## Captive Aerosol Growth and Evolution (CAGE) chamber system to investigate particle growth due to secondary aerosol formation

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## Supplement

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Figure S1. First generation CAGE chambers, which were referred to as OUALITY chambers in some publications.



25 Figure S2. Close-up photo of a CAGE chamber highlighting the overall clarity.



Figure S3. Pictures of MAQL instrumentation and research trailer setup at JSF during field campaign. (a): JSF sampling site within an open clearing of the forest facing southwest, where the following are shown from left to right and are numbered from 1 to 5: (1) University of Houston/Rice University MAQL, (2) Sandia National Laboratories trailer, (3) Texas A&M University trailer, (4) University of Houston/Rice University secondary trailer, and (5) Baylor University trailer, (b): TCEQ CAMPS698 tower located at the site from which wind speed and wind direction data are collected, (c): MAQL showing access to the rear instrumentation bed, and (d): Trace gas/meteorology/PM<sub>1</sub> sampling arm of the MAQL while in stationary sampling mode. Reprinted from Fig S 29 in Alexander Bui's PhD thesis 2018.



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Figure S4. Wind rose calculated for the period of the field study (August 15 – October 14, 2016) at the nearby Conroe airport. Courtesy of the Western Regional Climate Center (wrcc.dri.edu).



40 Figure S5. Relationship of the correlation between time series of NO<sub>y</sub> concentration i) measured in the chambers and ii) determined from the ambient measurements assuming the chambers can be modeled as CSTRs with exchange flow rate  $Q_{ex}$ . The  $Q_{ex}$  used for all subsequent calculations is that at which  $r^2$  was highest. The average of the results for Chambers A and B is shown.



45 Figure S6. Time series of lognormal fit diameter to all modes tracked during the 2016 study. The color bar towards the top of the graph indicates the periods during which at least one mode was tracked and growth rate could be determined. The gap from 9/9/2016 to 9/12/2016 was the three-day gas-phase comparison experiment discussed in Sect. 4.1.



Figure S7. Sub-1 µm non-refractory composition of ambient aerosol measured with an HR-ToF-AMS. The colors indicate the following species: green = organics, red = sulfates, yellow = ammonium, and blue = nitrates.



55 Figure S8. Relationship between the early evening average growth rate and average O<sub>3</sub> mixing ratio. The correlation between the two and the general increase in both during the fall suggests that some of the increased nighttime particle growth towards the end of the project was associated with increased O<sub>3</sub>.

Table S1. Reactions included i	in the	CSTR-0D	model
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Reaction	Rate coefficient (cm <sup>3</sup> s <sup>-1</sup> unless stated otherwise)	Notes
$NO + O_3 \rightarrow NO_2 + O_2$	$1.40 \ge 10^{-12} \exp(-1310/T)$	
$NO_2 + O_3 \rightarrow NO_3 + O_2$	$1.40 \ge 10^{-12} \exp(-2470/T)$	

$NO_2 + NO_3 \cdot (+M) \rightarrow N_2O_5$	1.9 x 10 <sup>-12</sup> (T/300) <sup>0.2</sup>	High pressure limit rate constant and expression		
$N_2O_5 + M \rightarrow NO_2 + NO_3$	1.30 x $10^{-3}$ (T/300) <sup>-3.5</sup>			
	exp(-11000/T)			
$NO_3$ · + $NO \rightarrow 2NO_2$	1.80 x 10 <sup>-11</sup> exp(110/T)			
$OH$ · + NO (+ M) $\rightarrow$ HONO	1.50 x 10 <sup>-11</sup> (T/300) <sup>-0.5</sup>	High pressure limit rate constant and expression		
$OH$ · + NO <sub>2</sub> (+ M) $\rightarrow$ HNO <sub>3</sub>	2.40 x 10 <sup>-11</sup> (T/300) <sup>-1.3</sup>	High pressure limit rate constant and expression		
$NO_3 \cdot + RO_2 \cdot \rightarrow NO_2$	2.00 x 10 <sup>-12</sup>			
$RO_2 \cdot + RO_2 \cdot \rightarrow \text{products}$	2.00 x 10 <sup>-12</sup>			
$OH$ + acetaldehyde $\rightarrow RO_2$ ·	$5.55 \ge 10^{-12} \exp(287/T)$			
	Photolysis react	tions		
$NO_2 + hv \rightarrow NO + O_3$	J <sub>NO2</sub>	Calculated assuming photostationary state for		
		ambient NO, NO <sub>2</sub> , and O <sub>3</sub> concentrations		
NO <sub>3</sub> · + hv → 0.12NO + 0.88NO <sub>2</sub>	J <sub>NO3</sub>	Calculated from measured spectral intensity		
$+0.88O_{3}$				
$O_3 + hv \rightarrow 0.3OH + O_3$	J <sub>O1D</sub>	Calculated from measured spectral intensity.		
		Assumes 15% of $O(^1D)$ produced reacts with $H_2O$		
		to form $OH$ . $O_3$ is conserved because any shift this		
		would cause would be captured in the estimated		
		J <sub>NO2</sub> .		
$HONO + hv \rightarrow OH + NO$	J <sub>HONO</sub>	Calculated from measured spectral intensity		
Secondary aerosol forming reactions				
	Secondary aerosol form	ing reactions		
$OH \cdot + \alpha$ -pinene $\rightarrow RO_2 \cdot$	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T)	ing reactions		
$OH \cdot + \alpha \text{-pinene} \rightarrow RO_2 \cdot$ $OH \cdot + \beta \text{-pinene} \rightarrow RO_2 \cdot$	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T)	ing reactions		
$\begin{array}{c} OH \cdot + \alpha \text{-pinene} \rightarrow RO_2 \cdot \\ \hline OH \cdot + \beta \text{-pinene} \rightarrow RO_2 \cdot \\ \hline OH \cdot + \text{ isoprene } \rightarrow 0.6RO_2 \cdot + \end{array}$	Secondary aerosol form           2.54 x 10 <sup>-11</sup> exp(410/T)           1.21 x 10 <sup>-11</sup> exp(444/T)           2.38 x 10 <sup>-11</sup> exp(357/T)	ing reactions		
OH· + $\alpha$ -pinene $\rightarrow$ RO <sub>2</sub> · OH· + $\beta$ -pinene $\rightarrow$ RO <sub>2</sub> · OH· + isoprene $\rightarrow$ 0.6RO <sub>2</sub> · + 0.4MVK+MACR	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T) 2.38 x 10 <sup>-11</sup> exp(357/T)	ing reactions		
OH· + $\alpha$ -pinene $\rightarrow$ RO <sub>2</sub> · OH· + $\beta$ -pinene $\rightarrow$ RO <sub>2</sub> · OH· + isoprene $\rightarrow$ 0.6RO <sub>2</sub> · + 0.4MVK+MACR OH· + toluene $\rightarrow$ RO <sub>2</sub> ·	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T) 2.38 x 10 <sup>-11</sup> exp(357/T) 1.81 x 10 <sup>-12</sup> exp(280/T)	ing reactions		
$OH· + \alpha$ -pinene $\rightarrow RO_2$ · $OH· + \beta$ -pinene $\rightarrow RO_2$ · $OH· + isoprene \rightarrow 0.6RO_2· + 0.4MVK+MACR$ $OH· + toluene \rightarrow RO_2·$ $OH· + sO_2 \rightarrow SO_4$	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T) 2.38 x 10 <sup>-11</sup> exp(357/T) 1.81 x 10 <sup>-12</sup> exp(280/T) 1.50 x 10 <sup>-12</sup>	ing reactions		
$OH· + \alpha$ -pinene $\rightarrow RO_2$ · $OH· + \beta$ -pinene $\rightarrow RO_2$ · $OH· + isoprene \rightarrow 0.6RO_2· + 0.4MVK+MACR$ $OH· + toluene \rightarrow RO_2·$ $OH· + sO_2 \rightarrow SO_4$ $OH· + sO_2 \rightarrow SO_4$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T) 2.38 x 10 <sup>-11</sup> exp(357/T) 1.81 x 10 <sup>-12</sup> exp(280/T) 1.50 x 10 <sup>-12</sup> 6.30 x 10 <sup>-16</sup> exp(-580/T)	ing reactions		
$OH· + \alpha$ -pinene $\rightarrow RO_2$ · $OH· + \beta$ -pinene $\rightarrow RO_2$ · $OH· + isoprene \rightarrow 0.6RO_2· + 0.4MVK+MACR$ $OH· + toluene \rightarrow RO_2·$ $OH· + sO_2 \rightarrow SO_4$ $OH· + SO_2 \rightarrow SO_4$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$	Secondary aerosol form           2.54 x 10 <sup>-11</sup> exp(410/T)           1.21 x 10 <sup>-11</sup> exp(444/T)           2.38 x 10 <sup>-11</sup> exp(357/T)           1.81 x 10 <sup>-12</sup> exp(280/T)           1.50 x 10 <sup>-12</sup> 6.30 x 10 <sup>-16</sup> exp(-580/T)           1.20 x 10 <sup>-15</sup> exp(-1300/T)	ing reactions		
$\begin{array}{c} OH \cdot + \alpha \text{-pinene} \rightarrow RO_{2} \cdot \\ OH \cdot + \beta \text{-pinene} \rightarrow RO_{2} \cdot \\ OH \cdot + \text{ isoprene} \rightarrow 0.6RO_{2} \cdot + \\ 0.4MVK + MACR \\ OH \cdot + \text{ toluene} \rightarrow RO_{2} \cdot \\ OH \cdot + SO_{2} \rightarrow SO_{4} \\ O_{3} + \alpha \text{-pinene} \rightarrow RO_{2} \cdot \\ O_{3} + \beta \text{-pinene} \rightarrow RO_{2} \cdot \\ O_{3} + \text{ isoprene} \rightarrow 0.6RO_{2} \cdot + \end{array}$	Secondary aerosol form $2.54 \times 10^{-11} \exp(410/T)$ $1.21 \times 10^{-11} \exp(444/T)$ $2.38 \times 10^{-11} \exp(357/T)$ $1.81 \times 10^{-12} \exp(280/T)$ $1.50 \times 10^{-12}$ $6.30 \times 10^{-16} \exp(-580/T)$ $1.20 \times 10^{-15} \exp(-1300/T)$ $1.03 \times 10^{-14} \exp(-1995/T)$	ing reactions		
$OH· + \alpha$ -pinene $\rightarrow RO_2$ · $OH· + \beta$ -pinene $\rightarrow RO_2$ · $OH· + isoprene \rightarrow 0.6RO_2· + 0.4MVK+MACR$ $OH· + toluene \rightarrow RO_2·$ $OH· + SO_2 \rightarrow SO_4$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + isoprene \rightarrow 0.6RO_2· + 0.4MVK+MACR$	Secondary aerosol form           2.54 x 10 <sup>-11</sup> exp(410/T)           1.21 x 10 <sup>-11</sup> exp(444/T)           2.38 x 10 <sup>-11</sup> exp(357/T)           1.81 x 10 <sup>-12</sup> exp(280/T)           1.50 x 10 <sup>-12</sup> 6.30 x 10 <sup>-16</sup> exp(-580/T)           1.20 x 10 <sup>-15</sup> exp(-1300/T)           1.03 x 10 <sup>-14</sup> exp(-1995/T)	ing reactions		
$\begin{array}{c} & \\ \hline OH^{\cdot} + \alpha \text{-pinene} \rightarrow RO_2^{\cdot} \\ \hline OH^{\cdot} + \beta \text{-pinene} \rightarrow RO_2^{\cdot} \\ \hline OH^{\cdot} + \text{isoprene} \rightarrow 0.6RO_2^{\cdot} + \\ \hline 0.4MVK + MACR \\ \hline OH^{\cdot} + \text{toluene} \rightarrow RO_2^{\cdot} \\ \hline OH^{\cdot} + SO_2 \rightarrow SO_4 \\ \hline O_3 + \alpha \text{-pinene} \rightarrow RO_2^{\cdot} \\ \hline O_3 + \beta \text{-pinene} \rightarrow RO_2^{-} \\ \hline O_3 $	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T) 2.38 x 10 <sup>-11</sup> exp(357/T) 1.81 x 10 <sup>-12</sup> exp(280/T) 1.50 x 10 <sup>-12</sup> 6.30 x 10 <sup>-16</sup> exp(-580/T) 1.20 x 10 <sup>-15</sup> exp(-1300/T) 1.03 x 10 <sup>-14</sup> exp(-1995/T) 1.20 x 10 <sup>-12</sup> exp(490/T)	ing reactions		
$OH· + \alpha$ -pinene $\rightarrow RO_2$ · $OH· + \beta$ -pinene $\rightarrow RO_2$ · $OH· + isoprene \rightarrow 0.6RO_2· +$ $0.4MVK+MACR$ $OH· + toluene \rightarrow RO_2·$ $OH· + SO_2 \rightarrow SO_4$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$ $AMVK+MACR$ $NO_3· + \alpha$ -pinene $\rightarrow RO_2·$ + $0.8NO_2$	Secondary aerosol form           2.54 x 10 <sup>-11</sup> exp(410/T)           1.21 x 10 <sup>-11</sup> exp(444/T)           2.38 x 10 <sup>-11</sup> exp(357/T)           1.81 x 10 <sup>-12</sup> exp(280/T)           1.50 x 10 <sup>-12</sup> 6.30 x 10 <sup>-16</sup> exp(-580/T)           1.20 x 10 <sup>-15</sup> exp(-1300/T)           1.03 x 10 <sup>-14</sup> exp(-1995/T)	ing reactions		
$OH· + \alpha$ -pinene $\rightarrow RO_2$ · $OH· + \beta$ -pinene $\rightarrow RO_2$ · $OH· + isoprene \rightarrow 0.6RO_2· + 0.4MVK+MACR$ $OH· + toluene \rightarrow RO_2·$ $OH· + sO_2 \rightarrow SO_4$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + isoprene \rightarrow 0.6RO_2· + 0.4MVK+MACR$ $NO_3· + \alpha$ -pinene $\rightarrow RO_2· + 0.8NO_2$ $NO_3· + \beta$ -pinene $\rightarrow RO_2· + 0.8NO_2$	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T) 2.38 x 10 <sup>-11</sup> exp(357/T) 1.81 x 10 <sup>-12</sup> exp(280/T) 1.50 x 10 <sup>-12</sup> 6.30 x 10 <sup>-16</sup> exp(-580/T) 1.20 x 10 <sup>-15</sup> exp(-1300/T) 1.03 x 10 <sup>-14</sup> exp(-1995/T) 1.20 x 10 <sup>-12</sup> exp(490/T) 2.50 x 10 <sup>-12</sup>	ing reactions		
$OH· + \alpha$ -pinene $\rightarrow RO_2$ · $OH· + \beta$ -pinene $\rightarrow RO_2$ · $OH· + isoprene \rightarrow 0.6RO_2· + 0.4MVK+MACR$ $OH· + toluene \rightarrow RO_2·$ $OH· + sO_2 \rightarrow SO_4$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \beta$ -pinene $\rightarrow RO_2·$ $O_3 + \alpha$ -pinene $\rightarrow RO_2·$ $O_3 - \alpha$ -pinene $\rightarrow RO_2·$	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T) 2.38 x 10 <sup>-11</sup> exp(357/T) 1.81 x 10 <sup>-12</sup> exp(280/T) 1.50 x 10 <sup>-12</sup> exp(280/T) 1.50 x 10 <sup>-16</sup> exp(-580/T) 1.20 x 10 <sup>-15</sup> exp(-1300/T) 1.03 x 10 <sup>-14</sup> exp(-1995/T) 1.20 x 10 <sup>-12</sup> exp(490/T) 2.50 x 10 <sup>-12</sup>	ing reactions		
$OH· + \alpha$ -pinene $\rightarrow RO_2$ : $OH· + \beta$ -pinene $\rightarrow RO_2$ : $OH· + isoprene \rightarrow 0.6RO_2$ · + $0.4MVK+MACR$ $OH· + toluene \rightarrow RO_2$ : $OH· + sO_2 \rightarrow SO_4$ $O_3 + \alpha$ -pinene $\rightarrow RO_2$ : $O_3 + \beta$ -pinene $\rightarrow RO_2$ : $AMVK+MACR$ $NO_3$ : $+ \alpha$ -pinene $\rightarrow RO_2$ : $+$ $0.8NO_2$ $NO_3$ : $+ \beta$ -pinene $\rightarrow RO_2$ : $+$ $0.3NO_2$ $NO_3$ : $+ isoprene \rightarrow 0.95RO_2: +$	Secondary aerosol form 2.54 x 10 <sup>-11</sup> exp(410/T) 1.21 x 10 <sup>-11</sup> exp(444/T) 2.38 x 10 <sup>-11</sup> exp(357/T) 1.81 x 10 <sup>-12</sup> exp(280/T) 1.50 x 10 <sup>-12</sup> exp(280/T) 1.50 x 10 <sup>-16</sup> exp(-580/T) 1.20 x 10 <sup>-15</sup> exp(-1300/T) 1.03 x 10 <sup>-14</sup> exp(-1995/T) 1.20 x 10 <sup>-12</sup> exp(490/T) 2.50 x 10 <sup>-12</sup> 3.15 x 10 <sup>-12</sup> exp (-450/T)	ing reactions		
$OH' + \alpha$ -pinene $\rightarrow RO_2'$ $OH' + \beta$ -pinene $\rightarrow RO_2'$ $OH' + isoprene \rightarrow 0.6RO_2' + 0.4MVK+MACR$ $OH' + toluene \rightarrow RO_2'$ $OH' + sO_2 \rightarrow SO_4$ $O_3 + \alpha$ -pinene $\rightarrow RO_2'$ $O_3 + \beta$ -pinene $\rightarrow RO_2'$ $O_3 + \beta$ -pinene $\rightarrow RO_2'$ $O_3 + isoprene \rightarrow 0.6RO_2' + 0.4MVK+MACR$ $NO_3' + \alpha$ -pinene $\rightarrow RO_2' + 0.8NO_2$ $NO_3' + \beta$ -pinene $\rightarrow RO_2' + 0.3NO_2$ $NO_3' + isoprene \rightarrow 0.95RO_2' + 0.3NO_2 + 0.05MVK+MACR$	Secondary aerosol form           2.54 x 10 <sup>-11</sup> exp(410/T)           1.21 x 10 <sup>-11</sup> exp(444/T)           2.38 x 10 <sup>-11</sup> exp(357/T)           1.81 x 10 <sup>-12</sup> exp(280/T)           1.50 x 10 <sup>-12</sup> 6.30 x 10 <sup>-16</sup> exp(-580/T)           1.20 x 10 <sup>-15</sup> exp(-1300/T)           1.03 x 10 <sup>-14</sup> exp(-1995/T)           1.20 x 10 <sup>-12</sup> exp(490/T)           2.50 x 10 <sup>-12</sup> 3.15 x 10 <sup>-12</sup> exp (-450/T)			

$N_2O_5 + H_2O(\ell) \rightarrow 2HNO_3$	2.67 x 10 <sup>-7</sup> x S (μm <sup>2</sup> cm <sup>-</sup>	$10^{-7}$ x S (µm <sup>2</sup> cm <sup>-</sup> Aerosol surface area, S, assumed to be 100 µ			
	<sup>3</sup> )	cm <sup>-3</sup>			
Tuning reactions					
$O_3$ + wall $\rightarrow$ 0.65acetaldehyde +	1.00 x 10 <sup>-4</sup> s <sup>-1</sup> for A	Used to match observed O <sub>3</sub> loss and acetaldehyde			
0.12acetone	$0.70 \ x \ 10^{\text{-4}} \ \text{s}^{\text{-1}}$ for B	and acetone production. Wall "concentration"			
		dependent upon solar intensity to adjust for			
		increase in contact frequency with increased			
		convective mixing.			
OH· + X →	2 s <sup>-1</sup>	Used to produce reasonable daily peak OH·.			
		Partially constrained by daytime difference			
		between chamber and ambient isoprene resulting			
		from reaction with OH.			
$(N_2+O_2) + wall \rightarrow HONO$	$1.0 \ge 10^7 \text{ s}^{-1}$	Recognized radical source in Teflon® chambers.			
		Partially constrained by pattern of daytime			
		difference between chamber and ambient isoprene			
		resulting from reaction with OH.			

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- OH is conserved for all "OH + reactant  $\rightarrow$ " reactions because an independent overall OH reactivity is assumed.

- Species in parentheses involved in reaction but not needed for calculated rate.

- Chamber temperature assumed to be the same as ambient temperature.

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