Supplement of

Spatial and Diurnal Variations of Aerosol Organosulfates in Summertime Shanghai, China: Potential Influence of Photochemical Process and Anthropogenic Sulfate Pollution

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S1. Classification of OSs

A total of 212 organosulfates (OSs) were detected in $PM_{2.5}$ samples using UPLC-MS. The quantified OSs were distributed over a wide mass range (m/z 167–489). The OSs and nitrooxy-organosulfates (NOSs) were initially identified from their molecular formulas based on the following equation (1) (Wang et al. 2019; Tao et al. 2014).

$$n_O / (4n_S + 3n_N) \geq 1 \qquad (1)$$

where the n_O, n_S, and n_N denote the number of oxygen, sulfur, and nitrogen atoms in a molecular formula, respectively.

The groups of isoprene-derived, monoterpene-derived, and C_2-C_3 OSs were further classified according to previous laboratory and filed studies (Hettiyadura et al. 2019; Wang et al. 2021a). Specifically, the isoprene-derived OSs (OS_i) were identified using the following method: (1) molecules with 4 and 5 carbon atoms were selected; (2) compounds with 4 carbon atoms have a molecular unsaturation degree (SD) of 1–2, $n_O \leq 6$, and $n_H \geq 6$; and (3) compounds with 5 carbon atoms have a SD of 0–2, $n_O \leq 7$, and $n_H \geq 8$. The detailed explanations about the workflow were provided by Xu et al., (2021) It should be noted that $C_7H_9O_7S^-$ was classified as OS_i based on Nozière et al., (2010).

For monoterpene-derived OSs (OS_m), they were grouped according to the following criteria. The compounds have the characteristics of 10 carbon atoms, effective oxygen atoms ($n_{Oeff} = n_O - 2n_N$) of more than 4, and $2 \leq DBE \leq 4$ (Guo et al. 2022; Ehn et al. 2012; Yan et al. 2016; Jokinen et al. 2014; Boyd et al. 2015; Berndt et al. 2016; Berndt et al. 2018). Moreover, $C_7H_{11}O_6S^-$, $C_7H_{11}O_7S^-$, $C_8H_{13}O_7S^-$, $C_9H_{15}O_6S$

$^{-}$, and $C_9H_{14}NO_8S^{-}$ were assigned to OS_m based on previous reports (Yassine et al. 2012; Nozière et al. 2010; Wang et al. 2017; Surratt et al. 2008). Moreover, correlation analysis between these target OSs and typical OS_m (e.g., $C_{10}H_{17}O_5S^{-}$) was conducted, showing the significant correlations between them ($r = 0.83$–0.99, $P < 0.01$). This result further confirms the reliability of the above classification.

Previous studies have suggested that OSs with double bond equivalent (DBE) ≥ 2 and aromaticity equivalent (Xc) ≥ 2.5 can be assigned to aromatic OSs (Jiang et al. 2022; Xie et al. 2021; Xie et al. 2020; Ma et al. 2022). The DBE and Xc value were calculated via the following equations, respectively (Yassine et al. 2014).

$$\mathrm{DBE} = 1 + n_C - n_H/2 + n_N/2 \qquad (2)$$

$$X_C = [3(\mathrm{DBE} - (f_m n_O - f_n \times n_s)) - 2]/[\mathrm{DBE} - (f_m n_O - f_n n_s)] \qquad (3)$$

where the n_C, n_H, n_N, n_O, and n_S denote the number of carbon, hydrogen, nitrogen, oxygen, sulfur atoms in a molecular formula, respectively. The f_m and f_n refer to the fractions of oxygen and sulfur atoms involved in the π-bond structure of the compound, respectively (Yassine et al. 2014). In this study, the negative mode was used in UPLC-ESI-QToF-MS analysis, which preferentially identified the compounds containing carboxylic acids and esters (Ye et al. 2021). Thus, the calculation for Xc of organosulfates can be simplified as the following equation (Ye et al. 2021).

$$X_C = [3(\mathrm{DBE} - 0.5(n_O - 4)) - 2]/[\mathrm{DBE} - 0.5(n_O - 4)]) \qquad (4)$$

However, the products of monoterpene oxidation by •OH or NO_3• also include the compounds with a DBE value of 2 (Yan et al. 2016; Ehn et al. 2014; Trostl et al. 2016). This means that OS_m and aromatic OSs overlapped partly. Thus, aromatic OSs with a

DBE value of 2 were further identified through correlation analysis between unidentified aromatic OSs and known OS_m and aromatic OSs (Bryant et al. 2021). The compounds were adopted with r value greater than 0.6.

The OSs with DBE less than 2, including alkanes and some unsaturated compounds, were assigned to aliphatic OSs (Xie et al. 2021; Xie et al. 2020; Tao et al. 2014). Recently, some aliphatic oxygenated organic molecules were found to have a DBE value of 2 (Wang et al. 2021b). Thus, OSs with a DBE value of 2 were further identified through correlation analysis between unidentified aliphatic OSs and known aliphatic subgroups. The compounds were adopted with r value greater than 0.6. It should be noted that $C_9H_{15}O_6S^-$ was divided into OS_m based on the abovementioned classification rules for OS_m, while $C_9H_{15}O_7S^-$ was not classified as OS_m. This is because $C_9H_{15}O_7S^-$ showed a stronger correlation with typical anthropogenic OS (e.g.,$C_8H_{17}O_4S^-$; $r = 0.98$) than typical OS_m (e.g., $C_{10}H_{17}O_5S^-$; $r = 0.96$). In this study, aliphatic and aromatic OSs were referred to anthropogenic OSs (OS_a). Several repotred OSs (non aromatic or aliphatic compounds) were classified as other anthropogenic OS (OS_a-other) (Wang et al. 2021a; Berndt et al. 2016).

Table S1. Organosulfate quantification using UPLC-ESI(-)-QToFMS.

Formula [M-H]-	MW (Da)	Standard for quantification	Reference	r^a
Isoprene-derived OSs				
$C_4H_7O_5S^-$	167.0014	Lactic acid sulfate (LAS)	(Schindelka et al. 2013)	
$C_4H_7O_6S^-$	182.9963	LAS	(Shalamzari 2013)	
$C_5H_9O_6S^-$	197.0120	LAS	(Riva et al. 2016a)	
$C_4H_7O_7S^-$	198.9912	LAS	(Hettiyadura et al. 2015)	
$C_5H_{11}O_6S^-$	199.0276	LAS	(Riva et al. 2016a)	
$C_5H_7O_7S^-$	210.9912	LAS	(Hettiyadura et al. 2015)	
$C_5H_9O_7S^-$	213.0069	LAS	(Riva et al. 2016a)	
$C_5H_{11}O_7S^-$	215.0225	LAS	(Surratt et al. 2010)	
$C_7H_9O_7S^-$	237.0069	LAS	(Nozière et al. 2010)	
$C_5H_{10}NO_9S^-$	260.0076	LAS	(Surratt et al. 2007)	
$C_5H_8NO_{10}S^-$	273.9869	LAS	(Nestorowicz et al. 2018)	
$C_5H_7O_8S^-$	226.9862	LAS		0.90
$C_5H_9O_8S^-$	229.0018	LAS		0.94
$C_4H_7O_8S^-$	214.9862	LAS		0.72
$C_4H_5O_7S^-$	196.9756	LAS		0.67
$C_4H_6NO_9S^-$	243.9763	LAS		0.87
$C_4H_8NO_7S^-$	214.0021	LAS		0.94
$C_5H_8NO_7S^-$	226.0021	LAS		0.90
$C_5H_9N_2O_{11}S^-$	260.0076	LAS		0.75
Monoterpene-derived OSs				
$C_7H_{11}O_6S^-$	223.0276	Glycolic acid sulfate (GAS)	(Yassine et al. 2012)	
$C_7H_{11}O_7S^-$	239.0225	GAS	(Nozière et al. 2010)	
$C_{10}H_{17}O_5S^-$	249.0797	α-Pinene sulfate	(Wang et al. 2017)	
$C_8H_{13}O_7S^-$	253.0382	GAS	(Schindelka et al. 2013)	
$C_{10}H_{15}O_7S^-$	279.0538	GAS	(Surratt et al. 2007)	
$C_{10}H_{17}O_7S^-$	281.0695	α-Pinene sulfate	(Nozière et al. 2010)	
$C_{10}H_{16}NO_7S^-$	294.0647	α-Pinene sulfate	(Surratt et al. 2008)	
$C_9H_{14}NO_8S^-$	296.0440	Limonaketone sulfate	(Surratt et al. 2008)	
$C_{10}H_{16}NO_{10}S^-$	342.0495	Limonaketone sulfate	(Yassine et al. 2012)	
$C_9H_{15}O_6S^-$	251.0589	Limonaketone sulfate	(Wang et al. 2017)	
$C_{10}H_{15}O_5S^-$	247.0640	α-Pinene sulfate		0.81
$C_{10}H_{15}O_6S^-$	263.0589	α-Pinene sulfate		0.87
$C_{10}H_{17}O_6S^-$	265.0746	α-Pinene sulfate		0.94

$C_{10}H_{17}O_8S^-$	297.0644	α-Pinene sulfate		0.96
$C_{10}H_{15}O_7S^-$	279.0538	α-Pinene sulfate		0.97
$C_{10}H_{15}O_8S^-$	295.0488	α-Pinene sulfate		0.98
$C_{10}H_{17}NO_9S^-$	326.0546	α-Pinene sulfate		0.85
C_2-C_3 OSs				
$C_3H_5O_4S^-$	136.9909	GAS	(Yassine et al. 2012)	
$C_2H_3O_5S^-$	138.9701	GAS	(Yassine et al. 2012)	
$C_3H_5O_5S^-$	152.9858	GAS	(Hettiyadura et al. 2015)	
$C_2H_3O_6S^-$	154.9650	GAS	(Olson et al. 2011)	
$C_3H_7O_5S^-$	155.0014	GAS	(Hettiyadura et al. 2019)	
$C_3H_5O_6S^-$	168.9807	LAS	(Olson et al. 2011)	
Aliphatic-OSs				
$C_8H_{17}O_4S^-$	210.0926	Sodium octyl Sulfate (SOS)	(Wang et al. 2021a)	
$C_{12}H_{21}O_7S^-$	309.1008	SOS		0.94
$C_{14}H_{29}O_5S^-$	309.1736	SOS		0.79
$C_{16}H_{33}O_5S^-$	337.2049	SOS		0.93
$C_6H_{13}O_4S^-$	181.0535	SOS		0.98
$C_7H_{15}O_4S^-$	195.0691	SOS		0.91
$C_7H_{15}O_5S^-$	211.064	SOS		0.89
$C_9H_{19}O_4S^-$	223.1004	SOS		0.70
$C_{10}H_{21}O_4S^-$	237.1161	SOS		0.7
$C_{24}H_{51}N_2O_{13}S^-$	607.3112	SOS		0.79
$C_7H_{13}O_5S^-$	209.0484	SOS		0.86
$C_9H_{17}O_5S^-$	237.0797	SOS		0.89
$C_{10}H_{19}O_5S^-$	251.0953	SOS		0.70
$C_9H_{17}O_7S^-$	269.0695	SOS		0.72
$C_{12}H_{23}O_5S^-$	279.1266	SOS		0.90
$C_9H_{17}O_4S^-$	221.0848	SOS		0.68
$C_{10}H_{19}O_5S^-$	251.0953	SOS		0.72
$C_9H_{17}O_6S^-$	253.0746	SOS		0.90
$C_{11}H_{21}O_5S^-$	265.1110	SOS		0.68
$C_{10}H_{19}O_6S^-$	267.0902	SOS		0.73
$C_{13}H_{25}O_5S^-$	293.1423	SOS		0.84
$C_{14}H_{27}O_5S^-$	307.1579	SOS		0.94
$C_{13}H_{25}O_6S^-$	309.1372	SOS		0.72
$C_{14}H_{27}O_6S^-$	323.1528	SOS		0.75
$C_{16}H_{31}O_5S^-$	335.1892	SOS		0.96
$C_{17}H_{33}O_5S^-$	349.2049	SOS		0.94
$C_{16}H_{31}O_6S^-$	351.1841	SOS		0.7
$C_{18}H_{35}O_5S^-$	363.2205	SOS		0.76

$C_{21}H_{41}O_5S^-$	405.2675	SOS		0.94
$C_8H_{15}O_5S^-$	223.0640	SOS		0.69
$C_7H_{13}O_6S^-$	225.0433	SOS		0.76
$C_6H_{11}O_7S^-$	227.0225	SOS		0.94
$C_8H_{15}O_6S^-$	239.0589	SOS		0.89
$C_6H_{11}O_8S^-$	243.0175	SOS		0.87
$C_{11}H_{21}O_5S^-$	265.1110	SOS		0.69
$C_{10}H_{19}O_6S^-$	267.0902	SOS		0.88
$C_7H_{13}O_9S^-$	273.0280	SOS		0.70
$C_{15}H_{29}O_5S^-$	321.1736	SOS		0.88
$C_{26}H_{51}O_{12}S^-$	587.3101	SOS		0.95
$C_{10}H_{17}O_6S^-$	265.0746	SOS		0.85
$C_9H_{15}O_5S^-$	235.0640	SOS		0.95
$C_{10}H_{17}O_5S^-$	249.0797	SOS		0.93
$C_9H_{15}O_6S^-$	251.0589	SOS		0.91
$C_{10}H_{17}O_6S^-$	265.0746	SOS		0.72
$C_9H_{15}O_5S^-$	235.0640	SOS		0.98
$C_{10}H_{17}O_5S^-$	249.0797	SOS		0.94
$C_9H_{15}O_6S^-$	251.0589	SOS		0.92
$C_{10}H_{17}O_6S^-$	265.0746	SOS		0.69
$C_{11}H_{19}O_6S^-$	279.0902	SOS		0.78
$C_{12}H_{21}O_6S^-$	293.1059	SOS		0.72
$C_{14}H_{25}O_6S^-$	321.1372	SOS		0.63
$C_8H_{13}O_6S^-$	237.0433	SOS		0.75
$C_9H_{15}O_7S^-$	267.0538	SOS		0.73
Aromatic-OSs				
$C_6H_5O_4S^-$	172.9909	Methyl sulfate	(Wang et al. 2021a)	
$C_7H_7SO_4S^-$	218.9786	Methyl sulfate	(Wang et al. 2021a)	
$C_{11}H_{19}O_{11}S^-$	359.0648	Methyl sulfate		0.77
$C_{10}H_{17}O_{12}S^-$	361.0441	Methyl sulfate		0.64
$C_7H_{11}O_9S^-$	271.0124	Methyl sulfate		0.65
$C_7H_{11}O_{10}S^-$	287.0073	Methyl sulfate		0.69
$C_8H_{13}O_9S^-$	285.0280	Methyl sulfate		0.70
$C_8H_{13}O_{10}S^-$	301.0229	Methyl sulfate		0.81
$C_{11}H_{17}O_{11}S^-$	357.0492	Methyl sulfate		0.86
$C_8H_{12}NO_{11}S^-$	330.0131	Methyl sulfate		0.90
$C_8H_7O_5S^-$	215.0014	Methyl sulfate		0.91
$C_8H_7NO_5S^-$	229.0045	Methyl sulfate		0.89
$C_8H_7O_4S^-$	199.0065	Methyl sulfate		0.98
$C_9H_7O_7S^-$	258.9912	Methyl sulfate		0.89
$C_9H_7O_6S^-$	242.9963	Methyl sulfate		0.74

$C_{34}H_{49}O_5S^-$	569.3301	Methyl sulfate		0.64
$C_{43}H_{63}O_5S^-$	691.4396	Methyl sulfate		0.72
$C_7H_{11}O_9S^-$	271.0124	Methyl sulfate		0.74
$C_{10}H_7O_{11}S^-$	334.9709	Methyl sulfate		0.86
$C_{10}H_5O_{11}S^-$	332.9553	Methyl sulfate		0.77
$C_{10}H_5O_{10}S^-$	316.9603	Methyl sulfate		0.72
$C_{12}H_7O_{13}S^-$	390.9607	Methyl sulfate		0.65
$C_8H_7O_5S^-$	215.0014	Methyl sulfate		0.77
$C_7H_5SO_5S^-$	200.9858	Methyl sulfate		0.65
$C_9H_9O_4S^-$	213.0222	Methyl sulfate		0.72
$C_7H_6SO_4S^-$	185.9987	Methyl sulfate		0.73
$C_{18}H_{13}O_6S^-$	357.0433	Methyl sulfate		0.69
$C_{23}H_{19}O_7S^-$	439.0851	Methyl sulfate		0.65
$C_{25}H_{21}O_7S^-$	465.1008	Methyl sulfate		0.77
$C_{24}H_{17}O_4S^-$	401.0848	Methyl sulfate		0.73
$C_{27}H_{21}O_7S^-$	489.1008	Methyl sulfate		0.90
$C_{12}H_{21}N_2O_{11}S$	401.0866	Methyl sulfate		0.77
OS$_a$-other				
$C_4H_7O_4S^-$	151.0065	Methyl sulfate	(Wang et al. 2021a)	
$C_5H_7O_6S^-$	194.9963	GAS	(Wang et al. 2021a)	
$C_6H_9O_6S^-$	209.0120	GAS	(Berndt et al. 2016)	

[a] Unreported OSs were further identified through correlation analysis between unreported OSs and known OSs. Aliphatic and aromatic OSs were referred to anthropogenic OSs (OSa). The compounds were adopted with R value greater than 0.6. Details were shown in **Sect. S1** (Classification of OSs).

Table S2. The mean mass concentrations (± SD) of identified OS_i, OS_m, and C_2-C_3 OSs in $PM_{2.5}$ collected in urban and suburban Shanghai in daytime and nighttime.

Formula $[M^{-}H]^{-}$	MW (Da)	Urban (ng m^{-3})		Suburban (ng m^{-3})	
		Daytime	Nighttime	Daytime	Nighttime
Isoprene-derived OSs					
$C_4H_7O_5S^-$	167.0014	0.70 ± 0.75	0.49 ± 0.55	0.53 ± 0.44	0.34 ± 0.17
$C_4H_7O_6S^-$	182.9963	5.93 ± 6.30	1.94 ± 2.83	3.80 ± 4.18	0.96 ± 1.13
$C_5H_9O_6S^-$	197.0120	4.36 ± 5.49	1.93 ± 2.95	2.20 ± 2.51	1.26 ± 1.52
$C_4H_7O_7S^-$	198.9912	6.09 ± 7.93	2.20 ± 3.56	3.20 ± 4.19	1.30 ± 1.76
$C_5H_{11}O_6S^-$	199.0276	1.60 ± 2.46	0.66 ± 0.96	0.77 ± 0.94	0.39 ± 0.41
$C_5H_7O_7S^-$	210.9912	11.99 ± 16.15	4.88 ± 6.49	6.38 ± 7.97	3.67 ± 4.46
$C_5H_9O_7S^-$	213.0069	12.32 ± 12.2	6.91 ± 4.83	9.47 ± 4.36	6.61 ± 2.05
$C_5H_{11}O_7S^-$	215.0225	53.44 ± 72.16	22.74 ± 36.10	25.64 ± 28.99	13.91 ± 14.14
$C_7H_9O_7S^-$	237.0069	0.81 ± 1.01	0.44 ± 0.85	0.46 ± 0.51	0.29 ± 0.44
$C_5H_7O_8S^-$	226.9862	9.89 ± 10.69	4.58 ± 5.60	6.22 ± 6.87	2.13 ± 3.01
$C_5H_9O_8S^-$	229.0018	4.47 ± 5.96	2.29 ± 3.67	2.09 ± 2.64	0.98 ± 1.37
$C_4H_7O_8S^-$	214.9862	0.52 ± 0.46	0.40 ± 0.44	0.53 ± 0.54	0.26 ± 0.29
$C_4H_5O_7S^-$	196.9756	0.48 ± 0.31	0.34 ± 0.27	0.44 ± 0.31	0.33 ± 0.19
$C_4H_6NO_9S^-$	243.9763	0.18 ± 0.19	0.13 ± 0.07	0.13 ± 0.01	0.11 ± 0.01
$C_4H_8NO_7S^-$	214.0021	0.38 ± 0.47	0.23 ± 0.24	0.25 ± 0.17	0.18 ± 0.11
$C_5H_8NO_7S^-$	226.0021	0.49 ± 0.83	0.17 ± 0.20	0.28 ± 0.28	0.15 ± 0.09
$C_5H_{10}NO_9S^-$	260.0076	1.79 ± 3.03	0.95 ± 2.06	0.49 ± 0.86	0.37 ± 0.71
$C_5H_8NO_{10}S^-$	273.9869	1.14 ± 2.09	0.73 ± 1.47	0.41 ± 0.44	0.23 ± 0.35
$C_5H_9N_2O_{11}S^-$	260.0076	1.02 ± 1.96	3.06 ± 6.16	0.37 ± 0.42	0.82 ± 1.60
Monoterpene-derived OSs					
$C_7H_{11}O_6S^-$	223.0276	8.24 ± 12.05	3.07 ± 4.24	4.13 ± 5.16	1.82 ± 1.72
$C_7H_{11}O_7S^-$	239.0225	11.04 ± 17.59	3.29 ± 5.02	4.40 ± 5.84	1.69 ± 1.61
$C_{10}H_{17}O_5S^-$	249.0797	0.20 ± 0.20	0.11 ± 0.05	0.16 ± 0.05	0.10 ± 0.05
$C_9H_{15}O_6S^-$	251.0589	0.47 ± 0.47	0.31 ± 0.21	0.30 ± 0.16	0.22 ± 0.09
$C_8H_{13}O_7S^-$	253.0382	10.33 ± 16.91	3.18 ± 4.92	4.35 ± 5.81	8.71 ± 25.31
$C_{10}H_{15}O_7S^-$	279.0538	8.24 ± 12.34	4.94 ± 6.61	4.68 ± 4.50	2.88 ± 2.64
$C_{10}H_{17}O_7S^-$	281.0695	0.14 ± 0.09	0.10 ± 0.03	0.14 ± 0.03	0.10 ± 0.01
$C_{10}H_{15}O_5S^-$	247.0640	0.07 ± 0.02	0.08 ± 0.03	0.08 ± 0.01	0.07 ± 0.01
$C_{10}H_{15}O_6S^-$	263.0589	0.04 ± 0.05	0.04 ± 0.04	0.03 ± 0.04	0.05 ± 0.03
$C_{10}H_{17}O_6S^-$	265.0746	0.08 ± 0.03	0.06 ± 0.01	0.07 ± 0.02	0.06 ± 0.01
$C_{10}H_{17}O_8S^-$	297.0644	0.13 ± 0.05	0.10 ± 0.02	0.13 ± 0.02	0.11 ± 0.01
$C_{10}H_{15}O_7S^-$	279.0538	0.19 ± 0.14	0.13 ± 0.06	0.16 ± 0.04	0.12 ± 0.02
$C_{10}H_{15}O_8S^-$	295.0488	0.13 ± 0.06	0.10 ± 0.02	0.14 ± 0.01	0.11 ± 0.01
$C_{10}H_{17}NO_9S^-$	326.0546	0.16 ± 0.07	0.13 ± 0.07	0.16 ± 0.03	0.12 ± 0.03
$C_{10}H_{16}NO_7S^-$	294.0647	0.34 ± 0.3	1.98 ± 1.89	0.24 ± 0.14	0.87 ± 0.58
$C_9H_{14}NO_8S^-$	296.0440	0.12 ± 0.11	0.39 ± 0.23	0.12 ± 0.11	0.26 ± 0.07
$C_{10}H_{16}NO_{10}S^-$	342.0495	0.22 ± 0.20	0.41 ± 0.41	0.17 ± 0.14	0.26 ± 0.13
C_2-C_3 OSs					
$C_3H_5O_4S^-$	136.9909	1.36 ± 0.94	0.98 ± 0.59	1.17 ± 0.48	0.95 ± 0.37
$C_2H_3O_5S^-$	138.9701	1.73 ± 1.13	1.69 ± 1.25	1.70 ± 0.64	1.60 ± 1.17
$C_3H_5O_5S^-$	152.9858	9.33 ± 10.64	4.51 ± 4.51	5.91 ± 5.50	3.18 ± 2.54
$C_2H_3O_6S^-$	154.9650	9.69 ± 8.65	4.55 ± 4.55	6.89 ± 6.07	4.07 ± 3.58
$C_3H_7O_5S^-$	155.0014	1.44 ± 1.07	1.69 ± 1.42	1.76 ± 1.28	1.37 ± 0.93
$C_3H_5O_6S^-$	168.9807	3.19 ± 2.61	2.18 ± 3.08	2.64 ± 1.94	1.60 ± 0.88

Table S3. The mean mass concentrations (± SD) of identified anthropogenic OSs in $PM_{2.5}$ collected in urban and suburban Shanghai in daytime and nighttime.

Formula $[M\text{-}H]^-$	MW (Da)	Urban (ng m^{-3})		Suburban (ng m^{-3})	
		Daytime	Nighttime	Daytime	Nighttime
Aliphatic-OSs[a]					
$C_{12}H_{21}O_7S^-$	309.1008	0.13 ± 0.09	0.10 ± 0.05	0.12 ± 0.03	0.10 ± 0.02
$C_8H_{17}O_4S^-$	210.0926	0.12 ± 0.03	0.13 ± 0.05	0.10 ± 0.02	0.08 ± 0.01
$C_{14}H_{29}O_5S^-$	309.1736	0.07 ± 0.01	0.06 ± 0.01	0.08 ± 0.01	0.07 ± 0.01
$C_{16}H_{33}O_5S^-$	337.2049	0.06 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_6H_{13}O_4S^-$	181.0535	0.05 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.05 ± 0.01
$C_7H_{15}O_4S^-$	195.0691	0.06 ± 0.01	0.06 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_7H_{15}O_5S^-$	211.064	0.06 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_9H_{19}O_4S^-$	223.1004	0.07 ± 0.01	0.06 ± 0.01	0.08 ± 0.01	0.07 ± 0.01
$C_{10}H_{21}O_4S^-$	237.1161	0.08 ± 0.01	0.08 ± 0.02	0.10 ± 0.01	0.09 ± 0.01
$C_{24}H_{51}N_2O_{13}S^-$	607.3112	0.06 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.05 ± 0.01
$C_7H_{13}O_5S^-$	209.0484	0.08 ± 0.03	0.06 ± 0.02	0.08 ± 0.02	0.06 ± 0.01
$C_9H_{17}O_5S^-$	237.0797	0.14 ± 0.08	0.08 ± 0.03	0.11 ± 0.05	0.07 ± 0.02
$C_{10}H_{19}O_5S^-$	251.0953	0.15 ± 0.25	0.06 ± 0.02	0.08 ± 0.02	0.06 ± 0.01
$C_9H_{17}O_7S^-$	269.0695	0.07 ± 0.03	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_{12}H_{23}O_5S^-$	279.1266	0.28 ± 0.24	0.18 ± 0.14	0.16 ± 0.10	0.09 ± 0.04
$C_9H_{17}O_4S^-$	221.0848	0.09 ± 0.02	0.07 ± 0.01	0.10 ± 0.02	0.07 ± 0.01
$C_{10}H_{19}O_5S^-$	251.0953	0.15 ± 0.25	0.06 ± 0.02	0.08 ± 0.02	0.06 ± 0.01
$C_9H_{17}O_6S^-$	253.0746	0.08 ± 0.04	0.06 ± 0.02	0.08 ± 0.01	0.06 ± 0.01
$C_{11}H_{21}O_5S^-$	265.1110	0.17 ± 0.13	0.13 ± 0.08	0.14 ± 0.08	0.09 ± 0.03
$C_{10}H_{19}O_6S^-$	267.0902	0.07 ± 0.02	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_{13}H_{25}O_5S^-$	293.1423	0.31 ± 0.27	0.20 ± 0.16	0.16 ± 0.08	0.11 ± 0.04
$C_{14}H_{27}O_5S^-$	307.1579	0.25 ± 0.25	0.13 ± 0.09	0.11 ± 0.05	0.08 ± 0.02
$C_{13}H_{25}O_6S^-$	309.1372	0.06 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_{14}H_{27}O_6S^-$	323.1528	0.06 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_{16}H_{31}O_5S^-$	335.1892	0.05 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_{17}H_{33}O_5S^-$	349.2049	0.07 ± 0.01	0.08 ± 0.02	0.11 ± 0.03	0.13 ± 0.05
$C_{16}H_{31}O_6S^-$	351.1841	0.08 ± 0.03	0.06 ± 0.01	0.08 ± 0.01	0.06 ± 0.01
$C_{18}H_{35}O_5S^-$	363.2205	0.05 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.05 ± 0.01
$C_{21}H_{41}O_5S^-$	405.2675	0.06 ± 0.01	0.05 ± 0.01	0.08 ± 0.01	0.06 ± 0.01
$C_8H_{15}O_5S^-$	223.0640	0.08 ± 0.02	0.06 ± 0.02	0.08 ± 0.01	0.06 ± 0.01
$C_7H_{13}O_6S^-$	225.0433	0.06 ± 0.02	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_6H_{11}O_7S^-$	227.0225	0.07 ± 0.04	0.05 ± 0.01	0.07 ± 0.01	0.05 ± 0.01
$C_8H_{15}O_6S^-$	239.0589	0.08 ± 0.03	0.06 ± 0.01	0.08 ± 0.01	0.06 ± 0.01
$C_6H_{11}O_8S^-$	243.0175	0.05 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.05 ± 0.01
$C_{11}H_{21}O_5S^-$	265.1110	0.11 ± 0.05	0.09 ± 0.03	0.11 ± 0.02	0.09 ± 0.02
$C_{10}H_{19}O_6S^-$	267.0902	0.06 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.05 ± 0.01
$C_7H_{13}O_9S^-$	273.0280	0.05 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.05 ± 0.01
$C_{15}H_{29}O_5S^-$	321.1736	0.15 ± 0.16	0.07 ± 0.04	0.08 ± 0.02	0.06 ± 0.01
$C_{26}H_{51}O_{12}S^-$	587.3101	0.08 ± 0.02	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_{10}H_{17}O_6S^-$	265.0746	0.07 ± 0.02	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_9H_{15}O_5S^-$	235.0640	0.10 ± 0.04	0.06 ± 0.02	0.09 ± 0.02	0.06 ± 0.01
$C_{10}H_{17}O_5S^-$	249.0797	0.07 ± 0.02	0.06 ± 0.01	0.08 ± 0.01	0.06 ± 0.01
$C_9H_{15}O_6S^-$	251.0589	0.08 ± 0.05	0.06 ± 0.02	0.08 ± 0.02	0.06 ± 0.01
$C_{10}H_{17}O_6S^-$	265.0746	0.07 ± 0.02	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_{11}H_{19}O_6S^-$	279.0902	0.07 ± 0.02	0.05 ± 0.01	0.07 ± 0.01	0.06 ± 0.01
$C_{12}H_{21}O_6S^-$	293.1059	0.06 ± 0.01	0.05 ± 0.01	0.07 ± 0.01	0.05 ± 00.01
$C_{14}H_{25}O_6S^-$	321.1372	0.18 ± 0.19	0.08 ± 0.04	0.09 ± 0.02	0.07 ± 0.01
$C_8H_{13}O_6S^-$	237.0433	0.08 ± 0.03	0.06 ± 0.02	0.09 ± 0.02	0.07 ± 0.02
$C_9H_{15}O_7S^-$	267.0538	0.13 ± 0.14	0.06 ± 0.04	0.08 ± 0.02	0.06 ± 0.01

Aromatic-OSs					
$C_{11}H_{19}O_{11}S^-$	359.0648	0.14 ± 0.01	0.12 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_{10}H_{17}O_{12}S^-$	361.0441	0.15 ± 0.02	0.12 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_7H_{11}O_9S^-$	271.0124	0.16 ± 0.04	0.13 ± 0.02	0.19 ± 0.02	0.15 ± 0.01
$C_7H_{11}O_{10}S^-$	287.0073	0.22 ± 0.10	0.15 ± 0.04	0.23 ± 0.06	0.17 ± 0.03
$C_8H_{13}O_9S^-$	285.0280	0.15 ± 0.01	0.12 ± 0.01	0.18 ± 0.02	0.15 ± 0.01
$C_8H_{13}O_{10}S^-$	301.0229	0.15 ± 0.02	0.12 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_{11}H_{17}O_{11}S^-$	357.0492	0.14 ± 0.01	0.12 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_8H_{12}NO_{11}S^-$	330.0131	0.15 ± 0.02	0.13 ± 0.02	0.19 ± 0.02	0.15 ± 0.01
$C_7H_7SO_4S^-$	218.9786	0.15 ± 0.01	0.13 ± 0.02	0.18 ± 0.01	0.15 ± 0.01
$C_8H_7O_5S^-$	215.0014	0.18 ± 0.05	0.15 ± 0.05	0.20 ± 0.03	0.16 ± 0.02
$C_8H_7NO_5S^-$	229.0045	0.14 ± 0.01	0.12 ± 0.01	0.17 ± 0.01	0.14 ± 0.01
$C_8H_7O_4S^-$	199.0065	0.15 ± 0.02	0.12 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_9H_7O_7S^-$	258.9912	0.14 ± 0.01	0.12 ± 0.01	0.18 ± 0.01	0.14 ± 0.01
$C_9H_7O_6S^-$	242.9963	0.14 ± 0.01	0.12 ± 0.01	0.18 ± 0.01	0.14 ± 0.01
$C_{34}H_{49}O_5S^-$	569.3301	1.34 ± 0.51	0.59 ± 0.35	0.26 ± 0.11	0.18 ± 0.02
$C_{43}H_{63}O_5S^-$	691.4396	0.27 ± 0.07	0.18 ± 0.06	0.19 ± 0.01	0.16 ± 0.01
$C_7H_{11}O_9S^-$	271.0124	0.24 ± 0.14	0.16 ± 0.08	0.22 ± 0.06	0.17 ± 0.04
$C_{10}H_7O_{11}S^-$	334.9709	0.20 ± 0.09	0.15 ± 0.07	0.20 ± 0.05	0.16 ± 0.03
$C_{10}H_5O_{11}S^-$	332.9553	0.15 ± 0.01	0.12 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_{10}H_5O_{10}S^-$	316.9603	0.15 ± 0.02	0.12 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_{12}H_7O_{13}S^-$	390.9607	0.14 ± 0.01	0.12 ± 0.01	0.18 ± 0.01	0.14 ± 0.01
$C_8H_7O_5S^-$	215.0014	0.20 ± 0.07	0.17 ± 0.06	0.22 ± 0.04	0.18 ± 0.03
$C_7H_5SO_5S^-$	200.9858	0.20 ± 0.07	0.15 ± 0.05	0.21 ± 0.04	0.17 ± 0.03
$C_9H_9O_4S^-$	213.0222	0.21 ± 0.08	0.16 ± 0.05	0.25 ± 0.06	0.17 ± 0.03
$C_7H_6SO_4S^-$	185.9987	0.16 ± 0.02	0.13 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_{18}H_{13}O_6S^-$	357.0433	0.14 ± 0.01	0.12 ± 0.01	0.18 ± 0.01	0.15 ± 0.01
$C_{23}H_{19}O_7S^-$	439.0851	0.19 ± 0.04	0.13 ± 0.01	0.21 ± 0.02	0.17 ± 0.01
$C_{25}H_{21}O_7S^-$	465.1008	0.14 ± 0.01	0.13 ± 0.03	0.17 ± 0.01	0.15 ± 0.01
$C_{24}H_{17}O_4S^-$	401.0848	0.19 ± 0.03	0.15 ± 0.02	0.23 ± 0.03	0.18 ± 0.01
$C_{27}H_{21}O_7S^-$	489.1008	0.14 ± 0.01	0.12 ± 0.01	0.17 ± 0.01	0.14 ± 0.01
$C_6H_5O_4S^-$	172.9909	0.15 ± 0.01	0.12 ± 0.01	0.18 ± 0.01	0.14 ± 0.01
$C_7H_7O_4S^-$	187.0065	0.15 ± 0.01	0.13 ± 0.02	0.18 ± 0.01	0.15 ± 0.01
$C_{12}H_{21}N_2O_{11}S^-$	401.0866	0.19 ± 0.03	0.15 ± 0.02	0.23 ± 0.03	0.18 ± 0.01
OS$_a$-other					
$C_4H_7O_4S^-$	151.0065	0.93 ± 0.77	0.83 ± 1.17	0.55 ± 0.43	0.44 ± 0.50
$C_5H_7O_6S^-$	194.9963	3.68 ± 3.75	1.80 ± 1.66	2.29 ± 1.80	1.35 ± 0.72
$C_6H_9O_6S^-$	209.0120	4.44 ± 6.52	3.00 ± 3.69	2.52 ± 3.24	1.90 ± 1.80

[a] Aliphatic and aromatic OSs were grouped into anthropogenic OSs (OS$_a$) (Riva et al. 2016b; Riva et al. 2015). Several unidentified aromatic and aliphatic OSs were classified as other anthropogenic OSs (OS$_a$-other).

Table S4. The mean mass concentrations of various OSs in $PM_{2.5}$ at different locations.

Typical Environment	Sampling site	Period	Season	OS_i (ng m^{-3})	OS_m (ng m^{-3})	OS_a (ng m^{-3})	C_2-C_3 (ng m^{-3})	Total (ng m^{-3})	Ref.
Urban	Pearl River Delta, China	2012	Summer	0.68	-	-	-	0.68	(He et al. 2014)
	Shanghai, China	2012	Spring	0.017	0.20	0.219	-	0.44	(Ma et al. 2014)
			Summer	23.1	26.7	0.77	-	50.57	
			Autumn	0.063	0.77	0.79	-	1.62	
			Winter	0.82	0.90	0.72	-	2.44	
	Beijing, China	2016	Spring	16.2	13.0	-	19.5	48.7	(Wang et al. 2018)
	Beijing, China	2016	Summer	12.3	15.1	-	-	27.4	(Wang et al. 2020)
			Winter	2.0	6.0	-	-	8.0	
	Atlanta, GA, USA	2014	Summer	1122.98	67.9	-	58.5	1249.38	(Hettiyadura et al. 2019)
	Guangzhou, China	2019	Summer	181.8	10.8	-	-	192.6	(Bryant et al. 2021)
	Tianjing, China	2019	Winter	600	-	-	-	600	(Ding et al. 2022)
			Summer	400	-	-	-	400	
	Patra, Greece	2018	Winter	31.5	20.7	3.1	-	55.3	(Kanellopoulos et al. 2022)
			Spring	75.7	32.4	1.3	-	109.4	
			Summer	658	37.7	1.6	-	697.3	
			Autumn	53.2	25.4	1.1	-	79.7	
	Hong Kong, China	2016	Summer	163.19	2.95	-	-	166.14	(Wang et al. 2022)
		2017	Winter	97.96	17.26	-	-	115.22	
	Guangzhou, China	2016	Summer	460.2	26.22	-	-	486.42	
		2017	Winter	88.03	20.96	-	-	108.99	
	Beijing, China	2016	Summer	236.64	21.7	-	-	258.34	
		2017	Winter	176.32	36.01	-	-	212.33	
	Shanghai, China	2016	Summer	326.4	34.9	-	-	361.3	
		2017	Winter	70.31	32.32	-	-	102.63	

	Copenhagen, Denmark	2011	Summer	11.31	0.87	-	-	18.18	(Nguyen et al. 2014)
	Xi’an, China	2014	Winter	-	-	0.14	77.3	77.44	(Huang et al. 2018)
Suburban	Centreville, AL, USA	2013	Summer	15.40	-	1.16	20.6	37.16	(Hettiyadura et al. 2017)
	Zion, Illinois, USA		Spring	121.1	8.7	-	-	129.8	(Hughes et al. 2021)
Regional	Beijing, China	2016	Summer	5.9	16.1	-	-	22.0	(Wang et al. 2020)
			Winter	69.5	18.7	-	-	88.2	
Forest	Centreville, Alabama	2013	Summer	16.5	20.6	-	-	37.1	(Hettiyadura et al. 2017)
Polar sites	Arctic sites		Spring	47	-	-	-	47	(Hansen et al. 2014)
Urban	Shanghai, China	2021	Summer	85.38	30.61	19.31	23.38	158.68	In this study
Suburban				48.98	19.30	15.73	18.25	102.26	

[a] The symbol of “-” denotes no data.

Table S5. The mean values (± SD) of the major parameters observed in different periods in daytime and nighttime.

	Urban- period A		Urban- period B		Suburban-period A		Suburban-period B	
	Daytime	Nighttime	Daytime	Nighttime	Daytime	Nighttime	Daytime	Nighttime
T (°C)	33.64 ± 0.98	29.16 ± 0.25	30.93 ± 0.78	27.95 ± 0.62	33.64 ± 0.98	29.16 ± 0.25	30.93 ± 0.78	27.95±0.62
RH (%)	73.45 ± 3.57	89.84 ± 1.83	75.88 ± 3.84	90.59 ± 1.85	73.45 ± 3.57	89.84 ± 1.83	75.88 ± 3.84	90.59±1.85
Wind speed	2.08 ± 0.66	1.97 ± 0.46	2.61 ± 0.19	2.17 ± 0.42	2.08 ± 0.66	1.97 ± 0.46	2.61 ± 0.19	2.17±0.42
PBLH (m)	459.17 ± 59.02	333.61 ± 36.65	919.17±189.63	574.63 ± 83.41	459.17 ± 59.02	333.61 ± 36.65	919.17 ± 189.63	574.63±83.41
UV (w/m^2)	21.87 ± 3.97	0.38 ± 0.01	21.17 ± 3.13	0.37 ± 0.01	21.87 ± 3.97	0.38 ± 0.01	21.17 ± 3.13	0.37±0.01
VC	951.89 ± 294.19	668.68 ± 220.05	2377.4 ± 378.6	1258.67 ± 334.63	951.89 ± 294.19	668.68 ± 220.05	2377.4 ± 378.6	1258.67 ± 334.63
NO_2 (ppb)	8.13 ± 1.80	9.25 ± 1.65	3.76 ± 1.31	5.01 ± 1.31	9.14 ± 1.26	9.76 ± 2.99	3.82 ± 1.75	5.80 ± 2.28
NO (ppb)	0.47 ± 0.56	1.91 ± 0.38	0.70 ± 0.51	1.25 ± 1.02	2.13 ± 0.55	1.93 ± 0.29	1.98 ± 0.31	2.37 ± 0.53
O_3 (ppb)	70.28 ± 8.44	28.98 ± 8.70	31.79 ± 9.08	15.55 ± 7.48	59.73 ± 11.47	24.69 ± 7.79	29.43 ± 8.07	16.21 ± 8.68
SO_2 (ppb)	3.08 ± 0.71	2.20 ± 0.36	1.65 ± 0.50	1.68 ± 0.72	2.37 ± 0.09	2.21 ± 0.08	2.21 ± 0.11	2.19 ± 0.13
ALW (ug m^{-3})	3.26 ± 1.64	5.14 ± 3.53	1.40 ± 0.80	2.57 ± 2.20	1.29 ± 1.15	2.15 ± 1.24	1.70 ± 1.26	2.55 ± 2.86
pH	2.08 ± 0.48	2.14 ± 0.31	3.10 ± 1.06	2.68 ± 0.98	2.21 ± 0.62	2.13 ± 0.35	2.56 ± 1.09	1.99 ± 0.43
OM (ug m^{-3})	23.39 ± 4.64	15.38 ± 4.26	7.02 ± 2.78	3.91 ± 1.25	12.23 ± 3.51	8.72 ± 4.24	4.18 ± 2.59	2.55 ± 1.11
EC (ug m^{-3})	4.34 ± 1.24	3.71 ± 1.05	1.05 ± 0.94	0.38 ± 0.29	2.16 ± 0.92	1.51 ± 0.92	0.49 ± 0.86	0.21 ± 0.22
NO_3^- (ug m^{-3})	2.60 ± 0.71	3.09 ± 2.18	1.44 ± 0.25	0.92 ± 0.14	2.28 ± 0.59	2.29 ± 2.12	1.11 ± 0.20	1.09 ± 1.02
SO_4^{2-} (ug m^{-3})	5.29 ± 1.65	4.10 ± 1.22	2.41 ± 1.18	2.00 ± 1.28	3.08 ± 1.02	2.23 ± 0.52	2.29 ± 1.21	2.13 ± 1.82
$PM_{2.5}$ (ug m^{-3})	31.75 ± 8.68	25.67 ± 9.12	12.54 ± 2.22	10.80 ± 3.10	27.91 ± 10.49	20.82 ± 8.25	10.75 ± 2.08	10.46 ± 3.95
Total OSs (ng m^{-3})	498.72 ± 249.22	272.38 ± 147.84	67.13 ± 24.53	48.55 ± 17.15	241.83 ± 123.49	175.04 ± 96.13	64.65 ± 18.61	49.4 ± 18.08
OS_i (ng m^{-3})	289.18 ± 145.14	146.30 ± 103.80	32.04 ± 16.37	19.43 ± 11.77	132.56 ± 75.86	75.98 ± 42.35	29.21 ± 13.08	20.41 ± 12.23
$C_5H_{11}O_7S^-$ (ng m^{-3})	136.47 ± 71.59	66.77 ± 55.60	11.93 ± 8.75	8.07 ± 7.06	54.26 ± 36.67	30.08 ± 19.65	11.33 ± 6.48	8.52 ± 6.89
NOS_i (ng m^{-3})	13.33 ± 11.18	19.24 ± 13.08	0.81 ± 0.25	0.62 ± 0.30	4.19 ± 2.53	5.58 ± 3.81	0.8 ± 0.28	0.62 ± 0.35
OS_m (ng m^{-3})	105.86 ± 70.67	55.97 ± 26.08	8.09 ± 3.16	8.73 ± 3.96	42.14 ± 27.33	51.62 ± 52.67	8.88 ± 1.88	7.64 ± 2.34
$C_8H_{13}O_7S^-$ (ng m^{-3})	27.70 ± 21.05	9.95 ± 6.43	1.64 ± 0.75	0.93 ± 0.27	10.29 ± 7.28	32.00 ± 49.38	1.39 ± 0.40	0.95 ± 0.34
NOS_m (ng m^{-3})	1.57 ± 0.58	6.63 ± 0.48	0.46 ± 0.26	1.66 ± 1.18	1.12 ± 0.20	2.44 ± 0.29	0.49 ± 0.24	1.21 ± 0.55
C_2-C_3 OS (ng m^{-3})	63.51 ± 19.82	41.10 ± 11.74	12.55 ± 4.68	9.27 ± 3.57	41.73 ± 13.50	27.81 ± 8.65	12.38 ± 4.39	9.85 ± 4.15
OS_a (ng m^{-3})	40.17 ± 16.25	29.01 ± 6.92	14.45 ± 1.64	11.12 ± 1.91	25.40 ± 8.12	19.63 ± 3.45	14.18 ± 1.06	11.5 ± 0.90

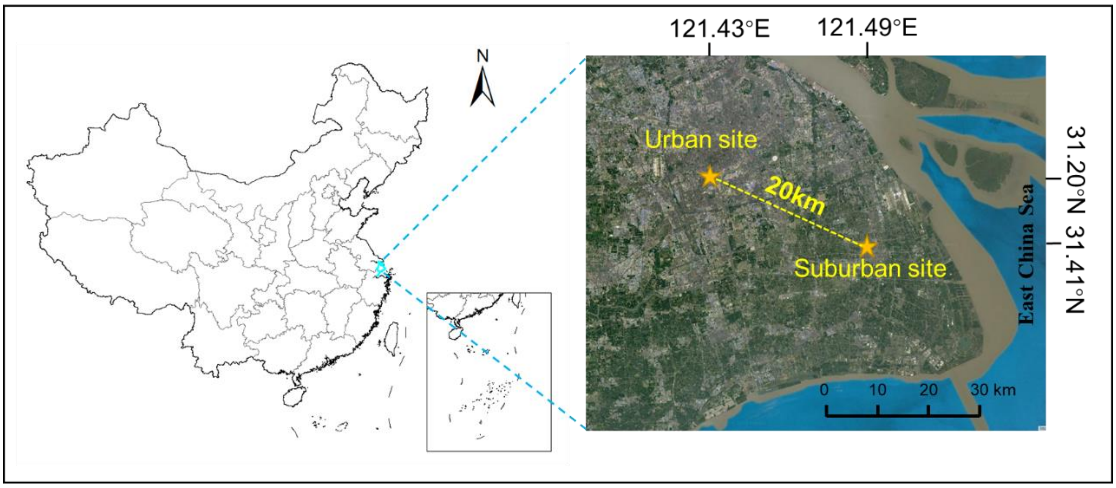

Figure S1. The locations of the sampling sites. The map was derived from Baidu Maps (BIDU, China).

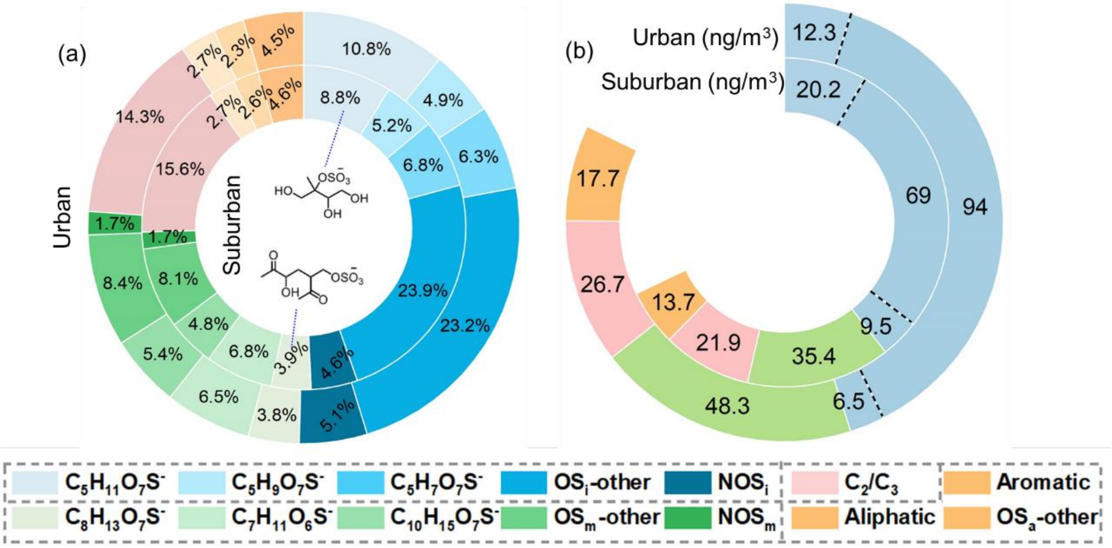

Figure S2. Average distributions in (a) mass concentrations and (b) mass fractions of various OSs in $PM_{2.5}$ collected in urban and suburban Shanghai in September 2020.

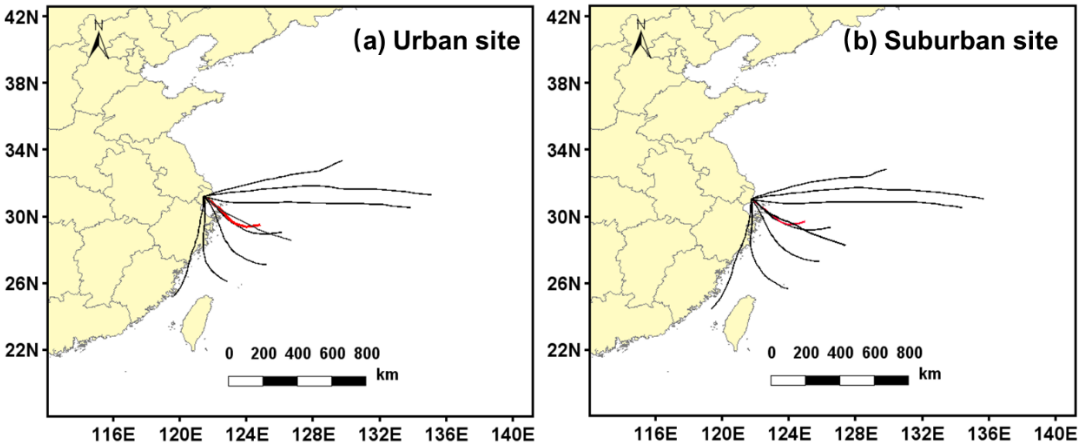

Figure S3. The 1-day (24 h) backward trajectories showing the daily air mass flows to the (a) urban and (b) suburban sites during period B.

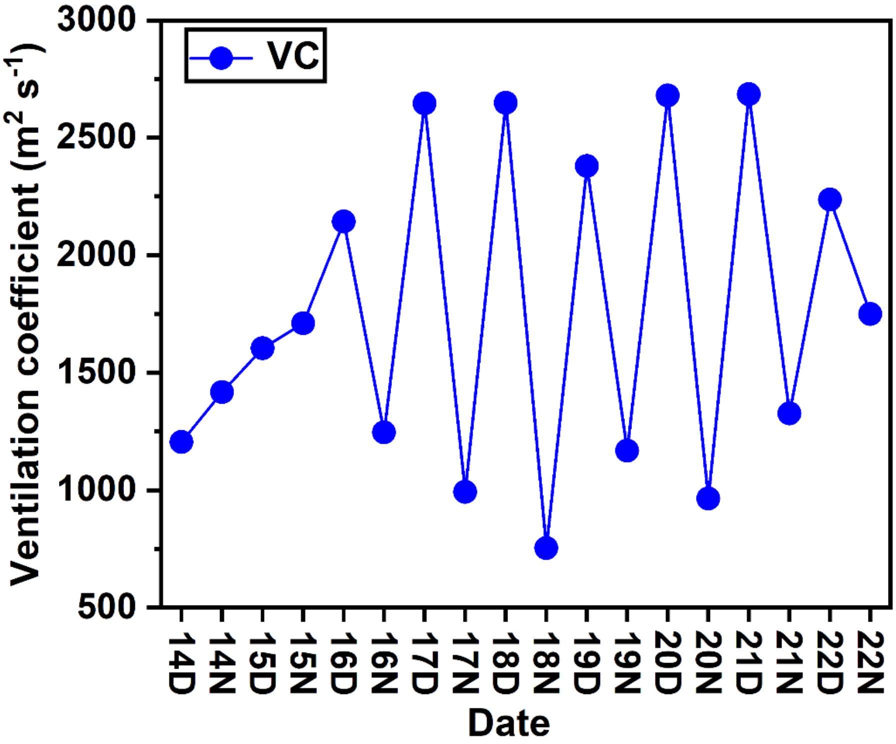

Figure S4. The temporal variations of ventilation coefficient (VC) during the sampling period.

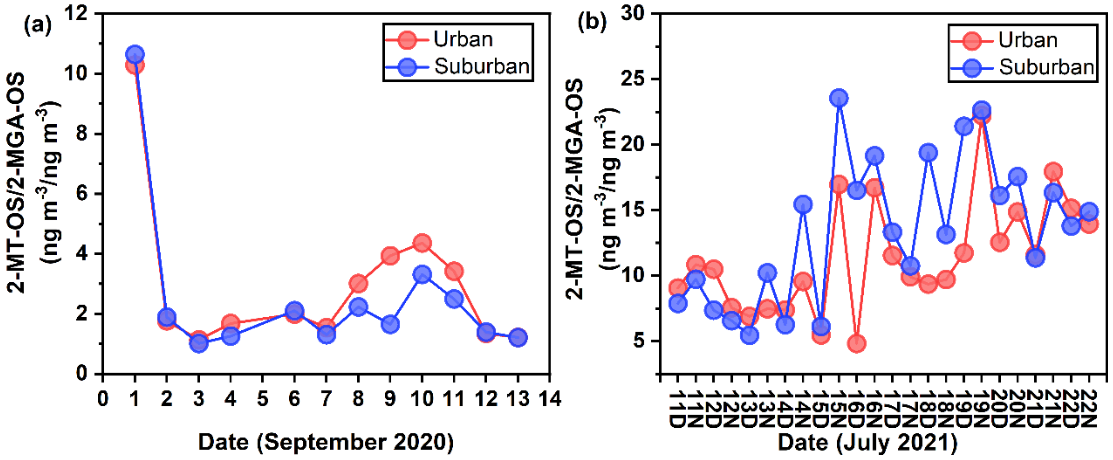

Figure S5. The temporal variations of the ratio of 2-MT-OS to 2-MGA-OS during (a) 2020 and (b) 2021 sampling campaigns.

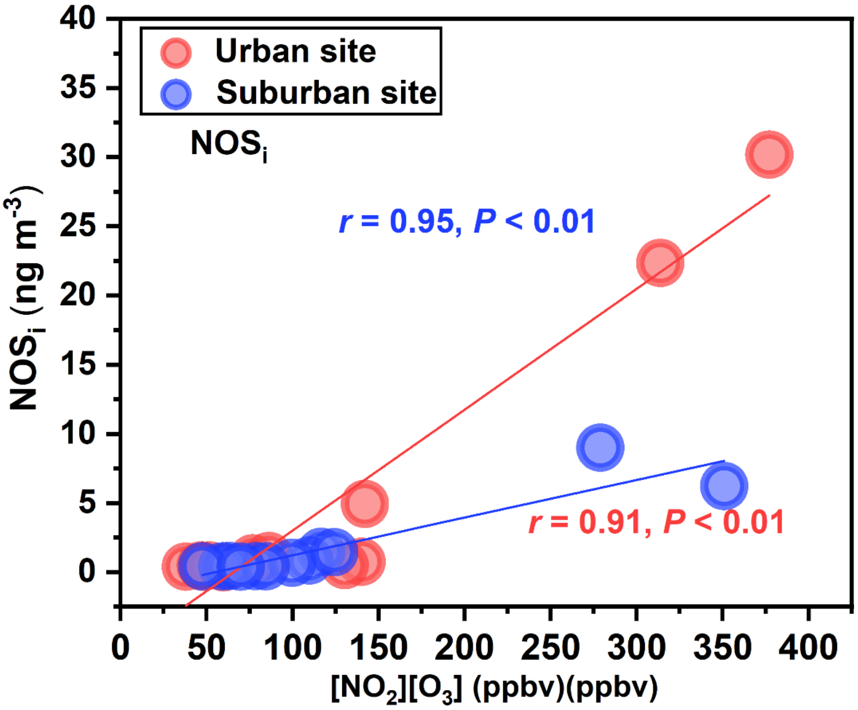

Figure S6. Scatterplots of the products of NO_2 and O_3 concentrations with the mass concentrations of NOS_i in $PM_{2.5}$ collected in the urban and suburban areas. Red and blue lines show regression lines at the urban and suburban sites, respectively.

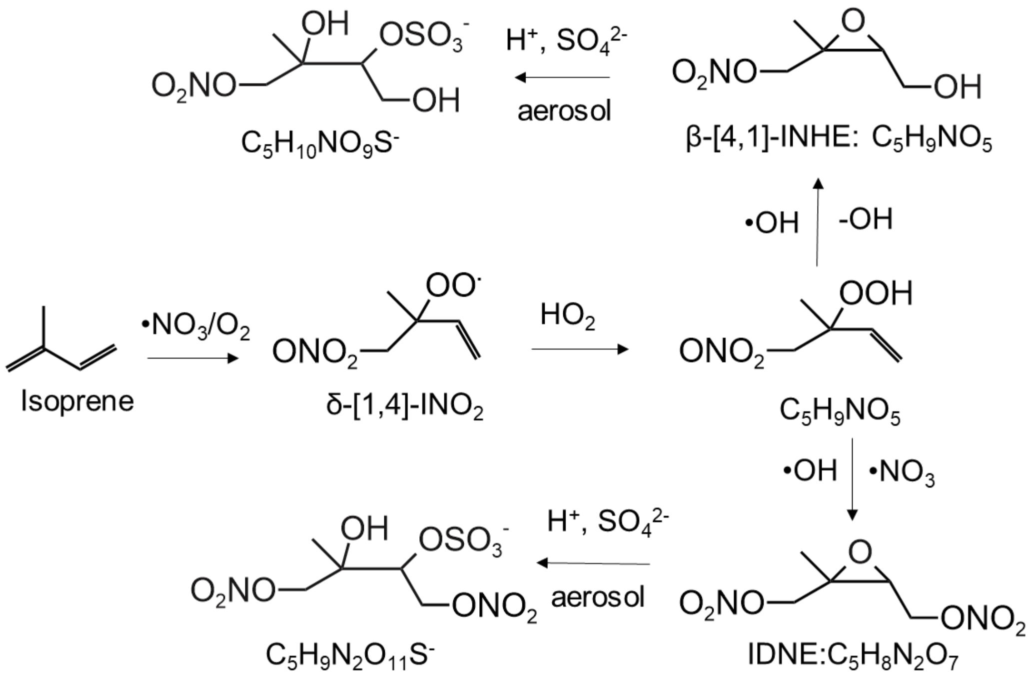

Figure S7. Possible formation mechanisms of $C_5H_{10}NO_9S^-$ and $C_5H_9N_2O_{11}S^-$.

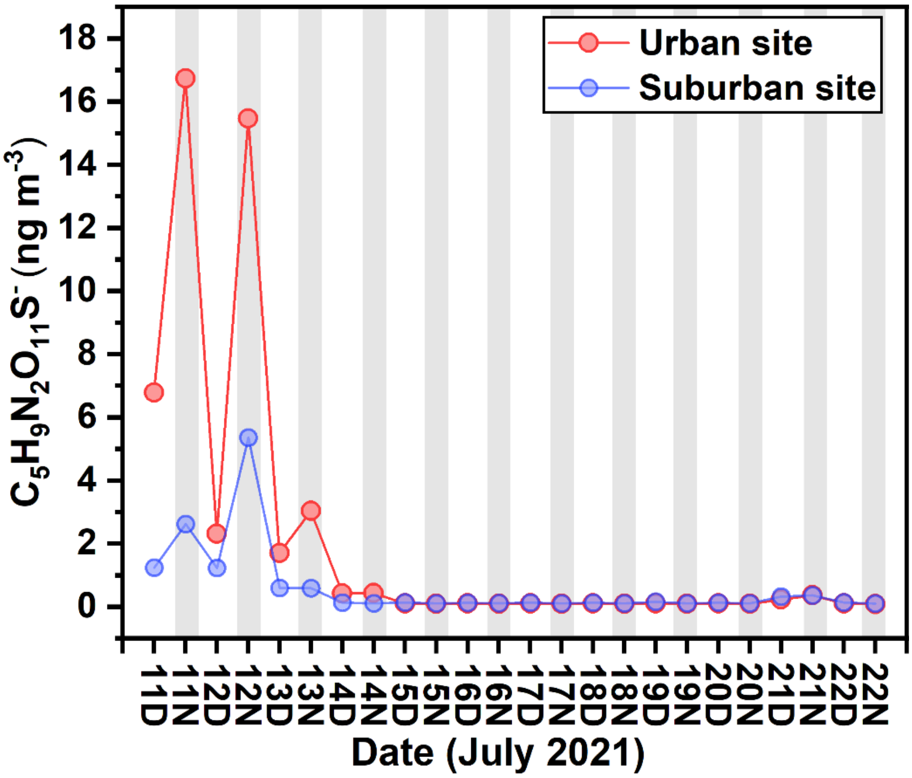

Figure S8. Temporal variations in the concentrations of $C_5H_9N_2O_{11}S^-$.

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