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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.

Bradley Visser et al.

bradley.visser@fhnw.ch

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The authors thank the reviewers for their time in reviewing the manuscript and their constructive questions and comments. The manuscript will most certainly be improved by implementing the suggested changes. On a personal level, the authors also very much enjoyed the high level discussion of our instrument and PTI in general.

Addressing the comments and questions of Anonymous Reviewer #1 in order:

1) Previous iterations of PTI instruments have relied upon lock-in detection for the evaluation of the PTI signal. With lock-in detection the signal is multiplied by a reference

sine wave and the amplitude component (R) is interpreted as the PTI signal. Information regarding the shape of the heating curves could potentially be extracted from the phase signal provided by the lock-in amplifier, though the authors are not aware of any PTI study of aerosols in which the phase of the PTI signal has been examined. It is assumed that the shape of the heating curve does not change with absorption. If this assumption is correct then the instrument response is linearly dependent on absorption (Sedlacek 2006, Figure 3). In the current work, the linear fit to the heating curves was employed as a simple alternative to lock-in detection. The linear fit is equivalent to lock-in detection. The measurement of the lock-in amplifier is the product (in the space of orthogonal sine functions) of the heating curve and the sine with the same frequency. If the shape of the heating curve changes, this will be observed in the amplitude of the product (and thus potentially interpreted as a change of absorption) and the measured signal phase. Therefore, unless the phase of the PTI signal with respect to the laser excitation is investigated, linearity (or at least a consistent heating curve form) is assumed and both lock-in detection and the linear fitting of the PTI data are equivalent. The authors acknowledge that a change in the shape of the heating curve would lead to a change in the proportionality of the linear and non-linear fits. It is also agreed, that this proportionality could change with a significant change in the composition of the bath gas, pressure and potentially RH, which would all result in considerably different thermal properties of the sample air stream. These effects have so far not been observed in the experiments. We aim to investigate the proportionality in a future work and examine whether additional information can be extracted from the sample.

2) This is correct. It is expected that a thermal lens forms due to the temperature distribution in the bath gas along the laser path, in particular around the laser focus and affects the paths of the laser beams through the instrument. The effect of a thermal lens would be two fold; firstly it would change the optical path length of the interferometer arm, thus influencing the phase measurement, Secondly, it would alter the size of the laser beam focus, which feeds back into the PTI measurement and the thermal lens. Experimentally, the authors have not been able to observe the effects of a thermal

lens or at least decouple this effect from, for example, the non-linearity observed in the heating curves due to loss of heat out of the measurement volume during the heating phase. In comparison to previous instruments and their response, it is believed that the energy density in the laser focus in the current study is very similar and that the effect of a thermal lens would be comparable in each case.

3) In choosing the units for presentation of the data, the authors referred to previously published works on photothermal interferometry, such as (Sedlacek 2006). Here equivalent plots are presented in terms of (fractions of) volts, which is the direct measurement unit in the case where a lock-in amplifier is used. Figure 9 presents the response of the MSPTI signal due to the addition of 1 ppm NO2 in both flow schemes. As the instrument is calibrated with NO2, the authors feel that it would be best to present this plot with two y-axes, one with the internal PTI units (rad s-1) and the other with units of absorption (Mm-1). The plot will be updated in the manuscript to add this second y-axis. In Figure 10, the standard deviation of a background measurement is presented. The y-axis of this plot will be changed in the manuscript to be in terms of absorption (Mm-1).

4) The reviewer makes a very good point. The instrument does indeed measure light absorption. References to black carbon were only meant for comparison purposes and to facilitate an easier understanding of the quantities of BC measured by presenting the measured values as a mass concentration by assuming a stable MAC. The manuscript will be changed to be more clearly framed in terms of the measurement of absorption, with eBC as a secondary parameter.

5) We have chosen the frequency based on the frequency dependence of the signal-to-noise ratio. The maximum of this function lies at 91 Hz. If the instrument were reliant on intensity modulation by changing the current applied to the laser head, then yes, even reaching 91 Hz would not have been possible. The AOM however, allows much higher modulation frequencies, even approaching 1 MHz. The non-linearity seen in the plot of PTI signal vs heating period (Figure 8) begins for heating periods above approximately

5 ms. We attempted experiments with the heating times slightly below this (modulation frequencies between 100 and 130 Hz) but encountered significantly increased noise in this frequency range due to other lab equipment. 91 Hz was subsequently chosen as it provided the optimum signal to noise ratio for the current instrument and the calibration was not required to be transferred to a different modulation frequency.

Addressing the specific comments of Anonymous Reviewer #1:

- With regards to the line 49 and the MAC not being extrapolated with AAE=1 The reviewer is correct. This line of text will be corrected in the manuscript.

- With regards to the text on lines 155-185 being a rather long explanation The authors agree with the reviewer. The text from lines 155-185 will be reworked in the manuscript to improve the readability.

- With regards to Figure 5 and dotted vs solid line Thank you to the reviewer for finding this error. The caption in the manuscript will be updated from dotted to solid line.

- With regards to line 293 and the precision of commercial beam splitters The instrument shows considerable sensitivity to non-50:50 beam splitting. A large variance has been observed for the polarisation angle. The sensitivity to angle of incidence has not been investigated as the very little variation is possible given the design of the instrument. The polarisation of the laser beam was adjusted to ensure 50:50 splitting of the beam as measured using a power meter as well as the interferometric contrast. It was found that the highest interferometric contrast was obtained when the laser beam intensity was split very close to 50:50, due to equal losses in both beam paths. Subsequent use of a 532 nm laser-line beam splitter that is much less polarisation sensitive has shown that the drift of the baseline measurement is not due to changes in the splitting ratio of the beam splitter (i.e. the splitting ratio appears to remain stable during the measurements).

- With regards to line 421 and the pressure drop across the filter Yes, the use of a filter did result in a pressure drop of around 1 mBar from the measurement chamber to the reference chamber. This was not accounted for in the volumes of the pressure chambers. In recent work we have instead changed the gas line configuration such that there is no longer a pressure difference between the sample and reference chambers.

- With regards to Figure 9 and colouring the data points differently This is a good suggestion to improve the clarity of Figure 9. The Figure will be updated in the manuscript to have different coloured points for the ramp up and down measurements.

- With regards to line 439 and the necessity of measuring the background drift The authors respectfully disagree with this statement. There are two potential sources of the background change: gas absorption and photothermal effects in the optical elements in the interferometer. Two-beam interferometers must in theory measure both changes or resort to another way of determining the concentration of gaseous species or employ e.g. a scrubber. We account for this with the reference chamber. The absolute baseline must continuously be monitored in any PTI instrument as it arises primarily from light absorption by optical elements in the interferometer, which may change over time. This type of baseline change is different to that arising from the absorption due to gaseous species and changes thereof. Two beam configurations with glancing angle type configurations are rather sturdier in this respect. Our single beam configuration features a perfect overlap between the pump and the probe beams – they are the same laser beam, but this is also true for the effects in the optical elements. Hence the need to measure the background.

- With regards to Figure 11 and the Allan deviation The authors feel that the standard deviation is more appropriate than the Allan deviation in expressing the uncertainty in PTI measurements. It is however acknowledged that presentation of the Allan deviation would better enable comparison to other measurement techniques. A plot of the Allan deviation using the same data as Figure 11 will be added to the supplementary information.

- With regards to lines 492-495 and charcoal scrubbers This is correct and a note to this effect will be added to the manuscript. It is however always advantageous to treat the aerosol as little as possible before measurement, in order to avoid changing any of its characteristics.

References Sedlacek, A., Real-time detection of ambient aerosols using photothermal interferometry: Folded Jamin interferometer, Rev. Sci. Inst., 2006, 77, 064903

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