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Can 3D Models Predict The Observed Fractions Of Modern And Fossil Carbon In and Near Mexico City?

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A 3D chemistry-transport model has been applied to the Mexico City metropolitan area in attempt to investigate the origin of elevated levels of non-fossil (NF) carbon aerosols observed in this highly urbanized region. Rapid measurements of the fine aerosol concentration and composition, and 12 or 24 hours integrated 14C measurements of modern carbon have been performed in and near Mexico City during the March 2006 MILAGRO field experiment. The modern carbon fraction (fCM) contained in PM2.5 carbonaceous aerosol samples was reported to range from 42 to 76% at the downtown location (T0), and from 55 to 96% at the suburban site T1. Substantially lower values (i.e. 27-54%) were reported for PM10 filters within the city suggesting higher than expected uncertainties in the measurement techniques of 14C. A 10-20% increase in fCM was observed for both sets of filters during high wildfire activity in comparison to periods when fires were suppressed by rain. Modeling results for organic carbon aerosols have shown similar spatial and temporal variations/tendencies for fCM between urban and suburban area, fire and no-fire periods, although the exact values of fCM were often underpredicted. The model simulation that included the secondary organic aerosols (SOA) formation from semi-volatile and intermediate volatility vapors showed a better skill in modeling both the total OA mass and its modern carbon fraction in comparison to the traditional two-product SOA approach. For the first time, the combined analysis of aerosol mass spectrometer (AMS) and radiocarbon measurements were used to assess the model performance in predicting source-specific organic material including primary and secondary organic aerosols from fossil (FF) and non-fossil (NF) origin.