



Mexico City Aerosol Analysis during MILAGRO using High Resolution Aerosol Mass Spectrometry

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Submicron aerosol was analyzed during the MILAGRO field campaign in March 2006 at the T0 urban supersite in Mexico City with a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS). Mass concentrations, diurnal cycles, and size distributions of inorganic and organic species are similar to results from the CENICA supersite in April 2003. Positive Matrix Factorization (PMF) analysis of the high resolution organic aerosol (OA) spectra identifies chemically-reduced urban primary emissions (hydrocarbon-like OA, HOA), oxygenated OA (OOA, mostly secondary OA or SOA), biomass burning OA (BBOA) that includes several very large plumes from regional fires, and a small local nitrogen-containing reduced OA component (LOA). OOA accounts for almost half of the OA on average, consistent with previous observations. Results from Chemical Mass Balance of organic molecular markers (CMB-OMM) are overall consistent with the PMF-AMS results. Two datasets of 14C analysis show significant differences, but by comparing high vs. low regional fire periods, both datasets suggest a similar BBOA contribution to the AMS and CMB results. During a low-fire period, 43% organic carbon (OC) and 30% total carbon (TC) are from modern sources, which suggest the importance of urban modern carbon sources. BBOA correlates with satellite fire counts, levoglucosan, and other fire tracers such as acetonitrile. Satellite fire counts are combined with emission and dispersion models to derive a fire impact index for T0 which captures most trends of the measured BBOA. This indicates that wildfires on the nearby mountains surrounding Mexico City are the dominant source of BBOA at T0 during MILAGRO. Comparing the concentrations of multiple tracers for the high versus low fire impact periods confirms the attribution of most BBOA to regional fires, and shows that OOA appears to be dominated by urban sources. Overall, the forest fires are estimated to contribute 15-23% of the OA and 7-9% of the fine PM at T0 during MILAGRO, and 2-3% of the fine PM as an annual average. The 2004 Mexico City emissions inventory underestimates the urban primary PM by a factor of approximately 3.5 (and it is approximately 17x lower than afternoon concentrations when secondary species are included) and the wildfire contribution by a factor of approximately 20.