STATUS AND PREDICTION OF SULFUR DIOXIDE AS AN AIR POLLUTANT IN TEHRAN, IRAN

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ABSTRACT

In the present study, air quality analyses for sulfur dioxide (SO₂) were conducted in Tehran, Capital of Iran. The measurements were taken in four different locations to prepare average data in the city. The averages concentrations were calculated for every 24 hours, each month and each season. Results showed that the highest concentration of sulfur dioxide occurs generally in the mid-night and beginning of morning while the least concentration was found at the afternoon. Monthly concentrations of the sulfur dioxide showed the highest value in August while the least value in April. The seasonal concentrations showed the least amounts in spring while the highest in summer. Then 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, dew point and rainfall were considered as independent variables. The level relationships between concentration of pollutant and meteorological parameters were expressed by multiple linear and nonlinear 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: *sulfur dioxide, 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 (SO_2) 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 on 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 SO_2 . 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 pollutant' behavior is necessary (Asrari et al., 2007). Current scientific evidence links short-term exposures to SO_2 , 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 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 (SOx). SO_2 is the component of greatest concern and is used as the indicator for the larger group of gaseous sulfur oxides (SOx). 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 SOx. Control measures that reduce SO_2 can generally be expected to reduce people's exposures to all gaseous SOx. This may have the important co-benefit of reducing the formation of fine sulfate particles, which pose significant public health threats.

SOx 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).

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 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 show 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 NO₂ and O₃ occurred at humidity $\leq 40\%$ indicative for strong vertical mixing. For CO, SO₂ and PM_{10} the highest average concentrations occurred at humidity above 80%. In another research, data on the concentrations of seven air pollutants (CH₄, NMHC, CO, CO₂, NO, NO₂ and SO₂) 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 SO₂ 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 is estimated to be: PM_{10} >SO₂>NO₂>CO>O₃, indicating that PM₁₀ 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

DOI: 10.6092/issn.2281-4485/7420

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 (Kinnear, 2002).

Materials and Methods

Study Area

The research area, Tehran is the capital of Iran located between $35^{\circ} 35'$ N to $35^{\circ} 50'$ N latitudes $51^{\circ} 05'$ E to $51^{\circ} 35'$ E Longitudes and the elevation is 1280 m above the mean sea level. Area of Tehran is 730 km². It has moderate climate and residential population was 8.5 million in 2011. There are about one million cars in city and many factories and industrials place around the city. So, Tehran is one of the most polluted cities in Iran and needs to carry out an ambient air quality analysis in this city.



Figure 1

Two photos graphs from the same place in Tehran city showing impacts of air pollution during recent years. (right one in clean condition and left one in worse condition).

Data and methodology

Four available sampling stations in the city called, Azadi, Gholhak, Tajrish and Sorkhe-Hesar, belong 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 2009 and 2010. Among the measured data in the four stations dioxide was chosen. Then the averages were calculated for every hour, each month and each season for the four stations by Excel. Finally averages of data at four stations were used to show air pollution situation as diurnal, monthly and seasonal graphs of concentration 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 & mean), ratio of humidity (min, max and mean), precipitation, sunshine hours, dew point (mean), wind direction (max) and wind speed (max and mean).

In the next step, daily average data at four stations in 2010 was considered as dependent variable for statistical analysis while daily data of meteorological parameters during this year were selected as independent variables in SPSS programme and the multiple regression equations showed that the concentration of sulfur dioxide depends on the kind of meteorological parameters and also give an idea about the levels of these relations. The relationship between the dependent variables and each independent variable has been considered for both linear and nonlinear techniques. The significant values in output are based on fitting a single model. Also linear regression equation was made for different seasons maybe show those relationships which are not observed using annual data.

The model for predicting sulfur dioxide 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 figure 2, 3 and 4, the diurnal, monthly and seasonal variations in concentration of sulfur dioxide have been presented. As shown in fig. 2 the high concentration of sulfur dioxide occurs in the mid-night and beginning of morning while the least concentration occurs in the afternoon.





Stability of atmosphere and inversion during these times may be responsible for this high concentration. Monthly concentration of sulfur dioxide showed the highest values in August and the least in April (Fig. 3). Seasonal concentration of the Nitrogen dioxide showed the highest values in summer and the least in spring (Fig. 4). Unfortunately, all graphs showed that the concentrations of sulfur dioxide are upper than Primary Standards of sulfur dioxide recommended by National Ambient Air Quality Standards (NAAQS) of Iran (0.007 ppm), protecting human health. These results are almost in good agreement with results obtained in other

DOI: 10.6092/issn.2281-4485/7420

Iranian cities of Shiraz (Ordibeheshti and Rajai, 2014), Esfahan (Gerami, 2014) and Ahvaz (Asadifard, 2013).



Table 1 shows the relationships between sulfur dioxide and other air pollutants. For example the concentration of sulfur dioxide shows negative correlation with CO while it shows positive correlation with O_3 and PM_{10} which all of the relationships are significant. SO₂, O_3 and PM_{10} are related to industrial activity and other resources till to traffic volume while CO is related to auto exhausts. These results are almost in good agreement with other results regarding SO₂ assessment in Ahvaz (Asadifard, 2014) but show weak agreement with results of SO₂ in Shiraz (Ordibeheshti and Rajai, 2014) and Esfahan (Gerami, 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).

	СО	NO ₂	NO _x	O ₃	PM_{10}	Table 1	
Pearson Correlation	-0.141**	0.006	-0.001	0.153**	0.310**	between air	
Sig.(2-tailed)	0.008	0.913	0.991	0.004	0.000	pollutants and	
Ν	357	357	357	357	357	suijui atoxiae.	

Table of analysis of variance (Table 2) shows that both regressions of 'enter' and 'stepwise' methods for annual condition are highly significant, indicating a significant relation between the different variables.

	Model	Sum of Squares	df	Mean Square	F	Sig.	Table 2Analysis of
	Regression	17805.465	12	1483.789	9.685**	0.000	variance for both
$(a)^1$	Residual	53621.111	350	153.203			regressions of 'enter' (a) and
	Total	71426.576	362				<i>'stepwise'</i> (b)
	Regression	7721.108	2	3860.554	21.816**	0.000	methods for annual condition
$(\mathbf{b})^2$	Residual	63705.468	360	176.960			
	Total	71426.576	362				_

 ¹ 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), Dew point. Dependent Variable: sulfur dioxide
² Predictors: (Constant), Sunshine Hours, Wind speed (mean)

Dependent Variable: sulfur dioxide

In Tables 3 the coefficients of sulfur dioxide 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.

Model		Unstandardized Coefficients		Standardized Coefficients	t	Sig.
		B Std. Error		Beta		
	(Constant)	-13.118	11.120		-1.180	0.239
	Temperature (mean)	-12.232	5.933	-9.543	-2.062*	0.040
(a) ¹	Temperature (max)	8.266	2.984	6.978	2.770**	0.006
	Temperature (min)	5.485	3.010	3.973	1.822	0.069
	Wind speed (mean)	-0.639	0.354	-0.128	-1.807	0.072
	Wind speed (max)	0.081	0.168	0.035	0.483	0.629
	Wind direction (max)	-0.003	0.010	-0.013	-0.255	0.799
	Ratio of Humidity (mean)	0.941	0.196	1.369	4.794**	0.000
	Ratio of Humidity (max)	-0.247	0.088	-0.409	-2.797**	0.005
	Ratio of Humidity (min)	0.098	0.133	0.118	0.742	0.459
	Rain	-1.415	0.429	-0.187	-3.298**	0.001
	Dew point	-1.613	0.353	-0.580	-4.572**	0.000
	Sunshine Hours	0.544	0.285	0.139	1.907	0.057
(b) ¹	(Constant)	47.349	2.077		22.802	0.000
	Sunshine Hours	1.214	0.196	0.310	6.187**	0.000
	Wind speed (mean)	-0.729	0.249	-0.146	-2.924**	0.004
¹ Dependent Variable: sulfur dioxid						

Table 3. Coefficients of sulfur dioxide pollution model and regression lines for bothenter (a) and stepwise (b) methods for annual condition.

DOI: 10.6092/issn.2281-4485/7420

The linear regression equations show that the sulfur dioxide pollution depends on the meteorological parameters and also give an idea about the levels of relations. The linear model equations after using 'enter method' [1] and 'stepwise method' [2] for annual condition are:

Sulfur dioxide amount (ppb) using 'enter method' for annual condition = (-13.118) + (-12.232) Temperature_(mean) + (8.266) Temperature_(max) + (5.485) Temperature_(min) + (.098) Ratio of humidity_(min) + (-0.247) Ratio of Humidity_(max) + (0.941) Ratio of Humidity_(mean) + (-1.415) Rain + (0.544) Sunshine Hours + (-0.003) Wind direction_(max) + (0.081) Wind speed_(max) + (-0.639) Wind speed_(mean) + (-1.613) [1] Dew point R= 0.449 (significant at 0.01)

Sulfur dioxide amount (ppb) using 'stepwise method' for annual condition = 47.349 + (1.214) Sunshine Hours + (-0.729) Wind speed (mean) R = 0.329 (significant at 0.01) [2]

Results of linear regression model show that wind speed (mean), temperature (mean), ratio of humidity (max), rain and dew point have reverse effect on concentration of sulfur dioxide. So that when this parameters increase, the concentration of sulfur dioxide decreases. While, when Sunshine Hours, Temperature (max) and Ratio of Humidity (mean) increase, the concentration of sulfur dioxide significantly increases (Table 3). Other meteorological parameters show different effects on sulfur dioxide amounts although these results are not significant. For example Wind direction has reverse effect (more western winds) on concentration of sulfur dioxide (Table 3a). These results are almost in good agreement with other results regarding sulfur dioxide measurements in Shiraz (Ordibeheshti and Rajai, 2014) and other studies (Elminir, 2005; Li et al., 2014).

Actually some of these events happen in real condition. Increasing 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 predicting sulfur dioxide amount. The amount of Adjusted R² in both equations is almost 0.22 and 0.10 for enter and stepwise methods showing that different parameters used can calculate almost 15% variability of sulfur dioxide. This result indicates for predicting most of air pollutants like sulfur dioxide, 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).

Beta in table 3 shows those independent variables (meteorological parameters) which have more effect on dependent variable (sulfur dioxide). The beta in the both tables 3 shows a highly significant effect of some variables like sunshine hours and ratio of humidity (mean) compared to other meteorological parameters

for measuring the sulfur dioxide which is close to the results of Masoudi et al. (2014) for ozone. Parameter Sig (P-value) from Table 3 shows that amount of relation between sulfur dioxide and meteorological parameters. For example, table 3a shows wind speed (max) has higher effect on sulfur dioxide than wind direction. On the other hand, in table 4 the linear regression equations of sulfur dioxide amount are presented for both enter and stepwise methods for different seasonal condition. Almost all of the models except summer model of enter method are significant. Stepwise methods show those meteorological parameters which are most important during these seasons for estimating the pollution. Among the models, spring models have the highest R while R of summer models shows the least. R in winter, spring and autumn models are higher than in annual models model, 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 SO₂ assessment in Shiraz (Ordibeheshti and Rajai, 2014) and Ahvaz (Asadifard, 2014).

Season	Enter method	R	Stepwise method	R
Spring	= (-10.118) + (-13.729) Tmean + (9.249) Tmax + (5.635) Tmin + (0.741)	0 745	- (- 6 045) +	0 691
	WSmean + (-0.124) WSmax + (0.019)	(significant	(3.209) Tmax +	(significant
opring	WDmax + (0.181) RHmean + (-0.162)	at 0.01)	(-1.822) Tmean +	at 0.01)
	RHmax + (0.507) RHmin + (0.740) Rain		(0.280) RHmean	
	+ (0.355) Dew $+$ (0.240) Sunshin			
	= 57.992 + (0.535) Tmean + (-0.092)			
	Tmax + (-0.289) Tmin + (-0.539) 0.422		- 44 370 -	0.323
Summor	WSmean + (0.008) WSmax + (0.006)	VSmean + (0.008) WSmax + (0.006) (not		(significant
Summer	WDmax + (0.014) RHmean + (-0.222) (not significant)		(-0.855) w Sinear +	at 0.01)
	RHmax + (-0.274) RHmin + (-14.246)	significant)	(0.0) (0.0)	
	Rain + (0.977) Dew + (0.561) Sunshin			
	=(-42.028) + (-40.269) Tmean $+ (22.750)$		48 628 +	
	Tmax + (20.157) Tmin + (-0.677)	0.672	(2.878) Tmay \pm	0.612
Autumn	WSmean + (0.019) WSmax + (0.017)	(significant	(2.070) That + (-4.792) Dev +	(significant
Autuilli	WDmax + (0.670) RHmean + (-0.026)	at 0.01)	(-4.7)2 DCw + (0.036) RHmean+	at 0.01)
	RHmax + (.278) RHmin + (0.120) Rain +		(0.930) Krinicali+ (-0.908) Sunshin	
	(-4.470) Dew + (-0.546) Sunshin		(-0.908) Sulisiili	
Winter	= 117.232 + (-9.162) Tmean $+ (4.685)$			
	Tmax + (2.013) Tmin + (-1.209) WSmean	0.702	= 65.990 +	0.642
	+ (0.158) WSmax $+ (0.001)$ WDmax $+ (-$	(significant	(-0.680) Tmin +	(significant
	0.448) RHmean + (-0.301) RHmax +	at 0.01)	(-1.111) WSmean +	at 0.01)
	(0.257128) RHmin + (-1.471) Rain +		(-1.477) Rain	
	(2.227) Dew + (-0.440) Sunshin			

Table 4. Sulfur dioxide amount (ppb) using two methods of enter and stepwise for different seasonal condition.

Note: Tmean=Temperature (mean), Tmax =Temperature (max), Tmin=Temperature (min), WSmean = Wind speed (mean), WSmax =Wind speed (max), WDmax =Wind direction (max), RHmean = Ratio of Humidity (mean), RHmax =Ratio of Humidity (max), RHmin= Ratio of Humidity (min), Dew =Dew point, sunshin= Sunshine Hours

Also the nonlinear multiple regression equation of sulfur dioxide amount using parameters of linear stepwise method for annual condition is calculated:

Sulfur dioxide amount (ppb) using nonlinear regression for annual condition =

$$40.729 \times (1.028^{(Sunshin)}) + 48.992 + (15.959/WSmean)$$
 R²= 0.108 [3]

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 and nonlinear model. 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:

$$RMSE = \sqrt{\frac{\sum_{i}^{M} = 1(O_{obs} - O_{cal})}{n}}$$
[4]

where: O_{obs} = observed SO₂ value and Ocal = predicted SO₂ value usin model.

Conclusion

The values of RMSE in both linear models of enter (24.63) and stepwise (26.73) show capability of them in predicting sulfur dioxide amount compared to nonlinear model value (73.68). On the other hand, RMSE in enter method is almost equaled to stepwise method, showing no different significant. This result which is the same as the results of Asadifard (2013) and Masoudi et al. (2014) indicates for predicting most of air pollutants like sulfur dioxide, we may take into consideration only linear models of stepwise which need less data compared to enter model and also its calculation is easier than nonlinear model.

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