PAUL SCHERRER INSTITUT

PSI

Martin Gysel :: Laboratory of Atmospheric Chemistry :: Paul Scherrer Institute

Characterization of black carbon properties by means of laser-induced incandescence techniques

Sino-German symposium “Soot and its climatic, environmental and health impacts”
27 June 2016, Beijing, China

Selected methods for the measurement of "black carbon" mass

Method/Principle	Quantity	Commercial Instruments (incomplete)	Comments
Evolved carbon methods (with optical correction for pyrolysis)	elemental carbon (EC)	• Sunset lab thermal-optical OC-EC analyzer	Various protocols (EUSAAR-2, Improve, NIOSH,…)

Absorption photometers	equivalent black carbon (eBC)	• Filter-based:
		  • Aethalometer
		  • MAAP
		  • PSAP
		  • COSMOS
		• Photo-acoustic
		  • PASS-3
			"mass" inferred from absorption coefficient with assumptions on the MAC of BC.

Laser-induced incandescence (LII) ➔ thermal emission of incandescent BC	refractory black carbon (rBC)	• Single particle soot photometer (SP2) • Artium LII-300	• Continuous-wave single particle LII • time-resolved LII
Aerosol mass spectrometry	«carbon content» (➔ EC? or rBC?)	• SP-AMS • ATOFMS	Usually not quantitative if stand-alone
Raman spectroscopy (detection graphite-like microstructure)	elemental carbon (EC)	• Commercial instruments?	Not addressed in this presentation.

Adapted from Petzold et al., Atmos. Chem. Phys., 2013

Refractory black carbon mass measurement by laser-induced incandescence (LII) methods

Laser-induced incandescence is NOT a light-absorption based method!
Some light-absorption by the investigated material is required to heat it in the laser beam. However, the light-absorption cross section of the material has no direct influence on the measured thermal radiation signal.

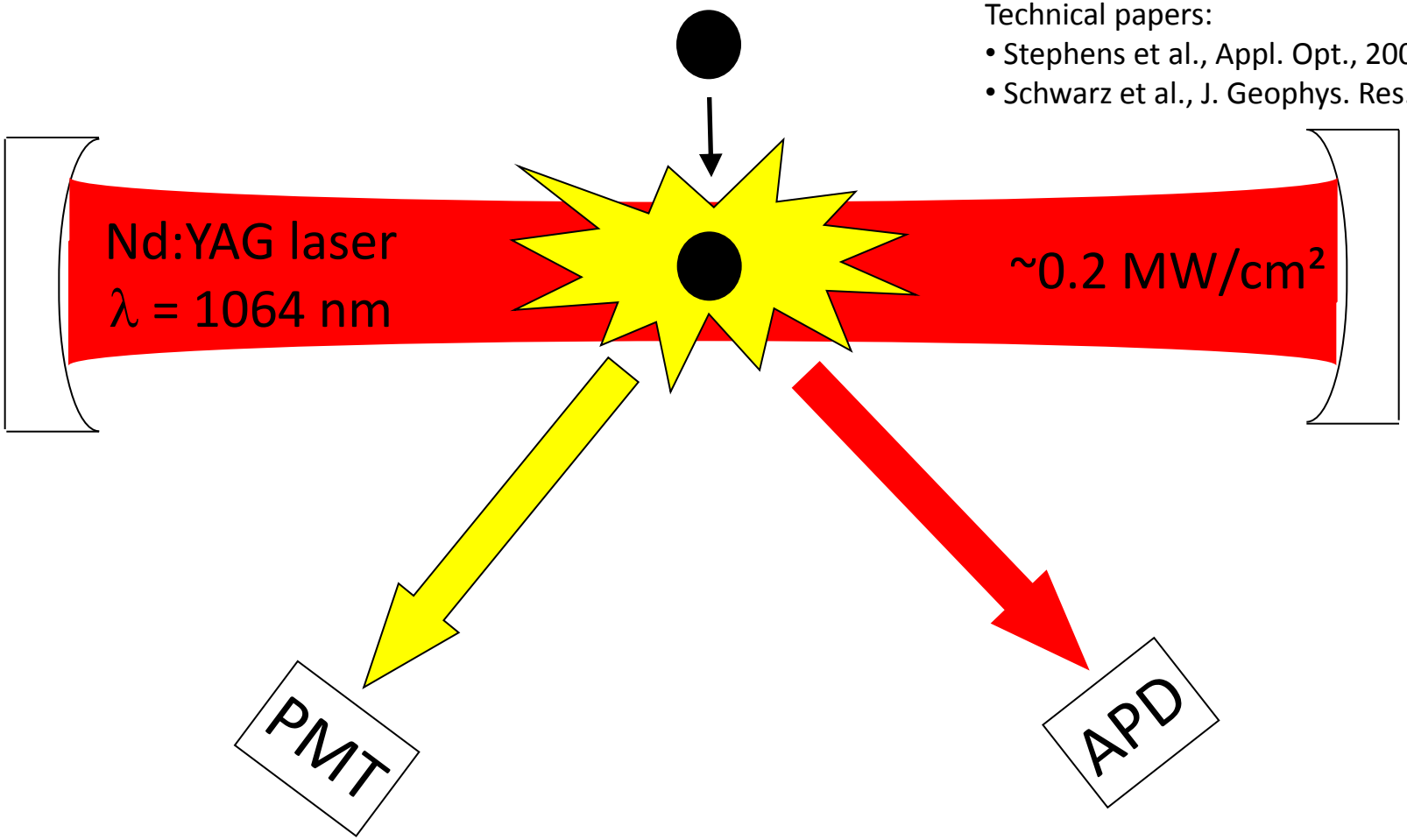
«BC core» = aggregate of primary particles, composed of "elemental carbon"

- almost «pure carbon» (>90% C)
- graphitic microstructure with some disorder
- fractal-like aggregate of primary spheres
- insoluble in any solvent
- strongly light-absorbing («black»)
- extremely refractory ($T_{sublim} \approx 4000\ ^{o}C$)

Laser-induced incandescence (LII):
measurement of **thermal radiation** emitted by the incandescent «BC core»
➔ **indirect measure of refractory black carbon (rBC) mass**

Single Particle Soot Photometer (SP2)

Technical papers:
- Stephens et al., Appl. Opt., 2003.
- Schwarz et al., J. Geophys. Res., 2006.



detection of
thermal radiation
➔ **BC mass**

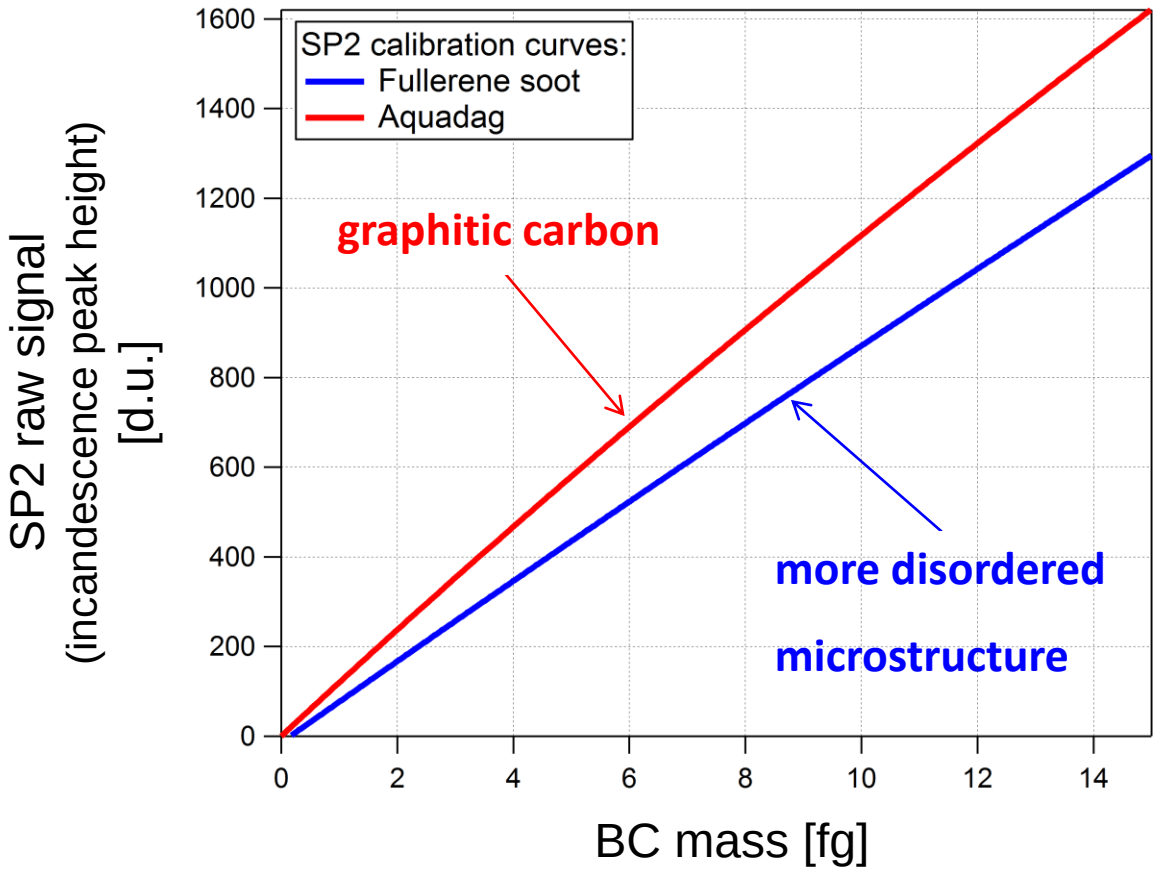
detection of
scattered light
➔ **optical sizing**

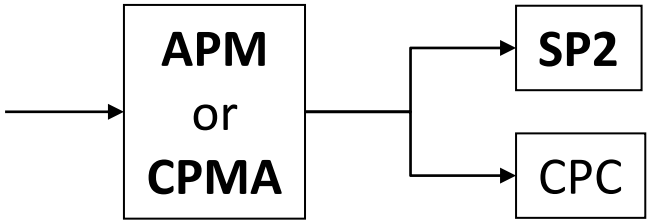
«Single particle continuous-wave LII»

«Single particle continuous-wave LII» (e.g. Schwarz et al., 2006)**:**

- *Principle*:
  - Continuous-wave intra-cavity laser used to heat the particles
  - Detection of peak thermal radiation emitted by individual particles at sublimation point
    ➔ **single particle properties**
  - Jet nozzle used to direct particle beam through the laser
    ➔ **100% detection efficiency** (within given BC particle size limits)
  - **Empirical calibration needed**
- *Application*:
  - **Extremely sensitive**, as every **single particle** is detected
  - **Limited to atmospheric BC mass concentrations** due to coincidence errors at high particle number concentrations.
  - **Provides additional information on BC mixing state**
  - Also applied for detection of BC in ice core, snow and rain samples (e.g. McConnell et al., 2007; Schwarz et al., 2012; Ohata et al., 2011; Wendl et al., 2014)
- *The only commercially available instrument* (to my knowledge):
  - **Single Particle Soot Photometer (SP2)** from Droplet Measurement Technology (DMT) (Stephens et al., 2003; Schwarz et al., 2006)

Calibration of the SP2



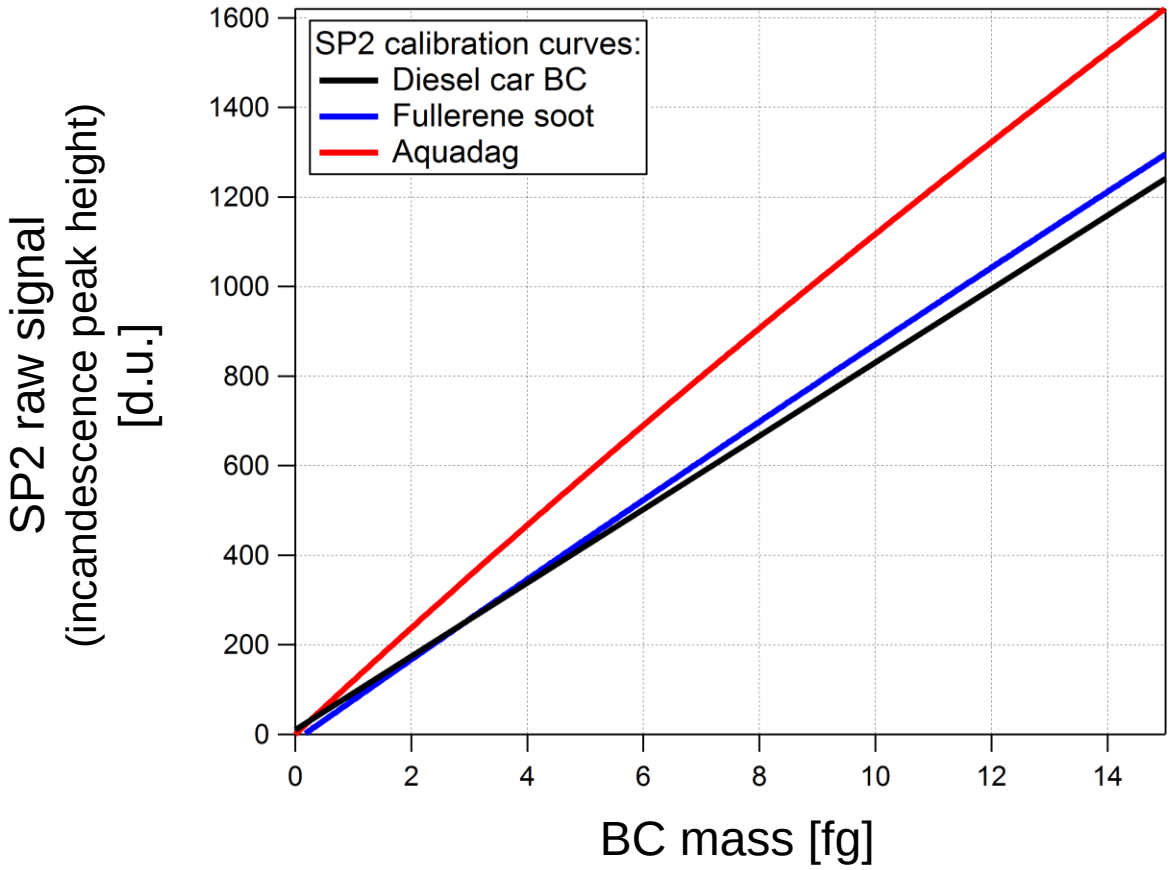


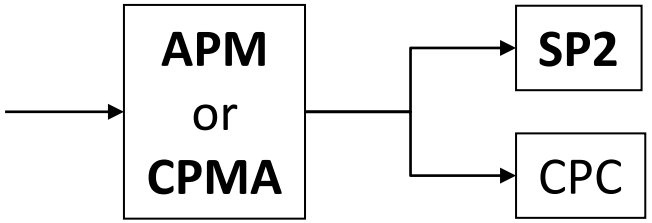
Laborde et al., 2012

➔ **Bad news: SP2 calibration curves for different BC types can differ by at least 30-40%**

➔ **Good news: the rBC mass measurement by the SP2 is not influenced by BC mixing state!**
(e.g. Moteki and Kondo, 2007; Slowik et al., 2007)

Calibration of the SP2





Laborde et al., 2012

➔ **Calibration curve for diesel car BC agrees well with that of fullerene soot**

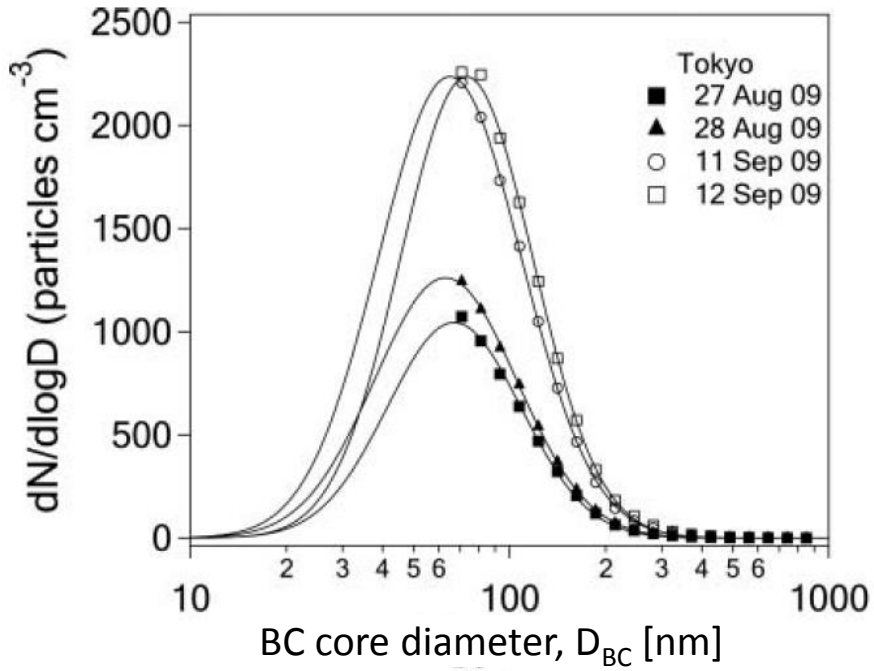
➔ **Diesel car BC calibration is also representative of SP2 sensitivity to BC from wood combustion and BC in ambient aerosol**
(Moteki and Kondo, 2010; Laborde et al., 2012; Baumgardner et al., 2012)

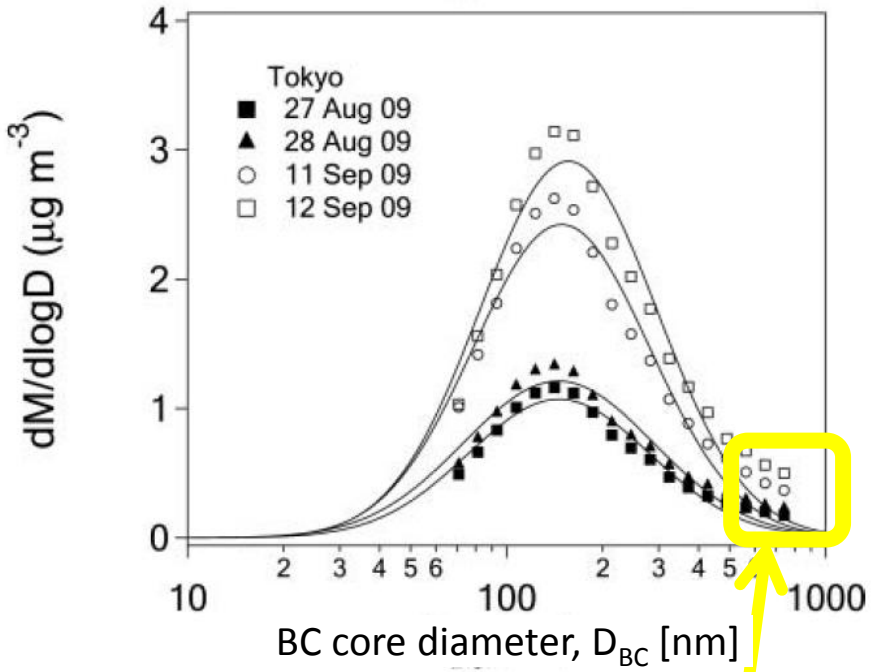
BC core size distributions measured by the SP2

Number

Example from Kondo et al., 2011

Mass





BC number size distribution:

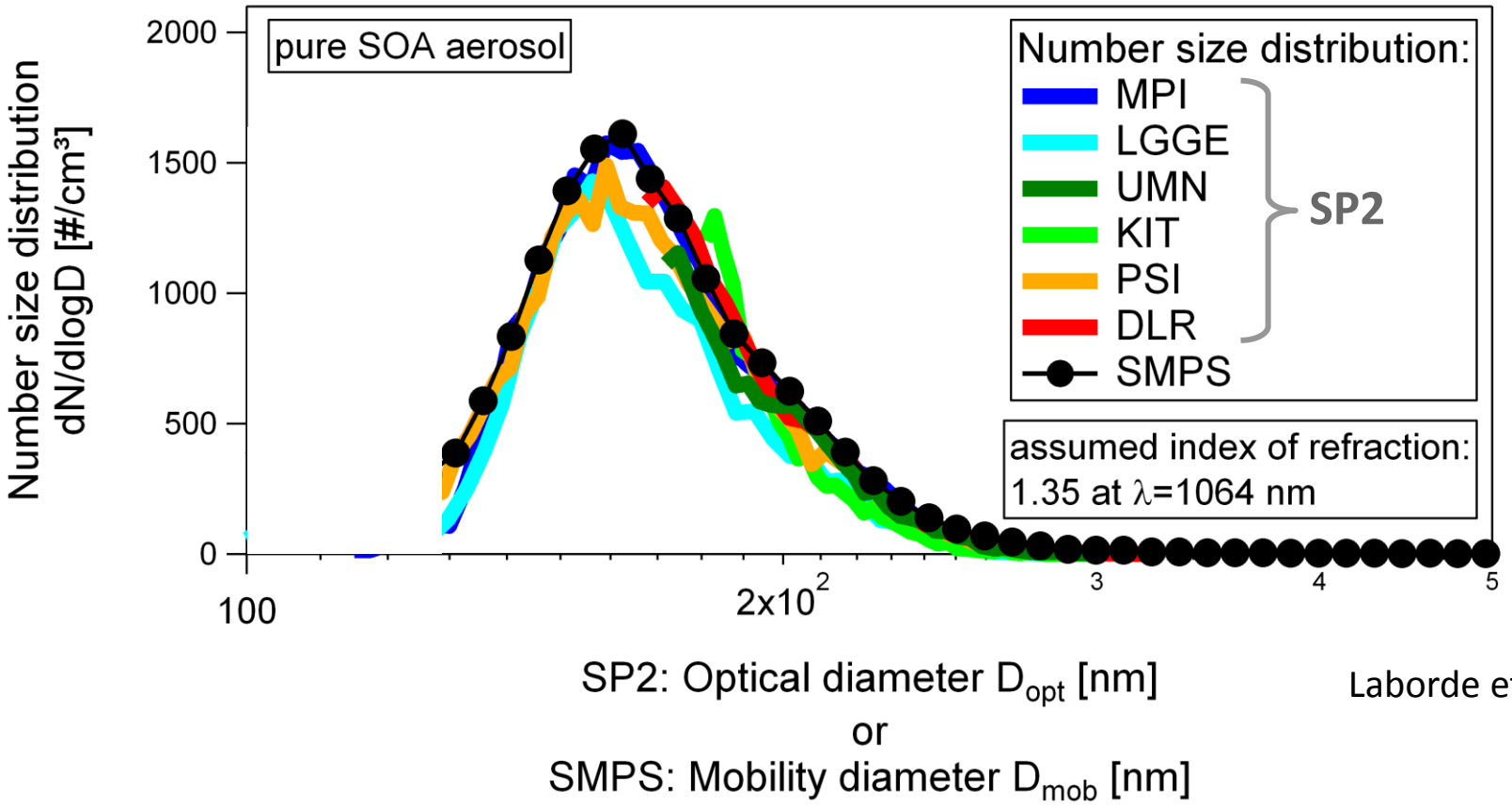
- Mode diameter is near the lower detection limit of the SP2
  ➔ **more than ~50% of the particles are typically below the detection limit**

BC mass size distribution:

- Mode diameter is typically around D_{BC} ≈ 150 nm (between 100 nm and 200 nm)
  ➔ **dominant fraction of submicron BC mass falls within the detection limits of the SP2**
  (this may not be true for fresh exhaust from efficient combustion sources)

Note: the upper detection limit is typically at D_{BC} ≈ 500 – 800 nm, or even larger (depends on settings)

Optical sizing: intercomparison of SP2 with SMPS for secondary organic aerosol



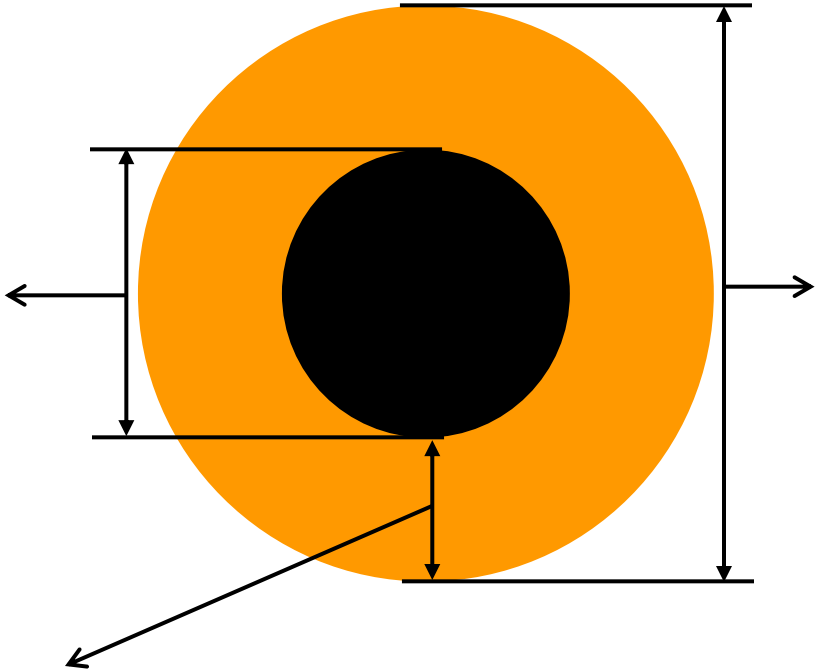
Laborde et al., 2012

Good agreement between SP2 and SMPS ➔ SP2 is also a reliable optical particle sizer

Required for meaningful optical sizing by the SP2:
PSL calibration of detector + Mie theory + known index of refraction of the particles

Black carbon mixing state derived from SP2 data

Incandescence:
BC mass
⇓
BC core diameter
D_{BC}



Light scattering:
Optical diameter
D_{opt}

Coating thickness

$$\Delta_{coat}=0.5\ (D_{opt}-D_{BC})$$

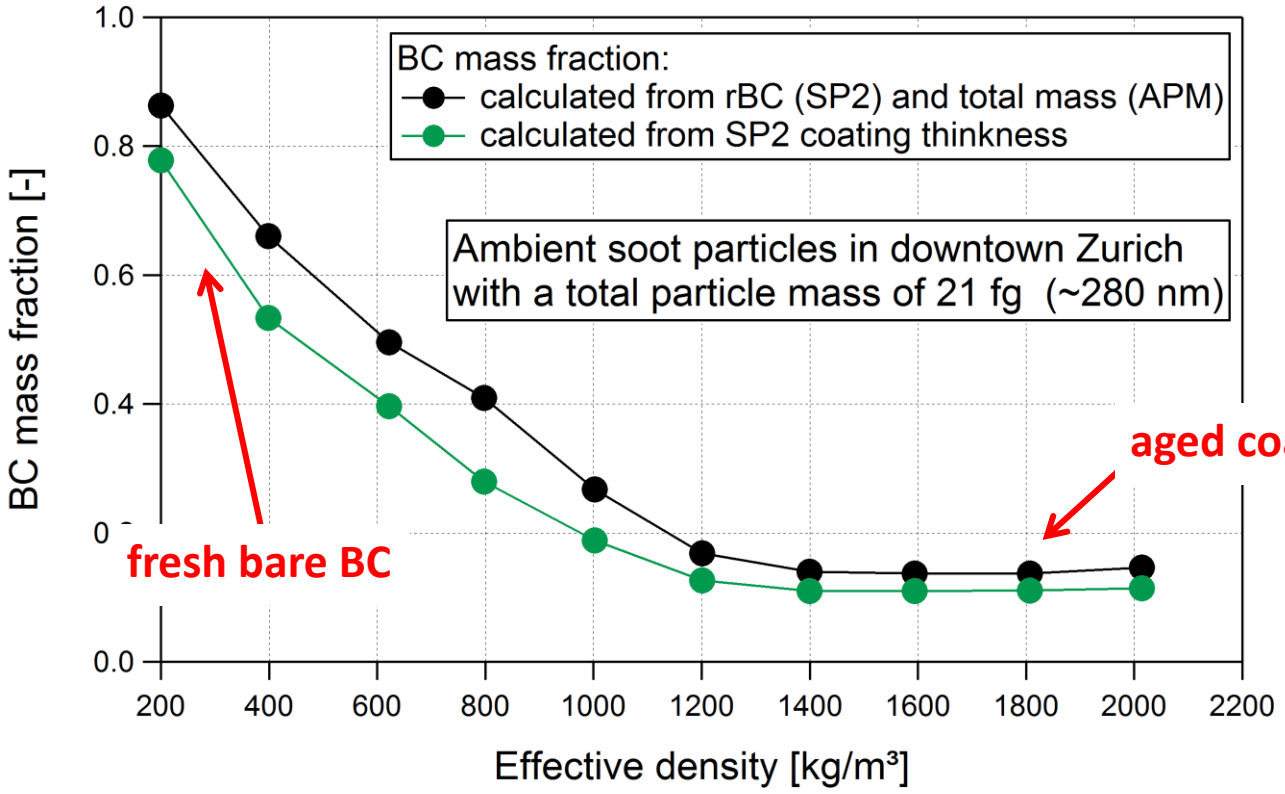
Methodology ("LEO-fit"):

- Gao et al., Aerosol Sci. Technol., 2007
- Schwarz et al., Geophys. Res. Lett., 2008
- Moteki and Kondo. J. Aerosol Sci., 2008
- Laborde et al., Atmos. Meas. Tech., 2012

Concentric spheres core-shell morphology is assumed for inferring the coating thickness

BC mass fraction in soot particles: Comparison of SP2 vs Aerosol Particle Mass Analyser (APM)

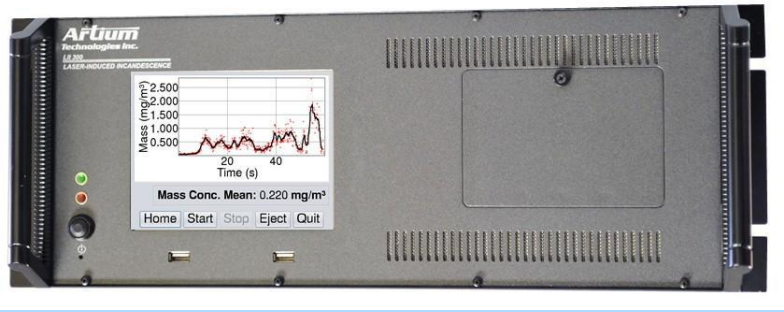
DMA → APM → SP2



unpublished data

➔ **Good agreement for BC mass fraction calculated from SP2 coating thickness with that calculated with total particle mass taken from APM.**
(Quantification of mixing state requires careful calibration and sophisticated data analysis)

“Time-resolved LII”

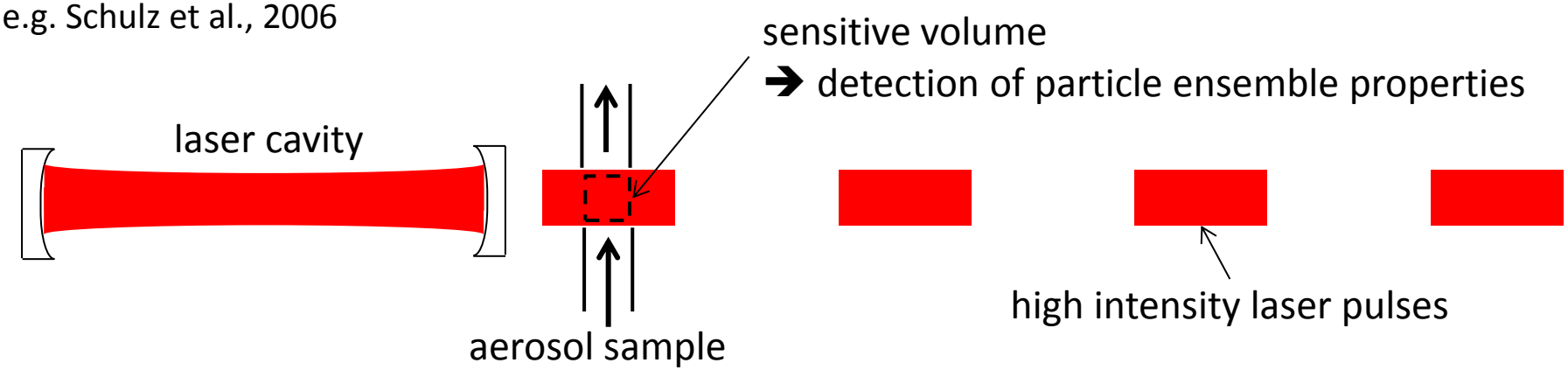


«Time-resolved LII» (e.g. Schulz et al., 2006)**:**

- *Principle*:
  - **Pulsed laser used to heat the particles**
  - Detection of the thermal radiation from all particles in the sensitive volume
    ➔ **BC mass concentration**
  - **No empirical calibration with BC reference material needed (?)** (Snelling et al., 2005)
- *Application*:
  - **Most commonly used for detection of BC in combustion exhaust** in laboratory applications
  - More recently also used for field measurements of BC in atmospheric aerosols
- *Field deployable instruments*:
  - **New high-sensitivity LII 300 from ARTIUM Technologies Inc.**:
    Wide dynamic range for BC mass concentration: < ~0.1 µg/m^3 – 20 g/m^3
    ➔ **just about suitable for atmospheric application, except for clean environments**

“Time-resolved LII” on “bulk samples”

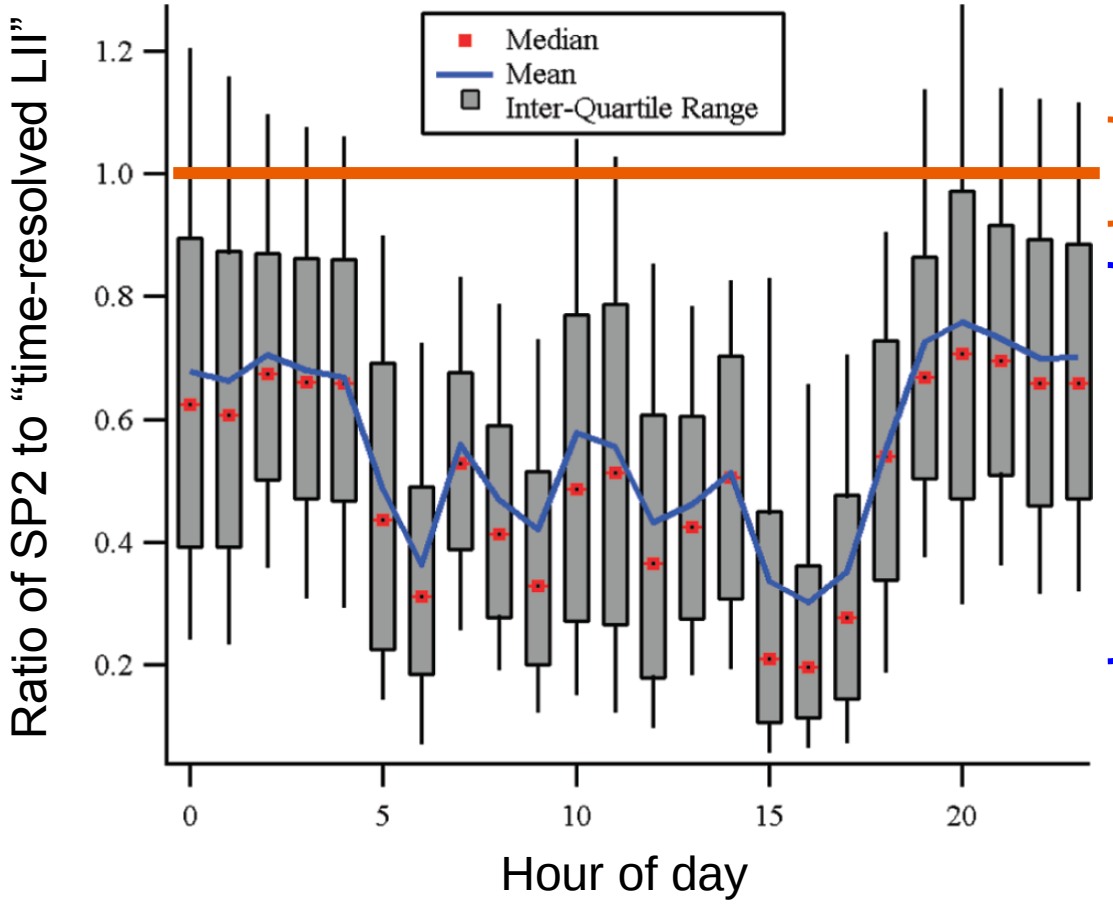
e.g. Schulz et al., 2006



Time-resolved measurement of the thermal radiation emitted after the laser pulse:

- Detection at two different wavelength bands ➔ **particle temperature**
- Detection of thermal emission at known temperature ➔ **BC mass concentration**
  No empirical calibration needed (?) (Snelling et al., 2005)
- Decay curve of thermal emission ➔ **effective primary particle size**
- Extremely high laser fluence
  ➔ **no lower detection limit in terms of BC mass per particle** (for relevant sizes)
- Detection of thermal radiation emitted by all particles in the sensitive volume
  ➔ wide dynamic range and **suitable for high BC mass concentrations**
  ➔ no single particle data

Comparison SP2 vs "time-resolved LII" (atmospheric BC)



agreement would be here

disagreement

Liggio et al. 2012

(Study by Environment Canada and National Research Council Canada)

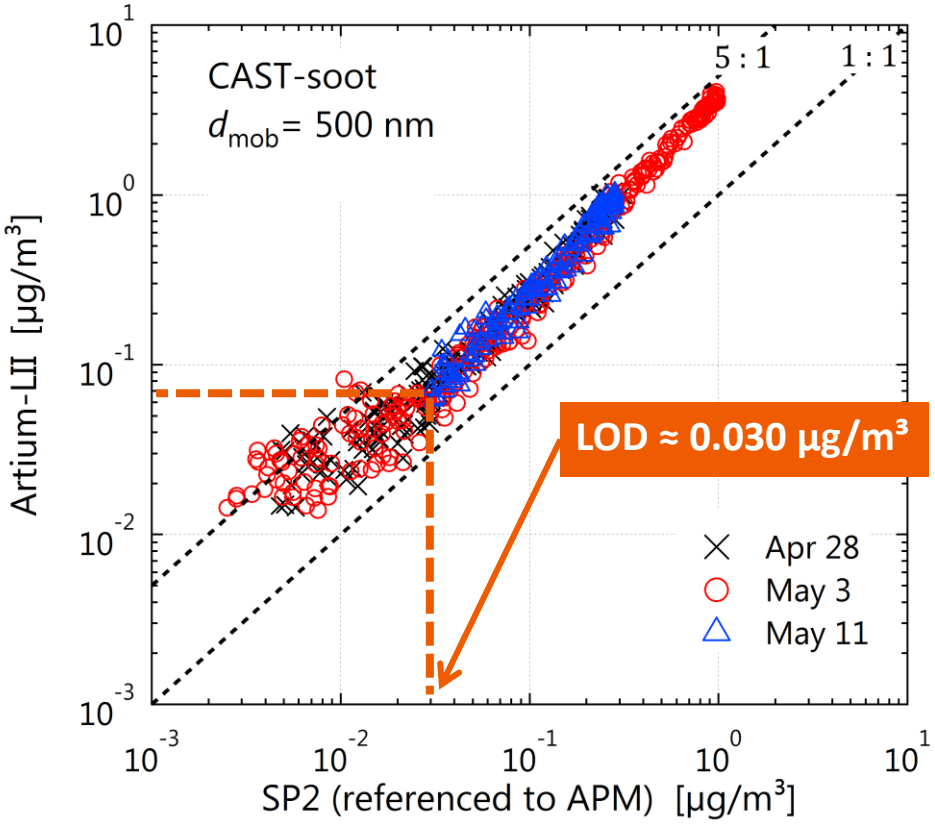
The time-resolved LII instrument and the SP2 **differ by up to a factor 5!**

➔ Difference can only partly be attributed to small BC cores below SP2 detection limit.

➔ **Calibration issues?** (currently my "best guess")

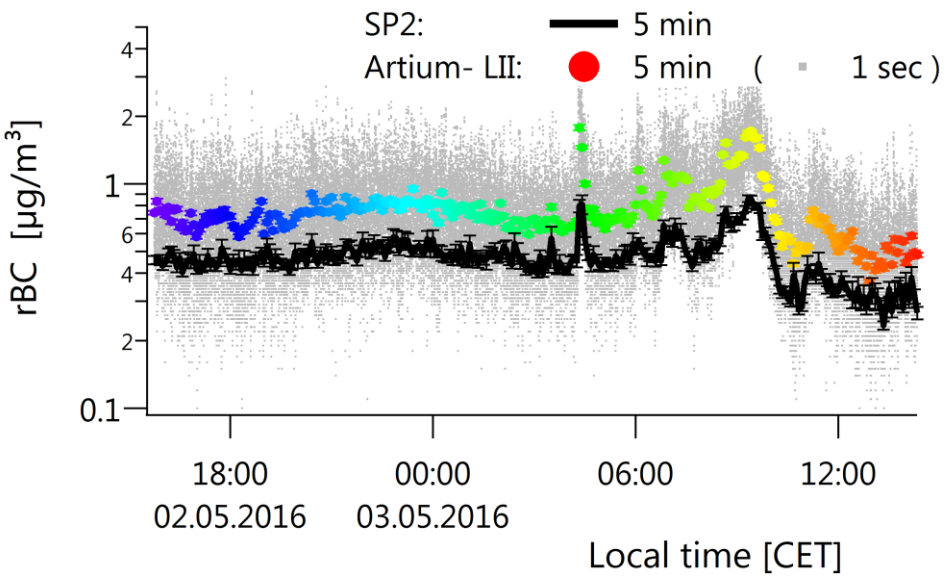
➔ More fundamental methodological problem?

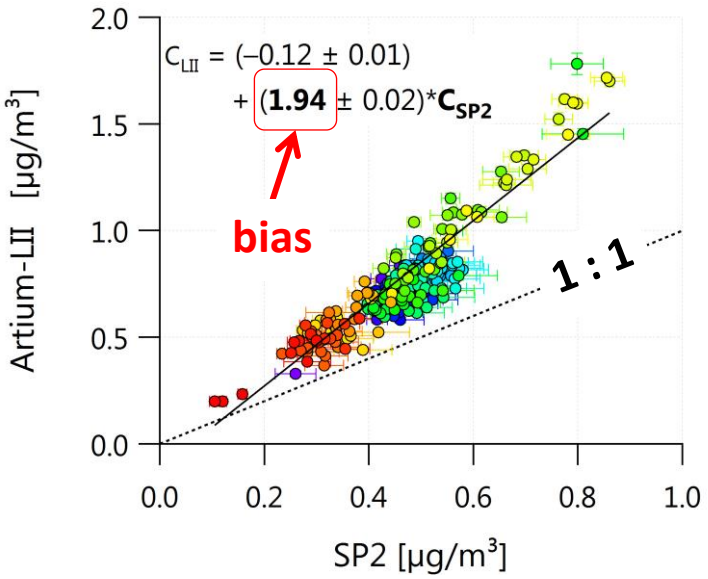
Comparison of Artium-LII 300 with SP2 (size-selected CAST soot)



- **Lower limit of detection of Artium-LII is approx. 0.030 µg/m³**
- **Ratio of Artium-LII to SP2 is far from unity and depends on concentration!?**

Comparison of Artium-LII 300 with SP2 (ambient air)



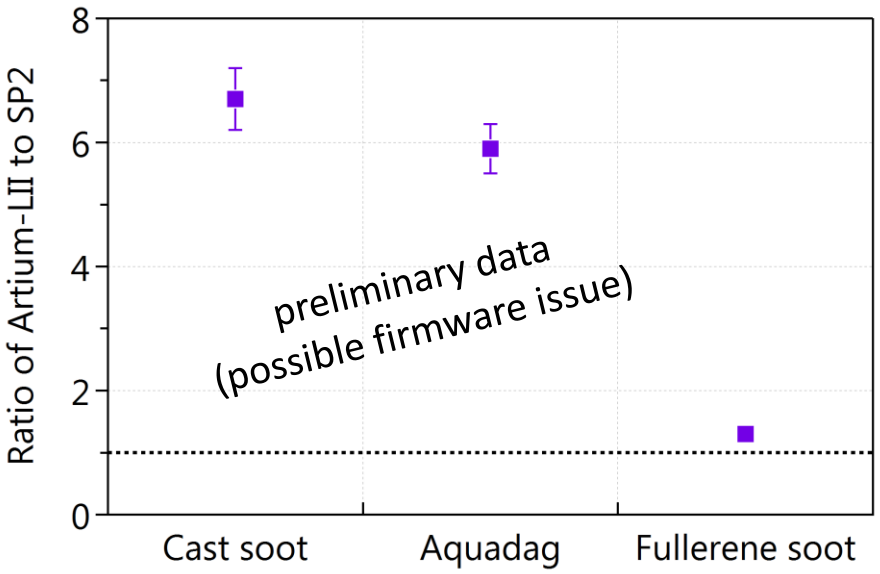


Factor of ~2 difference ➔ calibration and/or data analysis not yet finally settled!?

- **Ratio between the two instruments does not seem to be stable.**
  - ➔ Variable sensitivity to different BC types?
  - ➔ Size effects?
  - ➔ Mixing state effects?

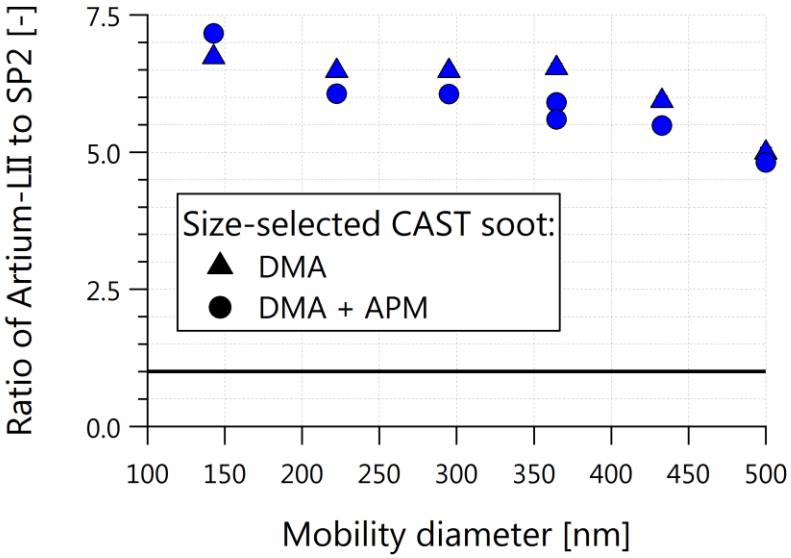
Comparison of Artium-LII 300 with SP2

Different BC types



➔ **Sensitivity of Artium-LII seems to depend on BC type**

Different particle sizes

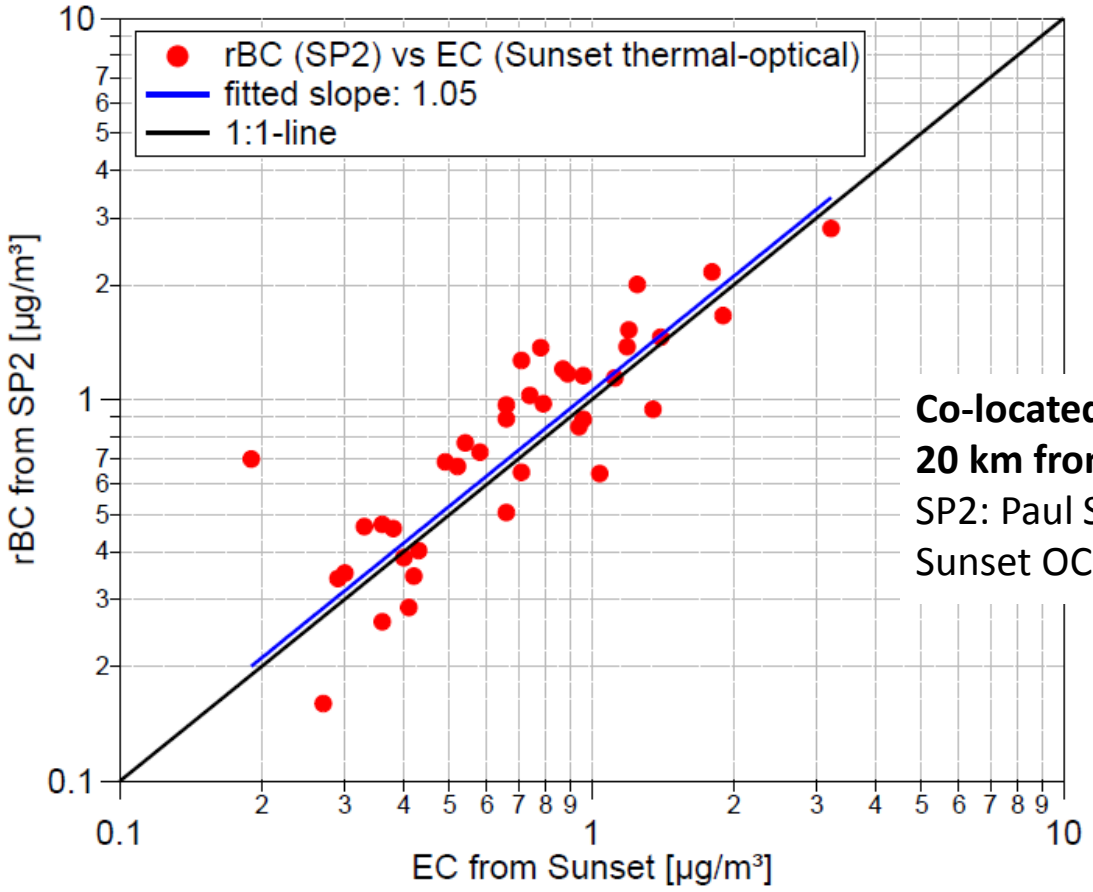


➔ **Sensitivity of Artium-LII seems to depend on BC type**

Note:

- SP2 calibrated against APM for each BC type ➔ reliable reference
- **Artium-LII data analysis approach for new «high-sensitivity»-mode might need some improvement?**

Comparison between SP2 rBC and Sunset EC (thermal-optical method)



Co-located measurements at SIRTA site, 20 km from Paris city centre
SP2: Paul Scherrer Institute
Sunset OCEC analyser: LGGE Grenoble

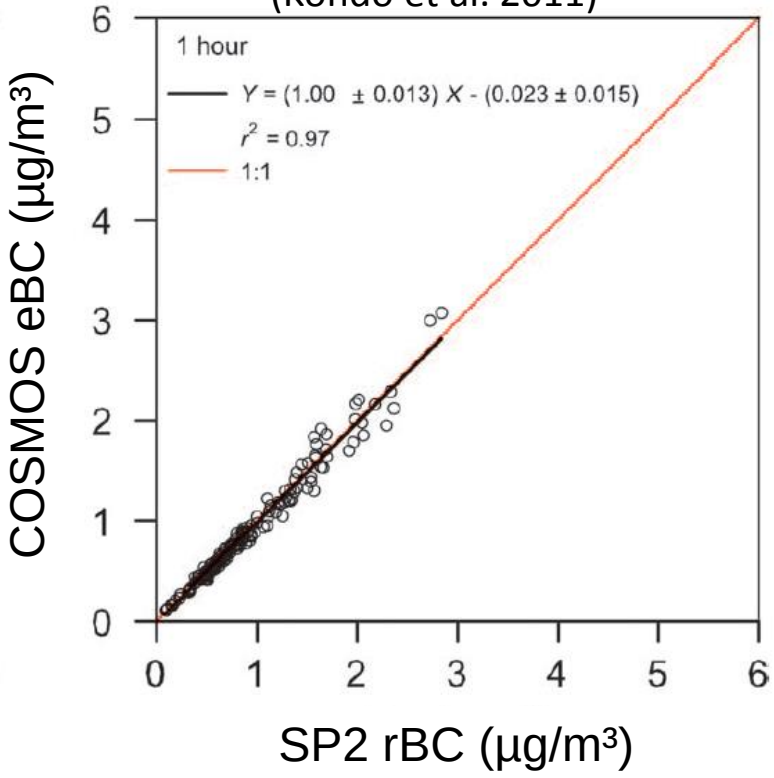
Laborde et al., 2013

SP2 and Sunset OC-EC analyzer (EUSAAR-2I protocol) agree within 5% on average
➔ this is well within the uncertainty of either method

Comparison of COSMOS with SP2 and thermal-optical EC

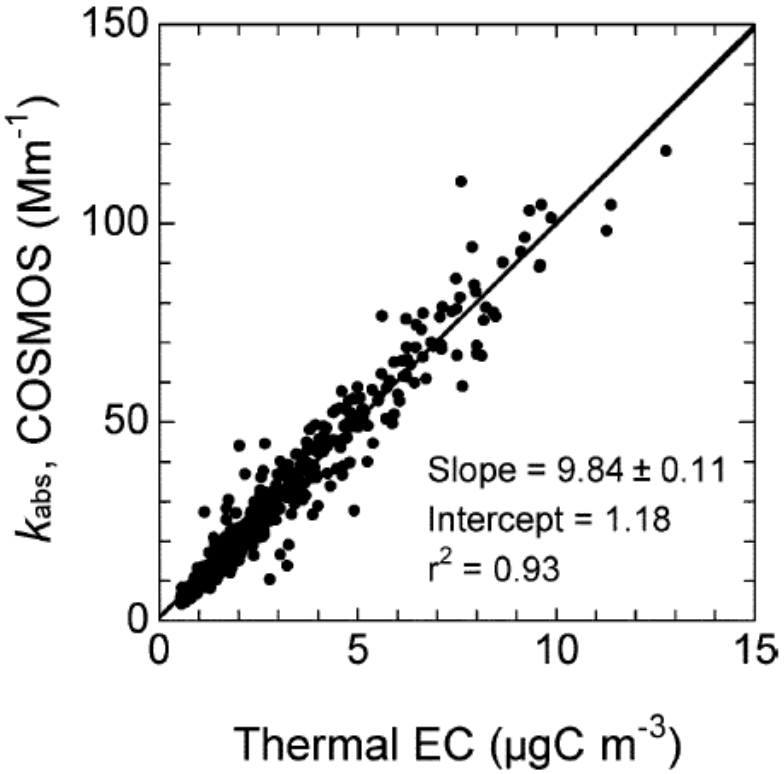
COSMOS vs SP2

(Kondo et al. 2011)



COSMOS vs EC (thermal-optical method)

(Miyazaki et al., 2008)



Consistent results can be achieved between SP2 (rBC), thermol-optical method (EC) and COSMOS (eBC; needs calibration against rBC or EC) (Kondo et al., 2011)

➔ **EC, rBC and eBC are more similar than what one might expect!**

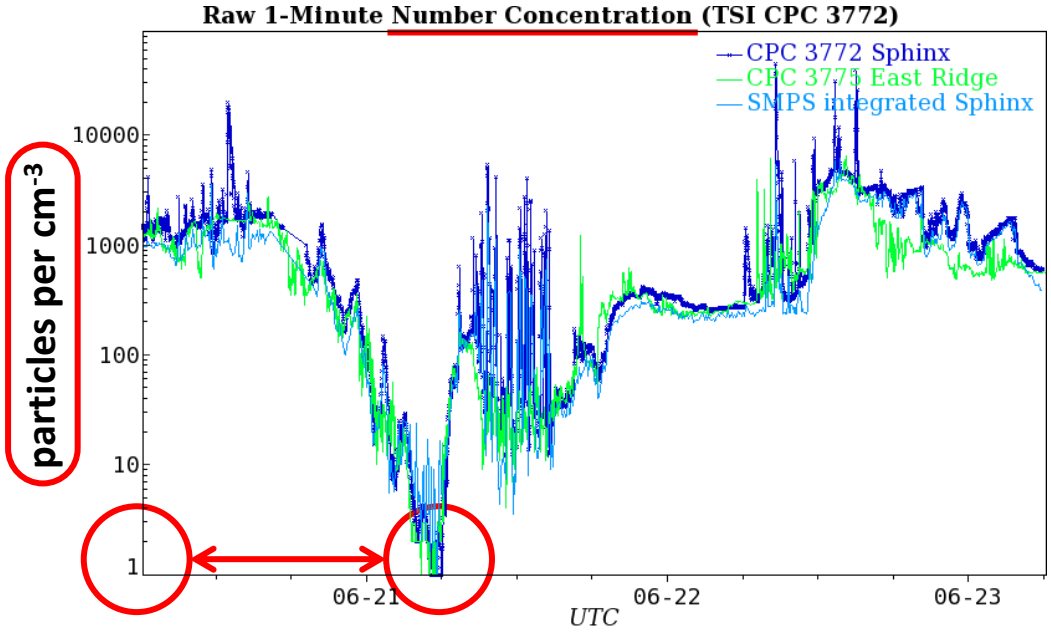
Take home messages

- **The SP2:**
  A highly sensitive instrument that provides additional insight into BC mixing state

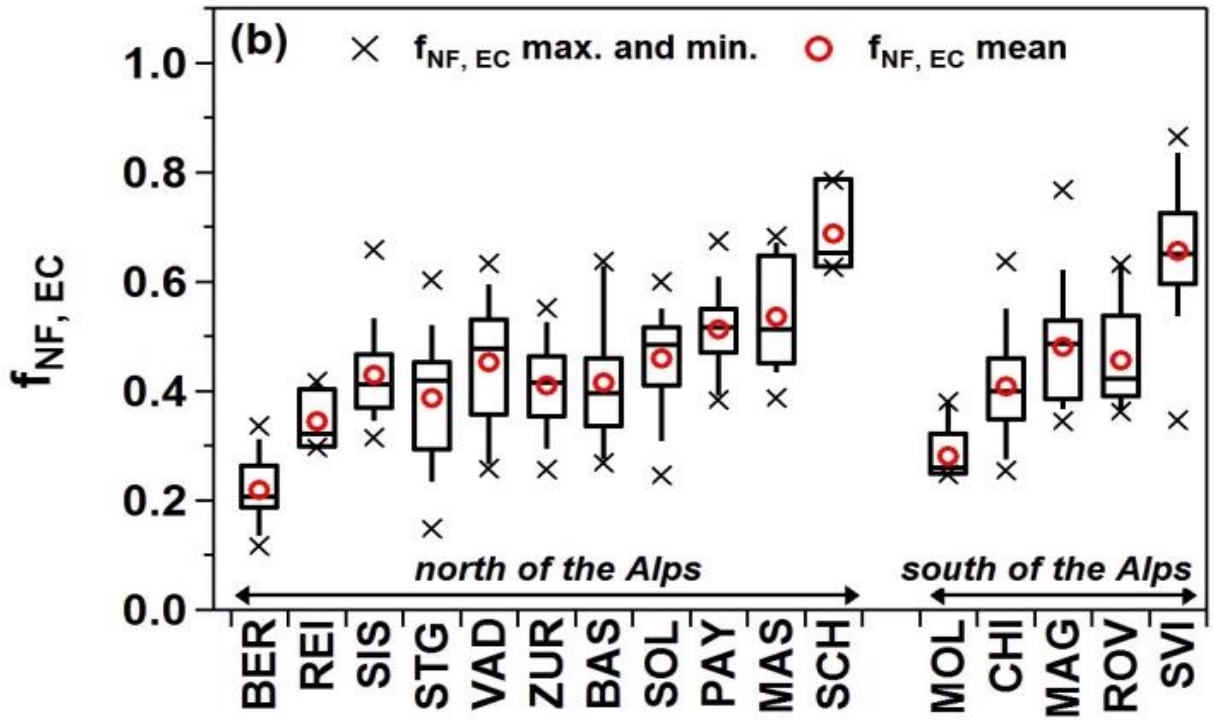
- **Time-resolved LII:**
  Testing Artium LII-300 performance for atmospheric aerosols and comparison with SP2 is "work in progress"

- **rBC, EC and eBC (from COSMOS):**
  More similar than what one might expect
  (or "systematic differences between these quantities might be as small as differences between results obtained by multiple users of the same method")

Don’t miss the Jungfraujoch research station when visiting Switzerland!



Sources of black carbon in Switzerland during winter smog episodes



Zotter et al., 2014

Fraction of non-fossil (wood burning) elemental carbon:

- 40-50% for most stations in Switzerland during winter smog episodes
- >80% at extreme locations (valleys in the Swiss alps)

Source apportionment of black carbon

Research question:

- Is source apportionment based on spectral dependence of absorption possible?

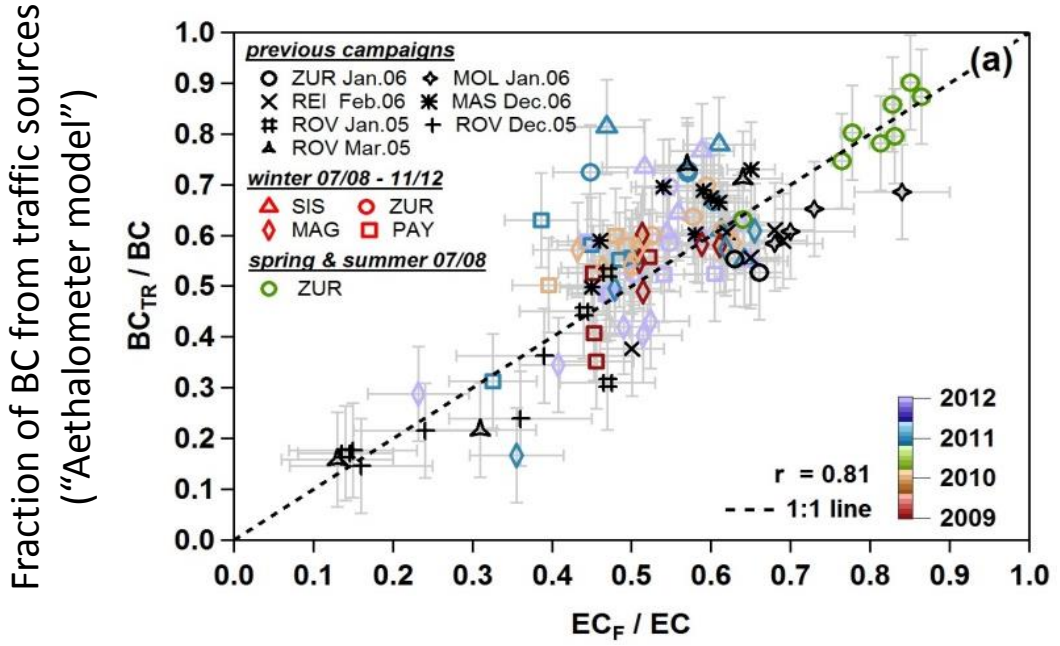
Radiocarbon method ("^{14}C method"):

- Differentiates between fossil and non-fossil sources **based on the ratio of ^{14}C to ^{12}C isotopes**.
- Key limitations: low time resolution and large analytical effort.
- e.g. Szidat, Chimia, 2009

"Aethalometer model":

- Based on different **spectral dependence of absorption coefficient** of BC from different sources.
- Key limitations: can only work for 2 sources if at all
- Sandradewi et al., Environ. Sci. Technol. 2008

Comparison of radiocarbon method with "aethalometer model" (Zotter et al., in prep.)



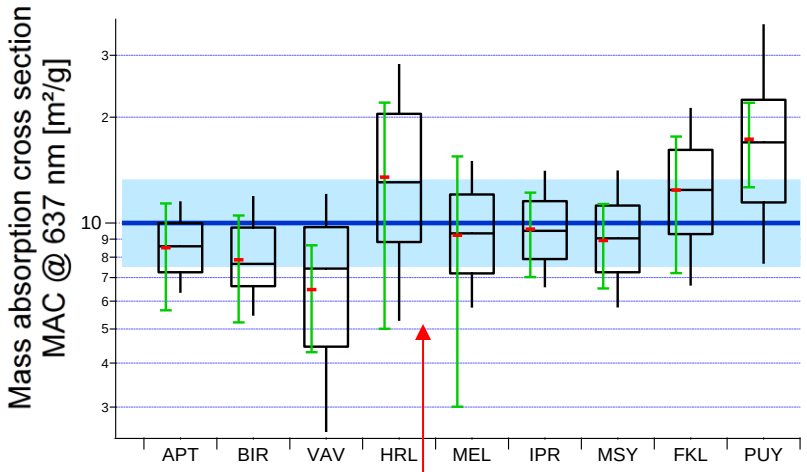
The "aethalometer model" compares well with the ^{14}C method:

- mean bias: <2%
- average precision (1σ): <10%

Caveats:

- For Switzerland, where traffic and wood burning are the only two sources.
- **Only after using updated parameters for aethalometer model.**

Mass absorption cross section (MAC) of black carbon



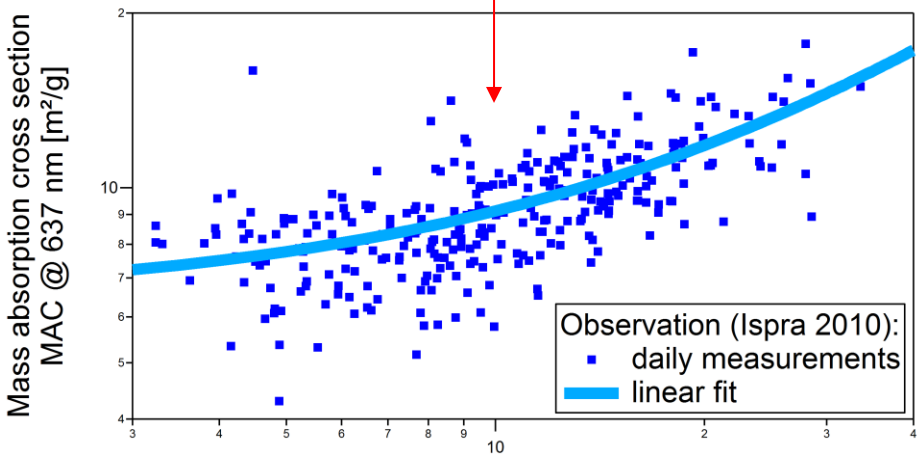
Sites of the ACTRIS research infrastructure

Only moderate variability of MAC at European background sites:
MAC_{BC} = 10.0 ± 30% m^2/g @ 637 nm

The MAC relates BC mass with absorption coefficient
Relevant for:
- modelling aerosol-radiation interactions
- absorption based black carbon measurements

Open research questions:
- What is the MAC of atmospheric BC?
- Does the MAC depend on BC mixing state?

Evidence for "lensing effect"



Zanatta et al., Atmos. Env., in review

Are black carbon particles important heterogeneous ice nuclei?

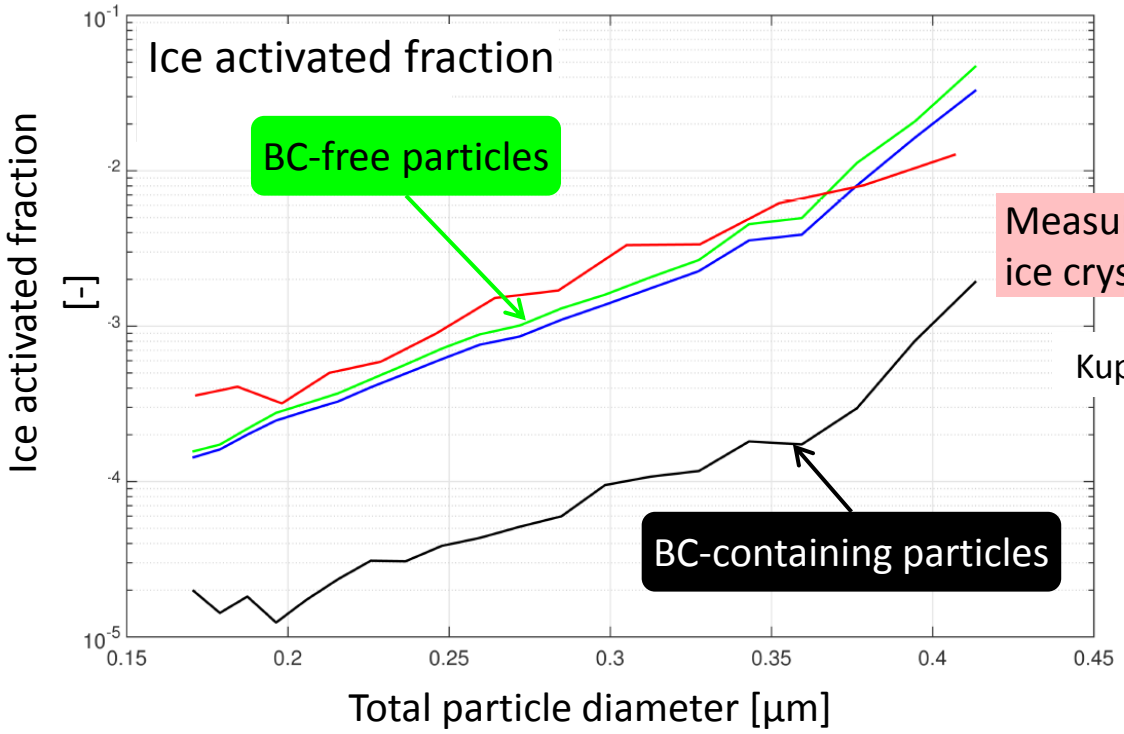
Motivation:

Heterogeneous ice nuclei can massively change the properties of supercooled clouds

Open research question:

Controversial results in existing literature concerning importance of black carbon

mostly anthropogenic!



Measurement of BC in residual particles of ice crystals in mixed-phase clouds

Kupiszewski et al., J. Geophys. Res., in review

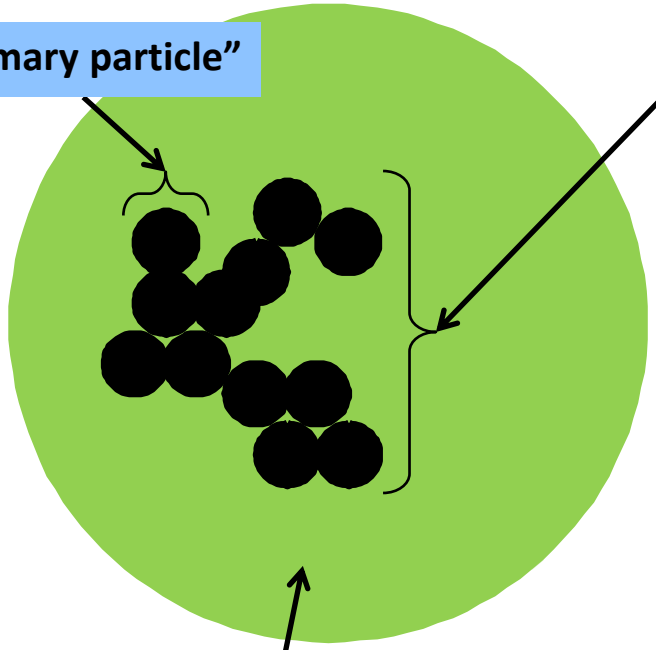
BC-containing particles depleted by more than one order of magnitude compared to BC-free particles

➔ Black carbon particles are unimportant for glaciation of clouds observed at the Jungfraujoch

Terminology for “black carbon” particles

“black carbon containing” particle
(often called soot particle)

“primary particle”



«coating»
organic carbon and/or inorganic salts

«BC core» = aggregate of primary particles, composed of black carbon (elemental carbon, …)

- almost «pure carbon» (>90% C)
- graphitic microstructure with some disorder
- fractal-like aggregate of primary spheres
- insoluble in any solvent
- strongly light-absorbing («black»)
- extremely refractory (T_{sublim} ≈ 4000 ^{o}C)

Recommendations for terminology:
Petzold et al. (2013) for terminology

«Single particle continuous-wave LII»

«Single particle continuous-wave LII» (e.g. Schwarz et al., 2006)**:**

- *Principle*:
  - Continuous-wave intra-cavity laser used to heat the particles
  - Detection of peak thermal radiation emitted by individual particles at sublimation point
    ➔ **single particle properties**
  - Jet nozzle used to direct particle beam through the laser
    ➔ **100% detection efficiency** (within given BC particle size limits)
  - **Empirical calibration needed**
- *Application*:
  - **Extremely sensitive**, as every **single particle** is detected
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  - **Provides additional information on BC mixing state**
  - Also applied for detection of BC in ice core, snow and rain samples (e.g. McConnell et al., 2007; Schwarz et al., 2012; Ohata et al., 2011; Wendl et al., 2014)
- *The only commercially available instrument* (to my knowledge):
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Artium LII-300 schematic

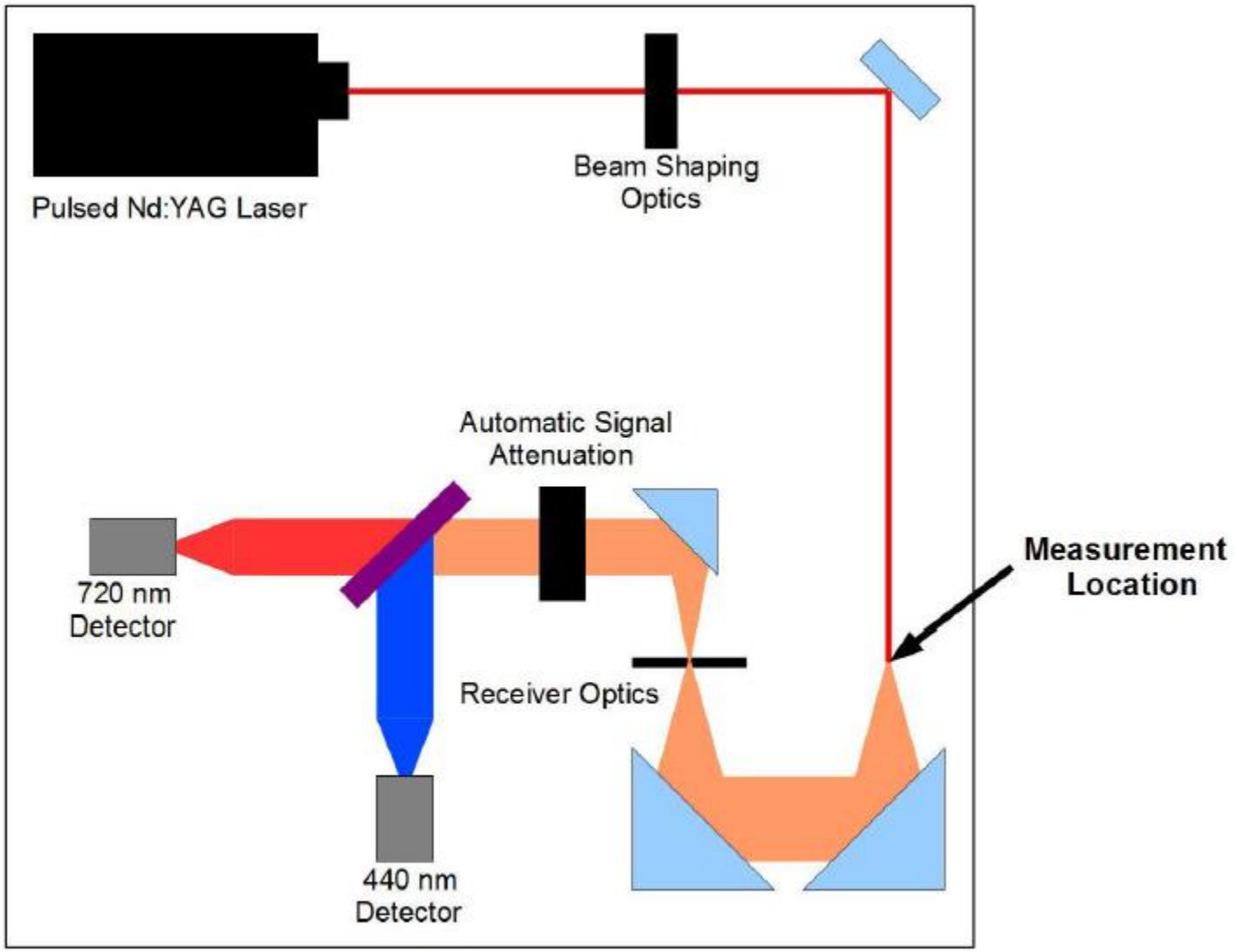
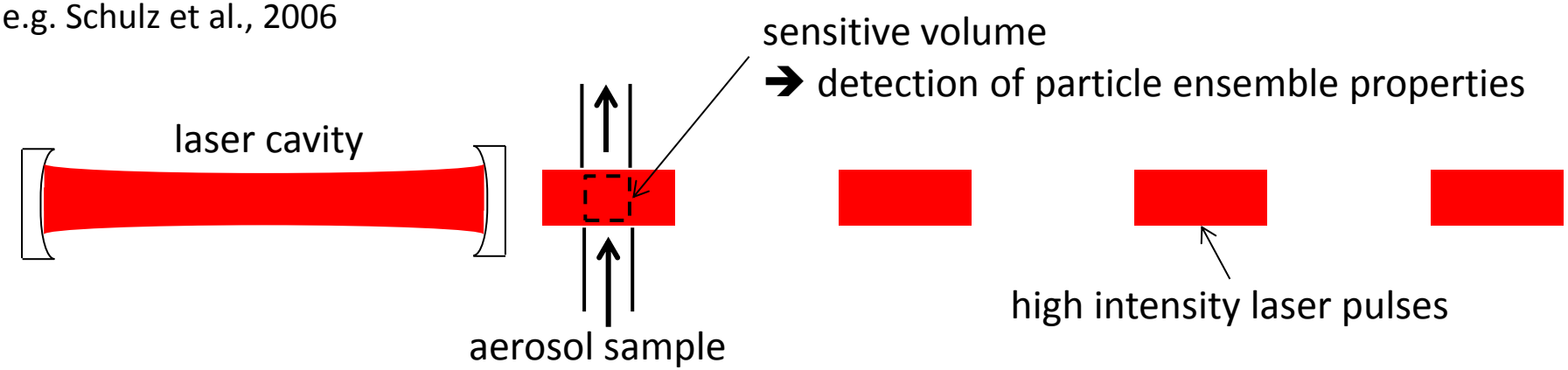


Figure 10.1: Schematic Layout of the LII 300.

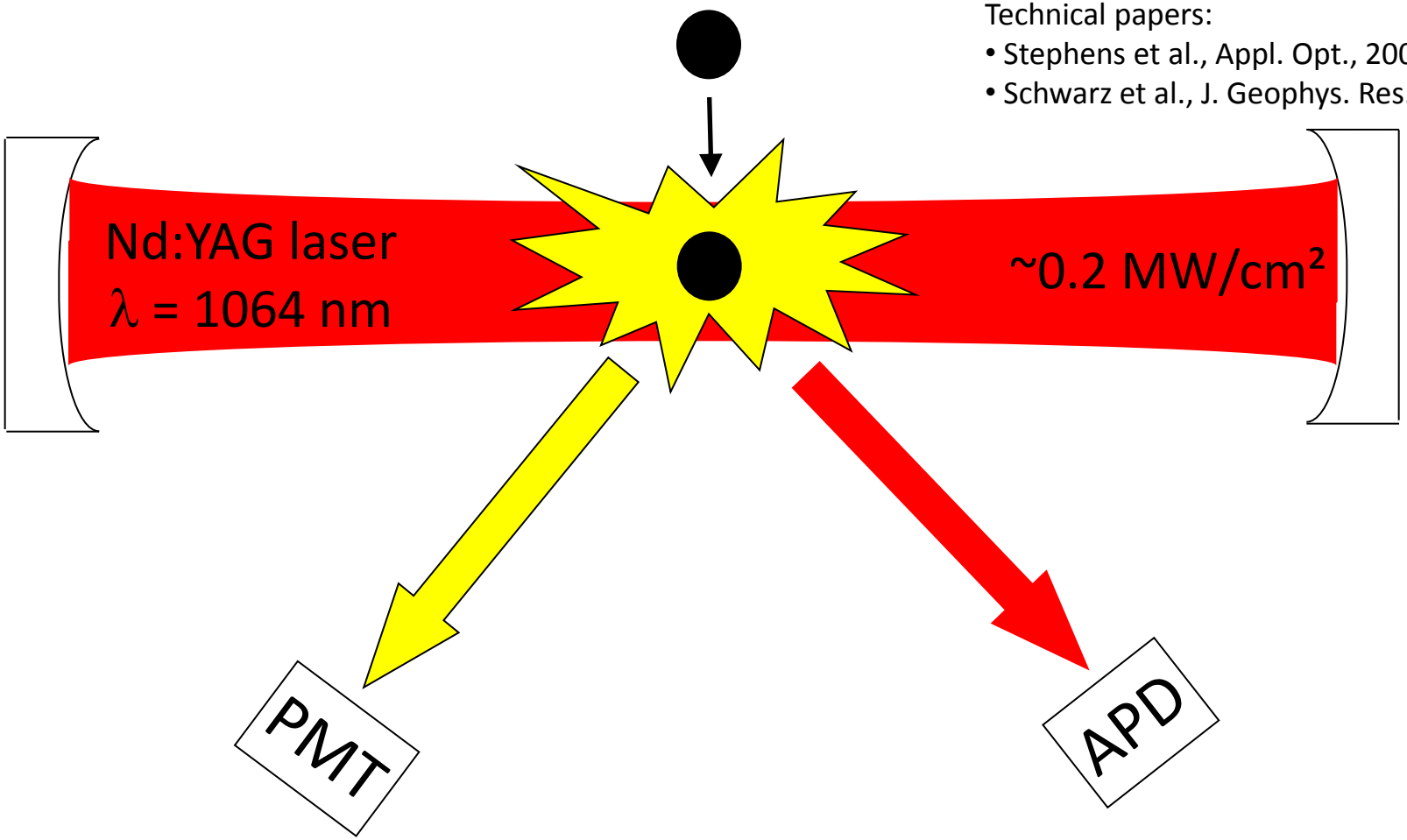
“Time-resolved LII” on “bulk samples”

e.g. Schulz et al., 2006



Single Particle Soot Photometer (SP2)

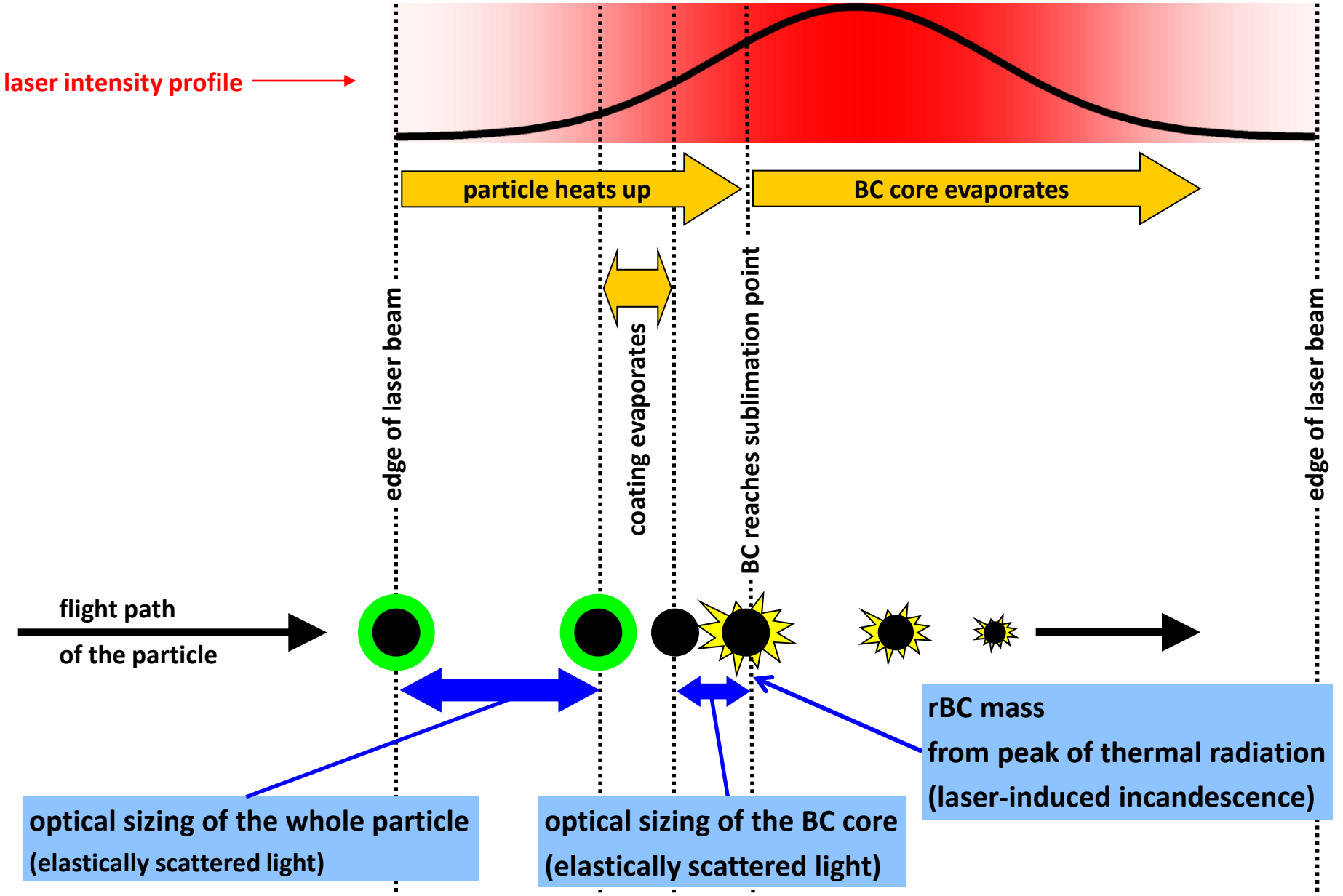
Technical papers:
- Stephens et al., Appl. Opt., 2003.
- Schwarz et al., J. Geophys. Res., 2006.



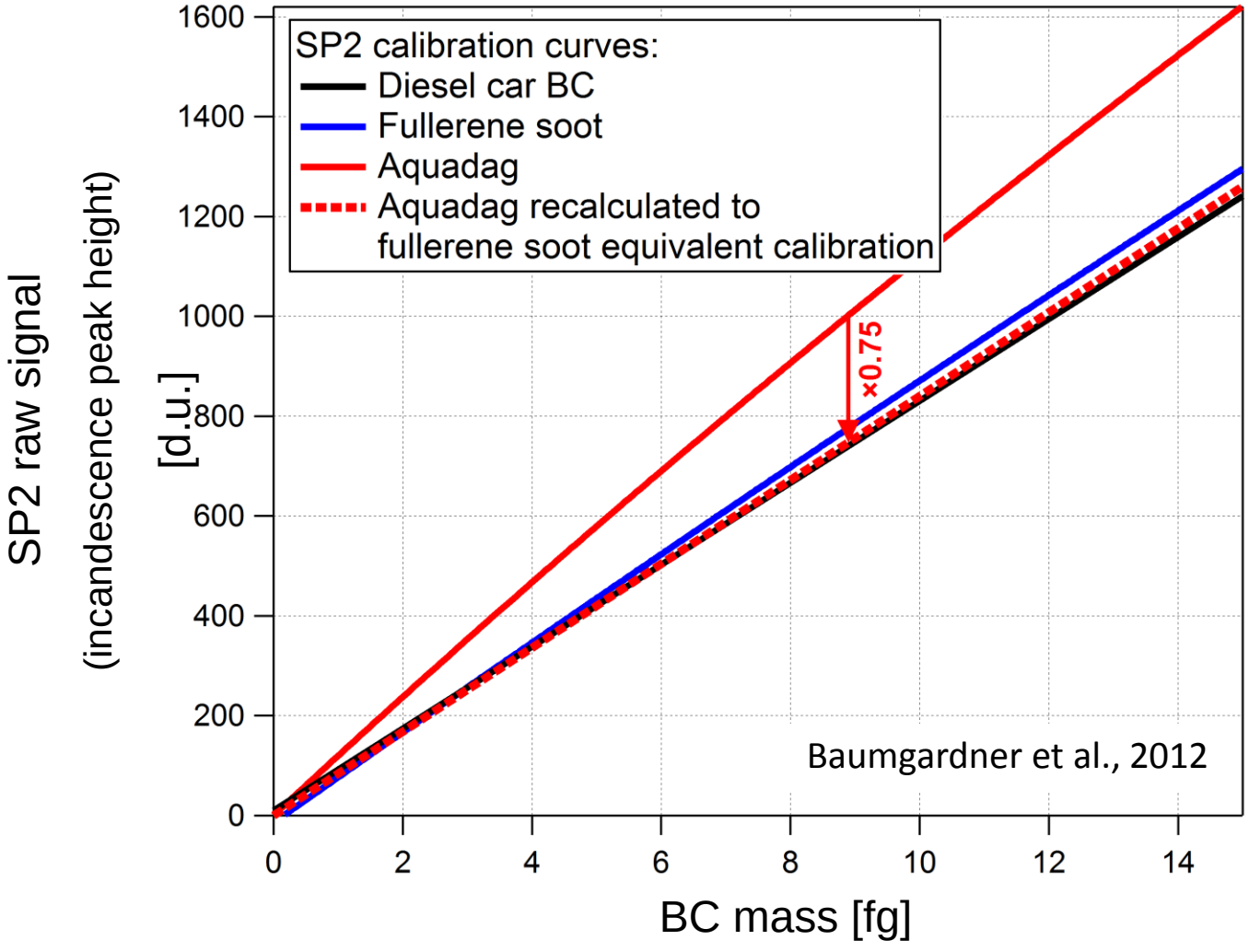
detection of
thermal radiation
➔ **BC mass**

detection of
scattered light
➔ **optical sizing**

SP2 signals of a coated BC particle



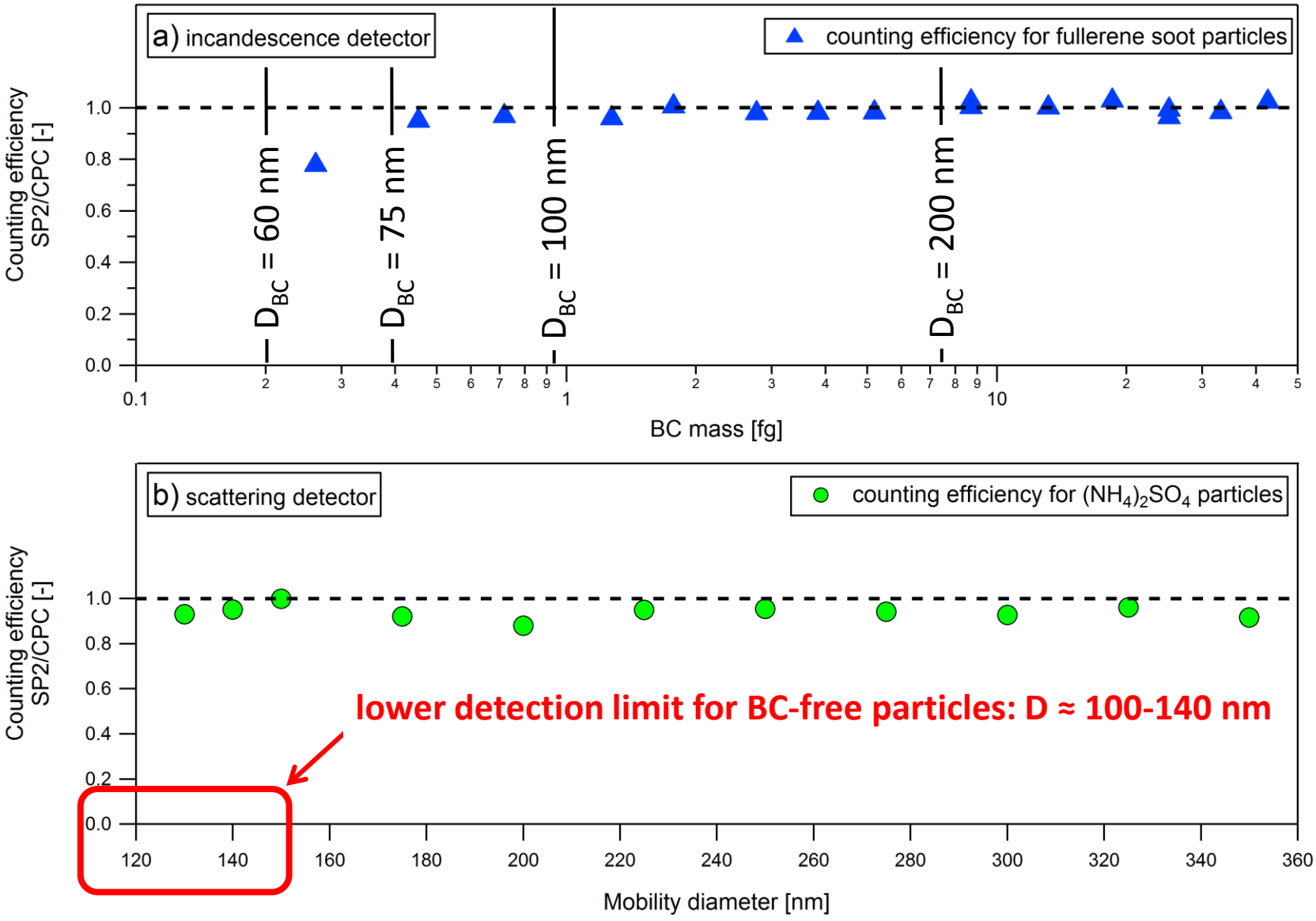
Calibration standard for the SP2 and associated uncertainty



➔ **Calibration standard: fullerene soot** (batch FS12S011) **or Aquadag** (needs recalculation)

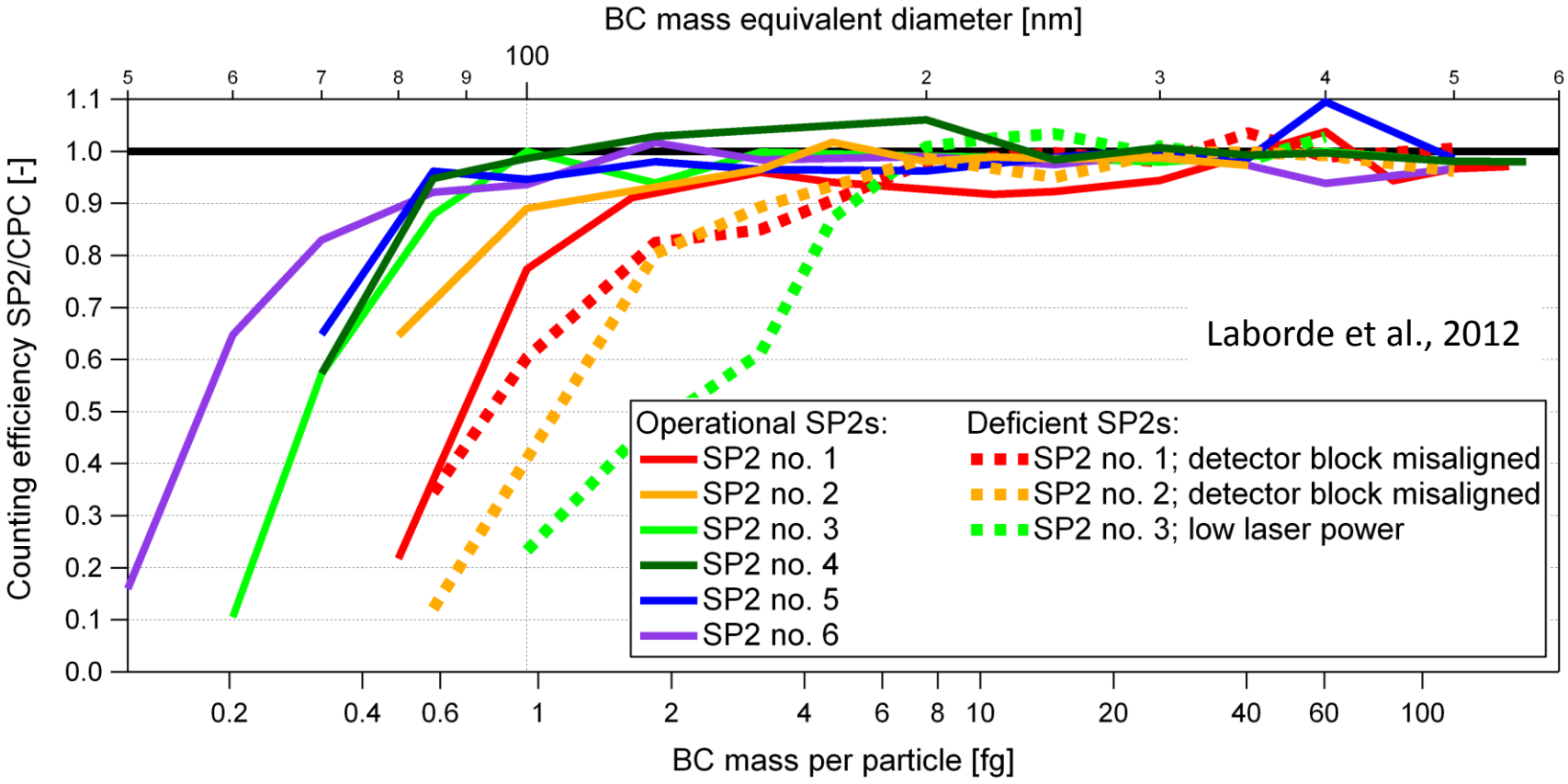
➔ **Absolute BC mass calibration uncertainty for atmospheric aerosols: ~±20-30%**
(can occasionally be larger or smaller for certain soot aerosol types)

Counting efficiency of the SP2



➔ **100% counting efficiency** (above lower detection limit; "no" upper limit for counting)

Lower detection limit of the SP2 for BC-containing particles



- **Lower detection limit for BC: m_{rBC} ≈ 0.3-1.0 fg BC / D_{BC} ≈ 70-100 nm** ("no" upper limit) (see Schwarz et al., 2010; Laborde et al., 2012).
- **Careful preparation of SP2 is critical for detection of particles with small BC mass!**
- The above detection limit applies for sufficiently compact particles. **PALAS soot is not detected by the SP2 even at 2 fg BC per particle** due to its extremely low effective density**!** (Gysel et al., 2012).

«Concentration detection limits» of the SP2

- **«No» lower concentration detection limit** (in the absence of leaks)
- **Coincidence imposes upper concentration detection limit**

Optical sizing:
- Coincidence occurs at particle number concentrations above ~1'000-3'000 cm^{-3} (with D_{opt}> ~120 nm) (this can be influenced within certain limits via instrument settings)

BC number concentration:
- Coincidence can be avoided up to at least ~10'000 cm^{-3}, when skipping the optical sizing
- When optical sizing is done, then the limit for optical sizing becomes relevant

BC mass concentration:
- Above coincidence limits translate to BC mass concentrations of ~1-10 µg m^{-3}
  ➔ **coincidence can become an issue in polluted environments**
- However, the bias due to coincidence is smaller for "BC mass" than for «BC number», as mostly small BC particles with negligible mass are lost

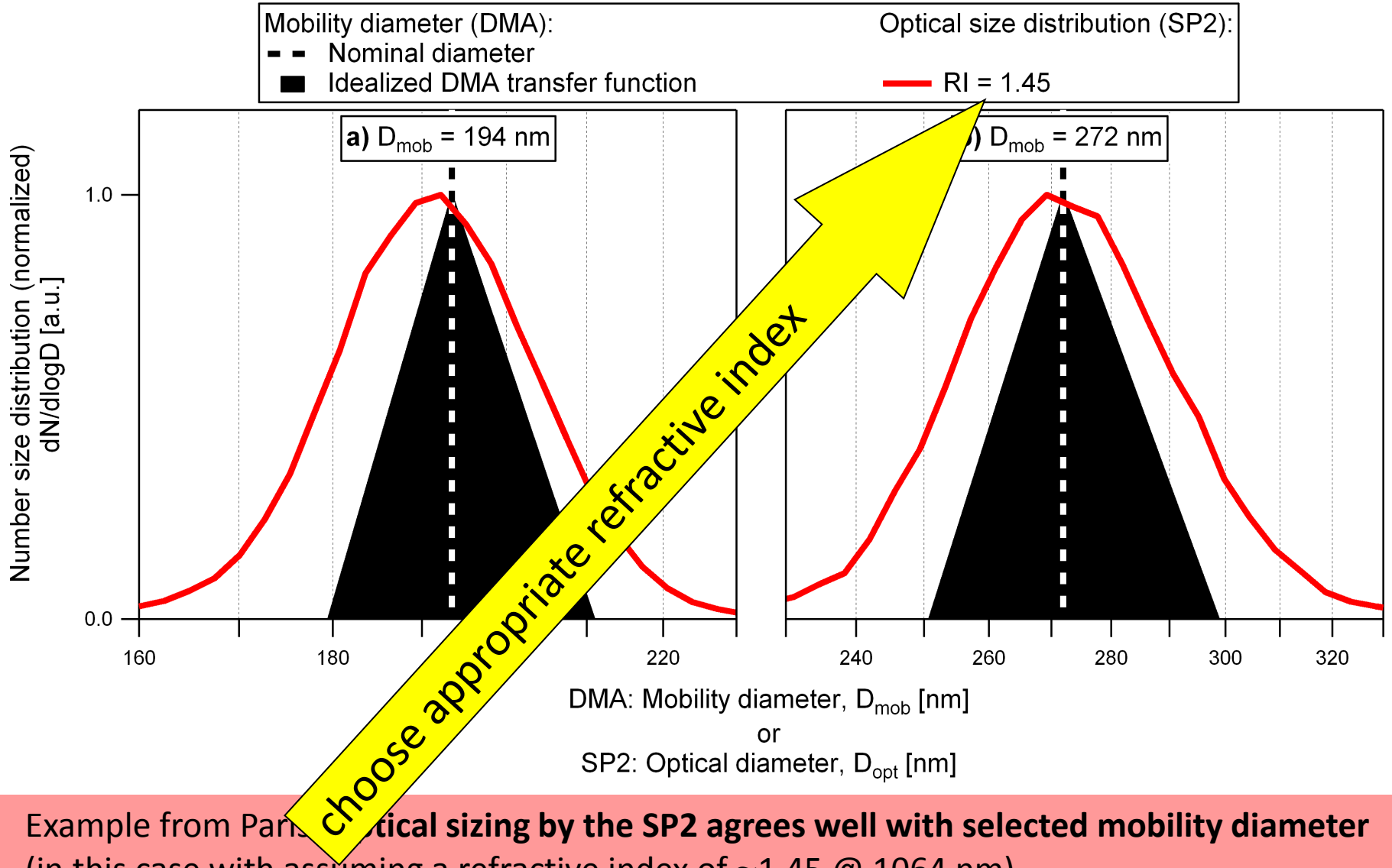
BC mixing state information:
- Coincidence is a critical issue for reliable mixing state measurements
  ➔ number concentration of particles above lower size detection limits of the SP2 must be kept below around ~1'000-3'000 cm^{-3}

Note: these numbers are all just a very crude guess!

Optical sizing of BC-free particles

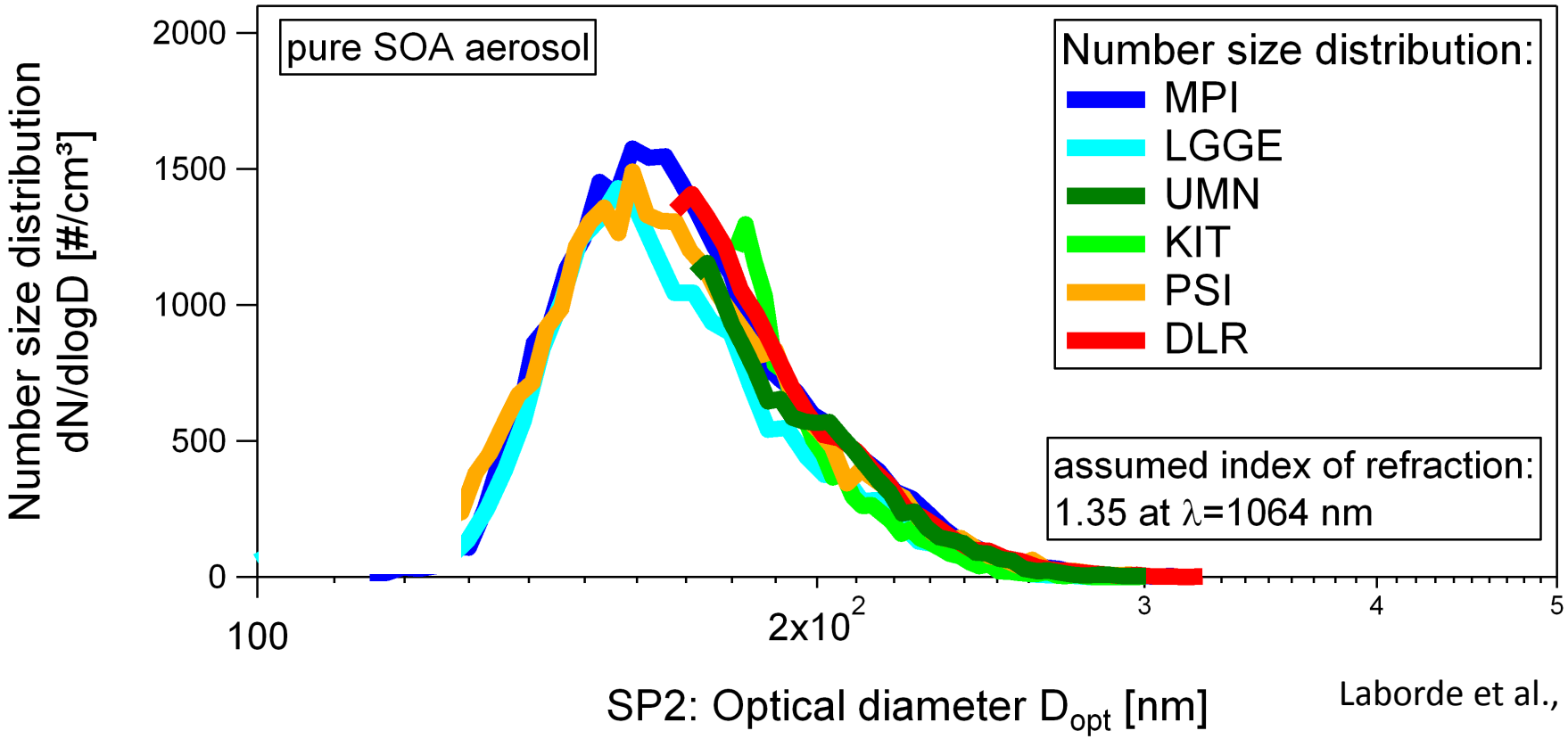
- **The elastically scattered laser light is used for optical sizing**
- **Absolute calibration** of measured (partial) scattering cross section is done **with certified PSL spheres** (only 1 size needed)
- **Mie theory for homogenous spheres** is used to infer optical diameters from measured scattering cross sections
  ➔ optical diameters are only meaningful when using appropriate index of refraction
- Measurement of size-selected ambient aerosol can be used to determine the appropriate index of refraction

Optical sizing of BC-free particles: Comparison of SP2 with DMA (mobility diameter)



Example from Pari**tical sizing by the SP2 agrees well with selected mobility diameter**
(in this case with assuming a refractive index of ~1.45 @ 1064 nm)

Optical sizing: Intercomparison of multiple SP2s for secondary organic aerosol



Laborde et al., 2012

➔ **SP2 instruments agree well with each other**

SP2 signals used to derive mixing state

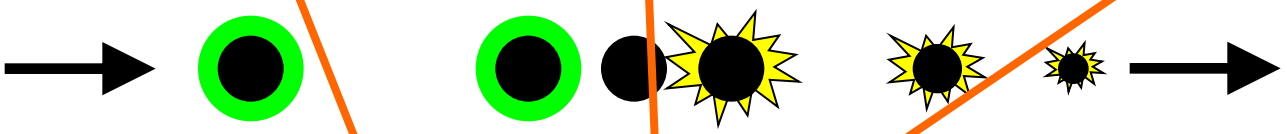
laser intensity profile →

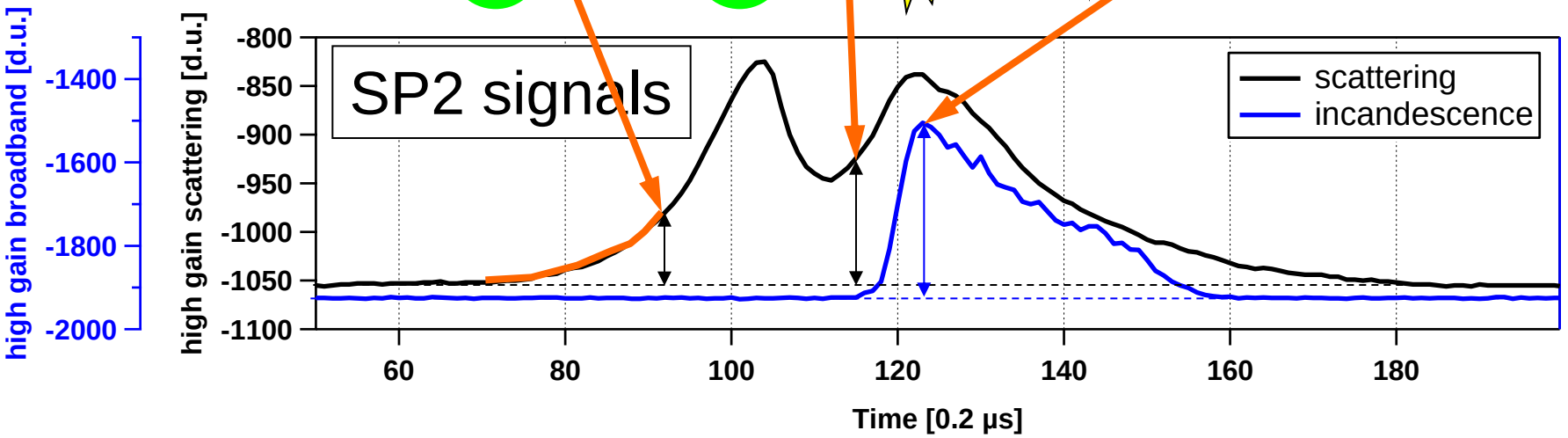
optical sizing of the bare BC core

cross-check between mass and optical size of the bare BC core

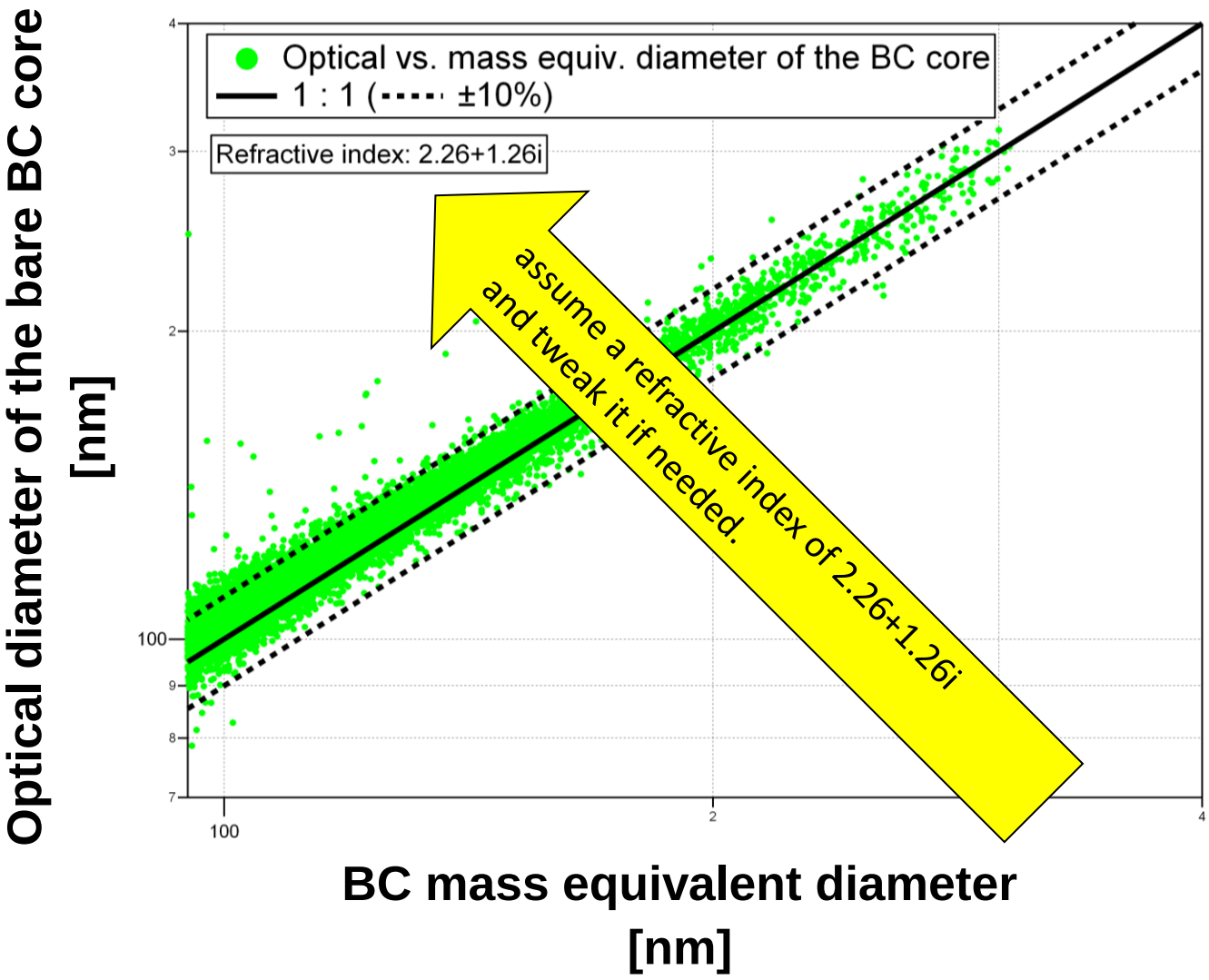
optical sizing of total particle (LEO-fit)

BC mass measurement





Optical sizing of the bare BC core



Excellent agreement between mass equivalent and optical diameter of the bare BC core
➔ this ensures correct coating thickness measurement for uncoated BC particles.

«BC» mass fraction in the soot particles: Comparison of SP2 vs Sunset analyser

«BC» mass fraction of CAST soot determined with:

- **SP2** (calculated from measured coating thickness)
- **Sunset analyser** (thermal-optical method; NIOSH 5040)

Date/time	$\varepsilon_{EC} \approx$ EC/TC (Sunset)	ε_{BC} (SP2)
23 Nov. 2010 10:19	0.38	Not available
23 Nov. 2010 16:50	0.36	0.3
24 Nov. 2010 09:01	0.39	0.35

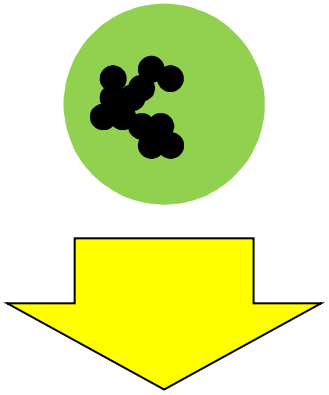
Laborde et al., 2012

➔ **Good agreement for "BC" mass fraction determined with SP2 and Sunset analyser**

Morphology of internally mixed BC particles

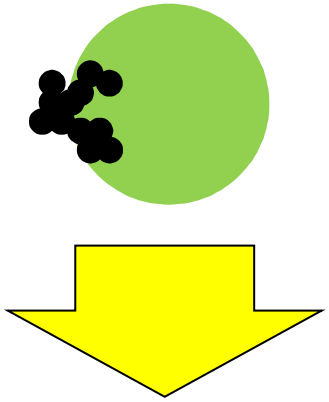
Some BC-containing particles disintegrate into the incandescing BC core and a BC-free chunk when crossing the SP2 laser
(Sedlacek III et al., 2012; Dahlkötter et al., 2014; Moteki et al., 2014)

BC core fully embedded in the «coating»



No disintegration (except for extreme coatings)
➔ **complete (substantial) coating evaporation**

"BC core" attached to the «coating»



BC disintegrates from «coating» in the SP2 laser
➔ **almost no evaporation of the coating**

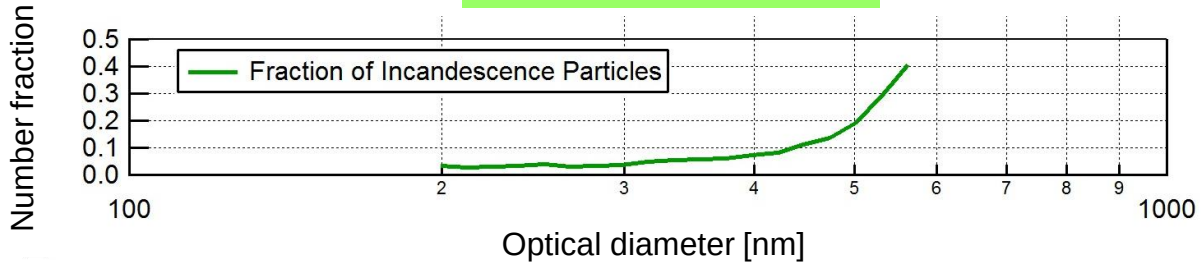
Shown by Moteki et al., 2014

➔ **The SP2 makes it possible to distinguish between these two morphologies.**

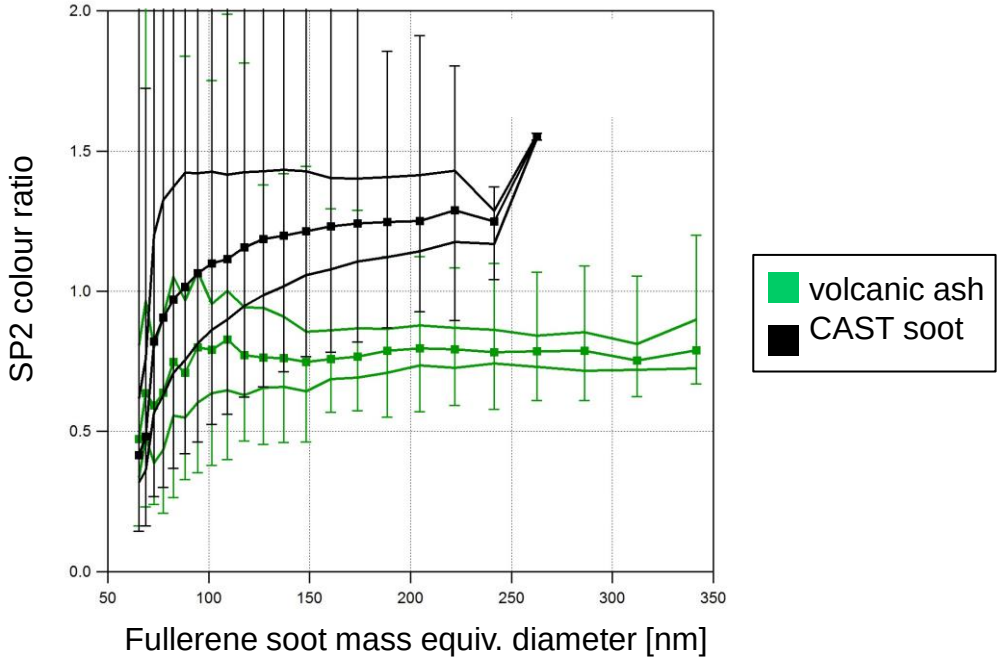
Interference from other incandescing particle types

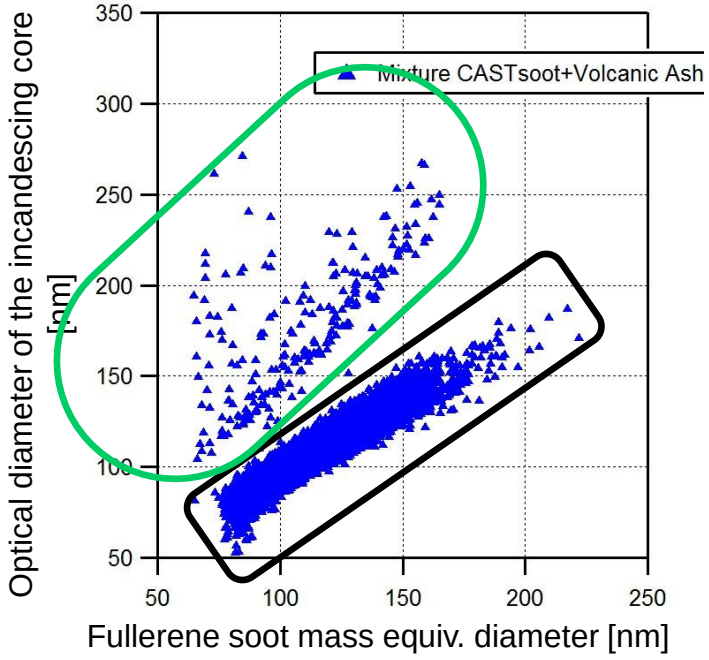
Highly refractory and moderately light-absorbing materials can also incandesce in the SP2 laser:
➔ potential interference from metals, volcanic ash, hematite (in pure form), dust (rarely), …

Example: volcanic ash



Heimerl et al., in prep.





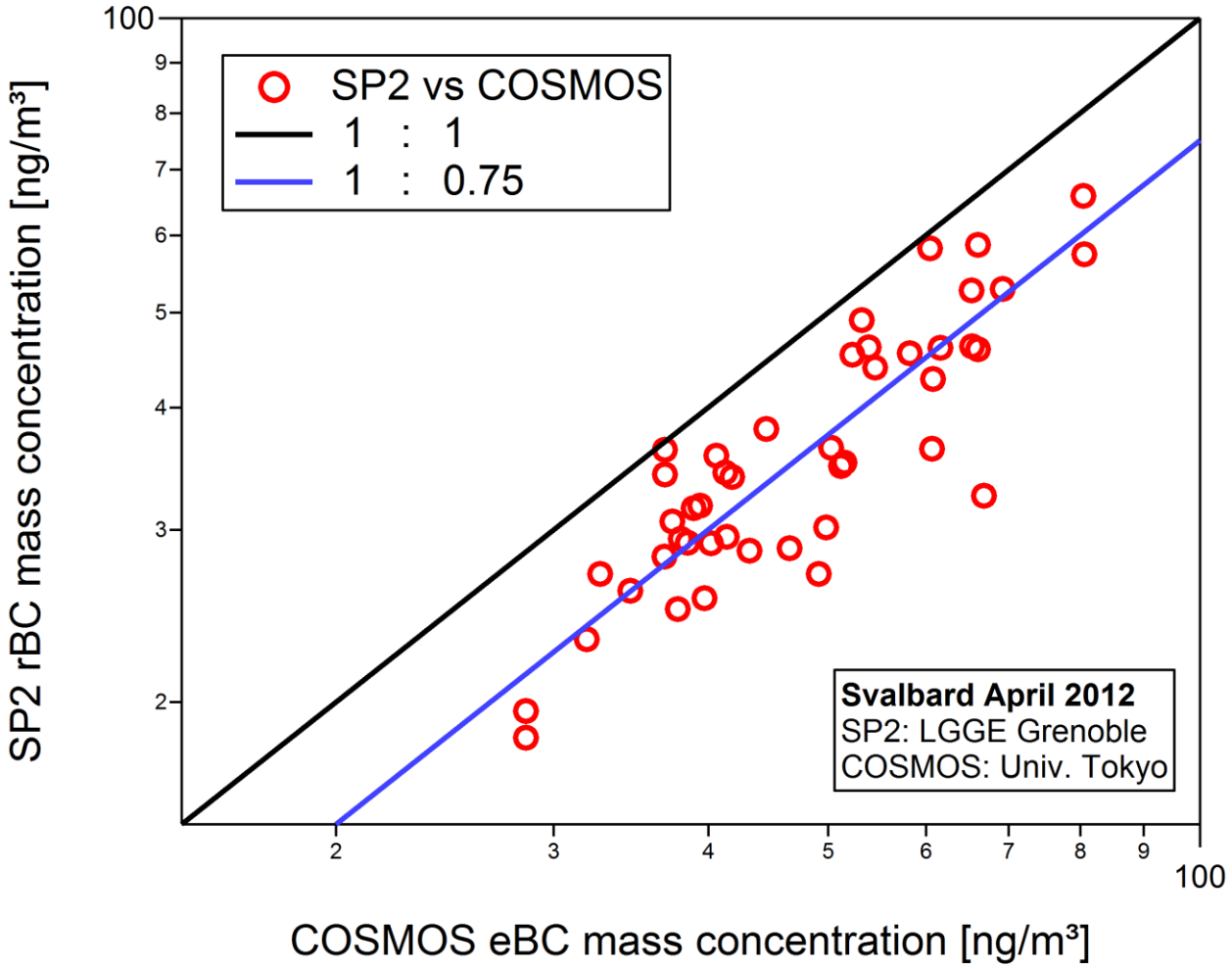
➔ It is possible to distinguish BC particles from other incandescing particles.

«Time-resolved LII»

«Time-resolved LII» (e.g. Schulz et al., 2006)**:**

- *Principle*:
  - **Pulsed laser used to heat the particles**
  - Detection of the thermal radiation from all particles in the sensitive volume
    ➔ **properties of the whole particle ensemble, mainly BC mass concentration**
  - **No empirical calibration with BC reference material needed (?)** (Snelling et al., 2005)
- *Application*:
  - **Most commonly used for detection of BC in combustion exhaust** in laboratory applications
  - More recently also used for field measurements of BC in atmospheric aerosols
- *Some field deployable instruments*:
  - **LII 300 from ARTIUM Technologies Inc.** (http://www.artium.com/):
    Dynamic range for BC is around <0.2 µg/m^3 – 20 g/m^3 ➔ suitable for combustion exhaust, while **not quite sensitive enough for clean atmospheric environments**
  - **NRC's LII instrument** (Smallwood, PhD thesis, 2008; Chan et al., 2008):
    Lower detection limit: 0.015 µg/m^3 ➔ **suitable for atmospheric applications**.

Comparison between SP2 and COSMOS (independently calibrated)



(see Miyazaki et al., 2008, for COSMOS description)

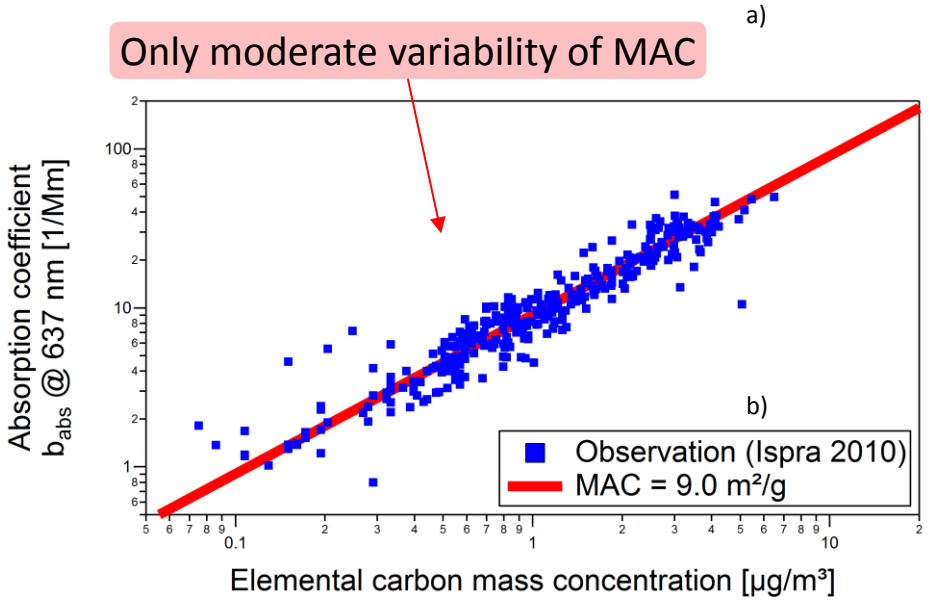
Zanatta et al., in prep.

SP2 (rBC) is ~25% lower than COSMOS (eBC)

➔ this is just about within experimental uncertainty, given independent calibration

Mass absorption cross section (MAC) of black carbon

Only moderate variability of MAC



The MAC relates BC mass with absorption coefficient

Relevant for:

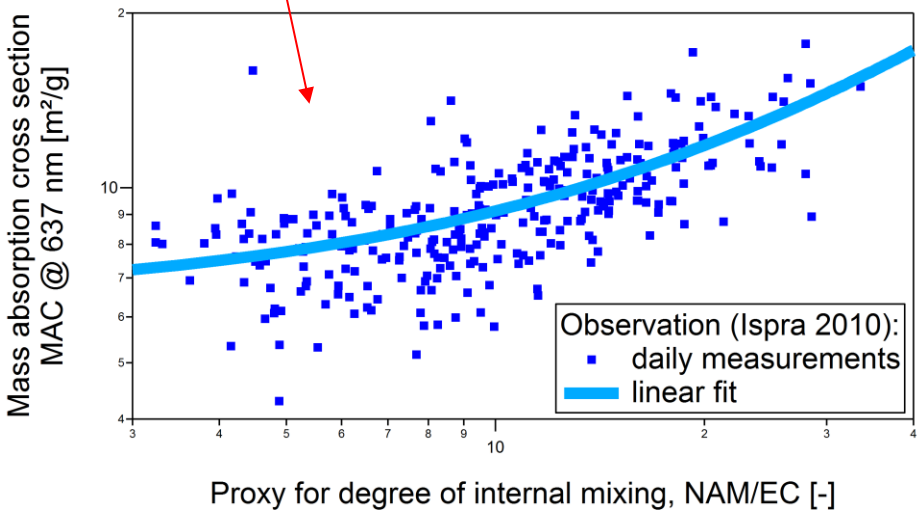
- modelling aerosol-radiation interactions
- absorption based black carbon measurements

Open research questions:

- What is the MAC of atmospheric BC
- Does the MAC depend on BC mixing state (lensing effect).

Observations at European background sites of the ACTRIS research infrastructure suggest[c]:
MAC_{BC} = 10.0 ±30% m^2/g @ 637 nm

Evidence for "lensing effect"



Example data taken from:
Zanatta et al., Atmos. Env., in review

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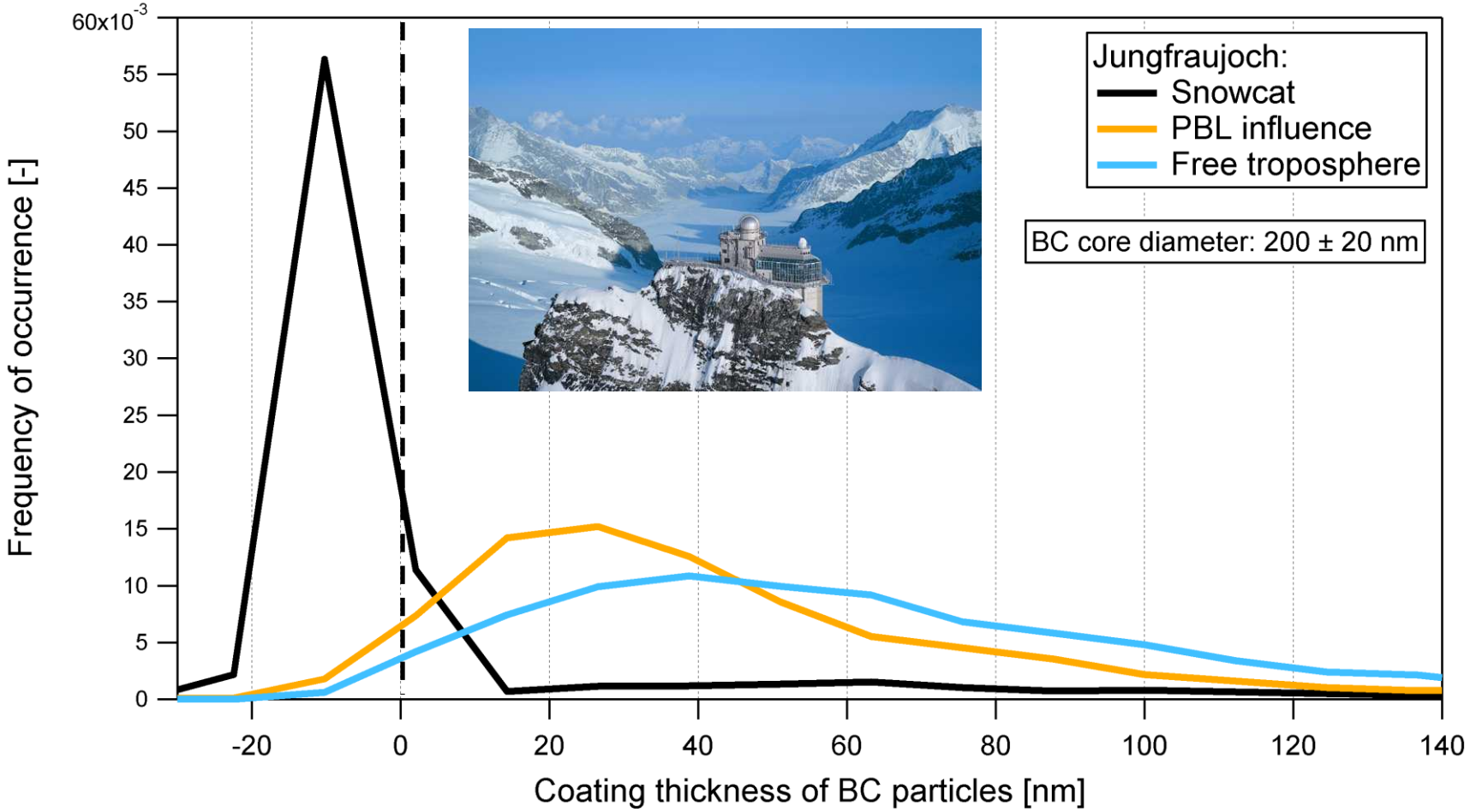
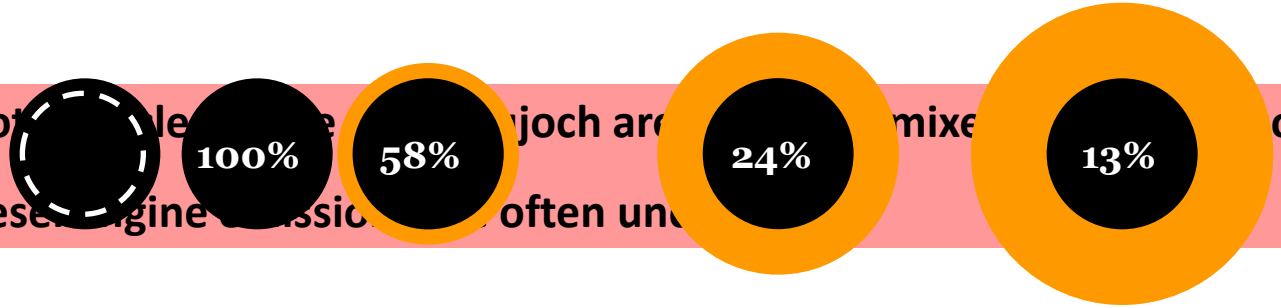
Swiss National Science Foundation

Capacity Building and Twinning for Climate Observing Systems

Histogram of BC coating thickness for different aerosols at the Jungfraujoch





➔ Aged soot [illegible] joch ar[illegible] mixe[illegible] oluble coatings.

➔ Fresh diesel [illegible] often un[illegible]

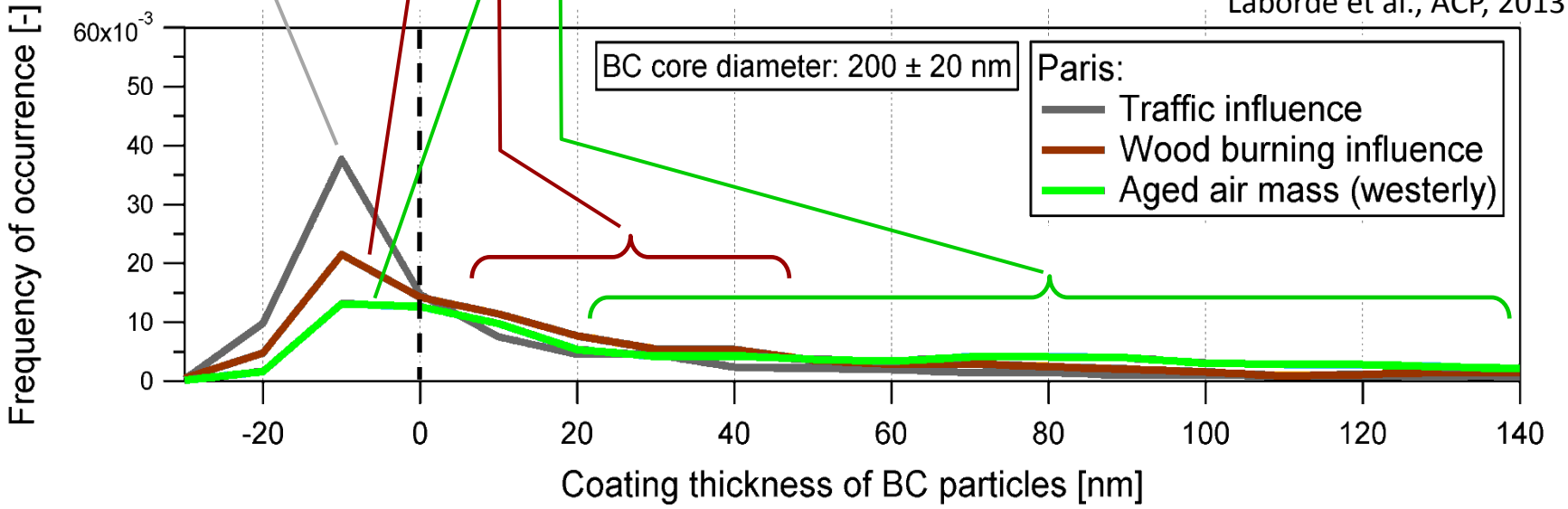
Mixing state of individual BC particles in Paris

Traffic influence:
➔ fresh traffic BC is uncoated
(note: negative value is «zero within uncertainty»)

Wood burning influence:
➔ traffic still dominates the BC mass
➔ BC from wood burning has medium coatings

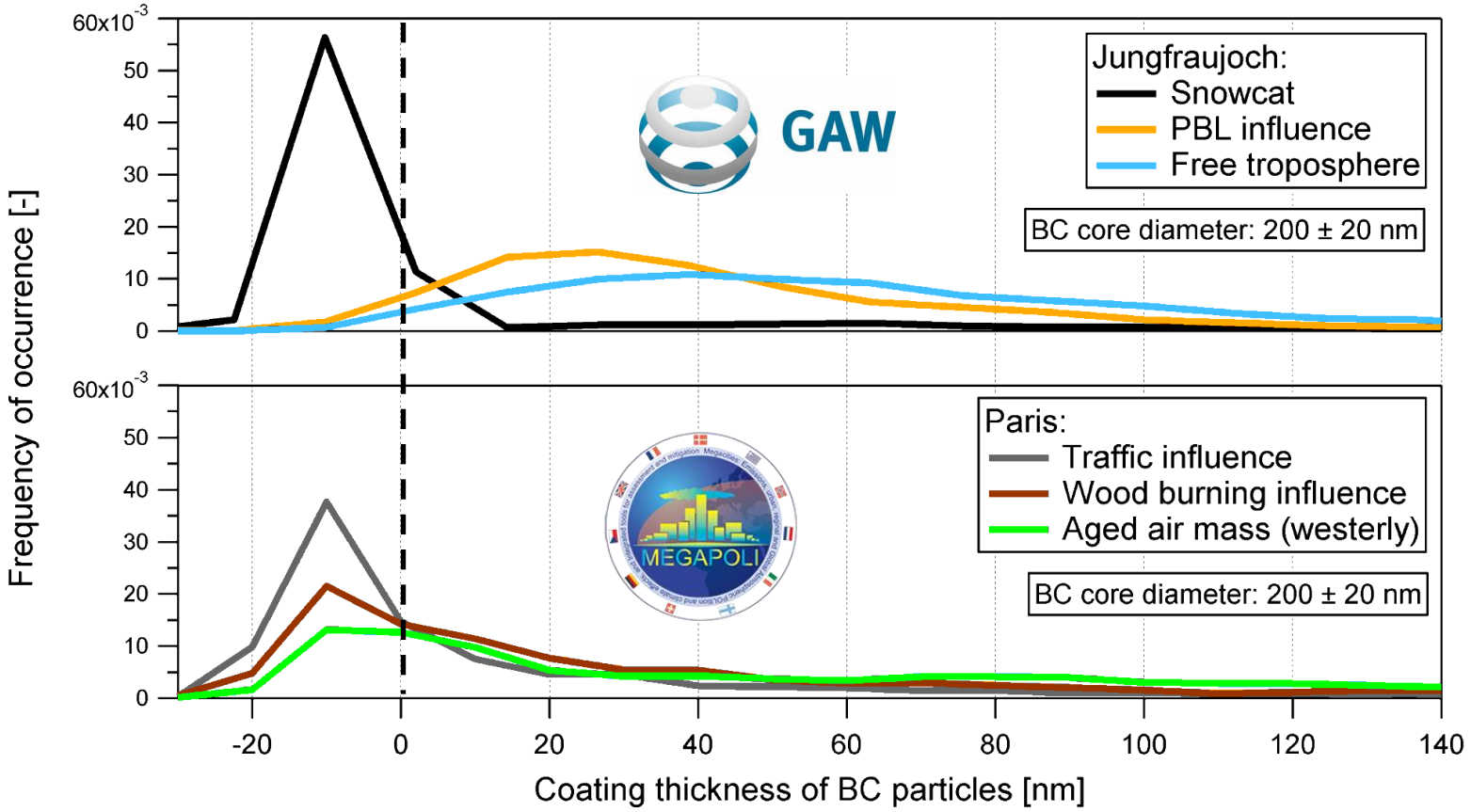
Aged air mass:
➔ substantial fraction of BC is from local/regional traffic
➔ BC in background air has considerable coatings

Laborde et al., ACP, 2013



➔ Most fresh BC is uncoated as traffic contribution dominates

Coating of BC particles: Jungfraujoch versus Paris

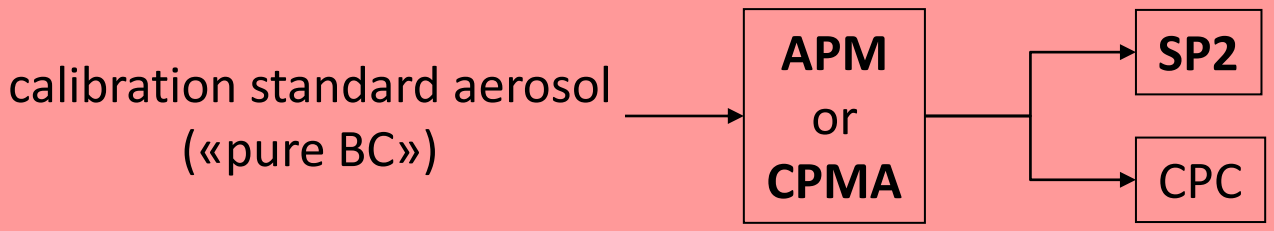


Coating thickness of BC particles increases substantially during transport "from Paris to the Jungfraujoch"

➔ **dominant fraction of BC particles at Jungfraujoch has substantial coatings**

SP2 calibration method

- **Particle Mass Analyser, APM or CPMA, is used as a mass reference** (they select particles directly by their mass)
- **«Pure BC» is used as calibration material:**
  - Thermo-denuded diesel, wood combustion or ambient soot
  - Fullerene soot or Aquadag (commercially available BC)



Alternative approach:

- **Use a Differential Mobility Analyser (DMA)** for size-selecting fullerene soot or Aquadag
- **Use effective density data from literature** to calculate mass from mobility diameter (Moteki and Kondo, 2010; Gysel et al., 2010)
  ➔ **~10% additional calibration uncertainty**