Aerosol aging and in-cloud scavenging over the southeast Atlantic

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Southern Africa produces \(\sim\)1/3 of all biomass burning aerosol
- Thousands of small fires
- Smoke advects over SE Atlantic at 2 to 6 km
- Obscures stratocumulus cloud deck
- Challenging region for satellites and models
- Aerosol direct effect is uncertain
- Ideal area for studying indirect effects of aerosol on cloud

2018/09/08 MODIS data
https://worldview.earthdata.nasa.gov/
Primarily radiation and cloud instrumentation. Limited chemistry payload:
HR-ToF-AMS
SP2
CO, CO₂, O₃, water isotopes

(No PTR-MS, no VOC, no NOx)
Results—Plume Structure
Discussion: Aerosol Reactions: Conserved BB tracers

- CO, CO$_2$, and BC are all conserved tracers
- Use CO and CO$_2$ to calculate **Modified Combustion Efficiency** (MCE)
  - MCE = $\Delta$CO$_2$ / ($\Delta$CO + $\Delta$CO$_2$)
- MCE for ORACLES 2016 = 0.98-0.995
- Very narrow range; efficient combustion

Collier et al. (2016)
What happens as BB aerosol ages?

- Lab and field studies often cover the first minutes to a day or so.
- Much harder to look at the decay after a few days—identifying sources and age, mixing with other plumes.
- Lagrangians are an obvious way, but really tough to pull off, particularly over several days.
- ORACLES is a unique situation, with massive, fairly uniform sources, no other large sources, and a range of ages.
BC:CO fairly constant

OA:BC less so
- ORACLES near endpoint of BB oxidation.
- f60 no good as aging tracer
- Some variation in f44

From Cubison et al., 2011, ACP 11(23):12049–12064
Results – Plume Structure

09/24/16 RF12

Vertical profile

09/24/16 RF12 Vertical Profile
UTC 12:44 - 13:01

upper plume
lower plume

Altitude (m)

Latitude °N

NO3:OA
Marker sized to OA

12%
6%
8%
5%
11%
70%
68%
8%
5%
11%
8%

BC
SO4
NH4
NO3
OA
Acid/Base chemistry indicates NO3 is important, organic acids are not

- Neglecting NO3 yields basic aerosol which doesn’t happen
- If organic acids were important, there would be red points above the 1:1 line
- Apparently an excess of NO3, available to neutralize NH4, but driven off by SO4
- Perhaps the higher SO4:OA plumes are older, as SO2 reacts to form H2SO4
Without chemical age tracers, check models. Low odds: tough problem.

Pablo Saide implemented an experimental age tracer in WRF-Chem.
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It appears there is a correlation between model age and f44. Gives some confidence in model ages.
Thus we can look at OA loss as a function of model time.

Exponential fit with time constant of 0.18, about 6 days. Other flights range from 3 to 8 days.
Cloud Droplet Residual Particles in Biomass burning Influenced Stratocumulus Clouds

Counterflow Virtual Impactor (CVI)
A mechanism to sample cloud droplet residual particles

Small particles caught by counterflow, rejected.
Large particles penetrate the stagnation zone, are sampled.

https://www.esrl.noaa.gov/gmd/aero/instrumentation/cvi.html
Overall about 7 instances with data above, below, and in cloud with AMS on CVI

30 August, 12:30 to 14:30 UTC

1. Overflight/LIDAR curtain
2. Spiral down, complete profile
3. Low altitude MBL run
4. Just above cloud leg
5. In cloud leg
6. Pollution leg
What affects composition of droplet nuclei in stratocumulus cloud?

Particle population in the MBL
Range of supersaturations
Particle size and hygroscopicity
Particles entrained from above
Precipitation scavenging
(Instrument artifacts)
* Total aerosol mass would include sea salt, dust, and refractory organics, ignored here.
An example: ease of activation appears to control composition here.

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But something else is going on here. Looks like aerosol above, but CO rules out entrainment.
Most likely this reflects scavenging of hygroscopic salts, leaving less soluble organics behind.
But wouldn’t that also leave even less hygroscopic rBC behind? It’s reduced instead. Must be on large particles.
Another example of apparent precipitation scavenging across convective cells
CONCLUSIONS

Aged BBOA has a decay timescale of ~6 days (half life 3.5 days)

Precipitation scavenging has a strong effect on composition, removing large particles and soluble material.

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