Evaluating biases in filter-based aerosol absorption measurements using photoacoustic spectroscopy

Nicholas W. Davies, Cathryn Fox, Kate Szpek, Michael I. Cotterell, Jonathan W. Taylor, James D. Allan, Paul I. Williams, Jamie Trembath, Jim M. Haywood, and Justin M. Langridge

We would like to thank the reviewers for taking the time to read our manuscript and for highlighting some important issues, which will be addressed in turn below.

In the responses below, an italic font highlights text from the manuscript, and underlined font and strike-through font highlight additions and deletions to the manuscript's text in response to the reviewers' comments, respectively.

Review 1 – John Ogren

1. p.1, line 24: It's more useful here to give typical values, not extremes

We have amended original page 1, lines 23-24, which now state:

"*Filter-based absorption measurement biases led to aerosol single-scattering albedos that were biased low by values in the range ~~up to~~ 0.00–0.07 and absorption Ångström exponents (AAE) that were in error by ± (0.03–0.54).*"

2. p.2, line 27: Good to cite the earlier work, but Lin et al did not measure transmittance continuously during sampling, only before and after sampling.

We have amended original page 2, lines 25-27, which now state:

"Filter-based instruments measure the light transmittance across a filter continuously, which changes as particles are deposited onto the filter, providing a measure of aerosol absorption (see Sect. 2.1) (~~Lin et al., 1973~~ e.g. Bond et al., 1999)."

3. p.3, line 28: recommended notation is "refractory BC" (rBC, Petzold et al., 2013)

We have amended original page 3, lines 26-28, which now state:

"More recently, Subramanian et al. (2010) derived the BC mass absorption coefficient (MAC) at 660 nm for fresh and 1–2 day-old aerosol emissions in and around Mexico City by dividing the absorption coefficients measured using a PSAP by the

refractory BC mass concentrations measured using a single particle soot photometer (SP2, Droplet Measurement Technologies).”

4. p.5, line 3: Indeed, this is the first such study.

We have amended original page 5, lines 3-5, which now state:

“~~To our knowledge,~~ This is the first study to simultaneously evaluate the Bond et al. (1999), Virkkula (2010) and Müller et al. (2014) correction schemes for ambient aerosol sampling across multiple aerosol types.”

5. p.5, line 13: Recommend calling this the attenuation coefficient.

We have amended original page 5, line 13, which now states:

“The ~~absorption~~ attenuation coefficient can thus be determined using”

We have also amended original page 9, line 31, which now states:

“Uncorrected ~~absorption~~ attenuation coefficients, b_{ap}^{raw}*, were calculated at 1 Hz, and the average and standard deviation for…”*

6. p.8, line 16: This is indeed a correct way to do the calculation, and actually was written this way in the AMTD version of the Müller et al (2014) paper (https://www.atmos-meas- tech-discuss.net/6/11093/2013/amtd-6-11093-2013.pdf). The equation 5 in Müller et al (2014) is correct, as long as one realizes that the summation is over all the particles that have been deposited on the filter, but the equation used by Davies et al. represents a practical way to do the summation.

We have amended original page 8, lines 17-19, which now state:

“~~This~~ Equation 24 is ~~different~~ a practical way to apply ~~the~~ equation 5 presented ~~by~~ in Müller et al. (2014) who ~~recommended~~ instead ~~using~~ used an equivalent method, which utilised individual particle scattering cross sections (as opposed to ensemble scattering coefficients). We used Eq. 24 as opposed to the recommended formulation because nephelometer measurements represent an ensemble.”

7. p.8, line 23: Very confusing to use this nomenclature for the backscatter fraction, as just six lines above you use it to denote the scattering coefficient. Also, a newer version of equation 24, which is also more broadly applicable, is given by Moosmüller and Ogren (2017; Atmosphere 2017, 8, 133; doi:10.3390/atmos8080133)

We have replaced “b_{sp}” with “$b_{back-sp}$” in equation 25 (original equation 24) on original page 8, line 22 and on original page 8, line 23 to represent the backscattering coefficient. The scattering coefficient is still represented by “b_{sp}” throughout the manuscript.

We have also updated our analysis to use a newer version of equation 25 (original equation 24), i.e. using equation 10 presented in Moosmüller and Ogren (2017). Using the updated equation 25 (original equation 24) only changed the results relating to the Müller et al. (2014) correction by a maximum of 1 % (i.e. the slopes in Table 2). Thus using an updated parameterisation for the asymmetry parameter does not change the overall conclusions of this manuscript. We have updated the relevant numbers in Tables 2 – 4 and throughout the manuscript to reflect this alteration (these manuscript changes are listed under point 1 in “Additional corrections” below), while our conclusions remain unchanged. Line 22 now states:

“$g_p = -\cancel{7.143889}\ 6.347b_{back-sp}^3 + \cancel{7.464439}\ 6.906b_{back-sp}^2 - \cancel{3.96356}\ 3.859b_{back-sp} + \cancel{0.9893}\ 0.9852,$

where $b_{back-sp}$ is the backscattering ratio measured using a nephelometer ~~(Andrews et al., 2006; Müller et al., 2014)~~ (Moosmüller and Ogren, 2017).”

8. p.9, line 30: Was the filtered air noise test done in-flight, on the aircraft in the hangar, or in the laboratory? If either of the latter two, are these results representative of the noise level in-flight?

The filtered noise test was performed in the laboratory. Original page 9, line 30 has been amended, which now states:

“To understand the impact of this on instrument sensitivity, the TAP was run for ~3 h in the laboratory while it sampled filtered room air to characterise the noise in the system.”

It is unclear how the noise levels will vary between laboratory and in-flight filter measurements.

9. p.10, line 4: This is unsubstantiated speculation. The difference could just as likely be differences between the TAP and CLAP. Since the authors have the ability to repeat the noise test with and without the digital filter, they can readily determine the effect of the digital filter on 30-s averages. Another factor suggesting a difference between the TAP and CLAP is that the present study found a pronounced difference (nearly factor of two) in the noise level of the three wavelengths, whereas Ogren et al (2017) reported that the results for the three wavelengths were very similar.

We have amended original page 10, lines 3-5 to state:

“The difference between the detection limits measured in this study and that presented in Ogren et al. (2017) ~~is likely~~ could be due to running without low-pass digital filtering in the current study and/or due to differences between the TAP and CLAP.”

10. p.10, line 8: Is there a manufacturer/vendor to go along with this product number?

Yes – Sigma Aldrich. We have amended page original 10, line 8, which now states:

“nigrosin (Sigma Aldrich, product number 198285-100G)”.

11. p.10, line 11: How does this area compare to the spot size recommended by the manufacturer? How do the less-defined spots in ambient samples affect the uncertainty of the ambient TAP measurements?

We have added the following information to original page 10, lines 11-16:

“The manufacturer-recommended spot sizes are 30.7210 mm^2. Filter spot sizes were determined using nigrosin rather than from the ambient aerosol samples themselves as the spot edges were more clearly defined. The spot edges of the deposited ambient aerosol were difficult to detect as the filter spot was changed at the start of each day when measurements were taken. It was possible to detect the aerosol spot for measurements that corresponded to high loadings of absorbing aerosol. In these cases there was evidence of aerosols spreading across the filter and the area of the spots was larger by 5–20 %. However, this observation is based on a limited sample of three aerosol spots and the timescale for spread across the filter is unclear. This analysis used the areas determined using the clearly defined nigrosin spots, and therefore provides a lower limit of area ~~and thus~~, absorption coefficient (see Eq. 2), and as will be shown in Sect. 3, the TAP absorption bias.”

12. p.10, line 30: Only four? Is there a path length correction to deal with a purge flow to prevent contamination of the PAS cell optical windows?

The phrase “determined” in line 30 (page 10) is intended to mean that the uncertainty in PAS measurements is governed by the three factors listed, i.e. (i) uncertainty in the ozone calibration, (ii) uncertainty in corrections applied to account for the PAS microphone pressure sensitivity and (iii) uncertainty in subtraction of background noise which arose primarily from laser heating of the PAS cell optical windows. Other corrections to the PAS data are relatively small and therefore contribute negligibly to the overall uncertainty in PAS measurements.

We have amended original page 10, line 30, which now states:

“*The accuracy of PAS absorption measurements was determined primarily by three factors: …”*

We do not use a purge flow to prevent contamination of the PAS cell windows, as contamination during normal ambient sampling is not significant. Even then, we record window-generated background noise at relevant intervals to remove any such contribution.

13. p.11, line 21: Are measurements on filtered air in the lab comparable to measurements on filtered air in-flight? Are there additional contributions to instrument noise from the aircraft environment, such as engine noise, vibration, turbulence, electrical interference, etc?

The noise performance was no worse than a factor of 2 larger for airborne operation, which bounded the error introduced by the background correction to be 0.27 – 0.54 Mm^{-1}. This updated uncertainty range (0.27–0.54 Mm^{-1} compared to the originally-stated 0.2 Mm^{-1}) reflects a more robust uncertainty analysis, which was derived using a larger range of absorption coefficient data for multiple wavelength PAS channels.

We have amended original page 11, lines 28-29, which now state:

"The uncertainty ~~*depended on the strength of aerosol absorption and*~~ *was found to be* ~~*0.2, 2.0 and 20.5 % at 100, 10 and 1 Mm^{-1} respectively*~~ *0.27–0.54 Mm^{-1}, which led to larger percentage uncertainties for lower absorption coefficients. The noise performance was no worse than a factor of 2 larger for airborne operation."*

We have also amended original page 11, line 26-27, which now states:

"The uncertainty in the background noise correction was found to be normally distributed, with a 1σ width of ~~*1.27 %*~~ *1.81–2.30 % across the range of cells used."*

We have also amended original page 11, lines 33-34, which now state:

"These factors were combined in quadrature, leading to total PAS measurement uncertainties of ~~*23.1 %*~~ *29.0–55.0 % for 1 Mm^{-1} absorption coefficient measurements across the range of cells used (independent of pressure) and approximately* ~~*8.0*~~ *8.1 % for 100 Mm^{-1}."*

14. p.11, line 29: Wouldn't it be simpler, and more useful, to express this percentage uncertainty in terms of an absorption coefficient?

We have amended original page 11, line 29, which now states:

"The uncertainty ~~*depended on the strength of aerosol absorption and*~~ *was found to be* ~~*0.2, 2.0 and 20.5 % at 100, 10 and 1 Mm^{-1} respectively*~~ *0.27–0.54 Mm^{-1}, which led to larger percentage uncertainties for lower absorption coefficients."*

15. p.12, line 14: This seems like a large difference in time response. How much of the difference can be attributed to tubing lengths? Was the response difference a pure lag (i.e., corresponding to plug flow), or was there also a difference in rise/fall times that could be indicative of differences in response times due to mixing?

The originally stated 20 seconds lag time between the TAP and PAS cells was dominated by inaccurate synchronisation between the two computers used to run the TAP and PAS instruments, respectively. The rise and fall times of the TAP and PAS were comparable. We have removed reference to the lag time between the two instruments.

We have deleted the following line from original page 12, line 14:

"The delay time between the TAP relative to the PAS was 20 s."

16. p.13, line 16: Not sure why you need a citation here, you could just look out the window and confirm that you were dealing with a near-source smoke plume.

We have removed the citation on original page 13, line 16, which now states:

"Mean in-flight CO concentrations were 175 ppbv although concentrations greater than 14000 ppbv were measured when flying through plumes close to the aerosol source, indicative of fresh biomass burning emissions ~~(Dentener et al., 2001)~~."

17. p.14, line 4: Interpreting a regression slope as a bias requires that the regression intercepts are very close to zero. Please justify this implicit assumption.

We have added the following text to original page 13, line 1:

"All linear regressions between TAP and PAS measurements were forced through the origin."

This information is also stated in the captions of Table 2 and Figure 3.

18. p.14, lines 20-23: These comparisons provide very helpful guidance for users of M2014 in deciding which parameterisation to use for black particles. Please include the corresponding comparison of biases if you use the B1999 parameterisation for black particles in the M2014 scheme. Also, on p.6, you reported that the difference between the two flavors of V2010 (wavelength-dependent vs. independent) was minimal, but here it appears that there is a substantial difference between parameterisations for black particles. Please elaborate.

We have updated Tables 2 to 4 and Figures 3 to 5 and 7 to 8 to reflect use of the CTS-B1999 parameterisation.

We have also included the M2014 (B1999 parameterisation) into the manuscript, replacing equation 14. The following text has been added to original page 7, line 19:

"Using the B1999 empirical correction,

$$F_{a,B1999}^{exp} = \frac{1}{\delta_{ap}} ln\left(\frac{e^{c_2\delta_{ap}+ln(c_1+c_2)}-c_1}{c_2}\right), \qquad (13)$$

where $c_1 = 1.555$ *and* $c_2 = 1.023$*, which were derived in Bond et al. (1999); see the alternative formulation of the B1999 correction in Müller et al. (2014).”*

The other equation numbers have been updated accordingly.

We have also amended the text of original page 14, lines 17–23, which now state:

“For the M2014 (B1999 parameterisation) correction scheme, the range of TAP biases across all aerosol sources was 1.04–1.26 and for the M2014 (V2010 parameterisation), the range of TAP biases was 1.01–1.18. The M2014 (V2010 parameterisation) scheme reduced TAP biases relative to the B1999 and V2010 schemes by 7–38 % and 7–25 %, respectively, dependent on the aerosol source and wavelength. The most significant reductions in TAP biases were for urban aerosol emissions and had the most impact on measurements at 652 nm. As discussed in Sect. 2.1.3, the M2014 (V2010 parameterisation) correction scheme applied here used the wavelength-dependent Virkkula (2010) parameterisation, in contrast to Müller et al. (2014), who applied the Virkkula et al. (2005) parameterisation.”

We have also amended the text on original page 14, line 29, which now states:

“The M2014 (V2010 parameterisation) scheme reduced the biases with 10 % of measurements biased greater than 1.27–1.41, 1.20–1.30 and 1.18–1.29 for urban, fresh BBA and aged BBA, respectively, dependent on wavelength.”

We have also amended the text on original page 14, lines 33-34 and page 15, lines 1-2, which now state:

“The TAP biases exhibited a strong wavelength dependence. In general, the lowest biases were seen at 652 nm and the largest biases at 467 nm when the V2010 and M2014 schemes were applied to TAP measurements for all aerosol sources. The exceptions to this trend were when the M2014 scheme (V2010 parameterisation) was applied to urban aerosol measurements, which led to the largest biases at wavelength 528 nm. The M2014 scheme (B1999 parameterisation) also led to the largest biases at 528 nm for all aerosol types.”

We have amended original page 16, line 2, which now states:

“Correcting the TAP data using the M2014 (V2010 parameterisation) correction scheme reduces the positive correlation between TAP biases and both R_{OA} *and* $R_{OA/LAC}$ *as shown in Fig. 6 (g–i).”*

We have amended original page 17, lines 8-10, which now state:

"The M2014 (B1999 parameterisation) led to mean AAE values that were in closest agreement with AAE values derived using PAS measurements for all aerosol types. The V2010 scheme led to mean AAE values that were in second-closest agreement with the AAE values derived using PAS measurements for urban aerosols, whereas the M2014 (V2010 parameterisation) scheme provided the second-closest match for fresh BBA and the B1999 scheme for aged BBA."

We have also amended original page 18, lines 4-7, which now state:

"Best agreement with AAE values derived using PAS measurements was obtained when TAP measurements were corrected using the M2014 (B1999 parameterisation) correction scheme and when ~~of~~ (i) urban aerosol measurements were corrected using the V2010 scheme, (ii) fresh BBA measurements were corrected using the M2014 scheme and (iii) aged BBA measurements were corrected using the B1999 scheme."

We agree with the reviewer's comment relating to the difference between the wavelength-dependent and wavelength-independent flavours of the CTS-V2010 parameterisation and have updated the text on page 6, lines 22-23 accordingly, which now states:

"This was found to have a ~~minor~~ moderate impact on the results of this study as discussed in Sect. 3".

We have also amended the caption of figure 6 to state:

"using the M2014 (V2010 parameterisation) correction scheme"

19. p.14, lines 27-31: How does the bias depend on filter transmittance? Is there any relationship between these 10% of points with larger biases and the filter transmittance? Ditto for the 10% of measurements with low biases.

We evaluated the TAP bias as a function of filter transmittance on a point-by-point basis for (i) all data, (ii) highest 10% of TAP biases and (iii) lowest 10% of TAP biases, and found no correlation for any dataset. Assessing the difference in the mean filter transmittance associated with the top 10% of TAP biases compared to the lowest 10% of TAP biases revealed absolute differences in filter transmittance up to 0.12, which depended on the measurement wavelength and correction scheme applied.

We have added the following text to page 14, line 32:

"An analysis of the dependence of TAP bias as a function of filter loading revealed no point-by-point dependence but potentially a weak signal in the large-scale mean such that the difference in absolute filter transmittance associated with the

highest 10 % of TAP biases compared to the lowest 10 % of biases across all channels and wavelengths was up to 0.12. The filter transmittance changed over the course of a flight by a maximum of 0.21."

20. p.16, lines 11-12: This finding suggests that a possible contribution of BrC is not the source of the discrepancy with Lack's results, but rather that the source of the discrepancy is the correction scheme.

Arguably, Figure 6 shows that the different aerosol sources led to a greater discrepancy with Lack's results than by applying a different correction scheme than applied in Lack (i.e. applying V2010 and M2014 instead of B1999). Ultimately, we will never be able to reconcile the differences between our results and those of Lack due to the different aerosol mixtures measured.

We have added the following text to page 16, line 12:

"This finding suggests that the source of discrepancy between the results presented in this study and the results of Lack et al. (2008) (i.e. Fig. 6) may be caused by the less advanced correction scheme applied to the Lack et al. (2008) data. However, given the strong dependence of R_{abs} on the aerosol type and source in Fig. 6, the bias dependence on organic fraction in the Lack et al (2008) data may well persist, independent of the correction scheme used, because of the different aerosol sources and source locations being studied."

21. p.17, line 12: Please justify the claimed importance to climate of AAE. For example, what climate model, or radiative transfer model, uses AAE in the calculation of radiative forcing? I would argue that the parameter of more importance to climate is the wavelength-dependent SSA, and the results in Table 3 show that the difference between PAS and TAP+M2014 measurements of this parameter is negligible for all wavelengths and aerosol types studied. AAE is useful for inferring aerosol type, and relative contributions of BrC or dust to absorption, and it is here that the differences among the measurement approaches become important.

We have amended the text on original page 17, line 11-13, which now states:

"However, what is clear from this analysis is that there are large uncertainties in this important ~~climate~~ parameter when calculated from filter-based absorption measurements, and that these uncertainties are strongly source and correction scheme dependent. This cautions that significant uncertainties could be introduced if using the AAE to differentiate between types of aerosol.

22. p.18, line 16: This should also apply to the code used to implement the various corrections, especially M2014. A brief mention of that code (i.e., what programming language) would be appropriate here. Given the conclusions of this paper, it seems likely that other users of the TAP would welcome publication of the source code for your implementation of the M2014 correction (perhaps as supplemental information?).

At present, we are unable to invest the time to get the code to a position suitable for sharing in the public domain but will consider this as something for the future. The code used to run the analysis presented in this manuscript was implemented in Python. We have added the following text to original page 5, line 19:

"*The code used to run the analysis presented in this manuscript, i.e. relating to the equations presented throughout this section, was implemented in Python.*"

23. p.33: Interesting to see the outliers that are far below the regression line only appear in the red channel. Why don't they show up in the green channel?

It is unclear why the outliers only appear in the red channel in corresponding to urban aerosol emission data, shown in Figure 3. We were unable to determine the cause of these outliers and attributed this to instability in the 652 nm PAS cell.

Review 2

1. Page 2 line 29: Particles are mainly collected in fibre filters. The penetration depth also depends on the particle size and influences the sensitivity of the photometer(Nakayama et al., 2010; Moteki et al. 2010). This circumstance should be taken into account in the discussion of the results, since different aerosols were present during the three measuring phases.

We have added the following text to original page 2, line 34:

"The sensitivity of filter-based absorption photometers is also affected by the penetration depth of particles within the filter matrix, which depends on particle size (Moteki et al., 2010; Nakayama et al., 2010)."

We have also added the following text to original page 15, line 3:

"As highlighted in the introduction, filter-based absorption photometers are sensitive to the particle penetration depth, which is dependent on particle size. Indeed, this sensitivity may have contributed in part to the variation in TAP biases observed for the three types of aerosol investigated during this study."

2. Page 9 line 11: The scrubber to remove gases may not be known to all readers. Can the author explain the function, also with the background that potentially present volatile organic material could be removed from the particles.

The function of the scrubber, i.e. to remove gases, is described on page 9, line 12, which states:

"The aerosol-laden stream was first dried to < 20 % relative humidity (Permapure, PD100T-12MSS) and then **passed through a scrubber** (MAST Carbon) **to remove absorbing gaseous impurities such as ozone and nitrogen dioxide**."

The carbon monolith will not denude the aerosol organics effectively. We may get some absorption of gaseous organics in the monolith that will perturb the aerosol/gas phase semi-volatile partitioning. However, the residence time in the scrubber is too short to result in significant mass loss from the aerosol via this process. In measurements by others using carbon monoliths to remove semi-volatile aerosol components, the aerosol sample is heated to approximately 300 degrees Celsius within the monolith to volatilise organic components to the gas phase to effectively denude the particles.

3. Page 13 line 13: The authors speculate that dust does not influence the measurements due to the impactor. Could this thesis be supported by other measurements? The reviewer assumes that the cutting characteristics refers to 1.3 µm aerodynamic diameter?

The reviewer is correct – the diameter is indeed an aerodynamic diameter. We have updated page 13, line 13 (relating to fresh BBA), which now states:

"~~There was little influence~~ The impact of dust on our PAS, TAP and CRDS measurements was minimised because of the 1.3 µm aerodynamic impactor used. Based on the scattering Ångström exponent, there was likely a dust influence on this fresh BBA dataset."

4. Chapter 3.3 and Figure 8: The Angström exponent strongly dependents on the source and correction schema as the author points out. The reasons cannot be clarified, but the author can deduce further motivation for this manuscript.

   It is noticeable that the B1999 and also the V2010 method have the tendency to suppress large absorption Angström exponents. For the "urban" case with high R_OA/LAC ratio (c.f. figure 6) it means that the determination of the organic fraction by means of the Angström exponent would show large errors. For the "Fresh BBA" case, high absorption Angström expeonenten are measured. The R_OA/LAC ratio, on the other hand, is very low. Where do the large values for the Angström exponent come from? Could it be an indicator for mineral dust? In this case, all TAPs correction methods show large values for the Angström exponents.

   A deeper aerosol characterization is not the focus of this manuscript. However, the presented results provide another good reason why this manuscript is so important. The differentiation of aerosol types by absorption Angström exponents is becoming increasingly important, but as the data show, only with great uncertainties if filter-based photometers are used.

We have emphasised the importance of the findings of this manuscript in relation to the absorption Ångström exponent on original page 17, lines 11-13, which state:

"However, what is clear from this analysis is that there are large uncertainties in this important parameter when calculated from filter-based absorption measurements, and that these uncertainties are strongly source and correction scheme dependent. This cautions that significant uncertainties could be introduced if using the AAE to differentiate between types of aerosol."

Indeed, the findings of this manuscript in relation to the variability in AAE determined using filter-based absorption measurements may serve as a suitable motivation for studying this further.

We agree with the reviewer in that the large absorption Ångström exponents (AAE) tend to be suppressed for the B1999 and V2010 correction schemes for all aerosol types (except for the V2010 scheme for fresh BBA measurements). However, the AAE values presented in Figure 8 will not affect the results of Figure 6. The organic aerosol (OA) mass concentrations were determined from aerosol mass spectroscopy and the light absorbing carbon (LAC) mass concentrations were determined by converting the mass absorption coefficient (MAC) value of black carbon (BC) at 532 nm to 528 nm using a standard AAE value for BC of 1. Neither of these measurements used the AAE measurements of Figure 6 and, therefore, cannot have led to large uncertainties in $R_{OA/LAC}$.

We agree that large AAE values were measured for fresh BBA. As the reviewer has pointed out, this result could be an indication that the measurements were contaminated with mineral dust. However, large AAE values could also suggest the presence of absorbing organic aerosols. An analysis of the scattering Ångström exponent revealed a potential dust contribution to the MOYA aerosol sample. Please refer to the amendment made in response to point 3 above.

Additional corrections

1. In addition to the above changes, we have updated the processing script to account for a minor coding issue related to processing of scattering coefficients and subsequent application of these scattering measurements in the M2014 correction scheme. The impact of these corrections on the results is a maximum of 4 % and, therefore, the conclusions of this manuscript are unchanged. This has a minor impact on a subset of the single scattering albedos, which, similarly, does not change the results of the manuscript. There was no impact on the AAE values. The Tables and Figures have been updated accordingly. The corresponding numbers have been updated throughout the manuscript, as detailed below.

We have amended the text on original page 1, line 20, which now states:

"… consistently reduced biases to ~~0–17~~ 0–18 % at all wavelengths"

We have amended the text on original page 14, line 11, which now states:

"… it increased the bias at 467 nm by ~~2–5~~ 3–5 %, …"

We have amended the text on original page 14, line 17, which now states:

"… the range of TAP biases was ~~0.99–1.17~~ 1.01–1.18 …"

We have amended the text on original page 14, line 18, which now states:

"… relative to the B1999 and V2010 schemes by ~~7–40~~ 7–38 % and ~~7–27~~ 7–25 %, …"

We have amended the text on original page 14, lines 27-31, which now states:

"… were biased by greater than ~~1.71–1.79~~ 1.67–1.80, 1.46–1.70 and 1.39–1.42 for urban, fresh BBA and aged BBA when corrected using the B1999 scheme, respectively, dependent on wavelength. The M2014 (V2010 parameterisation) scheme reduced the biases with 10 % of measurements biased greater than 1.27–1.41, ~~1.17–1.24~~ 1.20–1.30 and ~~1.18–1.30~~ 1.18–1.29 for urban, fresh BBA and aged BBA, respectively, dependent on wavelength."

We have amended the text on original page 14-15, lines 33-34 and 1-2, which now states:

"The ~~one~~ exceptions to this trend were when the M2014 scheme (V2010 parameterisation) was applied to urban aerosol measurements, which led to the largest biases at wavelength 528 nm. The M2014 scheme (B1999 parameterisation) also led to the largest biases at 528 nm for all aerosol types."

We have amended the text on original page 15, line 9, which now states:

"The biases of ~~1–45~~ 1–45 % observed in this study …"

We have amended the text on original page 16, line 23, which now states:

"… which typically show a ~~~0–45~~ 1–45 % high bias in absorption."

2. To determine TAP absorption coefficients, we first averaged the light transmitted through the TAP filter spots to 30 seconds and then input these averaged intensity values into the standard equations used to generate TAP absorption coefficients (i.e. equations 1 to 9). To clarify that the TAP absorption coefficients *were not* calculated based on 1 Hz light transmission values, we have added the following to original page 12, line 11:

"In the case of TAP measurements, the intensities of light transmitted through a filter were first averaged to 30 seconds and then input into Eq. 1–9 to determine the corresponding absorption coefficients."

Original page 17, line 18:

"*... reduced the TAP mean bias to within ~~0 to +45~~ 1 to +45 % of ...*"

3. To account for a fault with a calibration unit, the AMS mass concentrations have been lowered by 38 %. This update only affects the results presented in Figure 6, which has the effect of moving the points to lower OA mass concentrations (i.e. moving points towards the left). This update does not alter the conclusions presented in section 3.2 or of the overall conclusions of this paper. We have updated Figure 6 accordingly.

4. Since this manuscript has been under review, a relevant paper has been published, which we have added reference to. We have added the following to page 11, lines 10-12:

"Moreover, our recent work has demonstrated that the calibration accuracy of PAS using ozone is optimal when the gas phase composition closely resembles that of ambient air (Cotterell et al., 2019), as is the case for calibrations performed for this work."

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Abstract. Biases in absorption coefficients measured using a filter-based absorption photometer (Tricolor Absorption Photometer, or TAP) at wavelengths of 467, 528 and 652 nm are evaluated by comparing to measurements made using photoacoustic spectroscopy (PAS). We report comparisons for ambient sampling covering a range of aerosol types including urban, fresh biomass burning and aged biomass burning. Data are also used to evaluate the performance of three different TAP correction schemes. We found that photoacoustic and filter-based measurements were well correlated, but filter-based measurements generally overestimated absorption by up to 45 %. Biases varied with wavelength and depended on the correction scheme applied. Optimal agreement to PAS data was achieved by processing the filter-based measurements using the recently developed correction scheme of Müller et al. (2014), which consistently reduced biases to 0–18 % at all wavelengths. The biases were found to be a function of the ratio of organic aerosol mass to light-absorbing carbon mass although applying the Müller et al. (2014) correction scheme to filter-based absorption measurements reduced the biases and the strength of this correlation significantly. Filter-based absorption measurement biases led to aerosol single-scattering albedos that were biased low by values in the range 0.00–0.07 and absorption Ångström exponents (AAE) that were in error by ± (0.03–0.54). The discrepancy between the filter-based and PAS absorption measurements is lower than reported in some earlier studies, and points to a strong dependence of filter-based measurement accuracy on aerosol source type.

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1 Introduction

Aerosol-radiation interactions are estimated to contribute a global mean effective radiative forcing of −0.45 (−0.95 to +0.05) W m^{-2}, offsetting a potentially significant but poorly constrained fraction of the positive effective radiative forcing associated with greenhouse gases (2.26 to 3.40) W m^{-2} (Myhre et al., 2013a). One of the major factors governing the uncertainty in estimates of aerosol direct radiative forcing is the poorly constrained aerosol single scattering albedo (SSA), defined as the ratio of aerosol scattering to total extinction (Loeb and Su, 2010; McComiskey et al., 2008; Sherman and

McComiskey, 2018). Accurate determination of aerosol SSA is limited by uncertainties in aerosol absorption estimates, which could potentially be underestimated by up to a factor of two in global climate models (Shindell et al., 2013; Stier et al., 2007).

The main types of absorbing aerosol include black carbon (BC) and light-absorbing organic carbon, commonly referred to as brown carbon (BrC) (e.g. Myhre et al., 2013a). On a global scale, aerosol absorption is dominated by BC, a carbonaceous product formed during incomplete combustion, which may exert the next largest positive radiative forcing after carbon dioxide (Stocker et al., 2013). BC absorbs strongly across visible wavelengths and contributes an estimated 0.71 (0.09 to 1.26) W m^{-2} to aerosol direct radiative forcing (Bond et al., 2013). In recent years, BrC has received increasing attention as a climate forcing agent (e.g. Feng et al., 2013). Sources of BrC include primary emissions during biomass and biofuel combustion as well as secondary production via photo-oxidation of volatile organic compounds (Andreae and Gelencsér, 2006; Wang et al., 2018). BrC has been found to absorb strongly towards ultraviolet wavelengths, although the strength and wavelength dependence of this absorption is uncertain, due in part to the wide range of compounds that this term encompasses, many of which are poorly characterised (Andreae and Gelencsér, 2006; Lack et al., 2012b; Pokhrel et al., 2017). Climate models generally only crudely represent the optical properties of BC and BrC and their evolution with time. For example, while the Met Office Hadley Centre HadGEM3 model treats the internal mixing of aerosol components, the real and imaginary parts of the refractive index of organic carbon that are used to calculate the radiative properties of the composite aerosol are fixed (e.g. Johnson et al., 2016). In order to address this deficiency, stronger observational constraints are first required (e.g. Alexander et al., 2008; Bond et al., 2013; Liu et al., 2014; Myhre et al., 2013b; Saleh et al., 2014; Wang et al., 2018).

Over the course of several decades, filter-based absorption photometry has been used to measure aerosol absorption coefficients. The approach has considerable benefits including that it is relatively inexpensive, portable and capable of unattended measurements for long periods of time (Baumgardner et al., 2012). Filter-based instruments measure the light transmittance across a filter continuously, which changes as particles are deposited onto the filter, providing a measure of aerosol absorption (see Sect. 2.1) (e.g. Bond et al., 1999). Absorption coefficients determined using filter-based absorption photometry can be subject to measurement artefacts due to (i) scattering of light away from the light-detector leading to erroneous apparent absorption (Bond et al., 1999) and (ii) enhanced absorption as particles are deposited onto the filter. The latter leads to multiple scattering between the particles and the filter medium, providing multiple opportunities for absorption. The enhancement is complex to characterise and depends on the filter loading such that an increase in the number of deposited absorbing particles reduces the multiple scattering between the filter and particle layers (Bond et al., 1999; Liousse et al., 1993; Weingartner et al., 2003) leading to lower absorption coefficients for highly loaded filters (Weingartner et al., 2003). The sensitivity of filter-based absorption photometers is also affected by the penetration depth of particles within the filter matrix, which depends on particle size (Moteki et al., 2010; Nakayama et al., 2010). A number of

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empirical and semi-empirical correction schemes have been derived to correct for the aforementioned artefacts. This study will focus on correction schemes derived for use with glass-fibre Pallflex E70-2075W filters that have been used widely with the Particle Soot Absorption Photometer (PSAP, Radiance Research) (Bond et al., 1999; Müller et al., 2014; Virkkula, 2010; Virkkula et al., 2005). These correction schemes are also valid for similar instruments using this filter substrate, for example the Tricolor Absorption Photometer (TAP, Brechtel Manufacturing) used in this study and described in Sect. 2.3.2 (Ogren et al., 2017).

Another potentially significant measurement artefact is due to liquid-like organic aerosols spreading across the filter fibres (Lack et al., 2008). The mechanisms proposed for this artefact include a change in the physical shape and therefore optical properties of deposited particles, or a coating effect whereby deposited particle absorption is enhanced via a lensing effect (Cappa et al., 2008; Lack et al., 2008; Subramanian et al., 2007). Although recognised as potentially significant, there are no empirical corrections to account for these artefacts.

Previous work has examined the magnitude of biases in filter-based absorption measurements. For example, Lack et al. (2008) found PSAP absorption coefficients were biased high in the range 12 % to over 200 % at 532 nm compared to photoacoustic spectroscopy measurements for aerosols over the Gulf of Mexico, which included BC, nitrate, sulphate and organic aerosols from shipping emissions. The PSAP biases were found to be positively correlated to the organic aerosol mass concentration and even more strongly correlated to the ratio of the organic aerosol to light-absorbing carbon mass. To verify these measurements, Cappa et al. (2008) performed laboratory experiments using secondary organic aerosol (SOA) derived from the ozonolysis of α-pinene, which had a SSA > 0.998 at 532 nm. A key finding of this study was that for external mixtures of SOA and soot, the PSAP absorption could be biased high by a factor 2.6, consistent with the findings of Lack et al. (2008). Cappa et al. (2008) also found that the magnitude of the absorption bias was strongly dependent upon the filter transmittance and that the bias was both immediate (clean filter) and cumulative (filter previously exposed to absorbing material). The results from both of these studies (Cappa et al., 2008; Lack et al., 2008) were independent of the correction scheme applied (Bond et al., 1999; Virkkula et al., 2005).

More recently, Subramanian et al. (2010) derived the BC mass absorption coefficient (MAC) at 660 nm for fresh and 1–2 day-old aerosol emissions in and around Mexico City by dividing the absorption coefficients measured using a PSAP by the refractory BC mass concentrations measured using a single particle soot photometer (SP2, Droplet Measurement Technologies). For the fresh emissions, they found a ~50 % enhancement in their measured BC MAC relative to the value reported by Bond and Bergstrom (2006), whose review was based on an extensive range of measurements. The BC MAC bias was attributed in part to an over-estimation of the absorption coefficients measured by the PSAP due to externally mixed liquid-like organic matter. However, the BC MAC values for the relatively thickly coated, aged BC further from the city were in line with those estimated by Bond and Bergstrom (2006), which the authors suggested may indicate that biases in filter-based measurements relating to high organic aerosol loading may only be present when organic aerosol is externally mixed with BC (Subramanian et al., 2010).

Using a similar methodology, McMeeking et al. (2011) derived the BC MAC at 550 nm using PSAP and SP2 measurements for urban pollution aerosols around the UK, reporting organic aerosol mass concentrations in the range 1–7 μg m^{-3}. The work by Lack et al. (2008) indicates that a positive absorption bias of up to 50 % would be expected at these loadings, however no bias in the BC MAC was observed. McMeeking et al. (2011) postulated that this result could be due to limitations in the PSAP and SP2 measurements or a physical effect whereby absorption enhancements due to coatings were offset by the collapse of fractal BC aggregates (McMeeking et al., 2011). Indeed, another explanation for this discrepancy could have been that the organic aerosol sampled here was not quasi-liquid like and contributed different biases to those seen in previous studies.

Biases in filter-based absorption photometry measurements can limit the accurate determination of key climate-relevant parameters including, for example, the aerosol SSA and AAE (e.g. Sherman and McComiskey, 2018). Mason et al. (2018) compared PAS to filter-based absorption measurements of wildfires and agricultural fires over the continental U.S.A. during August and September 2013, which included a PSAP and a Continuous Light Absorption Photometer (CLAP) (Ogren et al., 2017). All PSAP and CLAP data were corrected using the Bond et al. (1999) and Ogren (2010) corrections. Biases in filter-based measurements were evaluated by comparison to PAS measurements, which were in the range 0.61 to 1.24, dependent on measurement wavelength (405, 532 and 660 nm). Mean SSA and AAE values derived using filter-based absorption photometry were found to be in error by up to 0.03 and 0.7, respectively, compared to PAS.

Further, Backman et al. (2014) assessed the sensitivity of the PSAP-derived AAE to the Bond et al. (1999) and Virkkula (2010) correction schemes for measurements recorded on the central Highveld in South Africa, where emissions were dominated by fossil-fuel burning activities including from coal-fired power plants. They found that the AAE varied between 1.34 to 1.96 dependent upon the PSAP correction scheme applied, which led to different conclusions regarding the aerosol composition and source (Backman et al., 2014).

Despite this body of previous work, there remains significant uncertainty related to the magnitude of biases in filter-based absorption measurements, particularly regarding dependence on source type and the correction scheme applied. The aim of this study is to address this gap. We assess biases by comparing absorption coefficients determined using multi-wavelength TAP and photoacoustic instruments during a series of research flights aboard the UK Facility for Airborne Atmospheric Measurements (FAAM) BAe-146 aircraft. Aerosol sources sampled include urban aerosol emissions over London, fresh biomass burning aerosol (BBA) over West Africa and aged BBA over the Southeast Atlantic Ocean. We follow the methodology of Lack et al. (2008) by looking at the absorption biases as a function of organic aerosol concentration, extending their study by looking at a greater range of wavelengths and aerosol sources as well as evaluating additional correction schemes, namely those developed by Virkkula (2010) and Müller et al. (2014). We then assess the impact that biases in filter-based absorption photometry have on the aerosol SSA and AAE. This is the first study to simultaneously evaluate the Bond et al. (1999), Virkkula (2010) and Müller et al. (2014) correction schemes for ambient aerosol sampling across multiple aerosol types.

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2. Methodology and measurements

2.1 Principles of filter-based absorption photometry

Filter-based absorption photometers measure the light transmitted through a filter as particles are deposited onto the filter such that the attenuation can be defined as

$$I = -ln\left(\frac{I_s}{I_r}\right), \tag{1}$$

where I_s and I_r are the intensities of light transmitted through a filter corresponding to a sample spot (i.e. an area of the filter with deposited aerosols) and reference spot (i.e. an area of the filter without deposited aerosols), respectively (Ogren et al., 2017). The attenuation coefficient can thus be determined using

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$$b_{ap}^{raw} = \frac{A}{Q\Delta t}\left(I(t+\Delta t) - I(t)\right), \tag{2}$$

where A is the area of the aerosol deposited onto a filter, Q is the flow rate of the aerosol-laden stream pulled through a filter, Δt is the time between successive measurements of light attenuation and $I(t)$ and $I(t+\Delta t)$ are the light attenuations at times t and $t+\Delta t$ (Ogren et al., 2017). To correct b_{ap}^{raw} for apparent and enhanced absorption, we applied the correction schemes developed by Bond et al. (1999), Virkkula, (2010) and Müller et al. (2014), which will be referred to as b_{ap}^{B1999}, b_{ap}^{V2010} and b_{ap}^{M2014} respectively. See Sect. 2.1.1–2.1.3. The code used to run the analysis presented in this manuscript, i.e. relating to the equations presented throughout this section, was implemented in Python.

2.1.1 The Bond et al. (1999) correction scheme (B1999)

The Bond et al. (1999) correction scheme was developed empirically by comparing PSAP absorption coefficients to reference absorption coefficients determined using the difference between extinction as measured by an optical extinction cell and scattering coefficients measured using a nephelometer. Calibration aerosols included polydisperse nigrosin and ammonium sulphate. This correction scheme was updated by Ogren (2010). Bond et al. (1999) found that

$$b_{ap}^{B1999} = f(Tr)b_{ap}^{raw} - sb_{sp}, \tag{3}$$

with

$$f(Tr) = \frac{0.85}{K_2(1.0796Tr+0.71)}, \tag{4}$$

$$s = \frac{K_1}{K_2}, \tag{5}$$

and where b_{sp} is the scattering coefficient, K_1=0.02, K_2=1.22 and Tr is the normalised filter transmittance, defined as (Ogren et al., 2017)

$$Tr = \frac{I_s(t)/I_r(t)}{I_s(0)/I_r(0)}. \qquad (6)$$

This correction scheme was derived at the wavelength 550 nm and is generally assumed to apply over the entire range of visible wavelengths, though there is no empirical basis for this (Bond et al., 1999; Ogren, 2010).

2.1.2 The Virkkula (2010) correction scheme (V2010)

The Virkkula et al. (2005) correction scheme and the subsequent Virkkula (2010) erratum were derived for the PSAP wavelengths 467, 530 and 660 nm, which is reflected by the $f(Tr,\lambda)$ term described below. The scheme was derived by comparing absorption coefficients determined using a multi-wavelength PSAP to those measured using either photoacoustic spectroscopy or to absorption derived by subtracting scattering from extinction measurements (Virkkula et al., 2005). Calibration aerosols included kerosene soot, graphite, diesel soot, ammonium sulphate and polystyrene latex spheres. Virkkula (2010) proposed that

$$b_{ap}^{V2010} = f(Tr,\lambda)b_{ap}^{raw} - sb_{sp}, \qquad (7)$$

where

$$f(Tr,\lambda) = k_0 + k_1(h_0 + h_1\omega_0)ln(Tr), \qquad (8)$$

and where k_0, k_1, h_0, h_1 and s are wavelength dependent constants and ω_0 is the wavelength dependent SSA. The values of the constants used in this study were taken directly from Table 1 in Virkkula (2010), which are provided in Table 1. The wavelengths at which these constants were derived differ to those used in the TAP by 2 nm and 8 nm at the green and red wavelengths, respectively. It is unclear how these constants depend on wavelength. To assess the impact that this wavelength mismatch might have on the absorption coefficients derived using the V2010 correction scheme, the single-wavelength V2010 constants were also applied to TAP measurements. These were taken from Table 1 in Virkkula (2010) and are provided in the fifth column of Table 1. This was found to have a moderate impact on the results of this study as discussed in Sect. 3. The Virkkula (2010) correction is an iterative correction scheme due to its dependence on the SSA. Hence the algorithm was run 10 times for each time-step, which was sufficient for the absorption coefficient to converge to a single value with a precision better than 0.001 Mm^{-1}.

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2.1.3 The Müller et al. (2014) correction scheme (M2014)

The constrained two-stream (CTS) algorithm developed by Müller et al. (2014) includes a two-stream radiative transfer model that explicitly accounts for the optical properties of the filter substrate and deposited particles and is constrained by either the Bond et al. (1999), Virkkula et al. (2005) or Virkkula (2010) parameterisations. This section covers the key equations from Müller et al. (2014) to show how they have been implemented in this study and the reader is referred to

Müller et al. (2014) for a full derivation. The M2014 correction scheme makes use of the relationship between the absorption coefficient and the change in particle absorption optical depth, δ_{ap}, on the filter medium between two measurements separated by a time-step Δt, as represented by:

$$b_{ap}^{M2014} = \frac{A}{Q\Delta t}\left(\delta_{ap}(t+\Delta t) - \delta_{ap}(t)\right), \tag{9}$$

For each time-step, δ_{ap} was calculated iteratively by minimising the difference between the measured total optical depth, δ_{tot} (filter + particles) and the relative optical depth, δ_{CTS}, which is the change in total optical depth of the filter system after collecting a particle relative to the unloaded filter. A Newton-type solver was applied, as suggested by Müller et al. (2014), and required ten iterations to converge to a precision better than 0.001 Mm^{-1}. δ_{tot} was calculated from measurements of the filter, with and without aerosol, using Eq. 1. The equations outlined in Müller et al. (2014) were used to calculate δ_{CTS} and are included here for clarity.

$$\delta_{CTS} = \frac{F_s^{exp}\delta_{sp} + F_a^{exp}\delta_{ap}}{F_f^{mod}}, \tag{10}$$

where $\delta_{sp}$is the particle scattering optical depth, calculated using

$$\delta_{sp} = \frac{Q\Delta t}{A}\sum_{t=0}^{t} b_{sp}(t), \tag{11}$$

$$F_s^{exp} = a_5 + \left(a_0 + a_1 g_p\right)e^{-\left(\frac{ln(\delta_{sp}) + a_4^2}{a_3 + a_4 g_p}\right)^4}, \tag{12}$$

where $a_0 = 0.1509$, $a_1 = -0.1611$, $a_2 = 4.5414$, $a_3 = -5.7062$, $a_4 = -1.9031$, $a_5 = 0.01$ and g_p is the average weighted particle asymmetry parameter (see Eq. 24). Using the B1999 empirical correction,

$$F_{a,B1999}^{exp} = \frac{1}{\delta_{ap}} ln\left(\frac{e^{c_2\delta_{ap} + ln(c_1+c_2)} - c_1}{c_2}\right), \tag{13}$$

where $c_1 = 1.555$ and $c_2 = 1.023$, which were derived in Bond et al. (1999); see the alternative formulation of the B1999 correction in Müller et al. (2014). Alternatively, using the V2010 empirical correction,

$$F_{a,V2010}^{exp} = \frac{1}{\delta_{ap}}\sqrt{\left(\frac{c_1}{c_2 h_0}\right)^2 - \frac{2\delta_{ap}}{c_2 h_0}} + \frac{c_1}{c_2 h_0}, \tag{14}$$

where c_1, c_2, h_0, h_1 and s correspond to the wavelength dependent constants k_0, k_1, h_0, h_1 and s as defined in Sect. 2.1.2, corresponding to the Virkkula (2010) parameterisation. Finally,

$$F_f^{mod}\left(\delta_{ap}, \delta_{sp}, g_p\right) = \frac{\delta(\delta_{ap}=0, \delta_{sp}, g_p) + \delta(\delta_{ap}, \delta_{sp}=0, g_p)}{\delta(\delta_{ap}, \delta_{sp}, g_p)}, \tag{15}$$

where

$$\delta\left(\delta_{ap}, \delta_{sp}, g_p\right) = -ln\left(T_{2L}\left(\delta_{ap}, \delta_{sp}, g_p\right)\right) + ln\left(T_{2L}\left(\delta_{ap} = 0, \delta_{sp} = 0, g_p = 0\right)\right), \tag{16}$$

$$T_{2L} = \frac{T_1 T_2}{1 - R_1(1 - T_2)}, \tag{17}$$

and T_1 and T_2 represent the filter transmittances of the particle-loaded and particle-free layers, respectively. These are represented by layers 1a and 1b in Müller et al. (2014), respectively. The filter transmittance and reflectance are given by

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$$T = \frac{2}{[2-\omega_0(1+g)]sinh(K\delta_e/\mu_1)/K+2cosh(K\delta_e/\mu_1)} \quad (18)$$

and

$$R = \frac{\omega_0(1-g)sinh\left({}^{K\delta_e}/_{\mu_1}\right)/K}{[2-\omega_0(1+g)]sinh\left({}^{K\delta_e}/_{\mu_1}\right)/K+2cosh\left({}^{K\delta_e}/_{\mu_1}\right)}, \quad (19)$$

where

$$\delta_e = \chi\delta_{sf} + \delta_{sp} + \chi\delta_{af} + \delta_{ap}, \quad (20)$$

$$K = \sqrt{(1-\omega_0)(1-\omega_0 g)}, \quad (21)$$

$$\omega_0 = \frac{\chi\delta_{sf}+\delta_{sp}}{\chi\delta_{sf}+\delta_{sp}+\chi\delta_{af}+\delta_{ap}}, \quad (22)$$

and

$$g = \frac{\chi g_f\delta_{sf}+g_p\delta_{sp}}{\chi\delta_{sf}+\delta_{sp}}. \quad (23)$$

The filter scattering optical depths used in this study were $\delta_{sf}^{467} = 7.76$, $\delta_{sf}^{530} = 7.69$ and $\delta_{sf}^{660} = 7.34$ and the filter absorption optical depths used were $\delta_{af}^{467} = 0.033$, $\delta_{af}^{530} = 0.038$ and $\delta_{af}^{660} = 0.019$, as measured by Müller et al. (2014) for the same type of filters. Small differences between wavelengths that the filter optical properties were measured at by Müller et al. (2014) (467, 530, 660 nm) compared to those at which the TAP measures (467, 528, 652 nm) were assumed to be negligible. Following the nomenclature of M2014, for filter layer 1 (the particle-loaded filter layer) $\chi = 0.2$ and for layer 2 (the unloaded filter layer) $\chi = 0.8$. This assumes that the particle penetration depth into the filter was 20 % and accounts for the fractional filter optical depths corresponding to each layer. The value used for μ_1 was ${}^{1}/_{\sqrt{3}}$. The value g_p is the average weighted asymmetry parameter of all particles deposited onto the filter, given by

$$g_p = \frac{\sum_i b_{sp}^i g_p^i}{\sum_i b_{sp}^i} \quad (24)$$

where i represents the i th ensemble of particles with scattering coefficient b_{sp}^i. Equation 24 is a practical way to apply equation 5 presented in Müller et al. (2014) who instead used an equivalent method, which utilised individual particle scattering cross sections (as opposed to ensemble scattering coefficients). We used Eq. 24 as opposed to the recommended formulation because nephelometer measurements represent an ensemble. In this study, bulk asymmetry parameters (i.e. corresponding to an ensemble of particles) were calculated for each time-step using the parameterisation

$$g_p = -6.347b_{back-sp}^3 + 6.906b_{back-sp}^2 - 3.859b_{back-sp} + 0.9852, \quad (25)$$

where $b_{back-sp}$ is the backscattering ratio measured using a nephelometer (Moosmüller and Ogren, 2017).

To confirm the accuracy of the implementation of the M2014 algorithm used during this analysis, equations 16–23 were used to reproduce the results in Fig. 6 of the Müller et al. (2014) study, which were verified against intermediate results (T. Müller, personal communication, 2016).

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2.3 Measurements and instrumentation

All measurements presented in this study were made aboard the UK's BAe-146-301 large Atmospheric Research Aircraft (ARA) operated by the Facility for Airborne Atmospheric Measurements (FAAM; www.faam.ac.uk). The aircraft is capable of carrying 3 crew, 18 scientists and a total scientific payload of up to 4000 kg with a range up to 3700 km. This section provides information on the filter-based, photoacoustic, nephelometer and aerosol composition instrumentation used aboard the aircraft and introduces the environments in which measurements were made.

2.3.1 Aerosol sampling and conditioning

An important strength of this dataset is that the TAP, PAS and cavity ring-down spectrometer (CRDS) instruments used to sample aerosol optical properties all shared a common sample inlet and were subject to the same flow conditioning. Aerosols were drawn into the aircraft through a modified Rosemount inlet (Trembath et al., 2012). The aerosol-laden stream was first dried to < 20 % relative humidity (Permapure, PD100T-12MSS) and then passed through a scrubber (MAST Carbon) to remove absorbing gaseous impurities such as ozone and nitrogen dioxide. An impactor removed particles with aerodynamic diameter > 1.3 µm (Brechtel, custom design). A series of flow splits (Brechtel 1110 and 1104) evenly distributed the aerosol-laden stream between the suite of instruments, which each sampled the aerosol at a flow rate of 1 L min^{-1}, as shown in Fig. 1. All measurements were corrected to standard temperature and pressure (PAS, CRDS and TAP: 20 °C and 1013 mb).

2.3.2 Tricolor Absorption Photometer

The TAP is a commercially available (Brechtel) version of the Continuous Light Absorption Photometer (CLAP), described by Ogren et al. (2017). The TAP comprises eight sample filter spots and two reference filter spots. The aerosol-laden air passes through one sample spot at a time, which allows for eight times the filter lifetime compared to single-spot photometers. The filtered air is re-circulated through one of the reference spots to enable the attenuation calculation (see Eq. 1) (Ogren et al., 2017). Upon reaching a pre-defined filter transmittance set point, the TAP automatically changes to the next available sample filter spot. We used 47 mm diameter Pallflex (E70-2075W) glass-fibre filters, which were nominally identical to the filters used to derive the correction schemes applied in this study (see Sect. 2.1.1–2.1.3). The TAP provides measurements at three wavelengths with peaks centred at 467, 528 and 652 nm, which allows the spectral dependence of climate relevant parameters such as the SSA and AAE to be evaluated (Sect. 3.3). The LEDs are cycled through each wavelength once per second, providing absorption measurements at 1 Hz at all wavelengths. The inlet of the TAP is heated to 35.2 ± 0.2 °C to minimise the effects of changing temperature and to prevent water condensing onto the filter. The built-in digital low-pass filter was disabled in all of our measurements to enable calculation of the absorption coefficients from the raw photodiode measurements, as it was unclear how the low-pass filter impacted the measurements. To understand the impact of this on instrument sensitivity, the TAP was run for ~3 h in the laboratory while it sampled filtered room air to characterise the noise in the system. Uncorrected attenuation coefficients, b_{ap}^{raw}, were calculated at 1 Hz, and the average and

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standard deviation for each time interval Δt ($1 < \Delta t < 1000$ s) were calculated. The 1-sigma detection limits at 30 s averaging time were 0.71, 1.37 and 0.89 Mm^{-1} at wavelengths 658, 528 and 467 nm, respectively. Ogren et al. (2017) calculated the mean 1-sigma detection limit for their 28 instruments over all three wavelengths to be 0.33 Mm^{-1}. The difference between the detection limits measured in this study and that presented in Ogren et al. (2017) could be due to running without low-pass digital filtering in the current study and/or differences between the TAP and CLAP. TAP internal particle losses were estimated to be < 1 % for particles with diameters in the range 0.03–2.5 µm (Ogren et al., 2017).

To determine the areas of the spots resulting from particle deposition onto the filter, nigrosin (Sigma Aldrich, product number 198285-100G) was atomised from solution, dried to < 10 % relative humidity using a silica gel diffusion drier (Topas, DDU-570) and sampled by the TAP. The areas of the eight sample spots were determined by measuring the number of pixels corresponding to the diameters in a magnified digital photograph, which yielded areas in the range 32.4–36.8 mm^2. The manufacturer-recommended spot sizes are 30.7210 mm^2. Filter spot sizes were determined using nigrosin rather than from the ambient aerosol samples themselves as the spot edges were more clearly defined. The spot edges of the deposited ambient aerosol were difficult to detect as the filter spot was changed at the start of each day when measurements were taken. It was possible to detect the aerosol spot for measurements that corresponded to high loadings of absorbing aerosol. In these cases there was evidence of aerosols spreading across the filter and the area of the spots was larger by 5–20 %. However, this observation is based on a limited sample of three aerosol spots and the timescale for spread across the filter is unclear. This analysis used the areas determined using the clearly defined nigrosin spots, and therefore provides a lower limit of area absorption coefficient (see Eq. 2), and as will be shown in Sect. 3, the TAP absorption bias.

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2.3.3 Photoacoustic and cavity ring-down spectrometers

The photoacoustic and cavity ring-down spectrometers used in this study were based on the designs by Lack et al. (2012) and Langridge et al. (2011), respectively and are described in detail in Davies et al. (2018) and Szpek et al. (in preparation). PAS measures absorption directly for aerosols in their suspended state (Arnott et al., 1999). The PAS principle relies on converting energy from a light source into sound. Light-absorbing media, such as aerosol, transfer electromagnetic energy into thermal energy that heats the surrounding air. This gaseous heating generates a pressure wave, which is detected by a microphone located within the PAS cell. The amplitude of the microphone signal is related to the sample absorption coefficient through calibration (Arnott et al., 1999; Davies et al., 2018; Moosmüller et al., 2009).

Much of this analysis relies on accurate PAS absorption measurements and thus we focus here on describing the uncertainty associated with these measurements. The total PAS measurement uncertainty is comprised of the measurement precision and accuracy. The PAS measurement precision was derived by evaluating the minimum sensitivities of the suite of PAS instruments in a similar way to the TAP, as described in Sect. 2.3.2, and were in the range 0.01–0.06 Mm^{-1} for 30 s averaging across the range of cells used. The minimum sensitivities of the suite of CRDS cells were evaluated in the same way and were found to be 0.02–0.05 Mm^{-1} across the range of cells used.

The accuracy of PAS absorption measurements was determined primarily by three factors: (i) uncertainty in the ozone calibration, (ii) uncertainty in corrections applied to account for the PAS microphone pressure sensitivity and (iii) uncertainty in subtraction of background noise which arose primarily from laser heating of the PAS cell optical windows. We consider each of these in turn below.

The accuracy of the PAS ozone calibration has previously been evaluated in laboratory experiments that compared measured and modelled absorption and extinction cross sections of strongly-absorbing nigrosin aerosol. This analysis showed the PAS calibration accuracy to be better than 8 % and the accuracy of the CRDS instruments used in this study to be better than 2 % (Davies et al., 2018). Moreover, our recent work has demonstrated that the calibration accuracy of PAS using ozone is optimal when the gas phase composition closely resembles that of ambient air (Cotterell et al., 2019), as is the case for calibrations performed for this work.

The second source of PAS measurement uncertainty was due to the PAS microphone sensitivity to pressure, which was evaluated by performing ozone calibrations at several pressures in the range 600–1000 mb (typical of those encountered during airborne operation). The measured PAS microphone sensitivities were fit to a linear trend across this range and normalised to yield a correction factor that varied from 0.83 (600 mb) to 1.00 (1000 mb). The uncertainty introduced by applying this pressure-dependent correction to PAS calibrations was estimated by propagating the 1σ fitting uncertainties in the linear regression between the calibration factors to in-flight PAS measurements, which led to uncertainties in PAS absorption coefficient measurements of 0.0–1.2 %. The smallest uncertainties were associated with measurements around 1000 mb where there was no correction applied and largest for relatively low pressures where the largest correction was applied.

The third source of PAS measurement uncertainty was due to subtraction of window-generated background noise, which is unstable for airborne operation due to its dependence on pressure. To account for this, in-flight background noise is typically characterised by periodically measuring a filtered-air stream for 30 s every 300 s. These measurements are then used post-flight to derive a background correction as a function of pressure. To evaluate the uncertainty introduced by this background noise correction, we took continuous PAS measurements of filtered-air in the laboratory and varied the pressure within the PAS cells over the range encountered during airborne operation. This laboratory PAS dataset was then processed to mimic in-flight conditions, with 30 s windows of data every 300 s being used to derive a continuous pressure-dependent background correction. Examining the difference between the continuous filtered-air measurements and the synthetically generated background data series provided the uncertainty in the background noise correction under variable pressure conditions. The uncertainty in the background noise correction was found to be normally distributed, with a 1σ width of 1.81–2.30 % across the range of cells used. This uncertainty was propagated through in-flight PAS data processing to derive

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the uncertainties introduced to airborne PAS absorption coefficient measurements from the background noise subtraction. The uncertainty was found to be 0.27–0.54 Mm^{-1}, which led to larger percentage uncertainties for lower absorption coefficients. The noise performance was no worse than a factor of 2 larger for airborne operation.

The total uncertainty in PAS measurements is the combination of the measurement precision and accuracy, including the PAS calibration accuracy, the pressure-dependent calibration correction uncertainty and the background noise correction uncertainty. These factors were combined in quadrature, leading to total PAS measurement uncertainties of 29.0–55.0 % for 1 Mm^{-1} absorption coefficient measurements across the range of cells used (independent of pressure) and approximately 8.1 % for 100 Mm^{-1}. These uncertainties are in-line with previous estimates for airborne PAS measurements, which were found to be ± 5 % for ground-based measurements with an additional ± 0.5 Mm^{-1} for airborne measurements (Lack et al., 2012a).

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2.3.4 Additional measurements

Nephelometer measurements (TSI 3563) were used to derive the aerosol asymmetry parameter needed to apply the Müller et al. (2014) correction scheme (see Sect. 2.1.3) and were corrected according to Müller et al. (2011). A Time-Of-Flight Aerosol Mass Spectrometer (TOF-AMS) (e.g. Drewnick et al., 2005) measured the aerosol composition. The TOF-AMS was run as described in previous publications (e.g. Morgan et al., 2010).

2.3.5 Data Averaging

All absorption, scattering and extinction coefficient data measured using the PAS, TAP, CRDS and nephelometer were recorded at 1 Hz. Data were subsequently averaged to 30 seconds during post-flight analysis to reduce the noise in these measurements and to aid temporal alignment of the PAS and TAP for direct comparisons. In the case of TAP measurements, the intensities of light transmitted through a filter were first averaged to 30 seconds and then input into Eq. 1–9 to determine the corresponding absorption coefficients. To account for time lags between the PAS and TAP, an optimisation routine was run that maximised the correlation coefficient (R^2) between the absorption coefficients determined using the PAS and TAP by delaying one instrument relative to the other. There was no time lag between the PAS and CRDS when using an averaging time of 30 seconds. Time alignment was verified by visually confirming that the rising and falling edges of the peaks in the absorption coefficients aligned.

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2.3.6 Flights and meteorology

This study uses data collected aboard the FAAM aircraft during 30 research flights (each 3-4 hours duration) in three distinct regions: London (three flights, 17 to 20 July 2017, from 1.7° W to 2.0° E and from 50.6° to 52.9° N), West Africa (three flights, 28 February to 1 March 2017, from 14.2° to 17.6 °W and from 9.6° to 14.8° N) and the Southeast Atlantic Ocean (24 flights, 16 August to 7 September 2017, from 8.0° to 18.6° W and from 4.6° N to 10.9° S). Figure 2 shows a map with the flight tracks indicated. All flights involved straight and level runs as well as deep profiles. Also shown in Fig. 2 are the mean aerosol optical depths (AODs) measured using the Moderate Resolution Imaging Spectroradiometer (MODIS) instruments aboard the Terra and Aqua satellite platforms (Remer et al., 2013) for each measurement period. The mean AOD for each region is shown corresponding to all satellite overpasses during the flight periods for both MODIS instruments. Figure 2 also shows time series of the columnar AOD values measured using the Aerosol Robotic Network (AERONET) for the Chilbolton and Oxford (~ 95 km southwest and northwest of London respectively), Dakar (West Africa) and Ascension Island (Southeast Atlantic Ocean) sites.

Urban emissions: during 17-20th July 2017, back trajectory analysis shows north-westerly flow brought air masses from over the Irish Sea to London (Rolph et al., 2017; Stein et al., 2015; available at http://ready.arl.noaa.gov/HYSPLIT_traj.php). Flights provided measurements of regional background aerosol (Northwest London) as well as the London pollution plume (Southeast London). AOD values of ~0.00–0.13 were measured using the AERONET sites at Chilbolton and Oxford during the measurement period, as shown in Fig. 2. Mean in-flight carbon monoxide (CO) concentrations were 98 ppbv indicating the presence of fossil fuel burning, for example from transport emissions and industrial processes (e.g. Dentener et al., 2001). These flights predominantly sampled the boundary layer with a maximum aircraft altitude of 2.2 km.

Fresh biomass burning emissions: flights over West Africa were dominated by freshly emitted BBA encountering similar conditions to those sampled during previous FAAM flight campaigns at the same time of year (e.g. DABEX; Haywood et al., 2008). Low-level flying through visible smoke plumes enabled measurements of fresh BBA within a few minutes of emission. During the measurement period, MODIS measured mean AOD values ~ 0.5–0.7 over large swaths of West Africa, > 1.0 near to the coast and ~ 0.5–1.0 over the Atlantic Ocean offshore of West Africa and AERONET reported AOD values in the range ~ 0.5–0.9 over Dakar, as shown in Fig. 2. Many flights targeted measurements close to the source and were dominated by fresh BBA emissions. The impact of dust on our PAS, TAP and CRDS measurements was minimised because of the 1.3 µm aerodynamic impactor used. Based on the scattering Ångström exponent, there was likely a dust influence on this fresh BBA dataset. Mean in-flight CO concentrations were 175 ppbv although concentrations greater than 14000 ppbv were measured when flying through plumes close to the aerosol source, indicative of fresh biomass burning emissions.

Aged biomass burning emissions: flights around Ascension Island sampled aged biomass burning aerosols transported from mainland Southern Africa in a general anticyclonic circulation (e.g. Garstang et al., 1996; Zuidema et al., 2016). East of ~ 8° W, MODIS reported mean AOD values generally between 0.1–0.5 and up to ~ 0.8 in the east of the area in which flights occurred. AERONET consistently measured AOD values between 0.1–0.5 over Ascension Island (the campaign base) during the entire four week measurement period. Mean CO concentrations were 126 ppbv, confirming that emission likely originated from a combustion source. Flights were performed in both the boundary layer and free troposphere. Based on HYSPLIT back trajectories, aerosols had generally undergone ~ 1 week of atmospheric transport since emission (Haywood et al., in preparation).

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3 Results and discussion

3.1 TAP-PAS comparisons

The primary result of this study is that the absorption coefficients determined using a TAP and PAS are linearly correlated and that the slope (R_{abs}) is dependent upon the aerosol source, measurement wavelength and the correction scheme applied to the TAP measurements. Scatter plots showing the relationship between absorption coefficients measured simultaneously by the TAP and PAS for urban, fresh and aged BBA are shown in Fig. 3–5 respectively. Tight correlations between TAP and PAS measurements were observed across all aerosol sources and for all correction schemes. All linear regressions between TAP and PAS measurements were forced through the origin. A summary of R_{abs} can be found in Table 2.

For the B1999 correction scheme, the range of TAP biases across all aerosol sources was 1.18–1.45. The smallest biases were consistently associated with 467 nm or 652 nm wavelength measurements and largest for 528 nm wavelength measurements. An interesting feature of this result is that the B1999 scheme led to the largest biases at 528 nm, which is the wavelength closest to that at which the scheme was derived.

For the V2010 correction scheme, the range of TAP biases across all aerosol sources was 1.08–1.38. The largest biases were consistently at 405 nm and smallest at 652 nm. Relative to the B1999 correction scheme, the V2010 scheme reduced the biases at 528 and 652 nm by 5–15 % while it increased the bias at 467 nm by 3–5 %, dependent on the aerosol source. The sensitivity of TAP biases to the wavelength dependent constants used in the V2010 scheme was investigated due to the mismatch in the TAP wavelengths and those for which the V2010 correction scheme was derived. Applying the single-wavelength V2010 correction scheme (i.e. applicable at all wavelengths) decreased TAP biases by 7–9 % at 467 nm, increased biases by 1 % at 528 nm and increased biases by 6–8 % at 652 nm.

For the M2014 (B1999 parameterisation) correction scheme, the range of TAP biases across all aerosol sources was 1.04–1.26 and for the M2014 (V2010 parameterisation), the range of TAP biases was 1.01–1.18. The M2014 (V2010 parameterisation) scheme reduced TAP biases relative to the B1999 and V2010 schemes by 7–38 % and 7–25 %, respectively, dependent on the aerosol source and wavelength. The most significant reductions in TAP biases were for urban aerosol emissions and had the most impact on measurements at 652 nm. As discussed in Sect. 2.1.3, the M2014 (V2010 parameterisation) correction scheme applied here used the wavelength-dependent Virkkula (2010) parameterisation, in contrast to Müller et al. (2014), who applied the Virkkula et al. (2005) parameterisation. Although not shown, applying the Virkkula et al. (2005) parameterisation to TAP data in this study would act to decrease TAP biases by 3–4 % at 467 nm, increase biases by 1–2 % at 528 nm and by 3 % at 652 nm.

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The R_{abs} from Figures 3–5 provide the mean TAP absorption coefficient biases for all measurements corresponding to each measurement wavelength and aerosol source, but it is pertinent to examine the range of biases corresponding to individual 30-s average measurements. Examining the 10th and 90th percentiles of each dataset (see Table 2) revealed that 10 % of TAP measurements were biased by greater than 1.67–1.80, 1.46–1.70 and 1.39–1.42 for urban, fresh BBA and aged BBA when corrected using the B1999 scheme, respectively, dependent on wavelength. The M2014 (V2010 parameterisation) scheme reduced the biases with 10 % of measurements biased greater than 1.27–1.41, 1.20–1.30 and 1.18–1.29 for urban, fresh BBA and aged BBA, respectively, dependent on wavelength.

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An analysis of the dependence of TAP bias as a function of filter loading revealed no point-by-point dependence but potentially a weak signal in the large-scale mean such that the difference in absolute filter transmittance associated with the highest 10 % of TAP biases compared to the lowest 10 % of biases across all channels and wavelengths was up to 0.12. The filter transmittance changed over the course of a flight by a maximum of 0.21.

The TAP biases exhibited a strong wavelength dependence. In general, the lowest biases were seen at 652 nm and the largest biases at 467 nm when the V2010 and M2014 (V2010 parameterisation) schemes were applied to TAP measurements for all aerosol sources. The exceptions to this trend were when the M2014 scheme (V2010 parameterisation) was applied to urban aerosol measurements, which led to the largest biases at wavelength 528 nm. The M2014 scheme (B1999 parameterisation) also led to the largest biases at 528 nm for all aerosol types.

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As highlighted in the introduction, filter-based absorption photometers are sensitive to the particle penetration depth, which is dependent on particle size. Indeed, this sensitivity may have contributed in part to the variation in TAP biases observed for the three types of aerosol investigated during this study.

Perhaps the most important and robust observation is that the M2014 scheme consistently led to the lowest biases across all measurement wavelengths and aerosol sources investigated. The largest biases were associated with TAP measurements corrected using the B1999 scheme at wavelengths 528 and 652 nm and when using the V2010 scheme at wavelength 467 nm for all aerosol sources.

3.2 Evaluating TAP biases as a function of the organic aerosol mass concentration

The biases of 1–45 % observed in this study are at the lower end of those measured by Lack et al. (2008) and Cappa et al. (2008), who reported biases of 12 % to ~200 % dependent upon the OA concentration. To investigate this apparent discrepancy, we evaluated the TAP biases as a function of the OA mass concentration measured using an Aerodyne Aerosol Time of Flight Mass Spectrometer (TOF-AMS, Aerodyne Research Inc.) (e.g. Drewnick et al., 2005).

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Figure 6 (a–c) shows how TAP biases vary with OA mass concentration for TAP measurements corrected using the B1999 correction scheme, for direct comparison with the Lack et al. (2008) study. The linear relationship between the PSAP biases and OA observed by Lack et al. (2008) is superimposed for reference. For urban emissions (Fig. 6a), TAP biases and OA mass are positively correlated and the trend is broadly consistent with that observed by Lack et al. (2008). There is however no correlation for fresh (Fig. 6b) or aged BBA (Fig. 6c).

TAP biases were also plotted as a function of the ratio of the mass concentrations of OA to light-absorbing carbon (LAC), denoted by $R_{OA/LAC}$. This was calculated using the method outlined by Lack et al. (2008) by (i) assuming all absorbing mass was black carbon, (ii) converting the mass absorption coefficient (MAC) of black carbon (BC) at 532 nm (7.75 m^{2}g^{-1}) to the PAS measurement wavelength 528 nm by using a BC AAE of 1 and the method outlined by Moosmüller et al. (2011) and (iii) dividing the PAS-measured absorption coefficient at wavelength 528 nm by the BC MAC at 528 nm. Hence the mass concentration of LAC was calculated as $LAC = {b^{PAS}_{abs,528\,nm}}/{MAC^{BC}_{528\,nm}}$ such that $R_{OA/LAC} = {OA}/{LAC}$ (Bond and Bergstrom, 2006; Lack et al., 2008). Figure 6(d) shows that the TAP bias is positively correlated with $R_{OA/LAC}$ for urban aerosol emissions when TAP measurements were corrected using the B1999 correction. This is consistent with the Lack et al. (2008) observation although our study shows lower biases. A likely contributor to this difference is that, for consistency with the Lack et al. (2008) study, this analysis assumed all absorption was due to BC. In reality this is a poor assumption for BBA emissions (e.g. Andreae and Gelencsér, 2006) and provides a maximum bound on the MAC value, a minimum bound on absorption attributed to LAC and therefore a maximum bound on $R_{OA/LAC}$. A more realistic approach would be to use the MAC value corresponding to BC plus BrC. Using a lower MAC to account for absorption contributions from both BC and BrC would lead to smaller $R_{OA/LAC}$ values than those shown in Fig. 6 (d–f) and better agreement with the Lack et al. (2008) study. Correcting the TAP data using the M2014 (V2010 parameterisation) correction scheme reduces the positive correlation between TAP biases and both R_{OA} and $R_{OA/LAC}$ as shown in Fig. 6 (g–i). This further demonstrates the improvement provided by using the M2014 scheme.

This analysis was repeated at wavelengths of 467 nm and 652 nm. For measurements at 652 nm, where BrC absorbs relatively weakly (e.g. Andreae and Gelencsér, 2006), stronger correlations between TAP biases and R_{OA} and $R_{OA/LAC}$ were seen compared to 528 nm measurements. This improved the agreement with Lack et al. (2008). For measurements at 467 nm, where BrC absorbs relatively strongly, weaker correlations between TAP biases and $R_{OA/LAC}$ were seen compared to 528 nm measurements. This reduced the agreement with Lack et al. (2008) for reasons described above. As for observations at 528 nm, TAP biases showed little dependence on R_{OA} and $R_{OA/LAC}$ when corrected using the M2014 scheme at 652 nm and 467 nm. This finding suggests that the source of discrepancy between the results presented in this study and the results of Lack et al. (2008) (i.e. Fig. 6) may be caused by the less advanced correction scheme applied to the Lack et al (2008) data. However, given the strong dependence of R_{abs} on the aerosol type and source in Fig. 6, the bias dependence on organic fraction in the Lack et al. (2008) data may well persist, independent of the correction scheme used, because of the different aerosol sources and source locations being studied.

A key result of this analysis is to show that biases observed in filter-based aerosol absorption measurements are strongly dependent on the type of aerosol being sampled. Correlating biases to aerosol composition information may provide tight constraint for a single source study, such as that observed by Lack et al. (2008) for aerosol emissions over the Gulf of Mexico, but care must be taken when applying these findings more broadly to other aerosol types.

3.3 Impact of TAP biases on climate relevant parameters

We now assess the impact that the observed TAP biases may have on climate relevant parameters including the aerosol single scattering albedo and absorption Ångström exponent. Figure 7 shows histograms of the SSA derived using PAS or TAP absorption data together with CRDS extinction data for the aerosol sources described in Sect. 2.3.6 and for the TAP corrections described in Sect. 2.1.1–2.1.3. The SSA is biased towards lower values when derived using TAP measurements, consistent with the results in Fig. 3–5 which typically show a ~1–45 % high bias in absorption. Campaign-mean SSA values derived using PAS and CRDS measurements for each measurement campaign are summarised in Table 3. The mean SSA values derived using TAP and CRDS measurements matched those derived using PAS measurements most closely for fresh BBA, which were biased low by 0.00–0.03, dependent on measurement wavelength and the TAP correction scheme applied. The SSA values were most different for urban aerosols, which were biased low by 0.01–0.07, dependent on wavelength and the TAP correction scheme applied. This is consistent with the results in Table 2, which highlights that TAP biases were largest for urban aerosol measurements. The wavelength dependence of the TAP-derived SSA values depended on the correction scheme applied. SSA values derived using the M2014 correction scheme agreed most closely with those derived using PAS measurements for all measurement wavelengths and correction schemes.

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Similarly, Fig. 8 shows histograms of the AAE values derived by performing linear regressions between the logarithms of the PAS-measured absorption coefficients and the PAS measurement wavelengths (405–658 nm) (Moosmüller et al., 2011). It also shows the same information for the TAP-derived AAE values. The AAE values were calculated for the aerosol sources outlined in Sect. 2.3.6 and TAP correction schemes outlined in Sect. 2.1.1–2.1.3.

The AAE values were strongly dependent on the TAP correction scheme applied. Campaign-mean AAE values are summarised in Table 4, which highlights that the highest mean AAE values were associated with fresh BBA emissions and the lowest for aged BBA emissions. TAP-derived AAE values were in absolute error by ± 0.54. The M2014 (B1999 parameterisation) led to mean AAE values that were in closest agreement with AAE values derived using PAS measurements for all aerosol types. The V2010 scheme led to mean AAE values that were in second-closest agreement with the AAE values derived using PAS measurements for urban aerosols, whereas the M2014 (V2010 parameterisation) scheme provided the second-closest match for fresh BBA and the B1999 scheme for aged BBA. It is unclear why the different TAP correction schemes perform so differently for the different aerosol sources sampled. However, what is clear from this analysis is that there are large uncertainties in this important parameter when calculated from filter-based absorption measurements, and that these uncertainties are strongly source and correction scheme dependent. This cautions that significant uncertainties could be introduced if using the AAE to differentiate between types of aerosol.

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4 Conclusions

Measurement artefacts in a commercially available filter-based absorption photometer (TAP) were evaluated as a function of wavelength and aerosol source. A range of correction schemes have been proposed in the literature to account for these artefacts and thus to maximise the accuracy of aerosol absorption coefficients determined using this technique, although biases can remain. Three correction schemes were evaluated, which all reduced the TAP mean bias to within 1 to +45 % of the PAS absorption, dependent upon aerosol source and wavelength. The largest biases were associated with urban aerosols and the lowest for aged BBA. The M2014 correction scheme consistently led to the lowest biases across all wavelengths and aerosol sources. To our knowledge, this is the first study to demonstrate the improved performance of the M2014 correction scheme as a function of wavelength and across multiple aerosol sources for ambient aerosol sampling.

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Biases in filter-based absorption measurements were strongly source dependent. On no occasion were the very large biases of over 200 % noted in the Lack et al. (2008) study observed. However, we note that the aerosol types measured in the Lack et al. (2008) study were very different to those studied here, and therefore this result may well be consistent with the strong source dependence observed in the current study.

The positive bias in filter-based absorption measurements resulted in a low bias in determinations of single scattering albedos of up to 0.07. The largest biases in SSA values were for urban aerosol measurements at wavelength 652 nm. The M2014 scheme consistently led to SSA values that were closest to those derived using PAS measurements across all wavelengths and aerosol sources.

Large discrepancies were seen between AAE values derived from PAS versus TAP measurements, the latter depending strongly on the correction scheme applied. The largest discrepancies in AAE values were for TAP measurements of urban aerosols corrected using the B1999 scheme, which were biased low by a mean absolute value of 0.54. Best agreement with AAE values derived using PAS measurements was obtained when TAP measurements were corrected using the M2014 (B1999 parameterisation) correction scheme and when (i) urban aerosol measurements were corrected using the V2010 scheme, (ii) fresh BBA measurements were corrected using the M2014 scheme and (iii) aged BBA measurements were corrected using the B1999 scheme. This highlights that the AAE is strongly source and correction scheme dependent.

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The strong aerosol source dependence of biases observed in this study cautions against extrapolating results more widely to other aerosol types. Further analyses exploring biases in filter-based absorption coefficient measurements may help to address this issue. However, given the empirical nature of filter-based correction schemes and strong source and wavelength dependencies, even this is unlikely to fully bound uncertainties associated with filter-based absorption measurements to the high level of confidence that can be achieved using alternative methods, such as photoacoustic spectroscopy.

Data availability. For data related to this paper please contact Justin Langridge (justin.langridge@metoffice.gov.uk).

Competing interests. The authors declare that they have no conflict of interest.

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	467 nm	**530 nm**	**660 nm**	**Ave**
k_0	0.377	0.358	0.352	0.362
k_1	−0.640	−0.640	−0.674	−0.651
h_0	1.16	1.17	1.14	1.159
h_1	−0.63	−0.71	−0.72	−0.687
s	0.015	0.017	0.022	0.018

Table 1: The values of the constants used in the Virkkula (2010) correction scheme (Virkkula, 2010).

Aerosol source	Wavelength	B1999				V2010				M2014 (B1999 parameterisation)				M2014 (V2010 parameterisation)			
		Slope	R^2	P_{10}	P_{90}	Slope	R^2	P_{10}	P_{90}	Slope	R^2	P_{10}	P_{90}	Slope	R^2	P_{10}	P_{90}
Urban	467	1.35	0.88	0.99	1.67	1.38	0.87	0.99	1.73	1.19	0.90	0.93	1.41	1.17	0.91	0.92	1.39
	528	1.45	0.89	1.12	1.80	1.37	0.88	1.04	1.70	1.26	0.90	1.00	1.50	1.18	0.91	0.95	1.40
	652	1.40	0.67	1.17	1.76	1.27	0.69	1.04	1.58	1.11	0.68	0.88	1.38	1.02	0.68	0.82	1.27
Fresh BBA	467	1.25	0.97	1.11	1.46	1.30	0.97	1.13	1.54	1.05	0.95	0.70	1.23	1.13	0.95	0.77	1.30
	528	1.30	0.97	1.17	1.54	1.23	0.97	1.08	1.44	1.10	0.95	0.74	1.26	1.11	0.96	0.76	1.25
	652	1.24	0.96	1.19	1.70	1.09	0.97	0.92	1.32	1.04	0.95	0.67	1.24	1.01	0.95	0.66	1.20
Aged BBA	467	1.18	0.99	1.10	1.39	1.21	0.99	1.11	1.42	1.06	0.98	0.97	1.27	1.11	0.98	0.99	1.29
	528	1.21	0.99	1.12	1.42	1.16	0.99	1.05	1.35	1.09	0.99	0.99	1.30	1.07	0.98	0.95	1.26
	652	1.18	0.99	1.11	1.41	1.08	0.99	1.00	1.28	1.05	0.99	0.94	1.24	1.01	0.99	0.89	1.18

Table 2: A summary of the slopes (R_{abs}) between PAS and TAP absorption coefficients. Correlation coefficients (R^2) are also provided. P_{10} and P_{90} are the 10th and 90th percentiles of each dataset. All absorption coefficients correspond to > 1 Mm^{-1}. All linear regressions were forced through the origin.

Aerosol source	Wavelength	Mean SSA				
		PAS	B1999	V2010	M2014 (B1999 parameterisation)	M2014 (V2010 parameterisation)
Urban	467	0.89	0.86	0.86	0.87	0.87
	528	0.88	0.84	0.85	0.86	0.87
	652	0.88	0.81	0.83	0.86	0.87
Fresh BBA	467	0.92	0.89	0.89	0.91	0.91
	528	0.93	0.90	0.91	0.92	0.92
	652	0.93	0.91	0.93	0.93	0.93
Aged BBA	467	0.84	0.80	0.80	0.82	0.81
	528	0.83	0.79	0.80	0.81	0.81
	652	0.81	0.77	0.79	0.80	0.81

Table 3: Campaign-mean single scattering albedo (SSA) derived using PAS and CRDS measurements and TAP and CRDS measurements.

Aerosol source	**Mean AAE**				
	PAS	**B1999**	**V2010**	**M2014 (B1999 parameterisation)**	**M2014 (V2010 parameterisation)**
Urban	1.51	0.97	1.35	1.54	1.74
Fresh BBA	1.91	1.50	2.27	1.97	2.22
Aged BBA	1.06	0.99	1.32	1.14	1.36

Table 4: Campaign-mean absorption Ångström exponent (AAE) derived using PAS and TAP measurements.

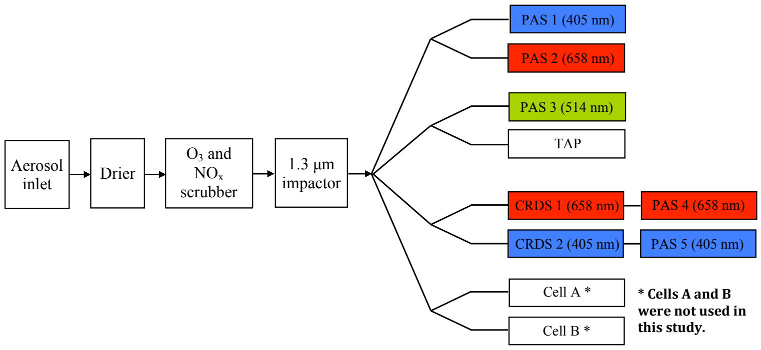

Figure 1: Schematic diagram highlighting the flow conditioning and how the aerosol-laden stream was distributed between the PAS and CRDS cells and the TAP. All PAS and CRDS wavelengths were centred at 405, 514 and 658 nm respectively.

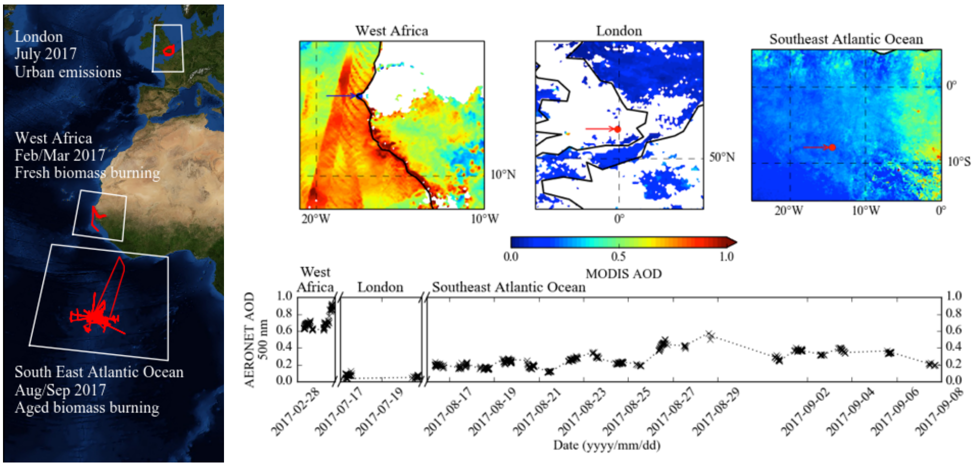

Figure 2: FAAM research aircraft flight tracks (red) over London in the United Kingdom (July 2017), West Africa (February and March 2017) and the Southeast Atlantic (August and September 2017). For each of the geographical areas highlighted in the white boxes, the mean aerosol optical depths (AODs) measured using the Moderate Resolution Imaging Spectroradiometer (MODIS) satellite instruments are displayed. A time series of Aerosol Robotic Network (AERONET) data shows AODs at 500 nm corresponding to each measurement period. Note the discontinuous AERONET AOD time axis. AERONET sites are shown on the MODIS AOD plots by arrows.

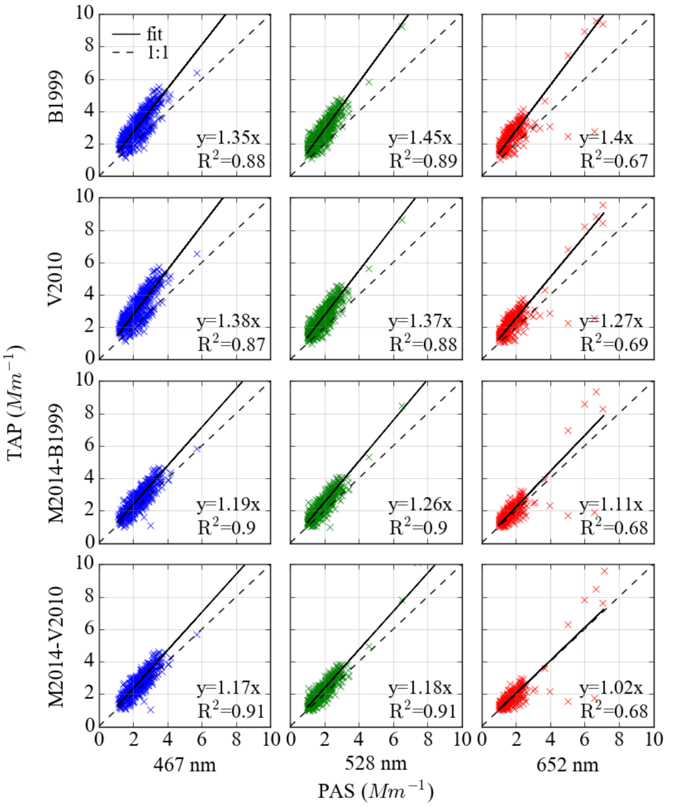

Figure 3: Absorption coefficients measured by PAS versus TAP for urban emissions around London in July 2017. The columns correspond to: column 1: 467, column 2: 528 nm, and column 3: 652 nm wavelengths and the rows correspond to the B1999, V2010 and M2014 corrections. All absorption coefficients correspond to > 1 Mm^{-1}. All linear regressions were forced through the origin.

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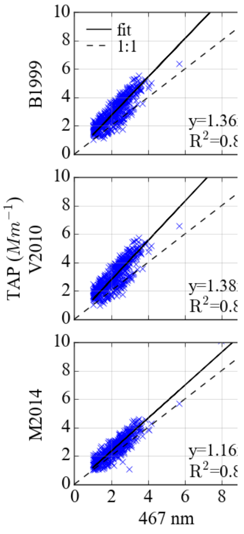

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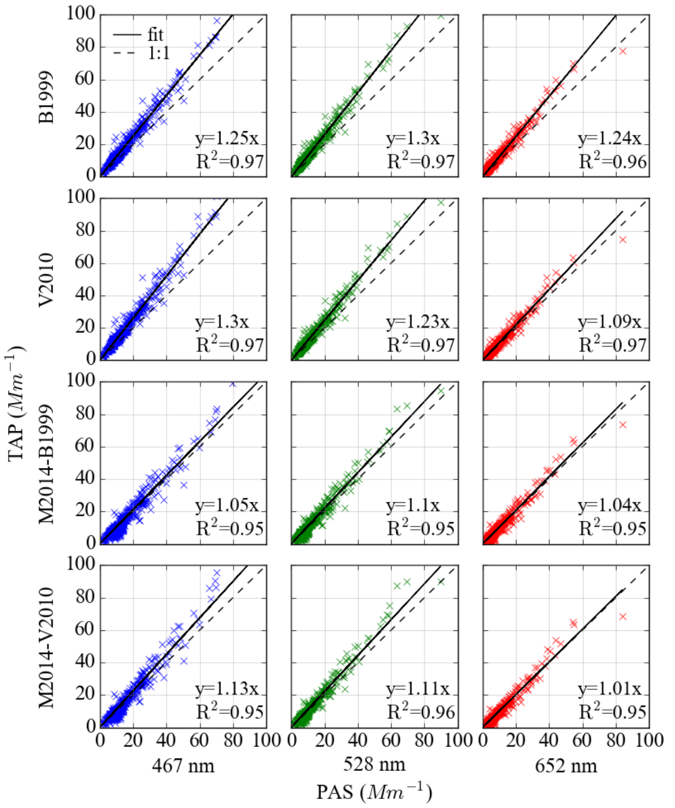
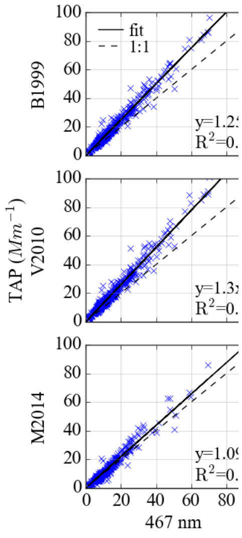


Figure 4: As Fig. 3 but for fresh biomass burning aerosol over Senegal in February and March 2017.

Nicholas Davies 9/4/2019 08:31



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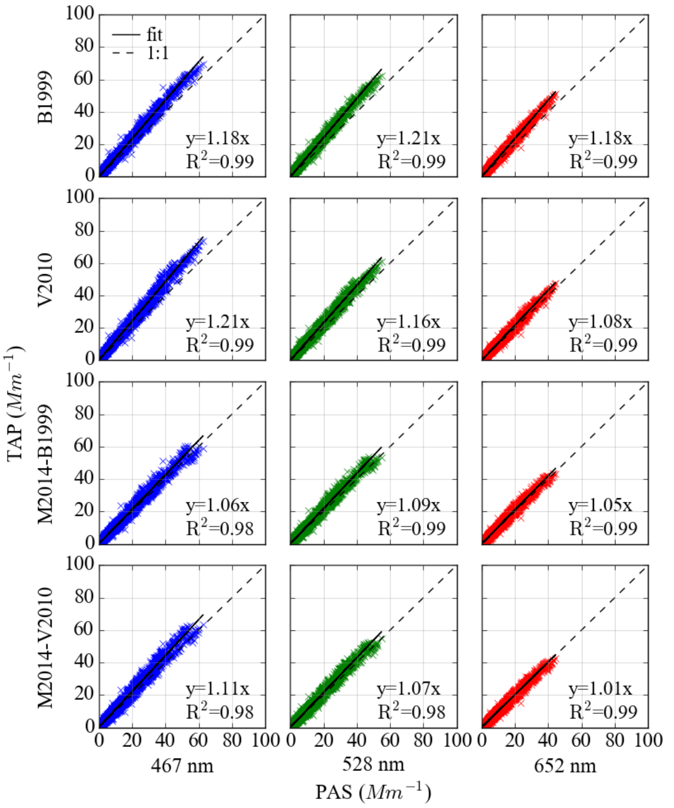

Figure 5: As Fig. 3 but for aged biomass burning aerosol over the Southeast Atlantic Ocean in August and September 2017.

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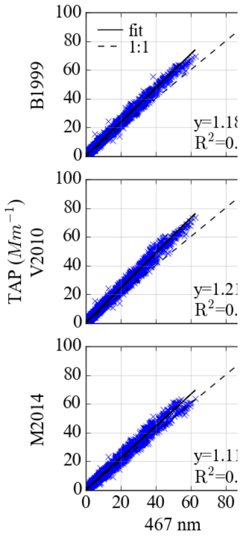

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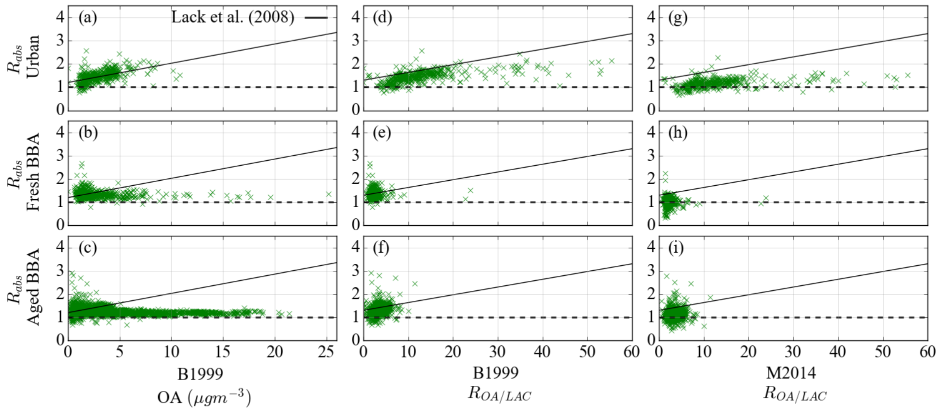

Figure 6: The ratio of TAP to PAS absorption coefficients at 528 nm as a function of the organic aerosol mass concentration using the B1999 correction scheme (a-c) and as a function of the ratio of the organic aerosol to light-absorbing carbon mass concentrations when using the B1999 correction scheme (d-f) and using the M2014 (V2010 parameterisation) correction scheme (g-i). All absorption coefficients correspond to > 1 Mm^{-1}.

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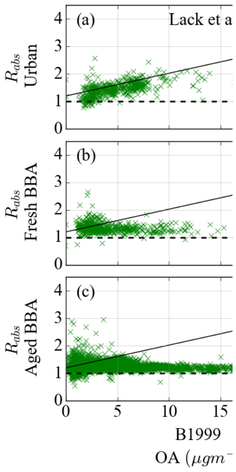

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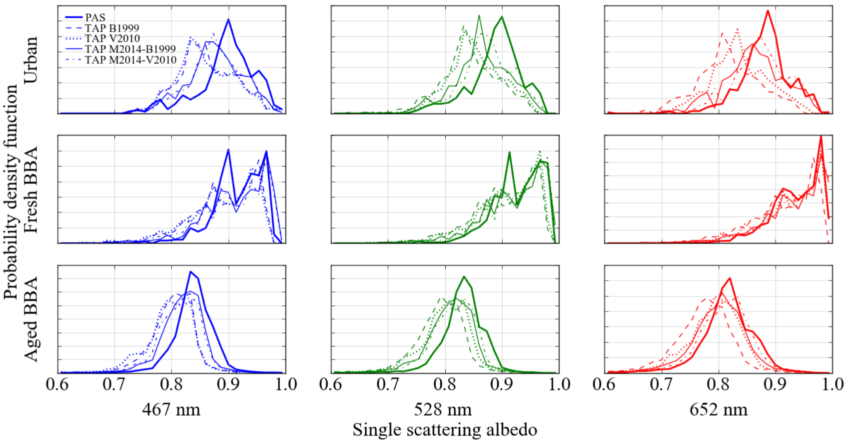

Figure 7: Probability density functions of the single scattering albedo derived using (i) PAS and CRDS and (ii) TAP and CRDS for the range of TAP correction schemes outlined in Sect. 2.1.1–2.1.3 at wavelengths 467, 528 and 652 nm. All absorption coefficients correspond to > 1 Mm^{-1}.

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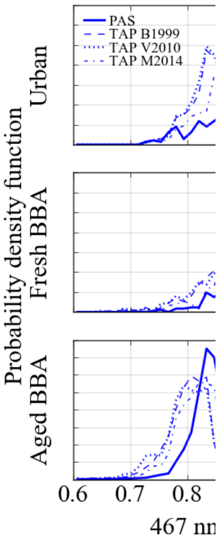

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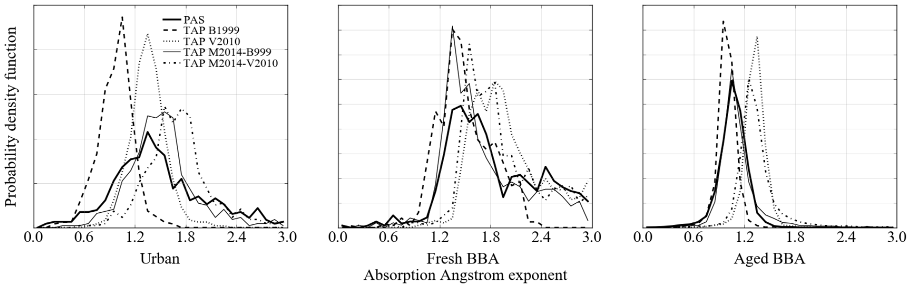

Figure 8: Probability density functions of the absorption Ångström exponents derived for PAS and TAP measurements using the range of TAP correction schemes as outlined in Sect 2.1.1–2.1.3. All absorption coefficients correspond to > 1 $\mathrm{Mm^{-1}}$.

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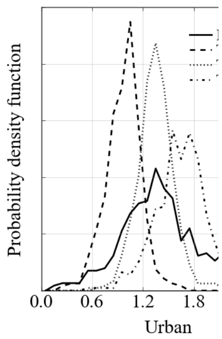

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