RETHINKING THE LIFETIME OF BIOMASS-BURNING-AEROSOL IN THE FREE TROPOSPHERE

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DOE ASR June 24
OVERVIEW

- ORACLES 2016-2017 HR-ToF-AMS DATA
- WRF_AAM Mean Age
- F44 as age tracer for in situ aerosol

Still seeing oxidation with age for 4-14 days.
BOX PLOTS: ORGANIC AEROSOL LOSS WITH AGE

OA/BC vs Age

TOC/BC vs Age

SO₄/BC vs Age

NO₃/BC vs Age

NH₄/BC vs Age
Implications with NO$_3$ thermodynamics with respect to age

- NO$_3$ in the particle phase favors high humidity and low temperatures
- Will return to gas phase at low RH and high temp.
- Are we seeing loss due to thermodynamics or loss due to oxidation reactions?
TWO FLIGHT COMPARISON – COASTAL VS ROUTINE
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- MOTIVATION WAS WU ET AL., 2020
- REPORT INORGANIC NITRATE NEAR ASCENSION ISLAND
- ORACLES APPEARS TO BE DOMINANTLY ORGANIC NITRATE
- ORACLES DATA NEAR ASCENSION SHOWS <50% ORGANIC NITRATE
- HYPOTHESIS: AS AEROSOL AGES THE ORGANIC NITRATE IS LOST, THUS LEAVING HIGHER FRACTIONS OF INORGANIC NITRATE.
- SAFARI 2000 REPORTS INORGANIC NITRATE
TWO FLIGHT COMPARISON – COASTAL VS ROUTINE

• CAN WE DISTINGUISH HOW MUCH OF ORGANIC AEROSOL LOST IS DRIVEN BY THERMODYNAMICS AND HOW MUCH IS DRIVEN BY PHOTOCHEMISTRY?

• CAN WE SEE THESE THERMODYNAMIC AND PHOTOCHEMICAL PROCESSES WITH AEROSOL OLDER THAN 9 DAYS?

MOVING FORWARD: ASCENSION ISLAND AEROSOL IS PREDICTED TO BE OLDER THAN ORACLES AEROSOL.
Can chemical differences be resolved between 3 campaigns? Higher % of BC toward Ascension Island?

ORACLES Sept. 2016

ORACLES August. 2017

CLARIFY FT Period 2

LASIC 08/07-08/17

CLARIFY data courtesy of HuiHui Wu
Does the higher % of BC relative to OA affect optical properties?

What about size?
<table>
<thead>
<tr>
<th>Instrument</th>
<th>Data collected</th>
<th>Campaign(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aerosol Mass Spectrometer (AMS)</td>
<td>Bulk Chemical Species (NO$_3$, OA, OM SO$_4$, NH$_4$) ($\mu$g/m$^3$), HR ratios (O/C, H/C)</td>
<td>LASIC (ACSM), ORACLES (HR-AMS), CLARIFY(ACSM)</td>
</tr>
<tr>
<td>Single Particle Soot Photometer (SP2)</td>
<td>Black Carbon (BC) ($\mu$g/m$^3$)</td>
<td>LASIC, ORACLES, CLARIFY</td>
</tr>
<tr>
<td>Los Gatos Research CO/CO$_2$ Analyzer</td>
<td>CO, CO$_2$, O$_3$ (ppbv)</td>
<td>LASIC (CO only), ORACLES, CLARIFY</td>
</tr>
<tr>
<td>TSI Nephelometer</td>
<td>Scattering (450,530,700nm) Mm$^{-1}$</td>
<td>LASIC, ORACLES, CLARIFY</td>
</tr>
<tr>
<td>Particle Soot Absorption Photometer (PSAP)</td>
<td>Absorption (450nm, 530nm, 660nm) Mm$^{-1}$</td>
<td>LASIC, ORACLES, CLARIFY</td>
</tr>
<tr>
<td>Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)</td>
<td>Aerosol size between 10nm and 1000nm</td>
<td>LASIC, ORACLES, CLARIFY</td>
</tr>
</tbody>
</table>

- Use Positive Matrix Factorization along with high resolution AMS analysis techniques to distinguish chemical differences between the LASIC/ORACLES/CLARIFY campaigns.
- Relate the results of the chemical analysis to differences in optical and physical properties between the three campaigns.
- Establish how well the differences can be attributed to an irreversible aging process (e.g., photooxidation).