



Characteristics of Volatile Organic Compounds in Nanjing and Suzhou, Two Urban Sites in the Yangtze River Delta, China

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Abstract

A field measurement study of volatile organic compounds (VOCs) was performed in January 2015 in the urban areas of two of the most important cities of the Yangtze River Delta: Nanjing and Suzhou. The objectives of this study included comparing the characteristics of VOC concentrations, comparing the impacts of emissions on VOCs, using species ratios to assess air mass age, and evaluating ozone formation potential (OFP) and secondary organic aerosol formation potential (SOAP) in air masses. The VOC concentrations in Nanjing (34.6 ± 5.8 ppbv) were higher than those of Suzhou (28.1 ± 5.6 ppbv). The most abundant VOC measured was ethane (6.6 ppbv in Nanjing and 3.6 ppbv in Suzhou). Relevant analysis shows that motor vehicle emissions in Suzhou were dominant, whereas industrial emissions in Nanjing also contributed to VOCs. During rush hour, the VOC concentrations in Nanjing were the highest (35.3 ppbv). The *T/B* ratio (0.92–1.79) for the two sites was observed to be relatively low in the other studied cities, indicating the source impact of traffic emissions in the two sites. Indicators *X/B* (0.26–0.39) and *X/E* (0.33–0.66) also confirmed an aging air mass was transported at the two sampling sites. According to principal component factor analysis results, vehicle emissions (44.8% in Nanjing and 30.6% in Suzhou) were the most important contribution to the two sites. Industrial sites were not only likely to experience industrial emissions but were affected by traffic emissions. Using the OFP method, both sites showed the largest percentage of alkenes (59.9% in Nanjing and 62.0% in Suzhou). When comparing SOAP, both sites showed an absolute majority of aromatics (97.6% in Nanjing and 98.3% in Suzhou). To control the formation of O₃ and SOA in the two sites, it is necessary to reduce the concentration of alkenes and aromatics, respectively. By CPF analysis, pollutants transported from the SE and NE have significant effects on the Nanjing site. In the Suzhou, roads and industrial parks in the SE and S of Suzhou have significant impacts on the site.

Volatile organic compounds (VOCs), as important precursors of atmospheric ozone (O₃) and secondary organic aerosols (SOA), are considered to be important pollutants in the atmosphere and have received increasing attention (Barletta et al. 2005; Liu et al. 2008). Many recently published articles have discussed the role of VOCs in atmospheric O₃

and SOA through experimental and numerical model studies (Atkinson and Arey 2003; Kroll and Seinfeld 2008; Leuchner and Rappenglück 2010; Kim et al. 2011). In addition, some components of VOCs, such as benzene, toluene, and 1,3-butadiene, affect human health and lead to a high risk of cancer (US EPA 2008).

The sources of VOCs in the atmosphere are both natural and anthropogenic. On a global scale, natural sources are the main source (Guenther et al. 2006). However, in urban areas, anthropogenic sources are the main source (Sosa et al. 2009). VOCs can be classified into subclasses such as alkanes, alkenes, alkynes, and aromatic hydrocarbons. Alkanes are mainly derived from the incomplete combustion of fuels in vehicles and the volatilization of fuels (Watson et al. 2001). Alkenes and alkynes are mainly emitted from combustion processes, whereas aromatic hydrocarbons are mostly emitted from motor vehicles, fuel volatilization, and solvent use (Chen et al. 2001). It should be noted that although natural gas and liquefied petroleum

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gas are considered “green energy,” they also will emit a certain amount of lighter alkanes (C₂–C₄).

Research on VOCs has been performed extensively around the world. In China, research on related VOCs has been performed in large cities, such as Beijing (Barletta et al. 2005), Shanghai (Cai et al. 2010), Guangzhou (Liu et al. 2008), and Hong Kong (Guo et al. 2011). Compared with large cities, the research on VOCs in medium-sized and small cities is sparse, and there are many remaining questions about local VOC characteristics. Moreover, with the industrial structure transformation of these megacities in recent years, many factories and enterprises that were originally concentrated in megacities have been relocated to medium-sized and small cities nearby, making the sources of VOCs in small- and medium-sized cities more complicated, with an increasing difficulty of governance.

The 56 important VOCs proposed by the photochemical assessment monitoring stations (PAMS), including C₂–C₁₂ alkanes, alkenes, alkynes, and aromatic hydrocarbons, play a key role in the formation of atmospheric ozone and secondary organic aerosols and are internationally recognized as photochemical smog monitoring species. Because of the large differences in the concentrations and activities of these substances in different regions, the contribution of local photochemistry also is very different. For example, alkenes and aromatic hydrocarbons emitted by industrial solvents and motor vehicles in the Pearl River Delta account for 59.4% and 25.8% of the total VOCs, respectively (Guo et al. 2011), whereas in the agricultural provinces of Anhui and Heilongjiang, agricultural activities can contribute 36% and 40%, respectively (Mo et al. 2017). Therefore, it is necessary to formulate appropriate local environmental control measures for the differences in VOC emissions caused by the energy structure and industrial layout of different regions.

Nanjing and Suzhou are located in the western and central parts of the Yangtze River Delta, respectively. They are important economic and industrial centers in this region. However, after years of industrial adjustment and environmental governance, ozone overshoot and haze continue to occur. For example, in 2017, the amount of excess ozone in Nanjing and Suzhou increased by 30% and 25%, respectively, compared with 2016. Therefore, it is necessary to perform relevant research on the concentrations of VOCs to provide a basis for environmental governance policies. In this study, a 1-month continuous VOC observation of Nanjing and Suzhou was conducted to compare the concentrations of VOCs, the impact of different emissions on VOCs in the two cities, the source differences of VOCs, and the ozone formation potential (OFP) and secondary organic aerosol formation potential (SOAP). A conditional probability function (CPF) was used to facilitate source identification. The results obtained in this study are expected to improve the

development and implementation of VOC control strategies in this region.

Measurements and Methods

Sampling Site

The YRD region consists of 14 different cities, surrounded by urban centers and surrounded by areas and/or towns directly managed by the city. Two of the cities are Nanjing and Suzhou (Fig. 1). Nanjing (31° 14' N–32° 36' N, 118° 22' E–119° 14' E), the capital of Jiangsu, is divided into 11 districts. Its land area comprises only 6% of Jiangsu Province, yet Nanjing accounted for 14% of the province's GDP in 2017. The Nanjing chemical industry park is located approximately 5 km northeast of the observation site. There are cogeneration power plants, iron and steel plants, and factories associated with the paint industry. This site contains both industry and traffic.

Suzhou (30° 47' N–32° 02' N, 119° 55' E–121° 20' E) has experienced rapid economic growth in the past two decades, becoming the economic center of Jiangsu Province. Suzhou is divided into five districts and has direct administration of four towns, where the majority of economic activities are located. Commercial centers and residential areas are located around the sampling site.

Description of Measurements and Instruments

VOCs and carbon monoxide (CO) continuous data were obtained in January 2015, and hourly data were provided after strict quality control: measured, identified, and quantified 56 environmental VOC species, including 29 alkanes, 10 alkenes, 16 aromatics, and 1 alkyne (ethyne), using an automated online gas chromatography-flame ionization detector (GC 5000, produced by AMA Instruments GmbH, Ulm, Germany). The GC 5000 is a chromatograph with automatic sampling, enrichment, and analysis capabilities, including two independent analyzers: the GC 5000 VOC analyzer (two-stage enrichment), and the GC 5000 BTX analyzer (single-stage enrichment). This system has been described in detail elsewhere (An et al. 2014; Shao et al. 2016). In brief, the GC 5000 VOC analyzer is a low boiling point VOC analyzer for C₂–C₆ VOC species, and the GC 5000 BTX analyzer is a high boiling point VOC analyzer for C₂–C₆ VOC species, such as benzene, toluene, and *m*-, *p*-, and *o*-xylene (BTX). The VOC low boiling point analyzer was concentrated at 13 °C, adsorbed twice at 20 °C, and desorbed when the temperature increased to 200 °C; separation was obtained through two-dimensional chromatography. An Al₂O₃/Na₂SO₄ porous-layer open-tubular (PLOT) column (diameter: 0.32 mm; thickness of membrane: 5 μm;

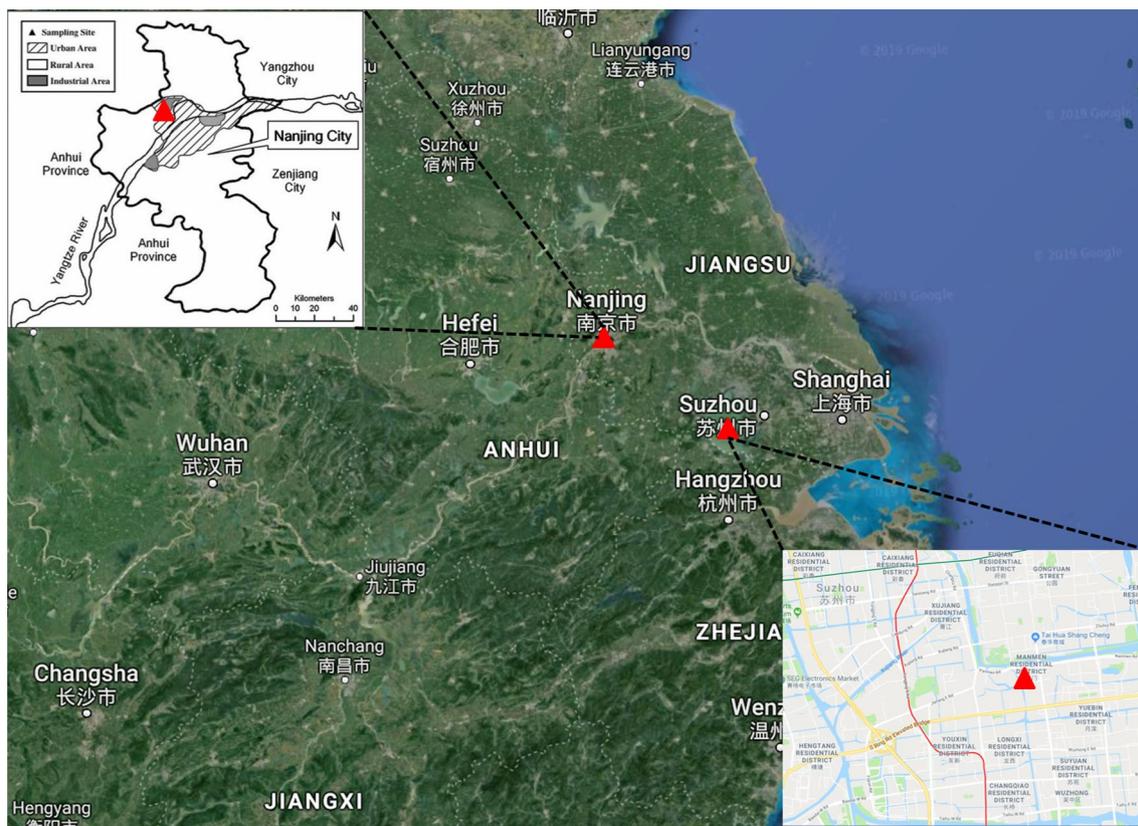


Fig. 1 Location of the sampling site

and length: 60 m) and carbowax backflush column (diameter: 0.32 mm; thickness of membrane: 0.25 μm ; and length: 30 m) were used separately as the chromatographic columns. The function of the backflush column was to remove the water in the component and the high boiling point component. Both types of chromatographic columns were polar columns. After twice conducting thermal desorption, the low boiling point hydrocarbons could be separated. A flame ionization detector (FID) was used as the detector, and the BTX high boiling point analyzer pre-concentrated the volatile organic compounds at 30 $^{\circ}\text{C}$ and then thermally desorbed them; afterwards, the analyzer separated the compounds in the DB-1 column (diameter: 0.32 mm; thickness: 10 μm ; and length: 60 m) to achieve optimum separation and prevent interference from related compounds. The 56 compounds designated by the U.S. Environmental Protection Agency (EPA) were used as the target compounds. The time resolution was 1 cycle/h to ensure the accuracy of the test data. Before monitoring, the U.S. EPA-approved ozone precursor standard photochemical gases were used to calibrate equipment. Standard gases were obtained from the Photochemical Evaluation Monitoring Station (PAMS) and included 56 gases at a concentration of 1 ppbv. Every 2 weeks, the standard gases were passed through the 5-point method for

equipment calibration and verification. Detailed information on the target analyses involved in this study and their corresponding linearity of calibration (r^2), measurement relative standard deviation (RSD), and method detection limit (MDL) is presented in Table S1.

The CO concentrations were continuously measured using a trace gas instrument 48i CO analyzer (manufactured by Thermo Environmental Instruments Inc., USA). An et al. (2015) provide more information on detailed calibration of instruments and instruments. The 48i CO analyzer operates on the principle that CO absorbs infrared radiation at a wavelength of 4.6 μm . The calibration of 48i was conducted by a dynamic gas calibrator (Model 146) in cooperation with the zero air supplier (Model 111), and a standard reference gas mixture (CO in N_2) was used for multipoint calibration and regular zero/span checks. Besides, the gas of CO was compared with National Institute of Standards and Technology (NIST)-traceable standards from Scott Specialty Gases of USA for traceability.

Principal Component Factor Analysis

Principal component factor analysis (PCA) was performed to determine the primary VOC source. The purpose of factor

analysis is to describe the covariance relationships among many variables (Johnson and Wichern 2008). Among many interesting variables, some can be grouped according to their relevance, so that they represent a potential factor. That is, all variables in a particular group are highly correlated between them but have a relatively small correlation with variables in different groups (Johnson and Wichern 2008). In this study, principal component solutions were used in factor analysis to perform parameter estimation. In PCA, a varimax rotation was applied to simplify the interpretation of the factors. Varimax represents the orthogonal rotation standard, which maximizes the variance of the square factor load as much as possible, with the intention of identifying groups of large or negligible coefficients in any column of the factor load matrix.

OFP and SOAP of VOCs

Carter (1994) proposed MIR method is compared each kind of VOC species of ozone formation potential (OFP) good indicator, and by the type definition:

$$\text{OFP}(i) = \text{conc}(i) \times \text{MIR}_{\text{coefficient}}(i) \quad (1)$$

where $\text{OFP}(i)$ is defined as the ozone formation potential of a single hydrocarbon i , and the $\text{MIR}_{\text{coefficient}}(i)$ is the maximum incremental reaction coefficient of compound i , as defined by Carter (2010) (Table 1).

Secondary organic aerosol formation potential (SOAP) multiplied by the corresponding SOAP factor by VOC concentration to calculate, as shown in Eq. (2), as follows:

$$\text{SOAP}(i) = \text{conc}(i) \times \text{SOAP}_{\text{coefficient}}(i) \quad (2)$$

where $\text{SOAP}(i)$ is defined as a single hydrocarbon i potential of SOA, and $\text{SOAP}_{\text{coefficient}}(i)$ each VOC (i) is relative to the toluene to form the potential of SOA, as proposed by Derwent et al. (2010) (Table 1).

Conditional Probability Functions

Conditional probability function (CPF) analysis was performed to determine the most likely location of the resolution sources resolved by PCA (Ashbaugh et al. 1985; Xie and Berkowitz 2006; Leuchner and Rappenglück 2010; Mo et al. 2017). Therefore, from the source analysis source contribution to the results obtained with the receptor combination location measurement of wind speed and wind direction data. The CPF is the fraction of the total sample in the fan area $\Delta\theta$, which has a higher source contribution than the threshold standard value. The equation used for the analysis is as follows:

$$\text{CPF} = \frac{m_{\Delta\theta}}{n_{\Delta\theta}} \quad (3)$$

where $m_{\Delta\theta}$ is the number of samples in a particular fan zone ($\Delta\theta = 22.5^\circ$), it is more than the 75th percentile scores from each source contributions, and $n_{\Delta\theta}$ is the total number of observations in the same wind sector. This is to ensure that only the high contribution is used to determine the source directionality. If the sample in the sector is insufficient or the wind speed is less than 1 m/s, the sector is discarded. For the CPF analysis of a PCA factor, the concentration refers to the value in the source contribution time series, and the high values also refer to the high values in that time series. A plot in polar coordinates, with the radial distance defined by the magnitude of CPF and the angle defined by the associated wind direction, visually illustrates the fraction of samples coming from a given direction that have “high” values and point to regions associated with these high values. Wind directions for this part of the analysis were binned into 16 sectors of 22.5° per sector. This approach can improve the accuracy of source identification, especially for a major stationary source.

Results and Discussion

General Characteristics of VOCs

Table 1 gives the statistical results of VOC concentrations during observations in Nanjing and Suzhou. It can be seen from the table that the average concentration of atmospheric VOCs in Nanjing is 34.6 ± 5.8 ppbv, while that in Suzhou is 28.1 ± 5.6 ppbv. The concentration of VOCs in Nanjing is 23.1% higher than that in Suzhou. The top five VOCs in the concentration categories are ethane, ethylene, propane, toluene, and *n*-butane in Nanjing, whereas those in Suzhou are ethane, propane, ethylene, toluene, and *n*-butane. The top five components of VOCs in the two sites are the same, but the order of concentration is different. Ethane, with the highest VOC concentration in the two sites is 6657 ± 92 pptv (Nanjing) and 3658 ± 285 pptv (Suzhou), and the concentration in Nanjing is 1.8 times that of Suzhou.

Because certain pollutants are emitted from specific source of pollution, this type of pollutant can act as a “tracer” of the source of the pollution. For example, CO can be used as a tracer for emissions from combustion processes, and the correlation between VOCs and CO can be examined to determine whether these components are emissions from the combustion process. Table 2 gives the correlation between CO and certain VOC components. It can be seen that in Nanjing and Suzhou, CO and most VOC components (except for isoprene, 1-butene, and benzene) have a good correlation ($R^2 = 0.40\text{--}0.61$), especially alkanes, such

Table 1 Mean, minimum (min), maximum (max), and standard error (SE) of the VOCs measured in Nanjing and Suzhou

	MIR ^a	SOAP ^b	Nanjing (<i>n</i> = 659)				Suzhou (<i>n</i> = 501)			
			Mean	Min	Max	SE	Mean	Min	Max	SE
CO (ppbv)	–	–	1086	290	9054	34	1076	277	3529	18
Ethane	0.28	0.1	6657	40	14,560	92	3658	10	15,520	285
Propane	0.49	0	4263	19	10,380	69	3454	10	13,900	152
<i>i</i> -Butane	1.23	0	1437	14	4160	28	1126	10	7240	63
<i>n</i> -Butane	1.15	0.3	2014	10	6070	40	1538	10	10,370	89
<i>i</i> -Pentane	0.93	0.2	1028	11	3920	24	749	10	6740	64
<i>n</i> -Pentane	0.88	0.3	741	12	3070	18	974	10	5010	50
Cyclopentane	0.09	0	98	11	380	23	116	10	2960	14
Methylcyclopentane	2.19	0	123	18	500	4	13	10	90	1
2,3-Dimethylbutane	0.97	0	236	14	3380	17	1823	10	7530	81
2,2-Dimethylbutane	1.17	0	29	20	120	1	123	10	720	7
2-Methylpentane	1.5	0	234	14	870	6	459	10	2090	17
3-Methylpentane	1.8	0.2	217	20	960	8	321	10	1580	12
2,4-Dimethylpentane	1.55	0	60	10	200	2	72	10	520	3
2,3-Dimethylpentane	1.34	0.4	131	20	610	5	404	10	1830	15
<i>n</i> -Hexane	1.24	0.1	383	20	1860	12	939	10	4150	35
Cyclohexane	1.25	0	237	20	2010	12	414	10	1970	15
3-Methylhexane	1.61	0	38	20	160	1	284	10	1010	11
2,2,4-TMP	1.26	0	34	10	170	1	162	10	700	6
2,3,4-TMP	1.03	0	17	10	140	1	122	10	650	5
<i>n</i> -Heptane	1.07	0.1	145	10	580	4	233	10	1150	9
Methylcyclohexane	1.7	0	89	10	360	3	112	10	378	18
2-Methylheptane	1.07	0	215	10	2980	22	67	10	233	11
3-Methylheptane	1.24	0	27	10	140	1	82	10	460	3
<i>n</i> -Octane	0.9	0.8	228	10	1410	11	98	10	67	4
<i>n</i> -Nonane	0.78	1.9	21	10	190	1	26	10	590	1
<i>n</i> -Decane	0.68	7	76	30	420	2	39	10	880	2
<i>n</i> -Undecane	0.61	16.2	85	10	360	3	11	10	390	1
Ethene	9	1.3	5057	40	15,880	115	2615	10	3545	184
Propene	11.66	1.6	1683	15	11,350	67	641	10	2910	29
<i>trans</i> -2-Butene	15.16	4	74	11	320	11	221	10	1520	15
<i>cis</i> -2-Butene	14.24	3.6	68	13	290	2	572	10	2790	30
1-Butene	9.73	1.2	356	12	1230	9	29	10	190	1
<i>trans</i> -2-Pentene	10.56	3.1	29	13	140	1	59	10	540	3
<i>cis</i> -2-Pentene	10.38	3.1	20	15	80	1	112	10	650	5
1-Pentene	7.21	0	33	15	130	1	98	10	850	7
Isoprene	10.61	1.9	53	10	280	2	153	10	694	9
1-Hexene	4.4	0	23	20	70	1	11	10	34	5
Benzene	0.72	92.9	1980	20	12,290	75	1315	10	2770	17
Toluene	4.01	100	2409	10	11,760	89	2366	10	7850	63
Ethylbenzene	3.04	111.6	1622	10	7420	49	910	10	3050	34
<i>m,p</i> -xylene	7.8	84.5	572	10	2440	16	556	10	2160	21
<i>o</i> -xylene	7.64	95.5	399	10	1880	12	307	10	1720	11
Styrene	1.73	212.3	156	10	960	7	395	10	1700	15
<i>i</i> -Propylbenzene	2.52	95.5	33	10	160	1	19	10	270	1
<i>n</i> -Propylbenzene	2.03	95.5	47	10	190	1	55	10	900	3
<i>p</i> -Ethyltoluene	4.44	69.7	278	20	910	7	18	10	540	1
<i>m</i> -Ethyltoluene	7.39	100.6	54	20	290	2	32	10	600	1
<i>o</i> -Ethyltoluene	5.59	13.5	69	10	230	2	25	10	490	1

Table 1 (continued)

	MIR ^a	SOAP ^b	Nanjing (<i>n</i> = 659)				Suzhou (<i>n</i> = 501)			
			Mean	Min	Max	SE	Mean	Min	Max	SE
1,3,5-TMB	11.76	94.8	56	10	190	14	25	10	560	1
1,2,4-TMB	8.87	20.6	685	60	2740	14	92	10	970	5
1,2,3-TMB	11.97	43.9	44	10	130	1	21	10	640	1

Units are pptv, unless otherwise specified

TMP trimethylpentane, *TMB* trimethylbenzene

^aUnits are (g O₃ g⁻¹ VOC) (Carter 2010)

^bSOA formation expressed on a mass emitted basis as SOAP relative to toluene = 100 (Derwent et al. 2010)

Table 2 Coefficient of determination (R^2) calculated for selected VOCs with CO and toluene, in Nanjing and Suzhou

	CO		Toluene	
	Nanjing	Suzhou	Nanjing	Suzhou
CO (ppbv)	–	–	0.52	0.45
Ethane	0.52	0.31	0.36	0.42
Propane	0.48	0.30	0.49	0.40
<i>i</i> -Butane	0.46	0.56	0.59	0.60
<i>n</i> -Butane	0.43	0.43	0.48	0.52
<i>i</i> -Pentane	0.60	0.50	0.57	0.46
<i>n</i> -Pentane	0.61	0.10	0.57	0.30
<i>n</i> -Hexane	0.52	0.33	0.51	0.46
<i>n</i> -Heptane	0.59	0.50	0.71	0.50
<i>n</i> -Octane	0.41	0.35	0.51	0.29
2-Methylpentane	0.52	0.31	0.62	0.21
3-Methylpentane	0.53	0.34	0.60	0.23
Ethene	0.54	0.55	0.56	0.54
1-Butene	0.31	0.10	0.34	0.16
Isoprene	0.20	0.32	0.27	0.42
1-Pentene	0.40	0.45	0.37	0.52
Benzene	0.38	0.68	0.39	0.53
Toluene	0.52	0.45	–	–
Ethylbenzene	0.50	0.52	0.61	0.67
<i>m,p</i> -xylene	0.50	0.44	0.68	0.77
<i>o</i> -xylene	0.45	0.49	0.66	0.68

as propane and butane. Therefore, the source of combustion can be considered an important source of VOCs in Nanjing and Suzhou, which may be related to the increasing number of vehicles in Nanjing and Suzhou (An et al. 2014; Shao et al. 2016; Mo et al. 2017; Xu et al. 2017). Similar studies in other cities also have shown that motor vehicles are the main source of urban VOCs. For example, studies in both Guangzhou and Shanghai have shown that motor vehicles are an important source of VOCs (40–65%) (Barletta et al. 2008; Cai et al. 2010).

Many cities have studied the source of toluene, but toluene sources in urban areas are different. Ho et al. (2009) attributed the high toluene levels measured in Hong Kong

to evaporation of gasoline because of the high toluene content in gasoline supplied in Hong Kong. Cai et al. (2010) reported high levels of toluene in Shanghai, presumably emitted from industrial sources. Different sources of toluene in different cities reflect differences in fuel types and industrial structures between cities. In this paper, a correlation analysis of toluene and related VOCs in Nanjing and Suzhou was performed. The results (Table 2) show that there is a good correlation between toluene in Nanjing and most VOC components ($R^2 > 0.40$), especially for *n*-heptane ($R^2 = 0.71$), methylpentanes ($R^2 = 0.60–0.70$), butanes ($R^2 = 0.48–0.59$), pentanes ($R^2 = 0.57$), and the aromatic fraction ($R^2 = 0.61–0.68$). Research on atmospheric VOCs in Nanjing shows that most of these pollutants are related to industrial activities (An et al. 2014; Shao et al. 2016). Comparing the correlations between toluene and CO, as well as propane and pentane in the atmosphere of Suzhou, we find a weak correlation. In addition, the correlation between toluene and benzene increased from 0.39 in Nanjing to 0.53 in Suzhou. These results reflect that industrial activity also is an important source of toluene in the atmosphere of Nanjing.

Comparison of Rush Hour Periods and Non-rush Hour Periods

To compare the impact of traffic sources on VOCs in the two sites, Table 3 compares the composition and concentration of VOCs in Nanjing and Suzhou during rush hour (07:00–09:00) and non-rush hour (14:00–16:00). Most VOC concentrations in Nanjing and Suzhou are consistent. VOCs in both areas are higher in rush hour than in non-rush hour. The difference in concentration between Suzhou during rush hour and non-rush hour is higher than that in Nanjing.

During rush hour, Suzhou VOCs are mainly affected by traffic sources. In addition to the impact of traffic sources, Nanjing also receives industrial emissions, thus showing a higher concentration of Nanjing VOCs. During non-rush hour, the traffic flow of the two sites decreases to varying degrees, which leads to a decrease in the concentration of VOCs from traffic sources. This was evident in Suzhou. In addition to transportation sources, Nanjing

Table 3 VOCs concentrations of the major species in Nanjing and Suzhou during rush hour and non-rush hour (average \pm standard deviation, ppbv)

Rank	Nanjing				Suzhou			
	Rush hours		Non-rush hours		Rush hours		Non-rush hours	
	Species	Conc.	Species	Conc.	Species	Conc.	Species	Conc.
1	Ethane	7.02 \pm 2.45	Ethane	6.23 \pm 2.46	Ethane	4.87 \pm 8.24	Propane	3.25 \pm 3.18
2	Ethene	5.81 \pm 2.97	Ethene	4.23 \pm 2.83	Ethene	3.95 \pm 7.20	Ethane	2.47 \pm 4.30
3	Propane	4.67 \pm 1.81	Propane	3.90 \pm 1.87	Propane	3.04 \pm 2.89	Toluene	1.94 \pm 1.45
4	Toluene	2.89 \pm 2.25	Benzene	1.91 \pm 2.07	Toluene	2.44 \pm 1.59	2,3-Dimethylbutane	1.75 \pm 1.80
5	<i>n</i> -Butane	2.17 \pm 1.01	Toluene	1.75 \pm 1.72	2,3-Dimethylbutane	1.90 \pm 1.75	Ethene	1.43 \pm 1.72
6	Propene	2.15 \pm 2.05	<i>n</i> -Butane	1.74 \pm 0.92	<i>n</i> -Butane	1.61 \pm 2.09	Benzene	1.20 \pm 0.52
7	Benzene	1.71 \pm 1.45	Propene	1.62 \pm 1.97	Benzene	1.36 \pm 0.47	<i>n</i> -Butane	1.00 \pm 1.27
8	Ethylbenzene	1.70 \pm 1.29	Ethylbenzene	1.50 \pm 1.19	<i>i</i> -Butane	1.21 \pm 1.53	Ethylbenzene	0.87 \pm 0.64
9	<i>i</i> -Butane	1.58 \pm 0.75	<i>i</i> -Butane	1.24 \pm 0.75	<i>n</i> -Pentane	1.14 \pm 1.37	<i>n</i> -Hexane	0.86 \pm 0.64
10	<i>i</i> -Pentane	1.14 \pm 0.57	<i>i</i> -Pentane	0.94 \pm 0.70	<i>i</i> -Pentane	0.97 \pm 1.89	<i>n</i> -Pentane	0.75 \pm 1.07
11	1,2,4- TMB	0.79 \pm 0.38	<i>n</i> -Pentane	0.66 \pm 0.51	<i>n</i> -Hexane	0.91 \pm 0.90	<i>i</i> -Butane	0.68 \pm 0.95
12	<i>n</i> -Pentane	0.78 \pm 0.45	1,2,4- TMB	0.65 \pm 0.44	Propene	0.83 \pm 0.87	<i>i</i> -Pentane	0.59 \pm 0.32
13	<i>m,p</i> -xylene	0.61 \pm 0.43	<i>m,p</i> -xylene	0.49 \pm 0.35	Ethylbenzene	0.80 \pm 0.62	<i>m,p</i> -xylene	0.47 \pm 0.41
14	<i>n</i> -Hexane	0.43 \pm 0.30	<i>o</i> -xylene	0.37 \pm 0.25	<i>cis</i> -2-Butene	0.67 \pm 0.76	<i>cis</i> -2-Butene	0.46 \pm 0.72
15	<i>o</i> -xylene	0.41 \pm 0.31	2-Methylheptane	0.33 \pm 0.66	<i>m,p</i> -xylene	0.53 \pm 0.42	2,3-Dimethylpentane	0.43 \pm 0.46
16	1-Butene	0.39 \pm 0.22	<i>n</i> -Hexane	0.32 \pm 0.28	2-Methylpentane	0.51 \pm 0.49	Cyclohexane	0.42 \pm 0.50
17	<i>p</i> -Ethyltoluene	0.29 \pm 0.17	1-Butene	0.31 \pm 0.24	Cyclohexane	0.48 \pm 0.47	2-Methylpentane	0.40 \pm 0.53
18	3-Methylpentane	0.26 \pm 0.20	2,3-Dimethylbutane	0.28 \pm 0.54	Styrene	0.41 \pm 0.37	Propene	0.39 \pm 0.37
19	2-Methylpentane	0.25 \pm 0.15	<i>p</i> -Ethyltoluene	0.26 \pm 0.18	2,3-Dimethylpentane	0.38 \pm 0.43	Styrene	0.32 \pm 0.39
20	Cyclohexane	0.25 \pm 0.31	Cyclohexane	0.26 \pm 0.41	3-Methylpentane	0.32 \pm 0.30	3-Methylpentane	0.29 \pm 0.23
Total VOC		35.28		28.98		28.33		19.98
Alkanes		18.54 (53%)		15.88 (55%)		17.33 (61%)		12.90 (65%)
Alkenes		8.35 (23%)		6.17 (21%)		5.46 (19%)		2.28 (11%)
Aromatics		8.39 (24%)		6.94 (24%)		5.55 (20%)		4.80 (24%)

TMB trimethylbenzene

also has continuous emissions from industrial sources. The decline in VOC concentrations is not as dramatic as in Suzhou.

Alkanes have a large contribution ratio (53–65%) to VOCs in both places. This is mainly because alkanes are the main process emissions of typical urban atmospheric fuel combustion and volatilization (An et al. 2014; Mo et al. 2017), including ethane, ethane, propane, toluene, and benzene. These components are typical products of motor vehicle combustion and fuel evaporation (Shao et al. 2016). This result is consistent with the large proportion of ethane, ethane, propane, toluene, and benzene of Chengdu (Mo et al. 2017) and Zhengzhou (Li et al. 2019).

VOC Characteristic Ratios

Due to daytime photochemical reactions, the more active VOCs species will be reduced in concentration during daytime due to photochemical degradation, while the lower activity components will be relatively increased, due to low photochemical consumption. For example, in an atmospheric environment in which the 12 h mean concentration of the OH free radical is 0.06 ppbv (Barletta et al. 2005), the residence times for benzene, toluene, ethylbenzene, and *m,p*-xylene are 12.5 days, 2 days, 23 h, and 7.8 h, respectively. To compare the degree of aging of VOCs affecting these two locations and the transport distance of contaminants, the activity differences of some typical VOCs often

are used (Barletta et al. 2005). Therefore, the characteristic ratios of toluene/benzene (T/B), m,p -xylene/benzene (X/B), and m,p -xylene/ethylbenzene (X/E) were analyzed and compared with values from other countries (Table 4). Relevant research indicates that the T/B ratio is close to 1, indicating that it is mainly affected by traffic sources. In addition, the closer to the source of traffic, the greater the T/B ratio (Gentner et al. 2009). A low X/B ratio means that the air mass has undergone an aging process. An increase in the X/E ratio indicates an increase in the intensity of photochemical reactions, whereas a decrease in the ratio indicates the aging of the gas mass (Guo et al. 2011).

Comparing the results in Table 4, it can be seen that the T/B ratios of Nanjing and Suzhou are between 0.92 and 1.79, which is small compared with the observations in other cities in Table 4 (0.52–12.21). The T/B ratio is approximately equal to 1, indicating that the traffic source has an impact on both sites. The T/B ratios of the two time periods in Suzhou are 1.79 and 1.62, respectively, but the change is not significant, while the Nanjing T/B ratio is 1.69 in rush hour, which is roughly equivalent to the Suzhou value. The T/B ratio during non-rush hour was 0.92, which was 45% less than that in rush hour. This may be related to the discharge of benzene from industrial sources in Nanjing during non-rush hour. The X/B ratios of both sites are lower (0.26–0.39), which is consistent with the observations of Wuhan (Lyu et al. 2016) and Kaohsiung (Liu et al. 2008), reflecting the

air mass transmission. The aging process has gone through the path. At the same time, the smaller X/E (0.33–0.66) also illustrates the effect of the aging process of the air mass through photochemical reactions on the two places. These ratios indicate that even at industrial sampling points, the transported traffic sources contribute to the concentration of VOCs.

Principal Component Factor Analysis

Factor analysis is widely used in the source analysis of VOCs, and many research efforts use PCA for source analysis (Nelson and Quigley 1983; Guo et al. 2004; Geng et al. 2009; An et al. 2014). Table 5 shows the results of PCA for the main components of VOCs in the two sites. The factor loading matrix is determined using the criteria of the eigenvalue > 1 (Kaiser Standard). Factor loading > 0.5 is listed in Table 5 to emphasize the significant contribution of this species.

The results of PCA of Nanjing show that Factor 1 contributes 44.8% of the source of VOCs, and the main contributing component is the group of alkanes emitted by motor vehicles. It therefore can be determined that Factor 1 is the motor vehicle source. Aromatic hydrocarbons are the major constituents of Factor 2, so it can be determined that industrial sources are the main source of Factor 2 (22.1%). The olefins in Factor 3 have a high loading, while

Table 4 VOCs characteristic ratios at urban and industrial areas in different cities

Sampling cities	Observation session	Characteristic ratio			References
		T/B	X/B	X/E	
Urban area					
Macau	November 1995	1.79	1.52	3.98	Wang et al. (2002)
Seoul	November 1997 to July 1999	5.49	1.73	3.29	Na et al. (2005)
Kaohsiung (rush hours)	July to October 2003	8.13	0.83	0.60	Liu et al. (2008)
Kaohsiung (non-rush hours)	July to October 2003	7.43	0.68	0.65	Liu et al. (2008)
Guangzhou	July to November 1996	1.09	0.88	4.68	Wang et al. (2002)
Chengdu	August to October 2016	2.25	0.89	1.02	Deng et al. (2019)
Wuhan	February 2013 to October 2014	1.17	0.24	0.80	Lyu et al. (2016)
Shanghai	January 2007 to March 2010	2.59	0.77	1.13	Cai et al. (2010)
Suzhou (rush hours)	January 2015	1.79	0.39	0.66	This study
Suzhou (non-rush hours)	January 2015	1.62	0.39	0.54	This study
Industrial area					
Edmonton	January 2003 to December 2009	1.28	0.60	NA	McCarthy et al. (2013)
Shizuoka	August to December 2010	4.92	0.86	0.94	Ohura et al. (2006)
Thane	March 1993 to May 1994	0.52	0.04	1.67	Mohan Rao et al. (1997)
Ulsan	June 1997	1.59	1.36	4.21	Na et al. (2001)
Kaohsiung (rush hours)	July to October 2003	12.21	0.69	0.38	Liu et al. (2008)
Kaohsiung (non-rush hours)	July to October 2003	6.48	0.44	0.43	Liu et al. (2008)
Nanjing (rush hours)	January 2015	1.69	0.36	0.36	This study
Nanjing (non-rush hours)	January 2015	0.92	0.26	0.33	This study

Table 5 PCA for VOCs concentrations of the major species in Nanjing and Suzhou

Compound	Nanjing			Suzhou			
	Factor 1	Factor 2	Factor 3	Factor 1	Factor 2	Factor 3	Factor 4
Ethane	0.827			0.875			
Ethene	0.550		0.574	0.862			
Propane	0.804			0.883			
Toluene		0.654				0.755	
<i>n</i> -Butane	0.870						0.794
Benzene		0.595		0.914			
Propene			0.699		0.654		
Ethylbenzene		0.743		0.879			
<i>i</i> -Butane	0.798						0.900
<i>i</i> -Pentane	0.782					0.767	
<i>n</i> -Pentane	0.823				0.697	0.548	
1,2,4-TMB		0.738		0.823			
<i>m,p</i> -xylene		0.756		0.810			
<i>o</i> -xylene		0.823					0.894
<i>n</i> -Hexane	0.648		0.579			0.816	
1-Butene			0.661		0.852		
<i>p</i> -Ethyltoluene		0.713					
Cyclohexane			0.938				0.822
2,3-Dimethylbutane			0.876		0.876		
2-Methylpentane	0.618			0.619			
3-Methylpentane	0.534		0.619	0.737			
Styrene			0.631		0.597		
2,3-Dimethylpentane	0.524	0.629					0.599
<i>cis</i> -2-Butene					0.605		
Eigenvalue	13.162	2.178	1.626	7.332	4.439	2.614	2.076
Proportion	44.8%	22.1%	8.7%	30.6%	23.5%	10.9%	9.3%
Cumulative	44.8%	66.9%	75.6%	30.6%	54.1%	64.9%	74.2%

TMB trimethylbenzene

Extraction method: principal component analysis

Rotation method: Varimax with Kaiser normalization. Rotation converged in nine iterations

Only factor loadings ≥ 0.500 listed. Only factors with eigenvalue ≥ 1 shown

olefins are generally used in nonpolar solvents (Kwon et al. 2007), so it can be determined that Factor 3 is from solvent usage (8.7%).

At the Suzhou site, the results show that Factor 1 explains 30.6% of the variability and were dominated by alkanes and aromatics. Previous studies have shown that motor vehicle emissions include not only C10–C15 VOCs but also C2–C9 VOCs, especially ethylene, which is a typical motor vehicle emission (Liu et al. 2008). Therefore, Factor 1, containing a large amount of C2–C9 VOCs is used as a vehicle emission source. *N*-pentane has a high load (0.697) in Factor 2. Additionally, the other components have a small load, and it can be confirmed that Factor 2, composed mainly of *N*-pentane, is a hydrocarbon volatilization source (23.5%). Toluene and cyclohexane in Factors 3 and 4 have large loads, respectively, and are the main emissions of industrial production and solvent usage; therefore, it can be determined

that Factor 3 is industrial production (10.9%) and Factor 4 is solvent usage (9.3%).

Evaluation of OFP and SOAP

Photochemical ozone formation and secondary organic aerosol formation are triggered by the reaction between the OH radical and VOCs, while the extent of ozone formation and secondary organic aerosol formation varies greatly among VOC reactions. Figure 2 shows the VOC concentrations, OFP, and SOAP at the two sites. The concentrations of VOCs in Nanjing and Suzhou showed similar composition characteristics: the highest proportion of VOCs was comprised of alkanes (54.3% in Nanjing and 61.1% in Suzhou), followed by aromatics (24.4% in Nanjing and 22.8% in Suzhou), and alkenes comprised the smallest proportion (21.3% in Nanjing and 16.1% in Suzhou). Comparing OFPs in various

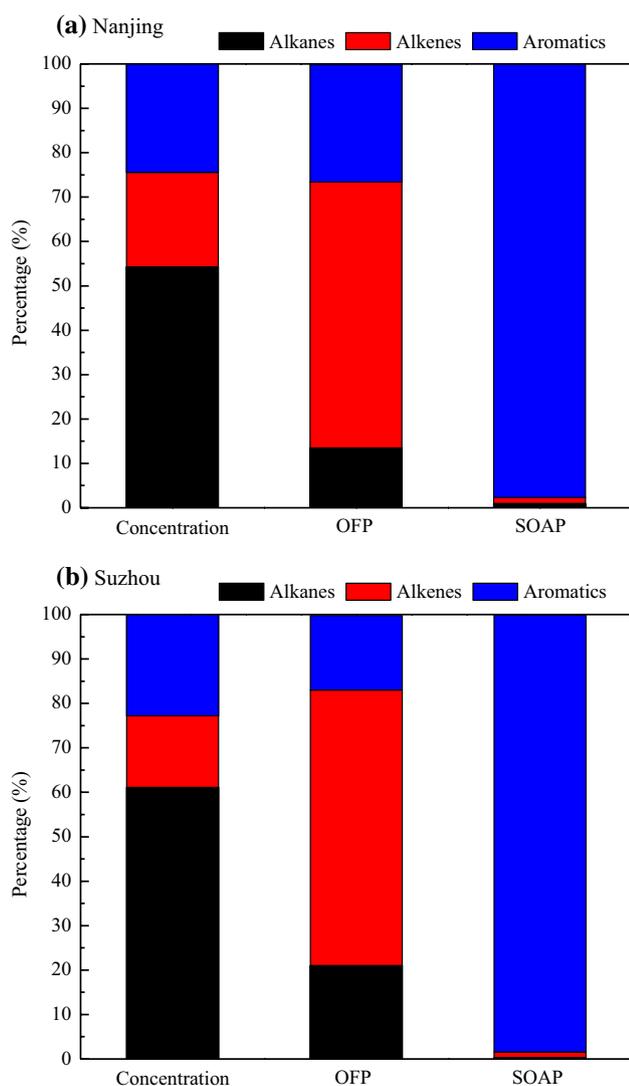


Fig. 2 Contribution of different groups of VOCs to concentration, OFP, and SOAP in **a** Nanjing and **b** Suzhou

categories of VOCs, in Nanjing, alkenes (59.9%) > aromatic (26.5%) > alkanes (13.5%), whereas Suzhou showed alkenes (62.0%) > alkanes (21.0%) > aromatics (16.8%). When comparing SOAP, both Nanjing and Suzhou showed an absolute majority of aromatic hydrocarbons, with 97.6% (Nanjing) and 98.3% (Suzhou). Although alkanes are the highest of the three types in terms of concentration, due to their low OH radical activity, they have a very low contribution to OFP. Although the concentration of alkenes is much lower than that of alkanes, they have a higher OFP contribution due to their higher OH radical activity. Aromatic hydrocarbons are the most important to SOAP in both Nanjing and Suzhou. Therefore, to control the formation of O₃ and SOA in the two sites, it is necessary to reduce the concentration of alkenes and aromatics, respectively.

Table 6 Top 10 VOCs species in terms of concentration, OFP, and SOAP in Nanjing and Suzhou

Rank	Nanjing				Suzhou			
	Compound	ConC (%)	OFP (%)	SOAP (%)	Compound	ConC (%)	OFP (%)	SOAP (%)
1	Ethane	19.2	38.0	30.0	Toluene	13.1	36.5	36.8
2	Ethene	14.5	16.1	23.1	Benzene	12.3	7.9	19.0
3	Propane	12.3	6.6	22.6	Ethylbenzene	9.3	7.3	15.8
4	Toluene	6.9	6.1	6.1	<i>m,p</i> -xylene	8.4	5.1	13.0
5	<i>n</i> -Butane	5.8	4.4	4.7	<i>o</i> -xylene	6.5	3.9	7.3
6	Benzene	5.7	4.3	4.2	<i>trans</i> -2-Butene	5.4	3.7	4.6
7	Propene	4.8	3.2	1.8	<i>m,p</i> -xylene	4.6	2.7	0.8
8	Ethylbenzene	4.6	2.6	0.8	<i>o</i> -Xylene	4.0	2.4	0.5
9	<i>i</i> -Butane	4.1	2.1	0.7	<i>n</i> -Butane	3.4	2.4	0.4
10	<i>i</i> -Pentane	2.9	2.1	0.6	<i>n</i> -Pentane	3.3	2.2	0.3

TMB trimethylbenzene, *DMB* dimethylbutane

Table 6 lists the top 10 VOCs species in concentration, OFP, and SOAP in Nanjing and Suzhou. In terms of concentration, the top ten components contributed are composed of 5 alkanes, 2 alkenes, 3 aromatics (Nanjing), and 6 alkanes, 1 alkene, and 3 aromatics (Suzhou), respectively. The higher first four components are ethane, ethene, propane, and toluene in both sites. For the OFP, ethene comprises the largest contribution ratio in both sites, accounting for 38.0% (Nanjing) and 36.5% (Suzhou), respectively. Propene, toluene, xylenes, and n-Butane have large contributions in both sites. Among the SOAP contributions, the benzene series has a significant contribution. In the top 10 in Nanjing, the benzene series accounted for 9 species, and in the top 10 in Suzhou, the benzene series accounted for 8 species. The cumulative contribution rate of BTEX accounted for 86.5% (Nanjing) and 83.4% (Suzhou) respectively, especially toluene, which contributed 30.0% (Nanjing) and 36.8% (Suzhou), respectively.

Direction Dependence of Sources

Using CPF analysis, certain physical locations of the different source categories can be identified. Figures 3 and 4 show the directional dependence of sources in Nanjing and Suzhou.

In Nanjing, the CPF plot as shown in Fig. 3a (for Factor 1) shows elevated VOCs from the ESE (53% on average) and SE (46% on average), suggesting a local motor vehicle sources. There are several major commuting routes, such as Provincial Expressway 205 and State Road 104 across the study area, where cars, trucks, and scooters, especially during rush hours, may significantly contribute to ambient VOCs. The CPF plot (Fig. 3b for Factor 2) shows high probabilities from ESE and SE (58% on average). The results apparently match the location of the local industrial sources: coal-fired power plant, Jiangbei High-tech zone, and Nanjing Iron & Steel industrial park. The solvent

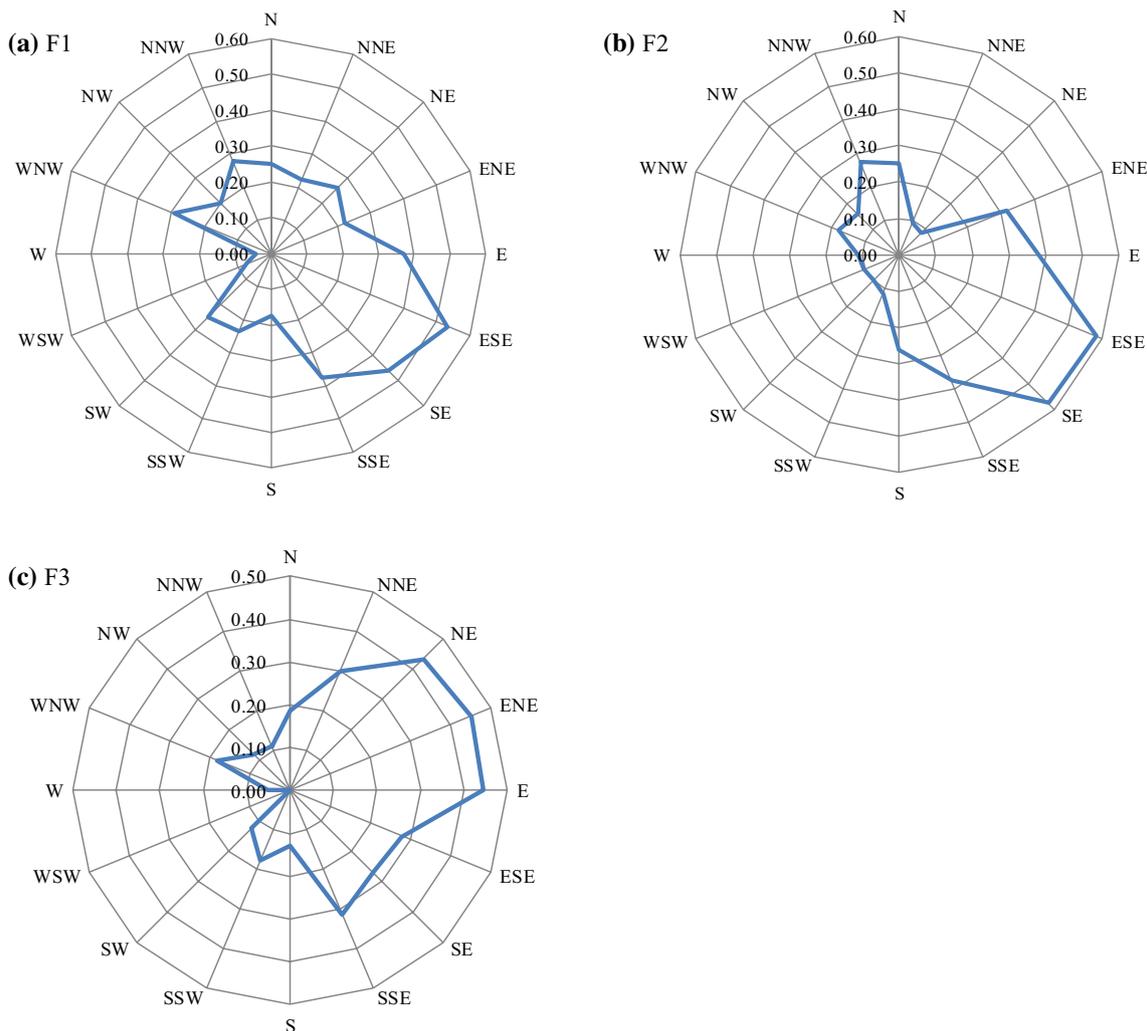


Fig. 3 CPF plots of VOCs sources in Nanjing

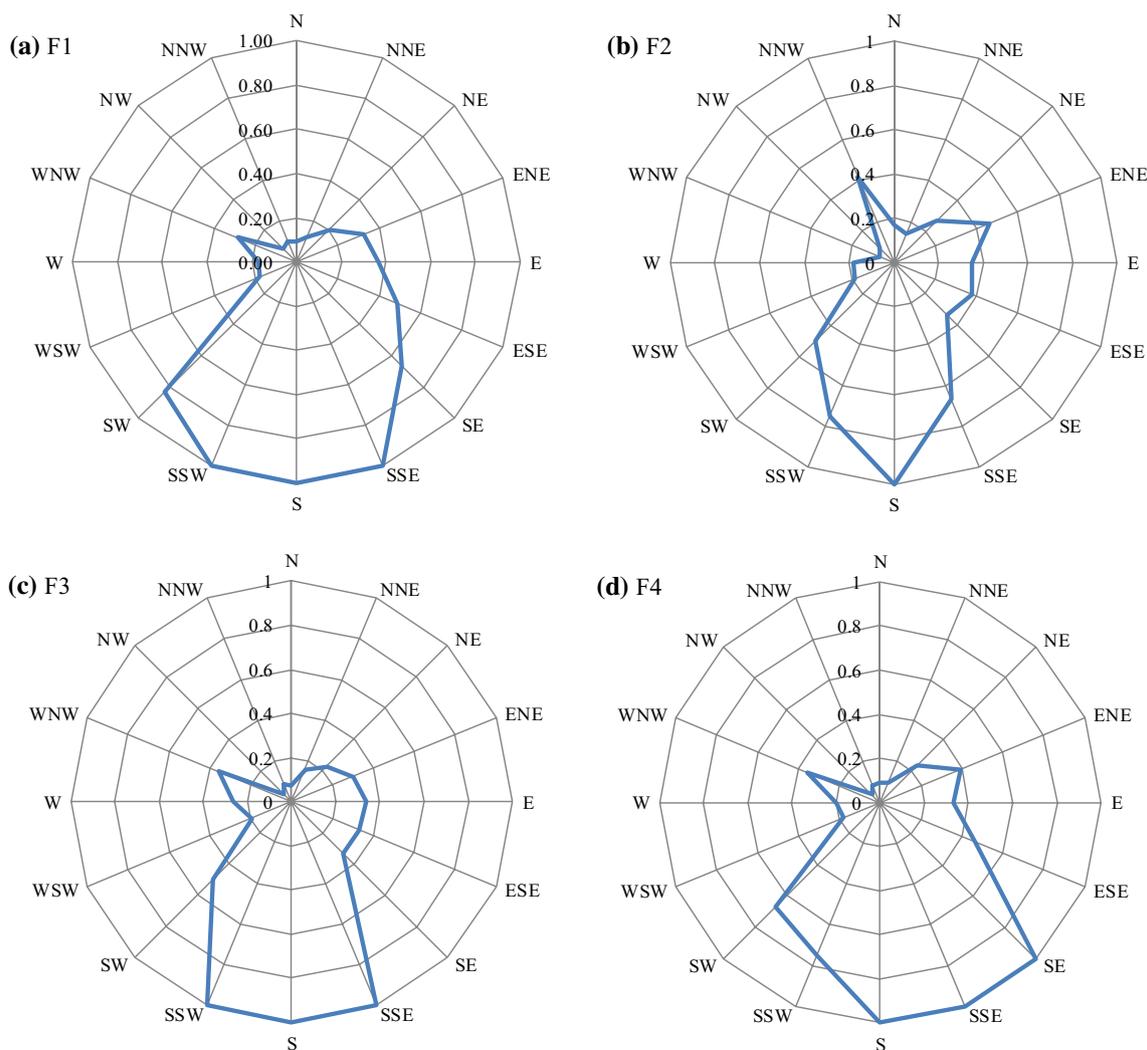


Fig. 4 CPF plots of VOCs sources in Suzhou

usage sources (Fig. 3c for Factor 3) displays a very different directional pattern, with highest values from the NE, ENE, and E (45% on average) directions, pointing towards the chemical industrial complex.

In Suzhou, high contributions of vehicular emissions (Fig. 4a for Factor 1) was more correlated with the SSW, S, and SSE directions, suggesting a local origin of traffic-related emission such as the second ring road of Suzhou and State Road 58 across area. The CPF plot (Fig. 4b for Factor 2) demonstrates a high probability from the S direction, suggesting a local origin of hydrocarbon evaporation in the surrounding gasoline stations, industrial pipes, and vehicle tanks. Industrial sources (Fig. 4c for Factor 3) were more correlated with the SSW, S, and SSE directions, pointing towards the Suzhou industrial park. The solvent usage sources (Fig. 4d for Factor 4) exhibit strong directional dependence from S, SSE, and SE directions, pointing to Suzhou chemical industrial complex.

Conclusions

VOCs were measured in the ambient air of the two major cities of Nanjing and Suzhou in the YRD region. VOCs concentration in Nanjing was 34.6 ± 5.8 ppbv, while that in Suzhou was 28.1 ± 5.6 ppbv. The concentrations of VOCs in Nanjing were 23.1% higher than that in Suzhou. The highest VOC measured in Nanjing and Suzhou was ethane, with concentrations of 6.6 ppbv and 3.6 ppbv, respectively. Relevant analysis shows that motor vehicle emissions in Suzhou were dominant, while industrial emissions in Nanjing also contributed to VOCs. In Nanjing, emissions of VOCs from industrial sources continued to increase and were more frequent during the non-rush hour than in Suzhou. Both sites may be severely affected by fuel combustion and evaporative emissions. The *T/B* ratio (0.92–1.79) for the two sites was observed to be relatively low in the other studied cities, indicating the impact of traffic emission sources.

According to PCA results, vehicle emissions and industrial sources accounted for more than 65% of the changes in the two sites, while major VOCs explained the importance of vehicle sources by matching VOCs fingerprints of vehicle exhaust and gasoline evaporation, even in nearby industrial areas. Using the OFP method, the alkenes was highest in both sites: 59.9% (Nanjing) and 62.0% (Suzhou), respectively. Ethene was the largest species in both sites. When comparing SOAP, both sites showed an absolute majority of aromatics: 97.6% (Nanjing) and 98.3% (Suzhou). To control the formation of O₃ and SOA in the two sites, it was necessary to reduce the concentration of alkenes and aromatics, respectively. CPF analysis showed that pollutants transported from urban traffic sources, industrial production sources, and solvent sources emitted by factories in the SE and NE directions have significant effects on the observation site in Nanjing. Ring/State roads and industrial parks in SE and S directions have significant impacts on the observation site in Suzhou.

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