

SPATIOTEMPORAL VARIABILITY OF INDOOR ATMOSPHERIC EMISSIONS IN THE MEGA-CITY OF PAKISTAN

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Abstract. Many sites of the world are currently faced with the prospects of air pollution. It is well established that photocopying machines generally contribute to the indoor emissions of Volatile Organic Compounds (VOCs) and Ozone that strongly influence the health of individuals. The current study aimed to discern the spatiotemporal variations of indoor Total Volatile Organic Compounds (TVOCs) and Ozone using a real-time monitoring approach in photocopying centers over mega-city of Pakistan. The real-time monitoring transcribed the diurnal variability of TVOCs and Ozone in photocopying centers (n = 200) during working hours (8:00 AM to 8:00 PM). Spatially, a high degree fluctuation was observed in the indoor level concentrations of TVOCs and Ozone. A typical diurnal cycle with a minimum value in the morning and a maximum in the noon with a gradual decrease until night was observed for TVOCs and Ozone suggesting that the photocopying emissions were a major source of generation. A significantly strong positive correlation was found between TVOCs and Ozone ($r = 0.627$, $p < 0.001$). The findings were compared with the available standards indicating the potential of health hazards to workers.

Keywords: *air pollution, ozone, photocopying machines, indoor environment, total volatile organic compounds*

Introduction

In the current era of mechanization, photocopying machines are among the imperative accessories in the office setup. Globally, photocopying industry is a source of earning for millions of workers. A prominent modification with regards to the technical aspects, basic material, convenience, yield and skillfulness in the photocopying process has also encountered (Elango et al., 2013; Kasi et al., 2018).

In spite of commercial ease and viability, photocopiers are considerable initiators of air pollution. During functional mode, photocopiers release particles of toner, volatile and semi-volatile organic compounds, particulate matter, particles of paper, nano-particles, ammonia, ozone, oxides of nitrogen, metals including arsenic, iron, chromium, titanium, zinc, nickel, cadmium, elemental sulphur and silicon (Durga Ch and Gokhale, 2015; Lee et al., 2006; Bar-Sela and Shoenfeld, 2008; Barthel et al., 2011; Bello et al., 2013; Wensing et al., 2008; Morawska et al., 2009; Adetunji et al., 2009; Pirela et al., 2013; Kleinsorge et al., 2011).

The tracking of Volatile Organic Compounds is required to evaluate the air quality. Spatiotemporal fluctuation of VOCs with respect to source is crucial in alleviating photochemical pollution (Huang et al., 2014). The chemical deterioration of VOCs gives rise to secondary air pollutants including peroxyacetyl nitrate (PAN), secondary organic aerosols and aldehydes. Sunlight as an auxiliary component in the presence of nitrogen oxides and volatile organic compounds promotes the generation of ground level ozone. The profile of VOCs and ozone reveals that the generation of ozone is VOC restricted (Srivastava et al., 2005; Lee et al., 2002; Toro et al., 2006). The association between the atmospheric emissions in terms of Nitrogen oxides and VOCs couples with the generation of ozone (Xu et al., 2017).

Ozone is a predominant component of the atmosphere that is of crucial importance to global climatic variation and tropospheric chemistry (Reddy et al., 2012). The precursors and elevated ozone levels are of great concern owing to the deleterious effects on the human health, forests, plants, natural and anthropogenic materials (Mavroidis and Iliá, 2012; Fann and Risley, 2013; Szyszkowicz et al., 2012; Kheirbek et al., 2013; Kumar et al., 2014a, b; Singh et al., 2014).

The traditional methods for the detection of organic compounds can require weeks to systematize, execute, delineate and interpret results. Direct reading methods (DRMs) have been initiated in workplaces as a substitutive approach with the benefits of swift measurement of the concentration (Brown et al., 2016). Direct reading methods using a portable real-time monitor for the detection of VOCs is a convenient tool (Coy et al., 2000; Peng et al., 2007). For the measurement of Volatile Organic Compounds at workplace, commercially available PID has been used for several decades (Driscoll, 2013).

Various kinds of compact portable VOC monitors are available nowadays. A monitor employing photoionization detector (PID) is able to detect Total Volatile Organic Compounds (TVOCs) with the intervening time of few seconds. The sensitivity of the measurement was outlined to range from ppb to ppm (RAE Systems, 2005). The PID-supported VOC monitor is anticipated to be a beneficial tool for the accurate measurement of TVOC exposure (Mizukoshi et al., 2010). As a segment of an effort in comprehending the role of photocopying process in the generation of TVOC and Ozone, the current study aims to perform real-time monitoring of TVOC and Ozone in photocopying centers and to evaluate the temporal and spatial variation in different areas of Lahore, Pakistan.

Materials and methods

Sampling area

Lahore is a megacity of Pakistan with the inhabitants of about 10+ million people (Razi et al., 2017). It is situated between 30.25° N to 31.5° N and 73° E to 74.5° E and a provincial capital of Punjab covering an area of 1, 172 km² (ul-Haq et al., 2016). Lahore is characterized by the semi-arid climatic conditions with very hot and wet summer and ordinarily cool and dry winter. In the month of June, the recorded highest maximum temperature was 48 °C; whereas, the lowest temperature was recorded to be -2 °C in January. Lahore is documented with an annual rainfall of 489 mm (Ali et al., 2014). The photocopying centers (n = 200) in the vicinity of Lahore were selected randomly. For the convenience, the sampling areas were divided into two different zones. The

sampling areas of the current study are outlined in *Figure 1*. For the collection of baseline data of photocopying centers, a self-structured questionnaire was designed.

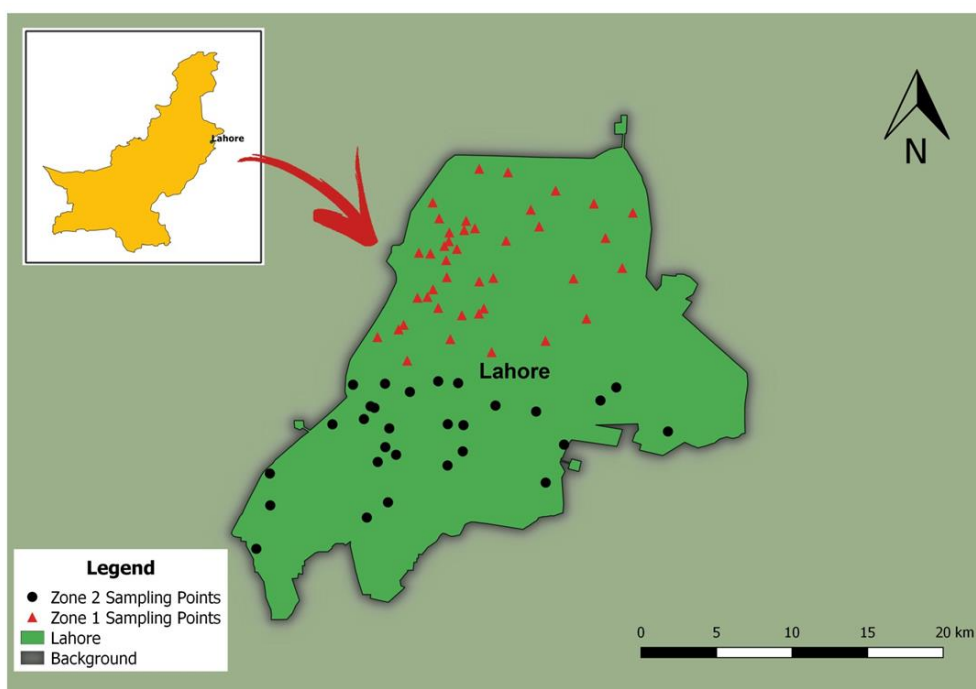


Figure 1. Sampling areas of Lahore, Pakistan

Monitoring instruments and technique

Monitoring was performed for the Total Volatile Organic Compounds (TVOCs) and Ozone (O_3) during the working hours in photocopying centers. A hand-held digital real-time VOC monitor with PID detector [ToxiRAE Pro PID (RAE Systems by Honeywell, 4.6" H X 2.4" W X 1.2" D, 235 g)] was employed as shown in *Figure 2a*. The monitor was characterized by the photoionization sensor with 10.6 eV lamp. The response time of the VOC monitor was <15 s. The monitor was turned on and the self-testing was performed by the instrument. Following the normal start-up process, the main measurement screen was displayed. The instrument specified the readings in parts per million (ppm). The documented values were converted to isobutylene equivalent concentrations in mg/m^3 . Since Isobutylene is conventionally used for the calibration of PID detectors (RAE Systems, 2005).

For the monitoring of Ozone, portable single gas detector [BW Technologies GAXT-G-DL Gas Alert Ozone (1.1" X 2.0" X 3.75", 82 g, by BW Technologies)] was employed with the concentration range of 0–1.00 ppm as shown in *Figure 2b*. The response time of Ozone monitor was 5 s. The self-testing was executed by the detector after being turned on. The normal start-up screen was displayed. The readings of Ozone were converted to $\mu g/m^3$. The TVOC and Ozone monitors can measure the concentrations every second. For the average concentration of TVOC and Ozone in an hour, the average concentration of 10-min interval derived from original 1-min interval reading was utilized. The TVOC and Ozone measuring instruments were placed inside the photocopying centers at the height of approximately 1.1–1.5 m above the ground

(Meciarova et al., 2017). Meanwhile, it was ensured that the instruments are away from the direct pollutant sources and ventilation openings (Moreno-Rangel et al., 2018).



Figure 2. Monitoring instrument for a TVOC and b Ozone

Data analysis

The mean TVOC and Ozone concentrations at a particular sampling point were shown in the maps. In order to analyze the spatial variability of TVOC and Ozone, the local interpolation method i.e., Inverse Distance Weighting (IDW) was employed using the QGIS Desktop 2.18.12.

Statistical analyses were executed by using SPSS [IBM SPSS Statistics Version 20 for Windows, SPSS Inc., Chicago, IL, USA]. The normal distribution of the data was determined by using Shapiro-Wilk Test. For categorical variables, Chi-square test was executed. For the data with no normal distribution, Mann Whitney U test was employed. Spearman's Rank correlation test was performed to quantify the correlation between testing variables. The Friedman Test was applied for assessing the difference in the concentrations of TVOC and Ozone with time of the day. The p-value < 0.05 was considered to be significant.

Results and discussion

The baseline attributes from photocopying centers were documented using a self-structured questionnaire as presented in *Table 1*. No significant difference was observed in terms of average working hours, number of machines and nature of area in photocopying centers located in both zones.

Spatial variability of total volatile organic compounds and ozone

The spatial variations of Total Volatile Organic Compounds (TVOCs) in mg/m^3 and Ozone in $\mu\text{g}/\text{m}^3$ are presented in *Figures 3a, b* and *4a, b*. A high degree fluctuation was observed in the concentrations of both variables.

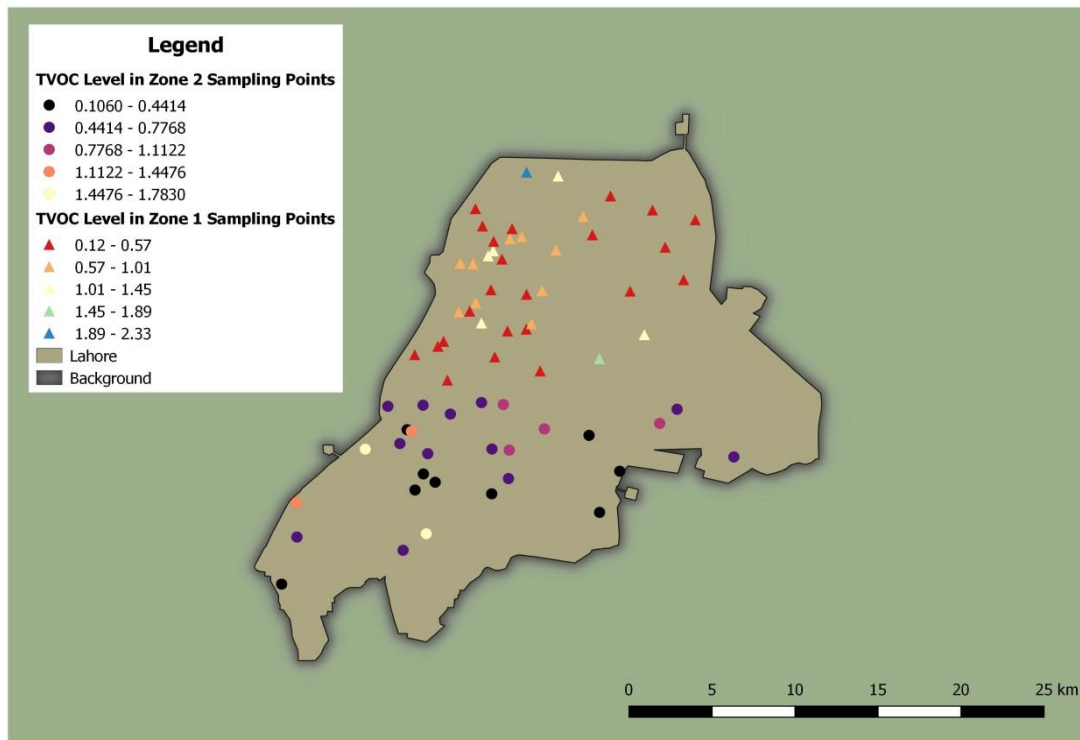
Table 1. Baseline attributes of photocopying centers

	Total (n = 200)	Zone 1 (n = 92)	Zone 2 (n = 108)	p-value
Average working hours	11.97 ± 0.47	12.02 ± 0.55	11.93 ± 0.38	0.194 *
No. of workers	2.30 ± 1.80	2.21 ± 2.25	2.38 ± 1.31	0.027 *
No. of machines in the working centers	2.90 ± 1.52	2.72 ± 1.61	3.05 ± 1.44	0.063 *
No. of black and white machines	2.63 ± 1.24	2.47 ± 1.29	2.76 ± 1.183	0.052 *
No. of colored machines	0.29 ± 0.58	0.25 ± 0.59	0.31 ± 0.57	0.206 *
No. of machines in working condition	2.73 ± 1.57	2.59 ± 1.62	2.84 ± 1.52	0.144 *
No. of machines not in working condition	0.19 ± 0.50	0.13 ± 0.40	0.25 ± 0.57	0.093 *
Presence of computer	Yes = 189 (94.5%) No = 11 (5.5%)	Yes = 82 (89.1%) No = 10 (10.9%)	Yes = 107 (99.1%) No = 1 (0.9%)	0.002 **
Nature of area	Well ventilated = 30 (15%) Ventilated = 55 (27.5%) Congested = 68 (34%) Dark and congested = 34 (17%) Dark and very congested = 13 (6.5%)	Well ventilated = 14 (15.2%) Ventilated = 22 (23.9%) Congested = 30 (32.6%) Dark and congested = 21 (22.8%) Dark and very congested = 5 (5.4%)	Well ventilated = 16 (14.8%) Ventilated = 33 (30.6%) Congested = 38 (35.2%) Dark and congested = 13 (12%) Dark and very congested = 8 (7.4%)	0.331 **
Air conditioner	Yes = 12 (6%) No = 188 (94%)	Yes = 9 (9.8%) No = 83 (90.2%)	Yes = 3 (2.8%) No = 105 (97.2%)	0.038 **
Average No. of copies per day	2860.63 ± 5601.65	2280.71 ± 6650.59	3354.63 ± 4495.12	0.000*

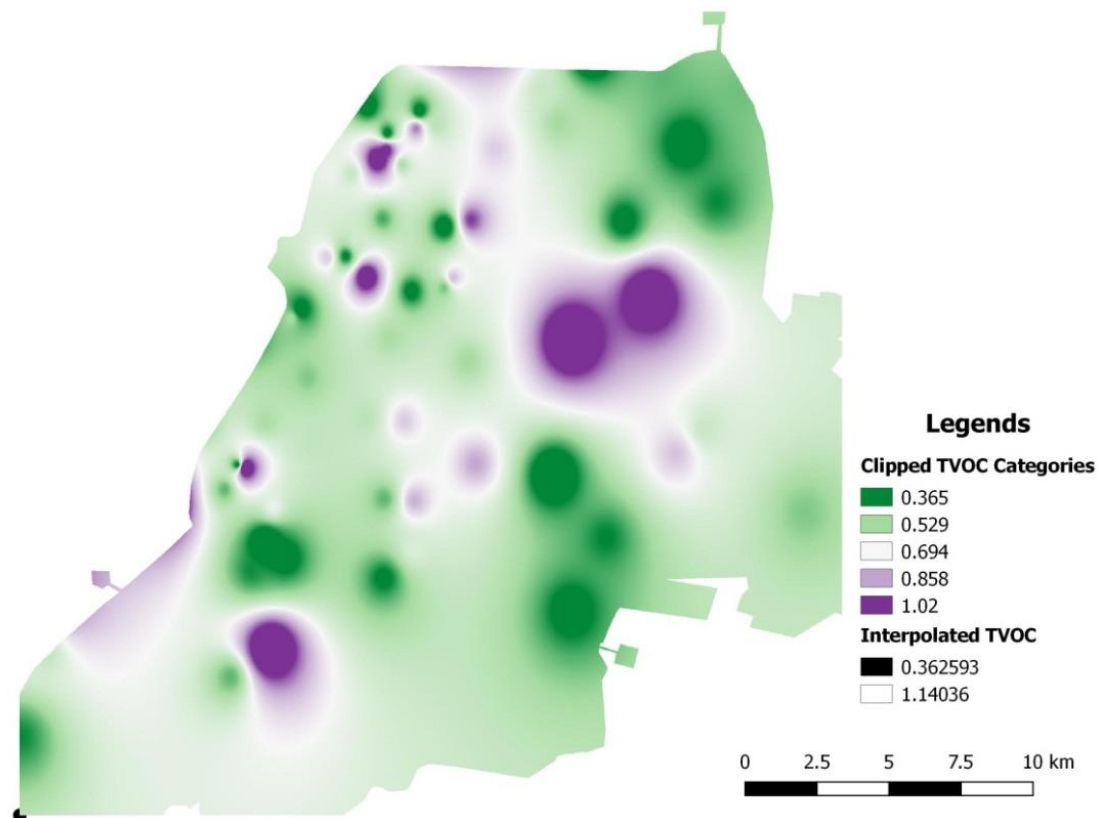
*Mann Whitney U test, **Chi square test

Diurnal variability of total volatile organic compounds

The diurnal variation of Total Volatile Organic Compounds (TVOC) in mg/m³ is presented in *Figure 5*. The average TVOC (mg/m³) during working hours in photocopying center of Zone 1 and Zone 2 was found to be 0.674 ± 0.699 and 0.691 ± 0.786, respectively. The average TVOC concentration in photocopying centers located in Zone 1 was marginally lower than the centers in Zone 2. However, regarding the concentration of TVOCs, no significant difference was observed in the photocopying centers located in two different zones. In Zone 1, the diurnal variation revealed a similar pattern throughout the day with the lowest value observed at 8:00 am in the morning when the photocopying centers were opened. In case of Zone 2, the highest value of TVOC was observed at 12 Noon. Thereafter, it decreases gradually with the lower value during closing time of the photocopying centers. Several studies reported the generation of Volatile Organic Compounds in the photocopying centers (Stefaniak et al., 2000; Lu et al., 2015; Kowalska et al., 2015). A minimum TVOC concentration was observed during morning that gradually started increasing till noon. A progressive decline was found with the passage of time with the lower value at 8:00 pm.

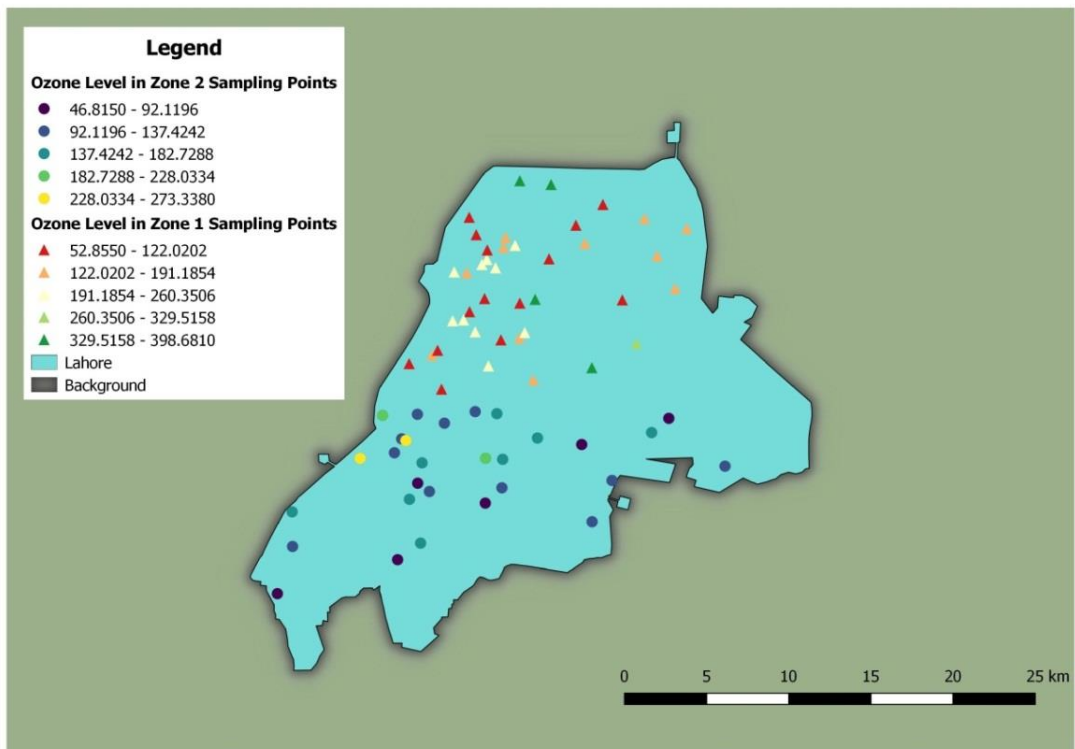


a

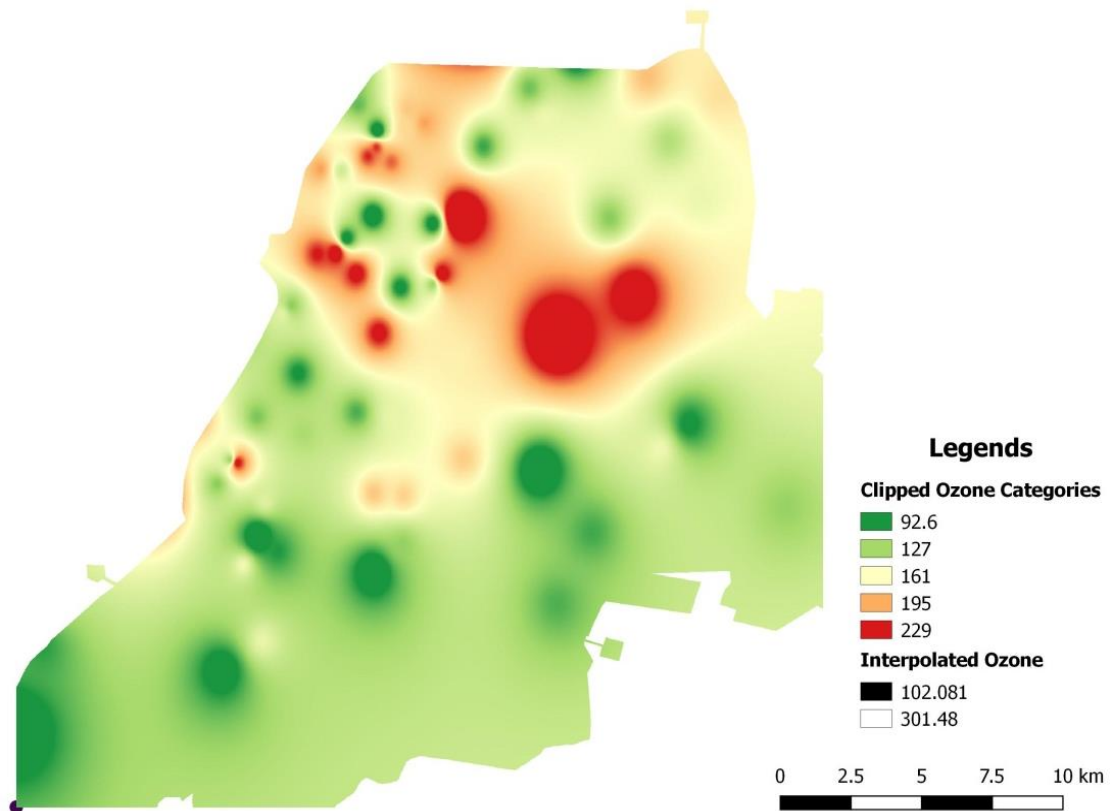


b

Figure 3. a Spatial distribution of mean TVOC concentrations in photocopying centers of Lahore. b Spatial distribution of TVOC concentrations by IDW method



a



b

Figure 4. a Spatial distribution of mean Ozone concentrations in photocopying centers of Lahore. b Spatial distribution of Ozone concentrations by IDW method

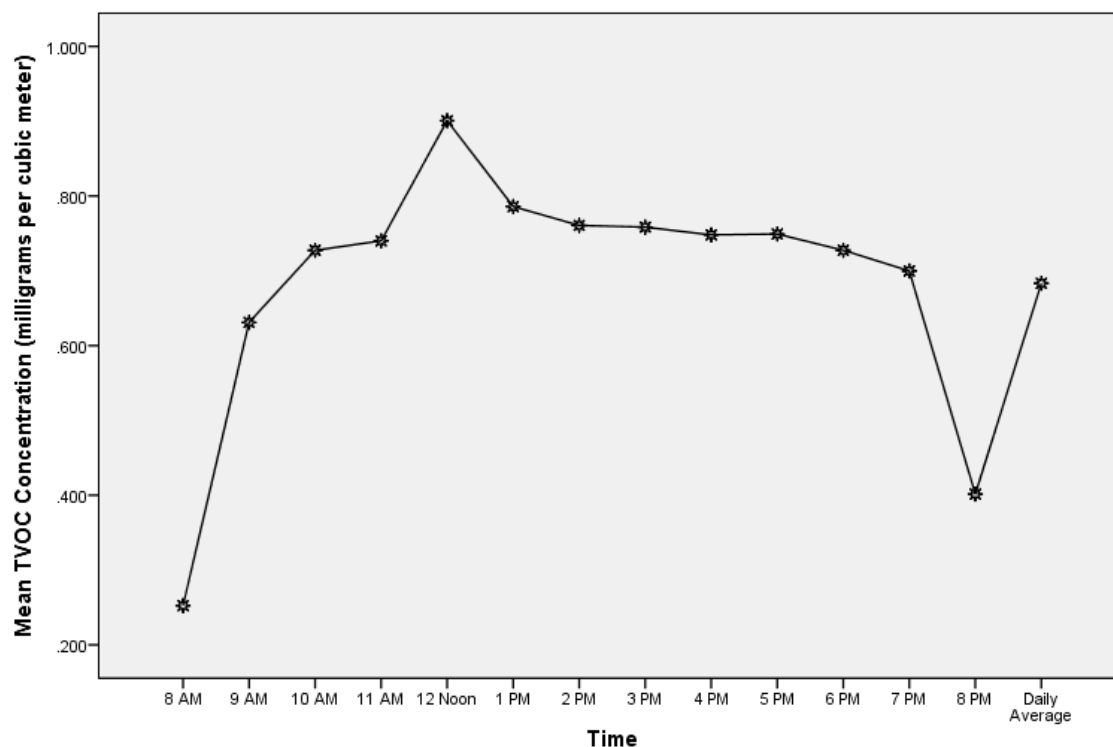


Figure 5. Diurnal pattern of TVOC (mg/m^3) in photocopying centers

Diurnal variability of Ozone

The observed diurnal variability of Ozone at different photocopying centers is shown in *Figure 6*. The average concentration of Ozone ($\mu\text{g}/\text{m}^3$) in Zone 1 and Zone 2 was found to be 187.50 ± 119.28 and 139.08 ± 83.56 respectively with the significant difference ($p < 0.05$) between both the zones. A relatively steep slope was recorded for the concentration of ozone. The diurnal variation of Ozone revealed that the minimum concentration was observed at 8:00 in the morning. It indicates that the maximum concentration was observed at 12 noon. Thereafter, it gradually declines and reaches the minimum value during the night. The similar diurnal variability was documented in other studies (Xue et al., 2017; Adame Carnero et al., 2010; Kumar et al., 2014b).

The Spearman's correlation was determined to evaluate the association between the TVOC and Ozone concentration (*Fig. 7*). A positive and highly significant correlation exists between the concentrations ($r = 0.627$, $p = 0.000$). The ozone precursors including oxides of nitrogen (NO_x) and Volatile Organic Compounds (VOCs) vigorously affect the ozone profile (Filella and Penuelas, 2006).

In order to verify whether the time of the day makes any significant impact on the concentrations of TVOC and Ozone in photocopying centers, the Friedman test was employed. The significant results for both the concentrations indicated that the time of the day did have a significant impact on the concentrations of TVOC ($\chi^2 = 599.311$, Df = 13, Asym. Sig 0.000) and Ozone ($\chi^2 = 1357.799$, Df = 13, Asym. Sig 0.000).

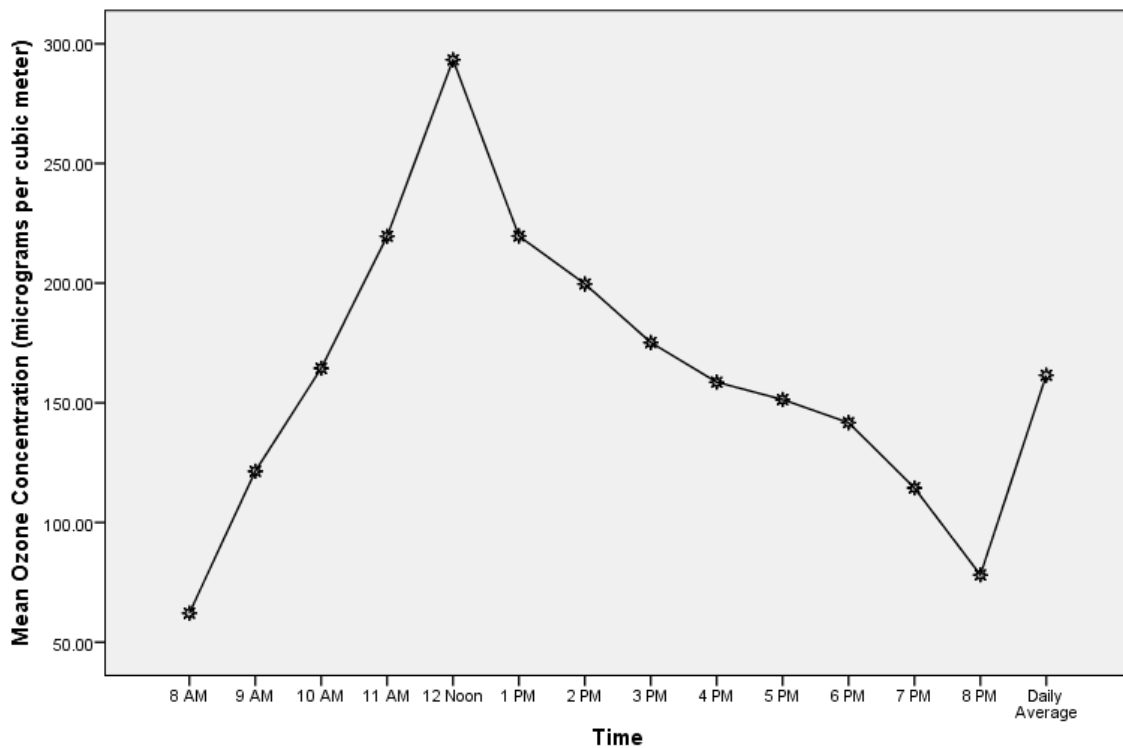


Figure 6. Diurnal pattern of Ozone ($\mu\text{g}/\text{m}^3$) in photocopying centers

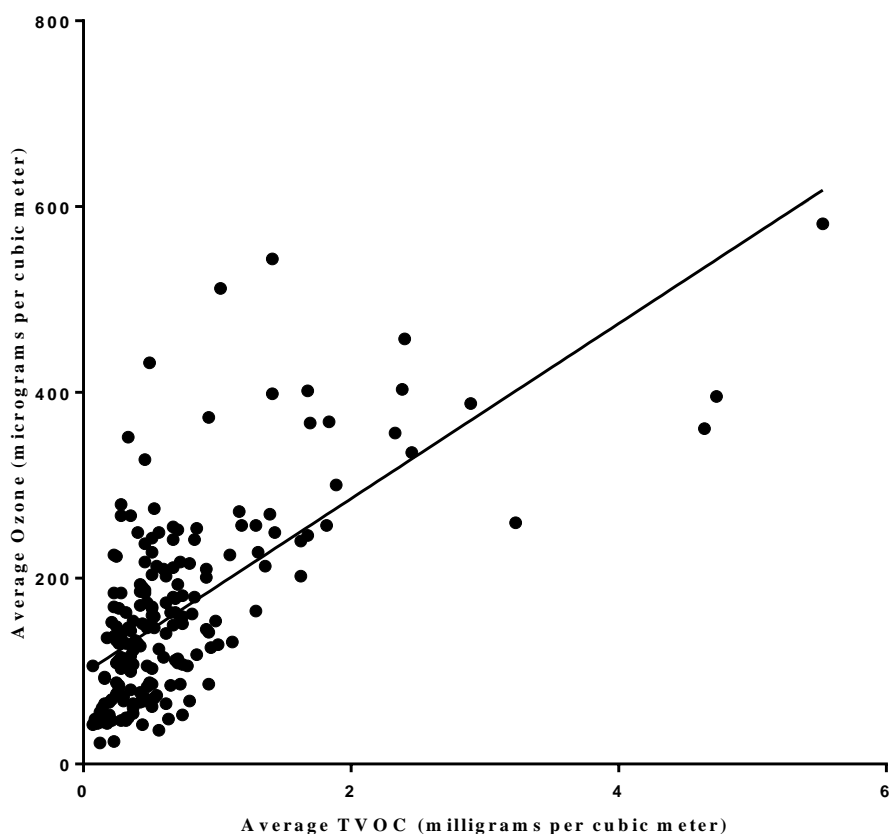


Figure 7. Correlation between average TVOC (mg/m^3) and Ozone ($\mu\text{g}/\text{m}^3$)

The average number of copies made in each photocopying center per day was found to be positively correlated with the concentrations of TVOC and Ozone. A highly significant and positive association was found between the average number of copies per day and the concentration of TVOC ($r = 0.556$, $p = 0.000$). Likewise, the average number of copies per day was computed to be positively and significantly correlated with the Ozone concentrations ($r = 0.333$, $p = 0.000$).

Concentration of TVOC and Ozone with respect to nature of area

In order to compute the association between the concentration of the testing variable and the nature of the area, Kruskal-Wallis test was employed. Based on the observation, the nature of the area was divided into 5 main categories i.e., Well Ventilated, Ventilated, Congested, Dark and Congested, Dark and very Congested. A highly significant association ($p < 0.01$) was observed between the nature of area and the concentrations of TVOC and Ozone (Figs. 8 and 9).

The observations of the current study revealed the variation in TVOC concentration at photocopying centers with respect to the nature of area. The maximum concentrations of TVOCs and ozone were observed in dark and very congested photocopying centers representing the significant role of ventilation in the declined TVOC concentration. Hori et al. (2013) reported the reduced concentration of TVOCs with the elevated ventilation.

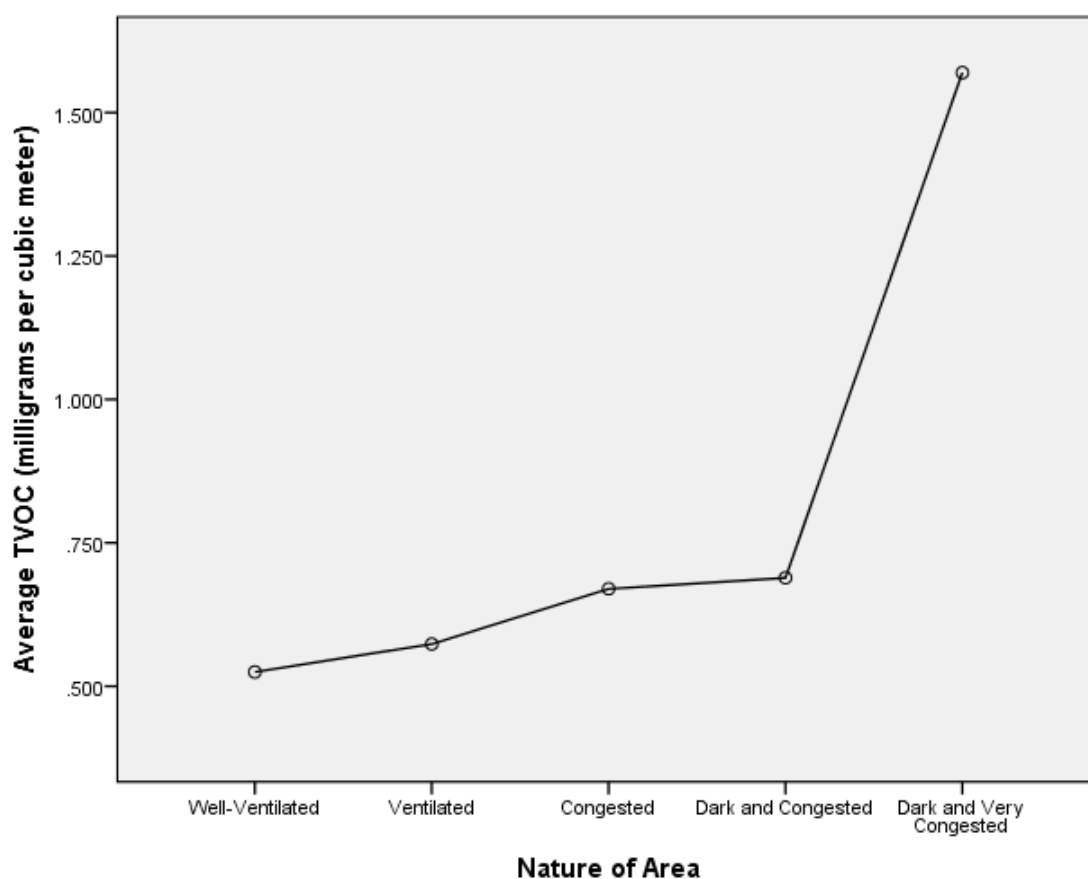


Figure 8. Association between the nature of area and average TVOC (mg/m^3)

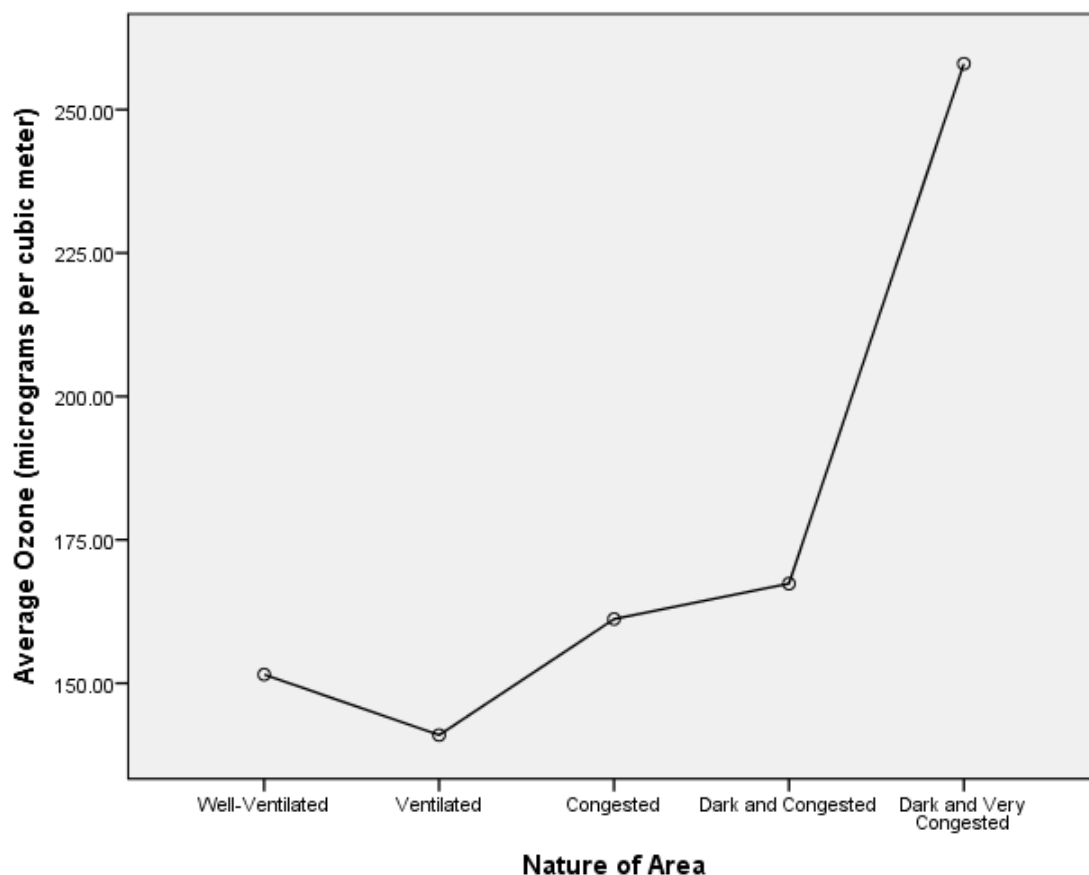


Figure 9. Association between the nature of area and average Ozone ($\mu\text{g}/\text{m}^3$)

The concentration of ozone and its precursors may have a significant effect on the health of human beings (Kumar et al., 2014b). Keeping this in view, the results were compared with the available standards. In non-industrial indoor environment, discomfort and irritability ranges are defined by Molhave (1991). The concentration of VOCs below about $0.2 \text{ mg}/\text{m}^3$ is considered to be the comfort range. Between $0.2\text{--}3.0 \text{ mg}/\text{m}^3$, Irritation and discomfort is possible if other exposures are encountered. The results of the current study with reference to the concentration of TVOCs indicate that it lies within the range of multifactorial exposures i.e. $0.2\text{--}3.0 \text{ mg}/\text{m}^3$. As specified by the National Environmental Quality Standards for Ambient Air by Pak- EPA, the one hour average allowable concentration is $130 \mu\text{g}/\text{m}^3$. However, in the current study, the average concentration exceeded this allowable limit that may result in ill-health of the workers.

Conclusion

In this research, the spatial and temporal variations of the total volatile organic compounds and ozone concentrations measured at photocopying centers in Lahore are presented. The variations in the concentrations of TVOC and Ozone between sampling sites indicated the role of photocopying emissions in generating indoor air pollution. The daily cycle presents variations with respect to the time of the day. The maximum

concentrations of TVOC and Ozone was observed at 12:00 Noon. The daily minimum concentration was monitored at 8:00 am and 8:00 pm with fluctuating values throughout the day. The findings from this research highlight the importance of monitoring strategy to ultimately develop effective control measures. The present research can be used to find the association with the health impacts in the workers. Furthermore, it is recommended to perform comprehensive research using an appropriate risk assessment strategy.

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