Supplement of

Organosulfates in atmospheric aerosols in Shanghai, China: seasonal and interannual variability, origin, and formation mechanisms

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Table S1. Recoveries of OS standards spiked in the blank filter.

	Limonaketone sulfate	α-Pinene sulfate	Δ-Carene sulfate	β-Caryophyllene sulfate	Octyl sulfate	Methyl sulfate	Phenyl sulfate	Camphor-sulfonate
Recovery	88.5%	88.7%	66.4%	84.2%	82.6%	88.0%	87.7%	94.3%

Table S2. The ratios of the signal response of OS standards in different sample extracts to that in pure solvent.

Species	Exp. 1	Exp. 2	Exp. 3	Exp. 4
Limonaketone sulfate	0.93	0.93	1.09	1.15
α-Pinene sulfate	0.90	0.98	1.02	1.06
Δ-Carene sulfate	0.81	0.91	1.02	1.08
β-Caryophyllene sulfate	0.96	1.08	1.07	1.10
Octyl sulfate	0.87	1.00	1.10	1.14
Methyl sulfate	0.20	0.16	0.51	0.55
Phenyl sulfate	1.01	0.96	1.06	1.12
Camphorsulfonate	0.89	1.07	1.06	1.11
Lactic acid sulfate	0.93	0.86	1.35	1.38
Glycolic acid sulfate	0.17	0.31	0.45	0.53

Exp. 1: 10 mg L^{-1} OS concentration and a polluted sample with 22 μg m^{-3} of OM, 34 μg m^{-3} of NO_3^-, and 15 μg m^{-3} of SO_4^{2-}; **Exp. 2:** 63 mg L^{-1} OS concentration and a polluted sample with 22 μg m^{-3} of OM, 34 μg m^{-3} of NO_3^- and 15 μg m^{-3} of SO_4^{2-}; **Exp. 3:** 63 mg L^{-1} OS concentration and a clean sample with 4 μg m^{-3} of OM, 0.4 μg m^{-3} of NO_3^- and 2 μg m^{-3} of SO_4^{2-}; **Exp. 4:** 63 mg L^{-1} OS concentration and a clean sample with 4 μg m^{-3} of OM, 0.7 μg m^{-3} of NO_3^- and 2 μg m^{-3} of SO_4^{2-}.

Table S3. Summary of average (standard deviation) values of meteorological parameters, trace gases, aerosol liquid water content (ALWC), aerosol [H^+], $PM_{2.5}$ and components of $PM_{2.5}$ in four seasons and throughout the year in 2015/2016 and 2018/2019.

	Spring		Summer		Autumn		Winter		Annual	
	2015	**2019**	**2015**	**2019**	**2015**	**2018**	**2016**	**2019**	**2015-2016**	**2018-2019**
T	16.6 (3.8)	17.2 (3.8)	30.2 (2.3)	30.5 (2.3)	16.4 (3.1)	17.9 (1.9)	7.2 (3.1)	6.2 (1.7)	18.2 (9.2)	18.4 (9.3)
RH	0.6 (0.1)	0.7 (0.1)	0.7 (0.1)	0.7 (0.1)	0.8 (0.1)	0.7 (0.1)	0.7 (0.1)	0.7 (0.1)	0.7 (0.1)	0.7 (0.1)
wind speed	3.8 (1.5)	4.0 (1.2)	4.4 (1.3)	4.4 (1.2)	3.6 (1.0)	3.4 (0.7)	3.3 (1.6)	3.4 (1.0)	3.8 (1.4)	3.8 (1.1)
NO_2	30.4 (11.5)	22.4 (6.1)	14.9 (5.2)	11.7 (4.0)	28.5 (7.4)	20.1 (7.1)	36.2 (14.1)	32.6 (9.9)	27.0 (13.0)	21.3 (10.3)
O_3	35.9 (8.9)	41.2 (11.0)	38.5 (18.9)	30.7 (13.8)	19.2 (8.8)	31.7 (7.8)	21.1 (9.5)	14.9 (6.7)	29.8 (15.2)	29.6 (13.9)
SO_2	6.3 (2.4)	2.2 (0.6)	3.5 (2.1)	1.5 (0.3)	4.8 (1.8)	3.5 (1.3)	8.4 (3.5)	2.8 (1.1)	5.7 (3.1)	2.5 (1.1)
ALWC	14.0 (15.4)	12.0 (8.5)	9.6 (4.5)	5.0 (3.3)	52.7 (43.7)	14.6 (32.4)	31.7 (16.8)	29.2 (19.8)	24.4 (27.0)	14.8 (20.4)
[H^+]	2.4E-4 (3.5E-4)	7.0E-5 (7.5E-5)	7.3E-3 (6.7E-3)	2.0E-3 (2.4E-3)	1.9E-4 (1.2E-4)	9.7E-5 (9.7E-5)	6.1E-5 (7.2E-5)	3.0E-3 (1.1E-2)	2.3E-3 (4.8E-3)	1.4E-3 (5.7E-3)
OM	15.9 (7.4)	10.6 (5.2)	7.4 (5.5)	6.2 (4.5)	11.5 (6.3)	8.3 (5.4)	16.2 (8.9)	11.1 (6.0)	12.7 (8.0)	9.0 (5.5)
EC	3.9 (1.7)	3.4 (1.2)	2.5 (1.1)	1.5 (0.8)	3.2 (1.2)	1.8 (0.8)	4.0 (1.8)	2.2 (1.0)	3.4 (1.6)	2.2 (1.2)
Cl^-	0.5 (0.7)	0.4 (0.2)	0.1 (0.1)	0.3 (0.2)	1.1 (0.8)	0.3 (0.2)	1.5 (1.0)	1 (0.6)	0.7 (0.9)	0.5 (0.4)
NO_3^-	9.4 (5.7)	9.9 (6.3)	1.0 (1.1)	3.4 (3.2)	9.6 (8.2)	6.7 (6.5)	16.6 (10.0)	14.1 (10.0)	8.8 (8.9)	8.4 (7.8)
SO_4^{2-}	8.9 (4.2)	5.3 (2.4)	7.4 (3.1)	4.2 (2.0)	9.4 (5.7)	4.0 (1.9)	11.1 (5.1)	9.2 (5.3)	9.1 (4.6)	5.7 (3.8)
NH_4^+	5.9 (3.1)	4.6 (2.6)	2.7 (1.7)	1.4 (1.1)	7.0 (4.8)	3.1 (2.6)	10.2 (5.1)	6.2 (4.1)	6.2 (4.6)	3.8 (3.3)
$PM_{2.5}$	59.1 (21.4)	44.6 (20)	30.8 (13.5)	22 (11.1)	59.7 (35.0)	34.2 (17.7)	91 (47.5)	55.9 (29.9)	59.0 (37.9)	38.6 (24.0)

Units: T (℃), wind speed (m s^{-1}), NO_2 (ppb), O_X (ppb), ALWC (μg m^{-3}), aerosol [H^+] (mol L^{-1}), OM (μg m^{-3}), EC (μg m^{-3}), Cl^- (μg m^{-3}), NO_3^- (μg m^{-3}), SO_4^{2-} (μg m^{-3}), NH_4^+ (μg m^{-3}) and $PM_{2.5}$ (μg m^{-3})

Table S4. Summary of individual organosulfate concentration (in ng m^{-3}) in four seasons in 2015/2016 and 2018/2019.

Category	Formula	Spring		Summer		Autumn		Winter	
	$[M-H]^-$	2015	2019	2015	2019	2015	2018	2016	2019
C_2/C_3 OS	$C_3H_5O_4S^-$	0.53	0.35	0.67	0.82	0.59	0.28	0.67	0.48
	$C_2H_3O_5S^-$	0.51	0.57	0.58	0.76	0.62	0.42	0.64	0.50
	$C_3H_5O_5S^-$	1.29	1.15	4.56	3.77	1.50	0.92	1.35	0.93
	$C_2H_3O_6S^-$	2.58	2.55	3.79	3.19	2.43	1.80	2.58	1.33
	$C_3H_7O_5S^-$	0.75	0.53	0.63	1.24	2.59	0.33	1.27	0.63
	$C_3H_5O_6S^-$	2.25	2.32	2.06	1.97	2.15	1.74	2.49	1.72
Anthropogenic OS	$C_4H_7O_4S^-$	2.29	2.18	2.31	1.30	1.75	1.23	1.77	2.33
	$C_5H_7O_6S^-$	0.85	0.63	0.79	1.14	0.69	0.48	0.95	0.69
	$C_6H_9O_6S^-$	0.86	0.46	1.43	1.47	1.24	0.34	2.20	0.25
	$C_8H_{17}O_4S^-$	0.94	0.54	0.37	0.34	0.99	0.54	1.86	1.95
	$C_6H_5O_4S^-$	0.35	0.08	0.37	--	0.41	0.13	0.34	0.12
	$C_7H_7O_4S^-$	0.33	0.11	0.32	0.28	0.43	0.11	0.33	0.16
Unknown source OS	$C_4H_5O_5S^-$	1.06	0.56	1.09	0.47	1.13	0.80	1.49	1.28
	$C_5H_8NO_8S^-$	1.40	1.06	3.34	2.98	0.90	1.04	0.70	0.68
Isoprene OS	$C_4H_7O_5S^-$	1.05	1.41	1.02	1.91	1.09	0.97	1.35	1.50
	$C_4H_7O_6S^-$	1.51	1.96	6.35	4.19	1.68	1.14	1.41	1.06
	$C_5H_9O_6S^-$	1.33	2.34	2.87	1.69	1.60	1.49	1.69	1.35
	$C_4H_7O_7S^-$	1.62	2.16	4.57	4.49	1.03	1.84	1.26	1.16
	$C_5H_{11}O_6S^-$	0.53	0.64	0.74	1.21	0.54	0.42	0.52	0.40
	$C_5H_7O_7S^-$	2.89	3.96	13.14	8.85	2.67	2.55	3.81	3.06
	$C_5H_9O_7S^-$	1.57	2.56	9.66	9.74	1.19	1.40	1.33	1.06
	$C_5H_{11}O_7S^-$	1.26	1.23	35.80	30.52	0.52	0.57	0.48	0.37
	$C_7H_9O_7S^-$	0.48	0.50	0.96	0.87	0.45	0.36	0.45	0.33
	$C_5H_{10}NO_9S^-$	0.82	0.60	2.64	6.82	--	0.40	0.20	0.24
	$C_5H_8NO_{10}S^-$	0.57	0.59	2.14	7.39	--	0.26	--	--
Monoterpene OS	$C_7H_{11}O_6S^-$	1.00	1.01	2.64	2.90	0.81	0.72	0.86	0.49
	$C_7H_{11}O_7S^-$	1.58	2.05	3.35	4.17	1.28	0.95	1.31	0.56
	$C_{10}H_{17}O_5S^-$	0.32	0.04	0.29	0.14	0.31	0.10	0.37	0.27
	$C_9H_{15}O_6S^-$	1.56	1.10	1.47	0.91	0.95	1.06	1.20	0.99
	$C_8H_{13}O_7S^-$	1.11	2.24	2.20	3.90	1.01	1.10	1.07	0.61
	$C_{10}H_{15}O_7S^-$	2.79	4.37	3.35	6.45	1.77	3.12	3.72	3.57
	$C_{10}H_{17}O_7S^-$	0.31	0.22	0.45	0.20	0.31	0.20	0.29	--
	$C_{10}H_{16}NO_7S^-$	11.20	6.57	6.50	4.92	3.00	5.52	3.04	5.39
	$C9H14NO_8S^-$	2.28	3.00	1.21	1.22	1.36	3.65	1.57	1.53
	$C10H16NO_{10}S^-$	1.70	2.06	1.24	1.60	1.13	1.59	1.11	1.11
	SUM	**51.04**	**51.53**	**114.13**	**102.09**	**38.15**	**37.98**	**44.48**	**35.99**

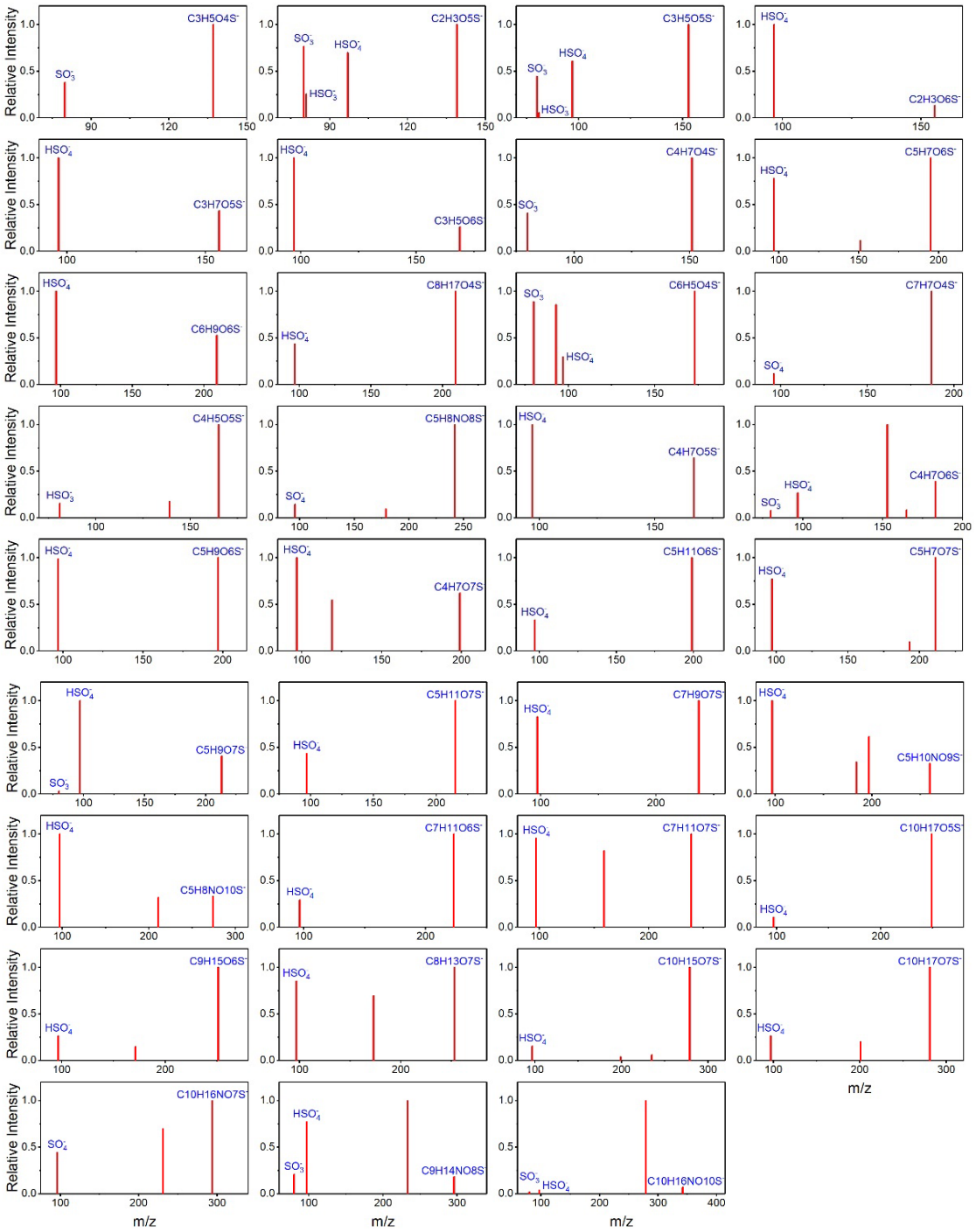

Figure S1. The MS^2 spectra of quantified OS species.