Advancing treatment of Secondary organic aerosols in CESM using the VBS-MAM approach

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Models often underpredict SOA loadings by a factor of ~10

- Discrepancy between model and data increases with photochemical age
- What is responsible for this discrepancy between models and measurements?
Model assumptions are inconsistent with laboratory and field data

### Traditional Modeling Approach Semivolatile SOA (SVSOA)
- Model treats SOA as rapidly mixing and evaporating solution
- Model assumes SOA particles are at equilibrium with the gas phase

### Recent Data Show
- SOA evaporates orders of magnitudes slower than assumed
- SOA is highly viscous semi-solid
- SOA is not at equilibrium with the gas phase

### New Modeling Paradigm-Nonvolatile SOA (NVSOA)
- Treat SOA as a non-volatile semi-solid
- Include fragmentation of gas-phase SOA precursors
SOA paradigms: nonvolatile vs semivolatile

- NVSOA, Highly viscous, non-absorbing, semi-solid

- SOA particles are almost non-volatile, highly viscous semi-solid and hence non-absorbing
While we know that fragmentation reactions play an important role in SOA formation, we lack the detailed data required to reliably represent the process.

- Fragmentation parameters correspond to the best model-measurement agreement during MILAGRO 2006 (Shrivastava et al. 2013)
- Because data show that precursor gas oxidation by OH yields higher and lower vapor pressure products, our model includes fragmentation reactions
Methodology

- CAM5 (1.9× 2.5°), nudged to ERA-Interim reanalysis
- Mozart chemistry coupled to modal aerosol module (MAM3)
- Daily fire emissions using GFED3.1
- 4-bin volatility basis-set (VBS) for biogenic SOA: Isoprene & Terpenes
- Semi-volatile and intermediate volatility organics (SIVOC) from anthropogenic and biomass burning emissions: 5-bin VBS×2
- 62 SOA species (42 SOA particle species and 20 gas phase organic species) + 104 gas-phase tracers from Mozart chemistry
- Flexible SOA modeling framework to easily change SOA species
- SOA species interact with clouds and the optical calculations
- Source-resolved SOA predictions from 3 precursor classes: anthropogenic, biomass burning and biogenic
- Treatment of gas-phase fragmentation and semi-solid SOA
Model configurations

1. Default SOA treatment that directly emits SOA (Liu et al. 2012, GMD)
2. Semi-volatile SOA, functionalization only (Func,no fragmentation)
3. Semi-volatile SOA, Func + Frag
4. Non-volatile semi-solid SOA, Func + Frag
Results: Annual average column burden

- New model configurations produce higher SOA burden and lifetimes than Liu et al. 2012
- Gas-phase fragmentation reduces average global SOA burden by a factor of ~2-3
- Treatments with fragmentation yield 25% higher SOA lifetime than without fragmentation

Previous SOA: 1.07 Tg, 3.8 days
Liu et al. GMD

SVSOA, Func only: 8.42 Tg, 5.6 days

SVSOA, Func + Frag: 2.18 Tg, 7.0 days

NVSOA, Func + Frag: 3.84 Tg, 7.14 days
Treating SOA as a non-volatile semi-solid increases global average SOA burdens by a factor of ~ 2

Largest increases in SOA burdens (> factor of 5) due to its non-volatile treatment correspond to pollution outflow over the oceans

Large potential implications on radiative forcing of climate
Results: Zonal average SOA

- New model configurations produce much higher SOA over both surface and the free troposphere compared to the previous model (explains higher lifetime)
Results: Source contributions to SOA

**Fragmentation and Nonvolatile SOA**

- **Anthropogenic SOA:** 0.39 mg m\(^{-2}\)
- **Biomass SOA:** 4.62 mg m\(^{-2}\)
- **Biogenic SOA:** 1.09 mg m\(^{-2}\)

- Biomass burning is the largest source of SOA globally (peaks over biomass burning regions)
- SOA from either biomass burning or biogenic sources is much larger than anthropogenic SOA
Comparison to measurements: biomass burning site Welgegund, South Africa

- Seasonal variation of measured OA captured by the model
- Neglecting fragmentation leads to large over-prediction of OA
- Model that includes fragmentation is closer to measurements

Obs. mean OC: 1.01 µg m⁻³

Model default: 3.01 µg m⁻³

NVSOA Func+Frag: 1.09 µg m⁻³

- Previous default treatment (Liu et al. 2012) overestimates OC
- Revised treatment with non-volatile NVSOA and fragmentation shows much better agreement
Conclusions

- Multi-generational aging of organic vapors increases SOA concentrations throughout the domain
- Fragmentation reduces SOA concentrations by a factor of 2-3
- Treating SOA as non-volatile semi-solid (glassy) increases SOA concentrations compared to its traditional semi-volatile treatment
- Biomass burning is the largest global source of SOA followed by natural biogenic emissions
- Including fragmentation significantly improves model-measurement agreement of OA at Welgegund, South Africa
- Treatment with fragmentation and non-volatile SOA shows much better agreement with IMPROVE network measurements

Future work:

- Improving treatment of particle nucleation and growth due to organics
- Evaluating the role of new SOA treatments on radiative forcing of climate
- Developing simpler but more accurate SOA treatments for use in global climate model simulations

Obs. mean OC: 1.01 µg m⁻³

SVSOA Func + Frag: 0.65 µg m⁻³
SVSOA Func only: 4.23 µg m⁻³

- Semivolatile SVSOA with fragmentation (Func+Frag) underestimates OC
- Semivolatile SVSOA neglecting fragmentation (Func only) significantly overestimates OC