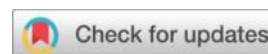




Investigation of VOCs in Autumn in the Lower Reaches of

the Yangtze River (Jiangsu Section)



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Abstract: There are many chemical enterprises, iron and steel enterprises, and coal-fired power plants along the Yangtze River, which are considered to be major sources of air pollution. For the first time, a mobile ship-based monitoring platform was used to continuously track and monitor atmospheric volatile organic compounds (VOCs) over the Jiangsu section of the Yangtze River during a period of similar and stable meteorological conditions. The monitoring results showed that the average concentration of TVOCs in the western section was $15.24 \mu\text{g}/\text{m}^3$, and the average concentration in the eastern section was $58.47 \mu\text{g}/\text{m}^3$. The pollution characteristics of VOCs were generally higher in the eastern section and lower in the western section, which was generally consistent with the distribution of chemical industry agglomeration areas along the river. The sources were mainly divided into four types: fossil fuel combustion, ports and terminals, shipbuilding industry, and solvent use, with distinct characteristics in each polluted section. This preliminary study is of great significance for regional joint pollution prevention and source tracing.

Keywords: Yangtze River; mobile monitoring; VOCs; SOAP

As a strong economic province in eastern China, Jiangsu Province has a strong demand for logistics and transportation. Convenient and low-cost shipping has made it the first choice for many factories to transport industrial products over long distances. The Yangtze River section in Jiangsu carries 70% of the total cargo traffic along the Yangtze River, with an annual transportation volume of more than 1.6 billion tons. There are many industrial concentrated areas distributed along the river, and it has trade contacts with ports in more than 100 countries and regions in the world. On the busy waterway, the impact of industrial parks along the Yangtze River and ship navigation on atmospheric environment quality cannot be ignored.

The emission inventory of anthropogenic air pollutants and Volatile Organic Compounds (VOCs) in the Yangtze River Delta compiled in 2011 shows that Suzhou, Wuxi, and Nanjing were in the top three for particulate matter and VOCs emissions. Power plants were the main sources of particulate matter emissions, while steel manufacturing, oil refineries, and paint accounted for the main part of VOCs sources^[1]. On the basis of the "Air Pollution Prevention and Control Action Plan" issued in 2013, The State Council issued the "Three-year Action Plan to Win the Blue Sky Defense War" in 2018, which put forward higher requirements for source control and precise measures against air pollution. Although the degree of primary PM_{2.5} pollution in the Yangtze River Delta region has decreased significantly, the pressure of secondary PM_{2.5} pollution and

VOCs emission reduction is still large^[2,3], At the same time, with the development of numerical models and the improvement of observation instrument performance^[4–8], the understanding of air pollution is also deepening^[9], Pollution control strategies have also evolved from single pollutant emission reduction in a single city to multi-dimensional pollutant emission collaborative control in regional urban agglomerations^[10–12].

The secondary organic aerosol generation potential of VOCs emitted by ships is greater than that of diesel and gasoline vehicles^[13], The "Ship Air Pollutant Emission Control Area Implementation Plan" issued by the National Ministry of Transport was implemented at the end of 2018, but some studies have found that after the replacement of fuel standards, VOCs emissions increased by about 14 times, and the components also changed greatly^[14]. In this context, research on the air pollution status of the Yangtze River waterway, as a spatial area distinct from the urban atmospheric background environment, is of great significance for regional joint prevention and control.

At present, there are few studies on air quality in the air corridor of the Yangtze River, which restricts the formulation and implementation of cross-regional joint prevention and control policies. This paper takes the Jiangsu section of the Yangtze River as the research object. The VOCs content and particulate matter concentration over the river section were monitored by a flight mass spectrometer. Combined with environmental monitoring data and meteorological data, the horizontal distribution and vertical structure of aerosols, and the concentration, components, and spatial distribution of VOCs were analyzed, to conduct a baseline survey of the air pollution status over the Yangtze River. This study provides a scientific basis for subsequent research and the formulation of joint prevention and control strategies for air pollution.

1 Materials and Methods

1.1 Mobile Monitoring Area and Time

This study started from Nanjing on October 9 and sailed 250 kilometers (about 12 hours) to Nantong. On October 10, the monitoring vessel sailed from Nantong along the river to near Taicang and then returned. It returned to Nanjing from Nantong on October 11. This mobile monitoring campaign passed through 20 industrial concentrated areas. The weather was fine with a force 3 southeast wind; details are shown in Figure 1. The monitoring schedule is detailed in Table 1.

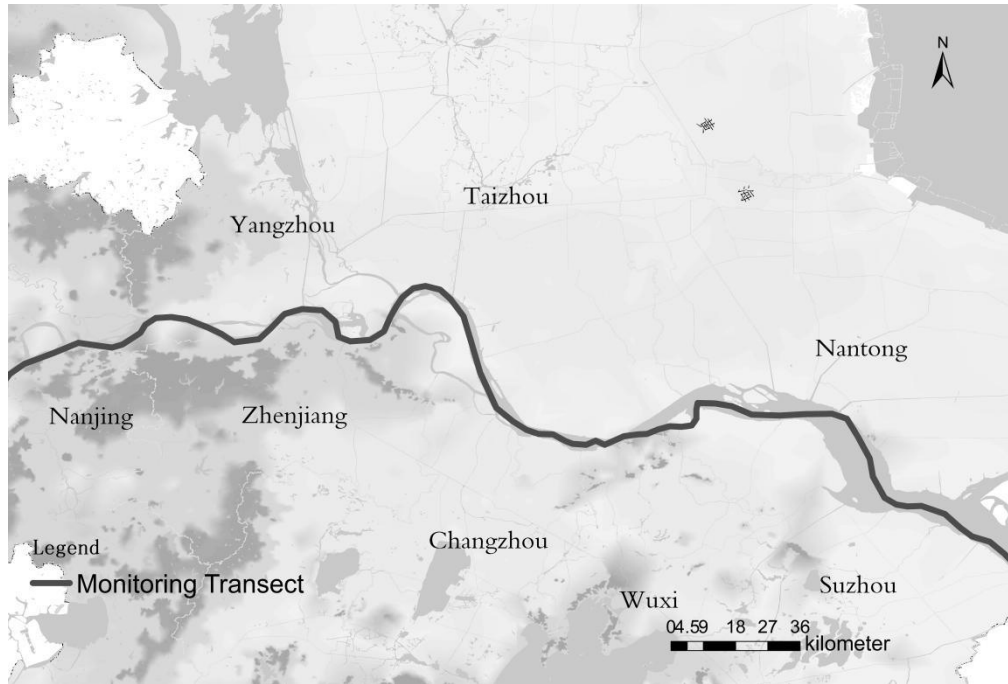


Fig. 1 Route of the mobile monitoring campaign during October 9-11

Table 1 Schedule of monitoring campaign

No.	Monitoring Time	Route
1	October 9, 9:56-17:18	Nanjing - Nantong
2	October 10, 9:35-12:04	Nantong - Taicang
3	October 10, 12:04-14:17	Taicang - Nantong
4	October 11, 9:43-15:42	Nantong - Yangzhou
5	October 11, 15:43-17:19	Yangzhou - Nanjing

1.2 Observation Equipment and Data

This study carried out monitoring activities on the river surface from Nanjing to Nantong, using a combination of mobile monitoring and fixed-point monitoring modes. Data from the mobile monitoring were analyzed for the period between 7:00~17:00 to avoid the cumulative effects of nighttime pollution. Data from two round trips were compared and verified. Three VOCs instruments were used for observation.

Table2 Schedule of monitoring

Instrument Model	Manufacturer	Monitoring Items	Power & Dimensions	Key Performance Parameters
SPIMS-2000	Hexin, Guangzhou	VOCs	Power: ~350 W Dimensions: 658(W)× 446(D)×580(H) mm	Principle: Time-of-flight mass spectrometry Detection limit: min. 0.1 ppb Detection components: PAMS, TO14, organic sulfur, etc.
HAPLINE M8800	BCT, Beijing	VOCs	Power: 50 W Size: 50×54×40 cm	Principle: GC-MS Can be used as mobile GC-MS in normal mode Detection limit: min. ppt level Detection component: full range VOCs (MW<300)
Vocus Scout CI-ToF	Zhongke Sanqing	VOCs	Power: 600 W Dimensions: 48×62×113 cm	Principle: Chemical ionization time-of-flight mass spectrometry Detection limit: min. 0.005 ppb Test components: aldehydes, ketones, esters, olefins,

1.3 Positive Matrix Factorization (PMF)

Positive Matrix Factorization (PMF) can identify the different characteristics of each factor, thus distinguishing between sources. Its powerful function is widely used in source analysis of particulate matter and volatile organic compounds^[15–17]. The specific principle is to decompose the input matrix X into a matrix G representing the contribution of pollutants to the source and a matrix F representing the composition of pollution sources. The formula is as follows:

$$X = G \times F + E \quad (1)$$

where E is the residual matrix.

The PMF version 5.0 developed by the US Environmental Protection Agency (EPA) was used in this study. According to the user manual, a concentration file and an uncertainty file are required. The uncertainty (Unc.) of different species should be calculated based on the method detection limit (MDL) and an error fraction (EF). MDL data were taken from the literature^[18], The Unc is calculated as follows:

$$Unc_i = \sqrt{(EF \times c_i)^2 + (0.5 \times MDL_i)^2} \quad (2)$$

where c_i is the concentration of the i th species, EF (error fraction) is the percentage of the estimated standard deviation, and MDL_i is the method detection limit of the i th component. Concentrations reported as 0 were replaced by 1/2 MDL, and their uncertainty was set to 5/6 MDL.

1.4 Secondary Organic Aerosol Potential (SOAP)

Toluene is widely recognized as one of the important anthropogenic emission sources of secondary organic aerosols^[19]. The equivalent toluene mass method reflects the tendency of each organic compound to form secondary organic aerosol (SOA) on an equal mass basis relative to toluene. This tendency is expressed as the Secondary Organic Aerosol Potential (SOAP). The SOAP_{*i*} for each organic compound is calculated as:

$$SOAP_i = \frac{\text{Incremental SOA concentration generated by unit concentration of VOC}_i}{\text{Incremental SOA concentration generated by unit concentration of toluene}} \times 100\% \quad (3)$$

SOAP_{*i*} data from the literature were used in this study^[20]. The SOA generation potential of different VOCs sources can be obtained by multiplying SOAP_{*i*} by the concentration of different VOCs components, calculated as follows:

$$SOAP = \sum c_i \times SOAP_i \quad (4)$$

2 Results and Discussion

2.1 VOCs Concentration Variation and Composition Characteristics

The monitoring results showed that the VOCs concentration in the Jiangsu section of the Yangtze River was lower in the upstream (west) and higher in the downstream (east), which was generally consistent with the distribution of chemical industry agglomeration areas along the river (see Figure 2). On the outbound journey, the average TVOCs concentration in the Nanjing-Jiangyin section was 26.92 $\mu\text{g}/\text{m}^3$, and the average concentration in the Jiangyin-Taicang section was 74.26 $\mu\text{g}/\text{m}^3$. On the return journey, the average concentration in the Taicang-Jiangyin section was 42.67 $\mu\text{g}/\text{m}^3$, and the average concentration in the Jiangyin-Nanjing section was 3.55 $\mu\text{g}/\text{m}^3$. The trend and range of VOCs concentration were generally consistent for both journeys. During the monitoring period, the meteorology was characterized by a southeast wind (force 3-4), high humidity, and stable pollutant diffusion.

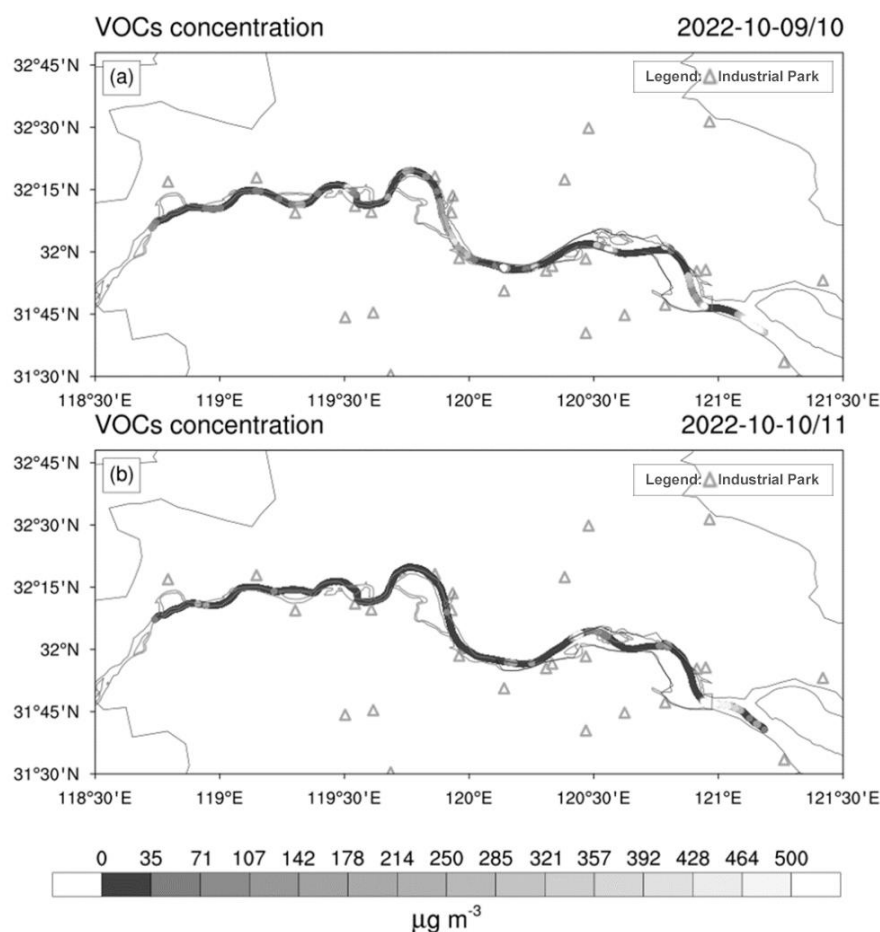


Fig. 2 Mobile monitoring profile of VOCs. (a) TVOCs concentration on October 9 (full day) and the morning of October 10; (b) TVOCs concentration on the afternoon of October 10 and October 11 (full day).

To further investigate the dominant species in high VOCs concentration areas, six time periods or points where VOCs concentrations exceeded $500 \mu\text{g}/\text{m}^3$ during the monitoring were selected chronologically (four on the outbound and two on the return). The top five dominant species by mass concentration percentage are listed in Table 3. The dominant VOCs species at the first three high points on the outbound journey were halogenated hydrocarbons, with trichloroethane and 1,1,2,2-tetrachloroethane together accounting for over 30%. The VOCs content in the Changshu section (Suzhou) varied little, with diethyl benzene being the highest. On the return journey, two high points were measured in the Jingjiang section (Taizhou). Pentene accounted for 40% at the first high point, and ethylbenzene accounted for 60% at the second high point.

Table 3 Dominant species in regions with high VOCs concentrations

Location	High VOCs Concentration ($\mu\text{g}/\text{m}^3$)	VOCs Dominant Species	Concentration ($\mu\text{g}/\text{m}^3$)	Proportion (%)	Dominant Species Category
Jiangyin Section, Wuxi	904.3	Trichloroethane	205.8	22.76	Halogenated
		1,1,2,2-Tetrachloroethane	110.97	12.27	hydrocarbons
		Diethyl benzene	92.08	10.18	
		Tetrachloroethylene	74.47	8.24	
		Trimethylbenzene	68.47	7.57	
Zhangjiagang Section,	860.9	Trichloroethane	198.58	23.07	Halogenated
		1,1,2,2-Tetrachloroethane	98.41	11.43	hydrocarbons

Suzhou		Tetrachloroethylene	66.19	7.69	
		Diethyl benzene	86.50	10.05	
		Trimethylbenzene	60.97	7.08	
Taicang	1201	Trichloroethane	272.91	22.72	Halogenated
Section,		1,1,2,2-Tetrachloroethane	145.87	12.15	hydrocarbons,
Suzhou		Trimethylbenzene	87.96	7.32	alkanes, aromatic
		Tetrachloroethylene	84.81	7.06	hydrocarbon
		Nonane	78.93	6.57	
Changshu	720.3	Diethyl benzene	71.43	9.92	Halogenated
Section,		1,1-Dichloroethylene	56.83	7.89	hydrocarbons,
Suzhou		Trimethyl phosphate	48.00	6.66	alkanes
		Nonane	47.46	6.59	
		n-Decane	47.33	6.57	
Jingjiang	1285	Pentene	516.45	40.19	Alkanes, alkenes
Section,		Hexene/Methylcyclopentane	182.67	14.22	
Taizhou		n-Octane	148.19	11.53	
		Butene	127.62	9.93	
		Isoprene	78.74	6.13	
Jingjiang	573.89	Ethylbenzene	349.19	60.85	Aromatic
Section,		Diethyl benzene	93.76	16.34	hydrocarbons,
Taizhou		Trimethylbenzene	72.46	12.63	alkanes
		Hexanethiol	9.83	1.71	
		n-Decane	7.10	1.24	

2.2 VOCs Source Analysis

Since, in addition to combustion processes, coatings and solvents are also important sources of toluene, the toluene/benzene ratio (T/B) is often used to identify different emission sources. Traffic emissions typically have a higher T/B ratio (usually >2) due to higher toluene content in gasoline combustion and exhaust^[21]. The T/B ratio in this mobile monitoring experiment was 2.017 (volume ratio), indicating significant contributions from ship fuel combustion emissions, although emissions from ports and terminals, shipbuilding industry, and coal-fired power plants also should not be overlooked.

The PMF model was used for VOCs source analysis. Based on existing research and the actual monitoring conditions, 15 VOCs components with good data quality and source specificity were selected, totaling 4592 data points. The PMF model uses the Q value to evaluate the results. The closer the Q_{true} and Q_{robust} values are, the better the fit between the model calculations and observations. By testing different factor numbers, the results showed that selecting 4 factors yielded Q_{true}/Q_{robust} = 1.06, indicating strong model interpretability. Accordingly, four emission sources were identified (see Figure 3).

In Factor 1, the major contributing organic compounds were 1,3-butadiene, benzene, toluene, styrene, and n-octane, primarily olefins and aromatic hydrocarbons. 1,3-Butadiene is a typical engine combustion tracer, n-octane is a fuel volatile, and styrene is a major component of VOCs emissions from coal-fired power plants^[22]. Thus, Factor 1 was identified as fossil fuel combustion emissions from thermal power plants and ships in the Yangtze River section.

In Factor 2, the highest contributing components were methylcyclohexane, nonane, diethylbenzene, n-decane, and undecane, predominantly alkanes. Xiao^[13] observed that methylcyclohexane is a major emission when ships switch to auxiliary engines after berthing. Additionally, this factor includes common combustion emissions like nonane, n-decane, and undecane. Since toluene and tetrachloroethylene also contribute, industrial coating emissions can be considered part of this factor. Therefore, Factor 2 was identified as ship berthing emissions at ports and terminals and industrial emissions.

In Factor 3, trichloroethane and tetrachloroethylene are common metal cleaning agents in industrial production, and trimethylbenzene and diethylbenzene are used as solvents in coatings and paints. This factor was attributed to shipbuilding industry emissions.

In Factor 4, ethylbenzene is used as a paint solvent and pharmaceutical raw material. This factor was determined to be solvent use.

In summary, fossil fuel combustion emissions from thermal power plants and ships, ship berthing and industrial emissions at ports and terminals, shipbuilding industry emissions, and solvent use are all significant contributors to air pollution over the Yangtze River.

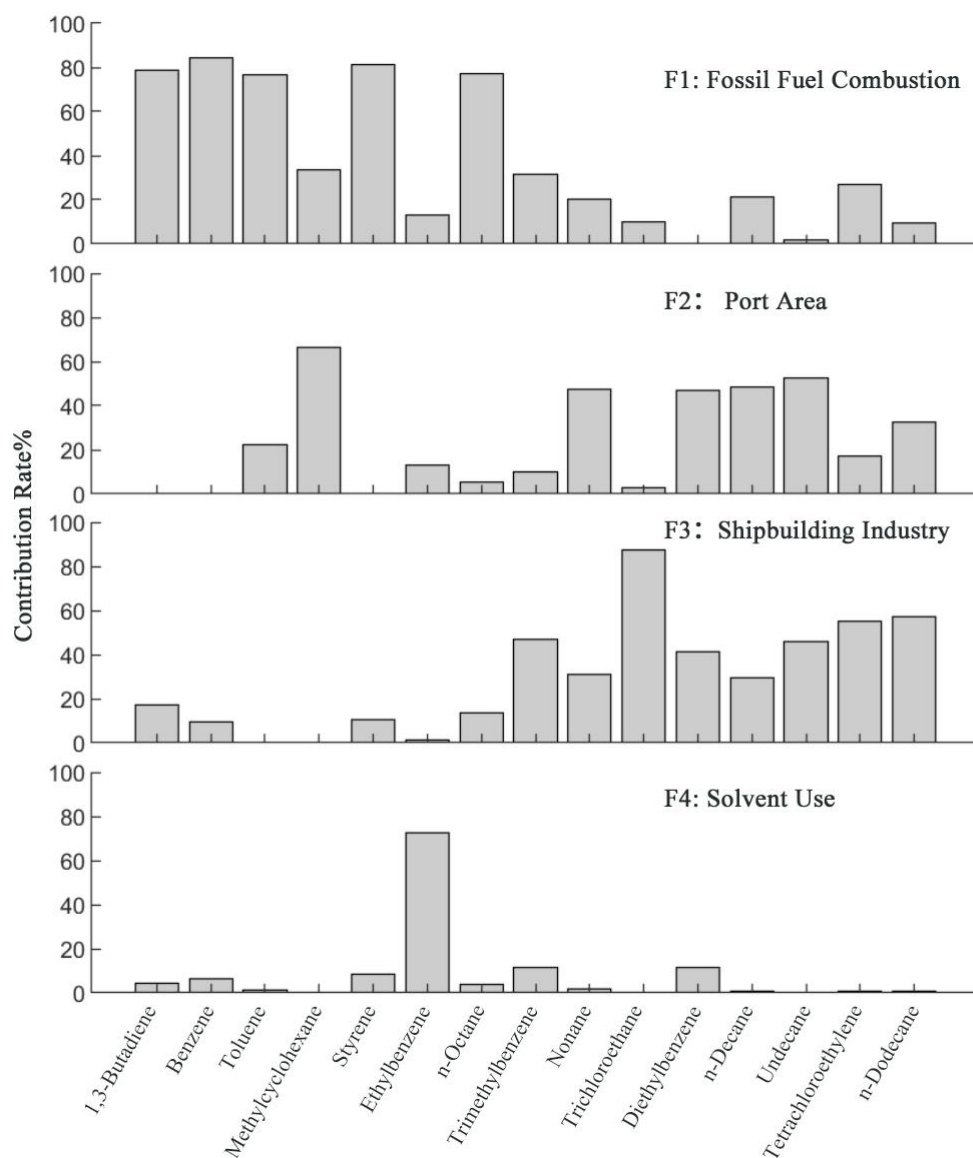


Fig. 3 Source apportionment of VOCs

2.3 SOA Generation Potential of Different VOCs Sources

VOCs are precursors of secondary organic aerosols and important precursors of PM_{2.5}. Based on the four VOCs sources classified in section 2.2, the SOA generation potential (SOAP) was used to further investigate the impact of different emission sources on SOA formation over the Yangtze River.

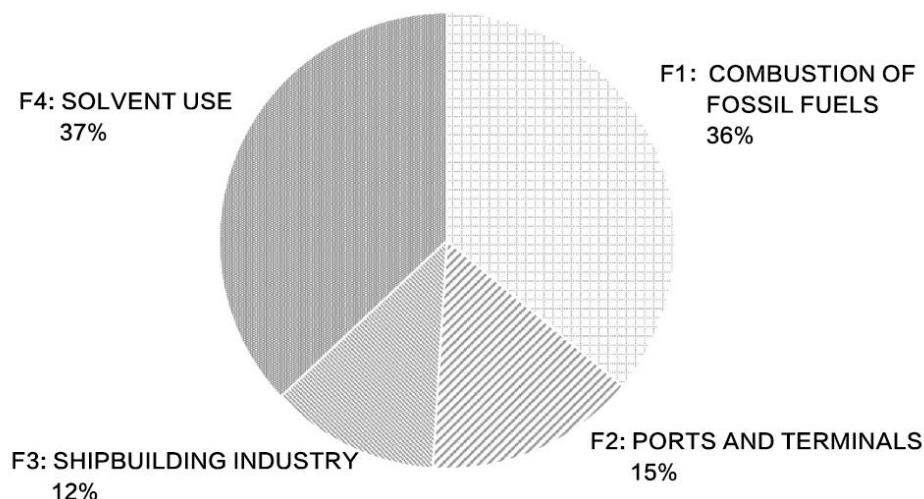


Fig. 4 Contribution of each factor to the total SOAP

Figure 4 shows the SOAP contributions of the four factors. Fossil fuel combustion is a major contributor to SOA formation, accounting for 36.1% and 37.1% of the total, respectively. Emissions from ports and terminals and the shipbuilding industry were relatively lower, accounting for 15.0% and 11.8%, respectively. Table 4 shows the concentration and SOAP of components within the four factors. The total SOAP reached 1645.95. Although the alkane concentration was higher than the aromatic hydrocarbon concentration in the fossil fuel combustion and port terminal factors, aromatic hydrocarbons were the absolute dominant contributors to SOAP generation from all VOCs sources, exceeding 22% contribution in each source category. Alkanes were important precursors for SOAP generation in the port terminal and shipbuilding industry factors, contributing 18% and 25%, respectively. In summary, fossil fuel combustion is the primary source of SOAP, while aromatic hydrocarbons are the dominant contributing species. Furthermore, the impact of alkane emissions on SOAP from ports and terminals and the shipbuilding industry should not be overlooked.

Table 4 Summary of SOAP from different VOCs sources

Component	SOAP _i	Fossil Fuel Combustion		Port Terminal		Shipbuilding Industry		Solvent Use	
		Conc.		Conc.		Conc.		Conc.	
		(μ g/m ³)	SOAP	(μ g/m ³)	SOAP	(μ g/m ³)	SOAP	(μ g/m ³)	SOAP
	1.80	0.65	1.17	0.00	0.00	0.14	0.25	0.04	0.06
1,3-Butadiene	92.90	0.92	85.34	0.00	0.00	0.10	9.40	0.07	6.72
Benzene	100.00	2.58	258.46	0.75	74.74	0.00	0.00	0.05	4.77
Toluene	0.00	0.56	0.00	1.10	0.00	0.00	0.00	0.00	0.00
Methylcyclohexane	212.30	0.22	46.75	0.00	0.00	0.03	6.13	0.02	4.80
Styrene	111.60	0.92	102.58	0.90	100.68	0.07	8.12	5.04	562.87
Ethylbenzene	0.80	5.49	4.39	0.38	0.30	0.97	0.77	0.26	0.21
n-Octane	43.90	1.91	83.70	0.59	26.06	2.80	123.01	0.70	30.66
Trimethylbenzene	1.90	0.58	1.09	1.36	2.59	0.88	1.68	0.05	0.09
Nonane	0.00	0.88	0.00	0.23	0.00	7.69	0.00	0.00	0.00
Trichloroethane	0.00	0.00	0.00	2.67	0.00	2.35	0.00	0.65	0.00
Diethyl benzene	7.00	0.82	5.74	1.86	13.02	1.15	8.03	0.03	0.21

n-Decane	16.20	0.03	0.55	1.06	17.18	0.93	15.02	0.00	0.00
Undecane	/	1.37	/	0.88	/	2.83	/	0.05	/
Tetrachloroethylene	34.50	0.11	3.73	0.36	12.57	0.65	22.26	0.01	0.27
n-Dodecane	/	6.55	576.83	4.92	201.47	5.36	146.67	6.53	609.81
Aromatic hydrocarbons	/	7.59	15.50	6.12	45.66	4.57	47.76	0.35	0.78
Alkanes	/	0.65	1.17	0.00	0.00	0.14	0.25	0.04	0.06
Olefins	/	0.88	0.00	0.23	0.00	7.69	0.00	0.00	0.00
Halogenated hydrocarbons	/	15.67	593.50	11.27	247.13	17.76	194.67	6.91	610.65
Sum			36.1%		15.0%		11.8%		37.1%

3 Conclusion

1) Mobile monitoring of volatile organic compounds (VOCs) was conducted over the Jiangsu section of the Yangtze River via a round-trip voyage, covering approximately 250 km from west to east. On the outbound journey, the average TVOCs concentration in the Nanjing-Jiangyin section (western Jiangsu section) was $26.92 \mu\text{g}/\text{m}^3$, and the average concentration in the eastern Jiangsu section was $74.26 \mu\text{g}/\text{m}^3$. On the return journey, the average concentration in the western Jiangsu section was $3.55 \mu\text{g}/\text{m}^3$, and the average concentration in the eastern Jiangsu section was $42.67 \mu\text{g}/\text{m}^3$. The VOCs concentration exhibited a trend of high in the west and low in the east, which was generally consistent with the distribution of chemical industry agglomeration areas along the Jiangsu section of the Yangtze River.

2) Based on the toluene/benzene ratio (T/B), PMF source apportionment, and the SOA generation potential (SOAP) contribution, it was determined that pollution in the Jiangsu section of the Yangtze River is primarily caused by ship fuel combustion exhaust emissions. Emissions from ports and terminals, shipbuilding industry, coal-fired power plants, and chemical industry are relatively lower, but their impact should not be overlooked. The results from the three analytical methods are generally consistent.

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