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Atmospheric Measurement Techniques Discussions

Interactive comment on "A single-beam photothermal interferometer for in-situ measurements of aerosol light absorption" *by* Bradley Visser et al.

Anonymous Referee #3

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Manuscript: A single-beam photothermal interferometer for in-situ measurement of aerosol light absorption (Visser et al.,)

Quantifying aerosol light absorption with high precision and accuracy remains an elusive but important need the climate change community trying to quantify the contribution of aerosols to the Earth's radiation budget. Measurement of light absorption is typically conducted using filter-based techniques which offer very high precision but are greatly hindered in accuracy due, in part, to their well-known measurement bias' which cannot be completely removed. Thus there exists a need to develop in situ techniques that can directly measure light absorption. Photothermal interferome-

try (PTI), like it photoacoustic cousin, is one class of measurement methodology can addresses this need by directly measuring aerosol light absorption through the dissipation of spectrally-absorbed energy by a particle. Another important hallmark of these photo thermal approaches is that their complete insensitivity to light scattering, which is a considerable advantage given that optical extinction by particles is dominated by scattering, save, of course, pure soot (e.g., black carbon).

The manuscript by Visser et al., describes a novel single-beam photothermal interferometer - called the modulated single beam PTI (or MSPTI) - to directly measure aerosol light absorption. The unique and clever wrinkle of the MSPTI is the single beam design that serves as both the interferometer (probe) laser AND as the excitation (pump) laser. Compared to its two-beam brethren - which uses a dedicated interferometer laser (typically a single-frequency HeNe laser) and a separate pump laser (405 nm, 532 nm, or 670 nm being typical wavelengths of interest) the single-beam configuration offers the significant improvement in the ease of instrument laser alignment. An additional advantage of the single-beam interferometer is the ability to remove, in realtime, signal contributions by light absorbing molecules by having the reference arm sample filter-free air. In contrast, two-beam PTIs require periodic acquisition of particle-free conditions via HEPA-filtered air to obtain a measure of the molecular contribution. Another unique advance in the Visser et al. design is the use of a pressure cell to help maintain phase quadrature lock as this feedback mechanism eliminates moving parts within the instrument thereby improving instrument robustness.

While the performance metrics for this "proof-of-principle" version of the MSPTI currently relegate its immediate utility to long-term, fixed ground-based measurements (for a 60-second integration a lower absorption detection limit ∼7.5 Mmˆ-1 or, assuming a mass absorption cross-section of 10 mˆ2/g, an equivalent black carbon (eBC) concentration of 0.75 ug/mˆ3), the instrument is a worthy addition to the measurement quiver in the aerosol direct effects community. With a couple of exceptions noted below, this manuscript is clearly written and requires only minor revisions. It is therefore

recommended that this manuscript be published.

Page 6, line 11. What was the criteria for selecting I_low and I_high? All that is stated is that the laser "is modulated between two sufficiently different intensity levels." Certainly there is a lower limit below which the ability to lock on to quadrature would be compromised. On the other hand, the larger the difference, would favor signal detection.

Page 8, lines 8 and 9: the use of "solid" and "external" noise sources is not very descriptive. Why not call this noise sources what they actually are: mechanical (vibrational) noise and acoustic noise. For those unfamiliar with PTI or, more generally interferometry, referring to a noise source as "solid" or "external" is a bit nebulous.

Page 8, Lines 15-17. The MSPTI utilizes a reference channel that samples filtered air - a necessary condition for the single-beam configuration to work. What is the impact of a sample containing a mixture of light absorbing and non-light absorbing particles at high concentrations, as might be encountered in a biomass burning event, where the refractive index (RI) of the particles could contribute to the sample ensemble RI but whose contributions would not be present in the particle filtered sample? What are thermal lensing implications under these conditions? In a two-beam PTI, the sample and reference arms probe the same particle-laden air simultaneously thereby enabling common mode rejection for such conditions.

Page 10, line 4. The authors are encouraged to cite Lack et al. (2006) here. This paper is already listed in their citations.

Page 11, line 16. The authors are encouraged to merge Figures 5 and 8. In a lot of ways, Figure 8 is far more informative as it beautifully captures how decreasing the modulation frequency - increasing the heating period - brings about significant departure from linearity due to energy diffusion outside the probe region.

Page 11, Line 22. The sentence "If the deviation from linearity of the PTI signal with

heating time due to heat loss out of the measurement volume could be excluded from the measurement, then both the measured signal and, by extension, the sensitivity determined from calibration measurements would be considerably higher." This is a very awkwardly worded sentence. I believe that the authors simply trying to say that the non-linear signal due to diffusional loss of heat outside the probe volume suppresses measurement of the total amount of energy deposited into the system. If so, please clarify. [As an aside from purely physics interest, this raises an interesting question with respect to a two-beam PTI: if the probe volume was configured to be slightly larger than the pump volume would this enable the 2-beam configuration to "delay" the onset of the departure from linearly and, in so doing, improve performance at lower modulation frequencies?]

Page 11, line 36. It seems to this reviewer that the two time series traces should be switched. Ideally, the authors should first show that their system can indeed detect NO2 (currently the right most trace) and THEN show how well their system does at removing the NO2 signal (currently the left most trace). The actual time stamps is immaterial here. This is a stylistic comment.

Page 12, line 2: The authors are reminded that there are chemical "denuders" for removing molecular species such as NO2 via MnO2.

Page 12, Comparison with Aethalometer. Philosophically, this reviewer has major concerns about the underlying assumption of a constant mass absorption cross-section (MAC) for black carbon (BC) in order to report an equivalent black carbon (eBC) concentration. There are a plethora of studies showing that the BC MAC (at 550 nm, for example) can vary from $\sim$7.5 m2/g for uncoated BC particles to 13-15 m2/g for coated particles. This reviewer understands that the Aethalometer reports a eBC value and that the authors are comparing their instrument to the Aethalometer. While this comment is well-beyond the scope of this present paper, one potential (and easy) solution that the authors might consider, is to compare absorption coefficients instead of mass concentrations - after all, this is what both instruments fundamentally measure.

Page 23, Figure 11. Is the departure observed in the variation from tˆ-1/2 due to the active quadrature lock feedback circuit? Also a more meaningful metric to the aerosol community would be an Allan variance plot of the absorption coefficient.

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