SOA Measurements vs. Models: A Status Report

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The advent of fast and chemically-resolved organic aerosol (OA) and VOC measurements in the last decade has allowed more detailed model-measurement comparisons for OA and secondary OA (SOA). Large model underpredictions have been reported for SOA at many locations, but this is not always the case. Here we summarize the patterns emerging from studies to date, focusing on studies that use highly time and/or chemically resolved OA measurements. The model-measurement comparisons exhibit clear patterns depending on the region of the atmosphere.

• At least 8 studies have reported a large (x5-10) underestimation of SOA for polluted regions when using traditional models (those developed until ∼2006) (Heald GRL05, Volkamer GRL06, Johnson ACP06, Kleinman ACP08, Matsui JGR09, Dzepina ACP09, Hodzic ACP09, Tsimpidi ACP09). This is especially obvious when models are evaluated with the OA/CO ratio.

• Close to pollution sources, discrepancies of an order-of-magnitude in SOA lead to smaller discrepancies (often x2-3) for total OA due to the presence of primary OA (de Gouw EST09). Such OA discrepancies have been repeatedly observed (e.g. Vutukuru JGR06, McKeen JGR07&09, Heald JGR07, Fast ACP09, Hodzic ACP09).

• The discrepancy is reduced when recently-updated yields for aromatics (Ng ACP07) and SOA from glyoxal (Volkamer GRL07) are used, and is eliminated when using SOA formation from S/IVOC (Robinson Sci07) although with an overprediction of SOA at long aging times (Dzepina ACP09; Hodzic ACP10), especially with the Grieshop (ACP09) update of the Robison mechanism (Hodzic10). It is not clear whether the urban discrepancy is removed for the right reasons.

• 4 evaluations of biogenic SOA formed in unpolluted regions find reasonable agreement between SOA from traditional models and field measurements (Tunved Sci06; Hodzic ACP09; Chen GRL09; Slowik ACPD09). One evaluation reports a significant underprediction (Capes ACP09), although the amount of precursor reacted was difficult to ascertain for that case. The difference with the systematic underprediction observed for anthropogenic SOA may be due to the lack of primary S/IVOC in biogenic emissions, or to other reasons (NOx, SO2, POA, etc.).

• Comparisons for biogenic SOA formed in polluted regions are more complex. Several studies have reported a lack of clear influence of biogenic VOCs in SOA formation in polluted regions (de Gouw JGR05, GRL09; Weber JGR07; Bahreini JGR09), but 14C studies suggest a large fraction of modern C (Weber JGR07). Synergistic effects of pollution and BVOCs appear likely (e.g. de Gouw JGR05; Weber JGR07; Goldstein PNAS09).

• Net SOA formation (above the POA mass emission) from biomass burning appears very variable in the field (Capes JGR09; Yokelson ACP09; DeCarlo ACPD10) as well as in the laboratory (Ortega AGU09; Prevot AGU09; Hennigan in prep.), likely due flaming vs. smoldering fraction and biomass identity, and perhaps also to prompt SOA formation triggered by HONO photolysis. Several studies report significant SOA formation, given enough photochemical processing. Models based on traditional precursors appear to underpredict SOA from BB sources (Grieshop ACP09; Hodzic ACP09).

• The very large SOA source in the free troposphere postulated by Heald (GRL05) has not been confirmed nor disproved by later studies. Dunlea (ACP09) did not find evidence of this source across the Pacific near North America, though precipitation removal precludes any strong conclusions. However Carlton (EST09) reported better comparisons after implementing in-cloud SOA formation.

• Evaluation of models against measured total OA or OC or SOA levels is thoroughly unsufficient to model verification. OA measurements can be matched through a large number of mechanism permutations, but investigators need to push further to determine whether the agreement is for the right reasons. Future model evaluations should compare POA, SOA precursors, OVOCs, oxidants, and boundary conditions. Multiple OA measurements

(WSOC, OC, AMS, molecular tracers, 14C, etc.) are necessary to overcome the limitations of any one method. Measurements of semivolatile species are critically needed to constrain models. These advanced diagnostics are needed in order to build confidence on SOA models, which is needed to predict the changes in SOA concentrations in response to precursor and climate changes.