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# Back-trajectory-based source apportionment of airborne sulfur and nitrogen concentrations at Rocky Mountain National Park, Colorado, USA

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ABSTRACT

The Rocky Mountain Atmospheric Nitrogen and Sulfur Study (RoMANS), conducted during the spring and summer of 2006, was designed to assess the sources of nitrogen and sulfur species that contribute to wet and dry deposition and visibility impairment at Rocky Mountain National Park (RMNP), Colorado. Several source apportionment methods were utilized for RoMANS, including the Trajectory Mass Balance (TrMB) Model, a receptor-based method in which the hourly measured concentrations are the dependent variables and the residence times of back trajectories in several source regions are the independent variables. The regression coefficients are estimates of the mean emissions, dispersion, chemical transformation, and deposition between the source areas and the receptors. For RoMANS, a new ensemble technique was employed in which input parameters were varied to explore the range, variability, and model sensitivity of source attribution results and statistical measures of model fit over thousands of trials for each set of concentration measurements.

Results showed that carefully chosen source regions dramatically improved the ability of TrMB to reproduce temporal patterns in the measured concentrations, and source attribution results were also very sensitive to source region choices. Conversely, attributions were relatively insensitive to trajectory start height, trajectory length, minimum endpoints per source area, and maximum endpoint height, as long as the trajectories were long enough to reach contributing source areas and were not overly restricted in height or horizontal location. Source attribution results estimated that more than half the ammonia and 30–45% of sulfur dioxide and other nitrogen-containing species at the RoMANS core site were from sources within the state of Colorado. Approximately a quarter to a third of the sulfate was from within Colorado.



## 1. Introduction

From 1985 to 2002, the concentrations of inorganic nitrate and ammonium in wet deposition increased approximately 25% and 75%, respectively, while the concentrations of sulfur compounds in wet deposition decreased about 40% at Rocky Mountain National Park, Colorado (RMNP) (Lehmann et al., 2005). Increasing deposition of nitrogen compounds has the potential to cause irreversible ecosystem changes (Porter et al., 2005). Additionally, airborne particulate matter, including ammonium sulfate and ammonium nitrate, causes visibility impairment in this protected area. These issues motivated the 2006 Rocky Mountain Atmospheric Nitrogen and Sulfur Study (RoMANS) (Malm et al., 2009; Levin et al., 2009; Beem et al., 2010; Rodriguez et al., 2011), which was designed to better determine the sources of these compounds. The field study consisted of two seasons of data collection to characterize airborne concentrations and wet and dry deposition during the two climatologically based peak precipitation and wet deposition seasons in northern Colorado east of the Continental Divide. Late March through April is the peak season for large-scale storms, while July through early August, is the peak season for convective storms.

As shown in Fig. 1, RMNP is located along the Continental Divide in northern Colorado. The prevailing wind direction in this area is westerly, but the largest population centers and agricultural activities are east of the park. The most densely populated region of the state is the Front Range Urban Corridor (FRUC), running north-south through the center of the state from southern Wyoming to

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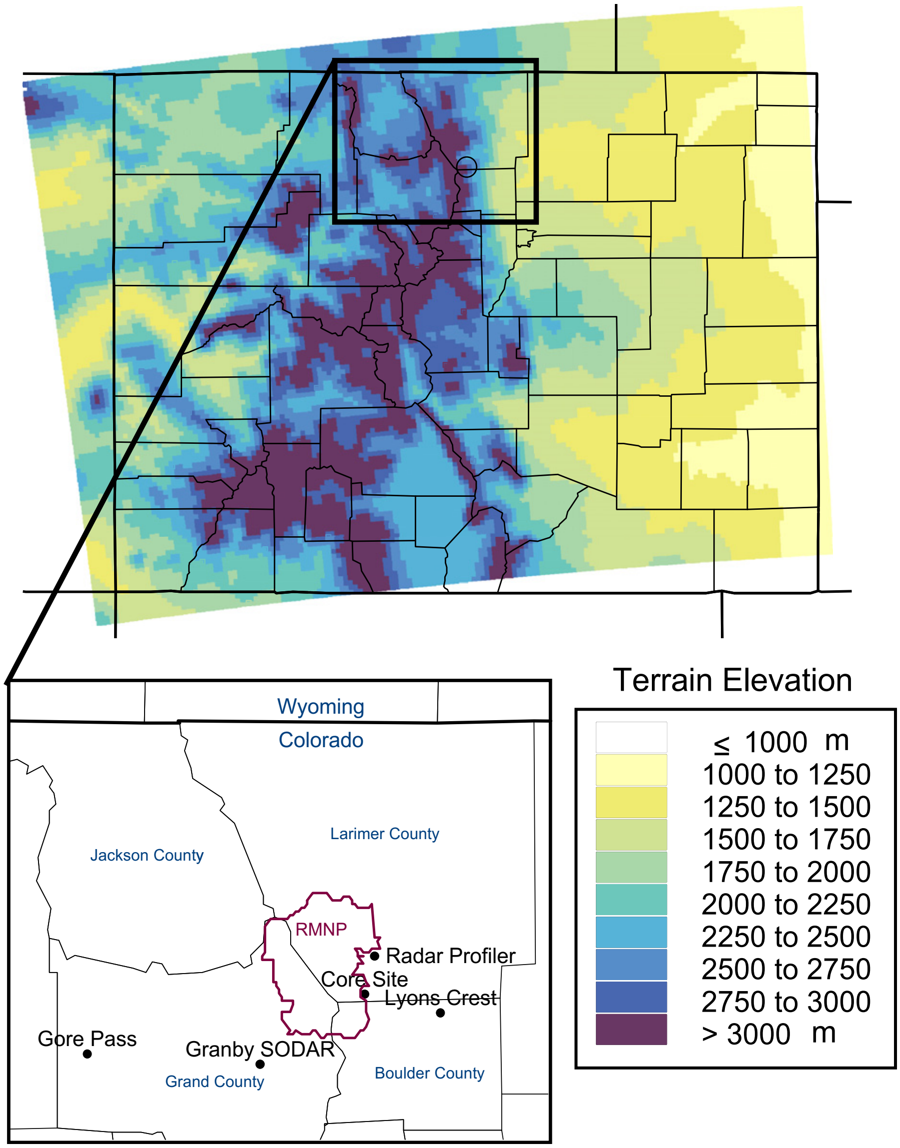


**Fig. 1.** The top map shows Colorado county boundaries and terrain heights used in the mesoscale meteorological model on the 4-km domain. The heavily outlined area in north-central Colorado is expanded on the bottom to show the approximate boundaries of Rocky Mountain National Park and locations of the RoMANS core site and other meteorological monitoring sites.

southern Colorado along a line where the terrain transitions between the foothills of the Rocky Mountains and the high plains of eastern Colorado. The city and county of Denver, the small county shown in Fig. 1 near the southeast corner of the heavy box, is the largest city, with a population of approximately 600,000. The population of the seven-county Denver metropolitan area is approximately 3 million, while the entire FRUC has about 4.3 million people. The eastern plains are more sparsely populated but have increasing oil and gas extraction and more agriculture, including fertilized crops and the state's largest confined animal feeding operations.

The core measurement site for RoMANS was located on the eastern slope of the park, which straddles the Continental Divide. East of this site, the terrain drops rapidly to the relatively flat eastern half of the state, with elevations of 1500–1800 m, while to the west are peaks of up to 4300 m above mean sea level. At the core measurement site, easterly upslope wind flow can occur due to either diurnal mountain valley circulations with winds near the surface blowing up valley during the day and reversing at night, or by synoptic weather patterns including low-pressure (counter-clockwise circulation) to the south or west or high pressure (clockwise circulation) to the north.

Recognizing that each model has different strengths and limitations, several source apportionment methods were utilized for RoMANS. These included a deterministic mesoscale chemical transport model, descriptive statistical back trajectory techniques, a hybrid deterministic-receptor model, and the Trajectory Mass Balance (TrMB) Model, which is the topic of this paper.

## 2. Methodology

### 2.1. Trajectory mass balance

The Trajectory Mass Balance (TrMB) Model (Pitchford and Pitchford, 1985; Malm, 1992; Gebhart et al., 2006) assumes that measured concentrations at a receptor are linearly related to the frequency of air mass transport from source areas to the receptor by the following relationship: