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Pollution Characteristics and Health Risk Assessment of VOCs in Jinghong

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Abstract: In order to investigate the seasonal variation in chemical characteristics of VOCs in the urban and suburban areas of southwest China, we used SUMMA canister sampling in Jinghong city from October 2016 to June 2017. Forty-eight VOC species concentrations were analyzed using atmospheric preconcentration gas chromatography–mass spectrometry (GC–MS), Then, regional VOC pollution characteristics, ozone formation potentials (OFP), source identity, and health risk assessments were studied. The results showed that the average concentration of total mass was 144.34 $\mu g \cdot m^{-3}$ in the urban area and 47.81 $\mu g \cdot m^{-3}$ in the suburban area. Alkanes accounted for the highest proportion of VOC groups at 38.11%, followed by olefins (36.60%) and aromatic hydrocarbons (25.28%). Propane and isoprene were the species with the highest mass concentrations in urban and suburban sampling sites. The calculation of OFP showed that the contributions of olefins and aromatic hydrocarbons were higher than those of alkanes. Through the ratio of specific species, the VOCs were mainly affected by motor vehicle exhaust emissions, fuel volatilization, vegetation emissions, and biomass combustion. Combined with the analysis of the backward trajectory model, biomass burning activities in Myanmar influenced the concentration of VOCs in Jinghong. Health risk assessments have shown that the noncarcinogenic risk and hazard index of atmospheric VOCs in Jinghong were low (less than 1). However, the value of the benzene cancer risk to the human body was higher than the safety threshold of 1×10^{-6}, showing that benzene has carcinogenic risk. This study provides effective support for local governments formulating air pollution control policies.

Keywords: volatile organic compounds; Jinghong city; ozone formation potentials; source identity; health risk

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1. Introduction

China's energy consumption has increased rapidly, and urban air environment problems have become increasingly prominent in recent years. Tropospheric ozone (O_3) has become one of the main pollutants affecting air quality. Volatile organic compounds (VOCs) have attracted widespread attention as important precursors of O_3. VOCs refer to a class of organic gas compounds that exist in the air, including alkanes, aromatic hydrocarbons, olefins, halogenated hydrocarbons, and oxygenated hydrocarbons [1,2]. Extensive participation in atmospheric photochemical reactions leads to the formation of ozone and secondary organic aerosols (SOAs), which have a strong risk of carcinogenicity, which

is harmful to ambient air quality and human health [3,4]. Health brings threats such as respiratory damage, teratogenicity, and carcinogenicity [5,6].

Presently, various countries in the world have carried out VOCs-related research. China's research is mainly concentrated in the Beijing-Tianjin-Hebei and surrounding areas [7–10], Yangtze River Delta [11–13], Pearl River Delta [14–16], and other economically developed and densely populated areas of urban agglomerations, using offline or online monitoring analysis instruments and analysis methods to conduct related research. For example, Jay used proton transfer reaction time-of-flight mass spectrometry (PTR-TOF-MS) to perform highly time-resolved measurements in Beijing to study the characteristics and sources of volatile organic compounds (VOCs) [7]. Wei and Wang studied Handan, Hebei Province, a typical industrialized city in China; they conducted online measurements of VOCs and discussed their impact on $PM_{2.5}$ in the atmosphere [8,9]. Gu analyzed the multiscale chemical characterization and source apportionment of volatile organic compounds (VOCs) in Tianjin, China, from 1 November 2018 to 15 March 2019 [10]. Wang conducted online measurements of VOCs in Nanjing during the epidemic and evaluated the impact of the COVID-19 lockdown on the mixing ratio and sources of VOCs [12]. Ma investigated the pollution characteristics of VOCs products from eight synthetic resin enterprises in Shanghai, China [13]. Wang conducted a field study on the specific VOC (including OVOC) emissions of six construction machinery and five inland ships in the Pearl River Delta (PRD) region [14].

Relatively less attention has been paid to the atmospheric environment in southwest China, especially in the border cities affected by Southeast and South Asia. Jinghong city is located in the south of Yunnan province, close to Myanmar, Laos, and other countries, with the characteristics of a tropical climate and dense vegetation. It is restricted by the natural geographical conditions of typical basin topography, less precipitation, high temperature, and more inversion weather in winter and low winds. Village units and residential edges have frequent straw and waste incineration, as well as downtown restaurants and barbecue stalls with no pollution control facilities. Surrounding the city, rubber and wood processing factories produce waste gases. Combined with this, vehicle use has risen in recent years; all of the above release emissions of nitrogen oxides (NOx) and VOCs into the city and increase the production of ozone. Ozone is a secondary photochemical pollutant and greenhouse gas. It is formed by the reaction of NOx and VOCs, and other precursors under high-intensity ultraviolet light. Favorable conditions for ozone generation are mainly found in suburban areas, where NOx-rich air from windward urban agglomerations mix with VOCs emitted by trees and at high altitudes due to enhanced UV radiation [17]. Phytotoxic ozone has caused significant damage to terrestrial vegetation worldwide [18]. Dry deposition on plant surfaces (cuticle, bark), soil and stagnant water, and deposition through stomata into leaves leads to oxidative damage, which is the cause of carbon dioxide absorption in photosynthesis and decreased forest productivity [19,20].

In this study, VOCs, an important precursor of ozone, were taken as the research object, as well as VOC species concentration, ozone formation potentials, and health risks and sources. These were studied to provide a scientific basis for VOCs and ozone pollution control in this region.

2. Materials and Methods

2.1. VOCs Sampling

The atmospheric sampling sites of this study were the Municipal Environmental Protection Bureau and the Olive Dam Water Quality Automatic Monitoring Station of Jinghong. As shown in Figure 1, the Jinghong Municipal Environmental Protection Bureau is located in the city's urban center, where transportation is more frequent. The Olive Dam Water Quality Automatic Monitoring Station is located in the suburb, with relatively little human activity and transportation; it was taken as the suburban sampling control site. By sampling the urban and suburban areas, we can understand the difference in VOC species concentration in the suburban and urban areas of Jinghong City.

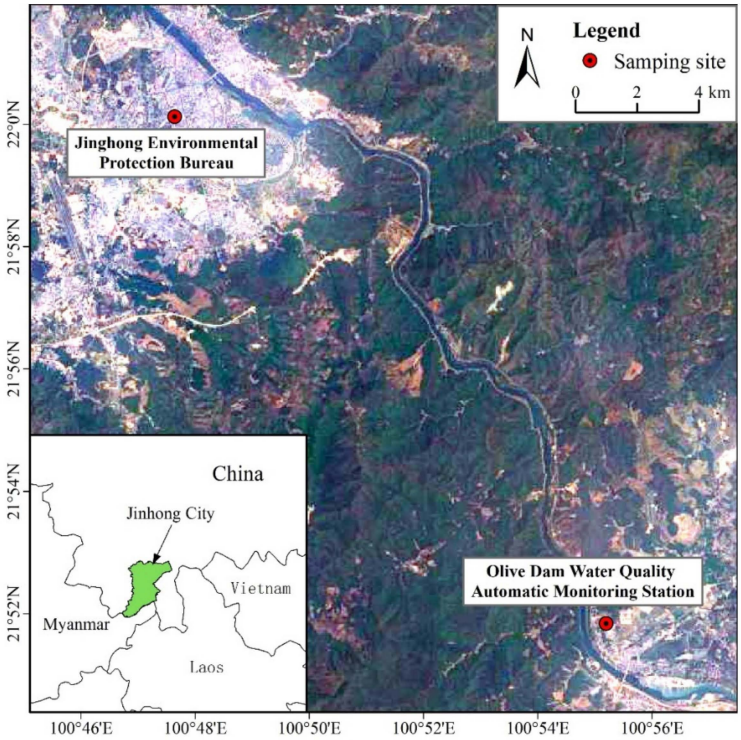

Figure 1. Sampling sites of VOCs in Jinghong.

2.2. Sample Collection and Component Analysis

According to the "Measuring Tank Sampling of Ambient Air Volatile Organic Compounds/Gas Chromatography-Mass Spectrometry (HJ759-2015)", the air samples were collected using a 3.2 L stainless-steel sampling tank produced by Entech Corporation of the United States. The inner wall of the SUMMA canister was cut and silanized, a flow valve controlled the sampling inlet flow, and a SUMMA canister sample was collected every 2 h. Sampling occurred in October 2016 and March, April, May, and June 2017, with 5 consecutive days of sampling at the beginning of each month. Meteorological conditions such as air temperature, air pressure, and wind speed during the sampling period were monitored and recorded online at a height of 3 m through the Vantage Pro2TM wireless weather station (Davis Instruments, Hayward, CA, USA). The rainfall in Jinghong varies significantly from month to month (abundant rainfall from June to October each year), so this paper defines the samples collected in October 2016 and June 2017 as the rainy season samples, and the samples collected in March, April, and May 2017 as the dry season samples.

This study analyzed 58 VOC species (alkanes, olefins, and aromatic hydrocarbons) by atmospheric preconcentration gas-conjugated methods. The main instruments were the atmospheric preconcentrator (Entech 7100, Entech Instruments, Simi Valley, CA, USA)-GC (Agilent 6890N, Agilent Technologies, Roseville, CA, USA)-MS (Agilent 5973N, Agilent Technologies, Roseville, CA, USA), and the column was the GS-GASPRO with a size of 60 m × 0.32 μm. The SUMMA canister was first connected to the autosampler, and 400 mL of the sample was pumped into the preconcentrator. The sample was concentrated by dewatering and using carbon dioxide interferences such as a 3-stage cold trap; then, the sample was transferred to GC–MS for detection. The carrier gas was high-purity helium, and the heating procedure was: initial temperature of 35 °C, held for 15 min, heated to 150 °C at 5 °C·min^{-1}; held for 7 min; heated to 200 °C at 10 °C·min^{-1}, maintained for 4 min. The inlet temperature was 140 °C, the carrier gas velocity was 1.0 mL·min^{-1}, and the solvent delay time was 5.6 min. Mass spectrometry conditions: interface temperature, 250 °C; ion source temperature, 230 °C; mass spectrometry detector ion source type, electron bombardment ionization (EI), operated by full scan (SCAN), with scanning range of 35~300 u. Five standard curves of different concentration gradients were established using

a PAMS calibration gas mixture (Spectra Gases USA) with volume fractions of 0.25×10^{-9}, 0.5×10^{-9}, 1×10^{-9}, 2×10^{-9}, and 3×10^{-9}.

2.3. Quality Assurance and Control

The SUMMA canister was cleaned 3 times with high-purity nitrogen with an automatic de-tanker (Entech 3100) before each sampling and pumped to a vacuum state, so that the pressure in the tank was less than 50 millitorr, and was ready for later. A cleaned SUMMA canister was injected with high-purity nitrogen as a laboratory blank, and the laboratory blank test was performed before the analysis of each batch of samples.

In the sample analysis process, 1 parallel sample was analyzed for every 10 samples measured, and the relative deviation in the target in the parallel sample was ≤30%. The correlation coefficients of the target compounds in the standard curve were >0.995. The retention time of the internal target in the sample deviated from the retention time of the internal standard in the continuous or recently drawn calibration curve of the day by no more than 20 s. Every 24 h, we analyzed the middle concentration point of the standard curve (1×10^{-9}); the measurement result was ≤30% of the initial mass concentration value; otherwise, the cause should be found or redrawn. The results showed no contamination during sample handling and collection, as assured by the quality assurance and control (QC/QA) procedures.

2.4. Analytical Methods

2.4.1. Ozone Formation Potentials (OFP)

Ozone formation potentials can be used to evaluate the potential of VOCs emissions participating in the reaction to generate ozone, and they provided some guidance for the formulation of VOCs control measures. In this study, the maximum incremental reactivity method (MIR) was used to determine the contribution of active components and key species in VOCs to O_3 production [21,22], calculated as in Equation (1):

$$OFP_i = VOCs_i \times MIR_i \quad (1)$$

where OFP_i represents the amount of ozone generation potential of species i in $\mu g \cdot m^{-3}$; VOC_{si} represents the mass concentration of species i in $\mu g \cdot m^{-3}$; and MIRi represents the MIR coefficient of species i [23].

2.4.2. HYSPLIT Model

The HYSPLIT model(https://ready.arl.noaa.gov/HYSPLIT_traj.php, accessed on 1 July 2021) is a comprehensive model system developed by the National Oceanic and Atmospheric Centre (NOAA) and the Australian Meteorological Agency (BOM). It can be used to calculate and analyze processes such as airflow movement, sedimentation, air pollutant transport, and diffusion trajectory [24]. At present, it has been widely used to study the transmission routes and source analysis of air pollutants [25–28].

In this paper, TrajStat follow-up software [29] was used to analyze and study the backward trajectory of air masses in Jinghong. As the 500 m height wind field accurately reflects the average flow field characteristics of the boundary layer [27], the simulated height was chosen as 500 m. Jinghong (100°47′38″ E, 22°00′07″ N) was the simulated receiving point, 8:00 (Beijing time) every day was the pushback start time, and the 72 h backward trajectory of the receiving point from July 2016 to June 2017 was calculated, to reflect the characteristics of the airflow.

2.4.3. Health Risk Assessment

In order to study the potential harm of VOCs to human health in Jinghong City, it is necessary to assess the health risk of VOCs. This study adopted a new health risk assessment method (EPA-540-R-070-002) proposed by the U.S. EPA in 2009 for inhaled route pollutants in specific places. The calculation formula is as follows:

$$EC = \frac{CA \times ET \times EF \times ED}{AT} \quad (2)$$

$$HQ = \frac{EC}{(RFC \times 1000)} \quad (3)$$

$$R = EC \times IUR \quad (4)$$

$$HI = \sum HQ_i \quad (5)$$

where EC is the exposure concentration in units of $\mu g \cdot m^{-3}$; CA is the ambient concentration of VOCs in $\mu g \cdot m^{-3}$; ET is the exposure time in $h \cdot d^{-1}$, with a value of 24; EF is the exposure frequency in $d \cdot a^{-1}$, with a value of 365; ED is the exposure time in a, with a value of 70; AT is the average time in h, with a value of $70 \times 365 \times 24$; HQ is the noncarcinogenic risk hazard quotient value; RfC is the reference concentration in $\mu g \cdot m^{-3}$; R is the lifetime risk value of carcinogenicity, IUR is the inhalation risk in $\mu g \cdot m^{-3}$; and HI is the hazard index. The RfC and IUR values are from the reference [30].

2.4.4. Data Sources

This study used daily averaged and hourly data on the concentrations of six atmospheric pollutants (SO_2, NO_2, CO, O_3, $PM_{2.5}$, and PM_{10}) at two environmental monitoring stations in Jinghong from July 2016 to June 2017. Contaminant data were from the National Environmental Monitoring Center of China (https://air.cnemc.cn:18007/, accessed on 1 July 2021). The data used in the backward trajectory model were 2016–2017 Global Data Assimilation System (GDAS) data provided by the NCEP (National Center for Environmental Prediction). Meteorological data were derived from the National Meteorological Science Data Sharing Service Platform (http://data.cma.cn/, accessed on 1 July 2021).

3. Results and Discussions

3.1. Species Composition Characteristics of VOCs

During the sampling period, 58 VOC compounds (alkanes, olefins, and aromatic hydrocarbons) were detected in the atmosphere at the urban and suburban monitoring sites in Jinghong, and a total of 48 VOC species were detected. As shown in Table 1, the total concentration of VOCs was (144.34 ± 36.15) $\mu g \cdot m^{-3}$ at the urban sampling site, which was much higher than at the suburban sampling site (48.21 ± 12.55 $\mu g \cdot m^{-3}$). Compared with the suburban sampling site, the urban sampling site in Jinghong was closer to various anthropogenic emission sources, which are susceptible to factors such as the emission of VOC sources of human activities. These emission sources have the characteristics of continuity, concentration, and high concentration, which make the concentration of VOCs in urban areas highly accumulated. As shown in Figure 2, in the dry season, the concentrations of alkanes, olefins, aromatic hydrocarbons, and TVOCs in the urban areas of Jinghong were (69.94 ± 9.25) $\mu g \cdot m^{-3}$, (37.53 ± 8.22) $\mu g \cdot m^{-3}$, (38.72 ± 17.32) $\mu g \cdot m^{-3}$, and 146.19 $\mu g \cdot m^{-3}$, respectively, with those in the suburban areas were (7.40 ± 2.34) $\mu g \cdot m^{-3}$, (32.22 ± 7.99) $\mu g \cdot m^{-3}$, (6.74 ± 3.43) $\mu g \cdot m^{-3}$, and TVOCs 47.15 $\mu g \cdot m^{-3}$, respectively. During the rainy season, the concentrations of alkanes, olefins, aromatic hydrocarbons, and TVOCs in urban areas were (62.93 ± 16.13) $\mu g \cdot m^{-3}$, (33.44 ± 8.22) $\mu g \cdot m^{-3}$, (46.13 ± 13.17) $\mu g \cdot m^{-3}$, and 142.5 $\mu g \cdot m^{-3}$, respectively, and those in suburban areas were (6.21 ± 1.99) $\mu g \cdot m^{-3}$, (37.48 ± 6.69) $\mu g \cdot m^{-3}$, (5.58 ± 2.65) $\mu g \cdot m^{-3}$, and 49.27 $\mu g \cdot m^{-3}$, respectively. The VOC groups at the urban sampling site were mainly alkanes, while the VOC groups at the suburban sampling site were mainly olefins. A nonparametric test (one-way analysis of variance, 95% confidence level) showed there were no significant differences in VOCs concentration and species composition between the dry season and rainy season ($p > 0.05$). The concentration of aromatic hydrocarbon groups in the urban sampling site was higher than that in the dry season, which may be related to the increase in benzene emissions due to the cooling of motor vehicle air conditioning and cooling in the rainy season.

Table 1. Concentration levels of atmospheric VOCs in different cities.

City	Period	Number of VOCs Species	Concentration of VOCs/(μg·m^{-3})	Alkanes/(%)	Olefins/(%)	Aromatic Hydrocarbons/(%)	Reference
urban of Jinghong	2016~2017	48	144.35 ± 36.15	46.03	24.58	29.39	This study
suburbs of Jinghong			48.21 ± 12.55	14.12	73.11	12.78	
Kunming	2014	35	30.22	68.58	7.78	23.64	[31]
Guangzhou	2009	31	114.50	59.97	15.18	39.24	[32]
Beijing	2016	99	44.00	36.80	7.00	11.80	[33]
Chengdu	2012	59	108 ± 52.43	47.08	11.53	44.52	[34]
Monterrey (Mexico)	2011	29	80.14	62.88	9.55	24.67	[35]
Nagoya (Japan)	2003~2004	48	62.28	50.08	9.28	40.64	[36]

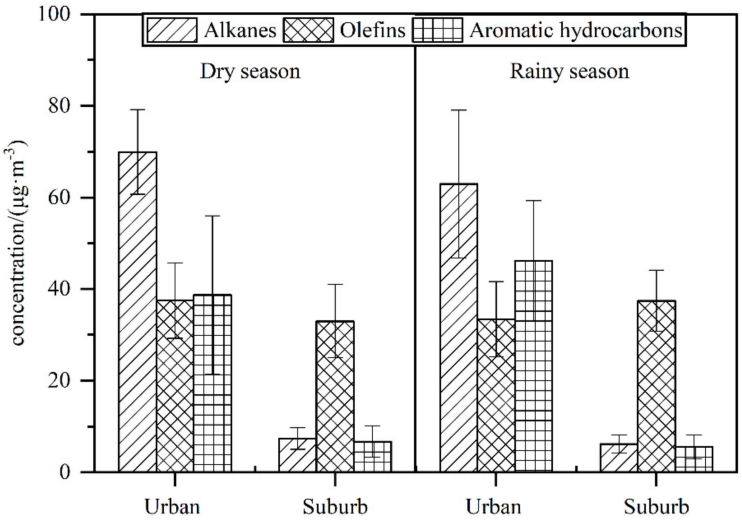

Figure 2. Concentration of alkanes, olefins, and aromatics in urban and suburban areas of Jinghong.

The comparison of VOC species concentrations in different atmospheric environments in Jinghong and other cities is shown in Table 1. The data sample collection equipment used a Suma tank with a volume of 6 L, and about 28 effective samples were in Kunming. The samples were collected with stainless-steel tubes, and about 12 effective samples were collected in Chengdu. VOC observations adopted syntech spectra gc955 online monitoring systems produced by synspec company in the Netherlands, and about 220 groups of effective data were from Guangzhou. The VOC sampling was conducted at the superstation for atmospheric environmental monitoring in Beijing Normal University, and the amount of valid data was not mentioned in Beijing. Samples were collected using ENTECH (Malvern, PA, USA) summa electro-polished stainless-steels container (6 L) following the United States Environmental Protection Agency analytical method (US EPA, 1999), and about 56 effective samples were collected in Monterey; an automatic measurement system for Non-methane Hydrocarbons (NMHCs) was constructed. The system was set-up at the Hydrospheric Atmospheric Research Center (HyARC) building in Nagoya University, and the amount of valid data was not mentioned in Nagoya (Japan). The sampling areas of the above cities were urban areas. Table 1 shows that there was a significant difference in the proportion of species between the urban and suburban area of Jinghong City. The concentration in the urban area was about three times that in the suburbs. The highest proportion of VOC species in the urban area was alkane, 46.03%, followed by olefin and aromatic hydrocarbon species, accounting for 24.58% and 29.39%, respectively. Its species composition was different from those of Kunming, Guangzhou, Beijing, Chengdu, Monterrey, Nagoya, and other cities. The proportion of olefins in the suburbs of Jinghong was highest (73.11%), followed by alkanes and aromatic hydrocarbons (14.12% and 12.78%, respectively), which may be related to the dense vegetation in the suburbs monitored by this study. During the whole observation period, the average concentration of VOCs in the

urban area of Jinghong city reached (144.35 ± 36.15) μg·m^{-3}, which was higher than those in other comparison cities, showing a high concentration level.

Figure 3 shows the distribution of VOC species concentrations in the dry and rainy seasons of the urban and suburban sampling site in Jinghong during the sampling period. As shown in Figure 2, the propane concentrations at the urban sampling site were highest during the entire sampling period. Higher alkane concentrations included propane, n-butane, isobutane, and isoamylene; alkene concentrations included isoprene and propylene species. Higher aromatic hydrocarbon concentrations were mainly toluene, which was much higher than the concentration of suburban points, reflecting the more significant impact of urban motor vehicle exhaust on the concentration of atmospheric VOCs. The concentration of isoprene in the suburban monitoring points was more prominent, which is closely related to suburban monitoring sites moving away from anthropogenic sources and closer to natural sources such as dense forests. This was followed by propane, probably related to petrochemical emissions. The concentrations of other species were low. The spectral characteristics of VOCs at the two sampling sites in Jinghong urban and suburban areas had a large difference, indicating that there were certain differences in the sources of VOCs between the two sampling sites in the city and suburb.

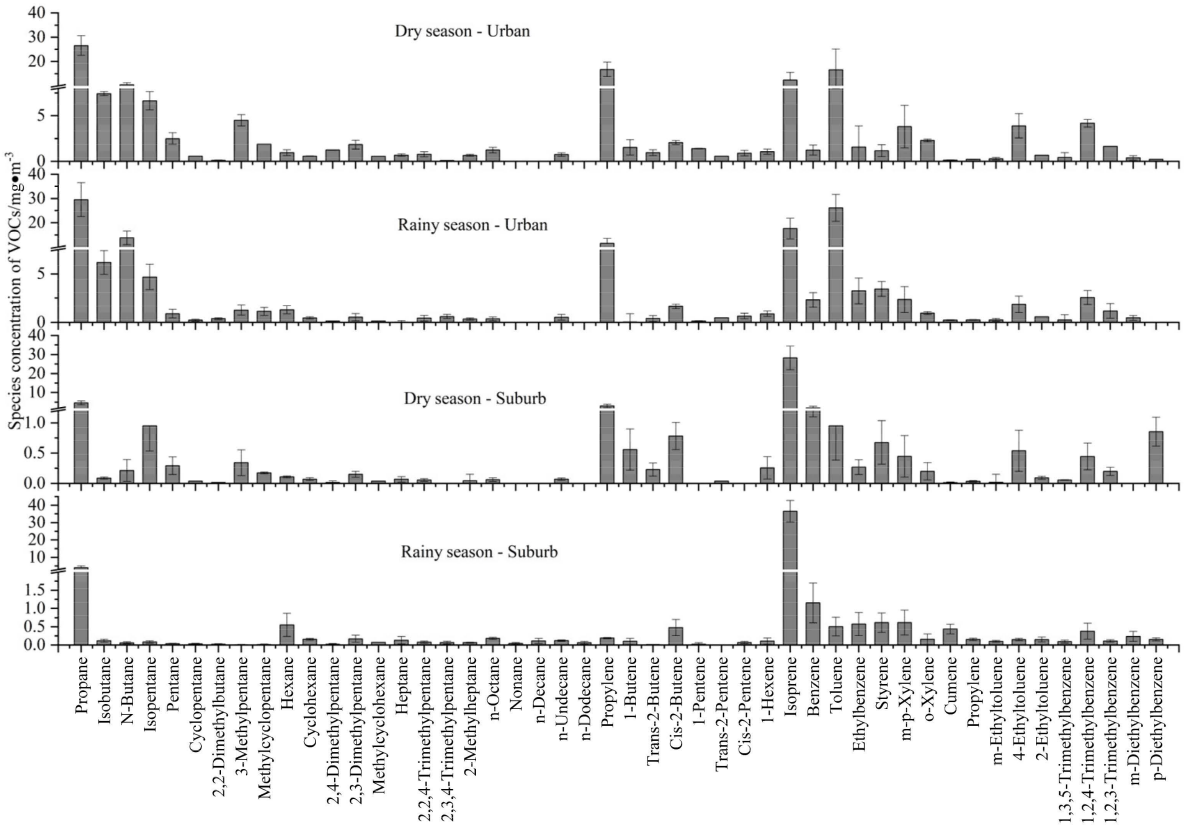

Figure 3. The level profiles of VOCs concentrations in Jinghong.

3.2. Ozone Formation Potentials (OFP) of VOCs

Table 2 lists the OFP values of VOCs species at urban and suburban sampling sites in Jinghong. The OFP of total VOCs in the atmosphere in the urban area of Jinghong was 588.07 μg·m^{-3} and 535.38 μg·m^{-3} in the dry season and 535.38 μg·m^{-3}, respectively, which was much higher than that of the suburb, indicating that the ozone generation capacity in the urban area of Jinghong was much higher than that in the suburban area. In the dry season, urban alkanes, olefins, and aromatic hydrocarbons accounted for 11.49%, 57.73%, and 30.78% of the OFP, respectively, and correspondingly accounted for 1.82%, 89.70%, and 8.48% of the OFP in the suburb. During the rainy season, alkanes, olefins, and aromatic hydrocarbons

accounted for 10.10%, 56.91%, and 32.99% of the OFP in the urban atmosphere, respectively, compared with 1.14%, 92.41%, and 6.45% in the suburb during the same period. Olefins are the key species of ozone formation potential in Jinghong, and the OFP ranking has always been manifested as: olefins > aromatic hydrocarbons > alkanes. The contribution of olefins to ozone generation in the suburban atmosphere is higher than that in urban areas, indicating that the suburban ozone generation is mainly affected by olefins.

Table 2. Potential Ozone Formation of VOCs in Jinghong.

Species	MIR Value	Ozone Formation Potential (μg·m^{-3})			
		DU	DS	RU	RS
Propane	0.48	12.75	2.21	14.18	1.92
Isobutane	1.21	8.99	0.11	7.47	0.14
N-Butane	1.02	10.64	0.22	14.03	0.06
Isopentane	1.38	9.17	1.31	6.45	0.11
Pentane	1.04	2.60	0.30	0.94	0.04
Cyclopentane	1.38	0.77	0.05	0.32	0.05
2,2-Dimethylbutane	0.82	0.10	0.01	0.3	0.02
3-Methylpentane	1.50	6.75	0.51	1.91	0.02
Methylcyclopentane	2.80	5.26	0.49	3.16	0.04
Hexane	0.98	0.94	0.11	1.27	0.54
Cyclohexane	1.28	0.72	0.09	0.58	0.20
2,4-Dimethylpentane	1.50	1.86	0.02	0.21	0.04
2,3-Dimethylpentane	1.31	2.40	0.20	0.71	0.22
Methylcyclohexane	2.80	1.48	0.10	0.38	0.20
Heptane	0.81	0.55	0.06	0.03	0.11
2,2,4-Trimethylpentane	0.93	0.71	0.05	0.4	0.07
2,3,4-Trimethylpentane	1.60	0.16	0.00	0.96	0.11
2-Methylheptane	0.96	0.63	0.04	0.34	0.06
n-Octane	0.60	0.74	0.04	0.22	0.11
Nonane	0.54	-	-	-	0.02
n-Decane	0.46	-	-	-	0.05
n-Undecane	0.42	0.31	0.03	0.22	0.05
n-Dodecane	0.38	-	-	-	0.02
Total alkanes		67.55	5.95	54.08	4.20
propylene	9.40	157.26	27.68	108.47	1.78
1-Butene	8.90	13.62	-	0.48	0.88
Trans-2-Butene	10.00	9.50	-	4	0.13
Cis-2-Butene	10.00	20.60	7.84	16.57	4.77
1-Pentene	6.20	8.68	0.00	0.89	0.16
Trans-2-Pentene	8.80	4.95	0.34	4.05	0.05
Cis-2-Pentene	8.80	7.88	0.00	5.78	0.59
1-Hexene	4.40	4.62	1.13	3.92	0.46
Isoprene	9.10	112.39	256.62	160.52	332.15
Total olefins		339.50	293.60	304.67	340.97
Benzene	0.42	0.52	0.82	0.98	0.49
Toluene	2.70	44.70	2.56	70.68	1.36
Ethylbenzene	2.70	4.24	0.72	8.74	1.55
Styrene	1.95	7.48	4.36	22.25	3.97
m-p-Xylene	7.40	7.45	0.87	4.6	1.20
o-Xylene	6.50	17.01	1.48	7.09	1.16
Cumene	2.20	0.90	0.09	1.56	2.85
Propylene	2.10	0.49	0.08	0.55	0.34
m-Ethyltoluene	7.20	0.61	0.04	0.55	0.20
4-Ethyltoluene	7.20	28.10	3.88	13.37	1.08
2-Ethyltoluene	7.20	4.86	0.67	4.15	1.08
1,3,5-Trimethylbenzene	10.10	2.99	0.39	1.8	0.67
1,2,4-Trimethylbenzene	8.87	42.20	4.50	25.86	3.83
1,2,3-Trimethylbenzene	8.90	14.63	1.76	10.38	0.96
m-Diethylbenzene	6.45	3.34	-	4.09	2.09
p-Diethylbenzene	6.45	1.51	5.51	-	0.98
Total aromatic hydrocarbons		181.01	27.75	176.64	23.81
Total VOCs		588.07	327.30	535.38	368.98

DU: Dry season—Urban; DS: Dry season—Suburb; RU: Rainy season—Urban; RS: Rainy season—Suburb.

From the analysis of specific VOCs compounds, in the dry season, the top 12 substances with the highest OFP in the atmosphere of Jinghong were propylene, isoprene, toluene,

1,2,4-trimethylbenzene, p-ethyltoluene, cis-2-butene, o-xylene, 1,2,3-trimethylbenzene, 1-butene, propane, n-butane, and trans-2-butene, and their mass concentration accounts for 67.86% of total volatile organic compounds (TVOCs). The top 12 substances in the suburban area regarding OFPs were isoprene, propylene, cis-2-butene, p-diethylbenzene, 1,2,4-trimethylbenzene, styrene, p-ethyltoluene, toluene, propane, 1,2,3-trimethylbenzene, o-xylene, and isopentane, which accounted for 88.75% of the TVOCs by mass. The ozone generation accounted for 97.28% of the total OFPs. In the rainy season, the top 12 substances of OFPs in the urban air were isoprene, propylene, toluene, 1,2,4-trimethylbenzene, styrene, cis-2-butene, propane, n-butane, p-ethyltoluene, 1,2,3-trimethylbenzene, ethylbenzene, and isobutane, which accounted for 83.34% of TVOCs by mass and 88.26% of total OFP by ozone generation. The top 12 substances of OFPs in the suburb were isoprene, cis-2-butene, styrene, 1,2,4-trimethylbenzene, isopropylbenzene, m-diethylbenzene, propane, propylene, ethylbenzene, toluene, p- and m-xylene, and o-xylene, which accounted for 90.69% of the TVOCs by mass and 97.20% of the total OFPs by ozone potential. Overall, isoprene accounted for the highest percentage of OFP in Jinghong, indicating that natural vegetation emission sources in Jinghong contributed prominently to ozone, followed by toluene, propylene, m-p-xylene, and other motor vehicle exhaust emissions that contributed significantly to ozone generation.

3.3. Source Analysis of VOCs

3.3.1. Ratio of Specific Species

The ratio of toluene to benzene is commonly used to determine the sources of traffic emissions, fuel combustion, and industrial and solvent use in the current regional atmospheric environment [37–40]. Isopentane and n-pentane have similar chemical reactions with free radicals, and their ratios can also be used to indicate different sources [38,41,42]. The ratio of m/p-xylene to ethylbenzene is often used to evaluate the degree of aging of gas clusters [42–44], and the length of photochemical age. Ethylbenzene is less reactive than m/p-xylene and the ratio decreases in atmospheric chemical reactions where m/p-xylene is consumed faster—the smaller the ratio, the higher the degree of aging of the gas cluster. Moreover, BTEX pollution may be affected by regional transmission [45]. Table 3 shows the degree of air mass aging for m- and p-xylene and ethylbenzene for the ranges of benzene to toluene and isopentane to n-pentane ratios, respectively, corresponding to the pollution sources. Benzene to toluene, isopentane to n-pentane, and m-p-xylene to ethylbenzene ratios were selected for the study; additionally, the emission share of isoprene was considered to study the emissions from natural sources, where a larger ratio indicates that the air mass is fresher, and a smaller ratio indicates that the air mass is aging.

Table 3. Ratio data of benzene to toluene and isopentane to n-pentane.

Characteristic Species Ratio	Value	Source	Reference
Benzene/Toluene	0.00–0.20	solvent usage	[39]
	0.50–0.60	vehicle emissions	
	1.05–2.20	coal emissions	
	2.50	biomass burning	
Isopentane/n-pentane	0.56–0.80	coal emissions	[41]
	1.50–3.00	liquid gasoline	
	1.84–4.60	fuel volatilization	
	2.93	motor vehicle exhaust	

Jinghong is located at the southern tail end of the longitudinal valley of the Hengduan Mountains, which has a tropical and subtropical humid monsoon climate with high precipitation and dense vegetation covering a wide area. As shown in Table 4, the concentration of isoprene in the suburban area (59.80% in the dry season and 74.10% in the rainy season) was higher than that in the urban area (8.45% in the dry season and 12.38% in the rainy season), indicating that the release of large amounts of VOCs from dense vegetation in

the suburb had an important impact on VOCs in the ambient air, and isoprene emission from vegetation exponentially increased with sunlight and temperature on rainy days. The benzene/toluene ratios of 2.42 and 2.70 in the dry and rainy seasons in the suburban area, respectively, showed that fresh emissions or exhaust emissions and biomass burning were the primary source. The ratios of 0.09 and 0.10 in the dry and rainy seasons in the urban area, respectively, which were less than 0.2, showed that solvent use was the primary source. From the ratios of isopentane/n-pentane, it shows that the ratios of 3.24 and 2.10 for the dry and rainy seasons in suburban areas, respectively, were mainly influenced by gasoline fuel volatilization, and the ratios of 2.66 and 5.19 for the dry and rainy seasons in urban areas, respectively, were mainly influenced by motor vehicle exhaust and fuel volatilization. The ratio of M-p-xylene/Ethylbenzene was small in the urban and suburban areas in the rainy season, which means that the air mass was relatively aging. In addition to local emissions, some of the benzene series pollution came from regional transmission. In the dry season, the ratio between urban and suburban areas was bigger, which means that the air mass was relatively fresh, and the pollution was mainly transmitted locally.

Table 4. Species ratio in dry and rainy seasons in urban and suburban areas of Jinghong.

Area	Benzene/Toluene	Isopentane/ n-Pentane	Isoprene/TVOCs	M-p-Xylene/Ethylbenzene
DU	0.09	2.66	8.45%	2.44
RU	0.10	5.19	12.38%	0.73
DS	2.42	3.24	59.80%	1.67
RS	2.70	2.10	74.10%	1.07

DU: Dry season—Urban; DS: Dry season—Suburb; RU: Rainy season—Urban; RS: Rainy season—Suburb; TVOCs: Total Volatile Organic Compounds.

3.3.2. The Long-Range Transport

As shown in Figure 4, the incoming air masses during the year of the sampling period were resolved using the HYSPLIT model to track the trajectories of air masses arriving in the region in the past 72 h. The air masses from the Southwest were the most abundant in the region. Almost all air masses originated from Southeast Asian countries, with the most air masses coming from Myanmar, accounting for the most southwestern air mass transport (Cluster 1 air masses, 48.09%) and the lowest transport altitude (pressure above 870 hPa).

The trajectory air masses in different directions may contain different levels of pollutants. The backward trajectory clustering and pollutant concentration information were combined to analyze the influence of each trajectory air mass on pollutants in the study area. The results are shown in Table 5. The pollutant concentrations in different trajectory air masses varied widely, with the largest values of pollutants being SO_2, NO_2, CO, O_3, $PM_{2.5}$, and PM_{10} in cluster 2 and the second largest values in cluster 5, mainly from the central region of Myanmar, becoming the most important transport path affecting the atmospheric pollutant concentrations in Jinghong. The airflow near Thailand and Laos (clusters 1 and 4) had the lowest ozone concentration values, compared to the other airflow (from Myanmar) trajectories, which had higher ozone concentration values, probably related to the more frequent biomass burning activities in Myanmar. It can be speculated that the outbound transport of VOCs may be mainly influenced by the transport from Myanmar.

3.4. Health Risk Assessment of VOCs

Benzene, toluene, ethylbenzene, m/p-xylene, and o-xylene are the important VOCs species in the atmosphere of Jinghong, and the proportion of aromatic hydrocarbons was ~58.70%. Table 6 compares the noncarcinogenic and carcinogenic risks of BTEX (benzene, toluene, ethylbenzene, m/p-xylene, o-xylene) mass concentration in Jinghong with other cities. The results show that the noncarcinogenic risk values of benzene > m/p-xylene > o-xylene > ethylbenzene > toluene in both the dry and rainy season of suburban sampling

sites, and benzene > m/p-xylene > o-xylene > toluene > ethylbenzene in both the dry and rainy season of urban sampling sites, ranging from 1.01×10^{-4} to 7.75×10^{-2}, were within the safe range (HQ < 1) as determined by the US EPA. The lifetime carcinogenic risk value of benzene ranged from 9.02×10^{-6} to 1.82×10^{-5}, which is higher than the safety range ($R < 1.00 \times 10^{-6}$) as determined by the US EPA. These results indicate the existence of carcinogenic risk and the possibility of inducing lymphatic system immune diseases and leukemia in humans exposed to the environment for an extended time, which should be taken more seriously.

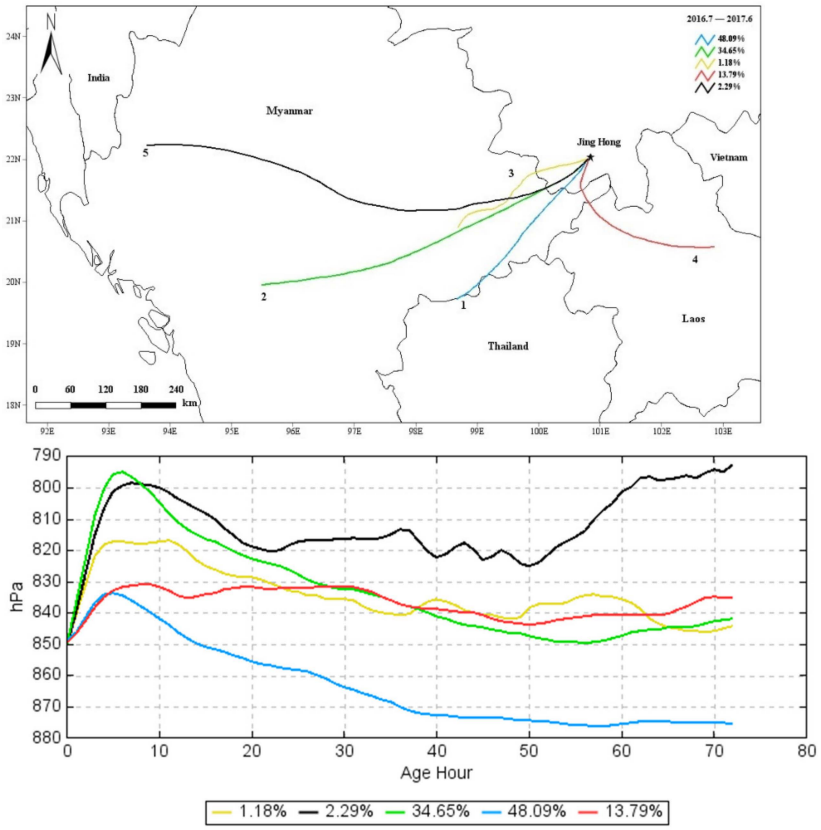

Figure 4. Trajectory clustering from July 2016 to June 2017.

Table 5. Statistical results of various trajectory pollutant concentrations from July 2016 to June 2017 in Jinghong.

Trajectory	Frequency of Occurrence/ (%)	SO_2/ ($\mu g \cdot m^{-3}$)	NO_2/ ($\mu g \cdot m^{-3}$)	CO/ ($mg \cdot m^{-3}$)	O_3/ ($\mu g \cdot m^{-3}$)	$PM_{2.5}$/ ($\mu g \cdot m^{-3}$)	PM_{10}/ ($\mu g \cdot m^{-3}$)
1	48.09	6.83	13.85	0.70	36.21	20.01	39.45
2	34.65	8.90	20.08	0.81	50.84	34.65	61.78
3	1.18	6.56	14.38	0.72	40.85	22.06	42.71
4	13.79	6.38	13.48	0.73	29.85	19.14	37.27
5	2.29	7.89	20.82	0.78	49.65	34.45	57.64

The changing trend of the noncarcinogenic risk quotient in Jinghong shows that benzene and toluene had higher noncarcinogenic risk quotient values, which was the same as those of Guangzhou [32] and Beijing [46]. However, it was shown that benzene and xylene had higher noncarcinogenic risk quotients in Chengdu [34] and Xiamen [47], while xylene had higher quotients in Lanzhou [48], and benzene had a higher quotient in Port Moody (Canada), Burnaby South (Canada) [49], and Tabriz (Iran) [50]. In this study, HI values were higher in urban areas than in suburban areas, and lower in Jinghong than in other cities. The lifetime carcinogenic risk of benzene was low compared to other cities,

except for the dry season suburban area and rainy season urban area, which were higher than that of the residential sampling site in Xiamen (1.23×10^{-5}). Overall, it was shown that the health risk level in Jinghong was lower than its urban counterparts, However, it should be noted that the carcinogenic risk value of benzene in this region has exceeded the safety threshold. Therefore, it is necessary to increase the control of benzene emissions to reduce its carcinogenic risk.

Table 6. Comparison of noncarcinogenic and carcinogenic risks of BTEX in Jinghong with other cities.

Sampling Site	*R* (Benzene)	HQ					HI	Reference
		Benzene	Toluene	Ethylbenzene	m/p-Xylene	O-Xylene		
dry season in suburb of Jinghong	1.52×10^{-5}	6.48×10^{-2}	1.90×10^{-4}	2.68×10^{-4}	4.48×10^{-3}	2.00×10^{-3}	7.18×10^{-2}	This work
dry season in urban of Jinghong	9.62×10^{-6}	4.11×10^{-2}	3.31×10^{-3}	1.57×10^{-3}	3.82×10^{-2}	2.30×10^{-2}	1.07×10^{-1}	
rainy season in suburb of Jinghong	9.02×10^{-6}	3.85×10^{-2}	1.01×10^{-4}	5.74×10^{-4}	6.16×10^{-3}	1.57×10^{-3}	4.70×10^{-2}	
rainy season in urban of Jinghong	1.82×10^{-5}	7.75×10^{-2}	5.24×10^{-3}	3.24×10^{-3}	2.36×10^{-2}	9.57×10^{-3}	1.91×10^{-1}	
Guangzhou	5.34×10^{-5}	2.28×10^{-1}	3.95×10^{-1}	4.26×10^{-3}	3.06×10^{-2}	2.42×10^{-2}	2.91×10^{-1}	[32]
urban area of Chengdu	6.77×10^{-5}	2.89×10^{-1}	2.36×10^{-3}	3.34×10^{-3}	1.18×10^{-1}	3.38×10^{-2}	4.47×10^{-1}	[34]
traffic area of Chengdu	6.98×10^{-5}	2.98×10^{-1}	3.09×10^{-03}	3.19×10^{-3}	9.08×10^{-2}	3.38×10^{-2}	4.29×10^{-1}	
Beijing	4.19×10^{-5}	1.57×10^{-1}	2.39×10^{-1}	3.29×10^{-3}	8.06×10^{-3}	3.53×10^{-3}	1.96×10^{-1}	[46]
residential area of Xiamen	1.23×10^{-5}	5.25×10^{-2}	9.73×10^{-4}	2.37×10^{-3}	1.63×10^{-2}	1.13×10^{-2}	8.34×10^{-2}	[47]
industrial area of Xiamen	3.08×10^{-5}	1.32×10^{-1}	4.29×10^{-3}	7.98×10^{-3}	5.57×10^{-2}	4.36×10^{-2}	2.43×10^{-1}	
lanzhou	8.09×10^{-6}	3.46×10^{-2}	1.27×10^{-4}	4.07×10^{-4}	6.18×10^{-3}	2.76×10^{-3}	4.42×10^{-2}	[48]
Port Moody (Canada)	-	2.02×10^{-1}	9.17×10^{-3}	2.62×10^{-4}	-	-	2.02×10^{-1}	[49]
Burnaby South (Canada)	-	1.51×10^{-1}	6.29×10^{-3}	1.32×10^{-4}	-	-	1.51×10^{-1}	
Tabriz (Iran)	-	1.07×10^{-1}	1.29×10^{-3}	-	-	-	2.21×10^{-1}	[50]

"-" means that there are no such data in the references.

4. Conclusions

In this study, VOCs samples were collected in Jinghong and analyzed for their mass concentration, ozone formation potential, source identity, and health risk assessments in dry and rainy seasons. During the sampling period, a total of 48 VOC species were detected at the urban and suburban monitoring sites. In the dry season, the TVOCs concentration was 146.19 $\mu g/m^3$ in the urban area and 47.15 $\mu g/m^3$ in the suburban areas. In the rainy season, the TVOCs concentration was 142.5 $\mu g/m^3$ in the urban area and 49.27 $\mu g/m^3$ in the suburban area. TVOCs in urban areas were much higher than those in suburbs, which was related to human activities. The concentration of VOCs in the dry season at the urban sampling site was higher than in the rainy season, while at the suburban sampling sites, the concentration in the rainy season was higher than that in the dry season. These results may be related to the emission of more olefins in the rainy season when the vegetation is dense and grows vigorously.

The species with high concentrations of atmospheric VOCs in Jinghong were propane, toluene, propylene, and isoprene, and their OFPs were olefins > aromatic hydrocarbons > alkanes. The species with the highest OFP was isoprene, indicating that the surrounding dense vegetation contributes significantly to the generation of O_3. The ratio of specific species was used to analyze

the primary sources of VOCs in the atmosphere of the main urban area of Jinghong: plant sources, motor vehicle exhaust, and oil and gas volatilization; combined with the study of air mass trajectories, the transmission of biomass combustion sources in the Myanmar region may influence VOCs in the study area. The health risk of VOCs in the main urban area of Jinghong was generally lower than those of other cities, and the noncarcinogenic risk was within the safety threshold. However, benzene's lifetime carcinogenic risk value in the atmosphere exceeded the safety range ($R < 1.00 \times 10^{-6}$), and there was a particular carcinogenic risk. The control of local VOC sources should be strengthened appropriately to reduce benzene emissions.

In this paper, chemical composition and source characteristics of VOCs in a plateau border city were first studied, and the main sources of VOCs in Jinghong were resolved. The results can provide scientific data to support VOCs pollution control in local and similar cities. In addition, due to high vegetation coverage, forest and vehicle exhaust emissions were prone to produce phytotoxic ozone, which reduced forest productivity and damaged terrestrial vegetation. Therefore, the local government needs to take corresponding measures. Biomass burning activities in Myanmar had an impact on VOCs concentration changes in Jinghong, and cross-border pollution issues also need attention, which has a certain guiding significance for our government to carry out international cooperation projects in Southeast Asia.

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