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Sources of pollution and interrelationships between aerosol and precipitation chemistry at a central California site

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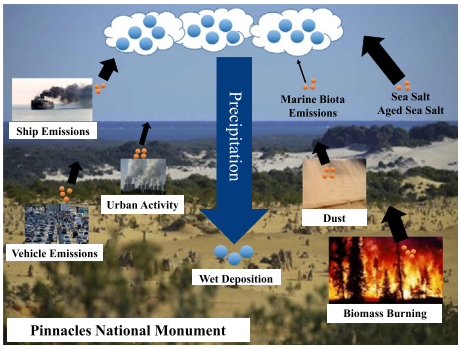
HIGHLIGHTS

- Characterization of aerosol and precipitation chemistry at a central California site
- Co-located aerosol and wet deposition data examined between 2010 and 2016.
- Positive matrix factorization and concentration weighted trajectory maps were used.
- Seven sources were identified with temporal profiles dependent on meteorology.
- Insights were gained into aerosol-precipitation interactions.

GRAPHICAL ABSTRACT



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ABSTRACT

This study examines co-located aerosol and precipitation chemistry data between 2010 and 2016 at Pinnacles National Monument ~65 km east of the coastline in central California. Positive matrix factorization analysis of the aerosol composition data revealed seven distinct pollutant sources: aged sea salt (25.7% of $PM_{2.5}$), biomass burning (24.2% of $PM_{2.5}$), fresh sea salt (15.0% of $PM_{2.5}$), secondary sulfate (11.7% of $PM_{2.5}$), dust (10.0% of $PM_{2.5}$), vehicle emissions (8.2% of $PM_{2.5}$), and secondary nitrate (5.2% of $PM_{2.5}$). The influence of meteorology and transport on monthly patterns of $PM_{2.5}$ composition is discussed. Only secondary sulfate exhibited a statistically significant change (a reduction) over time among the $PM_{2.5}$ source factors. In contrast, PM_{coarse} exhibited a significant increase most likely due to dust influence. Monthly profiles of precipitation chemistry are summarized showing that the most abundant species in each month was either SO_4^{2-}, NO_3^-, or Cl^-. Intercomparisons between the precipitation and aerosol data revealed several features: (i) precipitation pH was inversely related to factors associated with more acidic aerosol constituents such as secondary sulfate and aged sea salt, in addition to being reduced by uptake of HNO_3 in the liquid phase; (ii) two aerosol source factors (dust and aged sea salt) and PM_{coarse} exhibited a positive association with Ca^{2+} in precipitation, suggestive of directly emitted aerosol types with larger sizes promoting precipitation; and (iii) sulfate levels in both the aerosol and precipitation samples analyzed were significantly correlated with dust and aged sea salt PMF factors, pointing to the partitioning of secondary sulfate to dust and sea salt particles. The results of this work have implications for the region's air quality and hydrological cycle, in addition to demonstrating that the use of co-located aerosol and precipitation chemistry data can provide insights relevant to aerosol-precipitation interactions.

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1. Introduction

The largest uncertainty in quantifying global anthropogenic radiative forcing is linked to interactions of aerosol particles with clouds (IPCC, 2013), which is partly driven by the difficulty of conducting the required measurements and separating the influence of meteorology and aerosol pollution on clouds. The impact of aerosol particles on precipitation is challenging to address but is important for reasons extending from improving model representation of clouds and precipitation to identifying the impacts of wet deposition on aquatic and terrestrial ecosystems due to inputs of nutrients and contaminants that may have originated from particles serving as cloud condensation nuclei (CCN) or ice nuclei (IN). Precipitation has important effects on aerosol particles owing to scavenging and removing them from the air, in addition to also removing gases (e.g., MacDonald et al., 2018). In this regard, monitoring the composition of wet deposition is important not just for impacts on ecosystems (e.g., Bobbink et al., 1998; Driscoll et al., 2003; Pardo et al., 2011), but for evaluating chemical tracer transport models (e.g., Rodhe et al., 1995; Liu et al., 2001). As observational studies of aerosol-cloud-precipitation interactions relying on airborne in situ measurements and remote sensing have limitations (e.g., cost, statistics, temporal resolution and coverage), alternative methods of examining this complex system can provide much needed insight. One such method is to rely on long-term records of surface-based aerosol and precipitation chemistry data, as has been demonstrated in just a few studies focused on regions such as the southwestern United States (Sorooshian et al., 2013) and Mexico City (Mora et al., 2017). Examining simultaneously collected aerosol and precipitation chemistry data can provide knowledge about topics with important implications including (i) sources of pollutants, (ii) potential interactions between gases and particles with precipitation drops including uptake processes and wet scavenging, and (iii) aerosol types impacting clouds that produce the precipitation.

Aerosol-cloud-precipitation interactions have been the subject of extensive field projects across California, including for stratiform cloud decks off the coast (e.g., Russell et al., 2013; Sorooshian et al., 2018) and for the more convective clouds inland that provide important rainfall for the state (e.g., Fan et al., 2014; Ralph et al., 2016), including intense precipitation events associated with wintertime atmospheric rivers (e.g., Ralph et al., 2004, 2013). Many of these studies have tried to provide details about sources of pollution impacting the clouds and what the sign of the precipitation response was due to an increase in aerosol concentration. This study aims to fill a gap by examining for the first time long term datasets for aerosol and precipitation chemistry at a central California site, with results that can be contrasted with findings from other works that have relied on aircraft and remote sensing.

This study reports on long-term data collected at Pinnacles National Monument, which is distinguished from most other surface monitoring areas in California because of the co-location of monitoring instruments for both aerosol and precipitation chemistry for a long-term period. Another motivation for studying this site is that it represents an area impacted by a diverse set of sources (e.g., urban, agriculture, marine, shipping, wildfires, dust) due to its geographical position. Focus is first placed on a comprehensive analysis of the aerosol composition data to characterize temporal trends, sources of the pollutants, and the impact of meteorology and transport. Subsequently, the precipitation chemistry data are summarized and then compared with the aerosol composition data.

2. Methods

2.1. Site description

California is of interest in this study owing to its large population (~36.5 million as of 1 July 2017; US Census Bureau; https://www.census.gov/) that is impacted in various ways by aerosol pollution and precipitation. The Pinnacles National Monument (PNM) site was chosen for this analysis owing to its geographic location as an inland California site impacted by a diversity of sources (Fig. 1), and because there are co-located aerosol and precipitation monitoring stations operated by the Environmental Protection Agency (EPA) Interagency Monitoring of Protected Visual Environments (IMPROVE) network and the National Atmospheric Deposition Program (NADP) National Trends Network (NTN), respectively. The PNM site is ~65 km east of the ocean and ~115 km south of San Jose (Fig. 1). The California coastal range separates the marine atmosphere from the urban atmosphere over major cities

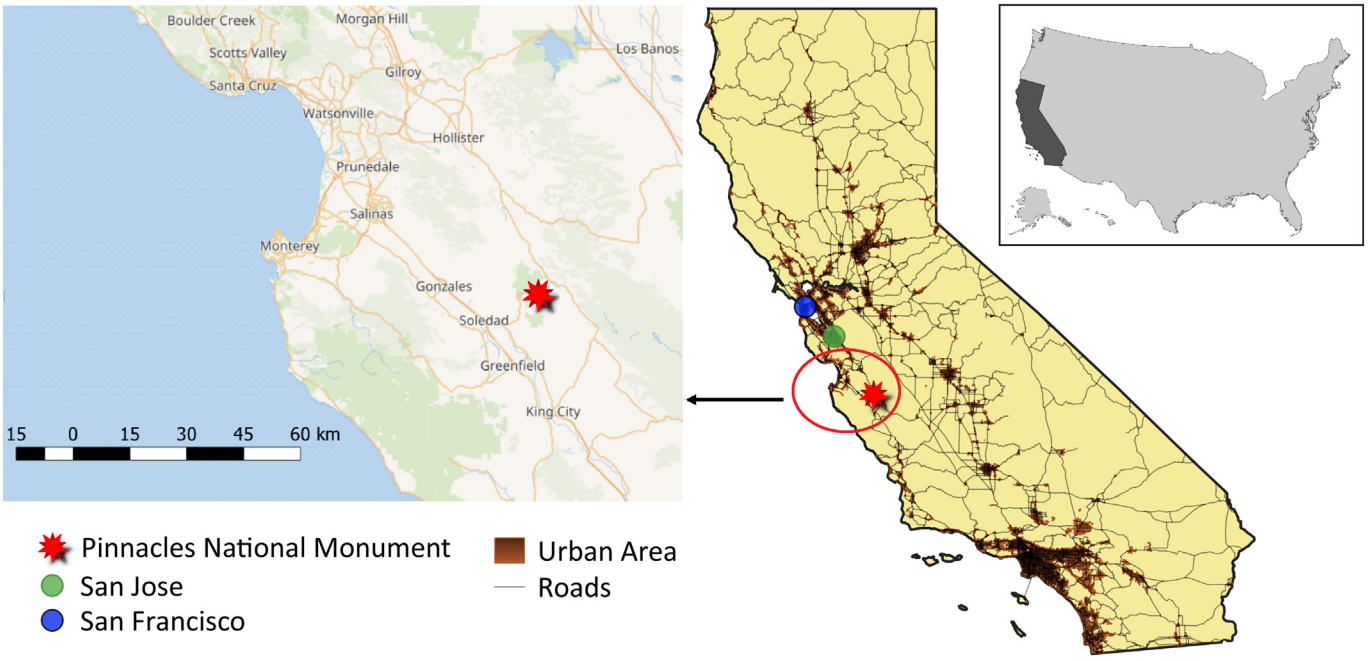

Fig. 1. Map showing the study region, including where the collocated IMPROVE and NADP/NTN stations are at Pinnacles National Monument relative to major urban centers and the coastline.