



Supplement of

Significant role of biomass burning in heavy haze formation in Nanjing, a megacity in China: molecular-level insights from intensive PM_{2.5} sampling on winter hazy days

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S1: The isomeric ratios of anhydrosugars

The isomeric ratios of anhydrosugars are good indicators of biofuels. For instance, the ratio of levoglucosan to mannosan (L/M) is generally higher for hardwood and crop residues compared to softwood (Engling et al., 2006; Sang et al., 2013; Schmidl et al., 2008; Zhu et al., 2015). Additionally, smoke particles from the combustion of crop straws, grasses, and biomass briquettes tend to contain more galactosan than mannosan (Fu et al., 2008; Oros et al., 2006; Sheesley et al., 2003). As shown in Fig. S8, the L/M and M/G ratios during the sampling period suggest mixed contributions from wood and crop burning to haze formation. Previous reports indicate that the impact of softwood combustion increases during winter (Cheng et al., 2013).

S2: The ratios of biogenic SOA tracers

2-MGA/2-MTs ratios can be used to assess the influence of anthropogenic pollutants on SOA formation. In this study, 2-MGA/2-MTs ratios showed little variation over time, except for a few peaks observed during the highest-PM_{2.5} episode (Fig. S13). This suggests that isoprene-SOA formation was significantly influenced by anthropogenic emissions during heavy haze events.

S3: Chloride in PM_{2.5} samples

Chlorine plays a crucial role in global tropospheric chemistry, influencing atmospheric oxidation capacity and, consequently, the formation of PM_{2.5} and O₃, particularly in winter (Wang et al., 2019, 2020; Yi et al., 2023). In general, the absolute abundance of particulate Cl⁻ decreased with falling PM_{2.5} levels, while its relative contributions to total PM_{2.5} increased (from 3.2% to 8.7%) (Fig. 1). Earlier research indicated that low temperatures and high RH facilitate the generation of PM_{2.5}-bound Cl⁻ (Wang et al., 2023), which may explain the relatively higher Cl⁻ concentrations during the first episode. Marine sources dominate the global tropospheric chlorine budget, primarily through the mobilization of chloride from sea-salt aerosols (Wang et al., 2019). This is supported by a correlation between Cl⁻ and Na⁺ ($r = 0.64$, $p < 0.01$) in this study. The rising Cl⁻ proportion with decreasing PM_{2.5} level may be linked to the growing influence of long-range transported sea-salt aerosols, as illustrated by the backward

trajectories of air masses (Fig. S1). In China, however, chlorine is mostly anthropogenic, such as coal combustion, BB, waste incineration, and industrial processes (Fu et al., 2018; Li et al., 2012; Yang et al., 2018). The positive relationship between Cl^- and levoglucosan ($r = 0.50, p < 0.05$) suggests that BB contributes to Cl^- levels at this site. Moreover, the higher mass ratios of Cl^-/Na^+ (about 5.5-15.5) in these samples than sea water (1.81) indicate a significant anthropogenic contribution to Cl^- during winter haze in Nanjing. The measured Cl^- concentrations in this study (average: $6.38 \pm 2.00 \mu\text{g m}^{-3}$) are comparable to those observed in winter in Beijing (Wang et al., 2005) but slightly higher than those in a coastal megacity of Shanghai (Wang et al., 2006; Ye et al., 2003).

S4: Monocarboxylic acids

Two monocarboxylic acids, formic and acetic acid, were also determined. Both acids showed a decreasing trend with decreasing $\text{PM}_{2.5}$ values, with peak concentrations occurring during the heaviest haze events (average: $0.18 \pm 0.05 \mu\text{g m}^{-3}$ for formic acid and $0.22 \pm 0.11 \mu\text{g m}^{-3}$ for acetic acid). The lower temperatures and higher RH during the first episode favor gas-to-particle conversion, resulting in elevated particulate concentrations. A strong relationship between formic and acetic acid was observed ($r = 0.88, p < 0.01$), suggesting similar sources. Additionally, both acids significantly correlated with levoglucosan ($r = 0.73, p < 0.01$ for formic acid and $r = 0.61, p < 0.01$ for acetic acid), indicating that BB is a significant source. Formic acid was also associated with EC ($r = 0.67, p < 0.01$), demonstrating that fossil fuel combustion may contribute as well. Moreover, secondary sources play an important role in the atmospheric budgets of formic and acetic acids (Paulot et al., 2011). The ratio of acetic to formic acid (A/F) can help estimate the relative importance of primary emissions (> 1) versus secondary photochemical transformations (< 1) for carboxylic acids (Wang et al., 2007). The average A/F ratios for three episodes were 1.15 ± 0.32 , 1.30 ± 0.52 , and 1.1 ± 0.30 , respectively, suggesting a relatively larger contribution from primary sources, such as BB, vegetation, coal burning, vehicular exhausts, and soil emissions during the heating season (Khare et al., 1999; Stavrakou et al., 2012; Wang et al., 2007).

S5: Dicarboxylic acids

Dicarboxylic acids predominantly exist as air particles rather than in the gas phase due to their low vapor pressures (Limbeck et al., 2001; Saxena and Hildemann, 1996). These acids, which contain two carboxyl groups, are major constituents of water-soluble organics in aerosols (Saxena and Hildemann, 1996), as evidenced by significant correlations between WSOC and dicarboxylic acids in this study ($r = 0.74\text{--}0.87$, $p < 0.01$). In addition to being directly emitted from incomplete combustion of fossil fuels, meat cooking, and BB, they can also form through secondary photochemical reactions (Mochida et al., 2003). For instance, isoprene and unsaturated fatty acids are proposed sources of dicarboxylic acids in the open ocean (Bikkina et al., 2014). In total, four saturated dicarboxylic acids (oxalic, malonic, succinic, and glutaric acid) and two unsaturated diacids (maleic and fumaric acid) were analyzed. Unsaturated dicarboxylic acids ($2.48\text{--}69.5 \text{ ng m}^{-3}$) were found in much lower concentrations than saturated diacids ($1.66\text{--}14.6 \text{ } \mu\text{g m}^{-3}$). Similar to biogenic SOA, dicarboxylic acids showed higher concentrations during episodes of elevated temperature and low RH, conditions that favor the photochemical oxidation of organic precursors.

Among the measured dicarboxylic acids, malonic acid (C3, $1.48\text{--}14.3 \text{ } \mu\text{g m}^{-3}$) was the most abundant, followed by oxalic acid (C2, $0.09\text{--}0.74 \text{ } \mu\text{g m}^{-3}$). C2 and C4 levels are comparable to those reported in $\text{PM}_{2.5}$ aerosols from other megacities such as Beijing (Ho et al., 2010) and Guangzhou (Liu et al., 2021). It was deduced that C2 and C3 diacids are formed through the oxidation of C4 and other longer-chain diacids, while longer-chain diacids (C5–C10) originate from the oxidation of unsaturated fatty acids (Kawamura and Gagosian, 1987; Kawamura and Sakaguchi, 1999). This is supported by significant correlations between C2 and C4 ($r = 0.86$, $p < 0.01$), C2 and C5 ($r = 0.77$, $p < 0.01$) and C4 and C5 ($r = 0.60$, $p < 0.01$) in the present study. The relatively higher levels of C2 and C3 compared to other diacids may result from considerable photodegradation of C4 and C5 during haze events, implying these urban aerosols have undergone extensive aging processes. The ratio of C3 to C4 is a useful indicator of elevated photochemical production of dicarboxylic acids, as C4 acts as a precursor for C3 formation (Kawamura and Ikushima, 1993). In this study, the abundance of C3 greatly exceeded that of C4, indicating strong photochemical processes, which is also supported by high WSOC/OC ratios. These findings highlight that secondary formation is a crucial pathway for

dicarboxylic acids on hazy days in urban Nanjing, in addition to primary emissions. Notably, both C2 and C5 were found to correlate well with levoglucosan ($r = 0.66\text{--}0.69$, $p < 0.01$), indicating BB is an alternative source of these diacids and may facilitate their oxidation reactions (Kawamura and Bikkina, 2016). Chlorine emissions from BB have been shown to increase oxidant levels, such as O₃ and OH radicals, significantly impacting atmospheric chemistry and oxidation processes (Chang et al., 2024).

Maleic acid (cis-isomer) and fumaric acid (trans-isomer) are two key aliphatic unsaturated diacids identified in this study, with concentration ranges of 0.86-41.9 ng m⁻³ and 1.61-27.6 ng m⁻³, respectively. A significant correlation between fumaric and maleic acid ($r = 0.81$, $p < 0.01$) suggests they may share common sources. Maleic acid, a likely product of the photochemical oxidation of benzene, can undergo photochemical isomerization in the presence of sunlight to form its trans isomer, fumaric acid. Consequently, the M/F ratios depend on the production of maleic acid and its subsequent transformation into fumaric acid. The M/F ratios were higher during the second episode with higher temperatures and low RH (average: 1.38 ± 0.35), but lower in the other two episodes (average: 0.71 ± 0.28 and 0.85 ± 0.44 , respectively). The higher M/F values may indicate superior photochemical generation of maleic acid over the cis-to-trans isomerization under high ambient temperatures and low RH, whereas lower ratios could be partly attributed to reduced maleic acid production at lower temperatures (Kawamura and Ikushima, 1993).

S6: Other identified chemicals

Methylglyoxal (MeGly) is an oxidation product of biogenic (e.g., isoprene/monoterpenes) and anthropogenic precursors (e.g., benzene, toluene, and xylenes) (Kampf et al., 2012). Typically, most carbonyls are present in the gas phase in the atmosphere. The high levels of MeGly observed during the highest PM_{2.5} episode (20.4 ± 29.2 ng m⁻³) may be due to lower temperatures, which facilitate the partitioning of gaseous carbonyls into the aerosol phase (Meng et al., 2018). In addition, fresh plumes from high BB activities during this episode may also contribute to the elevated levels (Kampf et al., 2012). In the other two episodes, MeGly abundances (10.1 ± 4.93 ng m⁻³ and 6.43 ± 3.04 ng m⁻³, respectively) were lower but comparable to those from Beijing PM_{2.5} samples (8.3 ± 7.9 ng m⁻³) (Zhao et al., 2018). A significant

correlation between MeGly and ^{14}C -WSOC was obtained in this study ($r = 0.74$, $p < 0.01$), suggesting that non-fossil sources, such as BB, may contribute to atmospheric MeGly levels.

Methanesulfonic acid (MSA) is an oxidation product of dimethylsulfide (DMS), which is primarily released by marine phytoplankton. Thus, MSA has been regarded as a valuable tracer for marine biogenic sulfur production (Chen et al., 2012; Legrand and Pasteur, 1998). Surprisingly, this study found higher levels of MSA during the most polluted episode ($0.09 \pm 0.02 \mu\text{g m}^{-3}$) compared to the other two episodes ($0.04 \pm 0.01 \mu\text{g m}^{-3}$ and $0.02 \pm 0.01 \mu\text{g m}^{-3}$, respectively), likely because anthropogenic sources, such as industrial emissions, could also contribute to MSA levels in urban atmospheres (Yuan et al., 2004). These MSA levels are similar to those reported in $\text{PM}_{2.5}$ aerosols from Beijing (Wang et al., 2005). The ratio of MSA to non-sea-salt sulfate (nss-SO_4^{2-}) can be used to assess the relative contributions of marine biogenic emissions compared to other sulfur sources (Legrand and Pasteur, 1998). The average MSA/ nss-SO_4^{2-} ratios for the three episodes were 0.003 ± 0.001 , 0.002 ± 0.00 , 0.002 ± 0.001 , respectively. These low values indicate that the influence of marine biogenic sources from coastal and oceanic areas on urban haze formation is relatively weak. Moreover, significant relationships were found between MSA and levoglucosan ($r = 0.75$, $p < 0.01$), suggesting that BB could be an important source of MSA, alongside industrial emissions. This is further supported by a significant relationship between non-fossil WSOC and MSA ($r = 0.79$, $p < 0.01$).

Table S1. Concentrations of other measured components in PM_{2.5} samples from urban Nanjing during polluted episodes.

Species	PM _{2.5} ($\mu\text{g m}^{-3}$)						PM _{2.5} ($\mu\text{g m}^{-3}$)					
	>200			100-200			<100			<100		
	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max
Saturated dicarboxylic acids ($\mu\text{g m}^{-3}$)												
oxalic, C2	0.46	0.16	0.23	0.74	0.34	0.11	0.23	0.51	0.18	0.06	0.09	0.30
malonic, C3	6.43	2.10	1.51	8.71	10.0	2.41	8.50	14.3	5.96	2.41	1.48	8.14
succinic, C4	0.04	0.02	0.01	0.07	0.03	0.02	0.01	0.06	0.01	0.01	nd	0.02
glutaric, C5	0.06	0.02	0.03	0.08	0.04	0.02	0.02	0.06	0.02	0.01	0.01	0.03
sum of saturated diacids	6.99	2.10	1.96	9.19	10.4	2.34	9.12	14.6	6.16	2.41	1.66	8.32
Unsaturated aliphatic diacids (ng m^{-3})												
maleic acid	8.32	5.35	0.86	20.2	21.3	9.11	11.5	33.1	10.79	13.1	1.00	41.9
fumaric acid	11.7	6.84	1.61	27.6	15.5	5.34	8.32	23.2	11.51	8.01	1.70	27.6
M/F	0.71	0.28	0.27	1.26	1.38	0.35	0.80	1.72	0.85	0.44	0.32	1.52
sum of unsaturated aliphatic diacids	20.0	11.8	2.48	47.8	36.8	13.9	21.0	56.3	22.3	20.5	2.70	69.5
α-Dicarbonyls (ng m^{-3})												
MeGly, methylglyoxal	20.4	29.2	7.47	103	10.1	4.93	6.55	18.7	6.43	3.04	2.12	10.4
Other species ($\mu\text{g m}^{-3}$)												
MSA, methanesulfonic acid	0.09	0.02	0.06	0.12	0.04	0.01	0.02	0.05	0.02	0.01	0.00	0.03
formic acid	0.18	0.05	0.08	0.25	0.12	0.01	0.11	0.14	0.05	0.02	0.02	0.08
acetic acid	0.22	0.11	0.07	0.44	0.16	0.07	0.08	0.27	0.05	0.01	0.03	0.06

nd: not detected.

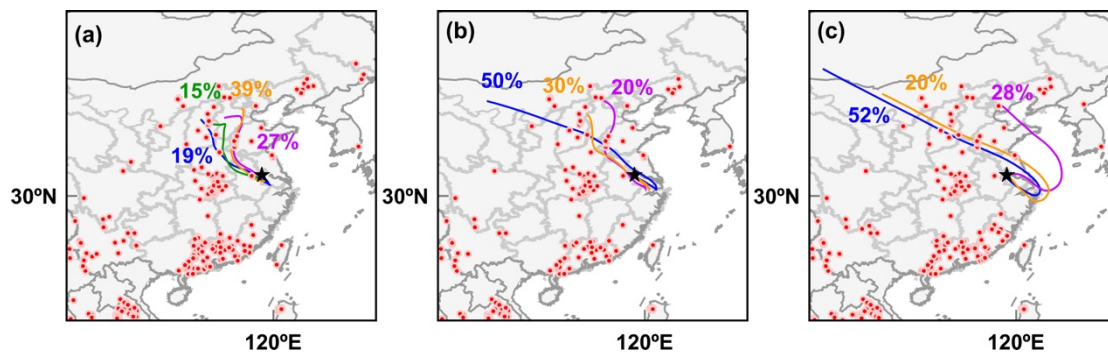


Figure S1. Three-day HYSPLIT backward trajectories initiated over Nanjing at altitudes below 500 m, combined with MODIS fire spots during three episodes with $PM_{2.5}$ concentrations of (a) > 200 , (b) $100-200$, and (c) $< 100 \mu g m^{-3}$. The black star represents the sampling site. MODIS active fire data can be accessed here: <https://firms.modaps.eosdis.nasa.gov/download/list.php>.

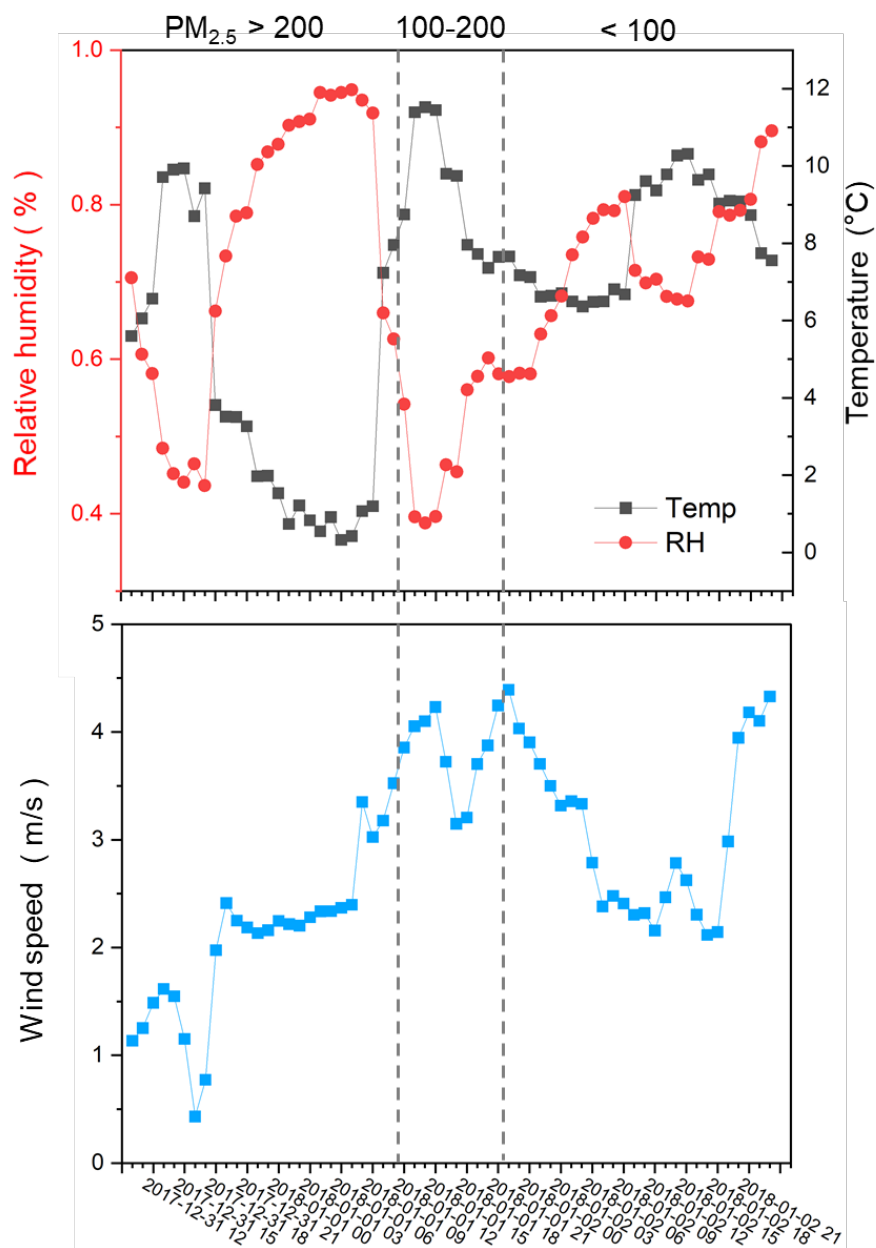


Figure S2. Time series of meteorological parameters (i.e., relative humidity, temperature, and wind speed) during the sampling period in urban Nanjing.

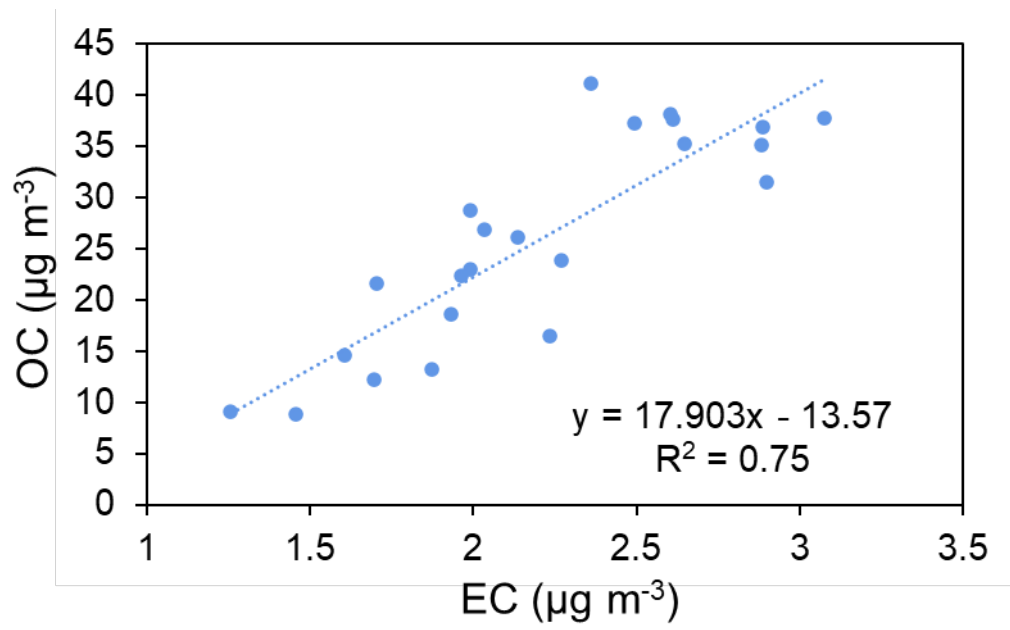


Figure S3. Relationship between OC and EC.

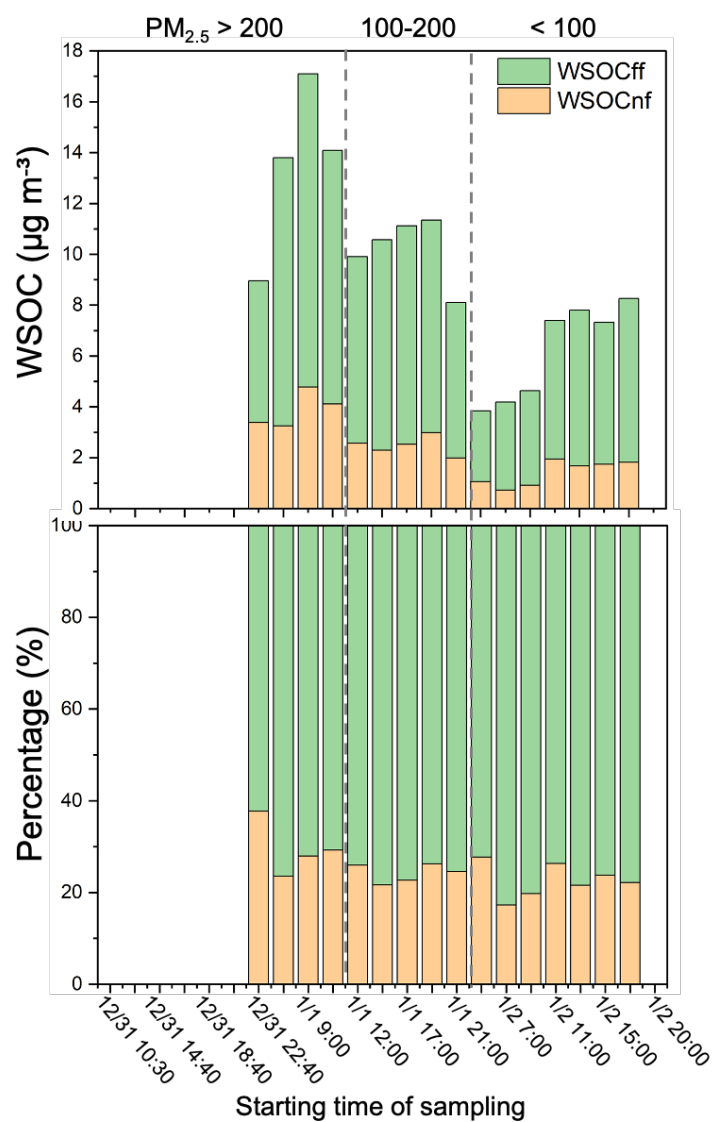


Figure S4. Temporal variations in fossil (ff) and non-fossil (nf) contributions to water-soluble organic carbon (WSOC) in PM_{2.5} samples from Nanjing.

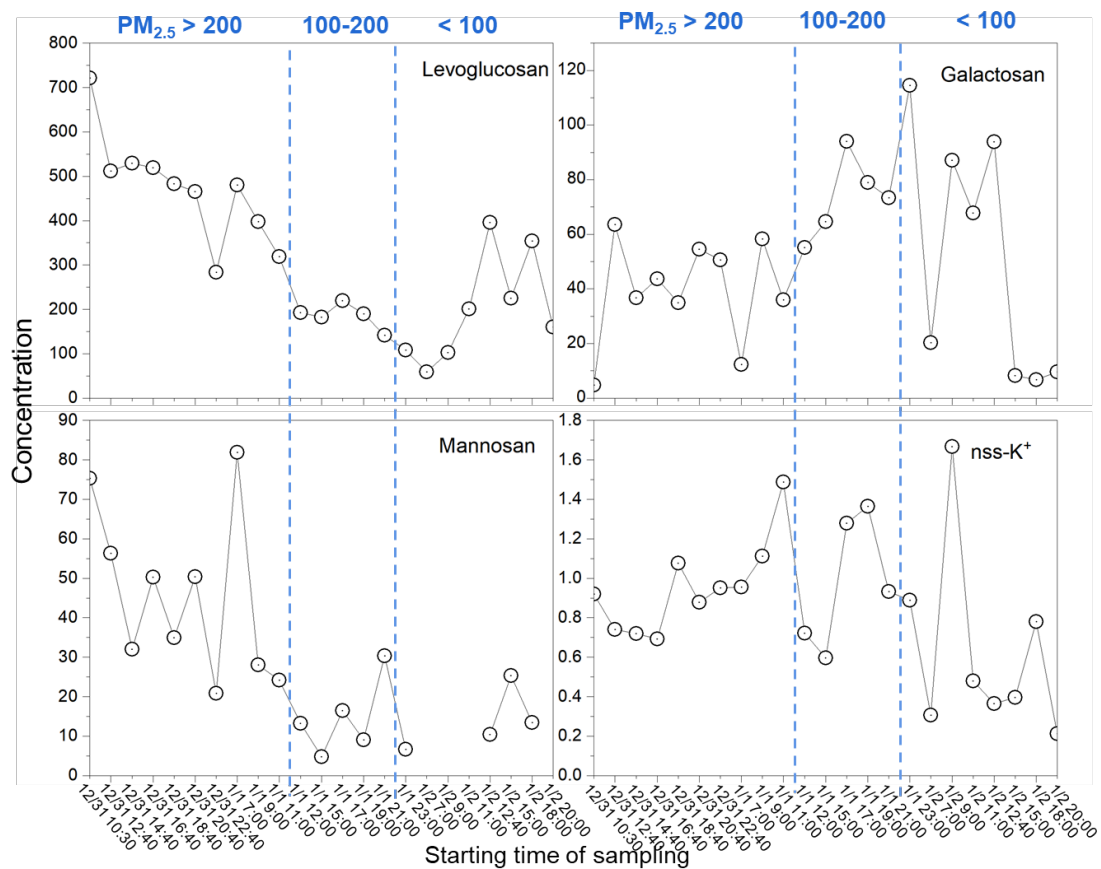


Figure S5. Temporal variations in the concentrations of biomass burning tracers, including anhydrosugars (levoglucosan, galactosan, and mannosan; ng m⁻³) and non-sea-salt K⁺ (μg m⁻³).

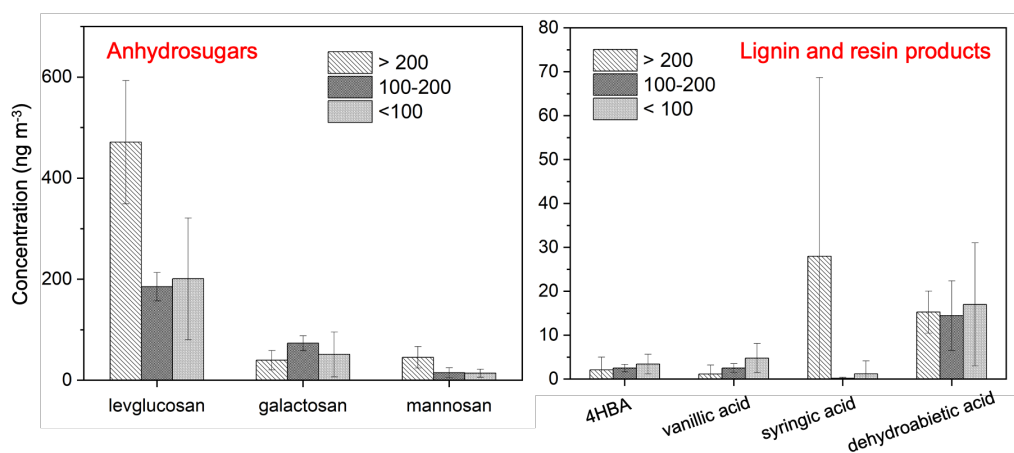


Figure S6. Average concentrations of biomass burning tracers measured during three episodes with $PM_{2.5}$ levels categorized as > 200 , $100-200$, and $< 100 \mu g m^{-3}$.

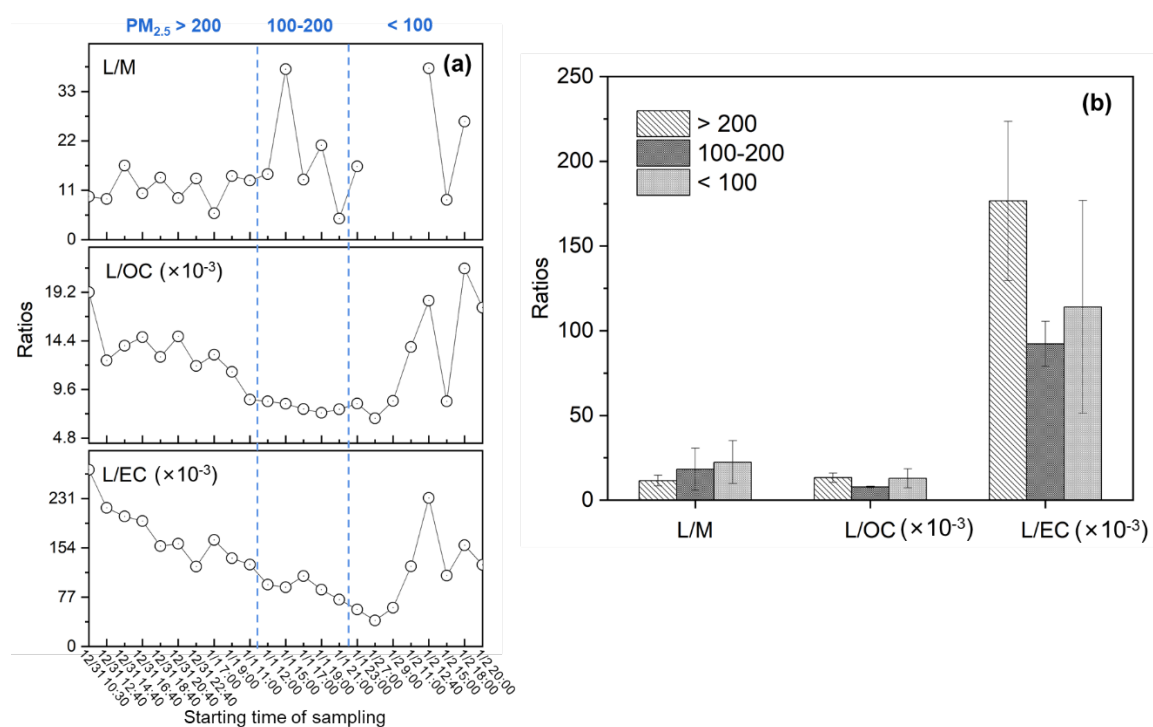


Figure S7. (a) Temporal variations in the ratios of levoglucosan to mannosan (L/M), levoglucosan to organic carbon (L/OC), and levoglucosan to elemental carbon (L/EC), and (b) average ratios during three pollution episodes (> 200 , $100-200$, and $< 100 \mu g m^{-3}$).

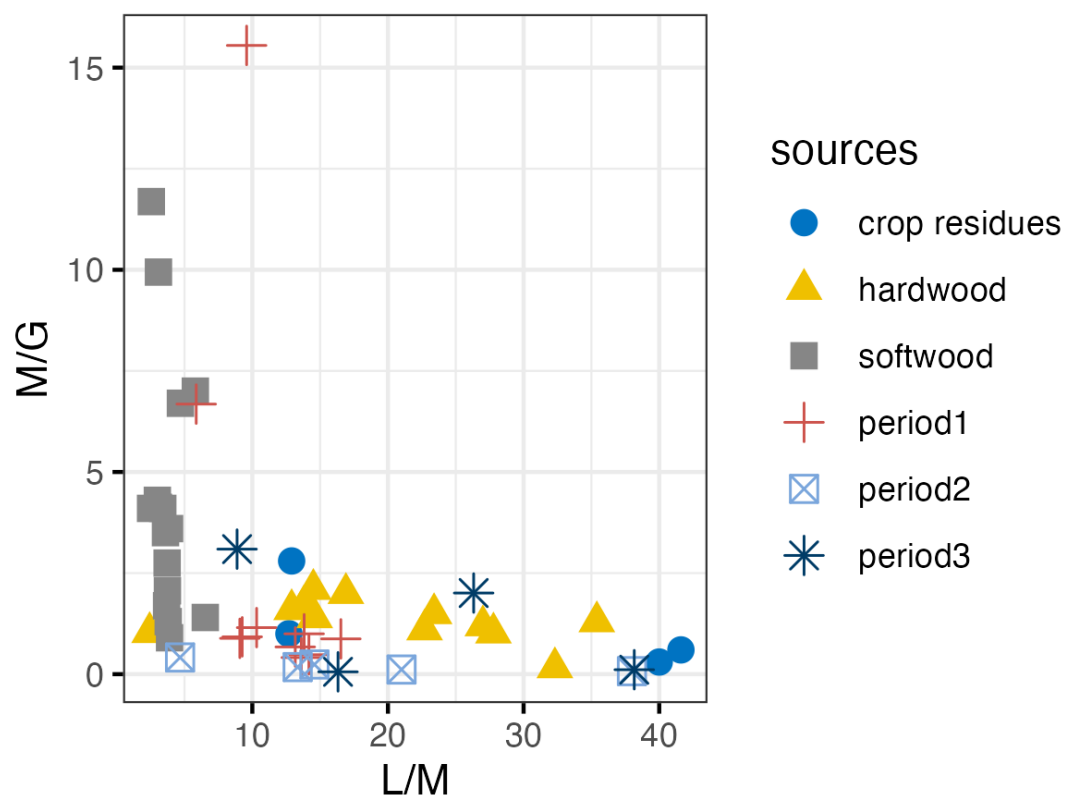


Figure S8. Comparison of L/M and M/G ratios from source emissions (literature values) and ambient aerosols in this study. L/M refers to the ratio of levoglucosan to mannosan, and M/G denotes the ratio of mannosan to galactosan.

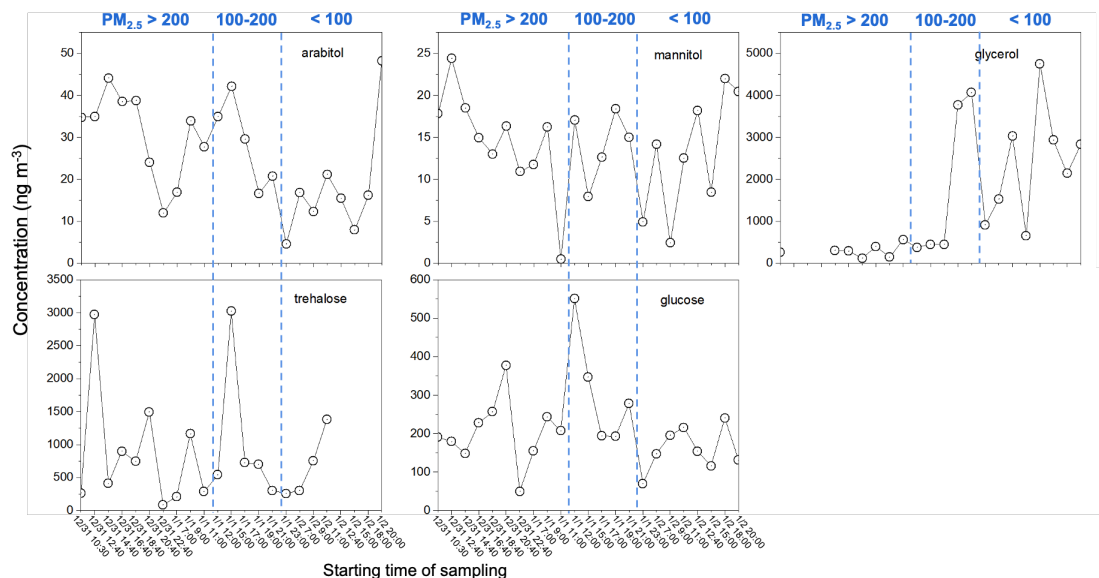


Figure S9. Temporal variations in concentrations of sugars and sugar alcohols (ng m⁻³).

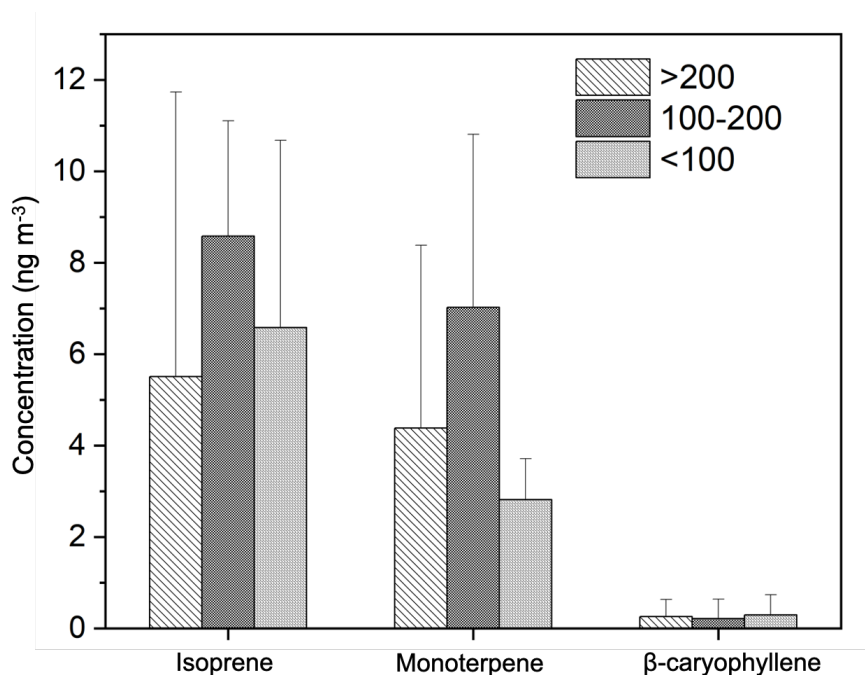


Figure S10. Average concentrations of isoprene, monoterpene, and β-caryophyllene SOA tracers detected during three episodes based on PM_{2.5} concentrations (> 200, 100-200, and < 100 μg m⁻³).

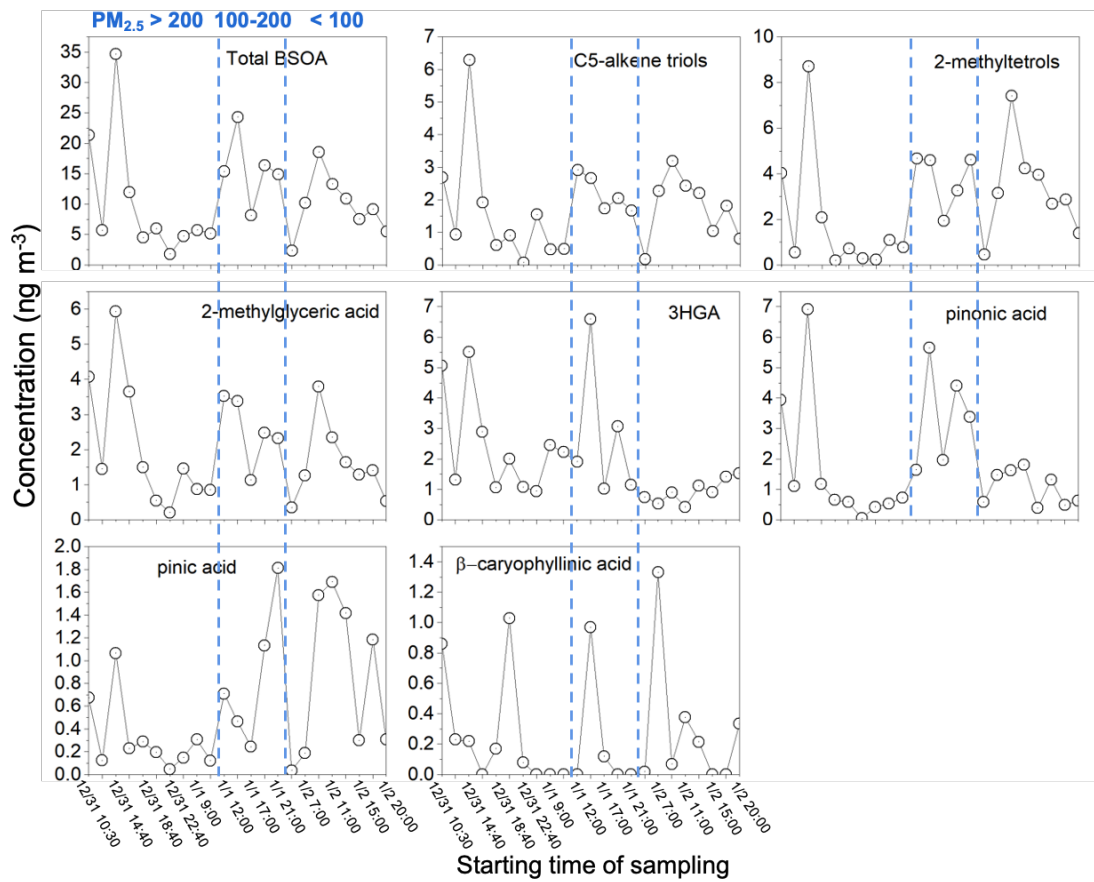


Figure S11. Temporal variations of biogenic SOA tracers in $PM_{2.5}$.

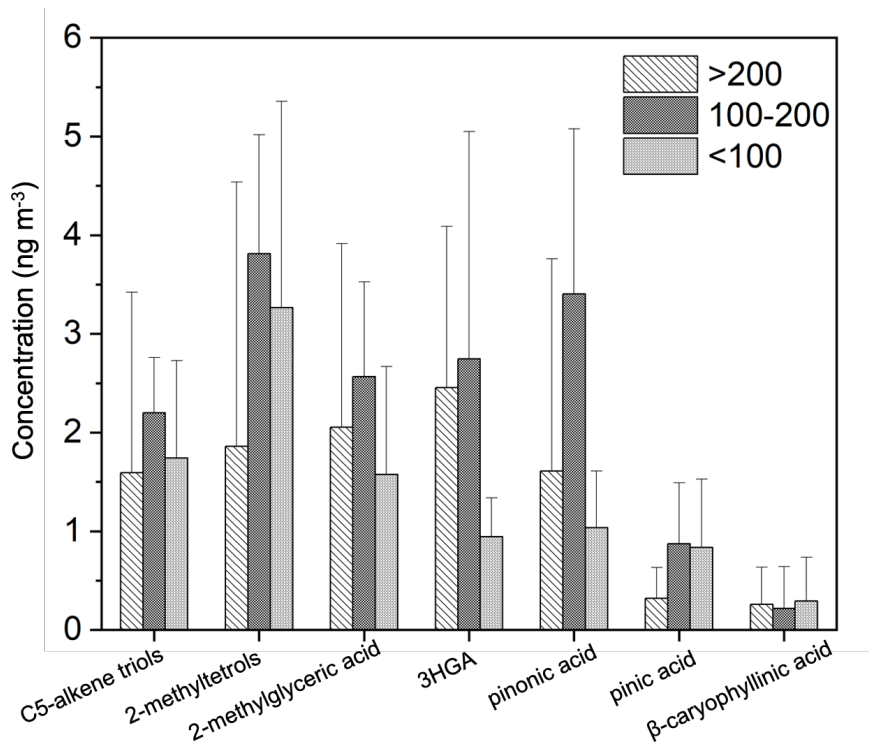


Figure S12. Average concentrations of biogenic SOA tracers detected during three episodes based on $\text{PM}_{2.5}$ concentrations (> 200 , $100-200$, and < 100 $\mu\text{g m}^{-3}$).

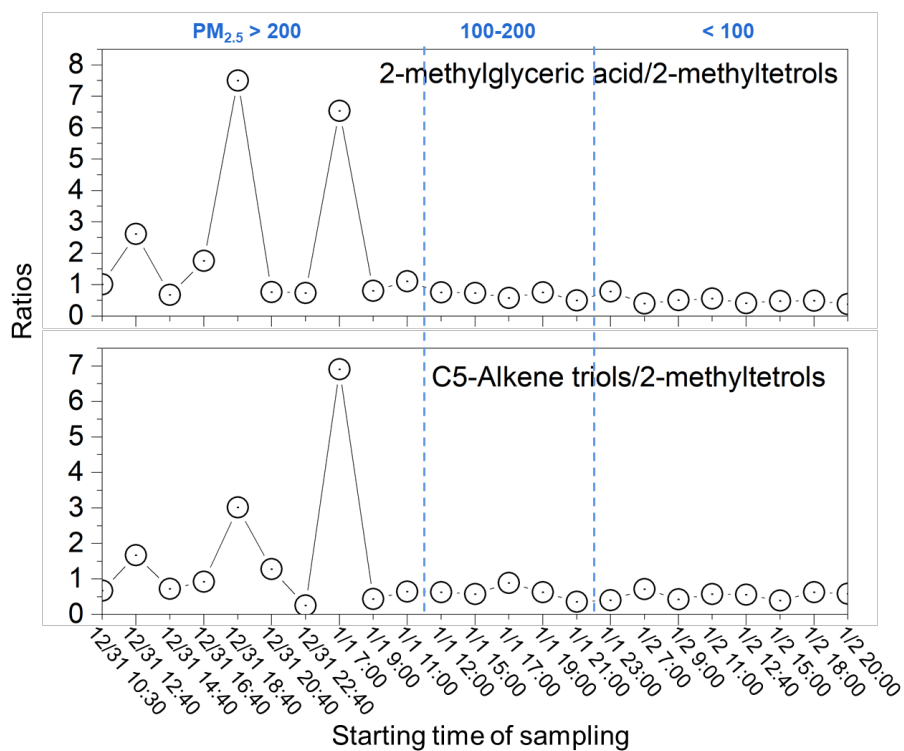


Figure S13. Temporal variations in the concentration ratios of isoprene oxidation products in $\text{PM}_{2.5}$.

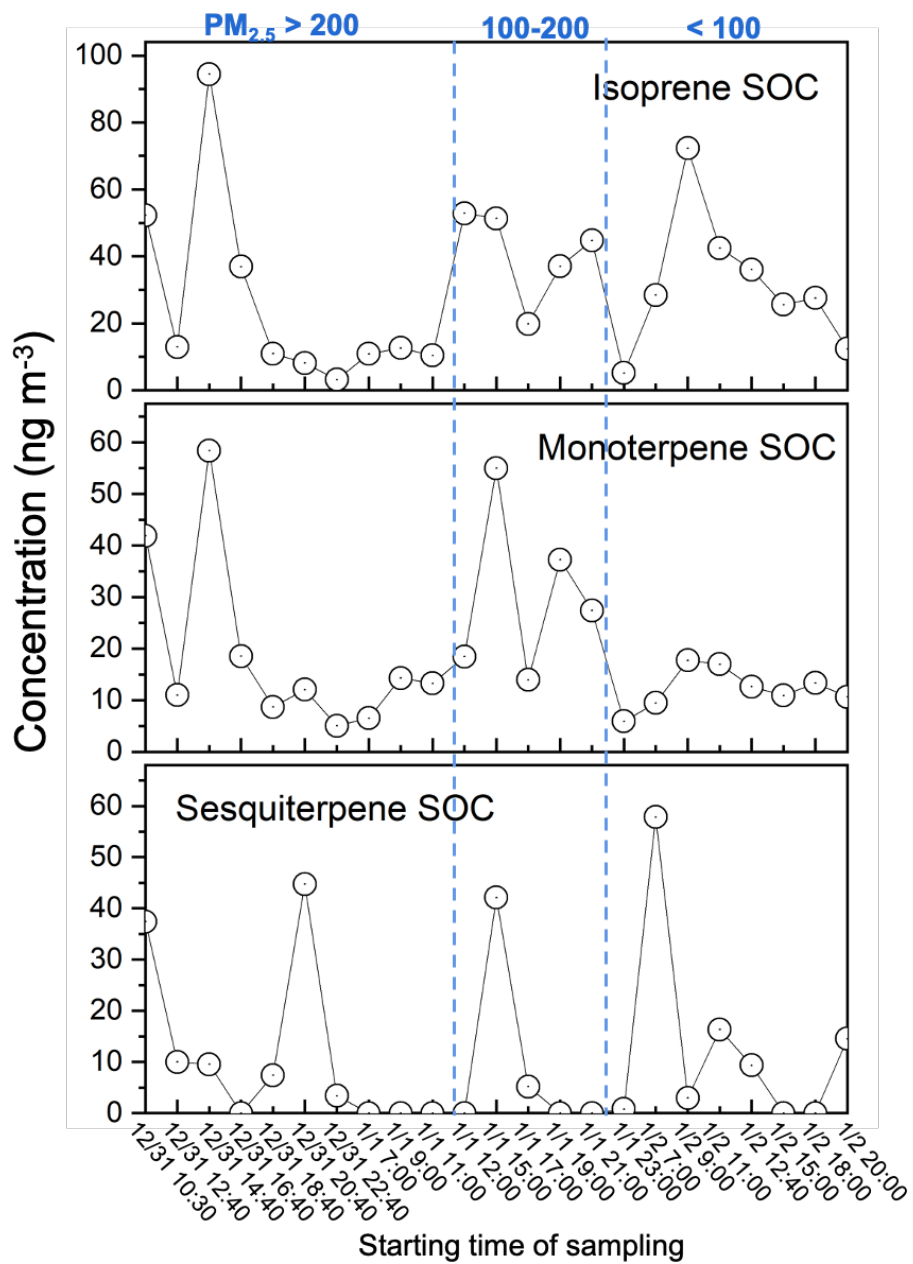


Figure S15. Temporal variations in biogenic secondary organic carbon (SOC) derived from isoprene, monoterpenes, and sesquiterpenes.

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