Investigation of brown carbon formation due to the drying of ambient aerosol in eastern United States

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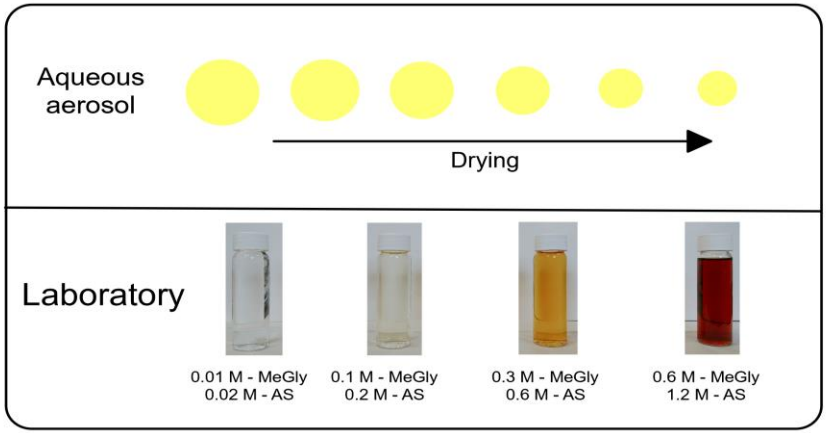


What is BrC and its formation

BROWN CARBON
Organic molecules like tar balls or fats, given off by long-smoldering fires

BLACK CARBON
Carbon particles given off by hot fires, like coal plants, forest fires, and combustion from cars

Argonne Nat. lab

Brown carbon

- Brown Carbon (BrC) is the light-absorbing organic carbon mostly generated in primary form by burning biomass.
- Brown carbon is defined operationally as the organic carbon which has strong absorbance in UV region (< 400 nm) – *First defined by Kirchstetter and Novakov, 2004, JGR*

Laskin et al., Chemical Reviews, 2015

Brown carbon

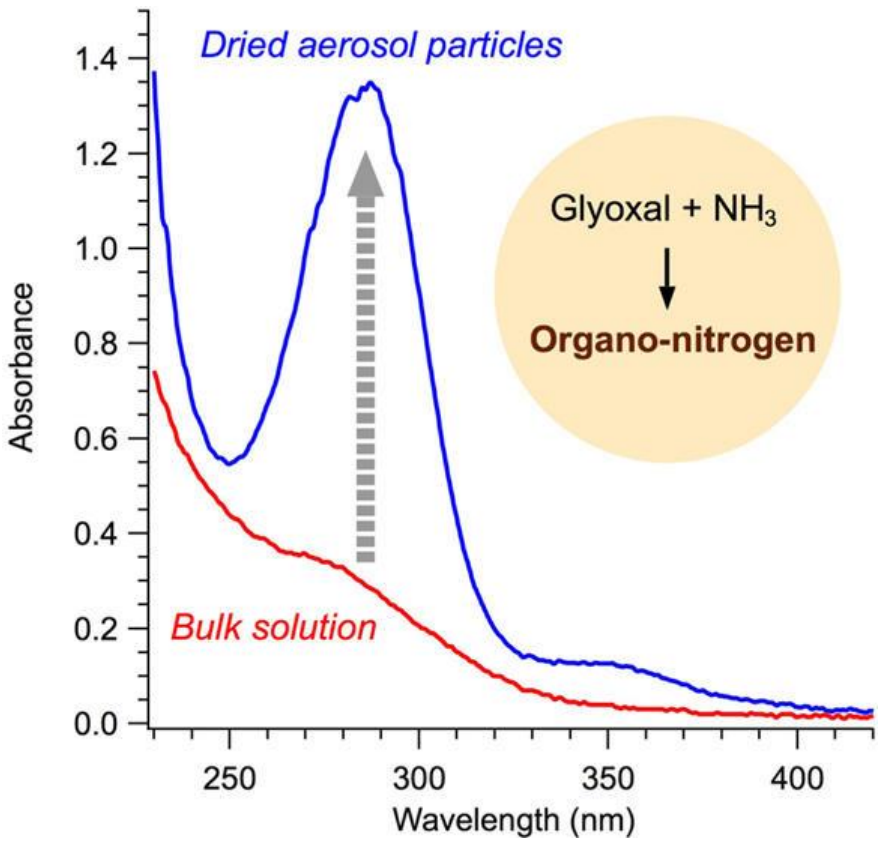
- BrC is formed through secondary processes too.
- Recent laboratory studies suggest BrC formation in aqueous particles containing organic compounds (like glyoxal, methylglyoxal) and $(NH_4)_2SO_4$ when dried partially.
- Ambient particles undergo drying and water intake cycles during their lifetime; therefore, may lead to BrC formation.
- The phenomenon, however, is never reported for ambient particles.

Laskin et al., Chemical Reviews, 2015

Motivation

- BrC formation observed with the drying of glyoxal + AS aerosol particles generated in the lab.
- The drying timescale of the order of few seconds yielded BrC formation.
- Drying was performed at ≤ 35% RH

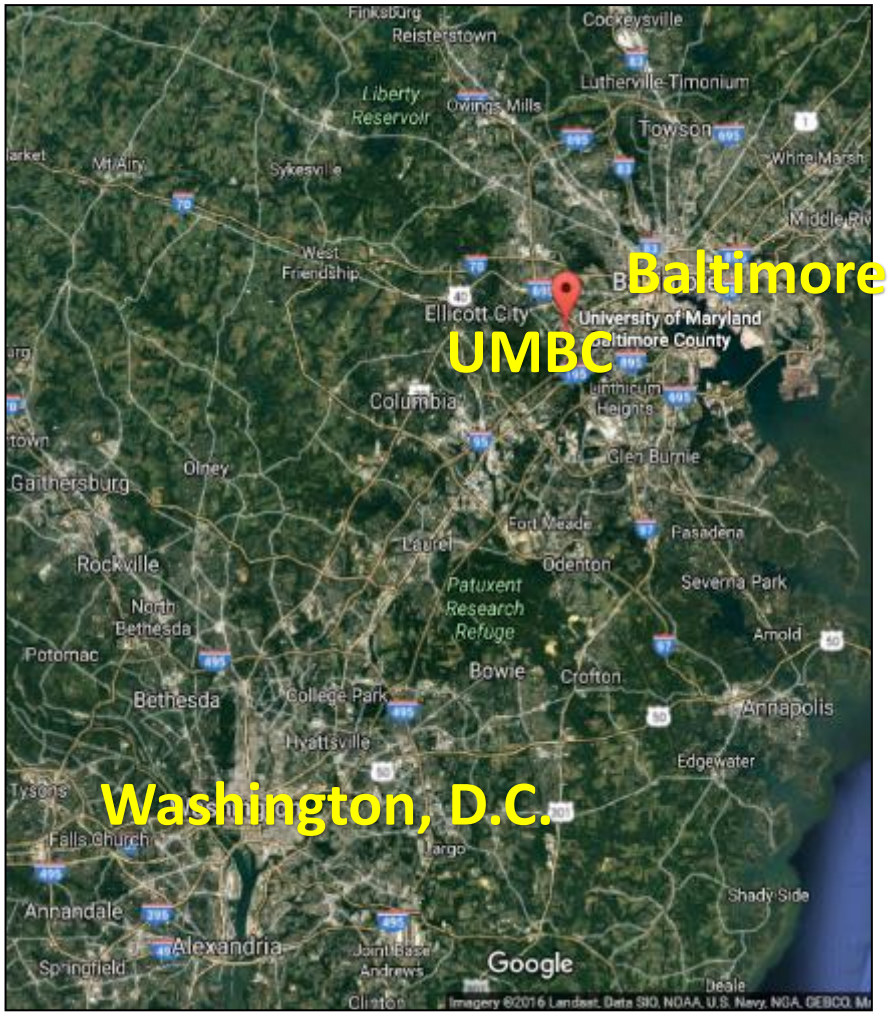
Lee et al., ES&T (2013)



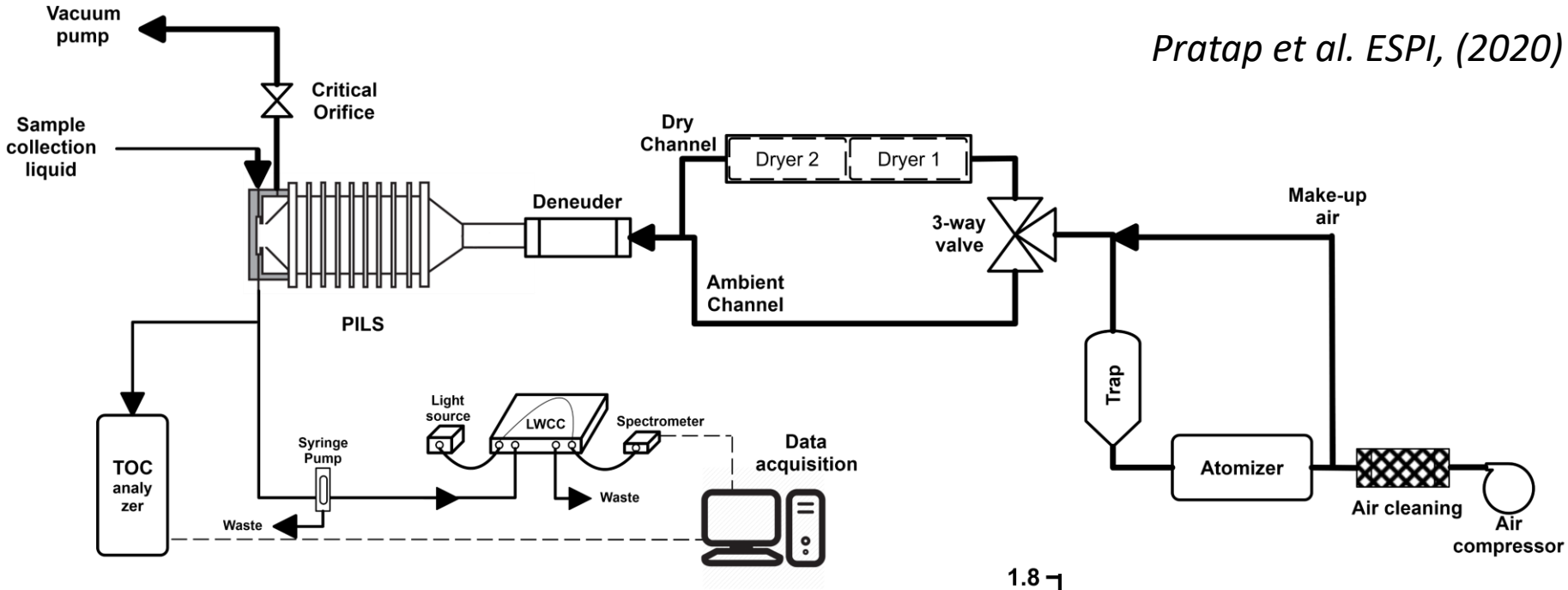
Investigate the phenomenon in ambient particles

Measurement location: Baltimore, MD, USA

- Summer: July – August, 2018, August 2019
- Total organic carbon (TOC), absorbance, NH_3 , and other inorganic ions were measured.



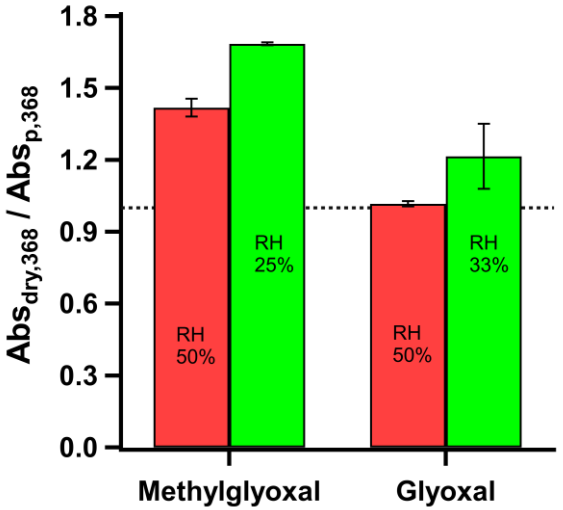
Lab Experiment - setup



Pratap et al. ESPI, (2020)

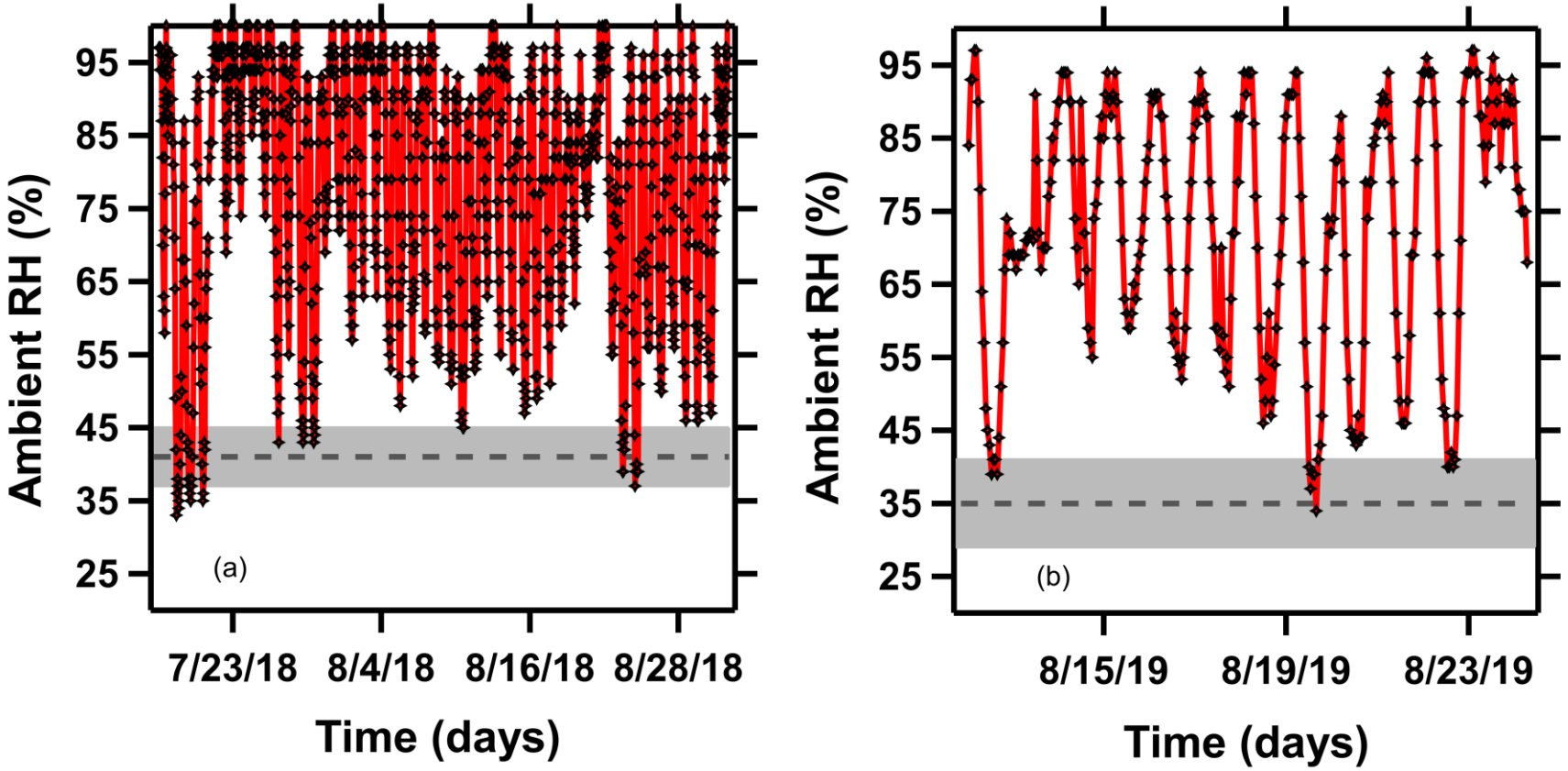
- 42% enhancement for MeGly at 50% RH.
- On further drying - ~19% absorbance enhancement for both Gly and MeGly at ~30% RH.

The setup demonstrates BrC formation on drying



Drying at what RH?

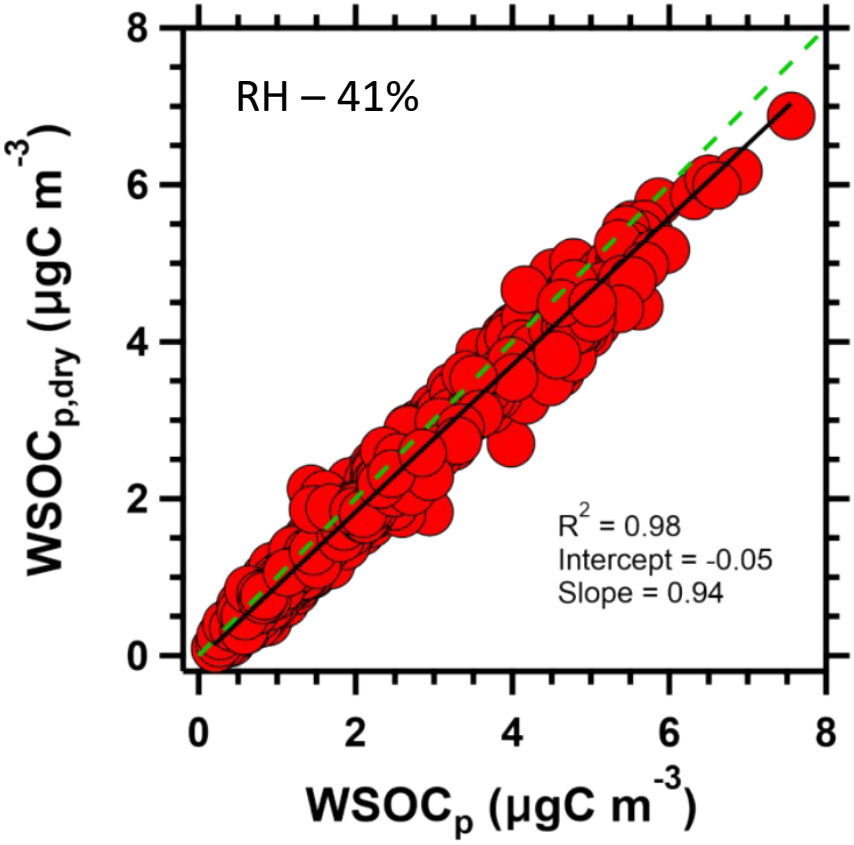
Pratap et al. ESPI, (2020)

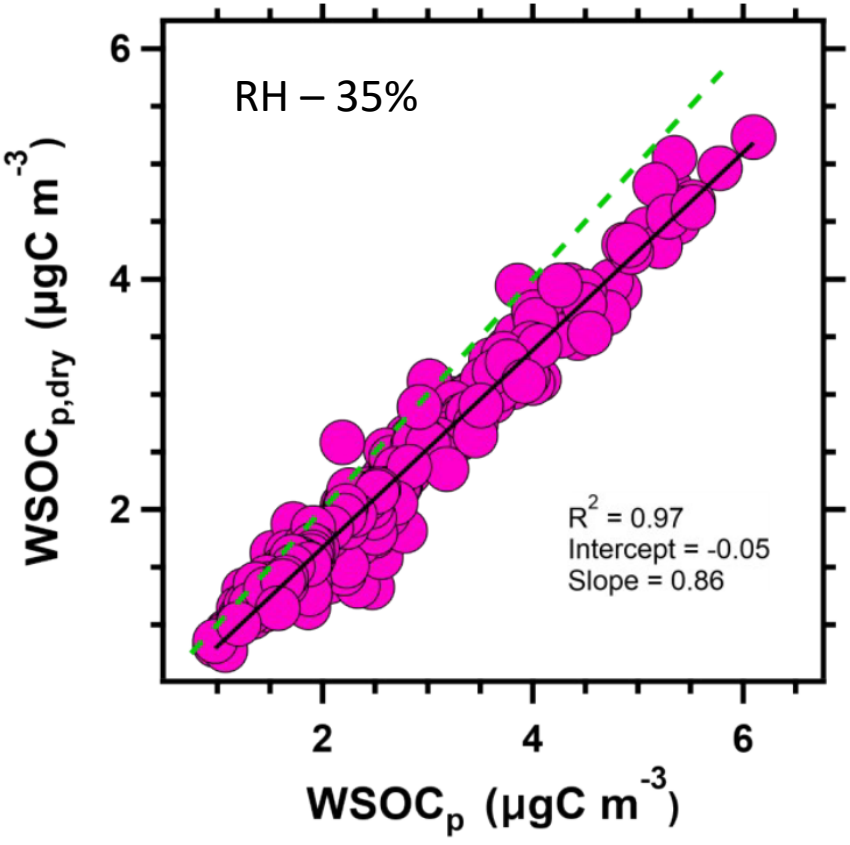


- Average daytime RH in Baltimore is ~ 55%.
- Keep drying RH atmospherically relevant
- Experiments performed at 41% RH (2018) and 35% RH (2019)

Results – evaporation of organics

Pratap et al. ESPI, (2020)

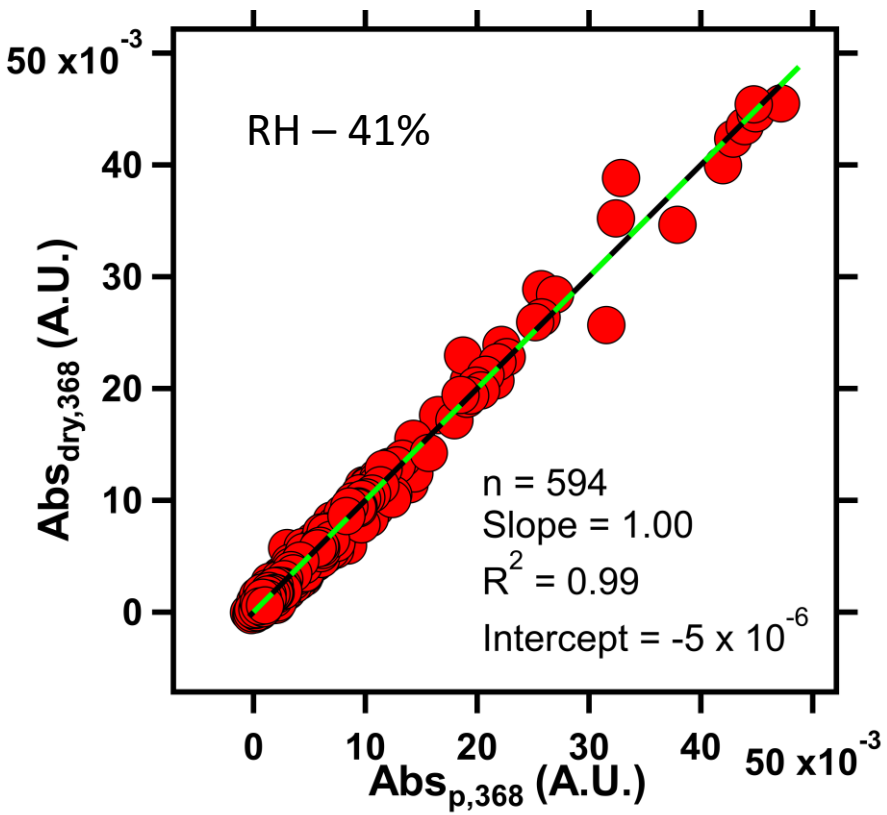


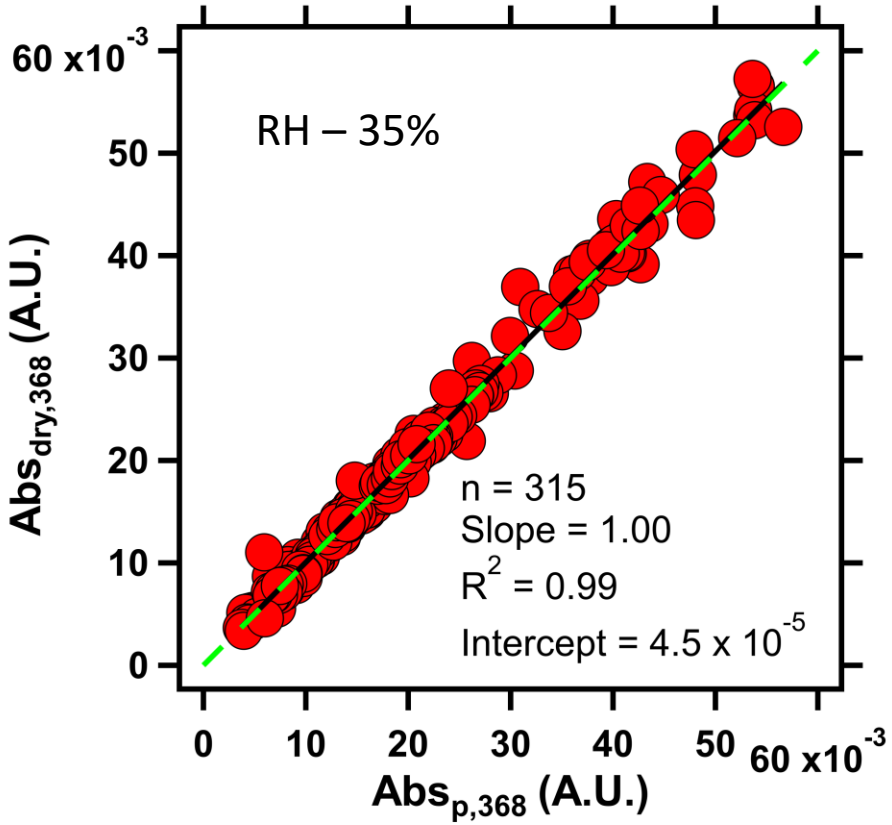


Evaporation of WSOC in both at RH 41% (2018) and 35% (2019)

Absorbance – dry vs ambient

Pratap et al. ESPI, (2020)



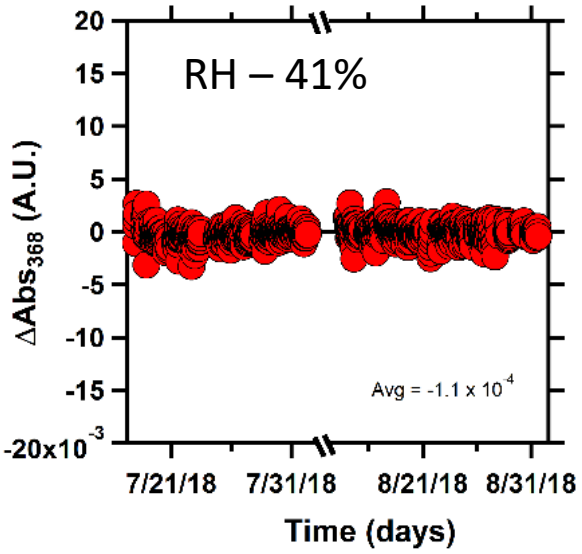


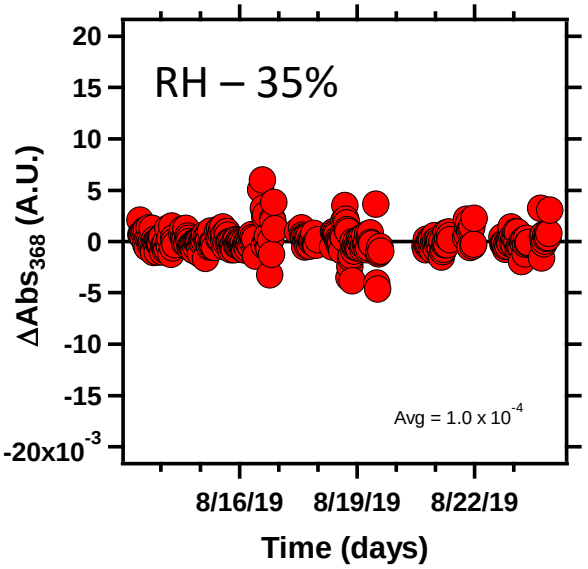
Pratap et al. ESPI, (2020)

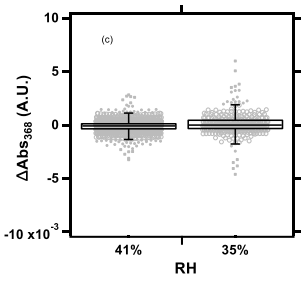
No difference in BrC due to particle drying.

Delta absorbance

Pratap et al. ESPI, (2020)



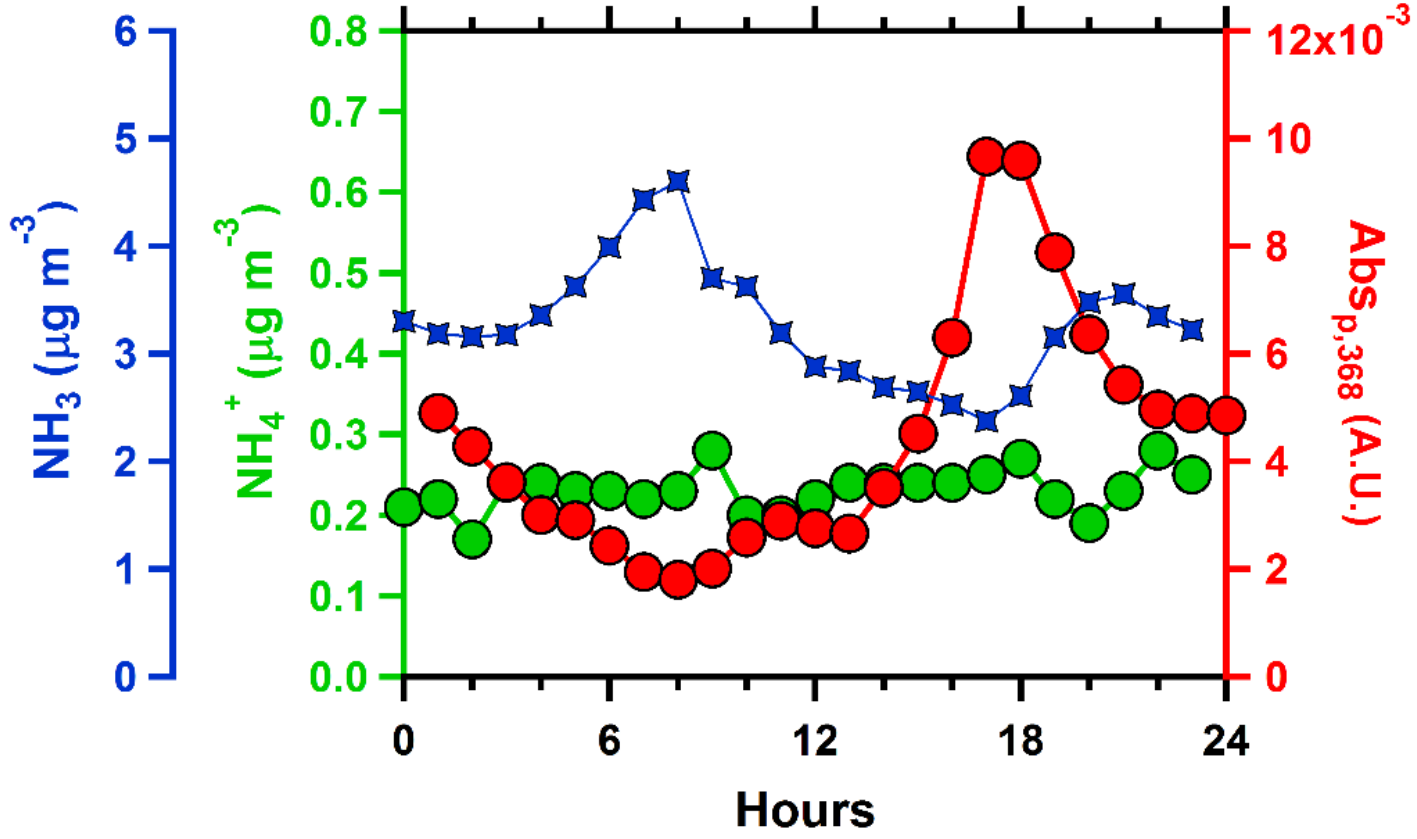




No difference in delta absorbance.

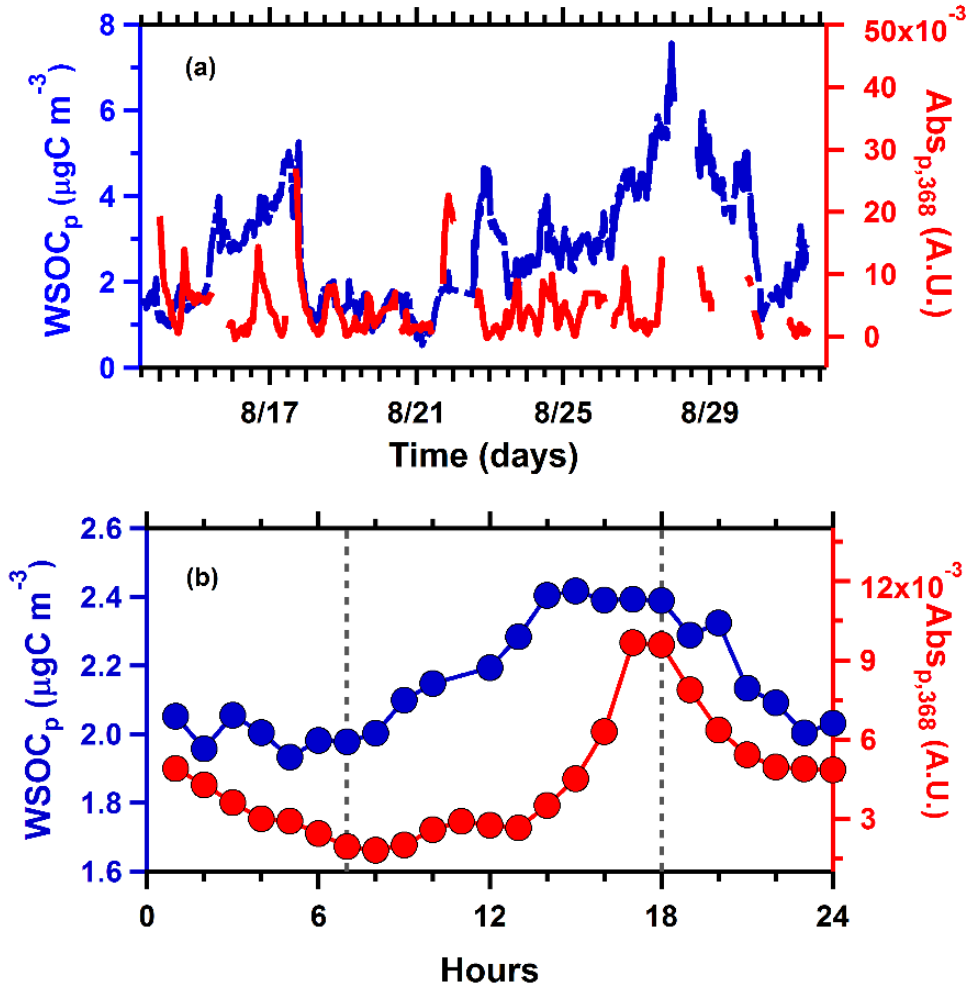
Ammonia/Ammonium

Pratap et al. ESPI, (2020)



- BrC formation does not seems to be limited by NH_4^+ conc
- BrC and NH3 conc. Are anti-correlated, but it is not likely causative

WSOC and Absorbance



Pratap et al. ESPI, (2020)

- Absorbance appears to be loosely correlated with WSOC.
- Photochemical production of WSOC and BrC.

Summary and Conclusions

- No evidence of BrC formation was found in dried ambient particles at 41% and 35% RH.
- Most probable cause is the extremely low conc of BrC precursors.
- Laboratory experiments demonstrate BrC formation in Gly and MeGly particles when dried.
- Diurnal ammonia concentration appears to be anti-correlated to BrC, though not likely causative.
- Further studies may be required to understand the effect of ammonia on BrC formation in ambient particles.

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No evidence for brown carbon formation in ambient particles undergoing atmospherically relevant drying†

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Recent laboratory studies have reported the formation of light-absorbing organic carbon compounds (brown carbon, BrC) in particles undergoing drying. Atmospheric particles undergo cycles of humidification and drying during vertical transport and through daily variations in temperature and humidity, which implies particle drying could potentially be an important source of BrC globally. In this work, we investigated BrC formation in ambient particles undergoing drying at a site in the eastern United States during summer. Aerosol BrC concentrations were linked to secondary organic aerosol (SOA) formation, consistent with seasonal expectations for this region. Measurements of water-soluble

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