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Supplement of

Development of a new correction algorithm applicable to any filter-based absorption photometer

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SUPPLEMENTARY TEXT

1. Developing the model described in Section 2.4.4

Statistical regression analyses were performed to predict $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ (dependent variable) in Eq. (9). The analyses were applied on a total of 2676 FIREX observations (PAX-derived $B_{\text{abs}}$, CLAP-derived $B_{\text{ATN}}$, Tr, SSA, and AAE) at three wavelengths (467 nm, 528 nm, and 652 nm). Table S1 summarizes the variables used in the analyses. The statistical software R was used for all analyses.

**Table S1** Descriptive statistics for the variables under consideration as inputs to the correction algorithm.

|       | $B_{\text{abs}}$ | $B_{\text{ATN}}$ | $B_{\text{abs}}/B_{\text{ATN}}$ | Tr  | SSA | AAE |
|-------|------------------|------------------|---------------------------------|-----|-----|-----|
|       | 467 nm | 528 nm | 652 nm | 467 nm | 528 nm | 652 nm | 467 nm | 528 nm | 652 nm | 467 nm | 528 nm | 652 nm |
| Min.  | 45.0   | 38.5   | 29.5   | 162.0   | 140.1   | 98.9   | 0.14   | 0.12   | 0.10   | 0.27   | 0.34   | 0.45   | 0.27   | 0.25   | 0.21   | 1.25   |
| 1st Qu.| 125.4  | 99.9   | 64.8   | 473.7   | 413.3   | 296.5   | 0.22   | 0.20   | 0.18   | 0.55   | 0.60   | 0.70   | 0.50   | 0.47   | 0.44   | 1.52   |
| Median | 216.5  | 169.5  | 112.1  | 843.9   | 721.8   | 528.5   | 0.25   | 0.23   | 0.21   | 0.66   | 0.71   | 0.79   | 0.72   | 0.72   | 0.70   | 1.72   |
| Mean   | 320.2  | 245.9  | 160.2  | 1276.7  | 1091.0  | 781.6   | 0.26   | 0.24   | 0.22   | 0.68   | 0.72   | 0.79   | 0.66   | 0.66   | 0.65   | 1.99   |
| 3rd Qu.| 407.5  | 316.2  | 203.7  | 1658.2  | 1418.6  | 1000.9  | 0.30   | 0.27   | 0.25   | 0.80   | 0.83   | 0.88   | 0.84   | 0.84   | 0.84   | 2.34   |
| Max.   | 2370.9 | 1689.2 | 1295.8 | 11227.0 | 9280.7  | 6391.2  | 0.45   | 0.43   | 0.40   | 1.00   | 1.00   | 1.00   | 0.95   | 0.96   | 0.97   | 4.07   |

Figure S1 shows the relationships between $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ and each independent variable. It is clear that $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ increases with decreasing Tr, SSA, and AAE. However, the relationships are nonlinear, and the data points scatter fairly widely. Moreover, $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ spans a wide range of values at a single value of Tr, which inspired us to investigate the interactions among Tr, SSA, and AAE.

**Figure S1.** Scatter plot of $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ against Tr, SSA, and AAE at 652 nm, 528 nm, and 467 nm.
• **Prediction of** \( \frac{B_{\text{abs}}}{B_{\text{ATN}}} \) **using multiple regression models**

Independent variables of \( \frac{B_{\text{abs}}}{B_{\text{ATN}}} \) were identified by “best subset regression” (using both adjusted R\(^2\) and Mallow’s Cp as the criterion) and “stepwise regression” (both forward and backward). Variables tested for significance included Tr, SSA, AAE, and three-way interactions (Tr: SSA, SSA: AAE, Tr: AAE, and Tr: SSA: AAE). Regardless of wavelength, the best-subset models selected the same form of regression, which included all independent variables and the intercept (Table S2). However, the stepwise regression models varied across wavelengths: at 652 nm, the forward stepwise model was the same as the best-subset models, but the backward stepwise model dropped two variables (Tr: AAE and Tr: SSA: AAE); at 528 nm and 467 nm, forward and backward stepwise regression produced the same model at each wavelength, but the selected variables were different at different wavelengths (see Table S2). Generally, the adjusted R\(^2\) of best-subset models was greater or equal to the adjusted R\(^2\) of stepwise models at each wavelength. As we placed a higher priority on prediction accuracy of \( \frac{B_{\text{abs}}}{B_{\text{ATN}}} \), we selected the models that result in the greatest adjusted R\(^2\) (the model including seven predictors).

Table S2 Predictors of \( \frac{B_{\text{abs}}}{B_{\text{ATN}}} \) using “best subset regression” and “stepwise regression”.

|                | 652 nm | 528 nm | 467 nm |
|----------------|--------|--------|--------|
|                | Best subset | Stepwise (forward) | Stepwise (backward) | Best subset | Stepwise | Best subset | Stepwise |
| R\(^2\)        | 0.620   | 0.620   | 0.607   | 0.544   | 0.510   | 0.531   | 0.531   |
| Intercept      | 1.08±0.15 | 1.08±0.15 | 0.84±0.03 | 1.21±0.15 | 0.84±0.03 | 1.39±0.16 | 1.39±0.16 |
|                | ***     | ***     | ***     | ***     | ***     | ***     | ***     |
| Tr             | -0.75±0.19 | -0.75±0.19 | -0.45±0.03 | -0.89±0.20 | -0.37±0.03 | -1.12±0.23 | -1.12±0.23 |
|                | ***     | ***     | ***     | ***     | ***     | ***     | ***     |
| SSA            | -0.86±0.15 | -0.86±0.15 | -0.53±0.04 | -0.89±0.15 | -0.46±0.04 | -1.07±0.17 | -1.07±0.17 |
|                | ***     | ***     | ***     | ***     | ***     | ***     | ***     |
| AAE            | -0.30±0.10 | -0.30±0.10 | -0.15±0.02 | -0.44±0.10 | -0.20±0.02 | -0.57±0.11 | -0.57±0.11 |
|                | **      | **      | ***     | ***     | ***     | ***     | ***     |
| Tr: SSA        | 0.79±0.19 | 0.79±0.19 | 0.38±0.04 | 0.77±0.21 | 0.17±0.04 | 0.99±0.25 | 0.99±0.25 |
|                | ***     | ***     | ***     | ***     | ***     | ***     | ***     |
| SSA: AAE       | 0.31±0.11 | 0.31±0.11 | 0.13±0.02 | 0.47±0.11 | 0.20±0.02 | 0.64±0.12 | 0.64±0.12 |
|                | **      | **      | ***     | ***     | ***     | ***     | ***     |
| Tr: AAE        | 0.19±0.13 | 0.19±0.13 |                | 0.34±0.14 |                | 0.51±0.16 | 0.51±0.16 |
|                |         |         |          | *        |          | **      | **      |
| Tr: SSA: AAE   | -0.24±0.13 | -0.24±0.13 |                | -0.38±0.14 |                | -0.59±0.17 | -0.59±0.17 |
|                |         |         |          | **      |          | ***     | ***     |

\*p < 0.001;  **p < 0.01;  *p < 0.05;  ·p < 0.1.

\(^{a}\) At 528 nm and 467 nm, forward and backward stepwise approaches output the same regression model.
• **Transformation of the regression models**

A nonlinear transformation of variables is commonly used if a non-linear relationship exists between the independent and dependent variables (e.g., (Benoit, 2011; Creamer et al., 1989; Lek et al., 1996)). As seen in the first column in Fig. S1, there appears to be a logarithmic relationship between B_{abs}^{BATN} and Tr, implying that logarithmic transformation of the regression model likely improve the performance of the regression model. We tried nonlinear transformation of the dependent variable. The results generally did not improve the regression results (the adjusted R^2 is smaller or equal to that of the original model); therefore, the original B_{abs}^{BATN} was retained. Then, we transformed the dependent variables. Using ln(Tr) instead of Tr in the models improved the adjusted R^2 from 0.54 to 0.57 (528 nm) and from 0.53 to 0.58 (467 nm), but no improvement at 652 nm. Moreover, ln(Tr) has a physical meaning in that ln(Tr) = -ATN, so the transformed results are easy to interpret. Consequently, we adopted ln(Tr) in the regression model. No improvement was found using the transformation of SSA and AAE; therefore, the original SSA and AAE were retained. We present the results of the regression models using logarithmic transformation of Tr in Table S3.

**Table S3** Predictors of B_{abs}^{BATN} (similar to Table S2, but using ln(Tr) instead of Tr in the model).

|         | 652 nm | 528 nm | 467 nm |
|---------|--------|--------|--------|
| R^2     | 0.62   | 0.57   | 0.58   |
| Intercept | 0.36±0.04 | 0.34±0.06 | 0.30±0.07 |
| ln(Tr)   | -0.61±0.15 | -0.73±0.14 | -0.87±0.15 |
| SSA      | -0.09±0.04 | -0.14±0.06 | -0.11±0.07 |
| AAE      | -0.12±0.03 | -0.11±0.04 | -0.06±0.05 |
| ln(Tr): SSA | 0.61±0.15 | 0.60±0.14 | 0.73±0.16 |
| SSA: AAE | 0.09±0.03 | 0.09±0.04 | 0.06±0.03 |
| ln(Tr): AAE | 0.19±0.10 | 0.33±0.10 | 0.45±0.10 |
| ln(Tr): SSA: AAE | -0.22±0.11 | -0.36±0.10 | -0.49±0.11 |

*** p < 0.001; ** p < 0.01; * p < 0.05; · p < 0.1.

• **Assessment of the fit of regression models**

After selecting the model, we performed “F-test” to determine whether the model with fewer variables predicted B_{abs}^{BATN} better than the model with all predictors. The F-tests indicated that dropping any predictor did not improve the fit of the model (F-ratios >> 1 and P-value < 0.05).
We then analyzed the residuals of the selected model to test the adequacy of prediction. We found that the residuals were well scattered in a random pattern against ln(Tr), SSA and AAE (Fig. S2), indicating that the models as presented in Eq. (9) in the main text give the best accuracy.

![Scatter plot of residuals against Tr, SSA, and AAE at 652 nm, 528 nm, and 467 nm.](image)

**Figure S2.** Scatter plot of residuals against Tr, SSA, and AAE at 652 nm, 528 nm, and 467 nm.

The above analyses were then repeated for TAP-related observations. The results were consistent with what we have found on the CLAP data. We present the final regression models to predict $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ for these TAP-related observations in Table S4.

**Table S4 Predictors of $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ for TAP-related observations**

|        | 652 nm | 528 nm | 467 nm |
|--------|--------|--------|--------|
| $R^2$  | 0.36   | 0.32   | 0.35   |
| Intercept | 0.03±0.11 | 0.02±0.12 | 0.16±0.11 |
| ln(Tr) | -1.20±0.39 | -0.97±0.33 | -0.53±0.28 |
|        | **     | **     | *      |
| SSA    | 0.48±0.11 | 0.40±0.12 | 0.25±0.12 |
|        | ***    | ***    | *      |
| AAE    | 0.20±0.07 | 0.18±0.08 | 0.10±0.08 |
|        | **     | *      |        |
| ln(Tr): SSA | 1.43±0.41 | 0.99±0.34 | 0.49±0.29 |
|        | ***    | **     |        |
| SSA: AAE | -0.28±0.08 | -0.23±0.08 | -0.15±0.08 |
**Interpretation of the regression models**

We can order the terms in Eq. (9) into two groups, the first group (terms that do not contain ln(Tr)):

\[
(G_0 + G_2 \times \text{SSA}(\lambda) + G_3 \times \text{AAE} + G_5 \times \text{SSA}(\lambda) \times \text{AAE})
\]

defines the intercept on a graph of $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ against ln(Tr); the second group (all terms that contain the ln(Tr)):

\[
\ln(\text{Tr}(\lambda)) \times (G_1 + G_4 \times \text{SSA}(\lambda) + G_6 \times \text{AAE} + G_7 \times \text{SSA}(\lambda) \times \text{AAE})
\]

defines the simple slope of the line (Dawson and Richter, 2006; Zedeck, 1971). As in this form, the new correction equation can be interpreted as following:

1. For a given wavelength, the relation between ln(Tr) and $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ varies across levels of SSA and AAE, and the combination of SSA and AAE.
2. Under different conditions of SSA and AAE, the same value of ln(Tr) may lead to various ratios between $B_{\text{abs}}$ and $B_{\text{ATN}}$, and the compensation and/or reduction of $B_{\text{ATN}}$ will be different to agree with the reference $B_{\text{abs}}$.

We also conduct simple slope analyses to explore the nature of the three-way interaction terms (Aiken et al., 1991). Specifically, we arbitrary assume four combinations of AAE and SSA: (1). SSA=0.95 and AAE=4; (2). SSA=0.8 and AAE=3; (3). SSA=0.8 and AAE=1.5; (4). SSA=0.4 and AAE=1. As seen in Fig. S3 (528 nm as an example), the $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$-ln(Tr) relationship is moderated by different combinations of AAE and SSA. For example, the slope of “Comb. 4” in Fig. S3(b) is significantly different (p<0.05) from the slopes of other three combinations of SSA and AAE. Moreover, the intercept of the four lines are inconsistent, indicating that even when the filter is slightly loaded (ln(Tr)$\rightarrow$0), the correction of $B_{\text{ATN}}$ should be different for the aerosols with various optical properties.

![Simple slopes analysis of cross-level interaction of SSA and AAE in predicting $\frac{B_{\text{abs}}}{B_{\text{ATN}}}$ as a function of ln(Tr) at 528 nm.](image_url)
Fig. S4 compares the simulated “g” term from our correction and the previous corrections. In panels S4a and S4b, we use the original coefficients reported in B1999 and V2005 to simulate the “g” term in Eq. 8. In panels S4c and S4d, we use the updated B1999 and V2005 coefficients from our Table S7 (FIREX-CLAP). When simulating Eq. 9 (panel S4e), we estimate AAE as a function of SSA (AAE = a + b×SSA), similar to the procedure of “Algorithm C” in our manuscript. Then, we plot the results of “g” derived by all corrections as a function of SSA (panels S4f – S4h: Tr = 0.9, 0.75, and 0.5).

In general, the values of “g” term from all corrections increase with decreasing Tr and SSA. However, the figure suggests that there are variations among the corrections for different combinations of Tr and SSA. For example, the original B1999 and V2005 corrections tend to yield greater values of “g” than the other corrections (eventually, insufficient correction), and the agreement between them gets worse as Tr and SSA decrease (panels S4f - S4h). Another observation from the figure is that our correction is in better agreement with the updated B1999 and V2005, but this agreement depends on both SSA and Tr. For example, when SSA > 0.95, our correction does not exhibit as strong of a non-linearity as the updated B1999 and V2005.

Figure S4. Simulated “g” term (528 nm) in Eq. (8) or Eq. (9). In panel c) and d), the grey regions correspond to “g” values less than 0.15.

2. Comparison of the corrections against different parameters

To further investigate how different algorithms apply to different aerosol properties, we generated Figures S5 and S6 (SGP and FIREX, respectively), in which the variable on y axis is \( \frac{B_{\text{abs}} \text{ ratio}}{B_{\text{abs}} \text{ from photoacoustic instruments (reference)}} \), and the parameters on x axis include relative humidity (RH), AAE, SAE, and SSA (528 nm). In general, an apparent association between the \( B_{\text{abs}} \) ratio and these parameters exists in the uncorrected data (raw \( B_{\text{ATN}} \)), and this association persists when using B1999 and V2005, especially for RH and SSA. However, these associations are reduced or eliminated when applying our algorithm on the
filter-based absorption measurements. Although RH and SAE are not included in our algorithm, our algorithm appears to account for any influence that these parameters have on the measurements.

**Figure S5.** $B_{\text{abs}}$ ratio vs. different aerosol properties from the FIREX campaign. In the first column, $B_{\text{abs}}$ ratio = uncorrected $B_{\text{ATN}}$ / $B_{\text{abs}}$ (photoacoustic). In the other columns, $B_{\text{abs}}$ ratio = corrected $B_{\text{abs}}$ from different corrections / $B_{\text{abs}}$ (photoacoustic).
Figure S6. B_{abs} ratio vs. different parameters at the SGP site. In the first column, B_{abs} ratio = uncorrected B_{ATN} / B_{abs} (photoacoustic). In the other columns, B_{abs} ratio = corrected B_{abs} by different corrections / B_{abs} (photoacoustic).

We also investigated how number-based geometric mean diameter (d_{pg}) of aerosols affects the corrections’ performance. Arguably, the pattern for the B1999 and V2005 data agrees with those reported in Moteki et al. (2010) and Nakayama et al. (2010), in that absorption tends to be over-estimated for smaller particles and that this effect is gradually reduced with increasing particle size. Unfortunately, the size distribution of SGP aerosols is unavailable during our target time period, so we cannot extend this analysis to those data.

Compared to B1999 and V2005, the B_{abs} ratio derived by our new correction is much close to unity when plotting against d_{pg}. Although d_{pg} was not considered when developing the algorithm, any effects related to particle size appear to be captured by our algorithm, potentially in one of the
interaction terms (like RH and SAE were above). Unfortunately, we do not have a strong qualitative physical explanation for this.

Figure S7. $B_{\text{abs}}$ ratio vs. $d_g$ from the FIREX campaign.

3. Application of the CTS correction algorithm on SGP aerosols

We applied the constrained two-stream (CTS) correction proposed in Müller et al. (2014) to our SGP-CLAP data. The parameters used by us (e.g., $\delta_{af}$, $\delta_{sf}$, $\mu_1$, $\chi$) are the same as those in Müller et al. (2014) and Davies et al. (2019). We first regenerate Fig. 6 and Fig. 7 in Müller et al. (2014) to validate our coding (see Fig. S8 below).

Figure S8. Simulated relative optical depth (panel a) and $F_f$ (panel b) as a function of scattering and absorption optical depths.

As seen in Figure S9, $B_{\text{abs}}$ corrected by the M2014 correction agrees fairly well with those derived by the original B1999 and V2005, but overestimates the photoacoustic measurements by factors ~2.5. Another observation is that the performance of M2014 increases as the wavelength decreases that (as seen by the $R^2$), which is consistent with the results for urban emissions in Davies et al. (2019).
Figure S9. Inter-comparison between the CLAP-derived $B_{abs}$ corrected by the M2014 correction and the reference $B_{abs}$ at 652, 528, and 467 nm for the SGP data. The relationships derived by the other corrections discussed in our manuscript are given in the figure.

4. The procedure for simulating the uncertainty of the new algorithms in Section 3.5

The simulation is performed in seven steps:
1. We arbitrarily set concentration ranges of “true” $B_{abs}$ at 652 nm from 100 to 2500 Mm$^{-1}$ and AAE from 0.5 to 4.5 (number of cases = 500).
2. For each combination of “true” $B_{abs}$ at 652 nm and AAE (500 × 500 in total), we calculate the $B_{abs}$ at 528 and 467 nm. For example, $B_{abs}(467\text{ nm}) = B_{abs}(652\text{ nm}) \times (467/652)^{AAE}$.
3. With the observed power relationship between AAE and SSA (similar to Fig. 6 in the main text, but using $B_{abs}$ instead of $B_{ATN}$ as the absorption measurements), we compute SSA for each AAE value. The derived SSA is then used to calculate $B_{scat}(B_{scat} = SSA/(1-SSA) \times B_{abs})$.
4. With the observed relationship between $B_{abs}$ and $B_{ATN}$ (Fig. 3 in the main text), we calculate the filter-based $B_{ATN}$ at all three wavelengths.
5. We simulate the measurements of filter-based $B_{ATN}$, photoacoustic $B_{abs}$, and NEPH-derived $B_{scat}$ by adding the measurement uncertainty of the instruments to the parameters described in Steps 1-4. The measurement uncertainties are forms of normal distribution (Table 1 in the main text). Figure S4 shows an example of a dataset derived by Steps 1-5.
6. We implement “Algorithm B” on the derived dataset from Step 5. The corrected filter-based results are then compared to the “true” $B_{abs}$.
7. The above procedure is repeated 1000 times using a Monte Carlo simulation to evaluate bias and power of our correction algorithms.
**Figure S10.** Distribution of the simulated measurements derived by Steps 1-5 in the above procedure.
SUPPLEMENTARY TABLES

Table S5 Relationship between the filter-based $B_{\text{abs}}$ (FIREX-TAP and SGP-PSAP) corrected by B1999 and V2005 algorithms and the reference $B_{\text{abs}}$ at 652, 528, and 467 nm. This table complements Table 3 from in the main text.

|                      | 652 nm                        | 528 nm                        | 467 nm                        |
|----------------------|-------------------------------|-------------------------------|-------------------------------|
| **FIREX - TAP**      |                               |                               |                               |
| B1999                | $y = -46 + 2.17x$ (0.83)      | $y = -55 + 1.88x$ (0.85)      | $y = -61 + 1.75x$ (0.86)      |
| V2005                | $y = -50 + 2.23x$ (0.85)      | $y = -62 + 2.03x$ (0.85)      | $y = -76 + 2.07x$ (0.86)      |
| B1999 (update coeffs)| $y = -12 + 1.00x$ (0.85)      | $y = -17 + 1.00x$ (0.86)      | $y = -19 + 0.99x$ (0.87)      |
| V2005 (update coeffs)| $y = -13 + 1.02x$ (0.87)      | $y = -16 + 1.00x$ (0.87)      | $y = -16 + 0.99x$ (0.87)      |
| **SGP - PSAP**       |                               |                               |                               |
| B1999                | $y = -6.40 + 5.86x$ (0.32)    | $y = -5.24 + 4.47x$ (0.43)    | $y = -4.10 + 3.88x$ (0.51)    |
| V2005                | $y = -7.10 + 6.10x$ (0.32)    | $y = -6.72 + 5.11x$ (0.40)    | $y = -5.43 + 5.43x$ (0.49)    |
| B1999 (update coeffs)| $y = -0.52 + 1.21x$ (0.40)    | $y = -0.57 + 1.18x$ (0.50)    | $y = -0.37 + 1.09x$ (0.55)    |
| V2005 (update coeffs)| $y = -0.89 + 1.40x$ (0.46)    | $y = -0.76 + 1.24x$ (0.52)    | $y = -0.45 + 1.11x$ (0.58)    |

Table S6 Inter-comparison between different filter-based $B_{\text{abs}}$ corrected by the same algorithm. The value in the bracket represents the coefficient of determination ($R^2$) of the linear relationship.

|                      | **FIREX: CLAP vs. TAP**                  | **SGP: CLAP vs. PSAP**                  |
|----------------------|------------------------------------------|------------------------------------------|
| **B1999**            |                                           |                                           |
| 652 nm               | $y = 7.00 + 1.29x$ (0.91)                 | $y = 0.61 + 0.77x$ (0.57)                |
| 528 nm               | $y = 13.61 + 1.36x$ (0.90)                | $y = 1.09 + 0.78x$ (0.53)                |
| 467 nm               | $y = 25.40 + 1.30x$ (0.89)                | $y = 1.61 + 0.75x$ (0.55)                |
| **V2005**            |                                           |                                           |
| 652 nm               | $y = 4.42 + 1.33x$ (0.90)                 | $y = 0.87 + 0.68x$ (0.54)                |
| 528 nm               | $y = 12.07 + 1.40x$ (0.89)                | $y = 1.39 + 0.69x$ (0.53)                |
| 467 nm               | $y = 28.84 + 1.32x$ (0.88)                | $y = 1.85 + 0.68x$ (0.56)                |
| **B1999 (update coeffs)** |                                           |                                           |
| 652 nm               | $y = 4.20 + 1.01x$ (0.88)                 | $y = 0.01 + 0.96x$ (0.61)                |
| 528 nm               | $y = 8.02 + 1.01x$ (0.86)                 | $y = 0.16 + 0.91x$ (0.65)                |
| 467 nm               | $y = 13.18 + 1.01x$ (0.85)                | $y = 0.21 + 0.90x$ (0.70)                |
| **V2005 (update coeffs)** |                                           |                                           |
| 652 nm               | $y = 3.30 + 1.02x$ (0.89)                 | $y = 0.08 + 0.92x$ (0.68)                |
| 528 nm               | $y = 6.81 + 1.01x$ (0.87)                 | $y = 0.18 + 0.90x$ (0.68)                |
| 467 nm               | $y = 11.00 + 1.01x$ (0.85)                | $y = 0.24 + 0.90x$ (0.72)                |
Table S7 Updated coefficients in the B1999 and V2005 algorithms using our data.

|        |      | C2   | C3   | C1   |
|--------|------|------|------|------|
| **B1999** | **CLAP** | 652 nm | 3.42 | 2.00 | 0.016 |
|         |       | 528 nm | 3.47 | 1.77 | 0.016 |
|         |       | 467 nm | 3.49 | 1.43 | 0.016 |
|         | **TAP** | 652 nm | 1.58 | 2.13 | 0.016 |
|         |       | 528 nm | 1.19 | 2.07 | 0.016 |
|         |       | 467 nm | 1.11 | 1.93 | 0.016 |
| **SGP** | **CLAP** | 652 nm | -1.39 | 6.12 | 0.016 |
|         |       | 528 nm | 0.88 | 4.03 | 0.016 |
|         |       | 467 nm | 1.51 | 3.46 | 0.016 |
| **PSAP** | **CLAP** | 652 nm | 1.730 | 3.930 | 0.016 |
|         |       | 528 nm | 2.230 | 3.180 | 0.016 |
|         |       | 467 nm | 2.590 | 2.940 | 0.016 |

| **V2005** | **CLAP** | 652 nm | 0.19 | -0.34 | 0.90 | -0.83 | 0.022 |
|          |       | 528 nm | 0.19 | -0.23 | 0.95 | -0.50 | 0.017 |
|          |       | 467 nm | 0.20 | -0.19 | 0.95 | -0.15 | 0.015 |
|          | **TAP** | 652 nm | 0.28 | -0.44 | 0.88 | -1.07 | 0.022 |
|          |       | 528 nm | 0.31 | -0.36 | 0.94 | -0.94 | 0.017 |
|          |       | 467 nm | 0.32 | -0.35 | 0.92 | -0.79 | 0.015 |
| **SGP**  | **CLAP** | 652 nm | 0.22 | -1.22 | 1.14 | -1.22 | 0.022 |
|          |       | 528 nm | 0.20 | -0.94 | 1.07 | -1.09 | 0.017 |
|          |       | 467 nm | 0.20 | -0.78 | 1.01 | -0.99 | 0.015 |
|          | **PSAP** | 652 nm | 0.19 | -0.55 | 0.98 | -0.92 | 0.022 |
|          |       | 528 nm | 0.19 | -0.48 | 1.00 | -0.90 | 0.017 |
|          |       | 467 nm | 0.18 | -0.44 | 0.96 | -0.84 | 0.015 |

*We update the coefficients in B1999 and V2005 using the Levenberg-Marquardt algorithm (Levenberg, 1944), which is different from the original approach to fitting the coefficients in those papers. Specifically, we hold $C_1$ to be the same as the value in B1999 and V2005 and iteratively fit the other coefficients until the chi-square of the coefficients are minimized.

**The general form of the B1999 algorithm:** $B_{\text{abs}} = B_{\text{ATN}} \times \frac{1}{C_2 \times \text{Tr} + C_3} - C_1 \times B_{\text{scat}}$

**The general form of the V2005 algorithm:** $B_{\text{abs}} = B_{\text{ATN}} \times C_4 + C_5 \times (C_6 + C_7 \times \text{SSA}) \times \ln (\text{Tr}) - C_1 \times B_{\text{scat}}$
**Table S8** Updated coefficients in the B1999 algorithm using different subsets of AAE and SSA for the FIREX measurements. The aerosols with different subranges of AAE and SSA result in different values of C2 and C3 for different wavelengths, which are different from the “default” values in B1999. The blank cells represent combinations with no available data.

| C2 | SSA | C2 | SSA |
|----|-----|----|-----|
| 652 nm | 4.384 | 652 nm | 3.419 |
| 528 nm | 3.988 | 528 nm | 3.196 |
| 467 nm | 3.778 | 467 nm | 2.954 |

**Table S9** Updated coefficients in the V2005 algorithm using different subsets of AAE and SSA for the FIREX measurements. The aerosols with different subranges of AAE and SSA result in different values of C4 - C7 for different wavelengths, which are different from the “default” values in V2005. The blank cells represent combinations with no available data.

| C4 | SSA | C4 | SSA |
|----|-----|----|-----|
| 652 nm | 0.194 | 652 nm | 0.276 |
| 528 nm | 0.197 | 528 nm | 0.260 |
| 467 nm | 0.204 | 467 nm | 0.285 |

| C6 | SSA | C6 | SSA |
|----|-----|----|-----|
| 652 nm | -0.353 | 652 nm | -0.057 |
| 528 nm | -0.384 | 528 nm | -0.175 |
| 467 nm | -0.362 | 467 nm | -0.146 |

| C5 | SSA | C5 | SSA |
|----|-----|----|-----|
| 652 nm | -0.070 | 652 nm | -2.349 |
| 528 nm | 0.188 | 528 nm | -4.758 |
| 467 nm | -0.083 | 467 nm | -1.298 |

| C7 | SSA | C7 | SSA |
|----|-----|----|-----|
| 652 nm | -0.277 | 652 nm | -0.202 |
| 528 nm | -0.392 | 528 nm | -0.093 |
| 467 nm | -0.216 | 467 nm | -0.216 |
Table S10 Updated coefficients in the B1999 algorithm using different subsets of AAE and SSA for the SGP measurements. The aerosols with different subrange of AAE and SSA result in different values of C2 and C3 for different wavelengths, which are different from the “default” values in B1999. The blank cells represent combinations with no available data.
Table S11 Updated coefficients in the V2005 algorithm using different subsets of AAE and SSA for the SGP measurements. The aerosols with different subranges of AAE and SSA result in different values of C4 - C7 for different wavelengths, which are different from the “default” values in V2005. The blank cells represent combinations with no available data.

| C4 | wavelength | SSA     | SSA     |
|----|------------|---------|---------|
|    |            | <0.8    | 0.8-0.9 | 0.9-1   |
| 652 nm | 0.210 | 0.202 | 0.152 |
| 528 nm | 0.196 | 0.172 | 0.152 |
| 467 nm | 0.193 | 0.159 | 0.145 |
|       |          | 0.184 | 0.194  |
|       | 528 nm | 0.179 | 0.182  |
|       | 467 nm | 0.176 | 0.176  |
|       | 528 nm | 0.176 |        |
|       | 467 nm | 0.172 |        |
|       |        | 0.176  |        |
|       |        | 0.182  |        |

| C5 | wavelength | SSA     | SSA     |
|----|------------|---------|---------|
|    |            | <0.8    | 0.8-0.9 | 0.9-1   |
| 652 nm | -0.596 | -2.247 | -1.376  |
| 528 nm | -0.517 | -1.712 | -1.681  |
| 467 nm | -0.571 | -1.161 | -1.102  |
|       | -1.376 | -3.306 |        |
|       | -0.267 | -1.799 |        |
|       | -0.182 | -1.509 |        |
|       | -2.233 |        |        |
|       | 1.733 |        |        |
|       | -0.094 |        |        |

| C6 | wavelength | SSA     | SSA     |
|----|------------|---------|---------|
|    |            | <0.8    | 0.8-0.9 | 0.9-1   |
| 652 nm | 0.922 | 1.027 | 1.204  |
| 528 nm | 0.943 | 1.043 | 1.242  |
| 467 nm | 0.922 | 1.125 | 0.998  |
|       | 1.183 | 1.276 |        |
|       | 0.965 | 1.215 |        |
|       | 0.952 | 1.104 |        |
|       | 1.415 |        |        |
|       | 1.162 |        |        |
|       | 0.967 |        |        |

| C7 | wavelength | SSA     | SSA     |
|----|------------|---------|---------|
|    |            | <0.8    | 0.8-0.9 | 0.9-1   |
| 652 nm | -1.124 | -1.216 | -1.256  |
| 528 nm | -1.084 | -1.579 | -1.313  |
| 467 nm | -1.111 | -1.194 | -1.036  |
|       | -1.371 | -1.426 |        |
|       | -0.862 | -1.336 |        |
|       | -0.718 | -1.187 |        |
|       | -1.500 |        |        |
|       | -1.335 |        |        |
|       | 0.065 |        |        |
**Table S12** Computation of the quartile deviation for the derived coefficient values in Algorithm A using half of the CLAP observation. The box-and-whisker plots of the derived coefficient values are presented in Fig. S7.

|       | FIREX 652 nm | FIREX 528 nm | FIREX 467 nm | SGP 652 nm | SGP 528 nm | SGP 467 nm |
|-------|--------------|--------------|--------------|------------|------------|------------|
| G0    | 0.012        | 0.012        | 0.015        | 0.043      | 0.037      | 0.043      |
| G1    | 0.057        | 0.047        | 0.049        | 0.428      | 0.333      | 0.309      |
| G2    | 0.023        | 0.022        | 0.025        | 0.059      | 0.050      | 0.056      |
| G3    | 0.008        | 0.009        | 0.010        | 0.035      | 0.029      | 0.032      |
| G4    | 0.094        | 0.077        | 0.074        | 0.573      | 0.409      | 0.388      |
| G5    | 0.013        | 0.013        | 0.015        | 0.050      | 0.041      | 0.042      |
| G6    | 0.044        | 0.036        | 0.035        | 0.340      | 0.244      | 0.224      |
| G7    | 0.068        | 0.036        | 0.054        | 0.460      | 0.334      | 0.285      |
SUPPLEMENTARY FIGURES

Figure S11. Inter-comparison between the filter-based $B_{\text{abs}}$ (FIREX-TAP and SGP-PSAP) corrected by different algorithms and the reference $B_{\text{abs}}$ at 652, 528, and 467 nm. The solid lines represent linear regressions, while the dashed line is a 1:1 line.
Figure S12. The AETH-derived $B_{abs}$ (corrected by Algorithm A in the present work) versus photoacoustic $B_{abs}$ for the FIREX aerosols. The solid lines represent a linear regression, while the dashed lines are 1:1 lines.
Figure S13. The distribution of derived coefficient values for Algorithm A using half of the CLAP observation. The red dots represent the coefficient values derived using all observations (as shown in Table 4).

Figure S14. Inter-comparison between the CLAP-derived $B_{abs}$ corrected by Algorithm C in the present work and reference $B_{abs}$ at 652, 528, and 467 nm for the subsamples of SGP measurements (AAE-SSA prediction error is within 30%). The solid lines represent a linear regression, while the dashed lines are 1:1 lines.
Figure S15. AAE vs. SAE for the SGP data. The three parameters are calculated using photoacoustic B$_{\text{abs}}$ and Nephelometer B$_{\text{scat}}$. The panels are overlaid with the classification scheme presented in Cappa et al. (2016) and Schmeisser et al. (2017). In panel a), the averaged values (and standard deviation) of AAE and SAE reported for the SGP site in Schmeisser et al. (2017) are illustrated by the brown marker and error bars. Our results are colored by the corresponding SSA. In panel b), the results in Schmeisser et al. (2017) are colored by the corresponding type of station location and our results are colored in grey.

Figure S16. The frequency distribution of SSA (528 nm) calculated for different instrument/correction combinations of B$_{\text{abs}}$ and B$_{\text{scat}}$. 
Figure S17. The frequency distribution of SSA (467 nm) calculated for different instrument/correction combinations of $B_{\text{abs}}$ and $B_{\text{scat}}$. 
Figure S18. The probability density of AAE and SSA computed by the new algorithms (A, B, C) for the FIREX and SGP CLAP data. The curves of Algorithm A and Algorithm B overlap in some panels.
Figure S19. The frequency distribution of AAE calculated by different wavelength combinations (derived by Algorithm A in the present work).

Figure S20. Inter-comparison of SGP-CLAP-\(B_{\text{abs}}\) derived by Algorithm A with different calculation of AAE.
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