



# Spatiotemporal variations of PM<sub>2.5</sub> organic molecular markers in five central cities of the Yangtze River Delta, East China in autumn and winter: Implications for regional and local sources of organic aerosols<sup>☆</sup>

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## ABSTRACT

Information on the spatiotemporal variations in the composition and sources of organic aerosols (OA) is needed to identify regional influences and to establish effective control measures. Here, 23-h PM<sub>2.5</sub> samples were collected in five central cities of the Yangtze River Delta in eastern China, including Nanjing, Suzhou, Wuxi, Changzhou, and Zhenjiang, every three days from 2020/09/01 to 2021/02/28. Each sample was analyzed for water-soluble inorganic ions, organic carbon (OC), elemental carbon (EC), and organic molecular markers (OMMs). Generally, the major components of PM<sub>2.5</sub>, including NH<sub>4</sub><sup>+</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, OC, and EC, exhibited similar temporal patterns across the five cities. In all OMM groups, the concentrations of PAHs, oxygenated PAHs, and secondary products of isoprene showed strong correlations ( $r = 0.79 \pm 0.050$ – $0.93 \pm 0.028$ ) and low coefficient of divergence (COD =  $0.22 \pm 0.024$ – $0.30 \pm 0.033$ ) between sampling sites, indicating a homogeneous spatial distribution of industrial emissions and biogenic secondary OA in autumn and winter. Other OMMs showed wider  $r$  (e.g., steranes and hopanes, 0.20–0.80) and COD (0.26–0.69) ranges for all site pairs, probably due to the influence of local emissions. Based on the source apportionment results using Positive matrix factorization, the biomass burning factor dominated the contribution to OC and EC in winter and showed strong correlations ( $r = 0.84 \pm 0.063$ ) between the sampling sites, indicating regional transport of emissions from biomass burning and fossil fuel combustion in the heating season. Traffic-related factors had the greatest spatial heterogeneity ( $r = 0.27 \pm 0.19$ – $0.51 \pm 0.16$ ) and contributed significantly to OC at their maximum levels.

## 1. Introduction

Organic aerosols (OA) consist of a complex mixture of compounds and make up 20–90% of particulate matter of less than 2.5 μm diameter (PM<sub>2.5</sub>; Zhang et al., 2007; Hallquist et al., 2009; Jimenez et al., 2009). The effects of OA on health and climate have been extensively investigated in recent decades (Maria et al., 2004; Mauderly and Chow, 2008; Pye et al., 2021; Jo et al., 2023). Deciphering the physicochemical features and atmospheric processes of OA depends to a large extent on how well they are characterized (Nozière et al., 2015). Several groups of semi-volatile organic compounds (SVOCs; e.g., *n*-alkanes, PAHs, and

anhydro sugars) were well known as organic molecular markers (OMMs) in receptor-based source apportionment studies (Jaekels et al., 2007; Lewandowski et al., 2008; Shrivastava et al., 2007). The recognized sources of secondary OA (SOA) were attributed to the transformation of volatile organic compounds (VOCs) during regional transport, and contributions from primary sources were expected to be associated with local emissions (e.g., motor vehicle emissions). Based on the source apportionment results, regulatory strategies were developed to control regional or local pollution. For example, leak detection in the petrochemical industry and the use of low-VOC adhesives and paints have been promoted to reduce SOA and ozone formation (Wei et al., 2011;

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Zhang et al., 2022), as well as smart traffic management systems and low-dust construction to control emissions from local traffic-related and dust sources (Wu et al., 2016; Padrón et al., 2022). However, emissions from multiple primary sources, such as biomass burning, fossil fuel combustion, and dust, can mix and undergo long-range transport (Zhou et al., 2018; Mukherjee et al., 2022), which then increases local PM<sub>2.5</sub> pollution for the specific study location. Therefore, the results of source apportionment determined at single sites cannot distinguish between regional and local sources.

Given the spatial variability of PM<sub>2.5</sub> constituents and their sources, the representativeness of PM<sub>2.5</sub> characterization and source apportionment at a single site has always been questioned. In previous studies, multiple-site sampling and measurements of PM<sub>2.5</sub> were conducted mostly at the city level (Krudysz et al., 2009; Xie et al., 2012a; Chow et al., 2022). Kim et al. (2005) showed that the spatial distribution of PM<sub>2.5</sub> elements and the contributions of local source categories (e.g., incinerators and cement kilns) were highly heterogeneous in St. Louis, United States, which could lead to exposure misclassification in epidemiologic studies where health outcomes were regressed over the time series of PM<sub>2.5</sub> components and source contributions. However, the spatial distribution of OA and its sources based on measurement results at the regional level has rarely been assessed (Zheng et al., 2006; Golly et al., 2019; Zheng et al., 2024). According to the available studies, biomass burning and the formation of SOA are major OA sources at the regional scale, although significant differences in source contribution distributions between urban and rural sites have been found.

The southern part of Jiangsu province in eastern China consists of Nanjing, Suzhou, Wuxi, Changzhou, and Zhenjiang, which are the central cities of the Yangtze River Delta. According to the Jiangsu Provincial Bureau of Statistics of China (2023), these five cities have an area of 28084 km<sup>2</sup> and a permanent population of 38.5 million in 2022, accounting for 26.3% and 45.2% of the total area and population of Jiangsu province, respectively. The total gross domestic product (GDP) of southern Jiangsu amounted to 70.28 trillion yuan in 2022, comparable to that of Zhejiang province (77.7 trillion yuan) and more than 50% higher than that of Shanghai (44.7 trillion yuan) — two neighboring developed areas in the Yangtze River Delta. Industry accounted for 40.8% of GDP in southern Jiangsu, and the industrial electricity consumption (2.8 trillion kWh) and highway passenger traffic (257 million) in this area dominated (60.2% and 78.8%) the entire Jiangsu province in 2022. Considering the high urbanization rate (80.2%) in southern Jiangsu, the urban areas in the above five cities can represent the highly developed region in the Yangtze River Delta.

PM<sub>2.5</sub> pollution in the Yangtze River Delta is a major concern and is usually associated with biomass burning, industrial and vehicle emissions, construction activities, and unfavorable weather conditions (Yang et al., 2018; Zhang et al., 2020). As reported by the Chinese Ministry of Ecology and Environment (<https://www.mee.gov.cn/hjzl/dqjh/qgkqzlk/>), the annual average concentrations of PM<sub>2.5</sub> in the Yangtze River Delta in 2021–2023 (31–32 μm<sup>-3</sup>) remained below the Class II limit (35 μm<sup>-3</sup>) of China's National Ambient Air Quality Standards (NAAQS) after the implementation of the “Air Pollution Prevention and Control Action Plan” (2013–2017) and the “Three-Year Blue Sky Action Plan” (2018–2020), but the average PM<sub>2.5</sub> concentrations in the winter months (December, January, and February) were always above 35 μm<sup>-3</sup> (38–66 μm<sup>-3</sup>) after 2020. Before implementing joint prevention and control measures at the regional level involving cooperation between local governments, industries, communities, and environmental authorities, information on the spatial distribution of PM<sub>2.5</sub> composition is needed, especially for OA where a significant proportion comes from primary emissions, to determine regional sources and their contributions. However, there are very few measurement studies on the spatial distribution of OA composition and sources in the Yangtze River Delta. Since PM<sub>2.5</sub> pollution in eastern China occurs more frequently in autumn and winter than in spring and summer (Zhang and Cao, 2015; Sun et al., 2019; Guo et al., 2020a), this work collected filter samples of ambient

PM<sub>2.5</sub> in five central cities of the Yangtze River Delta from September 1, 2020 to February 28, 2021. Water-soluble inorganic ions (WSIIs), organic carbon (OC), elemental carbon (EC), and a matrix of OMMs were speciated and compared among the cities to assess their spatial distribution. Finally, the OMM data were pre-selected for a positive matrix factorization (PMF) analysis, and regional and local OA sources were distinguished by analyzing the spatial variability of the time series of source contributions. The study results will benefit regional cooperation in the continuous reduction of PM<sub>2.5</sub> pollution in the Yangtze River Delta.

## 2. Methods

### 2.1. PM<sub>2.5</sub> sampling

From January 09, 2020 to 02/28/2021, 23-h PM<sub>2.5</sub> samples (10:00–9:00 a.m. the next day) were collected simultaneously every third day in Nanjing, Suzhou, Wuxi, Changzhou, and Zhenjiang, and the sampling frequency is about twice that of many previous studies on spatiotemporal variations of OMMs and OA (Krudysz et al., 2009; Cheung et al., 2012; Xie et al., 2012a, b; Chow et al., 2022; Zheng et al., 2024). The locations and sample numbers of the five sampling sites are shown in Fig. 1. All five sites are located in urban areas, and the surrounding pollutant sources are mainly residential areas and traffic emissions. A four-channel sampler (TH-16a, Wuhan Tianhong, China) was set up at each site to collect PM<sub>2.5</sub> samples at a flow rate of 16.7 L min<sup>-1</sup>. Two Teflon and two quartz filters (47 mm diameter) were installed for PM<sub>2.5</sub> sampling. Samples on the Teflon filters were analyzed for WSIs; OC, EC, and OMMs were determined from the quartz filter samples. Field blanks were taken each month to correct for possible contamination. All samples were stored in the dark at −20 °C before chemical analysis. During the whole sampling period, the temperature (°C) and relative humidity (RH, %) were recorded for each sampling day (Fig. S1), and the concentrations of the six criteria air pollutants (PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, CO, NO<sub>2</sub>, and MDA8 O<sub>3</sub>; Fig. S1) defined in the Chinese NAAQS were retrieved from the online platform for monitoring and analyzing air quality in China (Wang, 2013) for each city.

### 2.2. Chemical characterization

The analytical methods for WSIs, OC, EC, and OMMs in PM<sub>2.5</sub> samples were described in previous studies (Gou et al., 2021; Yang et al., 2021; Qin et al., 2021). WSIs were extracted from Teflon filter samples using ultrapure water (18.2 MΩ) and analyzed by ion chromatography. The recoveries of our target WSIs (NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, K<sup>+</sup>, and Ca<sup>2+</sup>) were in the range of 95.6 ± 1.76%–105 ± 5.86% with detection limits of less than 0.1 μg m<sup>-3</sup> (0.016–0.088 μg m<sup>-3</sup>). A ~0.5 cm<sup>3</sup> aliquot of each quartz filter was analyzed for OC and EC using a thermo-optical carbon aerosol analyzer (DRI model 2015) according to the IMPROVE-A protocol. The DRI OC-EC analyzer was calibrated daily with the standard solution of sucrose (5–15 μgC μL<sup>-1</sup>), and the detection limits of OC and EC were 0.40 and 0.20 μg m<sup>-3</sup>, respectively. After adding deuterated PAH solutions as internal standards (IS), two aliquots of each quartz filter were extracted with dichloromethane (DCM) and a DCM/methanol mixture (1:1, v:v), respectively. After filtration, the DCM extracts were rotary evaporated and blown down to ~200 μL for analysis of non-polar OMM using a gas chromatograph (GC, Agilent 7890B)-mass spectrometer (MS, Agilent 5977B). The DCM/methanol extracts were evaporated to dryness and reacted with N, O-bis(trimethylsilyl)trifluoroacetamide (BSTFA) at 70 °C for 3 h to convert the polar OMMs into trimethylsilyl ethers and esters for GC-MS analysis. Table S1 contains information on the non-polar and polar OMMs. All OMMs were quantified using six-point calibration curves following the internal standard method and corrected for field blank values. The recoveries of the authentic standards for non-polar (*n*-alkanes, PAHs, steranes, and hopanes) and polar species (cis-ketopinic acid, anhydrosugars, and sugar polyols; Table S1)

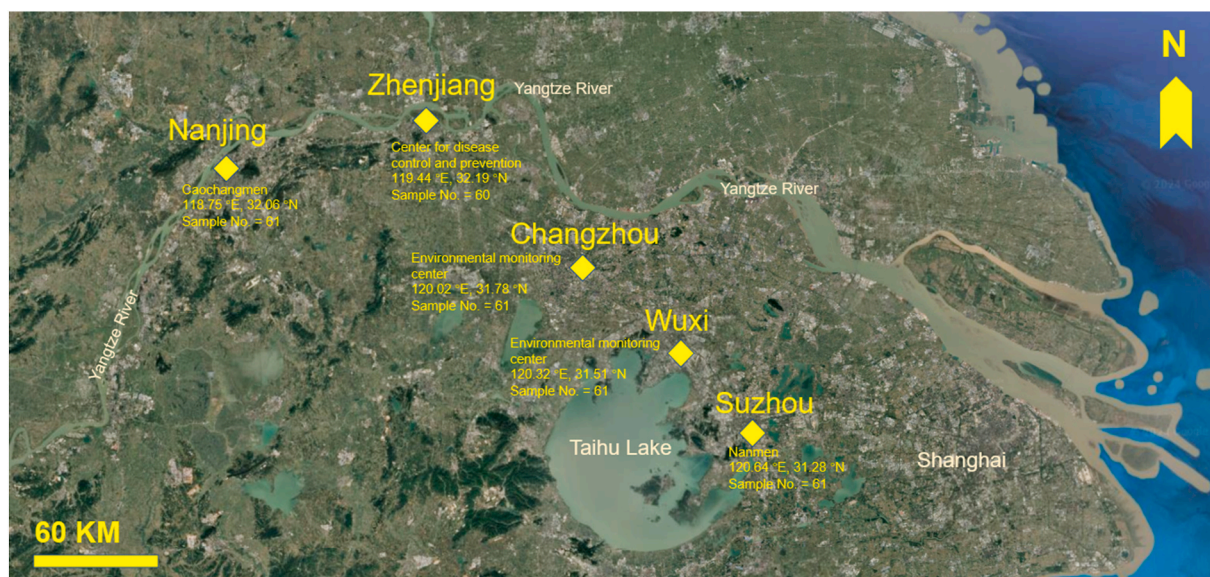


Fig. 1. Locations of sampling sites in five central cities of the Yangtze River Delta, eastern China. Map data: Google Maps.

were in the ranges of  $81.8 \pm 8.89\%$ – $112 \pm 10.9\%$  and  $67.2 \pm 3.50\%$ – $107 \pm 2.65\%$ , respectively, and the corresponding detection limits were in the ranges of  $0.012$ – $0.35 \text{ ng m}^{-3}$  and  $0.14$ – $0.71 \text{ ng m}^{-3}$ . The measurement results of the  $\text{PM}_{2.5}$ -bound OMMs were not adjusted by recoveries.

### 2.3. PMF analysis

The EPA PMF 5.0 model was applied to identify and quantify the sources of  $\text{PM}_{2.5}$  major components, especially for OC, in the five cities. Besides  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , OC, and EC,  $\text{Ca}^{2+}$  and  $\text{K}^+$  data were also included as they are commonly used as source indicators for dust and biomass burning, respectively (Huang et al., 2010; Urban et al., 2012; Fourtziou et al., 2017). The OMM input data were pre-selected based on the uniqueness of the source makers, the proportion of valid measurements ( $>30\%$ ), and the interpretability of the output base-case solutions (Xie et al., 2013; Feng et al., 2023). Due to the limited number of  $\text{PM}_{2.5}$  samples available at each site ( $N = \sim 61$ ), all sample data from the five cities were pooled for PMF analysis. However, the samples that did not contain OMM data were omitted. This resulted in a data set with 296 observations of 48 species (7 bulk components and 41 OMMs). The final factor number was determined primarily by referring to the physical meaningfulness of the output factors. In addition, the robustness of PMF solutions with 4–10 factors and their  $Q/Q_{\text{exp}}$  values were also considered (Table S2). Further details on the preparation of the input data set and the determination of the factor number for the PMF analysis are provided in Text S1 in Supporting Information.

### 2.4. Assessment of spatial variability

The correlation coefficient ( $r$ ) and the coefficient of divergence (COD) were calculated to assess the spatial variability of  $\text{PM}_{2.5}$  component concentrations and source contributions. The correlation coefficient is used to measure how the same species or sources vary together at two sampling sites, and the COD helps determine how different locations diverge in terms of species concentrations or source contributions, and is defined by the following formula

$$\text{COD}_{jh} = \sqrt{\frac{1}{n} \sum_{i=1}^n \left( \frac{x_{if} - x_{ih}}{x_{if} + x_{ih}} \right)^2} \quad (1)$$

where  $x_{if}$  and  $x_{ih}$  are the concentrations of the same species or the

contributions from the same source for the  $i$ th sampling day at sites  $f$  and  $h$ , and  $n$  is the number of sample pairs. A COD value of 0 indicates complete similarity between the two sampling sites, while a value close to 1 suggests complete differences.

## 3. Results and discussion

### 3.1. Overview of bulk speciation

$\text{NO}_3^-$  was the most abundant  $\text{PM}_{2.5}$  species in all five cities ( $9.54 \pm 10.1$ – $12.1 \pm 11.3 \mu\text{g m}^{-3}$ ; Table 1) during the sampling campaign, accounting for an average of  $30.0 \pm 12.8\%$  to  $34.5 \pm 12.0\%$  of reconstructed  $\text{PM}_{2.5}$  ( $\text{rPM}_{2.5}$ ) — the sum of all bulk components. As shown in Fig. S2,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{NH}_4^+$  (SNA) dominated the  $\text{PM}_{2.5}$  composition, especially after the heating season in northern China started on November 15, 2020. The mean contributions of  $\text{NO}_3^-$  to  $\text{rPM}_{2.5}$  in the five cities with  $\text{rPM}_{2.5} > 35 \mu\text{g m}^{-3}$  (Class II limit value for  $\text{PM}_{2.5}$  in the Chinese NAAQS) during the heating season ranged from  $36.3 \pm 7.39\%$  to  $47.3 \pm 4.61\%$ , significantly higher ( $p < 0.05$ ) than the mean values of the remaining observations ( $24.8 \pm 11.2\%$  to  $29.7 \pm 10.5\%$ ). Since the equivalent ratios of  $\text{NH}_4^+$  to  $\text{SO}_4^{2-} + \text{NO}_3^-$  were always close to unity, the sharp increase in  $\text{rPM}_{2.5}$  after November 15, 2020 was directly caused by the rapid formation of  $\text{NH}_4\text{NO}_3$ . The average OC concentrations were comparable to or higher than those of  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$ , contributing an average of  $18.7 \pm 7.39\%$ – $26.7 \pm 8.45\%$  to  $\text{rPM}_{2.5}$  (Table 1), indicating a necessity in controlling OA emissions and formation to further mitigate  $\text{PM}_{2.5}$  pollution in the Yangtze River Delta in autumn and winter.

Fig. S3 compares the concentration time series of individual bulk species in the five cities, and all components show similar temporal variations. Only  $\text{Ca}^{2+}$ , OC, and EC were obviously higher at their peak values in Changzhou than in the other four cities (Figs. S3c, f, and g). Since the combustion-related carbonaceous aerosols can mix with surface dust and be resuspended (Xie et al., 2022a; Feng et al., 2023), the sampling site in Changzhou may be more affected by dust resuspension than the other four sites. The average concentrations of  $\text{rPM}_{2.5}$ , which is dominated by SNA, showed no significant differences ( $p > 0.05$ ) among the sampling sites, and the time series of SNA and  $\text{rPM}_{2.5}$  in the five cities almost overlapped (Figs. S3a, d, e, and h). Except for  $\text{Ca}^{2+}$ ,  $\text{PM}_{2.5}$  bulk species had high  $r$  (mean,  $0.78 \pm 0.10$ – $0.90 \pm 0.05$ ) and low COD ( $0.15 \pm 0.03$ – $0.24 \pm 0.04$ ) values between sampling sites (Fig. 2), indicating high spatial homogeneity for  $\text{PM}_{2.5}$  major components. Therefore,  $\text{PM}_{2.5}$  pollution in the center of the Yangtze River Delta during the heating

**Table 1**  
Statistics for the concentrations of PM<sub>2.5</sub> bulk components (μg m<sup>-3</sup>) and OMM groups (ng m<sup>-3</sup>) in five central cities of the Yangtze River Delta.

	Nanjing		Suzhou		Wuxi		Changzhou		Zhenjiang	
	Median	Mean ± stdev	Median	Mean ± stdev	Median	Mean ± stdev	Median	Mean ± stdev	Median	Mean ± stdev
<b>Bulk components</b>										
NH <sub>4</sub> <sup>+</sup>	3.79	5.22 ± 3.86	3.54	4.32 ± 3.51	3.73	4.61 ± 3.45	3.50	4.58 ± 2.99	3.90	4.92 ± 3.98
K <sup>+</sup>	0.33	0.40 ± 0.19	0.27	0.31 ± 0.18	0.32	0.39 ± 0.21	0.39	0.42 ± 0.20	0.39	0.43 ± 0.20
Ca <sup>2+</sup>	0.19	0.23 ± 0.12	0.16	0.22 ± 0.15	0.16	0.21 ± 0.11	0.33	0.40 ± 0.30	0.16	0.22 ± 0.15
NO <sub>3</sub> <sup>-</sup>	7.92	12.1 ± 11.3	6.21	9.54 ± 10.1	6.87	10.0 ± 10.1	7.69	10.8 ± 9.15	7.28	10.9 ± 11.4
SO <sub>4</sub> <sup>2-</sup>	4.72	5.57 ± 3.24	4.08	4.69 ± 3.33	4.53	5.01 ± 3.15	4.33	5.05 ± 3.06	4.54	5.42 ± 3.41
OC	4.27	4.81 ± 2.64	4.48	5.03 ± 2.70	4.77	5.37 ± 3.12	6.88	8.09 ± 5.17	4.94	5.48 ± 2.50
EC	1.60	1.92 ± 1.33	1.48	1.95 ± 1.53	1.75	2.26 ± 1.70	2.17	2.97 ± 2.47	2.13	2.49 ± 1.47
rPM <sub>2.5</sub> <sup>a</sup>	21.9	30.1 ± 20.7	21.1	26.0 ± 19.6	22.0	27.8 ± 20.1	27.4	32.3 ± 20.5	23.3	29.8 ± 21.3
OC% <sup>b</sup>	17.8	18.7 ± 7.39	22.6	22.7 ± 7.90	21.9	21.8 ± 7.31	27.3	26.7 ± 8.45	21.2	22.0 ± 7.94
<b>Non-polar OMMs</b>										
<b>n-Alkanes</b>										
lg p <sup>0,*</sup> <sub>L</sub> > -8	2.72	3.26 ± 2.35	3.05	3.21 ± 1.76	0.54	1.96 ± 3.06	1.14	3.12 ± 5.02	0.56	1.24 ± 1.33
-10 < lg p <sup>0,*</sup> <sub>L</sub> < -8	6.76	11.1 ± 9.37	18.0	19.9 ± 8.99	9.56	13.2 ± 12.6	14.8	25.3 ± 27.2	9.75	12.7 ± 10.8
lg p <sup>0,*</sup> <sub>L</sub> < -10	28.1	32.1 ± 16.0	29.5	32.7 ± 18.2	19.8	25.5 ± 18.3	42.0	66.1 ± 53.8	20.7	24.5 ± 14.5
Subtotal	38.1	46.5 ± 25.9	52.6	55.8 ± 25.9	30.3	40.7 ± 32.2	63.3	96.0 ± 81.4	32.3	38.5 ± 24.7
<b>PAHs</b>										
lg p <sup>0,*</sup> <sub>L</sub> > -8	0.30	0.46 ± 0.52	0.31	0.48 ± 0.54	0.24	0.41 ± 0.52	0.41	0.70 ± 0.76	0.37	0.50 ± 0.53
-10 < log p <sup>0,*</sup> <sub>L</sub> < -8	1.04	1.49 ± 1.37	0.61	1.28 ± 1.41	0.95	1.50 ± 1.47	1.16	2.03 ± 2.03	1.29	1.79 ± 1.58
lg p <sup>0,*</sup> <sub>L</sub> < -10	4.20	5.03 ± 3.51	3.60	4.66 ± 3.93	4.82	5.94 ± 4.12	6.14	8.56 ± 7.55	6.02	6.77 ± 4.48
Subtotal	5.38	6.98 ± 5.19	4.43	6.42 ± 5.50	5.85	7.85 ± 5.77	7.85	11.3 ± 9.76	8.22	9.06 ± 6.33
<b>oxy-PAHs</b>										
lg p <sup>0,*</sup> <sub>L</sub> > -8	0.17	0.31 ± 0.38	0.13	0.27 ± 0.35	0.19	0.33 ± 0.38	0.24	0.40 ± 0.41	0.20	0.30 ± 0.30
lg p <sup>0,*</sup> <sub>L</sub> < -8	0.32	0.45 ± 0.43	0.38	0.47 ± 0.28	0.47	0.64 ± 0.50	0.58	0.86 ± 0.72	0.46	0.54 ± 0.44
Subtotal	0.51	0.76 ± 0.71	0.50	0.75 ± 0.58	0.66	0.97 ± 0.80	0.82	1.26 ± 1.03	0.67	0.84 ± 0.68
<b>Steranes and hopanes</b>										
Subtotal	0.38	0.55 ± 0.48	0.78	0.98 ± 0.63	0.96	1.17 ± 0.92	1.99	3.31 ± 3.24	0.64	0.79 ± 0.53
<b>Polar OMMs</b>										
Isoprene SOA tracers	2.54	8.57 ± 18.3	2.25	5.68 ± 12.2	2.00	6.51 ± 14.6	2.38	6.29 ± 11.4	2.47	5.43 ± 9.11
a-Pinene SOA tracers	7.21	11.5 ± 10.7	7.18	9.94 ± 8.52	8.62	13.8 ± 13.7	6.04	10.3 ± 11.6	6.74	8.40 ± 7.00
Biomass burning tracers	82.5	128 ± 215	65.1	91.0 ± 73.2	87.4	115 ± 98.1	111	171 ± 159	99.3	132 ± 104
Sugar alcohols	7.56	9.01 ± 4.83	7.12	8.69 ± 5.63	6.34	8.19 ± 4.86	13.3	15.8 ± 9.32	9.89	10.7 ± 6.75
Saccharides	13.5	15.3 ± 7.43	13.3	15.4 ± 9.53	11.6	13.8 ± 7.83	19.0	23.6 ± 13.3	12.8	13.5 ± 5.90

<sup>a</sup> rPM<sub>2.5</sub> = NH<sub>4</sub><sup>+</sup> + K<sup>+</sup> + Ca<sup>2+</sup> + NO<sub>3</sub><sup>-</sup> + SO<sub>4</sub><sup>2-</sup> + OC + EC.

<sup>b</sup> OC% = OC/rPM<sub>2.5</sub> × 100%.

<sup>c</sup> Vapor pressure (atm) at 298.15 K based on values from Gou et al. (2021).

season is not primarily caused by local direct emissions, arguing for the establishment of joint prevention and control mechanisms at the regional level.

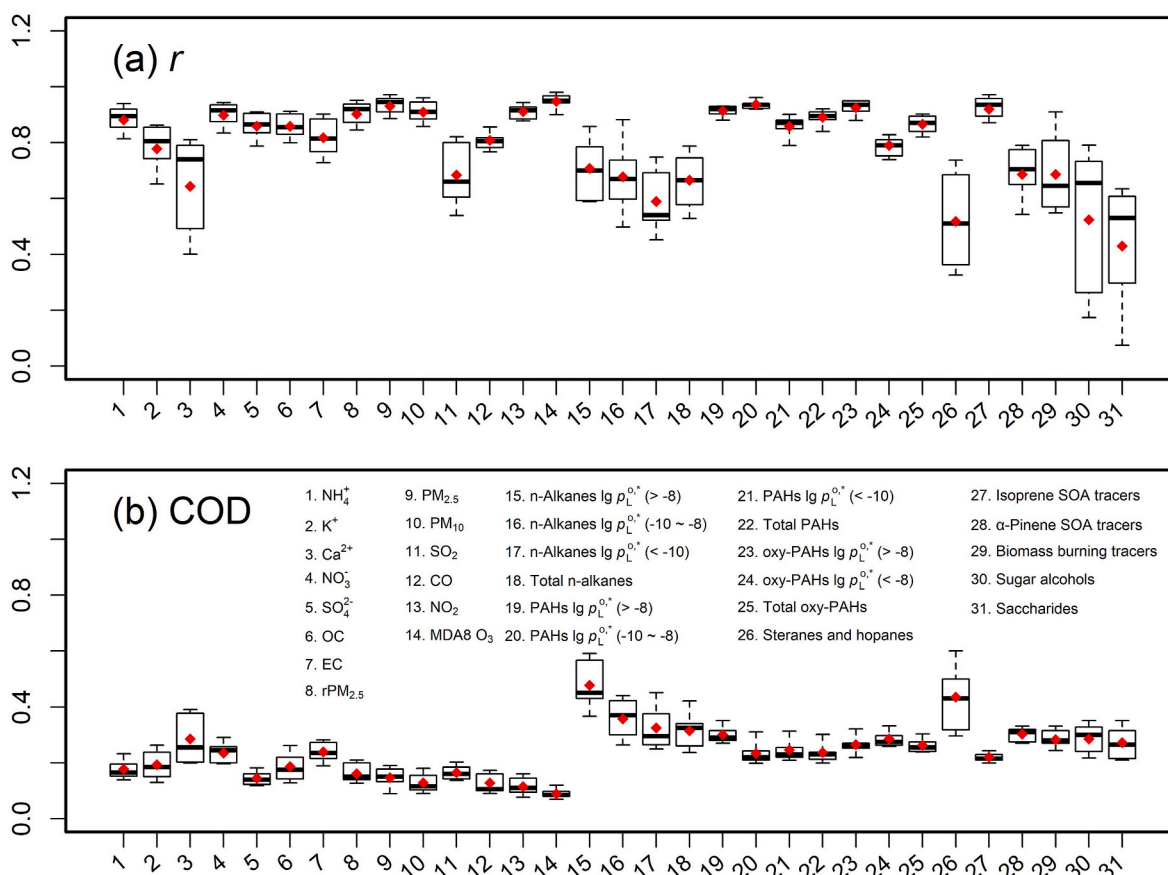
Fig. S1 shows that the concentrations of individual criteria air pollutants in the five cities also have similar temporal variations, and their COD values (0.09 ± 0.02–0.16 ± 0.03; Fig. 2b) suggest a high degree of similarity between the sampling sites. However, the *r* values of SO<sub>2</sub> concentrations (0.68 ± 0.11) were lower than those of other air pollutants (*r* = 0.81 ± 0.05–0.95 ± 0.03) and SO<sub>4</sub><sup>2-</sup> (0.86 ± 0.05, Fig. 2a), supporting that SO<sub>4</sub><sup>2-</sup> in PM<sub>2.5</sub> was not formed locally but during regional transport through atmospheric oxidation of SO<sub>2</sub> (van Donkelaar et al., 2008; He et al., 2012). Yu et al. (2020) and Xie et al. (2022b) found that the PM<sub>2.5</sub> mass in Nanjing city can be well reproduced by the sum of SNA, OC, and EC. Here, the PM<sub>2.5</sub> data collected for the five cities were in good agreement with rPM<sub>2.5</sub> (Fig. S4). These results validated our bulk speciation data and showed that rPM<sub>2.5</sub> can be used as a proxy measurement for PM<sub>2.5</sub> in the center of the Yangtze River Delta.

### 3.2. OMM composition

The median and mean concentrations of the measured OMM groups in the five cities are listed in Table 1. Tables S3 and S4 provide the median and mean concentrations of individual non-polar and polar OMMs, respectively. In all five cities, most *n*-alkanes and PAHs had a subcooled liquid vapor pressure at 298.15 K (*p*<sup>0,\*</sup><sub>L</sub>) of less than 10<sup>-10</sup> atm, and these low-volatility compounds dominated the concentrations of total *n*-alkanes and total PAHs in PM<sub>2.5</sub> (Table 1); the total concentrations of oxygenated PAHs (oxy-PAHs) with *p*<sup>0,\*</sup><sub>L</sub> < 10<sup>-8</sup> atm (1,8-NAA, ATQ, and 7H-BANT) were also higher than those with *p*<sup>0,\*</sup><sub>L</sub> > 10<sup>-8</sup> atm (ACE, FLO, PHL-O, and XA; Table 1S and S3). Because the gas + particle phase concentrations of individual *n*-alkanes and PAHs

generally follow a decreasing trend the lower the *p*<sup>0,\*</sup><sub>L</sub> value in northern Nanjing (Gou et al., 2021), the dominance of low-volatility species for non-polar OMM groups in PM<sub>2.5</sub> is largely due to the evaporation of low molecular weight (MW) compounds. Although PM<sub>2.5</sub> sampling was conducted at urban sites in autumn and winter, high MW *n*-alkanes showed a clear predominance of odd over even chain lengths (C27 – C35; Table S3), indicating contributions from epicuticular wax in dust (Rogge et al., 1993b). Among the five cities, Changzhou had the highest concentrations of all non-polar OMM groups, particularly for high MW *n*-alkanes (*p*<sup>0,\*</sup><sub>L</sub> < 10<sup>-10</sup> atm), steranes, and hopanes, and was more affected by fossil fuel combustion and road dust.

As shown in Fig. 3, the biomass burning tracers had the highest percentage (mean ± stdev, 1.07 ± 0.38%–1.41 ± 0.66%) of organic matter (OM = 1.6 × OC), followed by high MW *n*-alkanes with *p*<sup>0,\*</sup><sub>L</sub> < 10<sup>-10</sup> atm (0.28 ± 0.09%–0.52 ± 0.31%) and saccharides (0.19 ± 0.12%–0.23 ± 0.13%). The secondary products of isoprene and α-pinene exhibited noticeable contributions (up to ~2%) to OM only in the first half of September, when biogenic VOC emissions and solar radiation were still intense. Sugar alcohols and saccharides are typical source markers for biological PM (Simoneit et al., 2004) and can also originate from biomass burning in winter (Marynowski and Simoneit, 2022; Feng et al., 2023; Cui et al., 2024). Similar to the non-polar OMMs, the highest average concentrations of biomass burning tracers, sugar alcohols, and saccharides were also observed at the Changzhou site (Table 1), where anthropogenic emissions exerted the greatest influence. The secondary products of isoprene and α-pinene reached their highest concentrations in Nanjing (8.57 ± 18.3 ng m<sup>-3</sup>) and Wuxi (13.8 ± 13.7 ng m<sup>-3</sup>), respectively, but not in Changzhou. In addition to the fact that the precursors of the measured SOA products were mainly from biogenic sources (Claeys et al., 2004, 2007; Szmigielski et al., 2007), this could also be partly due to the spatial homogeneity of SO<sub>4</sub><sup>2-</sup> and NO<sub>2</sub> (Fig. 2),



**Fig. 2.** Distribution of (a) correlation coefficients ( $r$ ) and (b) coefficient of divergence (COD) for bulk  $PM_{2.5}$  components, criteria air pollutants, and individual groups of OMMs between two sampling sites. The boxes depict the median (dark line), the inner quartile (box), the 10th and 90th percentiles (whiskers), and the mean (red diamond) of  $r$  and COD for 10 pairs of sampling sites. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

as the biogenic SOA derived from isoprene and monoterpenes can be mediated by anthropogenic sulfate and  $NO_x$  (Xu et al., 2015).

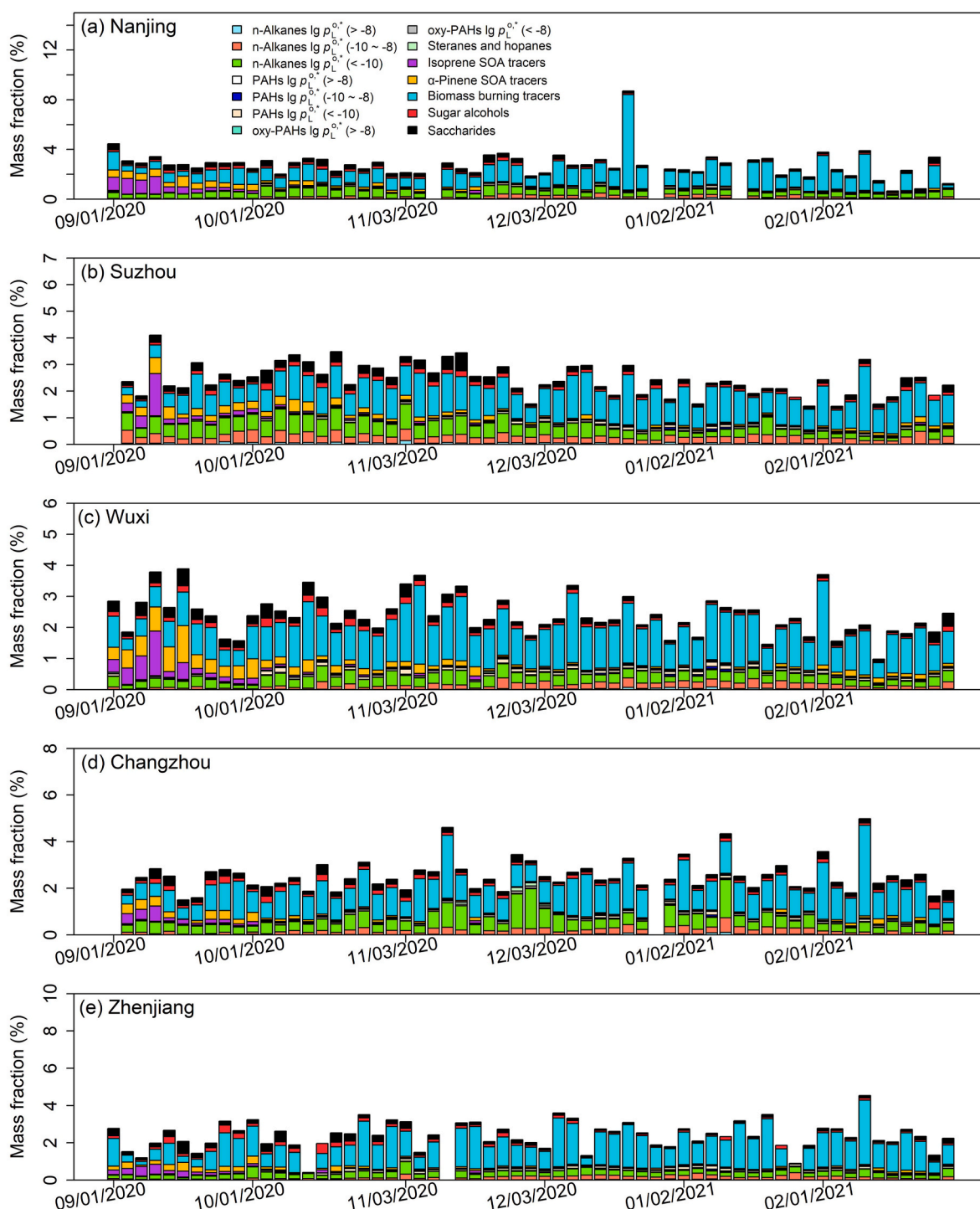
### 3.3. Spatiotemporal variations of OMMs

The temporal variations of the individual OMM groups in the five cities are illustrated in Figs. S5–S8. Fig. 2 visualizes the distribution of correlation coefficients ( $r$ ) and COD for each OMM group between two sampling sites using boxplots. In general, the non-polar OMMs and biomass burning tracers reached their maximum concentrations in December 2020 and January 2021, which is due to the increasing use of biomass and fossil fuels and the low boundary layer height in winter. Concentrations of isoprene and  $\alpha$ -pinene products showed a decreasing trend since September 2020 (Figs. S8a and b), as biogenic emissions and photochemical processes weakened after the end of summer. Compared to the temporal pattern of  $\alpha$ -pinene SOA tracers, which continuously decreased from autumn to winter, isoprene products rapidly diminished to a minimum level from September to October 2020. Since isoprene and  $\alpha$ -pinene are mainly emitted from deciduous and coniferous trees during the growing season, the temporal pattern of isoprene products may indicate a rapid decline in isoprene emissions due to the transition from the growing season to the dormant season of deciduous trees. Except for Changzhou, the sugar alcohols and saccharides in the other four cities did not show a clear temporal pattern (Figs. S8d and e). This is because sugar polyols are mainly derived from soil microorganisms and vegetation during the growing season and are affected by biomass burning in winter (Yttri et al., 2007; Verma et al., 2018).

Although the average concentrations of  $n$ -alkanes with  $p^{o,*}_L > 10^{-8}$

atm were comparable in Nanjing ( $3.26 \pm 2.35 \text{ ng m}^{-3}$ ), Suzhou ( $3.21 \pm 1.76 \text{ ng m}^{-3}$ ), and Changzhou ( $3.12 \pm 5.02 \text{ ng m}^{-3}$ ; Table 1), the Changzhou site showed much higher peak values on 12/21/2020 ( $31.3 \text{ ng m}^{-3}$ ) and November 01, 2021 ( $15.6 \text{ ng m}^{-3}$ ; Fig. S5a), when concentration peaks of  $n$ -alkanes with lower volatility ( $p^{o,*}_L < 10^{-8} \text{ atm}$ ) also appeared (Figs. S5b and c). Due to the large intermittent peaks of  $n$ -alkanes with  $p^{o,*}_L < 10^{-8} \text{ atm}$  from November 2020 to January 2021 in Changzhou, their average concentrations were significantly higher ( $p < 0.01$ ) than those in the other four cities. Considering that  $n$ -alkanes have multiple sources involving both biogenic emissions (e.g., plants and microorganisms; Rogge et al., 1993b, c) and anthropogenic activities (e.g., fossil fuel combustion, biomass burning, and cooking; Ren et al., 2020; Rogge et al., 1993a; Schauer et al., 1999, 2001, 2002), the wide ranges of  $r$  and COD values in Fig. 2 suggest considerable spatial heterogeneity of  $n$ -alkanes in all  $p^{o,*}_L$  ranges during autumn and winter.

Unlike  $n$ -alkanes, the concentrations of PAHs and oxy-PAHs in the five cities exhibited a consistent temporal pattern, peaking in December 2020 and January 2021 (Figs. S6–S7). Moreover, PAHs and oxy-PAHs had high  $r$  ( $0.79 \pm 0.050$ – $0.94 \pm 0.016$ ; Fig. 2a) and low COD values ( $0.23 \pm 0.048$ – $0.30 \pm 0.033$ ; Fig. 2b) with narrow distributions. Given the strong correlation ( $r > 0.90$ ,  $p < 0.01$ ) between total PAHs and total oxy-PAHs in each city, both groups of OMMs were expected to originate mainly from incomplete combustion of fossil fuels and to be homogeneously distributed in the center of the Yangtze River Delta in autumn and winter. However, large between-site differences were observed at peak concentrations of PAHs and oxy-PAHs in winter, and Changzhou had significantly ( $p < 0.01$ ) higher average concentrations of PAHs and oxy-PAHs than the other four cities (Table 1). One possible explanation



**Fig. 3.** Stacked time series of the contribution of each OMM group to organic matter ( $1.6 \times \text{OC}$ ) in (a) Nanjing, (b) Suzhou, (c) Wuxi, (d) Changzhou, and (e) Zhenjiang.

is that the sampling site in Changzhou is located near the industrial area in the northern part of the city, where large amounts of air pollutants were emitted and  $\text{PM}_{2.5}$  pollution was severe in winter (Tao et al., 2021a, b). The intermittent PAH emissions from potential local sources (e.g., biomass burning, fireworks, and waste incineration) could also contribute to the differences between sites in the peak concentrations of PAHs in winter (Peng et al., 2016; Dat and Chang, 2017; Pongpiachan et al., 2017a, b). Steranes and hopanes are enriched in fuels and lubricants derived from crude oil and coal (Kaplan et al., 2001). Their concentrations were strongly correlated with low volatile *n*-alkanes ( $\rho^{0*}_L <$

$10^{-10}$  atm;  $r > 0.65$ ) in the five cities, especially in Wuxi ( $r = 0.88$ ) and Changzhou ( $r = 0.80$ ). In Fig. 2, steranes and hopanes show a wide range of  $r$  values and the highest COD ( $0.44 \pm 0.14$ ). These results suggest a heterogeneous spatial distribution of hopanes and steranes in autumn and winter, which can be attributed to the difference in local emissions from motor vehicles and road dust.

Of all the OMM groups, the isoprene SOA tracers had the lowest average COD ( $0.22 \pm 0.024$ ) and a similar distribution of  $r$  values as PAHs and oxy-PAHs (Fig. 2). The homogeneous spatial distribution of isoprene products was probably due to their minimal formation after

September. While the  $\alpha$ -pinene SOA tracers exhibited greater spatial variability with average  $r$  and COD values close to those of the total  $n$ -alkanes. This could be ascribed to the fact that  $\alpha$ -pinene can also be emitted during biomass burning (Akagi et al., 2013). In this work, the biomass burning tracers showed moderate  $r$  values (0.53–0.91) and a narrow range of COD (0.19–0.34), indicating influences of both regional transport and local emissions. In the five cities, strong correlations ( $r > 0.70$ ) between sugar polyols and biomass burning tracers were observed only in Suzhou and Changzhou. Due to the spatial variability of the contributing sources (biomass burning and biogenic emissions) of sugar alcohols and saccharides, their  $r$  values in Fig. 2a show the widest distributions.

### 3.4. PMF analysis

After testing PMF solutions with 4–10 factors, an 8-factor solution was finally chosen as having the most physically meaningful factors (Fig. S9). The error estimation results confirmed the robustness of the 4- to 9-factor solutions (Text S1 in Supporting Information and Table S2), and the  $Q/Q_{exp}$  value changed less with the factor number varying from 8 to 10 factors (9.01%–9.05%). In Table S5, the measurement data of  $PM_{2.5}$  bulk species, particularly for the major components (SNA, OC, and EC), agree well with the PMF estimates. Thus, using an 8-factor solution to interpret the input data is appropriate. Referring to the dominant species in each factor profile (Fig. S9) and previous OMM-based source apportionment studies in Nanjing (Xie et al., 2022c; Feng et al., 2023), the resolved eight factors could be linked with biomass burning,

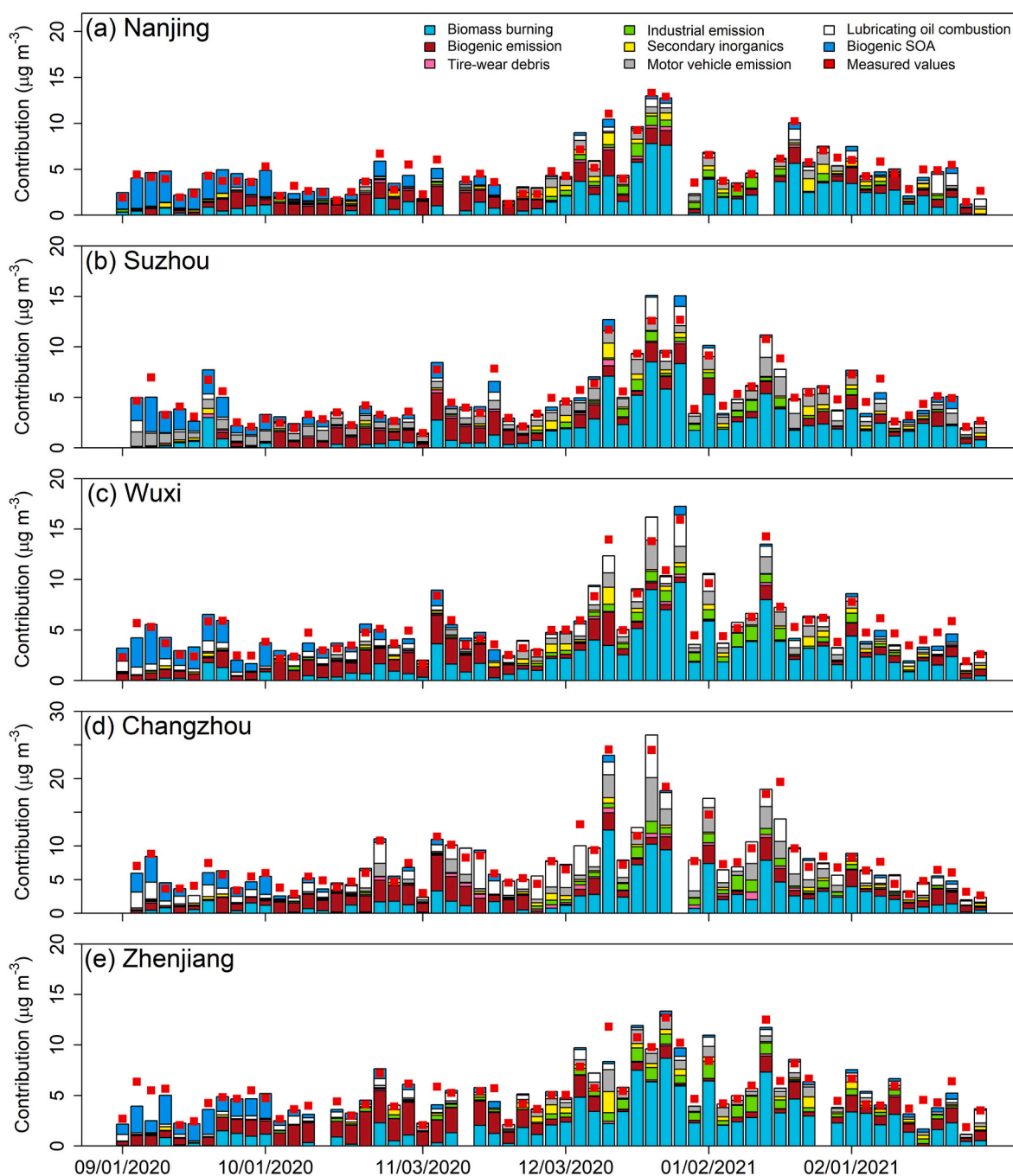


Fig. 4. Temporal variations of factor contribution distributions of OC in (a) Nanjing, (b) Suzhou, (c) Wuxi, (d) Changzhou, and (e) Zhenjiang.

biogenic emission, tire-wear debris, industrial emission, secondary inorganics, motor vehicle emission, lubricating oil combustion, and biogenic SOA. Their average contributions (%) to PM<sub>2.5</sub> major components in each city are shown in Table S6 in Supporting Information. The time series of the distribution of factors for the contribution of OC and total bulk components are shown in Fig. 4 and S10.

The biomass burning factor contained the largest proportion of galactosan, mannosan, and levoglucosan (53.9%–60.0%; Fig. S9a), which are typical byproducts of cellulose pyrolysis (Simoneit et al., 1999; Fabbri et al., 2009), and dominated the contributions to OC in December 2020 and January 2021 (Fig. 4). Its average relative contributions to OC and EC in the five cities were in the ranges of 27.9%–39.8% and 45.4%–57.9%, respectively (Table S6). Unlike the distribution of *r* values for the biomass burning tracers (Fig. 2a), the contributions of the biomass burning factor in the five cities showed strong correlations ( $r = 0.74\text{--}0.93$ ,  $p < 0.01$ ; Fig. 5a), indicating regional influences rather than local emissions. In addition to OC and EC, the contributions of the biomass burning factor at each site were also significantly correlated with NO<sub>3</sub><sup>-</sup> ( $r = 0.63\text{--}0.81$ ,  $p < 0.01$ ) and rPM<sub>2.5</sub> ( $r = 0.66\text{--}0.87$ ,  $p < 0.01$ ). Considering the high EC loading, the biomass burning factor was significantly affected by the long-range transport of biomass burning smoke and fossil fuel aerosols from residential heating in northern China in winter, and the use of clean energy (e.g., electricity and natural gas) should be increased to reduce emissions from winter heating. However, the influence of the aging process cannot be ruled out due to the lack of suitable organic tracers and source cycles (Hodshire et al., 2019; Qi and Wang, 2019).

The industrial emission, secondary inorganics, and biogenic SOA factors consisted mainly of PAHs, SNA, and secondary products of isoprene and  $\alpha$ -pinene, respectively (Figs. S9d, e, and h). The contributions of all these three factors showed strong correlations ( $r = 0.85 \pm 0.043\text{--}0.89 \pm 0.070$ ,  $p < 0.01$ ) between the sampling sites (Fig. 5a), and only the biogenic SOA factor contributed significantly to OC and total bulk species in September (Fig. 4). In southern China, the increase of PAHs in winter was expected to be mainly caused by regional and local emissions from fossil fuel combustion (Yan et al., 2019). In this case, the strong correlations among sampling sites suggested the predominance of regional transport over local emissions, and the differences in local combustion sources and dispersion conditions across the five cities were responsible for the greater spatial heterogeneity of the industrial emission factor (COD =  $0.46 \pm 0.045$ ) compared with the secondary inorganics (COD =  $0.31 \pm 0.053$ ) and biogenic SOA factors (COD =  $0.34 \pm 0.047$ ; Fig. 5b). Some of the input SOA products (e.g., 2-methyltetrols) may exist in the form of organosulfate by the reactive uptake of their gas-phase intermediates (e.g., isoprene epoxy diols; Surratt et al., 2010; Cui et al., 2018). In Fig. S9h, a notable fraction of SO<sub>4</sub><sup>2-</sup> (24.1%) is probably incorrectly assigned to the biogenic SOA factor, which could be due to the fact that the PMF model assumes constant factor profiles and the diurnal cycles of the input species are not available. Unlike the biomass burning factor, the regional sources represented by these three

factors were not the main contributors of the carbonaceous components in the five cities during autumn and winter (Table S6).

The biogenic emission factor was featured by significant proportions of sugar polyols (33.1%–55.1%) and also contained ~20% of biomass burning tracers (Fig. S9b). It dominated the contributions to OC from mid-September to mid-November in all five cities (Fig. 4). Besides the activities of microbiota and the decomposition of plant material in the soil, biomass burning also produces sugar polyols (Marynowski and Simoneit, 2022). In Fig. 5, the biogenic emission factor shows a broader *r* distribution (0.36–0.73) than the biomass burning factor with an average COD of  $0.40 \pm 0.024$ . Thus, this factor reflects combined contributions from local sources of soil resuspension and biomass burning in autumn and winter, which may explain the spatial heterogeneity of biomass burning tracers and sugar polyols (Fig. 2). Because the biomass burning and biogenic emission factors contained up to 28.8% of PAH compounds, regional transport and local emissions of biomass burning smoke were partly responsible for the elevated PAH levels in winter (Mandalakis et al., 2005; Pongpiachan, 2015, 2017a; Mao et al., 2018).

The tire-wear debris, motor vehicle emission, and lubricating oil combustion factors were characterized by high MW *n*-alkanes (*n*-C27–*n*-C35), *n*-alkanes with carbon numbers of 22–26, and steranes/hopanes, and were identified as traffic-related sources based on previous studies (Xie et al., 2013, 2022c; Feng et al., 2023). Among the five cities in southern Jiangsu, Suzhou had the highest number of motor vehicles (5.01 million) and highway passenger traffic (130 million) in 2022, followed by Nanjing and Wuxi (Jiangsu Provincial Bureau of Statistics of China, 2023), which cannot explain the highest contributions of traffic-related factors to OC (37.3%) and total bulk components (21.1%) at the Changzhou sampling site (Table S6). Several studies have found that the contributions of traffic-related factors and the concentrations of their source markers (e.g., steranes and hopanes) at each site are highly dependent on the surrounding traffic conditions (Xie et al., 2012a, b; Liu et al., 2013; Chow et al., 2022), and the three traffic-related factors identified in this study had lower mean *r* ( $0.27 \pm 0.19\text{--}0.51 \pm 0.16$ ) and higher COD ( $0.47 \pm 0.057\text{--}0.55 \pm 0.10$ ) values than other factors (Fig. 5). Even when their contributions in Changzhou were excluded for the spatial distribution analysis, these three traffic-related factors still exhibited greater spatial heterogeneity. Thus, the heterogeneous spatial distribution of traffic-related factors was caused by local emissions close to the sampling sites. As shown in Fig. 4, the motor vehicle emission and lubricating oil combustion factors have recognizable contributions to the total OC at its peak values in December and January, which can be mitigated by developing intelligent traffic management systems and promoting the adoption of new energy vehicles within each city (Guo et al., 2020b; Wang et al., 2021). Simply dividing the eight factors into two groups based on the distribution and mean values of their *r* values, regional sources, including biomass burning, industrial emission, secondary inorganics, and biogenic SOA, dominate the contributions to SNA (>80%) in all five cities. Except for Changzhou, where local emissions contributed more OC (55.7%) than regional transport, more

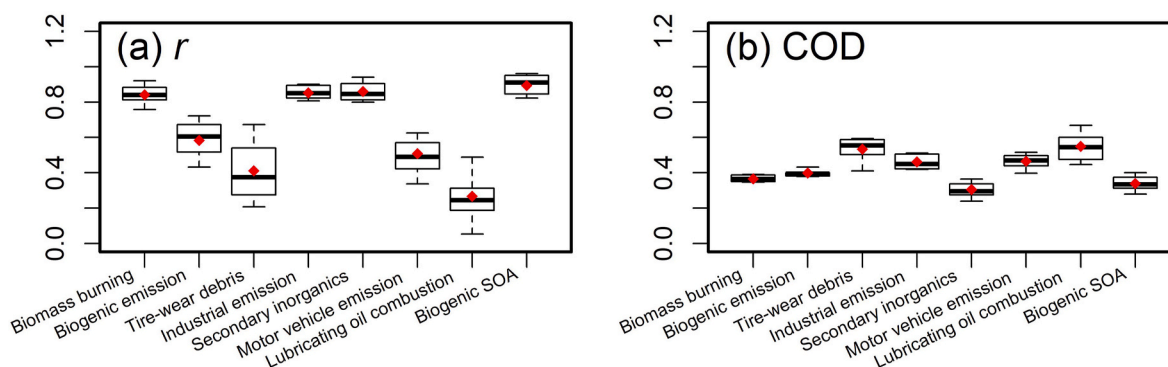


Fig. 5. Distribution of (a) *r* and (b) COD values for individual PMF factors between sampling sites.



than half of the OC in the other four cities came from regional sources (56.6%–65.6%) in autumn and winter.

#### 4. Conclusions

According to the speciation results of PM<sub>2.5</sub> from Nanjing, Suzhou, Wuxi, Changzhou, and Zhenjiang, OC has the second highest relative contribution (18.7 ± 7.39%–26.7 ± 8.45%) to the total concentrations of measured bulk species during autumn and winter, indicating a necessity in controlling OA emission and formation to further reduce PM<sub>2.5</sub> pollution in the Yangtze River Delta. Low volatile *n*-alkanes and PAHs ( $p^{0*}_L < 10^{-10}$  atm) dominated the composition of identified non-polar OMMs in all five cities due to the evaporation of homologues with higher volatility; biomass burning tracers accounted for the highest percentages of OM for most of the sampling period; biogenic SOA products contributed significantly to OM during the first few sampling days at the end of summer. Similar to SNA and OC, PAHs and oxy-PAHs in different  $p^{0*}_L$  ranges and isoprene SOA tracers showed a homogeneous spatial distribution in autumn and winter, while other groups of OMMs exhibited greater spatial heterogeneity due to the influence of local emissions. A regional biomass burning factor, reflecting the transport of biomass burning smoke and fossil fuel aerosols in the heating season, was resolved by PMF analysis of PM<sub>2.5</sub> speciation data, and the contributions of traffic-related factors showed the largest spatial variability. The overall contributions of regional sources dominated OC concentrations in winter, supporting the implementation of joint prevention and control measures (e.g., clean energy use) to reduce OA emissions from residential heating. Effective regulatory strategies are also needed to control local traffic-related OA sources in the Yangtze River Delta.

#### CRediT authorship contribution statement

**Wei Feng:** Writing – original draft, Investigation, Data curation. **Guihong Dong:** Investigation, Data curation. **Wanqing Qi:** Visualization, Investigation. **Yizhen Wang:** Validation, Investigation. **Xiangyu Zhang:** Investigation. **Ke Li:** Writing – review & editing, Resources, Funding acquisition. **Hong Liao:** Writing – review & editing, Resources. **Yuhang Wang:** Writing – review & editing, Resources. **Zhijuan Shao:** Writing – review & editing. **Mingjie Xie:** Writing – review & editing, Supervision, Project administration, Methodology, Conceptualization.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2024.125227>.

#### Data availability

I have shared the link to my data in the manuscript

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