# MATRIX-VBS: Condensing Organic Aerosols In An Aerosol Microphysics Model

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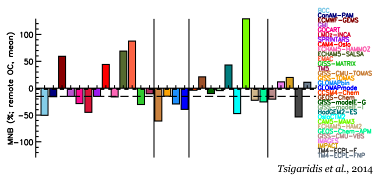
## Background & Motivation

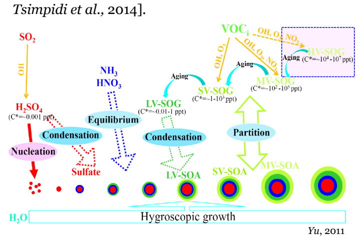
Aerosols play an important role in public health and climate [*Seinfeld and Pandis*, 2006]. Organic aerosol (OA) are ubiquitous and a major component of atmospheric aerosols [*Zhang et al.* 2007].

- **Problem:** Measurements imply that OA concentration are underestimated in models [*Heald et al.*, 2005, *Spracklen et al.*, 2011, *Tsigaridis et al.*, 2014].
- **Caused by:** missing amount organic aerosols.
- **Solution:** include semi-volatile primary organic aerosols (POA) and intermediate volatility organic compounds (IVOCs) as SOA precursors [*Robinson et al.*, 2007; *Hodzic et al.*, 2010].
- **Approach:** volatility-basis set (VBS) [*Donahue et al.* 2006]
- **Past studies:** Regional [*Hodzic et al.*, 2010] and global [*Farina et al.*, 2010; *Jathar et al.*, 2011; *Pye and Seinfeld*, 2010; *Shrivastava et al.*, 2008, *Tsimpidi et al.*, 2014].



*Tsigaridis et al.*, 2014

**Objective: to introduce the process of condensing organics in an aerosol microphysics model.**



*Yu*, 2011

**Important for aerosol size distribution:**

- Very low volatility organics play a key role in particle growth: condense on all sizes
- The range of volatilities contributing to aerosol size distribution increases with aerosol size [*Pierce et al.*, 2011; *Yu*, 2011]
- Affects aerosol size and mixing state & its impact on climate.

| Condensation | High Volatility | Low Volatility |
|---|---|---|
| Small particles | no | yes |
| Large particles | yes | yes |

## Model Description

**MATRIX**

- An aerosol microphysics model that can be used as a stand alone box model or within the GISS ModelE2 Earth System Model.
- Describes the mixing state of different aerosol populations [*Bauer et al.* 2008].
- POA : non-volatile
- No condensation of organics.

+

**VBS**

- Describes organic aerosols by separating low volatility organics into bins of effective saturation concentration, including gas-particle partition [*Donahue et al.* 2006].

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✓ **Our new version:**

- POA – semi-volatile, SVOCs and IVOCs represented with VBS
- Partition among different aerosol populations based on size and volatility, capturing particle growth via organic condensation

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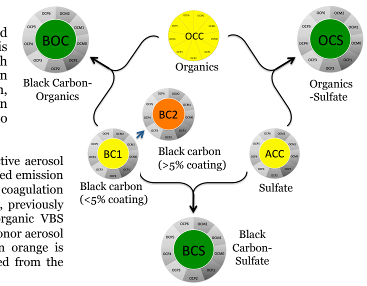
## Method & Results

### Stage 1: Model Development

In the original MATRIX model, organics grow or mix via coagulation, however, they can only be emitted as nonvolatile POA and cannot contribute to other aerosol populations via condensation. Implementing the VBS scheme fills this missing piece. This study introduces a ninth configuration in MATRIX with 15 selected aerosol populations, where eight populations (seven of which are in the illustration on the right and the eighth population is MXX, which accepts those who do not fit else where) contain organics as semi-volatile VBS species. Previously, each aerosol population carried a max of 5 tracers – sulfate, black carbon, nonvolatile organics, dust and sea salt, now they carry 9 additional VBS species, for a max of 14 available tracers per population for those containing organics. At this stage, we are only examining the BC-OA-Sulfate-Nitrate system(hence the decision to choose 8 of the populations to include organics), later in stage 3, organics will also be considered for dust and sea salt aerosols.

| Parameter | 9 Virtual VBS Species | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| C* (μg m⁻³ at 298K) | $10^{-2}$ | $10^{-1}$ | $10^{0}$ | $10^{1}$ | $10^{2}$ | $10^{3}$ | $10^{4}$ | $10^{5}$ | $10^{6}$ |
| Aerosol phase name | OCM2 | OCM1 | OCM0 | OCP1 | OCP2 | OCP3 | OCP4 | OCP5 | OCP6 |
| Gas phase name | gasm2 | gasm1 | gasm0 | gasp1 | gasp2 | gasp3 | gasp4 | gasp5 | gasp6 |
| Emission factors (Shrivastava et al., 2008) | 0.03 | 0.06 | 0.09 | 0.14 | 0.18 | 0.30 | 0.40 | 0.50 | 0.80 |

Left: Saturation concentration of the 9 VBS species, their respective aerosol and gas phase names used in this study, and the applied mass-based emission factors from *Shrivastava et al.*, 2008. Right: Schematic showing coagulation among organic-containing aerosol populations as colored circles, previously with no organics condensed (inner circles) and now with 9 organic VBS species condensed (grey outer circles). In yellow are the emitted donor aerosol populations, and green are the mixed recipient populations. In orange is population BC2, which contains >5% coating and quickly formed from the growth of BC1 population, which has <5% coating.



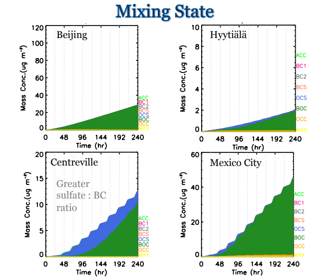

### Stage 2: Case Studies


| March 2006 | | Units | Beijing | Centreville | Hyytiälä | Mexico City |
|---|---|---|---|---|---|---|
| Parameters | Temperature | K | 278 | 289 | 268 | 289 |
| | Pressure | hPa | 1005 | 996 | 1009 | 797 |
| | RH | % | 46.6 | 77.7 | 79.5 | 62.5 |
| Gas emissions | $NO_x$ | pptv/hr | 222.2 | 90.3 | 172.0 | 143.1 |
| | CO | | 7188.1 | 1383.7 | 570.0 | 2481.6 |
| | Alkenes | | 4.4 | 0.3 | 0.1 | 1.4 |
| | Paraffin | | 8.5 | 2.3 | 0.6 | 10.8 |
| | Terpenes | | 4.1 | 41.6 | 15.2 | 34.5 |
| | Isoprene | | 24.0 | 106.1 | 0.2 | 34.5 |
| | $SO_2$ | | 577.5 | 197.7 | 24.7 | 551.0 |
| | $NH_3$ | | 187.4 | 24.5 | 52.0 | 63.0 |
| Aerosol emissions | sulfate | ug/m³/hr | 0.06 | 0.02 | 0.003 | 0.05 |
| | black carbon | | 0.09 | 0.01 | 0.008 | 0.03 |
| | organics* | | 0.19 | 0.03 | 0.015 | 0.11 |

*Organics 2.5x more in the new scheme.

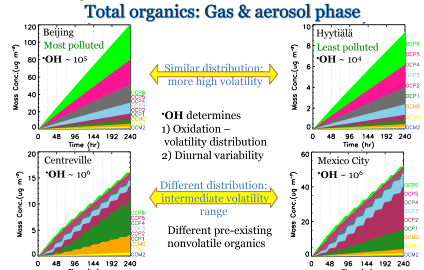
Four locations are used for case studies to examine the behavior of the newly developed model. One very polluted city Beijing, another cleaner yet still very polluted city with high altitude Mexico City, a very clean Finnish forest Hyytiälä, and a less clean forest in Centreville in the Southeast U.S. (see map) The experiments are set in March 2006 for ten days, and initial conditions and emission levels (listed in table above) from each location are taken from the GISS ModelE2 GCM output. Here, we examine and compare the mass concentrations and volatility distributions of total organics (the sum of gas and aerosol phase), gas phase organics and aerosol phase organics individually, as well as their amounts in different aerosol populations, thus taking a glimpse of their mixing states in different locations.

#### Mixing State



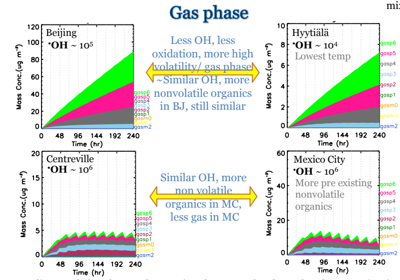

Most organics condensed or coagulated into BOC (black carbon-organics mixture) population, except those in Centreville, because there is higher sulfate to black carbon ratio and therefore some organics ended up in OCS (organics-sulfate mixture) population.

#### Total organics: Gas & aerosol phase



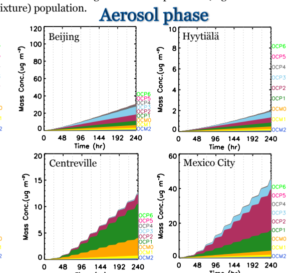

Mass concentrations of total organics are distributed by their volatilities as VBS species, from top green OCP6 to bottom blue OCM2, volatility decreases as they become more oxidized. Beijing has the highest mass concentration while Hyytiälä has the lowest, however, the two share similar volatility distribution, because the two locations have low •OH and the high volatility organics won't get oxidized as much. The difference in •OH levels also explains the diurnal variability in the other two locations, where the difference in the amount of pre-existing nonvolatile organics gives the two locations different volatility distributions .

#### Gas phase




As discussed for the total organics, low •OH levels make the organics in Beijing and Hyytiälä stay mostly in the high volatility range with high gas phase concentrations, whereas Centreville and Mexico City have low gas concentration because the high volatility gases are more efficiently oxidized by •OH and partitioned into the aerosol phase. Although Centreville and Mexico City share similar amount of •OH, the former's total reaches an equilibrium whereas the latter decreases because of the difference in the amount of pre-existing nonvolatile organics in the two locations as mentioned before.

#### Aerosol phase




Continuing the gas phase discussion, even though Beijing also has high amount of pre-existing nonvolatile organics compared to Hyytiälä, the two still share resemblance because the latter has very low temperature due to high latitude and volatility is temperature dependent.

Similar analyses can be made for the aerosol phase, which is the inverse of the gas phase.

**Stage 3: Sensitivity tests & include condensing organics in dust and sea salt aerosols**
**Stage 4: Simplification: Reduce number of tracers**
**Stage 4: Implementation: Simple Coupling with GCM -** box model is a module within GISS ModelE2.