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Association of airmass transport patterns and particulate sulfur concentrations at Big Bend National Park, Texas

Bret A. Schichtel*, Kristi A. Gebhart, Michael G. Barna, William C. Malm

National Park Service, Cooperative Institute for Research in the Atmosphere, Colorado State University, Fort Collins, CO 80523-1375, USA

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Abstract

The Big Bend Regional Aerosol and Visibility Observational (BRAVO) study was initiated to understand the causes of haze at Big Bend National Park. BRAVO included the measurement of aerosols throughout Texas from July to October 1999 and extensive modeling of these aerosols. In support of BRAVO, the potential contributions from source regions to particulate sulfur at Big Bend during the BRAVO period were examined via an airmass history analysis. This was done using residence time analysis and a new technique of decomposing the residence time probability density function into its basic components, an airmass transport directional frequency and inverse characteristic transport speed. Trajectory heights over potential source regions were also examined. The system was validated using inert perfluorocarbon tracers that were released from four Texas sites. Airmass transport to Big Bend was examined on days with high (>80th percentile), and days with low (<20th percentile), particulate sulfur. High particulate sulfur concentrations were associated with low-level and low-speed airmass transport from the eastern United States, eastern Texas, and northeastern Mexico. All three of these regions have high SO_2 emissions that could contribute to Big Bend's haze. Examination of individual trajectories showed that the highest particulate sulfur concentrations occurred when transport over several of these regions coincided. Low particulate sulfur concentrations coincided with low-level but high-speed airmass transport from the Gulf of Mexico and along the Mexico–Texas border. Precipitation often occurred along these trajectories. Low sulfur was also associated with transport from low SO_2 emission regions north and west of Big Bend. Days with high SO_2 or selenium concentrations were also examined. High SO_2 concentrations were associated with prior transport from nearby sources, particularly the Carbón power plants located in Mexico ~230 km southeast of Big Bend. High selenium concentrations were associated with prior transport over Carbón and eastern Texas.

Keywords: BRAVO; Sulfate; Regional haze; Long-range transport; Receptor model

1. Introduction

Big Bend National Park (NP) Texas is a place of rare scenic beauty where, on clear days, a person can see well over 100 miles, viewing the deep canyons cut by the Rio Grande River and the Chisos and Sierra Del Carmen mountains extending from the Chihuahuan desert floor. Unfortunately, during the 1990s, Big Bend's visibility decreased (Malm et al., 2000). During this same period, particulate sulfate increased, with concentrations on the 20% highest sulfate days increasing by 30%;

*Corresponding author. Tel.: +1 970 491 8581; fax: +1 970 491 8598.
E-mail address: schichtel@cira.colostate.edu (B.A. Schichtel).

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this was coincident with increases in SO_2 emissions in Texas and the southeastern United States, including Arkansas, Louisiana, Mississippi, and Alabama (Malm et al., 2002), and the building of the Carbón II coal-fired electric generating stations about 225 km to the southeast of Big Bend in Mexico (Fig. 1).

To investigate the sources of emissions associated with the reduced visibility, the Environmental Protection Agency (EPA) and National Park Service (NPS) initiated a field experiment referred to as the Big Bend Regional Aerosol and Visibility Observational study (BRAVO) (Pitchford et al., 2004, 2005; Schichtel et al., 2004, 2005a). This was an intensive monitoring study that sampled aerosol physical, chemical, and optical properties from July to October 1999 at Big Bend and throughout Texas. The monitoring was followed by a multi-year assessment of the causes of haze in Big Bend NP, with the primary purpose being to identify the source regions and source types responsible for the haze at the park. Analyses focused on the attribution of particulate sulfate since it accounted for about half of the particulate light extinction (Malm et al., 2003).

Sulfate concentrations at Big Bend are primarily the result of secondary formation from anthropogenic SO_2 emissions. Most SO_2 emissions are located to the east of Big Bend NP. Southeast of Big Bend in Mexico are the Carbón I & II power plants (Fig. 1) which emit 152,000–241,000 tonnes $SO_2\,yr^{-1}$, and the urban areas of Monterrey and Reynosa which also have high emissions. In Texas, the SO_2 emissions are concentrated in the east, where there are a number of coal-fired power plants and urban centers including Houston and Dallas, which have industrial and mobile sources of SO_2. In all, Texas emits about 1 million tonnes of $SO_2\,yr^{-1}$. The largest SO_2 emissions are located in the eastern United States., i.e., states to the east of Texas, where $\sim$14 million tonnes yr^{-1} of SO_2 are emitted. The areas surrounding Big Bend and to the north and west have low SO_2 emissions.

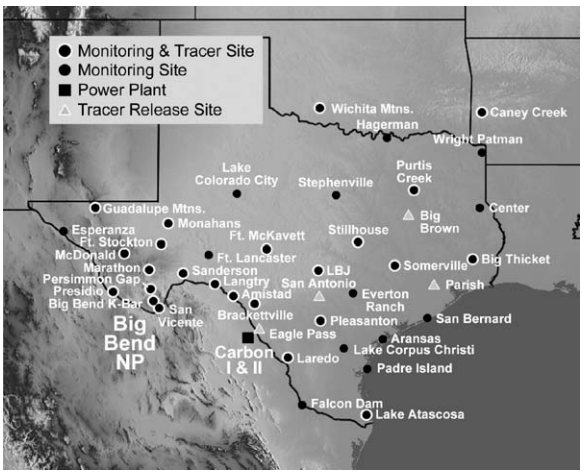

Fig. 1. The BRAVO monitoring network, perfluorocarbon tracer release sites, and terrain.

Apportionment of sulfate concentrations at Big Bend to these North American source regions was conducted using a number of qualitative and quantitative data analysis techniques, receptor models, and chemical transport models (CTMs). The CTMs were used to simulate the sulfate concentrations at Big Bend NP and other receptors and to assess the source contributions of sulfate to these receptors (Barna et al., 2005a, b; Knipping et al., 2005; Pun et al., 2005). Dispersion models were also used in forward and backward airmass history analyses to analyze airmass transport patterns and were incorporated into receptor models to apportion Big Bend's particulate sulfate to different source regions (Schichtel et al., 2004; Gebhart et al., 2005a). The source attribution techniques were compared and systematic differences were reconciled (Schichtel et al., 2005a). This led to the development of hybrid models to better account for biases in the different methods. One hybrid model was based upon an inverse modeling technique incorporating measured data and source attribution estimates from the CTMs to refine the initial model results (Schichtel et al., 2005b).

Airmass history analyses were conducted to examine the association of long-range transport pathways to Big Bend and particulate sulfur concentrations during the BRAVO study period. This was done using the residence time analysis technique (Ashbaugh, 1983; Poirot and Wishinski, 1986) which generates a probability density function identifying the likelihood that an airmass will traverse a given region en route to the receptor over a given time period. In addition, a new technique of decomposing the residence time into more basic processes based on airmass transport direction and transport speed was conducted. The decomposed residence time in conjunction with average airmass heights allowed for examining the processes leading to the residence time patterns.

The residence time analysis is a qualitative source attribution technique. The benefit of this technique