Volatile organic compounds and their contribution to ground-level ozone formation in a tropical urban environment

Mohd Faizul Hilmi Zulkifli [a,b], Nor Syamimi Sufiera Limi Hawari [a], Mohd Talib Latif [a,c,*], Haris Hafizal Abd Hamid [a], Anis Asma Ahmad Mohtar [a], Wan Mohd Razi Wan Idris [a], Nur Ili Hamizah Mustaffa [d], Liew Juneng [a]

[a] Department of Earth Sciences and Environment, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600, Bangi, Malaysia
[b] Air Division, Department of Environment, Ministry of Environment and Water, 62574, Putrajaya, Malaysia
[c] Department of Environmental Health, Faculty of Public Health, Universitas Airlangga, 60115, Surabaya, Indonesia
[d] Department of Environment, Faculty of Forestry and Environment, Universiti Putra Malaysia, 43400, UPM Serdang, Selangor, Malaysia

HIGHLIGHTS

- VOC concentrations recorded at the highest concentration in urban environment.
- n-Butane, ethene, ethane and toluene recorded as the most abundant species.
- Alkenes and aromatic hydrocarbons are the major contributors to O_3 formation.
- Relative humidity was found to influence the formation of VOCs.

GRAPHICAL ABSTRACT
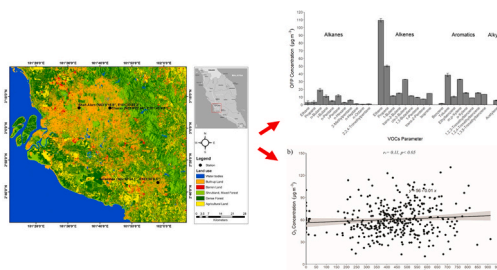
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ABSTRACT

This study aims to determine the trends of volatile organic compound (VOC) concentrations and their potential contribution to O_3 formation. The hourly data (August 2017 to July 2018) for 29 VOCs were obtained from three Malaysian Department of Environment continuous air quality monitoring stations with different urban backgrounds (Shah Alam, Cheras, Seremban). The Ozone Formation Potential (OFP) was calculated based on the individual Maximum Incremental Reactivity (MIR) and VOC concentrations. The results showed that the highest mean total VOC concentrations were recorded at Cheras (148 ± 123 μg m^{-3}), within the Kuala Lumpur urban environment, followed by Shah Alam (124 ± 116 μg m^{-3}) and Seremban (86.4 ± 89.2 μg m^{-3}). VOCs such as n-butane, ethene, ethane and toluene were reported to be the most abundant species at all the selected stations, with overall mean concentrations of 16.6 ± 11.9 μg m^{-3}, 12.1 ± 13.3 μg m^{-3}, 10.8 ± 11.9 μg m^{-3} and 9.67 ± 9.00 μg m^{-3}, respectively. Alkenes (51.3–59.1%) and aromatic hydrocarbons (26.4–33.5%) have been identified as the major contributors to O_3 formation in the study areas based on the overall VOC measurements. Relative humidity was found to influence the concentrations of VOCs more than other meteorological parameters. Overall, this study will contribute to further understanding of the distribution of VOCs and their contribution to O_3 formation, particularly in the tropical urban environment.

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* Corresponding author. Department of Earth Sciences and Environment, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Malaysia.
E-mail address: talib@ukm.edu.my (M.T. Latif).

1. Introduction

Atmospheric volatile organic compounds (VOCs) are a group of air pollutants that are often documented as being of global concern as they may lead to severe air pollution issues with further consequences for tropospheric ozone (O_3) formation. The composition of airborne VOCs is complex and dynamic due to the wide variety of sources and other factors, and the chemical reactivity of each species varies significantly (Zheng et al., 2014; Hui et al., 2018). The ubiquitous VOC emissions are derived mainly from two prominent sources; anthropogenic activities and natural processes (Hewitt, 1999; Tie et al., 2006; Kamal et al., 2016). Anthropogenic activities include burning fossil fuels (e.g., liquefied petroleum gas and natural gas) in power plants, fuel evaporation, motorised transportation, vehicular exhaust emissions, solvents utilisation and industrial processes, especially in urban areas (Sanchez et al., 2008; Ling et al., 2011; Zheng et al., 2018). Biomass burning is the largest emission source contributing to the global budget of anthropogenic VOCs (Karl et al., 2007; Yokelson et al., 2008). As for natural sources, plants (isoprene, monoterpene, sesquiterpene, oxygen compounds), oceans (methyl iodide, methyl nitrate, dimethyl sulfide) and soils (carbonyl compounds) also release biogenic VOCs (BVOCs) (Calfapietra et al., 2013; Guenther, 2013; Kashyap et al., 2019; Yáñez-Serrano et al., 2020). It was estimated that the global BVOC emissions were ten times higher than those of anthropogenic VOCs in the ambient air, with estimated emissions of 1150 Tg C yr^{-1} and 110 Tg C yr^{-1}, respectively (Guenther et al., 2012; Sindelarova et al., 2014).

VOCs are of particular interest as they are the key precursors to the formation of ground-level O_3 and secondary organic aerosol (SOA) through a series of photochemical reactions (Atkinson, 2000; Sahu, 2012; Li et al., 2020; Mozaffar et al., 2020). Atmospheric VOCs are removed via photochemical and deposition mechanisms over varying time periods of minutes to months (Atkinson and Arey, 2003). The interactions between O_3, oxides of nitrogen (NO_x) and VOCs are driven by complex non-linear photochemistry (Sillman and He, 2002; Cohan et al., 2005; Perera et al., 2019). As time progresses, NO_x contributions to hydroxyl radical (OH) reactivity become less important than those of proxy radicals from VOC oxidation during high O_3 episodes (Kim et al., 2018). The diverse VOC groups (i.e., aldehydes, alcohols, aromatic hydrocarbons, ketones, phenols, polymers and carbonyl compounds) reflect the differences in VOC reactivity and chemical structures towards O_3 formation (Srivastava and Mazumdar, 2011; Mellouki et al., 2015; Kamal et al., 2016; Montero-Montoya et al., 2018). Previous studies also reported that the distribution of ambient VOCs is influenced by meteorological variables such as ambient temperature, relative humidity, solar radiation, wind speed and wind direction (Lin et al., 2004; Ramsey et al., 2014; Radaideh, 2017; Manju et al., 2018; Maji et al., 2020). Besides, local time dependencies of emissions also control the diurnal distribution of VOCs, particularly in the tropical region (Sahu et al., 2016).

Tropospheric O_3 has a strong oxidising ability that has a negative effect on humans, plants, materials and the entire ecosystem (Lee et al., 1996; Ishii et al., 2007; Saarnio et al., 2008; Varotsos et al., 2012; Wilson et al., 2019). Long-term exposure to high O_3 concentrations can lead to an increase in annual morbidity and mortality rates because O_3 pollution has disproportionate effects on human health, such as pneumonia, asthma, cardiopulmonary dysfunction and respiratory diseases (Jerrett et al., 2009; Sahani et al., 2011; Lim et al., 2019; Maji and Namdeo, 2021). Using the American Cancer Society Cancer Prevention Study II (ACS CPS-II) cohort, Malley et al. (2017) estimated that approximately 1.04–1.23 million respiratory deaths in adults (<30 years old) were associated with long-term O_3 exposure. Meanwhile, many studies have also shown that O_3 pollution causes deterioration of crop yields and vegetation (Debaje, 2014; Ghude et al., 2014; Emberson et al., 2018; Agathokleous et al., 2020) with additional effects including damaged plant tissues, decrease in photosynthesis, rapid ageing and weakened biotic or abiotic defences (Dai et al., 2019).

In Asian countries, the tropospheric O_3 concentrations have increased rapidly due to the increasing trends in O_3 precursor emissions (Kulshrestha and Mishra, 2022; Tanimoto et al., 2009), as Asian megacities emit about 10–15% of the total anthropogenic VOC emissions and account for 30% of the Asian population (von Schneidemesser et al., 2010). Malaysia is one of the developing countries experiencing emerging air pollution due to the substantial anthropogenic emissions from various sectors (Sahani et al., 2016; Usmani et al., 2020). There are several studies of trace gases (O_3 and NO_x) and aerosol (PM_{10}, $PM_{2.5}$) that have been reported from different regions of Malaysia (Latif et al., 2012; Banan et al., 2013; Ahamad et al., 2014; Mohtar et al., 2018; Othman et al., 2020). According to the Malaysia Environmental Quality Report 2016 (DOE, 2017), the Klang Valley area recorded the highest O_3 concentrations in Malaysia. The deterioration of air quality due to increasing O_3 levels, particularly in the urban environment, is important and thus requires further exploration. High O_3 levels have been recorded in densely populated areas, typically due to the oxidation of high VOC concentrations (Ren et al., 2013; Tan et al., 2020). Previous studies have reported that the O_3 concentrations in urban areas in Malaysia exceeded the standard limit of 200 μg m^{-3} (100 ppbv) by the Malaysian Ambient Air Quality Standard (MAAQS) (Latif et al., 2012; Ahamad et al., 2014; Mohtar et al., 2018).

Given the importance of VOCs as the main contributors to O_3 in the atmosphere, this study aims to determine the concentrations of 29 VOCs as O_3 precursors recorded at three urban stations on the west coast of Peninsular Malaysia. The concentrations of VOCs have been associated with the formation of O_3 and the resulting concentration levels. The Maximum Incremental Reactivity (MIR) method has been used to determine the O_3 Formation Potential (OFP). The influence of ambient meteorological parameters on VOC concentrations was also determined during the observation period.

2. Methodology

2.1. Description of the study area

The study was conducted at three selected locations on the west coast of Peninsular Malaysia (Fig. 1). The three stations were Shah Alam (N03°6′16.9″, E101°33′22.3″), Cheras (N03°6′22.44″, E101°43′4.5″) and Seremban (N02°43′24.1″, E101°58′6.5″). All continuous monitoring stations were in urban environments. Shah Alam and Cheras are two areas within the vicinity of the Klang Valley, near Kuala Lumpur city centre, and Seremban is a city and state capital of Negeri Sembilan and is located to the south of Kuala Lumpur. The three stations are influenced by motor vehicle emissions, especially during rush hours in the morning and late afternoon when people commute to and from their workplaces. Due to its location within the city of Kuala Lumpur, Cheras station is most heavily affected by anthropogenic urban activities. Shah Alam station is located within residential areas surrounded by light industrial activities. All monitoring stations were situated within school compounds in residential areas surrounded by highly congested roads, especially during the rush hours.

2.2. VOC and O_3 measurements

The simultaneous data measurements of VOC concentrations, ground-level O_3 and meteorological parameters, including wind speed, relative humidity and ambient temperature, were collected from Continuous Air Quality Monitoring Stations (CAQMS) in Malaysia, operated by the Department of Environment (DOE), from August 2017 to July 2018 (12 months). Measurements of O_3 precursors were conducted using a continuous gas chromatography (GC) system GC5000 Analyzer (AMA Instruments, Germany). The instrument continuously detected 29 VOC target compounds of O_3 precursors at a temporal frequency of 1 h, consisting of eleven alkanes, nine alkenes, eight aromatics and one alkyne. The system consisted of two GC sets: GC5000 VOC for C2–C5 and GC5000 BTX for C6–C12. The GC5000 VOC analyser used

dual-column PLOT alumina columns (25 m × 0.32 mm x 0.8 μm) and AMA-Wax columns (15 m × 0.32 mm x 0.25 μm) as the backflush columns. The GC5000 BTX used capillary AMASep-1 columns (30 m × 0.32 mm x 1.5 μm) for the separation of the specific compounds. Both GC systems were equipped with thermal desorption, enrichment sorbent tube and Flame Ionization Detector (FID). The analyser was calibrated and validated every two weeks before measurements were taken. To perform a single-point calibration, a certified gas standard of 1 ppm was auto diluted with purified nitrogen to 10 ppb using the DIM 200 auto dilution system (AMA Instruments, Germany). Validation and the performance of the system were routinely checked based on the individual compound drift. During our sampling, the drift quality control (QC) evaluation was <20% for all compounds. The limit of detection (LOD) for the GC system for benzene was <0.03 ppb based on the manufacturer's certificate (PSTW, 2018a, b).

For the O_3 concentrations, the measurements were performed using a 49i-O_3 Analyzer with UV Photometric/US EPA Equivalent Method: EQOA-0880-047 principle detection (Thermo Environmental Instruments Inc., USA). Using this technique, O_3 concentrations can be measured from 0.05 ppb. Multi-point calibrations were performed bi-monthly during the sampling period. As part of the QC, auto programmed zero drift and span drift were monitored every 24 h, bypassed through the gaseous standard using the 146i multi-gas dilutor (Thermo Scientific, US). The zero drift and span drift were $< \pm 3.0$ ppb and $< \pm 7\%$, respectively, for the O_3 measurements (PSTW, 2018a, b).

2.3. Statistical data analysis

The hourly VOCs, O_3, NO_x ($NO + NO_2$) and meteorological data were processed to study the variation of the air pollutants at different time-scales. Statistical descriptive analysis, multivariate analysis and the data distribution at the three selected sampling locations were carried out using the R programming software, version 2.8.3 (Carslaw and Ropkins, 2012) and Statistical Package for Social Sciences software (SPSS version 28). All data are reported as mean ± standard deviation, unless otherwise indicated. Unit conversions for VOC, O_3 and NO_x concentrations (from ppb to μg m^{-3}) were also carried out. Any missing values from the study period were not considered in the computation of the statistics.

The Kolmogorov-Smirnov test and analysis of variance (Kruskal-Wallis *H* test) with pairwise comparisons using Dunn's Post Hoc test were employed to examine the normality of the data and the significant difference in VOC concentrations between the sampling sites. The non-normal distribution value was found to be $p < 0.05$ for all data and there was a statistically significant difference between the three stations ($p < 0.001$). In addition, Spearman's rank correlation analysis was also used to measure the monotonic association between the O_3 and OFP as well as the O_3 and NO_x concentrations. The interpretation of the Spearman correlation coefficient was scaled from -1 to $+1$. The Spearman correlation was calculated as shown in Eq. (1):

$$r_s = 1 - \frac{6 \sum d_i^2}{n(n^2 - 1)} \qquad \text{(Eq. 1)}$$

r_s = Spearman's rank correlation coefficient
d_i = Difference between the two ranks of each observation
n = number of observations.

2.4. Calculation of ozone formation potential (OFP)

The photochemical OFP was calculated based on the updated MIR scale that was provided by Carter (2010). This reactivity scale was established by Carter (1994) as a mechanism for quantifying the effects of various VOC compounds on O_3 formation and under various atmospheric conditions where O_3 is most sensitive to changes in VOC levels in the atmosphere. The formula can be calculated based on the following Eq. (2):

$$OFPi = Con(VOCi) \times MIRi \qquad \text{(Eq. 2)}$$

Where OFPi (μg m^{-3}) is defined as O_3 formation potential of the individual VOC hydrocarbon i, whereas Con(VOCi) is the concentration of the VOCi compound (μg m^{-3}) and MIRi (g O_3/g VOC) is the maximum incremental reactivity coefficient of compound i.
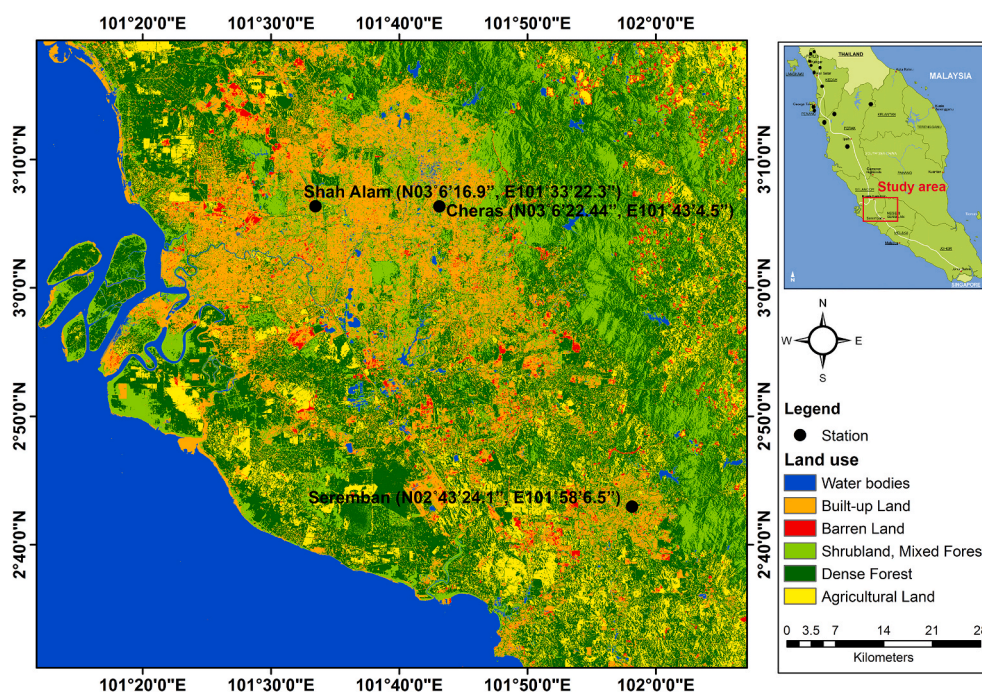

Fig. 1. Location of sampling stations (Shah Alam, Cheras and Seremban) in Peninsular Malaysia.

3. Results and discussion

3.1. Descriptive analysis of VOC concentrations

A descriptive analysis of the 29 measured VOC concentrations for Shah Alam, Cheras and Seremban stations during the study is presented in Table 1. The highest mean concentrations of total volatile organic compound (TVOC) levels were observed at Cheras (148 ± 123 µg m^{-3}), followed by Shah Alam (124 ± 116 µg m^{-3}) and Seremban (86.4 ± 89.2 µg m^{-3}). TVOC concentrations were relatively higher at Cheras compared to the other stations ($p < 0.05$) due to its location within the Kuala Lumpur urban environment and it being surrounded by roads and major highways with heavy traffic volume. Overall, the VOCs measured were dominated by alkanes (64.0 ± 54.9 µg m^{-3}, 53.8%), followed by alkenes (25.2 ± 27.9 µg m^{-3}, 21.1%), aromatics (24.5 ± 23.7 µg m^{-3}, 20.6%) and then alkyne (5.90 ± 3.20 µg m^{-3}, 5.0%).

Of the 29 individual VOCs measured, n-butane, ethane, ethene and toluene were reported as the most abundant species at all the selected stations, with overall mean concentrations of 16.6 ± 11.9 µg m^{-3}, 12.1 ± 13.3 µg m^{-3}, 10.8 ± 11.9 µg m^{-3} and 9.67 ± 9.00 µg m^{-3}, respectively. These light hydrocarbon fractions are released from similar anthropogenic sources, including liquefied petroleum gas (LPG) and natural gas productions, fuelled vehicle emissions and biomass burning (Tsai et al., 2006; Stauffer et al., 2008; Ying and Krishnan, 2010; Thompson et al., 2014; Rossabi and Helmig, 2018). Toluene was the most predominant species among the aromatic hydrocarbons measured in this study and is widely used as a gasoline additive, a paint solvent, and in printing inks (Kansal, 2009; Garzón et al., 2015). According to a prior study by Han et al. (2015), C3–C6 alkanes, benzene, toluene, ethylbenzene and xylene, constitute the majority of internal combustion engine emissions of VOCs. Motor vehicles were expected to contribute to the high amount of toluene in ambient air, as shown by previous studies of benzene, toluene, ethylbenzene and xylene (BTEX) in Malaysia (Latif et al., 2019). Cheras station, which is located within a busy area with high traffic volume, recorded the highest concentration of toluene (14.2 ± 10.4 µg m^{-3}).

The concentration of overall VOCs based on different groups has been compared to the results of other studies conducted in other urban environments, as shown in Supplementary 1a–1d. In general, alkanes recorded in this study were found to be higher compared to the studies conducted by Jia et al. (2016) in Lanzhou (China), Huang and Hsieh (2019) in Taichung (Taiwan), Wang et al. (2019) in Jinan (China) and Bari and Kindzierski (2018) in Alberta (Canada). The concentrations of alkanes recorded in this study (Supplementary 1a) were found to be lower compared to the study by Santos and Azevedo (2020) in a road tunnel in the urban tropical city of Rio de Janeiro, Brazil. Comparisons with another study by Ly et al. (2020) in Hanoi (Vietnam) shows that the concentrations of 2,2,4-trimethylpentane, n-hexane, n-heptane and n-octane recorded in Hanoi were far higher compared to the alkanes recorded in this study. This comparison indicated that the alkane concentrations are mostly contributed to by vehicles emission and other sources that may be influenced by tropical hot conditions.

The comparison between alkenes recorded in this study and other similar studies shows the trend of alkenes following the trend of alkanes (Supplementary 1b). Nevertheless, the concentrations of aromatic VOCs especially benzene recorded in this study (Supplementary 1c) were far below the concentrations of aromatics VOCs recorded in Hanoi, Vietnam by Ly et al. (2020) and Lanzhou, China by Jia et al. (2016). The measurement of VOCs recorded in Hanoi used a sorbent tube via active sampling and the sampling locations were very near to the roadside. The measurement of VOCs in Lanzhou, China by Jia et al. (2016) was conducted in a petrochemical city via an online gas chromatography system. The concentrations of aromatic VOCs recorded in this study are almost similar to the aromatic VOCs recorded by Huang and Hsieh (2019) in Taichung, Taiwan, Wang et al. (2019) in Jinan, China, and Santos et al. (2020) in Rio de Janeiro, Brazil. Toluene was found in abundance

Table 1
Description of the mean ($+/-$ standard deviation) of VOCs concentration for Shah Alam, Cheras and Seremban stations (n = 365).

Parameter	Shah Alam (µg m^{-3})	Cheras (µg m^{-3})	Seremban (µg m^{-3})	Mean (µg m^{-3})
Ethane	12.7 ± 11.5	12.9 ± 14.0	6.90 ± 10.1	10.8 ± 11.9
Propane	5.19 ± 5.59	10.7 ± 7.44	5.33 ± 4.74	7.07 ± 5.92
n-Butane	17.3 ± 12.5	19.6 ± 12.7	12.8 ± 10.6	16.6 ± 11.9
i-Butane	10.2 ± 8.35	11.3 ± 8.71	5.56 ± 5.45	9.02 ± 7.50
n-Pentane	3.74 ± 3.16	4.85 ± 3.64	2.83 ± 2.31	3.81 ± 3.03
i-Pentane	8.38 ± 7.34	10.5 ± 8.31	4.90 ± 6.43	7.94 ± 7.36
n-Hexane	2.31 ± 1.94	2.82 ± 2.18	1.43 ± 1.25	2.18 ± 1.79
2-Methylpentane	4.05 ± 3.66	5.05 ± 4.08	2.79 ± 2.34	3.96 ± 3.36
n-Heptane	1.39 ± 0.984	1.35 ± 0.931	0.796 ± 0.611	1.18 ± 0.842
n-Octane	0.867 ± 0.801	0.631 ± 0.522	0.325 ± 0.293	0.608 ± 0.539
2,2,4-Trimethylpentane	1.12 ± 0.921	0.955 ± 0.822	0.430 ± 0.461	0.834 ± 0.735
Alkanes (Total)	**67.2 ± 56.7**	**80.7 ± 63.3**	**44.1 ± 44.6**	**64.0 ± 54.9**
Ethene	14.3 ± 12.9	14.4 ± 15.7	7.75 ± 11.7	12.1 ± 13.3
Propene	5.30 ± 4.67	5.60 ± 5.66	1.96 ± 2.77	4.28 ± 4.37
1-Butene	1.42 ± 1.30	1.55 ± 1.18	0.565 ± 0.562	1.18 ± 1.01
trans-2-Butene	1.19 ± 0.972	1.34 ± 0.996	0.428 ± 0.405	0.992 ± 0.791
cis-2-Butene	1.92 ± 3.25	2.54 ± 2.85	2.44 ± 3.30	2.30 ± 3.13
1,3-Butadiene	1.13 ± 0.858	0.993 ± 0.702	0.623 ± 0.445	0.915 ± 0.668
1-Pentene	1.53 ± 3.37	1.68 ± 3.00	0.707 ± 2.55	1.31 ± 2.97
trans-2-Pentene	0.840 ± 0.639	0.879 ± 0.674	0.357 ± 0.297	0.692 ± 0.537
Isoprene	1.24 ± 0.990	1.41 ± 0.963	1.50 ± 1.24	1.38 ± 1.06
Alkenes (Total)	**28.8 ± 28.9**	**30.4 ± 31.7**	**16.3 ± 22.9**	**25.2 ± 27.9**
Benzene	2.13 ± 2.30	2.60 ± 2.40	2.42 ± 1.71	2.38 ± 2.14
Toluene	8.05 ± 10.0	14.2 ± 10.4	6.79 ± 6.55	9.67 ± 9.00
Ethylbenzene	4.62 ± 5.50	3.79 ± 3.34	1.99 ± 2.52	3.47 ± 3.79
m,p-Xylene	3.09 ± 4.00	4.17 ± 3.51	2.81 ± 3.07	3.36 ± 3.58
o-Xylene	1.59 ± 1.88	2.93 ± 2.04	1.52 ± 1.28	2.01 ± 1.73
1,2,3-Trimethylbenzene	0.759 ± 0.719	0.688 ± 0.706	0.786 ± 0.731	0.744 ± 0.719
1,2,4-Trimethylbenzene	2.13 ± 2.12	1.22 ± 1.28	1.88 ± 1.72	1.74 ± 1.71
1,3,5-Trimethylbenzene	1.22 ± 1.12	1.18 ± 1.21	1.05 ± 0.922	1.15 ± 1.08
Aromatics (Total)	**23.6 ± 27.7**	**30.7 ± 24.9**	**19.2 ± 18.5**	**24.5 ± 23.7**
Acetylene	4.84 ± 3.03	6.12 ± 3.35	6.74 ± 3.21	5.90 ± 3.20
Alkyne (Total)	**4.84 ± 3.03**	**6.12 ± 3.35**	**6.74 ± 3.21**	**5.90 ± 3.20**
Total VOC (TVOC)	**124 ± 116**	**148 ± 123**	**86.4 ± 89.2**	**119 ± 110**

compared to other aromatic VOCs compounds in Taichung, Taiwan, due to evaporation, particularly from motor vehicle emissions, surface coatings and other uses of toluene solvent (Huang and Hsieh, 2019). There are not many studies that have determined concentrations of acetylene (Supplementary 1d) but, based on the data available, the average concentration of acetylene recorded in this study was higher compared to the studies by Huang and Hsieh (2019), Wang et al. (2019) and Bari and Kindzierski (2018).

3.2. Monthly mean TVOC and different groups of VOC concentrations

The monthly means of the collective TVOC concentrations are shown in Fig. 2. The concentrations of TVOCs at Shah Alam and Cheras were found to be at higher concentrations compared to Seremban for the whole year. The concentrations at these two stations showed higher concentrations in September during the southwest monsoon. TVOC in Shah Alam reduced significantly during inter monsoon seasons in October. The concentrations of TVOCs at Cheras station continued to increase between October to January during the northeast monsoon. The results may reflect meteorological factors such as wind directions, temperature and relative humidity in these three areas. The southwest monsoon in Malaysia is usually associated with the dry period and transboundary emissions from biomass burning areas, while the northeast monsoon is associated with the rainy season in the beginning, followed by dry conditions (Mohtar et al., 2018). All these factors, in addition to the wind movement from the Kuala Lumpur city centre, especially during the northeast monsoon, may contribute to the high concentrations of TVOCs, especially at the Cheras station.

The monthly mean VOC concentrations based on the different groups are shown in Fig. 3 according to their groups (i.e., alkanes, alkenes, aromatics and alkyne). For Shah Alam station, the most abundant VOC groups recording the highest monthly mean concentrations were alkanes and alkenes, ranging from 18.4 to 86.8 $\mu g\ m^{-3}$. The alkanes and aromatics showed an increasing trend between May and September during the southwest monsoon and the northeast monsoon season (November to early March). The monthly mean concentrations of alkyne were the lowest and showed consistent values of 0.951–7.19 $\mu g\ m^{-3}$. The source of alkyne (acetylene) in ambient air is commonly incomplete combustion (Xiao et al., 2007; Li et al., 2018). The concentration of acetylene in ambient air might be relatively low due to its lower MIR reactivity or having lower OH reaction rates (Huang and Hsieh, 2019).

For Cheras station, alkanes, aromatics and alkenes showed the highest concentrations throughout the months, ranging from 20.2 to 98.7 $\mu g\ m^{-3}$ (Fig. 3). The monthly mean patterns of alkanes, alkenes and aromatics showed increasing trends between the southwest monsoon season (May to September) and the northeast monsoon season
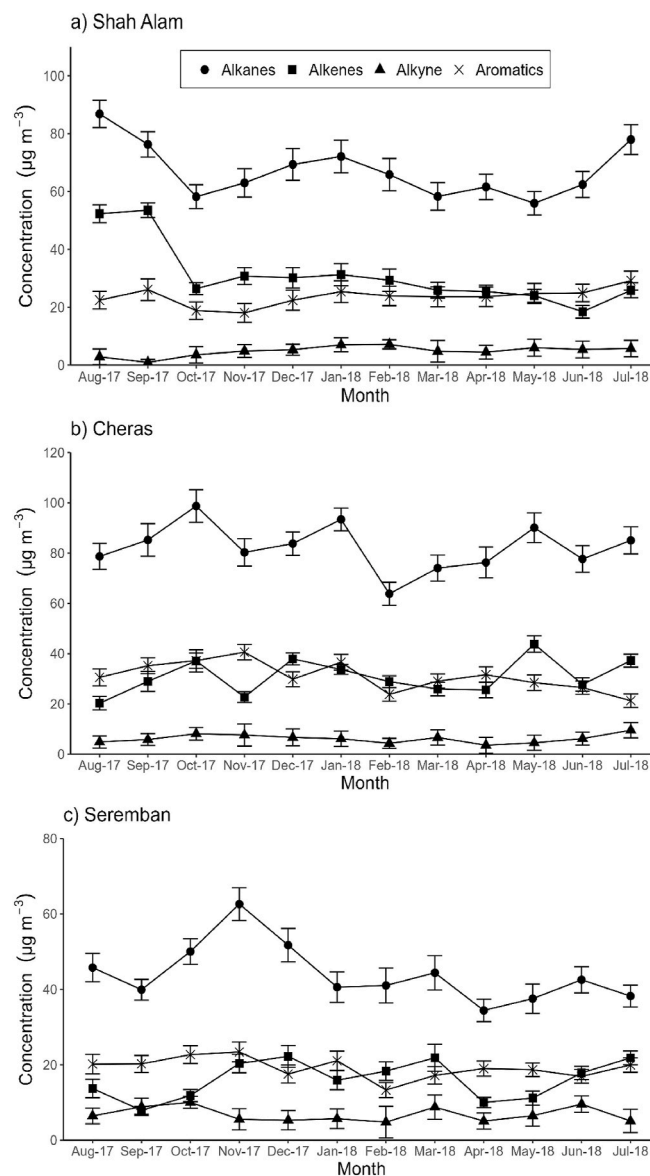

Fig. 3. Monthly mean VOC concentrations of each chemical group (alkanes, alkenes, aromatics and alkyne) measured at (a) Shah Alam, (b) Cheras and (c) Seremban stations (n = 12).

(November to early March). The monthly mean concentrations of acetylene were consistently between 3.59 and 9.57 $\mu g\ m^{-3}$. At Seremban station, alkanes showed the highest concentration during November with a concentration value of 62.6 $\mu g\ m^{-3}$. The VOC groups at that station showed a significant increasing trend between May and September during the southwest monsoon and the northeast monsoon seasons from November to early March. Overall, Shah Alam and Seremban stations showed a similar monthly trend of alkanes > alkenes > aromatics > alkyne during the sampling period.

3.3. Ozone formation potential (OFP)

The total OFP was calculated according to the mean concentrations of different VOC groups during the observation period at each of the selected stations (Table 2). The OFP is categorised into alkanes, alkenes, aromatics and an alkyne, to identify which hydrocarbon groups could theoretically contribute to the formation of VOC-generated O_3. Overall, this study demonstrated that the highest total OFP value was detected at Cheras (569 $\mu g\ m^{-3}$), followed by Shah Alam (502 $\mu g\ m^{-3}$), and
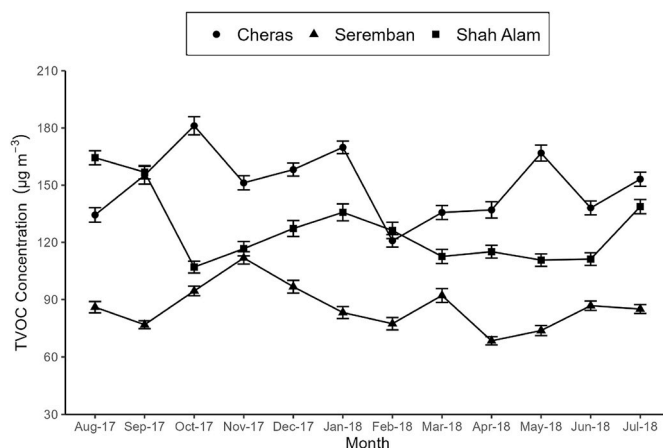

Fig. 2. Monthly mean of total volatile organic compounds (TVOC) concentrations at Shah Alam, Cheras and Seremban stations (n = 12).

Table 2
Maximum Incremental Reactivity (MIR) scales and Ozone Formation Potential (OFP) for 29 measured VOCs compound at Shah Alam, Cheras and Seremban stations (n = 365).

Parameter	MIR	Shah Alam		Cheras		Seremban	
	(g O_3/g VOC)	OFP ($\mu g\ m^{-3}$)	OFP (%)	OFP ($\mu g\ m^{-3}$)	OFP (%)	OFP ($\mu g\ m^{-3}$)	OFP (%)
Ethane	0.280	3.56	0.708	3.60	0.632	1.93	0.576
Propane	0.490	2.54	0.506	5.25	0.921	2.61	0.778
n-Butane	1.15	19.9	3.96	22.6	3.97	14.8	4.40
i-Butane	1.23	12.5	2.49	13.9	2.45	6.84	2.04
n-Pentane	1.31	4.90	0.98	6.35	1.12	3.71	1.11
i-Pentane	1.48	12.4	2.47	15.6	2.73	7.26	2.16
n-Hexane	1.24	2.86	0.569	3.49	0.614	1.77	0.528
2-Methylpentane	1.50	6.08	1.21	7.58	1.33	4.18	1.25
n-Heptane	1.07	1.49	0.296	1.45	0.254	0.852	0.254
n-Octane	0.900	0.780	0.155	0.568	0.100	0.292	0.087
2,2,4-Trimethylpentane	1.26	1.41	0.280	1.20	0.211	0.542	0.161
Alkanes (Total)	-	**68.4**	**13.6**	**81.6**	**14.3**	**44.7**	**13.3**
Ethene	9.00	128	25.5	130	22.8	69.7	20.8
Propene	11.7	62.0	12.3	65.5	11.5	22.9	6.84
1-Butene	9.73	13.9	2.76	15.1	2.64	5.50	1.64
trans-2-Butene	15.2	18.1	3.60	20.6	3.62	6.50	1.94
cis-2-Butene	14.2	27.3	5.44	36.1	6.34	34.7	10.3
1,3-Butadiene	12.6	14.2	2.83	12.5	2.20	7.85	2.34
1-Pentene	7.21	11.0	2.19	12.1	2.13	5.10	1.52
trans-2-Pentene	10.6	8.91	1.77	9.32	1.64	3.79	1.13
Isoprene	10.6	13.6	2.62	15.0	2.63	15.9	4.73
Alkenes (Total)	-	**297**	**59.1**	**316**	**55.5**	**172**	**51.3**
Benzene	0.720	1.53	0.305	1.87	0.328	1.74	0.519
Toluene	4.00	32.2	6.41	56.6	9.94	27.2	8.10
Ethylbenzene	3.04	14.1	2.80	11.5	2.03	6.04	1.80
m,p-Xylene	9.75	30.1	6.00	40.7	7.14	27.4	8.16
o-Xylene	7.64	12.1	2.42	22.4	3.94	11.6	3.45
1,2,3-Trimethylbenzene	12.0	9.11	1.81	8.25	1.45	9.43	2.81
1,2,4-Trimethylbenzene	8.87	18.9	3.77	10.8	1.90	16.7	4.97
1,3,5-Trimethylbenzene	11.8	14.4	2.87	13.9	2.44	12.3	3.68
Aromatics (Total)	-	**133**	**26.4**	**166**	**29.2**	**112**	**33.5**
Acetylene	0.950	4.60	0.915	5.81	1.02	6.40	1.90
Alkyne (Total)	-	**4.60**	**0.915**	**5.81**	**1.02**	**6.40**	**1.90**
OFP (Total)	-	**502**	**100**	**569**	**100**	**335**	**100**

Seremban (335 $\mu g\ m^{-3}$). The location of Cheras and Shah Alam stations are situated in densely urbanised areas and near congested roads where there is an excessive amount of potential sources of emissions of VOC compounds.

The overall outcomes also indicated that the OFP was in the order of alkenes > aromatics > alkanes > alkyne in all study areas. Alkanes and the alkyne contribute less to the O_3-forming potential due to their low MIR reactivity. Alkenes and aromatic hydrocarbons are the critical species that influence the total OFP for all three stations. This could be due to the MIR contribution of alkenes and aromatics being much higher compared with the other VOC groups. Ethene, *cis*-2-butene and propene were the most abundant species from the alkenes, whereas toluene was the largest species from the aromatics group. Alkenes are the by-product of evaporative sources, and aromatics are generally emitted from combustion. Those previously mentioned species can contribute to the OFP due to their capability to exhibit higher photochemical reactions (Hui et al., 2018, 2020).

Based on the results obtained for Shah Alam station, the alkenes and aromatic hydrocarbon groups were the major contributors to the formation of O_3, accounting for 59.1% and 26.4%, respectively. The species that recorded high OFP concentrations from the alkenes group were propene (62.0 $\mu g\ m^{-3}$), ethene (128 $\mu g\ m^{-3}$) and *cis*-2-butene (27.3 $\mu g\ m^{-3}$). As for the aromatic hydrocarbons, toluene (32.2 $\mu g\ m^{-3}$) was the most dominant species, followed by m,p-xylene (30.1 $\mu g\ m^{-3}$) and 1,2,4-trimethylbenzene (18.9 $\mu g\ m^{-3}$). At Cheras station, alkenes (55.5%) and aromatic hydrocarbons (29.2%) were also listed as the major contributors to the formation of O_3. The VOCs species with the highest OFP values from the alkenes group, were ethene (130 $\mu g\ m^{-3}$), propene (65.5 $\mu g\ m^{-3}$) and *cis*-2-butene (36.1 $\mu g\ m^{-3}$). For the aromatic compounds, it was found that toluene (56.6 $\mu g\ m^{-3}$), m,p-xylene (40.7 μg

m^{-3}) and o-xylene (22.4 $\mu g\ m^{-3}$) were recorded with the highest OFP values. The number of OFP TVOCs recorded at Cheras station was found to be 569 $\mu g\ m^{-3}$.

Similar to Shah Alam and Cheras stations, alkenes and aromatics carbon groups were also recorded as the highest OFP values at Seremban station, accounting for 51.3% and 33.5%, respectively. It was found that ethene (69.7 $\mu g\ m^{-3}$), *cis*-2-butene (34.7 $\mu g\ m^{-3}$) and propene (22.9 $\mu g\ m^{-3}$) were the most abundant from the alkene group, whereas m,p-xylene (27.4 $\mu g\ m^{-3}$), toluene (27.2 $\mu g\ m^{-3}$) and 1,2,4-trimethylbenzene (16.7 $\mu g\ m^{-3}$) were the major contributors for the aromatic carbon group. The total number of OFP TVOCs at Seremban Station was found to be 335 $\mu g\ m^{-3}$. Moreover, the results demonstrated that isoprene also contributed to the high OFP value, particularly at the Seremban station (15.9 $\mu g\ m^{-3}$). Isoprene is represented as the only biogenic volatile hydrocarbon in this study, and is largely emitted by trees and plants due to their large leaf photosynthetic capacity (Hanif et al., 2021). Isoprene is significantly associated with OFP, as it can react rapidly with atmospheric hydroxyl radicals (OH) (Ciccioli et al., 2014). For instance, oxidation of isoprene by OH radicals may produce hydroperoxides (RO_2), which can convert NO to NO_2, thus resulting in more O_3 production (Sharkey et al., 2007). The isoprene emission rate factors in tropical areas are higher than the other biomes (Guenther et al., 1995; Varshney and Singh, 2003; Taylor et al., 2018; Šimpraga et al., 2019). The findings can be further strengthened by a study conducted by Guenther et al. (2012), which stated that tropical trees contribute the highest (80%) of global annual BVOC emissions (including isoprene emissions), while both boreal and temperate trees accounted for only 10%. Therefore, this could be a probable explanation for why isoprene emissions are considered high in this study, given that all sample stations are located in tropical urban areas of Malaysia.

Our results are supported by previous studies that showed the alkane and aromatic fractions play a fundamental role in the formation of O_3. For instance, a similar trend related to the O_3-forming potential with aromatics compounds, particularly benzene, toluene, ethylbenzene, and xylene (BTEX), was found in Kuala Lumpur, Malaysia (Hamid et al., 2019) and New Delhi, India (Garg and Gupta, 2019). Both of the studies computed that the mean OFP of BTEX was $278.42 \pm 74.64 \, \mu g \, m^{-3}$ and $207.51 \pm 123.40 \, \mu g \, m^{-3}$, respectively. Kumar et al. (2018) also revealed that toluene and m,p-xylene were the major contributors among the aromatics species that influenced the O_3 formation in urban and rural sub-tropical sites in India. Similarly, a recent study by Mehta et al. (2020) in New Delhi, India, claimed that xylenes (m,p-xylene and o-xylene) and toluene possessed higher levels of OFP with mean values of 239.4, 188.9 and $98.2 \, \mu g \, m^{-3}$, respectively. In China (Foshan), alkenes (~49.5%) and aromatics (~28%) dominated the OFP with a total value of $863.4 \, \mu g \, m^{-3}$ (Tan et al., 2012). Huang et al. (2011) also observed that ethylene, o-xylene, toluene, 1,2,4-trimethylbenzene, ethylbenzene, propylene, 1-pentene and isoprene were the dominant contributors to OFP (77%) in the Yangtze River Delta area, China. Another comparable study in a sub-tropical urban city in Hanoi, Vietnam by Do et al. (2013) reported that the value of OFP was $1308 \, \mu g \, m^{-3}$. A recent study by Luo et al. (2020) in Guangzhou, China stated that the dominant VOC OFP groups (aromatic and aliphatic) with the total OFP ranged from 64.7 to $1165.3 \, \mu g \, m^{-3}$. Tunsaringkarn et al. (2014) found that roadside areas ($932.7 \, \mu g \, m^{-3}$) recorded a higher level of OFP concentrations compared with residential areas ($436.3 \, \mu g \, m^{-3}$) in Bangkok, Thailand. Compared with the previously mentioned Asian countries, the highest mean OFP calculated from the aromatic VOCs was observed in an urban street site in Dhaka, Bangladesh, with the value of $2200 \, \mu g \, m^{-3}$ (Do et al., 2015). The high levels of detected OFP in large urban areas are often linked to the exacerbation of O_3 pollution due to the VOC-sensitive regime present (Huy and Kim Oanh, 2020).

3.4. Correlation between OFP TVOCs, NO_x and O_3 concentrations

The Spearman correlation was applied to correlate the OFP of 29 TVOC parameters with ground-level O_3 at Shah Alam, Cheras and Seremban stations (Fig. 4). The correlation values of OFP TVOC and O_3 concentrations at Cheras station showed a positive r_s-value (0.11) with a p-value of less than 0.05, whereas the correlation values between OFP TVOCs and O_3 obtained at Shah Alam and Seremban were $r_s = -0.04$, $p > 0.05$ and $r_s = 0.02$, $p > 0.05$. The results from Cheras indicated a statistically significant correlation, which was interpreted as the maximum O_3 concentration increased as the OFP TVOC value increased. According to Wu et al. (2017), O_3 is susceptible to transportation, domestic solvent usage and open burning regarding the O_3-forming potential per unit VOC concentration. Numerous consistent studies showed that the VOC composition could cause tropospheric O_3 formation, enhance the atmospheric circulation by participating in photochemical processes and thus result in the production of other secondary pollutants (Carter, 1994; Song et al., 2007; Madronich et al., 2015; Mozaffar and Zhang, 2020; Zhang et al., 2020).

The correlation between OFP of individual VOC concentrations with maximum ground-level O_3 concentration at all stations was also discussed in this observation period (Supplementary 2). This correlation analysis was used to interpret, and provide a better understanding of which individual VOC compounds dominantly contributed to the photochemical O_3 at the selected sampling locations. Based on the results from Cheras station, the OFP from VOCs which have good correlations ($p < 0.05$) with the O_3 maximum is far higher compared to the other two stations. cis-2-Butene showed the strongest correlation (r_s-value more than 0.7) between its OFP and O_3 maximum ($r_s = 0.82$, $p < 0.01$), followed by propene ($r_s = 0.78$, $p < 0.01$), 1-3-butadiene ($r_s = 0.75$, $p < 0.01$) and 1-butene ($r_s = 0.74$, $p < 0.01$). Isoprene, which is usually associated with vegetation sources, also indicated a good correlation between its OFP and O_3 maximum ($r_s = 0.43$, $p < 0.01$). In
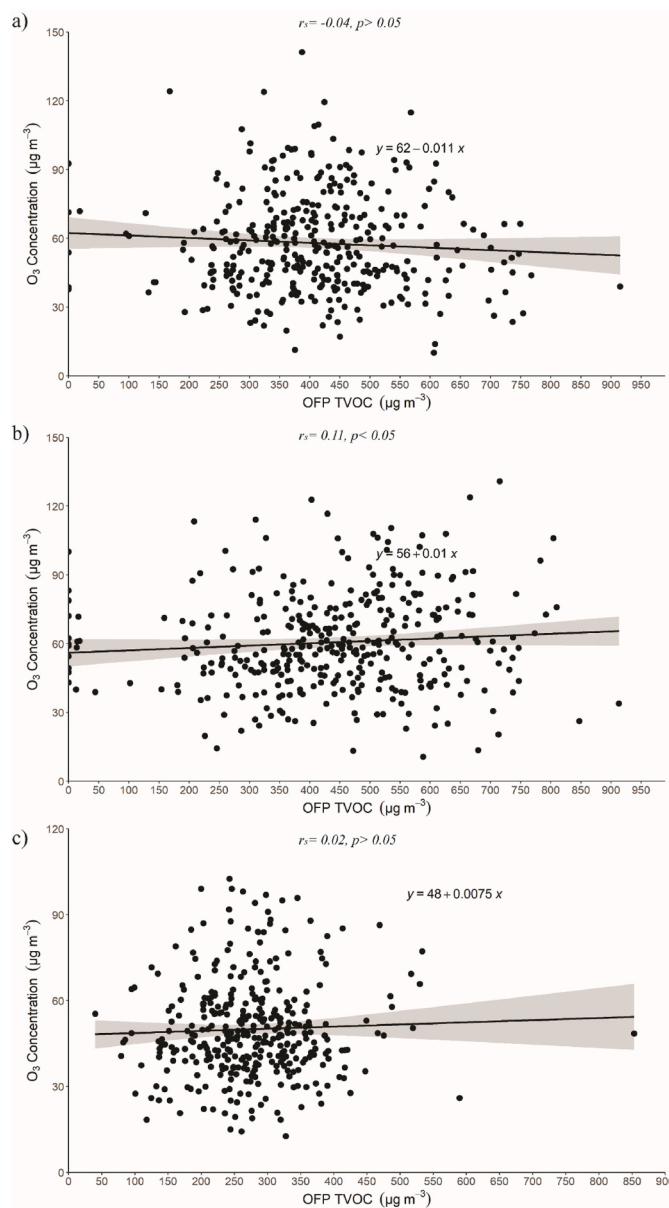

Fig. 4. The correlation between OFP TVOC and O3 concentration at (a) Shah Alam (b) Cheras and (c) Seremban stations during the observation period.

contrast, there was no significant correlation between OFP isoprene and O_3 concentration at Shah Alam and Seremban stations, with $r_s = 0.03$ ($p > 0.05$) and $r_s = 0.09$ ($p > 0.05$), respectively. The results of the correlation between individual OFP showed that VOCs from both anthropogenic and natural sources significantly contribute to the formation of O_3 maximum in Cheras compared to Shah Alam and Seremban stations.

To indicate the contribution of other O_3 precursors, especially NO_x, we have presented the correlation between NO_x and O_3 concentrations in Supplementary 3. The results showed the concentrations of NO_2 have significant positive correlations with the O_3 concentrations at Shah Alam ($r_s = 0.22$ ($p < 0.01$)) and Cheras ($r_s = 0.14$ ($p < 0.01$)) stations. At the same time, the NO concentrations showed negative but not significant correlations with O_3 concentrations at Shah Alam ($r_s = -0.04$, ($p > 0.05$)) and Cheras ($r_s = -0.01$ ($p > 0.05$)) stations. The concentrations of NO at Seremban station showed significant negative correlations to the O_3 concentrations ($r_s = -0.26$ ($p < 0.01$)). These results followed the results of previous studies which suggested NO_2 may contribute to the formation of O_3 as shown by correlations between NO_2 and O_3 at Shah

Alam and Cheras stations. NO reduces the concentration of O_3 due to titration processes as indicated by its correlation to O_3 at Seremban station. The results of the NO_x and O_3 correlation show that NO_2 influences the concentration of O_3 together with VOCs, especially at the Cheras and Shah Alam stations.

3.5. Correlation between TVOCs and meteorological variables

The preliminary analysis of meteorological parameters recorded at Shah Alam, Cheras and Seremban are presented in Supplementary 4. The correlation values for the three stations (Shah Alam, Cheras and Seremban) compared with TVOC concentrations corresponding to the meteorological variables such as wind speed, relative humidity and temperature are presented in Supplementary 5. The analysis obtained at Shah Alam station detected that the correlations between meteorological variables and TVOCs were not statically significant. For Cheras station, relative humidity showed a significant moderate correlation of $r_s = 0.443$ with a p-value less than 0.01. The Spearman correlation values at that station indicated that both temperature and wind speed had weak-moderate negative correlations of $r_s = -0.357$ and $r_s = -0.451$ ($p < 0.01$). A weak positive correlation between the concentration of TVOCs with relative humidity at Seremban station was also observed as $r_s = 0.141$ ($p < 0.01$), while wind speed obtained an inverse weak correlation value of $r_s = -0.272$ with a p-value less than 0.01. Herein, all correlation coefficients were regarded as weak-moderate and scattered. This might be due to meteorological conditions not directly influencing the VOCs in the study areas.

The correlation coefficients reported above show the association between the TVOCs and the meteorological variables. However, the air temperature, relative humidity and winds are related to each other and their compounded effect on TVOCs may be critically important. In order to examine the combined effect of temperature, relative humidity and wind speed on TVOCs, a multiple linear regression analysis was carried out by regressing the daily TVOCs on air temperature, relative humidity and average wind speed at the three stations, separately. Table 3 shows the regression coefficients and the intercepts computed at the three stations. The results indicate that TVOCs at Cheras station are affected by a strong meteorological influence as the meteorological variables explained ~36% ($R = 0.610$) of the day-to-day variance in the TVOCs. All three meteorological variables contributed significantly to the variance. This is followed by Seremban station where meteorology may have explained ~15% ($R = 0.380$) of the variance in TVOCs. However, this is largely contributed by the wind speed, and less so by the other two variables. Nevertheless, there is no statistically significant influence of meteorology on TVOCs when the three meteorological variables are analysed in coherence. This could be because Cheras and Seremban stations are located in the city centre and housing areas, where VOC sources are mainly contributed to by vehicles, which have peak activity in the morning and evening within a few hours. Station Shah Alam, on the other hand, is located in an industrial zone with a lot of point sources. As according to Do et al. (2013), other affecting factors such as road conditions, and possible other non-traffic sources of VOCs and meteorological conditions (season, wind speed, etc.), might also influence the level of VOCs. Therefore, the industrial and continual emitting sources may have masked the rather weak influence of meteorology on

the released VOCs.

Another interesting result of the regression analysis is the positive regression coefficients of air temperature, indicating higher temperatures result in higher TVOCs. For instance, at Cheras, a 1 °C increase in temperature is associated with a 12.5-unit (which is ~10% of the average TVOCs) increment in TVOCs. Nevertheless, the individual correlation coefficient between TVOCs and air temperature suggests a negative association. Therefore, the relative humidity (and potentially wind speed) is a crucial control for the association between temperature and TVOCs. It is known that the association between VOCs and temperature shows high non-linearity (Van der Wal et al., 1997; Song et al., 2019). Increased temperature may increase the evaporative emission of VOCs but at the same time also increase the decaying rate of VOCs. The results highlight the importance of considering the combined effect of meteorological elements in understanding the relationship between meteorology and air quality.

4. Conclusions

This study identified a total of 29 VOC compounds and the OFPs were measured from August 2017 to July 2018 at three selected continuous sampling stations in Malaysian urban tropical areas. Of the three sampling stations, the highest TVOC levels were observed at Cheras station (148 ± 123 µg m^{-3}), followed by Shah Alam (124 ± 116 µg m^{-3}) and Seremban (86.4 ± 89.2 µg m^{-3}). The VOC compounds were dominated by alkanes (64.0 ± 54.9 µg m^{-3}, 53.8%) and among individual compounds dominating the VOCs in the study areas are n-butane, ethane, ethene and toluene. Alkenes (51.3–59.1%) and aromatic hydrocarbons (26.4–33.5%) were the main contributors to O_3 formation. Based on OFP TVOC results, Cheras (569 µg m^{-3}) had the highest OFP values, followed by Shah Alam (502 µg m^{-3}) and Seremban (335 µg m^{-3}). From the observations, there was a statistically significant difference between the OFP TVOCs and O_3 concentrations at Cheras station. The study also suggested that meteorological factors, particularly relative humidity, influenced the distribution of VOC concentrations in ambient air. We also suggest that VOC concentrations contribute to the formation of surface O_3 in ambient air. Based on the location of the study areas and the highest concentrations of TVOCs and O_3 being recorded at Cheras station, emissions from motor vehicles would be expected to contribute to the high VOC concentrations that eventually produce surface O_3 due to photochemical interactions. To reduce VOCs and O_3 in ambient air, particularly in urban areas, the emission of VOCs, especially from motor vehicles, needs to be controlled. Future study on the influence of VOCs on O_3 concentration needs to consider the different scenarios of NO_x and VOC mixtures via the calculation of Maximum Ozone Incremental Reactivity (MOIR) and Equal Benefits Incremental Reactivity (EBIR), respectively. Additional research on relationships between VOC, NO_x and O_3 needs to incorporate meteorological factors and transboundary regional emissions.

Credit author statement

Mohd Faizul Hilmi Zulkifli: Conceptualization, Writing – original draft. **Nor Syamimi Sufiera Limi Hawari:** Writing – original draft. **Mohd Talib Latif:** Writing – review & editing, Supervision. **Haris**

Table 3

The regression coefficients obtained from the regression of TVOC concentrations on the air temperature, relative humidity and wind speed at the three stations. The R is the correlation coefficient between the observed TVOC and the predicted TVOC.

Stations	Air Temperature	Relative Humidity	Wind Speed	Intercept	R
Shah Alam	−5.48	−0.720	−6.34	338*	0.120
Cheras	12.5**	3.91**	−55.9**	−439**	0.610**
Seremban	2.17	0.490	−32.6**	−5.41	0.380**

Note: Significant values at 0.05 level are indicated **.

Hafizal Abd Hamid: Writing – review & editing, Supervision. **Anis Asma Ahmad Mohtar**: Writing – review & editing. **Wan Mohd Razi Wan Idris**: Writing – review & editing. **Nur Ili Hamizah Mustaffa**: Writing – review & editing, Supervision. **Liew Juneng**: Writing – review & editing, Supervision.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2022.134852.

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