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The influence of semi-volatile and reactive primary emissions on the abundance and properties of global organic aerosol

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Semi-volatile and reactive primary organic aerosols are modeled on a global scale using the GISS GCM II' "unified" climate model. We employ the volatility basis set framework to simulate emissions, chemical reactions and phase partitioning of primary and secondary organic aerosol (POA and SOA). The model also incorporates the emissions and reactions of intermediate volatility organic compounds (IVOCs) as a source of organic aerosol (OA), one that has been missing in most prior work. Model predictions are evaluated against a broad set of observational constraints including mass concentrations, degree of oxygenation, volatility and isotopic composition. A traditional model that treats POA as non-volatile and non-reactive is also compared to the same set of observations to highlight the progress made in this effort. The revised model predicts a global dominance of SOA and brings the POA/SOA split into better agreement with ambient measurements. This change is due to traditionally defined POA evaporating and the evaporated vapors oxidizing to form non-traditional SOA. IVOCs (traditionally not included in chemical transport models) oxidize to form condensable products that account for a third of total OA, suggesting that global models have been missing a large source of OA. Predictions of the revised model for the SOA fraction at 17 different locations compared much better to observations than predictions from the traditional model. Model-predicted volatility is compared with thermodenuder data collected at three different field campaigns: FAME-2008, MILAGRO-2006 and SOAR-2005. The revised model predicts the OA volatility much more closely than the traditional model. When compared against monthly averaged OA mass concentrations measured by the IMPROVE network, predictions of both the revised and traditional model lie within a factor of two in summer and mostly within a factor of five during winter. A sensitivity analysis indicates that the winter comparison can be improved either by increasing POA emissions or lowering the volatility of those emissions. Model predictions of the isotopic composition of OA are compared against those computed via a radiocarbon isotope analysis of field samples. The contemporary fraction, on average, is slightly under-predicted (20%) during the summer months but is a factor of two lower during the winter months. We hypothesize that the large wintertime under-prediction of surface OA mass concentrations and the contemporary fraction is due to an under-representation of biofuel (particularly, residential wood burning) emissions in the emission inventory. Overall, the model evaluation highlights the importance of treating POA as semi-volatile and reactive in order to predict accurately the sources, composition and properties of ambient OA.