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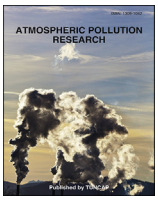

Source identification of coarse particles in the Desert Southwest, USA using Positive Matrix Factorization

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ABSTRACT

The Desert Southwest Coarse Particulate Matter Study was undertaken to further our understanding of the spatial and temporal variability and sources of fine and coarse particulate matter (PM) in rural, arid, desert environments. Sampling was conducted between February 2009 and February 2010 in Pinal County, AZ near the town of Casa Grande where PM concentrations routinely exceed the U.S. National Ambient Air Quality Standards (NAAQS) for both PM_{10} and $PM_{2.5}$. In this desert region, exceedances of the PM_{10} NAAQS are dominated by high coarse particle concentrations, a common occurrence in this region of the United States. This work expands on previously published measurements of PM mass and chemistry by examining the sources of fine and coarse particles and the relative contribution of each to ambient PM mass concentrations using the Positive Matrix Factorization receptor model (Clements et al., 2014).

Coarse particles within the region were apportioned to nine sources including primary biological aerosol particles (PBAPs - 25%), crustal material (20%), re-entrained road dust (11%), feedlot (11% at the site closest to a cattle feedlot), secondary particles (10%), boron-rich crustal material (9%), and transported soil (6%), with minor contributions from ammonium nitrate, and salt (considered to be NaCl). Fine particles within the region were apportioned to six sources including motor vehicles (37%), road dust (29%), lead-rich (10%), with minor contributions from brake wear, crustal material, and salt. These results can help guide local air pollution improvement strategies designed to reduce levels of PM to below the NAAQS.

1. Introduction

The U.S. National Ambient Air Quality Standards (NAAQSs) were established to protect human health from the damaging effects of air pollution (Federal Register, 2006). Numerous studies have demonstrated a link between ambient particulate matter (PM) and adverse human health effects (e.g. Peters et al., 2000; Mar et al., 2000; Anderson, 2009; Solomon et al., 2012; Calvo et al., 2013). Two air quality standards for particulate matter, one for PM_{10} (particles with an aerodynamic diameter [AD] less

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than or equal to a nominal 10 μm) and another for $PM_{2.5}$ (particles with an AD less than or equal to a nominal 2.5 μm) have been established to protect human health and welfare. PM_{10} is the sum of $PM_{2.5}$, also referred to as fine particulate matter (PM_f), and the coarse fraction (PM_c; particles with AD in the size range between 2.5 and 10 μm AD). Many counties in the southwest United States have measured ambient concentrations of PM_{10} in excess of the Federal standards. Exceedances of the PM_{10} NAAQS in the southwest US are often the result of spikes in the PM_c mass concentration because coarse particles dominate the overall PM_{10} mass within the region (Pinker et al., 2004; Cheung et al., 2011; Engelbrecht et al., 2015). When a region is out of compliance with the NAAQS, it is the responsibility of the State to develop a State Implementation Plan (SIP) designed to reduce PM concentrations to levels below the NAAQS (Federal Register, 2006). The development of a SIP requires knowledge of emission sources, emission strength, spatial and temporal variation in ambient PM concentrations, and source impacts.

In rural areas like the desert Southwest US, elevated coarse particle concentrations are often attributed to entrainment of crustal material from the arid native landscape leading to assumption that little can be done to manage periodic high concentrations of PM. Previous studies have found links between the entrainment of soil dust and weather conditions (Brazel and Nickling, 1986), wind speed (Holcombe et al., 1997; Hagen, 2004), soil type (Macpherson et al., 2008), as well as with soil moisture content (Ellis et al., 2006). Several studies have observed that crustal sources, in particular local sources, are major contributors to coarse particle mass concentrations in the desert southwest (Watson and Chow, 2001; Cheung et al., 2011; Upadhyay et al., 2011; Prabhakar et al., 2014; Gonzalez-Maddux et al., 2014).

Source apportionment, using tools like Positive Matrix Factorization (PMF) or Chemical Mass Balance (CMB), has been widely used to quantify the sources of PM in a variety of environments (Aldabe et al., 2011; Gummeneni et al., 2011; Banerjee et al., 2015). Source apportionment has also been used to distinguish between several similar soil types – for example, road dust and agricultural soils (Paode et al., 1999; Hwang et al., 2008). Other studies have measured the composition of soil source materials to identify chemical species unique to that source or source type to further distinguish among soil types (Rogge et al., 2007, 2012; Upadhyay et al., 2015). However, more work is needed to understand the magnitude of the crustal source which originates from mechanical entrainment of material to the environment that is measured specifically as PM_c, and to understand if some routes of crustal emissions, such as fugitive dust entrainment from agricultural activities, could be partially controlled through altered practices as part of a strategy to reduce PM levels below the PM NAAQS in areas where coarse particles drive the exceedance of the PM NAAQS. A few studies have been conducted within the Southwest US to better understand the range of source impacts, beyond crustal material, and their relative contribution to ambient particle concentrations (Gertler et al., 1995; Watson and Chow, 2001).

The Desert Southwest Coarse Particulate Matter Study was conducted in and around the town of Casa Grande in Pinal County, Arizona (Clements et al., 2013, 2014). This area experiences some of the highest PM_{10} concentrations in the region (U.S. EPA, 2014). This paper expands on work already published from this study (Clements et al., 2013, 2014; Upadhyay et al., 2015) by detailing the PMF modeling results for PM_c and PM_f within the region including source profiles, source identification, and spatially resolved yearly averaged and seasonally resolved source contributions to PM_f and PM_c mass within the region.

2. Methods

2.1. Ambient sampling and chemical characterization

PMF modeling was applied to the 1-in-6 day data set derived from filter-based ambient aerosol samples collected between February 2009 and February 2010 in Pinal County, AZ located south of Phoenix, AZ. Detailed information about the ambient sampling locations can be found in Clements et al. (2014). Briefly, samples were collected at three ambient monitoring locations on a common 1-in-6 day schedule. These included the town of Casa Grande (CG; population of roughly 50,000), a small urban location located in a business district more distantly surrounded by residential neighborhoods with trees; Cowtown (COW), a rural site located approximately 27 km to the northwest of the city of Casa Grande locally impacted by a cattle feedlot, a grain processing operation, and railroad and vehicle traffic, and agricultural fields; and Pinal County Housing (PCH), a rural site more closely impacted by active agricultural fields that is approximately 19 miles east of Casa Grande.

Each sampling site was equipped with three Sierra-Anderson Model 241 dichotomous samplers. These samplers simultaneously collected equivalent 24-hr samples of PM_f and PM_c on a one-in-six day schedule. Two of the three samplers employed Teflon filters in both PM_f and PM_c channels whereas the third sampler employed quartz-fiber filters. PM mass, ion, and elements were determined from the Teflon filters and elemental carbon (EC) and organic carbon (OC) from the quartz-fiber filters. Ambient concentrations were obtained by dividing the amount of material quantified on each filter by the sampled air volume.

PM mass concentrations were determined gravimetrically by difference between the post and pre-sample collection weights from each Teflon filter. The mean of the paired colocated PM_c filter and paired PM_f filter weights were used as the mass estimate unless one measurement was invalidated as specified in the project quality assurance plan. After gravimetric analysis, one set of Teflon filters was individually wetted with 150 μl of ethanol (Fisher HPLC grade), extracted into 10 ml of ultrapure water by ultrasonic agitation for 15 min, and analyzed the same day. Sample extracts were analyzed for five anion and five cation species by ion chromatography (Dionex IC20 system with CG12A and AS12A analytical columns). The second set of Teflon filters was analyzed for trace metals by the method described in Upadhyay et al. (2015). Briefly, filters were microwave digested into a 20 ml mixture of nitric, hydrochloric, and hydrofluoric acid and then analyzed for 63 trace elements by high-resolution inductively coupled plasma mass spectrometry (HR-ICP-MS, herein referred to as ICP-MS, Thermo Finnigan ELEMENT 2). OC and EC were determined from a 1 cm × 1.5 cm punch from the quartz-fiber filters by an analytical method slightly modified from Birch and Cary (1996) using a thermal-optical carbon analyzer (Sunset Laboratories, Tigard OR); details of the method modification can be found elsewhere (e.g. Clements et al., 2014 or Mancilla et al., 2015).

2.2. Positive Matrix Factorization framework

PMF modeling was conducted on the 64 daily samples collected over the 1-in-6 day sampling schedule using the EPA PMF model version 3.0 (U.S. EPA, 2008). This multivariate factor analysis tool decomposes the matrix consisting of the concentration of airborne components in ambient samples into two different matrices – one representing source profiles and the other representing the source contribution of each source profile to the measured PM mass. The contribution from each source profile is constrained to non-negative values.

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