Chemical and optical properties of black carbon particles in Toronto (CHEMBC)

Robert Healy

25 March 2015

Background- Black carbon

- "Black carbon" particles strongly absorb visible light across all wavelengths
- Sources include incomplete combustion of fossil fuels (vehicles, industry) and biomass (domestic burning, forest fires)
- Exert a positive direct radiative forcing (warming effect on climate)
- High uncertainty associated with climate impacts

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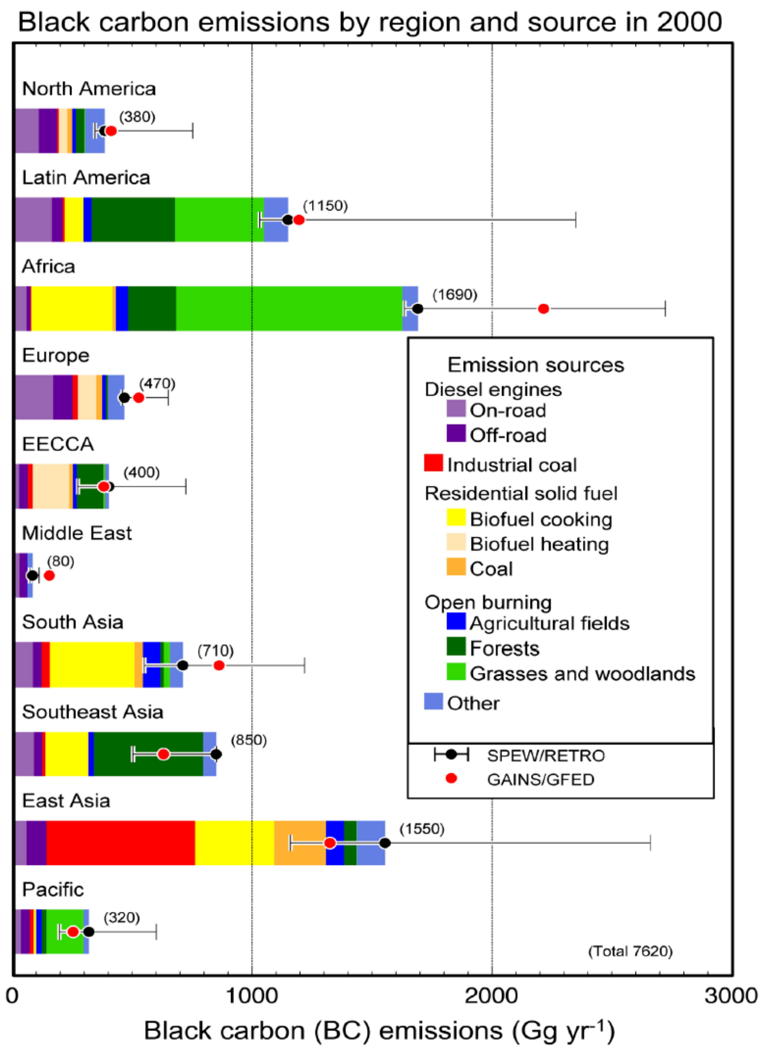
http://www.geog.ucsb.edu/ *http://www.catf.us/climate/*

Background- Black carbon



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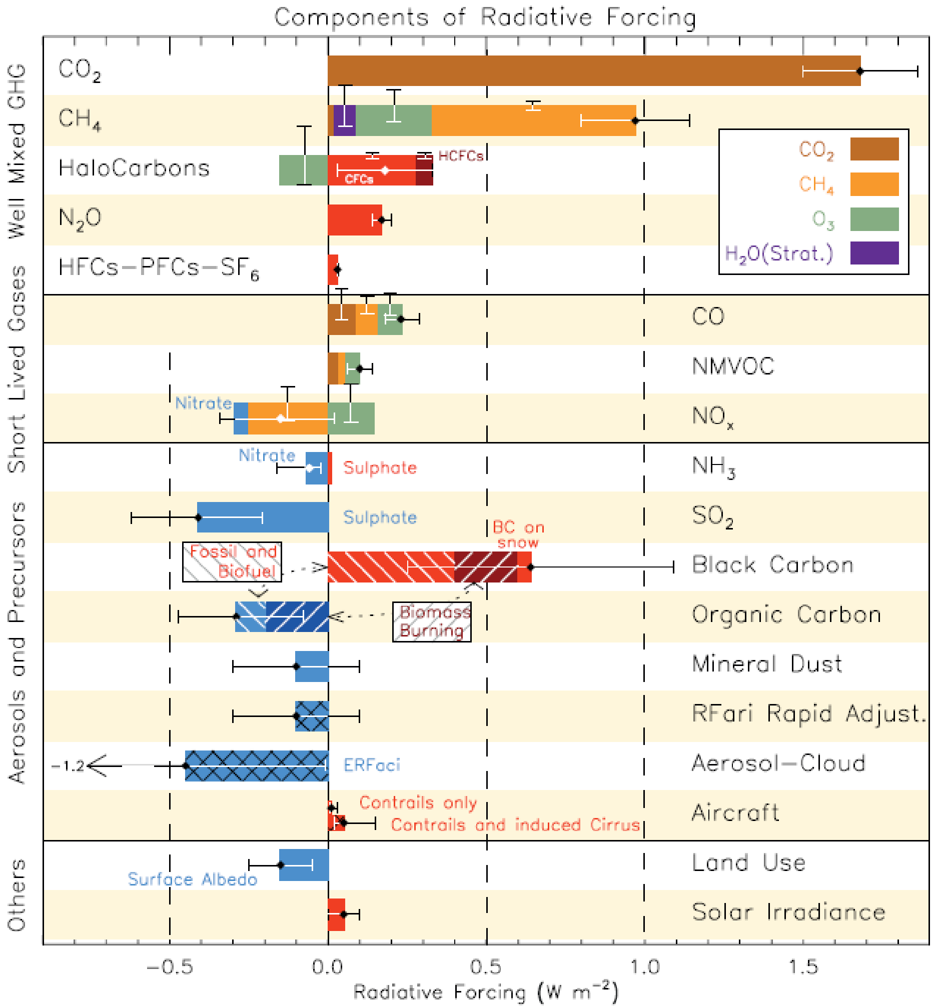
Bond et al. JGR 2013

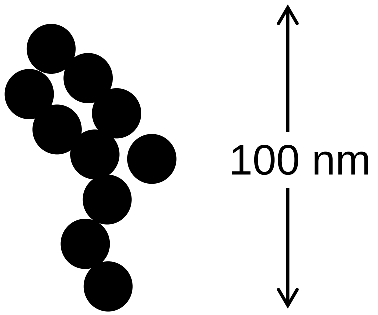
Background- Black carbon



Modern day radiative forcing relative to 1750

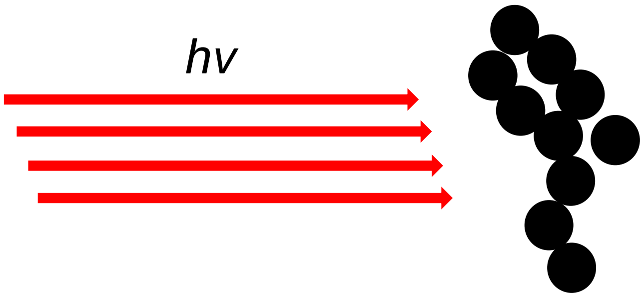
Black carbon absorption

- Black carbon (BC) particles are small, typically composed of multiple spherules, each with a diameter of about 20 nm



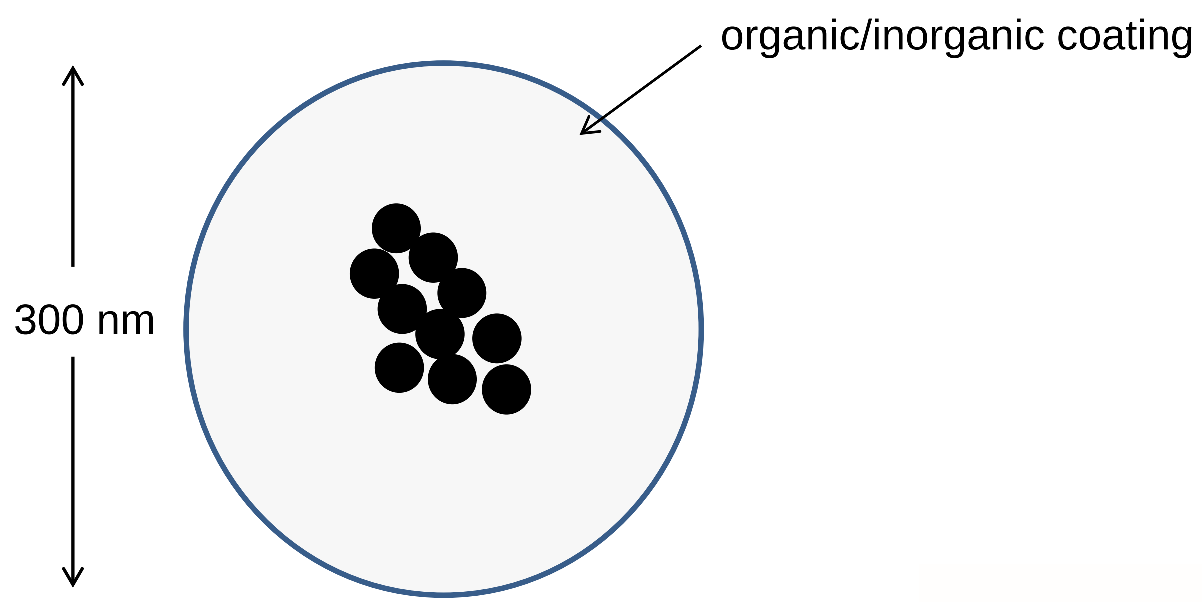
Black carbon absorption

- BC absorbs incoming solar radiation directly across a broad wavelength range



Black carbon absorption

- BC particles accumulate organic and inorganic secondary coatings during atmospheric transport



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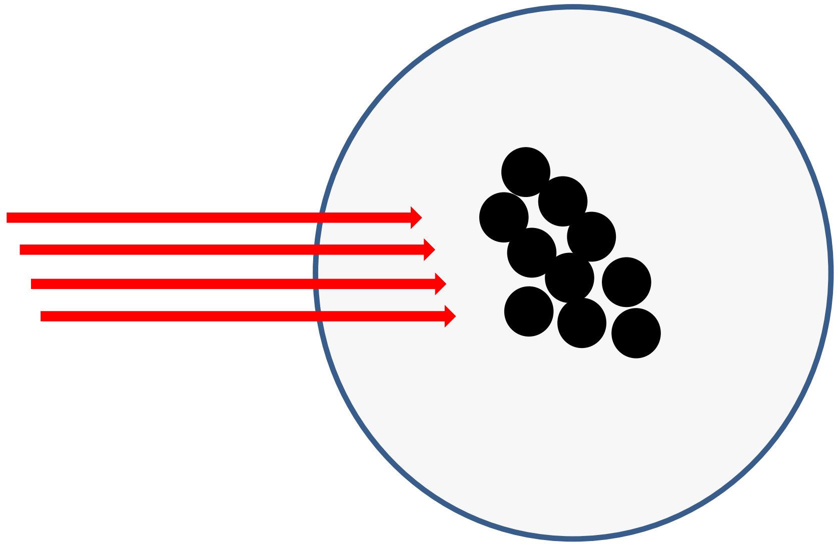
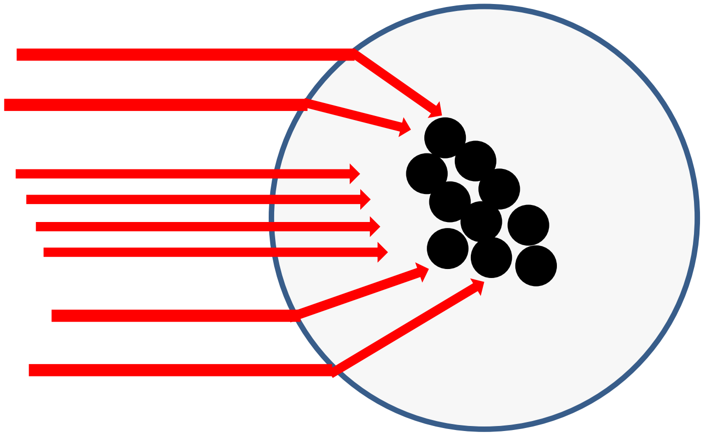
Black carbon absorption

- Coated black carbon can still absorb incoming radiation directly



BC absorption enhancement (E_{abs})

- Enhanced absorption of solar radiation has been proposed to occur through a "lensing effect" (refraction) for coated BC
- This enhancement effect has been demonstrated in laboratory studies
- But estimates of the enhancement for ambient BC vary greatly (6-50%), and thus measurements in different environments are needed

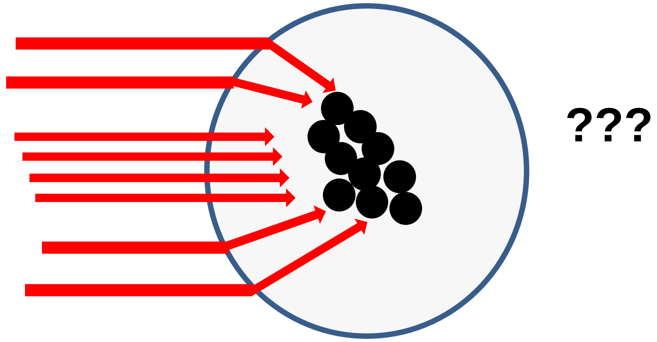


Cappa et al., Science 2012

Motivation

- Does BC coating composition and thickness affect absorption efficiency?

- If so, what is the impact in Toronto?



Campaign and instrumentation

Toronto, ON

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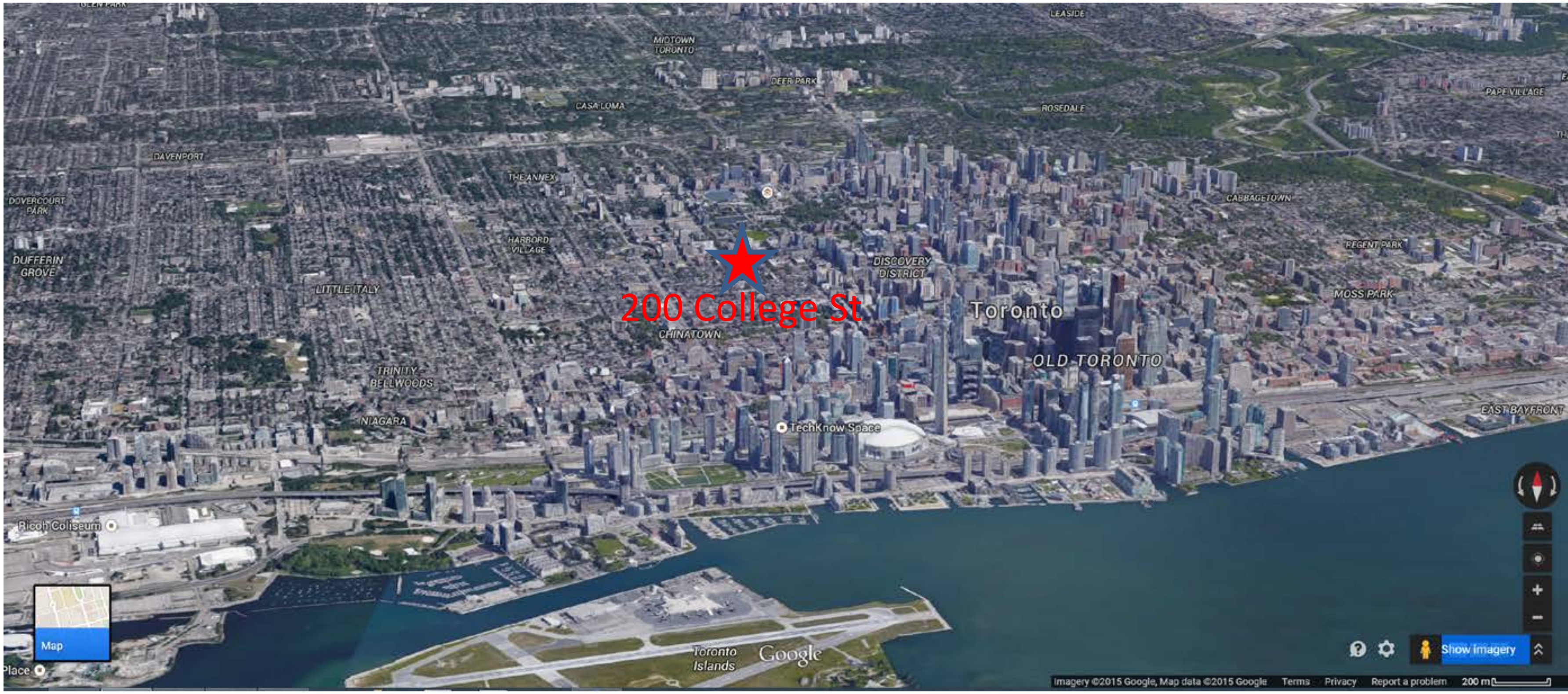
Campaign and instrumentation



Sampling took place over 3 weeks in June 2013

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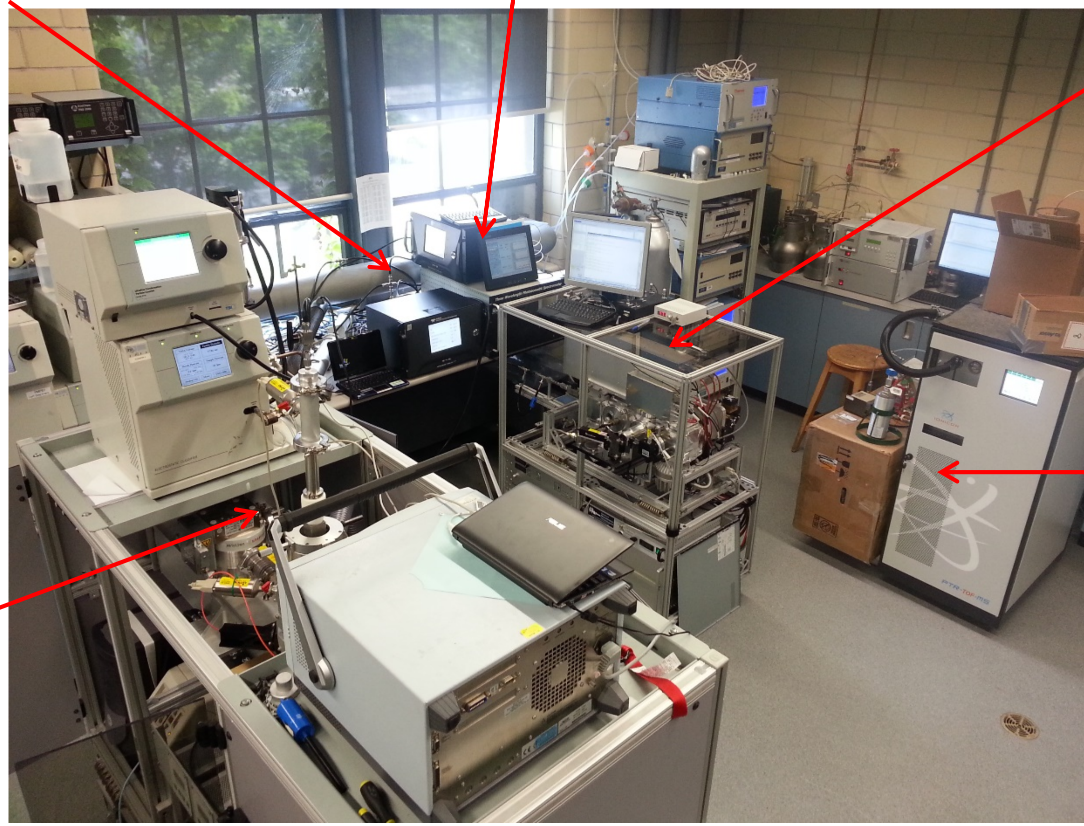
Campaign and instrumentation

Photoacoustic soot spectrometer (PASS)

Soot particle aerosol mass spectrometer (SP-AMS)

Thermodenuder



Aerosol time of flight mass spectrometer (ATOFMS)

Proton transfer reaction mass spectrometer (PTR-MS)

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Campaign and instrumentation

- Measurements were performed for three weeks in June 2013

Instrument	Function
Photoacoustic soot spectrometer (PASS-3)	Measures aerosol absorption/scattering at 405 nm and 781 nm
Soot particle aerosol mass spectrometer (SP-AMS)	Measures BC and coating material concentrations quantitatively
Aerosol time-of-flight mass spectrometer (ATOFMS)	Measures single particle composition and mixing state qualitatively

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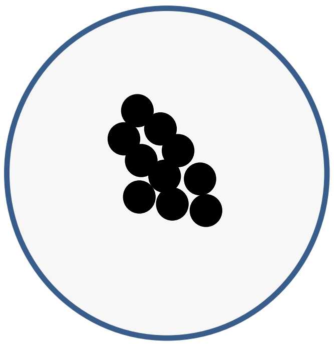
Experimental configuration

- Measure absorption for coated and uncoated (denuded) particles
- Is there a difference?

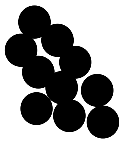
Measure absorption (b_{abs}) using PASS



Remove coatings using thermal denuder

250 °C

Measure absorption (b_{abs}) again using PASS



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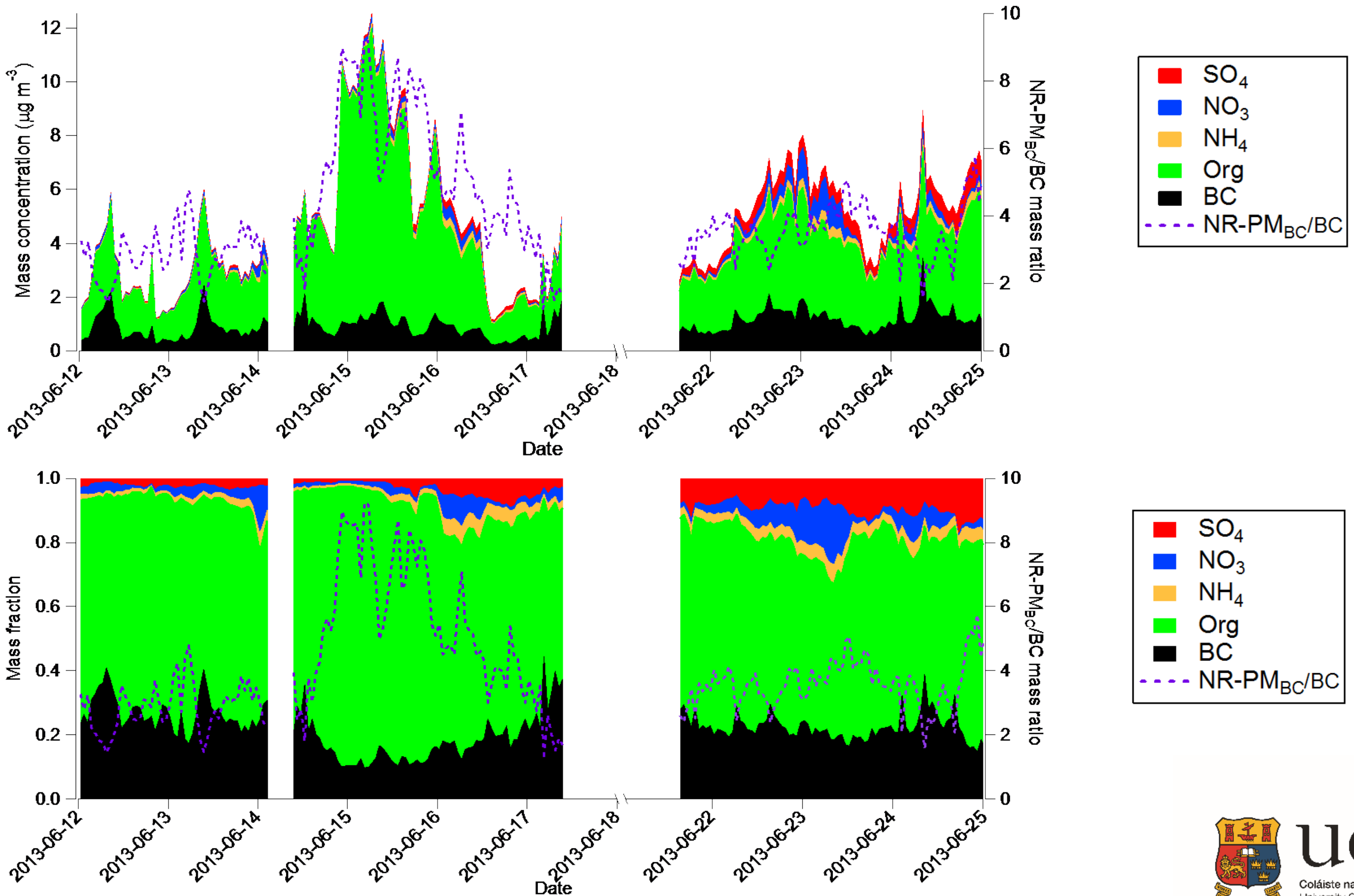
Results: BC-containing particle composition



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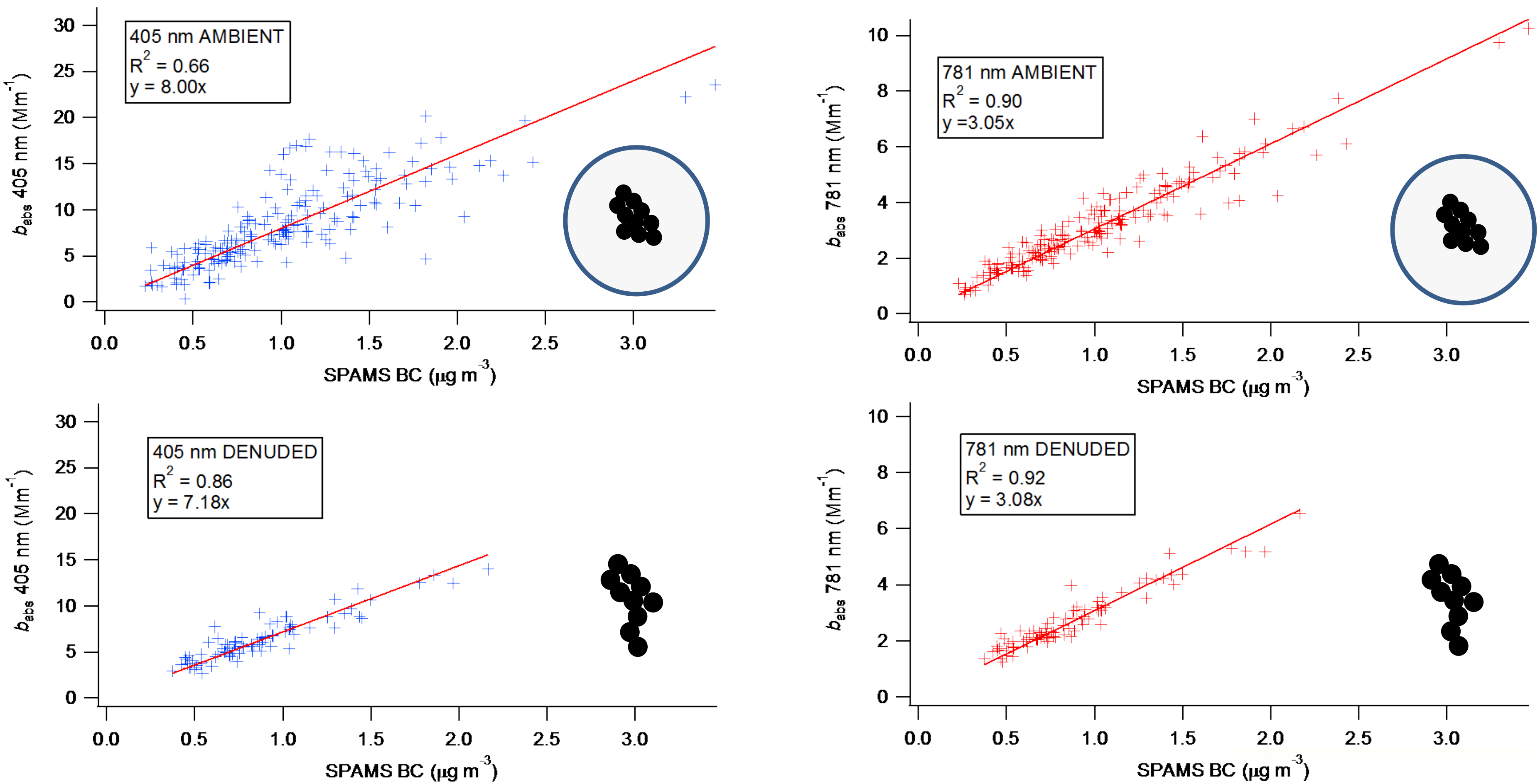
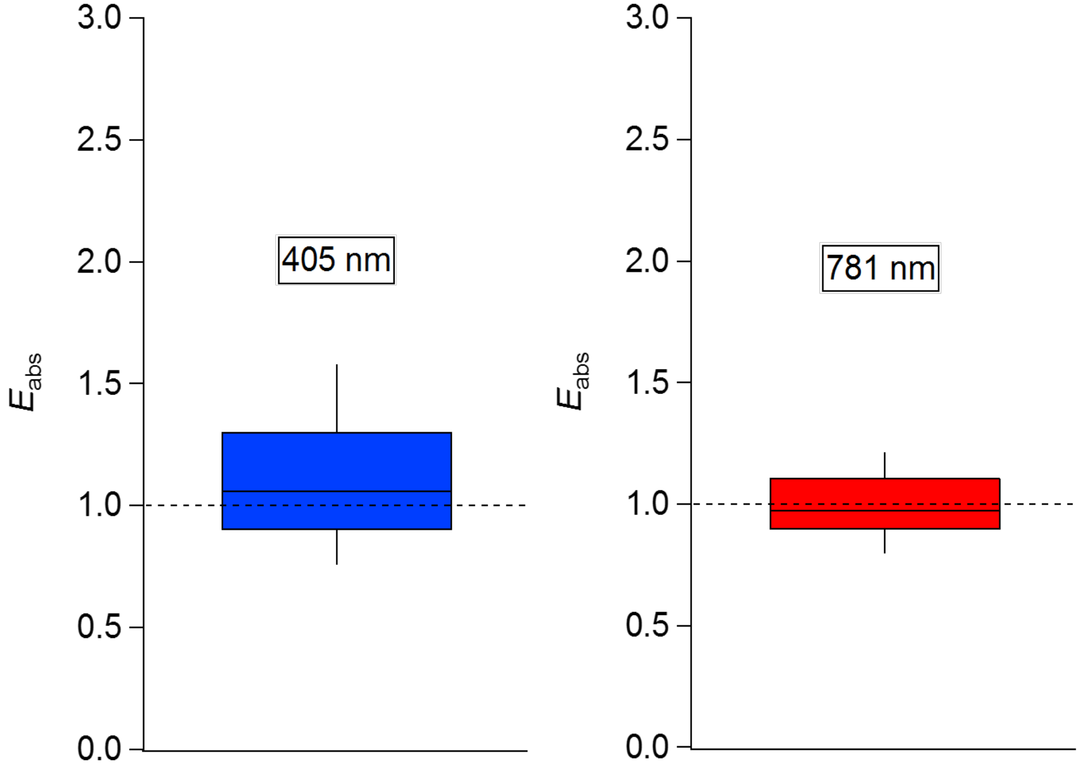
Healy et al., JGR (in review)

PASS b_{abs} vs SP-AMS BC (MAC values)



Absorption enhancement (E_{abs}) MAC approach



Mean enhancement at 405 nm is 12%, no enhancement at 781 nm

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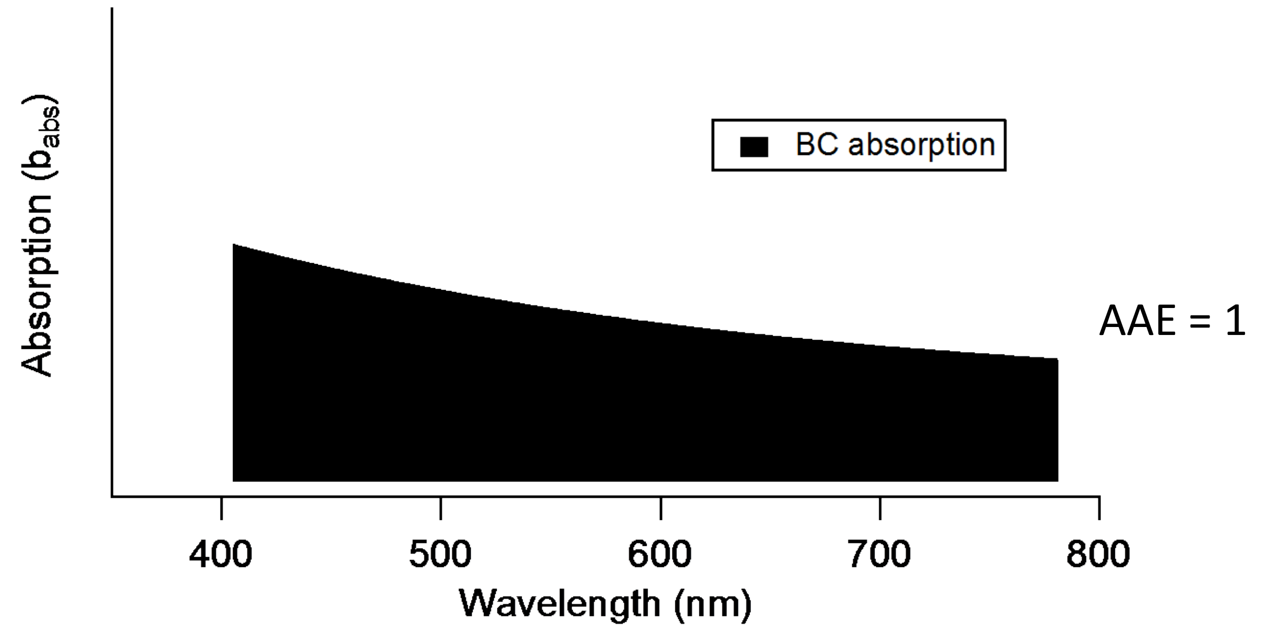
Why is the enhancement wavelength dependent?

Absorption Ångström exponent (AAE)

- Describes the wavelength dependence of aerosol absorption

$$AAE = -\frac{\log\left(\frac{b_{abs,\lambda 1}}{b_{abs,\lambda 2}}\right)}{\log\left(\frac{\lambda 1}{\lambda 2}\right)}$$



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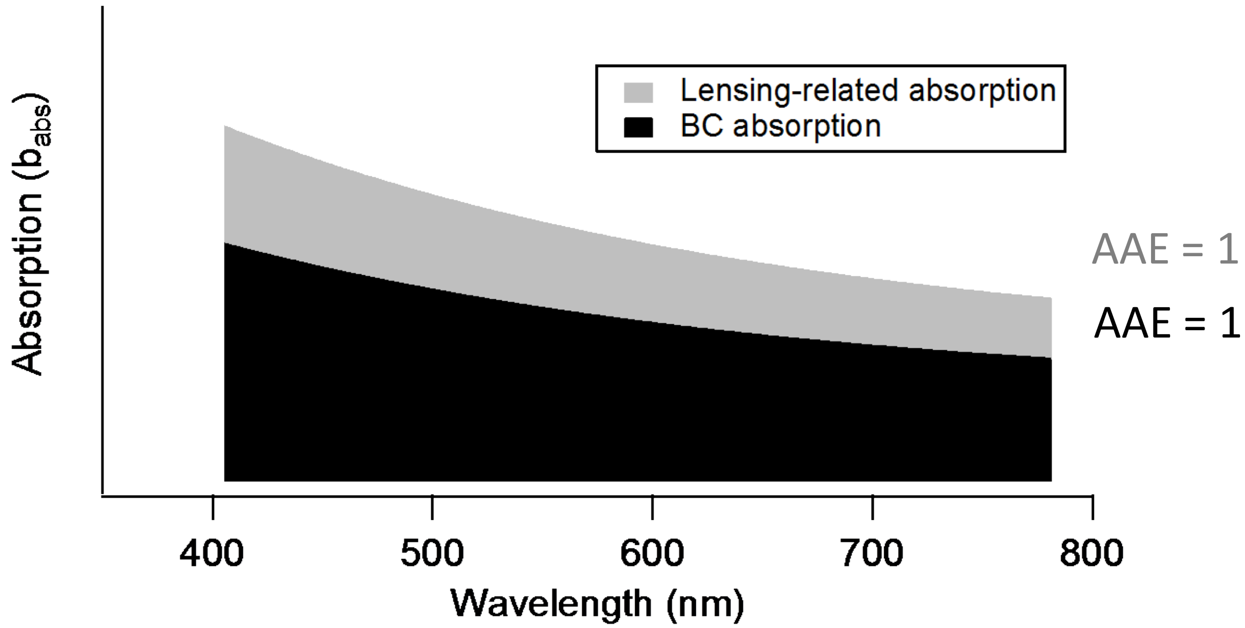
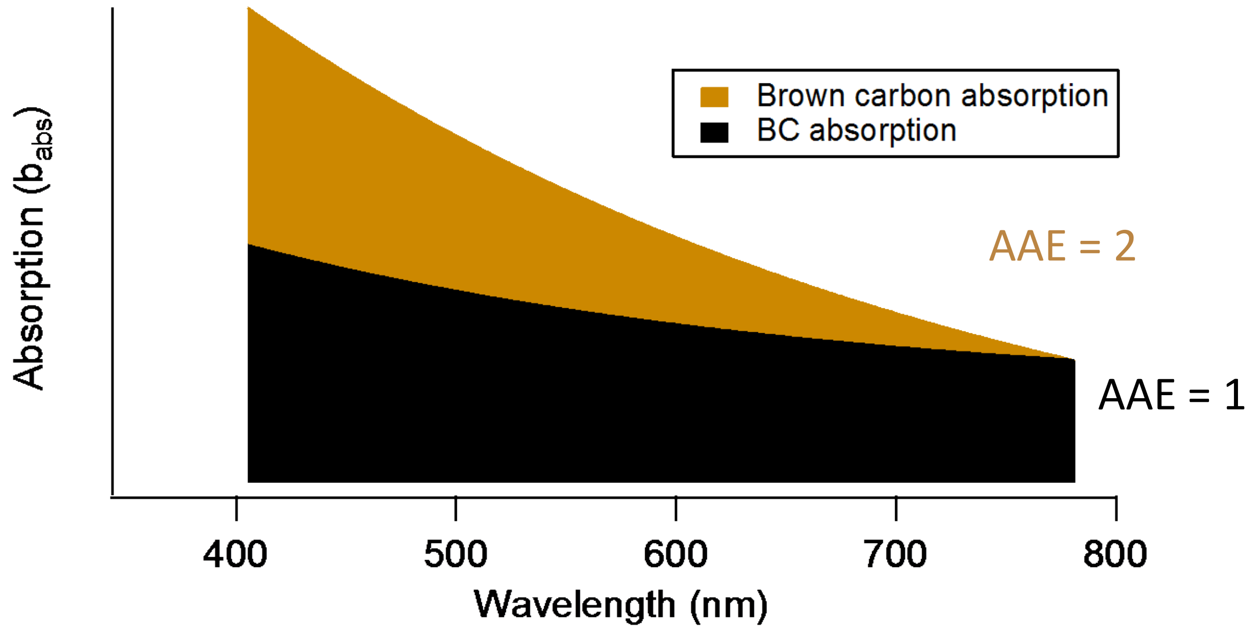
Absorption Ångström exponent (AAE)

- Impact of optical lensing absorption enhancement on AAE
- If lensing is occurring, we should observe an enhancement at 405 nm *AND* 781 nm



Absorption Ångström exponent (AAE)

- AAE of “brown carbon”, associated with biomass burning, is typically ~2 or greater
- Could explain the absorption enhancement being observed only at 405 nm



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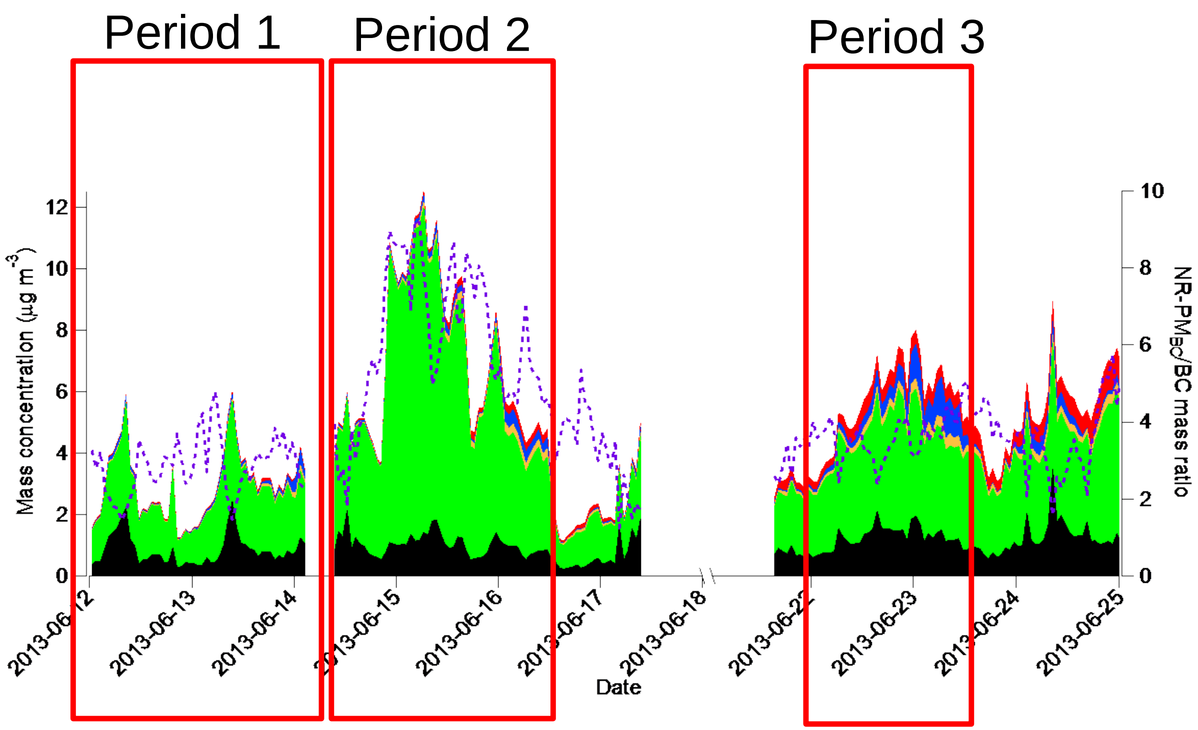
Dividing the campaign into periods of interest



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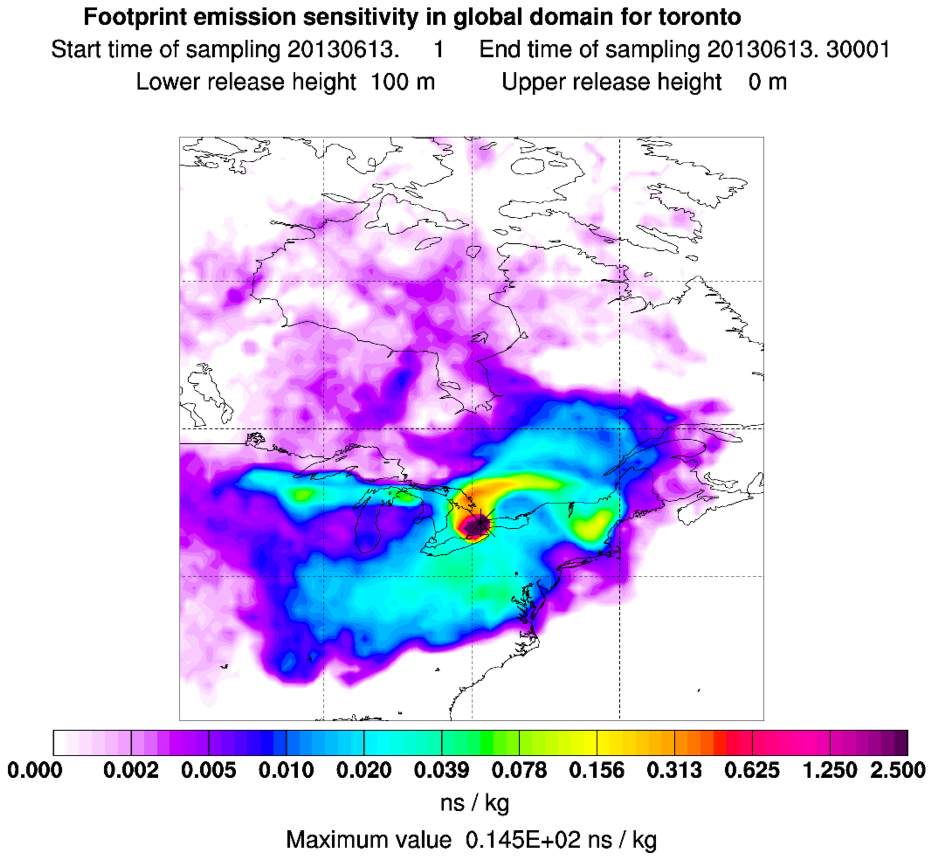
Dividing the campaign into periods of interest
Period 1



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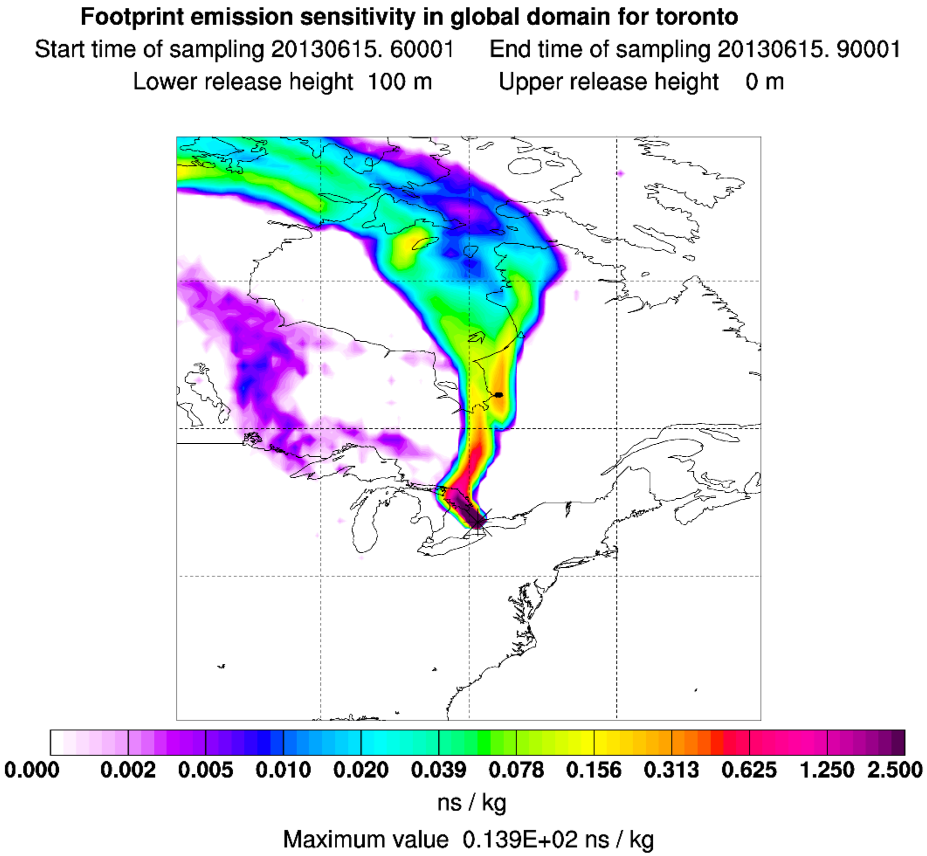
Dividing the campaign into periods of interest
Period 2



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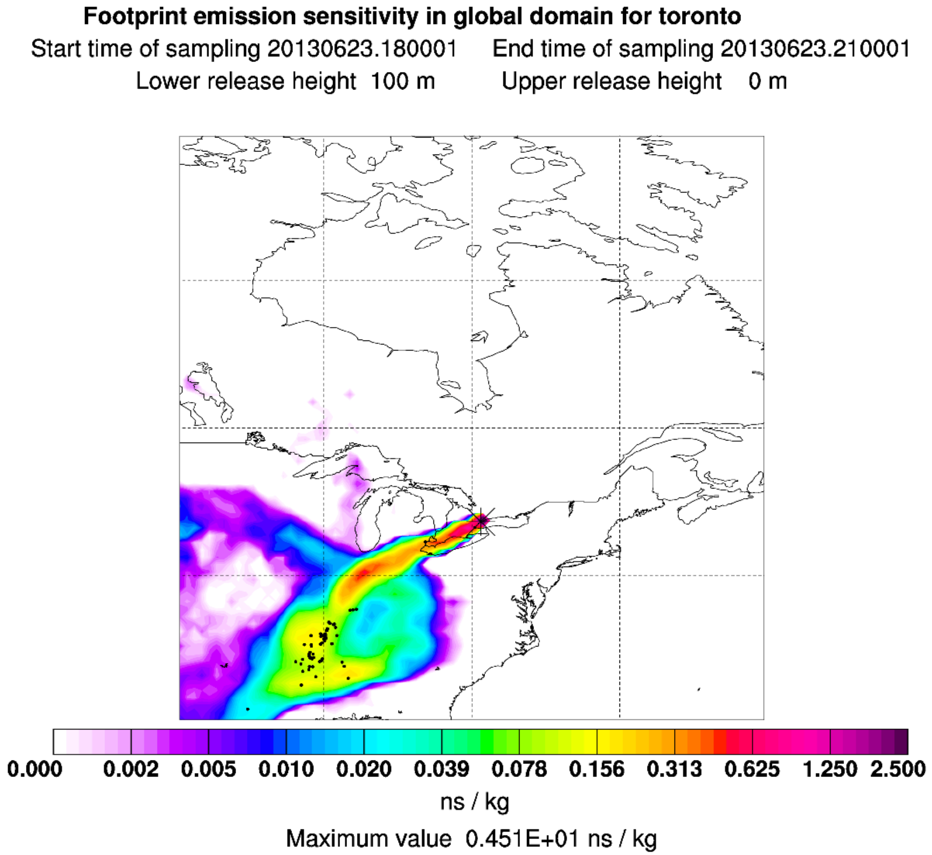
Dividing the campaign into periods of interest
Period 3



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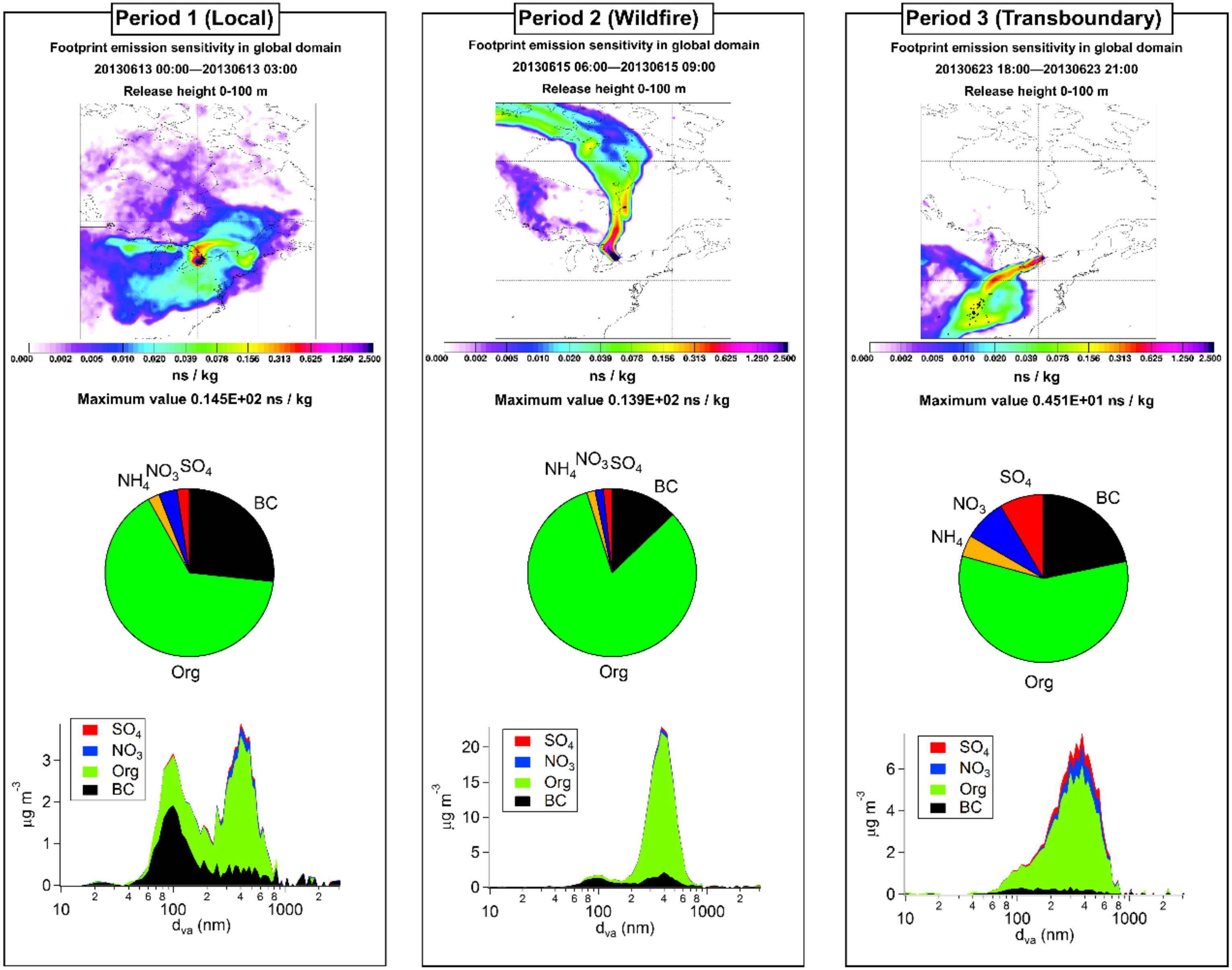
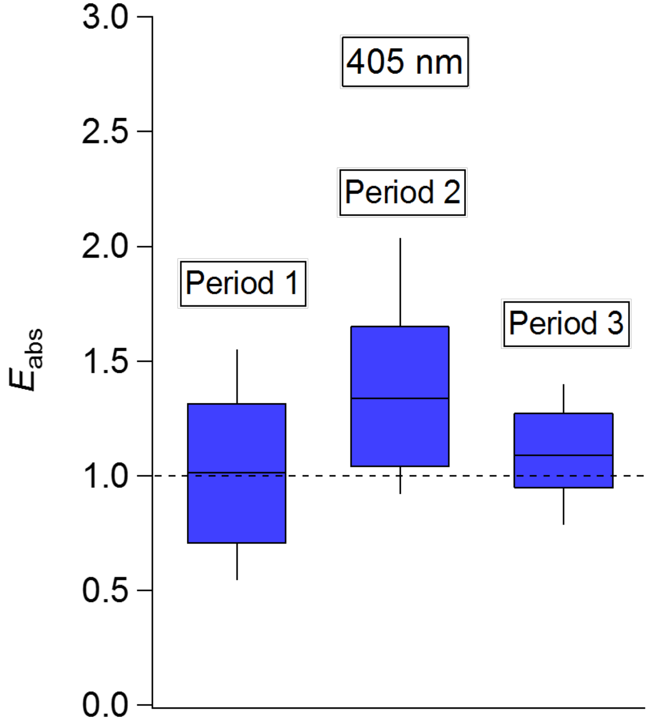
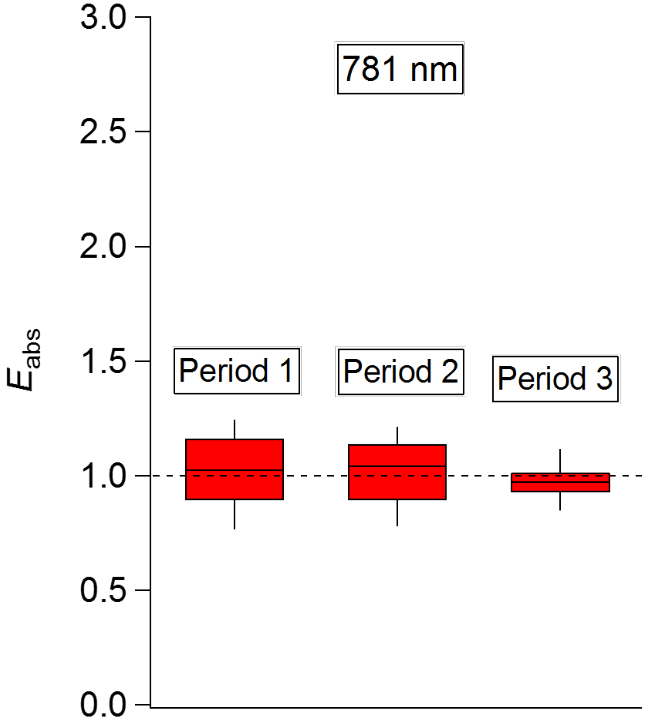


Period 1 (Local)
Footprint emission sensitivity in global domain
20130613 00:00—20130613 03:00
Release height 0-100 m
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.145E+02 ns / kg
NH4 NO3 SO4 BC Org
SO4 NO3 Org BC
μg m-3
dva (nm)
Period 2 (Wildfire)
Footprint emission sensitivity in global domain
20130615 06:00—20130615 09:00
Release height 0-100 m
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.139E+02 ns / kg
NH4 NO3 SO4 BC Org
SO4 NO3 Org BC
μg m-3
dva (nm)
Period 3 (Transboundary)
Footprint emission sensitivity in global domain
20130623 18:00—20130623 21:00
Release height 0-100 m
0.000 0.002 0.005 0.010 0.020 0.039 0.078 0.156 0.313 0.625 1.250 2.500
ns / kg
Maximum value 0.451E+01 ns / kg
SO4 NO3 NH4 BC Org
SO4 NO3 Org BC
μg m-3
dva (nm)

Absorption enhancement (E_{abs}) for each period





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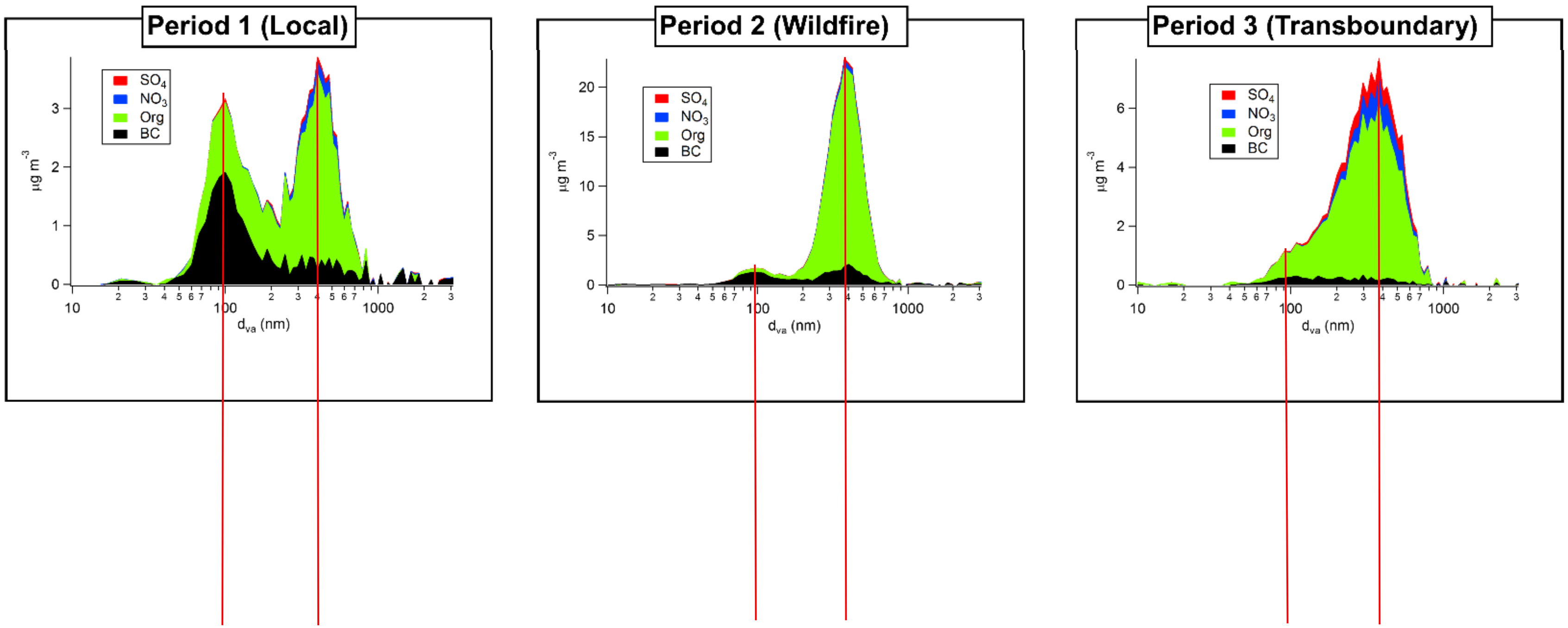
Mie theory predictions of lensing E_{abs} at 781 nm



1.1 2.0 1.1 1.9 1.2 2.0

By comparison we observe <u>1.0</u> for our campaign

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Conclusions

- Optical lensing is ***not relevant*** for BC in Toronto, even when the site is influenced by BC particles with large coatings

- Direct absorption by brown carbon ***is relevant*** when the site is influenced by wildfires, however, and is responsible for over 50% of direct absorption at 405 nm at times

- If radiative forcing is estimated using composition data and Mie theory, regional warming is substantially overpredicted

- Care must be taken when accounting for BC internal mixing in climate models, and further studies of absorption enhancement in other environments are necessary

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CHEMBC outcomes and future work

- Quantitative black carbon mixing state measurements using the ATOFMS and SP-AMS provide better input data for particle resolved models (Healy et al., ACP 2013, Lee et al., ACP 2015)

- A new approach for describing black carbon chemical mixing state through diversity measures (Healy et al., ACP 2014)

- Quantitative data for individual black carbon particles can be used to better predict hygroscopic growth and new cloud formation (Healy et al., JGR 2014)

- The next step is to develop a library of single particle mass spectral composition data from a variety of global field campaigns

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Thank you

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S. Eckhardt, A. Stohl

University College Cork
J.C. Wenger

MARIE CURIE ACTIONS

Questions?

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SUPPLEMENT

Aerosol absorption and the Beer-Lambert Law

$$I = I_0 e^{-\alpha l c}$$

I = outgoing light
I_0 = incident light
α = absorption cross section (m^2 g^{-1}) (α is often termed the “MAC” value)
l = path length (m)
c = concentration (g m^{-3})

b_{abs} is the product of α and c (m^{-1})
and is measured directly by the PASS

Scattering (and extinction) calculations are analogous to absorption

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Terminology

BC (black carbon, measured quantitatively by SP-AMS)

$\boldsymbol{b_{abs}}$ (absorption coefficient measured by PASS)

$\boldsymbol{E_{abs}}$ (absorption enhancement)

MAC (mass absorption cross-section)

NR-PM$_{BC}$ (non-refractory particulate matter on BC, measured by SP-AMS)

NR-PM$_{BC}$/BC (coating-to-core mass ratio for BC-containing particles)

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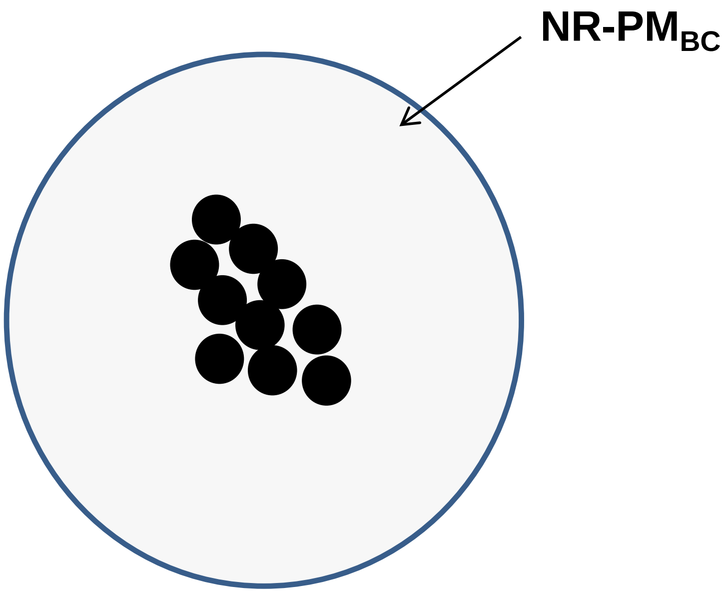
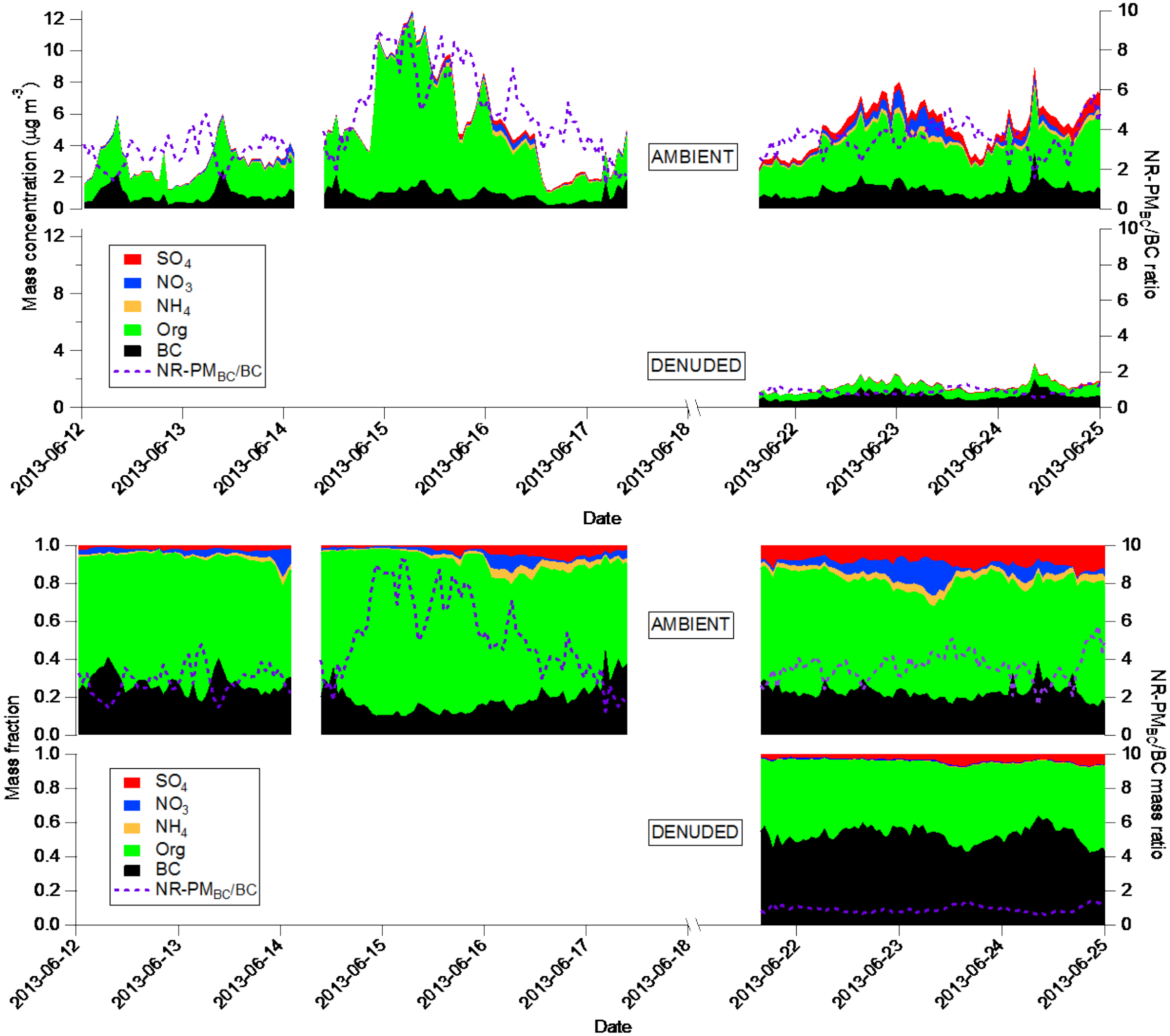
Terminology



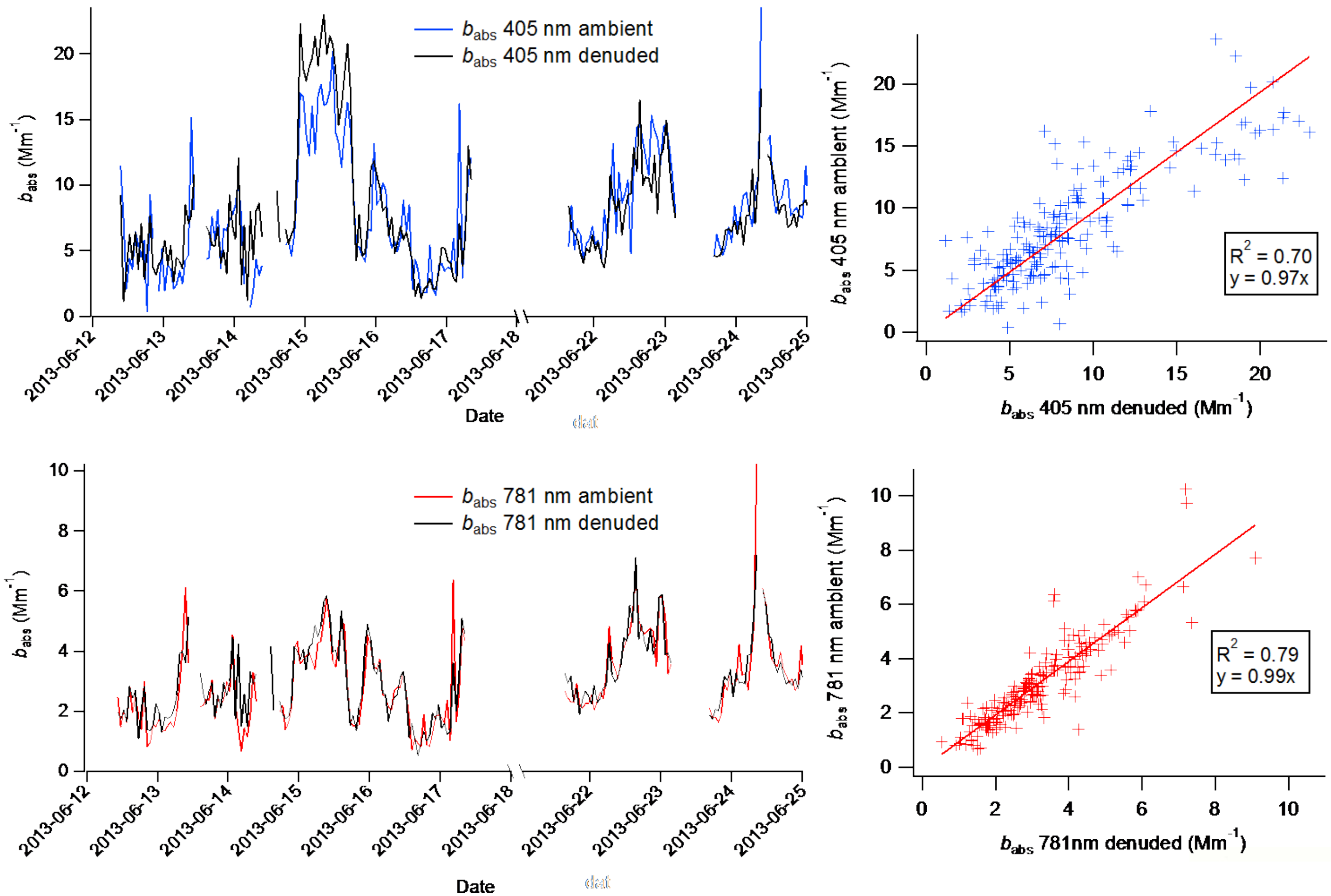
NR-PM$_{BC}$/BC is the coating-to-core ratio

Results: APPROACH 1: BC-containing particle composition

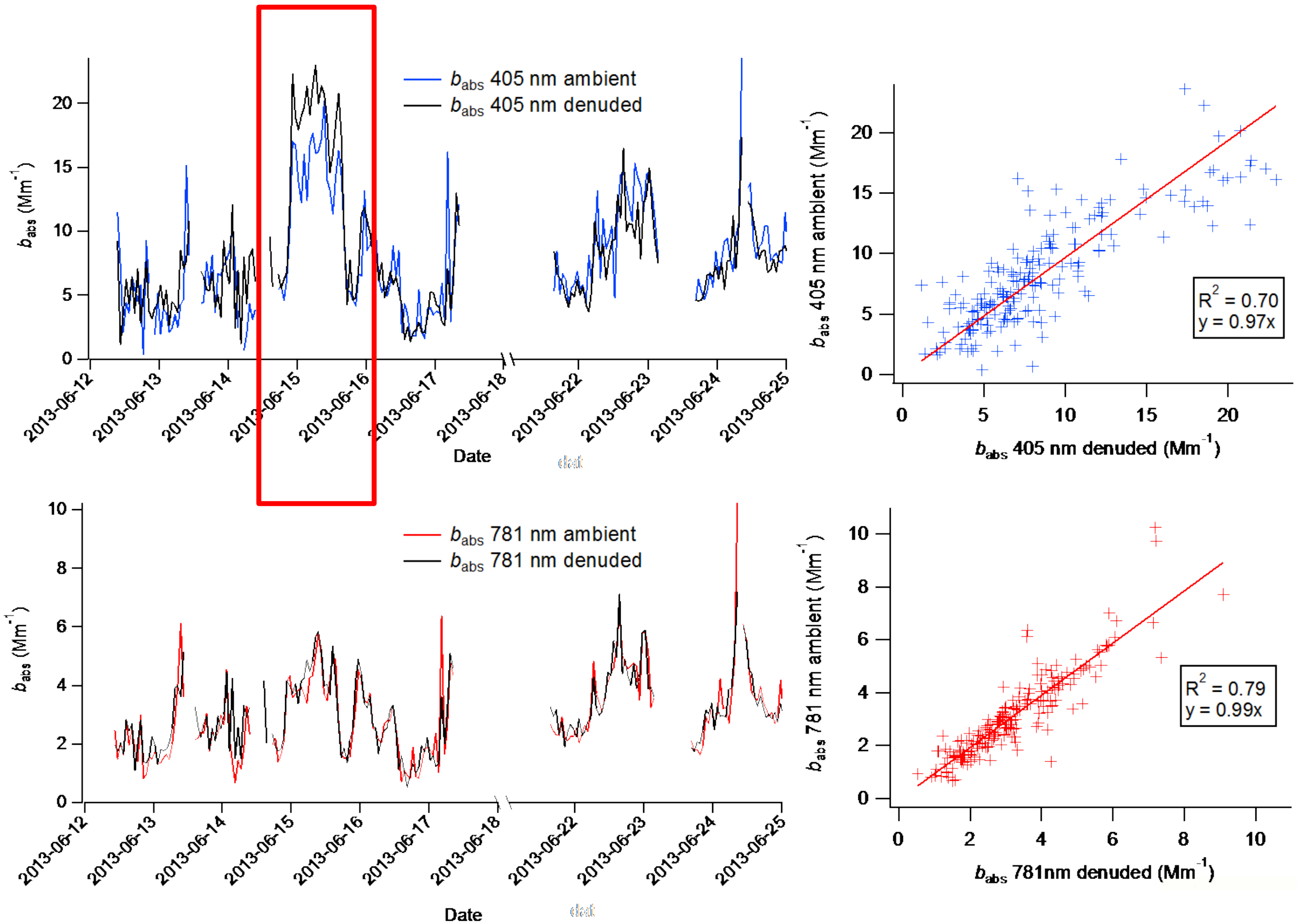


Healy et al., JGR (in review)

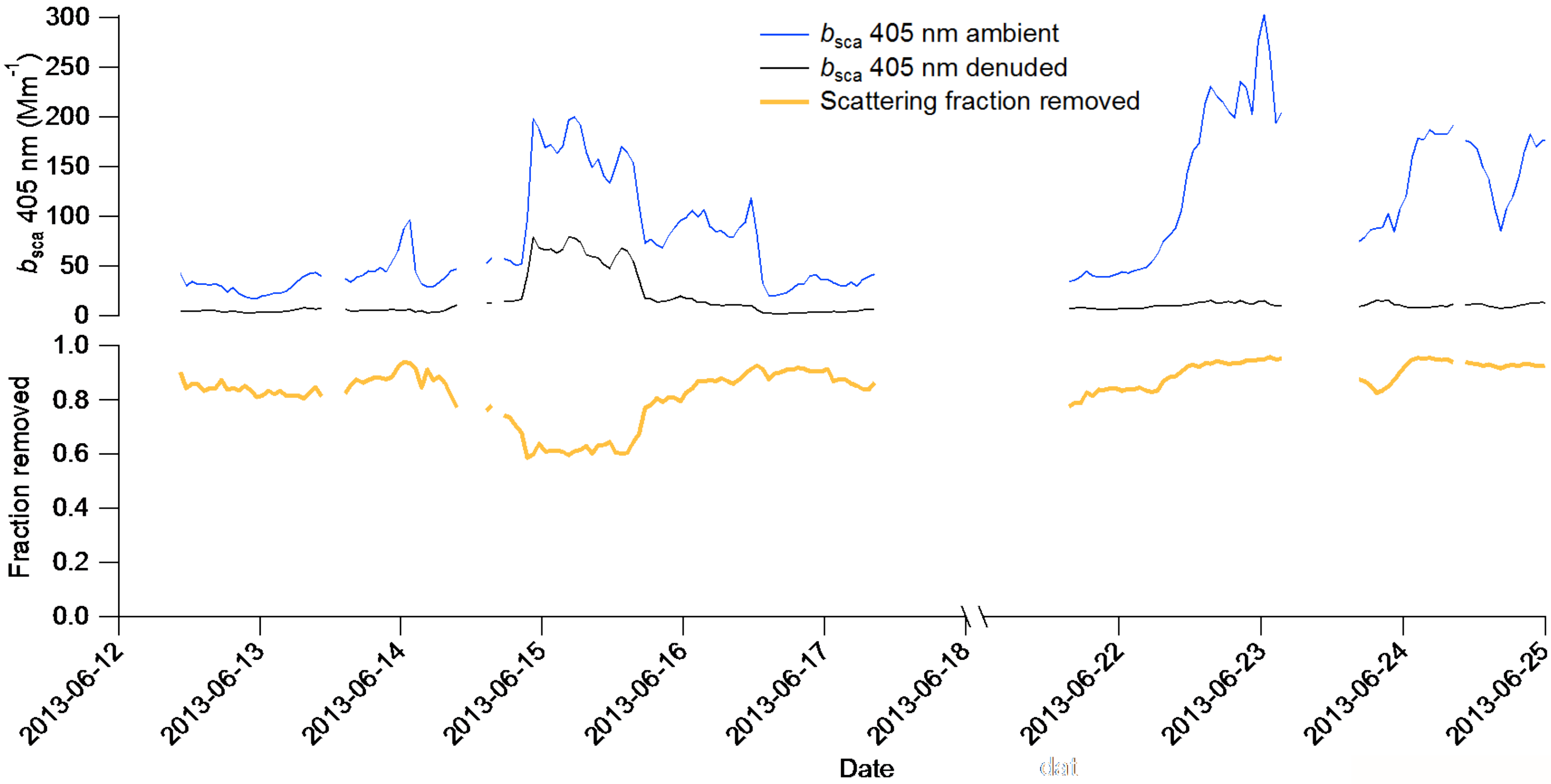
Results: APPROACH 1: Aerosol absorption (b_{abs})



Results: APPROACH 1: Aerosol absorption (b_{abs})



APPROACH 1: Aerosol scattering (b_{sca})



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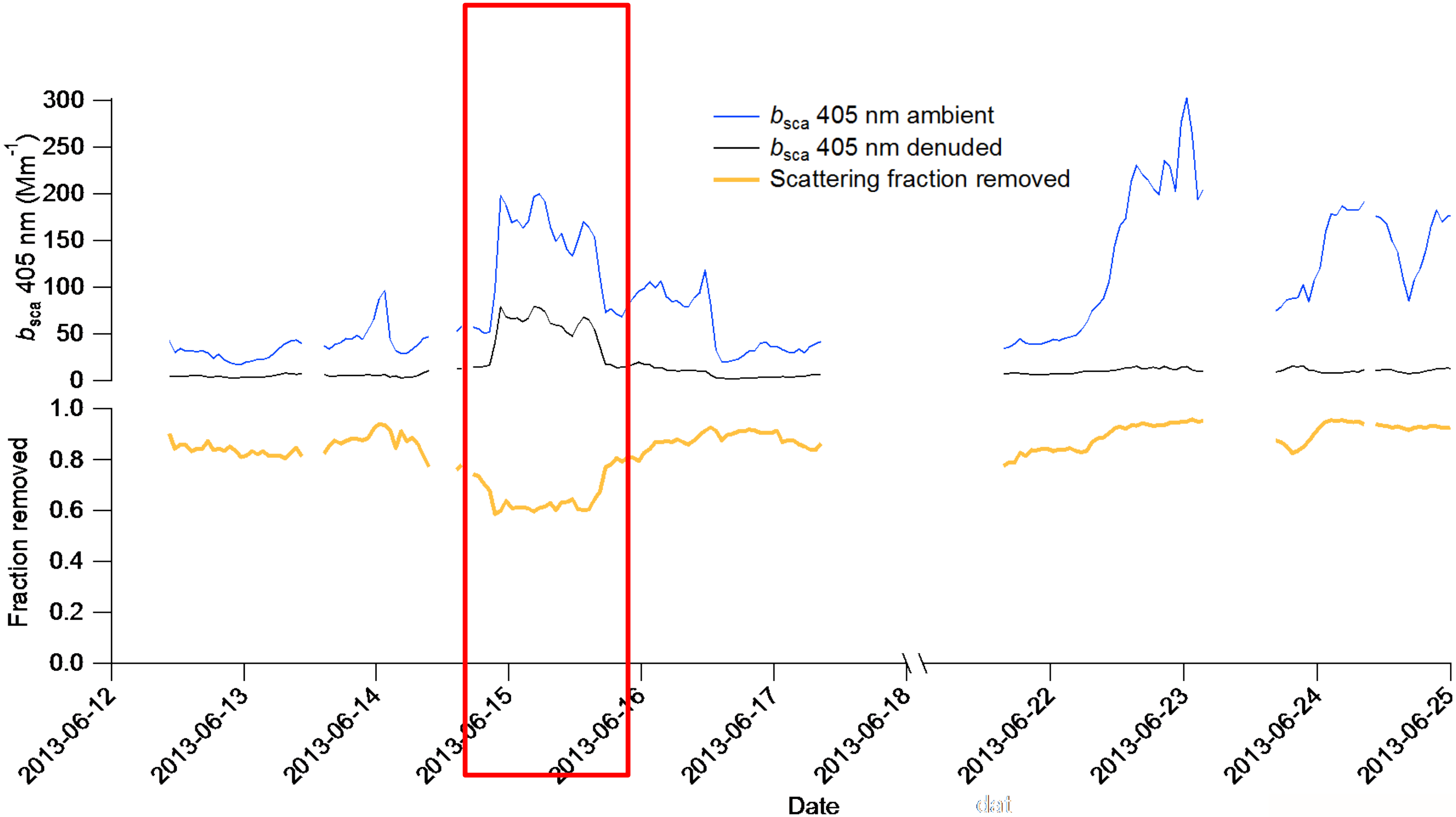
APPROACH 1: Aerosol scattering (b_{sca})



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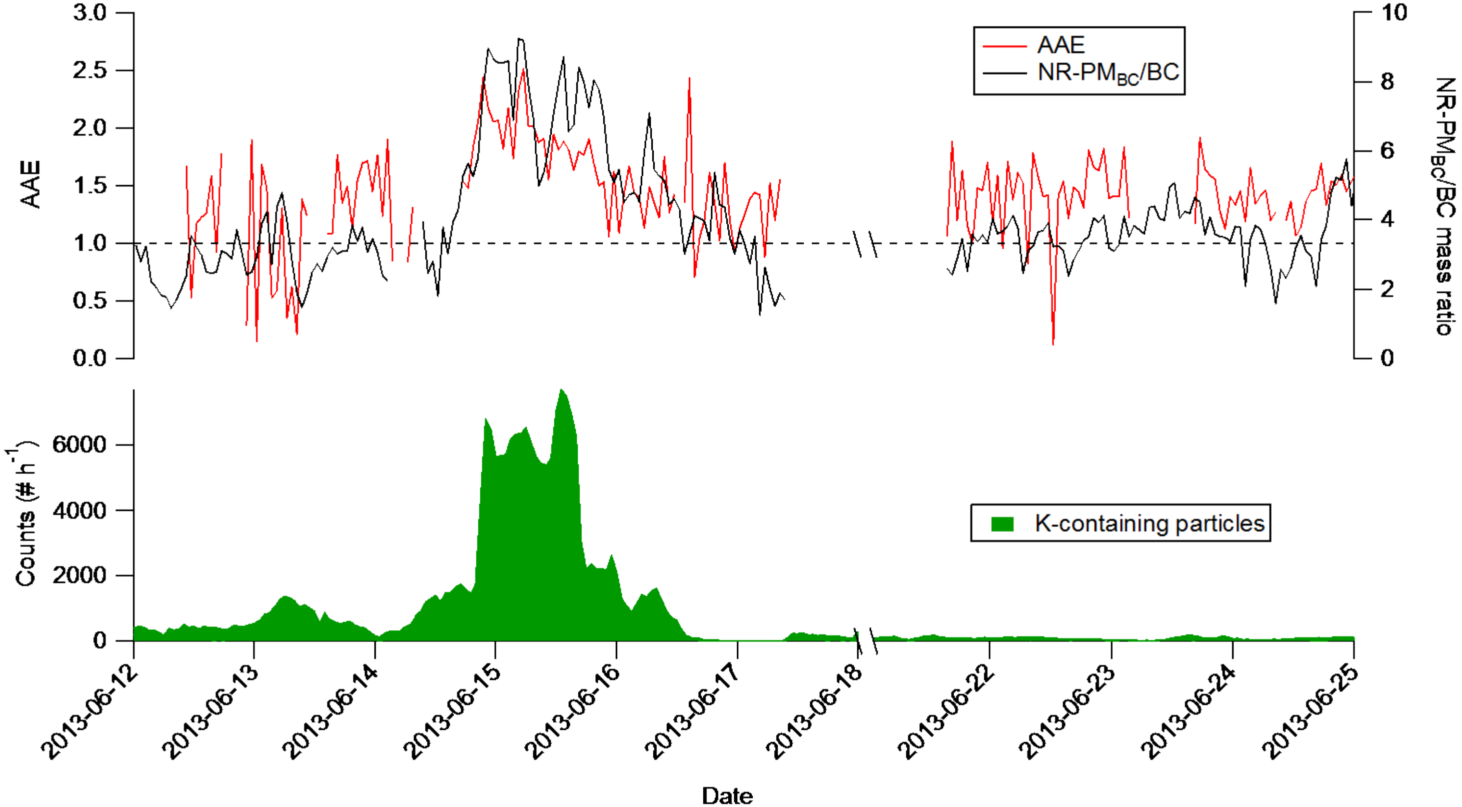
AAE, BC coating to core ratio, ATOFMS data



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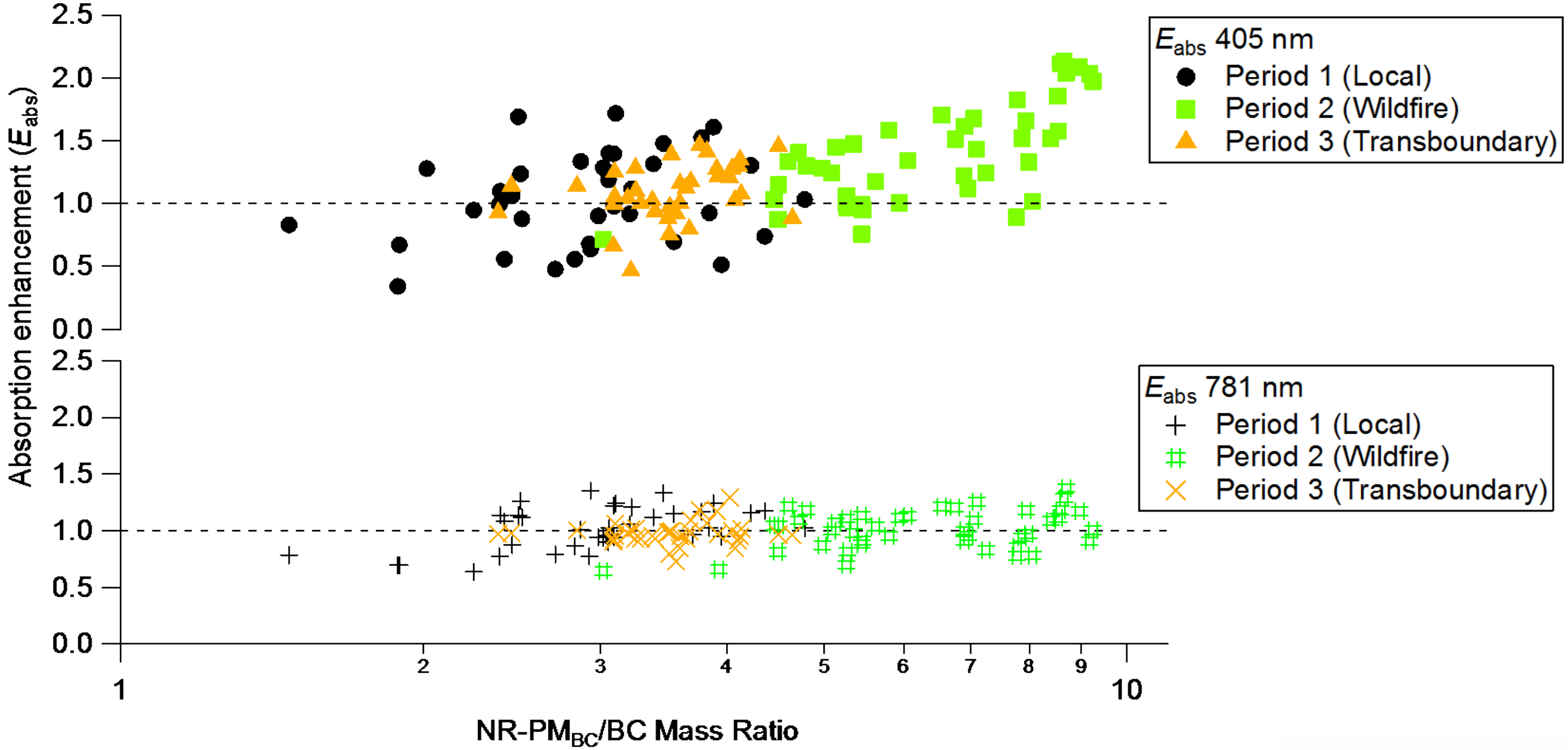
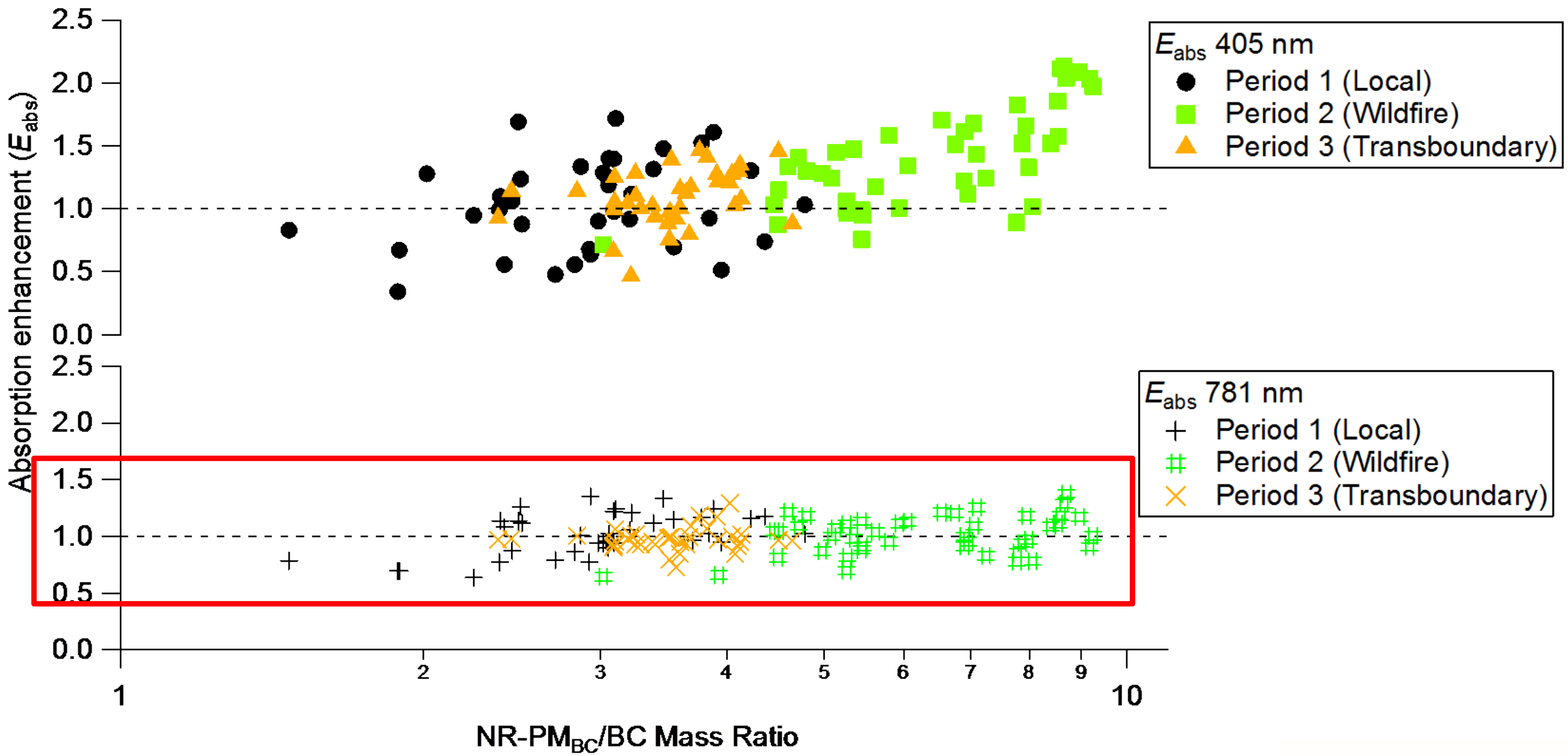
Absorption enhancement (E_{abs}) for each period



Absorption enhancement (E_{abs}) for each period



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