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Modeling the organic aerosol fraction within the Mexico City basin during the MILAGRO field experiment

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The meso-scale chemistry-transport model CHIMERE is used to asses our understanding of major sources and formation processes leading to a fairly abundant fraction of organic aerosols (OA, including primary OA (POA) and secondary OA (SOA)) observed in Mexico City during the MILAGRO field project (March 2006). Chemical analysis of submicron aerosols from aerosol mass spectrometers (AMS) indicate that carbonaceous particles found in the Mexico City basin have a large fraction of oxygenated organic species (OOA) which show strong correspondence with SOA, and that their production actively continues downwind of the city. The SOA formation is modeled according to the first-generation oxidation of anthropogenic (i.e. aromatics, alkanes) and biogenic (i.e. monoterpenes and isoprene) precursors and their partitioning into both organic and aqueous phases. The near-surface model evaluation shows that predicted OA correlates reasonably well with measurements during the campaign, however it remains a factor of 2-3 lower than the measured TOA. One of the reasons for this large gap is the inability of the model to simulate TOA peaks associated with the biomass burning events suggesting that near-city fires are not correctly represented in the emissions inventory (most likely too small to be detected by satellites). Fairly good agreement is found between observed and predicted POA within the city indicating that primary emissions are reasonable. Consistent with previous studies in Mexico City, large discrepancies are found for SOA species characterized by a factor of 5-10 model underestimate. When only anthropogenic SOA precursors were considered, the model was able to reproduce the sharp increase in SOA concentrations during the late morning at both urban and near-urban locations. However, predicted SOA concentrations were unrealistically low when photochemistry was not active, especially overnight. These discrepancies were not significantly reduced when greatly enhanced partitioning to the aerosol phase, or the condensation of the traffic-generated semi-volatile primary organics (SVOCs) were assumed. Model sensitivity results suggest that observed nighttime SOA concentrations are dominated by the regional background ($\tilde{2}$-3 μg/m3) from biogenic origin (mainly from isoprene) which are transported from the coastal regions into the Mexico City basin. The relative contribution of biogenic SOA increases at the regional scale (immediate vicinity of the city) where the influence of anthropogenic precursors is more limited. Our results confirm the large underestimation of SOA by traditional models in polluted regions, however it emphasizes for the first time the key role of biogenic precursors in this region and indicates that they cannot be neglected in modeling studies. According to our results, the contribution of biogenic SOA is comparable to the SOA production from anthropogenics, and more important than the influence of traffic SVOCs (<10%) in the vicinity of Mexico City.