Estimating the radiative effect and constraining the free parameter space of BrC aerosols in GISS ModelE

Presenting Author: *Maegan (Maggie) DeLessio[1,2]*

Co-Authors: *Kostas Tsigaridis[3,2], Susanne Bauer[2,3], Greg Schuster[4]*

Affiliations: [1]Department of Earth and Environmental Sciences at Columbia University, [2]NASA Goddard Institute for Space Studies, [3]Center for Climate Systems Research at Columbia University, [4]NASA Langley Research Center

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Previous work to improve representation of OA absorption

BrC introduced into One-Moment Aerosol Module (OMA)

- **Emissions:** 35% of OA from biomass burning are BrC
  - Range: 15-55%
- **Secondary BrC:** biogenic SOA (isoprene and terpenes) assumed brown
- **Optical properties:** non-zero imaginary refractive index (iRI)
  - Primary: 0.0165 (0.003-0.03)
  - Secondary: <0.002
- **Chemical aging:** primary BrC browns then bleaches

Estimating radiative effect and model sensitivity

- **TOA, annual average BrC direct effect:** 0.04 ± 0.01 W/m^2
  - Other ESMs: 0.029-0.13 W/m^2
  - CTMs: 0.04-0.57 W/m^2
- **Sensitivity tests:** 1 of 4 BrC parameters varied to see change in BrC radiative effect
  - Including SOA and chemical bleaching have distinguishable effects
  - Can't narrow range of iRI and emissions ratio

Evaluation of model with BrC scheme

- **Compared ModelE output to AERONET and MODIS total AOD and AAOD:** showed limited/no difference with and without BrC
  - Similar model skill regardless of BrC representation

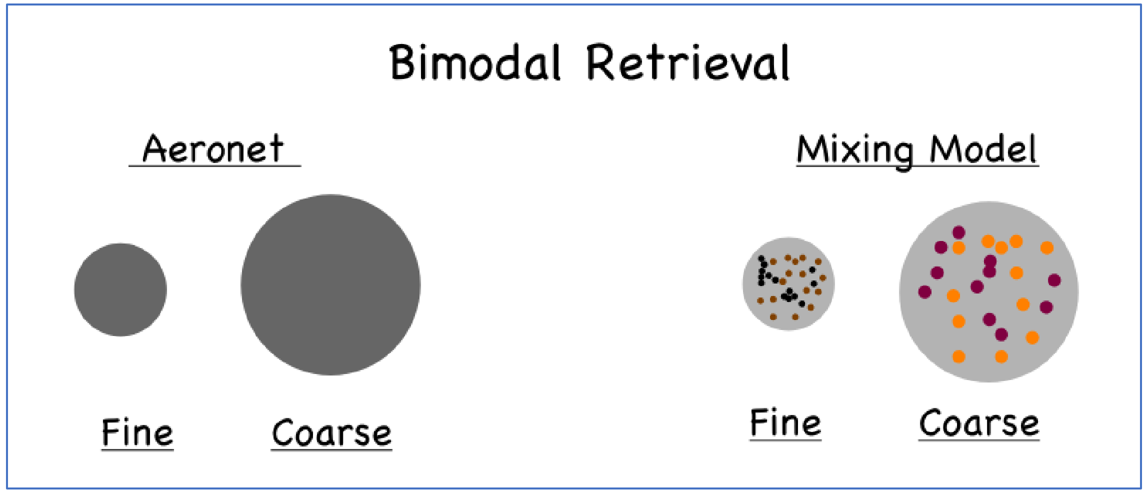
Have not been able to constrain the BrC free parameter space:

<u>Need to evaluate scheme against BrC data</u>

DeLessio et al., 2023 ACP

(https://doi.org/10.5194/egusphere-2023-2472)

AERONET retrieval of BrC properties



Retrieved size and complex refractive index → Internal mixture: non-absorbing organic host aerosol with absorbing inclusions

BrC data available for comparison

- Column burden: BrC, BC, organic host
- BrC AOD and AAOD

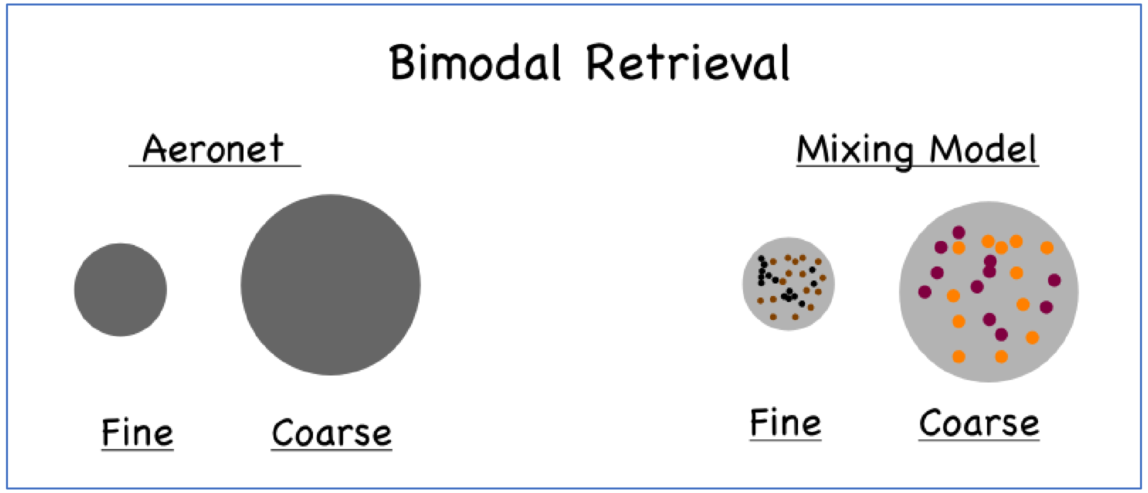
Schuster, G. L., Dubovik, O., and Arola, A.: Remote sensing of soot carbon – Part 1: Distinguishing different absorbing aerosol species, Atmos. Chem. Phys., 16, 1565–1585, 2016.

In-situ measurements of BrC absorption

BrC data available for comparison

- Absorption in Mm^{-1}, typically at 365 nm

AERONET retrieval of BrC properties



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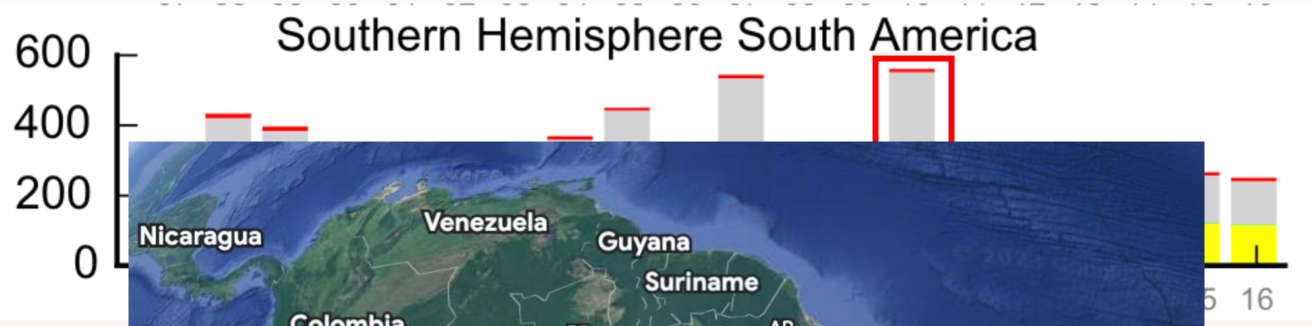
Annual fire carbon emissions



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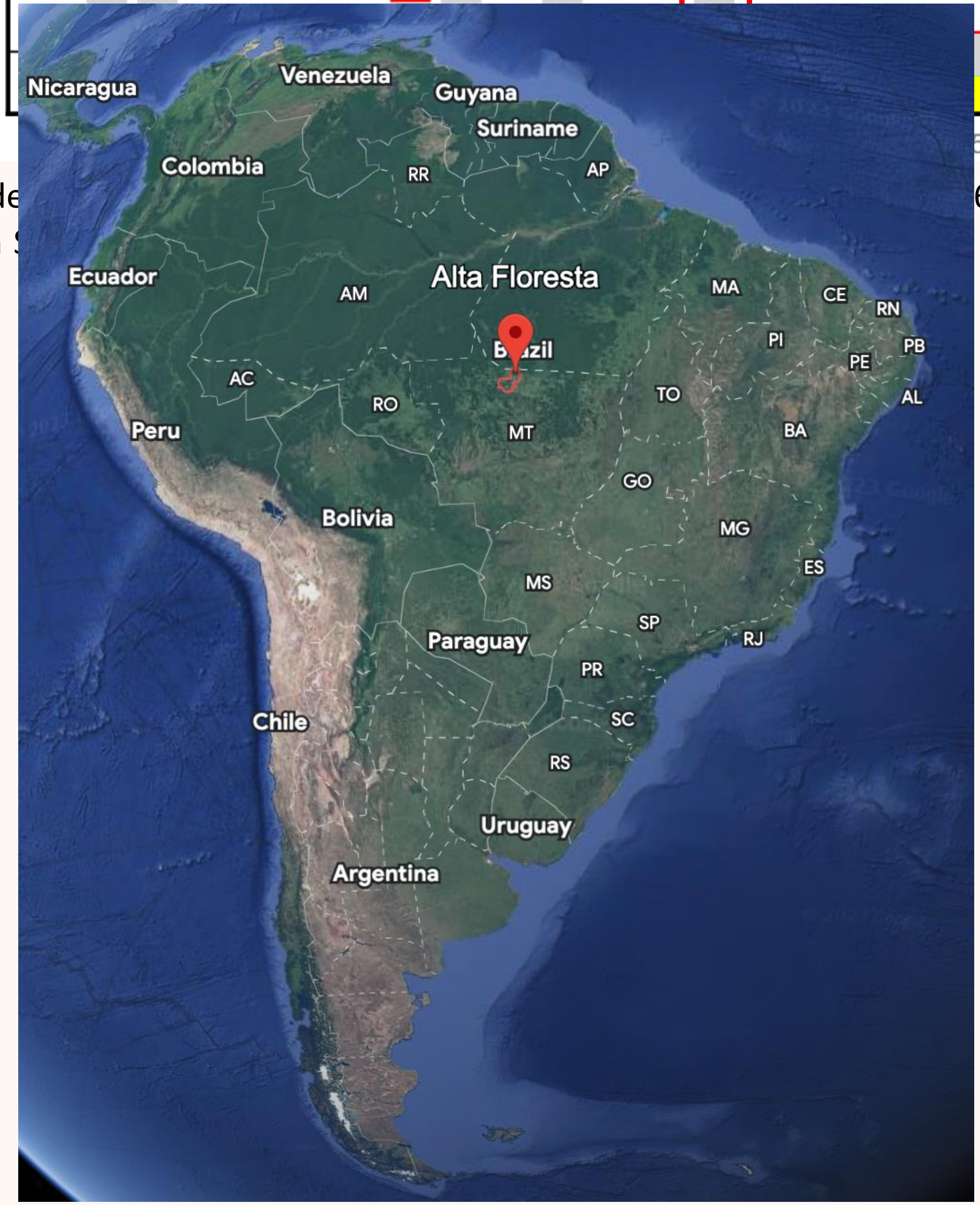
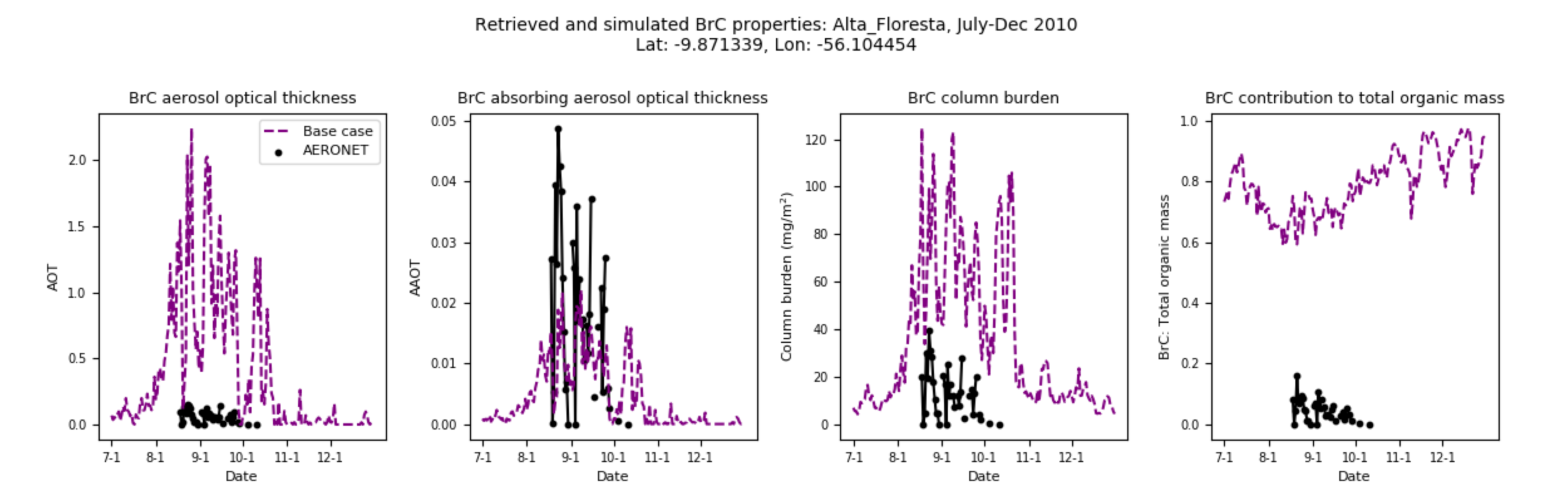




Initial Findings:

- Apparent inconsistency between retrieved BrC AOD/AAOD and simulated BrC properties
  - ModelE overestimates BrC AOD and mass, closer to BrC AAOD (or vice versa) → ModelE does not simulate as much BrC absorption with same mass

Exploring inconsistencies in BrC treatment between AERONET and ModelE

AERONET retrieval assumptions constrain 3 of 4 BrC properties

Key assumption: One, constant imaginary RI for BrC

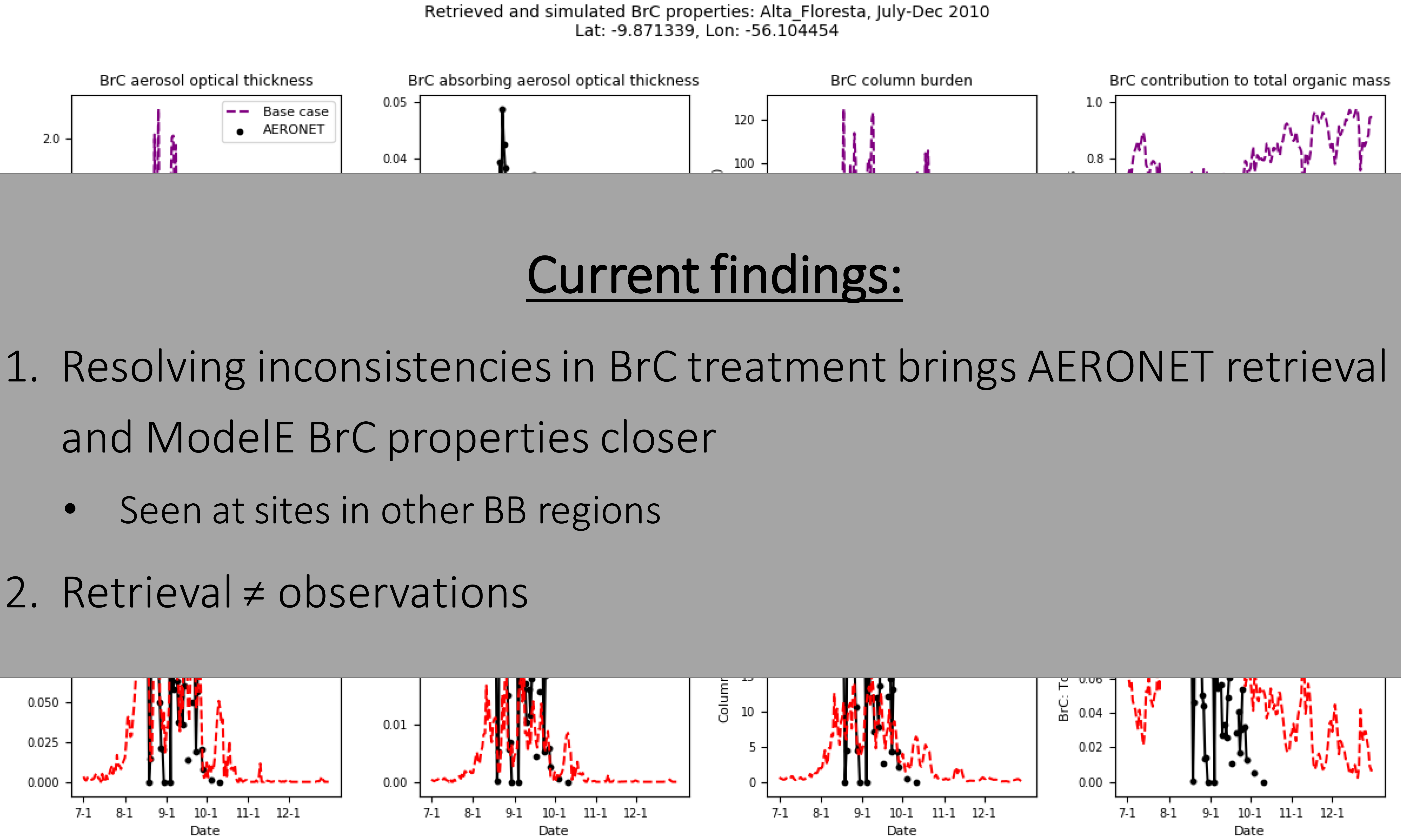
1. **Secondary BrC:** not included
2. **Optical properties:** k_{BrC} = 0.03
3. **Chemical aging:** none
4. **Emissions ratio (BrC-to-OA)**
   …we know mass tends to be overestimated

Run a "revised" simulation with:

1. No secondary BrC
2. More strongly absorbing primary BrC
3. No chemical aging
4. Reduction in emitted BrC: 18% BrC-to-OA

*****1.5 absorption enhancement factor applied

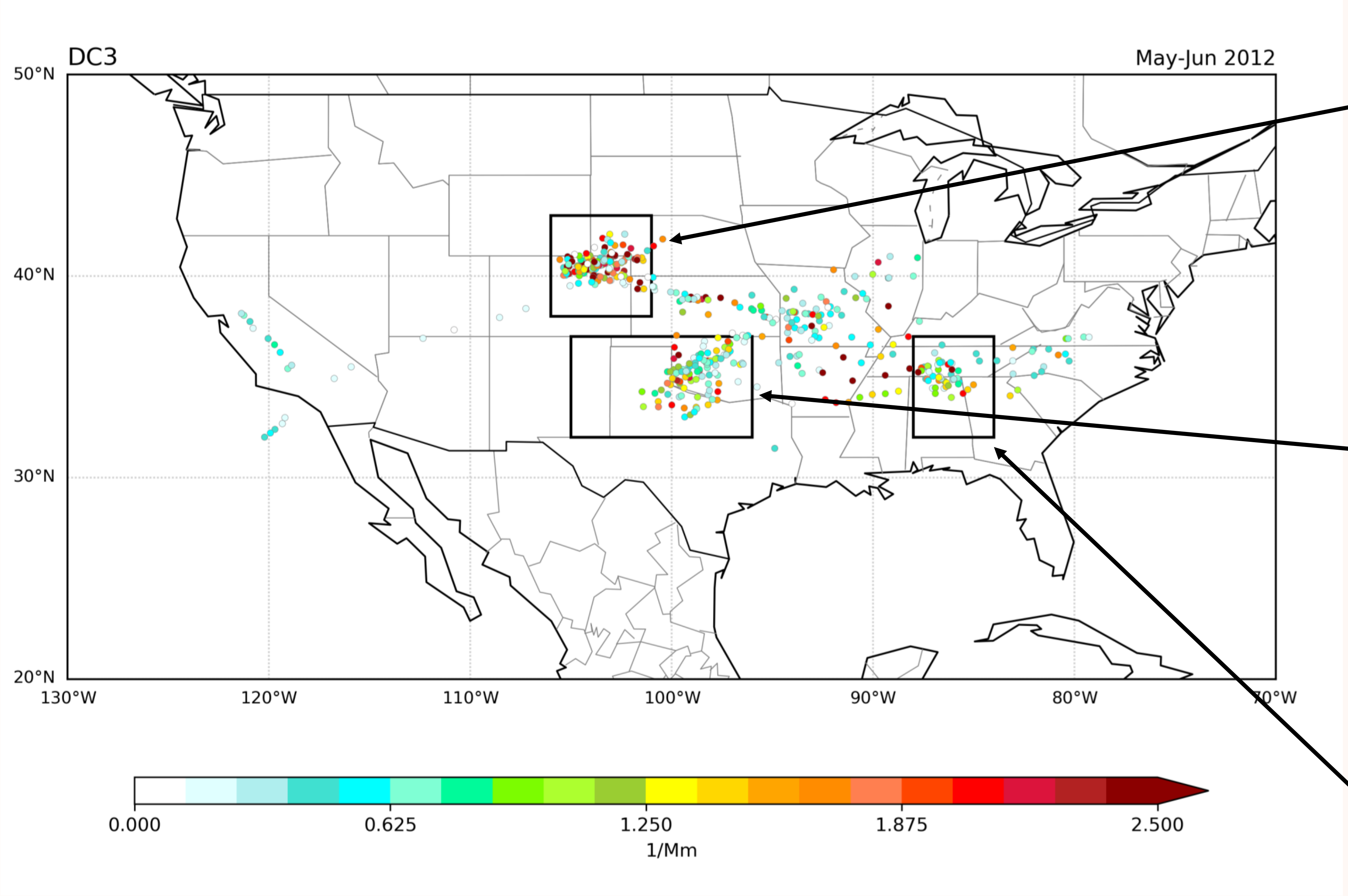
Goals of simulation: *reduce BrC mass (and BrC AOD), increase mass absorption efficiency (MAE)*



Current findings:

1. Resolving inconsistencies in BrC treatment brings AERONET retrieval and ModelE BrC properties closer
    - Seen at sites in other BB regions
2. Retrieval ≠ observations

Looking at in-situ BrC data from DC3 flight campaign



Northeastern Colorado:

- Urban corridor + agriculture/ranching
- Moderate-high anthropogenic VOCs, low BVOCs (except Rockies)
- PM1 mostly OA

Central Oklahoma to west Texas

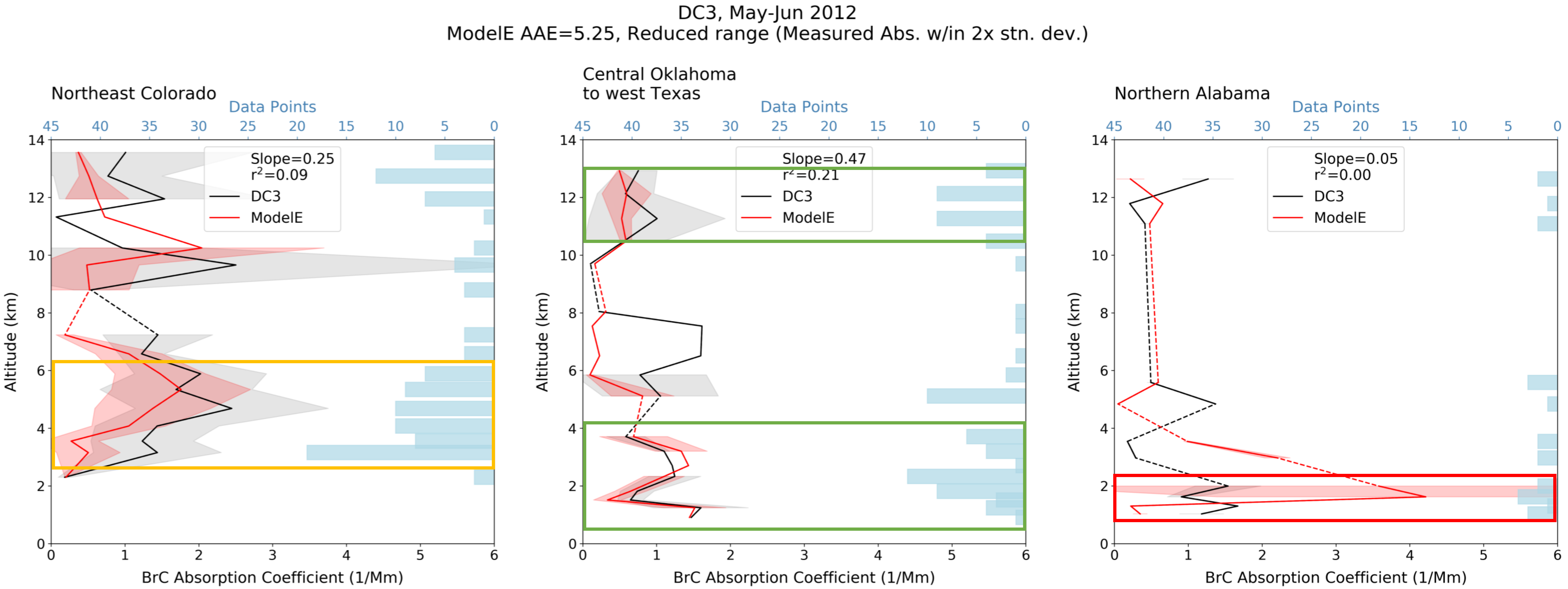
- Sparsely populated southern Great Plains: agriculture, pasture, grassland
- Metro area (Oklahoma city + Dallas-Ft. Worth outflow) affect central OK
- Low-moderate BVOCs, low-moderately high anthropogenic VOCs
- PM1 ~40-60% OA

Northern Alabama:

- High BVOCs from forested areas
- Regional sources/Birmingham: moderate anthropogenic VOCs
- PM1 ~40% OA, more sulfate

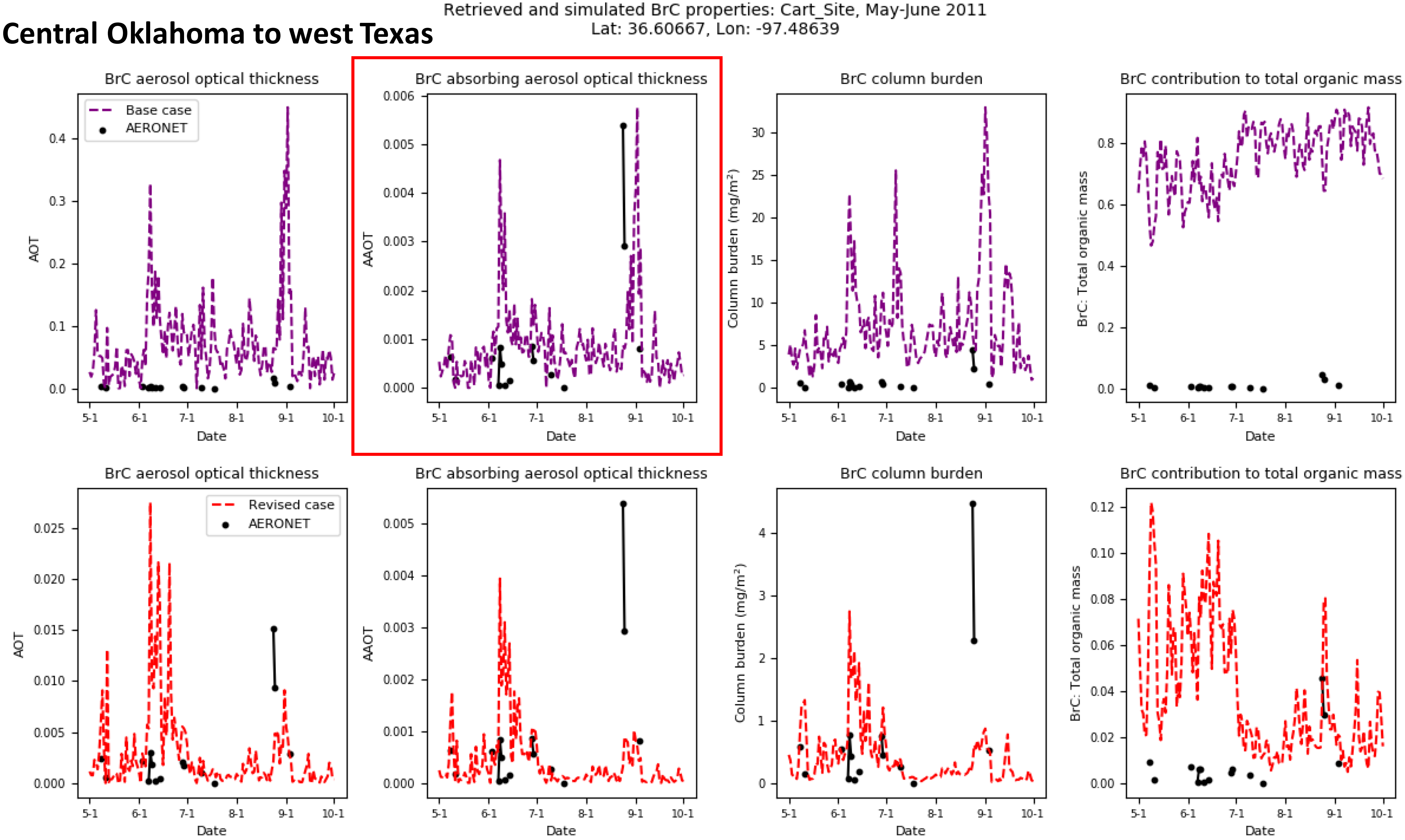
Zhang, Y. et al.: Top-of-atmosphere radiative forcing affected by brown carbon in the upper troposphere, Nature Geosci, 10, 486–489, 2017.
Barth, M. C. et al.: The Deep Convective Clouds and Chemistry (DC3) Field Campaign, Bulletin of the American Meteorological Society, 96, 1281–1309, 2015.

Vertical profiles within each sub-region:
gridded to model resolution (2º x 2.5º), averaged over entire measurement period

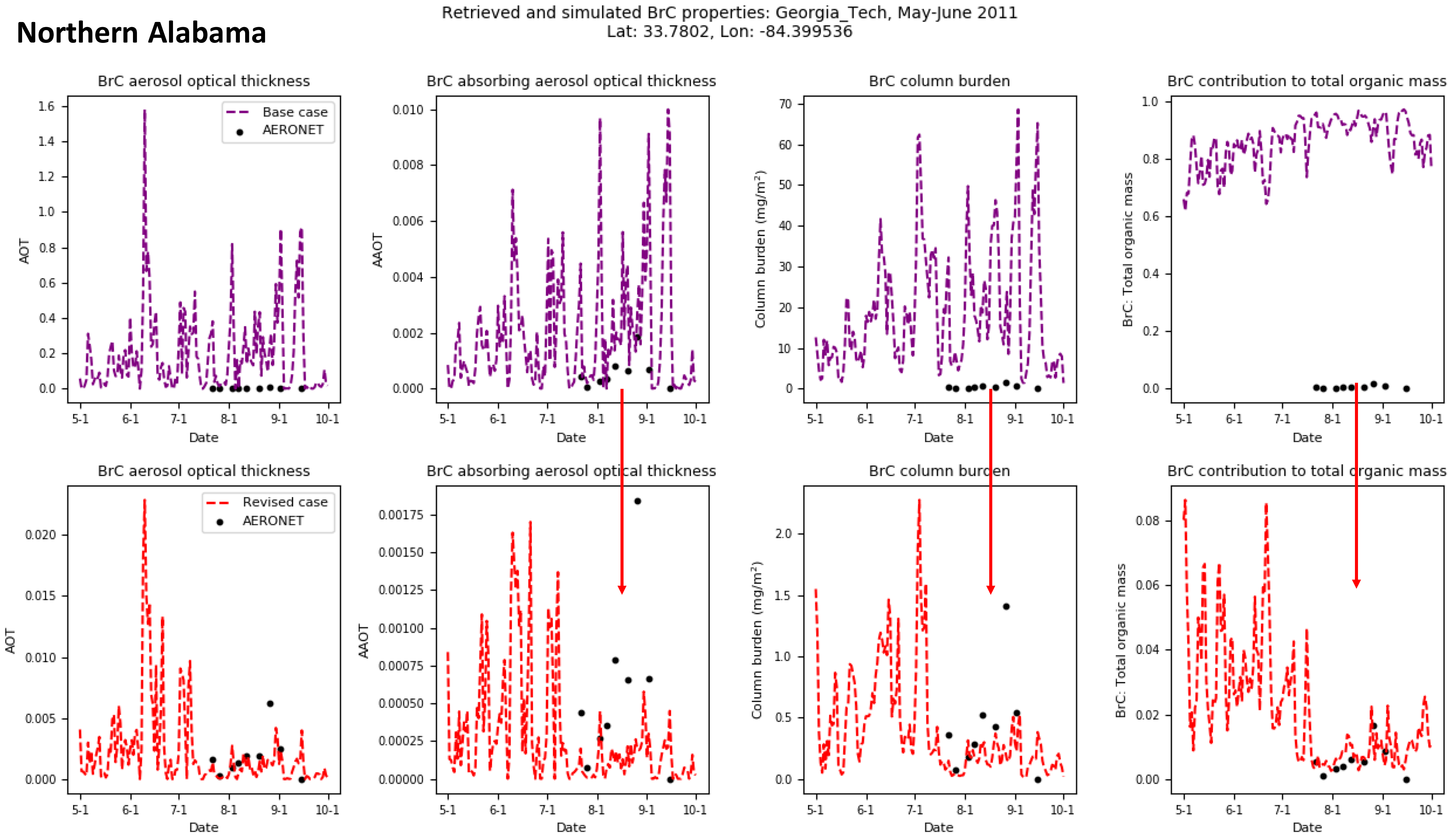


What if we apply the same analysis of AERONET sites to these regions?

Central Oklahoma to west Texas



Northern Alabama



What are we seeing and where do we go from here?

- Beginning to see regional differences in needed BrC representation emerge
  - Will explore co-located data to investigate this: WSOC mass, mixing ratio of VOCs (e.g. isoprene, toluene, pinene), number fractions of aerosols from different sources, etc.
- Will continue this study looking at other flight campaigns
  - Allows us to look at BrC in different regions and under different conditions

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Acknowledgements

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Contact: md3345@columbia.edu

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