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DOE/SC-ARM-TR-301

Single-Particle Soot Photometer (SP2) Black Carbon Number and Mass Concentrations

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April 2024

ARM

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How to cite this document:

Sedlacek, AJ, and RC Jackson. 2024. Single-Particle Soot Photometer (SP2) Black Carbon Number and Mass Concentrations. U.S. Department of Energy, Atmospheric Radiation Measurement user facility, Richland, Washington. DOE/SC-ARM-TR-301.

Work supported by the U.S. Department of Energy,
Office of Science, Office of Biological and Environmental Research

Executive Summary

The single-particle soot photometer (SP2) records particle-by-particle measurements of the intensity of both the scattering signature and incandescence signature of particles that enter its laser beam. These intensities are then used to calculate refractory black carbon (rBC) masses and particle diameters. In previous U.S. Department of Energy Atmospheric Radiation Measurement (ARM) user facility field campaigns, the SP2 data was difficult to process because the manufacturer-supplied code was not scalable to distributed machines, making it unusable for the large amounts of data output by the SP2. This prohibited the SP2 from becoming an operational instrument for ARM. Therefore, PySP2 was developed to solve this issue and enable SP2 to be an operational instrument for ARM. This technical document summarizes the test data sets from the ARM North Slope of Alaska (NSA) site and the Multidisciplinary Drifting Observatory for the Study of Arctic Climate (MOSAiC) field campaign that were used to develop and test PySP2.

Acknowledgments

This work was made possible by the support and patience of the scientific community. We acknowledge Eddie Schuman from the ARM Data Center for his help in implementing the SP2 ingest to production.

Acronyms and Abbreviations

ARM	Atmospheric Radiation Measurement
BC	black carbon
DMT	Droplet Measurement Technologies
EPCAPE	Eastern Pacific Cloud Aerosol Precipitation Experiment
MAOS	Mobile Aerosol Observing System
MOSAiC	Multidisciplinary Drifting Observatory for the Study of Arctic Climate
netCDF	Network Common Data Form
NSA	North Slope of Alaska
rBC	refractory black carbon
SAIL	Surface Atmosphere Integrated Field Laboratory
SP2	single-particle soot photometer
TRACER	Tracking Aerosol Convection Interactions Experiment

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1.0 Introduction

The ARM Mobile Aerosol Observing System (MAOS) contains a suite of instrumentation for sampling various aerosol properties, including the SP2. The SP2 provides particle-by-particle statistics of particles that enter its laser beam. As shown in Figure 1, the SP2 will measure two signals from rBC-containing particles that enter the laser. The scattering signal is related to the coated rBC particle diameter. As the particle is heated by the laser beam, the inner rBC layer underneath the coating incandesces. The SP2 measures this incandescence signature to derive the mass and diameter of the uncoated rBC particle to obtain the mixing state of the rBC-containing particle.

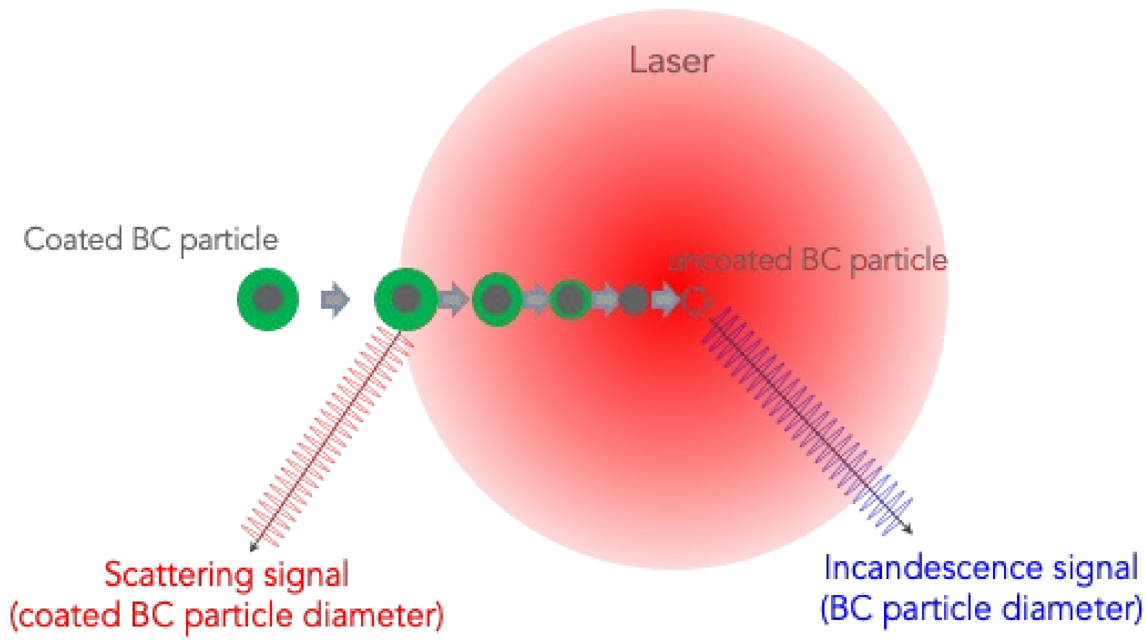

Figure 1. The basic operating principle of the SP2.

These scattering and incandescence signatures are recorded by the probe as raw voltages via the photodiode detectors for both the scattering and incandescence signals. Since these raw voltages are not interpretable by end users of the data, post-processing of the SP2 data is required to turn these voltages into interpretable rBC mass and diameters. Typically, the software provided by Droplet Measurement Technologies, based on the IGOR statistical analysis package, processed these raw signals into rBC masses and diameters. However, since millions of particles enter the SP2's sample volume per day for a typical field deployment, this translates into hundreds of gigabytes per day of waveform data to process. Data at this scale requires distributed computing to process, and the DMT-provided software was not capable of running on such systems. This prompted an effort to port the DMT-provided software to Python in a package called PySP2. This technical document demonstrates how PySP2 was used to develop the rBC mass and diameter data set for the ARM North Slope of Alaska (NSA) observatory and Multidisciplinary Drifting Observatory for the Study of Arctic Climate (MOSAiC) field experiment. PySP2 is available for use at https://ARM-DOE.github.io/PySP2.

2.0 Methodology

2.1 Deriving Particle-by-Particle Waveform Statistics

This section shows the methodology behind PySP2's calculations of the rBC particle mass and diameters. The SP2 records raw voltage from a 100-photodiode detector array for both the scattering and incandescence channels. In addition, each of the channels has an array of photodiode detectors with high gain for detecting smaller particles and low-gain detectors to better detect larger particles. In total, this results in eight different channels that are detecting the scattering and incandescence properties of each particle entering the SP2 sample volume.

- High- and low-gain broadband incandescence channels (~350-800 nm wavelength)
- High- and low-gain narrowband incandescence channels (~630-800 nm wavelength)
- High- and low-gain scattering channels
- High- and low-gain split-detector channels, used to determine particle position within the laser beam.

Figure 2 shows an example waveform from the high-gain scattering channel of the SP2 for a given particle. Since the intensity of light scattered by single particles approximately follows a Gaussian distribution, PySP2 will also calculate the Gaussian fit of the scattering particle waveforms as a function of photodiode number. The peak statistics such as height, position, and width are then calculated for each particle sampled by the SP2.

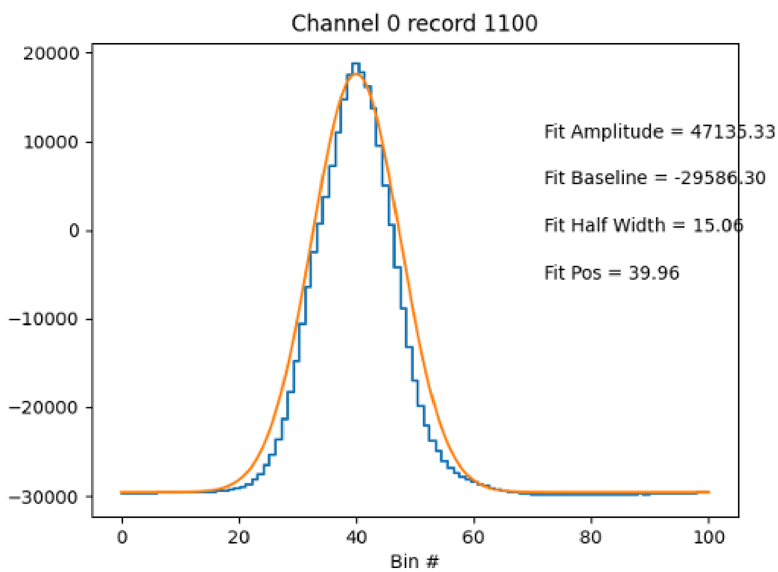

Figure 2. Example waveform of a particle that passed through the SP2's sample volume (blue). The orange line denotes the Gaussian fit to the waveform.

2.2 Artifact Elimination

Before the particles are used to derive rBC masses and sizes, certain artifacts in the measurements need to be removed. These articles are caused by particles entering the edges of the sample volume, multiple particles coincident in the laser beam, and other spurious signals. Therefore, the particles that did not meet the criteria listed in Table 1 were removed to eliminate such artifacts.

Table 1. Particle waveform thresholds used to eliminate artifacts from the SP2 data.

Particle Property Threshold	**Value**
Scattering Max Peak Height	60000
Scattering Min Peak Height	250
Scattering Peak Min Position	10
Scattering Peak Max Width	90
Scattering Min Peak Position	20
Scattering Max Peak Position	90
Incandescence Minimum Peak Height	200
Incandescence Maximum Peak Height	60000
Incandescence Minimum Peak Width	5
Incandescence Minimum Peak Position	20
Incandescence Maximum Peak Position	90
Incandescence Minimum Peak Ratio	0.1
Incandescence Maximum Peak Ratio	25
Incandescence Maximum Peak Offset	11

2.3 Calibration

The next step is to convert the particle peak heights, widths, and positions into rBC masses and diameters. This is accomplished by performing a calibration of the SP2 using fullerene soot or Aquadag particles with known diameters and masses (Gysel et al. 2011). For the calibrations used for the NSA and MOSAiC sites, Aquadag particles were used to calibrate the SP2. Figure 3 shows the results of the Aquadag black carbon (BC) mass calibration for the NSA site. For each particle, a linear fit of the Aquadag particle mass to the incandescence peak height is calculated to provide an empirical relationship between rBC particle mass and incandescence peak height. In Figure 3, the results of the calibration processed using both the original DMT-provided software and PySP2 are shown. The data from the calibration processed by both software products overlap, showing that PySP2 is well replicating the results provided by the DMT software.

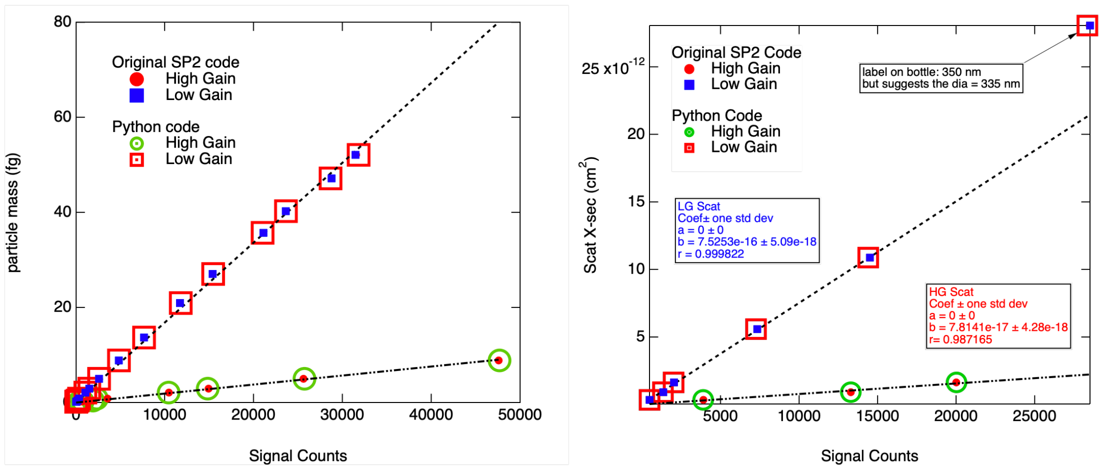

Figure 3. (left) The calibration of Aquadag particle mass as a function of SP2 incandescence signal counts (Ch 1). (right) as (left) but for the scattering cross-section and scattering channel (Ch 0). The results from both the IGOR-based processing and PySP2 are shown.

Figure 4 shows similar relationships derived for the scattering intensity as a function of scattering wave peak height. The scattering intensities are then converted to particle diameters assuming Mie scattering. Again, the DMT-provided software and PySP2 agree with regards to the particle peak heights.

2.4 Number and Mass Concentrations

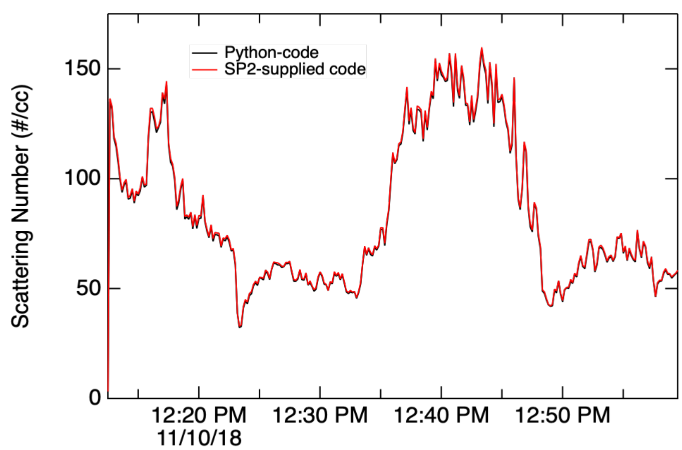

Figure 4. The scattering number concentration for a test period derived using PySP2 (black) and the DMT-provided code (red).

The number concentrations were derived by counting all the particles recorded by the scattering and incandescence channels over each 60-second rolling window and dividing this number by the sum of the sample flow over the 60-second period. Figure 4 shows a comparison of number concentrations derived by both the DMT-provided software and PySP2. In Figure 4, the number concentrations differ by less than 2 percent. This shows that PySP2 can replicate the number concentrations from the DMT-provided software.

Therefore, PySP2 generated the SP2 number and mass concentrations for the NSA and MOSAiC data sets. The files were generated on the ARM Stratus cluster with the help of the Dask package for scaling the PySP2 calculations to distributed computing. This enabled the NSA and MOSAiC data sets to be processed in under three days, a task that would have taken well over a month with the DMT-provided code on a standard laptop.

3.0 Data Object Description

PySP2 was used to derive the netCDF files generated for the aossp2bc60s.b1-level product for NSA, MOSAiC, Surface Atmosphere Integrated Field Laboratory (SAIL), and the Tracking Aerosol Convection Interactions Experiment (TRACER). Soon a product will also be available for the Eastern Pacific Cloud Aerosol Precipitation Experiment (EPCAPE). The description of the netCDF files provided in this data set is shown below. The file contains information about the BC particle size distribution and total mass.

```
netcdf epcaossp2bc60sM1.b1.20230403.000000 {
dimensions:
        time = UNLIMITED ; // (1416 currently)
        bin = 199 ;
        bound = 2 ;
variables:
        int base_time ;
                base_time:string = "2023-04-03 00:00:00 0:00" ;
                base_time:long_name = "Base time in Epoch" ;
                base_time:units = "seconds since 1970-1-1 0:00:00 0:00" ;
                base_time:ancillary_variables = "time_offset" ;
        double time_offset(time) ;
                time_offset:long_name = "Time offset from base_time" ;
                time_offset:units = "seconds since 2023-04-03 00:00:00 0:00" ;
                time_offset:ancillary_variables = "base_time" ;
        double time(time) ;
                time:long_name = "Time offset from midnight" ;
                time:units = "seconds since 2023-04-03 00:00:00 0:00" ;
                time:bounds = "time_bounds" ;
                time:standard_name = "time" ;
        double time_bounds(time, bound) ;
                time_bounds:long_name = "Time cell bounds" ;
                time_bounds:bound_offsets = -30., 30. ;
        double bin(bin) ;
                bin:long_name = "SP2 bin size median" ;
                bin:units = "um" ;
                bin:bounds = "bin_bounds" ;
        double bin_bounds(bin, bound) ;
                bin_bounds:long_name = "SP2 size bin bounds" ;
        double sp2_rbc_conc(time) ;
                sp2_rbc_conc:long_name = "Black carbon mass concentration" ;
                sp2_rbc_conc:units = "ng m-3" ;
```

```
		sp2_rbc_conc:_FillValue = -9999. ;
		sp2_rbc_conc:missing_value = -9999. ;
		sp2_rbc_conc:standard_name =
"mass_concentration_of_elemental_carbon_dry_aerosol_particles_in_air" ;
	double sp2_cnts(time, bin) ;
		sp2_cnts:long_name = "SP2 number concentration per bin" ;
		sp2_cnts:units = "m-3" ;
		sp2_cnts:_FillValue = -9999. ;
		sp2_cnts:missing_value = -9999. ;
	float lat ;
		lat:long_name = "North latitude" ;
		lat:units = "degree_N" ;
		lat:valid_min = -90.f ;
		lat:valid_max = 90.f ;
		lat:standard_name = "latitude" ;
	float lon ;
		lon:long_name = "East longitude" ;
		lon:units = "degree_E" ;
		lon:valid_min = -180.f ;
		lon:valid_max = 180.f ;
		lon:standard_name = "longitude" ;
	float alt ;
		alt:long_name = "Altitude above mean sea level" ;
		alt:units = "m" ;
		alt:standard_name = "altitude" ;

// global attributes:
		:command_line = "aossp2bc60s -s epc -f M1" ;
		:Conventions = "ARM-1.3" ;
		:process_version = "ingest-aossp2bc60s-1.2-0.el7" ;
		:dod_version = "aossp2bc60s-b1-1.0" ;
		:input_datastreams = "epcaossp2auxM1.a0 : 2.2 : 20230403.000000\n",
			"epcaossp2M1.a0 : 2.5 : 20230403.000000" ;
		:site_id = "epc" ;
		:platform_id = "aossp2bc60s" ;
		:facility_id = "M1" ;
		:data_level = "b1" ;
		:location_description = "Eastern Pacific Cloud Aerosol Precipitation Experiment
(EPCAPE), Scripps Pier, La Jolla, CA" ;
		:datastream = "epcaossp2bc60sM1.b1" ;
		:doi = "10.5439/1807910" ;
		:Calibration = "Fullerene equivalent AquaDag" ;
		:processed_with = "PySP2 0.1.0" ;
		:history = "created by user dsmgr on machine prod-proc1.adc.arm.gov at 2023-04-
04 01:39:06, using ingest-aossp2bc60s-1.2-0.el7" ;
}
```

4.0 Cases of Interest

An example in which smoke approaches the ARM NSA site is shown in Figure 5. During the evening of 15 February 2020, cleaner conditions persisted over the ARM NSA site. However, on the next day conditions become noticeably more polluted as the day progressed.

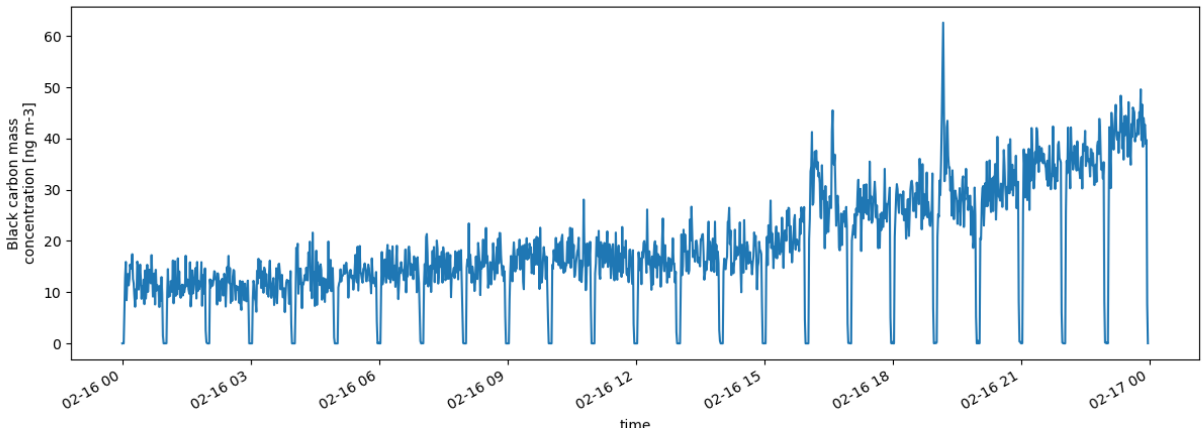

Figure 5. The BC mass concentration sampled by the SP2 at the ARM NSA site on 16 February 2020.

5.0 Future Work

Future work will involve incorporating the rBC coating thickness calculation into PySP2. We will launch this effort as an ARM Engineering Change Request during 2024. In addition, we will be processing more historical data sets from other ARM MAOS sites. The PySP2 package is available for use at https://ARM-DOE.github.io/PySP2.

6.0 References

Gysel, M. M Laborde, JS Olfert, R Subramanian, and AJ Gröhn. 2011. "Effective density of Aquadag and fullerene soot black carbon reference materials used for SP2 calibration." *Atmospheric Measurement Techniques* 4(12): 2851–2858, https://doi.org/10.5194/amt-4-2851-2011

Schwarz, JP, RS Gao, DW Fahey, DS Thomson, LA Watts, JC Wilson, JM Reeves, M Darbeheshti, DG Baumgardner, GL Kok, SH Chung, M Schulz, J Hendricks, A Lauer, B Karcher, JG Slowik, KH Rosenlof, ¨ TL Thompson, AO Langford, M Loewenstein, and KC Aikin. 2006. "Single-particle measurements of midlatitude black carbon and light-scattering aerosols from the boundary layer to the lower stratosphere." *Journal of Geophysical Research – Atmospheres* 111(D16): D16207, https://doi.org/10.1029/2006JD007076

Schwarz, JP, JR Spackman, RS Gao, LA Watts, P Stier, M Schulz, SM Davis, SC Wofsy, and DW Fahey. 2010. "Global-scale black carbon profiles observed in the remote atmosphere and compared to models." *Geophysical Research Letters* 37(18): L18812, https://doi.org/10.1029/2010GL044372

Slowik, JG, ES Cross, J-H Han, P Davidovits, TB Onasch, JT Jayne, LR Williams, MR Canagaratna, DR Worsnop, RK Chakrabarty, H Moosmuller, WP Arnott, JP Schwarz, R-S Gao, DW Fahey, GL Kok, and A Petzold. 2007. "An inter-comparison of instruments measuring black carbon content of soot particles." *Aerosol Science and Technology* 41(3): 295–314, https://doi.org/10.1080/02786820701197078

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