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Geochemical, isotopic, and mineralogical constraints on atmospheric deposition in the hyper-arid Atacama Desert, Chile

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Abstract

Modern atmospheric deposition across the Atacama was collected by an array of dust traps that stretched from the Pacific coast to the Andean altiplano, and the material was analyzed for its geochemical, mass and isotopic composition. The coastal trap had the second-highest insoluble mineral particle and highest soluble salt deposition rates due to significant inputs from the Morro Mejillones Range and the Pacific Ocean, respectively. The Andean trap had the highest insoluble mineral particle deposition owing to transport of weathered material, but the lowest deposition rate of soluble salts due to its distance from the ocean and anthropogenic sources. The removal of oceanic material was effective by the coastal mountains, while the westward transport of the Andean material was determined to be minimal. The atmospheric deposition in the inland traps was mainly from the local entrainment of surface material, inland anthropogenic emissions, and transport of marine aerosols. The nitrate isotopes ($\delta^{15}N$ and $\Delta^{17}O$) suggested that NO_x sources and NO_3^- chemistry shifted along the west–east transect, and were greatly impacted by anthropogenic emissions with soil NO_3^- being a minor source of deposited nitrogen.

1. INTRODUCTION

The influence of atmospheric deposition on soil development can be enhanced in arid environments and may be the dominant soil formation mechanism in hyper-arid regions such as the Atacama Desert. The Atacama Desert in northern Chile is one of the driest places on Earth, where the water limitation results in extremely low levels of organic matter and microorganisms in the soils, and little or no plant life across much of the desert (Ericksen, 1981; Navarro-Gonzalez et al., 2003; Quinn et al., 2005). These conditions restrict normal soil formation processes such as weathering, leaching, mass/chemical transport, and biological transformations. Massive nitrate deposits, rare iodate and perchlorate salts, and chloride, sulfate, and borate salts are ubiquitous in the Atacama (Ericksen, 1981). A wide range of theories have speculated on the origins of the salt deposits, but recent stable isotope evidence has indicated that a significant portion of the Atacama's nitrate, sulfate, and perchlorate salts are photochemically produced and deposited to the soil from the atmosphere (Böhlke et al., 1997; Bao et al., 2004; Michalski et al., 2004). This suggests that net mass gains from the atmospheric deposition of dust, including water-insoluble mineral particles and associated water-soluble salts, may be the major soil development mechanism in the Atacama (Dan and Yaalon, 1982; Gerson and Amit, 1987; Quade et al., 1995; Capo and Chadwick, 1999; Ewing et al., 2006; Amit et al., 2007).

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Atmospheric deposition also aids in the development of desert pavements, a ubiquitous geomorphic feature in arid environments. Desert pavements are characterized by a layer of closely packed, interlocking angular or rounded pebble- and cobble-sized clasts that protect the surface from wind erosion (Cooke, 1970). Insoluble mineral particles and soluble salts deposited from the atmosphere can fill between and beneath surface rock fragments, promoting the development and uplift of the pavement (McFadden et al., 1987; Anderson et al., 2002). In addition to dense desert pavements, gypsum crusts have developed in the Atacama. These gypsum crusts could potentially allow fine dust to migrate below the crust surface, trapping atmospherically deposited material in a fashion similar to desert pavements. This desert pavement/gypsum crust theory also suggests that atmospheric deposition is a key mechanism for soil formation in arid systems. Therefore, assessing the rates at which different types of material are deposited from the atmosphere is important for understanding soil formation in arid environments in general, and in the Atacama Desert in particular (McFadden et al., 1987; Reheis and Kihl, 1995).

While it is clear that atmospheric deposition plays an essential role in the Atacama's soil formation process, there still remains uncertainties about the rates, ionic composition, and sources of salts deposited in the Atacama. Rech et al. (2003) used strontium and sulfur isotopes to indicate that the Atacama gypsum/anhydrite development is mainly impacted by marine aerosols at coastal sites and the eolian reworking of Andean salar salts at inland sites, but the deposition of other material was not investigated. Ewing et al. (2006) analyzed the atmospheric deposition collected at three sites, but focused on evaluating atmospheric element flux along a north–south rainfall gradient without quantitative constraints of the relative importance of the origins of the atmospheric deposition and their importance as a function of distance inland from the coast. Also, the effect of aerosol size and composition on dry deposition rates and the role of fogs in wet deposition in the Atacama have not been systematically discussed. The extent of surface material that is recycled and the net dust flux across the Atacama remains unknown, as is the impact of modern human activities (*e.g.* mining, fossil fuel burning) on atmospheric deposition. Addressing these uncertainties is important for understanding how atmosphere-soil interactions influence soil formation in hyper-arid regions such as the Atacama. This may have implications for understanding surface processes on other planets, such as Mars where soil development in the absence of water has occurred for the past 600 million years (Pike et al., 2011).

In this study, we assessed the composition, rates and potential sources of atmospheric deposition along a west–east transect in the Atacama Desert. The objective was to investigate the spatial variations in modern atmospheric deposition characteristics such as mineralogy, ionic content and accumulation rates in order to explore atmospheric deposition of material from different sources that can potentially influence soil development. Previous studies indicated that there are two categories of material in atmospheric deposition in the Atacama (Rech et al., 2003; Michalski et al., 2004; Ewing et al., 2006). The first is primary aerosols (*i.e.* eolian material) consisting of marine aerosols from the adjacent Pacific Ocean, surface material (*e.g.* surface soil, crust, and playa salts) from local entrainment, weathered mountain material, long-range-transported dust, and direct volcanic emissions (Rech et al., 2003; Mather et al., 2004; Stuut et al., 2007). The second category is secondary aerosols such as nitrates and sulfates, produced from reactive atmospheric gases (Michalski et al., 2004; Ewing et al., 2006). We hypothesize that eolian material can be carried from different source regions by winds and deposited to a west–east transect, and the relative importance of each source is a function of its proximity to a given collector. We speculate that the distribution of secondary aerosols in the atmosphere is determined by the regional emissions of precursor gases, the residence time for converting the gases into aerosols, and the removal efficiencies of the resulting aerosols along their transport paths. In modern times, human activities are hypothesized to exert a significant enhancement in the production and deposition of secondary aerosols in the Atacama basin.

2. SAMPLING AND ANALYSIS METHODS

The physiography of the Atacama Desert consists of three major geologic units going from west to east: the Coastal Range, the Central Valley and the Andes (Figs. 1 and 2). The Coastal Range is a mountain range running north and south that abuts the Pacific Ocean, has altitudes generally less than 2000 m, and is mantled by Jurassic volcanic sequences imbedded with marine and continental conglomerates (SERNAGEOMIN, 2003). The Central Valley is a longitudinal depression typically with altitudes between 900 and 2500 m, comprising a few hundred meters thick layers of Upper Miocene-Pliocene piedmont clastic sediments (SERNAGEOMIN, 2003). The Andes consist of an altiplano about 4000 m in altitude surmounted by hundreds of volcanic peaks (~5000 m), and a succession of parallel Pre-Andes mountain ranges (*i.e.* the Cordillera Central in the north and the Cordillera Domeyko in the south) with intervening valleys and basins; the Andes are predominately underlain by Pliocene-Quaternary volcanic flow, tuffs, and breccia rocks (SERNAGEOMIN, 2003).

An array of ten dust traps (T1–T10) were installed along a west–east transect across the Atacama to investigate the spatial variations of atmospheric deposition in the desert (Figs. 1 and 2). The selection of the dust trap locations was based on the accessibility, absence of dirt roads or other artificially disturbed areas upwind, and inconspicuousness. The traps were mostly placed in flat, relatively open areas every 10–50 km inland from the western Pacific coast (see Supplementary Information for site pictures). The traps consisted of a single-piece Bundt cake pan (outer ring diameter: 25 cm, surface area: 477 cm^2) fitted with a circular piece of 0.25-inch mesh galvanized screen on which a layer of pre-washed glass marbles were placed to mimic desert pavements (Reheis and Kihl, 1995). The traps were mounted on ~1 m high poles above the ground to eliminate most saltating particles and left exposed from 7/10/2007 to 1/1/2010 (915 days). Trap 9 was destroyed during this period and is thus excluded from this study.