Supplement of

**Secondary Organic Aerosols from OH Oxidation of Cyclic Volatile Methyl Siloxanes as an Important
Si Source in the Atmosphere**

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Figure S1. Details of experimental setup

Table S1. Formula and structures of cyclic volatile methyl siloxanes

Name and Abbreviation	Formula	Structure
Hexamethylcyclotrisiloxane (D3)	$C_6H_{18}O_3Si_3$
Octamethylcyclotetrasiloxane (D4)	$C_8H_{24}O_4Si_4$
Decamethylcyclopentasiloxane (D5)	$C_{10}H_{30}O_5Si_5$
Dodecamethylcyclohexasiloxane (D6)	$C_{12}H_{36}O_6Si_6$

Table S2. Comparison of experimental results and conditions for D5-SOA in this study with those in other literatures

	D5-SOA yield	Reactor	D5 concentration	SOA concentration ($\mu\text{g m}^{-3}$)	Determination of OH concentration	Determination of reacted precursor	OH concentration (molecules cm^{-3})	OH exposure (molecules $\text{cm}^{-3} \text{ s}$)	Global average OH concentration (molecules cm^{-3})	Equivalent days	RH (%)	T ($^{\circ}\text{C}$)
Wu (2017)	0.08-0.16	photo- oxidation chamber (PC) (50 L)	5.4-13.3 ppbv	1.2-12	SO ₂ -OH	Calculation from the OH mixing ratio	10 ⁸	9×10 ¹⁰	—	—	8-10	27
Janecek (2019)	0.22	PAM OFR chamber (13.3 L)	502.55 $\mu\text{g m}^{-3}$	107.1	SO ₂ -OH	Solid-phase extraction (SPE) cartridges	—	1.6×10 ¹²	1.5×10 ⁶	12.4	25	24
	0.24		374.15 $\mu\text{g m}^{-3}$	84				2.3×10 ¹²		17.4		
	0.24		290.1 $\mu\text{g m}^{-3}$	68.4				2.7×10 ¹²		20.8		
	0.3		739.95 $\mu\text{g m}^{-3}$	219.7				4.8×10 ¹²		37.1	45	
	0.5		374.85 $\mu\text{g m}^{-3}$	180.7				5.1×10 ¹²		39.5		
This study (Low- NO _x and unseeded)	0.02± 0.02	ECCC- OFR	20-25 ppbv	0.5	MeOH-OH	PTR-MS	—	5.5×10 ¹⁰	1.5×10 ⁶	0.4	35±2	21± 1
	0.61± 0.03			169.7				9.0×10 ¹¹		7.0		
	0.70± 0.03			228.8				1.2×10 ¹²		9.0		
	0.75± 0.03			253.6				1.3×10 ¹²		10.2		
	0.79± 0.03			282.7				1.7×10 ¹²		12.9		
	0.80± 0.03			273.6				1.8×10 ¹²		14.2

Figure S2. The relationship between SOA yields and mass concentration for cVMS in low- NO_x experiments.

Figure S3. Comparison of low-NO_x (black) and high-NO_x (red) SOA yields for cVMS (a-d) for unseeded (square symbols) and seeded (circular symbols) experiments.

Figure S4. SOA yield enhancement ratio ($Y_{\text{seeded}}/Y_{\text{unseeded}}$) in the presence of seed particles. (a) Low-NO_x experiments; (b) high-NO_x experiments. The dotted lines represent $Y_{\text{seeded}}/Y_{\text{unseeded}}=1$.

Figure S5. HR-ToF-AMS mass spectra of D4 (a) and D5 (b) in high m/z range at OH exposure of 9.0×10^{11} molecules cm^{-3} s under low- NO_x conditions in unseeded experiments.

Figure S6. The weighted average values of Si/C (n/x) and Si/O (n/z) ratios for $\text{C}_x\text{H}_y\text{O}_z\text{Si}_n$ groups in SOA derived from the oxidation of D5 (a) and D6 (b) by OH radicals at different photochemical ages under low- NO_x and unseeded conditions.

Figure S7. HR-ToF-AMS mass spectra of cVMS SOA at OH exposure of 9.0×10^{11} molecules cm^{-3} s under high- NO_x conditions in unseeded experiments. (a-d) represent data of D3-D6, respectively.

Figure S8. Fraction of C_x , C_xH_y , $C_xH_yO_1$, $C_xH_yO_{>1}$ and $C_xH_yO_2Si_n$ groups for SOA derived from the oxidation of cVMS (a-d) by OH radicals at different photochemical ages under high- NO_x conditions.

Empty and solid triangles represent experimental data under unseeded and seeded conditions, respectively.

Figure S9. The weighted average values of Si/C (n/x) and Si/O (n/z) ratios for C_xH_yO_zSi_n groups in SOA

derived from the oxidation of D5 (a) and D6 (b) by OH radicals at different photochemical ages under

high-NO_x and unseeded conditions.

Table S3. Details about sites and concentrations of cVMS SOA. The concentrations of cVMS SOA were calculated on the basis of the cVMS concentrations reported from multiple literatures and SOA yields measured in this work. Here, the chosen concentrations of cVMS were maximum that have been reported in each site.

Site #	Site Name	Country	Site Type	D3 (ng/m ³)	D3-SOA (ng/m ³)	D4 (ng/m ³)	D4-SOA (ng/m ³)	D5 (ng/m ³)	D5-SOA (ng/m ³)	D6 (ng/m ³)	D6-SOA (ng/m ³)	Total SOA (ng/m ³)	Total Si-containing SOA (ng/m ³)	Fraction of Si-containing SOA (%)	References
1	Ny Alesund	Norway	PO	17	1.08	67	22.02	25	16.41	3.8	2.94	42.45	20.41	48.08	(Genualdi et al., 2011; Rauert et al., 2018)
2	Alert, NU	Canada	PO	13	0.83	72	23.66	26	17.06	39	30.16	71.71	35.47	49.47	
3	Barrow, AK	USA	PO	1.4	0.09	14	4.60	7.5	4.92	1.1	0.85	10.46	5.09	48.64	
4	Little Fox Lake, YK	Canada	BA	21	1.34	131	43.05	81	53.15	10	7.73	105.27	50.92	48.37	
5	West Branch, IA	USA	BA	—	—	14	4.60	29	19.03	2.3	1.78	25.41	12.29	48.36	(Yucuis et al., 2013)
6	Point Reyes, CA	USA	BA	16	1.02	66	21.69	35	22.97	3.7	2.86	48.54	23.33	48.06	(Genualdi et al., 2011; Rauert et al., 2018)
7	Hilo, HI	USA	BA	32	2.04	145	47.65	143	93.84	17	13.15	156.68	75.45	48.15	
8	Bratt's Lake, SK	Canada	BA	17	1.08	100	32.86	44	28.87	5.3	4.10	66.92	32.37	48.37	
9	Whistler,	Canada	BA	117	7.46	45	14.79	10	6.56	1.5	1.16	29.97	11.56	38.57	

	BC														
10	Fraserdale, ON	Canada	BA	15	0.96	53	17.42	16	10.50	2.8	2.17	31.04	14.89	47.96	
11	Ucluelet, BC	Canada	BA	81	5.16	121	39.76	31	20.34	3.5	2.71	67.97	31.28	46.02	
12	Sable Island, NS	Canada	BA	14	0.89	54	17.75	33	21.65	5.6	4.33	44.62	21.47	48.13	(Rauert et al., 2018)
13	Mount Revelstoke, BC	Canada	BA	2.4	0.15	30	9.86	33	21.65	9.1	7.04	38.70	18.87	48.77	(Genualdi et al., 2011; Rauert et al., 2018)
14	Tudor Hill	Bermuda	BA	8	0.51	76	24.98	56	36.75	66	51.05	113.28	56.21	49.62	
15	Storhofdi	Iceland	BA	3.5	0.22	14	4.60	13	8.53	1.5	1.16	14.51	6.98	48.08	
16	Malin Head	Ireland	BA	11	0.70	21	6.90	32	21.00	7.4	5.72	34.32	16.44	47.90	
17	Kosetice	Czech Republic	BA	25	1.59	61	20.05	297	194.89	169	130.71	347.24	170.07	48.98	
18	Cape Grim	Australia	BA	8.8	0.56	59	19.39	13	8.53	2.4	1.86	30.34	14.75	48.62	
19	Tibetan Plateau	China	BA	71.1	4.53	96.6	31.74	145.6	95.54	2.9	2.24	134.06	62.86	46.89	(Wang et al., 2018)
20	Toronto, ON	Canada	UR	18	0.32	77	6.73	247	82.96	22	13.22	103.23	28.81	27.91	(Krogseth et al.,

															2013; Ahrens et al., 2014)
21	Chicago, IL	USA	UR	—	—	190	16.60	1100	369.46	50	30.05	416.11	115.40	27.73	(Yucuis et al., 2013)
22	Cedar Rapids, IA	USA	UR	—	—	37	3.23	65	21.83	9.3	5.59	30.65	8.57	27.97	(Yucuis et al., 2013)
23	Sydney, FL	USA	UR	10	0.18	76	6.64	93	31.24	6.6	3.97	42.02	10.82	25.74	(Genualdi et al., 2011; Rauert et al., 2018)
24	Groton, CT	USA	UR	16	0.29	68	5.94	96	32.24	12	7.21	45.69	12.34	27.01	
25	Boulder, CO	USA	UR	—	—	—	—	65	21.83	—	—	21.83	6.01	27.52	(Coggon et al., 2018)
26	Zurich	Switzerla nd	UR	—	—	—	—	650	218.32	79	47.48	265.79	79.37	29.86	(Buser et al., 2013)
27	Catalan	Spain	UR	1166	21.01	676	59.07	1942	652.27	68	40.86	773.21	201.54	26.07	(Gallego et al., 2017)
28	Paris	France	UR	30	0.54	50	4.37	280	94.04	53	31.85	130.80	39.21	29.98	(Genualdi et al.,

															2011; Rauert et al., 2018)
29	Yantai	China	UR	——	——	22	1.92	38	12.76	——	——	14.69	3.69	25.10	(Xu et al., 2012)
30	Guangzhou	China	UR	11300	203.64	3300	288.34	——	——	——	——	491.97	26.70	5.43	(Wang et al., 2001)
31	Macau	China	UR	5800	104.52	4300	375.71	——	——	——	——	480.23	34.45	7.17	(Wang et al., 2001)
32	Foshan	China	UR	2300	41.45	3500	305.81	——	——	——	——	347.26	27.94	8.05	(Wang et al., 2001)
33	Dalian	China	UR	——	——	1950	170.38	1440	483.66	990	594.94	1248.98	390.18	31.24	(Li et al., 2020)
34	Kunming	China	UR	——	——	26	2.27	59	19.82	22	13.22	35.31	11.03	31.24	(Guo et al., 2019)
35	Lhasa	China	UR	44.8	0.81	54.6	4.77	464.6	156.05	1.3	0.78	162.41	43.70	26.91	(Wang et al., 2018)
36	Golmud	China	UR	26.8	0.48	48.6	4.25	208.1	69.90	2.8	1.68	76.31	20.31	26.61	(Wang et al., 2018)

Note: ‘——’ means that the data was not obtained for this site; ‘PO’ means polar sites; ‘BA’ means background sites; ‘UR’ means urban sites. Under the seeded conditions, the fractions

of $C_xH_yO_zSi_n$ in high- NO_x SOA at 8.5 equivalent days (D3- $C_xH_yO_zSi_n$: 0.002; D4- $C_xH_yO_zSi_n$: 0.091; D5- $C_xH_yO_zSi_n$: 0.275; D6- $C_xH_yO_zSi_n$: 0.406) were used to estimate Si-containing SOA concentrations at urban sites, whereas the fractions of $C_xH_yO_zSi_n$ in low- NO_x SOA under the unseeded conditions at 14.2 equivalent days (D3- $C_xH_yO_zSi_n$: 0.059; D4- $C_xH_yO_zSi_n$: 0.500; D5- $C_xH_yO_zSi_n$: 0.477; D6- $C_xH_yO_zSi_n$: 0.512) were applied in the calculation of Si-containing SOA at background and polar sites.

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