

Supplementary Information

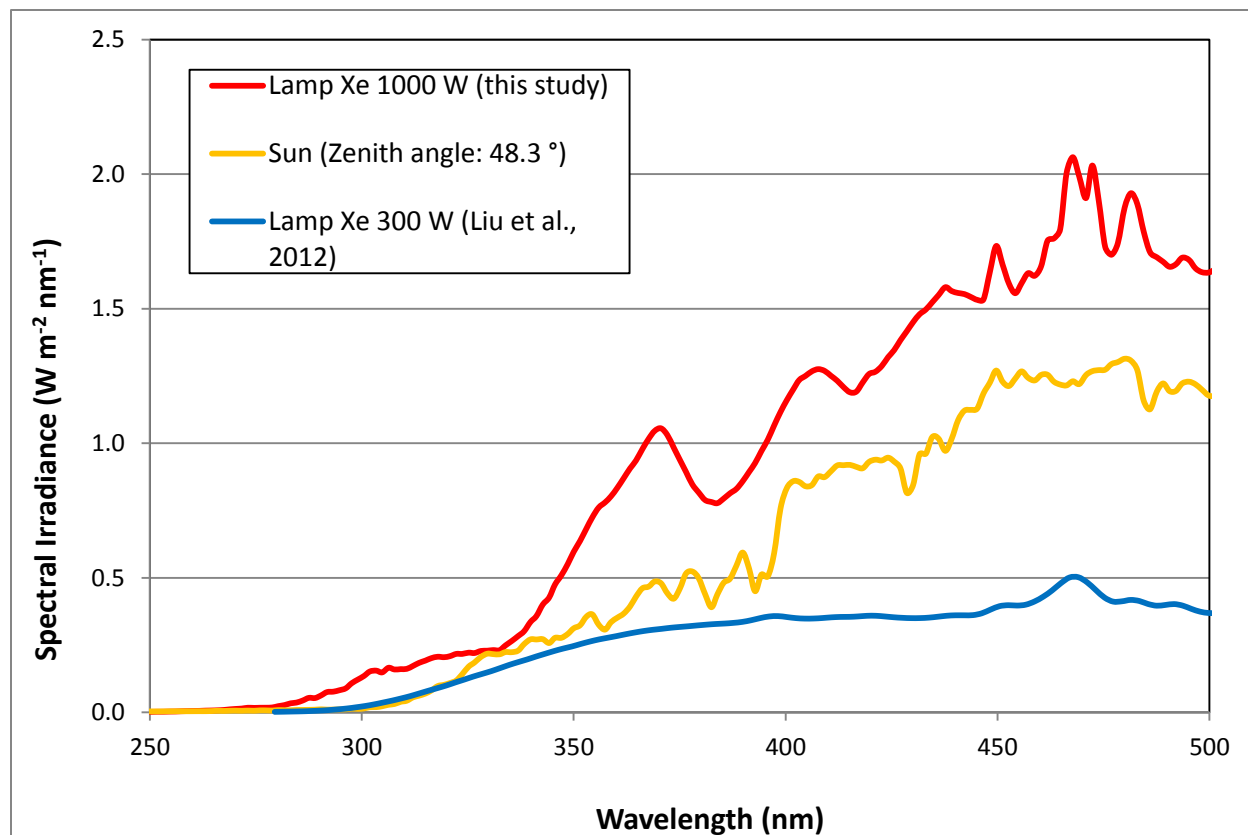
Supplementary information 1:

Calibration experiments of our set up were performed using aqueous solutions of three different compounds (NaCl, NH_4NO_3 , and succinic acid) at various concentrations covering those of the consumed MVK during its reaction (Table S1). In these calibration experiments, for each compound, the obtained numbers of particles increase with increasing solution concentrations, and the corresponding total particle mass (using the corresponding densities) increases linearly with the solution concentration. This result was used to evaluate the transmission efficiency of our set up. Assuming a similar behavior for the nebulized MVK-oligomers up to t_{max} (i.e. increasing oligomer concentrations with reaction time, assuming no major change in the oligomer composition), the calibration experiments were used to determine the SOA mass yields according to equations 2 and 3. The differences obtained between the three calibration experiments may be due to the different physical properties of the particles (e.g. volatility, surface tension and hygroscopicity) linked to their chemical composition. Table S1 shows that the solutions of NaCl generated a significantly higher transmission efficiency than the solutions of NH_4NO_3 and succinic acid. It is likely that the properties of the SOA generated from the nebulized solutions of oxidized MVK were closer to that of NH_4NO_3 and succinic acid than NaCl. Finally, the transmission efficiency obtained for NH_4NO_3 solutions was used to calculate the SOA mass yields according to equations 2 and 3.

Compound	Range of aqueous phase concentration (mg L^{-1})	Range of particle mass concentration after nebulization ($\mu\text{g m}^{-3}$)	Particle density (g cm^{-3})	Transmission efficiency (%)
Succinic acid	50 - 2000	20 - 1300	1.56	13.1 (\pm 2.6)
NH_4NO_3	30 - 3000	10 - 4500	1.73	17.2 (\pm 7.9)
NaCl	50 - 2000	100 - 4000	2.17	44.6 (\pm 2.3)

Table S1: Determination of particle transmission efficiency in the particle generation system

Figure S1: Average spectral irradiance of the Xe lamps 1000 W with a ASTM 490 AM 0 filter (red, used in the present study), 300 W with a pyrex filter (blue, Liu et al., 2012), as compared to the direct solar irradiance at sea level, for a 48.3° zenith angle (orange).



Supplementary information 3: Determination of $\cdot\text{OH}$ concentrations in the solution

$\cdot\text{OH}$ concentrations were calculated by means of two different methods (methods a) and b) below), which gave similar results:

a) Considering reactions R1 to R3, $\cdot\text{OH}$ concentrations were calculated assuming the steady state approximation at time 0:

$$[\text{OH}] = \frac{2 \times J_{\text{H}_2\text{O}_2} \times [\text{H}_2\text{O}_2]}{k_3 \times [\text{MVK}] + k_2 \times [\text{H}_2\text{O}_2]}$$



$J_{\text{H}_2\text{O}_2}$ was determined by fitting H_2O_2 concentration decay



$$k_2 = 2.95 \cdot 10^7 \text{ M}^{-1} \text{ s}^{-1} \text{ at } 25^\circ \text{ C} \quad (\text{Christensen et al., 1982})$$



$$k_3 = 7.3.(\pm 0,5) 10^9 \text{ M}^{-1} \text{ s}^{-1} \text{ at } 25^\circ \text{ C} \quad (\text{Schöne et al., 2014})$$

The obtained values are $[\cdot\text{OH}] = (6 - 5) \times 10^{-14} \text{ M}$ (for pH 6 - 3 respectively)

b) Fitting the initial MVK decay with an exponential decay:

$$[\text{MVK}]_t = [\text{MVK}]_0 \times e^{-(k_3 \times [\cdot\text{OH}] \times t)}$$

With $k_3 = 7,3.(\pm 0,5) 10^9 \text{ M}^{-1} \text{ s}^{-1}$ at 25° C (Schöne et al., 2014), we obtained similar values for $\cdot\text{OH}$ steady state concentrations ($[\cdot\text{OH}] = 2 \times 10^{-14} \text{ M}$) as for method a).

References

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