

Status and Prediction of SO₂ as an Air Pollutant in Shiraz, Iran

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Abstract: In the present study air quality analyses for sulfur dioxide (SO₂), 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 SO₂ occurs generally in the morning while the least concentration was found at the afternoon and mid-night. Monthly concentrations of SO₂ showed the highest value in June while the least value in February. The seasonal concentrations showed the least amounts in autumn while the highest amounts in summer. Relations between the air pollutants 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 are 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. RMSE test showed that among different prediction models, stepwise model is the best option.

Key words: SO₂, air pollution, meteorological parameters, regression model.

Introduction

Air sustains life. But the air we breathe is not pure. It contains a lot of pollutants and most of these pollutants are toxic (Sharma, 2001). While developed countries have been making progress during the last century, air quality has been getting much worse; especially in developing countries air pollution exceeds all health standards. For example, in Lahore and Xian (China) dust is ten times higher than health standards (Sharma, 2001).

Sulfur dioxide (SO₂) is one of the seven conventional (criteria) pollutants (including SO₂, CO, particulates, hydrocarbons, nitrogen oxides, O₃ and lead). These pollutants produce the highest volume of pollutants in the air and the most serious threat for human health and welfare. Concentration of these pollutants, especially in

cities, has been regulated by Clean Air Act since 1970 (Cunningham and Cunningham, 2002).

Sulfur dioxide (also sulphur dioxide) is the chemical compound with the formula SO₂. At standard atmosphere it is a toxic gas with a pungent, irritating and rotten smell. It is released naturally by volcanic activity (en.Wikipedia.org). The presence of pollutants in the atmosphere causes a lot of problems, thus the study of pollutants' behaviour is necessary (Asrari et al., 2007). Current scientific evidence links short-term exposures to SO₂, ranging from 5 minutes to 24 hours, with an array of adverse respiratory effects, including bronchoconstriction and increased asthma symptoms. These effects are particularly important for asthmatics at elevated ventilation rates. Studies also show a connection between short-term exposure and

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increased visits to emergency departments and hospital admissions for respiratory illnesses, particularly in at-risk populations including children, the elderly, and asthmatics.

EPA's National Ambient Air Quality Standard for SO_2 is designed to protect against exposure to the entire group of sulfur oxides (SO_x). SO_2 is the component of greatest concern and is used as the indicator for the larger group of gaseous sulfur oxides (SO_x). Other gaseous sulfur oxides (e.g. SO_3) are found in the atmosphere at concentrations much lower than SO_2 . Emissions that lead to high concentrations of SO_2 generally also lead to the formation of other SO_x . Control measures that reduce SO_2 can generally be expected to reduce people's exposures to all gaseous SO_x . This may have the important co-benefit of reducing the formation of fine sulfate particles, which pose significant public health threats.

SO_x can react with other compounds in the atmosphere to form small particles. These particles penetrate deeply into sensitive parts of the lungs and can cause or worsen respiratory disease, such as emphysema and bronchitis, and can aggravate existing heart disease, leading to increased hospital admissions and premature death. EPA's NAAQS for particulate matter (PM) are designed to provide protection against these health effects (www.epa.gov).

Status of pollutants concentration and effects of meteorological and atmospheric parameters on these pollutants compose the base of 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 CO 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 modelling 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 modelling. The observed behaviour 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. As expected, the pollutants associated with traffic were at highest ambient concentration levels when wind speed was low. At higher wind speeds, dust and sand from the surrounding desert was entrained by the wind, thus contributing to ambient particulate matter levels. 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. For CO, SO_2 and PM_{10} the highest average concentrations occurred at humidity above 80%.

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 day time ozone concentrations, the pollutants NO and SO_2 being emitted to the atmosphere were being depleted. However, the model did not predict the night time ozone concentrations as precisely as it did for the day time. 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.

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 showed 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: $\text{PM}_{10} > \text{SO}_2 > \text{NO}_2 > \text{CO} > \text{O}_3$, indicating that PM_{10} was most effectively cleaned by rainfall.

The present study exhibits diurnal, monthly and seasonal variations of concentration of sulfur dioxide and also a statistical model that is able to predict amount of sulfur dioxide. This is based on multiple linear and nonlinear regression techniques. Multiple Regression estimates the coefficients of the linear and nonlinear equations, involving one or more independent variables that best predict the value of the dependent variable (sulfur dioxide 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 (Kinneer, 2002).

Materials and Methods

Study Area

The research area, Shiraz is the biggest city in the southern part of Iran (Figure 1) located around $29^\circ 30' \text{ N}$ and $52^\circ 30' \text{ E}$ 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. There

are lots of cars driven in city and also many factories and industries around it. So, Shiraz is one of the most polluted cities in Iran and needs 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, belonging to Environmental Organization of Iran, were selected to represent different traffic loads and activities.

The sampling has been performed every 30 minutes daily for each pollutant during all months of 2011 and 2012. Among the measured data in the two stations sulfur dioxide was chosen. Then the averages were calculated for every hour, monthly and seasonally for the 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 sulfur dioxide in the city.

Studying correlation of sulfur dioxide 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, 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 programme and the linear regression equation showed that the concentration of sulfur dioxide depends on the kind of meteorological parameters and also gives



Figure 1: Two photographs from the same place in Shiraz showing impacts of dust pollution during recent years (left one in clean condition and right one in worse condition).

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 was made for different seasons showing those relationships which are not observed using annual data.

The model for predicting sulfur dioxide was determined using two multiple regression modelling 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 2, 3 and 4, the diurnal, monthly and seasonal variations in concentration of SO_2 have been presented. As shown in Figure 2 the high concentration of SO_2 occurs in the morning while the least concentration occurs in the afternoon and mid-night. Monthly concentration showed the highest values in June and the least amounts in February and November months (Figure 3). Seasonal concentration showed the highest values in summer and the least amounts in autumn (Figure 4). Unfortunately, results in the morning and warm months showed that the concentrations of sulfur dioxide are upper than Primary Standards of sulfur dioxide (0.007 ppm), recommended by National Ambient Air Quality Standards (NAAQS) of Iran respectively. These results are almost in good agreement with other results regarding SO_2 assessment in Tehran (Behzadi and Sakhaei, 2014) but differ somewhat with Iranian cities of Ahvaz (Asadifard, 2013) and Esfahan (Gerami, 2014) in monthly and seasonal condition.

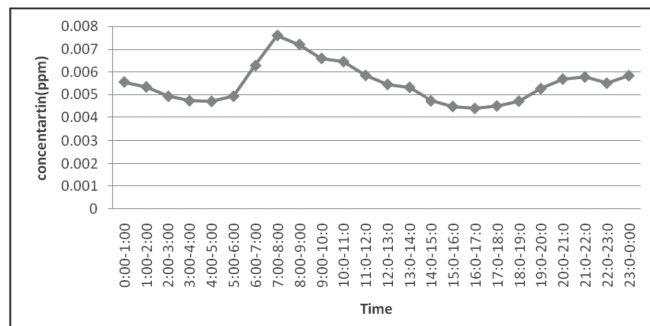


Figure 2: Diurnal variation of sulfur dioxide concentration in Shiraz (2011-2012).

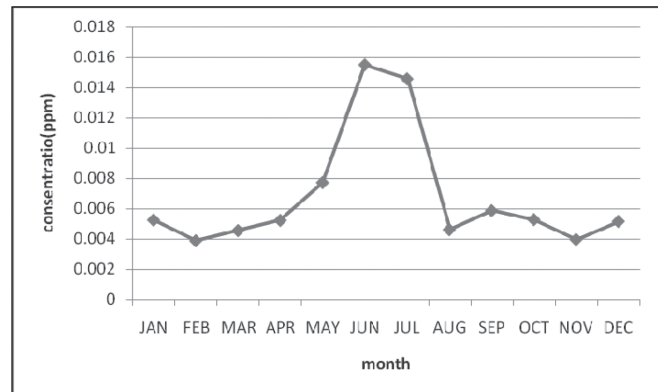


Figure 3: Monthly variation of sulfur dioxide concentration in Shiraz (2011-2012).

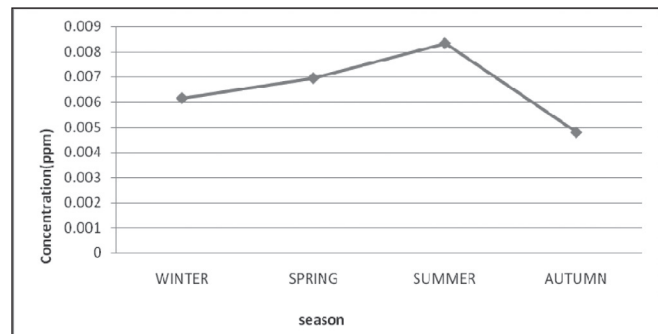


Figure 4: Seasonal variation of sulfur dioxide concentration in Shiraz (2011-2012).

Table 1 shows the relationships between SO_2 and other air pollutants. For example the concentration of SO_2 shows negative correlation with NO_2 , NO_x and O_3 while it shows positive correlation with PM and CO. These results are almost in good agreement with other results regarding SO_2 assessment in Esfahan (Gerami, 2014) but differ somewhat in other Iranian cities of Tehran (Behzadi and Sakhaei, 2014) and Ahvaz (Asadifard, 2014). Correlation coefficients significant at the 0.05 level are identified with a single asterisk (significant), and those significant at 0.01 level are identified with two asterisks (highly significant).

Table 1: Correlation between air pollutants and SO_2

	PM	NO_2	O_3	NO_x	CO
Pearson Correlation	.228**	-.532**	-.142*	-.402**	.447**
Sig. (2-tailed)	.001	.000	.035	.000	.000
N	221	221	221	221	221

Table of analysis of variance (Table 2) shows that both regressions of 'enter' and 'stepwise' methods in

annual condition are highly significant, indicating a significant relation between the different variables.

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	1086.954	11	98.814	36.330	.000**
Residual	935.638	344	2.720		
Total	2022.592	355			

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: Carbon monoxide

Analysis of variance (b)					
Model	Sum of squares	df	Mean square	F	Sig.
Regression	1067.756	5	213.551	78.278	.000**
Residual	954.836	350	2.728		
Total	2022.592	355			

Predictors: (Constant), Wind speed_(mean), Temperature_(max), Temperature_(min), Temperature_(mean), Rain. Dependent Variable: Carbon monoxide

In Tables 3 the coefficients of SO₂ 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.

The linear regression equations show that the SO₂ 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:

SO₂ amount (ppb) using 'enter method' for annual condition = .765 + (.147) Temperature_(min) + (.328) Temperature_(max) + (-.248) Temperature_(mean) + (.012) Ratio of humidity_(min) + (-.002) Ratio of Humidity_(max) + (.043) Rain + (.070) Sunshine Hours + (-.001) Wind direction_(max) + (-.051) Wind speed_(max) + (-.331) Wind speed_(mean) + (-.108) Evaporation

$$R = 0.733 \text{ (significant at 0.01)}$$

SO₂ amount (ppb) using 'stepwise method' for annual condition = 1.569 + (-.463) Wind speed_(mean) + (.320)

Temperature_(max) + (.158) Temperature_(min) + (-.293) Temperature_(mean)

$$R = 0.727 \text{ (significant at 0.01)}$$

Results of linear regression model show when wind speed_(mean) and temperature_(mean) have reverse effect on concentration of SO₂. So that, when these parameters increase, the concentration of SO₂ decreases. While, when temperature_(max) and temperature_(min) increase the concentration of SO₂ significantly increases (Table 3b). Other meteorological parameters show different effects on SO₂ amounts although these results are not significant. For example, wind direction has reverse effect on concentration of SO₂ (Table 3a). These results are almost in good agreement with other results regarding SO₂ measurements in Tehran (Behzadi and Sakhaei, 2014) and other regions (Elminir, 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 decrease most of air pollutants (Asrari et al., 2007).

The values and significance of *R* (multiple correlation coefficient) in both equations show capability of them in SO₂ amount. The amount of Adjusted *R*² in both equations is almost 0.53 showing that different parameters used can calculate almost 50% variability of SO₂. This result indicates for predicting most of air pollutants like SO₂, we should take into consideration consumption of fossil fuel. Major sources of fuel burning: Automobiles, thermal power plants, industrial processes, transportation and others. Burning of fossil fuels in thermal power plants produces 2/3 of SO₂. The automobile exhaust produces 75% of total air pollution and rapid industrialization are responsible for 20% of total air pollution (Sharma, 2001). On the other hand, *R* in enter method (0.733) is almost equal to stepwise method (0.728), showing no difference. Therefore, second equation based on stepwise method can be used to predict SO₂ 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 SO₂ in the city.

Beta in Table 3 shows those independent variables (meteorological parameters) which have more effect on dependent variable (SO₂). The beta in both Table 3 shows a highly significant effect of some variables like temperature compared to other meteorological parameters for measuring the SO₂. Parameter Sig (P-value) from Table 3 shows amount of relation

Table 3: Coefficients of carbon monoxide pollution model and regression lines for both enter (a) and stepwise (b) methods for annual condition**Coefficients (a)**

<i>Model</i>	<i>Unstandardized coefficients</i>		<i>Standardized coefficients</i>	<i>t</i>	<i>Sig.</i>
	<i>B</i>	<i>Std. Error</i>	<i>Beta</i>		
Constant	.765	1.243		.615	.539
Temperature (min)	.147	.049	.473	3.011	.003**
Temperature (max)	.328	.049	1.313	6.693	.000**
Temperature (mean)	-.248	.051	-.926	-4.831	.000**
Ratio of humidity (min)	.012	.015	.079	.805	.421
Ratio of humidity (max)	-.002	.010	-.022	-.251	.802
Rain	.043	.025	.085	1.747	.081
Sunshine hours	.070	.049	.085	1.415	.158
Evaporation	-.108	.064	-.191	-1.677	.094
Wind speed (max)	-.051	.062	-.046	-.823	.411
Wind direction (max)	-.001	.001	-.030	-.766	.444
Wind speed (mean)	-.331	.127	-.151	-2.602	.010**

Dependent Variable: SO₂**Coefficients (b)**

<i>Model</i>	<i>Unstandardized coefficients</i>		<i>Standardized coefficients</i>	<i>t</i>	<i>Sig.</i>
	<i>B</i>	<i>Std. Error</i>	<i>Beta</i>		
Constant	1.569	.480		3.269	.001
Temperature (max)	.320	.036	1.279	8.809	.000**
Wind speed (mean)	-.463	.087	-.212	-5.321	.000**
Temperature (mean)	-.293	.048	-1.094	-6.138	.000**
Temperature (min)	.158	.040	.508	3.906	.000**

Dependent Variable: SO₂

between SO₂ and meteorological parameters. For example, Table 3a shows wind speed (mean) has higher effect on SO₂ than wind direction.

On the other hand, in Table 4 the linear regression equations of SO₂ amount are presented for both enter and stepwise methods in different seasonal condition. Almost all of the models except summer model of stepwise method are significant. Stepwise methods show those meteorological parameters which are most important during these seasons for estimating the pollution. Among the models, autumn models have the highest *R* while the *R* of summer models shows the least. *R* in spring and autumn models are higher than in annual models, also indicating that relations between the pollutant and meteorological parameters are stronger than whole year during this season. These results are almost in good agreement with other results regarding

SO₂ assessment in Tehran (Behzadi and Sakhaei, 2014).

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:

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

where *O_{obs}* is observed SO₂ value and *O_{cal}* is predicted SO₂ value using model.

The values of RMSE in both linear models of enter (2.809) and stepwise (2.763) show capability of stepwise model in predicting SO₂ amount compared to enter model. This result which is the same as the results

Table 4: SO₂ amount (ppb) using two methods of enter and stepwise for different seasonal condition

Season	Enter method	R	Stepwise method	R
Winter	$= 2.044 + (-.132) T_{\min} + (.162) T_{\max} + (.017) RH_{\min} + (-.013) RH_{\max} + (.003) R + (-.035) SH + (-.046) E + (-.47) WS_{\max} + (-.001) WD_{\max} + (-.058) WS_{\text{mean}}$.561 (significant at 0.01)	$= .680 + (.260) T_{\max} + (.016) RH_{\min} + (-.238) T_{\text{mean}}$.521 (significant at 0.01)
Spring	$= .268 + (.021) T_{\min} + (.383) T_{\max} + (-.147) T_{\text{mean}} + (-.39) RH_{\min} + (-.014) RH_{\max} + (.194) R + (-.042) SH + (-.204) E + (-.013) WS_{\max} + (-.002) WD_{\max} + (.002) WS_{\text{mean}}$	0.834 (significant at 0.01)	$= -4.399 + (.459) T_{\max} + (.158) E + (-.150) T_{\text{mean}}$.820 (significant at 0.01)
Summer	$= 4.810 + (-.312) T_{\min} + (.124) T_{\max} + (.137) RH_{\min} + (-.009) RH_{\max} + (-.021) SH + (.442) E + (-.272) WS_{\max} + (.001) WD_{\max} + (.017) WS_{\text{mean}}$.453 (significant at 0.01)	-	-
Autumn	$= 16.323 + (-.244) T_{\min} + (.170) T_{\max} + (-.009) RH_{\min} + (-.126) RH_{\max} + (.001) R + (-.245) SH + (-.109) WS_{\max} + (.000) WD_{\max} + (.022) WS_{\text{mean}}$.913 (significant at 0.01)	$= 15.161 + (-.260) T_{\min} + (.214) T_{\max} + (-.204) SH + (-.130) RH_{\max}$.910 (significant at 0.01)

Note: T_{mean} = Temperature (mean), T_{\max} = Temperature (max), T_{\min} = Temperature (min), WS_{mean} = Wind speed (mean), WS_{\max} = Wind speed (max), WD_{\max} = Wind direction (max), RH_{mean} = Ratio of humidity (mean), RH_{\max} = Ratio of humidity (max), RH_{\min} = Ratio of humidity (min), SH = Sunshine hours, R = Rainfall, E = Evaporation

of Asadifard (2013) and Masoudi et al. (2014) indicates for predicting most of air pollutants like SO₂; we may take into consideration only linear models of stepwise which need less data and also its calculation is easier than enter model.

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