Global Modeling of Secondary Organic Aerosol Production from Reaction of NO_3 Radical with Speciated Monoterpenes

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Motivation

- On a global and regional scale, secondary organic aerosol (SOA) is known potentially to contribute to global cooling[1] and cause deleterious effects on human health[2].
- Historically, global modeling of SOA formation from monoterpenes has been based on a simplified lumped mechanism, which parameterizes all monoterpene-NO_3 reactions as β-pinene[3][4].
- The resulting global spatial patterns and annual budgets of organic aerosol gave poor matches with observations[5].
- Recent chamber studies reveal α-pinene reaction with NO_3 radical oxidant to have a much lower SOA yield than compared to the other bicyclic monoterpenes[6][7].
- To assess how a lower α-pinene-NO_3 SOA-producing pathway affects global organic aerosol concentrations, the global 3-D chemical transport model GEOS-Chem was updated with a new volatility basis set (VBS) based aerosol parameterization where α-pinene was removed from the lumped parameterized terpenes tracer and speciated with unique chemistry.

Model Configuration and Evaluation

GEOS-Chem Global Simulations

- GEOS-Chem v10-01i
- GEOS-5 Meteorology
- 4x5 resolution
- 2005 spinup/2006 output
- MEGAN v2.1 Biogenic Emissions

Monoterpene Speciation and VBS Parameterizations

- The monoterpene α-pinene (APIN) was removed from the bicyclic terpene tracer species (MTPA) and speciated with a VBS APIN-NO_3 yield of zero[6][7].

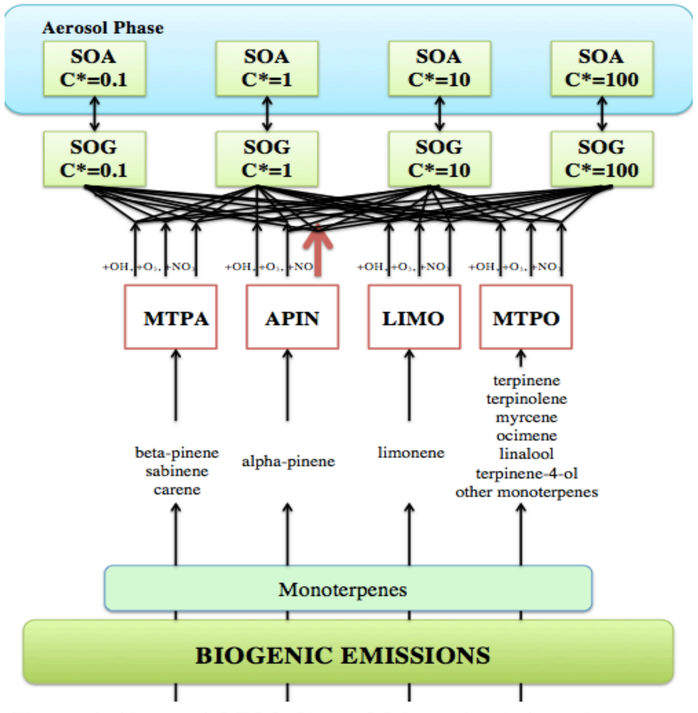

Figure 1: *Updated GEOS-Chem SOA mechanistic pathway*

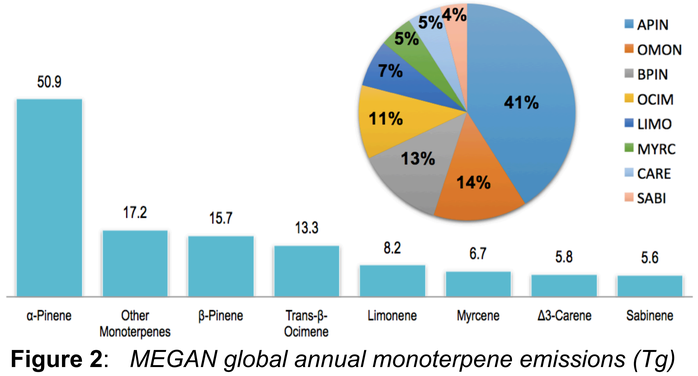

Figure 2: *MEGAN global annual monoterpene emissions (Tg) and relative percent contribution*

Model Results: Base Case-Test Case

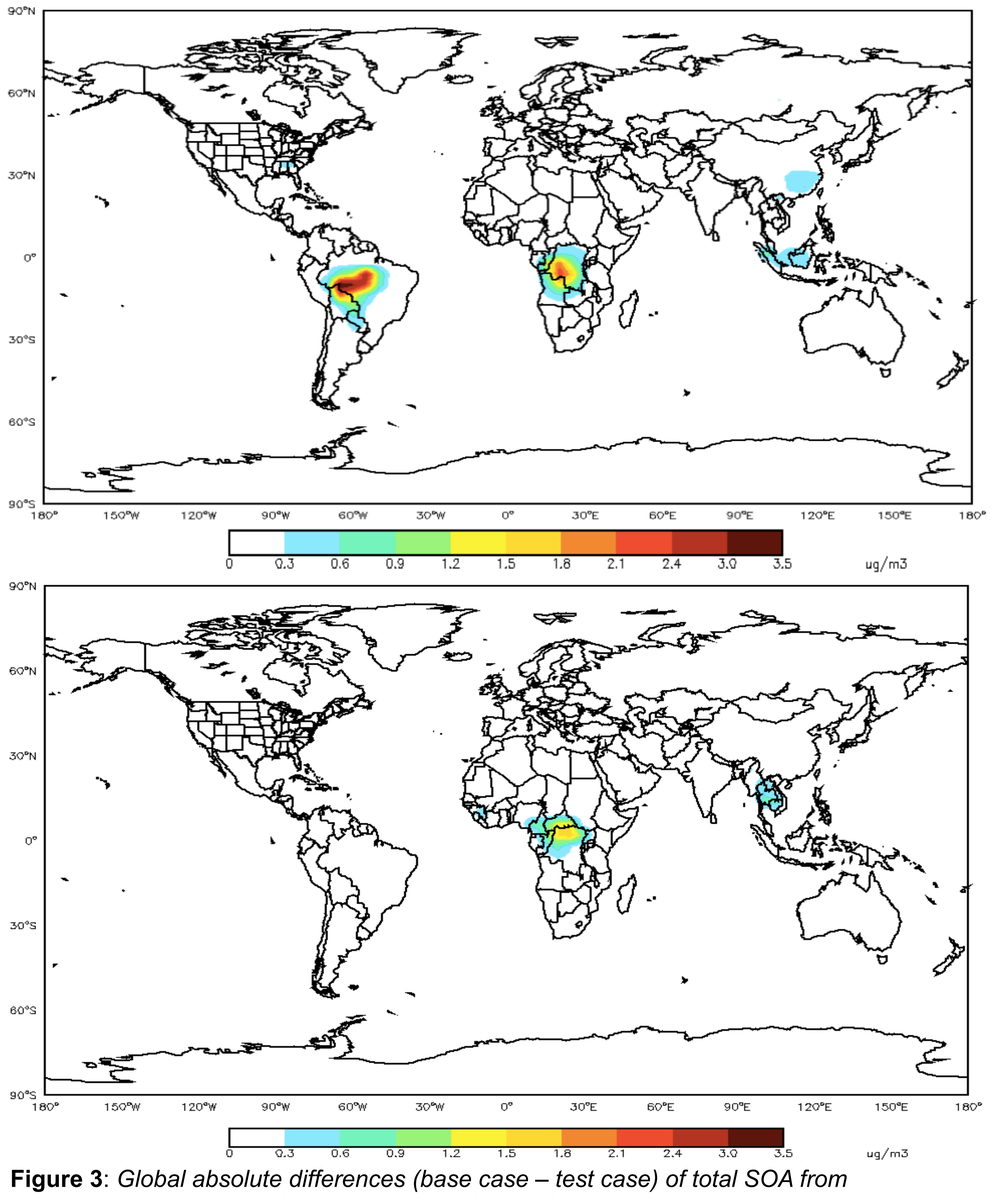

Figure 3: *Global absolute differences (base case – test case) of total SOA from monoterpenes in August (top) and January (bottom)*
Changes in SOA are relative to the base case model minus the α-pinene-NO_3 zeroed yield. Notice the largest differences occur in high VOC/SOA producing source regions such as the Congo forests and the Amazon. August is the greatest SOA-producing month due to increased VOC emissions and temperature.

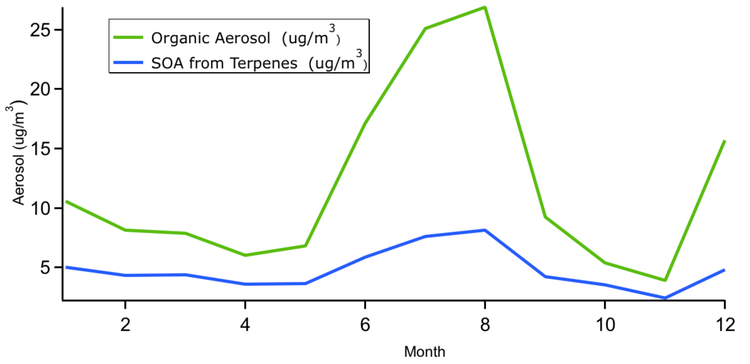

Figure 4: *Simulated monthly average total OA (aerosol products of terpene, isoprene, light aromatics, and POG oxidation) and SOA from terpenes in the Congo Forest*

Conclusions

- The major finding of this work is that for high SOA producing source regions such as the Amazon and the Congo, there is a 3.5 μg/m^3 decrease in predicted SOA in the summer months.
- The reduction in SOA concentrations due to updated chemistry leads to an annual difference of 2 Tg between the control and novel VBS mechanisms, a 10% change in terpene organic aerosol.

Future Work

- Analyze aircraft and satellite observations for model-to-measurement comparisons.
- Implement greater degree of speciation for the bicyclic monoterpenes and the sesquiterpenes.

References

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