

Observations of sesquiterpenes and their oxidation products in central Amazonia during the wet and dry seasons

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SI Tables

5 **Table S1: Composition by percent mass of total sesquiterpenes (% Σ SQT) and percent contribution to total ozone reactivity (% $\Sigma kO_3[SQT_i]$) from sesquiterpene ozonolysis at measurement site and in three commercial essential oils. Concentrations of each species estimated by using an average instrument response factor for several sesquiterpene standards unless otherwise noted that an authentic standard was used. For unidentified species, average kO_3 of all sesquiterpene species (from experimental determination or estimated as in Table 1) used for estimated % contribution to total ozone reactivity. BDL = Below Detection Limit, N/A = Not Applicable. Top three values for each column in bold print.**

Sample:	Feb 13 2014 06:47 AM UTC		Copaiba Oil		Copaiba Oil		Copaiba Oil		Andiroiba Oil	
Location/Origin:	T3, Manacapuru, AM, Brazil		Young Living Essential Oil, (origin Brazil)		Manacapuru, AM, Brazil		Bolivia		Manacapuru, AM, Brazil	
SQT	% Σ SQT	% ΣkO_3 [SQT _i]	% Σ SQT	% ΣkO_3 [SQT _i]	% Σ SQT	% ΣkO_3 [SQT _i]	% Σ SQT	% ΣkO_3 [SQT _i]	% Σ SQT	% ΣkO_3 [SQT _i]
α -copaene	2.5%	0.7%	5.6%	0.1%	4.5%	0.3%	4.8%	0.2%	6.3%	0.2%
cyperene	4.6%	4.0%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
β -gurjunene	1.4%	0.2%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
rimuene	6.4%	0.8%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
cis-calamene	9.2%	7.9%	1.1%	0.1%	5.0%	1.0%	3.3%	0.4%	BDL	N/A
α -cubebene	3.1%	2.2%	2.6%	0.2%	BDL	N/A	BDL	N/A	BDL	N/A
unidentified	1.2%	1.0%	BDL	N/A	4.9%	1.0%	10.9%	1.3%	14.8%	1.5%
cyclosativene	2.2%	0.3%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
β -elemene	2.5%	0.1%	1.1%	0.0%	2.5%	0.0%	2.5%	0.0%	BDL	N/A
α -cedrene	BDL	BDL	BDL	N/A	2.4%	0.0%	3.5%	0.0%	6.4%	0.0%
unidentified	2.8%	2.4%	BDL	N/A	3.9%	0.8%	7.4%	0.9%	7.5%	0.8%
unidentified	0.7%	0.6%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
unidentified	BDL	BDL	BDL	N/A	1.1%	0.4%	1.8%	0.4%	BDL	N/A
α -patchoulene	2.5%	0.3%	BDL	N/A	BDL	N/A	0.6%	0.0%	BDL	N/A
SQT202_A	0.7%	0.6%	BDL	N/A	BDL	N/A	1.1%	0.1%	BDL	N/A
γ -muurolene	4.3%	3.1%	1.8%	0.1%	BDL	N/A	BDL	N/A	BDL	N/A
α -amorphene	1.1%	1.6%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
β -selinene	3.4%	0.1%	2.0%	0.0%	13.8%	0.1%	3.1%	0.0%	9.1%	0.0%
α -muurolene	9.0%	12.9%	BDL	N/A	5.8%	1.9%	0.6%	0.1%	BDL	N/A
unidentified	5.7%	4.9%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
β -bisabolene	2.9%	4.2%	BDL	N/A	16.3%	5.5%	5.0%	1.0%	BDL	N/A
γ -cadinene	3.4%	2.5%	1.7%	0.1%	5.7%	1.0%	3.0%	0.3%	BDL	N/A
δ -cadinene	13.2%	35.7%	8.1%	1.9%	2.0%	1.2%	1.1%	0.4%	BDL	N/A
selinene <7-epi- α >	2.6%	7.0%	1.3%	0.3%	BDL	N/A	BDL	N/A	BDL	N/A
γ -cuprenene	0.6%	0.5%	BDL	N/A	BDL	N/A	0.6%	0.1%	BDL	N/A
α -cadinene	0.5%	0.8%	0.6%	0.1%	0.7%	0.2%	0.7%	0.1%	BDL	N/A
unidentified	BDL	BDL	0.6%	0.0%	BDL	N/A	0.7%	0.1%	BDL	N/A
selina-3,7(11)-diene	0.7%	1.9%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
pimaradiene	0.6%	0.1%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
sandaracopimaradiene	5.5%	0.7%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
kaurene	2.1%	0.0%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
SQT202_B	1.3%	1.2%	0.6%	0.0%	1.7%	0.3%	2.3%	0.3%	BDL	N/A
SQT202_C/cuparene	0.7%	0.6%	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A

isocaryophyllene	BDL	N/A	2.6%	0.2%	6.2%	1.1%	7.3%	0.7%	10.9%	1.0%
β -caryophyllene	BDL	N/A	56.2%	95.0%	17.8%	80.2%	32.9%	88.2%	41.5%	95.9%
trans- α -bergamotene	BDL	N/A	1.0%	0.1%	1.0%	0.3%	4.3%	0.9%	3.5%	0.6%
α -humulene	BDL	N/A	0.7%	1.1%	0.7%	3.3%	1.6%	4.3%	BDL	N/A
(-) alloaromadendrene	1.4%	0.0%	5.9%	0.0%	BDL	N/A	BDL	N/A	BDL	N/A
unidentified	BDL	N/A	1.2%	0.1%	BDL	N/A	BDL	N/A	BDL	N/A
unidentified	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
β -bisabolene	BDL	N/A	0.8%	0.1%	3.9%	1.3%	0.7%	0.1%	BDL	N/A
α -selinene	BDL	N/A	3.0%	0.2%	BDL	N/A	BDL	N/A	BDL	N/A
valencene- eremophilene	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A
(+) longifolene	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A	BDL	N/A

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Table S2: Comparison of % contribution of three sesquiterpenes: β -caryophyllene, α -copaene, and β -elemene to total sesquiterpenes in ambient air, essential oils, and tissue of Amazonian trees. Major component of sesquiterpene compounds also listed for reference. References listed as footnotes.

Analyte ^{Ref.}	Relative abundance raw integrated area basis				Relative abundance mass quantified basis			
	% β -caryophyllene	% α -copaene	% β -elemene	Major component	% β -caryophyllene	% α -copaene	% β -elemene	Major component
Ambient air, T3, 13 Feb 2014 06:47 AM UTC ¹	BDL	12.6	2.5	δ -cadinene	BDL	2.5	2.5	δ -cadinene
Copaiba oil, Manacapuru, Brazil ²	6.9	17.5	1.9	α -copaene	17.8	4.5	2.5	β -caryophyllene
Copaiba oil, Bolivia ²	12.4	18.3	1.9	α -copaene	32.9	4.8	2.5	β -caryophyllene
Andiroba oil, Manacapuru Brazil ²	13.7	20.8	BDL	α -cedrene	41.5	6.3	BDL	β -caryophyllene
Copaiba oil, Brazil, Young Living Essential Oils ²	30.0	30.0	1.0	α -copaene	52.5	5.3	1	β -caryophyllene
Copaifera officinalis, three-week old seedlings (leaves) ²	N/A	N/A	N/A	N/A	5.8	0.5	0.3	Germacrene D
Copaifera officinalis, three-week old seedlings (stems) ³	N/A	N/A	N/A	N/A	15.1	0.5	0.4	Germacrene D
Copaifera officinalis, three-week old seedlings (roots) ³	N/A	N/A	N/A	N/A	82.6	BDL	ND	β -caryophyllene
Copaifera officinalis, two-year old trees (leaves) ³	N/A	N/A	N/A	N/A	12	1.1	1	Germacrene D
Copaifera officinalis, two-year old trees (stems) ³	N/A	N/A	N/A	N/A	18	1	0.5	Germacrene D
Copaifera officinalis, two-year old trees (roots) ³	N/A	N/A	N/A	N/A	67.7	1.7	3.9	β -caryophyllene
Copaifera officinalis, three-week old seedlings injured, emissions ³	N/A	N/A	N/A	N/A	21.6	1.2	1.9	Germacrene D
Rosewood oil, <i>Aniba rosaedora</i> Ducke ³	3.4	16.3	6.3	α -copaene	N/A	N/A	N/A	N/A
C1 Commercial copaiba oil, Tarauacá, Acre state, Brazil ⁴	56.3	2.0	1.0	β -caryophyllene	N/A	N/A	N/A	N/A
C2 Commercial copaiba oil, Tarauacá, Acre state, Brazil ⁵	26.5	N/A	N/A	β -caryophyllene	N/A	N/A	N/A	N/A
C3 Commercial copaiba oil,	9.9	32.6	4.3	α -copaene	N/A	N/A	N/A	N/A

¹ This study

² Chen et al., 2009

³ Fidelis et al., 2011

⁴ Soares et al., 2013

Tarauacá, Acre state, Brazil ⁵								
C4-VF (volatile fraction, sesquiterpenes only content of C4 copaiba oil), Baía do Portel, PA, Brazil ⁵	42.4	11.2	2.0	β -caryophyllene	N/A	N/A	N/A	N/A
Mean	22.4	17.9	2.6		33.4	2.7	1.7	
Standard Deviation	17.9	9.4	1.8		24.9	2.1	1.2	
Median	13.7	17.5	2.0		21.6	1.7	1.5	

Table S3: Peaks attributed to BVOC oxidation products in shown in GCxGC chromatogram of main text (Figure 7). Best MS matches shown. Library names and source of MS are listed in Table S4.

Compound Name/Description	d-alkane LRI_I	d-alkane Library LRI_I	Best Match Library_Name	Library_Match_Factor	Library_Reverse Match_Factor	RTI_min	RTII_sec	Assigned Source Category
ISOP 2-methyltetrol 1	1574	1572	soasox	943	944	37.53	0.65	ISOPOX
ISOP isoprene SOA 1, Surratt 2006	1475	1475	soasox	921	921	34.09	0.79	ISOPOX
ISOP 2-methyltetrol 2	1555	1554	soasox	895	895	36.86	0.64	ISOPOX
ISOP isoprene SOA 2, Surratt 2006	1490	1489	soasox	888	890	34.60	0.82	ISOPOX
ISOP C5alkenetriol 1	1413	1413	soasox	859	868	31.94	0.61	ISOPOX
ISOP C5alkenetriol 2	1407	1406	soasox	786	806	31.70	0.64	ISOPOX
	1525	1523	limonox	855	856	35.81	0.74	MTOX
MT 2-hydroxyglutaric acid	1598	1597	soasox	897	898	38.35	0.98	MTOX
MT MBTCA	1776	1778	soasox	893	894	43.97	1.09	MTOX
	1615	1611	myrcox	815	818	38.89	1.26	MTOX
MT MBTCA isomer	1852	1853	soasox	803	810	46.20	1.14	MTOX
	1680	1679	copox	821	828	40.93	1.13	MTOX
MT pinic acid	1693	1693	soasox	778	796	41.35	1.09	MTOX
MT cis-norpinic acid	1630	1632	soasox2	833	840	39.36	1.98	MTOX
MT 3-hydroxyglutaric acid	1602	1602	soasox	774	785	38.50	1.00	MTOX
MT 3-hydroxy-4,4-dimethylglutaric acid	1637	1636	soasox	818	823	39.60	0.92	MTOX
MT Tricarballic acid	1757	-1	mainlib	704	731	43.35	1.16	MTOX
	1372	1335	myrcox	800	804	30.38	1.11	MTOX
MT 2,3-dihydroxy-4-oxo pentanoic acid	1533	1532	soasox	684	709	36.08	1.17	MTOX
	1653	1653	copox	914	914	40.10	1.35	SQTOX
β -caryophyllene aldehyde, TMS	1714	1714	bcpox2	899	900	42.02	2.21	SQTOX
	1763	1764	soasox	891	892	43.54	1.06	SQTOX
	1678	1677	copox	883	885	40.89	0.76	SQTOX
	1870	1870	ahumox2	882	883	46.71	0.71	SQTOX
	1744	1746	soasox2	867	870	42.96	1.58	SQTOX
β -nocaryophyllone aldehyde	1754	1757	soasox	861	864	43.27	0.49	SQTOX
	1582	1581	bcpox2	836	847	37.80	1.18	SQTOX
	1843	1850	bcpox3	681	687	45.93	1.14	SQTOX
α -copaene oxidation product	1719	1722	acopox	844	848	42.18	1.88	SQTOX
	2002	2001	copox	794	802	50.42	1.07	SQTOX

Compound Name/Description	d-alkane LRI_I	d-alkane Library LRI_I	Best Match Library_Name	Library Match_Factor	Library Reverse Match_Factor	RTI_min	RTII_sec	Assigned Source Category
	1790	1791	soasox	799	817	44.40	1.05	SQTOX
	1754	1753	aromox2	815	826	43.27	0.98	SQTOX
	2052.63	2053	copox	702	702	51.75	1.05	SQTOX
	1824.83	1824	ahumox2	813	817	45.42	0.66	SQTOX
	1983.45	1985	bcpox2	761	773	49.91	1.06	SQTOX
	1600	-1	mainlib	779	832	38.42	0.97	SQTOX
	1842.76	1844	acopox	767	773	45.93	1.05	SQTOX
	1797.52	1798	bcpox2	788	799	44.64	1.78	SQTOX
	2007.52	2005	bcpox3	794	820	50.58	0.64	SQTOX
	1726.71	1727	bcpox3	785	795	42.41	1.56	SQTOX
	1791.3	-1	mane2010.hp	651	667	44.44	1.62	SQTOX
	1875.86	1876	bfarnox	560	604	46.86	0.94	SQTOX
	1767.7	1768	ahumox	754	761	43.70	0.89	SQTOX
	1826.21	1821	soasox2	688	698	45.46	1.17	SQTOX
	1827.59	1838	bcpox2	626	647	45.50	1.22	SQTOX
	1846.9	1847	ahumox2	615	628	46.04	0.96	SQTOX
	1856.55	1855	bfarnox	656	673	46.32	1.20	SQTOX
β -nocaryophyllonic acid, TMS	1984.83	-1	bcpox	698	714	49.95	1.87	SQTOX
	1862.07	1872	acedox2	592	595	46.47	1.13	SQTOX
	1871.72	1872	aromox2	613	635	46.75	1.36	SQTOX
	1920	1919	ahumox	779	809	48.11	0.64	SQTOX
	1800	1800	ahumox	561	616	44.71	0.88	SQTOX
	1831.72	1828	acopox	568	585	45.61	0.86	SQTOX
	1808.28	1808	bcpox2	617	639	44.95	1.23	SQTOX
	1863.45	1864	soasox2	662	676	46.51	1.34	SQTOX
	2021.05	2020	bcpox2	655	683	50.93	1.09	SQTOX
	2028.57	2029	copox	658	664	51.12	1.08	SQTOX
	1878.62	1879	acedox	548	557	46.94	1.18	SQTOX
β -caryophyllonic acid, TMS	1931.03	1932	bcpox	663	692	48.43	1.43	SQTOX
	2061.65	2063	bcpox2	562	582	51.98	1.06	SQTOX
	1877.24	1879	soasox	890	890	46.90	0.74	TERPOX
	1633.54	1633	ahumox	882	883	39.48	1.57	TERPOX
	1786.34	1786	copox	891	894	44.28	0.75	TERPOX

Compound Name/Description	d-alkane LRI I	d-alkane Library LRI I	Best Match Library Name	Library Match Factor	Library Reverse Match Factor	RTI_min	RTII_sec	Assigned Source Category
	1479.78	1479	soasox2	870	873	34.24	1.31	TERPOX
	1513.48	1510	apinox	793	804	35.42	1.65	TERPOX
	1603.73	1602	soasox2	799	808	38.54	0.73	TERPOX
	1680.75	1682	soasox	820	829	40.96	2.30	TERPOX
	1503.37	1504	soasox	805	812	35.06	0.73	TERPOX
	1503.37	1504	soasox	825	835	35.06	0.70	TERPOX
	1561.8	1538	mainlib	669	900	37.10	1.22	TERPOX

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Table S4: Custom MS Library Names referred to in Table S3 with sources of mass spectra. Experimental conditions for sesquiterpene oxidation chamber experiments provided in Table S5.

Custom MS Library Name	Source of MS^{Previous Publication}
acedox	α -cedrene + O ₃
acedox2	α -cedrene + NO _x + hv ¹
acopox	α -copaene + O ₃ ²
ahumox	α -humulene + O ₃
ahumox2	α -humulene + NO _x + hv ⁶
apinox	α -pinene + O ₃ ³
apinox2	α -pinene + O ₃ ³
aromox	aromadendrene + O ₃ ⁷
aromox2	aromadendrene + NO _x + hv
bcpox	Synthesized β -caryophyllene oxidation products ⁴ : β -caryophyllene aldehyde, β -nocaryophyllone aldehyde, β -caryophyllonic acid, β -nocaryophyllonic acid, β -caryophyllinic acid, β -nocaryophyllinic acid
bcpox2	β -caryophyllene + NO _x + hv ⁷
bcpox3	β -caryophyllene + NO _x + acidic seed + hv
bfarnox	β -farnasene + NO _x + hv ⁷
copox	copaiba oil + O ₃
limonox	limonene + O ₃ ³
mainlib	NIST 14 Library
mane2010.hp	proprietary library from a flavour and fragrance company (MANE)
myrcox	myrcene + O ₃ ³
soasox/soasox2	Previously identified compounds in filters from Southern Oxidant and Aerosol Study (SOAS) ³

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¹ Jaoui, M., Kleindienst, T. E., Docherty, K. S., Lewandowski, M. and Offenberg, J. H.: Secondary organic aerosol formation from the oxidation of a series of sesquiterpenes: alpha-cedrene, beta-caryophyllene, alpha-humulene and alpha-farnesene with O-3, OH and NO3 radicals, Environ. Chem., 10(3), 178–193, doi:10.1071/en13025, 2013.

² Offenberg, J. H., Lewandowski, M., Kleindienst, T. E., Docherty, K. S., Jaoui, M., Krug, J., Riedel, T. P. and Olson, D. A.: Predicting Thermal Behavior of Secondary Organic Aerosols, Environ. Sci. Technol., 51, 9911–9919, doi:10.1021/acs.est.7b01968, 2017.

³ Zhang et al., PNAS

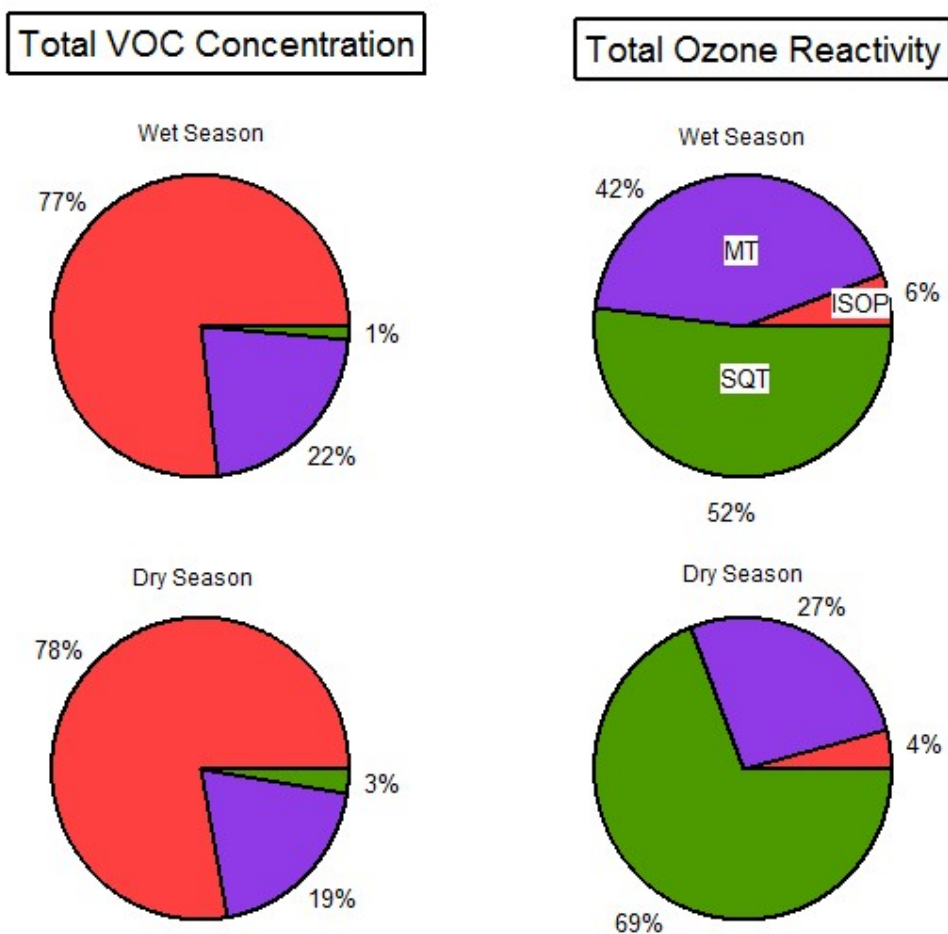
⁴ Be, A. G., Upshur, M. A., Liu, P., Martin, S. T., Geiger, F. M., Thomson, R. J. and Paulson, J. A.: Cloud Activation Potentials for Atmospheric α -Pinene and β -Caryophyllene Ozonolysis Products, ACS Cent. Sci., 3(7), 715–725, doi:10.1021/acscentsci.7b00112, 2017.

Table S5: Experimental conditions for sesquiterpene ozonolysis/photolysis experiments performed by U.S. EPA and used for making custom MS libraries as listed in Table S4.

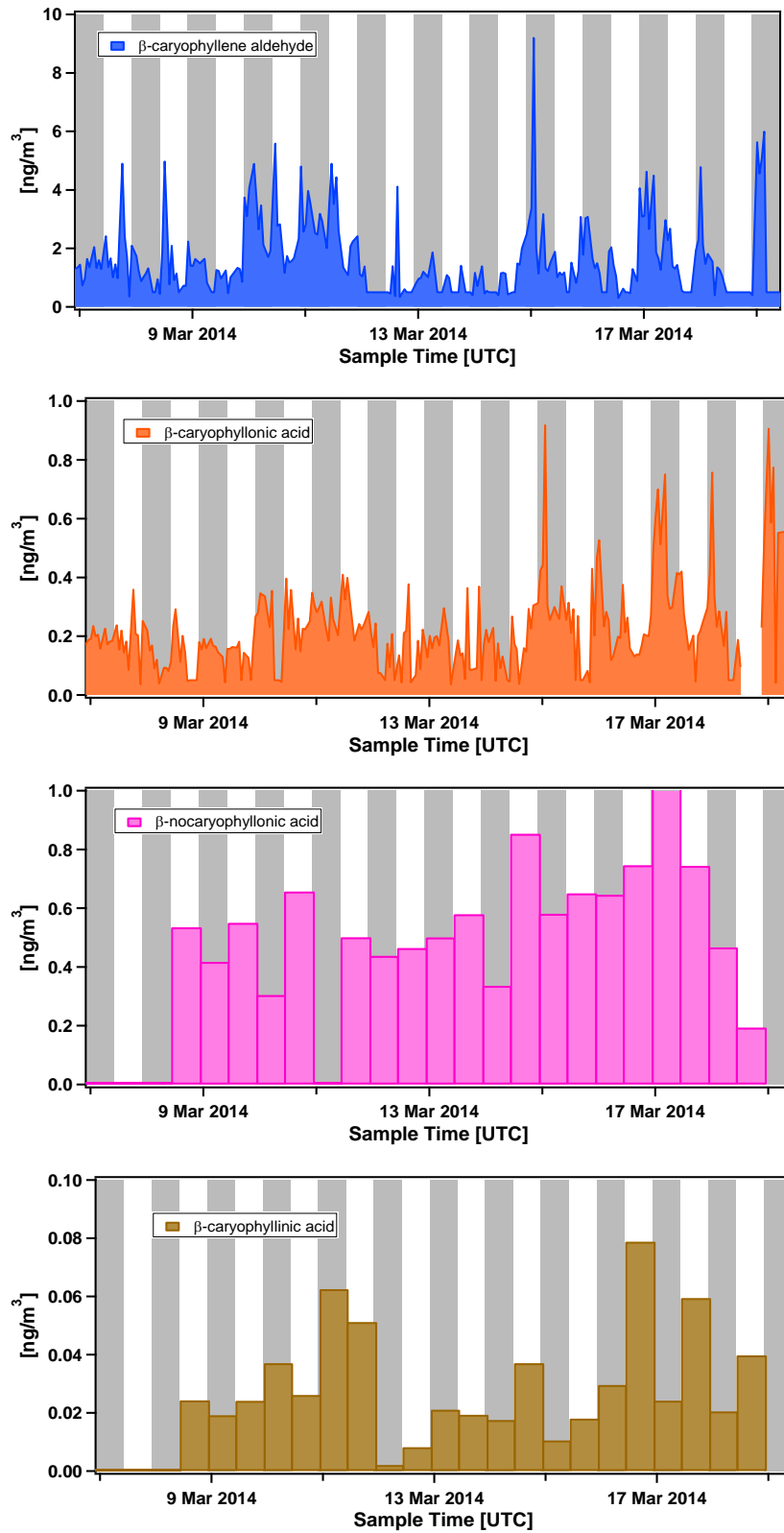
Custom MS Library Name	experiment ID	temp (°C)	RH (%)	hydrocarbon (HC)	initial/final [HC] (ppmC)	initial/steady-state [NO] (ppm)	initial/steady-state [O ₃] (ppm)	SOA mass (µg m ⁻³)	OM/OC
acedox	ER721	22.3	31.6	α-cedrene	0.68/0.10	N/A	0.203/0.166	230.5	1.4
acedox2	ER649	23.8	27.9	α-cedrene	0.64/0.39	0.108/0.059	0/0.002	33.9	1.4
acopox	ER734	22.1	31.7	α-copaene	0.66/0.01	N/A	0.213/0.156	271.3	1.4
ahumox	ER720	22.1	31.6	α-humulene	0.40/0.00	N/A	0.186/0.061	170.8	1.5
ahumox2	ER648	24.3	27.8	α-humulene	0.63/0.00	0.093/0.028	0/0.004	126.4	N/A
aromox	ER742	22.1	31.5	aromadendrene	0.65/0.06	N/A	0.201/0.147	87.8	1.4
aromox2	ER743	24.7	31.2	aromadendrene	0.66/0.34	0.055/0.039	0/0.004	43.7	1.5
bcpox2	ER716	24.7	29.3	β-caryophyllene	0.58/0.00	0.099/0.052	0/0.002	125.1	1.6
bcpox3	ER717	24.7	29.3	β-caryophyllene	0.54/0.03	0.103/0.061	0/0.002	160.1	1.9
bfarnox	ER711	24.8	31.1	β-farnesene	N/A	0.091/0.009	0/0.025	123.6	1.8
copox	ER722	22.2	31.6	Various in copaiba essential oil (Young Living Essential Oil)	0.25 ¹ /0.00	N/A	0.198/0.134	271.3	1.4

¹ Estimated from sum of all major (presumably SQT) peaks measured by GC-FID. Reported value will be underestimate of actual HC in experiment, thus SOA yield likely overestimated.

SI Figures



5 Figure S1: Average % of isoprene (ISOP, red), monoterpenes (MT, purple), and sesquiterpenes (SQT, green) during wet and dry seasons contributing to their summed VOC concentration and total ozone reactivity for chemical composition within/near the canopy. VOC concentrations are based on measurements from (Alves et al., 2016) and ozone reactivity calculated using monoterpene composition according to (Shu and Atkinson, 1994) and sesquiterpene composition according to copaiba essential oil (source: Young Living) in



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Figure S2: Selected timeline of gas-phase tracers (β -caryophyllene aldehyde, β -caryophyllonic acid) measured with SV-TAG and particle-phase tracers (β -nocaryophyllonic acid, β -caryophyllonic acid) from β -caryophyllene oxidation.

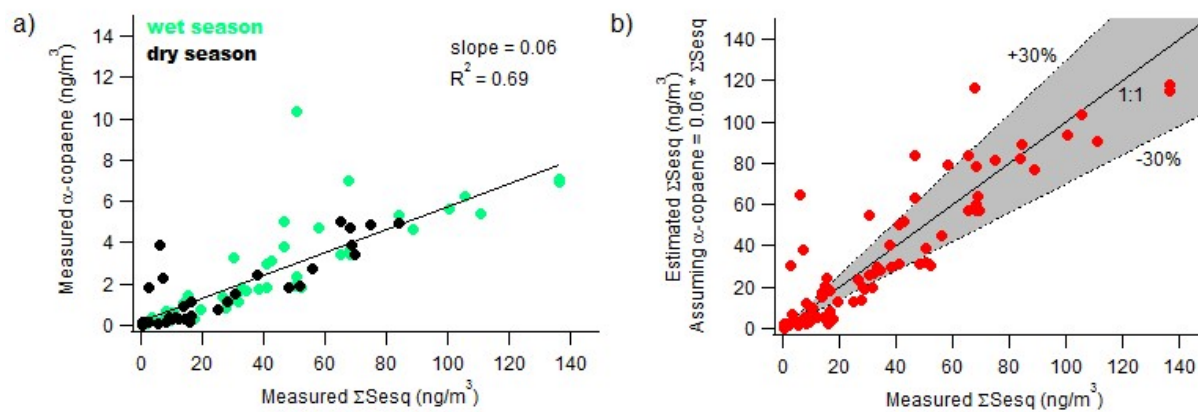


Figure S3: During underivatized runs ($n=79$), α -copaene on average accounts for 6% of total observed sesquiterpenes in panel a) within $\pm 30\%$ in panel b).