An Efficient Representation of Aerosol Mixing State for Atmospheric Models

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What is aerosol mixing state?

Aerosol mixing state: distribution of per-particle chemical species composition. [Riemer and West 2013]

externally mixed population
What is aerosol mixing state?

Aerosol mixing state: distribution of per-particle chemical species composition. [Riemer and West 2013]
Both CCN and optical properties depend on size and mixing state.
Objective

To reliably predict *CCN and optical properties* as a function of size and mixing state at a *reasonable computational cost*

- Develop a novel sectional framework to resolve aerosol mixing state: MOSAIC-mix.
- Apply the model using single particle measurements (SPLAT-II and SP2) during CARES to simulate aerosol mixing state evolution.
In each size bin, all particles have same BC mass fraction $w_{BC}$ and same hygroscopicity ($\kappa$).

Zaveri et al. (2008) MOdel for Simulating Aerosol Interactions and Chemistry
**MOSAIC-mix sectional framework**

**MOSAIC:**
Sectional approach resolving size only

In each size bin, all particles have same BC mass fraction $w_{BC}$ and same hygroscopicity ($\kappa$)

Zaveri et al. (2008)

**MOSAIC-Mix:**
Resolves size, BC mass fraction and hygroscopicity

Ching et al. (2016)
Approach

► Use 10 idealized urban scenarios to simulate aerosol mixing state evolution under different emissions and environmental conditions.

► Optimize the sectional framework using a high-resolution version of MOSAIC-mix and particle-resolved model PartMC-MOSAIC [Riemer et al. 2009].

10 Urban Plume Scenarios
- Gaseous emissions
- Black carbon emission
- Background particle concentration
- Solar radiation
- Temperature
- Relative humidity
Approach

High-resolution MOSAIC-mix with $24 \, D_{\text{dry}} \times 35 \, w_{\text{BC}} \times 30 \, \kappa$
Benchmarking

- High resolution MOSAIC-mix was evaluated against particle-resolved model PartMC-MOSAIC.
Approach

- Devise low-resolution MOSAIC-mix
Approach

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Approach

- Devise low-resolution MOSAIC-mix
About 2,000 low-resolution configurations in 1D, 2D, or 3D ($24D_{dry} \times 1-3w_{BC} \times 1-3\kappa$) with different choices of bin boundaries evaluated against the high-resolution configuration.
Performance

CCN concentrations

- Individual scenarios
- Average of 10 scenarios

Internally mixed
(24 x 1 x 1)

(24 x 2 x 2)

D\text{dry}

WBC

Proudly Operated by Battelle Since 1965
Performance

CCN concentrations

Optical properties

Internally mixed

(24 x 1 x 1)

(24 x 2 x 2)

Performance

CCN concentrations

Optical properties

Internally mixed

(24 x 1 x 1)

(24 x 2 x 2)

D_{dry}

W_{BC}

D_{dry}
CARES campaign

June 2 – 28, 2010

CARES Objectives:

- Investigate Anthropogenic-Biogenic Interactions in SOA formation.
- Investigate black carbon (BC) mixing state evolution.
- Quantify the effects of aerosol ageing on aerosol optical and CCN activation properties.

Zaveri et al. ACP 2012
Lagrangian simulations by MOSAIC-mix and PartMC-MOSAIC

- Used **5 trajectories** originating from T0 on June 15 from a FLEXPART-WRF simulations Fast et al. (2012).
- **Same gas and aerosol emission, initial and background conditions** were input to both PartMC-MOSAIC and MOSAIC-mix.
- Model simulations were compared to G-1 observations.
Model initialization using single particle measurements at T0

To derive initial conditions for model simulations, we used:

- SMPS and APS - size distributions
- SPLAT–II - size and mixing state distributions of non-BC containing particles
- SP2 – size and mixing state distributions of BC-containing particles
Deriving BC size and mixing state distribution from SP2

Assumed: Spherical shape
Core-shell structure

Total diameter: \( D_{\text{total}} = D_{\text{core}} + D_{\text{coat}} \times 2 \)
Deriving BC size and mixing state distribution from SP2

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Total diameter: \( D_{\text{total}} = D_{\text{core}} + D_{\text{coat}} \times 2 \)

Core diameter distribution

\[
dN / d\log_{10} D_{\text{core}} / \text{cm}^{-3}
\]

\[
D_{\text{core}} \quad (\text{Core diameter or Mass Equivalent Diameter})
\]

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Deriving BC size and mixing state distribution from SP2

Assumed: Spherical shape
Core-shell structure

Total diameter: $D_{\text{total}} = D_{\text{core}} + D_{\text{coat}} \times 2$

Core-coating thickness distribution

Core diameter distribution

$dN / d\log_{10}D_{\text{core}} / \text{cm}^{-3}$
We consider 7 particle classes from SPLAT, \textit{6 organic-sulfate and sea salt}.
Deriving size and mixing state distribution from SPLAT- II and SP2

Initial size distributions
Input to model

<table>
<thead>
<tr>
<th>dN / dlog10Dp / cm⁻³</th>
</tr>
</thead>
<tbody>
<tr>
<td>10⁴</td>
</tr>
<tr>
<td>10²</td>
</tr>
<tr>
<td>10⁰</td>
</tr>
<tr>
<td>10⁻²</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Diameter / μm</th>
</tr>
</thead>
<tbody>
<tr>
<td>10⁻²</td>
</tr>
<tr>
<td>10⁻¹</td>
</tr>
<tr>
<td>10⁰</td>
</tr>
<tr>
<td>10¹</td>
</tr>
</tbody>
</table>

- SMPS+APS
- SP2
- Organic-sulfate 1
- Organic-sulfate 2
- Organic-sulfate 3
- Organic-sulfate 4
- Organic-sulfate 5
- Organic-sulfate 6
- Sea Salt
Deriving size and mixing state distribution from SPLAT-II and SP2

Initial size distributions
Input to model

10 Initial size distributions input to the model derived from SP2

$\frac{dN}{d\log_{10}D_p} / \text{cm}^{-3}$

$Diameter / \mu m$

$10^{-2}$ $10^{-1}$ $10^0$ $10^1$

$10^{-2}$ $10^{-1}$ $10^0$ $10^1$
Aerosol mass concentrations simulations vs G-1 observations

- **BC**
- **POA**
- **SO\textsubscript{4}**
- **NO\textsubscript{3}**
- **NH\textsubscript{4}**
- **Total organics**

**Mass concentration / μg m\textsuperscript{-3}**

**Time (UTC) / h**

- **PartMC-MOSAIC simulations**
- **MOSAIC-mix simulations**
- **G-1 Observations**
Size-resolved vs mixing-state-resolved

Absorption coefficient

10 Idealized scenarios
(Ching et al., 2016)
5 CARES trajectories
× Average

NRMSD w.r.t. HR MOSAIC-mix / %

Internally mixed
(24 x 1 x 1)
(24 x 2 x 2)

Bin Configuration
Summary

► Developed a novel sectional framework that efficiently resolves mixing state: MOSAIC-mix.

► Showed that mixing-state-resolved simulations better predict the CCN and optical properties than size-resolved only simulations with a small number of mixing state bins.

► Applied the model using initial conditions from SPLAT-II and SP2 single particle measurements during CARES.

► MOSAIC-mix is being implemented in WRF-Chem to assess the impacts of aerosol mixing state at regional scale.

For more details, please refer to poster 71.