Improving secondary organic aerosol (SOA) in climate models:

Combining measurements, high resolution regional models, and global models

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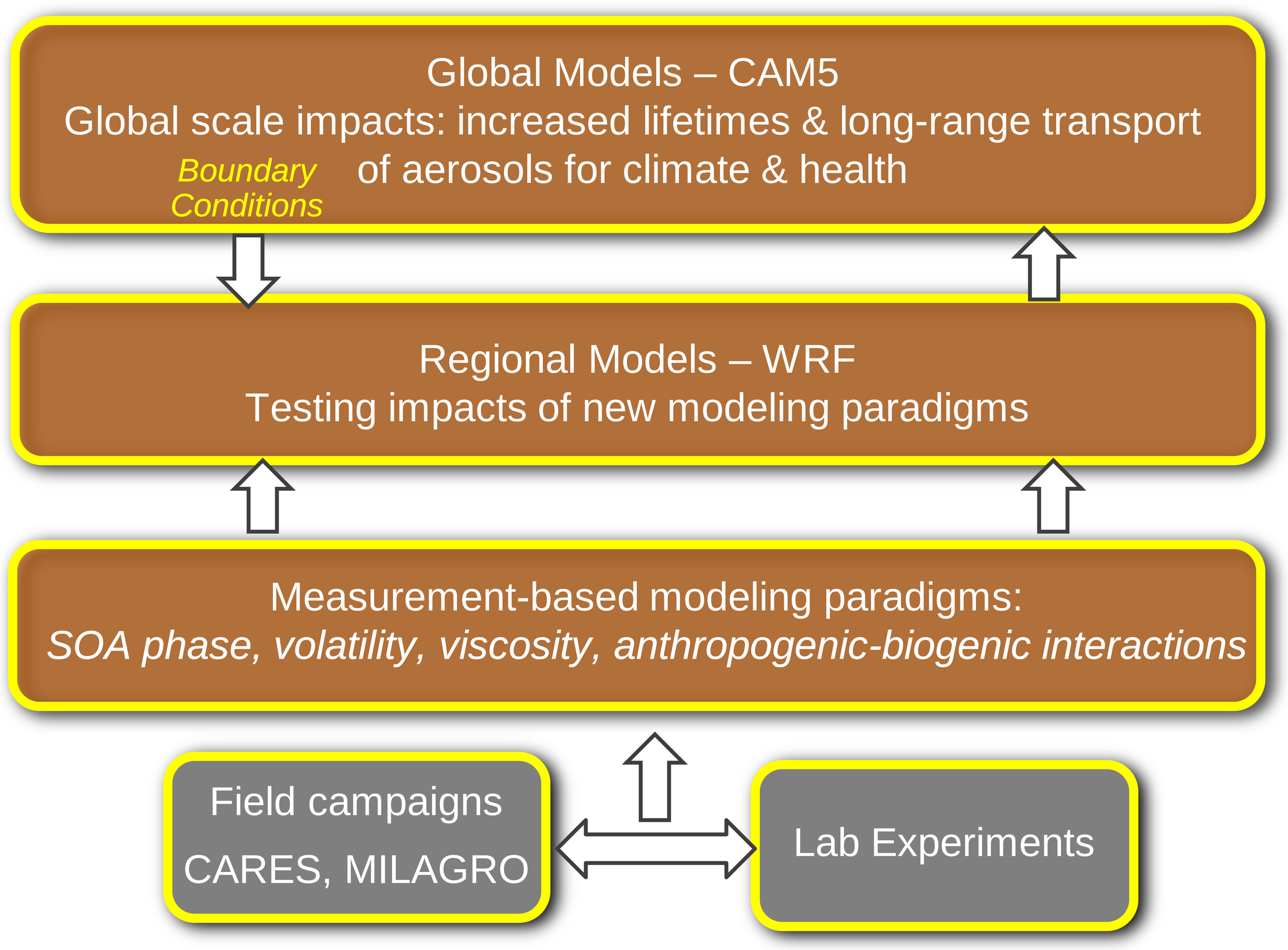
Introduction

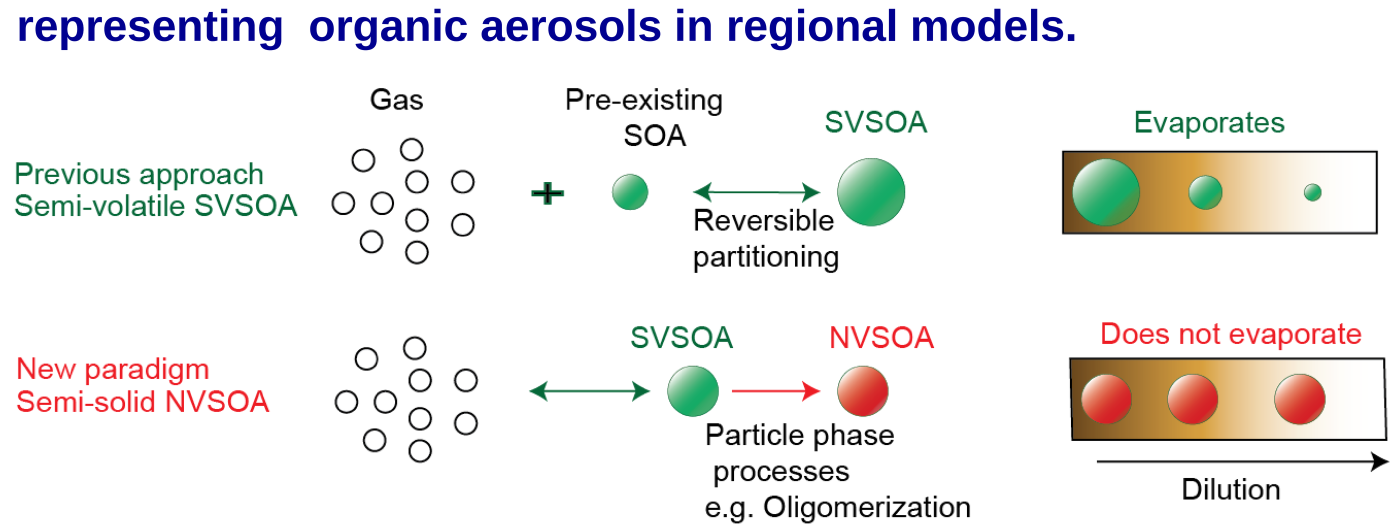
Recent measurements have shown that the processes governing the lifecycle of SOA particles and their properties (such as evaporation behavior, phase, and viscosity) are not represented accurately in regional and global climate models. As a part of the ASR program, we developed SOA modeling paradigms based on recent laboratory measurements that investigated the implications of extremely low effective volatility of SOA particles, and incorporating gas-phase fragmentation reactions using the regional model WRF-Chem. Results indicated large changes in loadings, and the spatial and temporal distribution of SOA.

Here, we take the next step connecting our ASR work to DOE's global Community Earth System Model (CESM).



Measurements suggest a paradigm shift in representation of SOA particles from semi-volatile liquid-like to low volatile semi-solids. New modeling paradigms were first investigated in high resolution regional model WRF-Chem, and then included within the global model CESM.

Importantly, the improved SOA representation in CESM will in turn be used to generate improved boundary conditions for representing organic aerosols in regional models.



Previous approach: *Semi-volatile liquid-like SOA rapidly evaporates*

New paradigm: *SOA is rapidly transformed to a very low volatility (effectively non-volatile) semi-solid that does not evaporate during transport.*

This shift in modeling paradigm could be explained based on particle-phase processes as oligomerization, and has large implications for the global loadings and lifetimes of SOA.

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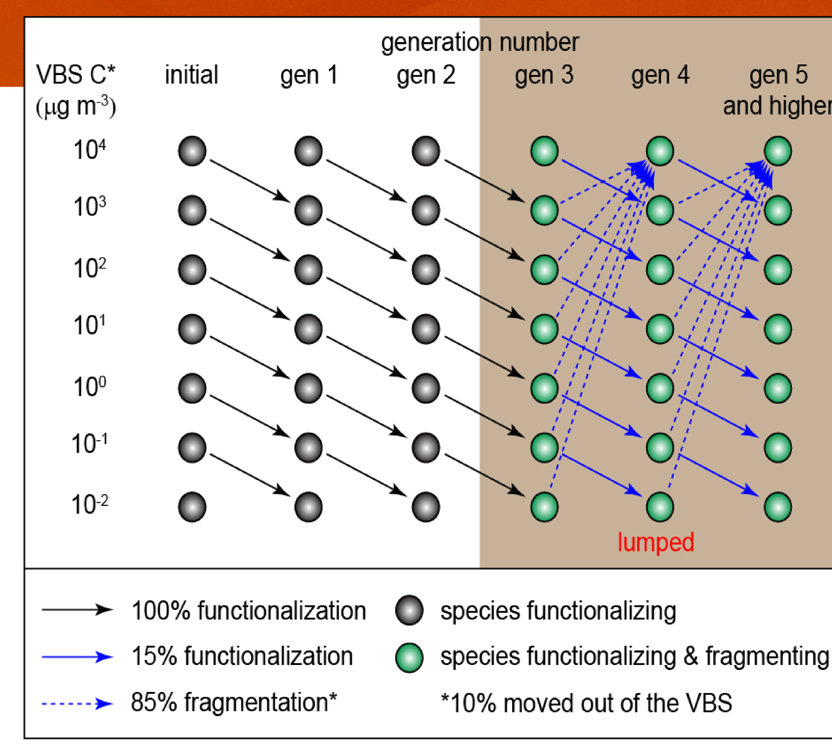
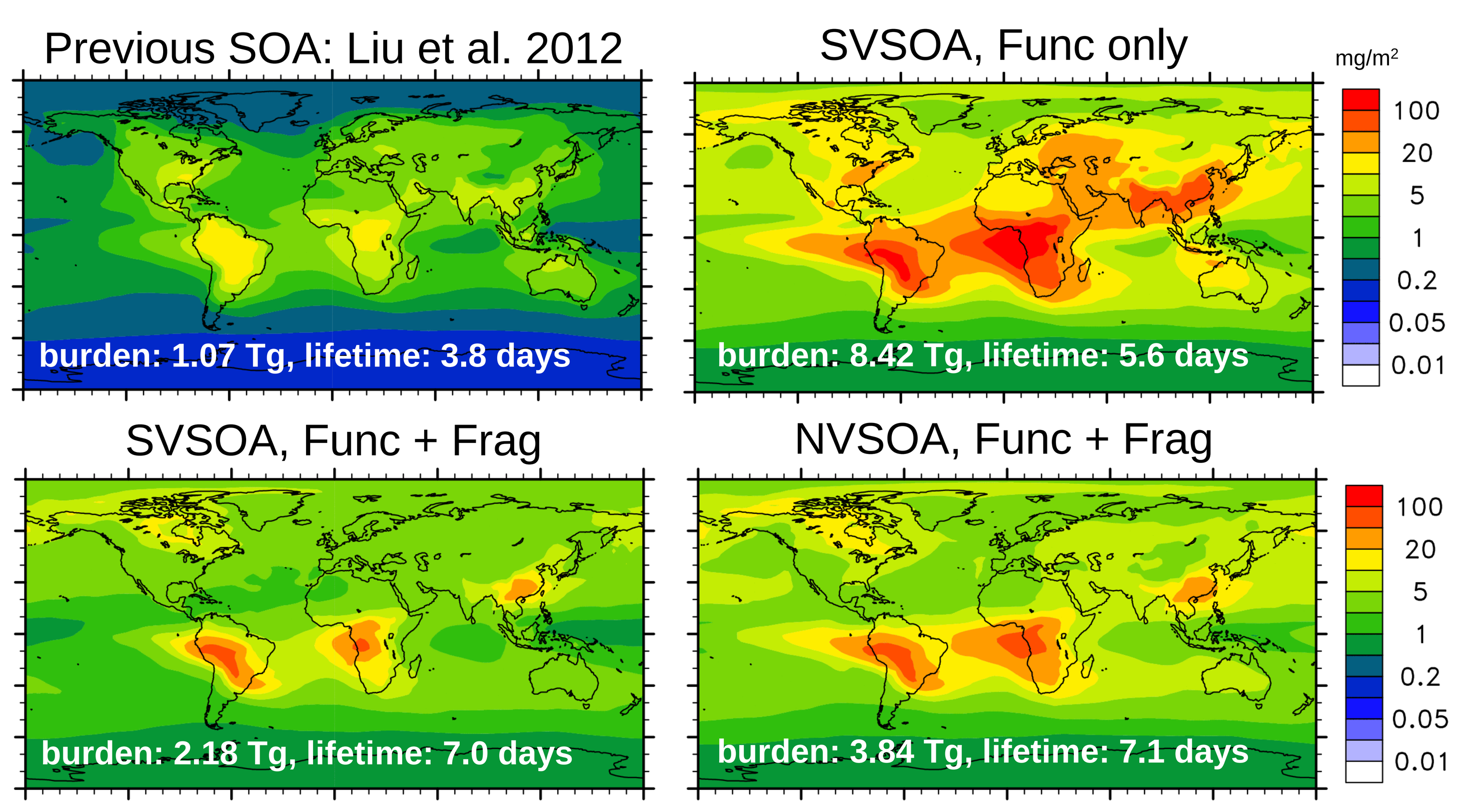


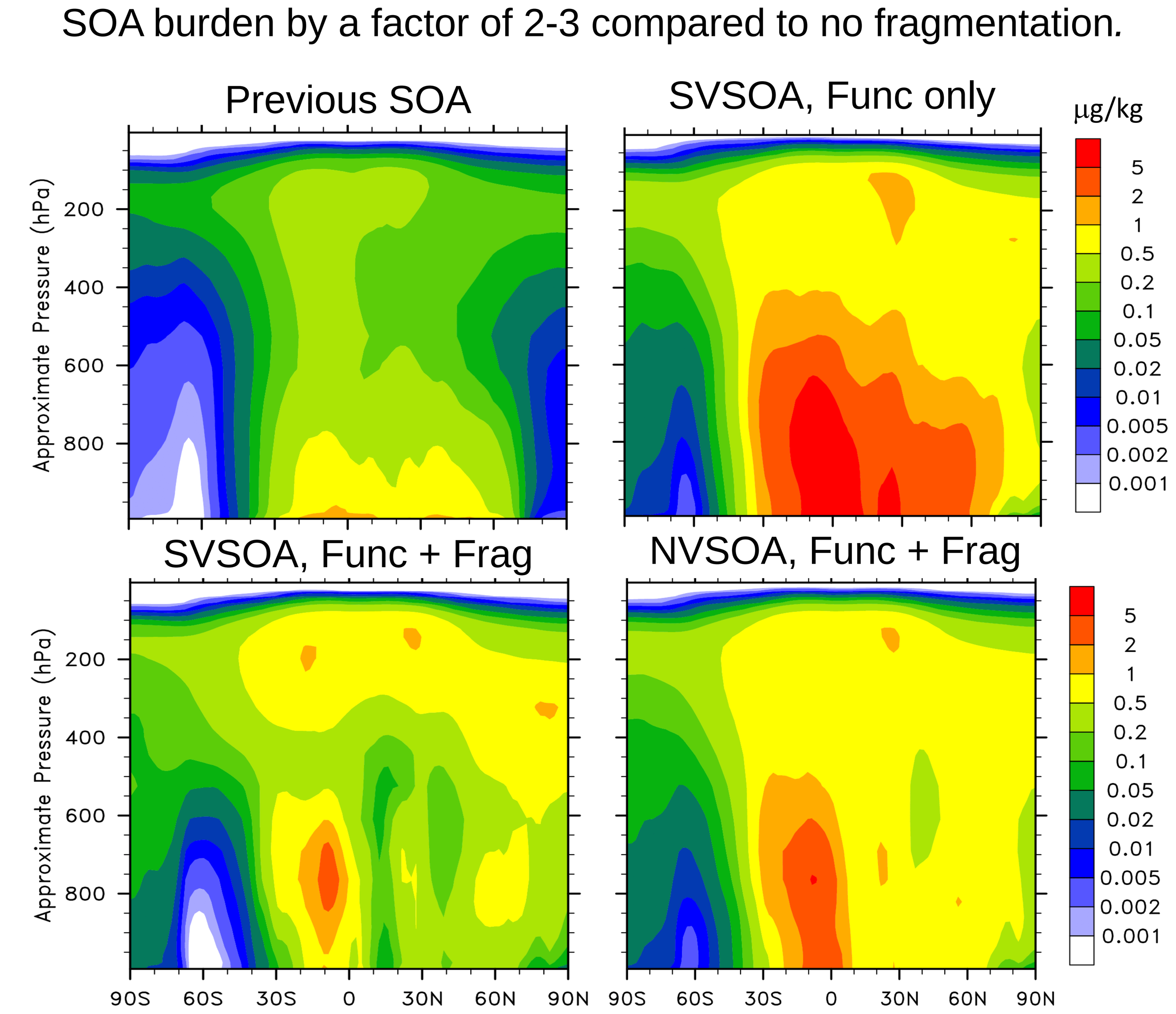
Although detailed data required to constrain the fragmentation of organic species during SOA formation is still lacking, we developed a simplified approach based on recent studies to represent this process in 3D chemical transport models (Shrivastava et al. 2013).

Results

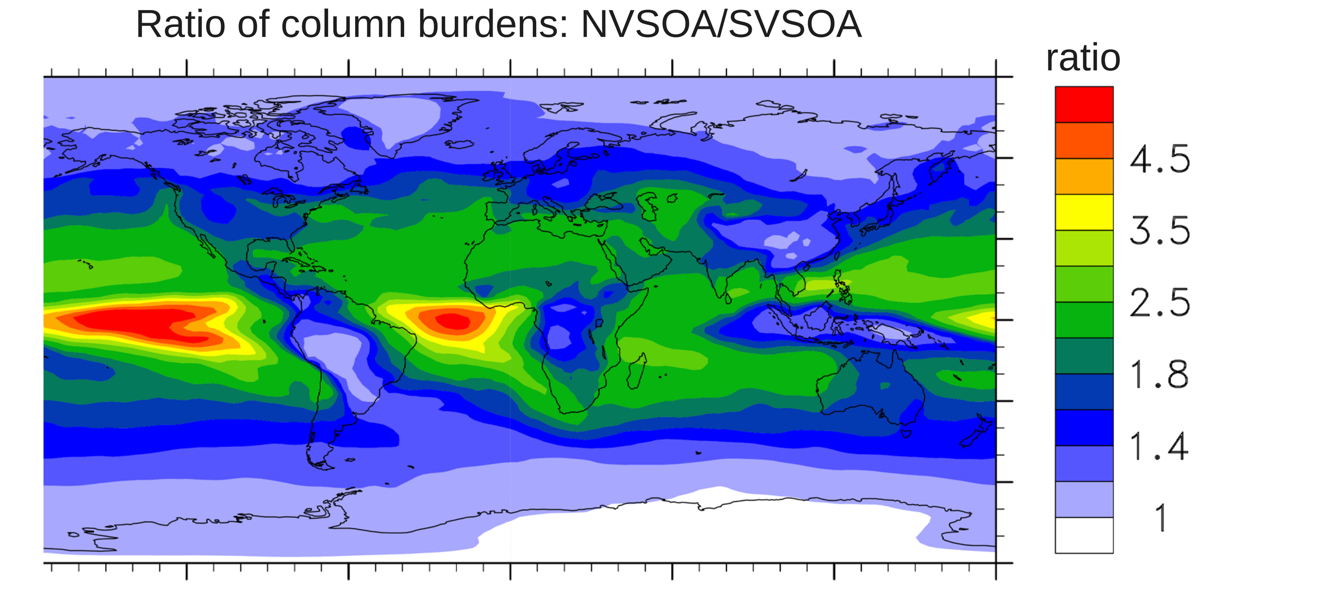


Functionalization only (Func only) refers to the simulation neglecting fragmentation, while Func+Frag refer to simulations that include fragmentation after first 2 generations of functionalization reactions.

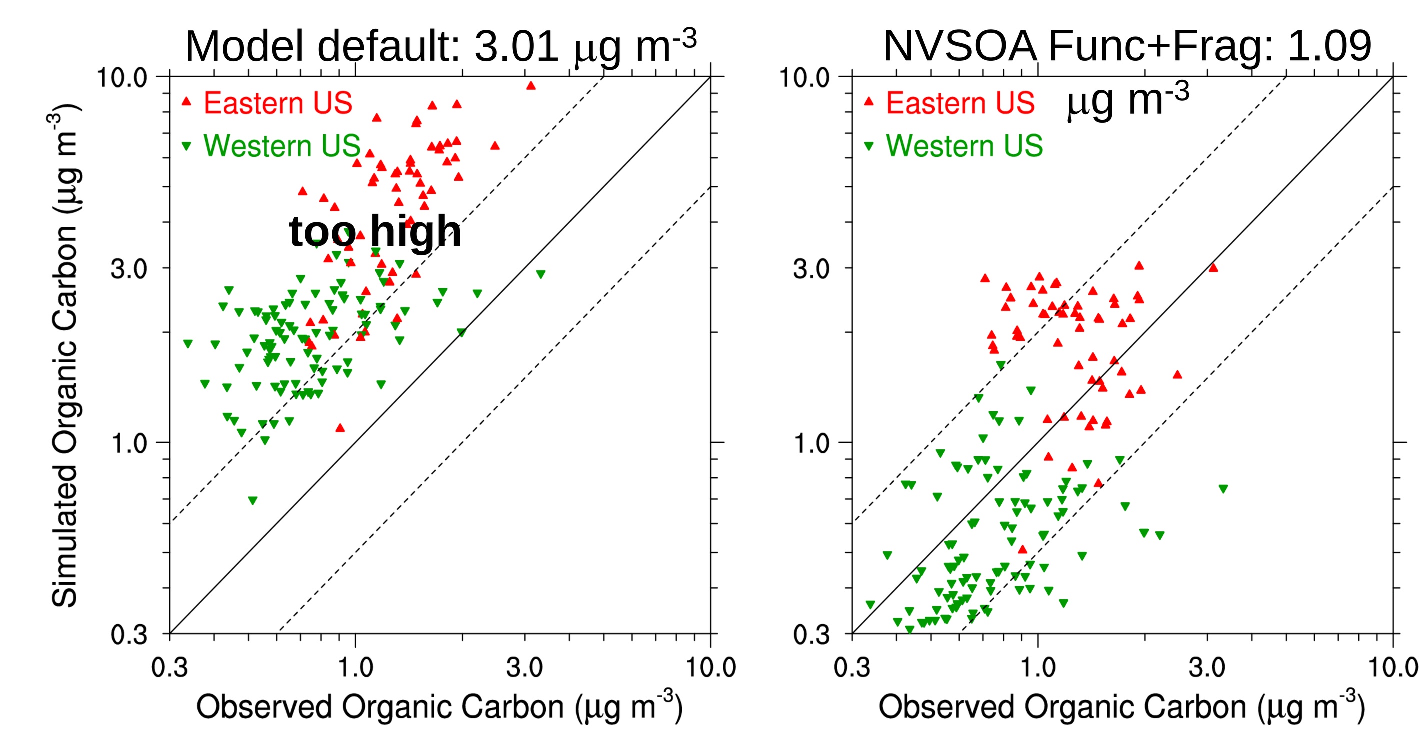
- New treatments predict a factor of 2-8 higher global SOA burdens compared to the previous model.
- Including gas-phase fragmentation in the new treatments reduces SOA burden by a factor of 2-3 compared to no fragmentation.



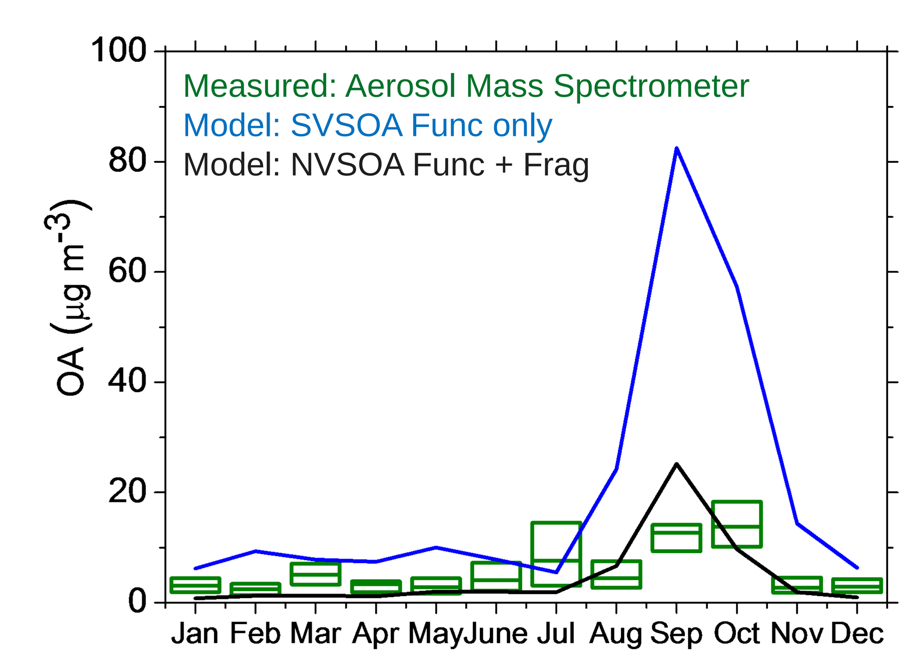
New model configurations simulate much higher SOA in the free troposphere compared to the previous model, consistent with previous studies reporting large model-measurement differences in the free troposphere (e.g. Volkamer et al. 2006).



Treating SOA as a non-volatile semi-solid increases global average SOA burdens by a factor of 2, and the largest increase corresponds to pollution outflow over oceans.



The revised treatment with non-volatile SOA and fragmentation shows much better agreement with U.S. IMPROVE network OC measurements



Comparison to AMS measurements at the biomass burning site Welgegund, South Africa. Model captures the seasonal variation of OA. Neglecting fragmentation (Func only) leads to large overprediction of OA . Including fragmentation improves agreement.

Conclusions

Our revised treatment includes 62 SOA tracers and accounts for semi-solid SOA and multi-generational gas-phase chemistry with fragmentation, and to the best of our knowledge, is the most advanced SOA treatment in a global model

- Multi-generational aging increases SOA concentrations throughout the domain
- Fragmentation reduces SOA burdens by a factor of 2-3
- Treating SOA as a non-volatile semi-solid increases its global burdens and lifetimes compared to the traditional semi-volatile treatment
- Neglecting fragmentation significantly overpredicts SOA at the biomass burning site in South Africa and over IMPROVE sites (not shown)
- Treatment with fragmentation and non-volatile SOA shows much better agreement with IMPROVE network measurements

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