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

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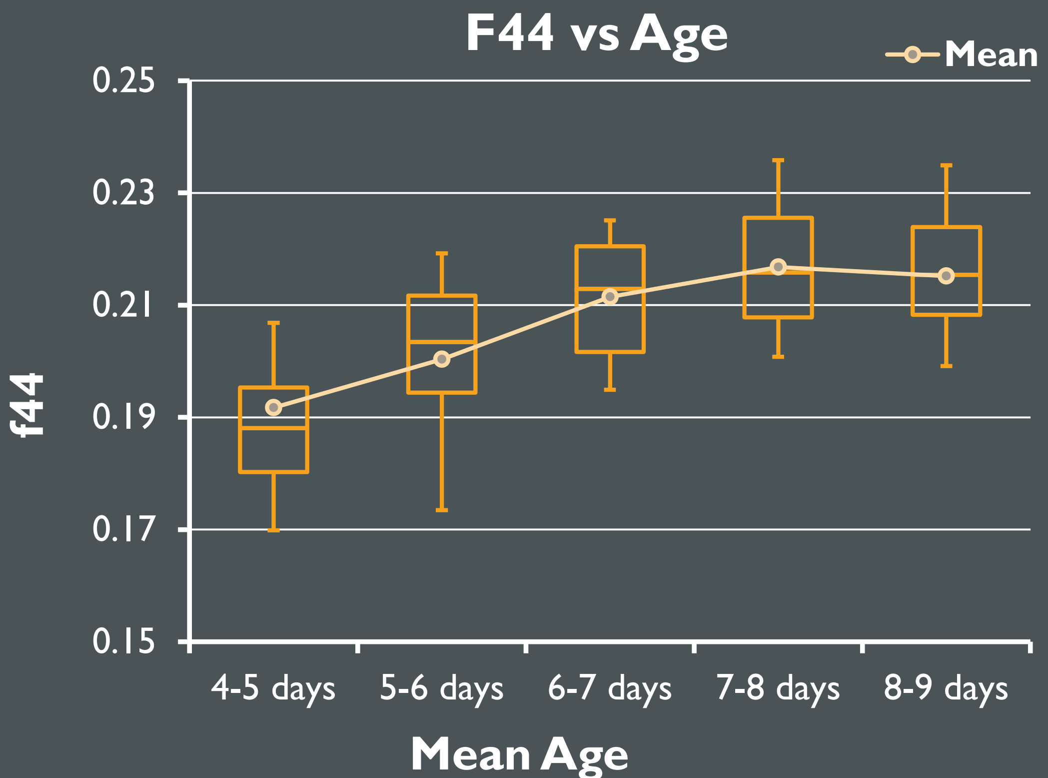
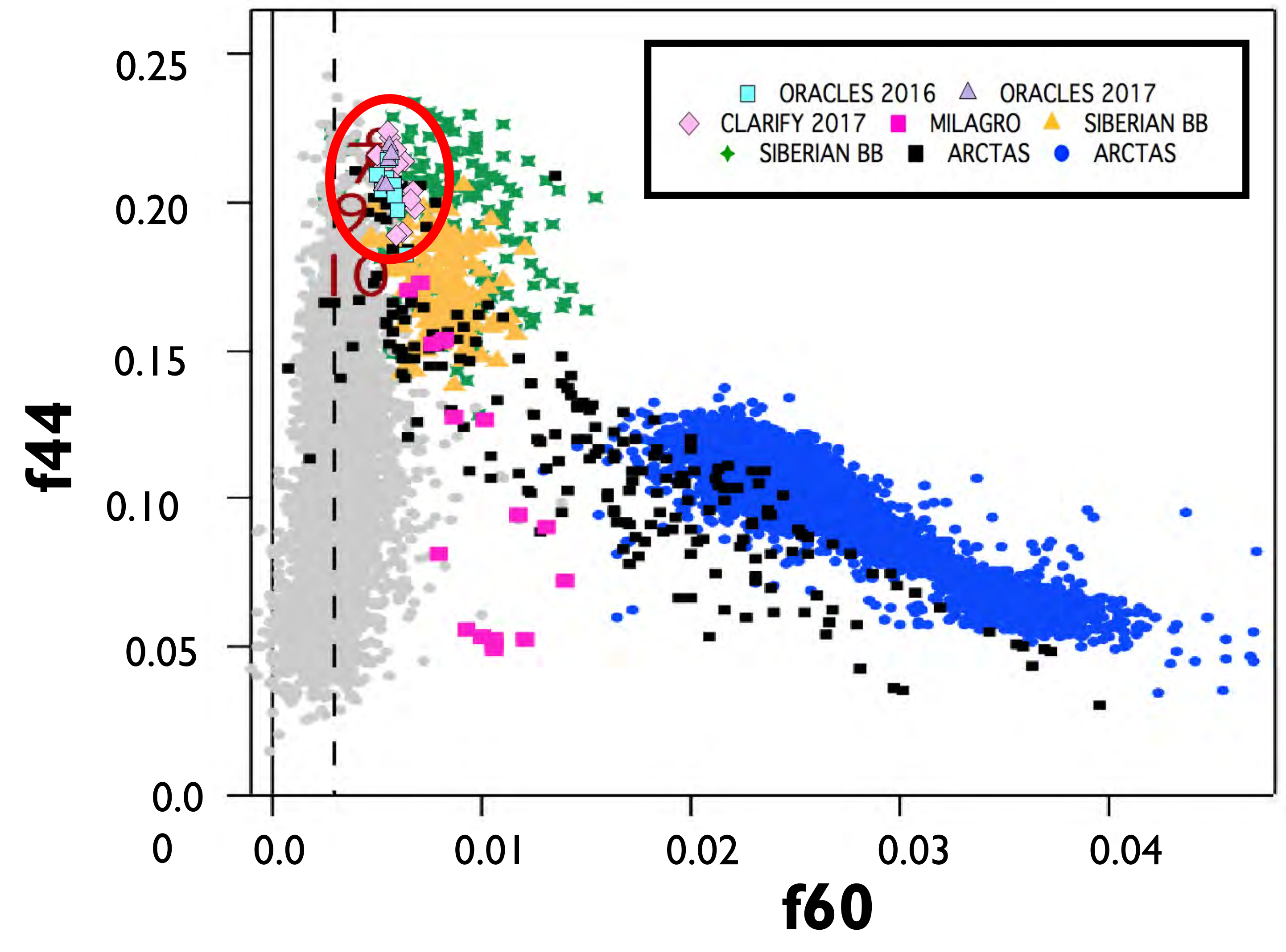
DOE ASR June 24

OVERVIEW

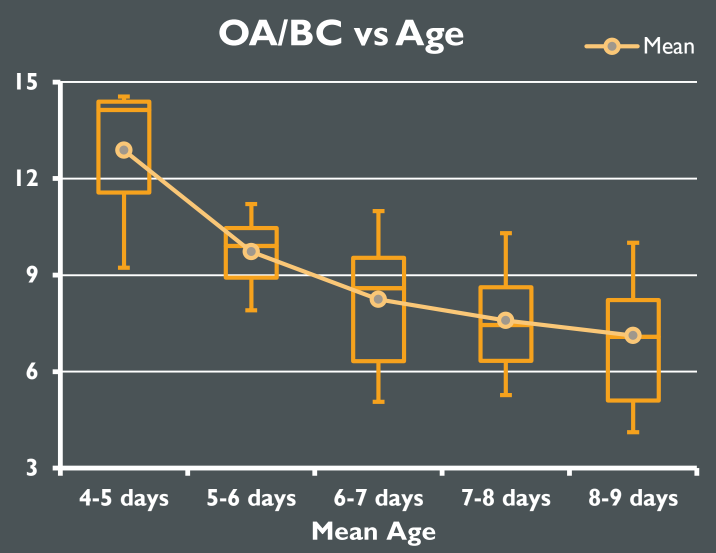
- ORACLES 2016-2017 HR-ToF-AMS DATA
- WRF_AAM Mean Age
- F44 as age tracer for in situ aerosol

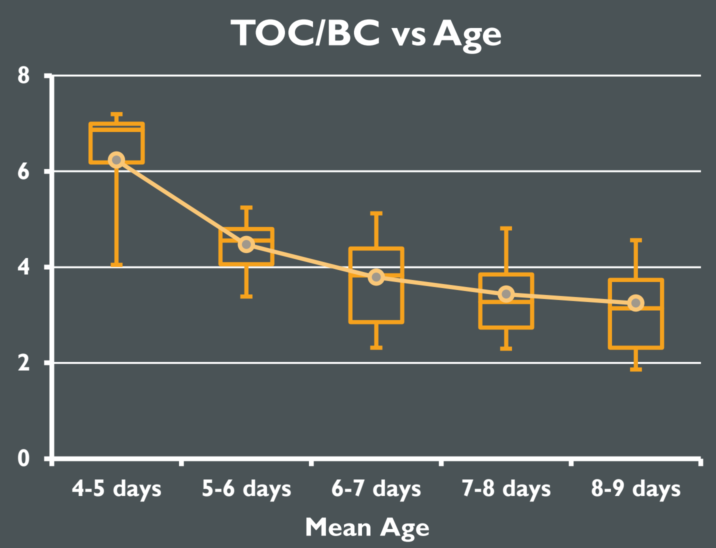


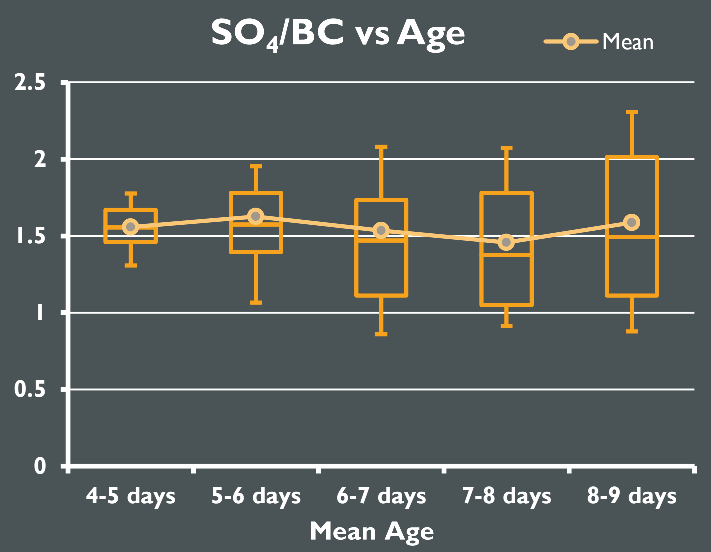


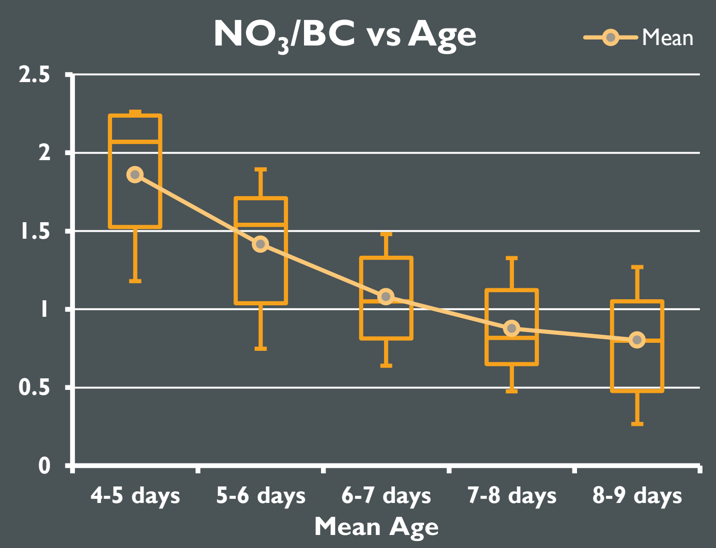
Still seeing oxidation with age for 4-14 days.

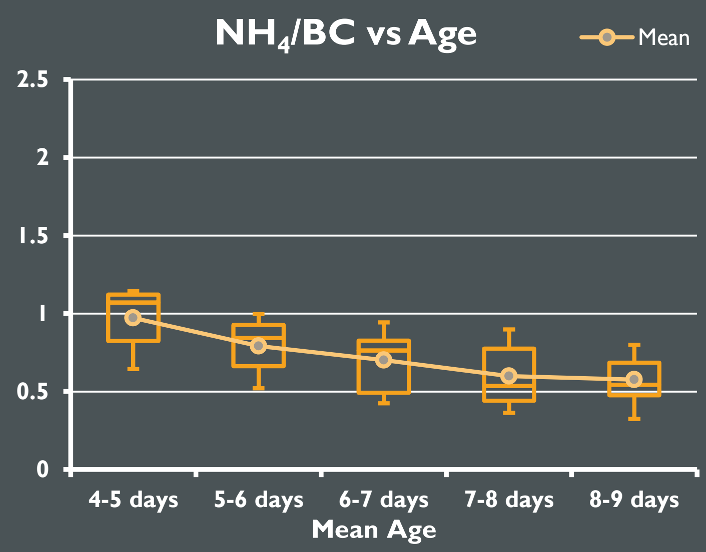
BOX PLOTS: ORGANIC AEROSOL LOSS WITH AGE







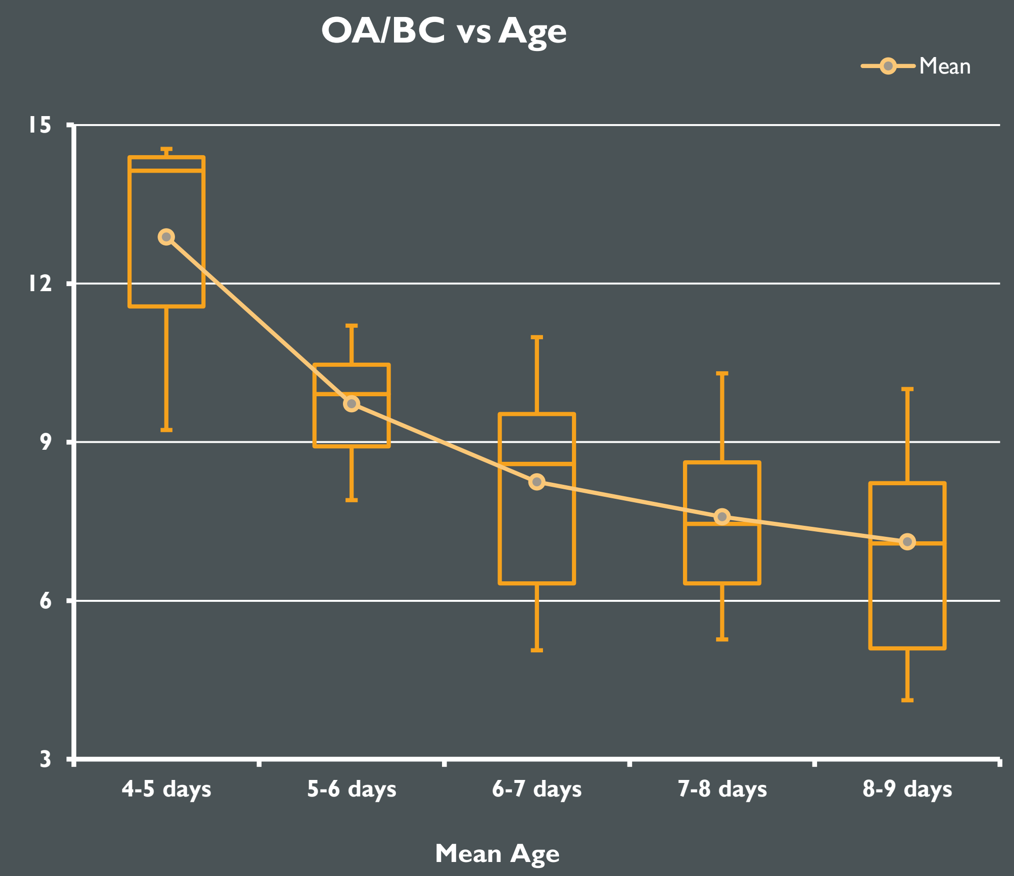


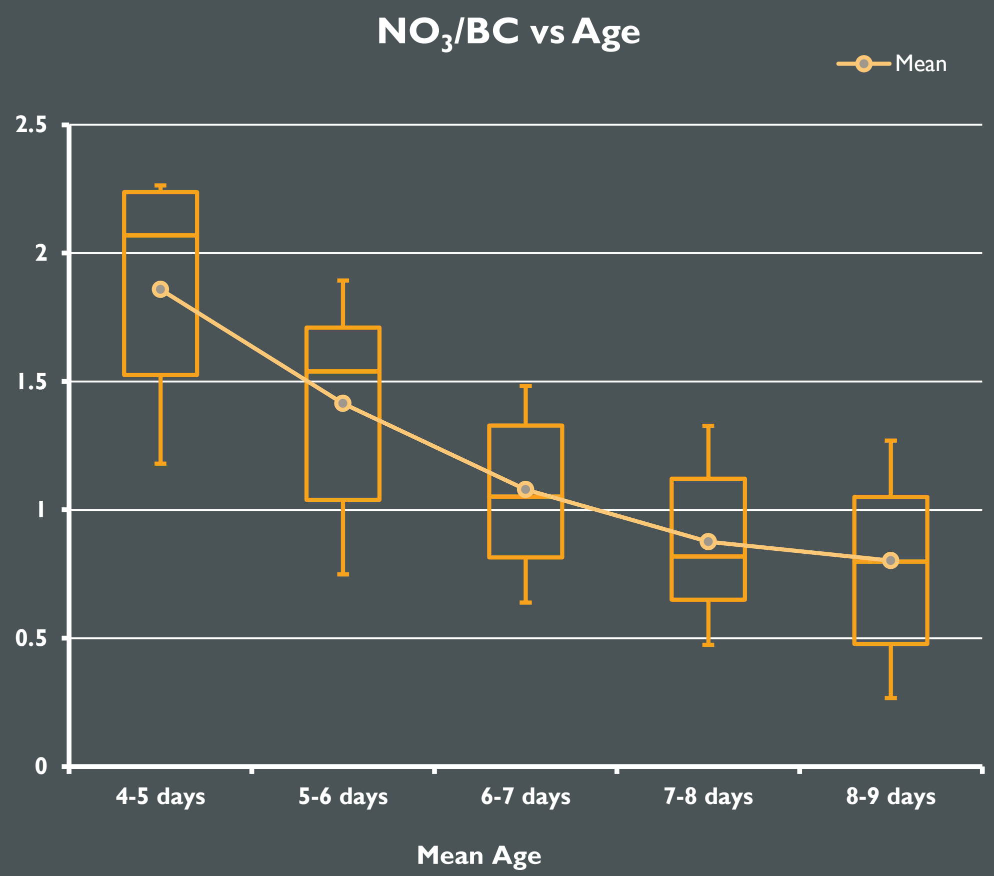


CROSSROADS BETWEEN THERMODYNAMICS AND PHOTOCHEMISTRY

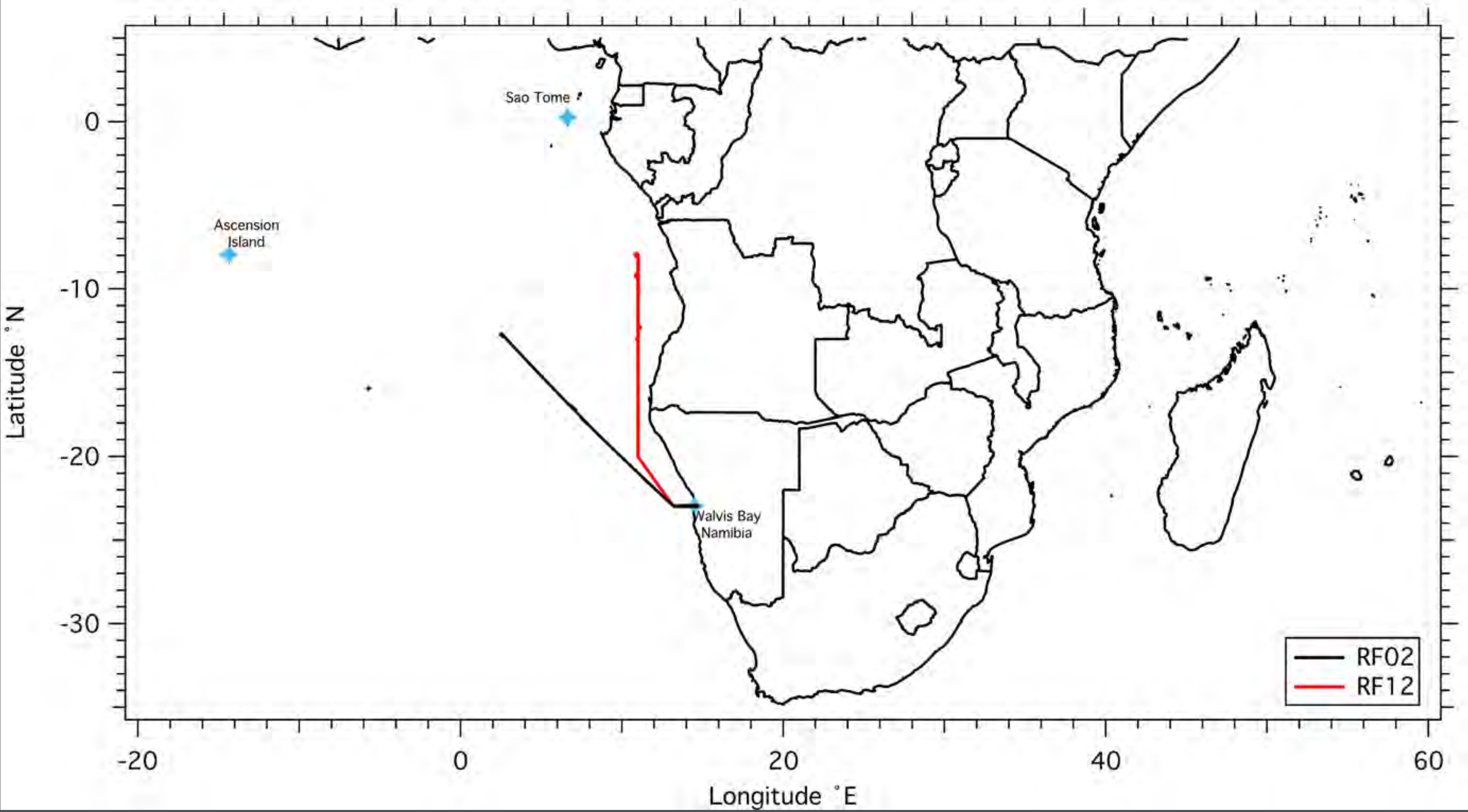
Implications with NO_3 thermodynamics with respect to age

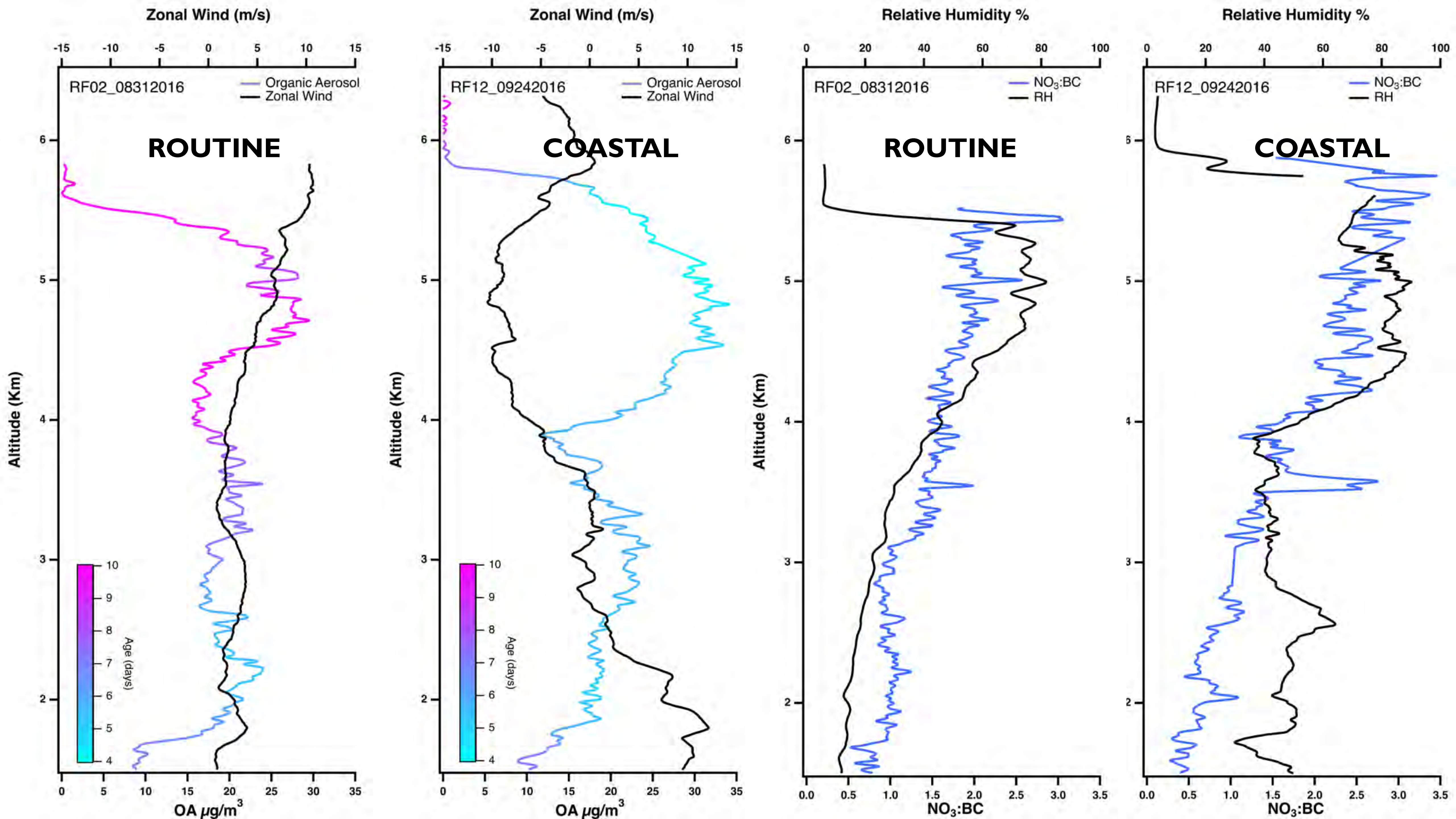
- NO_3 in the particle phase favors high humidity and low temperatures
  - Will return to gas phase at low RH and high temp.
- Are we seeing loss due to thermodynamics or loss due to oxidation reactions?





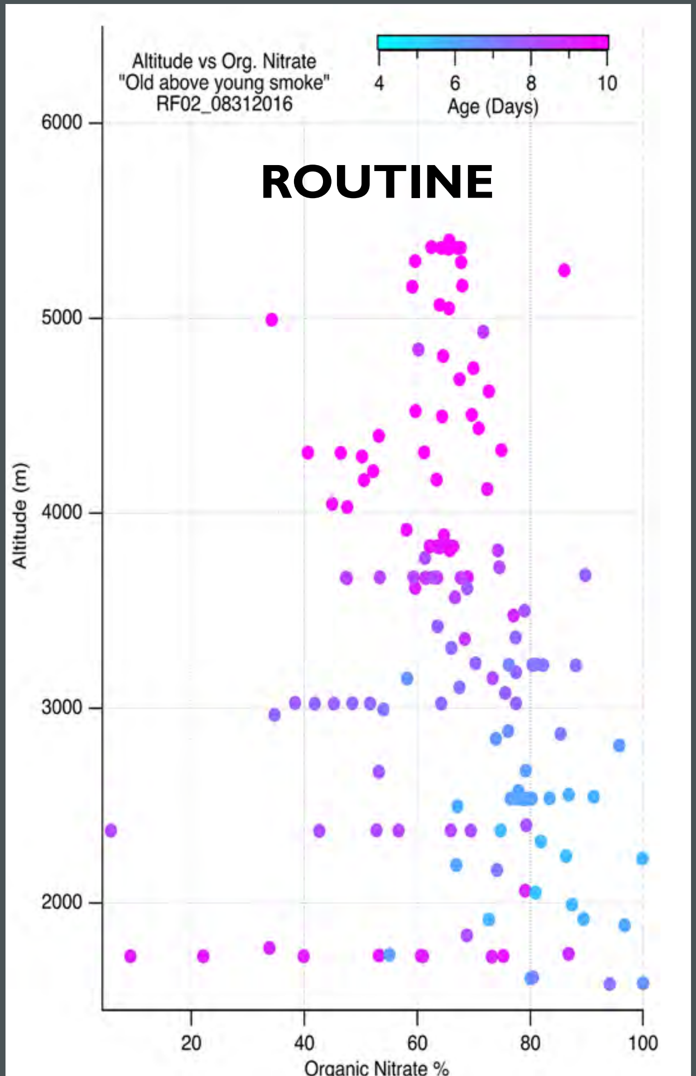
TWO FLIGHT COMPARISON – COASTAL VS ROUTINE

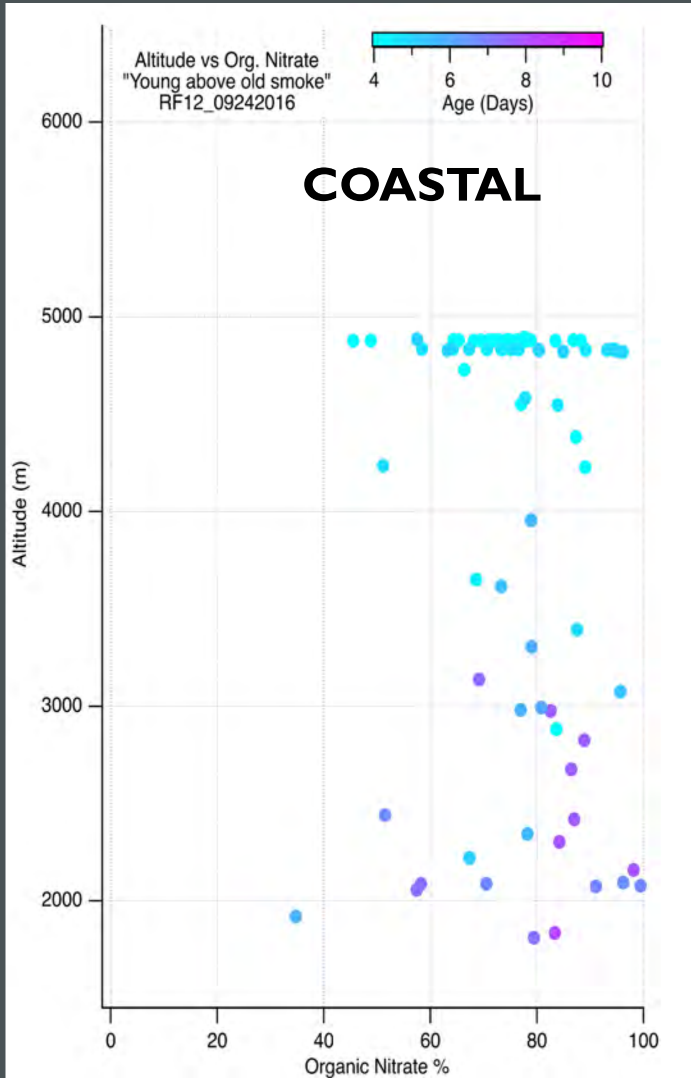


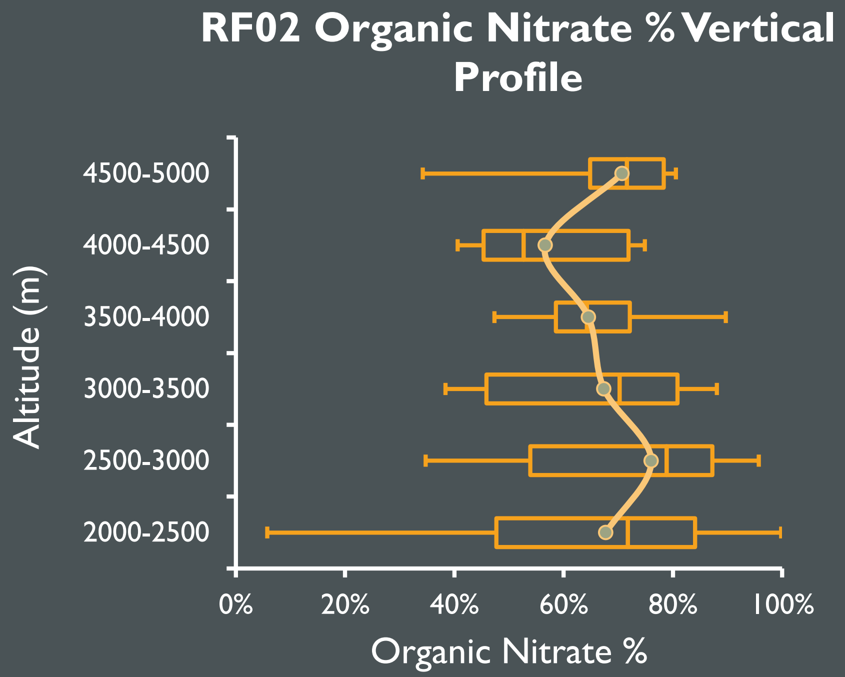


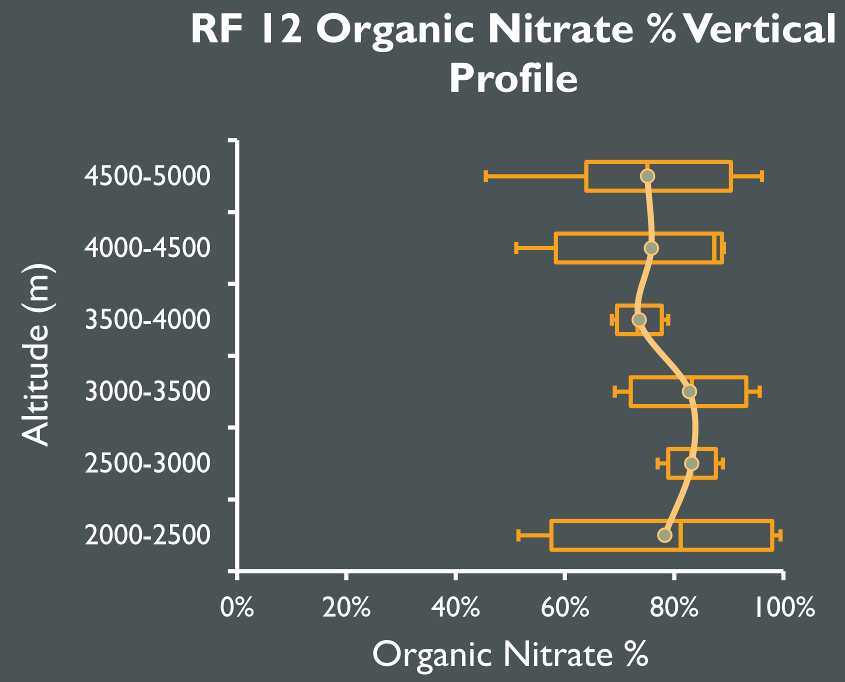
Zonal Wind (m/s)
RF02_08312016
Organic Aerosol
Zonal Wind
ROUTINE
Altitude (Km)
Age (days)
OA µg/m3
Zonal Wind (m/s)
RF12_09242016
Organic Aerosol
Zonal Wind
COASTAL
Altitude (Km)
Age (days)
OA µg/m3
Relative Humidity %
RF02_08312016
NO3:BC
RH
ROUTINE
Altitude (Km)
NO3:BC
Relative Humidity %
RF12_09242016
NO3:BC
RH
COASTAL
NO3:BC

TWO FLIGHT COMPARISON – COASTAL VS ROUTINE



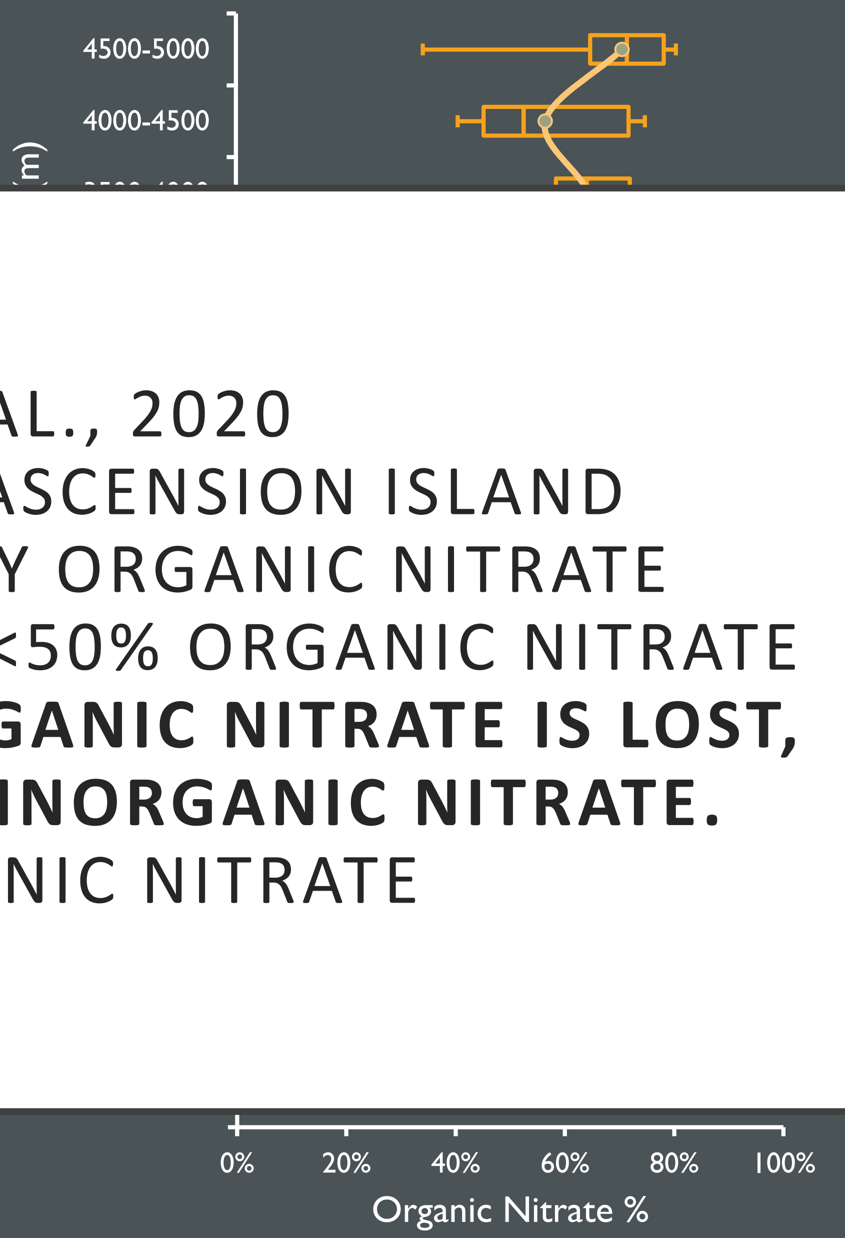




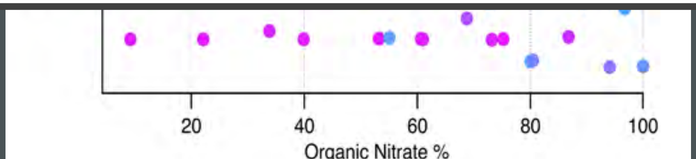


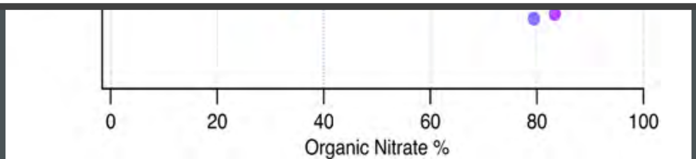
TWO FLIGHT COMPARISON – COASTAL VS ROUTINE

RF02 Organic Nitrate % Vertical Profile



- 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



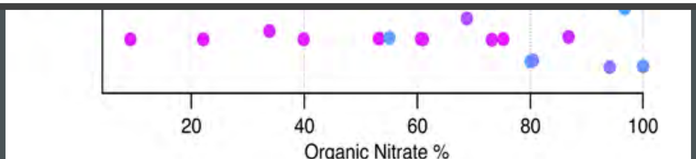


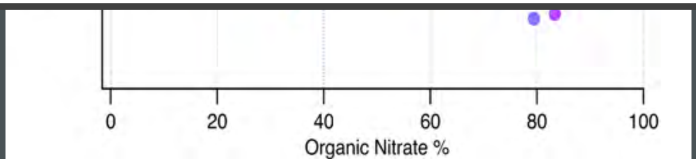
TWO FLIGHT COMPARISON – COASTAL VS ROUTINE

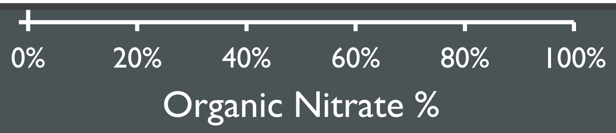
- CAN WE DISTINGUISH HOW MUCH OF ORGANIC AEROSOL LOST IS DRIVEN BY THERMODYNAMICS AND HOW MUCH IS DRIVEN BY PHOTOCHEMISTRY?

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

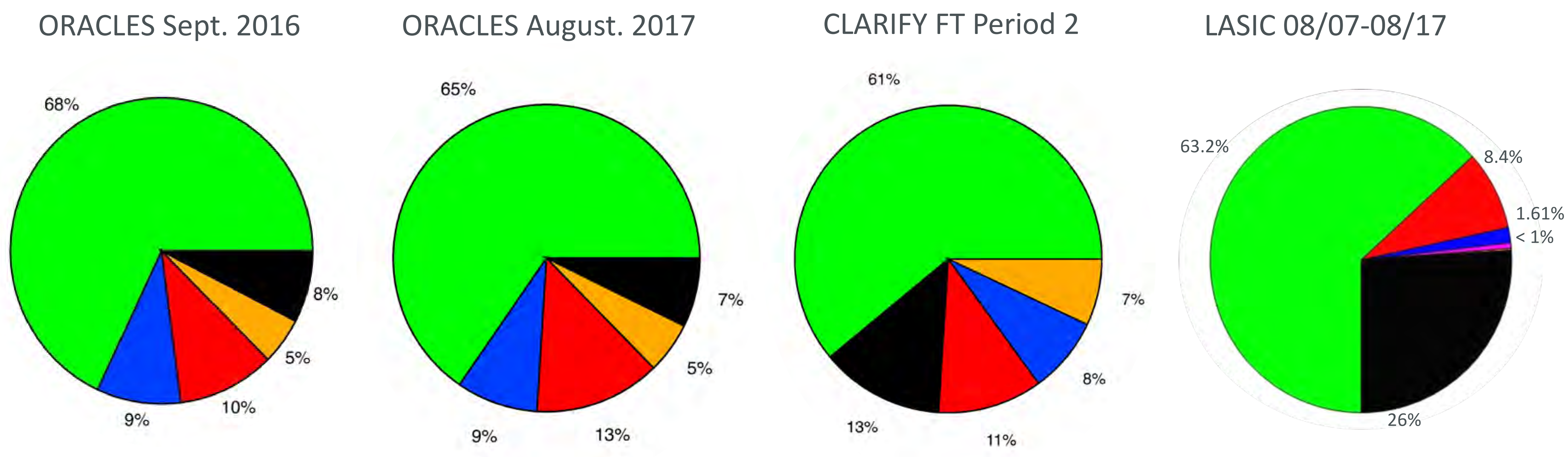
MOVING FORWARD: ASCENSION ISLAND AEROSOL IS PREDICTED TO BE OLDER THAN ORACLES AEROSOL.

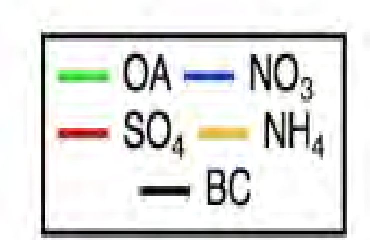






ORACLES – CLARIFY – LASIC BULK CHEMICAL SPECIES

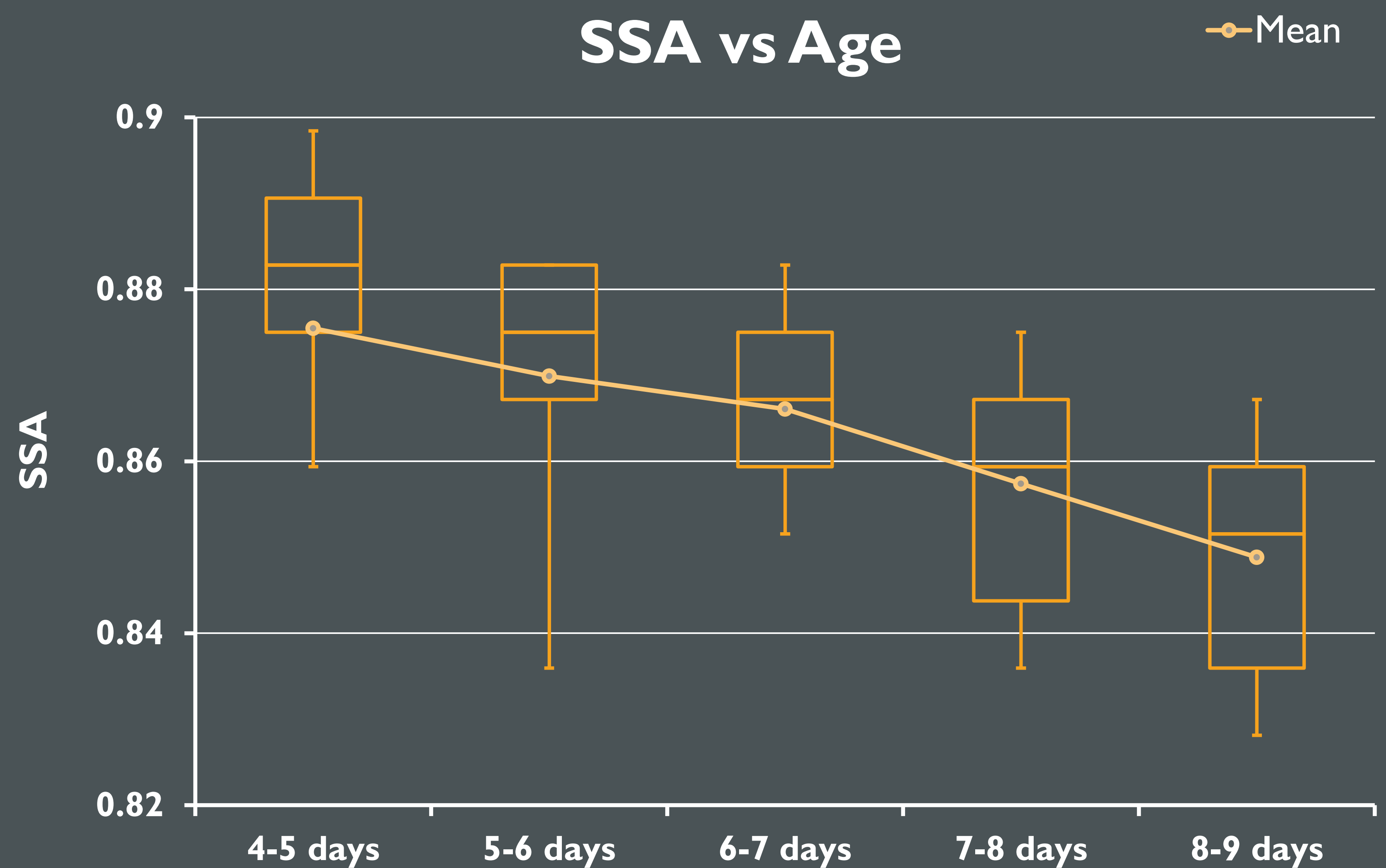




Can chemical differences be resolved between 3 campaigns? Higher % of BC toward Ascension Island?

CLARIFY data courtesy of HuiHui Wu

Does the higher % of BC relative to OA affect optical properties?
What about size?



FUTURE WORK AND COLLABORATION WITH THE DOE AND UK MET OFFICE

Instrument	Data collected	Campaign(s)
Aerosol Mass Spectrometer (AMS)	Bulk Chemical Species (NO_3, OA, OM SO_4, NH_4) (μg/m^3), HR ratios (O/C, H/C)	LASIC (ACSM), ORACLES (HR-AMS), CLARIFY(ACSM)
Single Particle Soot Photometer (SP2)	Black Carbon (BC) (μg/m^3)	LASIC, ORACLES, CLARIFY
Los Gatos Research CO/CO2 Analyzer	CO, CO_2, O_3 (ppbv)	LASIC (CO only), ORACLES, CLARIFY
TSI Nephelometer	Scattering (450,530,700nm) Mm^{-1}	LASIC, ORACLES, CLARIFY
Particle Soot Absorption Photometer (PSAP)	Absorption (450nm, 530nm, 660nm) Mm^{-1}	LASIC, ORACLES, CLARIFY
Ultra-High Sensitivity Aerosol Spectrometer (UHSAS)	Aerosol size between 10nm and 1000nm	LASIC, ORACLES, CLARIFY

- 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).