



Modeling Particulates and Direct Radiative Forcing from Urban to Synoptic Scales Downwind of Mexico City

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Mexico City is the largest metropolitan area in North America with a population of \sim 20 million. Field campaign measurements and modeling studies over the past twenty years have examined the local scale processes responsible for the high concentrations of ozone and particulates observed in the city. To examine how these pollutants affect air quality and climate downwind of Mexico City, extensive meteorological, chemical, and particulate measurements were collected during March 2006 over central Mexico and the Gulf of Mexico as part of the Megacity Initiative Local and Global Regional Observations (MILAGRO). In this study, we examine how particulate mass, composition, and size distribution evolves over several days downwind of Mexico City by combining MILAGRO measurements with simulations performed by a fully-coupled meteorology-chemistry-particulate model (WRF-Chem). Organic aerosols were observed to be a large fraction of the total particulate mass but traditional methods of simulating organic aerosols have been shown to produce too little mass. We have therefore coupled the recently developed volatility basis-set framework with the MOSAIC aerosol model within WRF-Chem to examine how well this framework represents the evolution of organic aerosols from anthropogenic and biomass burning sources. The performance of the model is evaluated with measurements obtained from surface, aircraft, and satellite instrument platforms. For example, measurements from several sunphotometers, NASA Langley's High Resolution Spectral Lidar deployed on an aircraft that flew over central Mexico, and MODIS instrument on the Terra and Aqua satellites were used to evaluate simulated aerosol optical thickness. The reduction in average net shortwave radiation over Mexico City at 18 UTC was -27 W m^{-2} , although the reduction on any given day was as large as -60 W m^{-2} . The day-to-day variation in direct radiative forcing over Mexico City depended largely upon the meteorology and the presence of biomass burning sources. The largest impact of aerosols on shortwave radiation downwind occurred over the coastal plain northeast of Mexico City. Relative humidity is usually much higher along the coast than over Mexico City, leading to the uptake of water on aerosols transported into the region. Consequently, aerosol water increased scattering considerably within the anthropogenic and biomass burning particulate plumes. Further downwind, the impact of aerosols on radiative forcing gradually decreases as the Mexico City plume becomes diluted.