

The authors would like to thank the reviewer1 for all their suggestions. We have addressed the comments and provide a point-by-point response to the recommendations made by the reviewer1 below. The reviewer1 comments are in black, and our responses are in red.

The manuscript "Correcting for filter-based aerosol light absorption biases at ARM's SGP site using Photoacoustic data and Machine Learning" by J. Kumar et al. shows that a random forest tree machine learning algorithm can correct particle absorption coefficients measured with filter-based instruments. The analysis was performed for a specific measurement site, but the study itself can be used as a blueprint for testing ML algorithms for other stations with various aerosol types.

The manuscript addresses a relevant topic and falls within the scope of AMT. It is well written and the conclusions are sound. The reviewer recommends the manuscript for publication after considering the following minor comments.

The authors would like to thank the reviewer for providing comprehensive and insightful suggestions. We have made appropriate changes to the manuscript as detailed in the reviewer's specific comments.

Specific comments:

Page 1, line 18: The reviewer does not fully agree with the chosen explanation or wording why filter-based instruments have problems in predicting the particle absorption coefficient. The reviewer believes that the reason for the limitations is not that fixed analytical (*) forms were chosen, but rather that there are hidden influencing parameters. There is also no algorithm that takes into account all known influencing parameters, e.g. Nakayama et al. (2010) present a correction for particle penetration depth, the restricted two-stream method (Mueller et al., 2014) takes into account particle asymmetry but not particle size. It should not be concluded that solvers with fixed analytical functions are generally unable to predict particle absorption coefficients with high accuracy.

(*The reviewer means that iterative solvers for fixed parameterised functions are also included in the class of fixed analytic functions.)

The author agrees with the reviewer. The references provided are cited and the statement is now correctly expressed in the updated version of the manuscript as follows – *“However, the inability of these algorithms to incorporate in their formulations the complex matrix of influencing parameters such as particle asymmetry parameter, particle size, and particle penetration depth, results in prediction of particle-phase absorption coefficients with relatively low accuracy.”*

Page 1, line 31: Does the RFR model use the particle size distribution as input? Cf. line 195, where it says that the total mass concentration is used as input. What does total mass concentration mean? Is the cumulative mass on a PSAP filter spot meant?

The RFR model is applied to two different cases in this paper:

1. The first application is on the SGP data where Input variables given to the RFR model are = [uncorrected BPSAP, PSAP transmission, Bscat, total mass concentration from ACSM which is

simply the sum of mass concentrations of Organics, Sulphate, Ammonium, Nitrate, and Chloride obtained from ACSM (Refer Fig. A2(a)) in now updated manuscript].

2. The second application is on data obtained from Lab-generated aerosols where the following Input variables to the RFR model are given = [Aerosol number size distribution parameters (N, u_g , σ_g) obtained from SMPS, uncorrected (TAP)-measured Babs, and nephelometer-measured scattering coefficient Bscat]. The output variable of RFR is the same in both cases, i.e., the corrected particle-phase absorption coefficient.

This is now updated and clearly described in the abstract (Page1; Lines 25-30) and Section 2.2.4 (Line 215-220) to avoid any confusion. *“For the application on the SGP dataset, the RFR model was trained with an uncorrected absorption coefficient derived from PSAP ($B_{abs_uncorrected_PSAP}$), PSAP transmission (Tr), scattering coefficient from nephelometer (B_{scat}), and total mass concentration obtained from the sum of ACSM measured concentrations of various species as input variables and particle-phase B_{abs} as the output variable.”*

Line 59: Can PASS be considered a first principle method? A few lines later the authors describe the problem with liquid or multiphase particles, which is a fundamental problem of the method?

We agree with the reviewer. This statement is now corrected to *“The PASS is a contact-free method to measure particle-phase aerosol light absorption coefficient (B_{abs})”*.

Line 98: Do the authors mean the absorption coefficients or the uncorrected absorption coefficients?

This statement is now updated to *“Figure A4 illustrates the timeseries of the aerosol absorption data as derived from PSAP ($B_{abs_uncorrected_PSAP}$) and PASS (B_{abs}) instruments.”* to avoid any further confusion.

Figure A4 (Previously Figure A3) illustrates the time series of the absorption coefficients as derived from the PSAP $\{(B_{abs_uncorrected_PSAP})\}$ and measured by the PASS. The PASS measurements are more accurate; hence, they are used as a reference to compare corrected filter-based absorption coefficients in this study. Figure A4 just presents all the data from PSAP and PASS so that the reader can be assured that the data correlates well across different instruments and is of good quality.

In Figures A3 and 1, the axis label and caption should indicate whether corrected or uncorrected absorption coefficients are shown.

Figure A4 (Previously Figure A3) represents the raw data used in this study for the SGP site from PASS {as measured} and PSAP $\{(B_{abs_uncorrected_PSAP}) = \text{uncorrected value derived after undo the automatic B1999 correction implemented by PSAP firmware}\}$.

The caption of Figure A4 is now updated to reflect this. We also made changes in the manuscript text by defining *“the uncorrected aerosol absorption data as derived from PSAP ($B_{abs_uncorrected_PSAP}$)”* in equation form to avoid any confusion. We have now updated the terminology in the manuscript. Now, B_{abs_PSAP} means the absorption coefficient output from the PSAP firmware which includes the automatic B1999 correction and $B_{abs_uncorrected_PSAP}$ refers to the back calculated uncorrected PSAP-based absorption coefficient before the B1999 correction that is auto applied by the Radiance Research’s PSAP firmware.

$$B_{abs_uncorrected_PSAP} = \frac{A_{PSAP}}{Q_{PSAP}\Delta t} \ln\left(\frac{I(t)}{I(t + \Delta t)}\right) = \frac{B_{abs_PSAP}}{f(Tr)} = \frac{B_{abs_PSAP}}{\left(\frac{1}{1.317 \times Tr + 0.866}\right)} \quad (1)$$

General comments to chapter 2:

Because of the known artefact due to light scattering particles, it would be informative if the authors presented single scatter albedos.

Thank you for this suggestion. To present the SGP data better, we now present descriptive statistics of all the instruments and the derived optical parameters like SSA, AAE and SAE obtained from the SGP data in the Appendix section.

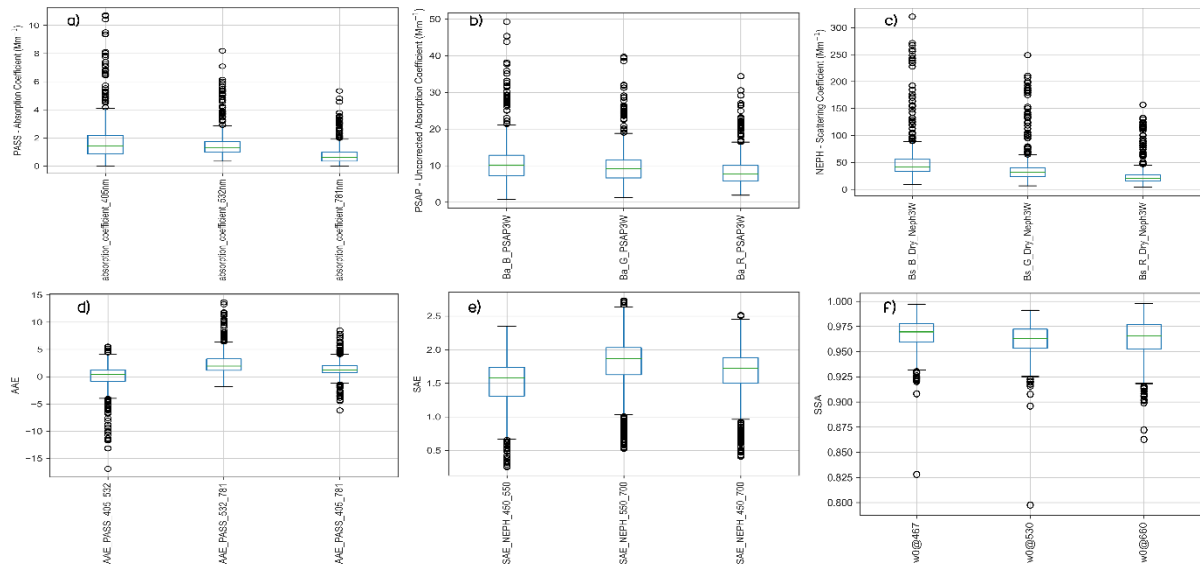


Figure A1: Summary of the SGP dataset. The boxplots of raw measurement data are shown as obtained from various instruments used in this study (a) PASS (b) PSAP{ $B_{abs_uncorrected_PSAP}$ } and (c) Nephelometer. The boxplots of parameters derived from the raw data are also shown (d) AAE (e) SAE and (f) SSA. The green line is the median of the data. The bottom line of box is 25% percentile of data and top line of box is 75% percentile of data, therefore, the box represents the middle 50% of all the datapoints which is the core of the data.

Why was the Virkkula (2010) correction revised but not the Ogren (2010)-Bond (1999) correction?

Thank you for pointing this out. We now show the performance of both Ogren-Bond and Virkkula separately before showing the “Average” performance of both algorithms. The reason for not revising Bond-Ogren is now also included in the manuscript as follows:- “The %RMSE for this (unrevised Bond-Ogren) algorithm on the SGP data is ~312% which is almost the same as the %RMSE of unrevised Virkkula (2010). Since the general equation form of Ogren (2010) modified Bond (1999) is similar to that of Virkkula (2010) and both the unrevised versions of algorithms perform with similar accuracy, hence, the improvement in accuracy of Ogren (2010) modified Bond (1999) with revised coefficients can be expected to be very similar to that in the case of Virkkula.”

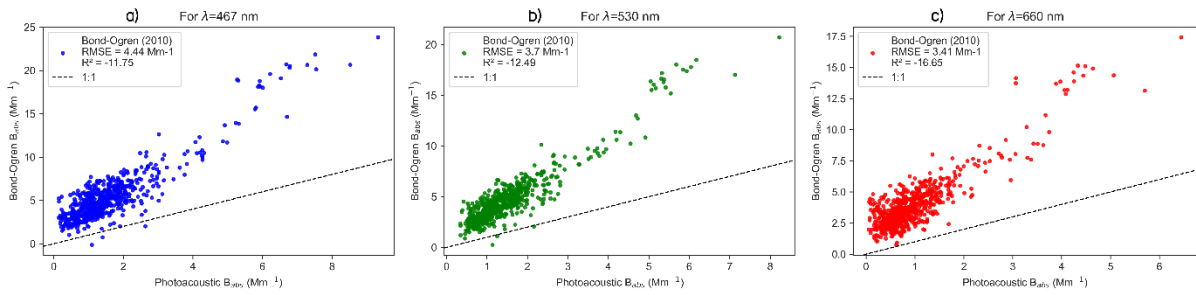


Figure 4: Comparison between PSAP absorption coefficients, corrected for using Bond-Ogren correction algorithm, and the reference PASS absorption coefficients measured at the SGP site corresponding to (a) 467nm, (b) 530nm and (c) 660nm wavelengths.

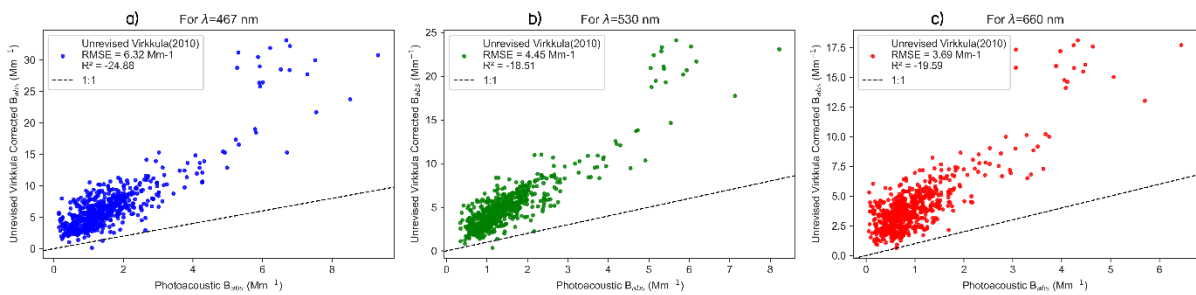


Figure 2: Comparison between PSAP absorption coefficients, corrected for using Virkkula (2010) algorithm with unrevised coefficients, and the reference PASS absorption coefficients measured at the SGP site corresponding to (a) 467nm, (b) 530nm and (c) 660nm wavelengths.

Figure 5: It seems that there are fewer data points in Figure 5 than in Figures 3 and 4. Is the split of the data into training and test data sets the only reason?

Yes, the sole reason for less number of points in Figure 6 and Figure 8 {previously Fig 5 and Fig 7} (application of RFR on SGP data and RFR on lab-generated aerosol data) is due to the fact that we are only showing RFR's performance on the test data (30% of randomly shuffled data) which the RFR did not encounter during training and was used only for prediction and calculating RMSE. We did this to show that RFR has lower RMSE scores even on unseen data as compared to the traditional algorithms which yield high RMSE values even when predicting on the same data that they used to train/revise coefficients.

Line 303: Does it have any influence that the laboratory dataset was taken with a TAP photometer and the data from the SPG site was taken with a PSAP photometer?

As mentioned earlier in detail the RFR is applied to two different cases in this paper, and both the RFR models were trained independently. The first RFR model was trained and tested with SGP data and the other RFR model was trained and tested on lab-generated aerosol data, to further establish RFR as a more accurate correction algorithm on aerosols from various sources. Since these models are trained independently, they do not influence from each other's input data.

Appendix A4: check sentence: "Tune the parameters of the ML model if the performance to achieve desired level of accuracy."

Thank you for pointing out, all grammatical mistakes found in the manuscript are now corrected.

References:

Nakayama, T., et al. (2010). "Size-dependent correction factors for absorption measurements using filter-based photometers: PSAP and COSMOS." *Journal of Aerosol Science* 41(4): 333-343.

Mueller, T., et al. (2014). "Constrained two-stream algorithm for calculating aerosol light absorption coefficient from the Particle Soot Absorption Photometer." *Atmos. Meas. Tech.* 7: 4049-4070.