

PREDICTION OF NO₂ CONCENTRATIONS IN A GAS REFINERY USING AIR DISPERSION MODELING

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Abstract. Nitrogen dioxide (NO₂) is considered as one of the most important ambient air pollutants in gas refineries. In this study, AERMOD dispersion model was applied for prediction of NO₂ ambient concentrations and dispersion patterns from point sources, including 13 stacks and six flares in a gas refinery located in Asaluyeh, Iran. For this purpose, the NO₂ concentrations exhausted from the stacks were measured by a portable emission analyzer and the NO₂ concentrations resulted from the flares were estimated using the emission factors. Moreover, the amounts of ambient NO₂ concentrations in 10 monitoring stations were measured in four seasons. Then, the ambient NO₂ concentrations and dispersion patterns were predicted using the AERMOD model within a domain of 10 × 10 km², in 1-hr and 12 months averages and the unhealthy zones in the study area were defined. The results revealed that for both annual observed and predicted values, ambient NO₂ concentrations were higher than WHO standard limits but they did not exceed the US EPA standard limits. However, the hourly observed and predicted concentrations were lower than the standard levels. Statistical methods were used for comparing the predicted and observed NO₂ concentrations. Simulation results indicated that the predicted concentrations were underestimated by a factor of 20 % in comparison to the measured ones which revealed the estimated contribution of other sources including mobile sources and neighbor sources located in the vicinity of the gas refinery.

Keywords: AERMOD, air quality, emission, performance, point source

Introduction

Air pollution will adversely affect the quality of life and human health. Therefore, it has been widely concerned by environmental experts in the world.

The World Health Organization (WHO) has reported that about 2.7 million people die due to the health effects of air pollution annually (WHO, 2014). The minimum concentrations of gasses in the atmosphere could be essential in determining the health status of the societies (Shooter et al., 1993). Meanwhile, the air quality management policies are important in order to reduce the acute effects of air pollutants. Identifying the emissions due to different sources and assessing their adverse effects are important for appropriate air quality management (Bhanarkar et al., 2005).

The NO_x concentrations exhausted from the gas refinery usually increase with rising ignition temperature that leads to producing NO, and then oxidized to nitrogen dioxide (NO₂) in the presence of oxygen in the atmosphere. NO₂ is an air pollutant with a 1–3 days

lifetime in mesoscale. Ambient NO₂ concentrations have been regulated by the U.S. EPA as one of the six primary air pollutants. Respiratory tract infections due to the pollutant's interaction with the immune system may be increased by NO₂ exposure. Furthermore, it has been associated with slight respiratory symptoms at low NO₂ concentrations and with death in indoor locations (Chen et. al, 2007). It is caused throat, eyes and nose irritation for inhalation in human (Perkins, 1974). Direct effects of nitrogen oxides can be considered as the creation of smog and photochemical ozone components (Akdemir et al., 2013). The U.S. EPA (2010) has determined that the hourly average ambient NO₂ concentrations should not exceed 200 µg/m³. In order to meet the regulation goals and for minimizing the adverse effects of air pollutants, it is essential to study the dispersions of air pollutants which are affected by several factors including atmospheric stability, orography, obstacles, roughness, maximum mixing height, wind speed and wind direction, and height of the release above ground level (Crowl and Louvar, 2002; Seangkiatiyuth et al., 2011).

Various methods including measurement, emission inventory and simulation studies are applied to assess air pollution impacts due to pollutants released into the atmosphere and to monitor whether pollutants' concentrations exceed the standard limits (Capelli et al., 2013). Air quality dispersion modeling is important for predicting the pollutants' concentrations, simulating the emission distribution patterns and the spatial allocation of outdoor air pollutants (Holmes and Morawska, 2006; Zhang et al., 2010). On the other hand, dispersion models are effective tools for predicting the contributions of emissions due to various pollutant sources including traffic, industry, residential and commercial sectors (Cimorelli et al., 2005). Advanced dispersion models such as American Meteorological Society/Environmental Protection Agency Regulatory Model (AERMOD) were often applied for predicting the air quality after a long time, e.g. at least 10 years (Ma et al., 2013).

As AERMOD model can combine various concepts and complex algorithms, it has been used for evaluating the ambient concentrations and dispersion patterns of some pollutants, including SO₂, PM₁₀, VOCs, hydrogen cyanide (HCN), and sulfur hexafluoride (SF₆) (Bhardwaj et al., 2005; Orloff et al., 2006; Venkatram et al., 2001, 2004, 2009; Zou et al., 2009, 2010).

Moreover, AERMOD can be used for simulating the dispersion of heavy metals, such as chromium and mercury (Sax and Isakov, 2003; Mazur et al., 2009). When upper climatological data are not accessible, AERMOD can be integrated with meteorological models such as MM5, WRF, and RAMS, etc (Caputo et al., 2003; Isakov et al., 2007; Kesarkar, 2007).

Some studies were carried out regarding prediction of NO₂ concentrations and dispersion patterns with AERMOD. The dispersion of NO₂ emitted from four cement plants in Thailand were studied using AERMOD model. In this study, the predicted ambient NO₂ concentrations were compared with the observed values in 12 receptors. The results showed that simulated and observed concentrations were in better agreement within 1-5 km than those at receptors situated in 5km further away from the reference point. Moreover, the results revealed that both observed predicted ambient NO₂ concentrations were not exceed the limit values set by the National Ambient Air Quality Standards (Seangkiatiyuth et al., 2011). Furthermore, AERMOD was used for modeling of NO_x, PM_{2.5} and SO₂ emissions from point and line sources in Halifax, Nova Scotia, Canada in 50 × 50 km² domains over 1-hr, monthly and annual periods averages. The evaluation of model performance was done by applying

statistical parameters and the results showed poor agreement among the observed and simulated results (Gibson et al., 2013). In another study, Cohan et al. (2011) examined the dispersion and predicted concentrations of NO_x and PM_{2.5} in port communities in California using AERMOD. The correlation coefficients for NO_x and PM_{2.5} were 50% and 43%, respectively (Cohan et al., 2011). AERMOD model was also used to simulate the NO₂ and SO₂ concentration in Beilun area. The results demonstrated that the ambient average concentrations of NO₂ and SO₂ were equivalent to 16.7%~58.3% and 26.7%~53.3% of the clean air quality standard respectively. However, concentrations of NO₂ and SO₂ were relatively higher in the upper air of Beilun region. NO₂ and SO₂ gasses in the upper air were a reason for the high potential of acid rain on Beilun region (Hasson et al., 2013).

In order to study the performance of applying emission control devices, AERMOD modeling system was applied for predicting the air pollution in Xuanwei, an important industrial city in China, and it was concluded that AERMOD can be used properly for simulating NO_x and SO₂ emissions (Ma et al., 2013). The emission, transport, dispersion, and concentration analysis of PM emitted from a big industrial complex in Malagueño city, Argentina, were also conducted using AERMOD model. The model was applied to 224 emission sources. The performance of the model was evaluated by comparing the simulated results with the observed Total Suspended Particulate matters (TSP) at two monitoring sites for 62 continuous days in winter. The results showed the impact of the industrial activities on the local PM concentrations, from which stockpiles and unpaved industrial roads were the main emission sources, straightly affecting two of the nearest neighborhoods in the study area (Abril et al., 2016). In another study, AERMOD model was used to predict SO₂, CO, NO_x and PM₁₀ emissions impacts at receptors due to calcining processes including handling and storage of raw petroleum coke in Argentina. The observed and predicted ambient pollutants' concentration levels were compared with public health standards. The results revealed that the exposures of the simulated NO_x, CO, PM₁₀ and SO₂ concentrations were lower than the air quality standards. However, the PM concentration level was higher than the standard limits (Singh et al., 2015).

The exposures of various pollutants including NH₃, CH₄, CO, CO₂, H₂S, NO₂, N₂O, SO₂ and organic dust were estimated for populations residing in close vicinity to ten poultry concentrated animal feeding operations (CAFOs) located in Central Poland. AERMOD model was used to predict the pollutants' concentrations in order to compute the Hazard Index (HI) for a mixture of chemicals. Results presented that the estimated hazard indexes which were less than unity; therefore there was low potential for adverse health consequences for the surrounding society for the combined mixture of chemicals (Pohl et al., 2016).

A comparative study regarding the health effects of emissions due to vehicles and industries were conducted in China and Pakistan. In this study, ambient CO, NO₂, and SO₂ concentrations were measured and compared with U.S. EPA, WHO and national clean air standards in China and Pakistan (Niaz et al., 2015).

The performances of ISCST-3, AERMOD, and CALPUFF models for NO_x and CO emissions in point, line and area sources were compared together using statistical analysis in Körfez. The results revealed that AERMOD predictions for NO_x concentrations were lower than those predicted by ISCST-3, and CALPUFF models. Meanwhile, CO concentrations predicted by AERMOD were among the concentration levels simulated by CALPUFF and ISCST-3 (Demirarslan and Doğruparmak, 2016).

The main goal of the present study was to simulate the ambient concentrations and dispersion patterns of NO₂ emitted from stacks and flares in a sour gas refinery located in a complex industrial region with special climatological and topographical conditions and to determine the contribution of the gas refinery in NO₂ emissions using AERMOD dispersion model.

In this study, ambient NO₂ concentrations in a gas refinery situated in Asaluyeh was measured seasonally in 10 receptors in and around the refinery from June 2014 to May 2015. Thereafter, the 1-hr and annual ambient concentrations and dispersion patterns of NO₂ were simulated by applying AERMOD dispersion model in the area of 10×10 km². Moreover, the predicted concentrations in receptors were compared with the measured ones using statistical methods. Then, unhealthy zones were determined in the study region using contour plots of the seasonal NO₂ distribution patterns. Finally, the contribution of emissions due to point sources in this refinery was determined.

Materials and Methods

Study Area

Asaluyeh, a sub-city of Kangan, belongs to Boushehr Province, which locates in the southwest of Iran. The desired gas refinery as shown in *Figure 1*, is situated on 27° 30' to 27° 31' north latitude and 52° 34' to 52° 36' east longitude in Asaluyeh. It was commissioned with a refining 50 million cubic meters per day sour gas and 80,000 barrel per day gas condensate to supply for the Iran's local strategic demand.

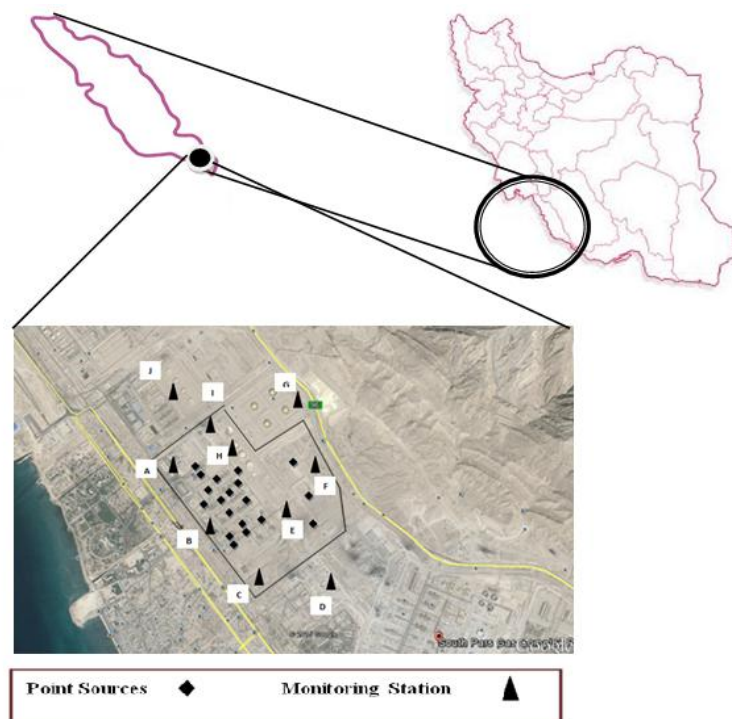


Figure 1. The study area, the point sources and monitoring stations

Field Measurements

Emission Sources

The point sources in the gas refinery included 13 stacks and six flares. The exit velocity of the flue gas and concentrations of NO₂ emitted from the stacks were measured by a portable emission analyzer (TESTO 350 XL), in four seasons (June 2014 - May 2015) and four times in each season (ASTM, 2011a). The emission rate of NO₂ resulting from the flares was also determined by emission factors according to EPA/AP-42 method.

The required AERMOD inputs data were including emission height (m), flue gas temperature (°C), inner diameter of stack (m), flue gas velocity (m/s), geographical coordinates (X,Y) and elevation above the ground level (m) (*Table 2*). The locations of point sources are illustrated in *Figure 1*. In the current study, the maximum NO_x emission for the continuous operation has been considered. The total NO₂ emission rate from 19 point sources which evaluated according to Tier2 in AERMOD guidelines (US EPA, 2009, 2010a) was 270 g/s.

Table 2. Characteristics of point sources in the gas refinery

Sources*	Cartesian Coordinates		Diameter	Height	Flue Gas Temperature	Flue Gas Velocity
	X (m)	Y (m)	(m)	(m)	(°C)	(m/s)
BO-1	0	0	3	42.7	160.5	6.1
BO-2	-17	-10	3	42.7	154	5.2
BO-3	-33	-26	3	42.7	159	5.2
BO-4	-49	-35	3	42.7	154	5.1
GTG-1	-183	145	4	30	550	32
GTG-2	-167	130	4	30	550	32
GTG-3	-153	111	4	30	550	32
GTG-4	-139	94	4	30	550	32
GTC-1	311	-110	3.15	17.3	520	30
GTC-2	325	-129	3.15	17.3	520	30
GTC-3	339	-129	3.15	17.3	520	30
X-1	129	-266	2.1	117	592	10
X-2	31	-138	2.1	117	500	10
F-1**	998	221	0.48	142.8	800	60
F-2**	1057	474	0.48	142.8	800	60

* BO: Boiler, GTG: Gas Turbine Generator, X: Incinerator, GTC: Gas Turbine Compressor, F: Flare

**According to Iowa Procedure the modified diameter and height were used for flares.

Ambient NO₂ Concentrations

Measurements of 1-hr average of ambient NO₂ concentrations were performed using a mobile device (LSI-Lastem Babuc A) in four seasons and four times in each season from June 2014 to May 2015 in 10 receptors (ASTM, 2011b). The locations of receptors from reference point (BO-1) have been presented in *Table 3*.

Table 3. Cartesian coordinates of the receptors

Receptor	Cartesian Coordinates	
	X (m)	Y(m)
A	402	-478
B	-127	-141
C	-286	712
D	-460	667
E	-201	982
F	-13	846
G	55	799
H	594	343
I	904	-4
J	691	-281

Meteorological Data

All Surface and upper-air weather data (wind speed, wind direction, temperature, cloud cover, dew point temperature, pressure at sea level, rainfall and humidity, solar radiation) in eight time steps (0:00, 3:00, 6:00, 9:00,12:00, 15:00, 18:00, 21:00) in a day were provided as meteorological inputs for AERMOD from June 2014 to May 2015 (IRIMO, 2015).

The annual wind rose was produced based on data received from Asaluyeh Weather Station (*Figure 2*). The prevailing wind direction in this station was from NW to SE in all seasons. The AERMET was employed to simulate the climatically conditions using the values of Bowen ratio, surface roughness length and Albedo parameters based on the types of the surrounding vegetation and land use in the study area. The values of these parameters have been presented in *Table 1*.

Table 1. The surface specifications according to meteorological data

Sector Number	Beginning Direction (Degree)	Ending Direction (Degree)	Albedo	Bowen Ratio	Roughness Length (m)
1	0	150	0.28	6	0.3
2	150	300	0.14	0.1	0.0001
3	300	360	0.28	6	0.3

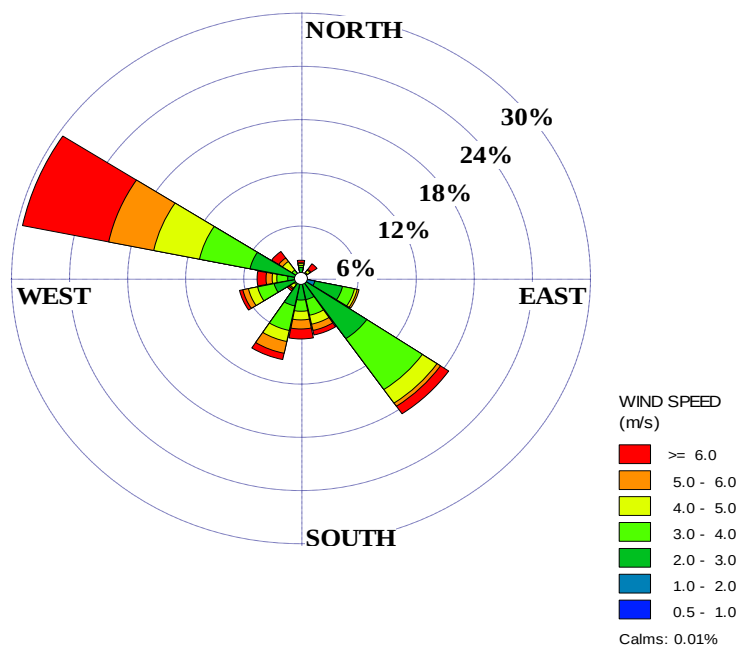


Figure 2. The annual wind rose in the study area

Elevation Data

In order to predict the NO₂ concentrations at receptors, elevation data was required as model input. In this study, the required digital elevation files were provided from Iran National Cartographic Center. The 3D view of topography in the study region produced by GIS with a resolution of 90 m has been shown in *Figure 3*.

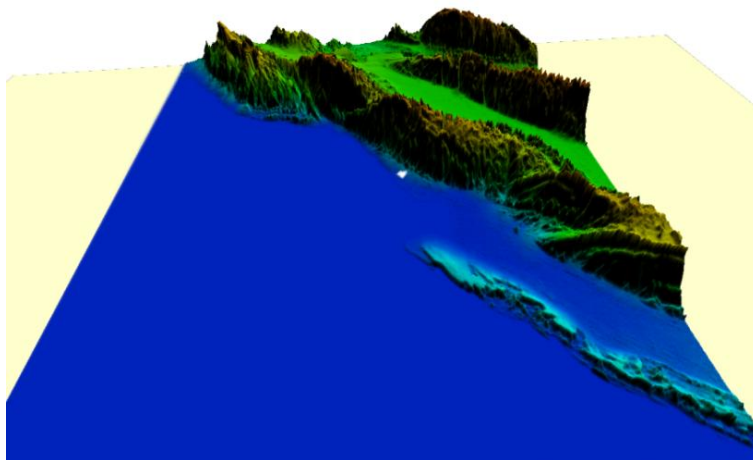


Figure 3. The 3D view of topography in the study region

AERMOD Modeling System

AERMOD dispersion model developed by the U.S. EPA is a steady-state Gaussian model for predicting air pollutants' concentrations and dispersion patterns due to various emission sources. AERMOD model consists of three sections: Meteorological Preprocessor (AERMET), Terrain Preprocessor (AERMAP) and AERMOD Gaussian Plume Model (US EPA, 2004a, 2004b, 2009). It is necessary to provide all emission rates due to various sources (point, line, area, and volume sources) and the meteorological and topographical data to predict pollutants' concentrations and dispersion patterns in rural and urban areas, calculating building downwash effect, terrain adjustment, etc. at short-range (up to 50 km) (Cimorelli et al., 2005; Perry et al., 2005). AERMOD makes distribution estimations by utilizing the meteorological characteristics of the study region, including stack diameter and height; stack temperature, flue gas velocity, wind direction and wind speed (Mokhtar et al., 2014; Huertas et al., 2014; Stein et al., 2007).

In this study, simulations for NO₂ ambient concentrations and dispersion patterns were performed for the total study area of 100 km², where a grid of 50 m × 50 m was built.

Evaluation of AERMOD Performance

In order to evaluate the model performance, the results of field measurement and model simulation were compared together using statistical methods. Statistical Parameters suggested by U.S. EPA (U.S. EPA, 2003, 2005) including: “correlation coefficient (COR)”, “Normal Mean Bias (NMB)”, “Normal Mean Error (NME)”, “fractional bias (FB)”, and also “Index of Agreement (IOA)” (Luhar, 2003) were applied (*Equations 1 ~ 5*). According to *Eq. (1)*, parameter COR indicates the relationship between the Simulated and the measured results. The more the value of COR is closer to 1, the performance of the model is more satisfactory.

$$COR = \frac{\sum_{i=1}^N (s_i - \bar{s})(m_i - \bar{m})}{(\sum_{i=1}^N (s_i - \bar{s})^2 \sum_{i=1}^N (m_i - \bar{m})^2)^{1/2}} \quad (\text{Eq. 1})$$

NMB and NME parameters were applied to evaluate the model performance for simulating NO₂ concentrations. The variation ranges for COR, NMB and NME were (-1 ~ + 1), (-1 ~ + ∞), (0 ~ + ∞), respectively (U.S. EPA, 2003).

$$NMB = \frac{\sum_{i=1}^N (s_i - m_i)}{\sum_{i=1}^N m_i} \times 100 \quad (\text{Eq. 2})$$

$$NME = \frac{\sum_{i=1}^N |s_i - m_i|}{\sum_{i=1}^N m_i} \times 100 \quad (\text{Eq. 3})$$

FB indicates the tendency of the model to predict values more or less than the measured concentrations in this study (Olesen, 2001). The acceptable value for FB was less than 0.3 (Ghannam and El-Fadel, 2013).

$$FB = \frac{(\overline{M}_i - \overline{S}_i)}{0.5(\overline{M}_i + \overline{S}_i)} \quad (\text{Eq. 4})$$

IOA is sensitive to differences between the measured and simulated results means as well as to certain changes in proportionality (Luhar, 2003). The variation range for IOA was (0,1).

$$IOA = 1 - \left[\frac{\sum_1^N (S - M)^2}{\sum_1^N (|S - \overline{M}| + |M - \overline{M}|)^2} \right] \quad (\text{Eq. 5})$$

Where, S_i : simulated concentrations, M_i : measured concentrations, $\overline{S}$: average simulated concentrations, $\overline{M}$: average measured concentrations, N : the total number of measurements.

Results and Discussion

Experimental Results

The 1-hr averages of ambient NO₂ concentrations in 10 receptors in four seasons from June 2014 to May 2015 have been illustrated in *Figure 4*. For all receptors except B in summer 2014, the measured ambient NO₂ concentrations were lower than the clean air U.S. EPA standard for 1-hr ambient concentration levels. However, in all seasons the simulated ambient NO₂ concentrations in all receptors were lower than the standard limit (200 µg/m³). Based on the safety regulation for hourly NO₂ concentrations (200 µg/m³) published by the WHO Guideline, NO₂ emissions from the gas refinery showed no significant health effects on personnel. However, the average annual concentration was 87 µg/m³ which was higher than the WHO standard limit (40 µg/m³) and they have not exceeded the U.S. EPA standard limits (100 µg/m³) (WHO, 2017; U.S. EPA, 2010b).

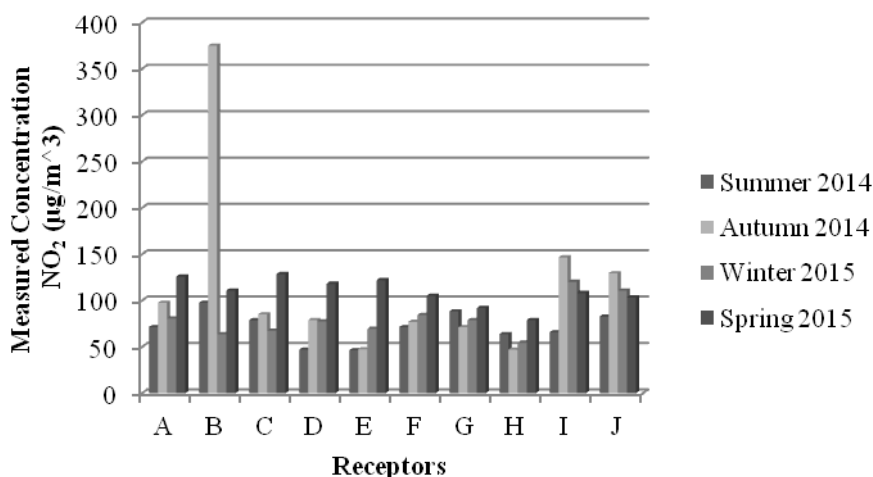
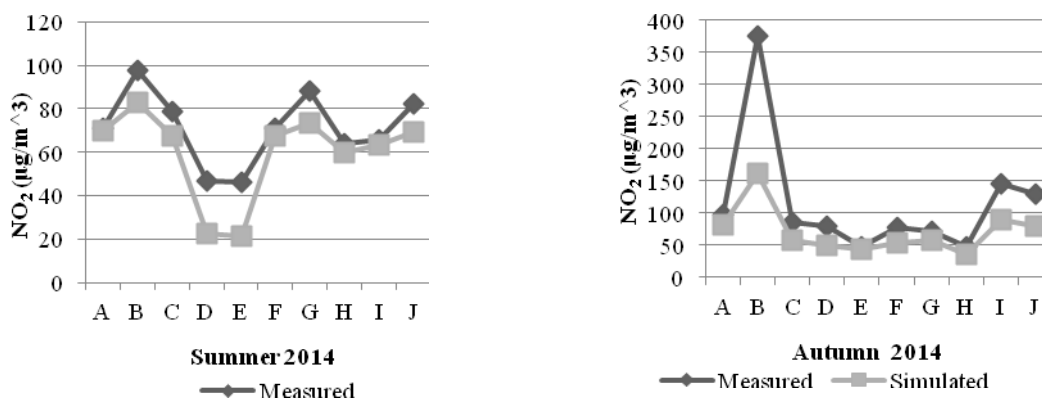


Figure 4. Ambient NO₂ concentrations in 10 receptors at 1-hr average period

The 1-hr averages of ambient NO₂ concentrations measured in 10 receptors varied from 46.4 to 97.6 $\mu\text{g}/\text{m}^3$ in summer 2014, from 47 to 375 $\mu\text{g}/\text{m}^3$ in autumn 2014, from 54.5 to 120.2 $\mu\text{g}/\text{m}^3$ in winter 2015 and from 78.9 to 125.8 $\mu\text{g}/\text{m}^3$ in spring 2015. As a result, the highest observed NO₂ ambient concentration was measured in autumn and the lowest amount was measured in summer for 1-hr average period. However, in all seasons the predicted ambient SO₂ concentrations in all receptors were lower than the US EPA clean air standard levels except for B monitoring stations in autumn 2014, due to the direction of prevailing wind (from NW to SE) and the neighboring refineries.

The simulated results also showed that the variations of NO₂ concentrations were ranged from 21.2 to 83.03 $\mu\text{g}/\text{m}^3$ in summer 2014, from 35.7 to 162 $\mu\text{g}/\text{m}^3$ in autumn 2014, from 34.5 to 87 $\mu\text{g}/\text{m}^3$ in winter 2015, and from 73 to 114.5 $\mu\text{g}/\text{m}^3$ in spring 2015, which presented the highest concentration in the autumn and the lowest value in the summer. Meanwhile, in all seasons the simulated ambient NO₂ concentrations in all receptors were lower than the clean air standard levels.

A comparison among the results of measured 1-hr average NO₂ concentrations and the simulated values for 10 monitoring stations (receptors) in four seasons in the study area is presented in *Figure 5*. As shown, the simulated ambient NO₂ concentrations were less than the measured ones for all receptors. The variations among the simulated and measured ambient NO₂ concentrations in D, I and J receptors were higher than those in other receptors. These receptors were situated on the border of the study area with other refineries and it might be resulted from emissions due to other neighboring refineries and other industries. The observed and simulated results were in good agreement. The values and locations of maximum simulated ambient NO₂ concentrations at 1-hr and annual averages are presented in *Table 4*.



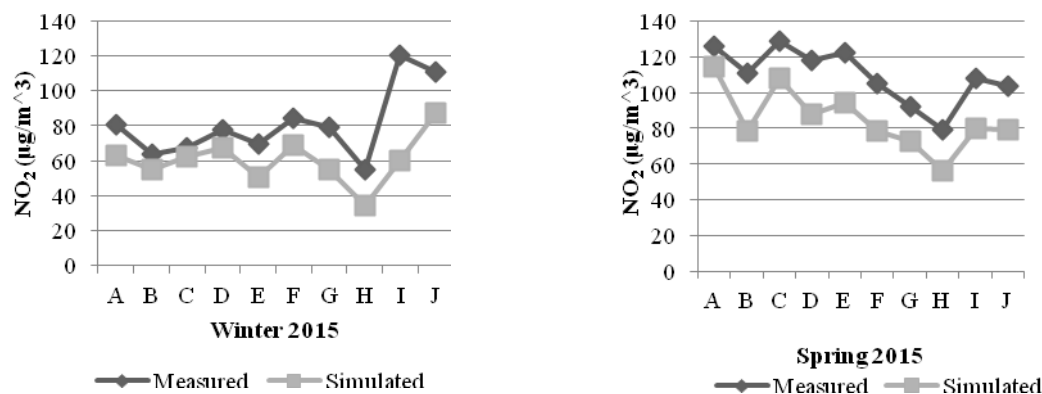
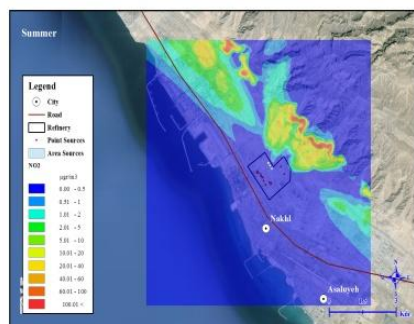


Figure 5. Comparison among measured and simulated 1-hr averages of ambient NO₂ concentrations for 10 receptors

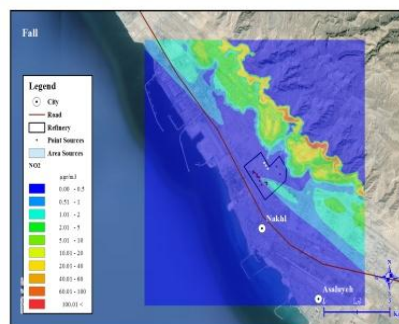
Table 4. Maximum simulated amounts of ambient NO₂ concentrations based on the average periods in a 10×10 km² domain

Time Scale	Max. Concentration (µg/m ³)	hour	day	month	X (m)	Y (m)	Z (m)
1-hr	10544.1	19	21	11	1650	2050	1.5
Annual	217.4	-	-	-	2550	1050	1.5

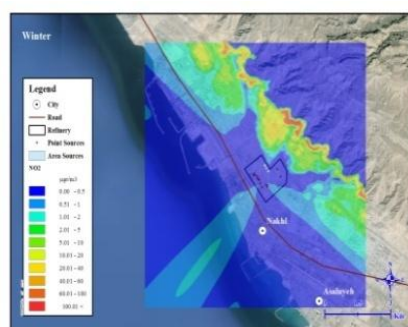
The NO₂ distribution maps based on annual simulated results among the four seasons are presented in *Figure 6*. In this figure, the areas with intensive color were more affected by NO₂ emissions and defined as unhealthy zones. As shown in the figure, the unhealthy zones were located in the right part of the simulating domain.



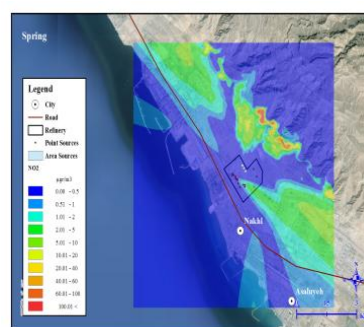
a) Summer 2014



b) Autumn 2014



c) Winter 2015



d) Spring 2015

Figure 6. NO₂ distribution maps for the simulated results in the study area

1-hr maximum concentrations of the simulated and measured values were used for the statistical analysis for evaluating the model performance. The values of correlation coefficients (COR) for NO₂ concentrations were 0.89 in summer 2014, about 0.87 in autumn 2014, about 0.67 in winter 2015 and about 0.73 in spring 2015. The results are shown in *Table 5*. As a result, there were acceptable ranges for statistical parameters and the model simulation indicated that the simulated concentrations were underestimated in compare to the measured ones.

Table 5. The results of statistical analysis

Season	COR (0,1)	NMB ≤%15	NME ≤%30	IOA (0,1)	FB <0.3
Summer (2014)	0.89	-16	16	0.86	-0.017
Autumn (2014)	0.87	-38	28	0.70	-0.047
Winter (2015)	0.67	-21	25	0.61	-0.023
Spring (2015)	0.73	-22	22	0.63	-0.025

Since there are some limitations for applying AERMOD modeling system, the simulation results may not be quite accurate. The limitations are such as: lack of module for considering NO₂ deposition reactions; high sensitivity of AERMOD to various time scales (Zou et al., 2010; Bhardwaj, 2005); lack of considering the dynamic emission rates (U.S. EPA, 2009); lack of considering the substantial time intervals for wind velocities less than 1 m/s; errors due to monitoring devices (Drew et al., 2007); complexity in topographical structure of the study area (El-Fadel et al., 2009), coastal evaporation, and the geographical effects (Yao et al., 2011).

A comparison made among the measured and predicted ambient NO₂ concentrations. The results indicated that the predicted concentrations were underestimated by a factor of 20 % in comparison to the measured ones which indicated the contribution of other sources including mobile sources and the neighboring gas refineries and other industries.

Conclusions

In the present study, ambient NO₂ concentrations in a gas refinery located in Asaluyeh was measured in 10 receptors in and around the refinery and the 1-hr and annual ambient NO₂ concentrations and dispersion patterns were simulated by AERMOD model in 10×10 km² domain. Then the unhealthy zones were determined in the study area using contour plots of the seasonal NO₂ distribution patterns. It was found that the hourly observed and predicted concentrations of NO₂ were lower than the U.S. EPA clean air standard except in receptor B in autumn 2014. However, there was no health risk due to NO₂ emissions for short time (1-hr) exposure in the gas refinery. Furthermore, a comparison made among the measured and the simulated NO₂ concentrations in 10 receptors. The results indicated the performance of the modeling was quite satisfactory with little disagreement in the values of predicted results. The discrepancy between the average simulated and measured concentrations varied from 2% to 56%; however the average predicted concentrations were underestimated by a factor of 20 % in comparison to the measured ones.

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