

# Black and Brown Carbon – local, regional and global relevance

WeBaSOOP, 3 Apr 2024

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# Thanks to co-workers!

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# BC and CM – two sides of the same coin

## Black Carbon

### BC

Primary,  
sources



## Carbonaceous matter

### CM

Primary and  
secondary,  
contribution to  
PM2.5



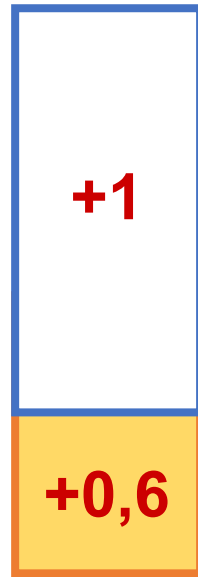
# A note on the terminology

- **Black Carbon mass concentration**  $BC$   $\mu\text{gm}^{-3}$ 
  - Optical meas. **equivalent** to mass **eBC** (filter photometers)
  
- **Aerosol absorption coefficient**  $b_{abs}$   $\text{Mm}^{-1}$ 
  - Optical measurements, in-situ, (in)direct

$$b_{abs} = BC \cdot MAC$$

# BC, absorption, heating

**+1,6 W/m<sup>2</sup>**

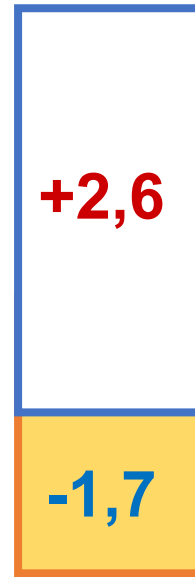


**CO<sub>2</sub>**

atmosphere

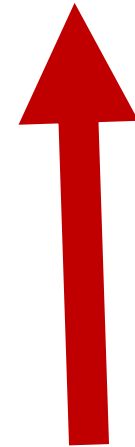
surface

**+0,9 W/m<sup>2</sup>**



**BC  
(direct)**

BC total:  
**+1,1 ± 0,7 W/m<sup>2</sup>**



**Oops!**

Bond et al., 2013



over Malaysia

BC @ ~10.000 ft

over Congo river



# Sources



# Regional pollution



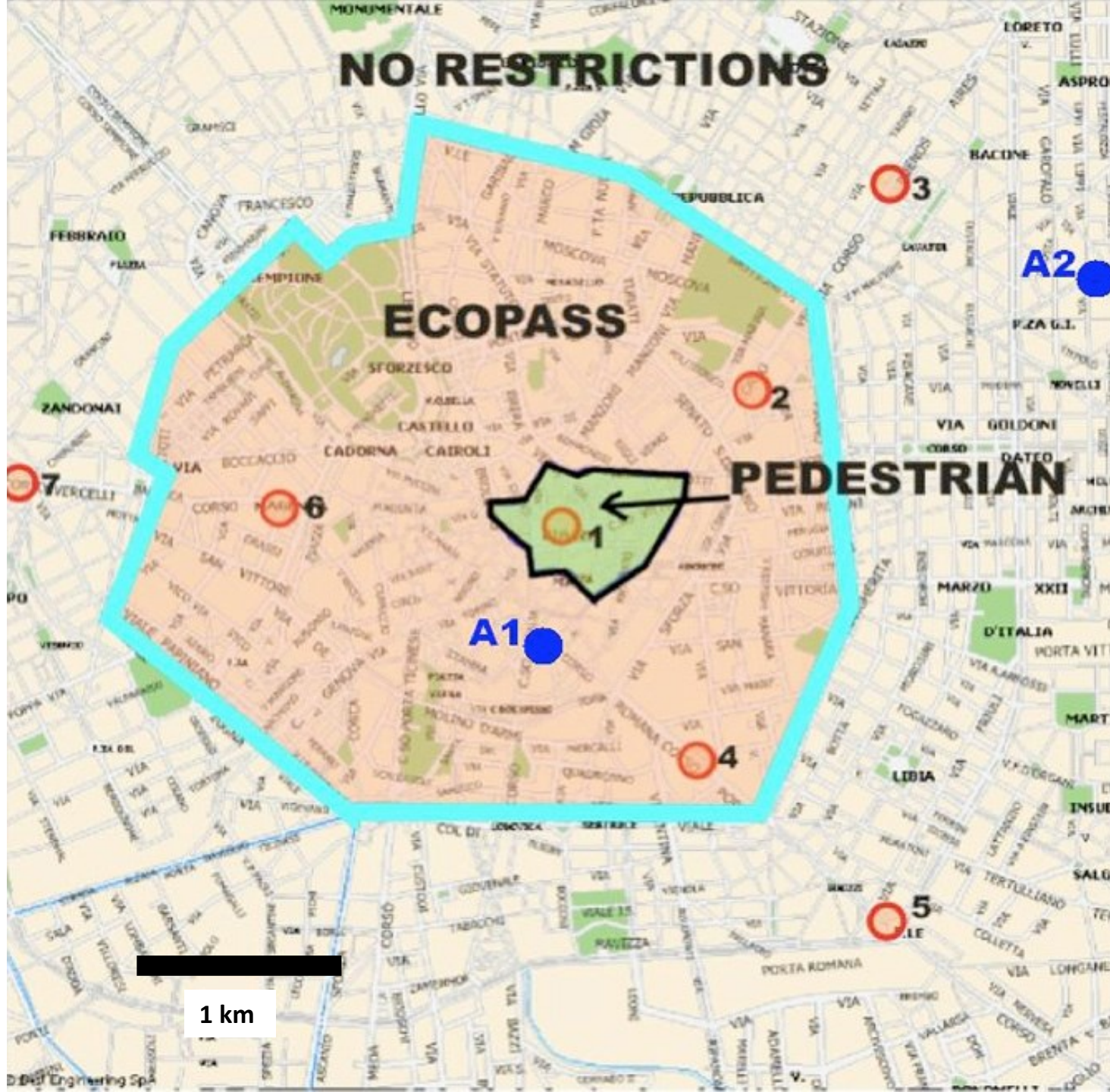


# Milano, Italy

- largest city in N Italy, in a valley
- 1.3 M inhabitants
- one of the most polluted cities in Europe
- major traffic in the Po valley
- traffic: 89,000 vehicles in the city center every day
- traffic restriction in place since Jan 2008



# Milano



## Radial measurements

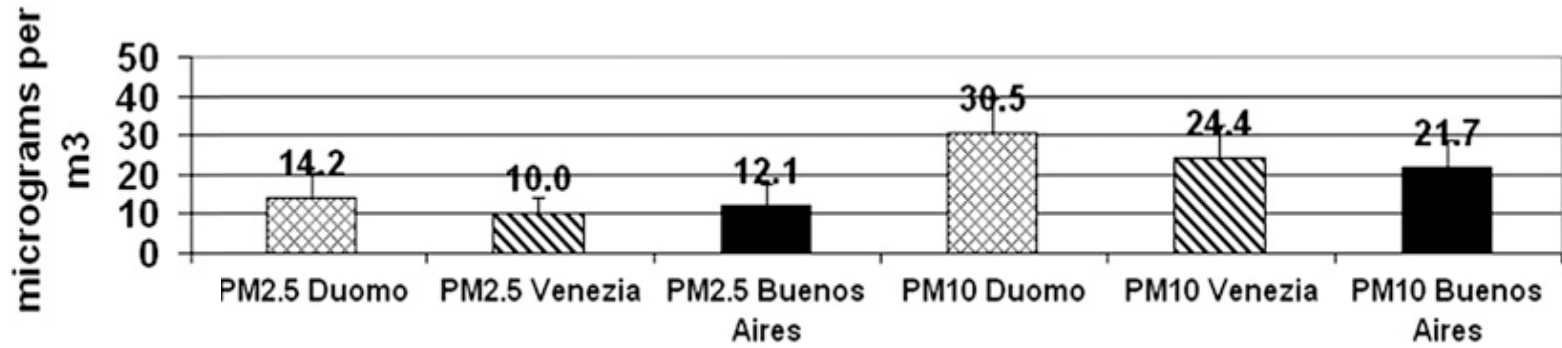
Each site:

- PM10, PM2.5, PM1
- Black Carbon
- RH, T

08:30 – 19:30

Summer:  
July 2010

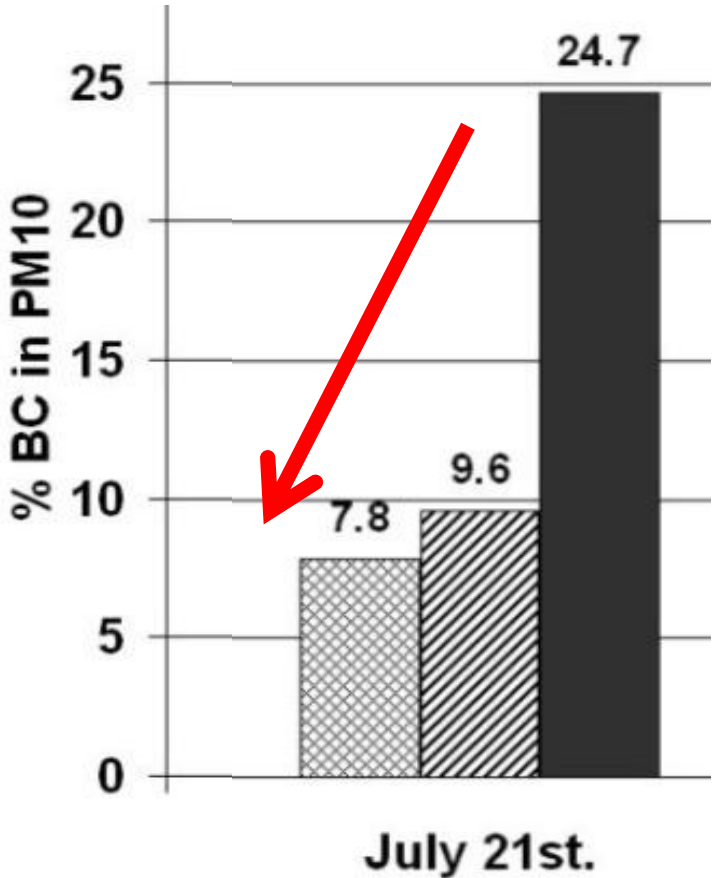
T = 30 C



**No difference!**

**PM**

**Big difference!**

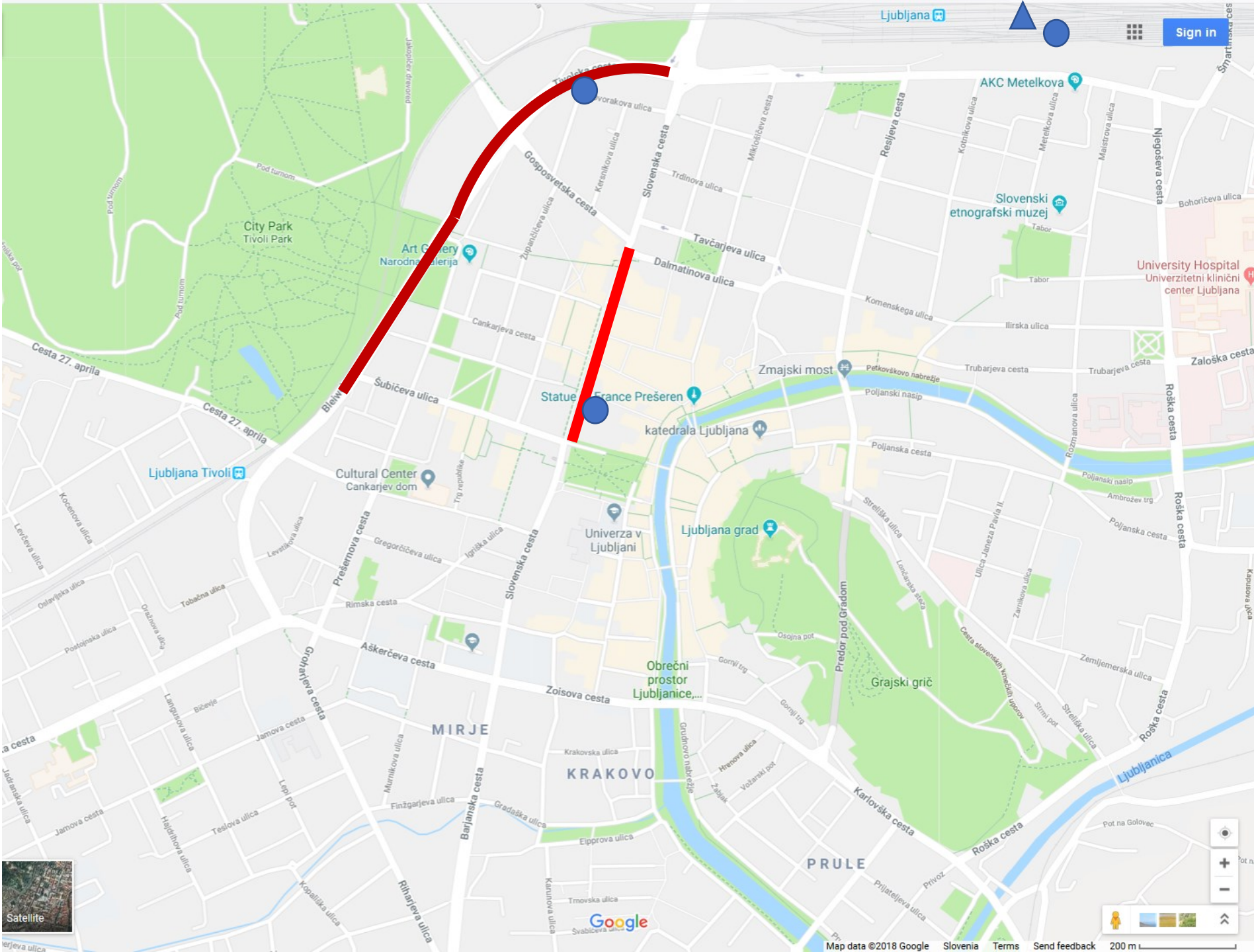


**BC/PM**

# Ljubljana, Slovenia



- largest city in Slovenia, in a basin
- 350k inhabitants
- traffic: 120,000 vehicles into the city every day
- traffic restriction in place since Sept 2013



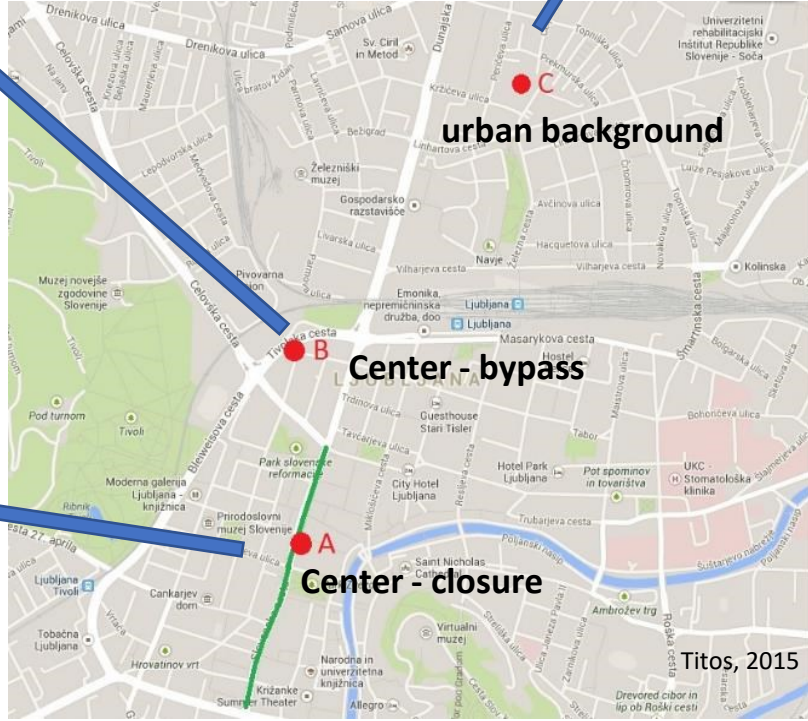
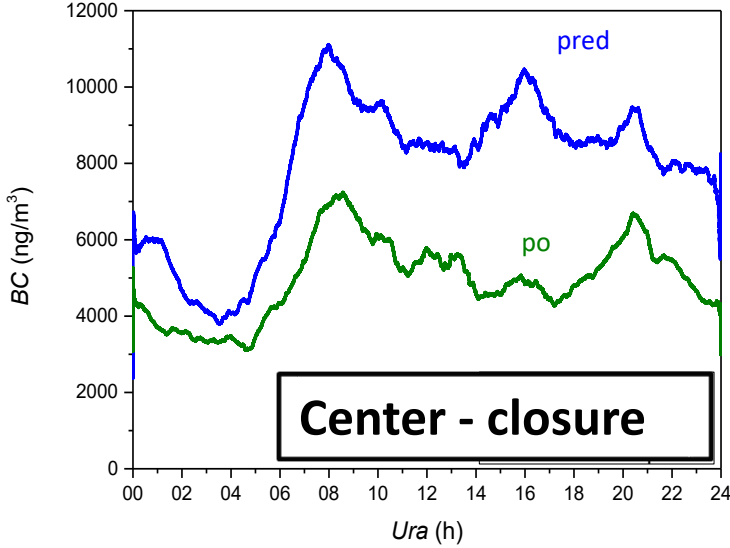
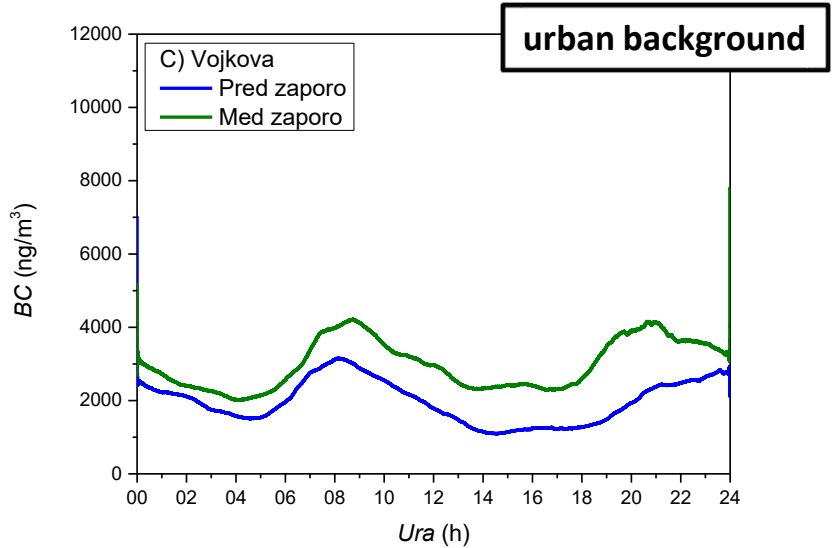
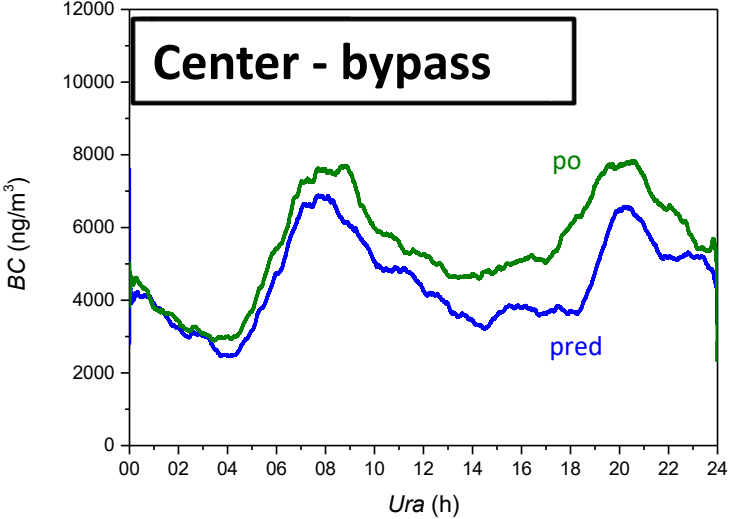
Sign in



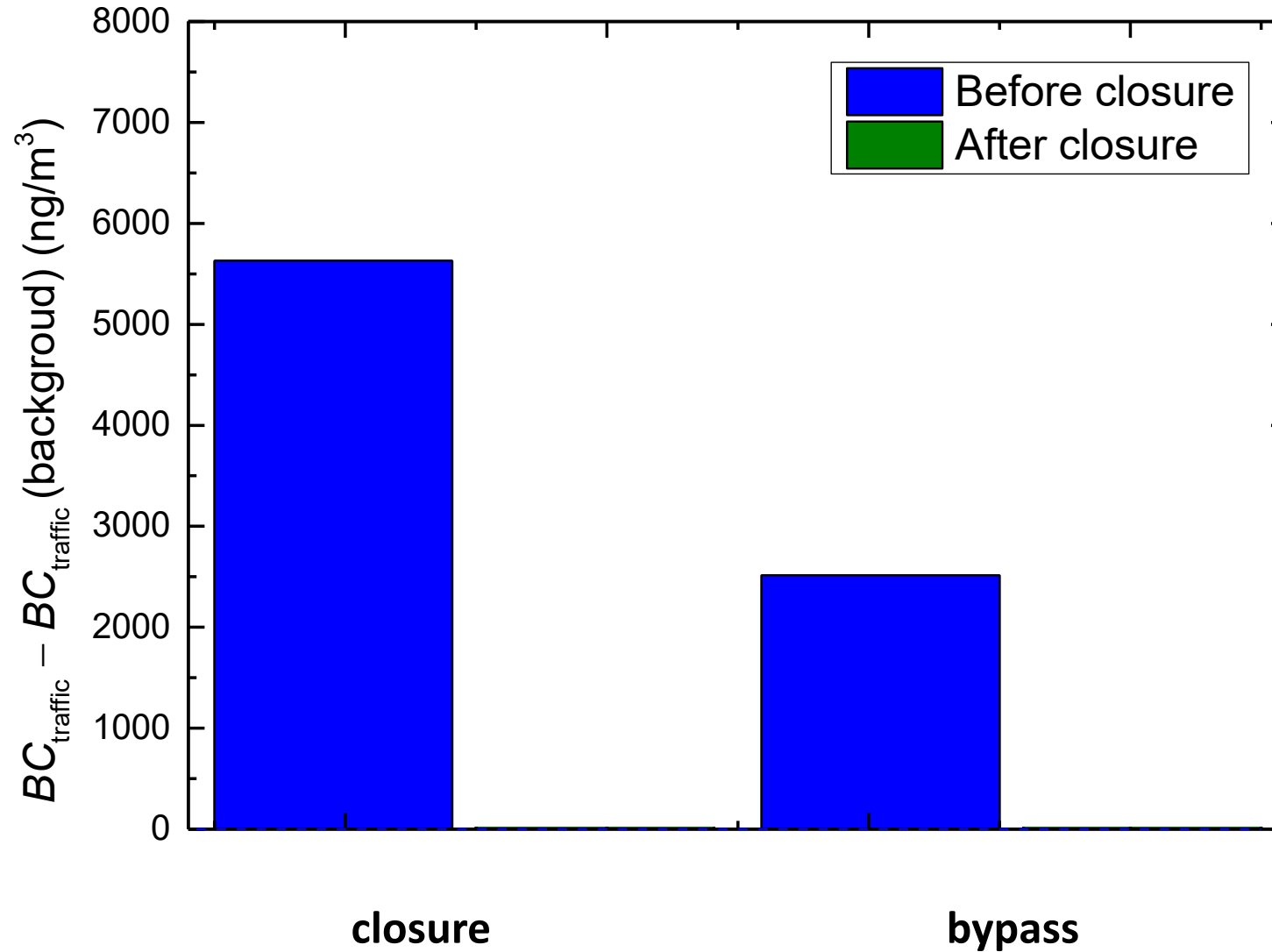
Satellite

Google

# Ljubljana (Aug – Oct 2013)



# Ljubljana ( $BC_{tr}$ : city-background)



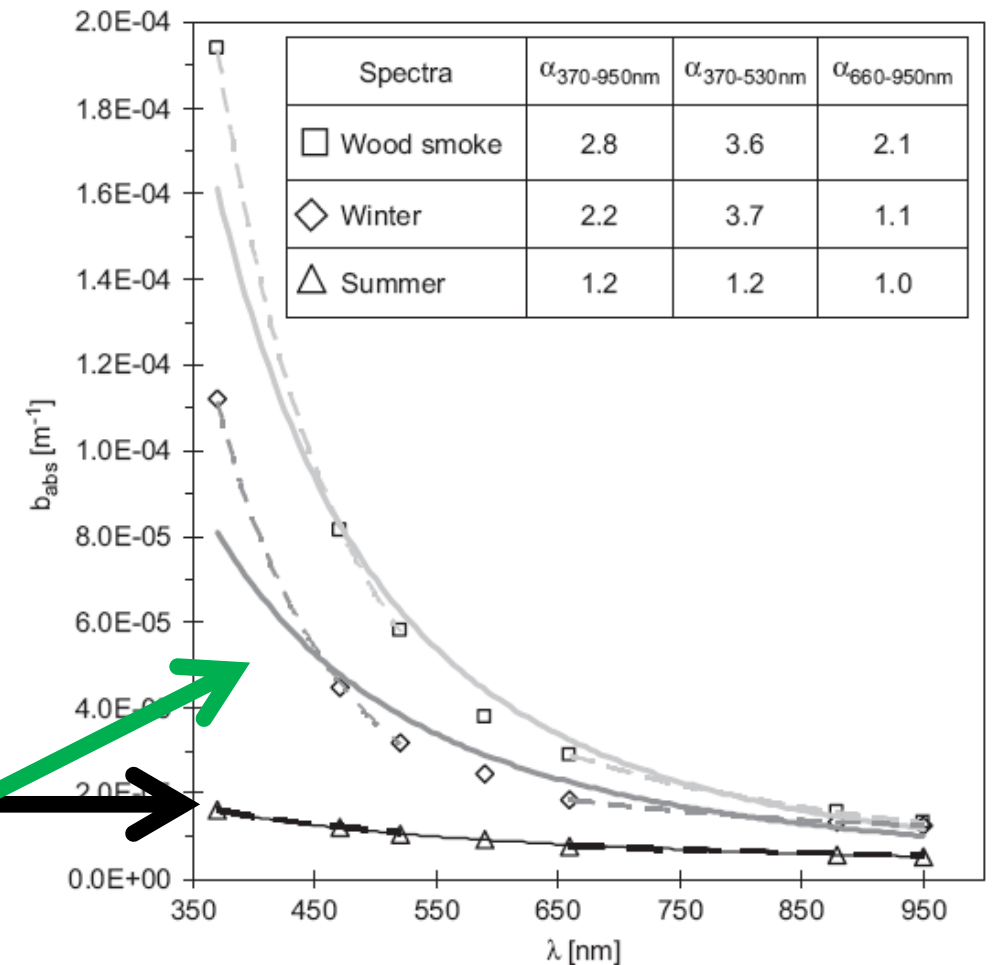
# Optical properties - source apportionment

- measure attenuation with the Aethalometer
- absorption coefficient -  $b_{abs}$
- for pure black carbon:  $b_{abs} \sim 1/\lambda$
- generalize **Angstrom exponent**:

$$b_{abs} \sim 1/\lambda^{AAE}$$

**diesel:**  $AAE \approx 1$

**biomass-smoke:**  $AAE \approx 2$  and higher



# Quantification – sources of BC - Sandradewi/Aethalometer model

$$b(\lambda) = b_{bb}(\lambda) + b_{ff}(\lambda)$$

$$\lambda = 470 \text{ nm}, 950 \text{ nm}$$

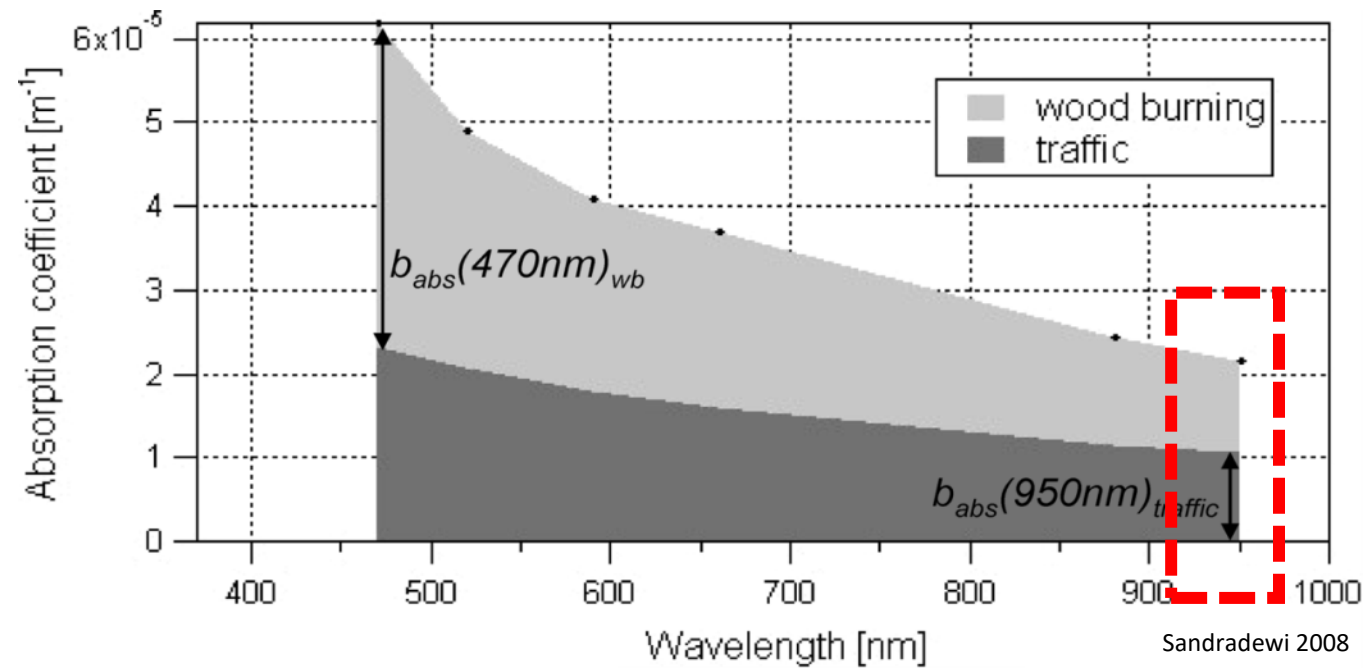
$$b_i(470 \text{ nm}) / b_i(950 \text{ nm}) = (470 \text{ nm} / 950 \text{ nm})^{-AAE_i}$$

$$AAE_{ff} = 1,0 \pm 0,1 \text{ (fossil)} \quad \text{Bond \& Bergstrom 2004}$$

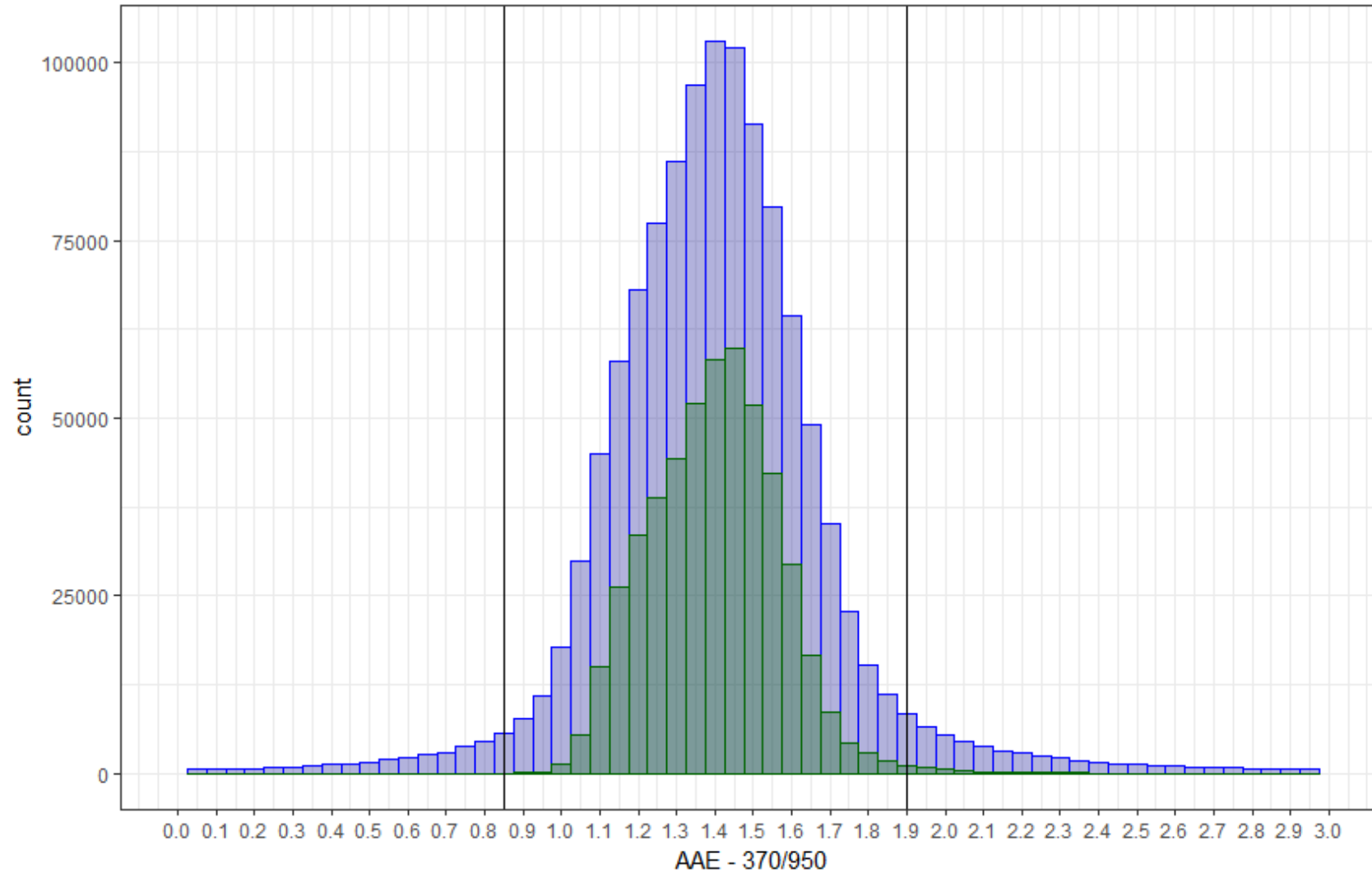
$$AAE_{bb} = 2,0 - 0,5 / +1,0 \text{ (wood)} \quad \text{Kirchstetter 2004,}$$

Day 2006,  
Lewis 2008

Zotter et al.: Atmos.  
Chem. Phys., 17,  
4229–4249, 2017  
**14C**



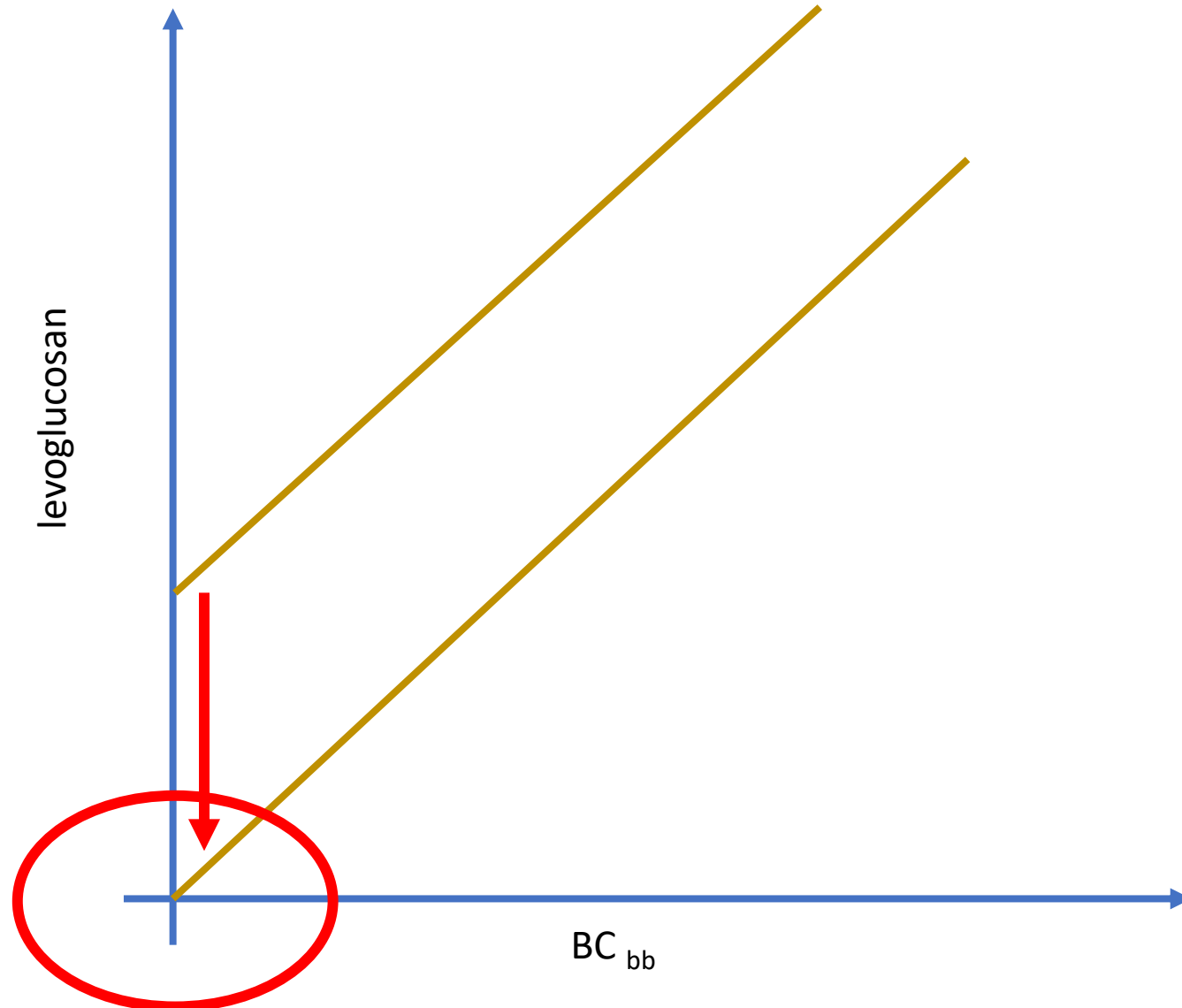
# AAE pair – $BC_{ff}$ $BC_{bb}$



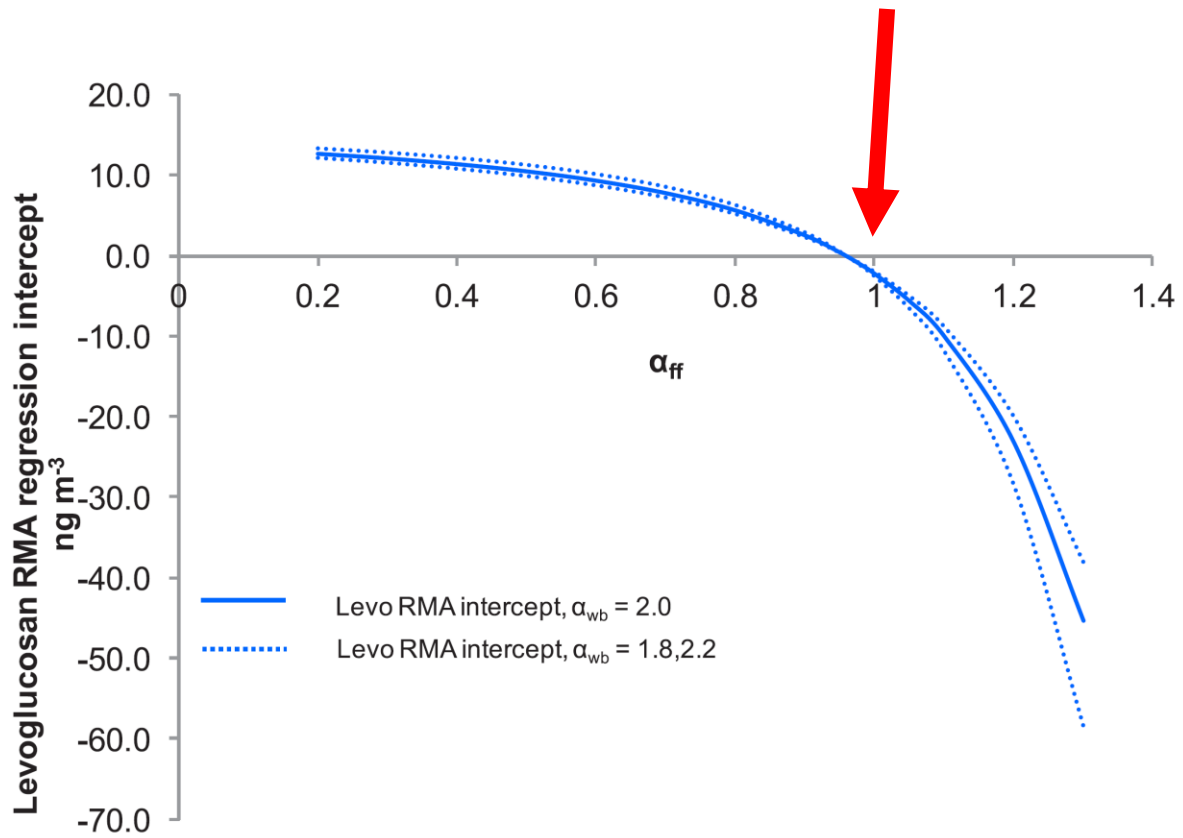
$AAE_{ff}$

Fuller et al., 2013

<http://dx.doi.org/10.1016/j.atmosenv.2013.12.037>

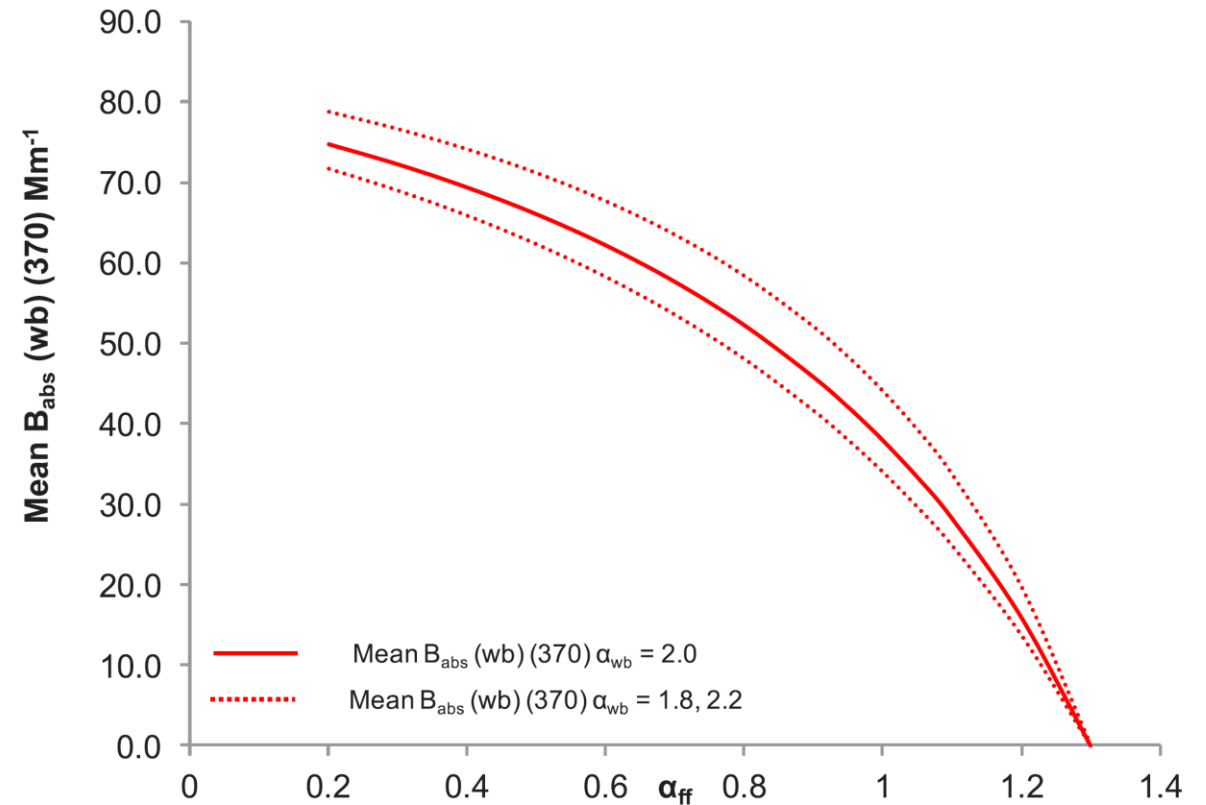


# AAE<sub>ff</sub>



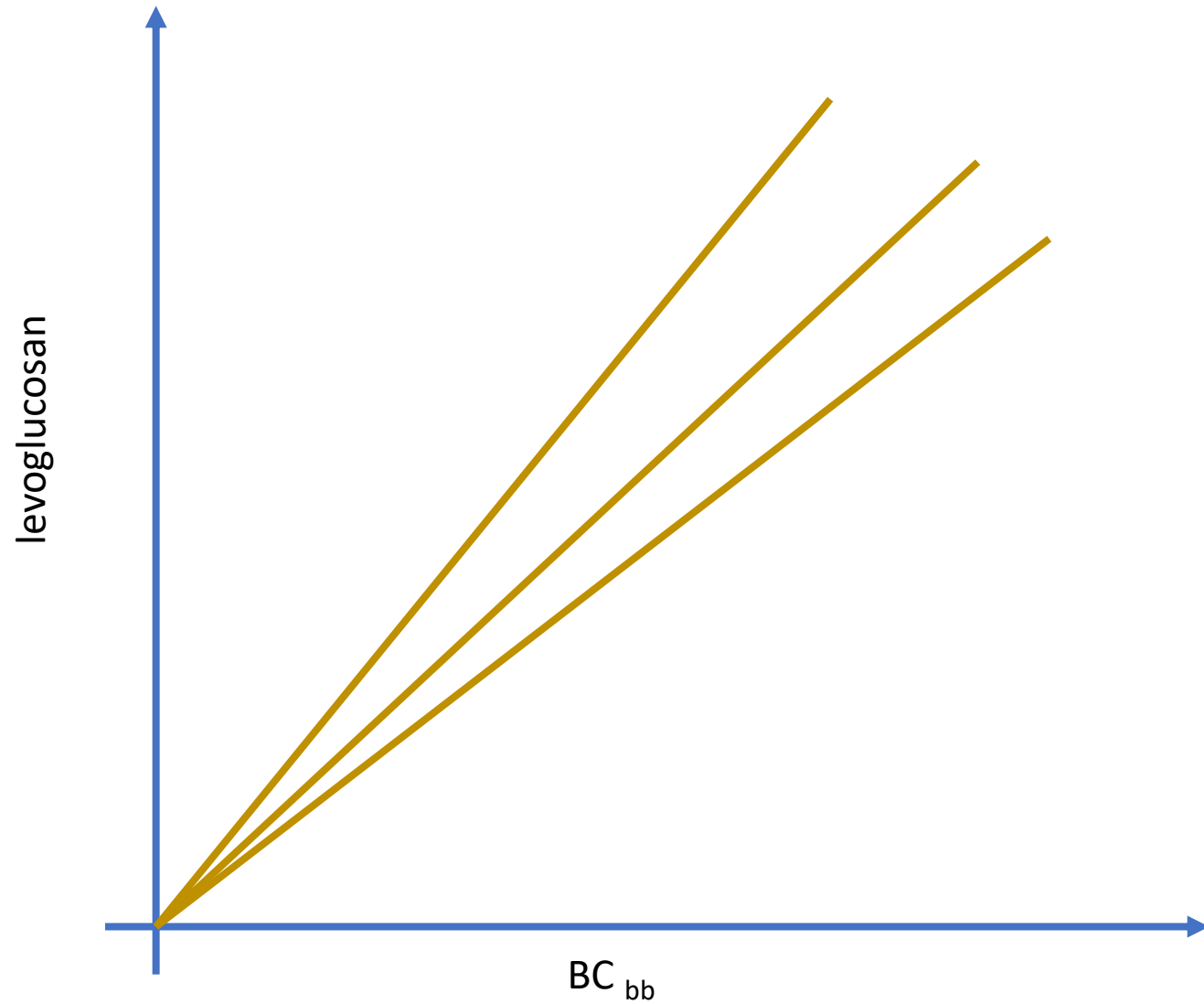
Fuller et al., 2013

<http://dx.doi.org/10.1016/j.atmosenv.2013.12.037>

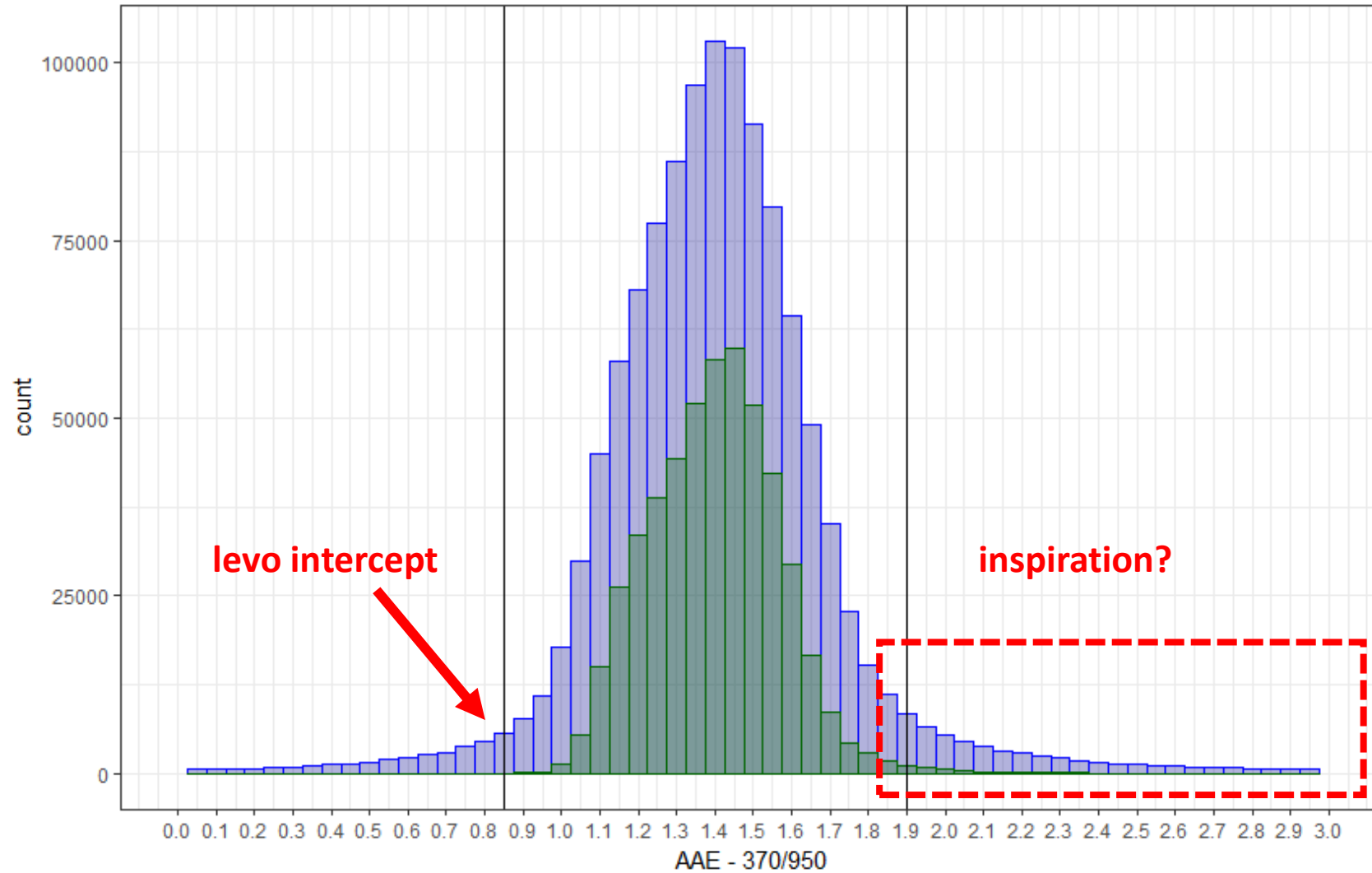


$AAE_{bb}$

Fuller et al., 2013  
<http://dx.doi.org/10.1016/j.atmosenv.2013.12.037>



# AAE pair – $BC_{ff}$ $BC_{bb}$



# Two-component models – Sandradewi/Aethalometer model

$$b(\lambda) = b_{ff}(\lambda) + b_{bb}(\lambda)$$

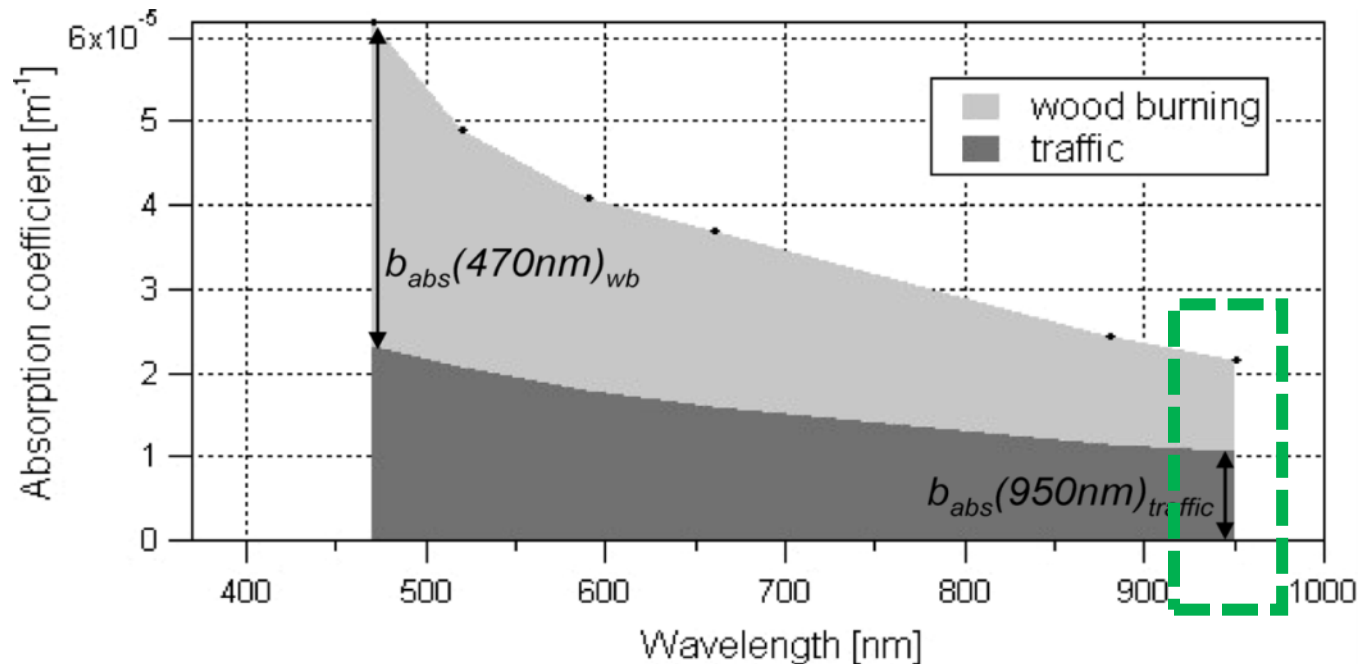
$$\lambda = 470 \text{ nm}, 950 \text{ nm}$$

$$\frac{b_i(470 \text{ nm})}{b_i(950 \text{ nm})} = \left[ \frac{470 \text{ nm}}{950 \text{ nm}} \right]^{-AAE_i}$$

$$\begin{matrix} AAE_{ff} \\ AAE_{bb} \end{matrix}$$

external parameter assumption

result



$$BC_{ff} = BC \frac{b_{ff}(950 \text{ nm})}{b(950 \text{ nm})}$$

$$BC_{bb} = BC - BC_{ff}$$

# Two-component models – Massabò/MWAA model

$$b(\lambda) = b_{BC}(\lambda) + b_{BrC}(\lambda)$$

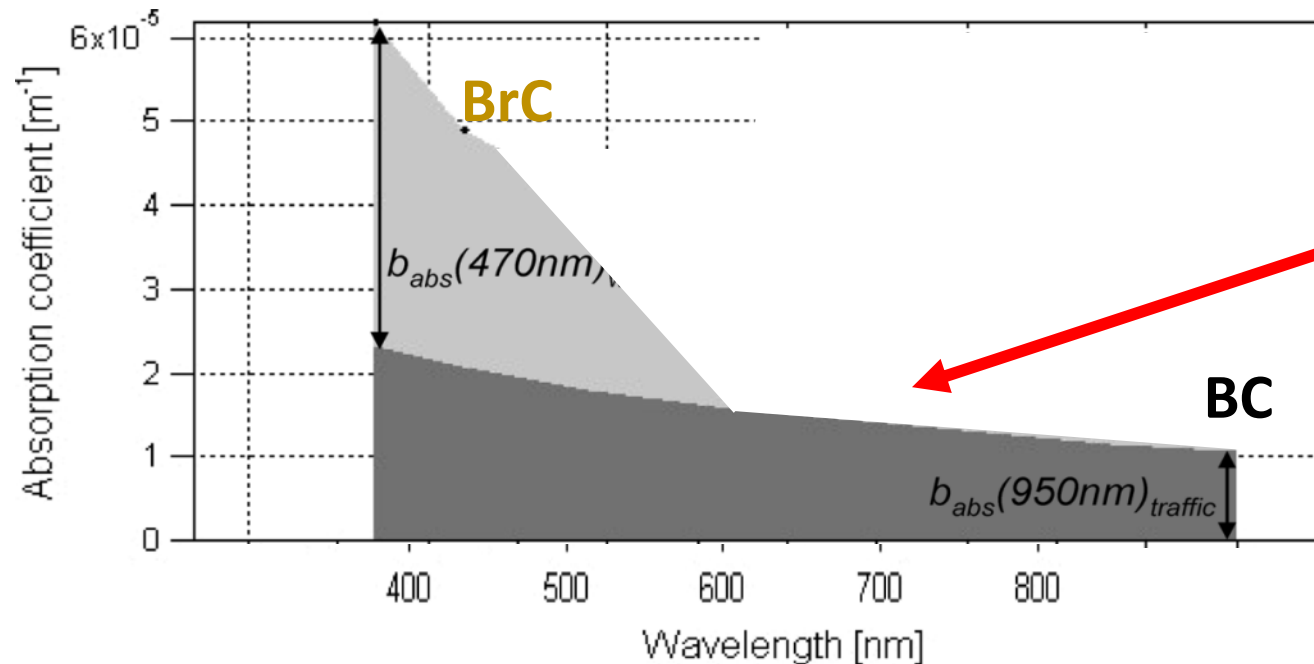
$$\lambda = 370 \text{ nm}, 950 \text{ nm}$$

$$\frac{b_{BC}(370 \text{ nm})}{b_{BC}(950 \text{ nm})} = \left[ \frac{470 \text{ nm}}{950 \text{ nm}} \right]^{-AAE_{BC}}$$

$$AAE_{BC} \sim 1$$

$$AAE_2$$

result



assumption

BrC does not absorb in red, IR

# C...c...c...calibration to determine $C \leftrightarrow MAC!$

Field: comparison w/ other filter photometers

**Aerosol absorption coefficient  $b_{abs}$   $Mm^{-1}$**

- need reference **sample**
- need **reference method to measure absorption**
- **Reference methods:**
  - extinction - scattering: low  $SSA$
  - photoacoustics: non-coated particles, no VOCs or water
  - **photothermal interferometry**

# Historical calibration – filter photometers

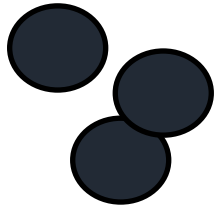
## **Black Carbon** mass concentration BC $\mu\text{gm}^{-3}$

- Optical meas. equivalent to mass (filter photometers)
- **BC mass „calibration“:**
  - sample filters
  - wash with different solvents (Soxhlet extraction)
  - analyze optically
  - analyze thermally for total carbon –  $\text{CO}_2$
  - assume all chemically refractory carbon also absorbs!
  - voilà : mass **attenuation** cross-section:  $\sigma$ ,  $\text{m}^2/\text{g}$

Gundel et al., 1984

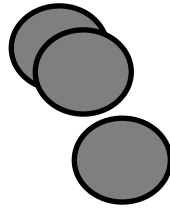
$$SSA = \frac{b_{sca}}{b_{sca} + b_{abs}}$$

SSA = 0.1



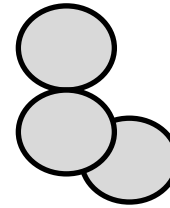
urban

0.5



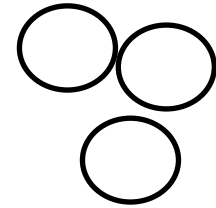
urban

0.8



regional

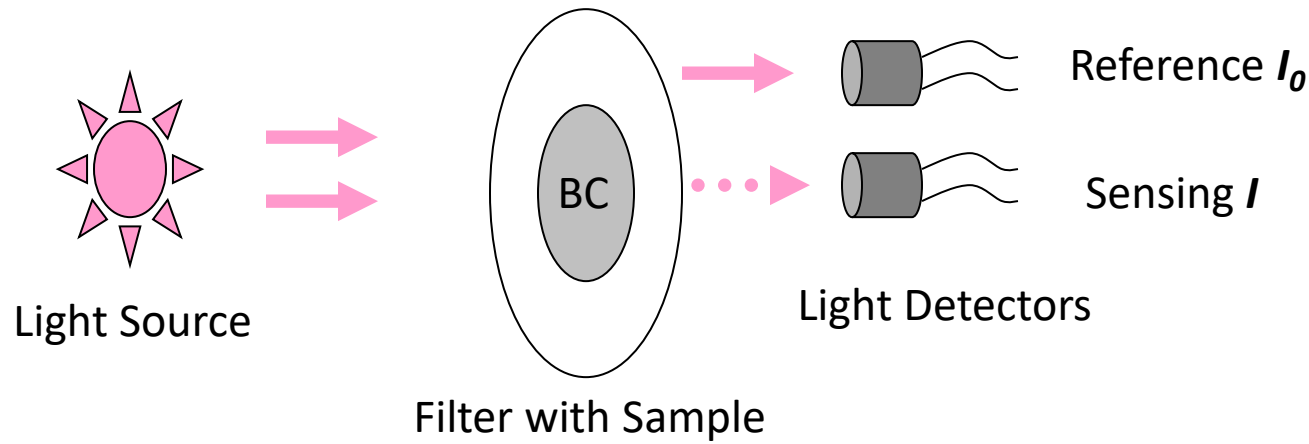
1



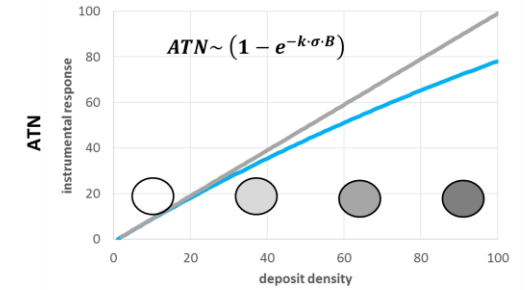
global

background

# Optical measurement of BC: filter photometers



$$ATN = \ln \frac{I_0}{I}$$



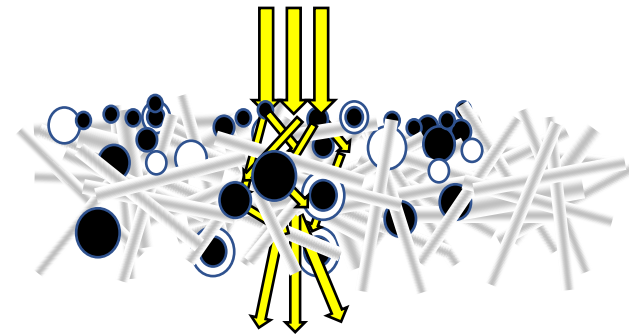
Non-linear response of the detection - saturation

$$b_{ATN} \sim \frac{\Delta ATN}{\Delta t} + \underline{m_s b_{sca}}$$

- Collect sample continuously.
- **Optical attenuation**  $\sim$  change in ATN.
- Convert **attenuation** to **absorption**:

$$b_{abs} = \frac{b_{ATN}}{c}$$

- Real-time data: **minutes**



# Filters: blessing or curse?

$$b_{ATN} = \frac{S \Delta ATN}{\Phi \Delta t} \cdot f(ATN) = BC \cdot \sigma + \underline{m \cdot b_{sca}}$$

$$\underline{b_{abs}} = \frac{\underline{b_{ATN}}}{\underline{C_f}} - \underline{m_s} \cdot \underline{b_{sca}}$$

Filter properties.

Need reference measurement.

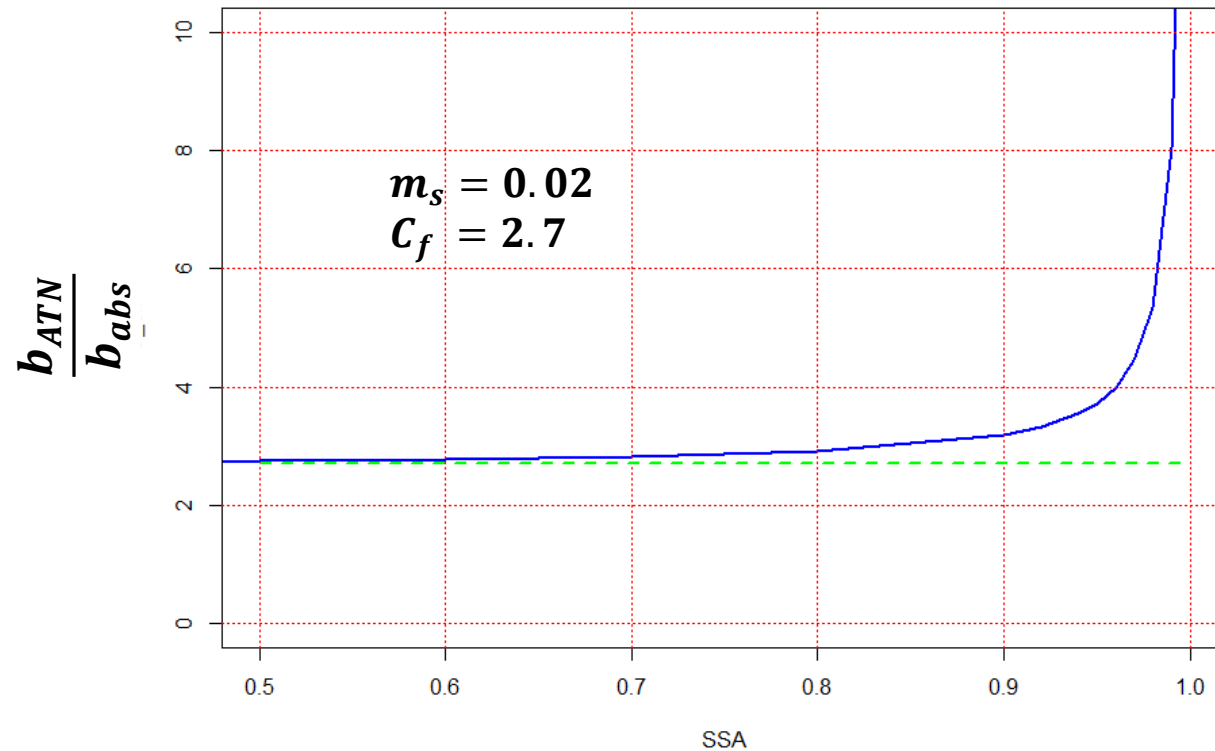
Additional measurement:  
scattering.

$b_{sca} \gg b_{abs}$   
( $SSA > 0.8 \dots$ )

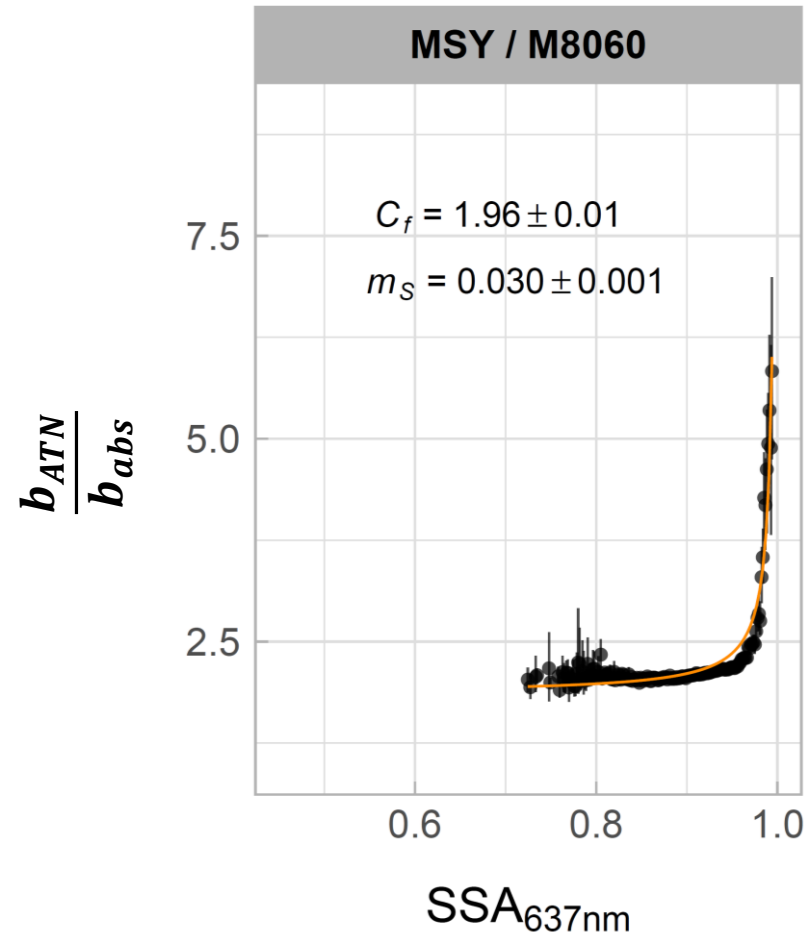
# SSA dependence

$$b_{abs} = \frac{b_{ATN}}{C_f} \cdot \frac{1 - SSA}{1 - SSA + m_s \cdot SSA}$$

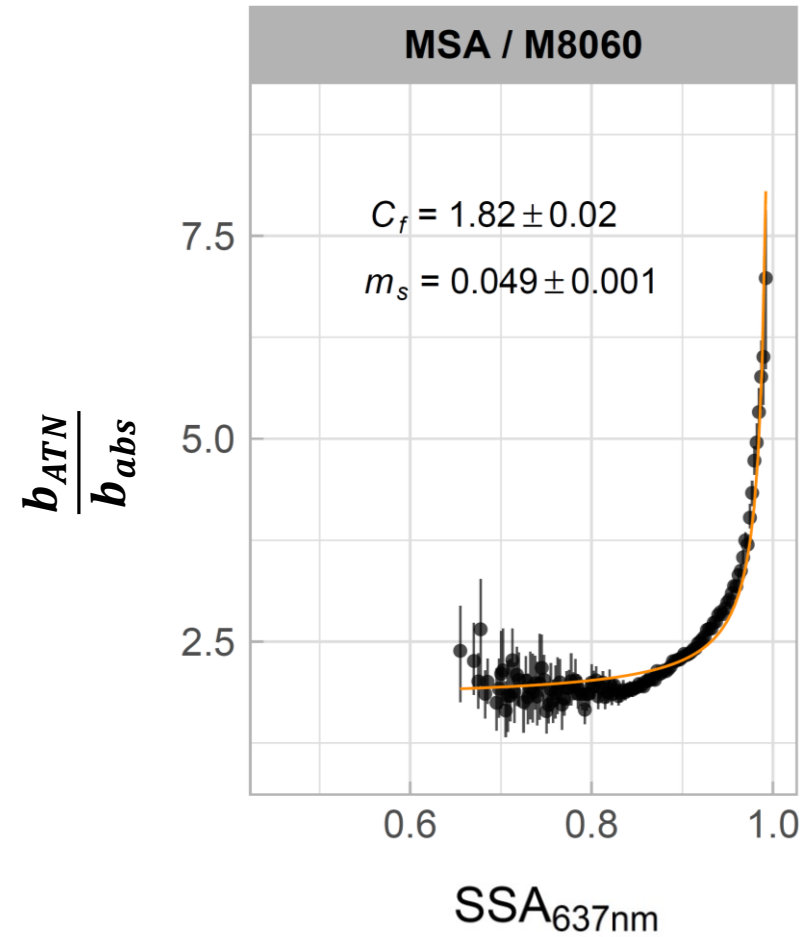
$$\frac{b_{ATN}}{b_{abs}} = C_f \cdot \frac{1 - SSA + m_s \cdot SSA}{1 - SSA}$$



# Montseny regional



# Montsec remote regional

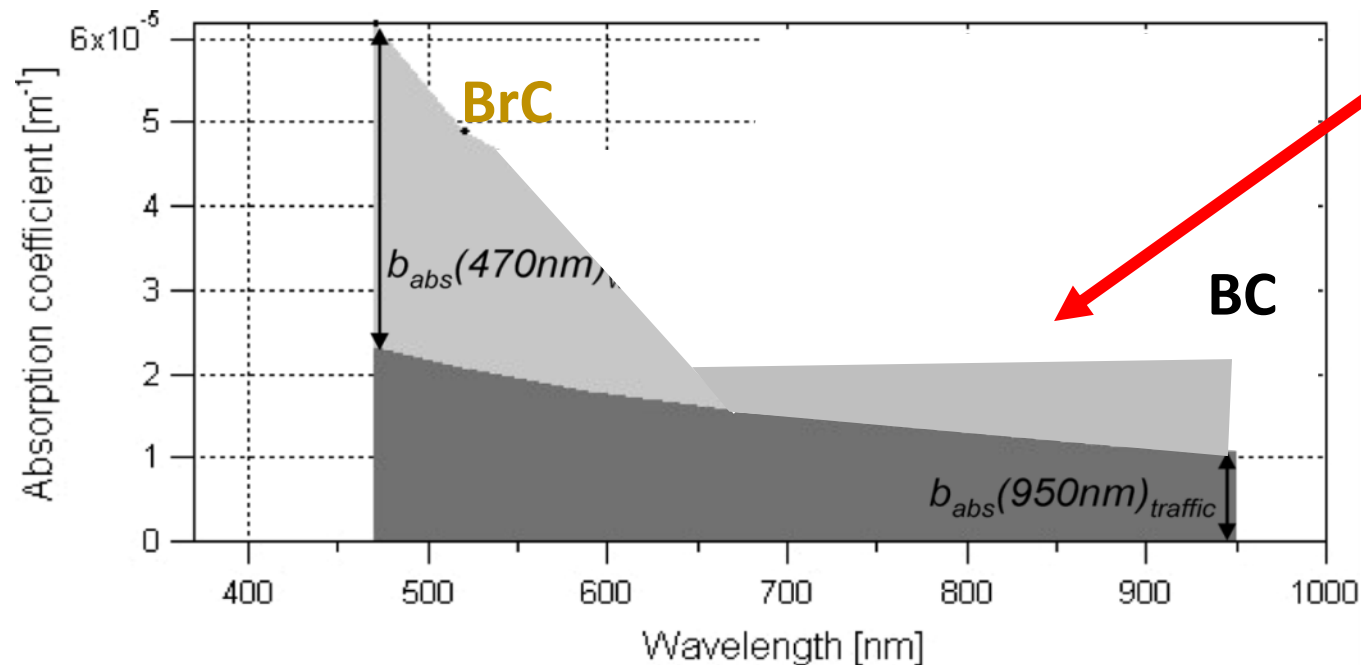


# Two-component models – Massabò/MWAA model

$$b(\lambda) = b_1(\lambda, \text{biomass}) + b_2(\lambda, \text{fossil}) \quad \lambda = 470 \text{ nm}, 950 \text{ nm}$$

$$b_i(470 \text{ nm}) / b_i(950 \text{ nm}) = (470 \text{ nm} / 950 \text{ nm})^{-AAE_i}$$

$AAE_1$   
 $AAE_2$



BrC does not absorb in red, IR

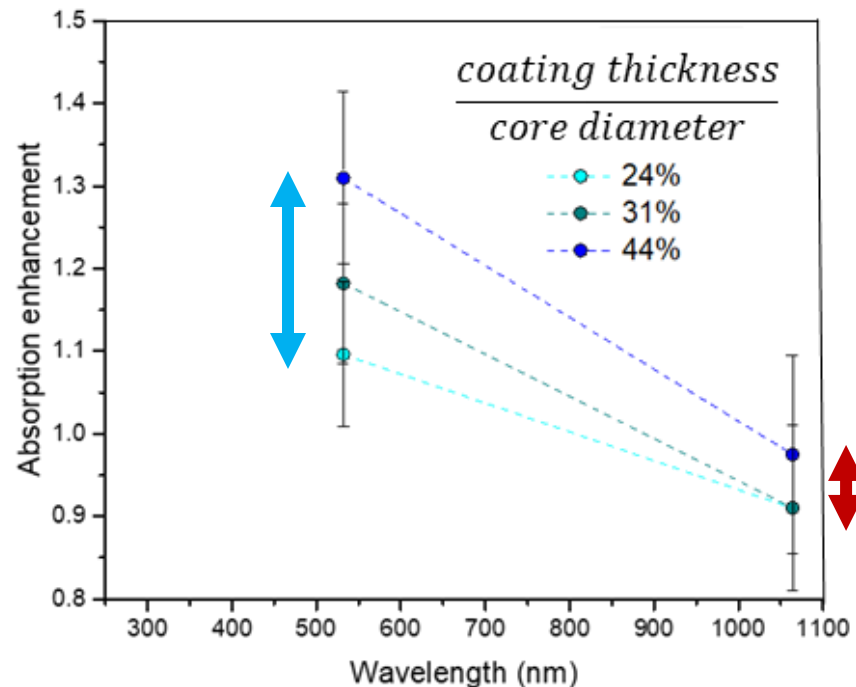
# BrC mass, MAC

**WRONG!**

- traditionally – measure OM, use OM mass

$$b_{abs,BrC} = OM \cdot MAC_{BrC}$$

BC particles coated with  $\alpha$ -pinene



→ change in AAE!

<https://doi.org/10.5194/amt-15-561-2022>  
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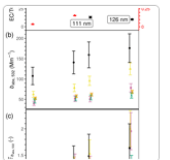
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01 Feb 2022

Comparing black-carbon- and aerosol-absorption-measuring instruments – a new system using lab-generated soot coated with controlled amounts of secondary organic matter

Daniel M. Kalbermatter, Griša Močnik, Luka Drinovec, Bradley Visser, Jannis Röhrbein, Matthias Oscity, Ernest Weingartner, Antti-Pekka Hyvärinen, and Konstantina Vasilatou

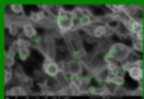


<https://doi.org/10.5194/amt-15-561-2022>

## WP1 (Objective 1)

Aerosolised nigrosin spherical particles

Refractive index + Mie theory



NO<sub>2</sub> Gas

Primary standards  
CALIBRATION

Extinction Minus Scattering (EMS)  
Photo thermal Interferometry (PTI)

Secondary standards  
EQUIVALENCE TESTING

Photoacoustic (PAX)

Traceable light  
absorption coefficient

$b$  ( $Mm^{-1}$ )

Target uncertain  
< 10% (95% confidence level)

Standardization  
stanBC

METROLOGY  
PARTNERSHIP



## WP4 (Objective 4)

New working group or  
CEN/TC 264 WG 35

Traceable reference methods for  
determining aerosol light absorption  
coefficients at multiple wavelengths

Materials, methods and correction factors  
for calibrating filter-based photometers  
against the reference method(s)

## WP2 (Objective 2)

Primary standards  
CALIBRATED Instruments

Extinction Minus  
Scattering (EMS)  
Photo thermal  
Interferometry (PTI)

$b$  ( $Mm^{-1}$ )

Mass absorption  
cross-section

$$MAC(m^2/g) = \frac{b}{EC_{mass}}$$

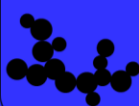
Standardised Method

EC mass determination  
(EN 16909:2017)

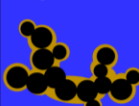
$EC_{mass}$   
( $g/m^3$ )

Reference aerosols

Fresh soot



Aged soot

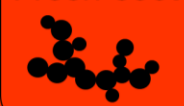


SSA < 0.7 & SSA < 0.3

## WP3 (Objective 3)

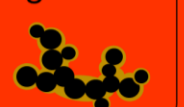
Vehicle engines

Fresh soot



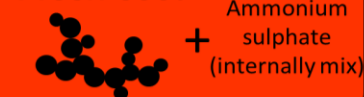
Suburban  
and rural site

Aged soot



Industrial and  
harbours site

Fresh soot



Ambient-like aerosol  
in urban sites

Aged soot



0,1 < SSA < 1

WP1

Traceable light  
absorption  
coefficient  $b$

WP2

Mass absorption  
cross-section  
 $MAC$

Target candidate method  
Uncertainty  
< 15% (95% confidence level)

eBC mass concentration

$$eBC(g/m^3) = \frac{b}{MAC}$$

Candidate Method

- Photo-acoustic instruments
- Filter-based photometers
- Wearable filter-based photometers

Measurements @ 4 km and 200 km/h



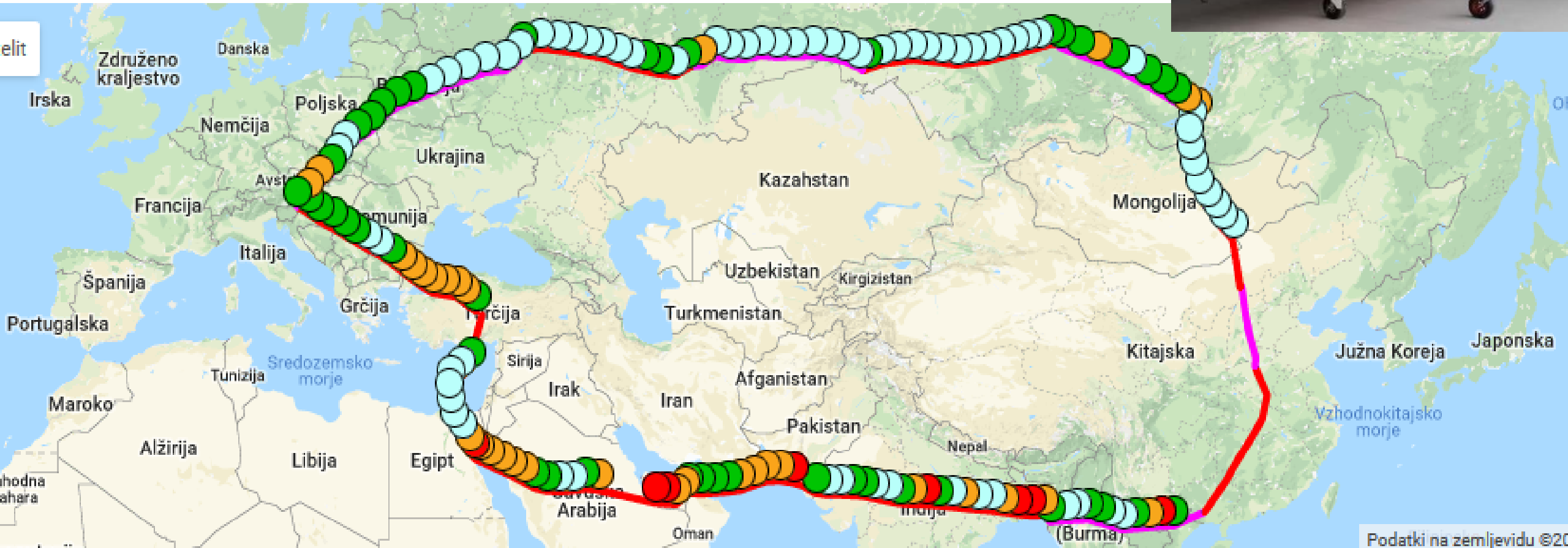
# Flight 2012



# Flight 2016

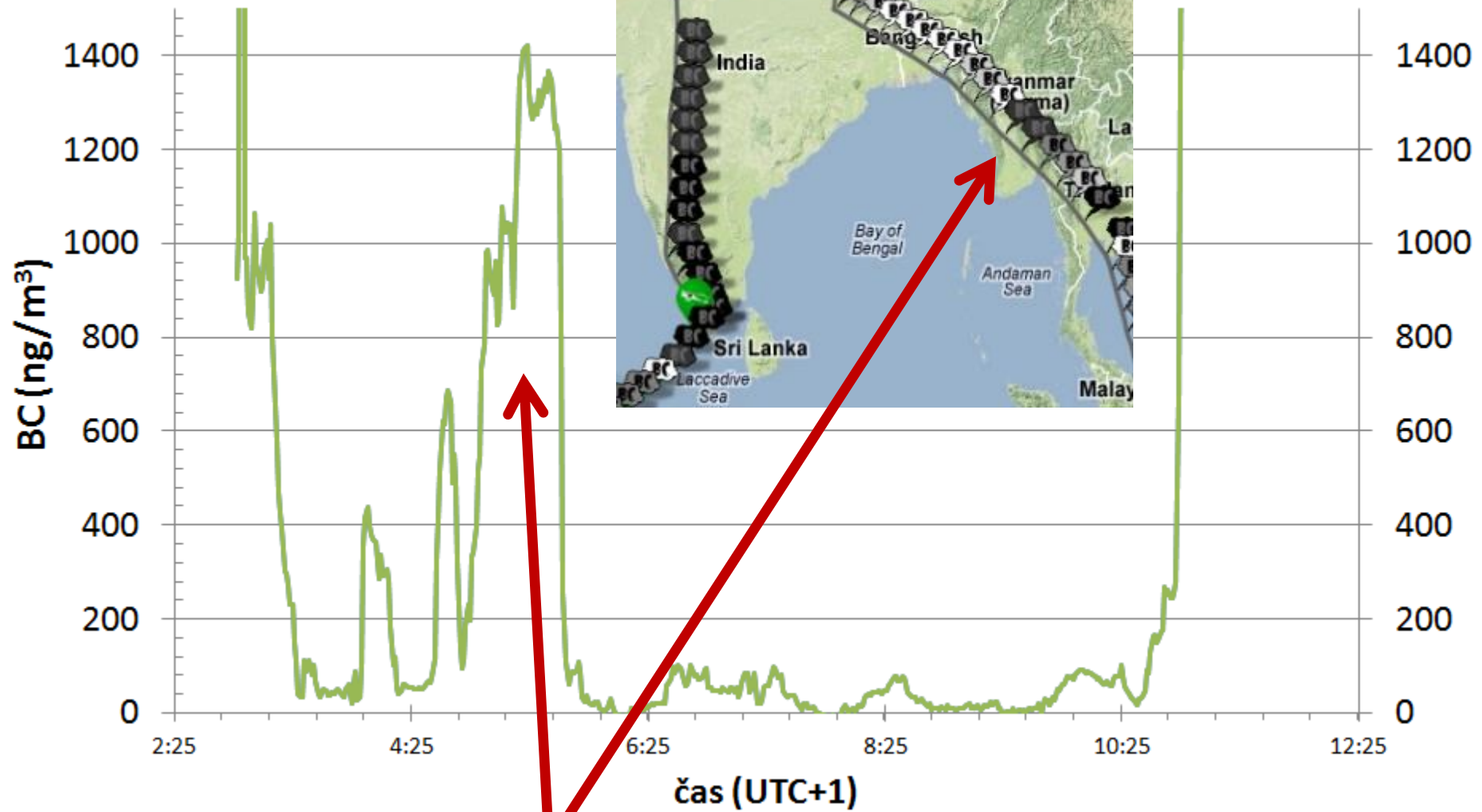


# Flight 2018



Thailand

India



**increased absorption**

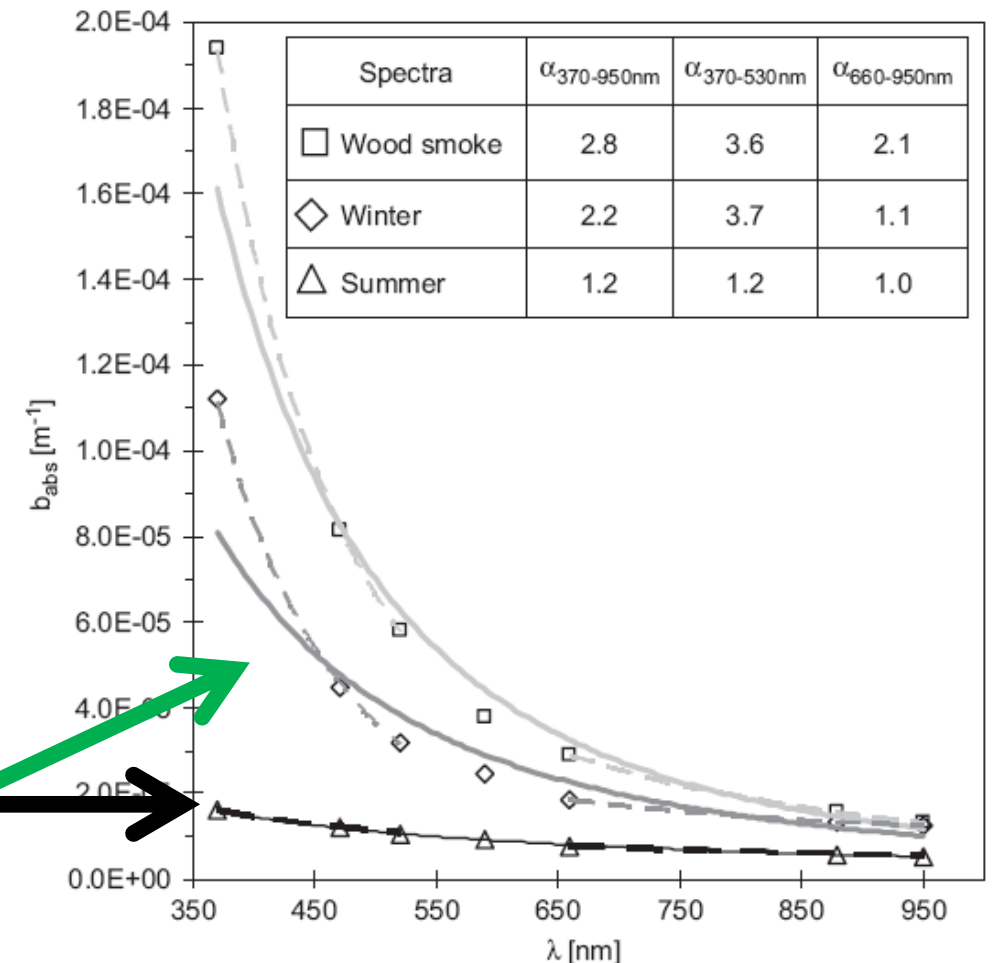
# Optical properties - source apportionment

- measure attenuation with the Aethalometer
- absorption coefficient -  $b_{abs}$
- for pure black carbon:  $b_{abs} \sim 1/\lambda$
- generalize **Angstrom exponent**:

$$b_{abs} \sim 1/\lambda^\alpha$$

**diesel:**  $\alpha \approx 1$

**biomass-smoke:**  $\alpha \approx 2$  and higher

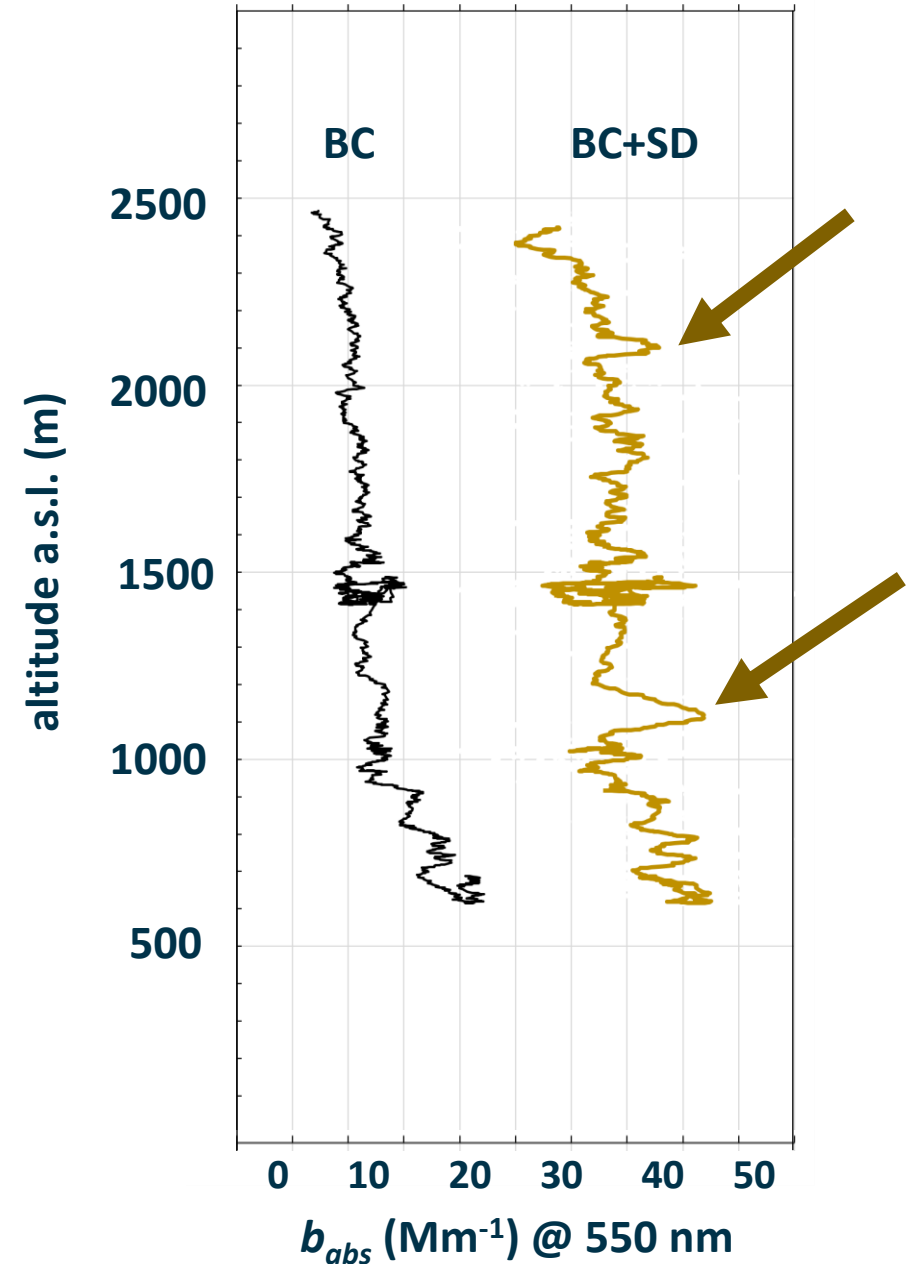


# Flight 2019 – Saharan dust events

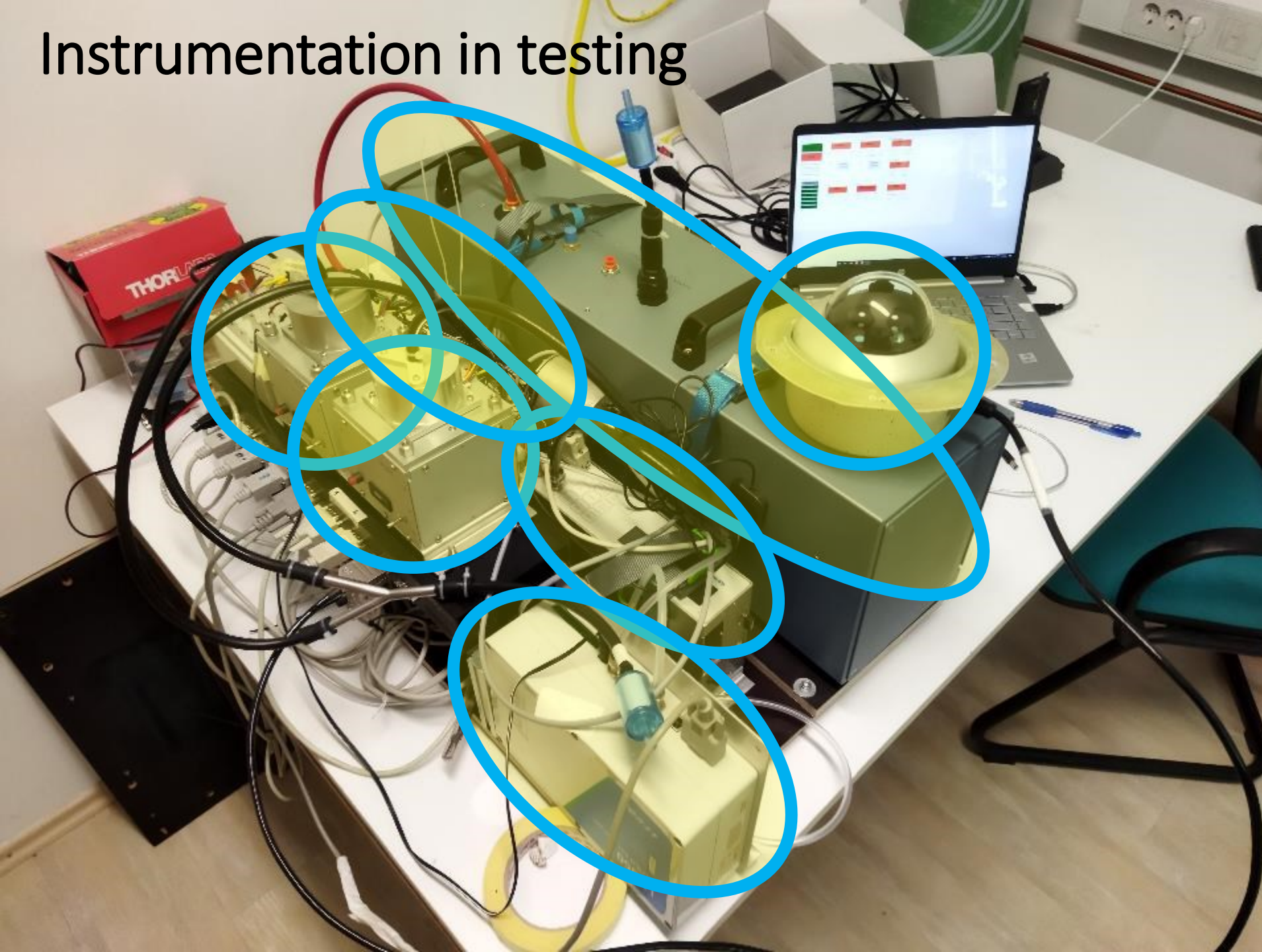


# Vertical profiles – NE Spain 2019

- **vertical profiles of optical properties:**
  - fine  $b_{abs}$
  - coarse  $b_{abs}$
  - size distribution
- **regional transport, atmospheric mixing**
- **layers not vertically, spatially homogeneous**
- **validation of Aeolus ALADIN lidar extinction, scattering @ Cape Verde**



# Instrumentation in testing



Absorption coeff.:

- fine
- coarse enhanced

Scattering coeff.:

- total

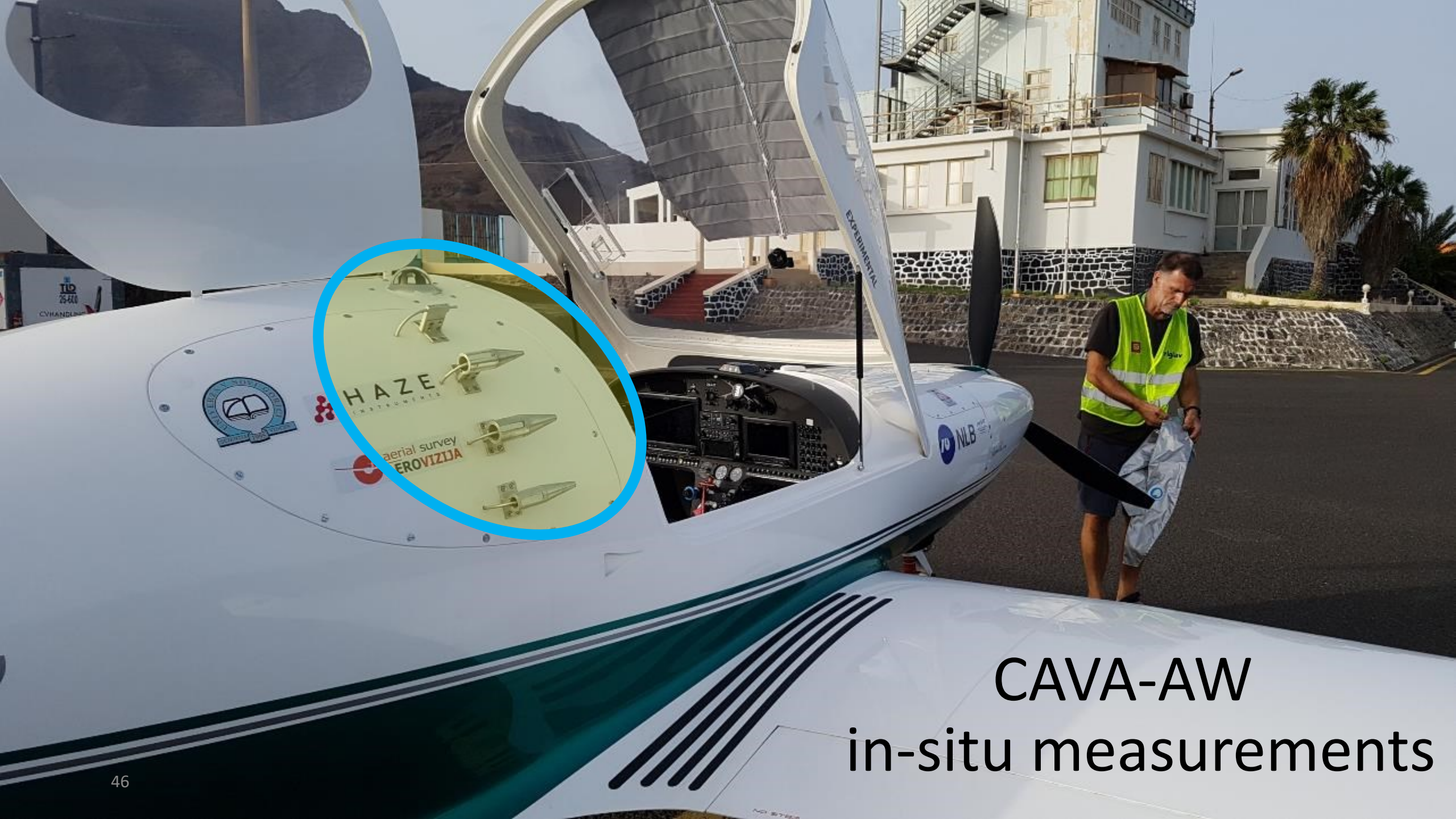
Size distribution:

- total
- coarse enhanced
- large particles

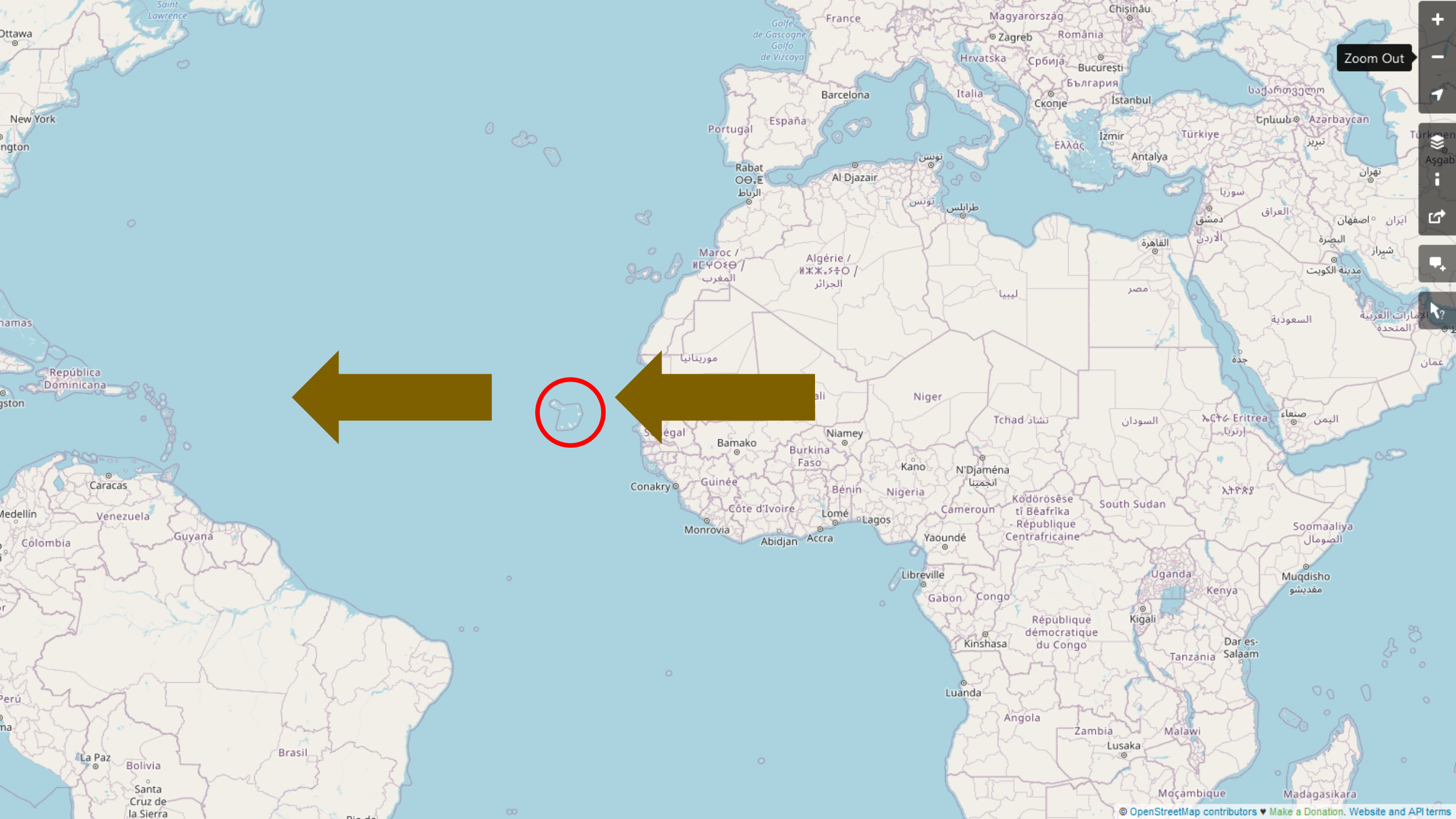
Solar irradiance

CO2 sensor





# CAVA-AW in-situ measurements








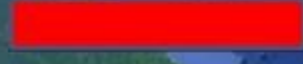
Zoom Out



# Aeolus Cal/Val



## Orbit Days

	Mon
	Tue
	Wed
	Thu
	Fri
	Sat
	Sun

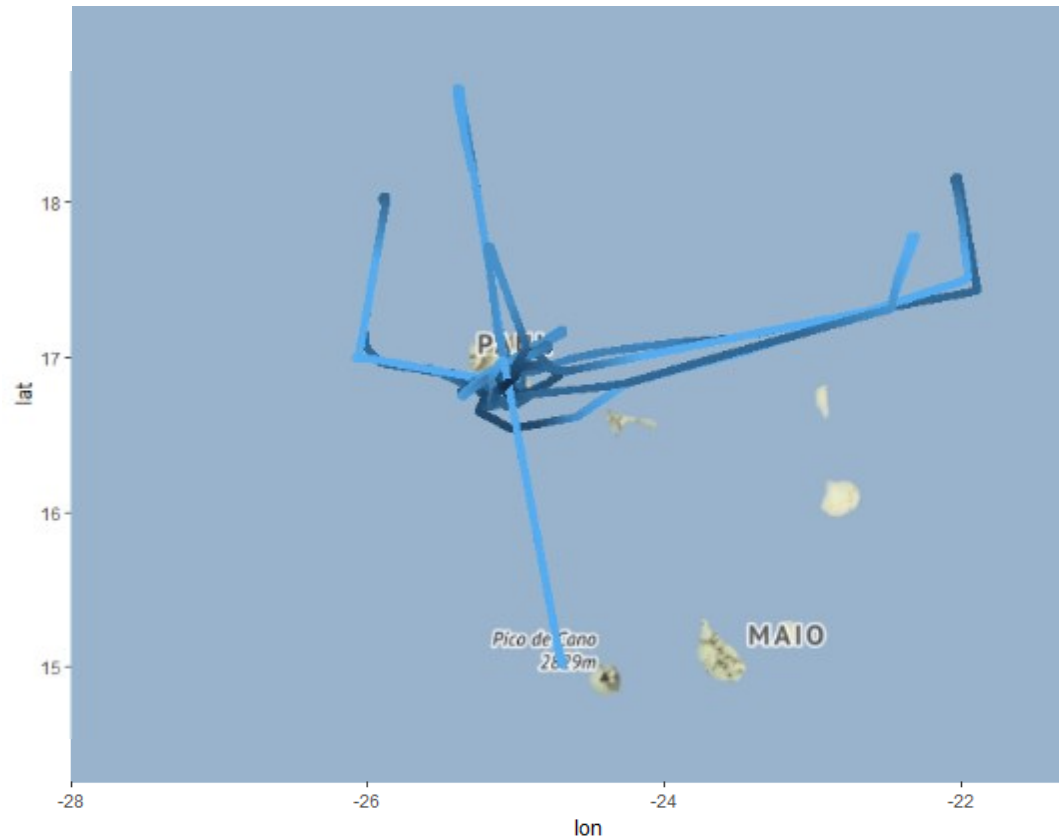


Data SIO, NOAA, U.S. Navy, NGA, GEBCO  
Image Landsat / Copernicus

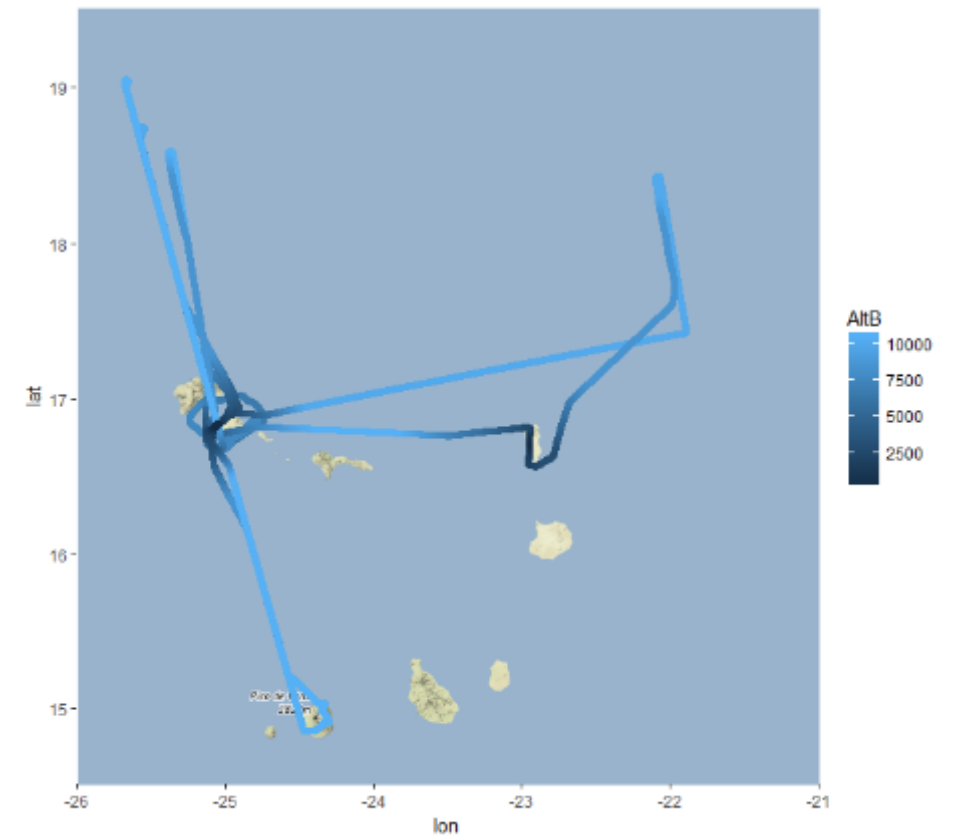
Google Earth

# Campaign overview

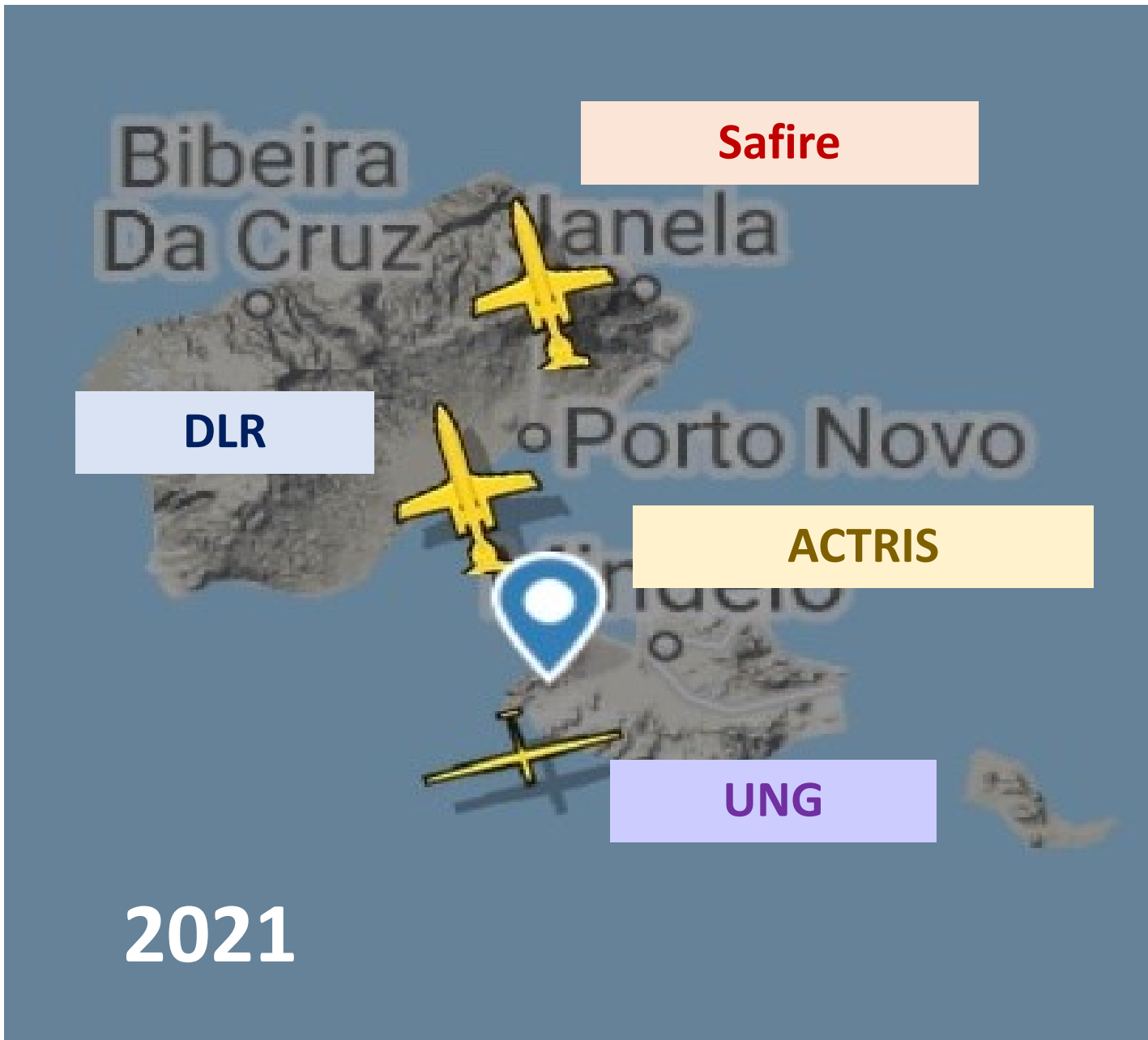
2021 – 18 flights



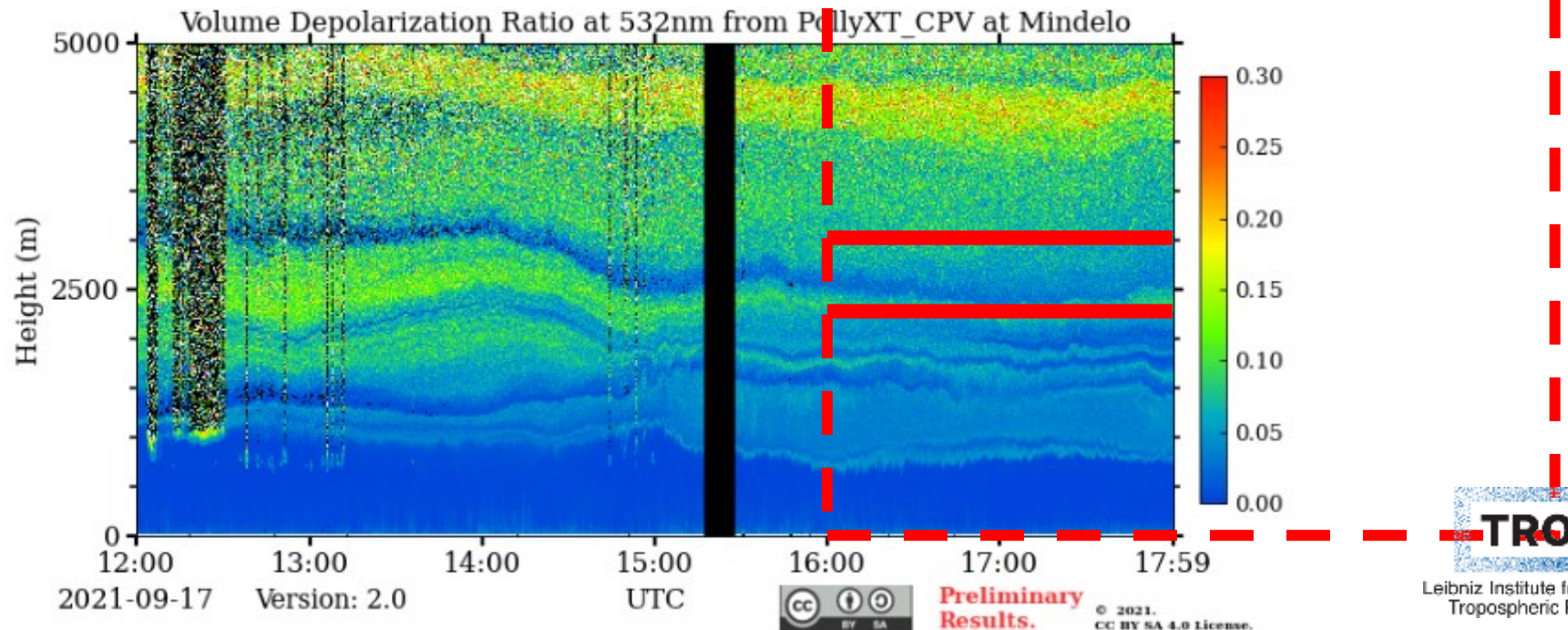
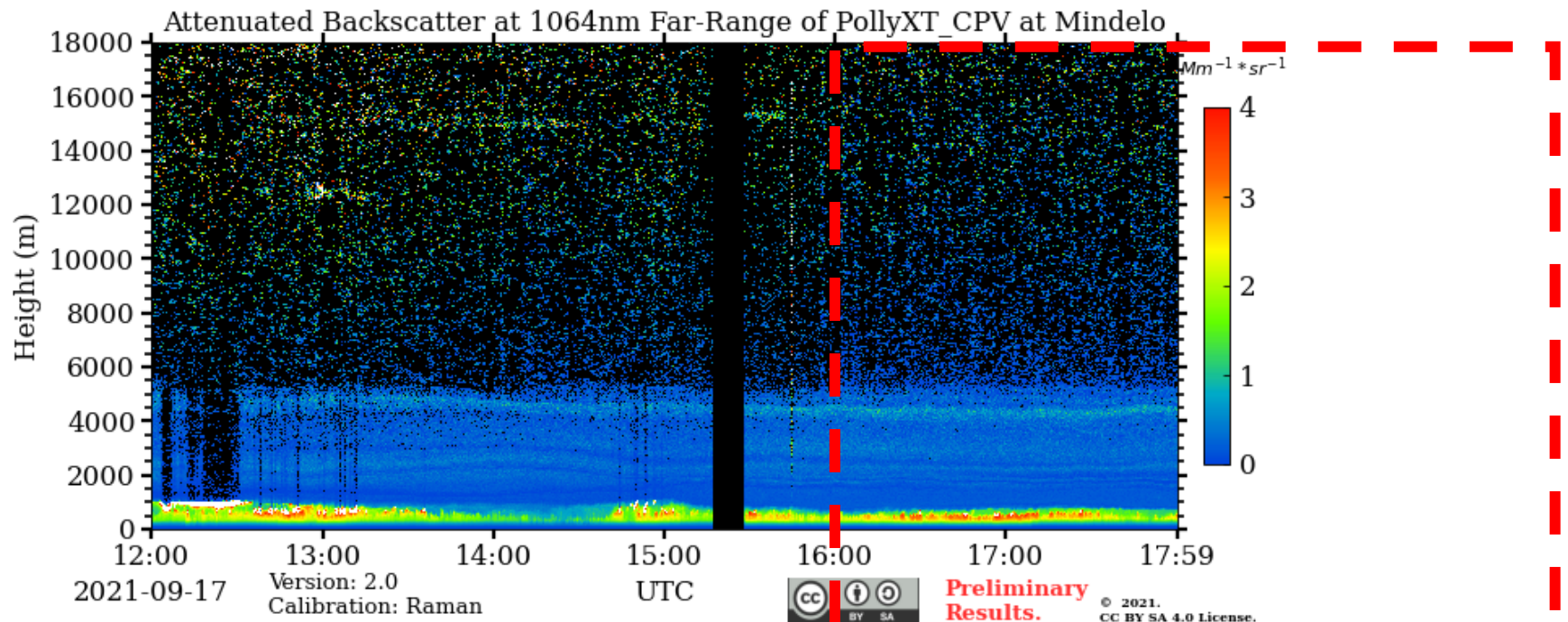
2022 – 9 flights



# Coordination with DLR, SAPHIRE, NASA DC-8







17 Sep 2021

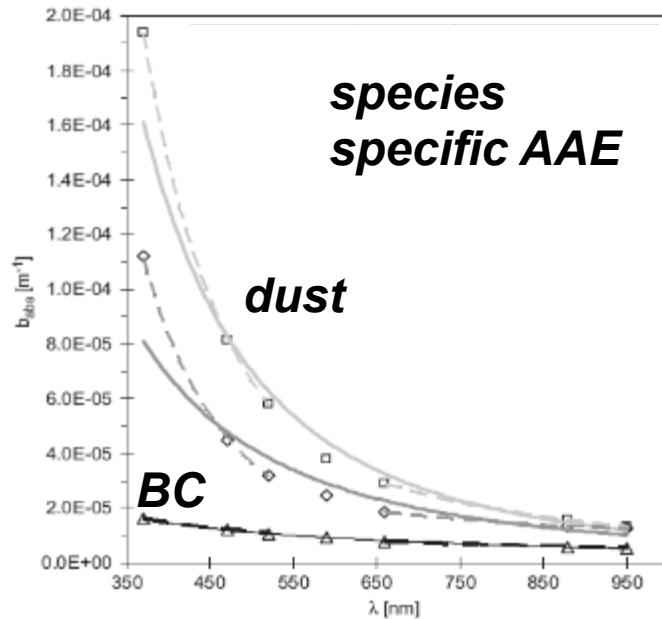


Leibniz Institute for  
Tropospheric Research

17 Sep 2021



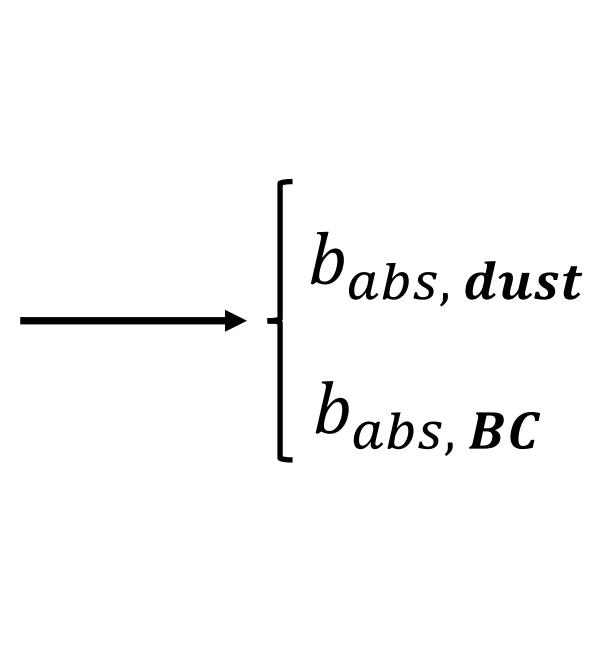
# Methodology – Dust & BC Absorption



pVI inlet  
&  
isokinetic inlet

+

wavelength  
dependence

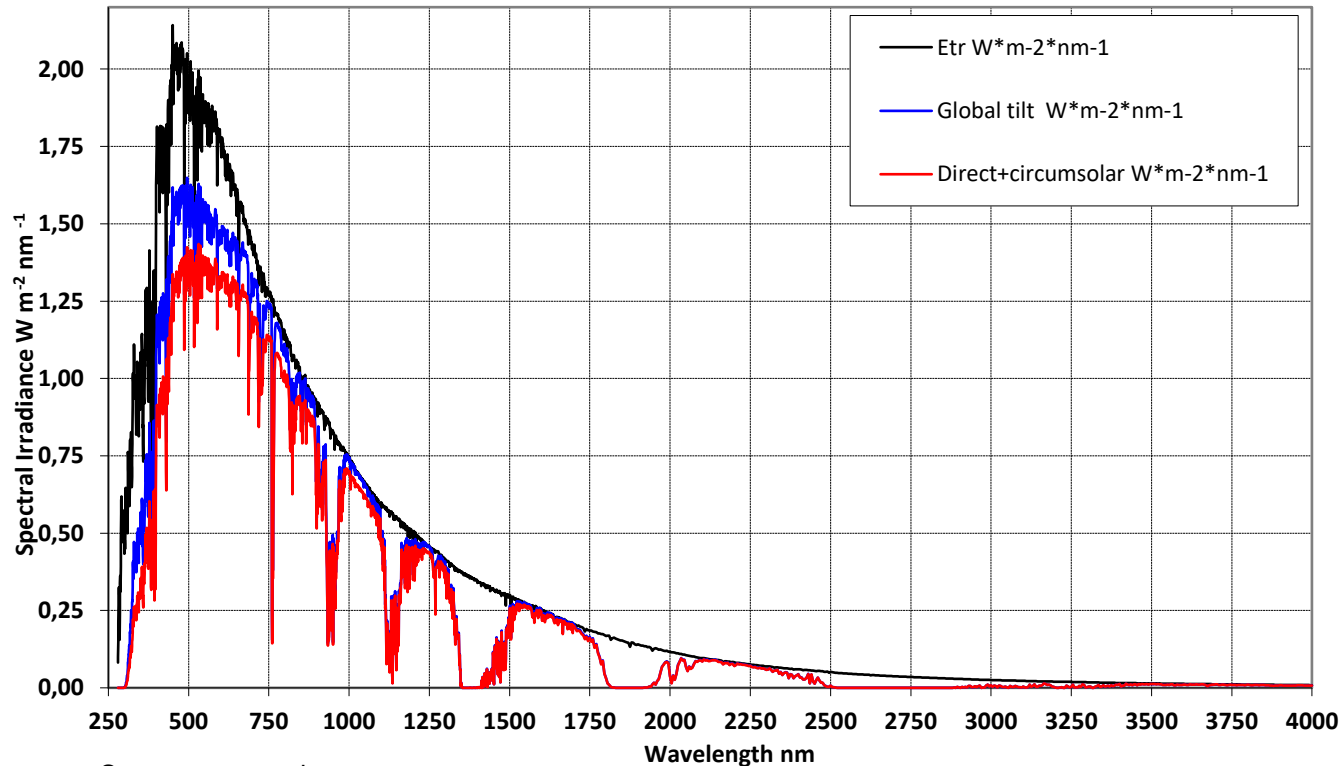


# Heating Rate

$$HR = HR_{\text{direct}} + HR_{\text{diffuse}} + HR_{\text{reflected}}$$

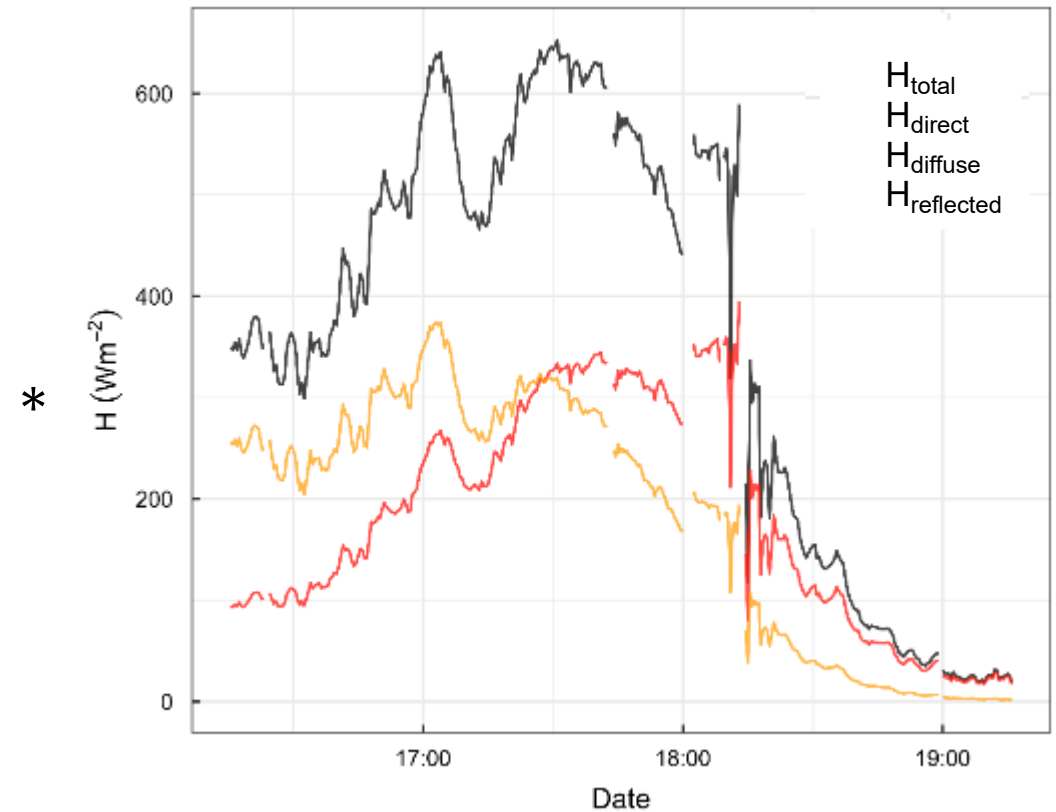
$$HR = \frac{1}{\rho C_p} \sum_{n=1}^3 \int_{\theta} \int_{\lambda} \frac{F_{n(\lambda, \theta)}}{\mu} b_{\text{abs}(\lambda)} d\lambda d\theta$$

ASTM G173-03 Reference Spectra



Source: www.nrel.gov

16 September 2021



**Thank you!**  
**Questions?**

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