AMT paper supplementary:

The following analysis is motivated by the large continuum optical depth observed in the high-wavenumber window at 1 µm in the 18 September Langley-derived best estimate of the continuum (Figure 7 in the main text). Here, it is supposed that there is a wavenumber-dependent offset $a(\nu)$ to the irradiance *I* observed by the spectrometer, such that:

(S1) $$I'(\nu) = a(\nu)\, I(\nu)$$

The Beer-Bouguer-Lambert law gives the irradiance observed by the spectrometer on the ground as a function of the incoming solar irradiance I_0, the airmass factor $m = \cos(\theta)$, where θ is the solar zenith angle, and the atmospheric column-integrated optical depth τ.

(S2) $$I = I_0 \exp(-m\,\tau)$$

and

(S3) $$I' = I_0 \exp(-m\,\tau')$$

Inverting both of these equations (i.e. Eq. (1) in the main manuscript):

(S4) $$\ln I = \ln I_0 - m\,\tau$$

(S5) $$\ln I' = \ln I_0 - m\,\tau'$$

Solving for τ, substituting $I' = a\,I$ and then taking the residual $\tau - \tau' = \Delta\tau$ i.e. the change in observed optical depth $\Delta\tau$ with the substitution *I* -> *I'*:

(S6) $$m\tau = \ln I_0 - \ln I$$

(S7) $$m\tau' = \ln I_0 - \ln aI = \ln I_0 - (\ln I + \ln a)$$

Then:

(S8) $$m(\tau - \tau') = \ln I_0 - \ln I - \ln I_0 + \ln I + \ln a$$

(S9) $$\Delta\tau = \frac{\ln a}{m}$$

Assuming *a* = 1.1 (i.e. I' is a factor of 10% larger than I), then for an observation taken at an airmass of ~1.5 the optical depth correction is ~ 0.06, which is a very large change considering a typical continuum optical depth in the windows is ~0.03. This has a potentially large impact on measurements using the radiative closure method.

However, in the Langley method, observations are taken at multiple airmasses. Any offset would therefore need to affect the gradient of the Langley slope, i.e. change each observation differently, in a way which propagates through to the logarithmic space in which a Langley plot is taken.

The Langley-derived optical depth τ is the gradient of the straight line:

(S10) $$\ln I = \ln I_0 - m\,\tau$$

With multiple observations, this is computed using the ordinary least squares method; the gradient in which has the general form (dropping the subscript of the summation *i* on each variable):

(S11) $$\beta_1 = \frac{n\Sigma xy - \Sigma x \Sigma y}{n\,\Sigma x^2 - (\Sigma x)^2}$$

Again, the offset $\tau' - \tau = -\Delta\tau$ is the target of interest here. Taking S10 as a linear equation of the form *y* = *ax* + *b* and substituting (S10) into (S11):

(S12) $$\tau = \frac{n\Sigma\,(\mathrm{m}\ln I) - \Sigma m\,\Sigma(\ln I)}{n\,\Sigma m^2 - (\Sigma m)^2}$$

(S13) $$\tau' = \frac{n\Sigma\,(\mathrm{m}\ln aI) - \Sigma m\,\Sigma(\ln aI)}{n\,\Sigma m^2 - (\Sigma m)^2}$$

$$= \frac{(n\Sigma(m\ln I) + n\,\Sigma(m\ln a))\ - \Sigma m\,\Sigma(\ln a) - \Sigma m\,\Sigma(\ln I)}{n\,\Sigma m^2 - (\Sigma m)^2}$$

Then subtracting τ' from τ:

(S14) $$\Delta\tau = \frac{n\Sigma\,(\mathrm{m}\ln I) - \Sigma m\,\Sigma(\ln I)\ - n\Sigma(m\ln I) - n\Sigma(m\ln a) + \Sigma m\,\Sigma(\ln a) + \Sigma m\,\Sigma(\ln I)}{n\,\Sigma m^2 - (\Sigma m)^2}$$

Several of these terms then cancel, leaving:

(S15) $$\Delta\tau = \frac{-\,n\Sigma(m\ln a) + \Sigma m\,\Sigma(\ln a)}{n\,\Sigma m^2 - (\Sigma m)^2}$$

Assuming that the adjustment factor is *angle independent*, i.e. the correction does not change at all with airmass allows us to take $\ln a$ out of the summation over *m,* as it is constant. Additionally, the summation over $\ln a$ simply gives $n\ln a$. Therefore:

(S16) $$\Delta\tau = \frac{-(n\ln a)\,\Sigma m + \Sigma m\,(n\ln a)}{n\,\Sigma m^2 - (\Sigma m)^2} = 0$$

So in the case where a is independent of angle, there is no change in the observed optical depth**.** This means that any correction to the calibration has no impact on the Langley result (as expected), even taking into the account the log scaling (in fact it is this log scaling that allows it to be independent of the calibration; if it was not for this shift the terms would not cancel).

In practice however, *a* is not completely independent of angle, as our calibration includes angular dependence (see Gardiner et al. (2012)). A change in calibration therefore will have an effect, although the effect will be rather small since it only takes into account the relative change with angle brought about by shifting the calibration factor.

This can be calculated numerically just using the ratios of the observed irradiances (or the ratios of the calibration functions at each angle). Figure S1 shows the case for two assumptions about the extrapolation of the mirror reflectance from the region in which there is data (4000-6600 cm^{-1}) to the regions 2000-4000 and 6600-10000 cm^{-1} in which there is no data. *a* therefore is the difference in observed irradiance arising from using each of these calibration methods. Here *a* varies from ~1.07 at low airmass to ~1.09 at high airmass.

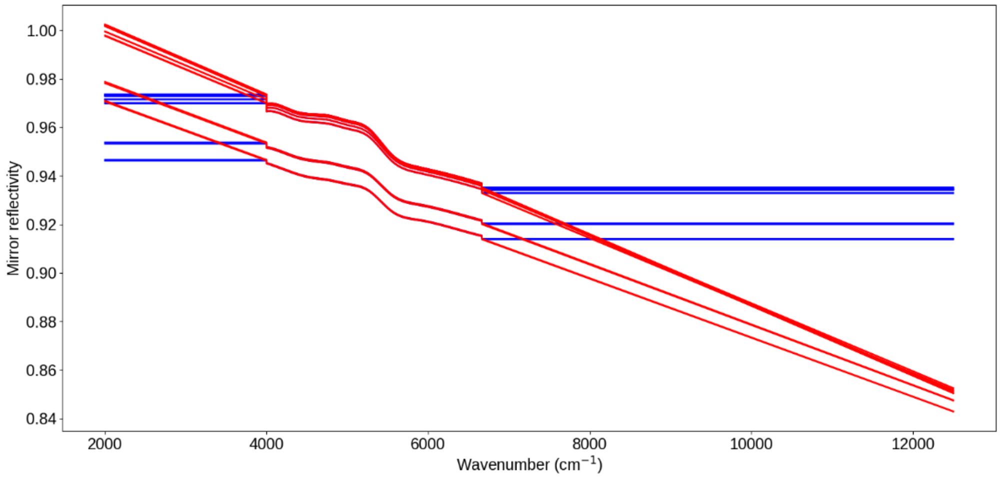

Figure S1: Mirror reflection correction as a function of wavenumber. The red line shows the mirror reflectivity correction used in this work (extrapolating through the observed data), while the blue line shows an alternative mirror reflectivity correction (using the maximum/minimum values).

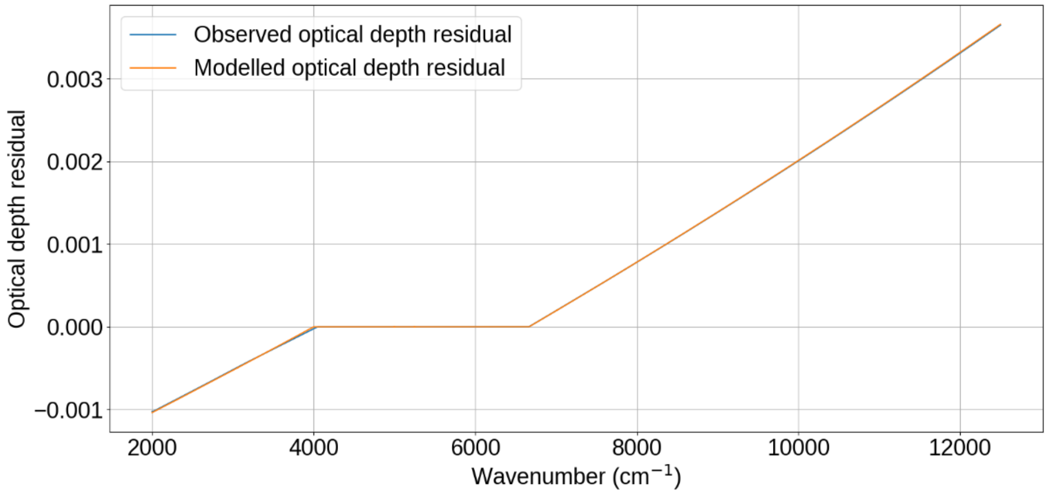

Figure S2: Optical depth residual for a change in mirror reflectance extrapolation (see Figure S1).

Given the optical depth in the centre of the windows at 10000 cm^{-1} and 12000 cm^{-1} in the Langley observations is about 0.10, for an angular dependence to explain all of this contribution (fitting the assumption that the continuum in these windows is close to zero), there would need to be some large, unexplained deviations with angle. Assuming a varies linearly with airmass, the angular dependence would have to account for a doubling of signal in this case. This is extremely unlikely to be the case, particularly since there is only one mirror in our optical setup which has this angular dependence. Therefore, we do not believe that the large optical depth observed in the 1 µm window can be explained via an issue with the mirror reflectivity calibration of our instrument. Additionally, the agreement between the Langley and closure data (Figure 8) indicates that our calibration is robust, since such a calibration issue would affect the closure method significantly but have little effect on a Langley analysis.

It was speculated that there may be some issues arising from the phase correction, as discussed in Section 2. For any such calibration issue to have a significant effect on the Langley-derived spectrum, such an effect would need to vary in time across the course of a day. Additionally, such an issue would need to have a strong wavenumber dependence in order to explain the spurious results at ~10000 cm^{-1}, but have less of an effect at lower wavenumbers. Figure S3 shows the phase correction with respect to time and wavenumber for 18 September 2008.

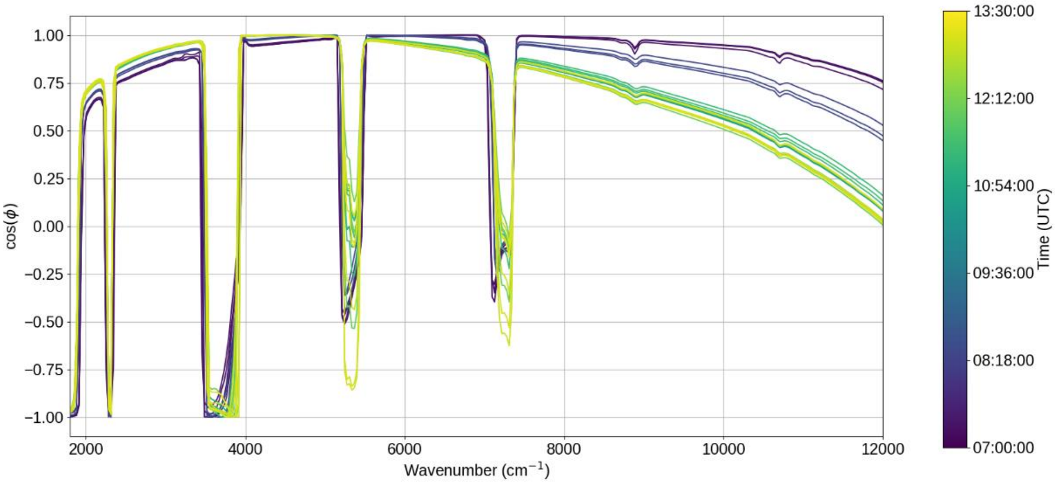

Figure S3: Phase correction calibration with wavenumber for 18 September 2008. The colour bar shows time of day; darker colours indicate earlier times (and higher solar zenith angles).

It is clear from Figure S3 that the correction changes markedly with both time and wavenumber. In principle, the phase correction should bring the output spectrum closer to the true value. However, any uncertainty in the phase angle (the cosine of which is the phase correction in this case) will have an effect on the optical depth. Figures S4 and S5 show the change in derived optical depth for a percentage systematic offset on the phase angle (S4) and for a fixed systematic offset in radians to the phase angle (S5).

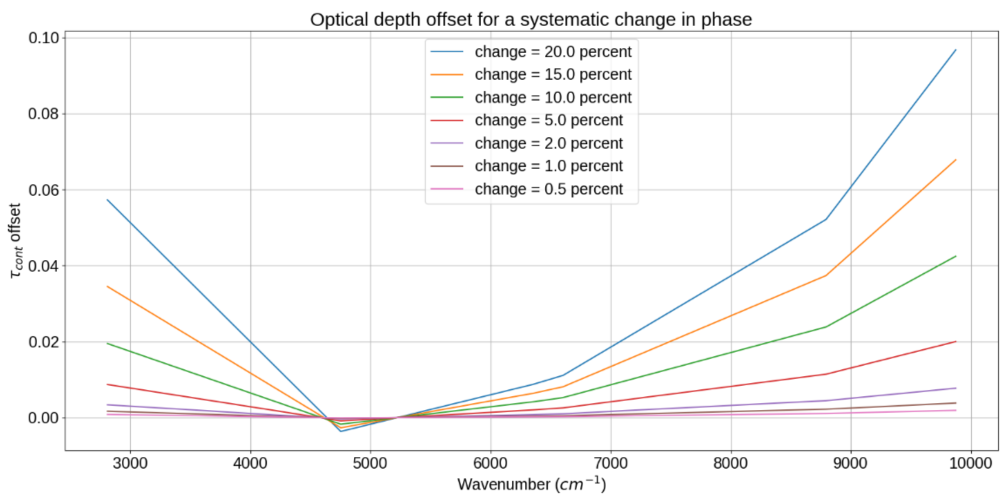

Figure S4: Optical depth offset for a systematic percentage change in phase angle.

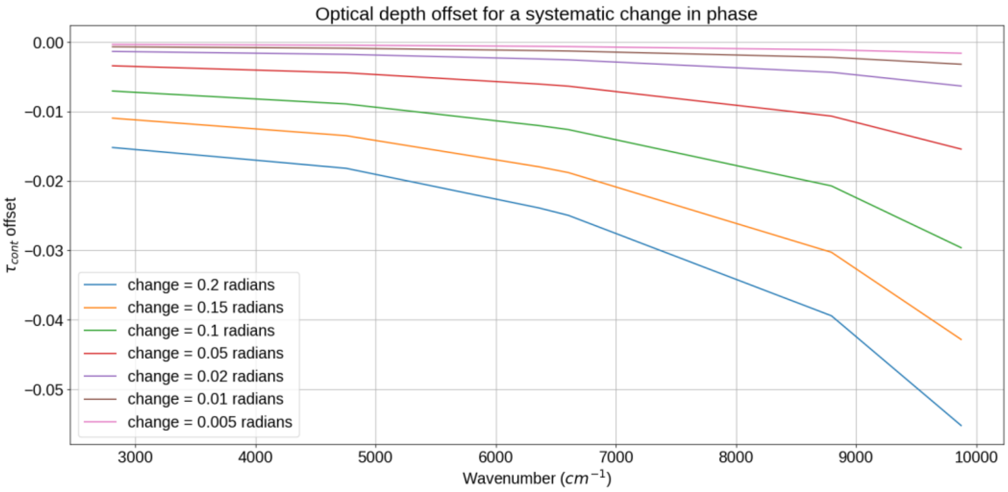

Figure S5: Optical depth offset caused by an absolute systematic change in the phase angle.

These Figures show that the effect on the Langley-derived optical depth is much more significant where there are uncertainties in the phase correction at higher wavenumbers. Interestingly, the pattern is different for a percentage, rather than an absolute offset. This is because of the phase angle itself, which at ~3000 cm^{-1} is offset by a factor of 2π over the course of the day (see Figure S6). This has no effect on the phase correction by itself, but a percentage perturbation to the phase angle results in a significantly different effect on the calibration depending on time of day, causing the large optical depth offset observed in Figure S4.

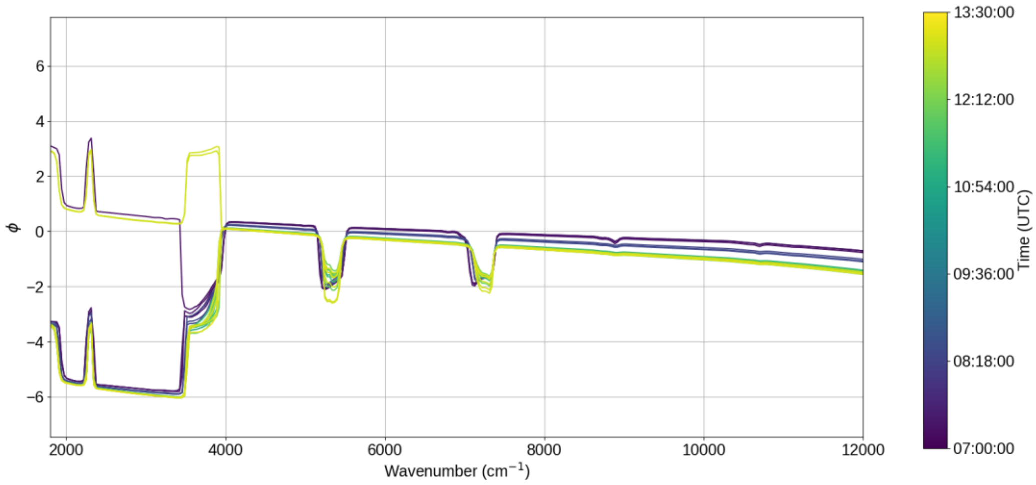

Figure S6: Phase angle used in the phase correction for the observations of 18 September 2008.

We believe that a combination of these factors, as explained in Section 2 of the paper, and the potential for a larger aerosol effect than measured by the Microtops sunphotometer may lead to an underestimate of the uncertainty at wavenumbers beyond ~7000 cm^{-1}. We do not have a means of calculating the magnitude of this uncertainty. Therefore, motivated by the lack of laboratory observations of the water vapour continuum at these wavenumbers, and potential inaccuracy/unreliability of our measurements, we restrict the analysis of our data to the 4, 2.1 and 1.6 µm windows. The introduction of field calibration capability that provided regular monitoring of the calibration across the spectral range would help resolve this issue in future measurements.