Response to anonymous referee #2 comments (RC2) on

A novel approach to calibrating a photo-acoustic absorption spectrometer using polydisperse absorbing aerosol (https://www.atmos-meas-tech-discuss.net/amt-2018-413/)

The authors greatly appreciate the comments, corrections, and suggestions from this anonymous referee. Please note that the individual responses for each comment below use the page and line numbers from the manuscript that was originally submitted in AMTD.

Comment #1:
Typical analyses of photoacoustic data have included the effect of the acoustic frequency, quality factor of the acoustic cavity, and laser power. Even if these quantities are constant for these experiments, the authors should include them in their formalism. Generally some more formalism such as the photoacoustic equation relating microphone signal to absorption coefficient would be a benefit.

Response to comment #1:
In the portion of the introduction that discusses photoacoustic spectrometry history and the different calibration approaches, the authors have added a discussion of the photoacoustic equation and the equation itself. These additions are

P3L1: "Theoretically, the absorption (b_{abs}) coefficient can be determined from a PAS as a function of absolute laser power (P_{Laser}), pressure at the microphone (P_{Mic}), resonator cross sectional area (A_{Res}), resonant frequency F_R, and quality factor (Q).

$$b_{abs} = \frac{P_{Mic}}{P_{Laser}} \frac{A_{Res}}{\gamma - 1} \frac{\pi^2 F_R}{Q} \quad (1)$$

For multi-pass instruments it is difficult (Fischer and Smith, 2018b) or not feasible given the instrument setup (Lack et al., 2012b) to know all of these terms accurately. This means the first principles approach of Arnott et al. (1999) is not possible for many instruments. The issue with a fundamental calibration is that the overlap integral of the laser, acoustic mode, and aerosol is not known accurately enough for calibrations. Additionally, the microphone sensitivity and laser power are not known accurately enough for calibration purposes in the design of Lack et al. (2012b)."

Comment #2:
The typical style is to leave a space between quantity and the unit. Please check this, in particular when wavelengths are reported.

Response to comment #2: The authors apologize for this error and appreciate the attention to detail from the referee. All such cases of a space missing have been found and corrected. Listed are the locations where a space needed to be added between the wavelength and unit:
p1L17;p3L21;p4L13;p6:L5,L24,L29,L31,L32;p7L10;p8L7,L29,L30;p10L2,L3,L10,L11,L12

Comment #3:
Some values and errors are reported with too many significant digits

Response to comment #3: The authors have carefully gone through the manuscript to confirm consistency of significant digits between values and errors within each analysis. Several cases of

excessive significant figures were found and have now been altered (P10L11,P10L13,P10L15,P10L31). This comment from the referee is much appreciated.

Comment #4:
The optical power in these multipass cells is not unknowable. It can be determined with few simple measurements. 1) Measure the transmission of the rear mirror. 2) place a calibrated optical power meter to measure the optical power leaking through the mirror. 3) account for the mirror transmission and a factor two for a similar amount of light leak through the front mirror to get the optical power in the acoustic cavity. The issue with a fundamental calibration is that the overlap integral of the laser, acoustic mode, and aerosol is not known accurately enough for calibrations. Or possibly the microphone sensitivity and/or the laser power are not known accurately enough for calibration purposes.

Response to comment #4:
We agree that the issues were oversimplified in the AMTD manuscript. We have adopted to reviewer's suggestion and the passage on P3L1 now reads: "For multi-pass instruments it is difficult (Fischer and Smith, 2018b) or not feasible given the instrument setup (Lack et al., 2012b) to know all of these terms accurately. This means the first principles approach of Arnott et al. (1999) is not possible for many instruments. The issue with a fundamental calibration is that the overlap integral of the laser, acoustic mode, and aerosol is not known accurately enough for calibrations. Additionally, the microphone sensitivity and laser power are not known accurately enough for calibration purposes in the design of Lack et al. (2012b)."

Comment #5:
P3L23: 'area' should be 'are'

Response to comment #5: corrected

Comment #6:
P4L11: extra space before "Lack"

Response to comment #6: corrected

Comment #7:
P4L19: Because this acoustic cavity consists of two high coupled resonators, there are two "primary" eigenmodes. In one mode the pressures at center of both resonators are in-phase and in the other the pressures are 180 deg. out of phase. Please rewrite the sentence to clarify which mode is being used.

Response to comment #7: Thank you for this input, this sentence has been reworded to explain that in our instrument, the antinodes are at the center of each cell and are 180 degrees out of phase.

P4L19: "The primary eigenmode of this instrument consists of one full wavelength across the two cells, such that the antinodes are at the center of each cell and 180 degrees out of phase."

Comment #8:
P5L4: either "each cell's resonant frequency" or "each cells' resonant frequencies" would be appropriate.

Response to comment #8: We appreciate this attention to detail. Pertaining to both this comment and comment #9, this sentence now reads:

"To obtain the maximum and consistent signal the lasers must be modulated at each cell's resonant frequency, which is dependent on temperature and pressure in the cell. Accordingly, a resonant frequency calibration is performed at regular intervals (typically at least every 5 minutes), to account for any drifts in temperature or pressure."

Comment #9:
P5L5: please replace "whatever interval" with a more formal phrase

Response to comment #9: Thank you, the sentence has been corrected with "a regular interval", and we clarify that we have not set a specific timing, but this frequency calibration is done at least every 5 minutes. See above for the new sentence.

Comment #10:
P5L6: *The microphone part number does not need to be repeated here.*

Response to comment #10: corrected

Comment #11:
P5L15: track changes indicator on this line

Response to comment #11: corrected, thank you

Comment #12:
P6L2-6: If known please state the concentration of solution that is used. Even if it is not critical, it is a good starting point for future projects and replication.

Response to comment #12: The starting amounts of Nigrosin/Regal Black/Aquadag were not recorded due to the variability between tests and the need to add substances or serially dilute. However, we have added an indication of the amount of substance that was initially added and the amount of water that was initially used.

P6L2: "A few crystals (solids) or a quarter spatula (slurry), of the given substance is mixed with Milli-Q water (Millipore system SimPak2) and progressively diluted (starting with a couple hundred ml of water) until the size distribution of the atomized aerosols is such that 99% of the mass is below 300 nm. More dilute solutions tend to yield aerosol with smaller sizes."

Comment #13:
P6L21: in this sentence please replace the 'extinction' with 'extinction channel'

Response to comment #13: corrected

P6L30: "The scattering channel for the CAPS PM_{SSA} is calibrated relative to the extinction channel, because the extinction does not require calibration."

Comment #14:
P7L25-30: Nigrosin has a complex absorption spectrum and is not appropriately modeled with an angstrom exponent model.

Response to comment #14: The authors agree that Nigrosin does not follow the relationship between wavelength and absorption modeled by an angstrom exponent. The discussion in section 3.2 has been modified to add additional discussion of the issues with Nigrosin. Given the small difference in wavelength between 405 and 450 nm, it is still reasonable to assess the error in the AAE method of calibrating with Nigrosin. Of the three substances, we demonstrate that Nigrosin has the least reliability for calibration, partially due to this complex relationship between absorption and wavelength. Modifications to the text begin on p7,L29:

"Additionally, the Nigrosin tested here yielded a negative AAE, which is inconsistent with figure 4 of Bluvshtein et al., (2017) in the wavelength range of 400-450 nm that shows a positive AAE. Nigrosin has been shown to have an index of refraction that significantly varies across the visible wavelengths (Bluvshtein et al., 2017), and does not have a relationship between absorption and wavelength that is appropriately modeled by AAE.. However, given that the adjustment is only over a small wavelength range (11% difference in wavelength between 450 nm and 405 nm), the error introduced by adjusting absorption measurements from 450 to 405 nm with the AAE technique is assessed here."

Comment #15:
P8L17: Use of the Allan deviation is common in the atmospheric community where it is implicitly presented as a detection limit as a function of averaging time. The Allan deviation is useful for identifying drifts, but detection limits and instrument stability are more accurately characterized using the standard deviation which can also be presented as a function of averaging time. I suggest the authors use the standard deviation instead of the Allan deviation.

Response to comment #15: We believe it is important to keep figure 4, the Allan deviation as a function of averaging time, in this paper so as to provide comparison between different instruments and to demonstrate the stability of the instrument. However, based on the comment, we also include a table in the supplement that demonstrates the standard deviation for each cell as a function of averaging time for 1 second, 30 second, and 60 second data. Additionally, the range of standard deviations measured for the four cells is now presented in the main body of the text. Added is Table 1 in the supplement, and the following text, beginning on P8L21:

"As an alternative noise assessment to the Allan deviation, the standard deviation of 1, 30, and 60 second average data are listed for all cells in Table S1 in the supplementary material. The 1 second data standard deviation varies between channels, from 0.01 to 0.12 Mm^{-1}, while at 30 seconds the range is 0.004 to 0.02 Mm^{-1}, and 60 seconds averaging has little change from 30 seconds."

cell	1 second data (Mm^{-1})	30 second average (Mm^{-1})	60 second average (Mm^{-1})
405 dry	.0092	.0039	.0035
660 dry	.0687	.0321	.0308
405 den	.0250	.0311	.0225
660 den	.1160	.0196	.0199

Comment #16:

P9L10: Please remove one of the periods.

Response to comment #16: corrected

Comment #17:
P9L6: This manuscript should assess the overall measurement accuracy for ambient measurements. This section concludes that the accuracy of these calibrations is roughly +/- 6%. Is this the expected overall accuracy for ambient measurements? If yes, please state that explicitly. If no, please explain why.

Response to comment #17: Yes, this is the expected measurement accuracy based on the calibration. However, the measurement accuracy could also be affected by baseline drifts or abnormally large noise in various situations (aircraft, mobile, etc.). Given this, we only state the accuracy of the calibration, not the accuracy of the actual ambient measurements, which will have to be assessed for every measurement campaign.

Comment #18:
P10L23: please replace '3' with 'three'

Response to comment #18: corrected, thank you

Comment #19:
P11L4: There seem to be a few problems with some of the references. I suggest the authors look over all them carefully. P11L24: Both AMT and AMTD versions of Bluvshtein are in the reference list P12L2: "K??rcher" P12L21: no journal listed P13L19&22: Initials for Lack should capitalized. P13L31: remove "(Julie)" P15L21: No Journal listed

Response to comment #19: thank you for this attention to detail, the full reference list has been worked over and corrected, with the specific problems addressed. The author also found an additional error and made the appropriate fix.

Comment #20:
Figure 1: Could you re-arrange this figure so the flow goes from left to right. Maybe add arrows.

Response to comment #20: Figure 1 has now been re-arranged so that flow goes from left to right, and arrows have been added to show the direction of flow.

Comment #21:
Figure 2: Please change the vertical axis to 'nominal scattering' to indicate it is uncalibrated.

Response to comment #21: The authors thank the referee for this helpful suggestion, and agree that 'nominal scattering' is more correct for this figure, given that the calibration slope has to be applied for true scattering. The title on the vertical axis of figure 2 has been changed.