Non-reversible aging can increase solar absorption in African biomass burning aerosol plumes of intermediate age

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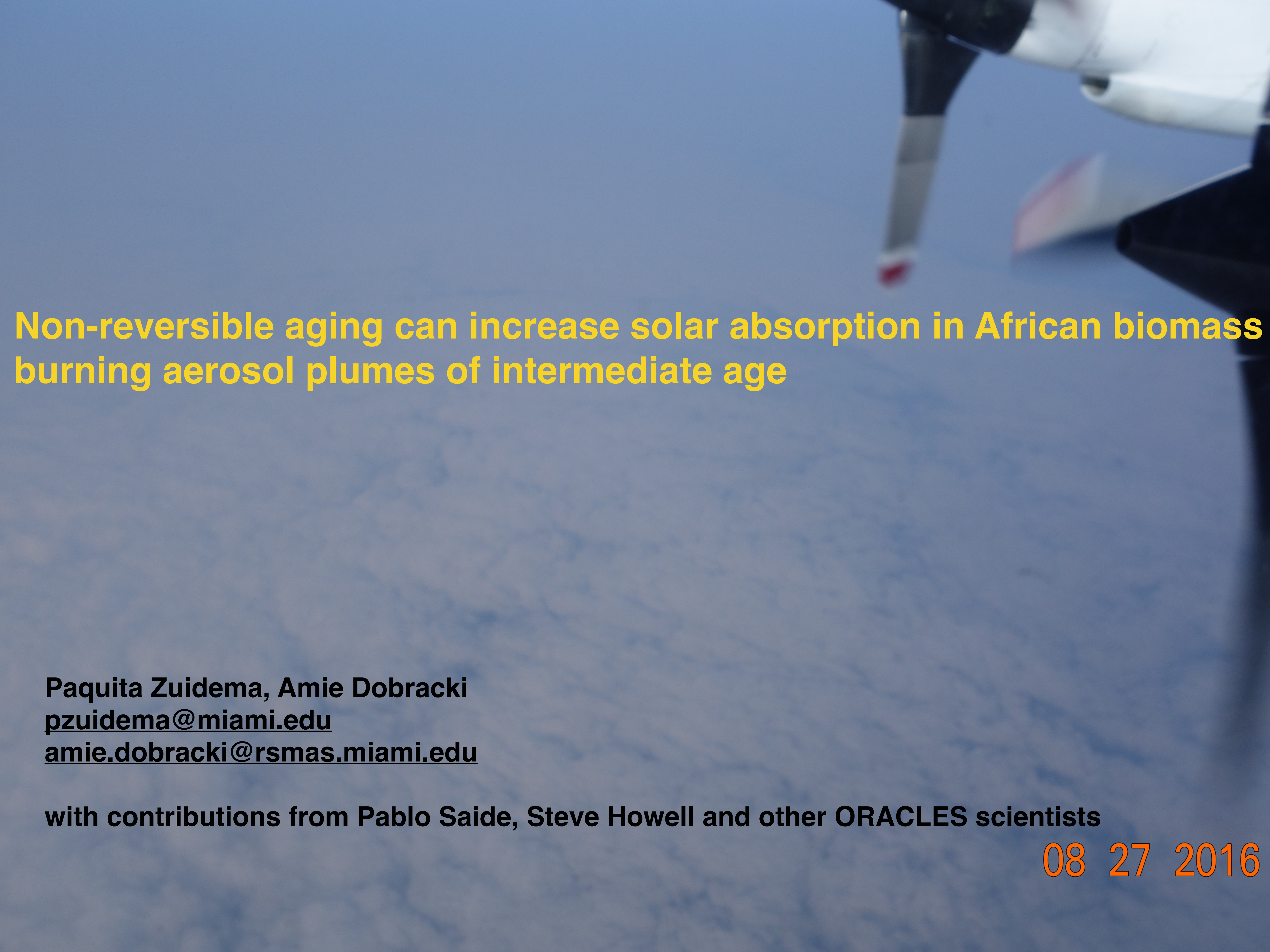
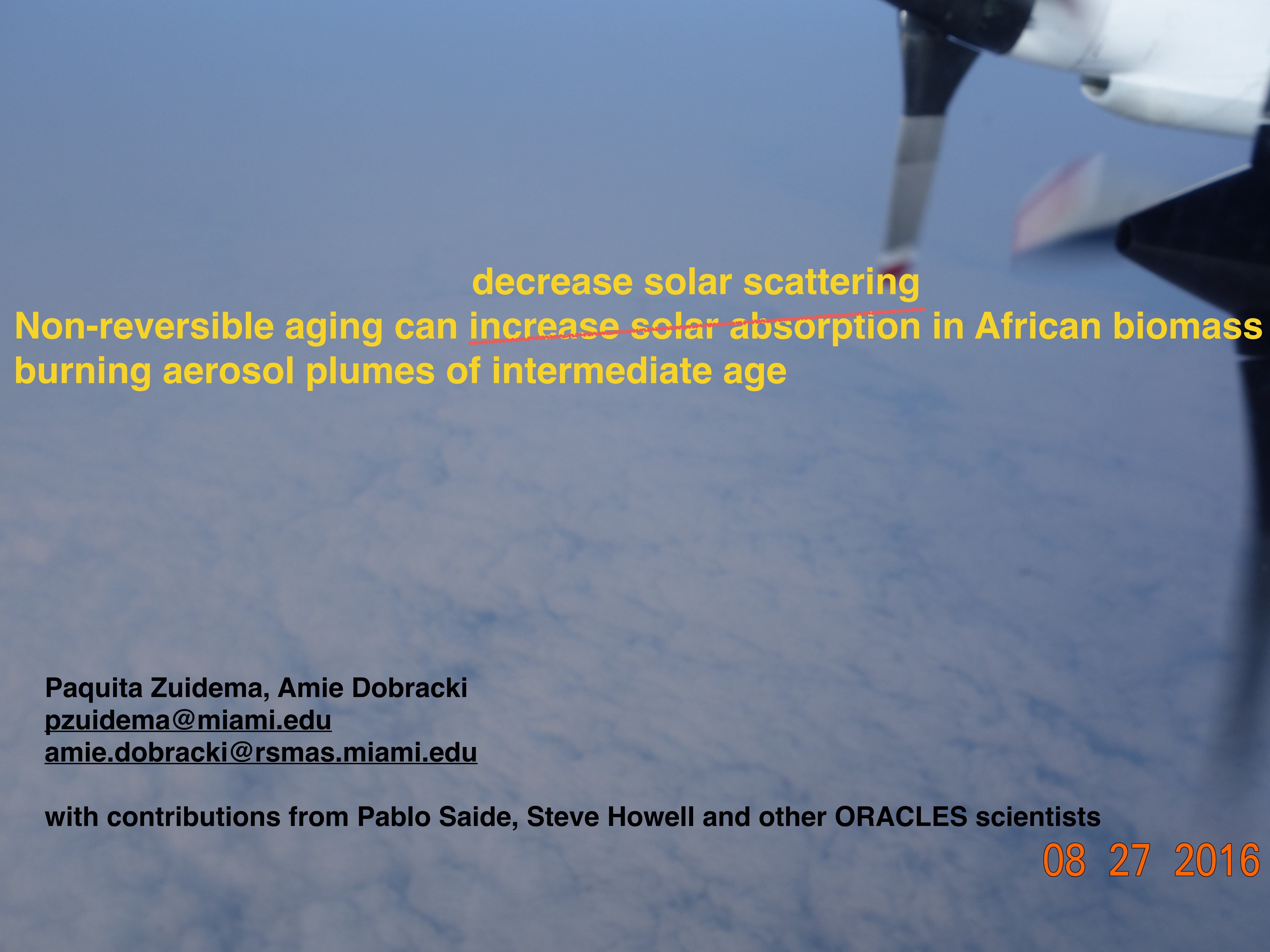
with contributions from Pablo Saide, Steve Howell and other ORACLES scientists





decrease solar scattering
Non-reversible aging can ~~increase solar absorption~~ in African biomass burning aerosol plumes of intermediate age

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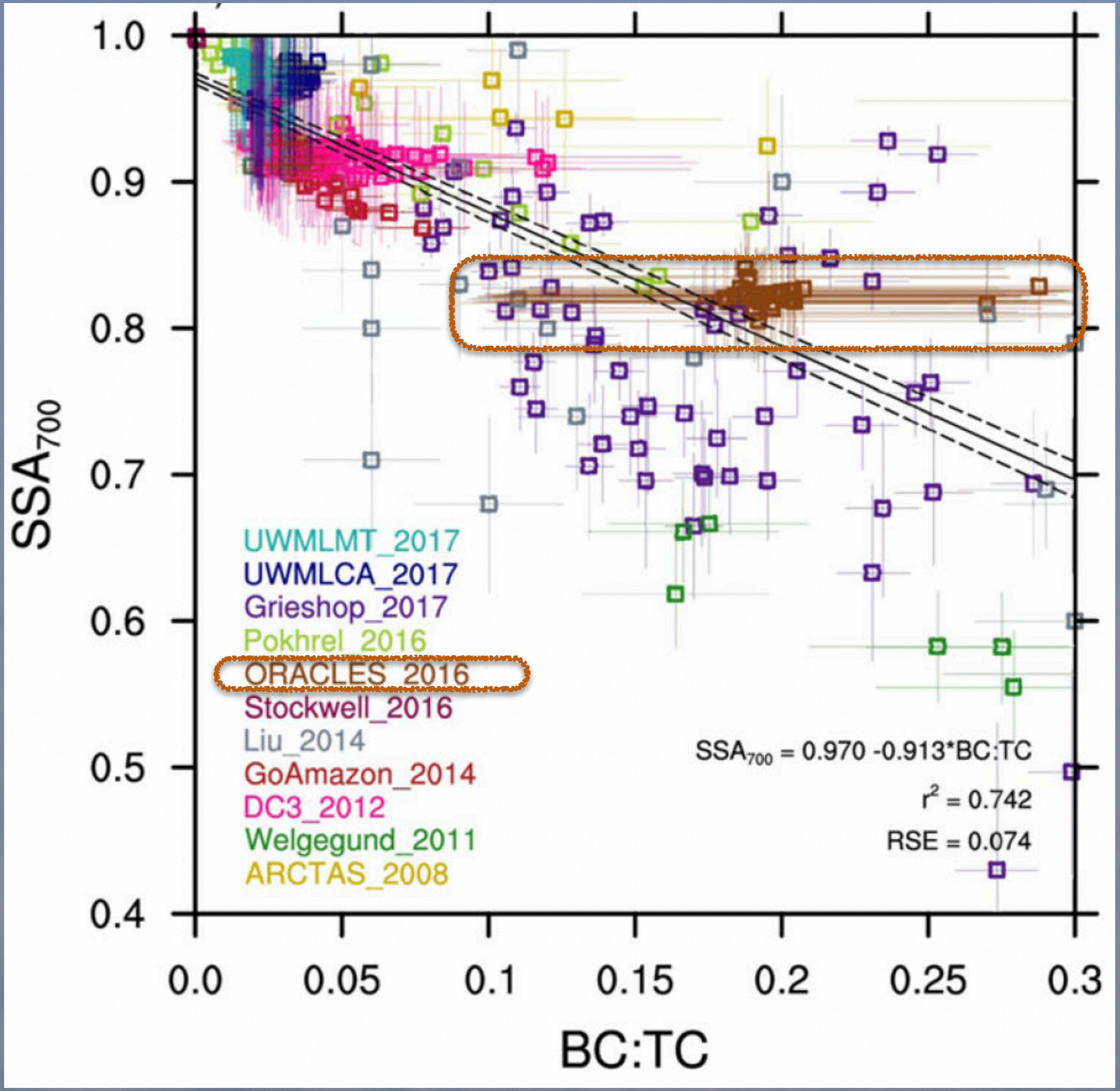
Our fundamental question: why does biomass-burning aerosol over the southeast Atlantic possess such a low single-scattering albedo compared to other regions?



Brown et al., 2021

And: do aging processes contribute?

Pro: ***we have data (aerosol composition, black carbon, optical properties)***

Con: ***we never sampled close to a fire (no Lagrangian tracking; can only indirectly infer fire source characteristics)***

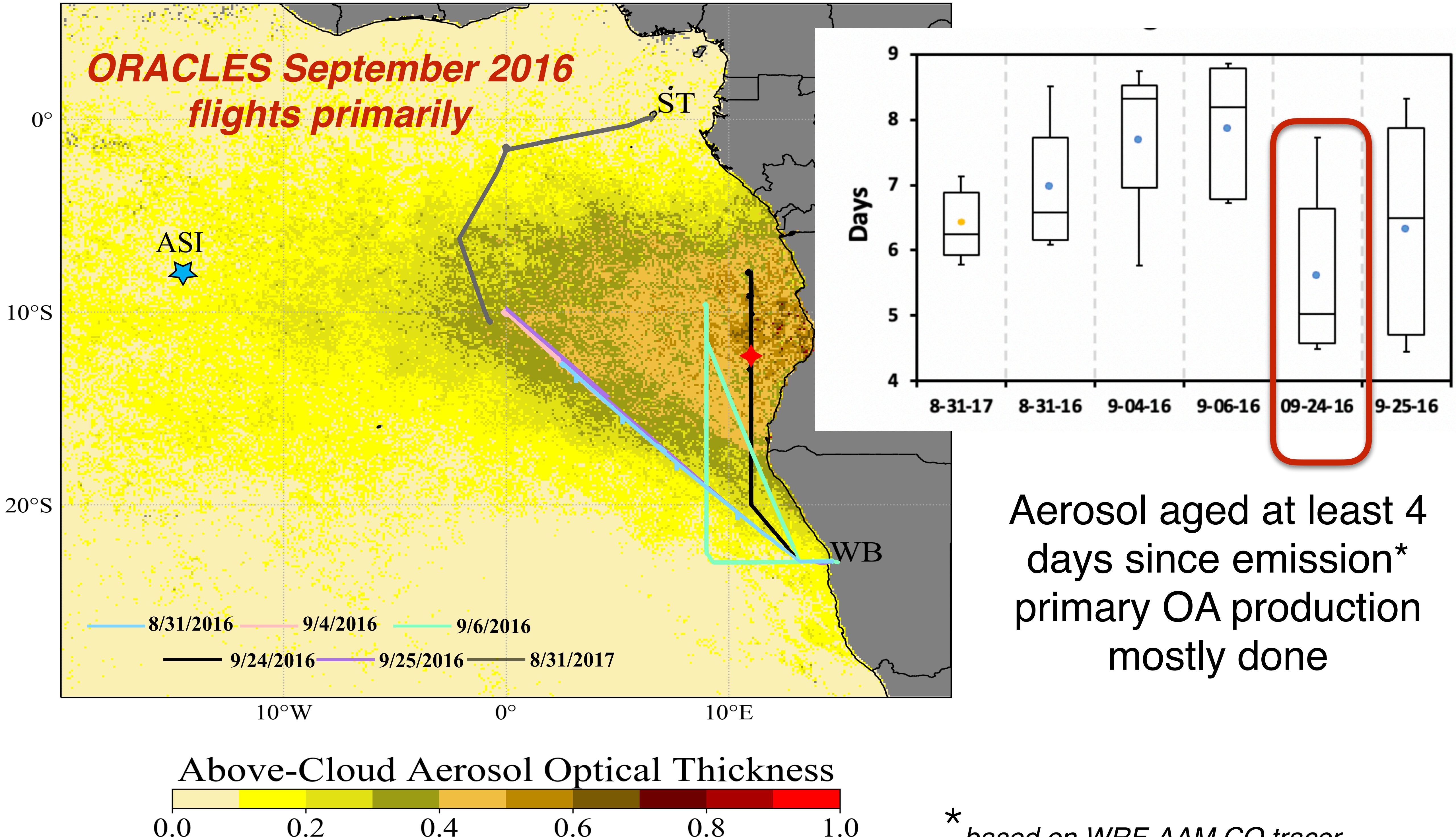


Figure 2. Terra MODIS Above Cloud Aerosol Optical Depth (Meyer, 2015) for September 2016 overlaid with the tracks of the 6 flights selected for this study. Locations of the profiles shown in Figs. 10- 11 are indicated with red diamonds. ST=Sao Tome; WB=Walvis Bay; ASI=Ascension Island.

Aerosol aged at least 4 days since emission* primary OA production mostly done

* *based on WRF-AAM CO tracer*

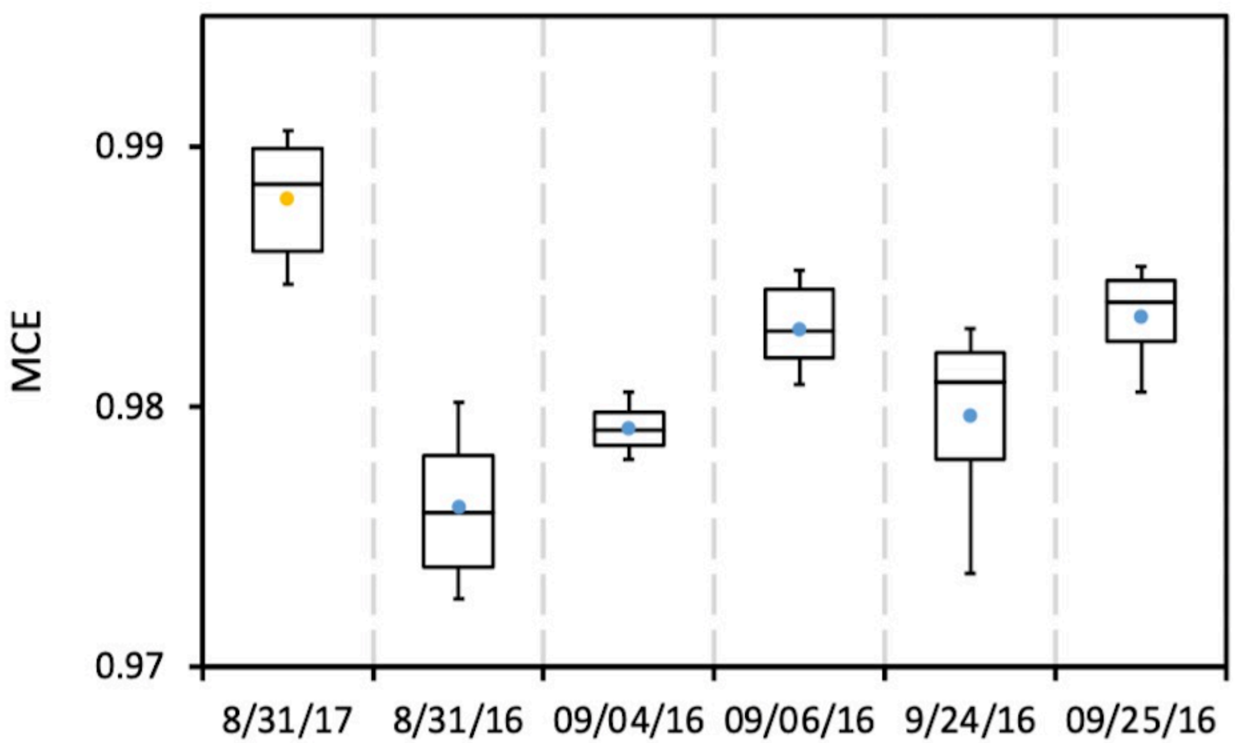
Redemann et al., 2021 doi:10.5194/acp-21/1507-2021
Dobracki et al., in prep

Modified combustion efficiency (MCE) values>0.975 indicate flame-efficient, dry, grass-dominated conditions.

Korontzi et al. 2003; Southern African Fire-AtmospheRre research Initiative SAFARI-92; JGR 1996 special issue

=> relatively high black carbon, low carbon monoxide efficiencies
=> lessened production of organic aerosols

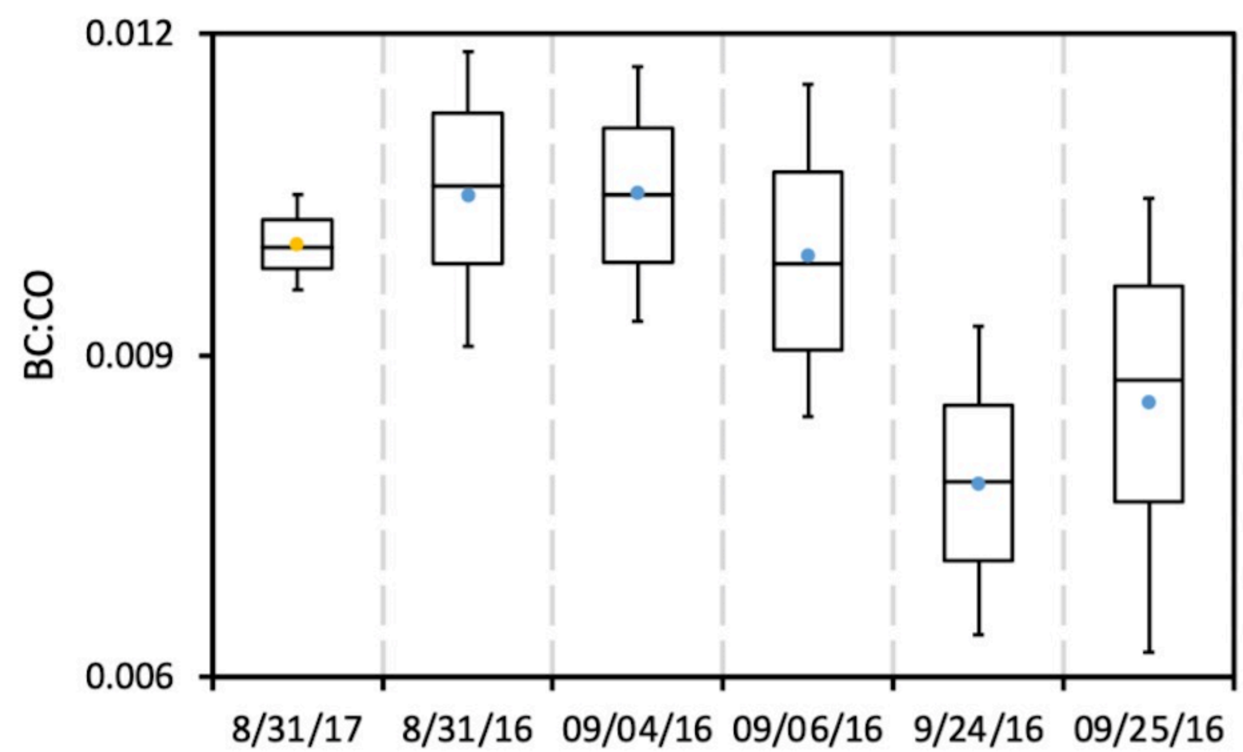
Modified combustion efficiency



$$MCE = \frac{\Delta CO_2}{\Delta CO + \Delta CO_2} = \frac{1}{1 + \Delta CO / \Delta CO_2}$$

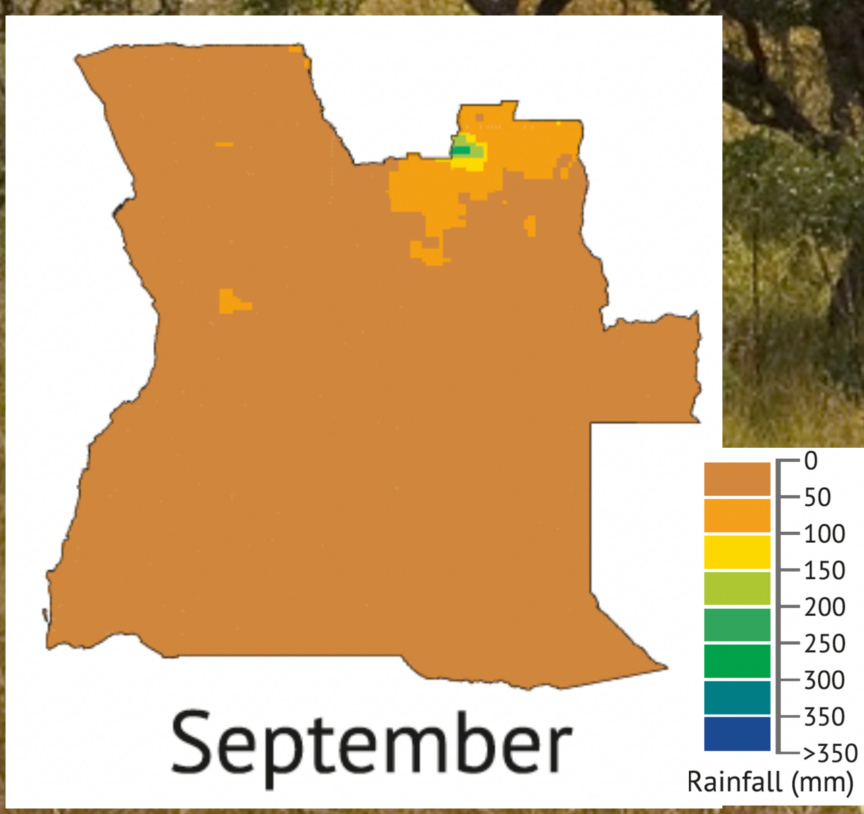
Yokelson et al., 2009

BC:CO (unitless)



7-11 *0.001 range at high end, more consistent with grass
(Vakkari et al., 2018)

Miombo woodlands the primary vegetation type: the grass burns first



Mendelsohn, 2013

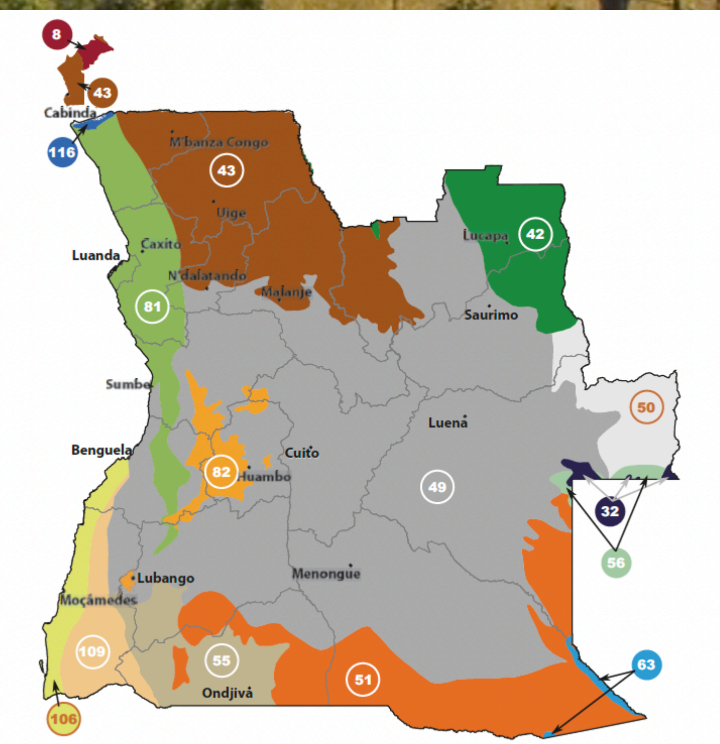
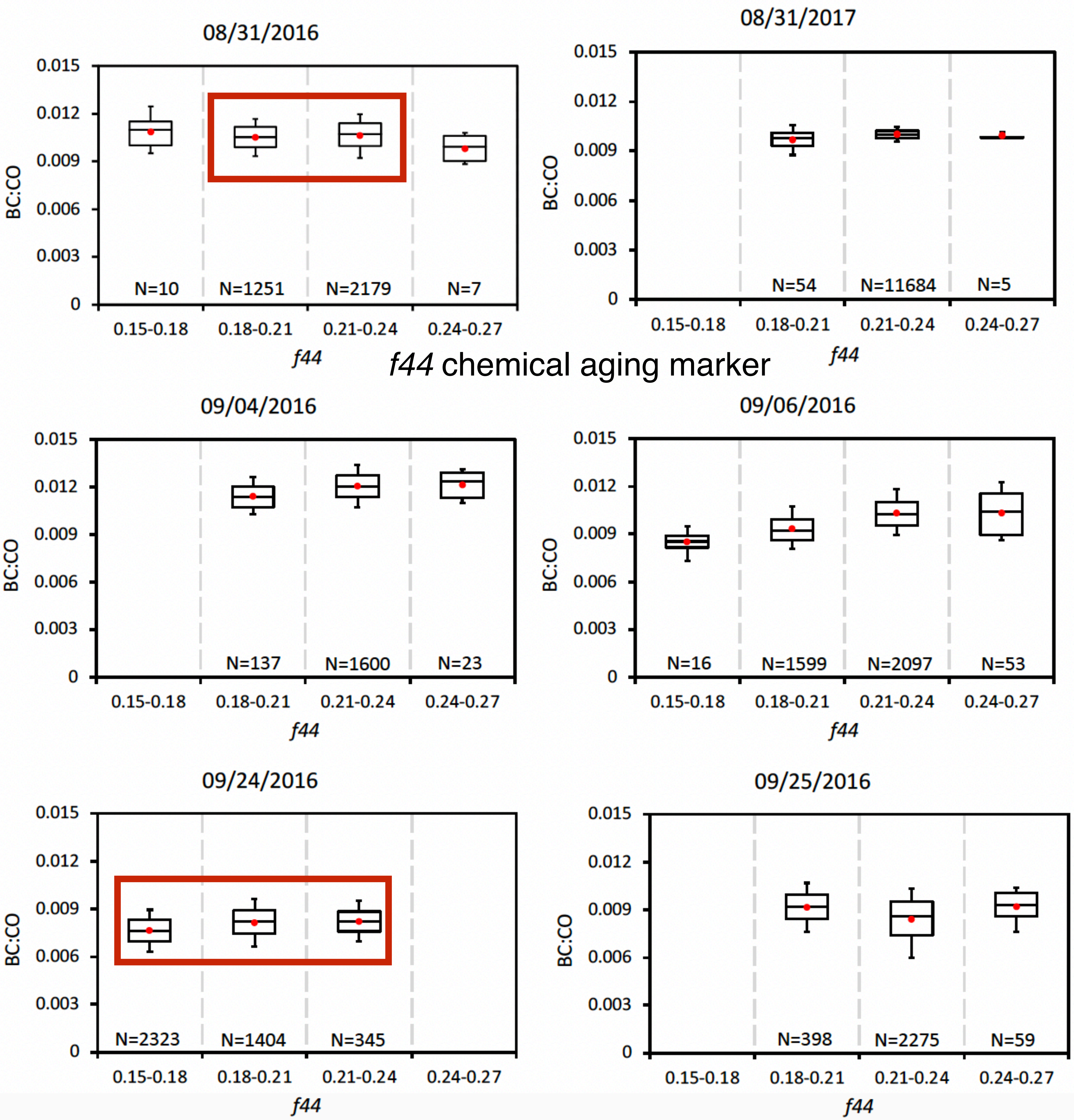
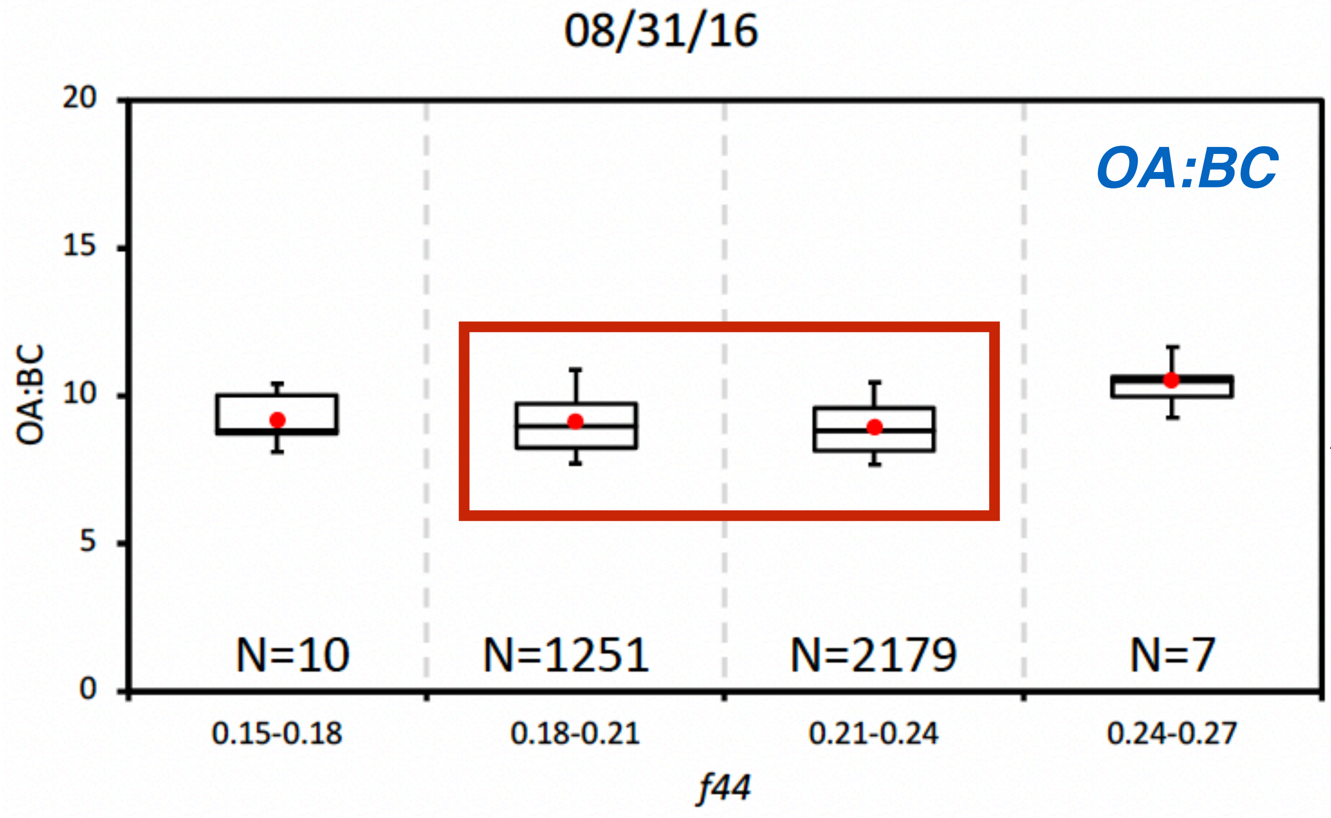


Fig. 2.8 Ecoregions of Angola
8 Atlantic Equatorial Coastal Forest • 32 Zambezian *Cryptosepalum* Dry Forest • 42 Southern Congolian Forest-Savanna Mosaic • 43 Western Congolian Forest-Savanna Mosaic • 49 Angolan Miombo Woodland • 50 Central Zambezian Miombo Woodland • 51 Zambezian *Baikiaea* Woodland • 55 Angola Mopane Woodland • 56 Western Zambezian Grassland • 63 Zambezian Flooded Grasslands • 81 Angolan Scarp Savanna and Woodland • 82 Angolan Montane Forest-Grassland Mosaic • 106 Kaokoveld Desert • 109 Namib Escarpment Woodlands • 116 Central African Mangroves. (After Burgess et al. 2004, map used with permission)

We use BC:CO to select for aerosol of similar composition at the fire source



- OA>20 ug m^{-3}
- N= # of seconds; > 5 minutes

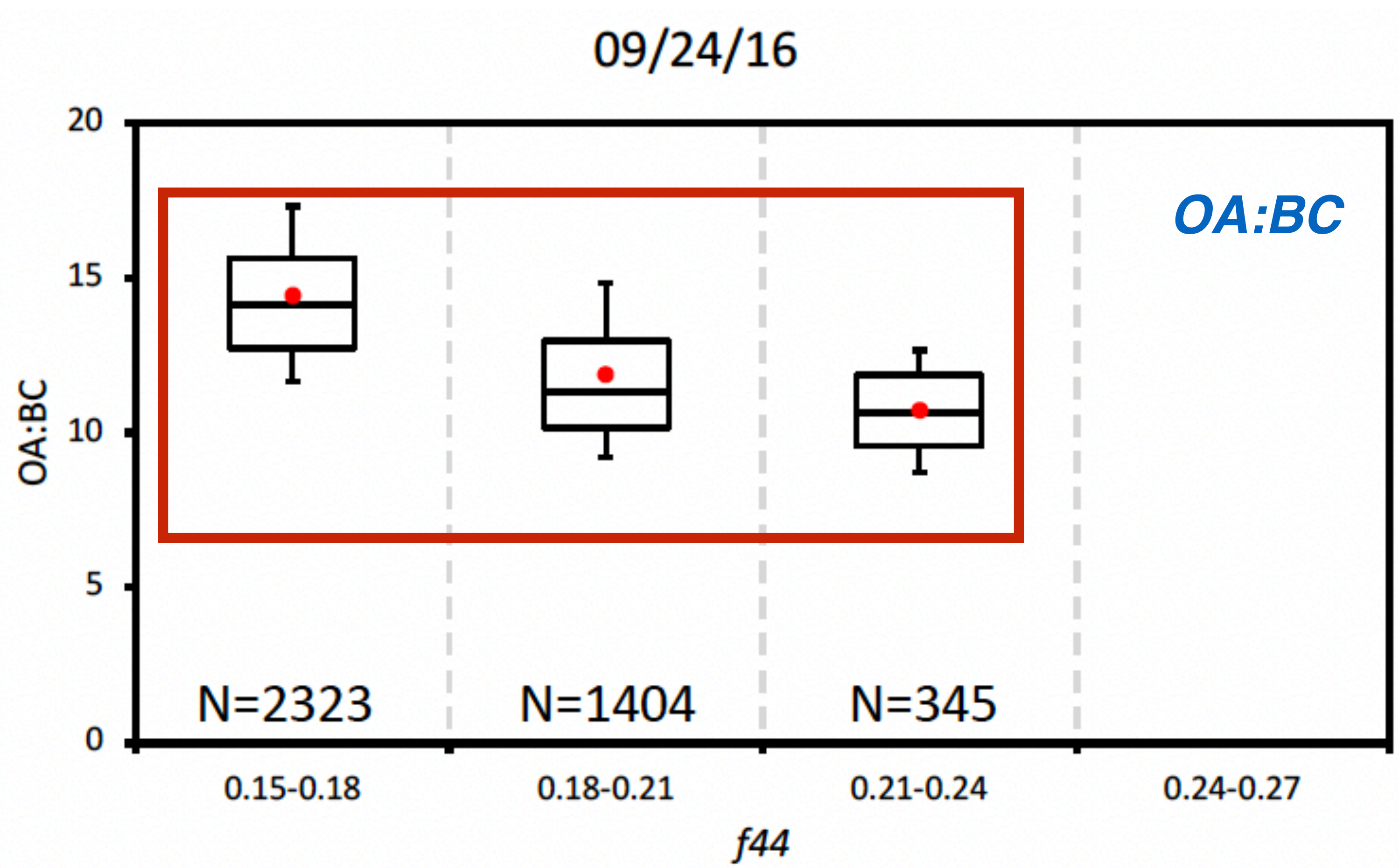


8/31/2016:

*no real change in OA:BC with f44**

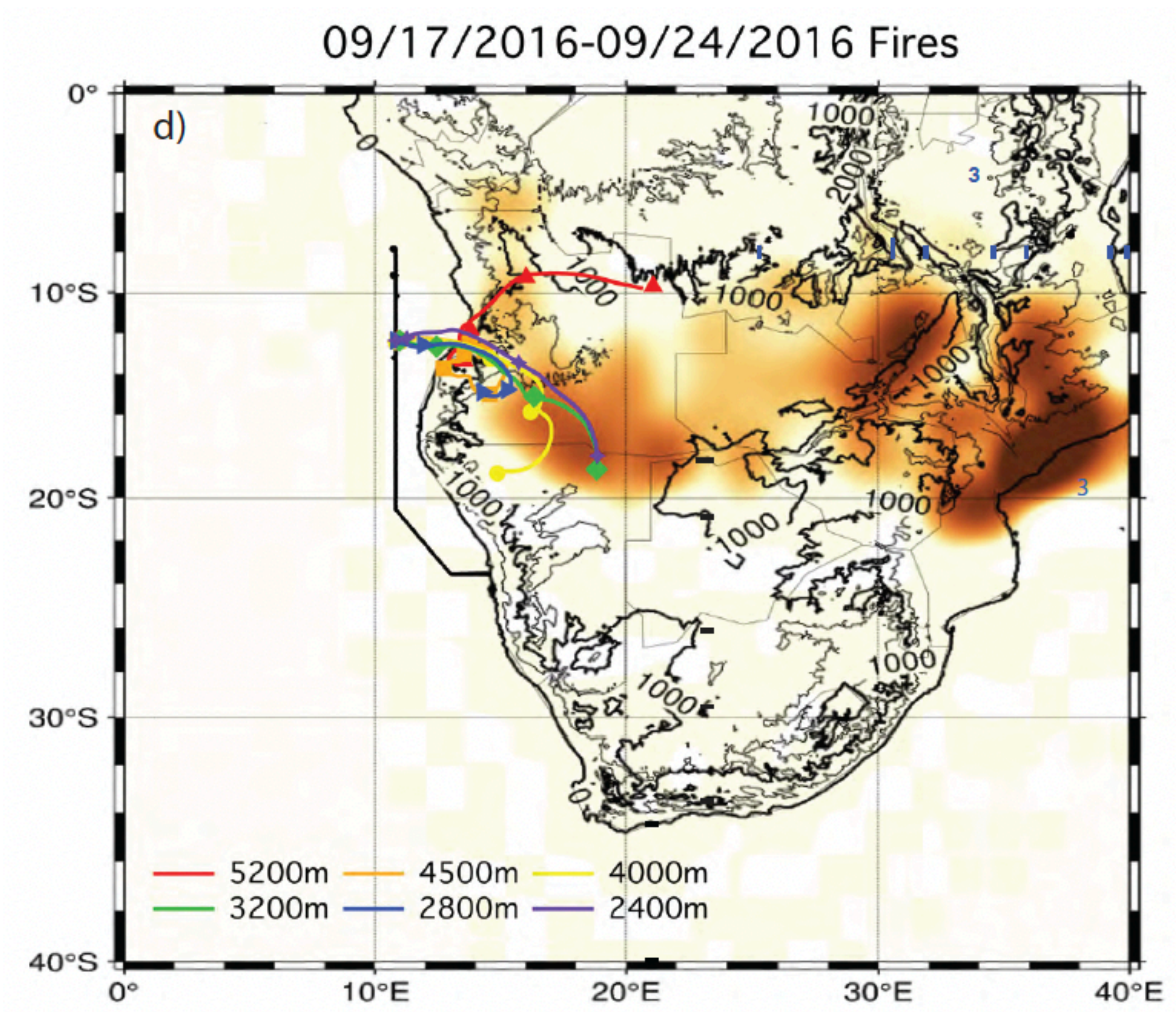
*misreported in https://acp.copernicus.org/preprints/acp-2021-1081/

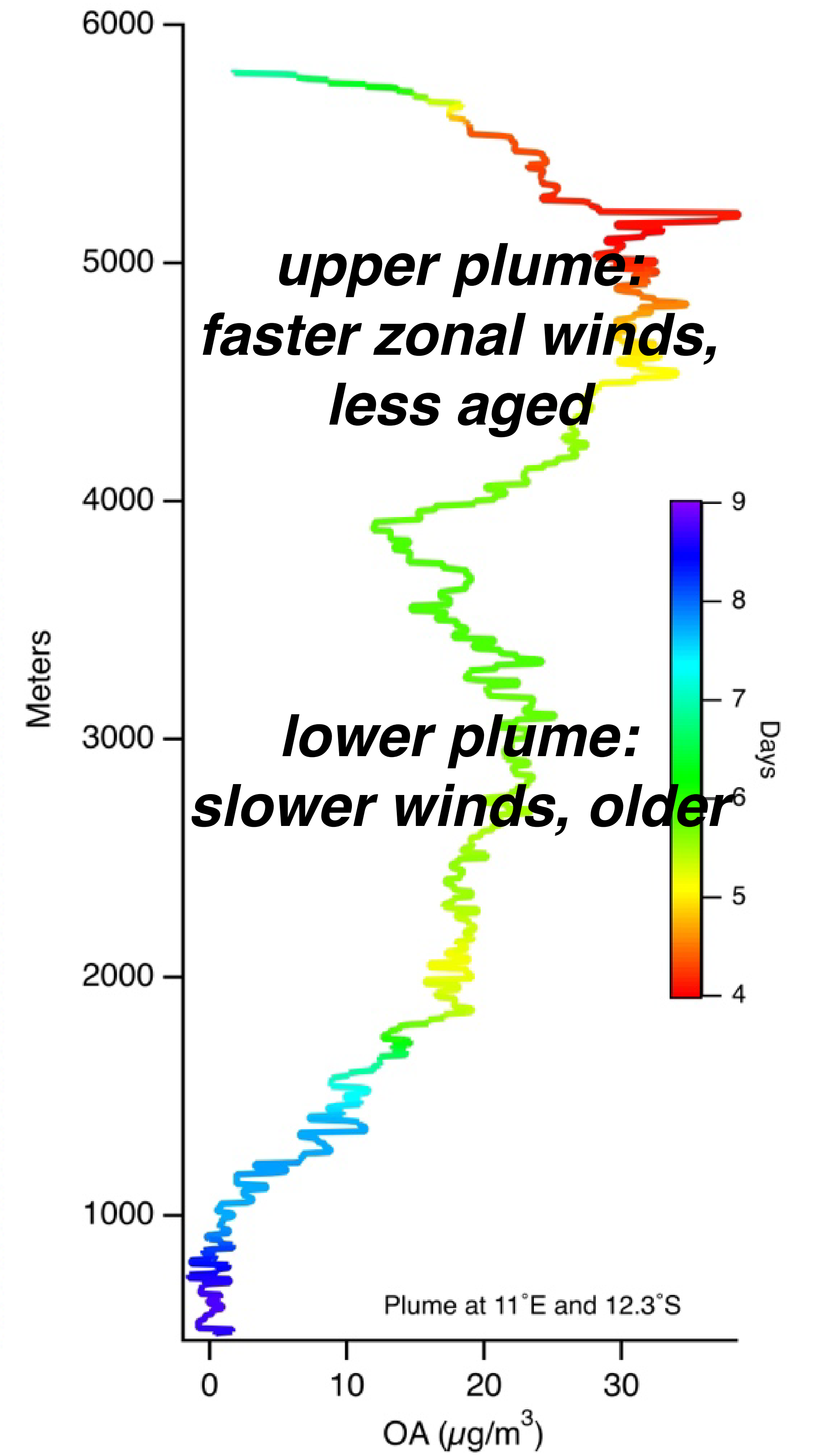
9/24/2016: only flight indicating loss of OA as chemical aging continues, reducing SSA - 0.89 to 0.865



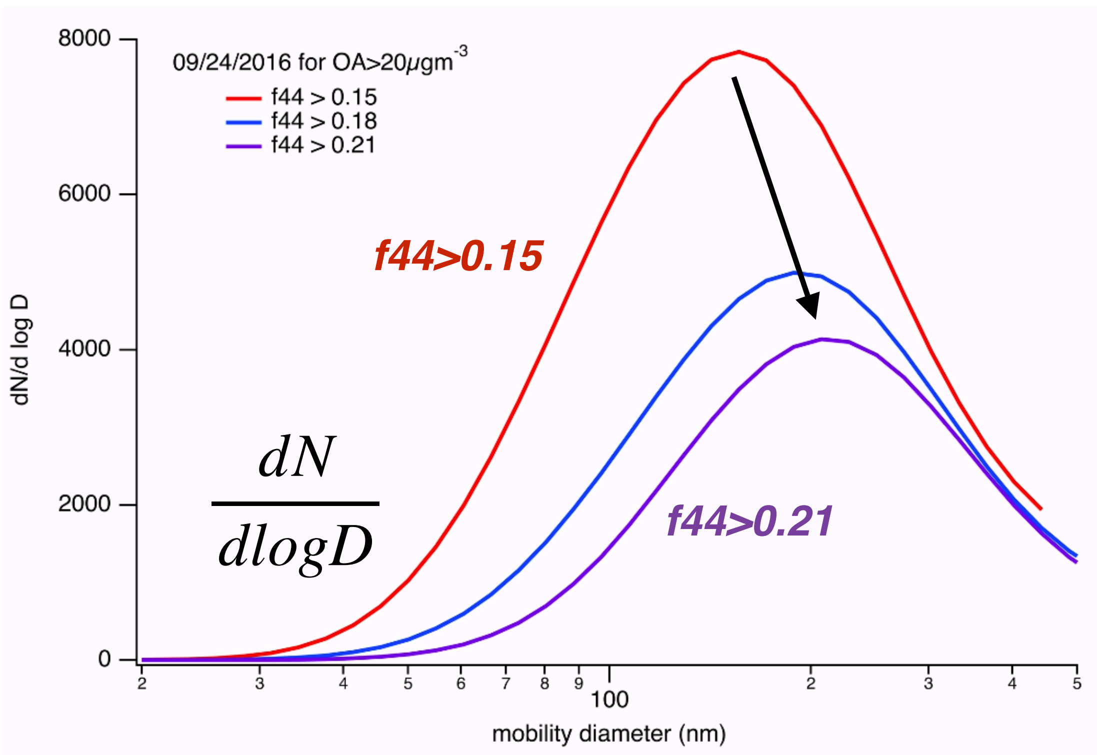
Two distinct smoke plumes: both directly advected from miombo woodland-grassland source regions.

upper smoke plume: faster zonal winds, less aged (by ~1 day)

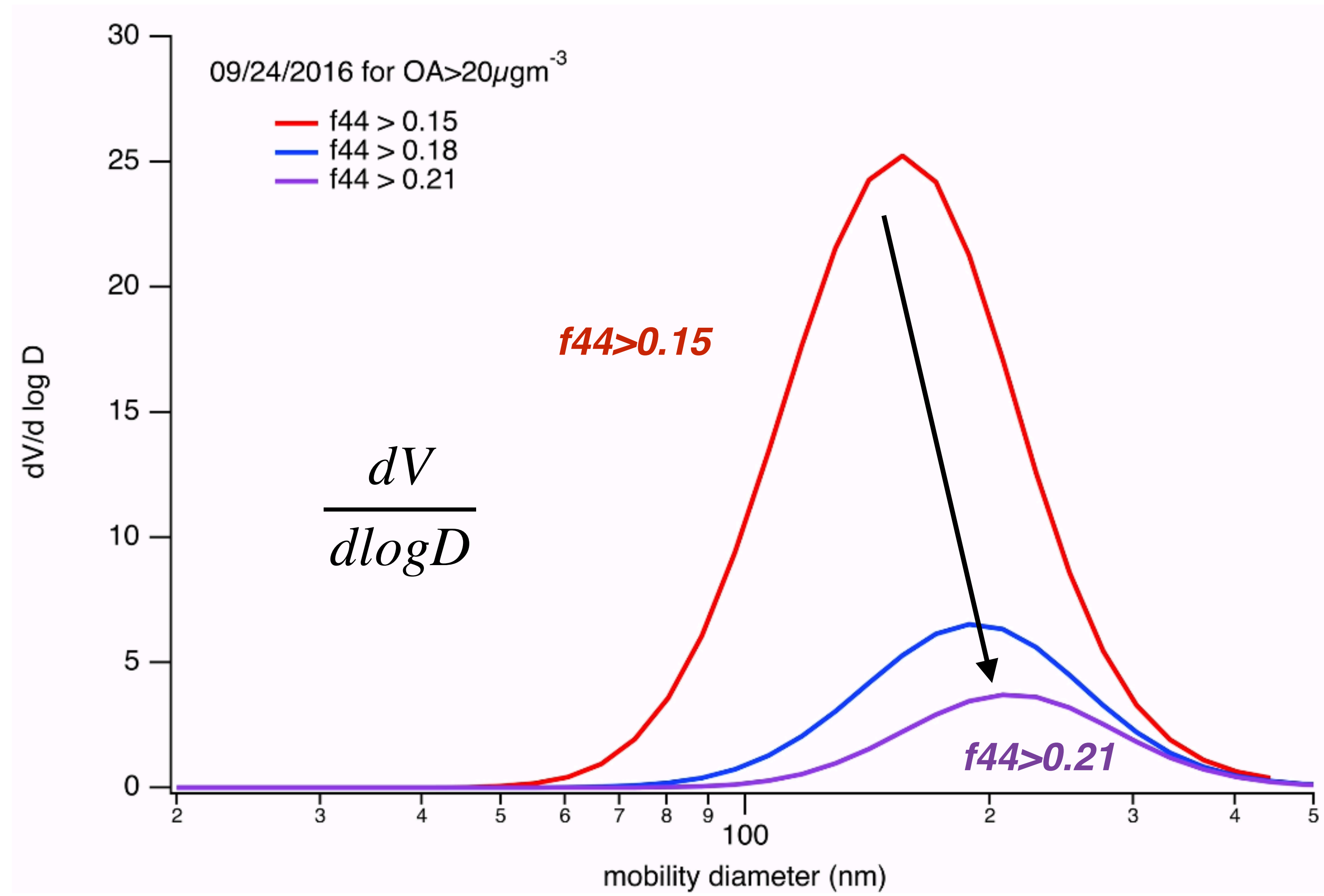




Continued coagulation. Genuine volume/OA loss* - with chemical aging marker f_{44} consistent w SSA reduction from 0.89 to 0.865

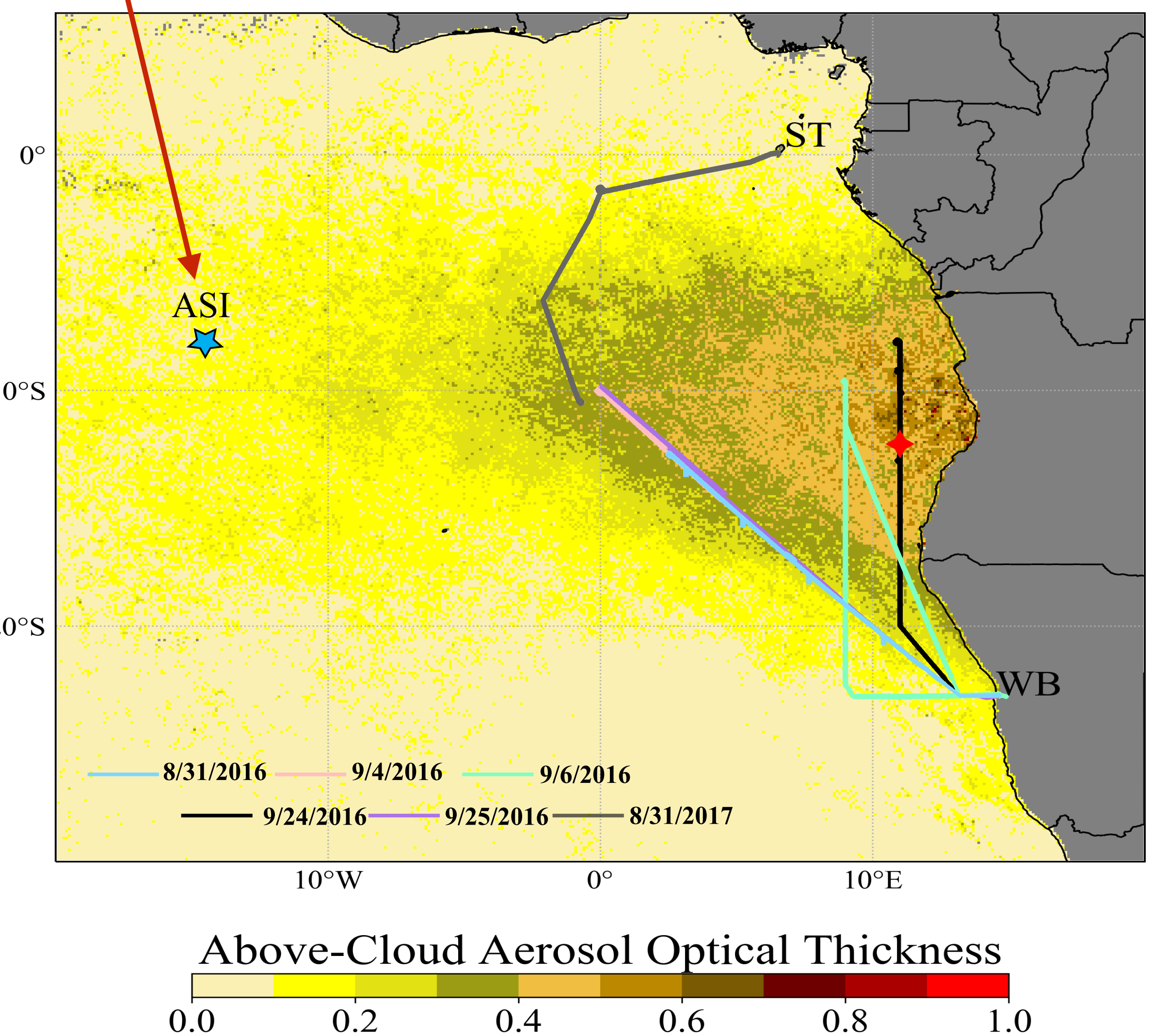


Long Differential Mobility Analyzer; 10-550 nm



*overall little thermodynamic repartitioning with height, in contrast to CLARIFY

CLARIFY* measurements at Ascension report proportionately more BC; yet larger particles, higher MACs & slightly more volatile aerosol



	CLARIFY	this study
BC mass frac.	13%	7%
BC # frac.	39 ±7%	15-40%
SSA_{530}	∼0.84	0.85-0.88
MAC_{660}	11-12 $m^2\ g^{-1}$	9.5-11.5 $m^2\ g^{-1}$
OA:BC mass	4-5	10 ± 2
median diameter	232 nm	140-200 nm
IN* fraction	100%	∼ 25%

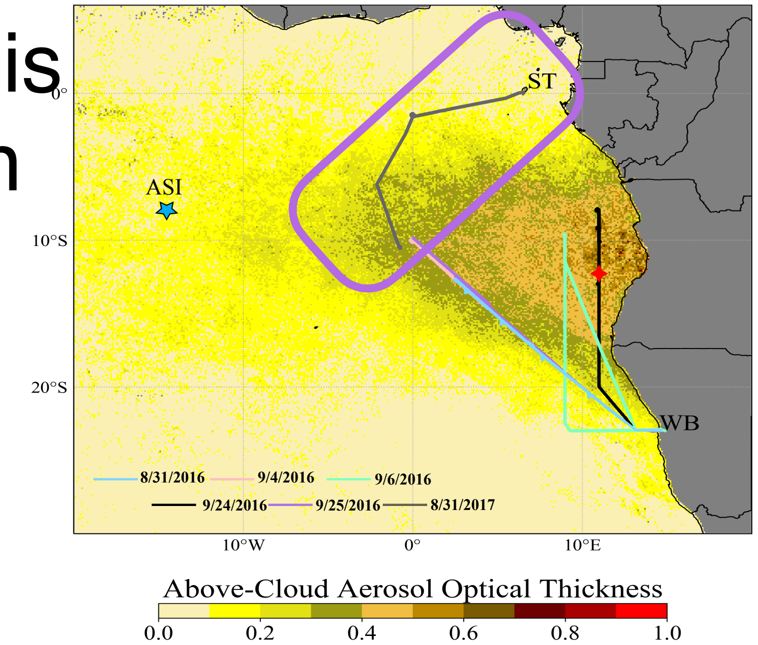
*inorganic nitrate

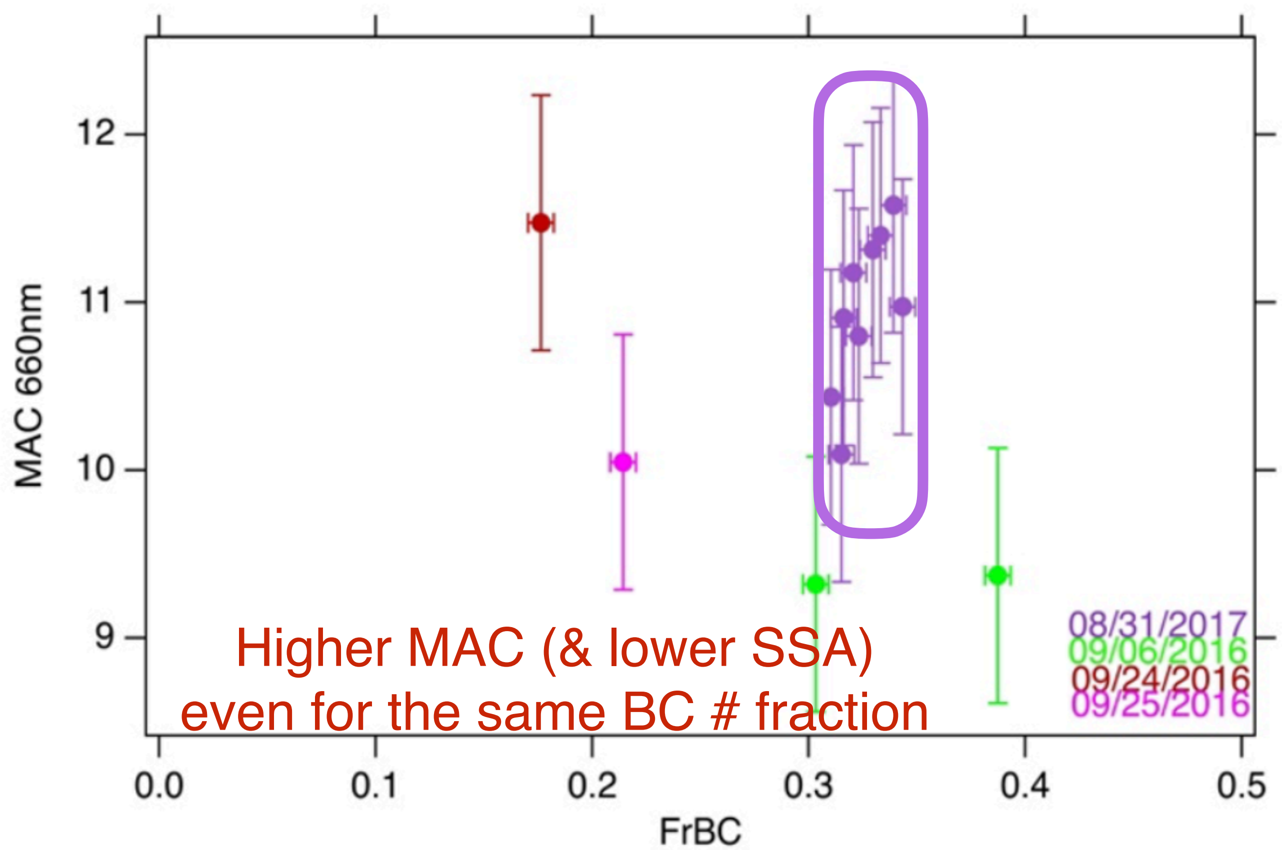
CLARIFY values taken from Wu et al., 2020; Taylor et al., 2020
Greater volatility at ASI also indicated in Dang et al., 2022

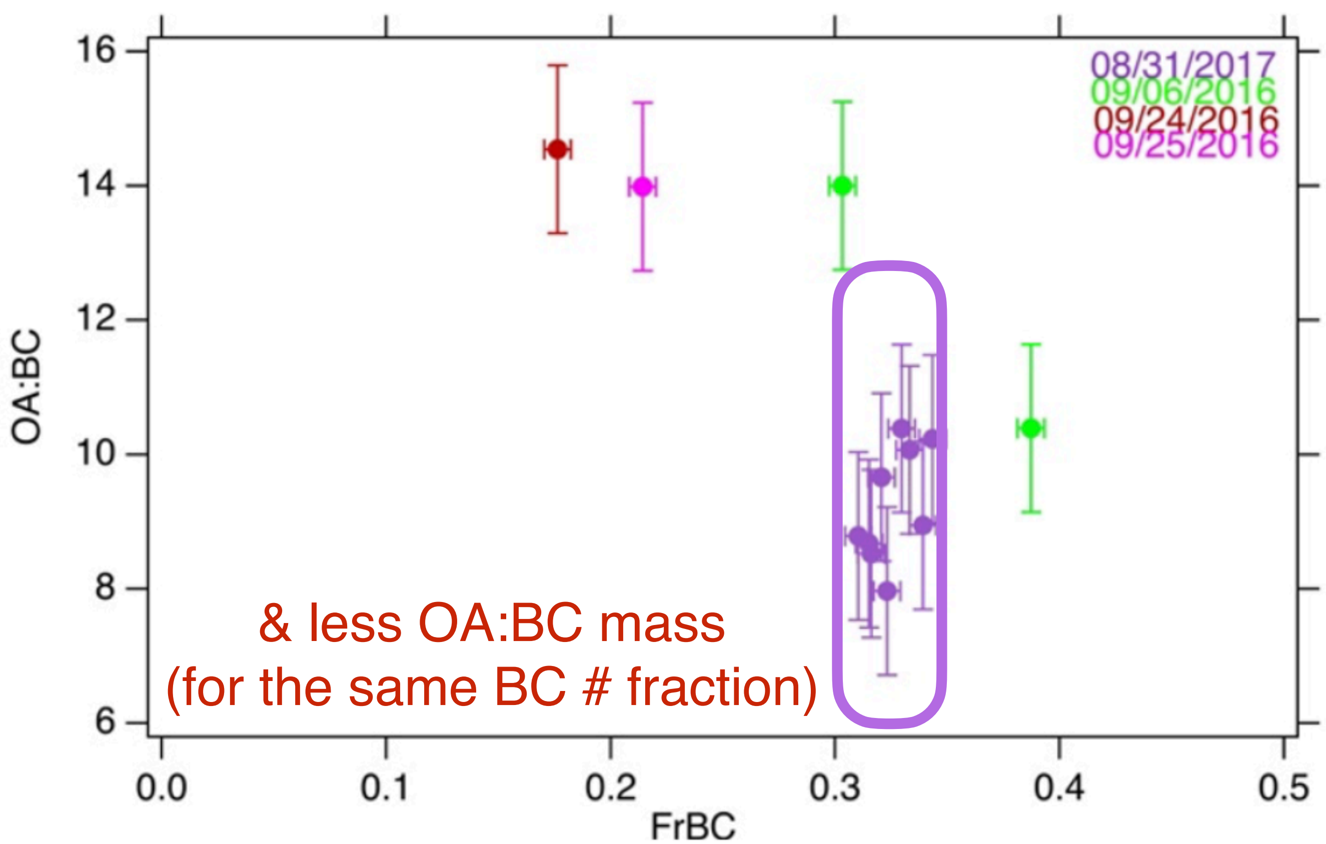
Consistent w thicker coatings generating more lens absorption during CLARIFY

*August 17-September 7, 2017

One flight overlapping w CLARIFY is consistent w this comparison - even after controlling for BC # fraction

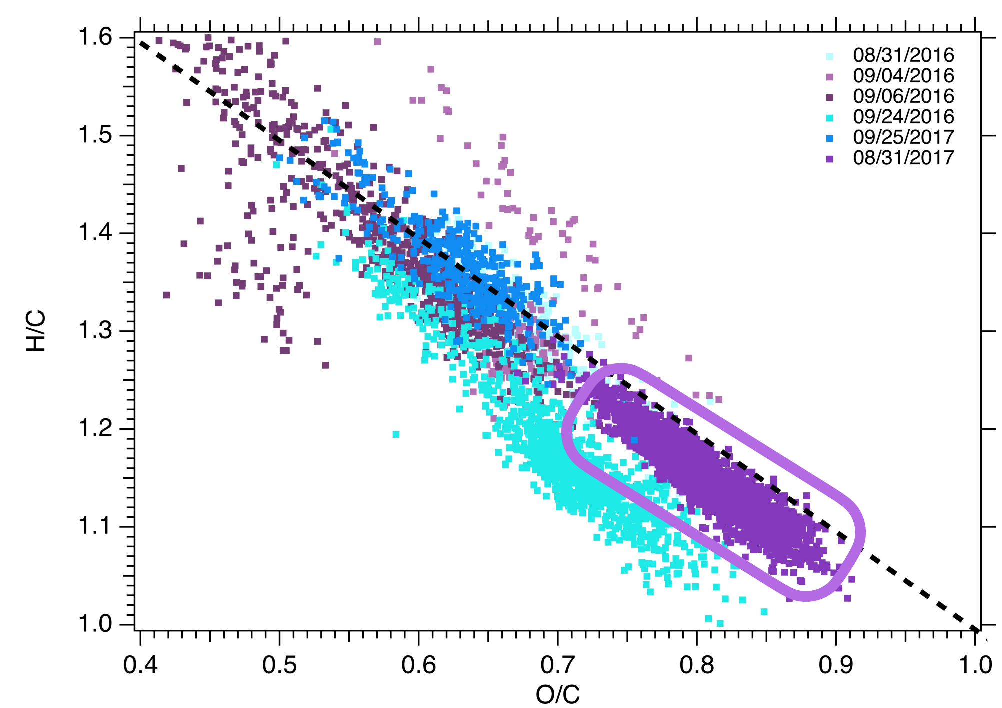






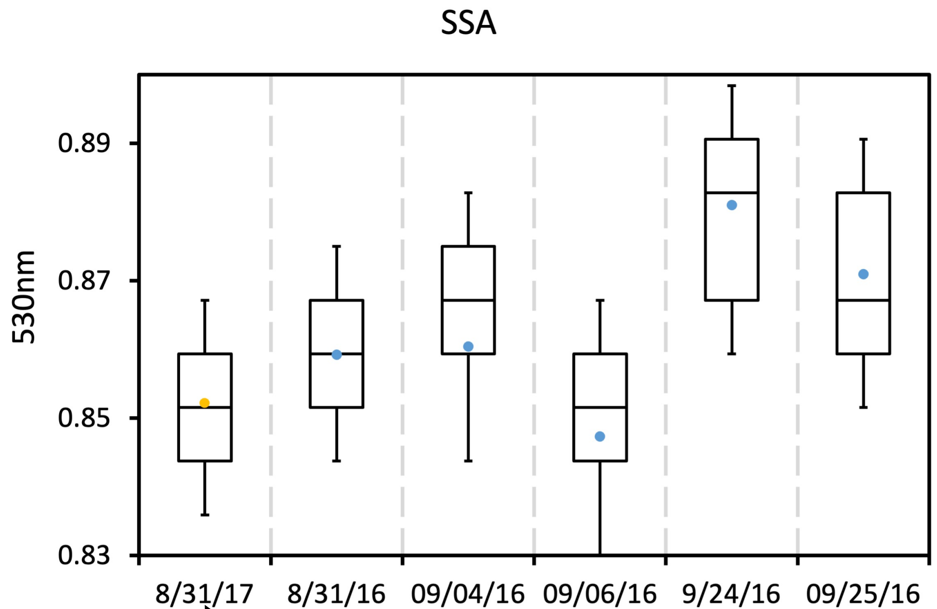
Source differences or chemical aging?

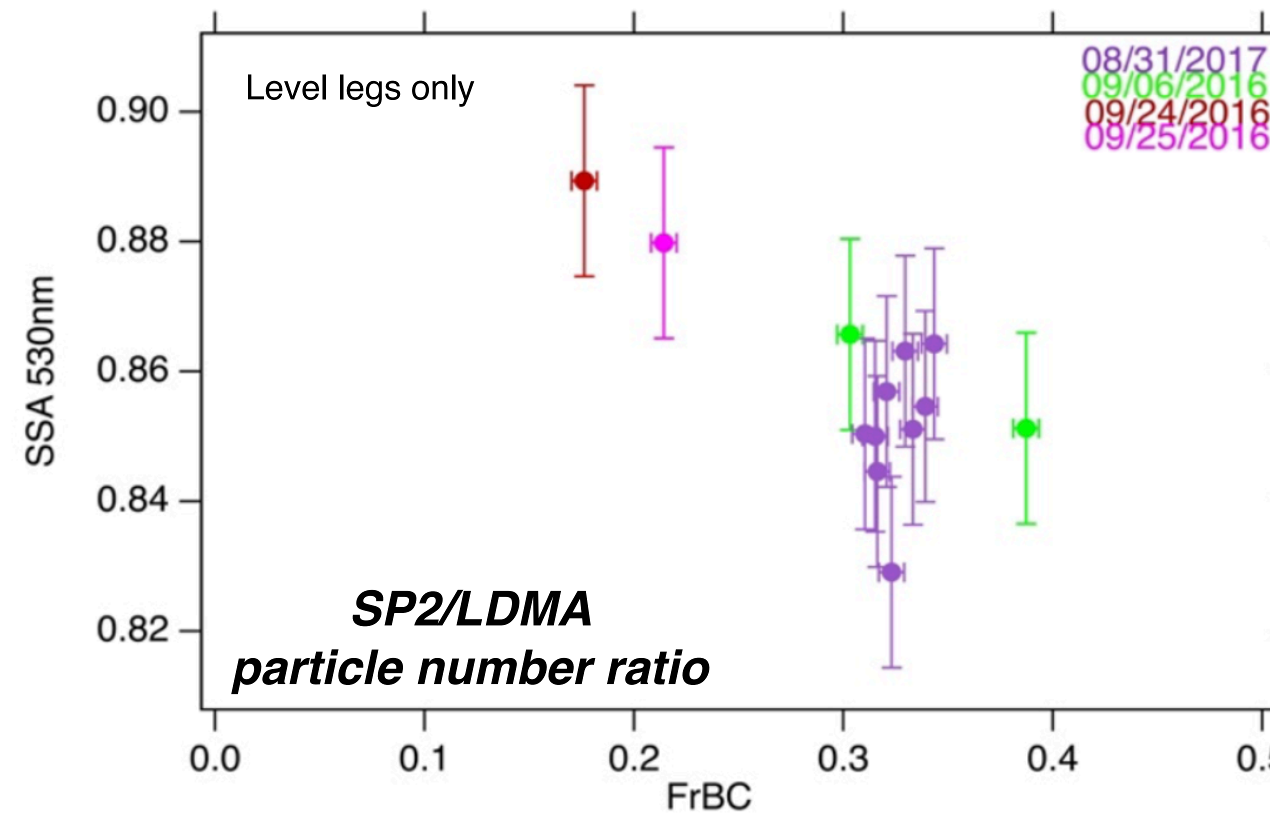
Highest OSC: 0.4-0.5; highest MCE of the flights



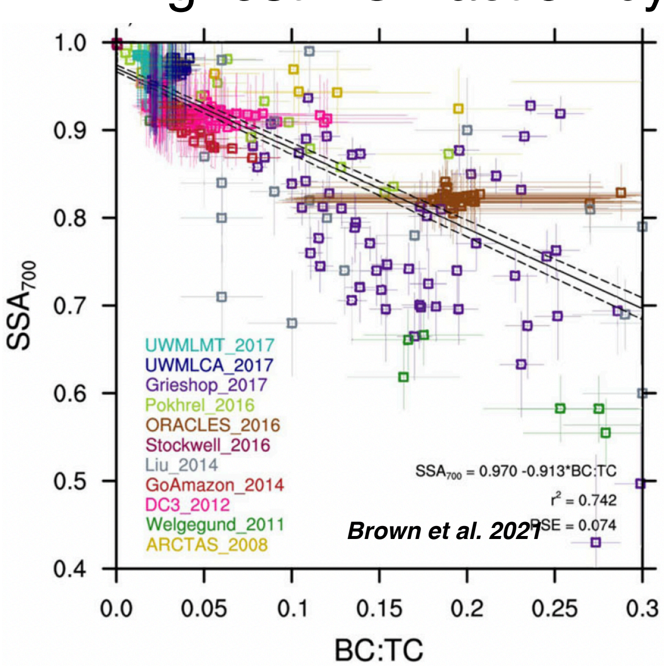
Summary:

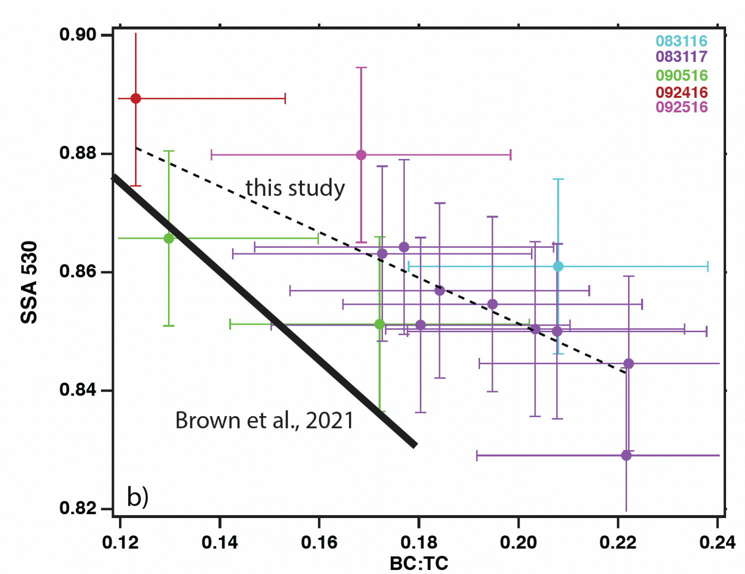
- most of SSA variability (0.85-0.87) explained by BC fraction at emission
- Evidence of net OA loss with age in one flight w/ constant BC:CO
- Intriguing difference between ORACLES-2016 and CLARIFY-2017 also seen in 8/31/2017 oracles flight, no robust explanations yet





Highest BC fraction by number





We find a weaker dependence on BC:total carbon than Brown et al. 2021

Interested in any comments, feedback:
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