TNA User Report

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Project title	BC intercomparison workshop
Name of the accessed calibration centre	Calibration Centre for Soot Measurements (CCSM)
Number of users in the project	12
Project objectives (max 100 words)	The project objective is to improve the reproducibility of refractory black carbon (rBC) measurements using the laser induced incandescence technique (LII). The workshop was organized around both a training and a calibration activity, with the overarching goal to promote the use of standard operational procedures for the handling, data processing and calibration of LII instruments. This goal will ensure the robustness of inter-comparability of future rBC measurements conducted by international groups. For example, 5 of the hosted SP2 instruments will participate in the MOSAiC project in 2019-20, allowing the capture of a pan-Arctic snapshot of rBC concentrations and properties.
Description of work (max 100 words):	The workshop consisted of a training session and experimental activity. The training session on the PSI-SP2 software toolkit for data processing included data loading, calibrations and processing of rBC concentration, size distribution and mixing state. The training sessions on SP2 hardware included alignment procedures and setting of acquisition parameters. Experimental activity was conducted in one of the PACS-C3 simulation chambers and included: 1) Common calibrations with artificial soot standards (Aquadag/Fullerene) and diesel exhaust; 2) Internal mixing experiment with diesel exhaust and fullerene; 3) External mixing experiment with fullerene soot and ammonium sulfate; 4) interference experiments with volcanic ashes and magnetite.

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[2] UNI= University and Other Higher Education Organisation;
RES= Public Research Organisation (including international research organisations and private research organisations controlled by public authority);
SME= Small and Medium Enterprise;
PRV= Other Industrial and/or Profit Private Organisation;
OTH= Other type of organization.

[3] UND= Undergraduate; PGR= Post graduate; PDOC= Post-doctoral researcher; RES= Researcher EXP= Engineer; ACA= Academic; TEC= Technician.

[4] Reproduce the table for each user who accessed the infrastructure

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RES= Public Research Organisation (including international research organisations and private research organisations controlled by public authority);
SME= Small and Medium Enterprise;
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OTH= Other type of organization.
[7] UND= Undergraduate; PGR= Post graduate; PDOC= Post-doctoral researcher; RES= Researcher EXP= Engineer; ACA= Academic; TEC= Technician.

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The report will be made available on the eurochamp.org website. Should any information be confidential or not be made public, please inform us accordingly (in this case it will only be accessible by the European Commission, the EUROCHAMP-2020 project partners, and the reviewers). Please include:

- Introduction and motivation
- Scientific objectives
- Reason for choosing the calibration facility
- Method and experimental set-up
- Data description
- Preliminary results and conclusions
- Outcome and future studies
- References

Name of the PI: Marco Zanatta
Calibration center's name and location: Calibration Centre for Soot Measurements based at PSI in Villigen, Switzerland
Campaign name and period: BC intercomparison workshop
Text:

1. Introduction and Motivation

Black carbon (BC) aerosol particles are emitted into the atmosphere by incomplete combustion of fossil and biogenic fuels. The unique optical properties of BC are responsible for substantial impacts on the Earth's climate system as BC is second only to CO_2 as a warming agent in the atmosphere. Consequently, accurate measurements of BC concentrations and properties that are standardized across the BC research community are required to fully assess these climate impacts.

The observation of atmospheric soot particles began in the '80s (Cachier et al., 1989), and over the years has involved a wide variety of measuring techniques that include light attenuation (Hansen et al., 1984), thermal (Cachier et al., 1989), thermo-optical (Johnson et al., 1981), photoacoustic (Petzold and Niessner, 1996) and laser-induced incandescence (LII; Filippov et al., 1999). The heterogeneity and, in some cases, measurement protocols, of these techniques has caused accuracy and precision issues, the most important being the nature/properties of the detected particles and subsequent inter-comparability of the measurements. Unfortunately, even to this day, the comparability of black carbon concentration derived from these different techniques is still hampered by unacceptably large technique-to-technique variability. For example, while the performance of thermo-optical analysis and transmission photometry has been investigated in depth (i.e. Cavalli et al., 2010; Müller et al., 2011), the uncertainties and precision of LII based measurements were assessed by in only one study (Laborde et al., 2012).

Measurement of BC using LII, now commonly referred to refractory BC (rBC) within the LII community (Petzold et al., 2013) have become increasingly common over the last decade due to the commercial availability of the Single Particle Soot Photometer (SP2; DMT Inc.). The SP2 is a particle-resolved measurement that not only provides rBC mass concentrations, but also rBC size distributions and its mixing state. Quantification of the derived rBC mass loadings is achieved through an empirical calibration that relates the incandescence signal amplitude to rBC particle mass. Considerable expertise is required to properly setup and calibrate an SP2, as well as to process and analyse the large amounts of the particle-resolved data. As a consequence, the use of SP2 has been limited to few working groups and deployed for short/intensive field campaigns (days to weeks) rather than for monitoring activity (months to years). Interestingly, a new version of the SP2 developed by DMT (SP2-XR) opens up the possibility of routinely extending particle-resolved measurements of rBC to the monitoring regime. However, the performance envelope of the SP2-XR are not yet fully characterized.

2. Scientific objectives

The objective of this study is to improve the instrument-to-instrument reproducibility of refractory black carbon (rBC) measurements that employ laser induced incandescence (LII). In addition, the workshop was organized around both a training and a calibration activity, so as to promote a standard operational procedure for the handling, calibration, and data processing of SP2 instruments and datasets. This will improve the robustness of rBC measurement inter-comparability across research groups and field campaigns. For example, five of the hosted SP2 instruments will be participating in the MOSAiC project in 2019-20, allowing the capture of a pan-Arctic snapshot of rBC concentrations and properties (e.g., size distributions and mixing state).

3. Reasons for choosing the calibration facility

The Calibration Centre for Soot Measurements (CCSM) at PSI was chosen for this activity because it is equipped with a simulation chamber where it is possible to perform common calibrations and measurement intercomparisons with a large number of LII instruments using both standard soot calibration materials (fullerene soot, Aquadag) and real-world BC emissions (diesel car emissions coated with secondary organic aerosol). Additionally, the staff of the CCSM have very strong expertise in the optimal setup of SP2 instruments, and the processing and analysis of SP2 data.

4. Summary of activity

The workshop took place over two weeks [13 to 24 May 2019] and was composed by five primary activities that included theoretical and scientific discussions on the radiative forcing impact of BC, an LII-centric data treatment workshop, SP2 set-up workshop, and chamber experiments (Table 1).

Table 1 Activity performed at the PSI facility during the BC intercomparison workshop. Theoretical and scientific discussion in yellow, SP2 data treatment in orange, SP2 hardware and software setting in green, laboratory experiments in pink.

	13. May	14. May	15. May	16. May	17. May	18. May	19. May
Morning	Beginning of workshop	SP2 data treatment 2	Scientific presentations	SP2 data treatment 5	SP2 set-up 2	Ambient sampling	Ambient sampling
Afternoon	SP2 data treatment 1	SP2 data treatment 3	SP2 data treatment 4	SP2 set-up 1	SP2 set-up 3		
Night				Workshop dinner			
	20. May	21. May	22. May	23. May	24. May		
Morning	YAG laser power test	PSL calibration	Diesel soot calibration	Detection efficiency	Non-rBC interference		
Afternoon	Fullerene calibration	AQUADAG calibration	Diesel soot mixing	Fullerene mixing	Instrument packing		
Overnight			Diesel soot mixing	Fullerene mixing			

4.1. SP2 data treatment workshop

The workshop began with the introduction to the use of the "PSI SP2 toolkit -V4.111" software, written in IGOR-Pro by PSI staff, for the analysis of SP2 data. The presentations in this portion of the workshop covered the main functionalities of the software including: loading of SP2 raw data, calibration treatment, basic data treatment, postprocessing and coating thickness quantification. The goal of the activity was to compare the approach of different users to data treatment and to establish common procedure to be implemented during the data analysis by all PSI-SP2-toolkit users.

4.2. SP2-settings workshop

On 16-17 May, a hands-on training session on the routine alignment of the SP2 was undertaken (Table 1). This activity was conducted under the guidance of PSI staff and DMT technical staff and included adjustment of the output couple mirror to ensure a TEM00 laser beam shape, alignment of the mode aperture to maximise output laser power, and horizontal alignment of sampling jet (particle stream) with the laser beam to maximise signal detection and sensitivity. Extra time was dedicated specifically to the alignment of the position sensitive scattering detector (known as the split detector) as this measurement is central for quantifying coating thickness. Taken together, all these adjustments in this activity are needed to ensure the proper detection of rBC and non-rBC particles.

4.3. LII intercomparison workshop

The LII instruments were installed at the 9 m^3 coolable simulation chamber, on 17 May and sampled ambient air during the weekend (18-19 May). The experimental work began on 20 May and ended on 24 May (Table 1).

4.3.1. LII instrumentation

The LII instrumentation included eight single particles soot photometers (SP2), two single particle soot photometers with extended range (SP2-XR) – both instrument types manufactured by Droplet Measurement Technologies (DMT, Longmont, USA), one pulsed laser LII (LII-300) manufactured by Artium Technologies, Inc. (Sunnyvale, CA, USA) and one custom-built pulsed laser LII (CNR-ICMATE).

Ten international institutes participated in the workshop:

- SP2: Alfred Wegener Institute (AWI); Brookhaven National Lab (BNL), Environment Canada (EC), Finnish Meteorology Institute (FMI), Institute of Geophyiscal Research (IGE), Lund University (LU), Karlsrhue Instiute of Technology (KIT), Paul Scherrer Institute (PSI)
- SP2-XR: Alfred Wegener Institute (AWI), Droplet Measurement Technologies (DMT)

- LII: National Council of Research (CNR), Paul Scherrer Institute (PSI)

4.3.2. Infrastructure and instrumentation provided by the CCSM

The CCSM offered a wide set of supporting tools such as standard calibration material, aerosol generators, mixing chamber, and supplementary aerosol measuring instruments. The characterization of the LII instruments performance envelope was accomplished using differing aerosol types such as Fullerene Soot (FS12S011, L20W054, LI8U002, W08A039), Aquadag, diesel vehicle emissions (Opel Combo 1.3 CDTI, common rail without particle filter; Euro 4), and metal containing particles (volcanic ash and magnetite). In support of the core chamber measuremnts, a set of supplementary instrumentation was deployed to characterize or select aerosol properties: scanning Mobility Particle Sizer (SMPS); Aethalometer model AE-33 (AE-33); Photoacustic extinctionmeter (PAX); Aerosol Particles Mass analyser (APM), electron microscopy samples.

4.3.3. Experimental setups

Three different setups were used in order to calibrate the SP2s (Figure 1a), to intercompare measurements of internally and externally mixed BC aerosols, including coating thickness quantification (Figure 1b), and to invesitgate potential interferences caused by non-rBC refractory materials (e.g., dust, see Figure 1c). For all chamber-based measurements, the SP2s, SP2-XRs, and PAX were connected to a common stainless steel inlet and thus sampled the air in parallel. The LII-pulsed measurements were installed on a separate line which also contained the AE-33. The SMPS was installed on an independent line. The APM was used mainly during the calibration experiments as mass selector and located immediatelly after the aerosol generator or the 9 m³ coolable simulation chamber.

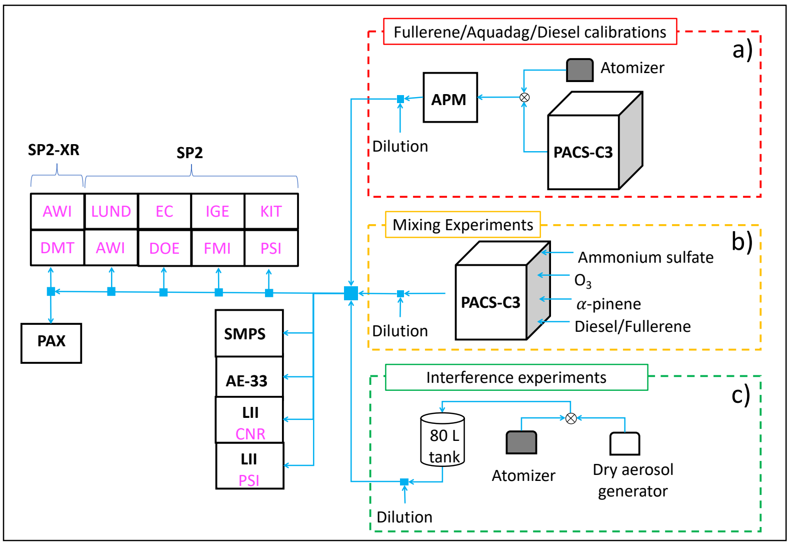

Figure 1: Instrumental setup implemented during the different intercomparison studies: a) SP2 calibration; b) internal and external mixing experiments; c) interference by metal containing particles.

5. Preliminary results

The data coverage obtained for the experimental part of the workshop was excellent. All LII instruments operated successfully over the full program of measurements with no major malfunctions. Reported here are preliminary results of the main experimentanl activities: calibration, rBC mixing state measurements, and mineral dust interference. Data analysis is currently ongoing by all groups and thus only example data is shown here: calibration curves for a subset of SP2/SP2-XR measurement (AWI, PSI, BNL, LGGE, KIT), rBC mixing state, and non-rBC particle (e.g., dust) interference (AWI SP2).

As highlighted above, common calibrations were performed using three different soot materials: fullerene soot, Aquadag and diesel soot. The soot particles were mass selected with the APM from 0.15 (~ 55 nm mass equivalent diameter) to 50 fg (~ 375 nm mass equivalent diameter) . The SP2 exhibited differing detection sensitivities to different particle types (Figure 2a) with detection sensitivity being highest for Aquadag followed Fullerene soot and then diesel exhaust. These findings are consistent with that reported by Laborde et al., (2012a). As expected, detection sensitivities for individual SP2s also differ (Figure 2a), since signal acquisition depends on instrument-dependent properties such as detector gain settings. However, application of intrument-specific calibration curves, such as those shown in Figure 2a, will remove instrument-specific settings and enable meaningful intercomparison of rBC mass concentrations and size distributions derived from the different instruments.

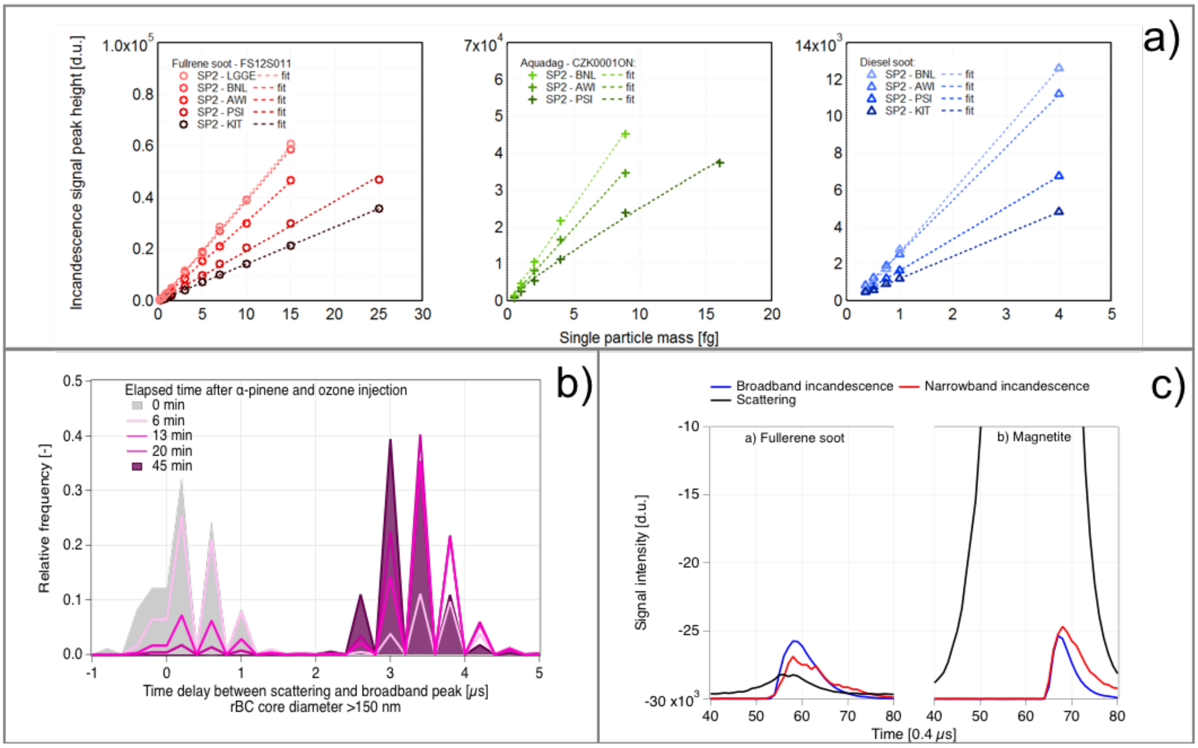

Figure 2 a) Calibration of the SP2 incandescence detector (broadband) with fullerene soot, Aquadag and diesel soot. Particles were mass selected with APM, no thermal treatment was applied. b) Temporal evolution of time delay (proportional to internal mixing degree) for diesel soot particles having a minimum core diameter of 150 nm. c) Raw scattering, broadband incandescence and narrowband incandescence signals measured by the AWI SP2 for fullerene soot and magnetite.

The internal mixing experiment was achieved by coating diesel soot with secondary organic aerosol generated from the ozonlysis of α-pinene. The SP2 "time-delay" between the scattering signal and the incandescence signal is a qualitative metric that can be used to assess the degree of internal mixing (large delay times indicate thick coatings on BC cores (Gao et al., 2007)). This method was applied to AWI SP2 measurments to show that thickly coated diesel BC cores were successfully generated during the experiment. Figure 2b indicates these coating were formed quite rapidly with internally-mixed rBC being formed within six minutes following injection of α-pinene and ozone into the PACS-C3 simulation chamber. After 45 minutes all of the diesel BC particles in the chamber were thickly coated. Work is now ongoing to quantitatively characterize coating thicknesses using the leading-edge-only (LEO) fit methodology with the goals of comparing mixing state analysis amongst the different instruments.

Finally, a non-rBC particle type was injected into the chamber as sampled by group so as to evaluate the effiacy of the SP2 to the detection of dust and the potential interference of this signal on the detection of rBC. Shown in Figure 2c is the raw scattering and incandescence (broadband and narrowband) signals for fullerene soot and magnetite recorded by the AWI SP2. Like soot, magnetite is refractory and capable of absorbing laser light at 1064 nm and thus incandesce. Comparison of the incandescence signals from rBC and magentite reveal that broadband incandescence signal intensity (blue curves), the shape, and the time of occurance of the incandescence signals are quite different, with magnetite incandescing later than the fullerene soot and more exhibiting more intense emission in the red part of the spectrum (narrowband detector). Morover, the comparison of scattering signals indicates that the magnetite particles are significantly larger than fullerene soot. Work is ongoing to further identify and characterize features that can distinguish magnetite from rBC signals, and to compare the extent to which the different instruments were able to detect and quantify (e.g. as a number fraction) the presence of interfering magnetite particles.

6. Outcome and future studies

As alluded to above, five of the SP2 instruments that participated in the workshop will be deployed at different sites around the Arctic as part of the year-long MOSAiC field campaign. The common calibrations performed during this workshop as well as data reduction methodologies will be applied to the in field measurements thereby ensuring that all derived rBC mass concentrations, size distributions, and mixing states are truly inter-comparable. In addition, the intercomparison results from the workshop (which remain to be analysed) will be used to assess the degree to which differences between the future measurements can be interpreted as real and not the result of measurement uncertainties.

One journal article is currently in preparation that will provide a campaign overview and present the SP2, SP2-XR and pulsed-shot LII intercomparison results from the workshop. This manuscript will include comparison of rBC mass concentrations, size distributions, and mixing state for each of the different aerosol systems measured. Particular focus will be placed on comparing the ability of the instruments to detect and potentially quantify interference from magnetite. A separate journal article is also planned that will focus more specifically on intercomparison of the two pulsed-laser LII systems. This manuscript will focus on comparing the relative sensitivity of the two instruments to the different rBC types that were measured.

Finally, given the complicated nature of SP2 data treatment, we plan to take advantage of the large number of participating users and instruments in the workshop to investigate the influence of individual operator decisions SP2 data treatment, as well as compare of different SP2 data processing codes (PSI SP2-toolkit, PAPI software, FMI code). These results of these investigations will be made available to the SP2 community through the campaign overview manuscript discussed above and presentations at future workshops and scientific conferences.

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