Simulating aerosols, radiative forcing, and impacts on marine stratocumulus during VOCALS REx

Scott Spak, Marcelo Mena-Carrasco, Pablo Saide, Greg Carmichael
Motivations: Policy Relevance

- How have anthropogenic emissions altered SEP Sc? Which sources matter most? How are they expected to change?
- Unintended regional/global climate consequences of Santiago’s urban development?
- What about geo-engineering proposals?
- The SEP, beyond clouds and climate
  - traditional criteria air pollution questions for human health
  - direct radiative forcing and BC:S strategies
  - acid deposition to the southern oceans
Motivation → Approach

Simulate & evaluate basic properties of chemical transport during REx

*then* aerosol effects on cloud microphysics & climate

*then* response to changes in human activities
Building on ... tracer modeling

- Daily variability in $SO_4$ driven by synoptic condition: SEP Subtropical High location & strength
- DMS importance by omission: anthropogenic emissions can’t account for observed offshore $SO_4$ & $SO_2$ concentrations
- Coastal and Altiplano emissions entrained from FT to clouds in WRF-STEM

$\Delta$ Cloud-level LPS $SO_4$ ($\mu g/m^3$)
RF03-RF06 & RF10 – other flights

Model Configuration

WRF-Chem v3.1.1 @ 12 km, 27 layers
- CBM-Z + MOSAIC (8 bins), Lin microphysics, Grell cumulus
- **No DMS yet!**
- **MYNN v2.5 PBL**
- Direct, 1\textsuperscript{st} & 2\textsuperscript{nd} indirect, semi-direct aerosol radiative forcing
- Restarted every 5 days with 1 day spin-up for continuity
- MOZART 4 boundary conditions
- **Emissions**
  - daily 1 km FINN fires (Wiedinmeyer et al., GMDD, 2010)
  - + MEGAN biogenics
  - + VOCA anthropogenic emissions **ON/OFF through 10/27**
### Campaign chemical transport evaluation

No DMS, too much OC (FINN + MOZART)

<table>
<thead>
<tr>
<th></th>
<th>Observed</th>
<th>Mean Bias</th>
<th>RMSE</th>
<th>FB*</th>
<th>FE*</th>
</tr>
</thead>
<tbody>
<tr>
<td>O3 (ppb)</td>
<td>37.25</td>
<td>-3.53</td>
<td>10.47</td>
<td>-0.12</td>
<td>0.26</td>
</tr>
<tr>
<td>CO (ppb)</td>
<td>65.68</td>
<td>2.60</td>
<td>20.40</td>
<td>0.06</td>
<td>0.13</td>
</tr>
<tr>
<td>SO2 (ppt)</td>
<td>52.71</td>
<td>-32.00</td>
<td>152.04</td>
<td>-1.25</td>
<td>1.49</td>
</tr>
<tr>
<td>SO4 (µg/m³)</td>
<td>0.43</td>
<td>-0.01</td>
<td>0.73</td>
<td>0.34</td>
<td>1.09</td>
</tr>
<tr>
<td>NO3 (µg/m³)</td>
<td>0.011</td>
<td>0.002</td>
<td>0.035</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>NH4 (µg/m³)</td>
<td>0.08</td>
<td>-0.01</td>
<td>0.11</td>
<td>0.35</td>
<td>1.31</td>
</tr>
<tr>
<td>OC (µg/m³)</td>
<td>0.13</td>
<td>0.37</td>
<td>0.52</td>
<td>1.58</td>
<td>1.81</td>
</tr>
</tbody>
</table>

*Fractional bias = (2 x bias)/(observed + modeled)

**Bolded values meet US EPA/community performance standards**

**Italicized values exceed targets**
20º S chemical transport evaluation
all C-130 flights @ 1 minute average, processed per Allen et al. (ACPD)

<table>
<thead>
<tr>
<th></th>
<th>Observed</th>
<th>Mean Bias</th>
<th>RMSE</th>
<th>FB*</th>
<th>FE*</th>
</tr>
</thead>
<tbody>
<tr>
<td>O3 (ppb)</td>
<td>34.84</td>
<td>-5.71</td>
<td>10.83</td>
<td>-0.22</td>
<td>0.30</td>
</tr>
<tr>
<td>CO (ppb)</td>
<td>65.89</td>
<td>0.83</td>
<td>16.37</td>
<td>0.03</td>
<td>0.11</td>
</tr>
<tr>
<td>SO2 (ppt)</td>
<td>34.31</td>
<td>-16.06</td>
<td>61.75</td>
<td>-1.21</td>
<td>1.52</td>
</tr>
<tr>
<td>SO4 (µg/m³)</td>
<td>0.47</td>
<td>-0.07</td>
<td>0.63</td>
<td>0.06</td>
<td>1.13</td>
</tr>
<tr>
<td>NO3 (µg/m³)</td>
<td>0.011</td>
<td>0.001</td>
<td>0.021</td>
<td>NA</td>
<td>NA</td>
</tr>
<tr>
<td>NH4 (µg/m³)</td>
<td>0.08</td>
<td>-0.02</td>
<td>0.10</td>
<td>-0.03</td>
<td>1.36</td>
</tr>
<tr>
<td>OC (µg/m³)</td>
<td>0.12</td>
<td>0.33</td>
<td>0.47</td>
<td>1.44</td>
<td>1.81</td>
</tr>
</tbody>
</table>

Bolded values meet US EPA/community performance standards
Italicized values exceed targets
WRF-Chem MBL Gases @ 20ºS vs. Allen et al. (2011)

Fig. 4. Sulphur dioxide (top panels), ozone (centre panels) and carbon monoxide (lower panels) concentration statistics in the MBL (left panels) and the FT (right panels) gridded into 84, degree longitudinal zones along the 86º South parallel averaged from aircraft measurements during VOCALS-REx. For each zone, thick centre lines indicate the median, dashed lines indicate the mean, boxes indicate upper and lower quartiles with upper and lower decile whiskers. Plotted above each longitude zone and for each panel, numbers before the comma indicate number of flights contributing to each statistic, followed by the number of straight and level runs, with the total sampling time in decimal hours in parentheses.
FT Gases: whiskers better than boxes
MBL aerosol composition

Figure 7. Longitudinally gridded aerosol mass concentrations for: organics (top panels); ammonium (middle panels); and sulphate (bottom panels) for the marine boundary layer between -85° and -70° longitude. For each zone, thick centre lines indicate the sample median, dashed lines indicate the mean, boxes indicate upper and lower quartiles with upper and lower decile whiskers. Median mass concentrations measured by the Ron Brown research vessel are plotted as purple circles for the marine boundary layer, with purple box and whisker plots for periods when the Ron Brown was anchored on station. Plotted above each longitude zone and for each panel, numbers before the comma indicate number of flights contributing to each statistic, followed by the number of straight and level runs, with the total sampling time in decimal hours in parentheses.
FT aerosol composition

Fig. 7. Longitudinally gridded aerosol mass concentrations for: organics (green top panels); ammonium (yellow middle panels); and sulphate (red bottom panels) for the marine boundary layer between -85° to -70° (left panels) and the free troposphere between -85° to -70° (right panels). For each zone, thick centre lines indicate the sample median, dashed lines indicate the mean, boxes indicate upper and lower quartiles with upper and lower decile whiskers. Median mass concentrations measured by the Ron Brown research vessel are plotted as purple circles for the marine boundary layer, with purple box and whisker plots for periods when the Ron Brown was anchored on station. Plotted above each longitude zone and for each panel, numbers before the comma indicate number of flights contributing to each statistic, followed by the number of straight and level runs, with the total sampling time in decimal hours in parentheses.
Cloud Condensation Nuclei

Fig. 10. Aerosol concentration and cloud physics measurement statistics averaged into 73.2° longitude zones below 6755 m altitude (left panels) and between 6755–8755 m altitude (right panels). For each vertical domain, cloud condensation nucleus concentration (CCN), cloud droplet number (CDN), accumulation mode aerosol concentration (ACN), and total condensation nuclei greater than 65 nm particle diameter (CN) are plotted as titled. With the exception of CCN data, for each longitude zone, thick center lines indicate the sample median, dashed lines indicate the mean, boxes indicate upper and lower quartiles with upper and lower decile whiskers. Only median data are presented for CCN, color-coded to supersaturation as labeled in the figure. Plotted above each longitude zone and for each panel, numbers before the comma indicate the number of flights contributing to each statistic, followed by the number of straight and level runs, with the total sampling time in decimal hours in parentheses.
A word about CCN in WRF-Chem...

- WRF-Chem assumes 0.2% supersaturation for aerosol coupling
- CCN not well simulated by any WRF supersaturation level: observed @ 0.55 – 0.75% < modeled CCN @ 0.5%
- Variability along flight paths encouraging
Average 10/15 – 10/27 ozone (ppb), cloud level

Base case

Anthropogenic impact

seq(from = -391.5 * 12000, length.out = 269, by = 12000)
Anthropogenic impact: cloud-level aerosol properties

Number concentration enhancement by LPS >> central Chile
Anthropogenic impact: cloud-level CCN (cm$^{-3}$)

0.2% supersaturation

0.5% supersaturation

Order of magnitude underestimate in local coastal effect in WRF-Chem
Water vapor (g/kg), cloud level

20° S changes due to LPS (coastal) and central Chile (offshore)
Radiative forcing (W/m²)

Downwelling SW @ SFC

Upwelling LW @ TOA
Anthropogenic impact on Temperature (K)

Coastal surface cooling by brighter clouds, offshore cloud-level cooling by longer cloud lifetime
Conclusions

- **Emissions & chemical transport: cautious but confident**
  - Most species of interest simulated as well or better than regulatory modeling standards, prior airborne field campaigns
  - Aerosols offshore @ 20S: central Chile + northern large point sources
  - Natural emissions are key to improvement: DMS & biomass burning essential, sea salt algorithms and OC too high

- **Modeling anthropogenic aerosol influence on clouds & climate: large but uncertain**
  - Very strong anthropogenic indirect radiative forcing
  - Average >0.5 °C surface & in-cloud cooling
  - CCN diverges from observations, insensitive to anthropogenic emissions
Ongoing extensions

- Santiago daily operational WRF-Chem PM$_{10}$/PM$_{2.5}$ forecast
- Adjoint WRF-Chem aerosol direct/indirect/semi-direct effects
- Effects of Santiago’s development on regional climate and SEP Sc
  - land use/density/transportation
  - vehicle/energy technology

Next steps

- 1 km simulations on new UI supercomputer to resolve clouds & changes in Santiago’s urban form
- Ocean-atmosphere-aerosol coupling WRF-Chem with ROMS
  - effects of aerosols on SEP SSTs, ENSO
  - aerosol vs. ocean contributions to Sc variability
- Adjoint applications
  - emissions sectors & aerosol composition impacting 20° s clouds
  - SO$_2$ emissions inversion: an initial test for 4DVAR
  - constraining box/column simulations with REx observations to isolate process errors in aerosol-microphysics interactions
Larger Questions

• How much skill must >LES models demonstrate to be useful?
  - primary: aerosols (concentrations, AOD, CCN, CDN, CN)
  - secondary: clouds (LWP, drizzle/rain rate, $\tau_{\text{CLOUD}}$, brightness temperature)
  - tertiary: climate (SST, ocean/atmosphere energy balance, radiative forcing)

• Beyond “lots of CCN” geoengineering, other scenarios of interest?

• VOCALS “summary for policymakers” on atmosphere-ocean-cloud interactions
  - confidence in processes, answers to hypotheses
  - community metrics for model performance: criteria & target
  - specific process improvements needed
  - REx findings, long-term reanalyses, projections
Acknowledgements

- VOCALS science and observational teams
- Data & Modeling
  - C. Wiedinmeyer, NCAR
  - L. Emmons, NCAR
  - J. Fast, PNNL

Funding
Building on...

VOCA Emissions Inventory

- Anthropogenic
  - EDGAR 3.2 FastTrack 2000 + Bond et al. (2004) BC/OC @ LandScan
  - CONAMA/CODELCO 2008 for Chile
    - 1,400+ point sources
    - Municipal-level residential, industrial, mobile area sources

- Volcanoes & Peruvian smelters from OMI SO2 during VOCALS REx inverted as in Cairn et al. (2007)
Episodic temperature (K): 10/15/08 only

Surface

Cloud top

Point source sulfate -> brighter coastal clouds, surface cooling

Less drizzle, inhibited convection -> offshore surface warming