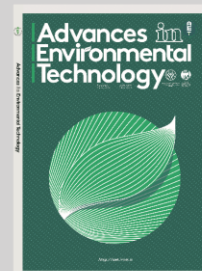




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## Statistical analysis of tropospheric ozone and its precursors using principal component analysis in an urban area of Surat, India

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### ABSTRACT

The objective of this study was to investigate the sources of tropospheric ozone ( $O_3$ ) precursors in an urban area using principal component analysis. Chemically reactive conventional pollutants such as carbon monoxide (CO), carbon dioxide ( $CO_2$ ), nitric oxide (NO), and nitrogen dioxide ( $NO_2$ ), as well as some selected meteorological parameters such as global solar radiation (SR), air temperature (AT), relative humidity (RH), wind speed (WS), and wind direction (WD), were incorporated in this analysis. Real-time observation data were obtained from two monitoring stations, Limbayat and Varachha, situated in Surat city, India. The occurrence of a peak  $O_3$  level in the summer at 5 p.m. proved the well-known fact of interconnection among the temperature, solar radiation, and increment in  $O_3$  concentration. The potencies of CO and NO were remarkable in either the first or second principal component (PC) observed at both locations with more than 45% concentration, which alluded that the main source of  $O_3$  was urban transportation and AT contributed with 50% weightage in the PC ascertained key role of photolysis process in the  $O_3$  formation.

### 1. Introduction

Surface ozone is a colorless, reactive oxidant gas and a secondary pollutant formed from photochemically active primary pollutants. In 2009, the Indian Central Pollution Control Board (CPCB) announced revised air quality standards to control  $O_3$  pollution, which included one-hourly ( $180 \mu\text{g}/\text{m}^3$ ) and eight-hourly ( $100 \mu\text{g}/\text{m}^3$ ) permissible limits for tropospheric  $O_3$ . Also,  $O_3$  was included in the list of criteria pollutants, as prescribed by the CPCB [1].  $O_3$  is becoming a major

concern as its concentration is increasing gradually, causing damage to plants and either directly or indirectly threatening human health even at low concentrations, as reported by Pleijel [2]. Therefore, to develop an emission control strategy, it is necessary to investigate the sources of  $O_3$  formation, i.e., whether  $O_3$  levels are governed by local sources (generated in the region itself) or foreign sources (transported from adjacent regions). The  $O_3$  levels were found to be influenced by intra-urban activities and also by long-range transport [3]. In the troposphere,  $O_3$

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variation is developed by its precursors and meteorological conditions [4,5]. Along with anthropogenic emissions (fossil fuel, solvents, transport), natural sources (lightning, wetlands, forests) also contribute to O<sub>3</sub> precursor gases such as NO<sub>x</sub>, CO, CH<sub>4</sub>, and NMVOC that interact with climatic factors [6]. The role of O<sub>3</sub> precursors can be revealed based on O<sub>3</sub> formation chemistry. Sunlight (accompanied by other O<sub>3</sub> forming compounds) is a directly proportional factor and mostly responsible for O<sub>3</sub> formation, as reviewed by Lu et al. [7]. In India, huge variations are found in sunlight and temperature; therefore, the measurement dataset is distributed according to the seasons to observe its temporal variations. The temporal variability in O<sub>3</sub> is governed by the atmospheric process of the seasons, as characterized by Roberts-Semple et al. [8]. The information gained regarding pollutant sources and their contribution to ambient air pollution levels is known as Source Apportionment (SA) [9]. Two source apportionment models, namely positive matrix factorization and chemical mass balance, have been applied to discover the sources of particulate pollutants [10]. To identify the source contribution of aerosol, chemical characterization, temporal trends, and positive matrix factorization were performed near Houston, TX, USA [11]. Among the several types of correlation coefficients, Pearson's correlation coefficient is commonly used to measure the relationship between independent and dependent variables in linear regression and in the case of the unknown nature of the dataset. Pearson's coefficient was recently used to assess air pollutants with statistical tools in Madrid, Spain [12]. It has been used to evaluate the association between O<sub>3</sub> and its influencing parameters in the form of values between -1 and +1. The interpretations of 0, >0, and <0 are no association, positive association, and negative association, respectively. Here, a p value <0.01 is considered for statistical significance. Principal component analysis (PCA) is a primary tool used to screen the sources that control O<sub>3</sub> concentration. A large number of datasets that correlate with each other are converted to a compressed number of non-correlated and orthogonal variables that constitute a cluster, as evaluated by Martínez and Polanco [13]. PCA is used as a dimensionality

reduction statistical technique in which datasets consisting of numerous interrelated variables are transformed to the most important uncorrelated new variable set, known as the principal component, with a large variance [14]. The highest possible variance occurs in the dataset represented in the first PC. In general, the size of the PC set is the same as the original dataset, and 80% of the total variance of the whole original dataset is considered in general. Standardization is used in the case of different unit measurements and where the data are measured on a large scale. According to the Kaiser criterion, PCs with an eigenvector greater than one are considered. Rotated PCs have a high impact in characterizing the potential group of non-correlated variables. The focus of this study was to investigate the potential sources of tropospheric O<sub>3</sub> with the help of PCA. Carbon monoxide (mg/m<sup>3</sup>), nitrogen dioxide (µg/m<sup>3</sup>), carbon dioxide (ppm), nitric oxide (µg/m<sup>3</sup>), air temperature (°C), relative humidity (%), global solar radiation (W/m<sup>2</sup>), wind direction (°), and wind speed (m/s<sup>1</sup>) were considered as the independent variables.

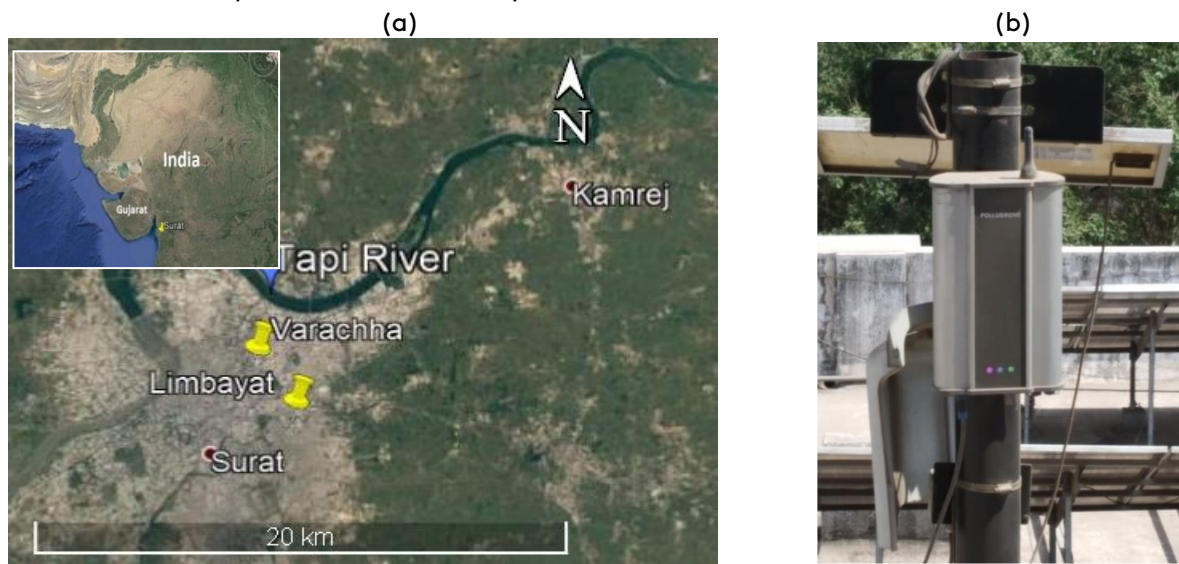
## 2. Materials and methods

### 2.1. Description of the study area and instruments used

The city of Surat is situated in the western region of India on the bank of the Tapi River at a latitude of 21.1702° N and a longitude of 72.8311° E. The city has an altitude of 13.0 m from the mean sea level and covers an area of 474 km<sup>2</sup>. Surat has a population more than 70 lacs, making it the eighth densely populated city (Census of India, 2011) in Gujarat. Real-time continuous air quality monitoring data were collected from the Surat Municipal Corporation at two locations, Limbayat and Varachha, situated at a distance of 3.6 km from each other. A continuous monitoring system was installed in both sites. The readings were recorded on an hourly basis. The monitoring point was kept on the terrace of the Surat Municipal Corporation building at the height of 15 m from the ground level in both locations. The sampling sites represent a typical urban atmosphere, surrounded by colossal roadside traffic, with an industrial area nearby. Polludrone's Continuous Ambient Air Quality Monitoring system (CAAQMS) was used to measure

the gaseous pollutants and the meteorological factors. The CO<sub>2</sub> was measured using the non-dispersive infra-red principle that has a 20-ppm detection limit. The CO, NO, NO<sub>2</sub>, and O<sub>3</sub> were determined based on the electrochemical principle that has detection limits of 100 ppb (11.45 µg/m<sup>3</sup>), 10 ppb (12.5 µg/m<sup>3</sup>), 10 ppb (18.8 µg/m<sup>3</sup>), and 10 ppb (20 µg/m<sup>3</sup>), respectively. An ultrasonic sensor was used to measure wind speed (0–60 m/s) and wind direction (360°). The detection ranges of the respective sensors were 0–40,000 µg/m<sup>3</sup> for O<sub>3</sub>, 0–1000 µg/m<sup>3</sup> for PM<sub>2.5</sub> and PM<sub>10</sub>, 0–1000 µg/m<sup>3</sup> for CO, 0–37,600 µg/m<sup>3</sup> for NO<sub>2</sub>, 0–52,400 µg/m<sup>3</sup> for SO<sub>2</sub>, and 0–5000 ppm for CO<sub>2</sub>. To get higher data accuracy, an innovative e-breathing approach was adopted by the company who installed the CAAQMS system at the site. Sensor calibration for all the parameters was carried out by the manufacturer in the National Accreditation Board Laboratories; the gas calibration was done with pure air and calibration gas at a high concentration range of 1-10 ppm and flow rate of 0.5L/min before installation. As per the information available by the manufacturer of the instrument, multi-level sensor calibration was performed at a factory with

a bump test calibration facility, to test the working efficiency. Spot calibration was performed for a limited data before installation of sensors. Figure 1 shows the two monitoring locations in Surat, India, and the instrument setup at Varachha. Surat has a minimum temperature of around 10°C during the winter nights and a maximum temperature of 46°C during summer in the afternoon. Therefore, considerable variations in O<sub>3</sub> concentrations are found throughout the year. The diamond industries of Surat occupy about 35%–40% of the total area (approximately 37 sq. km) of Varachha. The rest of the region is residential and commercial with heavy traffic areas. The Limbayat area is surrounded by chemical dyeing and power-loom industries, covering approximately 70% of the total area (approximately 19.6 sq. km). The textile industry emits a high pollution load in the form of CO, CO<sub>2</sub>, NO<sub>x</sub>, and particulate matter, as reviewed by Mia et al. and Tiwari et al. [15,16].



**Fig.1.** (a) Location of the ambient air quality monitoring stations (b) Real-time monitoring instrument setup at Varachha.

## 2.2. Data preprocessing

The concentrations of the above-stated parameters were recorded in the two monitoring sites from Feb 1, 2018, to Dec 14, 2019, at Limbayat and from Jan 30, 2018, to Dec 21, 2019, at

Varachha. The outlier data were removed from the O<sub>3</sub> data series after ascertaining that there was no occurrence of any O<sub>3</sub> episodic event on the corresponding day. Such events can be attributed to local photochemistry and transport [17]. In the past, research was focused on potential vorticity

(PV), which is a measure of air turbulence.  $PV > 1.5$  (PV unit;  $1 \text{ PVU} = 10^{-6} \text{ km}^2 \text{ kg}^{-1} \text{ s}^{-1}$ ) in the troposphere is an indicator of stratospheric  $\text{O}_3$  sinking; Lai et al. [18]. The global model of the European Centre for Medium-Range Weather Forecasts, using the Lagrangian method, found that the PV values at different pressure levels (300 hPa–975 hPa) change with altitude; Spreitzer et al. [19]. At the preceding and succeeding day when the highest  $\text{O}_3$  concentration occurred, PV was  $< 1.5$  PV at all pressure levels in the  $20^\circ\text{N}$ – $24^\circ\text{N}$  latitude and  $71^\circ\text{E}$ – $75^\circ\text{E}$  longitude ranges. These data suggest the absence of the vertical transport of  $\text{O}_3$  from the stratosphere during the corresponding days. The datasets of PV were acquired from the website: <http://apps.ecmwf.int/datasets/data/interim-full-daily/>; Sandhya et al. [20]. Moreover, high concentrations of  $\text{O}_3$  were not found in the photochemically inactive hours, which indicated that there was no long-range transportation during the aforesaid days. Hence, the high concentration values of a total number of eight days were removed from the whole dataset after verifying the above criteria. During the day for the respective hour, there was low-to-moderate intensity of global SR and wind speed; hence, the photolysis process in the local region must have been responsible for the observed  $\text{O}_3$  concentration. The missing values and non-detected (below detection limit of the particular sensor) values of the variables were found due to instrument maintenance in the entire dataset. For the missing data, the cubic spline interpolation method was applied, as suggested by Junninen et al. and Moshenberg et al. [21,22], for a 24-h diurnal cycle. The readings recorded as a below detection limit was substituted with a constant value, such as the LOD (limit of detection of the instrument) divided by the square root of 2 (thumb rule), to make the dataset reliable.

### 3. Results and discussion

#### 3.1. Monthly and seasonal $\text{O}_3$ variations

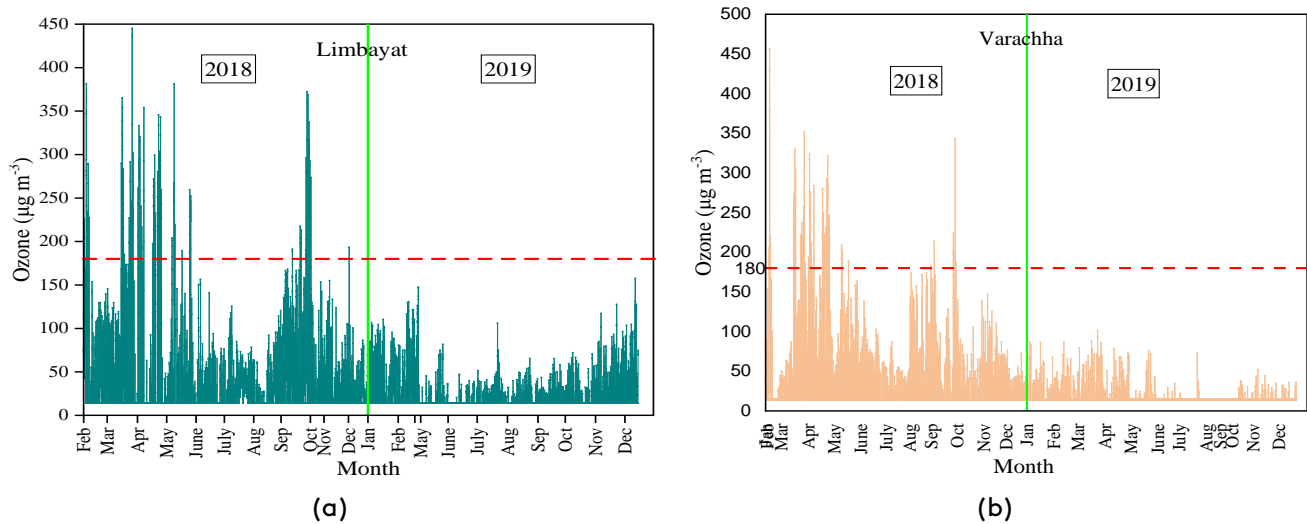
This paper attempts to analyze the  $\text{O}_3$  behavior in an urban area located in the industrial and

commercial zone. Table 1 presents a summary of the data for  $\text{O}_3$ , as evaluated by descriptive statistics. The average hourly data for 2018 and 2019 at Limbayat and Varachha were taken for further processing. The maximum concentrations of  $\text{O}_3$  were found in the summer season at both locations, except at Limbayat in 2019. The highest concentration was found at Limbayat in the summer of 2018 ( $445.22 \pm 65.53$ ), followed by Varachha in 2018 ( $351.4 \pm 64.75$ ) as shown in Table 1. The  $\text{O}_3$  concentration in Mumbai under normal conditions (before the COVID-19 lockdown) fluctuated between 40 and  $90 \mu\text{g}/\text{m}^3$  [23]. Hourly  $\text{O}_3$  variations can be observed by using a time-series plot, as shown in Figure 2. It indicated that hourly averaged  $\text{O}_3$  concentrations exceeded above the threshold ( $180 \mu\text{g}/\text{m}^3$ ) in 2018 at both locations. The data were further distributed season-wise, i.e., winter (January, February), summer (March, April, May), southwest monsoon (June, July, August, September), and post-monsoon (October, November, December), as defined by the Indian Meteorological Department, to examine the season-wise  $\text{O}_3$  behavior.

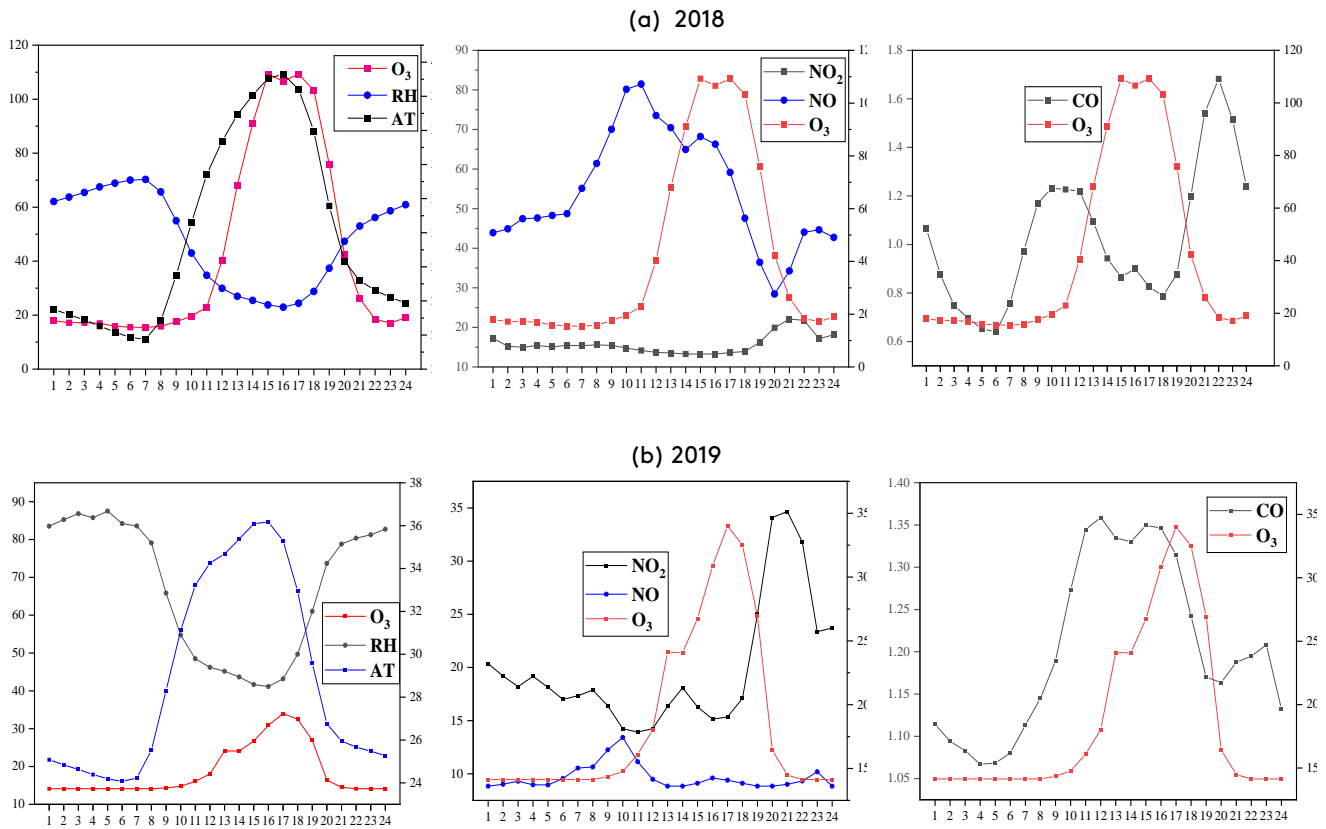
Out of the four seasons, the maximum  $\text{O}_3$  concentrations were observed in the summer (except at Limbayat, 2019). Therefore, to analyze further, summer diurnal variations in  $\text{O}_3$  were compared with other independent parameters at Limbayat, as illustrated in Figure 3. The red colour  $\text{O}_3$  concentration curve (Figure 3) depicts a strong correlation with AT in the summer. At mid-day, both concentrations reached their peak. In the morning, NO reached its maximum at about 10 a.m., indicating the high congestion in road transport. Consequently, the  $\text{O}_3$  concentration increased in the sunlight with NO titration as per ozone photochemistry. During the night, in the absence of SR,  $\text{NO}_2$  was not converted into NO and increased gradually, as shown in Figure 3. All these processes proved that photolysis was the governing process.

**Table 1.** Descriptive Statistics of O<sub>3</sub> (µg/m<sup>3</sup>) at Limbayat and Varachha for 2018 and 2019.

Monitoring station	Year	Seasons	N Sample size	Number of hours for O <sub>3</sub> concentration more than	Max. Mg/m <sup>3</sup>	Mean Mg/m <sup>3</sup>	Standard deviation Mg/m <sup>3</sup>
Limbayat	2018	Winter	576	22	381.18	51.74	58.98
		Summer	2184	118	445.22	33.74	65.53
		Southwest	4224	38	371.9	31.64	43.25
		Post-monsoon	1416	02	273.5	23.43	28.90
	2019	Winter	1152	0	130.59	25.31	26.32
		Summer	816	0	147.3	8.49	19.56
		Southwest	2880	0	105.86	8.86	11.23
		Post-monsoon	1791	0	157.39	15.53	22.83
Varachha	2018	Winter	432	21	219.7	31.04	64.35
		Summer	2208	142	351.4	41.93	64.75
		Southwest	2664	08	343.56	25.53	28.17
		Post-monsoon	1932	0	146.56	13.05	21.10
	2019	Winter	1416	0	93.72	8.16	13.99
		Summer	2208	0	101.61	5.69	13.79
		Southwest	2064	0	73.11	0.75	3.74
		Post-monsoon	1776	0	51.73	2.88	6.50



**Fig. 2.** Time-series plot at Limbayat and Varachha for 2018 and 2019.



**Fig. 3.** Diurnal pattern of O<sub>3</sub> with RH, AT, NO, NO<sub>2</sub> and CO during summer at Limbayat for (a) 2018 and (b) 2019.

**3.2. Correlation between O<sub>3</sub> and its precursors**

SPSS software (version 16.0) was used to analyze the strength of the correlation between the dependent and independent variables. Pearson’s correlation coefficient formula was used to establish the relationship between the variables. The formula involves the computation of the covariance of two variables that are divided by the standard deviation of each measurement.

$$r_{xy} = \frac{\sum_{i=1}^n (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_{i=1}^n (x_i - \bar{x})^2} \sqrt{\sum_{i=1}^n (y_i - \bar{y})^2}} \quad (1)$$

Table 2 shows that the correlation coefficient has a value >0.4 (|r| > 0.4) between O<sub>3</sub> and CO, NO<sub>2</sub>, CO<sub>2</sub>, NO, AT, RH, SR, WD, and WS during the four seasons at Limbayat and Varachha. The positive relationship between O<sub>3</sub> and AT and the strong and moderately inverse relationship with RH in most of the seasons indicate that the source of O<sub>3</sub> is mainly the photochemical processes of the primary pollutants.

**Table 2.** Pearson’s correlation coefficient (r) between O<sub>3</sub> and conventional air pollutants along with the meteorological parameters (|r| > 0.4).

<b>Winter</b>							
Limbayat				Varachha			
	2018	2019		2018	2019		2019
AT	0.468	AT	0.499	CO	0.463	AT	0.475
RH	-	RH	-				
<b>Summer</b>							
Limbayat				Varachha			
	2018	2019		2018	2019		2019
AT	0.654	CO	0.476	AT	0.579	-	-
RH	-	AT	0.583	RH	-		
		RH	-				
<b>Southwest monsoon</b>							
Limbayat				Varachha			
	2018	2019		2018	2019		2019
-	-	-	-	NO	-	-	-
<b>Post-monsoon</b>							
Limbayat				Varachha			
	2018	2019		2018	2019		2019
-	-	-	-	CO <sub>2</sub>	-	-	-
				RH	-		

Note: (-) indicates correlation coefficient values <0.4. The correlation values mentioned in the table are significant at the 0.01 level (2-tailed).

3.3. PCA on multivariate data

In this method, ten correlated elements were transformed into PCs. The total variance of the PCs was the same as the variance of the original variables. The PCs were arranged from the highest to the lowest. The first PC (PC1) represented the major variance in the data, and the tenth PC (PC10) had the lowest variance [24].

In this study, PCA functioned in the following way:

$$PC_i = \sum_{i=1}^n v_{ki}x_i \tag{2}$$

where  $x_i$  is the original element in the dataset and  $v_{ki}$  is the eigenvector. The PCA coefficients were calculated with the help of the correlation matrix, identity matrix, and eigenvalue. The PC scores obtained from the summation of the products of the original variables' value and their loadings were

utilized for further interpretation. PCA was performed using the R open access software along with packages such as FactomineR and FactoInvestigate. The Varimax function was used for rotating the components toward their principal axes to develop the rotated factor loadings. PCA was applied to ten different variables seasonally to analyze the dependence of O<sub>3</sub> concentration on other variables. The first two PCs of all the variables were used owing to high variances, as shown in Table 3. The results obtained from the PCA, correlation study, and a previous research paper (based on the chemical reaction involved in O<sub>3</sub> formation) have been included to identify the primary source of the secondary pollutant O<sub>3</sub>. As seen in Table 4, the release of CO moderately and strongly influenced the increment of O<sub>3</sub> throughout the year in both 2018 and 2019 at Limbayat.

Table 3. First two PCs of all elements for 2018 and 2019.

Elements	Winter							
	Limbayat				Varachha			
	2018		2019		2018		2019	
	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2
CO	-0.046	0.895	-0.033	0.838	0.315	-0.437	0.771	-0.139
NO <sub>2</sub>	0.042	0.112	-0.755	-0.108	-0.066	0	0.207	-0.737
O <sub>3</sub>	0.848	-0.134	-0.013	0.105	0.588	0.312	0.426	-0.179
CO <sub>2</sub>	-0.046	0.03	0.02	-0.192	-0.111	0.118	0.054	-0.053
NO	0.287	0.394	0.203	0.88	0.101	-0.063	0.768	0.35
AT	0.715	0.086	0.736	-0.046	0.9	0.03	0.296	0.528
RH	-0.691	-0.366	-0.277	-0.436	-0.814	0.307	-0.828	-0.047
Solar	0.386	0.147	0.799	0.142	0.74	0.201	0.428	0.746
WD	-0.457	-0.464	0.132	-0.092	-0.272	0.472	-0.248	0.012
WS	0.013	-0.141	0.043	-0.106	0.12	0.863	-0.105	0.106

In most urban areas, aerosol and CO are the leading gases emitted from gasoline fueled vehicles [25]. During 2007–2017, more than 28.87 lakh motor vehicles were registered in Surat City; it is the fastest growing city in the last ten years, followed by Kanpur and Pune [26]. Emission inventory was done in Bengaluru in 2015, and it was observed that vehicular exhaust contributed to 70.7% of the CO emission [27]. Regarding the season-wise formation of the PCs, CO and NO were found in a group either by PC1 or PC2 in most of the seasons. Hence, it could be stated that the main source of O<sub>3</sub> precursor was vehicular exhaust. In the summer season (Table 4) at Limbayat, O<sub>3</sub> was grouped with

CO and CO<sub>2</sub> along with AT and RH in 2019. Furthermore, particulate matter, nitrogen oxide, CO, and hydrocarbons are vehicle criteria pollutants [28]. In the commercial area of Varachha, WD and WS played important roles (Table 4). Hence, O<sub>3</sub> levels can be controlled by equipping the vehicle engines with the latest technologies. On the other hand, the Centre for Science and Environment reported increased levels of O<sub>3</sub> during the COVID-19 lockdown in 22 cities across India even though it flattened the NO<sub>2</sub> curve [29]. The data were unavailable for March and April in 2019 at Limbayat, owing to instrument maintenance.

**Table 4.** Summary of the variables with the highest contribution in the first two PCs.

Limbayat															
Winter				Summer				Southwest monsoon				Post-monsoon			
2018		2019		2018		2019		2018		2019		2018		2019	
PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2
O <sub>3</sub>	NO	WS	CO	O <sub>3</sub>	CO	CO	SR	AT	WD	NO <sub>2</sub>	CO	AT	NO	NO <sub>2</sub>	CO <sub>2</sub>
AT	CO	NO	NO <sub>2</sub>	AT	NO	O <sub>3</sub>		RH	WS	AT	NO	RH	WS	NO	O <sub>3</sub>
RH		O <sub>3</sub>		RH	WD	CO <sub>2</sub>	NO <sub>2</sub>	SR	CO	RH		SR	NO <sub>2</sub>	AT	
WD		AT		SR		AT			NO	SR			O <sub>3</sub>	RH	
WS		RH		WS		RH				CO <sub>2</sub>				SR	
		SR				SR									

Varachha															
Winter				Summer				Southwest monsoon				Post-monsoon			
2018		2019		2018		2019		2018		2019		2018		2019	
PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2
NO <sub>2</sub>	CO	CO	CO <sub>2</sub>	O <sub>3</sub>	CO <sub>2</sub>	CO	CO <sub>2</sub>	AT	CO	NO	O <sub>3</sub>	CO	O <sub>3</sub>	NO	NO <sub>2</sub>
AT	O <sub>3</sub>	NO	WS	AT	CO	NO	WS	RH	CO <sub>2</sub>	AT	NO <sub>2</sub>	NO	CO <sub>2</sub>	AT	WD
RH		AT		RH	NO	AT	WD	SR	WS	RH		AT		RH	WS
SR		RH		SR		RH				SR		SR	RH	SR	
		SR		WS		SR									

#### 4. Conclusions

In this research work, the O<sub>3</sub> concentration variations with seasons were analyzed at two locations, Varachha and Limbayat of Surat city. With PC variable loadings, the relationship of O<sub>3</sub> with other pollutants, and several meteorological factors were investigated. In a seasonal diurnal pattern, a higher concentration of surface O<sub>3</sub> was observed during the daytime, especially afternoon or early evening, when compared with the nighttime due to the higher temperature. In the entire year, the maximum O<sub>3</sub> concentration was noted in the summer, owing to the influence of meteorological parameters such as AT and SR. This result was also proven by the moderate correlation coefficient between tropospheric O<sub>3</sub> and temperature in the summer season at both locations. In the PC variable loadings, the group of NO and CO in the first or second PC revealed the effect of the local source, i.e., road transport is the main source. The meteorological factors with O<sub>3</sub> in the same cluster disclosed the presence of photochemical reactions of ozone. Conclusively, in the numerical or chemical source apportionment of ozone, PCA can be used as an effective pre-processor tool.

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