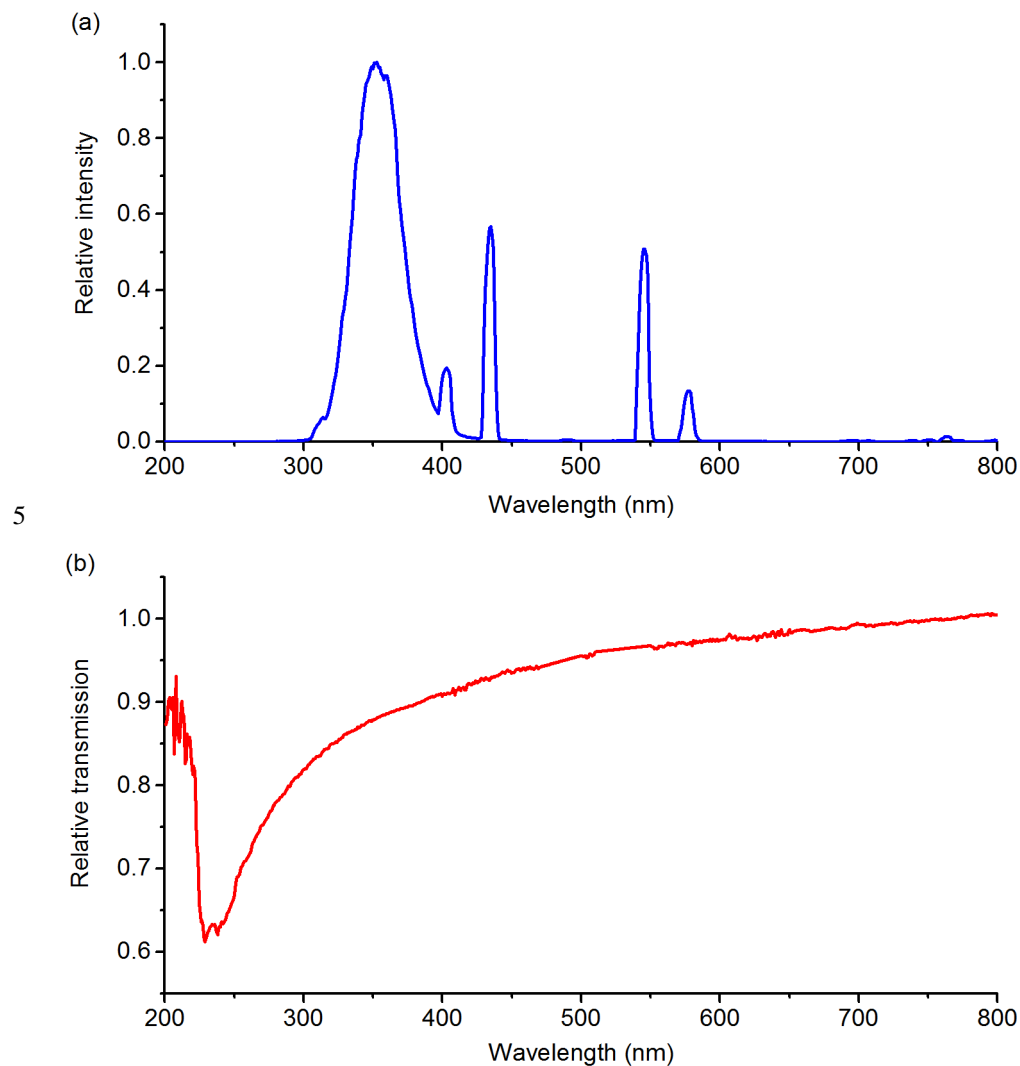


Supplementary material for **“Multiphase composition changes and reactive oxygen species formation during limonene oxidation in the new Cambridge Atmospheric Simulation Chamber (CASC)”** by P.J. Gallimore et al.

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### Spectra of chamber light sources

The spectrum of the tanning lamps used for photochemistry and transmission characteristics of the FEP used for the chamber walls is presented in Figure S1.



**Figure S1: (a) Spectrum of tanning lamps installed in CASC facility. (b) Relative transmission of FEP film used for chamber walls, demonstrating good transmission at wavelengths above 300 nm.**

### Photolysis frequency of NO<sub>2</sub>

A common approach to assess light intensity in atmospheric simulation chambers is to quantify the photolysis frequency of NO<sub>2</sub>,  $J_{\text{NO}_2}$ . Irradiation of NO<sub>2</sub> results in a photo-stationary state between NO, NO<sub>2</sub> and O<sub>3</sub>.  $J_{\text{NO}_2}$  can be calculated from these measured steady-state concentrations and the known bimolecular rate constant ( $k_{\text{NO}+\text{O}_3} = 2 \times 10^{-14} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ , (Atkinson et al., 2004)) according to Equation S1:

$$J_{\text{NO}_2} = \frac{k_{\text{NO}+\text{O}_3}[\text{NO}][\text{O}_3]}{[\text{NO}_2]} \quad (\text{S1})$$

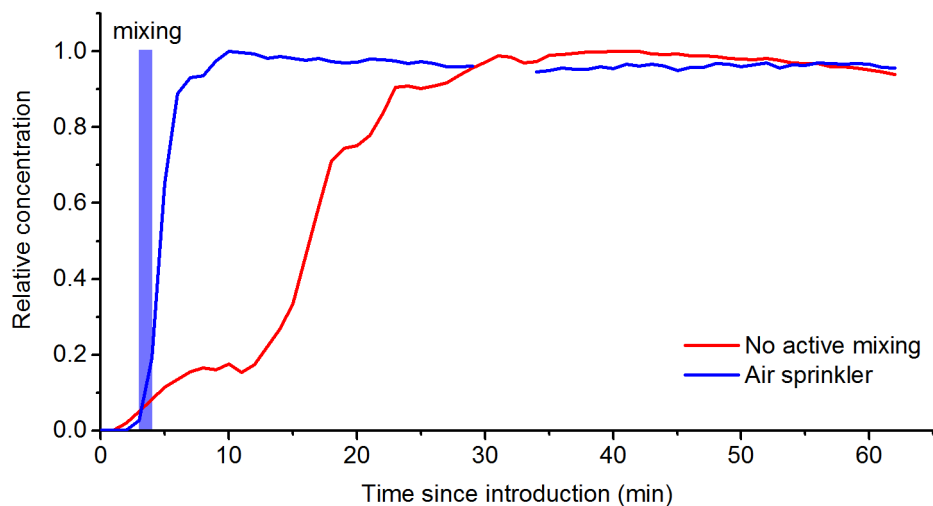
$J_{\text{NO}_2}$  was determined to be  $0.49 \pm 0.09 \text{ min}^{-1}$ . Values of  $J_{\text{NO}_2}$  from various chambers are compiled in Table S1.

Chamber	$J_{\text{NO}_2}$ (min <sup>-1</sup> )	Reference
CASC	0.49	This work
Caltech	1.5	Cocker et al. (2001)
PSI	0.12	Paulsen et al. (2005)
GIG-CAS	0.49	Wang et al. (2014)

**Table S1: Comparison of NO<sub>2</sub> photolysis rates for different indoor chambers using artificial light sources.**

## Chamber mixing

The mixing performance of the “air sprinkler” system was assessed by introducing  $\alpha$ -pinene into chamber via the reactant port and monitoring its concentration over time at the sampling port via PTR-MS.



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**Figure S2: Relative concentration of  $\alpha$ -pinene measured at the sampling chamber port after evaporation into the reactant chamber port. The use of  $3 \times 10$ s “bursts” of the air sprinkler mixes the chamber in a few minutes (blue curve) rather than 30-40 minutes by diffusion alone (red curve). The short gap in the blue data set at 30 minutes corresponds to an interruption to PTR-MS sampling.**

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## Particle wall losses

The lifetime of particles in a simulation chamber is affected by wall losses which are dependent on the size distribution of the aerosol as well as chamber-specific properties such as volume and wall material. Wall losses were determined for the CASC facility by monitoring the evolving size distribution of ammonium sulfate (AS) aerosol over ten experiments between 0 and 70 % RH. Particles were generated from a 0.01 M aqueous AS solution optionally dried prior to introduction. A Kr-85 neutraliser (TSI 3077) was used to ensure an equilibrium aerosol charge distribution in some experiments, but the wall loss rates were not found to be substantially different without the neutraliser.

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Deposition to the walls was treated as a first order process (Cocker et al., 2001). The total aerosol number and mass concentrations as a function of time can be described by Equations S2 and S3 respectively:

$$\frac{dN(D_p, t)}{dt} = -\beta_N N(D_p, t) \quad (\text{S2})$$

15 
$$\frac{dM(D_p, t)}{dt} = -\beta_M M(D_p, t) \quad (\text{S3})$$

Where  $\beta_N$  and  $\beta_M$  are the characteristic number and mass weighted first order wall loss coefficients. 10 aerosol experiments were used to calculate  $\beta_N = 0.201 \pm 0.025 \text{ h}^{-1}$  and  $\beta_M = 0.166 \pm 0.020 \text{ h}^{-1}$ , respectively. This corresponds to aerosol lifetimes of 5-6 hours, comparable to other chambers as illustrated in Table S2.

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Chamber	Volume (m <sup>3</sup> )	$\beta_N$ (h <sup>-1</sup> )	$\beta_M$ (h <sup>-1</sup> )	Reference
CASC	5.4	0.201	0.166	This work
Caltech	28	0.09- 0.18	-	(Cocker et al., 2001)
PSI	27	0.209	0.139	(Paulsen et al., 2005)
EUPHORE	200	0.18		(Martin-Reviejo and Wirtz, 2005)
SAPHIR	270	0.27	-	(Rollins et al., 2009)
CMU	12	0.40	-	(Donahue et al., 2012)
GIG-CAS	30	0.17	-	(Wang et al., 2014)

**Table S2: Comparison of number-weighted and mass-weighted particle wall loss rates for different Teflon chambers.**

- 5 The procedure for particle wall loss correction is similar to that described by Rollins et al., (2009) who assume that particle concentration changes between consecutive measurements (~ 2 minutes) can be decomposed into two separate terms: wall deposition and SOA production. By iteratively increasing the measured concentration at each time step to offset the losses calculated from Equation S3, a loss-free concentration is calculated.
- 10 The resulting loss-corrected time series (Figure 3) suggests that for limonene ozonolysis SOA production continues for 2-3 hours via multi-generational reactions in the chamber and reaches a maximum value of  $375 \mu\text{g m}^{-3}$ . This corresponds to an aerosol yield (defined as the mass of SOA formed per mass of reacted VOC) of 32 %.

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