

# Supplementary Materials

## Methodology to Create Reproducible Validation/Reference Materials for Comparison of Filter-Based Measurements of Carbonaceous Aerosols that Measure BC, BrC, EC, OC, and TC

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This SI provides our experimental procedure and our methodical stepwise process to choosing the printer and ink that provides results that met our expectations. The main text describes our detailed evaluation of the chosen printer, printing onto other substrates, and a round robin study where four independent laboratories measured optical attenuation on separate sample sets. It also provides results from thermal-optical analysis (TOA) used to determine amounts of OC and EC deposited onto quartz-fiber filters (QFF) at different print densities or print levels as specified by the software. Print levels ranged from G00 (darkest or highest density of ink deposited) to G223 (lightest or least density of ink deposited). The stability of deposited ink on filter substrates over time is presented as well as results of the analysis of the inorganic and major organic chemical composition of the ink used in the chosen printer.

### S1. Experimental Procedure

The concept consists of using a commercial printer and associated ink to print onto a relevant substrate and measuring the optical attenuation (ATN) through the substrate before after printing. Relevant substrates include paper and various filter media used in ambient air quality sampling for particulate matter. We started with measuring optical attenuation on blank paper to obtain a baseline followed by printing 47 mm diameter circles onto blank paper to evaluate printer reproducibility. Circles were removed from either blank paper or paper with printed samples using a 25-mm diameter paper punch. Samples also were punched from filter substrate, such as the filter tape, using a metal circular punch. Punches from filter material were either 25 mm or 47 mm in diameter. Individual filter samples were taped along the top and bottom edges (top going into the print first) onto sheets of paper using standard commercial adhesive paper tape (e.g., 3M™ Micropore™ Surgical Tape, St. Paul, MN), placed into the paper tray, and passed through the printer in the normal manner. Typically, 6-9 samples taped to a single sheet of paper, plus a field blank, was considered a sample set. Ink was deposited onto the sample at one or multiple print densities (print levels). Different print levels or print densities were applied by different software programs (Microsoft Word and Adobe Photoshop) to obtain varying amounts of ink deposited onto the substrates. This also allowed us to determine if there was a difference in printing using these two programs. Print levels were defined by the fill density of the printed circle, set through font color, more colors, custom and varying

RGB, with print-in-black only specified under printer properties. This final setting is critical to ensure that the printer only prints using its black ink source.

Optical attenuation was directly measured on blank or printed paper samples or was measured on blank or printed filter samples before and after removal from the sheet, respectively, they were taped onto. ATN on all samples was measured through the center 15 mm of each sample at 880 nm and 370 nm using a commercial transmissometer (SootScan™, Model OT21, Magee Scientific, Berkeley, CA). After optical attenuation was measured, paper and filter samples were re-attached to the same sheet, in the same locations, for storage in a file folder or in individually labeled petri dishes. Each sample set included one sample that passed through the printer but did not have ink deposited directly. These are referred to as field blanks and measured potential contamination due to the potential of ink being deposited where it should not be deposited. No evidence of this was noted. Once we started using inkjet printers, the optical attenuation of every sample was measured before (referred to as the sample blank) and after printing, and each sample was blank corrected based on its sample blank, including the field blank. Sample blanks accounted for blank variability among filter samples in a given roll or lot. In all cases, the field blank associated with a given sample set, which had been sample blank adjusted, was subtracted from each filter's result in that set. ATN was measured on all samples in triplicate by measuring all punches in a sample set (6-9) once and then repeating the process two more times. Punches were randomly rotated between each of the triplicate measurements for all substrates to avoid systematic error in a single punch. For paper, each triplicate specific value was used in the mean and standard deviation. For all filter samples, all triplicates were averaged to obtain a single value for that sample. This process was maintained throughout the study. The average standard deviation (propagated) across all print levels of the rotated triplicate measurements for filter tape using inkjet printer #VI (n = 1,248) was 0.46 ATN and 0.41 ATN at 880 nm and 370 nm, respectively. These mean values are within the uncertainty of the OT21 ( $\pm 1$  ATN) and indicate the best average reproducibility that can be achieved.

## **S2. Reproducibility of ATN of Blank Paper**

Printing onto paper was initially used to evaluate the variability of printing using different printers. Achieving this required determining the consistency of measured optical attenuation with clean paper. Different types of clean paper were examined as an initial step. The reproducibility in optical attenuation was determined at different locations on a sheet of paper, across multiple individual sheets within a ream, multiple reams within a box, and in some cases paper from different boxes of the same brand. The goal was to find the brand of paper that provided the best precision, measured as one standard deviation of the average optical attenuation and smallest range of ATN values, within a sheet and across multiple sheets. The most uniform paper would then be used to evaluate the variability of printing by different printers. One brand of 100 % post-consumer paper (recycled) and five brands of premium paper, as defined by the manufacturer, were examined. Nine 25-mm punches, taken from the same location on each sheet, were removed from each sheet of paper and optical attenuation through each sample was measured as described above. A summary of measured ATN by paper type is presented in Table S1. Results in this table are averaged across sheets (see footnotes b and c). Optical attenuation through blank paper varied by paper type from ~-1.8 to ~6.5 ATN at 880 nm and from approximately -6.0 to 42.9 at 370 nm indicating that optical properties vary with different types of paper. Using Hammermill® LaserPrint as an example, the precision, as one standard deviation of the mean across all positions of an individual sheet, for 8 sheets, ranged from 0.8 to 1.45 ATN at 880 nm and from 2.95 to 4.09 ATN at 370 nm. Precision (SD) across sheets, at a given position, ranged from 2.92 to 3.63 at 880 nm and 4.34 to 7.75 at 370 nm. Thus, while optical attenuation varied by paper type, within a given paper type, especially within a given box, the average variability across sheets at a given position or across positions of a given sheet was, in general, less than 1-2 ATN at 880 nm and slightly higher at 370 nm.

### **S3. Laser Jet Printers – Printed Paper**

Initially, the variability of printing was examined using three laser jet printers (Table S2 footnotes). Two paper types, one post-consumer and one premium, were used to determine if the intrinsic reproducibility of the blank paper was affected by printing. Nine 47 mm diameter circles were printed in the same location on each sheet and most sample sets included a field blank. A 25-mm circle was punched from the center of each 47 mm printed circle. Measurements were typically made within one or two days of printing. Optical attenuation was measured on each sample as described above. In general, the standard deviation of the numerous field blank triplicates was below 0.5 ATN. A limited number of circular samples were printed at the same print level in multiple locations on a sheet while keeping the positions consistent across several sheets to provide insight into the reproducibility and variability of the entire process, including both paper and printer. Using Hammermill LaserPrint paper and the Lexmark® V2A printer as an example (Table S2 footnotes), the standard deviation of the optical attenuation, averaged across the 9 positions of an individual sheet for each of the 10 sheets where the same print level was used, varied from approximately 1.72 – 5.17 ATN at 880 nm and from 2.89 – 7.99 ATN at 370 nm. The range of the average optical attenuation across the 9 positions of the 10 individual sheets was 10.7 ATN and 17.2 ATN at 880 nm and 370 nm, respectively. For the same printer and paper combination and across sheets at the same position and print level, the standard deviation of the optical attenuation varied from about 3.57 – 4.82 ATN (propagated average SD of 4.10 ATN) at 880 nm and from 4.93 – 8.51 ATN (propagated average SD of 9.37 ATN) at 370 nm. The average variability across sheets, based on one standard deviation was, on average about 3.5 (880 nm) to 1.6 (370 nm) times higher than that observed for blank paper. A summary of the mean measured ATN, at both wavelengths, by printer type and paper type is presented in Table S2. The average optical attenuation, at print level G170 using Hammermill LaserPrint paper, varied across the different printers from about 36 to 91 ATN at 880 nm and from about 36 to 108 ATN at 370 nm. These results indicate differences in the optical attenuation of the toner for different laser jet printers or that different printers deposit different amounts of toner at the same print level. Based on the toner specification sheets and likely slight differences in how the toner is deposited and adhered to the paper, both likely play a role. The ATN range was less than ~15% of the mean at 880 nm and less than 30% of the mean at 370 nm. Field blank ATN values (as defined above) were high for some paper types, especially at 370 nm, although consistent within a paper type as indicated by the standard deviation of the field blank.

### **S4. Printed Filter Material – Laser Jet Printers**

Commercial Aethalometer® roll filter tape (Part No. 8050, 3.0 cm x 1000 cm for Model AE33 Aethalometer, Magee Scientific Corporation, Berkeley, CA) was used throughout as the filter material substrate unless otherwise noted. Ten circular punches (~25 mm in diameter) were removed along approximately 30 cm segments of the roll filter tape and attached, in set positions, onto a standard sheet of paper. Sample blanks on pre-printed filters were not obtained on these samples. Four laser jet printers, three color and one black and white (Table S2 footnote) were used to print onto the roll filter tape material. Each sample set included a field blank and nine printed samples, and all samples in a set were printed at the same software print level (print density, G170). Using the Lexmark V2A printer as an example, the standard deviation of the optical attenuation, averaged across the 9 positions of an individual sheet for each of the 38 sheets where the same print level was used, varied from approximately 1.88 – 5.68 ATN at 880 nm and from 3.09 – 10.5 ATN at 370 nm. For the same printer and across sheets at the same position and print level, the average standard deviation varied from 6.50 – 8.15 ATN (propagated average SD of 7.40 ATN) at 880 nm and from 8.53 – 11.1 ATN (propagated average SD of 9.89 ATN) at 370 nm. A summary of mean, standard deviation, and range in measured optical attenuation at both wavelengths, obtained using Aethalometer filter tape, and printed using each of four laser jet printers is presented in Table S3. In general, mean ATN ranged from between 30 ATN (Ricoh) to about 82 ATN (Lexmark V2A) at 880 nm whereas reproducibility, as

standard deviation, ranged from about 0.4 ATN (n = 9 samples) to 3.5 ATN (n = 334 samples) (Lexmark V2A, 880 nm). Similar results were observed at 370 nm (Table S3). A large range in ATN (880 nm) across samples was observed varying from <5% to ~27% of the mean. Similar results were observed for measurements at 370 nm.

The relatively large variability in the ATN of the field blanks along the filter tape roll (Table S3, last column) indicated the need to obtain a sample blank value for each sample, including the field blank, before printing. Recently, Sreekanth et al. (2019) reported similar variability in blank Aethalometer filter tape. Measuring optical attenuation on the blank filter tape follows the standard approach used in the Aethalometer where the blank measurement for each filter spot is used as the baseline for that sample. Therefore, in the remainder of experiments conducted, a sample blank was obtained on each filter, including the field blank, to blank adjust that specific filter whereas a field blank associated with each sample set was obtained and used for all filters in a set.

During this initial set of experiments with roll filter tape, the position of the paper tape to hold down the filter material on the sheet of paper was varied either by placing paper adhesive tape on the left and right side of the sample circle or on the top and bottom of the sample circle (the top is the side entering the printer first). Taping on the top and bottom resulted in less distortion of the Teflon-coated glass-fiber filter samples as they passed through the printer so this approach was used with this filter material for the rest of the study. Quartz-fiber filter samples required taping the edges of the entire filter, likely the result of a thicker more fragile material. White areas along the edges of the filters shown in Figure 1 indicate where tape was placed around each type of filter.

## **S5. Printed Filter Material – Inkjet printers**

As described above, we initially used laser jet printers (SI3 and SI4) to evaluate our method because they were easily available and expected to provide consistent results as opposed to an inkjet printer, simply based on an expected higher print quality as a result of the printing process. Laser jet toner is a powder and the printer uses heat to bind the particles to the paper. An average toner includes plastic (polyester, up to 95% of the powder), polypropylene wax (to keep the toner from sticking to the printer rollers), high purity carbon black (black toner) or a colored pigment, fumed silica (to keep the particles from sticking to each other), and typically a metal oxide. The metal oxides are used to help attach toner particles to the paper where they act as charge control agents (e.g., particles of iron oxide to maintain the charge given to the particles) or have other uses (Matsushita, 2012; Bello et al., 2013; Pirela et al., 2015; Shara et al., 2018). However, metal oxides may act as combustion catalysts and influence the OC/EC split in thermal-optical analysis (TOA; Chow et al., 2001; Chow et al., 2005; Wang et al., 2010; Panteliadis et al., 2015). This is potentially problematic since TOA was used to measure the EC and OC content of the filter deposits (see Section 2.4 in main text: Experimental, Quartz-fiber Filters). Laser jet printers also had higher than desired variability, as standard deviation, in optical attenuation (Table S3). For these reasons, we subsequently tested inkjet printers.

Inkjet printers deposit picoliter size liquid droplets without heating to fix the ink. Inkjet inks (Table S4) are primarily composed of water and or at least one water-soluble organic compound from a large range of compounds, ink or pigment (the latter is a powder dispersed in the liquid and is usually carbon black for black pigmented inks), a water surfactant and or dispersant, and other components. Inkjet ink typically does not contain metal oxides, although some colored inks have metals associated with the organic compound that gives the ink color. The latter was one reason for only using printers that printed in black only and did not require colored ink, i.e., print in gray-scale that uses colored ink to obtain black color on a sheet.

The variability in optical attenuation (ATN) printed onto roll filter tape was tested using six inkjet printers, two of which were self-identified as “photo printers” (Table S4). Filters were processed in the same manner as described above. The results of these analyses are presented in Table S5, albeit at slightly different print levels. Four of the inkjet printers did not meet our criteria for reproducibility or overall range in optical

attenuation. For example, the ATN measured at 880 nm, for inkjet printer #I, decreased over time as the ink level decreased in the cartridge, although initially it appeared to be an excellent candidate. Since this printer used a print cartridge, we believe the pigment may have settled to the bottom of the cartridge during the period of use. However, shaking the cartridge did not improve results. One inkjet printer (#II) apparently did not include carbon black in the black ink (maximum ATN at 880 nm at the highest print level as 2.3 ATN). It did respond well at 370 nm however (ATN of 140), likely due to the organic dyes in the colored ink. Inkjet printers #IV and #V only printed in gray scale indicating they used colored ink along with black ink to print in gray to black. Based on results presented in Table S5, inkjet printer #VI was chosen for printing onto a variety of substrates attached to paper or directly onto paper for the remainder of the experiments. The inkjet pigment contained sufficient carbon black to reproducibly obtain optical attenuation values ranging from < 5 to >140 ATN at 880 nm and from <5 to >165 ATN at 370 nm, with low overall standard deviations at print levels of G223 to G00 (lightest to darkest, respectively, where G00 was the darkest print density achievable by a single pass through the printer with the software - printer combination).

## **S6. Software Programs**

Samples were printed using two common software programs (Adobe Photoshop and Microsoft Word) to determine whether the choice of software affected the consistency and reproducibility of the printed deposit. Six print levels and a field blank were used. Optical attenuation was measured at both wavelengths as described above on approximately 60 sample sets across three filter tape rolls. The average difference between ATN values obtained when printed with Adobe Photoshop versus Microsoft Word for each of the three filter tape rolls was less than 1.2 ATN at either wavelength except at the darkest print level used (G70), where ATN varied by <2.3 at 880 nm and <2.8 at 370 nm. The relative difference across print levels was less than 3.5% with the largest relative differences typically at the lowest print level (ATN of 5-7). There appeared to be no difference in results between samples created when using the two software programs: consequently, we used Adobe Photoshop for the remainder of the study.

## **S7. Multiple Operators**

Five undergraduate students, typically only one at a time, assisted with optical attenuation measurements related to printing on roll filter tape during the study. Optical attenuation was measured as described above. Each student was fully trained in the use of the optical transmissometer and each re-analyzed a series of samples previously measured by the lead operator. For these samples, ATN measurements ranged from about 6 ATN to 135 ATN at 880 nm and from 6 ATN to about 155 ATN at 370 nm. Once results obtained by the students were consistent with those obtained previously by the lead operator (within 1-2 ATN at lower ATN values and within 10% at higher ATN values), the students measured new samples, including the sample blank, field blank, and their respective printed samples. All students achieved this level of reproducibility. Two students overlapped in time and provided a series of seven sample sets where the two students and the lead operator analyzed the same samples. Two transmissometer instruments also were used. The two instruments and operators agreed to within 3% at 880 nm and 370 nm except at the lowest print level (lightest samples) at 880 nm. Overall, the average absolute difference was <2 ATN at 880 nm and <1 ATN at 370 nm across all print levels.

**Table S1.** Variability of optical attenuation (ATN) measured at 880 nm and 370 nm through blank paper across multiple reams and boxes as indicated. .

<b>Paper Type<sup>a</sup></b>	<b>Mean ATN<sup>b</sup></b>	<b>SD<sup>c</sup> of Mean ATN</b>	<b>Range ATN</b>	<b>n<sup>d</sup> (Reams/Boxes)</b>
<b>– 880 nm –</b>				
<i><b>100% Post-Consumer</b></i>				
Cascade Rolland Enviro100™	–1.75	1.17	10.5	31 (7/3)
<i><b>Premium</b></i>				
Hammermill® Great White® 30	0.51	0.95	3.49	5 (1/1)
Xerox® Premium Laser	2.32	0.98	10.2	9 (1/2)
WAUSAU by Neenah	2.56	1.16	4.17	8 (2/1)
Boise® ASPEN Premium	2.76	0.66	3.45	4 (1/1)
Hammermill® LaserPrint™	6.45	0.90	7.15	8 (2/2)
<b>– 370 nm –</b>				
<i><b>100% Post-Consumer</b></i>				
Cascade Rolland Enviro100	–5.97	2.46	11.25	31 (7/3)
<i><b>Premium</b></i>				
Hammermill Great White 30	–1.38	2.15	8.70	5 (1/1)
Xerox Premium Laser	7.36	1.81	23.8	9 (2/2)
WAUSAU by Neenah	4.36	3.25	16.4	8 (2/1)
Boise ASPEN Premium	7.74	1.96	7.12	4 (1/1)
Hammermill LaserPrint	42.9	3.19	12.3	8 (2/1)

a. 100 % Post-Consumer Paper

- Cascade Rolland Enviro100, 8.5 x 11, ream (500 sheets), 20#, 75 b/m<sup>2</sup>, blue white, 90 brightness, smooth finish, grain long, Rolland product number # 5101, Cascades, Kingsey Falls, Québec.

Premium Paper

- Hammermill Great White 30, 8.5 x 11, ream, 20#, 75 g/m<sup>2</sup>, soft white, 92 brightness, bond finish, 30% post-consumer content, 3 hole punch, product number HAM 86702, International Paper Company, Memphis, TN.
- Xerox Premium Laser, 8.5 x 11, ream, 24#, 98 g/m<sup>2</sup>, bright white, 98 brightness, smooth finish, 0% post-consumer content, product number XER3R11030, Xerox Corporation, Holland.
- WAUSAU by Neenah, Premium Cardstock, 8.5 x 11, 250 sheets per ream, 110#, 199 g/m<sup>2</sup>, white, 94 brightness, smooth finish, 30% post-consumer content, product number 49508, WAUSAU Paper Company acquired by EssityTork® Philadelphia, PA.
- Boise ASPEN Premium Recycled Laser, 8.5 x 11, ream, 24#, 90 g/m<sup>2</sup>, bright white, 96 brightness, smooth finish, 30% post-consumer content, product number BPL-2411-RC, Boise Paper, Lake Forest, Ill.
- Hammermill LaserPrint, 8.5" x 11", ream, 24#, 90 g/m<sup>2</sup>, radiant white, 96 brightness, super smooth finish, 0% post-consumer content, grain long, product number 00461-2, Hammermill Papers, Erie, PA.

b. Positions across a sheet were averaged and the reported mean is the average of the sheet averages across pages in multiple reams and boxes of the specific paper type as indicated.

c. Precision is the standard deviation of the mean as described in footnote b.

d. n = number of sheets, each with 9 positions for a total of 9 x n samples. The number of boxes and reams is indicated.

**Table S2.** Variability of optical attenuation (ATN) measured at 880 nm and 370 nm through printed paper when laser jet toner, associated with different laser jet printers, at one print level was deposited onto the paper samples. Results are for multiple (n) sheets of printed paper and may be from sheets in multiple reams and boxes as indicated. .

Printer Type <sup>a</sup>	Paper Type; Print Level <sup>b</sup>	Mean ATN <sup>c</sup>	SD <sup>d</sup> of Mean ATN	ATN Range	n <sup>e</sup> (Reams/Boxes)	Field Blank Mean $\pm$ SD
<b>– 880 nm –</b>						
Lexmark V2A	Cascade Rolland Enviro100, G160	104.7	3.58	9.81	6 (3/3)	4.24 $\pm$ 2.13
	Hammermill LaserPrint, G170	90.9	3.09	10.7	10 (2/1)	11.5 $\pm$ 2.66
Ricoh <sup>®</sup>	Hammermill LaserPrint; G170	36.4	1.67	3.69	6 (2/1)	11.7 $\pm$ 1.12
HP <sup>f</sup>	Hammermill LaserPrint; G170	49.1	3.77	7.60	6 (2/1)	6.7 $\pm$ 2.13
<b>– 370 nm –</b>						
Lexmark V2A	Cascade Rolland Enviro100, G160	115.6	5.98	15.3	6 (2/3)	5.03 $\pm$ 3.30
	Hammermill LaserPrint, G170	107.9	5.31	17.2	10 (2/1)	50.9 $\pm$ 7.49
Ricoh	Hammermill LaserPrint; G170	35.7	3.19	9.58	6 (2/1)	50.6 $\pm$ 3.28
HP	Hammermill LaserPrint; G170	59.4	5.45	16.4	6 (2/1)	42.9 $\pm$ 4.72

a. Lexmark V2A Color Printer; stand alone; Lexmark International, Inc., Australia.

Ricoh; B&W Laser Multifunction Printer; Stand alone; Ricoh USA, Inc., Malvern, Pennsylvania, USA.

HP Color LaserJet<sup>®</sup>; Desktop; Hewlett-Packard Company, Boise, Idaho.

b. Based on software settings in Microsoft Word.

c. Positions across a sheet were averaged and the reported mean is the average of the sheet averages.

d. Precision is the propagated standard deviation of the standard deviation of the individual sheet means as described in footnote c.

e. n = number of sheets, each with 9 positions for a total of 9 x n printed samples.

f. HP<sup>®</sup> - Hewlett Packard<sup>®</sup>

**Table S3.** Variability of optical attenuation (ATN) measured at 880 nm and 370 nm through printed Aethalometer filter tape, when laser jet toner at one print level was deposited onto the filter tape samples using different laser jet printers. .

Printer Type <sup>a</sup>	Print Level	Mean <sup>b</sup>	SD <sup>c</sup>	Range	n <sup>d</sup> (Rolls/Sheets)	Field Blank Mean $\pm$ SD
<b>– 880 nm –</b>						
Lexmark V2A	G170	81.8	3.51	22.7	334 (5/38) <sup>e</sup>	-4.38 $\pm$ 0.65
Lexmark V2B	G170	43.1	2.27	4.81	63 (3/7)	-4.91 $\pm$ 1.19
Ricoh	G170	30.2	0.40	5.01	9 (1/1)	1.58 (n = 1)
HP	G170	48.8	2.66	1.94	36 (1/4)	N/A <sup>f</sup>
<b>– 370 nm –</b>						
Lexmark V2A	G170	94.0	5.93	30.3	334 (5/38) <sup>e</sup>	2.35 $\pm$ 1.24
Lexmark V2B	G170	47.0	3.45	4.81	63 (3/7)	10.3 $\pm$ 2.44
Ricoh	G170	31.3	0.33	7.14	9 (1/1)	0.21 (n = 1)
HP	G170	53.2	3.34	0.31	36 (1/4)	N/A <sup>f</sup>

a. See Table S2 footnotes, Lexmark V2A is a standalone printer and Lexmark V2B is a desktop model. They differ only in the base that supports the printer.

b. Positions across a sheet were averaged and the reported mean is the average of the sheet averages.

c. Precision is the propagated standard deviation of the standard deviation of the individual sheet means as described in footnote b.

d. n = number of samples. The number of rolls is indicated. Seven to 9 samples were taped to a sheet for printing.

e. Lexmark V2A: Samples were fairly evenly split across 4 rolls with 24 samples from a 5<sup>th</sup> roll. Lexmark V2B: Most samples were from 1 roll and two sets (9 samples) were from one different roll each.

f. Field blanks were not obtained when using this printer.

**Table S4.** Inkjet printer specifications.

Printer #	Printer Type <sup>a</sup>	Ink or Pigment	Print Method (Drop-on-demand)	Minimum Drop Volume	Carbon Black <sup>b</sup>	Water <sup>b</sup>
I	HP Office Jet All in One	Pigment	Thermal	12.4 picoliters	< 5%	70-80%
II	Epson® Photo Printer -1	Pigment	MicroPiezo®	1.5 picoliters	< 10%	>63%
III	Brother	Ink	Piezo	1.5 picoliters	1-10%	LC103
IV	Brother	Ink	Piezo	1.5 picoliters	0%	40-50%
V	Epson Photo Printer - 2 <sup>c</sup>	Pigment	MicroPiezo	1.5 picoliters	1–5% or 0% <sup>d</sup>	<80% for both
VI	Epson	Pigment	MicroPiezo	3.3 picoliters	5–10 %	65–70%

a. All inkjet printers, except the Epson Photo Printer – 2, are color printers with 1 black cartridge and 3 color cartridges, yellow, cyan, and magenta.

b. Percentages are from material data safety sheets.

c. This photo printer includes 8 individual cartridges (photo black – 1-5% carbon black, matte black – 0% carbon black, cyan, magenta, yellow, red, orange, gloss optimizer) and only prints in gray scale.

d. Also, includes colorant at <1% per the material safety data sheet.

**Table S5.** Variability of optical attenuation (ATN) measured at 880 nm and 370 nm through printed Aethalometer filter tape using inkjet printers to deposit ink onto the filter tape samples at set print densities. .

Printer	Print Level	Mean <sup>a</sup>	SD <sup>b</sup>	Range	n <sup>c</sup> (Rolls/Sheets)	Field Blank Mean ± SD
<b>– 880 nm –</b>						
I	G90	76.0	9.2	7.3	24 (3/4)	4.2 ± 1.2
II <sup>d</sup>	G75	2.3	0.2	---	1 <sup>f</sup> (NA)	N/A
III	G70	65.4	1.86	3.90	20 (1/2)	–2.8 ± 1.8 <sup>g</sup>
IV <sup>e</sup>						
V <sup>e</sup>						
VI	G135	36.9	1.73	1.83	80 (2/8)	–1.59 ± 1.73
<b>– 370 nm –</b>						
I	G90	88.0	14.6	22.3	24	45.1 ± 17.3
II <sup>d</sup>	G75	140.7	1.3		1 <sup>f</sup> (NA)	N/A
III	G70	72.6	2.54	3.22	20 (1/2)	16.4 ± 11.8 <sup>g</sup>
IV <sup>e</sup>						
V <sup>e</sup>						
VI	G135	40.34	2.24	2.77	80 (2/8)	15.5 ± 3.9

a. Positions across a sheet were averaged and the reported mean is the average of the sheet averages.

b. Precision is the propagated standard deviation of the standard deviation of the individual sheet means as described in footnote b.

c. n = number of samples. The number of rolls is indicated. For inkjet printer #VI, results for all print levels are given in Table 2a (880 nm) and Table 2b (370 nm).

d. Did not contain carbon black at sufficient levels, although colored pigments provided an excellent range of ATN at 370 nm.

e. Only gray scale was available with this printer, indicating that it used a mix of cyan, yellow, magenta, and black ink to provide gray to black print. It was not used since we only wanted black ink deposited onto the samples.

f. See discussion in text.

g. Additional blanks were included with these samples and are included in the average and standard deviation (n = 10 rather than 2).

**Table S6.** Results from round robin quality assurance at 7 print levels.

	<b>Printed filters: Reference laboratory<sup>a</sup></b>	<b>Printed filters: Initial reference and participant group<sup>b</sup></b>	<b>Printed filters: Participant group and final by reference laboratory<sup>c</sup></b>	<b>Field Blanks: Reference laboratory and participant group<sup>d</sup></b>
		– <b>880 nm</b> –		
Mean <sup>e</sup> (ATN)	1.39	1.72	1.32	0.68
SD <sup>e</sup> (ATN)	1.18	1.51	1.60	0.46
		– <b>370 nm</b> –		
Mean <sup>e</sup> (ATN)	1.42	1.69	1.24	1.40
SD <sup>e</sup> (ATN)	1.38	1.60	1.73	1.18

- <sup>a.</sup> Mean absolute bias between ATN measured by the reference laboratory before and after measurement by the participant groups, on a per filter basis across all print levels.
- <sup>b.</sup> Mean absolute bias between ATN measured initially by the reference laboratory and ATN measured by the participant groups on a per filter basis across all print levels.
- <sup>c.</sup> Mean absolute bias between ATN measured by the participant groups and ATN measured by the reference laboratory, after printed samples were shipped back to the reference laboratory, on a per filter basis across all print levels.
- <sup>d.</sup> Mean absolute bias in field blanks between ATN measured by the reference laboratory and ATN measured by the participant groups, after printed samples were shipped back to the reference laboratory, on a per filter basis across all print levels.
- <sup>e.</sup> Field blanks were not included in the mean or standard deviation for printed filters.

**Table S7.** Chemical composition of ink used in inkjet printer #VI.

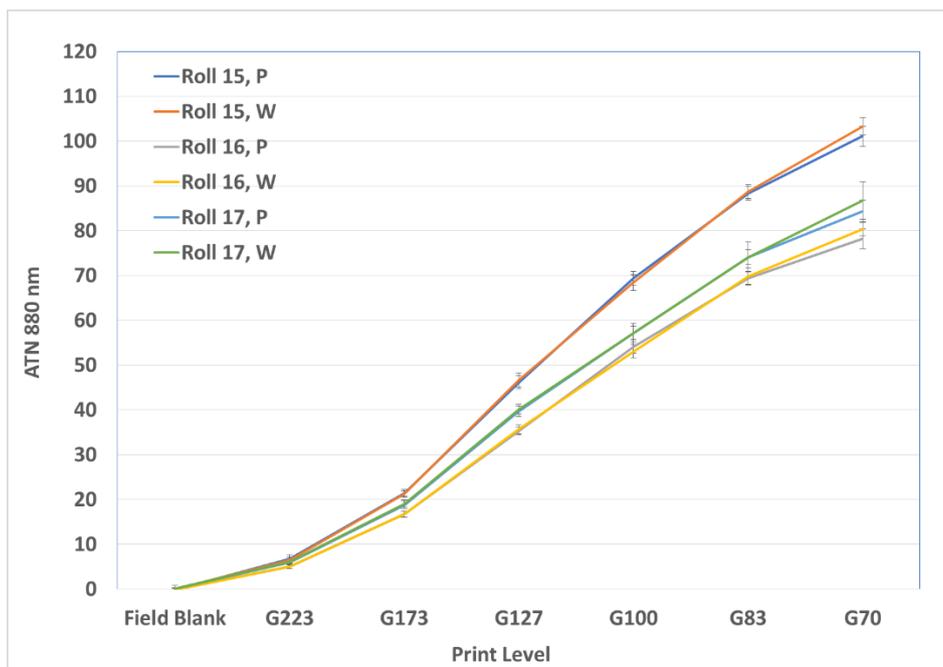
<b>-- Carbonaceous Components<sup>a</sup> --</b>			
	<b>Mass per Unit Volume (<math>\mu\text{g}/\mu\text{L}</math>)</b>	<b>Uncertainty<sup>b</sup> (<math>\pm \mu\text{g}/\mu\text{L}</math>)</b>	<b>% by Weight<sup>c</sup></b>
Carbon black reported as EC by TOA	35.75	1.89	3.38
Organic carbon as reported by TOA	162.5	8.22	15.3
Total carbon	198.25	10.11	18.7
<b>-- Trace Elements<sup>d</sup> --</b>			
	<b>Mass per Unit Volume (ng/mL)</b>	<b>Uncertainty<sup>b</sup> (<math>\pm \text{ng/mL}</math>)</b>	<b>% by weight<sup>c</sup></b>
K	1247732	111763	0.116564
Ca	51939	5090	0.004852
Na	40434	3441	0.003777
Al	6493	768	0.000607
Se	3718	5425	0.000347
Li	1592	235	0.000149
Fe	1345	107	0.000126
Mg	978	264	0.000091
Zn	348	261	0.000033
Rb	126	18	0.000012

a. OC/EC analysis – volume of ink used was 10  $\mu\text{L}$  with a 1:10 dilution. Samples were pipetted onto QFF and allowed to dry and analyzed by thermal-optical analysis using a slightly modified ACE-Asia method (Schauer et al 2003). The percent reported on the safety data sheet was between 5-10%.

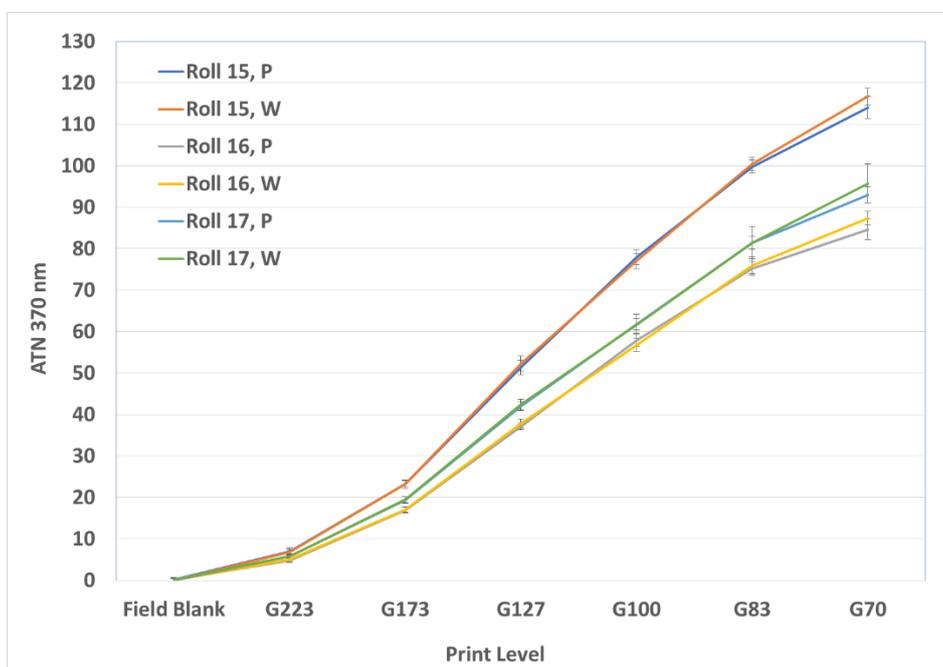
b. Uncertainty estimates were provided by the analysis laboratory.

c. Samples were weighed prior to analysis and percent of component in sample was based on this weight.

d. Elements – 51 elements were measured; the 10 with the highest mass/volume are shown here. The volume of ink used for SF-ICPMS analysis was an average of 0.2409 mL, including duplicates. Sample weight was 0.2579 gm based on gravimetric analysis. Samples were microwave digested in Teflon bombs using 5.00 mL 16M nitric acid + 5.00 mL 36M sulfuric acid + 0.2 mL 28M hydrofluoric acid and then diluted to 30.0 mL. The sample was further diluted 1:4 for SF-ICPMS analysis. Uncertainty was propagated based triplicate SF-ICPMS analyses, blank subtraction, and digest recovery based on long-term SRM recoveries. Results as given by the analysis laboratory.

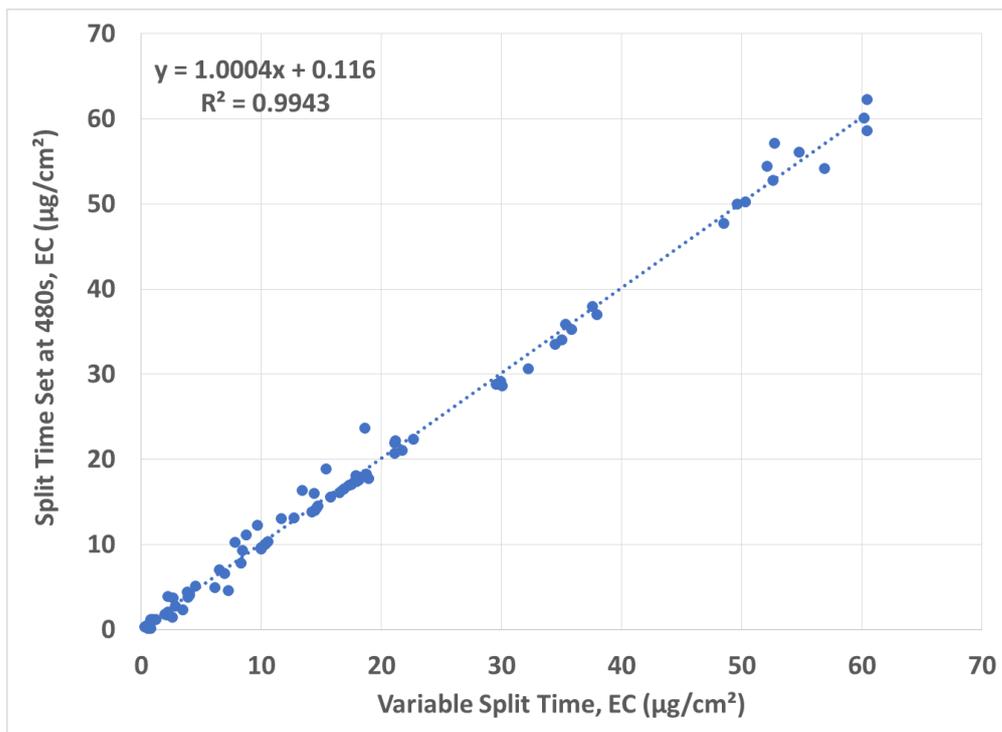


(a)

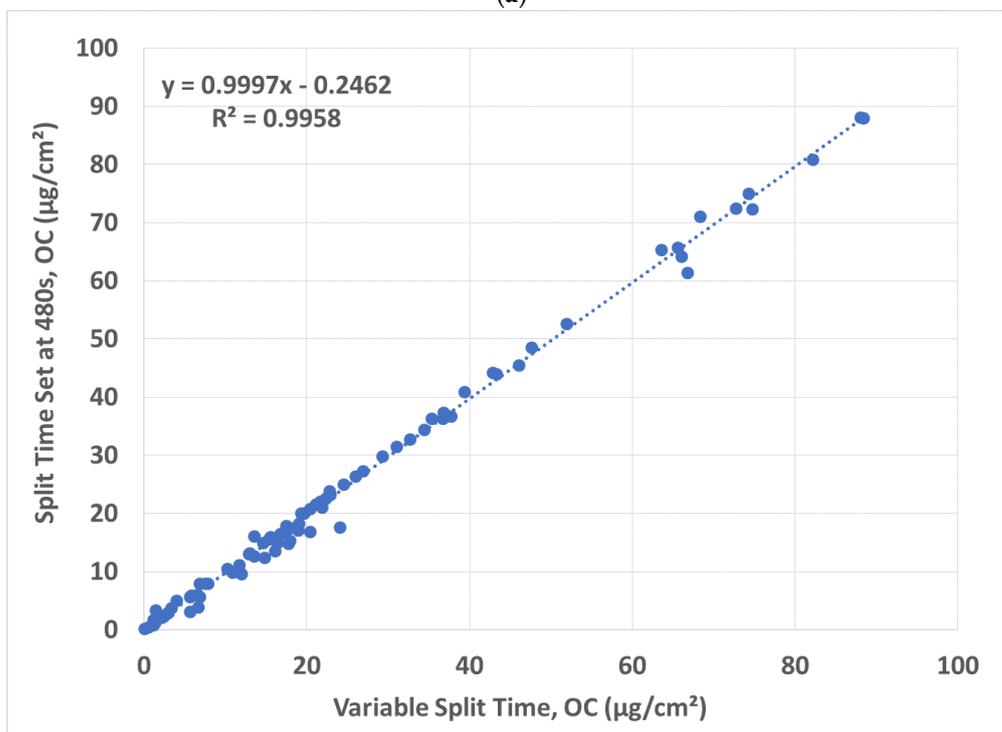


(b)

**Figure S1.** Comparison of optical attenuation measured at 6 print levels using different software programs – Adobe Photoshop (P) and Microsoft Word (W). ATN was measured on Aethalometer tape rolls 15, 16, and 17 at (a) 880 nm and (b) 370 nm. The number of sample sets analyzed in each roll was 15, 23, and 19, respectively. Samples were field blank corrected following the standard protocol applied in this method. Error bars are  $\pm 1$  standard deviation of the mean at that print level. Difference in mean optical attenuation among rolls reflects differences in the structure and morphology of the filter material among the tape rolls.

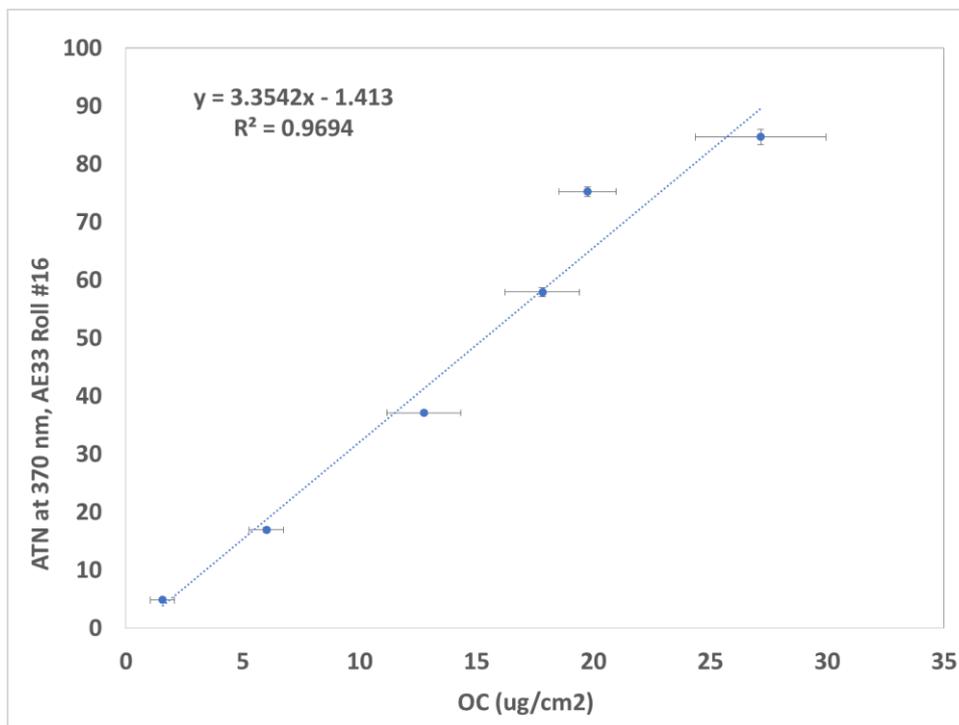


(a)

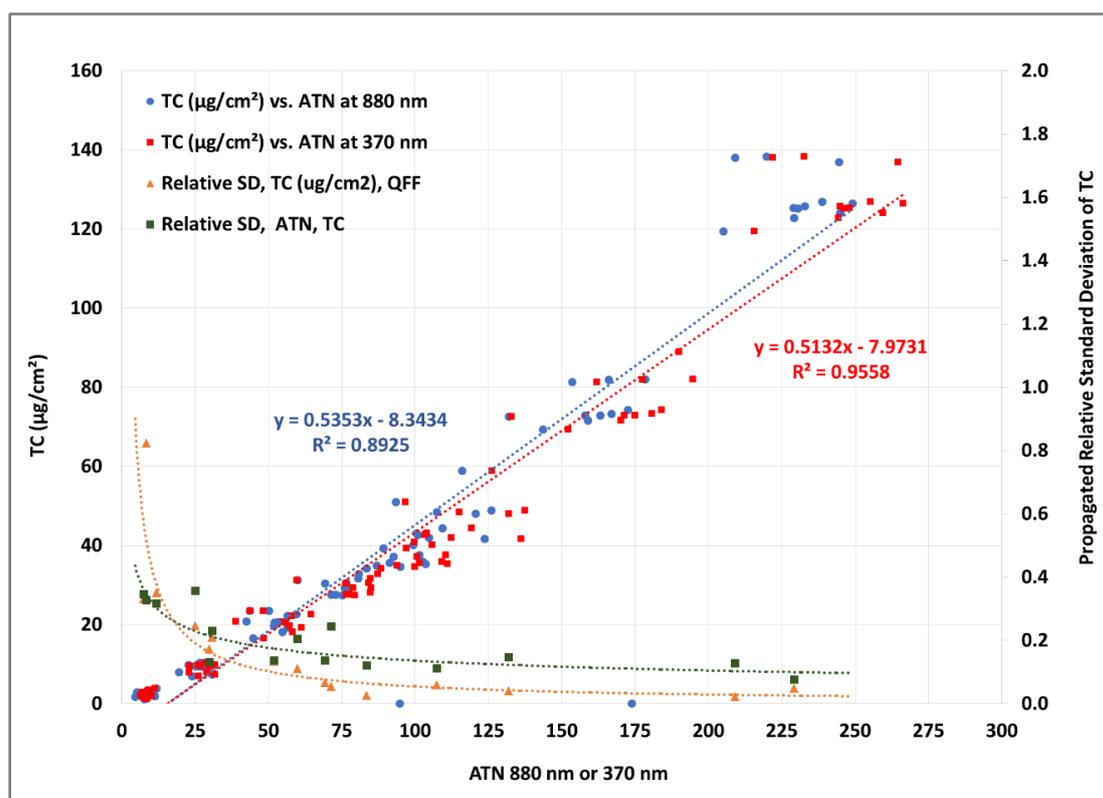


(b)

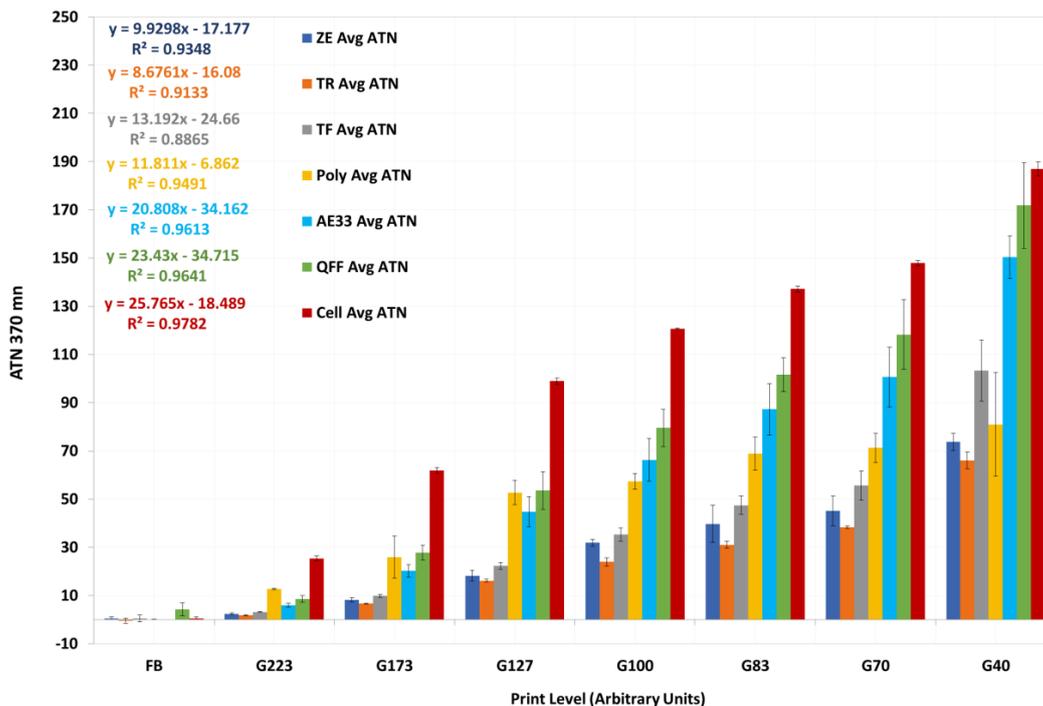
**Figure S2.** Comparison of the thermal-optical analysis results using the software-chosen split time (variable) versus using a fixed split time of 480 s for a) EC and b) OC analysis. .



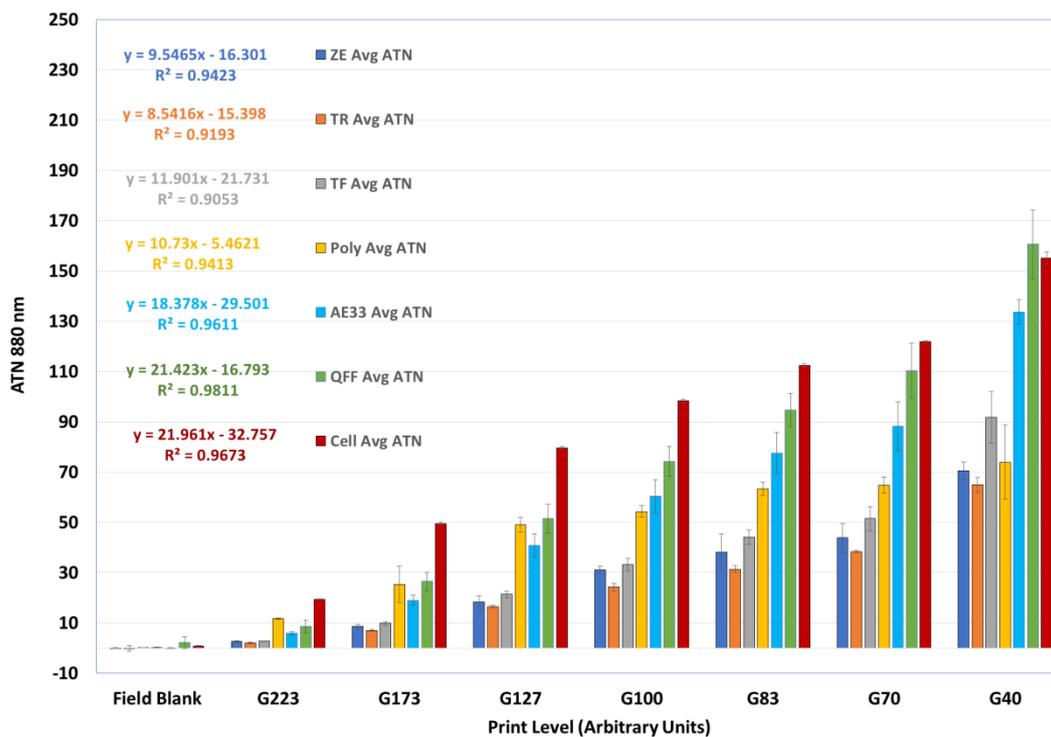
**Figure S3.** Comparison of optical attenuation measured at 370 nm on Aethalometer tape roll #16 and OC mass loading ( $\mu\text{g}/\text{cm}^2$ ) measured on QFF ( $n = 11$  sets). Error bars are  $\pm 1$  standard deviation of either the ATN across all sample sets for roll 16 at each print level (G123 – G70) (y-axis) or OC mass loading (x-axis) measured at each print level. Results for EC are presented in Figure 4.



**Figure S4.** Comparison of TC to ATN measured on QFF at 880 nm and 370 nm as a validation/reference material for comparison of filter-based methods that measure TC. Ink was printed onto QFF filters using inkjet printer #VI and Photoshop. Total carbon (TC) was the sum of the individual sample EC and OC values measured on the same QFF as ATN. EC and OC were obtained by the ACE-Asia protocol (Schauer et al. 2003). Uncertainty in ATN measurements for TC is propagated from OC and EC uncertainties. Error in ATN measurements are less than (lower ATN values) or about equal to the size of the data point (higher ATN values). Results for EC and OC are presented in Figure 5.



(a)



(b)

**Figure S5.** Relationship between ATN at (a) 880 nm and (b) 370 nm on various substrates and print levels (arbitrary units based on the printer-software combination) using inkjet printer #VI and Adobe Photoshop. Error bars are  $\pm 1$  standard deviation of the mean. Expanded results, including the number of samples measured for each substrate are presented in Table 1 at print level G70. ZE – Zefluor; TR - PTFE with support ring; TF – Teflon (TF-1000); poly – polypropylene; AE33 – Teflon-coated glass fiber; QFF – quartz-fiber; Cell – cellulose.