



Investigation of the Sources and Processing of Organic Aerosol over the Central Mexican Plateau from Aircraft Measurements during MILAGRO

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Organic aerosol (OA) represents approximately half of the submicron aerosol in Mexico City and the Central Mexican Plateau. This study uses the high time resolution measurements performed onboard the NCAR/NSF C-130 aircraft during the MILAGRO/MIRAGE-Mex field campaign in March 2006 to investigate the sources and chemical processing of the OA in this region. An examination of the OA/CO ratio evolution as a function of photochemical age shows distinct behavior in the presence or absence of substantial open biomass burning (BB) influence, with the latter being consistent with other studies in polluted areas. In addition, results from Positive Matrix Factorization (PMF) analysis of 12-second High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS) OA spectra will be presented. Four components were resolved. Three of the components contain substantial organic oxygen and are termed semivolatile oxygenated OA (SV-OOA), low-volatility OOA (LV-OOA), and biomass burning OA (BBOA). A reduced “hydrocarbon-like OA” (HOA) component is also resolved. LV-OOA is highly oxygenated (atomic O/C~1) and is aged organic aerosol linked to regional airmasses, with likely contributions from pollution, biomass burning, and other sources. SV-OOA is strongly correlated with ammonium nitrate, Ox, and the Mexico City Basin. We interpret SV-OOA as secondary OA which is nearly all (>90%) anthropogenic in origin. In the absence of biomass burning it represents the largest fraction of OA over the Mexico City basin, consistent with other studies in this region. BBOA is identified as arising from biomass burning sources due to a strong correlation with HCN, and the elevated contribution of the ion C₂H₄O₂⁺ (m/z 60, a marker for levoglucosan and other primary BB species). WRF-FLEXPART calculated fire impact factors (FIF) show good correlation with BBOA mass concentrations within the basin, but show location offsets in the far field due to model transport errors. This component is small or absent when forest fires are suppressed by precipitation. Since PMF factors represent organic species grouped by chemical similarity, additional postprocessing is needed to more directly apportion OA amounts to sources, which is done here based on correlations to different tracers. The postprocessed AMS results are similar to those from an independent source apportionment based on multiple linear regression with gas-phase tracers. During a flight with very high forest fire intensity near the basin OA arising from open BB represents ~66% of the OA mass in the basin and contributes similarly OA mass in the outflow. Aging and SOA formation of BB emissions is estimated to add OA mass equivalent to about ~32-42% of the primary BBOA over several hours to a day.