PAUL SCHERRER INSTITUT

PSI

ETH - NPC 2019

June 19th

ETH, Zurich

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The Single Particle Soot Photometer - Extended Range (SP2-XR) for black carbon measurements: an extensive comparison with the SP2

Michele Bertò[1], Rob L. Modini[1], Marco Zanatta[2], Jinfeng Yuan[1], Martin Irwin[1], Angela Marinoni[3], Michaela Ess[4], Hannes Schulz[2], Andreas Herber[2], Alexis Attwood[5], Fernando Velarde[6], Marcos Andrade[6], Birgit Wehner[7], Konstantina Vasilatou[4], Paolo Laj[3,8,9] and Martin Gysel-Beer[1]

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Droplet Measurement Technologies

INTRODUCTION

Measuring BC: techniques and terminology

- Accurately measuring BC mass is still a challenge…

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INTRODUCTION

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Measuring BC: techniques and terminology

- Accurately measuring BC mass is still a challenge…

PROPERTIES	DETECTION METHODS	NOMENCLATURE*
REFRACTORINESS	Thermo-optical (Sunset)	Elemental carbon (EC)
	Incandescence (SP2) Vaporization (SP-AMS)	Refractory BC (rBC)
LIGHT ABSORPTION	Photoacustic (PAS,…)	Equivalent BC (eBC)
	Filter based (PSAP, Aethalometer, MAAP, COSMOS,…)	

Adapted from Zanatta, 2016, PhD Thesis
*Petzhold et al., ACP, 2013

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How to measure the BC mass?

- The Single Particles Soot Photometer (SP[2], DMT)
  - ✓ **On-line single particle instrument**
  - ✓ It measures **refractory black carbon** (rBC) mass
  - ✓ Laser induced incandescence technique

48 cm W x 61 cm L x 26 cm H

~ 30 kg

SP2 Tecnical papers:
Schwarz et al., JGR, 2006
Stephens et al., AO, 2003

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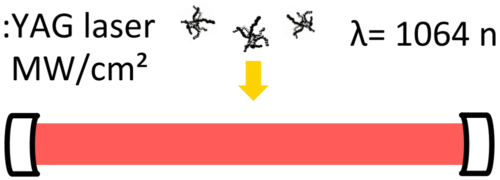
Nd:YAG laser
~3 MW/cm²



λ= 1064 nm

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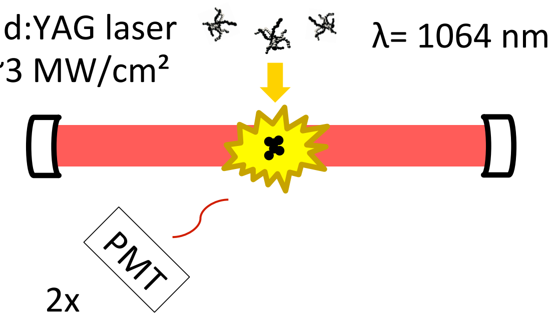
Detection of **incandescence** light

⬇ empirical calibration

-> BC mass concentration

⬇ density

-> BC size distribution



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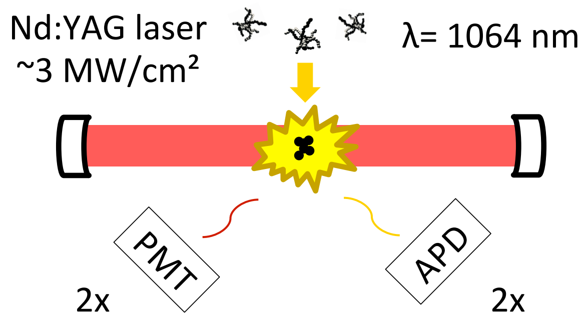

Detection of elastically **scattered** light

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optical sizing of **BC**

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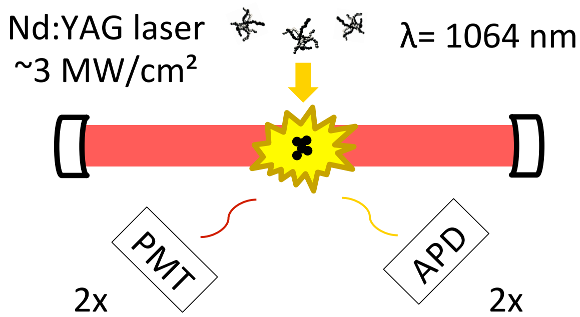

Detection of elastically **scattered** light

⬇ empirical calibration

optical sizing of **BC**

- ✓ Well-characterized (Laborde et al., AMT, 2012)
- ✓ Detection range: **~ 90 to 600 nm**
- ✓ **Dedicated operator** -> 1-2 months campaigns
- ✓ **Raw data analyses ->** based on "external" toolkits

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INTRODUCTION

DMT's new SP2-XR (Extended-Range)

- **S**ingle **P**article **S**oot **P**hotometer – Extended Range (**SP^2-XR**)
  - ✓ Same physical principles of the SP2

20 cm x 21.5 cm x 45 cm
(~ **4 times smaller**)
13 kg (~ **3 times lighter**)

INTRODUCTION

DMT's new SP2-XR (Extended-Range)

- **Single Particle Soot Photometer** – Extended Range (**SP^2-XR**)
  - ✓ Same physical principles of the SP2
  - ✓ Optimized hardware -> **improved stability** over time
  - ✓ Extended Detection range: nominally ~ **50 to 800 nm**
  - ✓ **Real-time automatic raw data processing**
  - ✓ Possibility for **long-term campaigns?** (currently testing)
  - ✓ Designed for use on new technology platforms (UAS, tethered sondes)
  - ❖ **2 detectors (broadband incandescence and scattering)** -> less info on the BC mixing state

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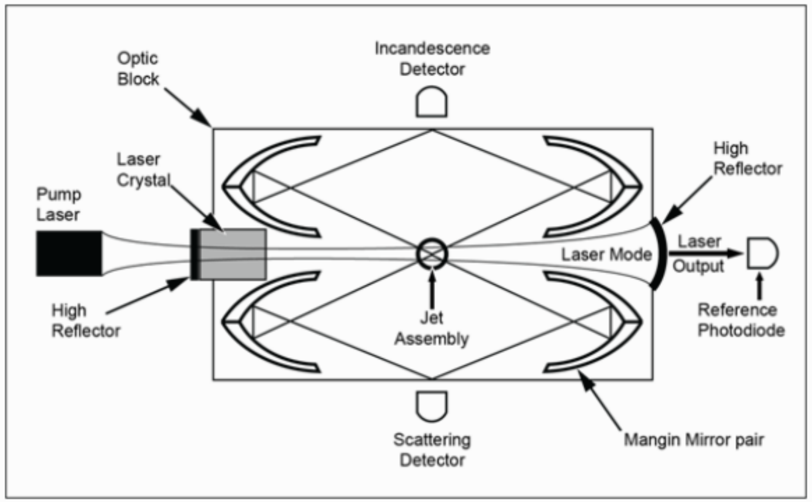


MOTIVATIONS

AIM:

Detailed characterization of the performance of the SP2-XR

HOW:

1. We tested 3 SP2-XR units from PSI, DMT and AWI
2. Verifying the incandesce/scattering **calibrations** stability and repeatability
3. Extensive **comparison with SP2s**:
    - **Sensitivities** to different calibration materials (fullerene soot/Aquadag)
    - **Detection/counting efficiencies** of absorbing and scattering particles
    - **rBC mass concentrations and size distributions** from laboratory and ambient measurements

METHODS

Laboratory and field experiments/measurements

1. CALIBRATIONS – SENSITIVITY - DETECTION EFFICIENCY

NEBULIZER
Aquadag, fullerene soot, ammonium sulfate

DRIER and NEUTRALIZER

DMA/APM or AAC

SP2

SP2-XR (DMT, PSI and AWI)

CPC

Data Analyses:
XR Calibrations: SP2-XR DMT's Toolkit
XR Data: PSI XR Toolkit (R. Modini)
SP2 Calibrations and data: PSI SP2 Toolkit (M. Gysel)

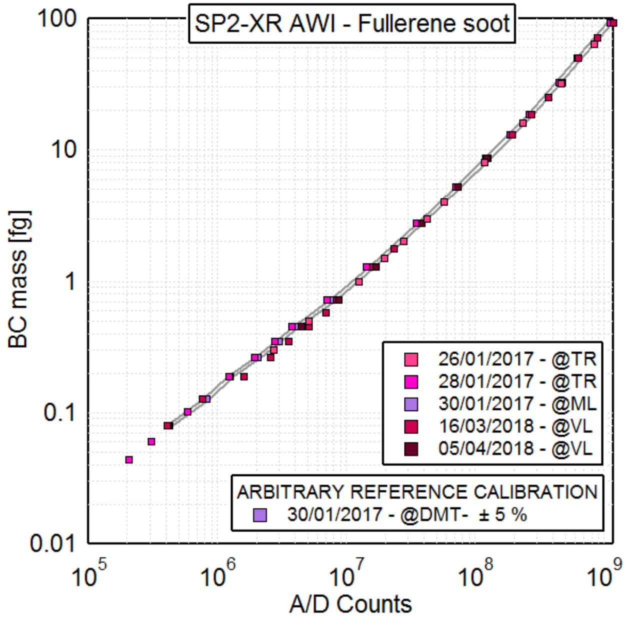
2. FIELD CAMPAIGN COMPARISONS

CHACALTAYA (Bolivia) – Spring 2018
High Altitude Station (~ 5200 m a.s.l.)

One week of parallel measurements with SP2 (LGGE) and 2 SP2-XRs (PSI and DMT)

RESULTS

Stability of Incandescence Calibration: fullerene soot (with equal SP2 response as diesel engine BC)



5 incandescence calibrations over a 1 year period for AWI's XR -> **variability of ~5 %**

NB: same for the other XR units

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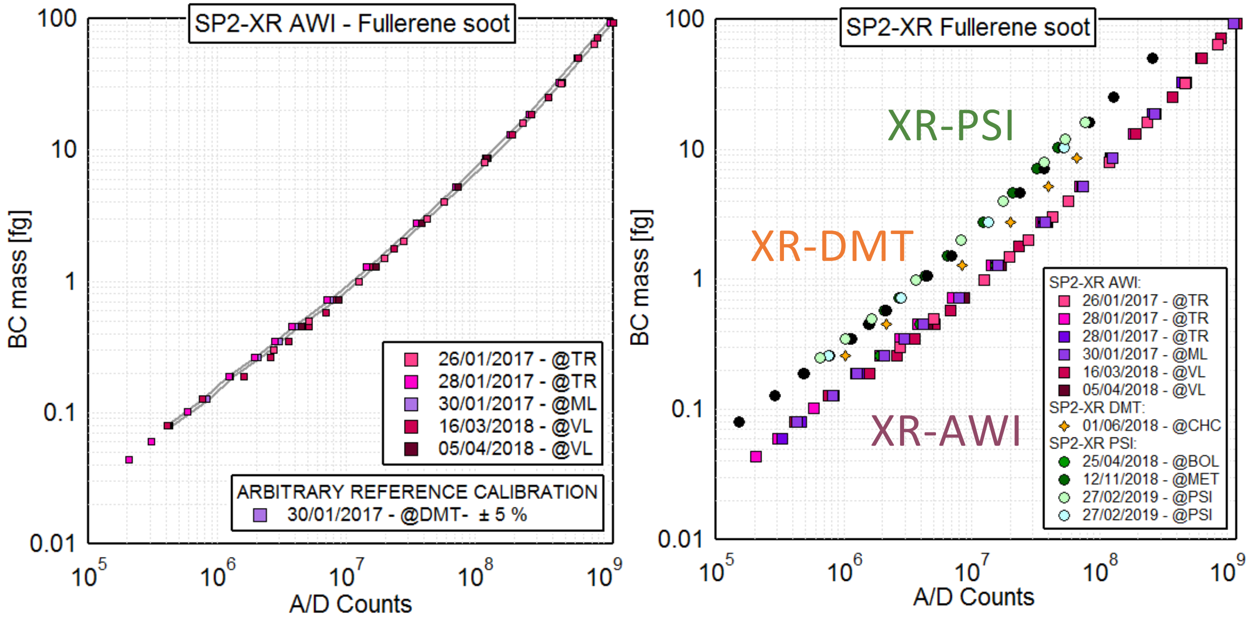

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- **3** XR units and **11** incandescence calibrations over a 2 years period

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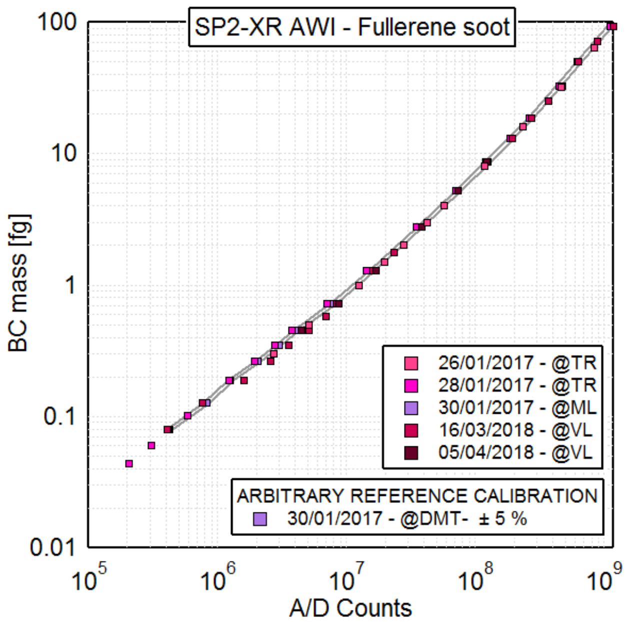
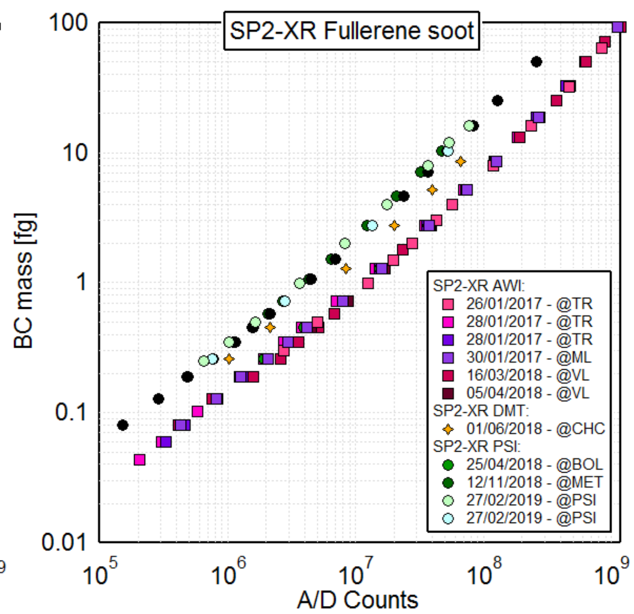
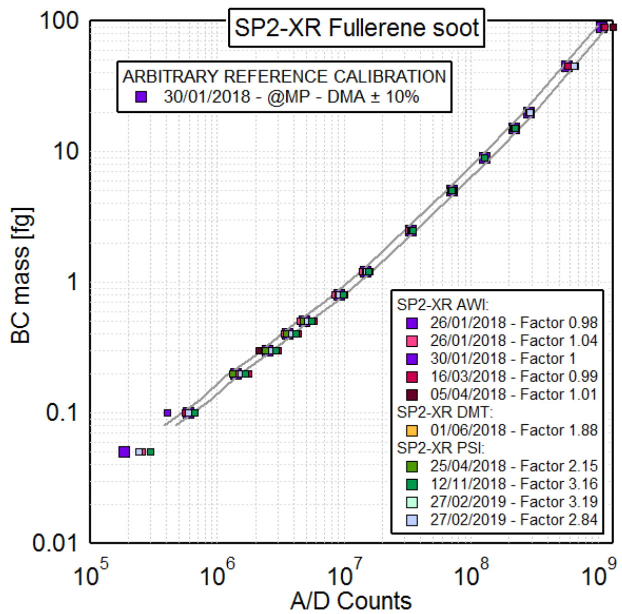






5 incandescence calibrations over a 1 year period for AWI's XR

-> **variability of ~5 %**

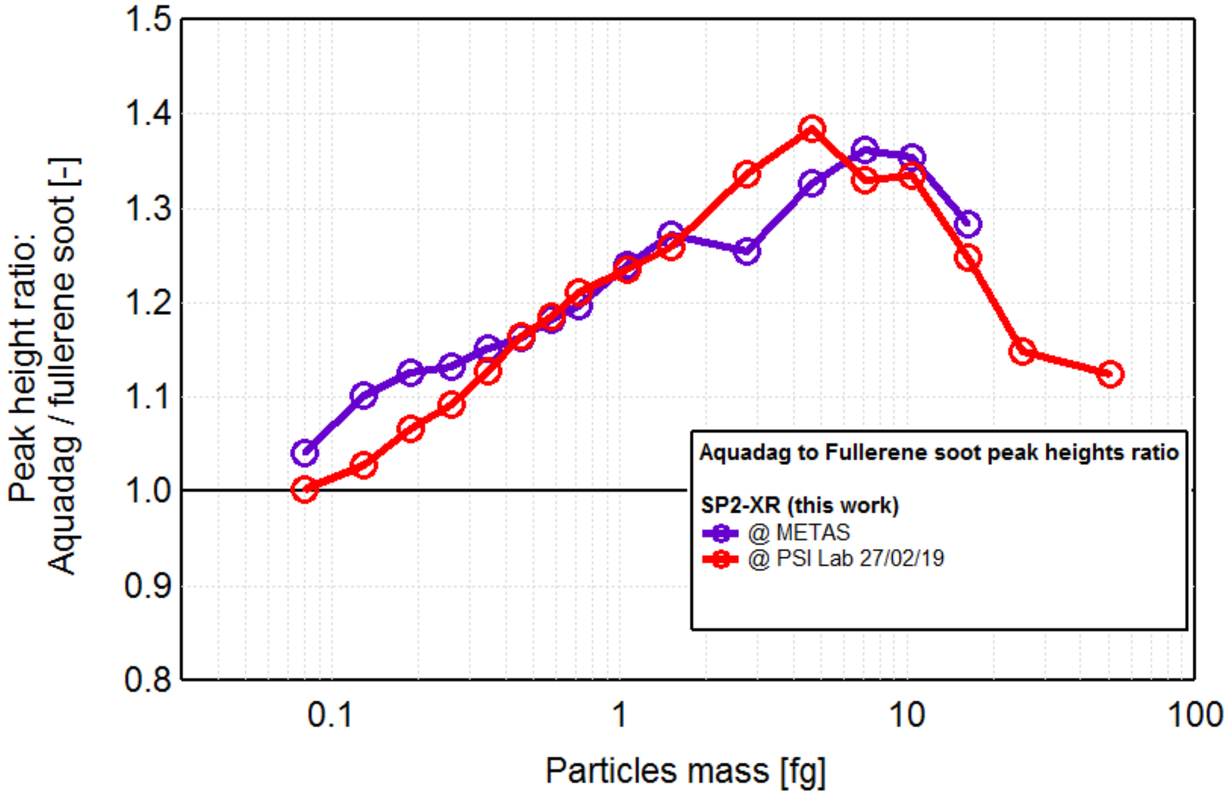
NB: same for the other XR units

- **3** XR units and **11** incandescence calibrations over a 2 years period
  - arbitrary offset factors:

    -> **slope/shape variability of ~10 %**

RESULTS

Relative Sensitivity to Different Calibration Materials



- As for SP2s, the SP2-XR is more sensitive to Aquadag than to fullerene soot

* Schwarz et al., GRL, 2008; Laborde et al., AMT, 2012; Baumgardner et al., 2012

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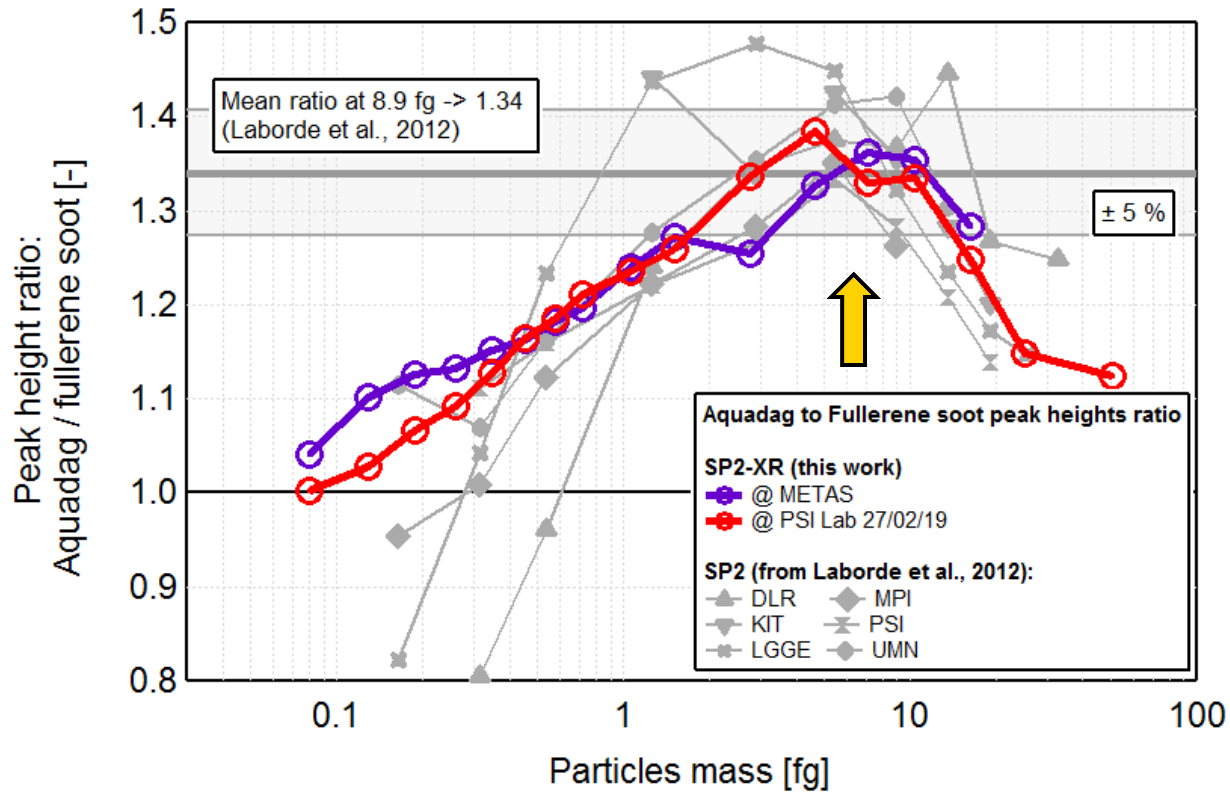

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- The **sensitivity ratio is comparable** to that of SP2s (within < 5% at 8.9 fg)

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RESULTS

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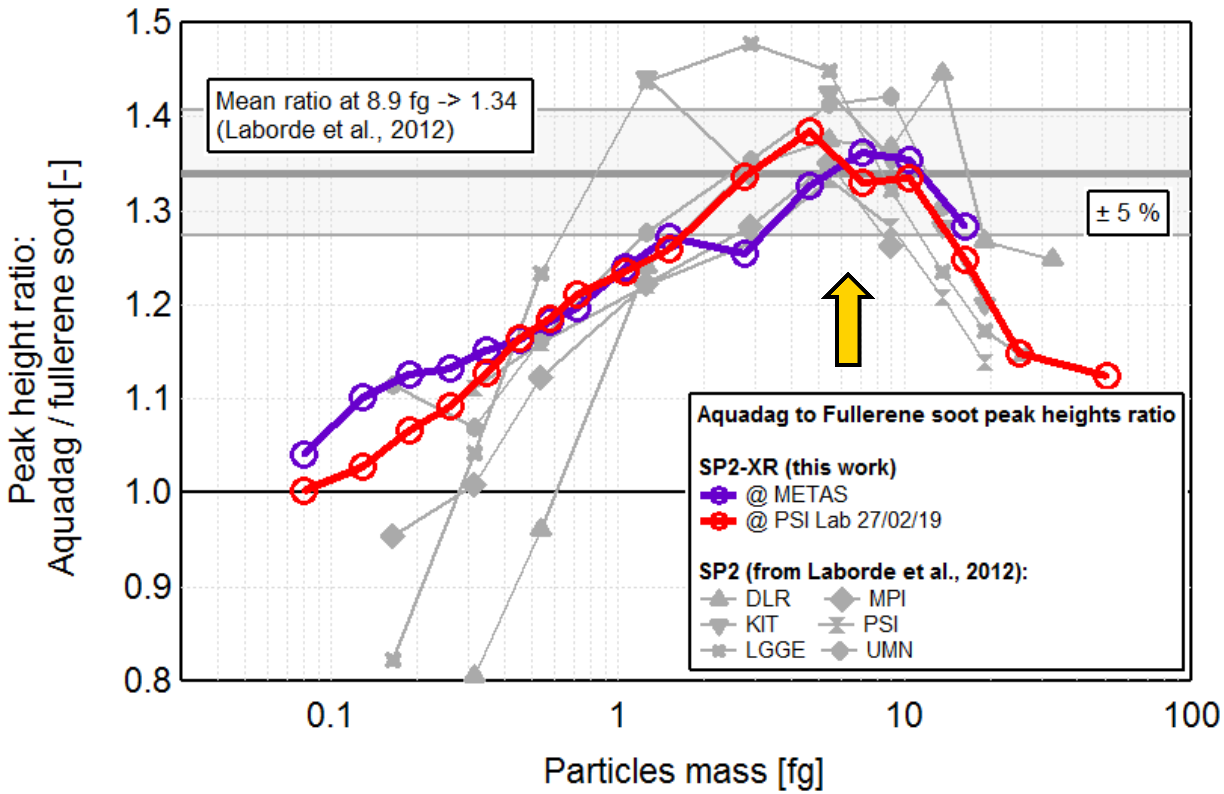

- As for SP2s, the SP2-XR is more sensitive to Aquadag than to fullerene soot
- The **sensitivity ratio is comparable** to that of SP2s (within < 5% at 8.9 fg)
- The sensitivity is **instrument-independent** (further data from other XR units currently being collected)

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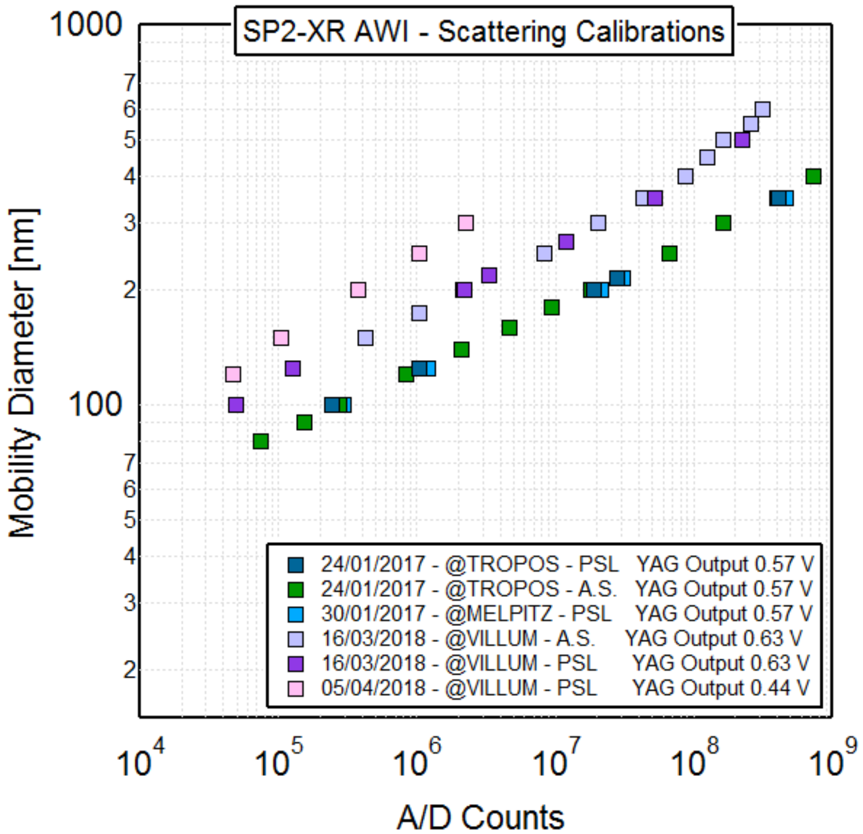
RESULTS

Scattering Calibration Stability

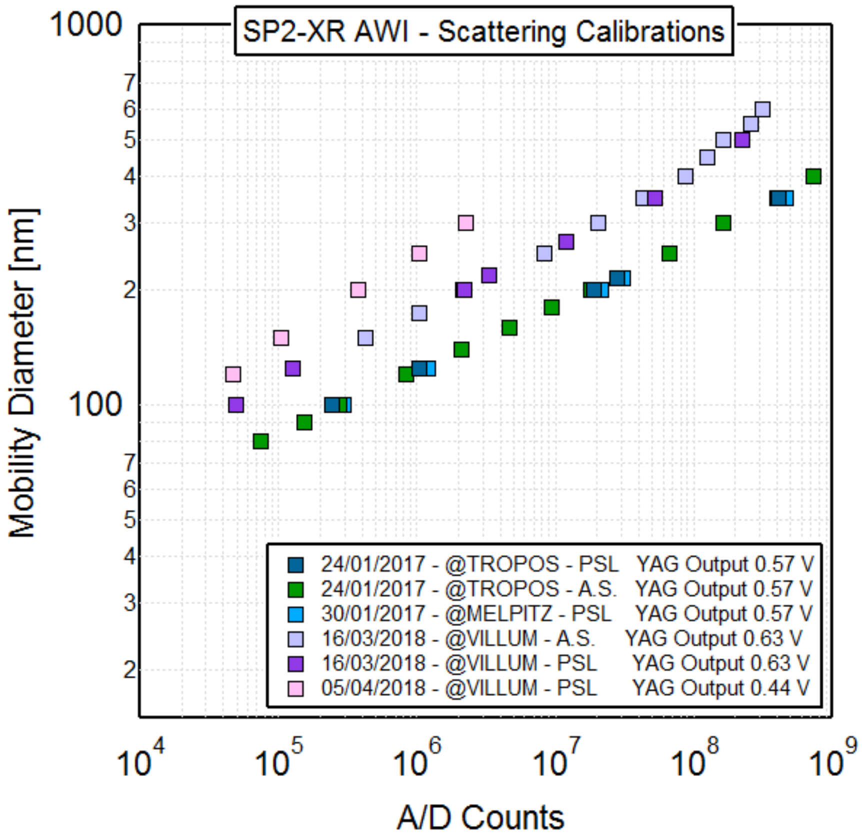


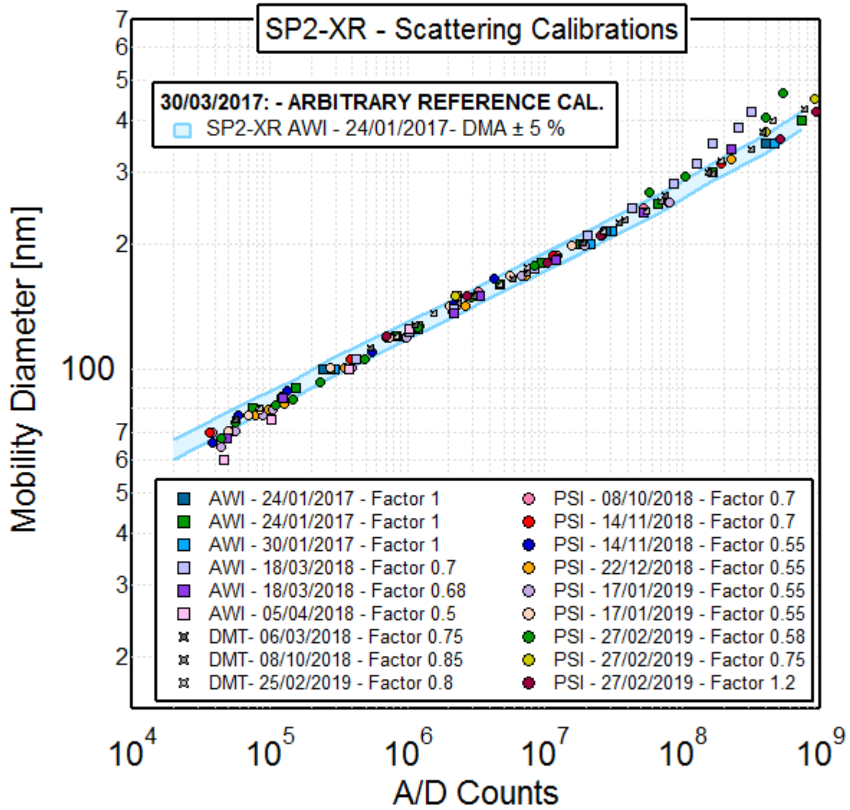
6 scattering calibrations over a 2 year period (XR AWI)

-> **sensitivity variability higher than 50 %**

RESULTS

Scattering Calibration Stability





5 scattering calibrations over a 2 year period (XR AWI)
-> **sensitivity variability higher than 50 %**

Arbitrary factor to match a reference calibration (**18** curves)
-> **slope/shape variability of ~5 %**
-> single point calibration

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RESULTS

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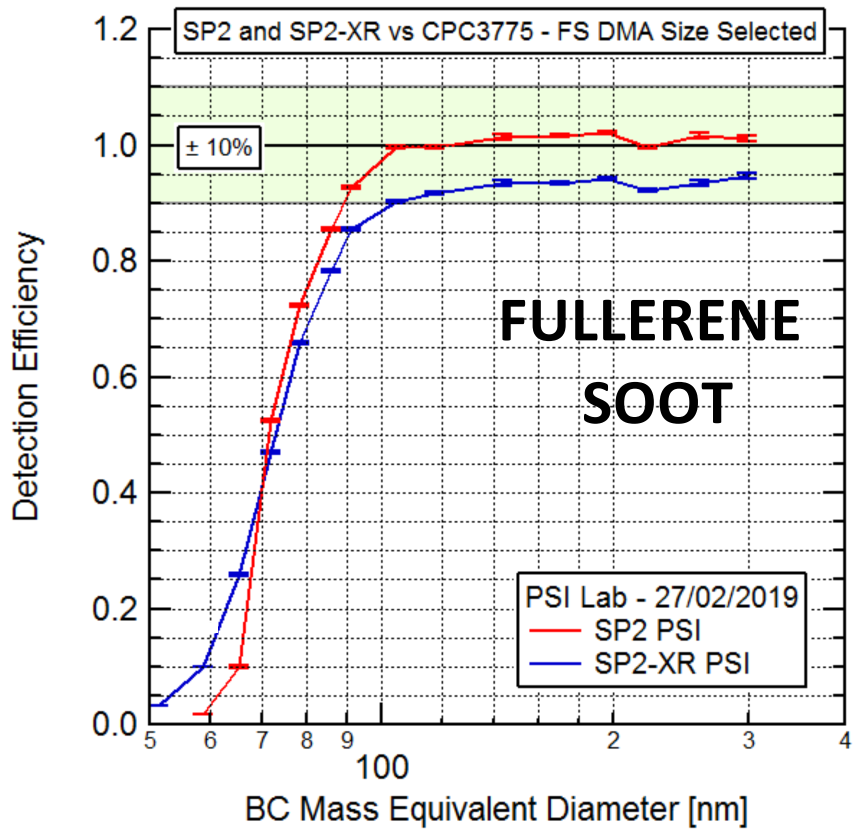
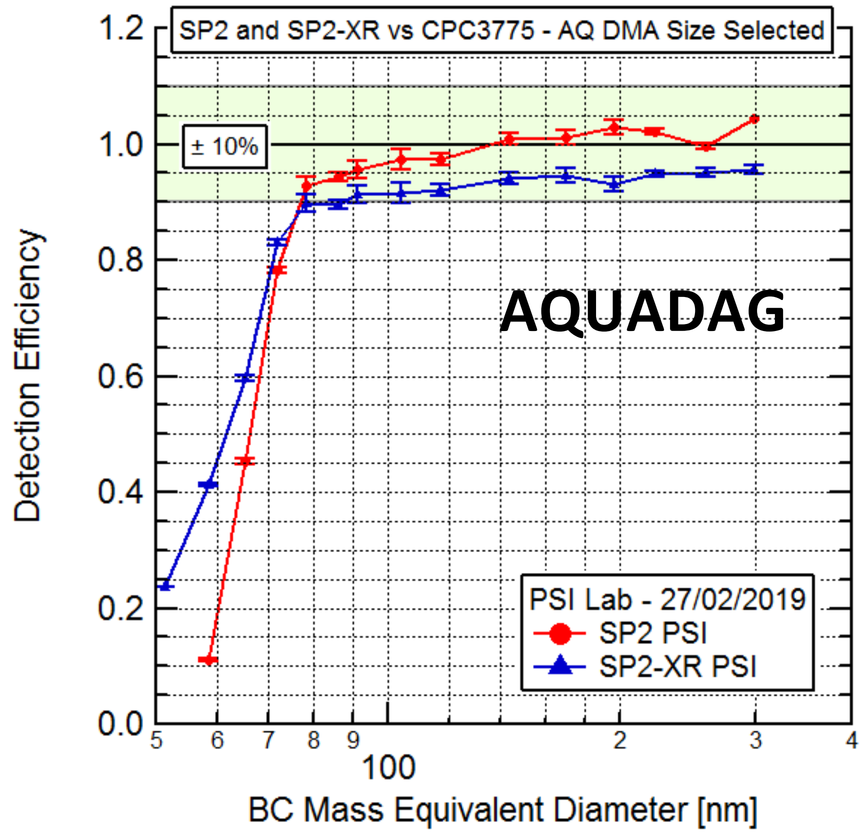
Detection Efficiency – Fullerene soot and Aquadag





1. The SP2-XR detection efficiency is **comparable** with that of an SP2

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RESULTS

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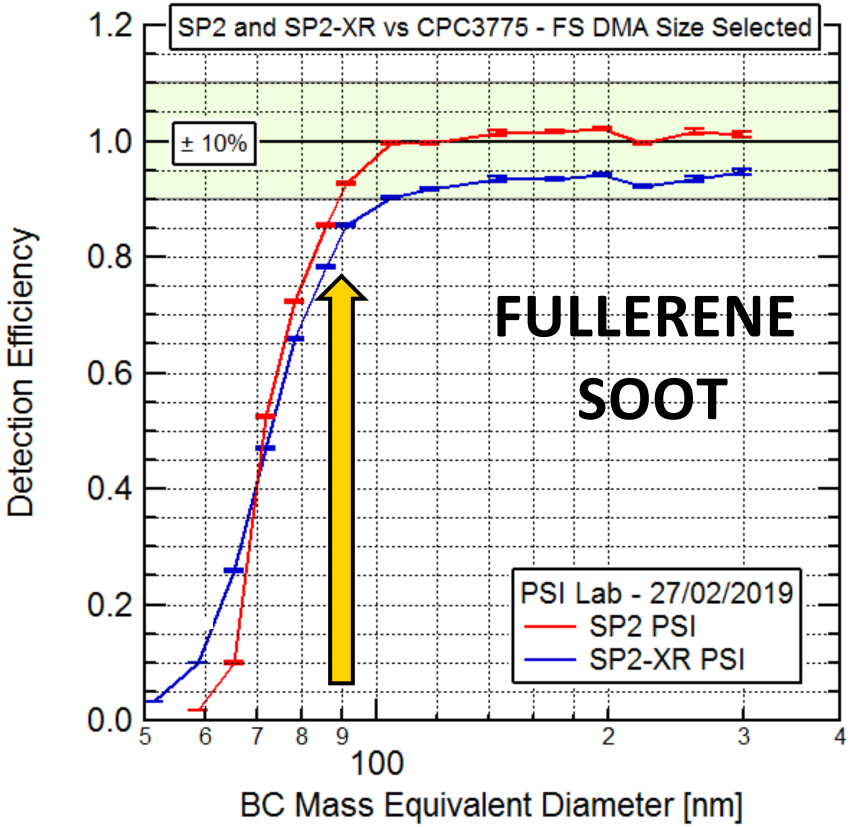

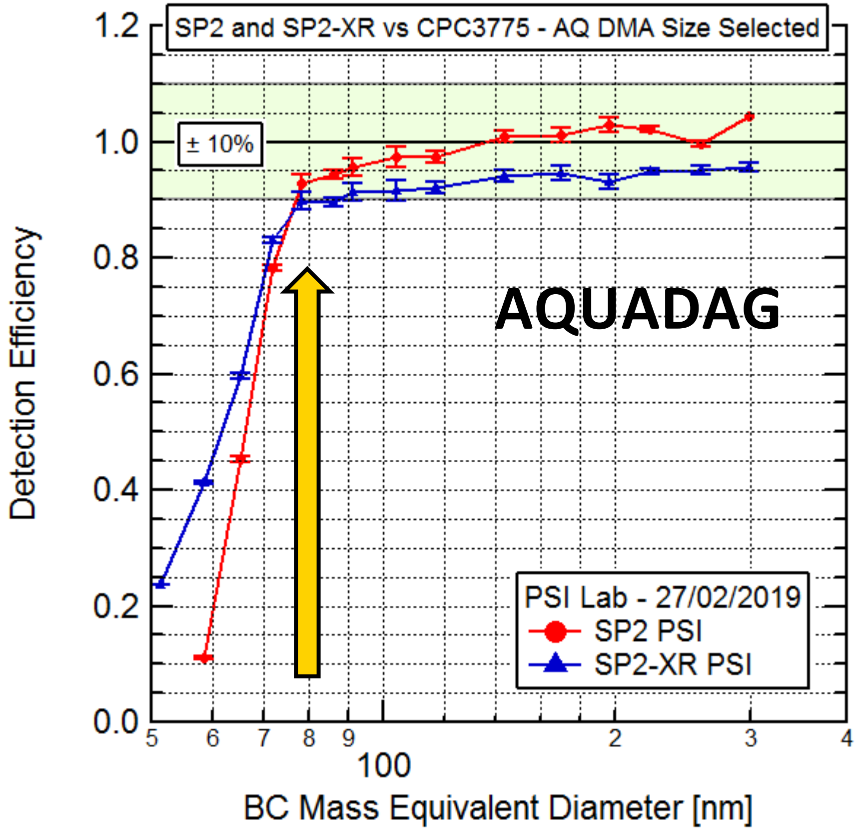

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2. 100±10% detection efficiency for diameters greater than **80-100 nm**

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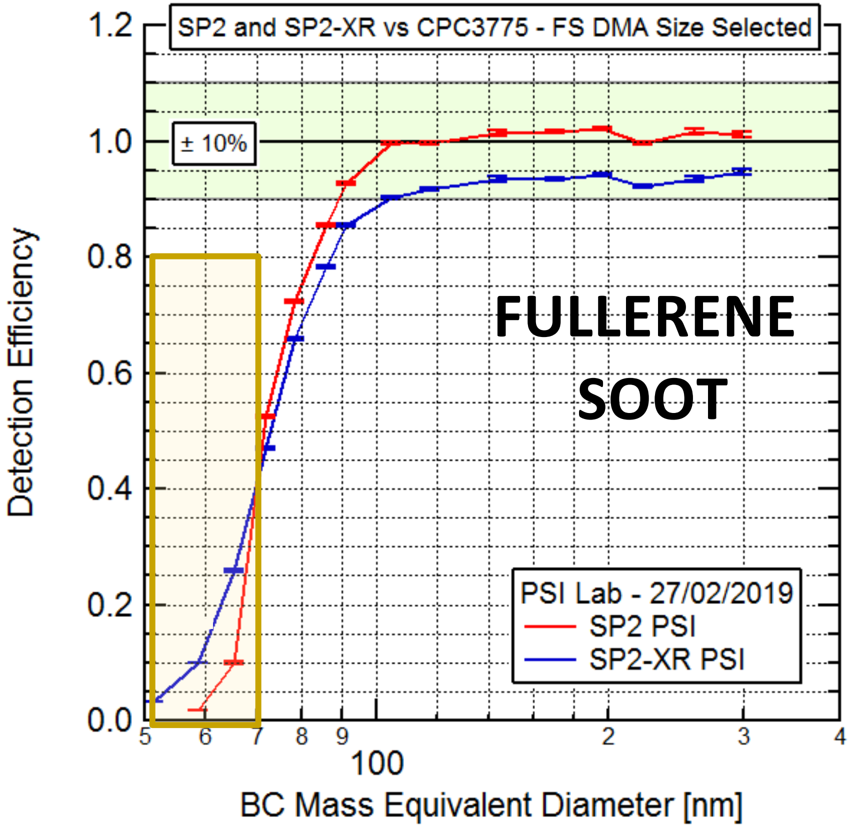

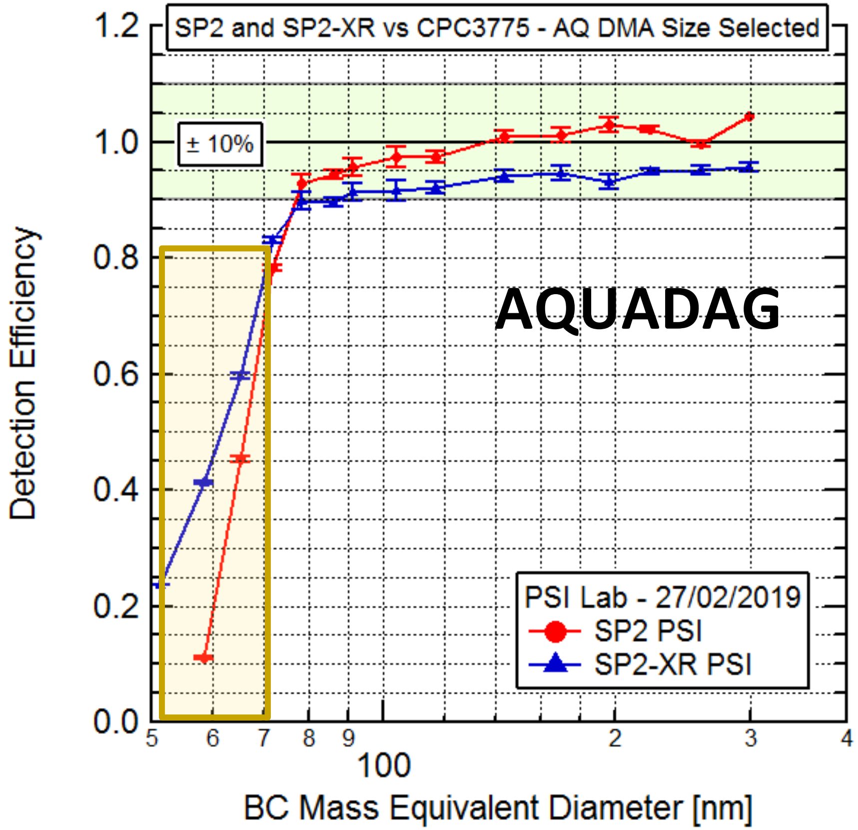

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2. 100±10% detection efficiency for diameters greater than **80-100 nm**
3. The SP2-XR works better for the smallest particles (< 70 nm)

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RESULTS

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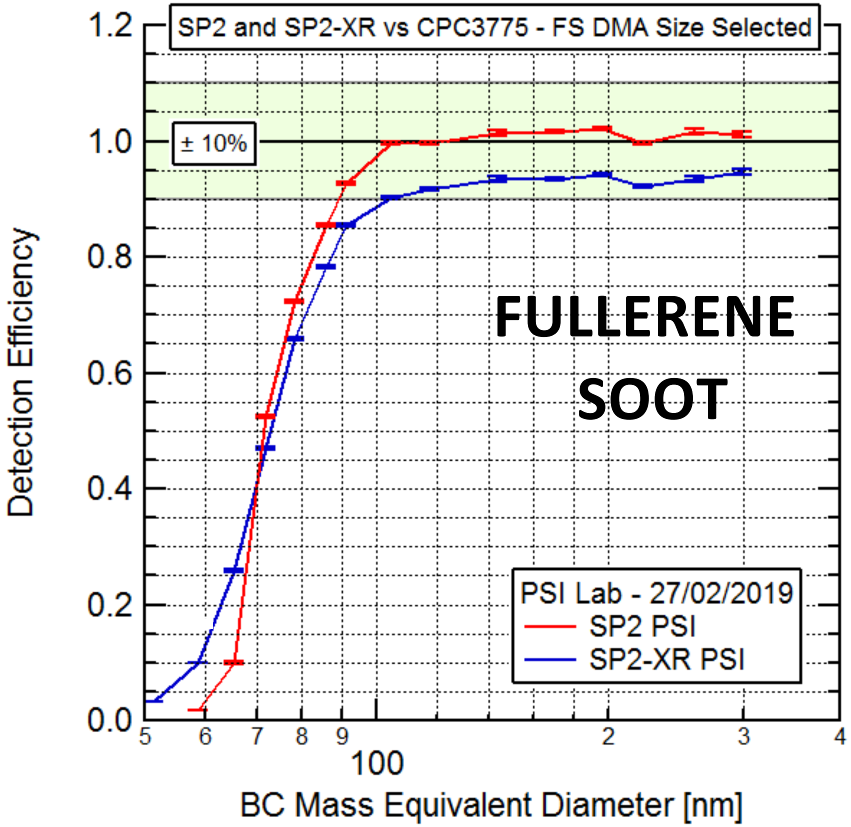
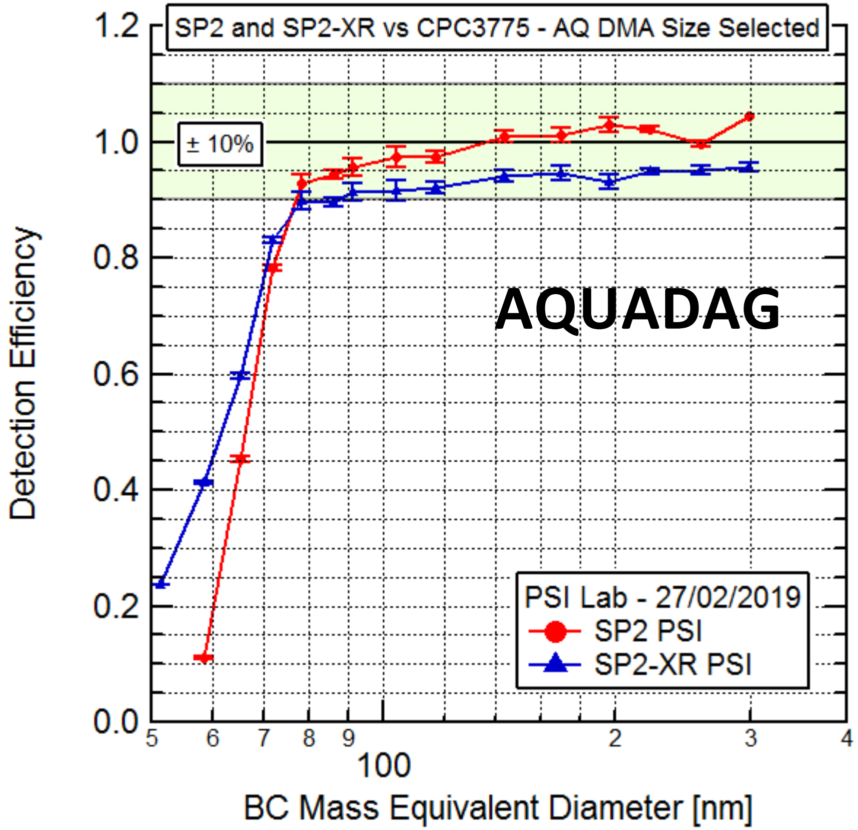
Detection Efficiency – Fullerene soot and Aquadag





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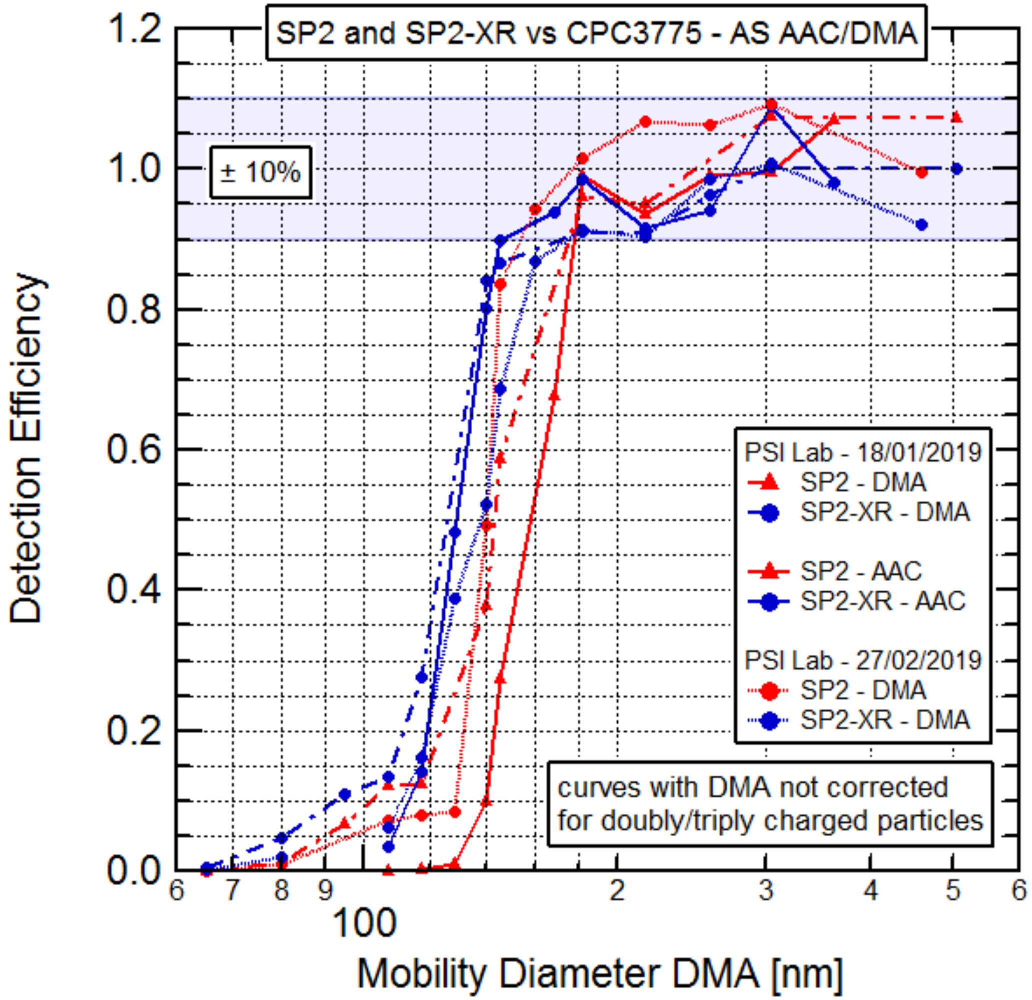
4. The SP2-XR (PSI) systematically measures 5-10 % less at the plateau (?)

RESULTS

Detection Efficiency – Ammonium Sulfate



1. **Comparable** DEs for XR and SP2

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RESULTS

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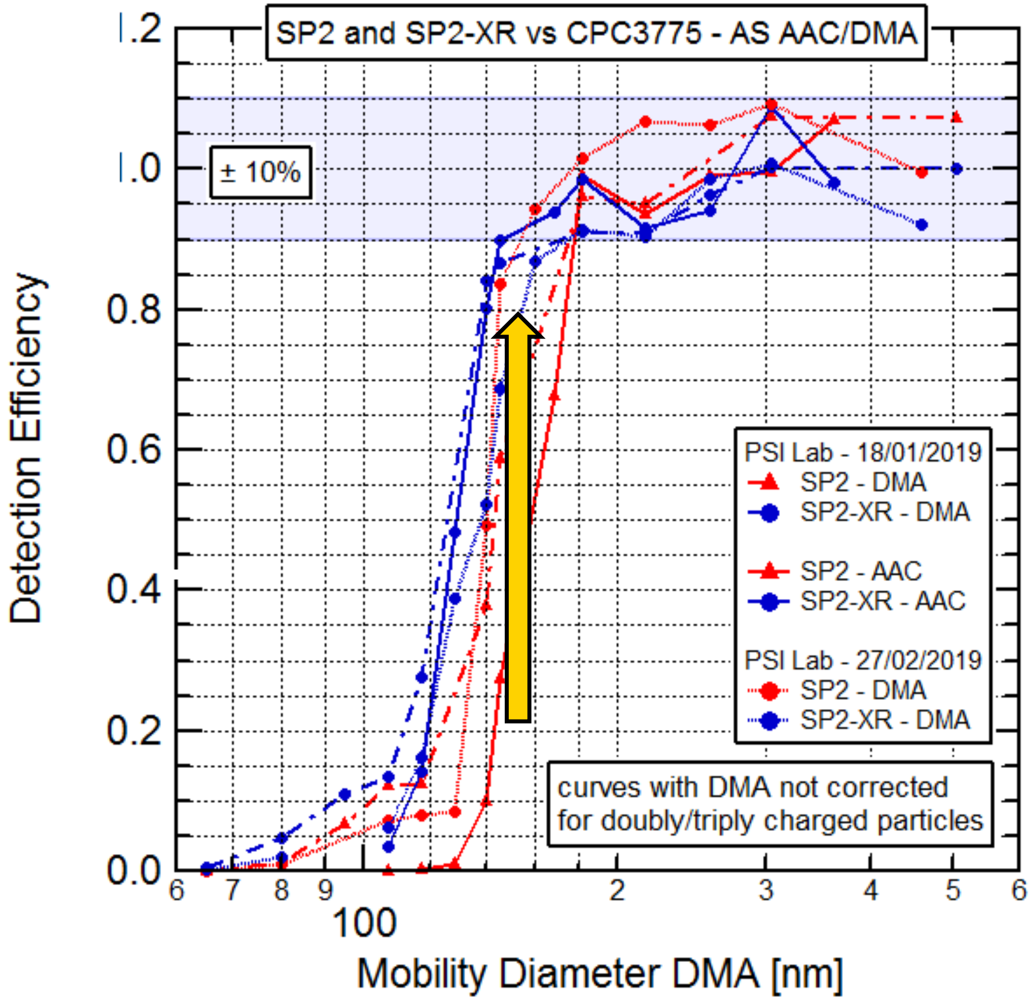


1. **Comparable** DEs for XR and SP2
2. 100±10% detection efficiency for diameters greater than **140-160 nm** (mobility diameter)

RESULTS

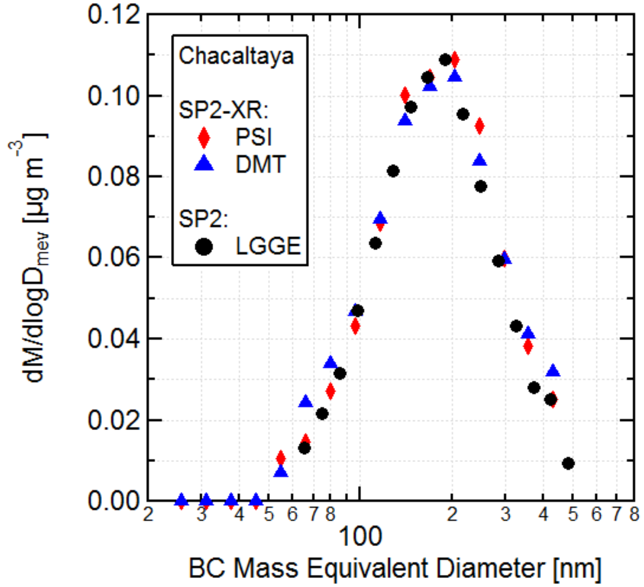
Ambient Measurements – the Bolivian campaign

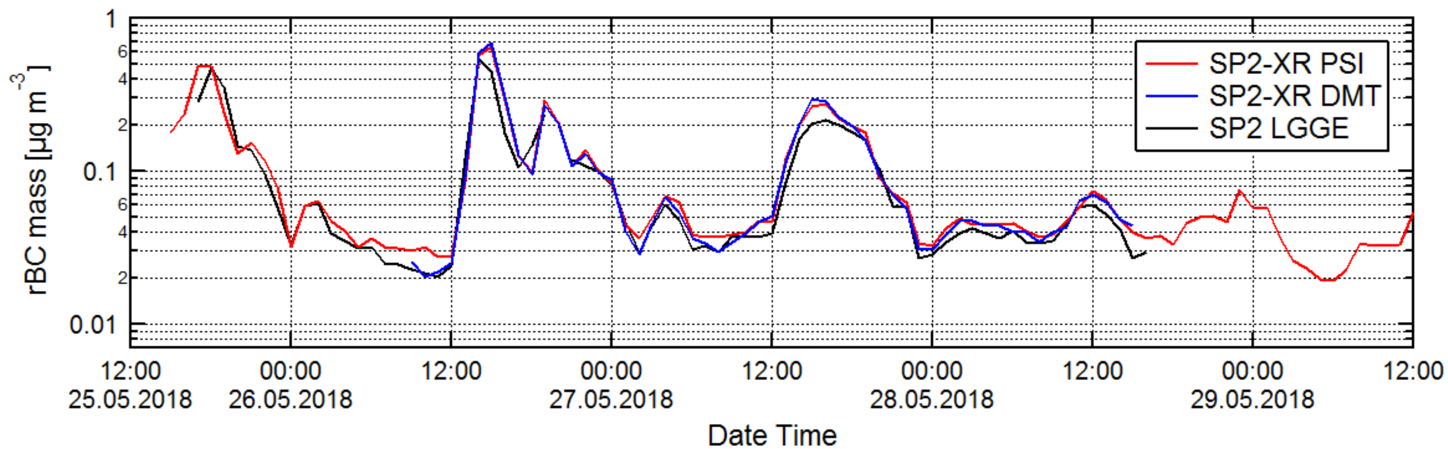
1 week of parallel measurements with SP2-XR (PSI and DMT) and SP2 (LGGE)

⇩

size distributions and rBC mass concentrations are in good agreement

Median size distribution over the period 26/05/2018 12:00:00 to 28/05/2018 12:00:00





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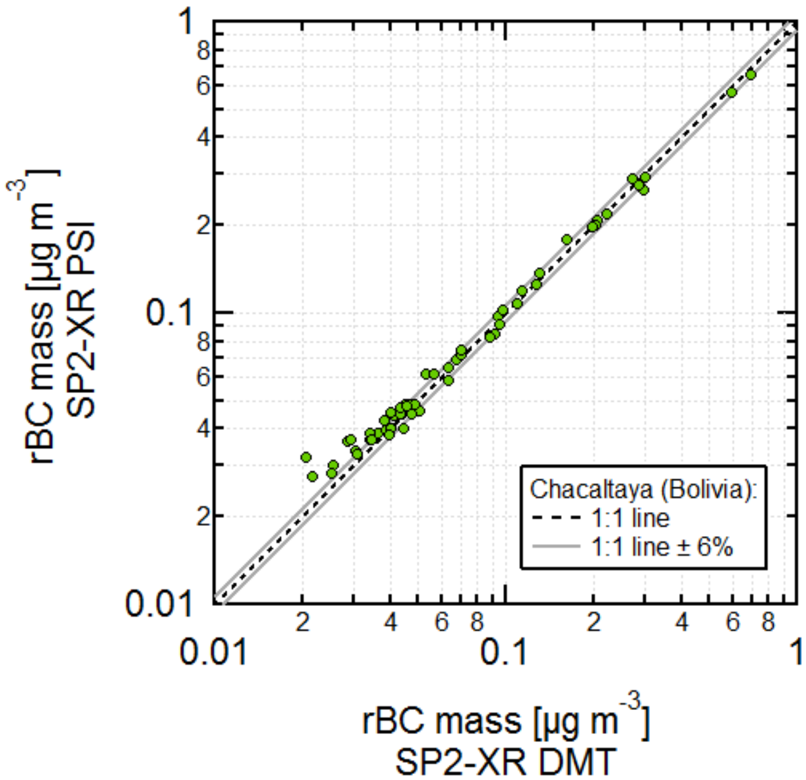
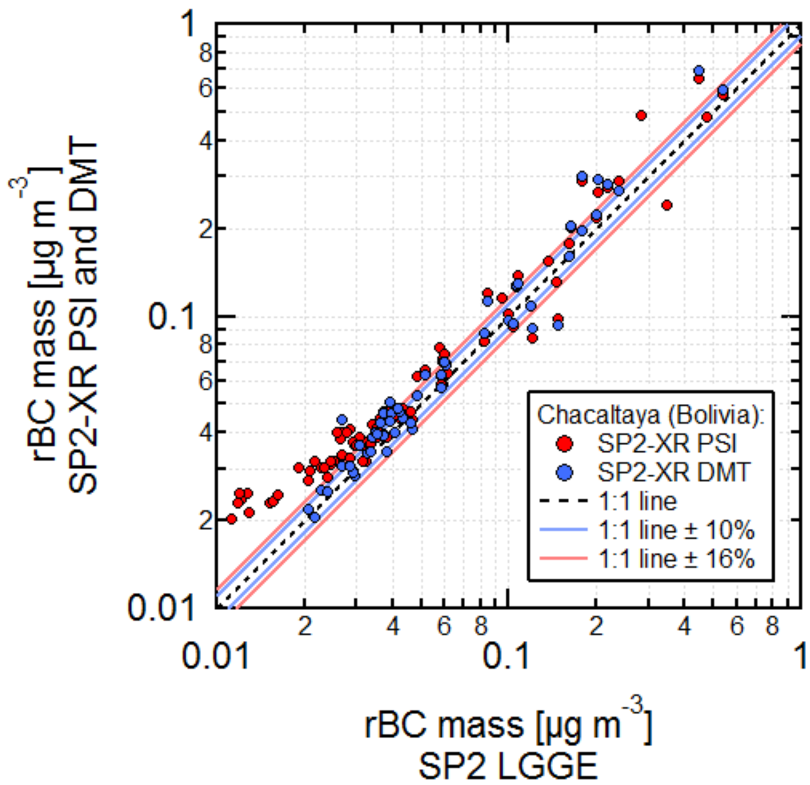
Ambient Measurements – the Bolivian campaign

SP2-XR PSI vs SP2-XR DMT



SP2-XR PSI vs SP2-XR DMT



SP2-XRs agree in 6% (3 – 9)

SP2 agrees with:

- XR-DMT in 10% (6 – 17)
- XR-PSI in 16% (9 – 21)

CONCLUSIONS

1. The SP2-XR **incandescence calibrations** are very stable in time (variability of ~ 5 %) and similar between different units (~ 10 %)

2. The XR sensitivity to **scattering** particles is highly variable (> 50 %), whereas the shapes of the calibration curves are stable in time (~ 5 %)

3. Comparison with SP2s:
    1. Comparable **sensitivity** to Aquadag and fullerene soot
    2. Similar **detection efficiency** (XR better for smallest particles)
        - Incandescence: 100±10% DE for diameters greater than 80-100 nm
        - Scattering: 100±10% DE for diameters greater than 140-160 nm

4. Good agreement for **rBC mass concentration** and **size distributions** during ambient measurements (~ 6-16 %)

The SP2-XR appeared to be a valid and easier alternative to the SP2

AKNOLEDGMENTS

- **Thanks to all the co-authors of this study**
  Rob L. Modini, Marco Zanatta, Jinfeng Yuan, Martin Irwin, Angela Marinoni, Michaela Ess, Hannes Schulz, Andreas Herber, Alexis Attwood, Fernando Velarde, Marcos Andrade, Birgit Wehner, Konstantina Vasilatou, Paolo Laj, Martin Gysel-Beer and to all who helped in organizing the campaigns.
- Thanks to G. Wehrle and R. Richter.

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REFERENCES

Schwarz, J. P., Gao, R. S., Spackman, J. R., Watts, L. A., Thomson, D. S., Fahey, D. W., Ryerson, T. B., Peischl, J., Holloway, J. S., Trainer, M., Frost, G. J., Baynard, T., Lack, D. A., de Gouw, J. A., Warneke, C., and Del Negro, L. A.: Measurement of the mixing state, mass, and optical size of individual black carbon particles in urban and biomass burning emissions, Geophys. Res. Lett., 35,L13810, doi: 10.1029/2008GL033968, 2008.

Stephens, M., Turner, N., and Sandberg, J.: Particle identification by laser-induced incandescence in a solid-state laser cavity, Appl. Optics, 42, 3726–3736, 2003.

Laborde, M., Schnaiter, M., Linke, C., Saathoff, H., Naumann, K.- H., M¨ohler, O., Berlenz, S., Wagner, U., Taylor, J. W., Liu, D., Flynn, M., Allan, J. D., Coe, H., Heimerl, K., Dahlk¨otter, F., Weinzierl, B., Wollny, A. G., Zanatta, M., Cozic, J., Laj, P., Hitzenberger, R., Schwarz, J. P., and Gysel, M.: Single Particle Soot Photometer intercomparison at the AIDA chamber, Atmos. Meas. Tech., 5, 3077–3097, doi:10.5194/ amt-5-3077-2012, 2012.

Petzold, A., Ogren, J. A., Fiebig, M., Laj, P., Li, S. M., Baltensperger, U., ... & Wehrli, C. (2013). Recommendations for reporting" black carbon" measurements. Atmospheric Chemistry and Physics, 13(16), 8365-8379.

SUPPLEMENTARY MATERIAL

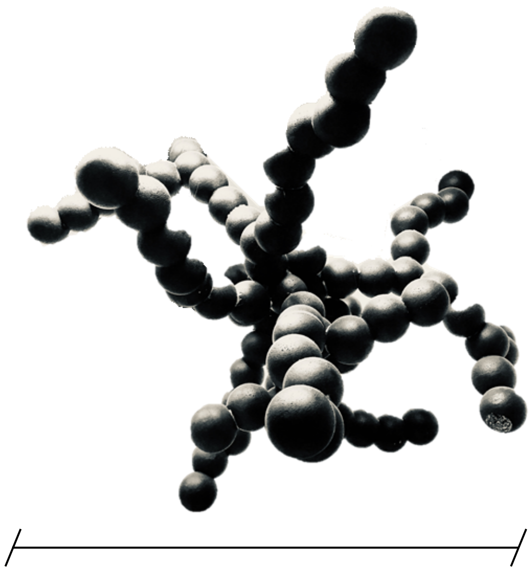
PAUL SCHERRER INSTITUT

LABORATORY OF ATMOSPHERIC CHEMISTRY

INTRODUCTION

Black Carbon (BC) in the atmosphere

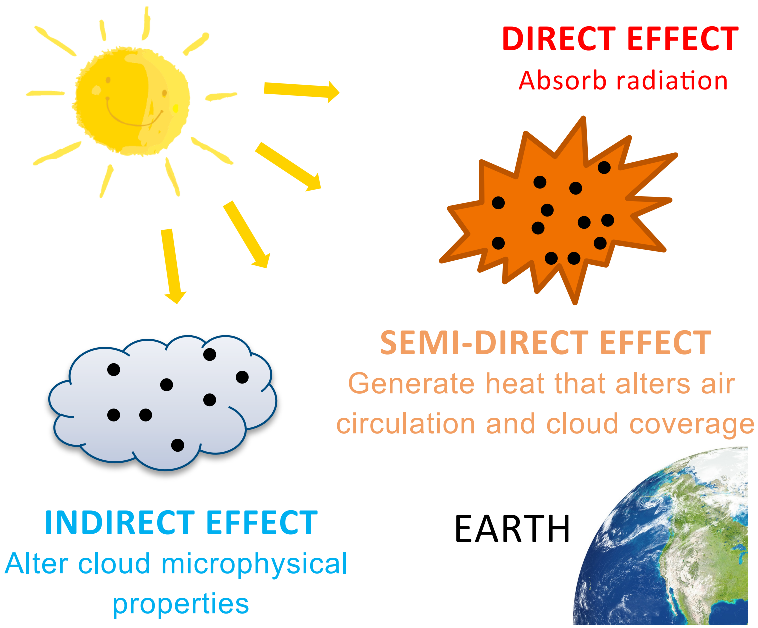
- **SOURCES**: combustion processes, mainly anthropogenic origins (Klimont et al., 2017)
- **TOTAL AEROSOL MASS FRACTION**: ~ 10% in Europe (Zanatta et al., 2016)
- **SIZE DISTRIBUTION**: from 10 to 300 nm (Bond et al., 2013)



~ 100/300 nm

Klimont et al., ACP, 2017
Zanatta et al., AE, 2016
Bond et al., JGR, 2013
BC picture from METAS

BC CLIMATE EFFECTS

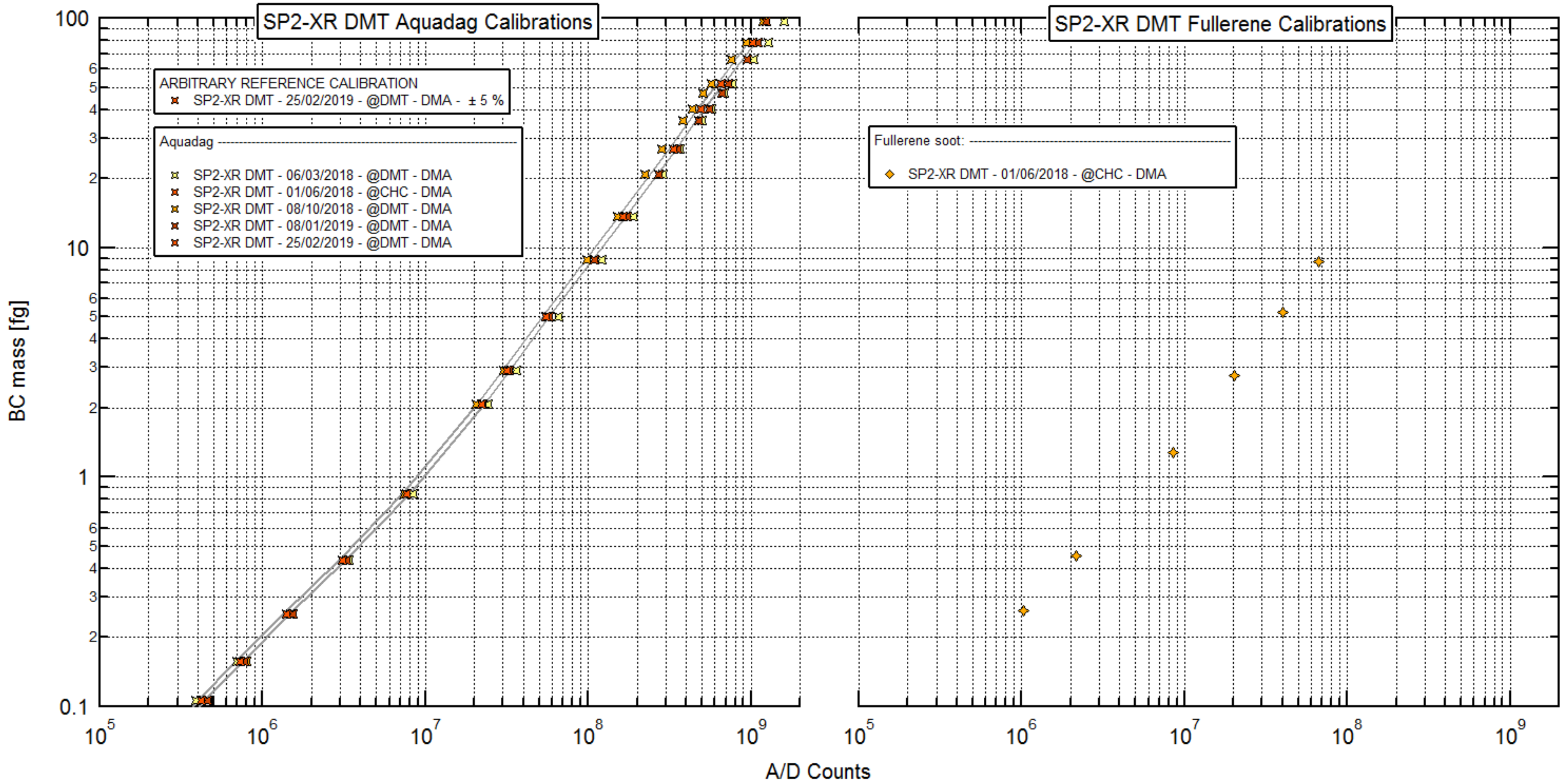


⇨ **POSITIVE RADIATIVE FORCING**

RESULTS

Incandescence Calibration Stability – SP2-XR DMT

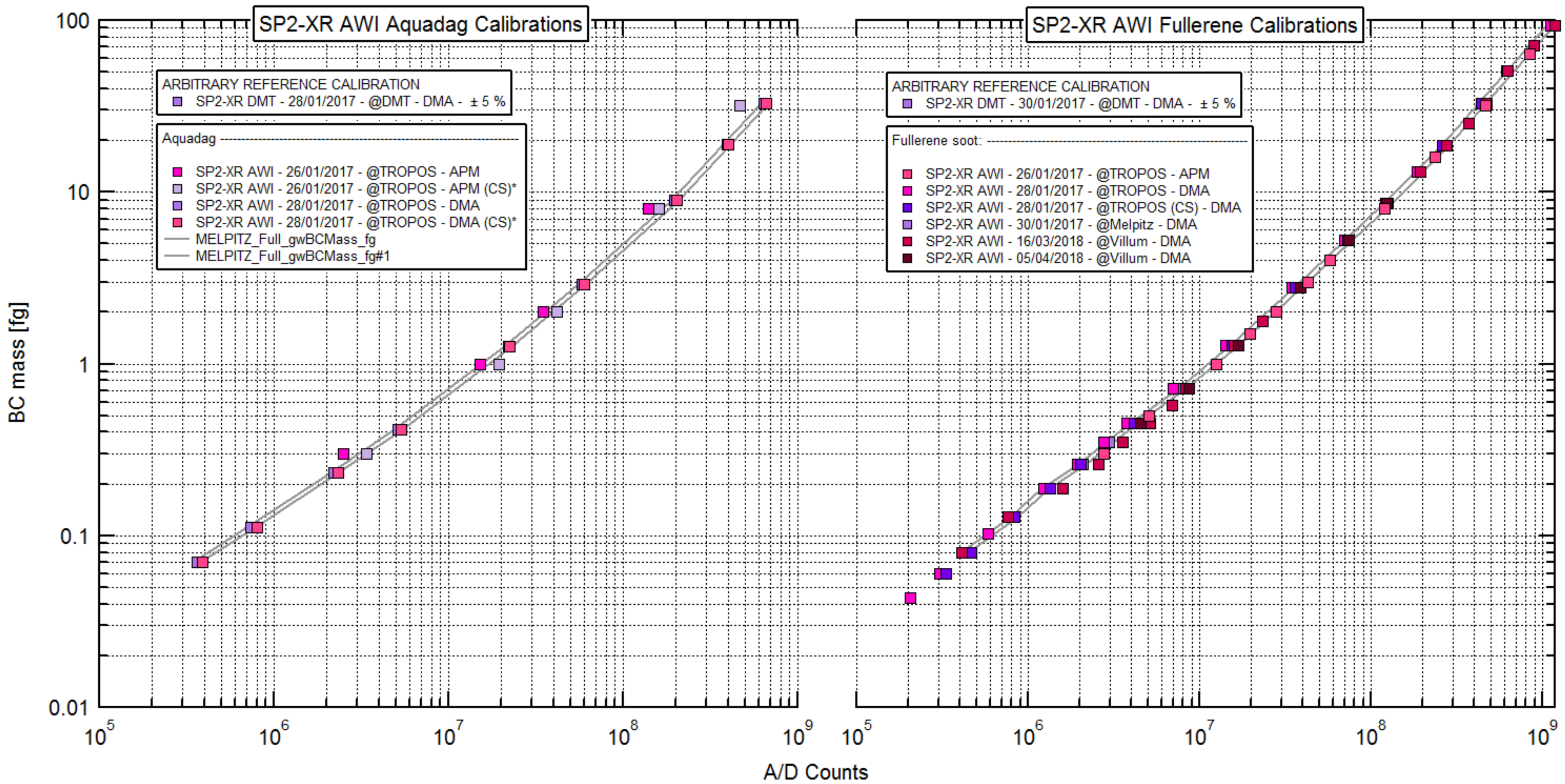
- **GOAL**: evaluate the stability of the Incandescence calibrations per SP2-XR unit



RESULTS

Incandescence Calibration Stability – SP2-XR AWI

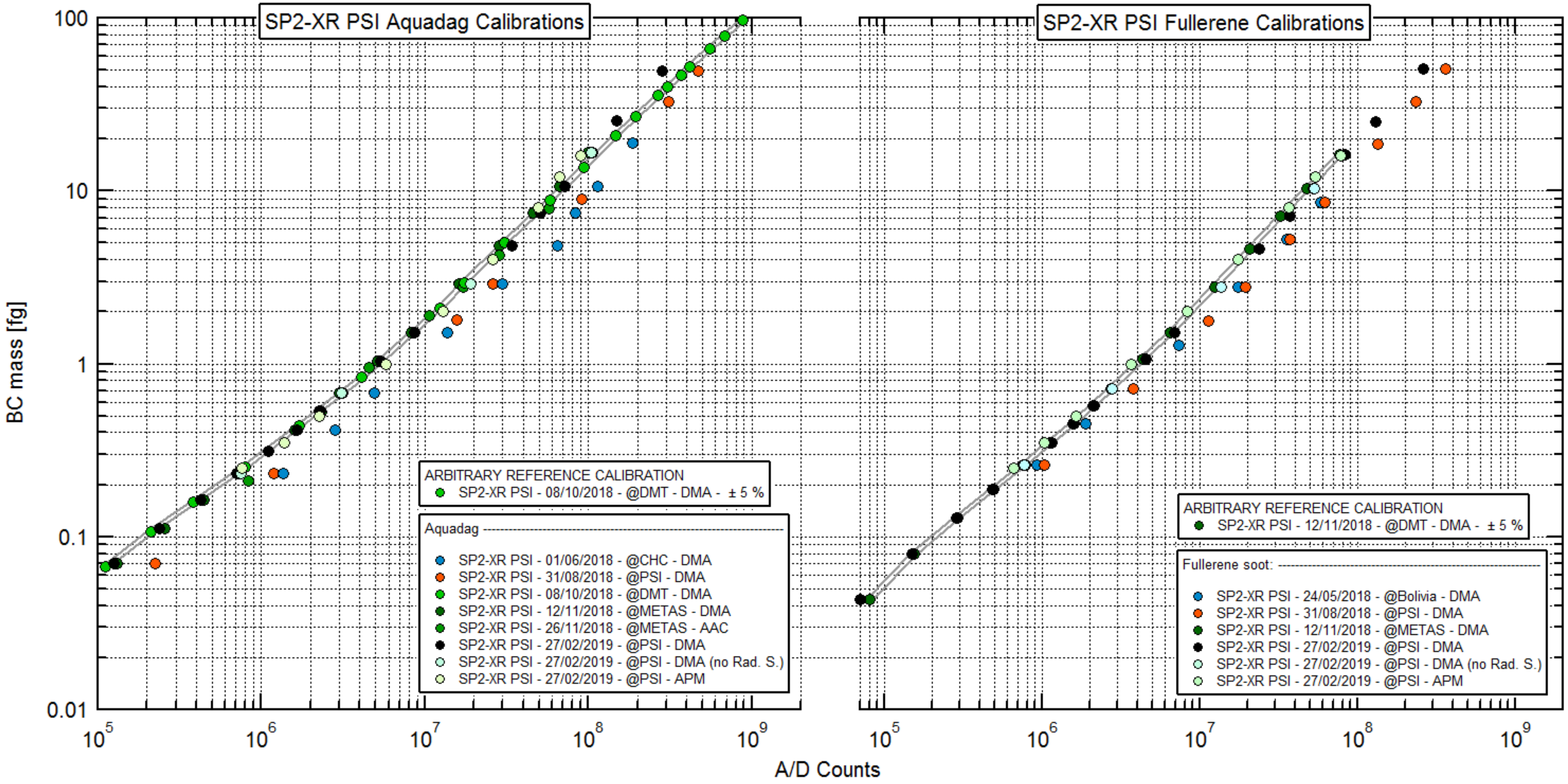
- **GOAL**: evaluate the stability of the Incandescence calibrations per SP2-XR unit



RESULTS

Incandescence Calibration Stability – SP2-XR PSI

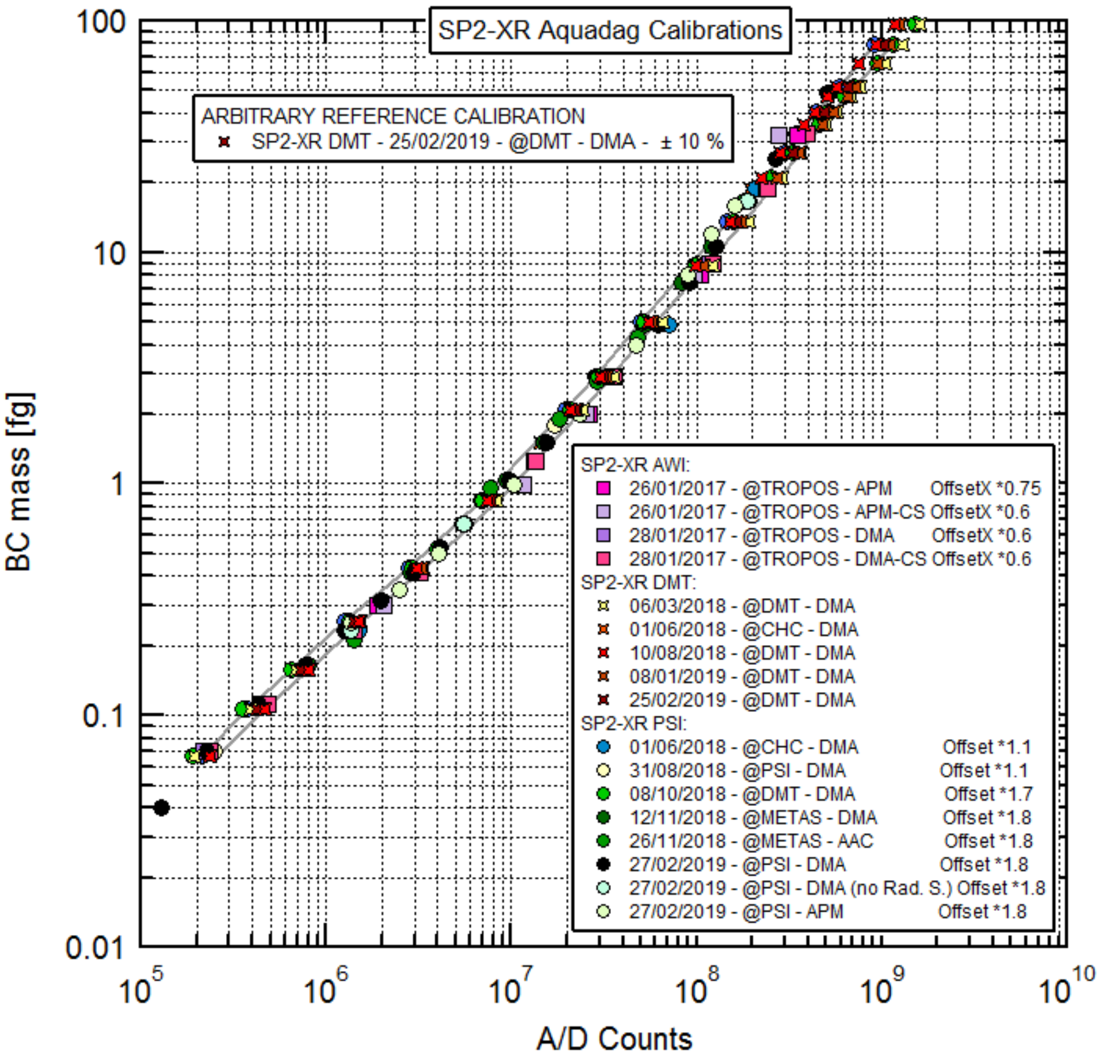
- **GOAL**: evaluate the stability of the Incandescence calibrations per SP2-XR unit



RESULTS

Incandescence Calibration Stability - AQ - SP2-XR PSI

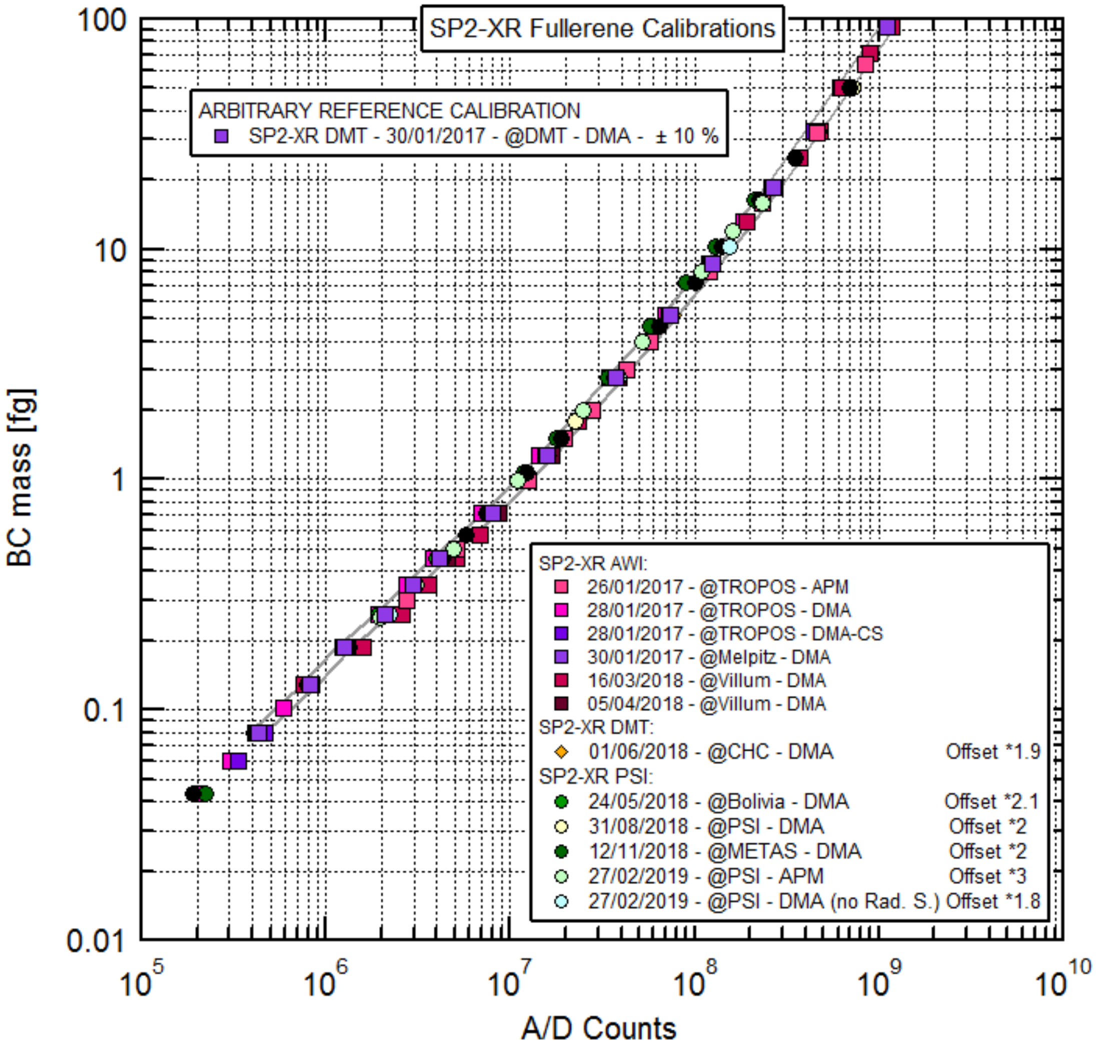
- **All the available Aquadag calibrations for the three XR units (with offset factors…)**



RESULTS

Incandescence Calibration Stability - FL - SP2-XR PSI

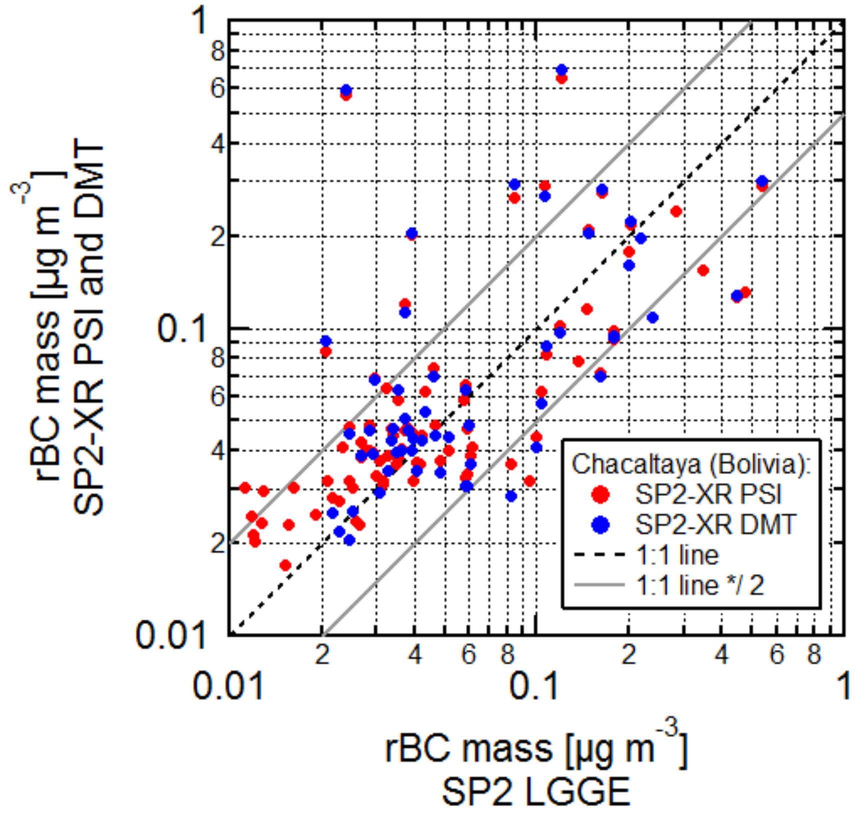
- **All the available Aquadag calibrations for the three XR units (with offset factors…)**

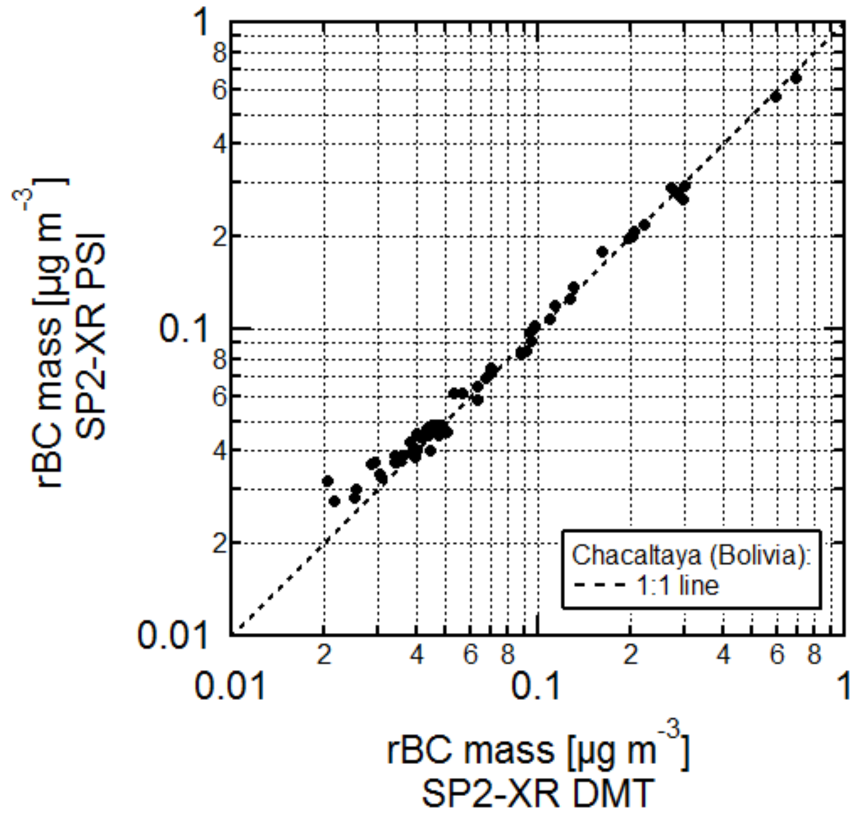


RESULTS

Ambient Measurements – the Bolivian campaign

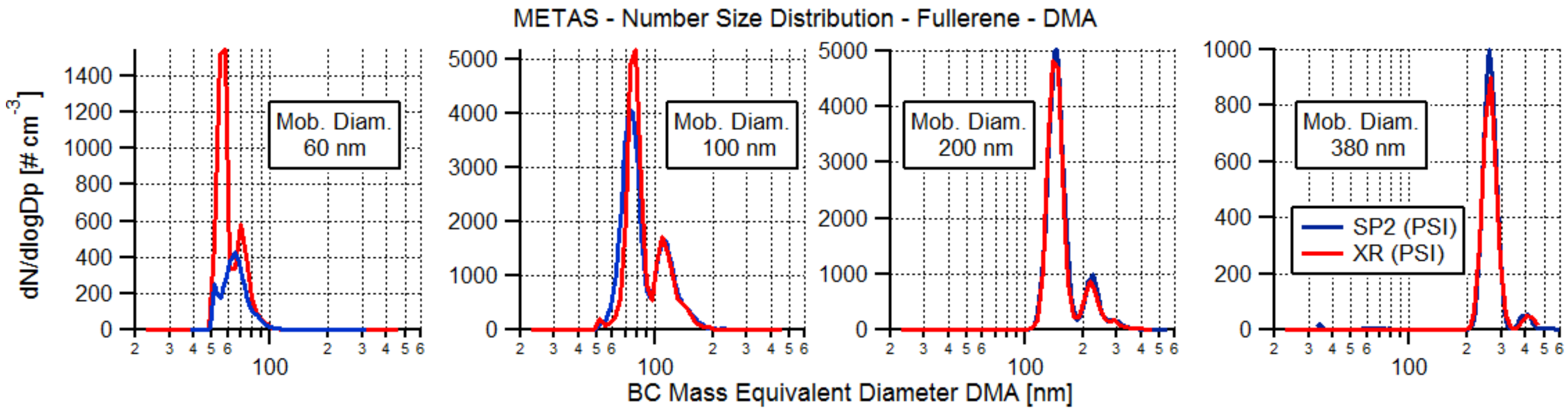
- **All the available Aquadag calibrations for the three XR units (with offset factors…)**

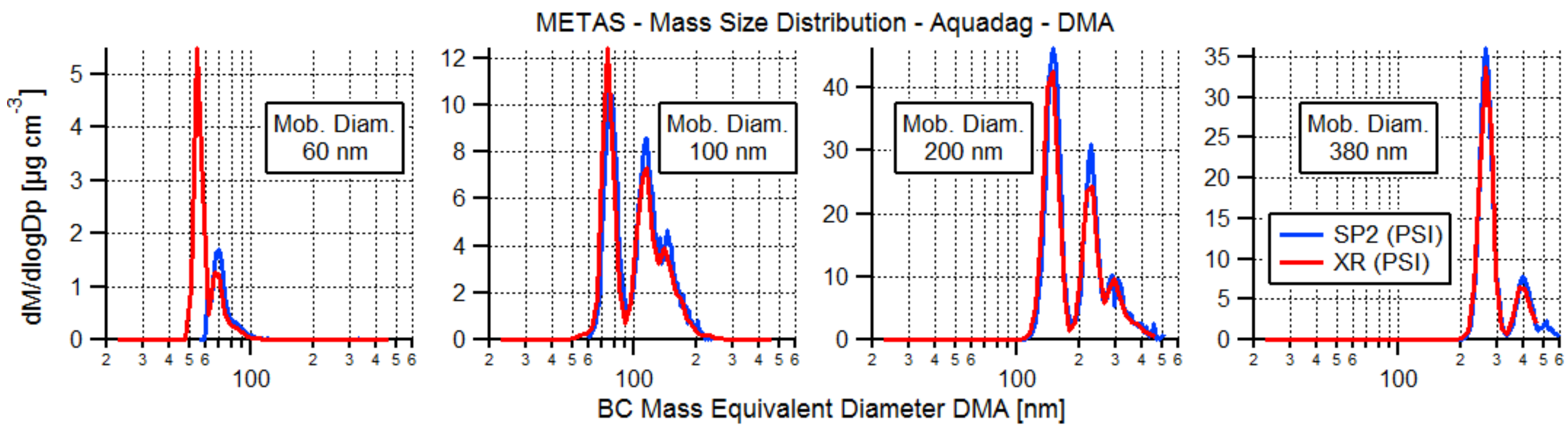




RESULTS

Mass/Number Size Distribution – Fullerene soot and Aquadag – Laboratory tests at METAS (using a DMA)





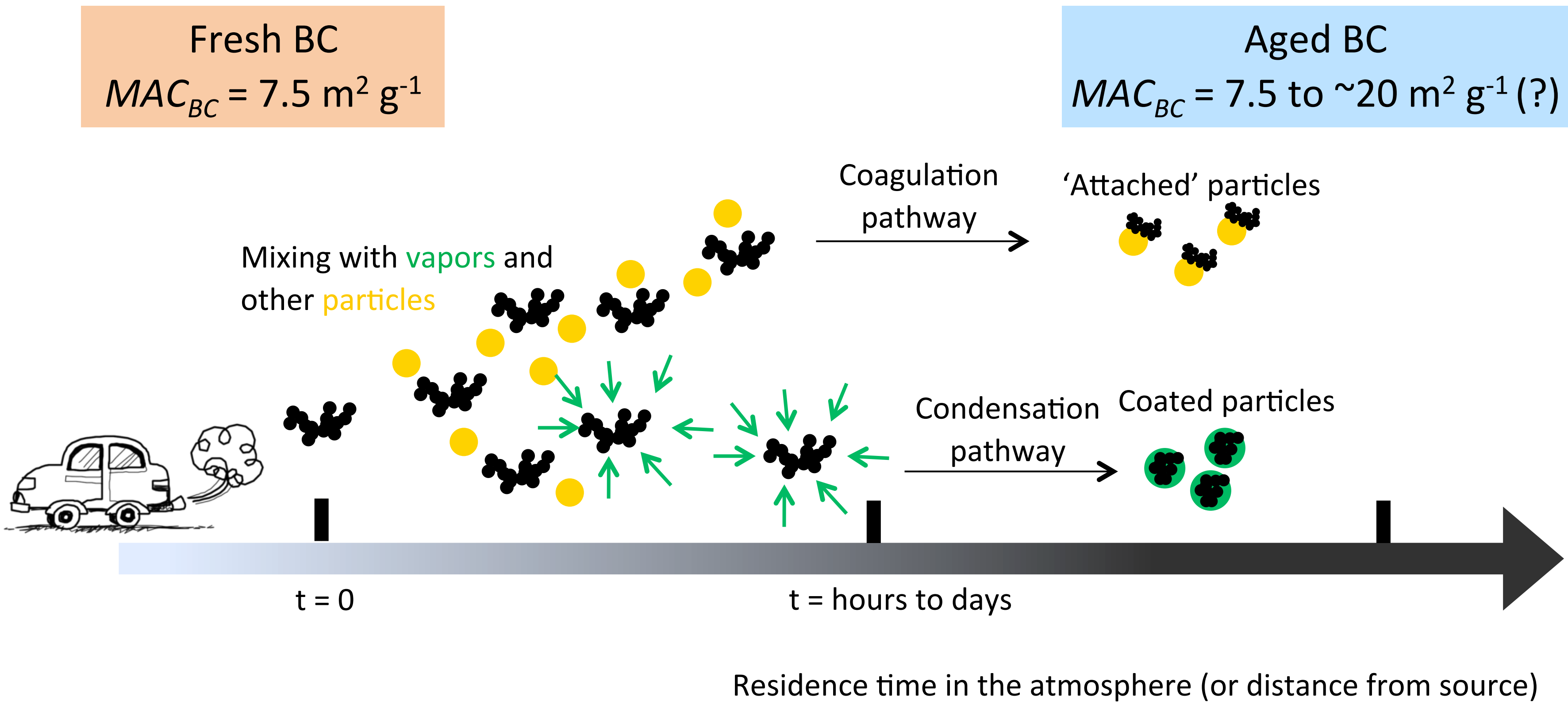
- The number and mass size distributions agree very well
- Below 100 nm (mob. diam.) the XR has a higher sensitivity than the SP2

INTRODUCTION

The BC «Mass Absorption Coefficient» (MAC)

General progression from an external to an internal mixture as BC particles age → MAC_{BC} is a function of particle age

Internal mixing can occur via competing pathways, which have different effects on MAC_{BC} → range of MAC_{BC} observed for aged BC in the atmosphere.

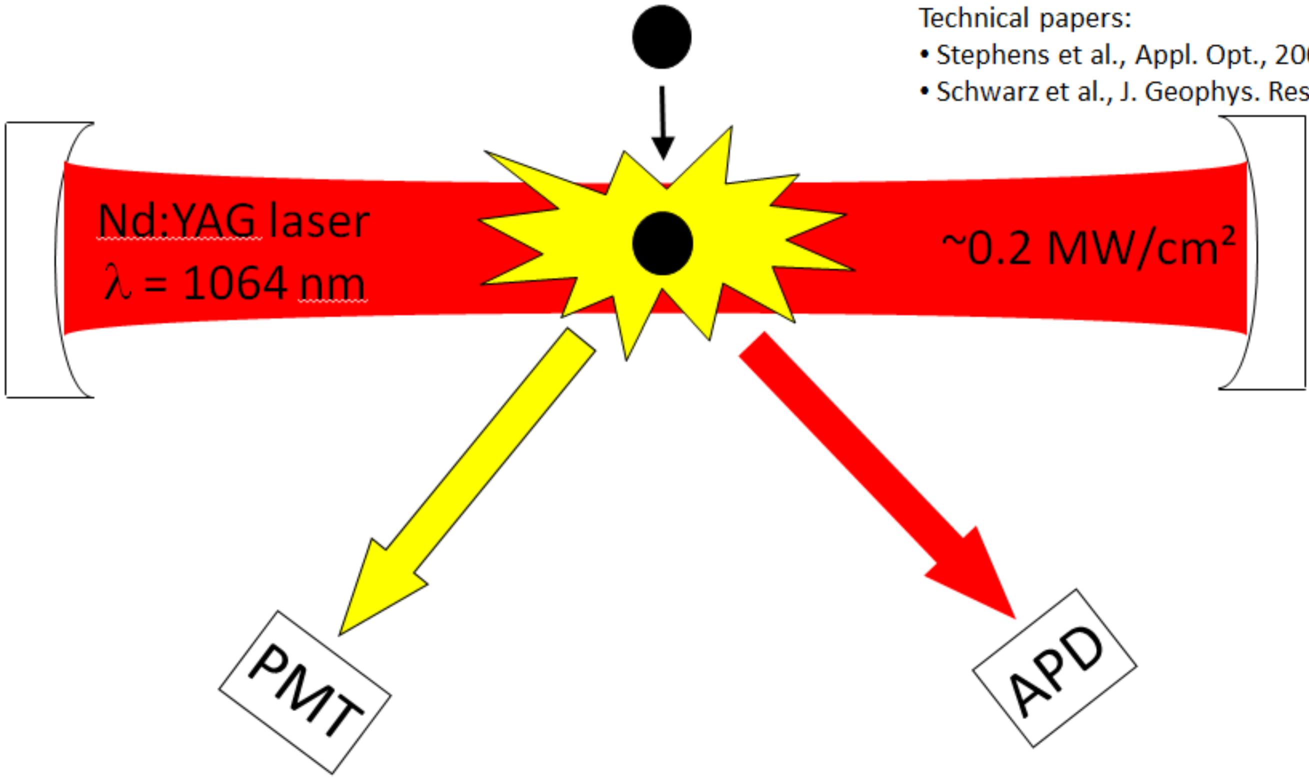


RESULTS

Ambient Measurements – the Melpitz campaign

The Single Particle Soot Photometer

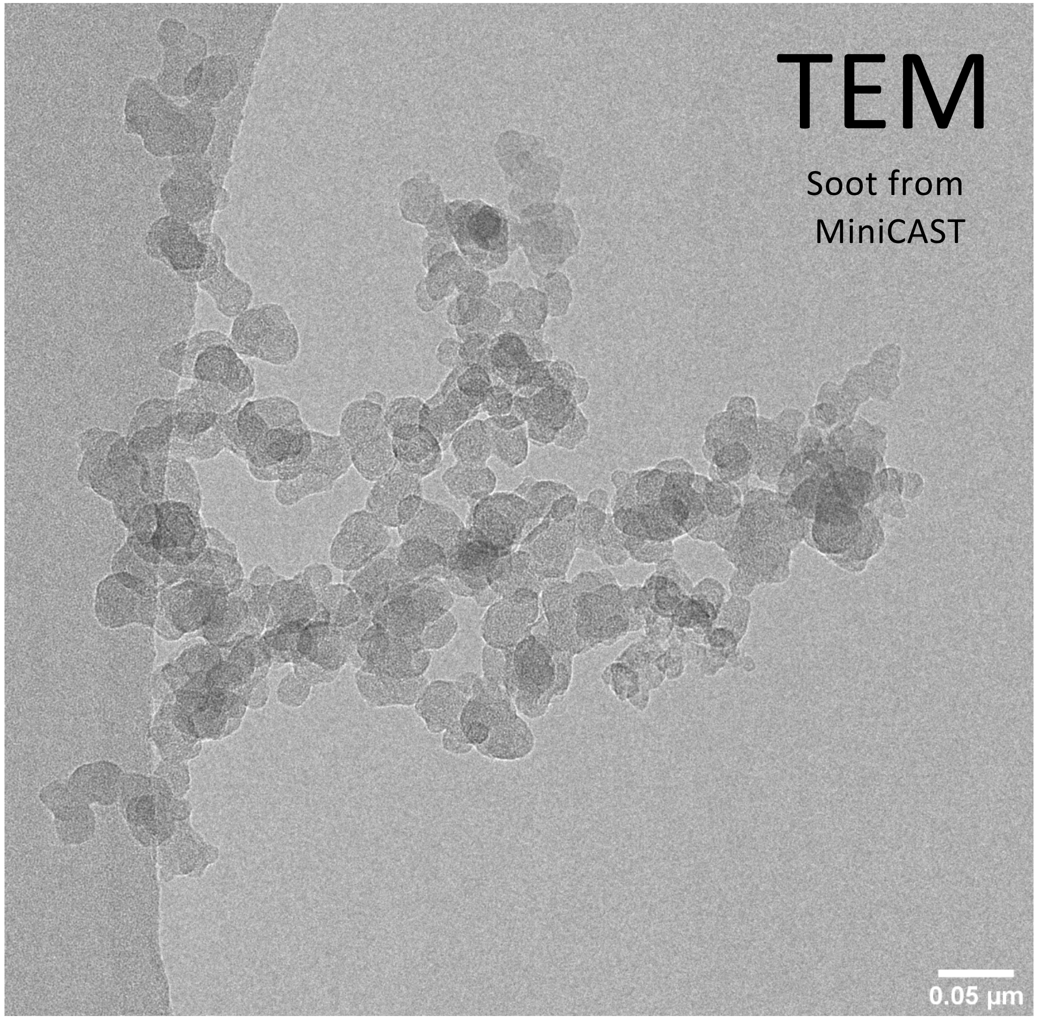
Technical papers:
- Stephens et al., Appl. Opt., 2003.
- Schwarz et al., J. Geophys. Res., 2006.



detection of
incandescent light
⇒ **BC mass**

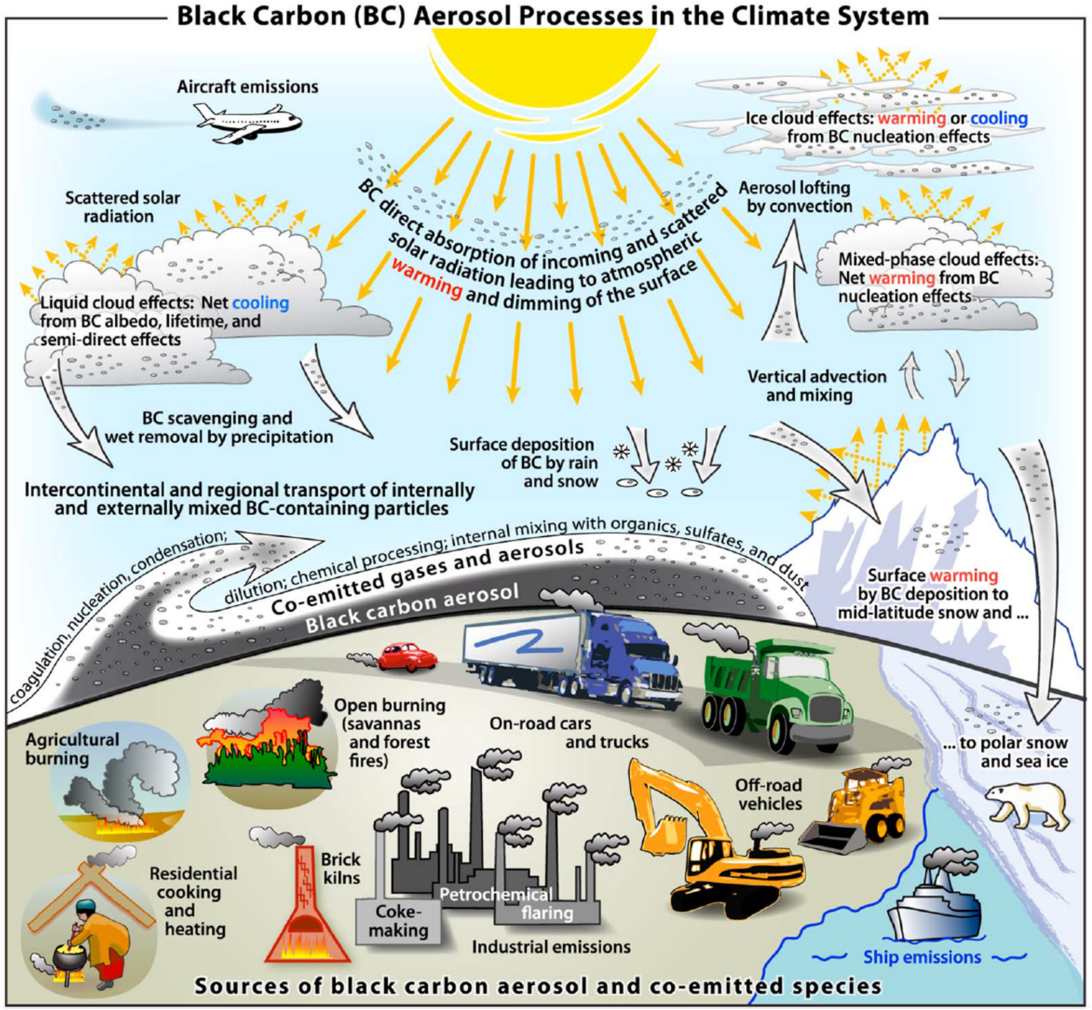
detection of
scattered light
⇒ **optical sizing**

BC in the Climate System



Ess et al., In preparation

BC in the Climate System



Bond et al., 2006

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INTRODUCTION

The BC «Mass Absorption Coefficient» (MAC)

- **Definition:**

$$MAC_{BC}(\lambda) = b_{abs,BC}(\lambda)/m_{BC} \quad [m^2 g^{-1}]$$

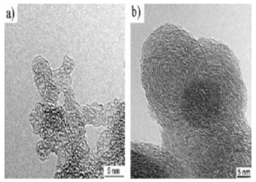
$b_{abs,BC}$: absorption coeff.

m_{BC} : BC mass conc. [g m^{-3}]

MAC_{BC} depends on:

Complex refractive index

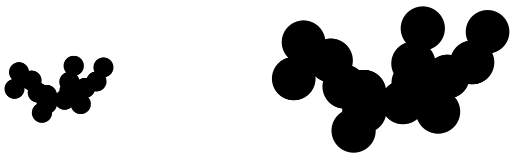
Measured rarely and with difficulty.



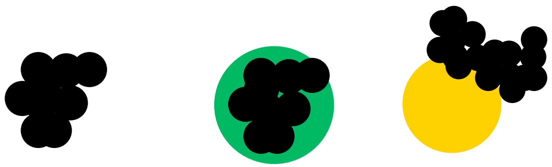
Müller et al., PCCP, 2007

Morphology

Size



Mixing state with other aerosols



Absorption Enhancement

INTRODUCTION

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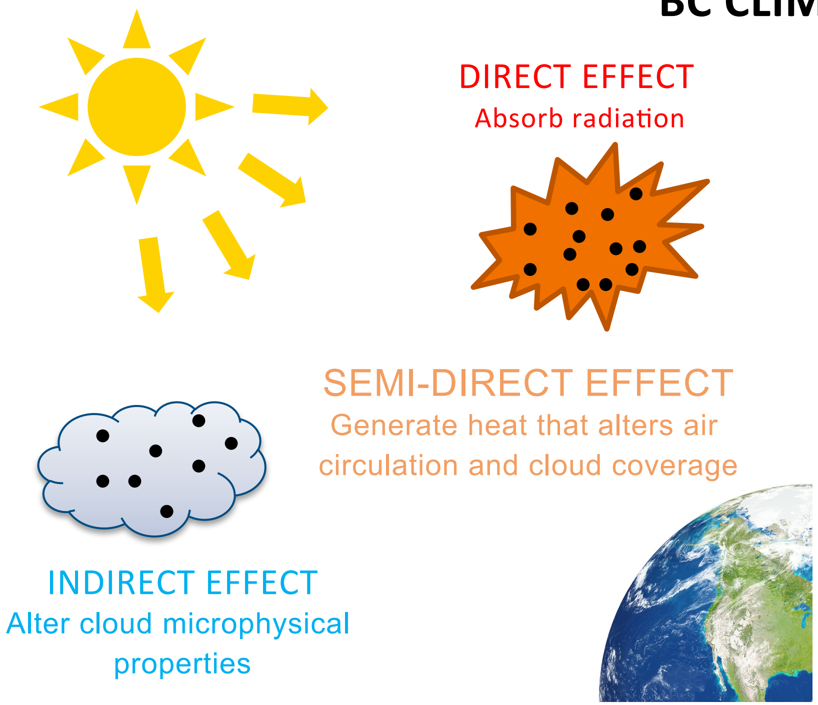
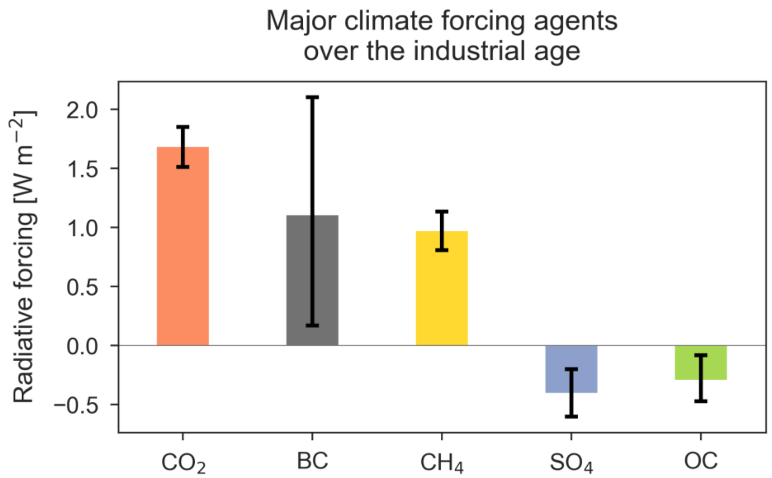
BC CLIMATE EFFECTS



⇨ **POSITIVE RADIATIVE FORCING**



BC radiative forcing is particularly uncertain

Data source: IPCC WG1 AR5 and Bond et al., JGR 2013

SP2 – Number Fraction of Thickly Coated Particles

Thickly Coated particle ➡ when the BC core volume is less than 30% of the total particle volume (methods based on time differences in the SP2 laser)