Supplement of Atmos. Chem. Phys., 23, 13433–13450, 2023
https://doi.org/10.5194/acp-23-13433-2023-supplement

Atmospheric Chemistry and Physics
Open Access

Supplement of

Spatial and diurnal variations of aerosol organosulfates in summertime Shanghai, China: potential influence of photochemical processes and anthropogenic sulfate pollution

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S1. Classification of OSs

Mass spectrometry data was processed to obtain the m/z information, formula, and intensity of detected molecular ions using the MassLynx v4.1 software. A molecular formula calculator was used to predict the possible formula of identified compounds that can be expressed as $C_nH_nO_nS_nN_n$ within the mass tolerance of ± 10 ppm. Specifically, a total of 212 organosulfates (OSs) were detected in $PM_{2.5}$ samples. The quantified (or semi-quantified) OSs were distributed over a wide mass range (m/z 167–691). 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 the characteristics of a double bond equivalent (DBE) value of 1–2, $n_O \leq 6$, and $n_H \geq 6$; and (3) compounds with 5 carbon atoms have the characteristics of a DBE value 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). The DBE value was calculated via the following equation (Yassine et al. 2014).

$$\mathrm{DBE} = 1 + n_{\mathrm{C}} - n_{\mathrm{H}}/2 + n_{\mathrm{N}}/2 \qquad (2)$$

where the n_{C}, n_{H}, and n_{N} denote the number of carbon, hydrogen, and nitrogen atoms in a molecular formula, respectively.

For monoterpene-derived OSs ($\mathrm{OS_m}$), they were grouped according to the following criteria. The compounds have the characteristics of 10 carbon atoms, effective oxygen atoms ($n_{\mathrm{Oeff}} = n_{\mathrm{O}} - 2n_{\mathrm{N}}$) of more than 4, and $2 \leq$ DBE value ≤ 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 $\mathrm{OS_m}$ 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 $\mathrm{OS_m}$ (e.g., $C_{10}H_{17}O_5S^-$) was performed, 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 a DBE value ≥ 2 and aromaticity equivalent (X_{C}) value ≥ 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 X_{C} value was calculated via the following equation (Yassine et al. 2014).

$$X_{\mathrm{C}} = [3(\mathrm{DBE} - (f_{\mathrm{m}} n_{\mathrm{O}} - f_{\mathrm{n}} n_{\mathrm{S}})) - 2]/[\mathrm{DBE} - (f_{\mathrm{m}} n_{\mathrm{O}} - f_{\mathrm{n}} n_{\mathrm{S}})] \qquad (3)$$

where the n_{O} and n_{S} denote the number of oxygen and 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 ion 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 X_C value of organosulfates can be simplified as the following equation (Ye et al. 2021).

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

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 ($P < 0.01$).

The OSs with a DBE value of 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 ($P <$ 0.01). 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.,$C_8H_{17}O_4S^-$; $r = 0.98$, $P < 0.01$) than typical OS_m ($r < 0.98$). In this study, aliphatic and aromatic OSs were referred to anthropogenic OSs (OS_a).

Several repotred OS_a (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	Phenyl sulfate	(Wang et al. 2021a)	
$C_7H_7SO_4S^-$	218.9786	Phenyl sulfate	(Wang et al. 2021a)	
$C_{11}H_{19}O_{11}S^-$	359.0648	Phenyl sulfate		0.77
$C_{10}H_{17}O_{12}S^-$	361.0441	Phenyl sulfate		0.64
$C_7H_{11}O_9S^-$	271.0124	Phenyl sulfate		0.65
$C_7H_{11}O_{10}S^-$	287.0073	Phenyl sulfate		0.69
$C_8H_{13}O_9S^-$	285.0280	Phenyl sulfate		0.70
$C_8H_{13}O_{10}S^-$	301.0229	Phenyl sulfate		0.81
$C_{11}H_{17}O_{11}S^-$	357.0492	Phenyl sulfate		0.86
$C_8H_{12}NO_{11}S^-$	330.0131	Phenyl sulfate		0.90
$C_8H_7O_5S^-$	215.0014	Phenyl sulfate		0.91
$C_8H_7NO_5S^-$	229.0045	Phenyl sulfate		0.89
$C_8H_7O_4S^-$	199.0065	Phenyl sulfate		0.98
$C_9H_7O_7S^-$	258.9912	Phenyl sulfate		0.89
$C_9H_7O_6S^-$	242.9963	Phenyl sulfate		0.74

$C_{34}H_{49}O_5S^-$	569.3301	Phenyl sulfate		0.64
$C_{43}H_{63}O_5S^-$	691.4396	Phenyl sulfate		0.72
$C_7H_{11}O_9S^-$	271.0124	Phenyl sulfate		0.74
$C_{10}H_7O_{11}S^-$	334.9709	Phenyl sulfate		0.86
$C_{10}H_5O_{11}S^-$	332.9553	Phenyl sulfate		0.77
$C_{10}H_5O_{10}S^-$	316.9603	Phenyl sulfate		0.72
$C_{12}H_7O_{13}S^-$	390.9607	Phenyl sulfate		0.65
$C_8H_7O_5S^-$	215.0014	Phenyl sulfate		0.77
$C_7H_5SO_5S^-$	200.9858	Phenyl sulfate		0.65
$C_9H_9O_4S^-$	213.0222	Phenyl sulfate		0.72
$C_7H_6SO_4S^-$	185.9987	Phenyl sulfate		0.73
$C_{18}H_{13}O_6S^-$	357.0433	Phenyl sulfate		0.69
$C_{23}H_{19}O_7S^-$	439.0851	Phenyl sulfate		0.65
$C_{25}H_{21}O_7S^-$	465.1008	Phenyl sulfate		0.77
$C_{24}H_{17}O_4S^-$	401.0848	Phenyl sulfate		0.73
$C_{27}H_{21}O_7S^-$	489.1008	Phenyl sulfate		0.90
$C_{12}H_{21}N_2O_{11}S$	401.0866	Phenyl 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 (OS_a). The compounds were adopted with r value greater than 0.6 ($P < 0.01$). 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 $[M\text{-}H]^-$	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 $[M-H]^-$	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.

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 site	Pearl River Delta, China	2012	Summer	0.68	-	-	-	0.68	(He et al. 2014)
	Shanghai, China	2012	Spring	0.02	0.20	0.22	-	0.44	(Ma et al. 2014)
			Summer	23.10	26.70	0.77	-	50.57	
			Autumn	0.06	0.77	0.79	-	1.62	
			Winter	0.82	0.90	0.72	-	2.44	
	Beijing, China	2016	Spring	16.20	13.00	-	19.50	48.70	(Wang et al. 2018)
	Beijing, China	2016	Summer	12.30	15.10	-	-	27.40	(Wang et al. 2020)
			Winter	2.00	6.00	-	-	8.00	
	Atlanta, GA, USA	2014	Summer	1122.98	67.90	-	58.50	1249.38	(Hettiyadura et al. 2019)
	Guangzhou, China	2019	Summer	181.80	10.80	-	-	192.6	(Bryant et al. 2021)
	Tianjing, China	2019	Winter	600.00	-	-	-	600.00	(Ding et al. 2022)
			Summer	400.00	-	-	-	400.00	
	Patra, Greece	2018	Winter	31.50	20.70	3.10	-	55.30	(Kanellopoulos et al. 2022)
			Spring	75.70	32.40	1.30	-	109.40	
			Summer	658.00	37.70	1.60	-	697.30	
			Autumn	53.20	25.40	1.10	-	79.70	
	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	460.20	26.22	-	-	486.42	
		2017	Winter	88.03	20.96	-	-	108.99	
	Beijing, China	2016	Summer	236.64	21.70	-	-	258.34	
		2017	Winter	176.32	36.01	-	-	212.33	
	Shanghai, China	2016	Summer	326.40	34.90	-	-	361.30	
		2017	Winter	70.31	32.32	-	-	102.63	

	Copenhagen, Denmark	2011	Summer	31.60	12.00	-	15.40	59.00	(Nguyen et al. 2014)
	Xi'an, China	2014	Winter	-	-	0.14	77.30	77.44	(Huang et al. 2018)
	Manaus, Brazil	2016	Summer	1200.90	116.30	-	194.20		(Riva et al. 2019)
	Birmingham, Al, USA	2013	Summer	179.00	-	-	29.10	208.10	
	Mexico City, Mexico	2006	Spring	1.50	-	-	-	1.50	(Olson et al. 2011)
	Rotterdam, Netherlands	2013	Winter	3.32	22.91	9.07	-	35.30	(Glasius et al. 2018)
	Bakersfield, CA, USA	2010	Summer	0.60	-	-	-	0.60	(Olson et al. 2011)
	Riverside, CA, USA	2005	Summer	0.80	-	-	-	0.80	
	Cleveland, OH, USA	2007	Summer	0.40	-	-	-	0.40	
	Lahore, Pakistan		Winter	3.80	-	2.02	-	5.82	(Kundu et al. 2013)
	Pasadena, USA	2010	Summer	-	-	1.03	-	1.03	(Riva et al. 2015)
	Dept of Environmental Qualify Fresno, CA, USA	2016	Winter	74.00	-	-	-	74.00	(Chen et al. 2021)
			Summer	44.00	-	-	-	44.00	
Suburban site	Zion, Illinois, USA	2013	Spring	121.10	8.70	-	-	129.80	(Hughes et al. 2021)
	Towson, MD, USA	2012	Spring	3.75	2.62	1.60	0.70	8.67	(Meade et al. 2016)
			Summer	19.96	0.80	1.40	0.24	22.4	
			Autumn	2.22	2.90	1.40	0.35	6.87	
			Winter	0.50	2.41	0.90	0.30	4.11	
	Lille Valby, Denmark	2009	Summer	0.27	2.40	-	-	2.67	(Yttri et al. 2011)
	Beijing, China	2016	Summer	5.90	16.10	-	-	22.0	(Wang et al. 2020)
			Winter	69.50	18.70	-	-	88.20	
	Wanqingsha, China	2010	Autumn	0.85	101.70	-	-	102.55	(He et al. 2014)
	Birkenes, Norway	2013	Winter	0.96	1.53	0.65	-	3.14	(Glasius et al. 2018)
	Risoe, Denmark		Winter	2.28	6.39	2.16	-	10.83	
Rural site	Cabauw, Netherlands		Winter	1.98	16.10	6.67	-	24.75	
	Melpitz, Germany		Winter	11.12	49.33	32.83	-	93.28	

	Vavihill, Sweden		Winter	2.75	6.39	4.15	-	13.29	
	Centreville, AL, USA		Summer	15.40	-	1.16	20.83	37.39	(Hettiyadura et al. 2017)
	Look Rock, TN, USA	2013	Summer	180.90	-	-	-	-	(Budisulistiorini et al. 2015)
	Wangdu, China	2014	Summer	-	36.70	-	-	36.70	(Brüggemann et al. 2019)
	Yorkville, GA, USA	2010	Summer	115.11	-	-	-	115.11	(Lin et al. 2013)
	Acadia National Park, ME, USA	2016	Summer	62.00	-	-	-	62.00	(Chen et al. 2021)
	Bondville, IL, USA		Summer	83.00	-	-	-	83.00	
	Londonderry, NH, USA		Winter	3.00	-	-	-	3.00	
			Summer	73.00				73.00	
	Great Smoky Mountains National Park, TN, USA		Winter	18.00	-	-	-	18.00	
			Summer	75.00	-	-	-	75.00	
	Hawaii Volcanoes National Park, HI, USA		Summer	3.00	-	-	-	3.00	
	Kenai Peninsula Borough, AK, USA		Summer	58.00	-	-	-	58.00	
	Lye Brook Wilderness Area, VT, USA		Summer	45.00	-	-	-	45.00	
	Mingo National Wildlife Refuge, MO, USA		Summer	98.00	-	-	-	98.00	
	Organ Pipe Cactus National Monument, AZ, USA		Winter	2.00	-	-	-	2.00	
			Summer	3.00	-	-	-	3.00	
	Point Reyes National Seashore, CA, USA		Winter	1.00	-	-	-	1.00	
			Summer	22.00	-	-	-	22.00	
	Presque Isle, ME, USA		Summer	2130.00	-	-	-	2130.00	
	Redwood National Park, CA, USA		Winter	3.00	-	-	-	3.00	
			Summer	42.00	-	-	-	42.00	

	Rocky Mountain National Park, CO, USA	2016	Winter	2.00	-	-	-	2.00	(Chen et al. 2021)
			Summer	19.00	-	-	-	19.00	
	Shining Rock Wilderness, NC, USA		Summer	1130.00	-	-	-	1130.00	
	Snoqualmie Pass, WA		Winter	2.00	-	-	-	2.00	
			Summer	24.00	-	-	-	24.00	
	Tallgrass Prairie National Preserve, KS, USA		Summer	62.00	-	-	-	62.00	
	UL Bend, MT, USA		Winter	12.00	-	-	-	12.00	
			Summer	16.00	-	-	-	16.00	
	White Pass, WA, USA		Summer	9.00	-	-	-	9.00	
Forest site	The Green Ocean Amazon	2014	Spring	1733.1	116.3	-	268.4	2117.8	(Riva et al. 2019)
	Brasschaat, Belgium	2007	Summer	12.07	1.51	-	-	13.58	(Gómez-González et al. 2012)
	Fichtelgebirge, Germany	2002	Summer	-	5.00	-	-	5.00	(Iinuma et al. 2010)
	Hyytiälä, Finland	2009	Summer	-	4.91	1.47	-	6.38	(Yttri et al. 2011)
	Silkeborg, Denmark	2008	Spring	0.03	0.52	-	-	0.55	(Kristensen andGlasius 2011)
Polar site	Station Nord, Greenland	2010	Winter	5.03	0.43	-	-	5.46	(Hansen et al. 2014)
	Zeppelin Mountain, Svalbard	2010	Winter	1.34	0.14	-	-	1.48	(Hansen et al. 2014)
Urban site	**Shanghai, China**	**2021**	**Summer**	**85.38**	**30.61**	**19.31**	**23.38**	**158.68**	**This study**
Suburban site				**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 (℃)	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

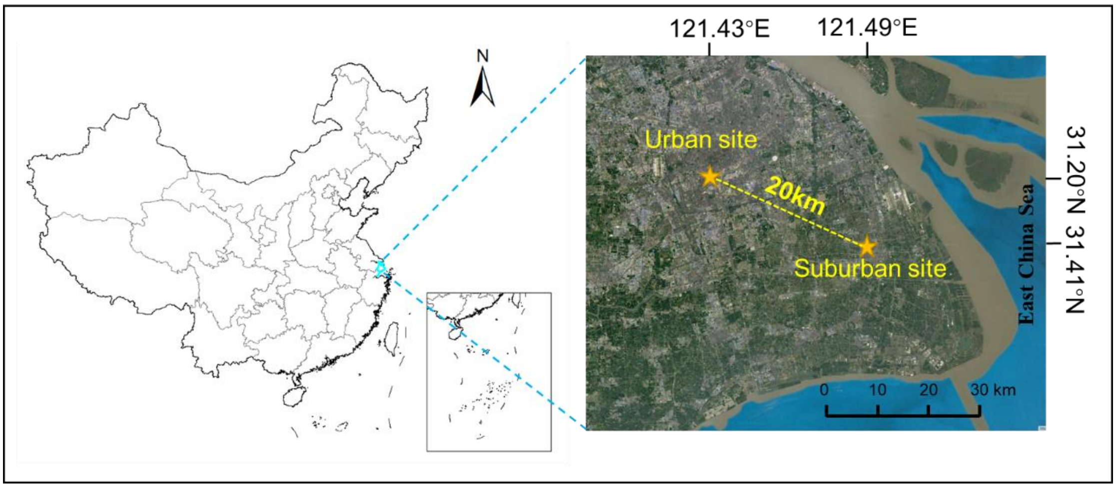

Figure S1. The locations of the sampling sites. The map was derived from ©Baidu Maps (BIDU, China).

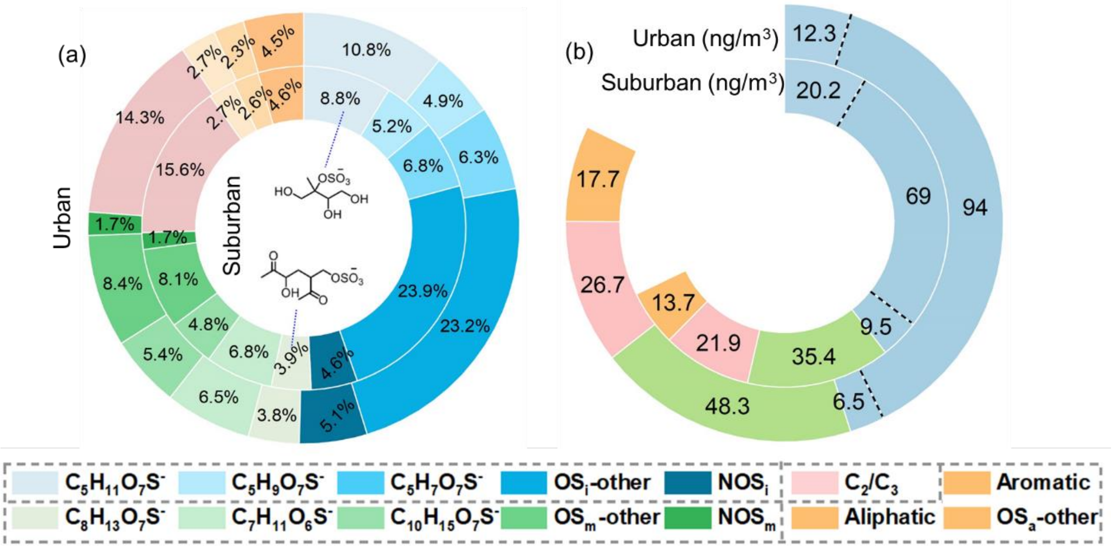

Figure S2. Average distributions in (a) mass concentrations and (b) mass fractions of various OSs in $PM_{2.5}$ collected in urban and suburban Shanghai in September 2020.

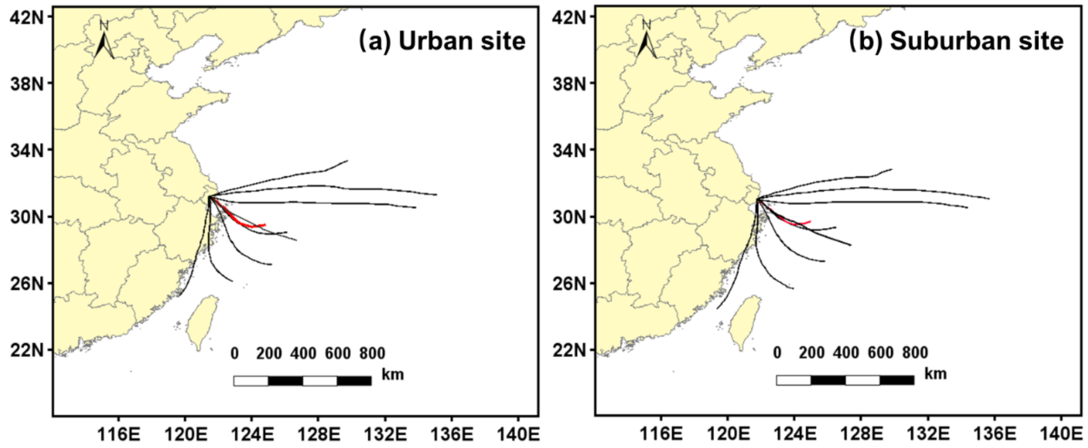

Figure S3. The 1-day (24 h) backward trajectories showing the daily air mass flows to the (a) urban and (b) suburban sites during period B.

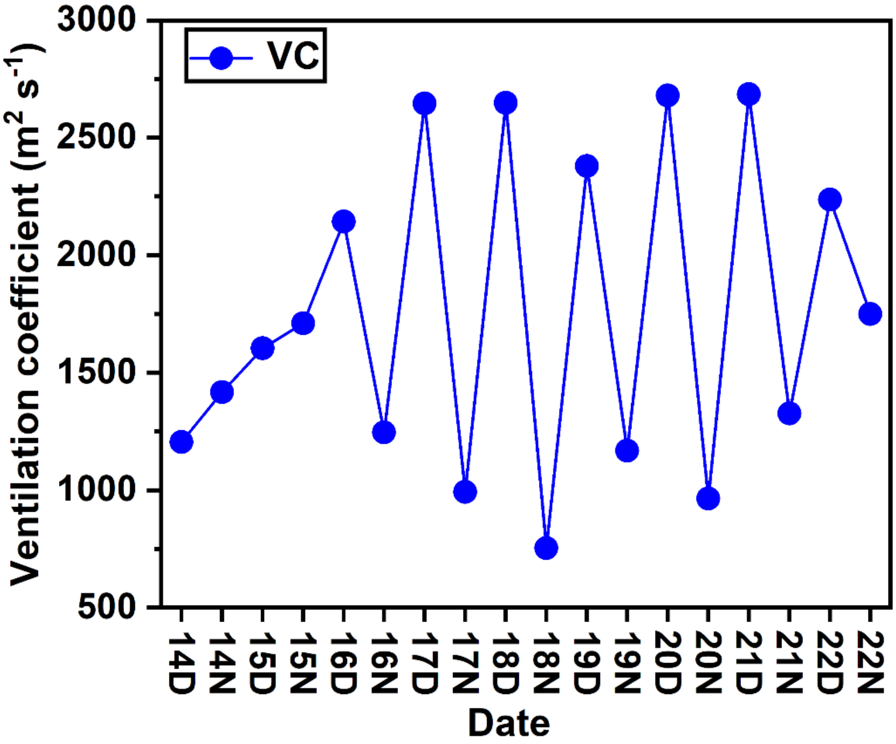

Figure S4. The temporal variations of ventilation coefficient (VC) during the sampling period.

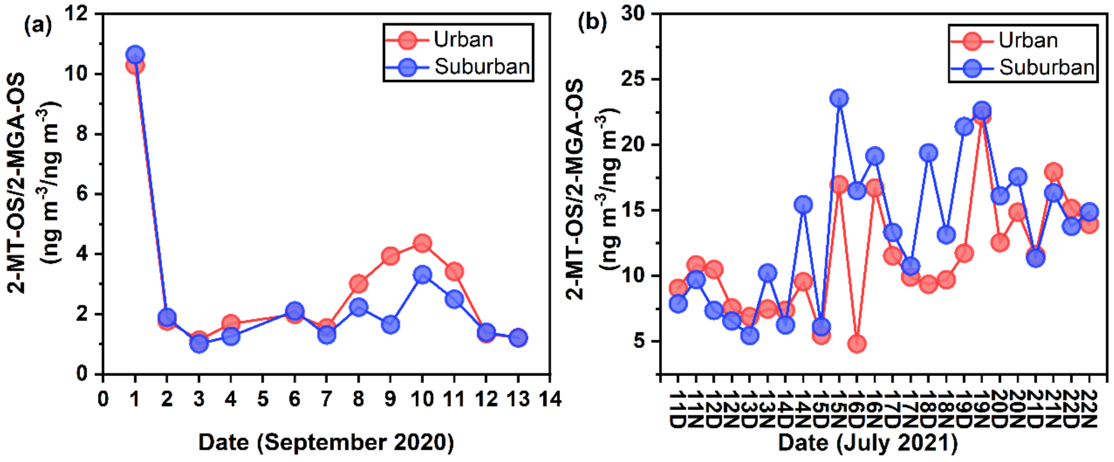

Figure S5. The temporal variations of the ratio of 2-MT-OS to 2-MGA-OS during (a) 2020 and (b) 2021 sampling campaigns.

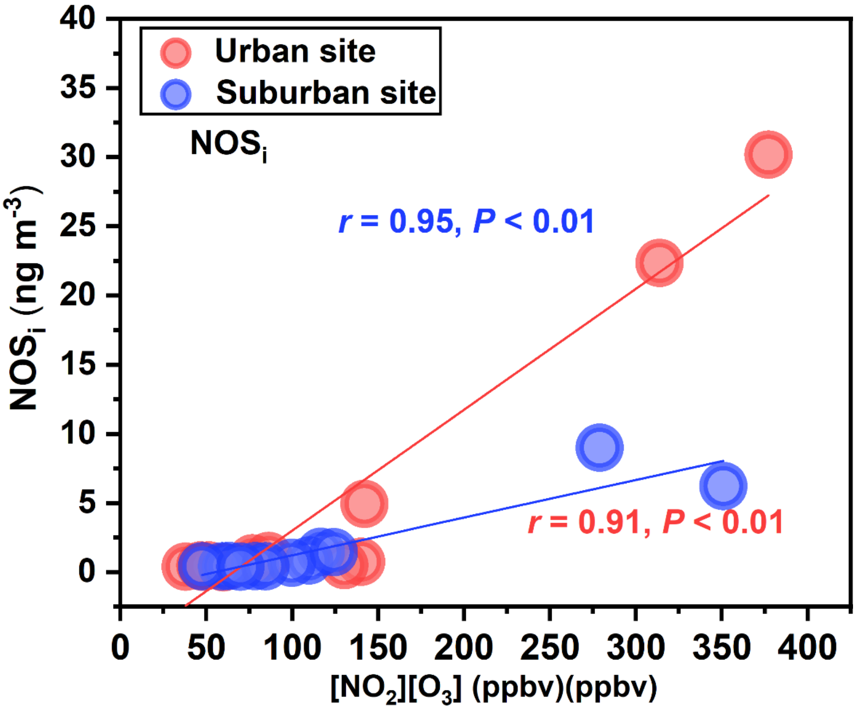

Figure S6. Scatterplots of the products of NO_2 and O_3 concentrations with the mass concentrations of NOS_i in $PM_{2.5}$ collected in the urban and suburban areas. Red and blue lines show regression lines at the urban and suburban sites, respectively.

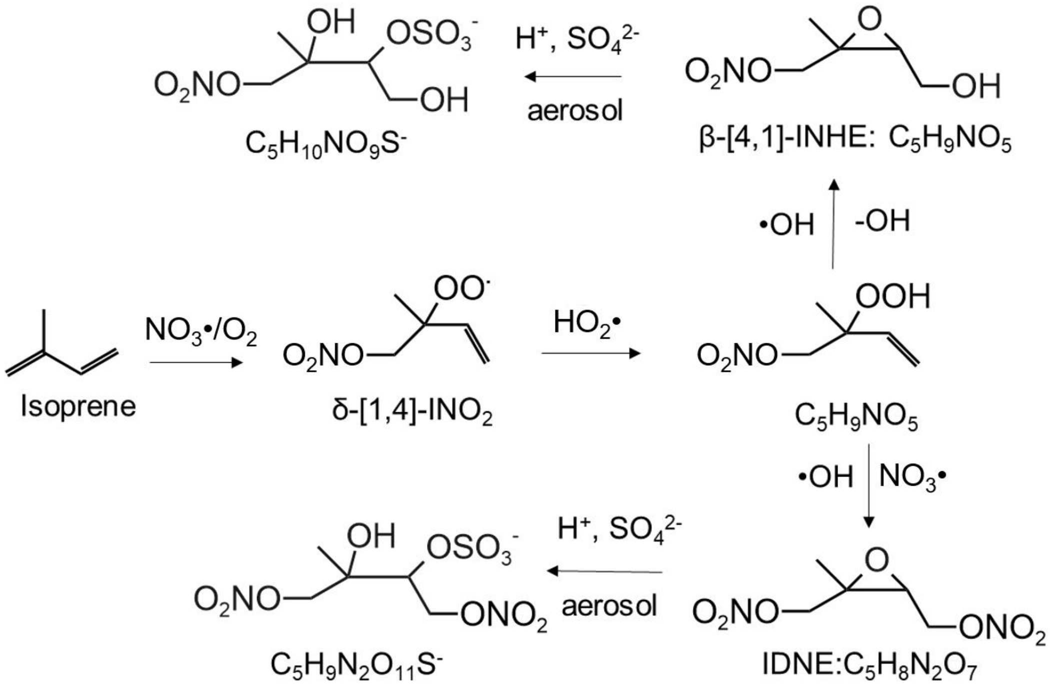

Figure S7. Possible formation mechanisms of $C_5H_{10}NO_9S^-$ and $C_5H_9N_2O_{11}S^-$.

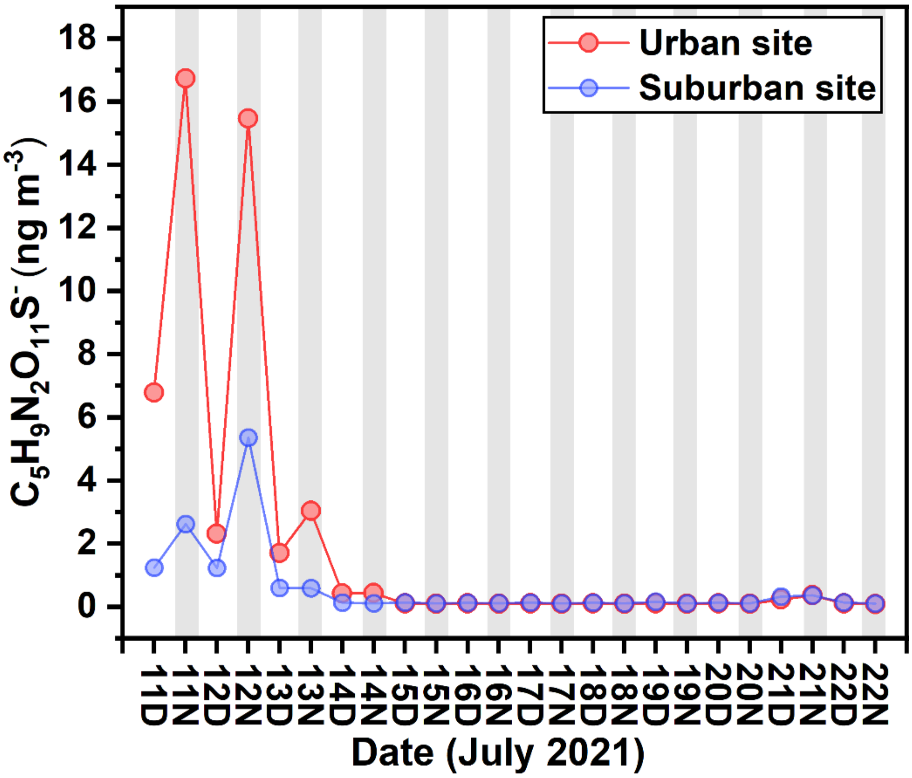

Figure S8. Temporal variations in the concentration of $C_5H_9N_2O_{11}S^-$.

REFERENCES

Berndt, T., Mentler, B., Scholz, W., Fischer, L., Herrmann, H., Kulmala, M., and Hansel, A.: Accretion Product Formation from Ozonolysis and OH Radical Reaction of alpha-Pinene: Mechanistic Insight and the Influence of Isoprene and Ethylene, Environ. Sci. Technol., 52, 11069-11077, 10.1021/acs.est.8b02210, 2018.

Berndt, T., Richters, S., Jokinen, T., Hyttinen, N., Kurten, T., Otkjaer, R. V., Kjaergaard, H. G., Stratmann, F., Herrmann, H., Sipila, M., Kulmala, M., and Ehn, M.: Hydroxyl radical-induced formation of highly oxidized organic compounds, Nat. Commun., 7, 13677, 10.1038/ncomms13677, 2016.

Boyd, C. M., Sanchez, J., Xu, L., Eugene, A. J., Nah, T., Tuet, W. Y., Guzman, M. I., and Ng, N. L.: Secondary organic aerosol formation from the β-pinene+NO<sub>3</sub> system: effect of humidity and peroxy radical fate, Atmos. Chem. Phys., 15, 7497-7522, 10.5194/acp-15-7497-2015, 2015.

Brüggemann, M., van Pinxteren, D., Wang, Y., Yu, J. Z., and Herrmann, H.: Quantification of known and unknown terpenoid organosulfates in PM10 using untargeted LC–HRMS/MS: contrasting summertime rural Germany and the North China Plain, Environmental Chemistry, 16, 10.1071/en19089, 2019.

Bryant, D. J., Elzein, A., Newland, M., White, E., Swift, S., Watkins, A., Deng, W., Song, W., Wang, S., Zhang, Y., Wang, X., Rickard, A. R., and Hamilton, J. F.: Importance of Oxidants and Temperature in the Formation of Biogenic Organosulfates and Nitrooxy Organosulfates, ACS Earth Space Chem., 5, 2291-

2306, 10.1021/acsearthspacechem.1c00204, 2021.

Budisulistiorini, S. H., Li, X., Bairai, S. T., Renfro, J., Liu, Y., Liu, Y. J., McKinney, K. A., Martin, S. T., McNeill, V. F., Pye, H. O. T., Nenes, A., Neff, M. E., Stone, E. A., Mueller, S., Knote, C., Shaw, S. L., Zhang, Z., Gold, A., and Surratt, J. D.: Examining the effects of anthropogenic emissions on isoprene-derived secondary organic aerosol formation during the 2013 Southern Oxidant and Aerosol Study (SOAS) at the Look Rock, Tennessee ground site, Atmospheric Chemistry and Physics, 15, 8871-8888, 10.5194/acp-15-8871-2015, 2015.

Chen, Y., Dombek, T., Hand, J., Zhang, Z., Gold, A., Ault, A. P., Levine, K. E., and Surratt, J. D.: Seasonal Contribution of Isoprene-Derived Organosulfates to Total Water-Soluble Fine Particulate Organic Sulfur in the United States, ACS Earth and Space Chemistry, 5, 2419-2432, 10.1021/acsearthspacechem.1c00102, 2021.

Ding, S., Chen, Y., Devineni, S. R., Pavuluri, C. M., and Li, X. D.: Distribution characteristics of organosulfates (OSs) in PM2.5 in Tianjin, Northern China: Quantitative analysis of total and three OS species, Sci. Total. Environ., 834, 155314, 10.1016/j.scitotenv.2022.155314, 2022.

Ehn, M., Kleist, E., Junninen, H., Petäjä, T., Lönn, G., Schobesberger, S., Dal Maso, M., Trimborn, A., Kulmala, M., Worsnop, D. R., Wahner, A., Wildt, J., and Mentel, T. F.: Gas phase formation of extremely oxidized pinene reaction products in chamber and ambient air, Atmos. Chem. Phys., 12, 5113-5127, 10.5194/acp-12-5113-2012, 2012.

Ehn, M., Thornton, J. A., Kleist, E., Sipila, M., Junninen, H., Pullinen, I., Springer, M., Rubach, F., Tillmann, R., Lee, B., Lopez-Hilfiker, F., Andres, S., Acir, I. H., Rissanen, M., Jokinen, T., Schobesberger, S., Kangasluoma, J., Kontkanen, J., Nieminen, T., Kurten, T., Nielsen, L. B., Jorgensen, S., Kjaergaard, H. G., Canagaratna, M., Maso, M. D., Berndt, T., Petaja, T., Wahner, A., Kerminen, V. M., Kulmala, M., Worsnop, D. R., Wildt, J., and Mentel, T. F.: A large source of low-volatility secondary organic aerosol, Nature, 506, 476-479, 10.1038/nature13032, 2014.

Glasius, M., Hansen, A. M. K., Claeys, M., Henzing, J. S., Jedynska, A. D., Kasper-Giebl, A., Kistler, M., Kristensen, K., Martinsson, J., Maenhaut, W., Nøjgaard, J. K., Spindler, G., Stenström, K. E., Swietlicki, E., Szidat, S., Simpson, D., and Yttri, K. E.: Composition and sources of carbonaceous aerosols in Northern Europe during winter, Atmospheric Environment, 173, 127-141, 10.1016/j.atmosenv.2017.11.005, 2018.

Gómez-González, Y., Wang, W., Vermeylen, R., Chi, X., Neirynck, J., Janssens, I. A., Maenhaut, W., and Claeys, M.: Chemical characterisation of atmospheric aerosols during a 2007 summer field campaign at Brasschaat, Belgium: sources and source processes of biogenic secondary organic aerosol, Atmospheric Chemistry and Physics, 12, 125-138, 10.5194/acp-12-125-2012, 2012.

Guo, Y., Yan, C., Liu, Y., Qiao, X., Zheng, F., Zhang, Y., Zhou, Y., Li, C., Fan, X., Lin, Z., Feng, Z., Zhang, Y., Zheng, P., Tian, L., Nie, W., Wang, Z., Huang, D., Daellenbach, K. R., Yao, L., Dada, L., Bianchi, F., Jiang, J., Liu, Y., Kerminen, V.-M., and Kulmala, M.: Seasonal variation in oxygenated organic molecules in urban Beijing and their contribution to secondary organic aerosol, Atmos. Chem. Phys., 22, 10077-10097, 10.5194/acp-22-10077-2022, 2022.

Hansen, A. M. K., Kristensen, K., Nguyen, Q. T., Zare, A., Cozzi, F., Nøjgaard, J. K., Skov, H., Brandt, J., Christensen, J. H., Ström, J., Tunved, P., Krejci, R., and Glasius, M.: Organosulfates and organic acids in Arctic aerosols: speciation, annual variation and concentration levels, Atmos. Chem. Phys., 14, 7807-7823, 10.5194/acp-14-7807-2014, 2014.

He, Q. F., Ding, X., Wang, X. M., Yu, J. Z., Fu, X. X., Liu, T. Y., Zhang, Z., Xue, J., Chen, D. H., Zhong, L. J., and Donahue, N. M.: Organosulfates from pinene and isoprene over the Pearl River Delta, South China: seasonal variation and implication in formation mechanisms, Environ Sci Technol, 48, 9236-9245, 10.1021/es501299v, 2014.

Hettiyadura, A. P. S., Al-Naiema, I. M., Hughes, D. D., Fang, T., and Stone, E. A.: Organosulfates in Atlanta, Georgia: anthropogenic influences on biogenic secondary organic aerosol formation, Atmos. Chem. Phys., 19, 3191-3206, 10.5194/acp-19-3191-2019, 2019.

Hettiyadura, A. P. S., Stone, E. A., Kundu, S., Baker, Z., Geddes, E., Richards, K., and Humphry, T.: Determination of atmospheric organosulfates using HILIC chromatography with MS detection, Atoms. Meas. Tech., 8, 2347-2358, 10.5194/amt-8-2347-2015, 2015.

Hettiyadura, A. P. S., Jayarathne, T., Baumann, K., Goldstein, A. H., de Gouw, J. A., Koss, A., Keutsch, F. N., Skog, K., and Stone, E. A.: Qualitative and quantitative analysis of atmospheric organosulfates in Centreville, Alabama, Atmos. Chem. Phys., 17, 1343-1359, 10.5194/acp-17-1343-2017, 2017.

Huang, R.-J., Cao, J., Chen, Y., Yang, L., Shen, J., You, Q., Wang, K., Lin, C., Xu, W., Gao, B., Li, Y., Chen, Q., Hoffmann, T., O'Dowd, C. D., Bilde, M., and Glasius, M.: Organosulfates in atmospheric aerosol: synthesis and quantitative analysis of PM2:5 from Xi'an, northwestern China, Atoms. Meas. Tech., 11, 3447-3456, 10.5194/amt-11-3447-2018, 2018.

Hughes, D. D., Christiansen, M. B., Milani, A., Vermeuel, M. P., Novak, G. A., Alwe, H. D., Dickens, A. F., Pierce, R. B., Millet, D. B., Bertram, T. H., Stanier, C. O., and Stone, E. A.: PM2.5 chemistry, organosulfates, and secondary organic aerosol during the 2017 Lake Michigan Ozone Study, Atmos. Environ., 244, 10.1016/j.atmosenv.2020.117939, 2021.

Iinuma, Y., Böge, O., Gräfe, R., and Herrmann, H.: Methyl-Nitrocatechols: Atmospheric Tracer Compounds for Biomass Burning Secondary Organic Aerosols, Environ. Sci. Technol., 44, 8453-8459, https://doi.org/10.1021/es070938t, 2010.

Jiang, H., Li, J., Tang, J., Cui, M., Zhao, S., Mo, Y., Tian, C., Zhang, X., Jiang, B., Liao, Y., Chen, Y., and Zhang, G.: Molecular characteristics, sources, and formation pathways of organosulfur compounds in ambient aerosol in Guangzhou, South China, Atmos. Chem. Phys., 22, 6919-6935, 10.5194/acp-22-6919-2022, 2022.

Jokinen, T., Sipila, M., Richters, S., Kerminen, V. M., Paasonen, P., Stratmann, F., Worsnop, D., Kulmala, M., Ehn, M., Herrmann, H., and Berndt, T.: Rapid autoxidation forms highly oxidized RO2 radicals in the atmosphere, Angew. Chem. Int. Ed. Engl., 53, 14596-14600, 10.1002/anie.201408566, 2014.

Kanellopoulos, P. G., Kotsaki, S. P., Chrysochou, E., Koukoulakis, K., Zacharopoulos, N., Philippopoulos, A., and Bakeas, E.: PM2.5-bound organosulfates in two Eastern Mediterranean cities: The dominance of isoprene organosulfates, Chemosphere, 297, 134103, 10.1016/j.chemosphere.2022.134103, 2022.

Kristensen, K. and Glasius, M.: Organosulfates and oxidation products from biogenic hydrocarbons in fine aerosols from a forest in North West Europe during spring, Atmos. Environ., 45, 4546-4556, 10.1016/j.atmosenv.2011.05.063, 2011.

Kundu, S., Quraishi, T. A., Yu, G., Suarez, C., Keutsch, F. N., and Stone, E. A.: Evidence and quantitation of aromatic organosulfates in ambient aerosols in Lahore, Pakistan, Atmospheric Chemistry and Physics, 13, 4865-4875, 10.5194/acp-13-4865-2013, 2013.

Lin, Y. H., Knipping, E. M., Edgerton, E. S., Shaw, S. L., and Surratt, J. D.: Investigating the influences of SO<sub>2</sub> and NH<sub>3</sub> levels on isoprene-derived secondary organic aerosol formation using conditional sampling approaches, Atmospheric Chemistry and Physics, 13, 8457-8470, 10.5194/acp-13-8457-2013, 2013.

Ma, J., Ungeheuer, F., Zheng, F., Du, W., Wang, Y., Cai, J., Zhou, Y., Yan, C., Liu, Y., Kulmala, M., Daellenbach, K. R., and Vogel, A. L.: Nontarget Screening Exhibits a Seasonal Cycle of PM2.5 Organic Aerosol Composition in Beijing, Environ. Sci. Technol., 56, 7017-7028, 10.1021/acs.est.1c06905, 2022.

Ma, Y., Xu, X., Song, W., Geng, F., and Wang, L.: Seasonal and diurnal variations of particulate organosulfates in urban Shanghai, China, Atmos. Environ., 85, 152-160, 10.1016/j.atmosenv.2013.12.017, 2014.

Meade, L. E., Riva, M., Blomberg, M. Z., Brock, A. K., Qualters, E. M., Siejack, R. A., Ramakrishnan, K., Surratt, J. D., and Kautzman, K. E.: Seasonal variations of fine particulate organosulfates derived from biogenic and anthropogenic hydrocarbons in the mid-Atlantic United States, Atmospheric Environment, 145, 405-414, 10.1016/j.atmosenv.2016.09.028, 2016.

Nestorowicz, K., Jaoui, M., Rudzinski, K. J., Lewandowski, M., Kleindienst, T. E., Spolnik, G., Danikiewicz, W., and Szmigielski, R.: Chemical composition of isoprene SOA under acidic and non-acidic conditions: effect of relative humidity, Atmos Chem Phys, 18, 18101-18121, 10.5194/acp-18-18101-2018, 2018.

Nguyen, Q. T., Christensen, M. K., Cozzi, F., Zare, A., Hansen, A. M. K., Kristensen, K., Tulinius, T. E., Madsen, H. H., Christensen, J. H., Brandt, J., Massling, A., Nøjgaard, J. K., and Glasius, M.: Understanding the anthropogenic influence on formation of biogenic secondary organic aerosols in Denmark via analysis of organosulfates and related oxidation products, Atmos. Chem. Phys., 14, 8961-8981, 10.5194/acp-14-8961-2014, 2014.

Nozière, B., Ekström, S., Alsberg, T., and Holmström, S.: Radical-initiated formation of organosulfates and surfactants in atmospheric aerosols, Geophys. Res. Lett., 37, n/a-n/a, 10.1029/2009gl041683, 2010.

Olson, C. N., Galloway, M. M., Yu, G., Hedman, C. J., Lockett, M. R., Yoon, T., Stone, E. A., Smith, L. M., and Keutsch, F. N.: Hydroxycarboxylic acid-derived organosulfates: synthesis, stability, and quantification in ambient aerosol, Environ Sci Technol, 45, 6468-6474, 10.1021/es201039p, 2011.

Riva, M., Budisulistiorini, S. H., Zhang, Z., Gold, A., and Surratt, J. D.: Chemical characterization of secondary organic aerosol constituents from isoprene ozonolysis in the presence of acidic aerosol, Atmospheric Environment, 130, 5-13, 10.1016/j.atmosenv.2015.06.027, 2016a.

Riva, M., Da Silva Barbosa, T., Lin, Y. H., Stone, E. A., Gold, A., and Surratt, J. D.: Chemical characterization of organosulfates in secondary organic aerosol derived from the photooxidation of alkanes, Atmos. Chem. Phys., 16, 11001-11018, 10.5194/acp-16-11001-2016, 2016b.

Riva, M., Tomaz, S., Cui, T., Lin, Y. H., Perraudin, E., Gold, A., Stone, E. A., Villenave, E., and Surratt, J. D.: Evidence for an unrecognized secondary anthropogenic source of organosulfates and sulfonates: gas-phase oxidation of polycyclic aromatic hydrocarbons in the presence of sulfate aerosol, Environ. Sci. Technol., 49, 6654-6664, 10.1021/acs.est.5b00836, 2015.

Riva, M., Chen, Y., Zhang, Y., Lei, Z., Olson, N. E., Boyer, H. C., Narayan, S., Yee, L. D., Green, H. S., Cui, T., Zhang, Z., Baumann, K., Fort, M., Edgerton, E., Budisulistiorini, S. H., Rose, C. A., Ribeiro, I. O., RL, E. O., Dos Santos, E. O., Machado, C. M. D., Szopa, S., Zhao, Y., Alves, E. G., de Sa, S. S., Hu, W., Knipping, E. M., Shaw, S. L., Duvoisin Junior, S., de Souza, R. A. F., Palm, B. B., Jimenez, J. L., Glasius, M., Goldstein, A. H., Pye, H. O. T., Gold, A., Turpin, B. J., Vizuete, W., Martin, S. T., Thornton, J. A., Dutcher, C. S., Ault, A. P., and Surratt, J. D.: Increasing Isoprene Epoxydiol-to-Inorganic Sulfate Aerosol Ratio Results in Extensive Conversion of Inorganic Sulfate to Organosulfur Forms: Implications for Aerosol Physicochemical Properties, Environ. Sci. Technol., 53, 8682-8694, 10.1021/acs.est.9b01019, 2019.

Schindelka, J., Iinuma, Y., Hoffmann, D., and Herrmann, H.: Sulfate radical-initiated formation of isoprene-derived organosulfates in atmospheric aerosols, Faraday Discuss., 165, 237-259, 10.1039/c3fd00042g, 2013.

Shalamzari, M. S., Ryabtsova, O., Kahnt, A., Vermeylen, R., Herent, M. F., Quetin-Leclercq, J., Van der Veken, P., Maenhaut, W., and Claeys, M: Mass spectrometric characterization of organosulfates related to secondary organic aerosol from isoprene, Rapid Commun. Mass Spectrom., 784-794, 10.1002/rcm.6511, 2013, 2013.

Surratt, J. D., Lewandowski, M., Offenberg, J. H., Jaoui, M., Kleindienst, T.E., Edney, E. O., and Seinfeld, J. H.: Effect of Acidity on Secondary Organic Aerosol Formation from Isoprene, Environ. Sci. Technol., 41, 5363–5369, 2007.

Surratt, J. D., Chan, A. W., Eddingsaas, N. C., Chan, M., Loza, C. L., Kwan, A. J., Hersey, S. P., Flagan, R. C., Wennberg, P. O., and Seinfeld, J. H.: Reactive intermediates revealed in secondary organic aerosol formation from isoprene, Proc. Natl. Acad. Sci. U.S.A., 107, 6640-6645, 10.1073/pnas.0911114107, 2010.

Surratt, J. D., Gómez-González, Y., Chan, A. W. H., Vermeylen, R., Shahgholi, M., Kleindienst, T. E., Edney, E. O., Offenberg, J. H., Lewandowski, M., Jaoui, M., Maenhaut, W., Claeys, M., Flagan, R. C., and Seinfeld, J. H.: Organosulfate Formation in Biogenic Secondary Organic Aerosol, J. Phys. Chem. A., 112, 8345-8378, 10.1021/jp802310p, 2008.

Tao, S., Lu, X., Levac, N., Bateman, A. P., Nguyen, T. B., Bones, D. L., Nizkorodov, S. A., Laskin, J., Laskin, A., and Yang, X.: Molecular characterization of organosulfates in organic aerosols from Shanghai and Los Angeles urban areas by nanospray-desorption electrospray ionization high-resolution mass spectrometry, Environ. Sci. Technol., 48, 10993-11001, 10.1021/es5024674, 2014.

Trostl, J., Chuang, W. K., Gordon, H., Heinritzi, M., Yan, C., Molteni, U., Ahlm, L., Frege, C., Bianchi, F., Wagner, R., Simon, M., Lehtipalo, K., Williamson, C., Craven, J. S., Duplissy, J., Adamov, A., Almeida, J., Bernhammer, A. K., Breitenlechner, M., Brilke, S., Dias, A., Ehrhart, S., Flagan, R. C., Franchin, A., Fuchs, C., Guida, R., Gysel, M., Hansel, A., Hoyle, C. R., Jokinen, T., Junninen, H., Kangasluoma, J., Keskinen, H., Kim, J., Krapf, M., Kurten, A., Laaksonen, A., Lawler, M., Leiminger, M., Mathot, S., Mohler, O., Nieminen, T., Onnela, A., Petaja, T., Piel, F. M., Miettinen, P., Rissanen, M. P., Rondo, L., Sarnela, N., Schobesberger, S., Sengupta, K., Sipila, M., Smith, J. N., Steiner, G., Tome, A., Virtanen, A., Wagner, A. C., Weingartner, E., Wimmer, D., Winkler, P. M., Ye, P., Carslaw, K. S., Curtius, J., Dommen, J., Kirkby, J., Kulmala, M., Riipinen, I., Worsnop, D. R., Donahue, N. M., and Baltensperger, U.: The role of low-volatility organic compounds in initial particle growth in the atmosphere, Nature, 533, 527-531, 10.1038/nature18271, 2016.

Wang, Y., Ren, J., Huang, X. H. H., Tong, R., and Yu, J. Z.: Synthesis of Four Monoterpene-Derived Organosulfates and Their Quantification in Atmospheric Aerosol Samples, Environ. Sci. Technol., 51, 6791-6801, 10.1021/acs.est.7b01179, 2017.

Wang, Y., Ma, Y., Kuang, B., Lin, P., Liang, Y., Huang, C., and Yu, J. Z.: Abundance of organosulfates derived from biogenic volatile organic compounds: Seasonal and spatial contrasts at four sites in China, Sci. Total. Environ., 806, 151275, 10.1016/j.scitotenv.2021.151275, 2022.

Wang, Y., Ma, Y., Li, X., Kuang, B. Y., Huang, C., Tong, R., and Yu, J. Z.: Monoterpene and Sesquiterpene alpha-Hydroxy Organosulfates: Synthesis, MS/MS Characteristics, and Ambient Presence, Environ. Sci. Technol., 53, 12278-12290, 10.1021/acs.est.9b04703, 2019.

Wang, Y., Zhao, Y., Wang, Y., Yu, J. Z., Shao, J., Liu, P., Zhu, W., Cheng, Z., Li, Z., Yan, N., and Xiao, H.: Organosulfates in atmospheric aerosols in Shanghai, China: seasonal and interannual variability, origin, and formation mechanisms, Atmos. Chem. Phys., 21, 2959-2980, 10.5194/acp-21-2959-2021, 2021a.

Wang, Y., Hu, M., Wang, Y.-C., Li, X., Fang, X., Tang, R., Lu, S., Wu, Y., Guo, S., Wu, Z., Hallquist, M., and Yu, J. Z.: Comparative Study of Particulate Organosulfates in Contrasting Atmospheric Environments: Field Evidence for the Significant Influence of Anthropogenic Sulfate and NOx, Environ. Sci. Technol.Lett., 7, 787-794, 10.1021/acs.estlett.0c00550, 2020.

Wang, Y., Hu, M., Guo, S., Wang, Y., Zheng, J., Yang, Y., Zhu, W., Tang, R., Li, X., Liu, Y., Le Breton, M., Du, Z., Shang, D., Wu, Y., Wu, Z., Song, Y., Lou, S., Hallquist, M., and Yu, J.: The secondary formation of organosulfates under interactions between biogenic emissions and anthropogenic pollutants in summer in Beijing, Atmos. Chem. Phys., 18, 10693-10713, 10.5194/acp-18-10693-2018, 2018.

Wang, Z., Ehn, M., Rissanen, M. P., Garmash, O., Quéléver, L., Xing, L., Monge-Palacios, M., Rantala, P., Donahue, N. M., Berndt, T., and Sarathy, S. M.: Efficient alkane oxidation under combustion engine and atmospheric conditions, Commun. Chem., 4, 10.1038/s42004-020-00445-3, 2021b.

Xie, Q., Su, S., Chen, J., Dai, Y., Yue, S., Su, H., Tong, H., Zhao, W., Ren, L., Xu, Y., Cao, D., Li, Y., Sun, Y., Wang, Z., Liu, C.-Q., Kawamura, K., Jiang, G., Cheng, Y., and Fu, P.: Increase of nitrooxy organosulfates in firework-related urban aerosols during Chinese New Year's Eve, Atmos. Chem. Phys., 21, 11453-11465, 10.5194/acp-21-11453-2021, 2021.

Xie, Q., Li, Y., Yue, S., Su, S., Cao, D., Xu, Y., Chen, J., Tong, H., Su, H., Cheng, Y., Zhao, W., Hu, W., Wang, Z., Yang, T., Pan, X., Sun, Y., Wang, Z., Liu, C. Q., Kawamura, K., Jiang, G., Shiraiwa, M., and Fu, P.: Increase of High Molecular Weight Organosulfate With Intensifying Urban Air Pollution in the Megacity Beijing, J. Geophys. Res.: Atmos. , 125, 10.1029/2019jd032200, 2020.

Xu, Z. N., Nie, W., Liu, Y. L., Sun, P., Huang, D. D., Yan, C., Krechmer, J., Ye, P. L., Xu, Z., Qi, X. M., Zhu, C. J., Li, Y. Y., Wang, T. Y., Wang, L., Huang, X., Tang, R. Z., Guo, S., Xiu, G. L., Fu, Q. Y., Worsnop, D., Chi, X. G., and Ding, A. J.: Multifunctional Products of Isoprene Oxidation in Polluted Atmosphere and Their Contribution to SOA, Geophys. Res. Lett., 48, 10.1029/2020gl089276, 2021.

Yan, C., Nie, W., Äijälä, M., Rissanen, M. P., Canagaratna, M. R., Massoli, P., Junninen, H., Jokinen, T., Sarnela, N., Häme, S. A. K., Schobesberger, S., Canonaco, F., Yao, L., Prévôt, A. S. H., Petäjä, T., Kulmala, M., Sipilä, M., Worsnop, D. R., and Ehn, M.: Source characterization of highly oxidized multifunctional compounds in a boreal forest environment using positive matrix factorization, Atmos. Chem. Phys., 16, 12715-12731, 10.5194/acp-16-12715-2016, 2016.

Yassine, M. M., Dabek-Zlotorzynska, E., Harir, M., and Schmitt-Kopplin, P.: Identification of weak and strong organic acids in atmospheric aerosols by capillary electrophoresis/mass spectrometry and ultra-high-resolution Fourier transform ion cyclotron resonance mass spectrometry, Anal. Chem., 84, 6586-6594, 10.1021/ac300798g, 2012.

Yassine, M. M., Harir, M., Dabek-Zlotorzynska, E., and Schmitt-Kopplin, P.: Structural characterization of organic aerosol using Fourier transform ion cyclotron resonance mass spectrometry: aromaticity equivalent approach, Rapid. Commun. Mass. Spectrom., 28, 2445-2454, 10.1002/rcm.7038, 2014.

Ye, Y., Zhan, H., Yu, X., Li, J., Wang, X., and Xie, Z.: Detection of organosulfates and nitrooxy-organosulfates in Arctic and Antarctic atmospheric aerosols, using ultra-high resolution FT-ICR mass spectrometry, Sci. Total. Environ., 767, 144339, 10.1016/j.scitotenv.2020.144339, 2021.

Yttri, K. E., Simpson, D., Nøjgaard, J. K., Kristensen, K., Genberg, J., Stenström, K., Swietlicki, E., Hillamo, R., Aurela, M., Bauer, H., Offenberg, J. H., Jaoui, M., Dye, C., Eckhardt, S., Burkhart, J. F., Stohl, A., and Glasius, M.: Source apportionment of the summer time carbonaceous aerosol at Nordic rural background sites, Atmospheric Chemistry and Physics, 11, 13339-13357, 10.5194/acp-11-13339-2011, 2011.