Modeling Regional-Scale Variability of Organic Aerosols in the Atmosphere

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Objectives

While organic matter (OM) comprises a large fraction of the total aerosol burden in many places of the world, large uncertainties remain in the prediction of secondary organic aerosol (SOA) formation and transformation that likely affects aerosol radiative forcing. Our current objectives are to:

- *Quantify the performance of simulated primary, secondary, and total OM based on the **volatility basis set** (VBS) approach. Since the VBS approach is currently being implemented in CAM5 (global model), it is important to assess its performance at scales compatible with ARM measurements.*
- *Adapt the VBS approach to investigate the important issues of particle-phase changes in **volatility** and gas-phase **fragmentation versus functionalization** reactions affecting the formation and evolution of SOA.*

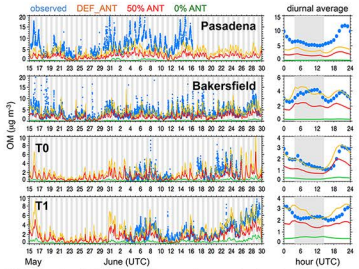
Evaluation of the VBS Approach for CARES

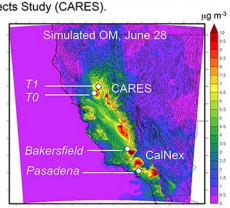
The VBS approach implemented in the WRF-Chem model (Shrivastava etla. 2011) is used to simulate OM in California during the 2010 Carbonaceous Aerosol and Ratiative Effects Study (CARES).

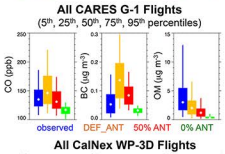
Three simulations performed for May – June 2010 with Δx = 4 km:

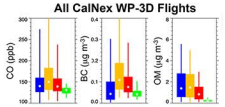
1) *Default 2008 CARB emission inventory, DEF_ANT*
2) *Anthropogenic emissions reduced by 50%, 50%_ANT*
3) *No anthropogenic emissions, 0%_ANT*

Many species (CO, NOx, BC) greatly improved for 2).

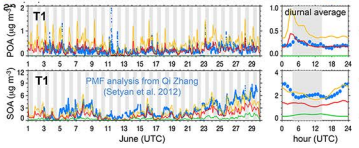








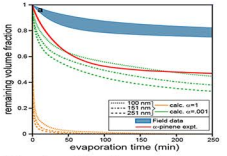
While simulated OM from 1) agrees better with data, it is probably fortuitous. Primary organic matter from simulation 2) is in better agreement with POA derived from PMF



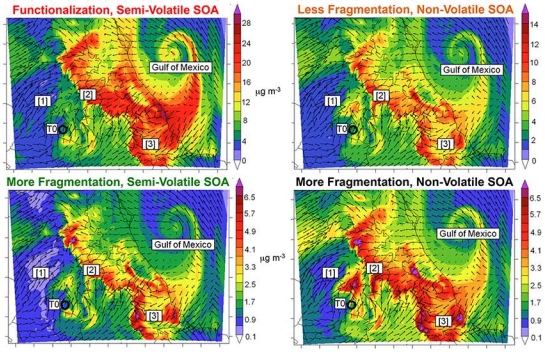
- While the current VBS approach reproduces the observed diurnal and multi-day trends in OM, it overestimates SOA downwind of Mexico city (Shrivastava et al., 2011) and underestimates SOA by a factor of 2-3 for the CARES case.
- The 2-species VBS already assumes that anthropogenic SOA is non-volatile, while contribution of biogenic SOA is much lower based on literature yields.
- **Next Steps:** Investigate increase of biogenic SOA through multi-generational chemistry, anthropogenically-enhanced biogenic SOA, and volatility of biogenic SOA.

Implications of Low Volatility and Fragmentation

- Shrivastava et al. (2013) show that under realistic assumptions of mass accommodation coefficient, measured evaporation rates of SOA imply significantly lower "effective volatility" than those derived from SOA growth from chamber data (Vaden et al., 2011), pointing to the role of particle phase changes after SOA formation. Thus, models may need to use different parameters to describe the formation and atmospheric evolution of SOA.

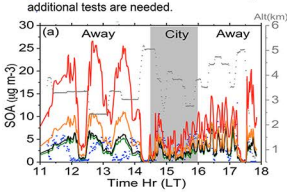


- We examine implications of low "effective volatility" of SOA and gas-phase fragmentation reactions; simplified parameterizations are developed to capture the first-order effects of fragmentation.
- Simulating the case study of MILAGRO 2006, we found complex variations in spatial and temporal distribution of SOA with varying degrees of gas-phase fragmentation reactions. The treatment of SOA as semi-volatile or non-volatile also caused variations in predicted SOA loadings in the atmosphere.



Shrivastava, M. A. Zelenyuk, D. Imre, R. Easter, J. Beranek, R.A. Zaveri, and J. Fast, 2013: Implications of Low Volatility SOA and Gas-Phase Fragmentation Reactions on SOA Loadings and their Spatial and Temporal Evolution in the Atmosphere. In Press, *J. Geophys. Res.*

- Including fragmentation also led to an improved prediction of SOA downwind of Mexico City, but additional tests are needed.



Acknowledgements: This research was supported by the U.S. DOE's Atmospheric System Research (ASR) program under contract DE-AC06-76RCO 1830 at PNNL with measurements supported by ARM Climate Research Facility. We thank the numeous CARES and CalNex investigators for providing measurements used in this study.

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