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Supplementary Materials for

Role of sesquiterpenes in biogenic new particle formation

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Table S1 List of experimental runs. Summary of the runs included in this study showing experimental conditions, injection mixing ratios and median measured concentrations of precursor vapours. Pure α-pinene runs (1, 2, 3) marked with a (*) are from Simon *et al.*(*30*). Most runs are performed at 5°C and 40% RH, experiments with different conditions are marked in **bold**. Steady state concentrations for runs 27 and 28 are approximated due to missing measurements and are marked with (•).

				Concentration (ppbv)		injected concentrations (pptv)			steady state concentration (ppt)		
System	**Run**	**RH (%)**	**Temp (°C)**	**NO_2**	**NO**	**α-pinene**	**isoprene**	**β-caryophyllene**	**α-pinene**	**isoprene**	**β-caryophyllene**
α-pinene	1*	40	5	0	0	600	0	0	323.3	0.0	0.0
	2*	40	5	0	0	1200	0	0	592.5	0.0	0.0
	3*	40	5	0	0	2400	0	0	1123.8	0.0	0.0
	4	40	5	0	0	1200	0	0	666.6	0.0	0.0
	5	40	5	0	0	2400	0	0	1165.7	0.0	0.0
α-pinene + isoprene	6	40	5	0	0	600	10000	0	310.2	3988.0	0.0
	7	40	5	0	0	1200	10000	0	609.8	3858.5	0.0
	8	40	5	0	0	2400	10000	0	1116.4	3584.6	0.0
β-caryophyllene	9	40	5	0	0	0	0	100	0.0	0.0	1.8
	10	40	5	0	0	0	0	200	0.0	0.0	3.3
	11	40	5	0	0	0	0	400	0.0	0.0	6.6
α-pinene + isoprene + β-caryophyllene	12	40	5	0	0	600	5000	100	300.0	1471.0	3.0
	13	40	5	0	0	1200	10000	200	585.3	2751.8	7.1
	14	40	5	0	0	2400	20000	400	974.1	4577.9	15.8
	15	40	**25**	0.26	0.01	600	5000	100	215.2	445.5	1.1
	16	40	**25**	0.24	0.01	1200	10000	200	468.3	851.6	2.3
	17	40	**25**	0.25	0.01	1200	10000	200	932.1	1994.2	5.1
	18	**65**	**25**	0.23	0.01	600	5000	100	241.0	437.7	3.8
	19	**65**	**25**	0.23	0.01	1200	10000	200	472.8	453.4	5.3
	20	**65**	**25**	0.23	0.02	2400	20000	400	947.1	564.9	9.5
	21	40	5	0.79	0.07	600	5000	100	235.5	1588.5	1.0
	22	40	5	0.35	0.03	600	5000	100	282.4	1780.9	1.1
	23	40	5	0.32	0.02	1200	10000	200	541.4	3389.2	2.3
	24	40	5	0.22	0.01	2400	20000	400	1049.0	6827.8	5.6
	25	40	5	0.21	0.01	1200	10000	200	670.7	4488.5	3.0
	26	40	5	0.25	0.01	1200	10000	200	662.4	4402.1	3.0
	27	**5**	5	0.23	0.00	1200	10000	200	665 •	9081.0	3.0 •
	28	**90**	5	0.31	0.02	1200	10000	200	665 •	1842.3	3.0 •

Table S 2 Volatilities for $C_{14}H_{22}O_x$ and $C_{15}H_{22}O_x$ oxidized compounds from FIGAERO thermogram analysis. Measured volatility of common $C_{14}H_{22}O_x$ and $C_{15}H_{22}O_x$ oxidation products of pure β-caryophyllene (orange) resulting in a β-caryophyllene monomer and of α-pinene + isoprene mixture (blue) resulting in an α-pinene+isoprene dimer. Dimers formed from α-pinene (C_{10}) and isoprene (C_5) result in $C_{14}H_{22}O_x$ and $C_{15}H_{22}O_x$ compounds which are less volatile than those formed from pure β-caryophyllene (C_{15}). Volatilities are given in units of $\mu g/m^3$ and cm^{-3} for each compound. The number densities in the table are calculated based on the individual molar mass of each compound.

	C* (β-caryophyllene:monomer)		C* (α-pinene+isoprene:dimer)	
	µg m^{-3}	cm^{-3}	µg m^{-3}	cm^{-3}
$C_{14}H_{22}O_7$			3.02E-04	6.02E+05
$C_{14}H_{22}O_8$	1.55E-03	2.93E+06	1.74E-04	3.29E+05
$C_{14}H_{22}O_9$	9.77E-04	1.76E+06	6.61E-05	1.19E+05
$C_{14}H_{22}O_{10}$	1.82E-04	3.13E+05	7.41E-06	1.27E+04
$C_{15}H_{22}O_6$	3.89E-03	7.85E+06		
$C_{15}H_{22}O_7$	2.14E-03	4.10E+06	7.94E-05	1.52E+05
$C_{15}H_{22}O_8$	1.74E-03	3.17E+06	6.31E-06	1.15E+04
$C_{15}H_{22}O_9$	6.76E-04	1.18E+06	5.89E-06	1.02E+04
$C_{15}H_{22}O_{10}$	5.50E-05	9.14E+04	3.16E-06	5.26E+03
$C_{15}H_{22}O_{11}$	2.09E-05	3.33E+04	5.50E-08	8.75E+01

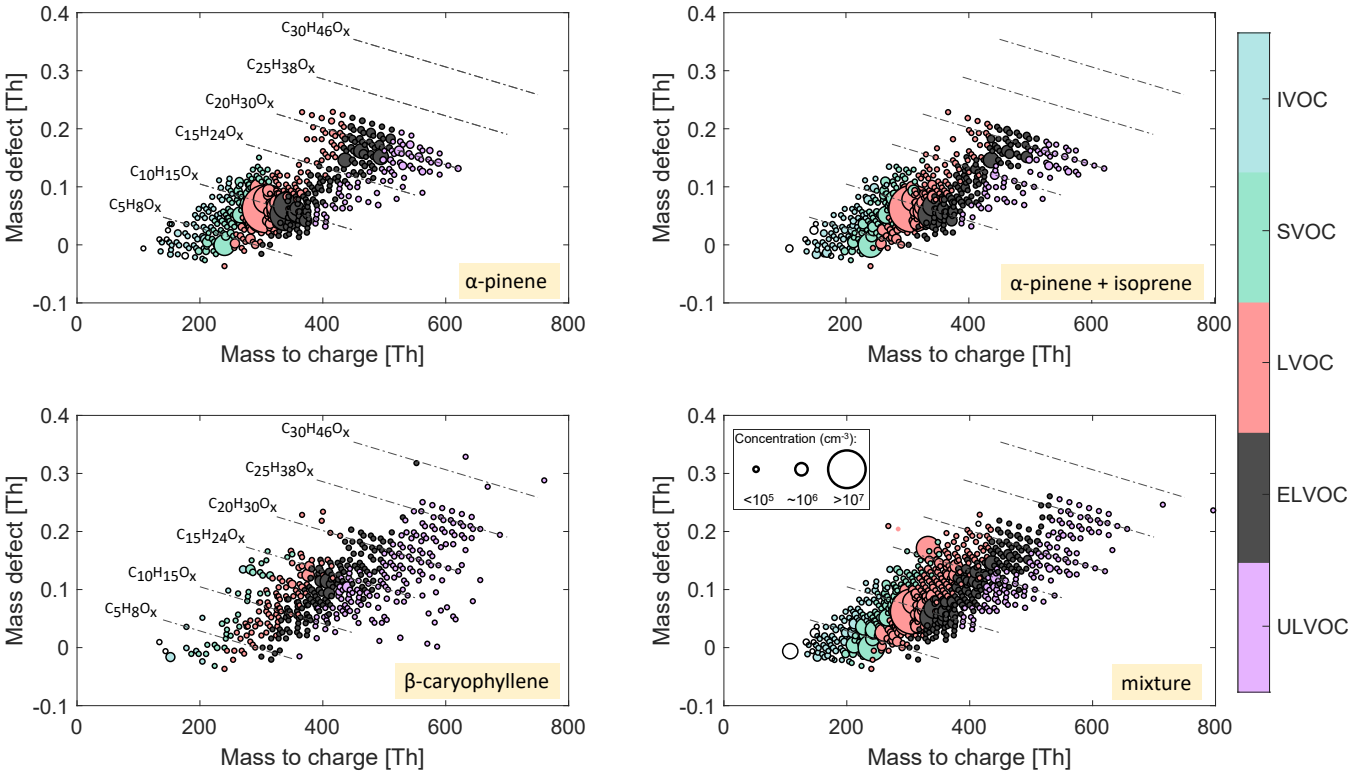

Figure S1 Chemical composition of BVOC oxidation results measured with NO_3-CIMS. Mass defect plots showing the oxidation products during different runs. Here we show example runs of pure components, α-pinene (run #4) and β-caryophyllene (run #10) as well as mixtures of α-pinene + isoprene (run #7) and α-pinene+isoprene+ β-caryophyllene (run #13).

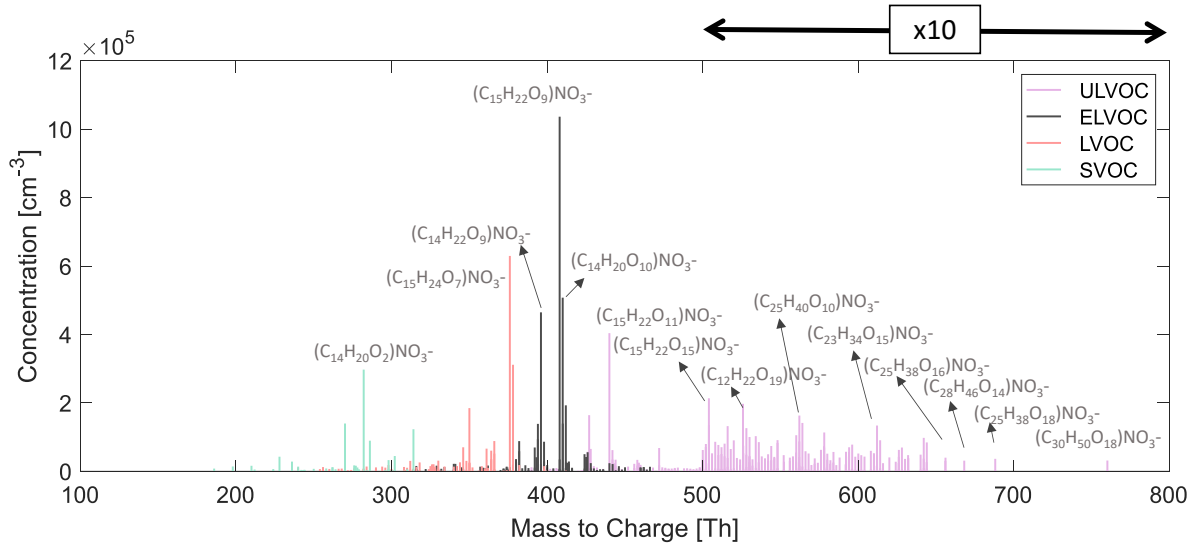

Figure S2 Chemical composition of β-caryophyllene oxidation products. Mass spectrum of β-caryophyllene oxidation products measured with NO_3-CIMS. Here we show β-caryophyllene ozonolysis run #10. Concentrations of compounds exceeding 500 Th are multiplied by 10.

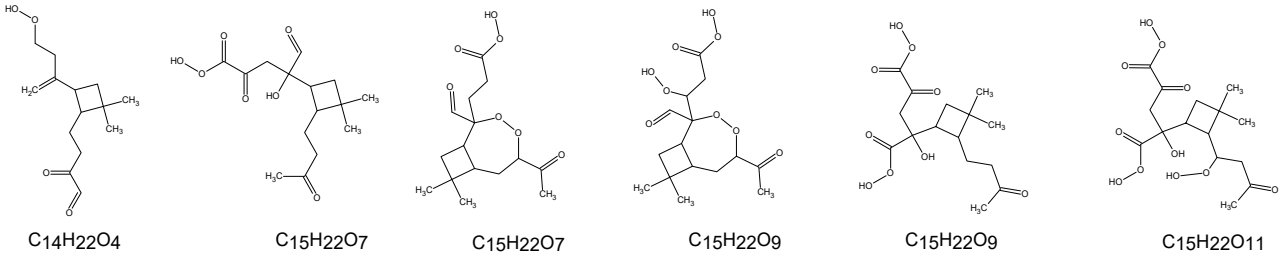

Figure S3 Assumed structures of β-caryophyllene oxidation products: $C_{14}H_{22}O_x$ and $C_{15}H_{22}O_x$. The β-caryophyllene autoxidation product structures were drawn according to prototypical reaction schemes based on previous literature on OOM formation in similar systems (*26,74-76*) and were used in the subsequent SIMPOL model. Briefly, the β-caryophyllene autoxidation subsequent to decomposition of the primary ozonide is propagated by peroxy radical H-shift reactions followed by O_2 additions, with the aldehydic and tertiary hydrogens being easiest to abstract, and the second double bond leading to rapid bridging of O_2 (i.e., endoperoxidation). The oxidation sequence is terminated by abstraction of a hydrogen from a carbon containing hydroperoxy functionality (i.e., C-OOH), leading to ejection of an OH and formation of a carbonyl functional group.

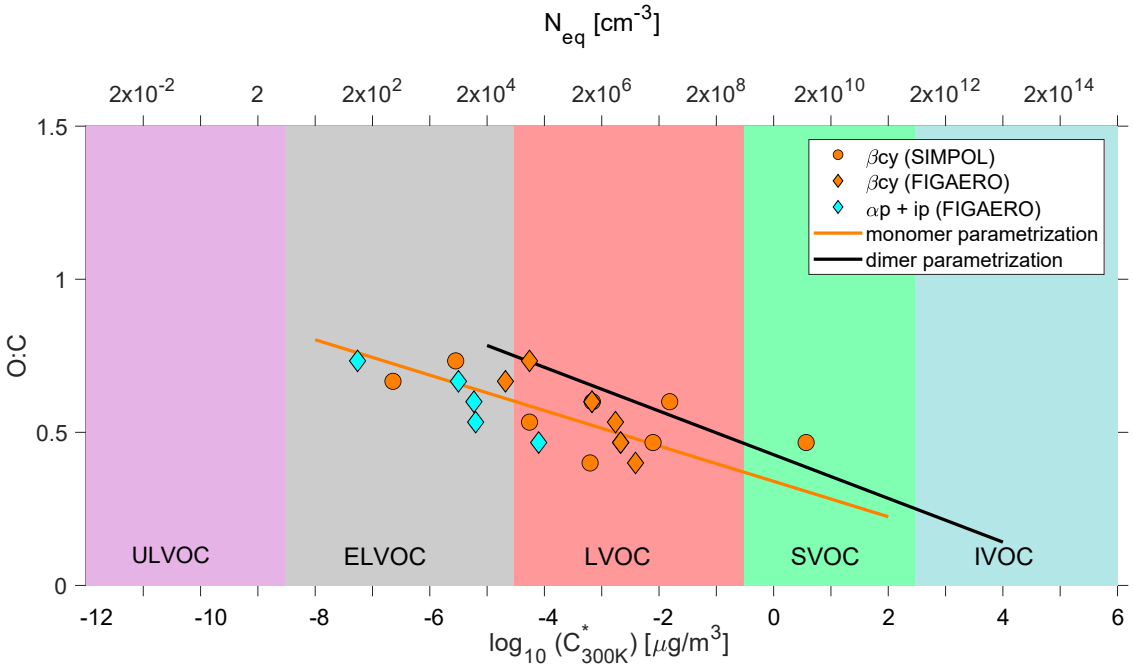

Figure S4 Volatility model at 300K used in this study. The volatility is calculated by the group-contribution method SIMPOL for $C_{15}H_{22}O_x$. We used the structures shown in Fig. S3 to know the functional groups to use SIMPOL (C_{15} monomer and C_{15} dimer, in the BVOC mixture most of the C_{15} are from the cross dimer). This figure validates our parametrization that in the BVOC mixture C_{15} species are monomers. The parametrization can be used if up to C_{15} are monomers. The FIGAERO confirms that the moleculesare closer to the monomer line. Species larger than C_{15} are dimers which include the peroxide groups.

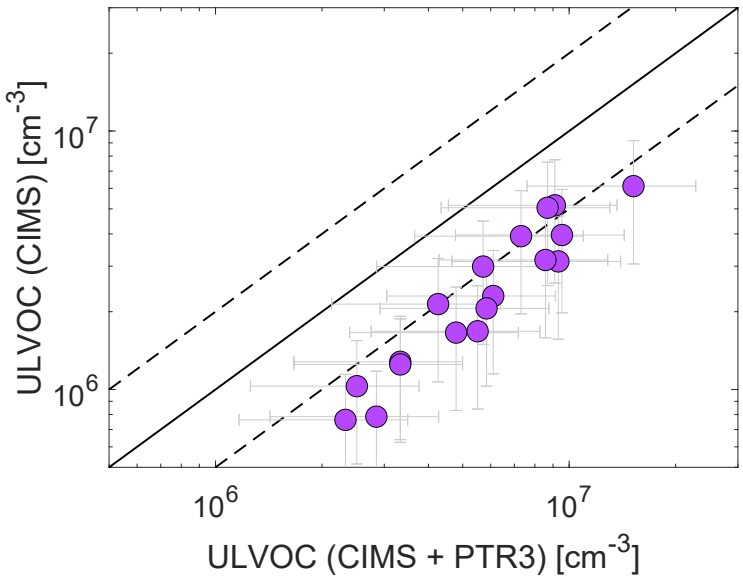

Figure S5 Concentration of ULVOC parametrized from NO_3-CIMS data only as a function of ULVOC acquired from NO_3-CIMS and PTR3 data combined. The difference appears to be within the error margin, an average of a factor of two higher when including the PTR3 data.

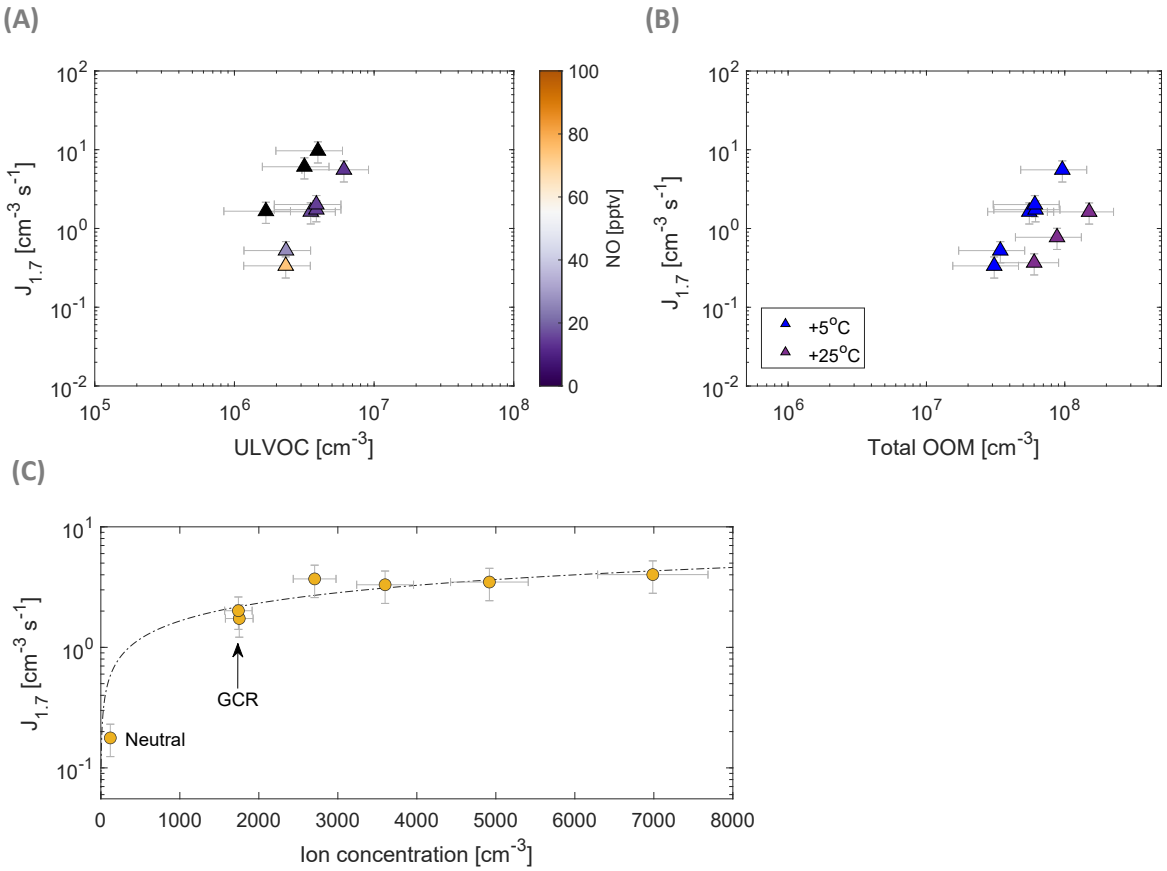

Figure S6 Effect of atmospheric conditions on particle formation rate. (A) Mixture runs (α-pinene+isoprene+β-caryophyllene) at temperature = 5°C and RH = 40%. Particle formation rate ($J_{1.7}$) as a function of ULVOC concentration, without NO are shown in black and coloured by NO concentration (colour bar). (B) Mixture runs (α-pinene+isoprene+ β-caryophyllene), RH = 40% and ~10 - 20 pptv NO, particle formation rate ($J_{1.7}$) as a function of OOM showing the temperature effect. (C) Mixture runs (α-pinene+isoprene+ β-caryophyllene), RH = 40% and ~10 - 20 pptv NO, particle formation rate ($J_{1.7}$) as a function of increasing ion concentration showing the importance of ions for pure biogenic nucleation. Neutral runs were achieved by turning on the high voltage field in the chamber, while increased ion concentration runs were performed using the adjustable π+ beam from the CERN Proton Synchrotron, see Methods section.

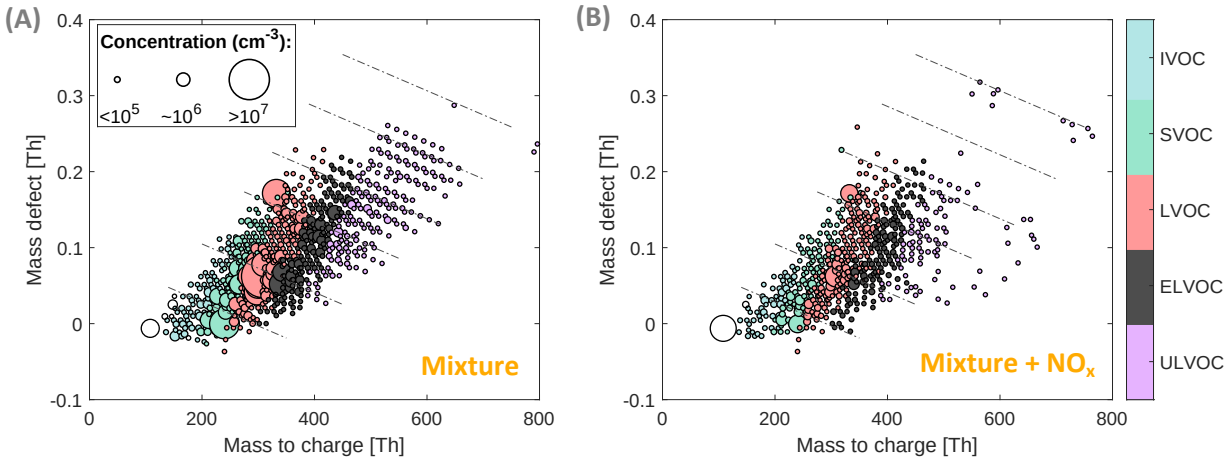

Figure S7 Chemical composition of BVOC oxidation results measured with NO_3-CIMS in the absence and presence of NOx. Here we show example of (A) mixture runs α-pinene+isoprene+ β-caryophyllene (run #14) and (B) α-pinene+isoprene+ β-caryophyllene with ~30 pptv NO (run #22).

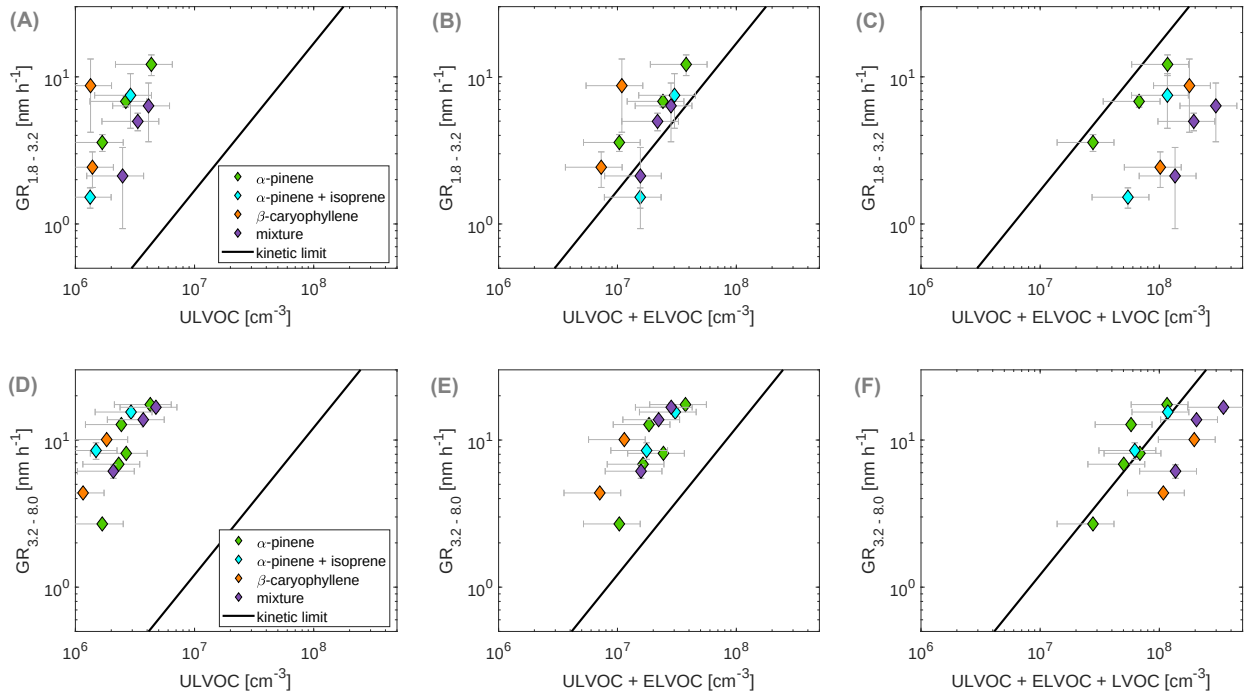

Figure S8 Particle growth rates as a function of vapour concentrations in different volatility classes. Particle growth rates during ozonolysis experiments for particle diameters between 1.8 and 3.2 nm as a function of (A) ULVOC, (B) ELVOC + ULVOC and (C) LVOC + ELVOC + ULVOC concentrations measured with NO_3-CIMS and PTR3 and particle growth rates during ozonolysis experiments for particle diameters between 3.2 and 8 nm as a function of (D) ULVOC, (E) ELVOC + ULVOC and (F) LVOC + ELVOC + ULVOC. The black line shows the geometric limit of kinetic condensational growth (*41*).

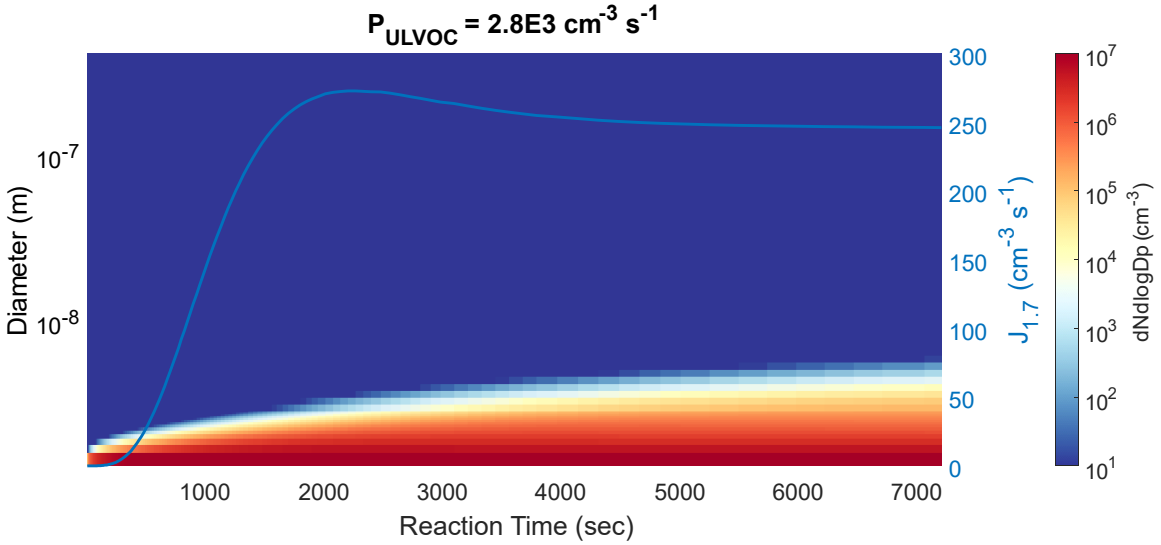

Figure S9 Model results example of kinetic limit ULVOC nucleation at constant production rate of ULVOC = 2.8x10^3 cm^{-3} s^{-1}. The calculation excludes evaporation to simulate the kinetic limit. The particle concentration is given as contour and $J_{1.7}$ as blue line (right axis).

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