000 m a.s.l. or more for the Precordillera near El Salvador. These high elevations may be attributed to intense folding and thrusting (Niemeyer and Munizaga, 2008) and crustal thickening (e.g., Haschke et al., 2002) affecting the region in the late Eocene.

The increasing δD values throughout the middle Miocene are contrary to the trend expected for an uplifting mountain range. However, the isotopic composition of meteoric water is not only controlled by orographic effects, but also by evaporation and recycling of meteoric water (Godfrey et al., 2003). Thus, we interpret the higher than expected Miocene δD values largely as an effect of evaporation. Bird et al. (1989) and Sillitoe (2005) suggested that high rates of evaporation are conducive for supergene alunite formation, providing support to our interpretation. Thus, the least negative δD values would coincide with the most intense evaporation and hyper arid conditions which likely persisted between about 15 and 8 Ma. The timing of the onset of hyper-arid conditions is also recorded by a
marked decrease of sediment accumulation in the Central Depression in the middle Miocene (Nalpas et al., 2008; Riquelme et al., 2007). Given that the Precordillera attained considerable elevations significantly prior to the middle Miocene hyper arid climate, the uplift of the mountain range probably does not by itself account for the climate desiccation in the southern Atacama Desert (see also Lamb and Davis, 2003). However, the eastward migration of the deformation into the Cordillera Claudio Gay in the late Oligocene (Mpodozis and Clavero, 2002) and the formation of the Preandean depression likely enhanced aridification of the Central Depression. We suggest that the widening rather than simply the uplift of the Andes likely has resulted in increased rain shadow effects at the western Andean slope.

Somewhat wetter conditions probably dominated the early Pliocene in the Precordillera when compared to the arid middle Miocene climate. Stable isotope evidence suggests that the Precordillera probably had attained elevations similar to the present and that evaporation effects were limited. This is interpreted as the result of increased capture of orographically controlled precipitation at that time. Moreover, sedimentological evidence in the Calama basin, some 400 km farther north (Fig. 1; Hartley and Chong, 2002), indicates that semiarid climatic conditions prevailed in the Precordillera and western Andes between about 6 and 3 Ma.

The present climate and hydrologic conditions in the eastern Precordillera are potentially still wet enough to permit the formation of supergene alunite group minerals, but significant evaporation likely affects the meteoric waters. Strong evaporation effects have been documented for meteoric waters in the internally drained basins of the Salar de Hombre Muerto and Salar de Atacama basins (Godfrey et al., 2003).

8. Conclusions

- Geomorphologic and stable isotope evidence strongly suggests that the Precordillera in the Southern Atacama Desert has attained elevations of at least 3000 m a.s.l. already in the early Oligocene and thus, significantly prior to the major uplift of the Altiplano.

- The climate evolved differently in the western Precordillera and Central Depression from the eastern Precordillera. The cessation of supergene processes at El Salvador around 13 Ma has been confirmed and is attributed to climate desiccation, an interpretation also supported by sedimentological and stable isotope evidence. However, conditions at Coya and El Hueso in the Eastern Precordillera, situated near 4000 m present day elevation, remained conducive for at least episodic supergene alunite formation until the early Pliocene, and possibly up to the present day. Uplift to elevations near 4000 m a.s.l. have led to increased capture of moisture and consequently increased availability of meteoric waters.

- The new 40Ar/39Ar age constraints presented herein provide evidence confirming the previously proposed protracted history of the Damiana exotic Cu deposit and indicate that the local geomorphologic and hydrologic configuration has remained relatively stable over 23 Ma.

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Fig. 8. Chart integrating landscape chronology, tectonic episodes, ages of supergene minerals and climate. Abbreviations: A1: early stage Atacama pediplain; A2: Main stage Atacama pediplain; A3: late stage Atacama pediplain; AS: Asientos surface; SC: Sierra Checos del Cobre surface. WP: Western Precordillera; EP: Eastern Precordillera. Supergene ages are plotted individually (black bars; bold correspond to this study) or as groups of ages (boxes; number of dates indicated). References as follows: supergene ages from El Hueso: Marsh et al. (1997); supergene ages from El Salvador: Mote et al. (2001); Pediment formation: Mortimer (1973), Si1ltoe et al. (1968), Bissig and Riquelme (2010); canyon incision: Riquelme et al. (2003, 2007); Gravel deposition: Riquelme et al. (2007); Tectonic episodes: Niemeyer and Munizaga (2008), Mpodozis and Clavero (2002), Riquelme et al. (2003, 2007).

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