Estimation of SOA formation rate

The change in OA concentration (C_{OA}) of the suspended fresh aerosol (prior to photo-oxidation) is due only to wall-deposition. The wall-loss rate usually exhibits a first-order decay:

$$C_{OA}(t) = C_{OA}(t_{reference}) \exp\left(\frac{-\Delta t}{\tau}\right)$$ (S1)

Where τ is the wall-loss time constant, and can be determined by fitting equation (S1) to the decay of the fresh emissions, as shown in Figure S5.

When photo-oxidation is initiated, SOA formation adds mass to the suspended C_{OA}, while it continues to decay by wall-deposition. Assuming that τ does not change much upon SOA formation, SOA concentration in the suspended aerosol can be calculated as the difference between measured C_{OA} and C_{OA} predicted by equation (S1):

$$C_{SOA}(t) = C_{OA}(t) - C_{OA}(t_{reference}) \exp\left(\frac{-\Delta t}{\tau}\right)$$ (S2)

Corrections to aethalometer measurements

Absorption coefficients were measured using an aethalometer (Magee Scientific, model AE-31). The measurements were corrected for two artifacts. The first is absorption enhancement due to multiple scattering in the collection filter. Weingartner et al. (2003) reported a correction factor of 2.14 for aethalometers. Bond et al. (1999) reported a correction factor of 2.18 for the particle soot absorption photometer (PSAP), which is another instrument that measures light absorption and is susceptible to multiple scattering bias. In this work, we used a correction factor of 2.14. The second artifact is the decrease in the aethalometer response as the particle loading increases. This can be due to diminishing the enhancement of filter scattering as particles deposit on it (Kirchstetter and Novakov, 2007; Arnott et al., 2005) or due to some particles being shadowed by others (Weingartner et al., 2003). We used the method of Kirchstetter and Novakov (2007) to correct for this artifact. Measurements of constant BC concentrations over a period long enough to witness a substantial decrease in instrument transmission (Tr) can be used to derive a linear correction factor as a function of Tr. To calibrate

the aethalometer used in this study, we generated aquadag (BC surrogate) by atomizing and drying a suspension in deionized water. We verified that the output concentration was constant by measuring the total concentration using a scanning mobility particle sizer (SMPS). Figure S6 shows original measured and corrected absorption coefficients. It is evident that the derived correction factor eliminates the artifact due to particle loading.

The combined correction due to multiple scattering and particle loading used in this study is:

$$b_{abs,AET} = \frac{b_{AET,raw}}{2.14(0.55Tr + 0.42)} \tag{S3}$$

where $b_{AET,raw}$ and $b_{abs,AET}$ are the measured and corrected absorption coefficients respectively.

References

Arnott WP, Hamasha K, Moosmuller K, Sheridan PJ, Ogren JA (2005) Towards light absorption measurements with a 7-wavelength aethalometer: evaluation with a photoacoustic instrument and a 3-wavelenght nephelometer. *Aerosol Sci Tech* 39: 17-29.

Bond TC, Anderson TL, Campbell D (1999) Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols. *Aerosol Sci Tech* 30: 582-600.

Kirchstetter TW, Novakov T (2007) Controlled generation of black carbon particles from a diffusion flame and applications in evaluating black carbon measurement methods. *Atmos Env* 41: 1874-1888.

Weingartner E *et al.* (2003) Absorption of light by soot particles: determination of the absorption coefficient by means of aethalometers. *J Aerosol Sci* 34: 1445-1463.
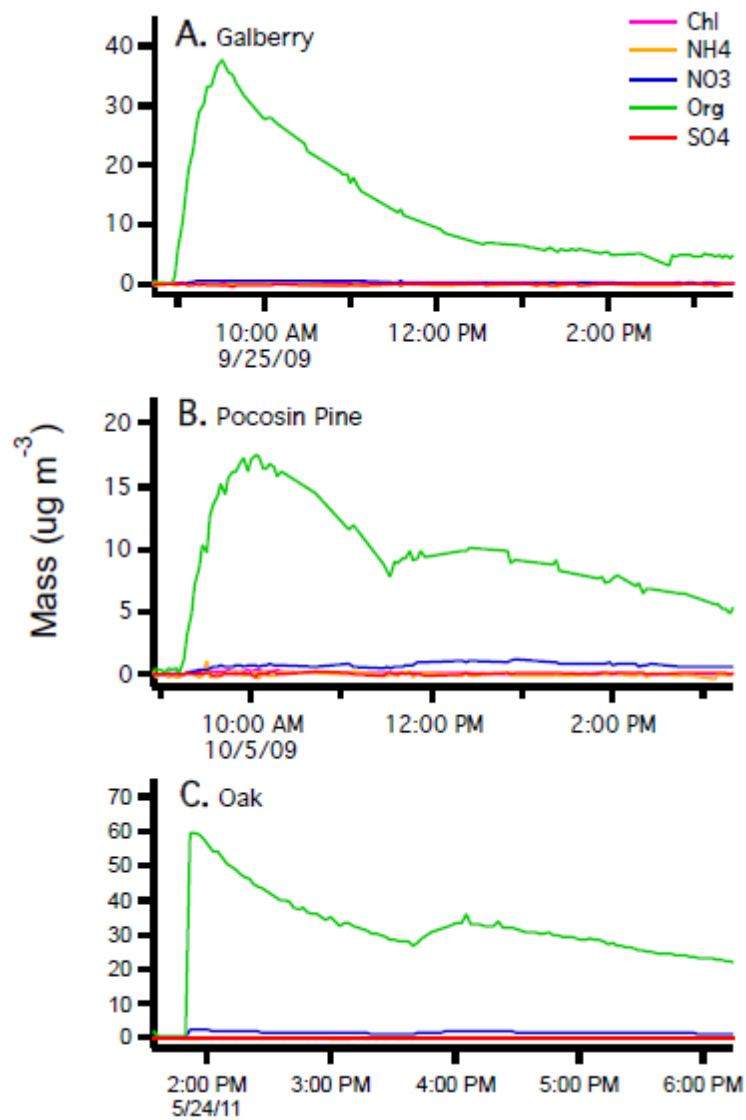
Figures

Figure S1 Time series of non-refractory aerosol concentration in biomass-burning emissions measured using an AMS.
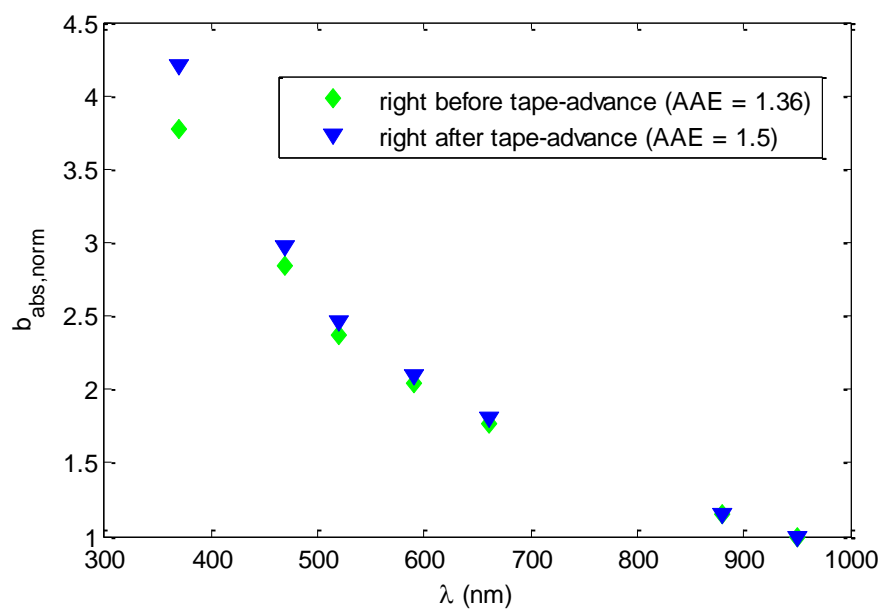
Figure S2 Normalized absorption coefficients obtained from Aethalometer measurements right before and right after tape-advance for oak-burning experiment.
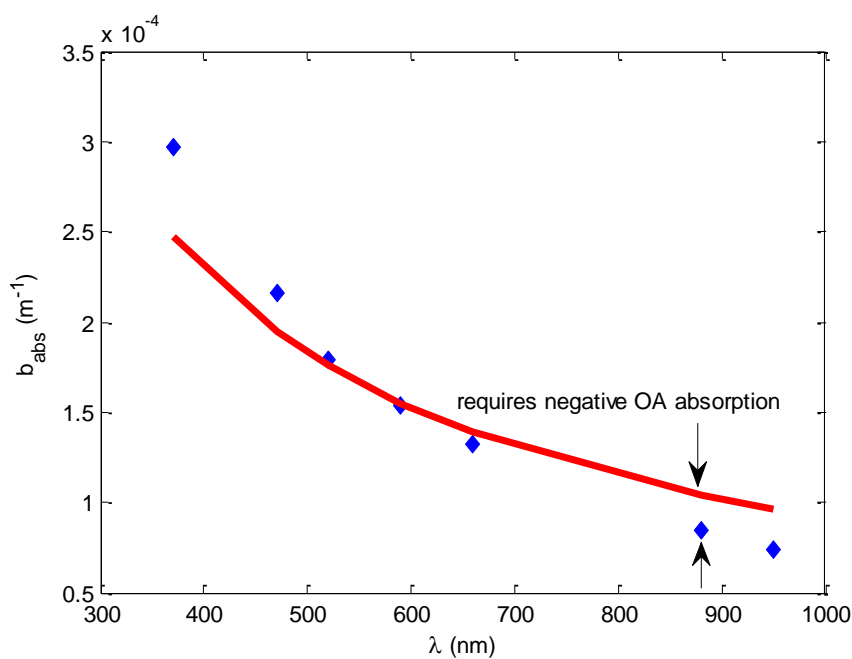

Figure S3 Absorption coefficients obtained from Aethalometer measurements and assuming that OA absorbs negligibly at 550 nm for oak POA experiment. The red curve is the absorption coefficient of BC. Evidently, the shape of the absorption curve does not allow the assumption of

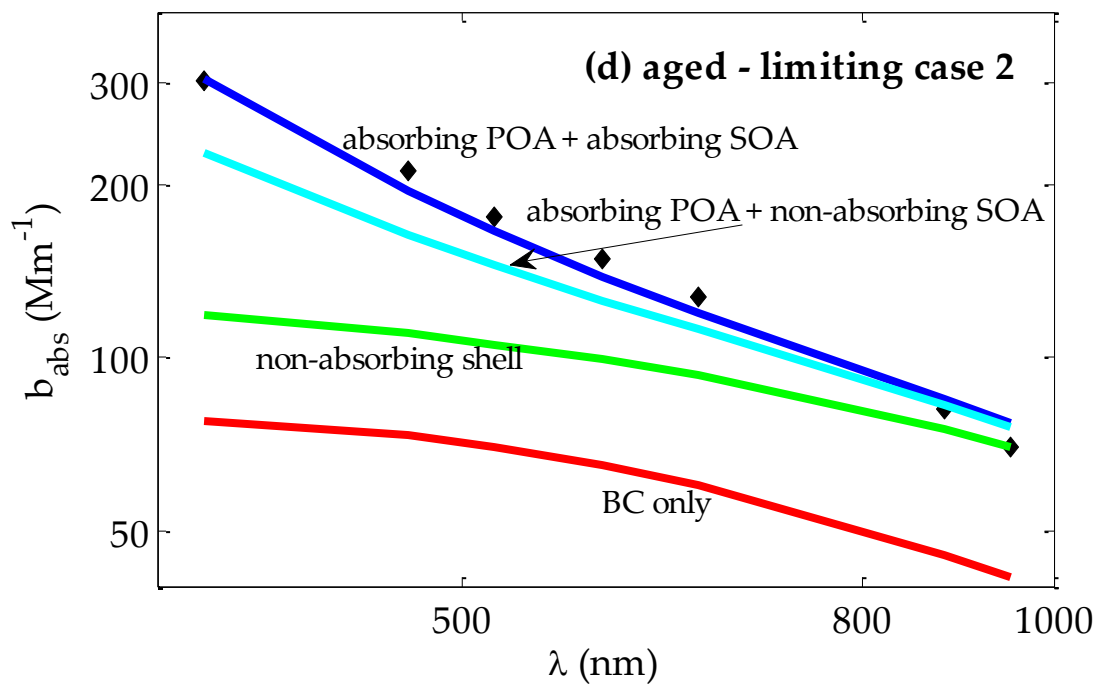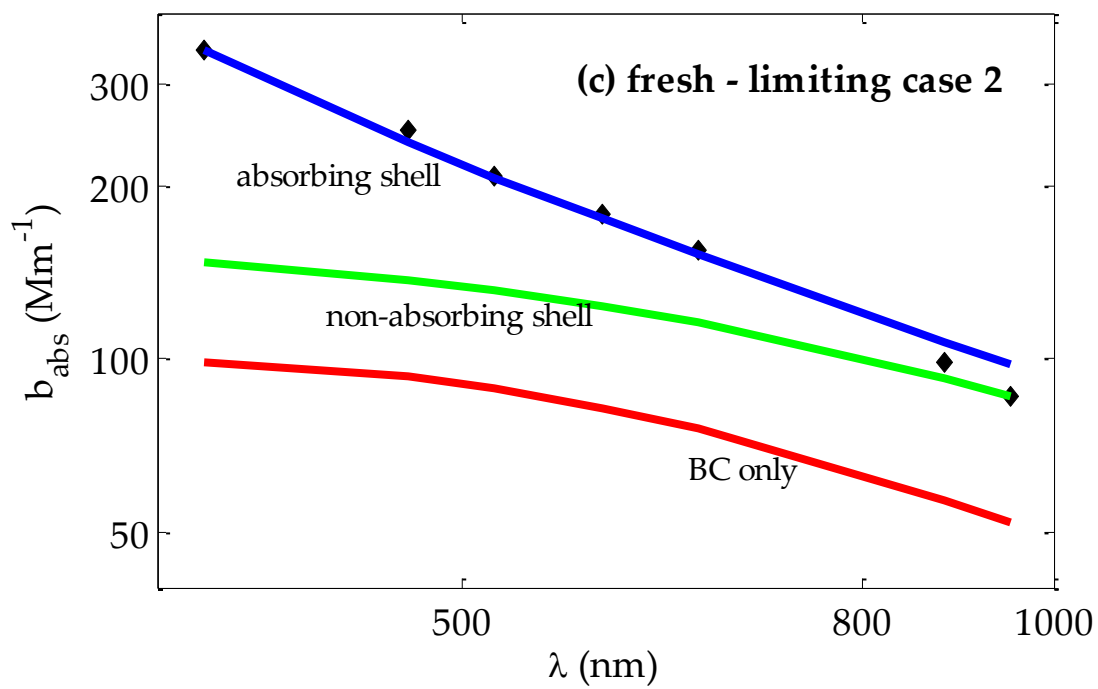negligible OA absorption at 550 nm, as it would require (unphysical) negative OA absorption at $\lambda < 550$ nm.
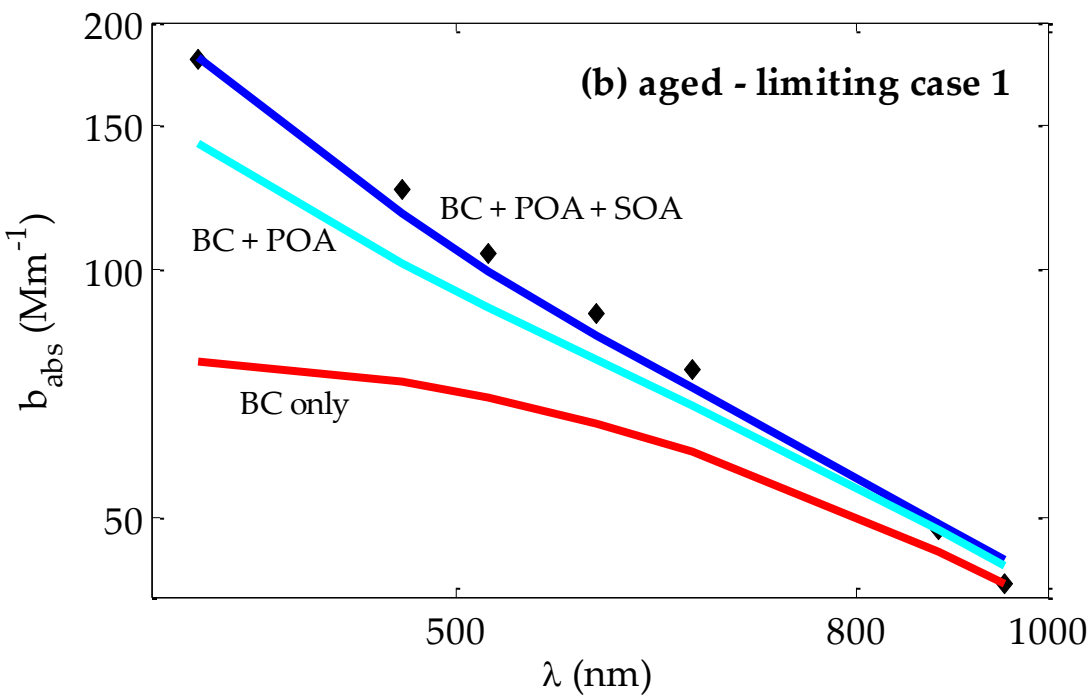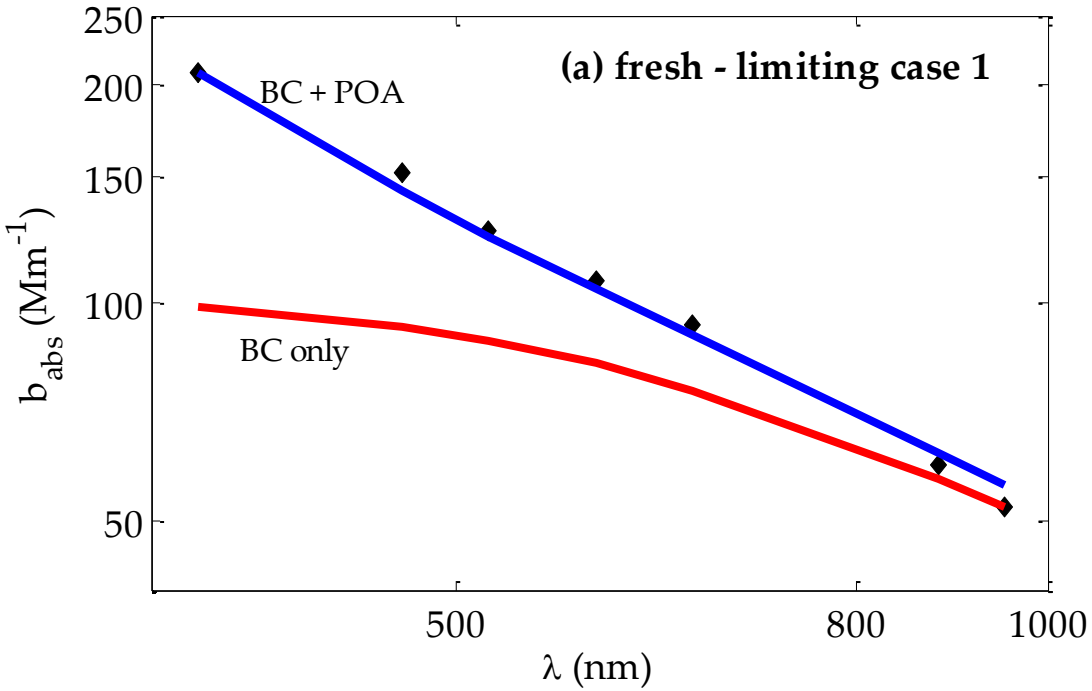
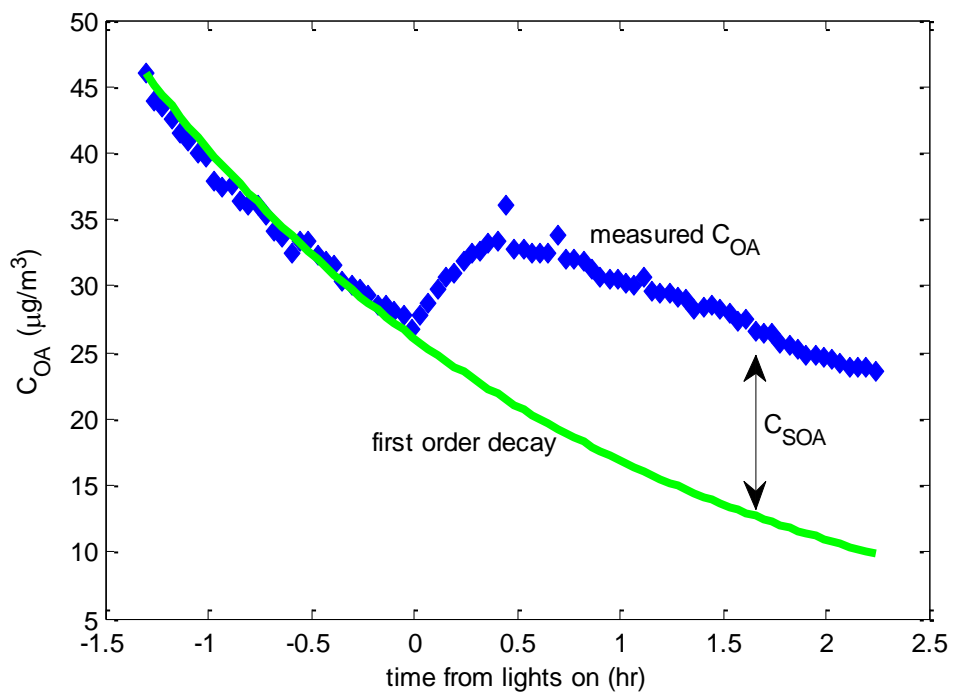**Figure S4** Same as Figure 5 in the main text, but plotted on a log-log scale.

Figure S5 Time series of suspended C_{OA} in oak-aging experiment. The green curve is a fit given by equation (1). The difference between measured C_{OA} and the first order decay fit is the suspended SOA mass concentration (C_{SOA}).
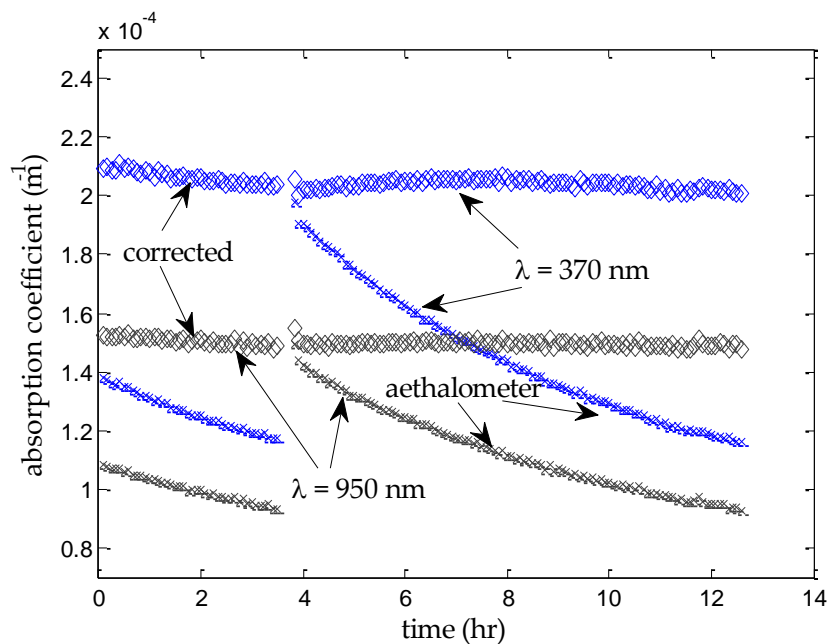
Figure S6 Raw and corrected (for particle loading) absorption coefficients measured by the aethalometer for a constant aquadag concentration. We show the measurements for the RI2 (950 nm) and UV (370 nm) channels only to avoid cluttering. The discontinuity in the raw measurements (at around 4 hr) corresponds to when the filter advances and the correction factor reduces to 1.