Pollution, 2(4): 387-397, Autumn 2016
DOI: 10.7508/pj.2016.04.002
Print ISSN 2383-451X Online ISSN: 2383-4501
Web Page: https://jpoll.ut.ac.ir, Email: jpoll@ut.ac.ir

Status and preparation of prediction models for ozone as an air pollutant in Shiraz, Iran

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Received: 28 Feb. 2016 Accepted: 1 Jun. 2016

ABSTRACT: In the present study, air quality analyses for ozone (O_3) were conducted in Shiraz, a city in the south of Iran. The measurements were taken from 2011 through 2012 in two different locations to prepare average data in the city. The average concentrations were calculated for every 24 hours, each month and each season. Results showed that the highest concentration of ozone occurs generally in the afternoon while the least concentration was found in the morning and at midnight. Monthly concentrations of ozone showed the highest value in August and June while the least value was in December. The seasonal concentrations showed the least amounts in autumn while the highest amounts were in spring. Relations between the air pollutant and some meteorological parameters were calculated statistically using the daily average data. The wind data (velocity, direction), relative humidity, temperature, sunshine periods, evaporation, dew point, and rainfall were considered as independent variables. The relationships between concentration of pollutant and meteorological parameters were expressed by multiple linear regression equations for both annual and seasonal conditions using SPSS software. Root mean square error (RMSE) test showed that among different prediction models, stepwise model is the best option.

Keywords: air pollution, meteorological parameters, ozone, regression model.

INTRODUCTION

At ground-level, ozone is a pollutant, but in the stratosphere, it screens UV radiation. Ozone (O_3) is one of the seven conventional (criteria) pollutants (including SO_2, CO, particulates, hydrocarbons, nitrogen oxides, O_3, and lead). These pollutants produce the highest volume of pollutants in the air and the most serious threat for human health and welfare. Concentration on these pollutants, especially in cities, has been regulated by Clean Air Act since 1970 (Cunningham and Cunningham, 2002).

Tropospheric ozone is a secondary pollutant and greenhouse gas (Lacis et al., 2014) produced by photochemical oxidation of its precursors such as carbon monoxide (CO), methane (CH_4), and non-methane volatile organic compounds (NMVOCs), and nitrogen oxides (NOx) in presence of hydroxyl radical (OH) (Rasmussen et al., 2012).

Ozone (O_3) is formed in the atmosphere when oxygen atoms (O) react with oxygen molecules (O_2). In the upper atmosphere, solar UV radiation of wavelengths shorter than 242 nm can photolyze O_2 molecules and

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thereby produce O atoms (Fabian and Martin, 2014). The breakdown of ozone in the stratosphere results in reduced absorption of ultraviolet radiation. Consequently, unabsorbed and dangerous ultraviolet radiation is able to reach the Earth's surface at a higher intensity. Ozone levels have dropped by a worldwide average of about 4% since the late 1970s (Bernhard et al., 2010).

In recent years, there have been increasing concerns on air temperature and ozone concentration due to climate change. Studies undertaken by Jacob et al. (1993), Ryan et al. (2007), and Camalier et al. (2007) ascertain that temperature constitutes a meteorological factor influencing surface ozone formation amongst other conditions. According to International Panel for Climate Change (IPCC, 2007), surface ozone is expected to rise with an increase in temperature.

Status of pollutants concentration and effects of meteorological and atmospheric parameters on these pollutants compose the basis for the following studies. Ho and Lin (1994) studied semi-statistical model for evaluating the NO_x concentration by considering source emissions and meteorological effects. Street level of NO_x and SPM in Hong Kong has been studied by Lam et al. (1997). In a study, the relationship between monitored air pollutants and meteorological factors, such as wind speed, relative humidity ratio, and temperature, was statistically analyzed using SPSS. According to the results obtained through multiple linear regression analysis, for some months there was a moderate and weak relationship between the air pollutants like O_3 level and the meteorological factors in Trabzon city (Cuhadaroglu and Demirci, 1997).

Mandal (2000) has shown the progressive decrease of air pollution from west to east in Kolkata. Statistical modeling of ambient air pollutants in Delhi has been studied by Chelani et al. (2001). Abdul-Wahab and Al-Alawi (2002) developed a neural network model to predict the tropospheric (surface or ground) ozone concentrations as a function of meteorological conditions and various air quality parameters. The results of this study showed that the artificial neural network (ANN) is a promising method for air pollution modeling. The observed behavior of pollution concentrations to the prevailing meteorological conditions has been studied for the period from June 13 to September 2, 1994, for the Metropolitan Area of Sao Paulo (Sánchez-Ccoyllo and Andrade, 2002). Results showed low concentrations associated with intense ventilation, precipitation, and high relative humidity. While high values of concentrations prevailed due to weak ventilation, absence of precipitation, and low relative humidity for some pollutants. Also for predicting CO, Sabah et al. (2003) used a statistical model.

Elminir (2005) mentioned dependence of air pollutants on meteorology over Cairo in Egypt. The results hint that wind direction was found to have an influence not only on pollutant concentrations but also on the correlation between pollutants. It was also found that the highest average concentration for NO_2 and O_3 occurred at humidity $\leq$40% indicative for strong vertical mixing.

In another research, data on the concentrations of seven air pollutants (CH_4, NMHC, CO, CO_2, NO, NO_2, and SO_2) and meteorological variables (wind speed and direction, air temperature, relative humidity, and solar radiation) were used to predict the concentration of ozone in the atmosphere using both multiple linear and principal component regression methods (Abdul-Wahab et al., 2005). Results showed that while high temperature and high solar energy tended to increase the daytime ozone concentrations, the pollutants NO and SO_2 being emitted to the atmosphere were being depleted. However, the model did not predict the nighttime ozone concentrations as precisely as it did for the daytime. Asrari et al. (2007) studied effect of meteorological factors for predicting CO. Also, variations in concentration of CO in different times have been shown in this study.

Kalabokas et al. (2012) presented seasonal variation characteristics of ozone during the four- year period 2001-2004 in the two major urban areas of Greece, Athens, and Thessaloniki. On the other hand though, they exhibit also characteristics encountered in ozone, like the broad mid-day peak at the peripheral stations as well as the existence of relatively high rural background levels around both urban areas.

Li et al. (2014) presented the spatial and temporal variation of Air Pollution Index (API) and examined the relationships between API and meteorological factors during 2001–2011 in Guangzhou, China. Relationships were found between API and a variety of meteorological factors. Temperature, relative humidity, precipitation, and wind speed were negatively correlated with API while diurnal temperature range and atmospheric pressure were positively correlated with API in the annual condition. Yoo et al. (2014) mentioned that all of the pollutants show significant negative correlations between their concentrations and rain intensity due to washout or convection. The relative effect of the precipitation on the air pollutant concentrations was estimated to be: PM_{10} > SO_2 > NO_2 > CO > O_3, indicating that PM_{10} was most effectively cleaned by rainfall. The analysis showed that the O_3 concentrations may increase due to vertical mixing leading to its downward transport from the lower stratosphere/ upper troposphere.

Wang et al. (2015) studied on air quality in Chongqing, the largest mountainous city in China. From 2002 to 2012, statistical analysis of SO_2, PM_{10}, and NO_2 concentrations was conducted. The analysis of Pearson correlation indicated that concentrations of SO_2, PM_{10}, and NO_2 were positively correlated with atmospheric pressure but negatively with temperature and wind speed. The analysis of Multi-Pollutant Index (MPI) showed that air quality in Chongqing was serious.

The climatology of tropospheric ozone at Irene was investigated using SHADOZ network data to assess the correlation between the observed seasonal ozone enhancement and meteorological factors (Mulumba et al., 2015). A multiple linear regression model was used to provide seasonal correlation between ozone and temperature and relative humidity. All seasons display strong regression coefficients between ozone and temperature. Similar trends are also observed for relative humidity and ozone concentrations in autumn, spring, and summer.

Statistical modeling of PM_{10} in Tehran was studied by Masoudi et al. (2016). According to the results obtained through multiple linear regression analysis, for seasonal and annual conditions there were significant relationships between PM_{10} level and the meteorological factors in Tehran.

The present study exhibits diurnal, monthly, and seasonal variations of concentration of ozone and also a statistical model that is able to predict amount of ozone. This is based on linear regression technique. Linear Regression estimates the coefficients of the linear equation, involving one or more independent variables that best predict the value of the dependent variable (ozone amount in this study). So, a large statistical and graphical software package (SPSS, Software Package of Social Sciences, V. 20) as one of the best known statistical packages has been used.

MATERIALS AND METHODS

Study area

The research area, Shiraz, is the biggest city in the southern part of Iran located around 29° 30' N and 52º 30' E and the elevation is about 1500 m above the mean sea level. Annual precipitation of Shiraz is about 330 mm. It has semi-arid climate and residential population was 1,500,000 in 2010. Recently, Ahvaz and some Iranian cities were considered as the worst polluted cities of the world according to a survey by the World Health Organization in 2011 (Guinness World Records, 2013). Shiraz is also one of the biggest cities of Iran and we need to carry out an ambient air quality analysis in this city.

Data and Methodology

Two available sampling stations in the city called Setad and Darvazah-Kazarun belonged to Environmental Organization of Iran were selected to represent different traffic loads and activities.

The sampling was performed every 30 minutes daily for each pollutant during all months of 2011 and 2012. Among the measured data in the two stations, ozone was chosen. Then the averages were calculated for every hour, monthly, and seasonally for both stations by Excel. Finally, averages of data at two stations were used to show air pollution situation as diurnal, monthly, and seasonal graphs of concentration of ozone in the city.

Studying correlation of ozone and metrological parameters of synoptic station of city was the next step. The metrological parameters studied include: temperature (min, max and mean), ratio of humidity (min and max), precipitation, sunshine hours, wind direction (max), wind speed (max and mean), and evaporation.

In the next step, daily average data at two stations in 2012 was considered as dependent variable in statistical analysis, while daily data of meteorological parameters during this year were selected as independent variables in SPSS software and the linear regression equation showed that the concentration of ozone depends on the kind of meteorological parameters and also gives an idea about the levels of this relation. The relationship between the dependent variables and each independent variable should be linear. The significant values in output are based on fitting a single model. Also, linear regression equation made for different seasons may show those relationships which are not observed using annual data.

The model for predicting ozone was determined using two multiple regression modeling procedures of ‘enter method’ and ‘stepwise method’. In ‘enter method’, all independent variables selected are added to a single regression model. In ‘stepwise’, which is better, all variables can be entered or removed from the model depending on the significance. Therefore, only those variables which have more influence on dependent variable are observed in a regression model.

RESULTS AND DISCUSSION

In Figures 1, 2 and 3, the diurnal, monthly, and seasonal variations in concentration of ozone have been presented. As shown in Figure 1, the high concentration of ozone occurs in the afternoon. Actually, ozone in the lower atmosphere is formed by reaction of NOx and VOC’s in the presence of sunlight. Monthly concentration of ozone showed the highest values in August and the least amounts in December (Fig. 2). Seasonal concentration of ozone showed the highest values in spring and the least amounts in autumn (Fig. 3). All graphs showed that the concentrations of ozone are lower than Primary Standards of ozone (0.12 and 0.05 ppm) recommended by National Ambient Air Quality Standards (NAAQS) of USA and Iran, respectively.

However, these graphs are almost about annual and monthly condition, not about hourly conditions while these amounts (0.12 and 0.05 ppm) are the Primary Standards for hourly condition; but provide useful information about the daily conditions. Therefore, it is assumed that some of these amounts in the Figures are more than the standards which show unhealthy condition. These results are almost in good agreement with results obtained in other cities like Tehran (Masoudi et al., 2014b) and Esfahan (Gerami, 2014) and Ahvaz (Masoudi et al., 2014a).

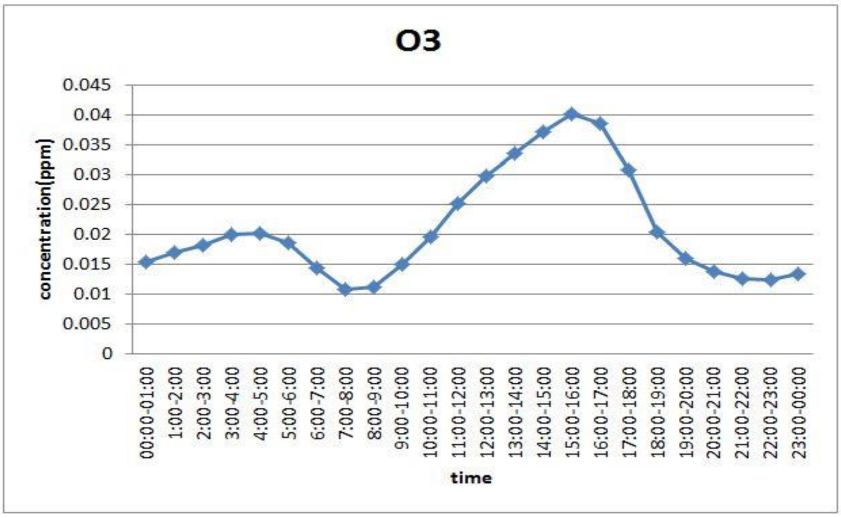

Fig. 1. Diurnal variation of ozone concentration in Shiraz (2011-2012)

Fig. 2. Monthly variation of ozone concentration in Shiraz (2011-2012)

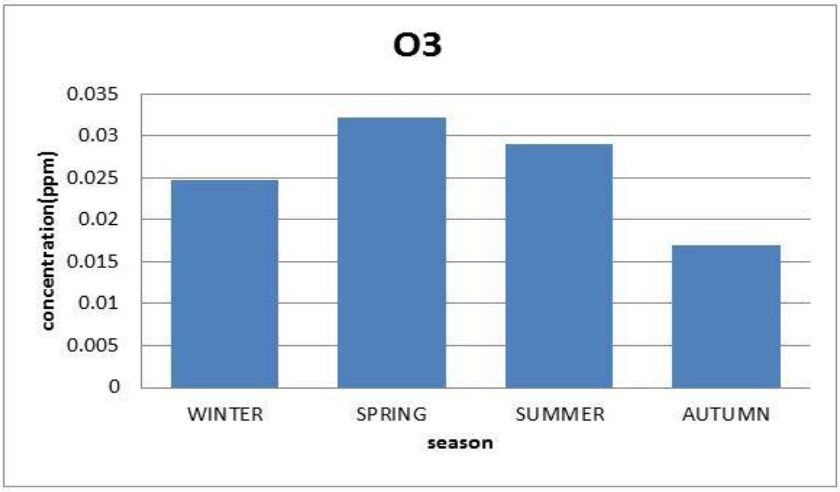

Fig. 3. Seasonal variation of ozone concentration in Shiraz (2011-2012)

Table 1 shows the relationships between ozone and other air pollutants. For example, the concentration of ozone shows negative correlation with CO and SO_2 while it shows positive correlation with NO_2, and PM_{10}. Ozone is increased with reaction between nitrogen oxides and sunlight. Therefore, it was that assumed ozone shows positive relation with nitrogen oxides. Ozone is increased when sunlight is increased while other pollutants are related to traffic volume that is observed more in the evening. Therefore, this negative relation is observed between ozone and other pollutant instead of PM_{10} which main source of it is detached soils from western neighbors like Iraq. These results are almost in good agreement with other results regarding relationships between ozone and other air pollutants in other regions (Abdul-Wahab and Al-Alawi, 2002; Masoudi et al., 2014b). Correlation coefficients significant in 0.05 level are identified with a single asterisk (significant), and those significant in 0.01 level are identified with two asterisks (highly significant).

Table 2 of analysis of variance shows that both regressions of ‘enter’ and ‘stepwise’ methods in annual condition are highly significant, indicating a significant relation between the different variables.

In Table 3, the coefficients of ozone pollution model and regression lines for both enter and stepwise methods in annual condition are presented. Regression coefficients, standard errors, standardized coefficient beta, t values, and two-tailed significance level of t have been shown in the Tables.

Table 1. Correlation between air pollutants and ozone

	CO	PM_{10}	NO_2	NOx	SO_2
Pearson Correlation	-0.715**	0.162*	0.130	-0.096	-0.142*
Sig. (2-tailed)	0.000	0.016	0.053	0.156	0.035
N	221	221	221	221	221

Table 2. Tables of analysis of variance for both regressions of ‘enter’ (a) and ‘stepwise’ (b) methods for annual condition

Analysis of variance (a)

Model	Sum of Squares	df	Mean Square	F	Sig.
Regression	23042.380	11	2094.762	26.551**	0.000
Residual	22801.130	289	78.897		
Total	45843.510	300			

Predictors: (Constant), Rain, Wind direction (max), Wind speed (max), Wind speed (mean), Temperature (max), Temperature (min), Temperature (mean), Sunshine Hours, Ratio of Humidity (min), Ratio of Humidity (max), Ratio of Humidity (mean), Evaporation.

Dependent Variable: Ozone

Analysis of variance (b)

Model	Sum of Squares	Df	Mean Square	F	Sig.
Regression	22969.771	7	3281.396	42.033**	0.000
Residual	22873.739	293	78.067		
Total	45843.510	300			

Predictors: (Constant), Sunshine Hours, Ratio of Humidity (min), Ratio of Humidity (max), Wind direction (max)

Dependent Variable: Ozone

Table 3. Coefficients of Ozone pollution model and regression lines for both enter (a) and stepwise (b) methods for annual condition

Coefficients (a)

Model	**Unstandardized Coefficients**		**Standardized Coefficients**	**t**	**Sig.**
	B	**Std. Error**	**Beta**		
(Constant)	54.527	7.069		7.714	0.000
Temperature (min)	0.835	0.277	0.479	3.017**	0.003
Temperature (max)	-1.474	0.276	-1.060	-5.342**	0.000
Temperature (mean)	-0.101	0.285	-0.069	-0.353	0.724
Ratio of Humidity (min)	-0.206	0.083	-0.267	-2.477*	0.014
Ratio of Humidity (max)	-0.243	0.056	-0.398	-4.361**	0.000
Rain	0.045	0.133	0.019	0.338	0.736
Sunshine Hours	0.721	0.275	0.174	2.618**	0.009
Evaporation	1.410	0.363	0.459	3.883**	0.000
Wind speed (max)	0.263	0.352	0.049	0.749	0.454
Wind direction (max)	0.002	0.006	0.016	0.348	0.728
Wind speed (mean)	2.017	0.719	0.187	2.804**	0.005

Dependent Variable: Ozone

Coefficients (b)

Model	**Unstandardized Coefficients**		**Standardized Coefficients**	**t**	**Sig.**
	B	**Std. Error**	**Beta**		
(Constant)	56.411	6.570		8.585	0.000
Ratio of Humidity (max)	-0.247	0.055	-0.404	-4.516**	0.000
Wind speed (mean)	2.424	0.526	0.225	4.609**	0.000
Temperature (max)	-1.533	0.244	-1.103	-6.277**	0.000
Temperature (min)	0.788	0.241	0.452	3.270**	0.001
Evaporation	1.414	0.339	0.461	4.175**	0.000
Sunshine Hours	0.681	0.269	0.164	2.537*	0.012
Ratio of Humidity (min)	-0.203	0.073	-0.263	-2.780**	0.006

Dependent Variable: Ozone

The linear regression equations show that the ozone pollution depends on the meteorological parameters and also give an idea about the levels of relations. The linear model equations after using ‘enter method’ and ‘stepwise method’ for annual condition are:

Ozone amount (ppb) using ‘enter method’ for annual condition = 54.527 + (0.835) Tmin + (-1.474) Tmax + (-0.101) Tmean + (-.206) RHmin + (-0.243) RHmax + (0.045) R + (0.721) SH + (0.002) WDmax + (0.263) WSmax + (2.017) WSmean + (1.410) E

R= 0.709 (significant in 0.01)

Note: Tmean=Temperature (mean), Tmax =Temperature (max), Tmin=Temperature (min), WSmean = Wind speed (mean), WSmax =Wind speed (max), WDmax =Wind direction (max), RHmax = Ratio of Humidity (max), RHmin= Ratio of Humidity (min), SH= Sunshine Hours, R=Rainfall, E=Evaporation

Ozone amount (ppb) using ‘stepwise method’ for annual condition = 56.411 + (2.424) WSmean + (-0.247) RHmax + (-0.203) RHmin + (-1.533) Tmax + (0.788) Tmin + (1.414) E + (0.681) SH
R= 0.708 (significant in 0.01)

Results of linear regression model show that ratio of humidity and temperatures (max) have reverse effect on concentration of ozone. So that, when these parameters increase, the concentration of ozone decreases. While sunshine hours, wind speed (mean), temperature (min), and evaporation increase, the concentration of ozone significantly increases (Table 3b). Other meteorological parameters show different effects on ozone amounts although these results are not significant. For example, wind direction has reverse effect on concentration of ozone (Table 3a). These results are almost in good agreement with other results regarding ozone measurements in Tehran (Masoudi et al., 2014b) and other regions (Abdul-Wahab et al., 2005; Li et al., 2014). Actually, some of these events happen in real condition. Increase in rainfall, wind speed, and temperature (inversion happens in low temperatures) usually decreases most of air pollutants (Asrari et al., 2007). But with increasing sunshine hours, we expect ozone amounts to increase. The following reactions occur in the lower atmosphere for producing O_3 (Sharma, 2001):

$N_2 + O_2 \Rightarrow 2NO$
$2NO + O_2 \Rightarrow 2NO_2$
$NO + VOC \Rightarrow NO_2$
$NO_2 + \text{sunlight} \Rightarrow NO + O$
$O + O_2 + \text{sunlight} \Rightarrow O_3$
$O_3 + NO \Rightarrow NO_2 + O_2$

The values and significance of R (multiple correlation coefficients) in both equations show capability of them in predicting ozone amount. The amount of Adjusted R^2 in both equations is almost 0.48 showing that different parameters can calculate almost 48% variability of ozone. This result indicates for predicting most of air pollutants like ozone, we should take into consideration consumption of fossil fuel, especially in motor vehicles. Half of emission of (VOC) Hydrocarbons and NOx in cities is produced by motor vehicles. The automobile exhaust produces 75% of total air pollution, releasing poisonous gases of CO (77%), NOx (8%), and Hydrocarbons (14%) (Sharma, 2001). On the other hand, R in enter method (0.709) is equal to stepwise method (0.708), showing no difference. Therefore, second equation based on stepwise method can be used to predict ozone in the city instead of using first equation which needs more data. On the other hand, no difference between the two R values indicates that the excluded variables in second equation have less effect on measuring of ozone in the city.

Beta in Table 3 shows those independent variables (meteorological parameters) which have more effect on dependent variable (ozone). The beta in the both Table 3a and b shows a highly significant effect of some variables like Temperature to other meteorological parameters for measuring the ozone which is close to the results of Masoudi et al. (2014b). Parameter Sig. (P-value) from Table 3 shows amount of relation between ozone and meteorological parameters. For example, Table 3a shows wind speed (mean) has higher effect on ozone than wind direction.

On the other hand, the linear regression equations of ozone amount were presented for both enter and stepwise methods in different seasonal condition. Almost all of the models are significant. Stepwise methods show those meteorological parameters which are most important during these seasons for estimating the pollution. Again, those parameters showing increasing in sun radiations like temperature and sunshine hours are observed as the most important among the others. Among the models, winter models have the highest R while the R of spring models shows the least. R in autumn and winter models are higher than in annual models, also indicating that relations between the pollutant and meteorological parameters are stronger than whole year during these seasons. These results are almost in good agreement with other results regarding ozone assessment in other cities like Tehran (Masoudi et al., 2014b) and Esfahan (Gerami, 2014) and Ahvaz (Masoudi et al., 2014a). The linear model equations after using 'enter method' and 'stepwise method' for seasonal condition are:

Ozone amount (ppb) using 'enter method' for winter season = 55.250 + (-1.436) Tmax + (1.314) Tmin + (0.718) WSmean + (0.304) WSmax + (0.002) WDmax + (-0.161) RHmax + (-0.298) RHmin + (0.065) R + (-0.011) E + (0.697) SH R= 0.827 (significant in 0.01)

Ozone amount (ppb) using 'stepwise method' for winter season = 56.335 + (-0.160) RHmax + (1.187) WSmean + (-2.737) Tmax + (-0.291) RHmin + (2.639) Tmean + (0.668) SH R= 0.824 (significant in 0.05)

Ozone amount (ppb) using 'enter method' for spring season = 61.386 + (1.322) Tmin+ (-1.256) Tmax + (-0.638) Tmean + (-0.379) RHmin + (-0.080) RHmax + (-0.989) R + (1.002) SH + (0.576) E + (-0.003) WSmax + (0.009) WDmax + (0.163) WSmean R= 0.481 (significant in 0.05)

Ozone amount (ppb) using 'stepwise method' for spring season = 15.541 + (-0.170) RHmin + (1.844) Tmin + (-1.246) Tmean + (0.663) SH R= 0.310 (significant in 0.05)

Ozone amount (ppb) using 'enter method' for summer season = -94.505 + (1.212) Tmin + (2.785) Tmax + (-0.623) RHmin + (0.533) RHmax +(0.076) SH + (-1.823) E + (-0.881) WSmax + (0.004) WDmax + (-0.881) WSmean R= 0.678 (significant in 0.01)

Ozone amount (ppb) using 'stepwise' for summer season = -25.073 + (1.805) Tmean R= 0. 513 (significant in 0.05)

Ozone amount (ppb) using 'enter method' for autumn season = 23.043 + (1.332) Tmin+ (-0.760) Tmax + (-0.865) Tmean + (-0.156) RHmin+ (-0.097) RHmax + (0.062) R + (0.908) SH + (-0.419) E + (0.587) WSmax + (0.001) WDmax + (-1.003) WSmean R= 0.781(significant in 0.01)

Ozone amount (ppb) using 'stepwise method' for autumn season = 51.924 + (-0.469) Tmean + (-0.520) RHmin R= 0.753 (significant in 0.01)

To test which annual model is better to use, RMSE (Root Mean Square of Error) is calculated for different linear models of enter and stepwise. Predicted amounts using the different annual models for 30 days during 2011 are calculated and compared with observed data during those days using RMSE equation:

$$\mathrm{RMSE} = \sqrt{\frac{\sum_{i=1}^{n}(O_{obs} - O_{cal})^2}{n}}$$

O_{obs}: observed ozone value

O_{cal}: predicted ozone value using model

The values of RMSE in both linear models of enter (10.72) and stepwise (10.43) show capability of stepwise model in predicting ozone amount compared to enter model. This result which is the same as the results of Masoudi et al. (2014a, 2014b), and Masoudi and Asadifard (2015) indicates for predicting most of air pollutants like ozone, we may take into consideration only linear models of stepwise which need less data and also its calculation is easier than enter model.

CONCLUSIONS

In the current research, air quality analyses for Shiraz, a city in the south of Iran, were conducted for ozone (O_3). Shiraz is one of the polluted cities in Iran. Hence a need was felt to carry out an ambient air quality analysis in the city. Results showed there were significant relationships between O_3 and some meteorological parameters. Based on these relations, different multiple linear regression equations for O_3 for annual and seasonal conditions were prepared. Results showed that among different prediction models, stepwise model was the best option. Also different variations in concentration during day, months, and seasons were observed. It is assumed that some of the amounts for concentration of O_3 in the afternoon (especially in summer and spring months) are higher than Primary Standards showing unhealthy condition. These results are almost in good agreement with other results regarding ozone assessment in other Iranian cities like Tehran (Masoudi et al., 2014b) and Esfahan (Gerami, 2014) and Ahvaz (Masoudi et al., 2014a).

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