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Alunite Supergroup Minerals from an Acid-Sulfate Alteration Assemblage in the Southern Atacama Desert as Indicators of Paleo-Hydrothermal and -Supergene Environments

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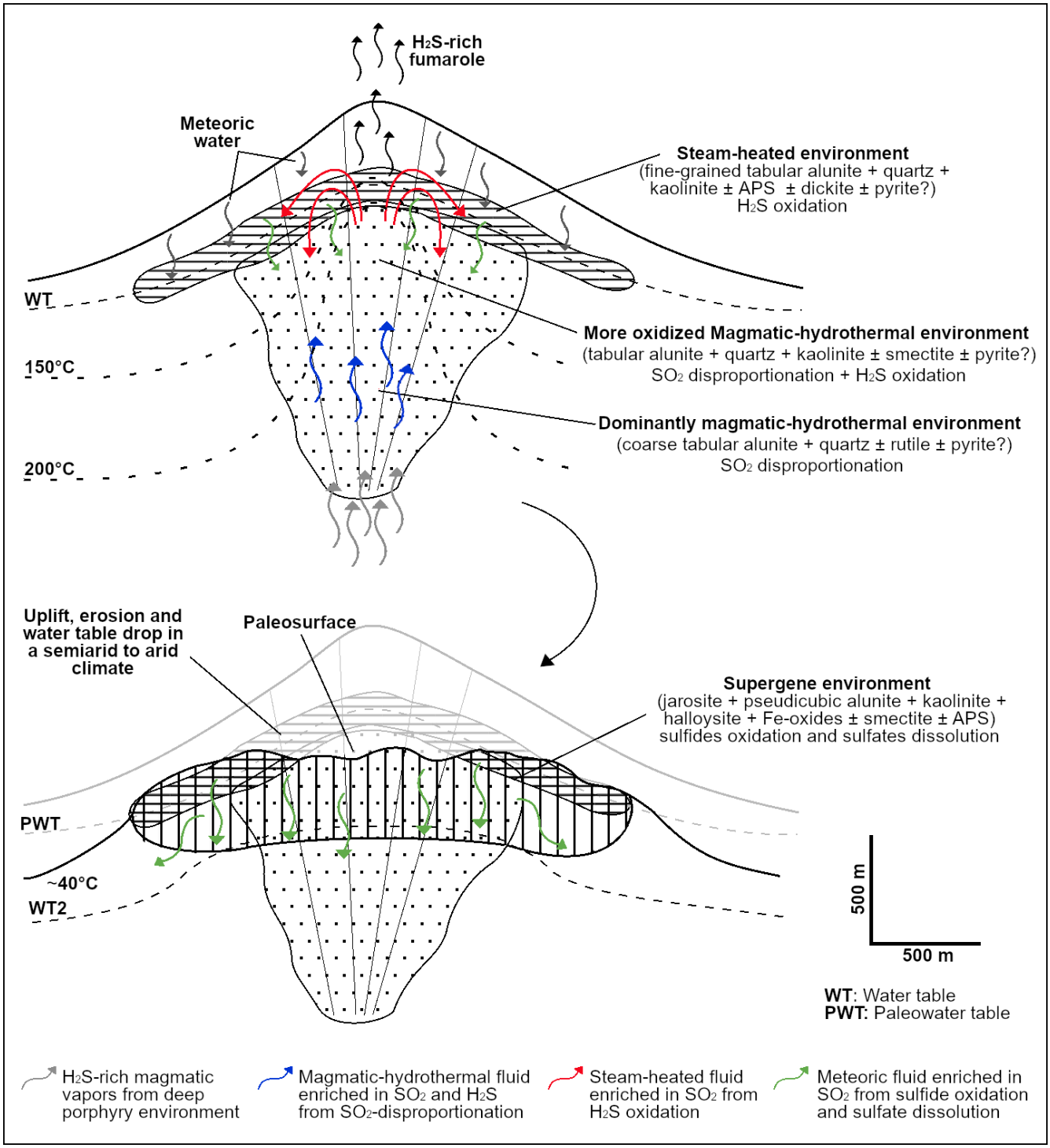
Hydrothermal activity genetically related to pluton intrusion during the Incaic tectonic phase affected the Potrerillos district in the Chilean Precordillera at the southern edge of the Atacama Desert which produced acidic fluid-driven alteration lithocaps, later overprinted by supergene processes. Outcrops with advanced argillic (AA) alteration commonly contain alunite supergroup minerals (e.g., alunite, jarosite, aluminum-phosphate-sulfate [APS] minerals) of both hypogene and supergene origin, indicating overprinting of multiple alteration events over the system's lifespan. Morphology, textural patterns, and composition of alunites, jarosite, and APS minerals related to hypogene and supergene processes in the Potrerillos district were investigated as proxies for physicochemical parameters as well as composition and source of the fluids producing AA alterations in the area. Field observations, optical microscopy, scanning electron microscopy (SEM), X-ray diffraction, short-wave infrared spectroscopy, in situ mineral chemistry (μ-XRF, EPMA), and sulfur isotope analyses define three different types of fluids, each related to a specific depositional environment (Fig. 1): The first is a magmatic-hydrothermal environment dominated by acidic fluids of magmatic origin, with sulfur species derived from SO_2 disproportionation at depth. This environment is characterized by a mineral assemblage consisting of tabular-shaped alunite, quartz, and traces of Ti oxides, indicative of precipitation at temperatures >200°C. In shallow and oxidized environments, the magma-derived fluids mix with meteoric water, producing fluid containing SO_4^{2-} originating from both disproportionation at depth and H_2S-oxidation from the vadose zone, thereby triggering the precipitation of tabular-shaped alunite, quartz, kaolinite, and small amounts of smectite. The second is a steam-heated, acidic fluid-dominated environment at a temperature of 150° to 200°C with highly oxidized and high-PO_4^{3-} activity solutions (pH ~1 to <5) containing sulfate generated upon oxidation of distilled H_2S in the vadose zone. This environment is characterized by fine-grained tabular alunite associated with quartz, kaolinite, traces of dickite, and APS minerals with svanbergite-woodhouseite composition. The third is an oxidized supergene environment with acid-sulfate meteoric waters (pH ~1), characterized by an assemblage of jarosite, pseudocubic alunite, kaolinite, halloysite, Fe-oxides, smectite, and low amounts of APS minerals. The acidic solutions therein originate from sulfuric acid produced upon oxidation of sulfide minerals and dissolution of other minerals such as alunite and APS minerals at the time of rock leaching, resulting in enrichment of the supergene fluid in Al^{3+}, Fe^{3+}, Fe^{2+}, H^+, K^+, SO_4^{2-}, Ca^{2+}, Sr^{2+}, and PO_4^{3-}. The textural relationships between alunite, jarosite, and APS minerals and SEM analyses suggest variations in physical and chemical characteristics of the fluid in the steam-heated and supergene environments. Multiple events responsible for AA alteration in the studied area have induced complex evolution of an epithermal system that grades from an early magmatic-hydrothermal stage to late leaching processes driven by meteoric waters. Furthermore, the capability of alunite supergroup minerals to record environmental physicochemical changes renders them suitable for the evolution of epithermal systems to be studied, including both hypogene and supergene processes.

Figure 1: Acid environments in the studied area. A) Magmatic-hydrothermal and steam-heated environments. B) Supergene environment shows the extinct epithermal system being leached by acid-meteoric fluids during supergene alteration.


H2S-rich fumarole
Meteoric water
Steam-heated environment
(fine-grained tabular alunite + quartz + kaolinite ± APS ± dickite ± pyrite?)
H2S oxidation
WT
150°C
More oxidized Magmatic-hydrothermal environment
(tabular alunite + quartz + kaolinite ± smectite ± pyrite?)
SO2 disproportionation + H2S oxidation
200°C
Dominantly magmatic-hydrothermal environment
(coarse tabular alunite + quartz ± rutile ± pyrite?)
SO2 disproportionation
Uplift, erosion and water table drop in a semiarid to arid climate
Paleosurface
Supergene environment
(jarosite + pseudicubic alunite + kaolinite + halloysite + Fe-oxides ± smectite ± APS)
sulfides oxidation and sulfates dissolution
PWT
~40°C
WT2
500 m
500 m
WT: Water table
PWT: Paleowater table
H2S-rich magmatic vapors from deep porphyry environment
Magmatic-hydrothermal fluid enriched in SO2 and H2S from SO2-disproportionation
Steam-heated fluid enriched in SO2 from H2S oxidation
Meteoric fluid enriched in SO2 from sulfide oxidation and sulfate dissolution