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ORIGINAL PAPER

Predicting ambient concentrations of NO_2 in a gas refinery located in South Pars Gas Complex

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Abstract In this study, AERMOD dispersion model has been applied for predicting the values of ambient concentrations of NO_2 emissions due to the stacks of fourth gas refinery located in South Pars Gas Complex in Asaluyeh, Iran. First, the values of NO_2 emissions from the stacks and the amounts of ambient concentrations of NO_2 in nine monitoring stations have been measured in four seasons in 2013. Then, dispersion of NO_2 emissions has been predicted by using AERMOD model in the region with the domain area of $10 \times 10\ km^2$, in average times of 1 h. Finally, the simulated and observed values of ambient NO_2 concentrations in the nine receptors have been compared. Comparison of 1-h concentrations of the observed and predicted results with the international ambient standard levels shows that NO_2 concentrations are higher than the standard value. The results show that AERMOD model can be used effectively for predicting the amounts of pollutants' concentrations in the study area.

Keywords AERMOD dispersion model · Air quality · Gas refinery · NO_2 · Statistical analysis

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Introduction

The problem of air pollution is inevitable in many countries around the world due to the increased use of energy, population growth, rapid development and industrialization. Moreover, this growth can have enormous economic and social costs in the absence of appropriate environmental policies (Arya 1999). Ambient air quality has deteriorated to such an extent that it can have a significant impact on human health and welfare, as the World Health Organization (WHO) has stated that about 2.7 million people die due to the health effects of air pollution every year (WHO 2014). The minimum concentrations of gases in the atmosphere could be important in specifying the health status of communities (Shooter et al. 1993). Therefore, air quality management strategy is essential in order to minimize the acute effects of air pollutants. Specifying the type of emissions from various sources and evaluating their effects are important for proper management of air quality (Bhanarkar et al. 2005). The amounts of NO_x exhaust gas from the flue of a gas refinery generally rise with increasing ignition temperature that its main component is NO, oxidized to NO_2 in the ambient temperature and in the presence of oxygen. NO_2 is a toxic gas for inhalation and causes eyes, nose and throat irritation in human and is an air pollutant with a lifetime of 1–3 days in mesoscale (Perkins 1974). Direct effects of oxides of nitrogen can be considered as the production of photochemical ozone components and smog. In addition to the health effects, ozone is entered into a series of chain reactions in the presence of hydrocarbons and OH radicals that results in a new pollutant called proxy acetyl nitrate (PAN) (Akdemir et al. 2013). The US Environmental Protection Agency (2010) has determined that the average amounts of hourly ambient concentrations of NO_2 should

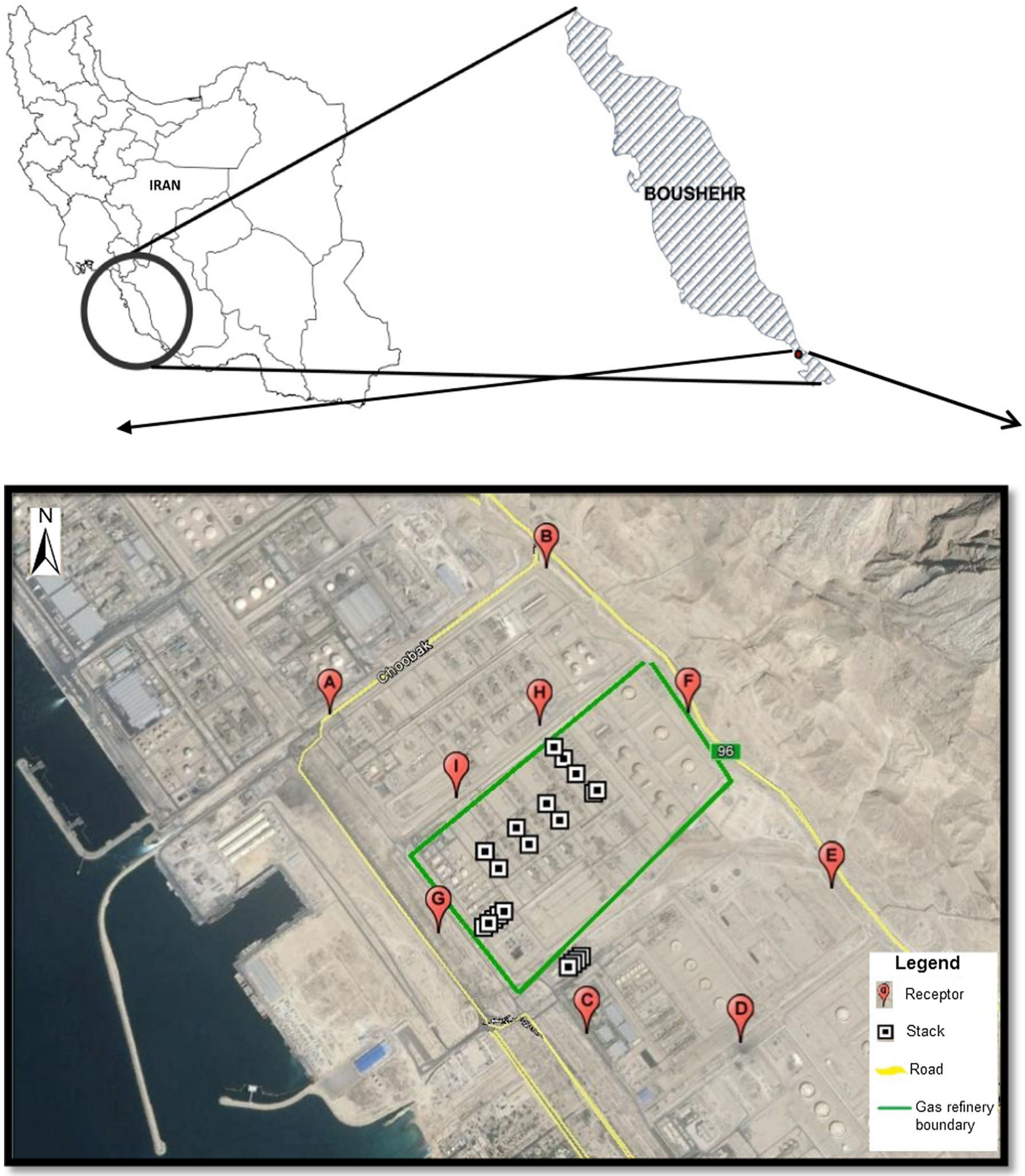


Fig. 1 Study area location and nine receptors of the fourth gas refinery

not exceed 200 μg/m^3. In order to meet the legislative goals and to minimize the negative effects of pollutants, it is important to examine the distribution of atmospheric pollutants. When a pollutant is emitted from a source, it is transported into the air by the wind. Accordingly, the maximum pollutant concentration is in emission source, and minimum concentration is downwind due to the mixture of pollutant gas with air. In general, the dispersion of pollutants is affected by many parameters including atmospheric stability, roughness, obstacles, orography, wind speed and direction and maximum mixing height (Seangkiatiyuth et al. 2011). Since the continuous monitoring and control of air pollutants is not possible from the two aspects of time and space, air quality modeling is necessary to estimate pollutants' concentrations in the interested areas. Sabah Abdul-Wahab and colleagues have

Table 1 Date of sampling and the number of stacks

Number of stacks	Month of sampling	Date of sampling
16	Spring 2013 (May)	05.05.2013
		12.05.2013
		17.05.2013
16	Summer 2013 (July)	03.07.2013
		10.07.2013
		19.07.2013
16	Fall 2013 (Oct)	07.10.2013
		13.10.2013
		20.10.2013
22	Winter 2013 (Dec)	08.12.2013
		16.12.2013
		22.12.2013

done a study to examine the patterns of SO_2 dispersion from Al-Fahal refinery by CALPUFF model in 2010. The main objective of this study was to make a comparison of the results generated by CALPUFF model with a previous study which was conducted for the same area using Industrial Source Complex Short Term (ISCST) model (Abdul-Wahab et al. 2011). Kanyanee Seangkiatiyuth and colleagues measured the emitted NO_2 concentrations due to four cement plants in Thailand for a period of seven consecutive days in 12 receptors in 2012. Then, they stimulated the NO_2 emissions from the cement plants in the dry and wet seasons by using AERMOD model and compared the measured results with the simulated results in the receptors (Seangkiatiyuth et al. 2011).

Previous studies show that comprehensive and scheduled researches have not yet been performed on modeling of air pollutants' emissions from stacks using AERMOD dispersion model for a gas refinery. The main goal of this study is to evaluate the capability of AERMOD model to simulate the ambient concentrations of NO_2 in the special topographical and climatological conditions of the study area. First, the amounts of NO_2 exhausted from the stacks and the ambient concentrations of NO_2 due to the emitted gases from stacks of fourth refinery located in South Pars Gas Complex in Asaluyeh have been monitored in nine receptors during four seasons (spring, summer, fall, winter) in 2013. Then the ambient concentration levels of NO_2 have been simulated for the receptors, using AERMOD dispersion model. Finally, the comparison of model prediction results and the monitored concentrations have been

Table 2 Stacks of NO_2 emissions for 2013 (g/s)

Stacks name	Spring	Summer	Fall	Winter
SP6-2-1104	1.7930E+02	4.2504E+01	5.8440E+01	2.3802E+02
SP6-2-2104	1.6025E+02	5.0571E+01	5.8986E+01	2.4648E+02
SP7-2-3104	2.5069E+02	4.5446E+01	5.1162E+01	1.8615E+02
SP7-2-4104	2.7135E+02	4.8014E+01	5.8440E+01	1.2015E+02
SP8-2-5104	3.7093E+02	3.5934E+01	4.5637E+01	3.5218E+02
SP8-2-6104	2.0433E+02	4.1491E+01	4.4774E+01	1.0136E+01
SP7-2-3107	0.0000E+00	0.0000E+00	3.0682E+02	5.5846E+01
SP7-2-4107	2.0589E+01	0.0000E+00	2.2504E+02	2.0260E+02
SP8-2-5107	2.8645E+01	0.0000E+00	0.0000E+00	2.0284E+02
SP8-2-6107	3.2990E+01	9.4391E+01	2.4071E+02	4.3273E+02
121-U-103 A	7.5191E+01	2.3802E+02	2.2962E+02	2.3610E+02
121-U-103 D	2.6587E+01	0.0000E+00	0.0000E+00	2.7368E+02
121-U-103 E	8.5049E+01	1.2630E+02	5.1283E+02	2.4602E+02
120-16-GTG1	0.0000E+00	8.2215E+02	0.0000E+00	5.7527E+02
120-17-GTG2	0.0000E+00	2.3828E+04	0.0000E+00	1.0912E+03
120-17-GTG3	0.0000E+00	0.0000E+00	0.0000E+00	3.2853E+03
1551-GT-101	8.4588E+01	3.8607E+02	1.1814E+01	1.2944E+03
3551-GT-101	0.0000E+00	3.6901E+02	1.9317E+03	1.7105E+03
4551-GT-101	8.2373E+01	7.8400E+02	0.0000E+00	1.7788E+03
1106-GT-101	0.0000E+00	3.9249E+02	4.2424E+02	1.2534E+03
2106-GT-101	0.0000E+00	4.0392E+02	3.3965E+03	1.6817E+03
3106-GT-101	3.9090E+01	0.0000E+00	8.0044E+02	2.1294E+03
4106-GT-101	8.5707E+01	0.0000E+00	0.0000E+00	0.0000E+00

Table 3 Description of Asaluyeh meteorological station

Meteorological station	Coordinates (UTM)		Elevation from sea level (m)	Distance from the reference point (km)	Meteorological parameters
	Latitude	Longitude			
Asaluyeh	671610.52E	3030046.94N	7	24	Surface air data: P, RH, T 10 m: WD, WS, CH, CV

RH (%), relative humidity; *T* (°C), dry bulb temperature; P (mbar), pressure; WS (m/s), wind speed; WD, wind direction (degree from the North); CH (m), ceiling height; CV (tenths), cloud cover; DWPT (°C), dew point temperature; H (m), height above sea level

done through statistical analysis, and considering the ambient standard levels, the healthy and unhealthy regions have been determined.

Materials and methods

Study area

Asaluyeh is one of the cities of Bushehr Province located in south of Iran which is considered as a huge industrial area. The fourth refinery of south pars gas is located in South Pars in the southeastern of Bushehr Province in Asaluyeh. The range of its longitude is 52°30′–52°55′ and the latitude range is 27°20′–27°37′. It is limited to Shirino village at the westernmost point, from the east to Chahmobarak village, from the north to the foothills of the Zagros Mountains and from the south to the Persian Gulf (SPGC 2012). The location of the study area is shown in Fig. 1.

The fourth gas refinery has been designed in three phases with harvesting and processing total capacity of 110 million cubic meters (about 36.7 million cubic meters for each phase). This gas refinery includes 33 stacks. The main sources of pollutants in South Pars Gas Complex are resulted from exhaust gases of the stacks which cause air pollution in the refinery area and its surrounding (SPGC 2012). The values of NO_2 emissions from the stacks of gas refinery have been measured by Testo 350 XL device in four seasons of spring, summer, fall and winter in 2013 and three times in each season (ASTM 2011b). The date of sampling and the emission data from stacks in each season have been presented in Tables 1 and 2.

Integrated 1-h ambient NO_2 concentrations were sampled at nine downwind receptor locations (Fig. 1) using a LSI-Lastem Babuc A apparatus on a height of 1.5 m above ground level (breathing height) (ASTM 2011a). In-stack and ambient concentration levels of NO_2 were field-sampled three times in each of the four seasons (spring, summer, fall, winter) in 2013.

The amounts of ambient NO_2 concentrations have been measured in nine monitoring stations in the area around the refinery. Measurements were performed based on the average 1 h. The locations of the air quality monitoring stations are presented in Fig. 1.

Model description

The American Meteorology Society–Environmental Protection Agency Regulatory Model (AERMOD) is a steady-state dispersion model used to determine the concentrations of various pollutants in urban and rural areas, smooth and rough regions, surface diffusion, considering the height of the point, area and volume sources (Huertas et al. 2012). It is suggested to simulate the dispersion of pollutants in a range of up to 50 km. In this model, it is assumed that the concentration distributions in the stable boundary layer in both vertical and horizontal directions are Gaussian functions similar to the concentration distributions in horizontal direction of the convection boundary layer (CBL) (Cimorelli et al. 2004, 2005; Perry et al. 2005). However, the concentration distributions in CBL in the vertical direction are defined with a bi-Gaussian probability density function (Willis and Deardorff 1967). This model consists of a processing core to estimate pollutant concentration from the two preprocessors: AERMET and AERMAP (Snyder et al. 1985).

AERMET is one of the preprocessors of AERMOD which is responsible for processing and providing meteorological data (US EPA 2004). In this study, the meteorological data including cloud cover, rainfall, pressure at sea level, standard pressure as surface characteristics, and wind speed and direction, ambient and dew point temperatures and relative humidity observed in Asaluyeh synoptic station (IRIMO 2014) have been provided. This is the nearest meteorological station to the study area and the emission sources. The AERMET processor has been used to calculate boundary layer parameters such as the mixing height, temperature scale, Monin–Obukhov length, surface heat flux and convective velocity scale (Freddy Kho et al. 2007).

Since the seasonal fluctuations especially variations of atmospheric parameters such as speed and direction of the wind, air temperature, boundary layer height and relative humidity in an area in different seasons will have a

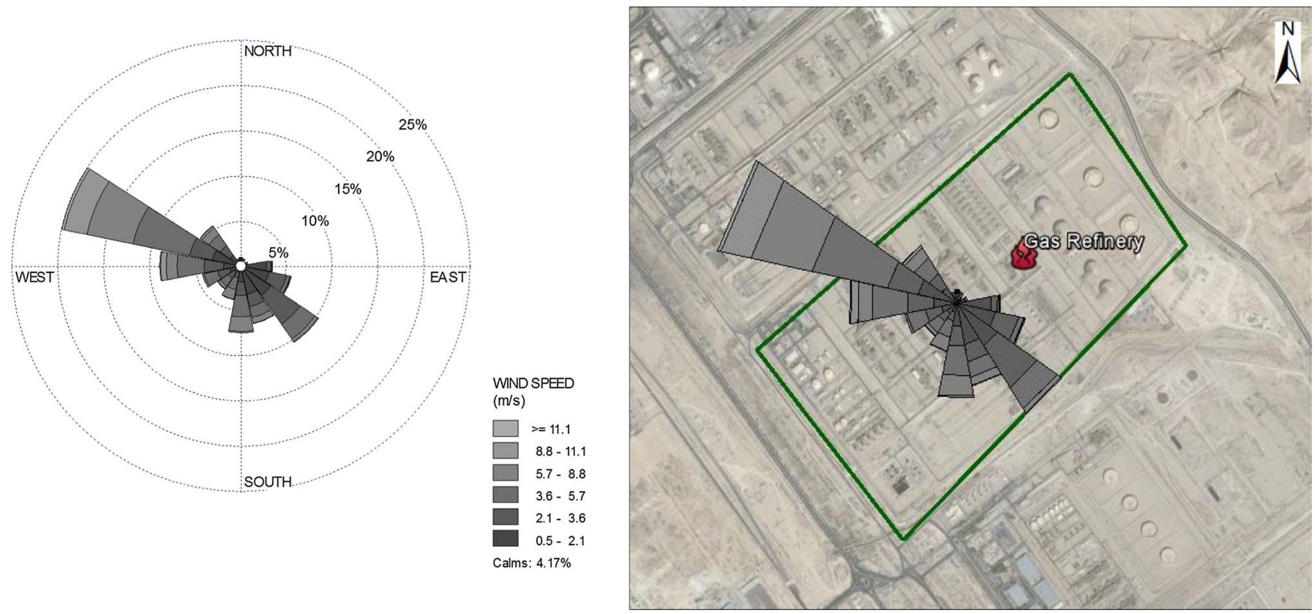


Fig. 2 Wind rose plot in the study area

Table 4 The used site characteristics based on the seasonal changes

Season	Beginning direction	Ending direction	Land use	Albedo	Bowen ratio	Roughness
Spring	0	220	Desert shrub land	0.30	3	0.30
Summer				0.28	4	0.30
Fall				0.28	6	0.30
Winter				0.45	6	0.15
Spring	220	360	Water (fresh and sea)	0.12	0.1	0.0001
Summer				0.10	0.1	0.0001
Fall				0.14	0.1	0.0001
Winter				0.20	1.5	0.0001

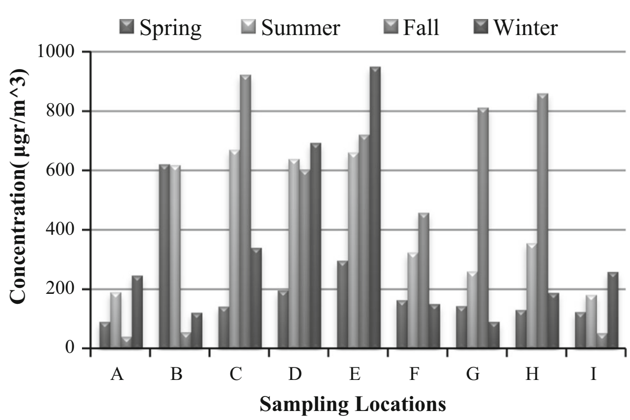
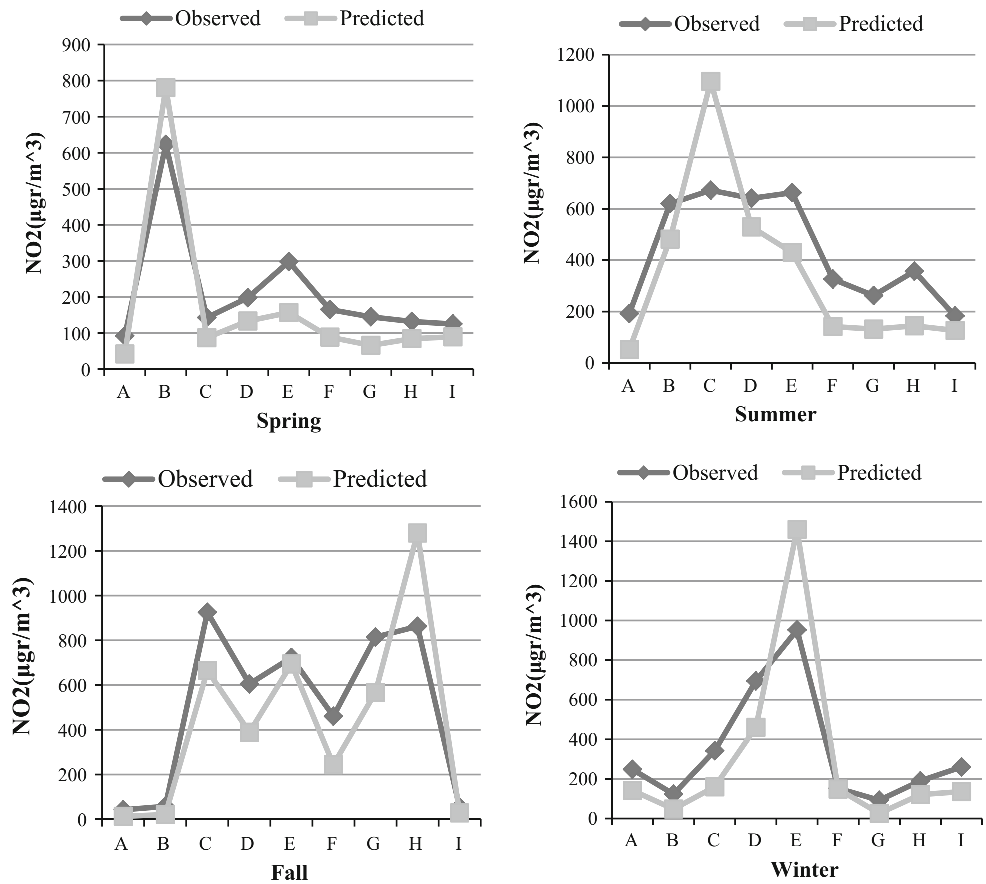


Fig. 3 Seasonal variations of NO_2 ambient concentrations in the fourth gas refinery

significant effect on the plume dispersion and air pollutants, thus simulations have been done in four seasons. All required data have been collected in order to evaluate the effective parameters in the study area. The location and specifications of Asaluyeh meteorological station are presented in Table 3. Wind rose for a period of 1 year has been presented based on meteorological data of Asaluyeh meteorological station in Fig. 2. As it is shown, the prevailing wind direction is from the northwest to the southeast in this station.

The AERMET preprocessor requires three surface characteristics of the study area as input including surface roughness, Bowen ratio and albedo. To specify the values, it is necessary to divide the study area into appropriate sectors in clockwise based on the type of land use and the surrounding vegetation. In the fourth section of user's

Fig. 4 Comparison of the observed and predicted hourly NO_2 concentrations for nine receptors



guide for the AERMOD meteorological preprocessor (AERMET), values of these parameters are presented based on user type and vegetation and theory of pain (1987). The used values in this study are presented in Table 4.

AERMAP uses gridded terrain data for the modeling area to calculate the influence of the terrain specifications and the height for each source and receptor. Digital files of unevenness elevation are the information sources of AERMAP that are provided from satellite images by some institutions (US EPA 2004). The related necessary data have been provided from Mapping Organization of Iran for this study.

Other required data for applying AERMOD model include information on the emission sources, location of the receptors and the specifications of weather files. In this study, the receptors have been introduced to the model in two gridded and discrete systems in order to cover all point sources located in study area. The network receptors have been defined in Cartesian coordinates in a domain with an area of 10×10 km^2 and grid distances of 50 m, in each direction of x and y. The location of the monitoring stations has been identified as discrete receptors in the model. The layout of all receptors with one stack in center among the gas refinery has been selected in such a way that covers all the sources and has the ability to express atmospheric phenomena in micro- and mesoscales and also the effects of topography and land use. Predicting the dispersion of the emitted NO_2 has been done for the average time of 1 h. A domain in the range of 10×10 km^2 has been considered for evaluating the health effects of this pollutant on the refinery's personnel. Thus, the modeling has been performed for the receptors at the height of 1.5 m above ground level (breathing height).The unhealthy regions have been determined as the regions with concentrations more than the amounts of ambient standard levels for NO_2.

Model validation

In this study, for the nine receptors comparison of the results of the simulation done by AERMOD model with the field

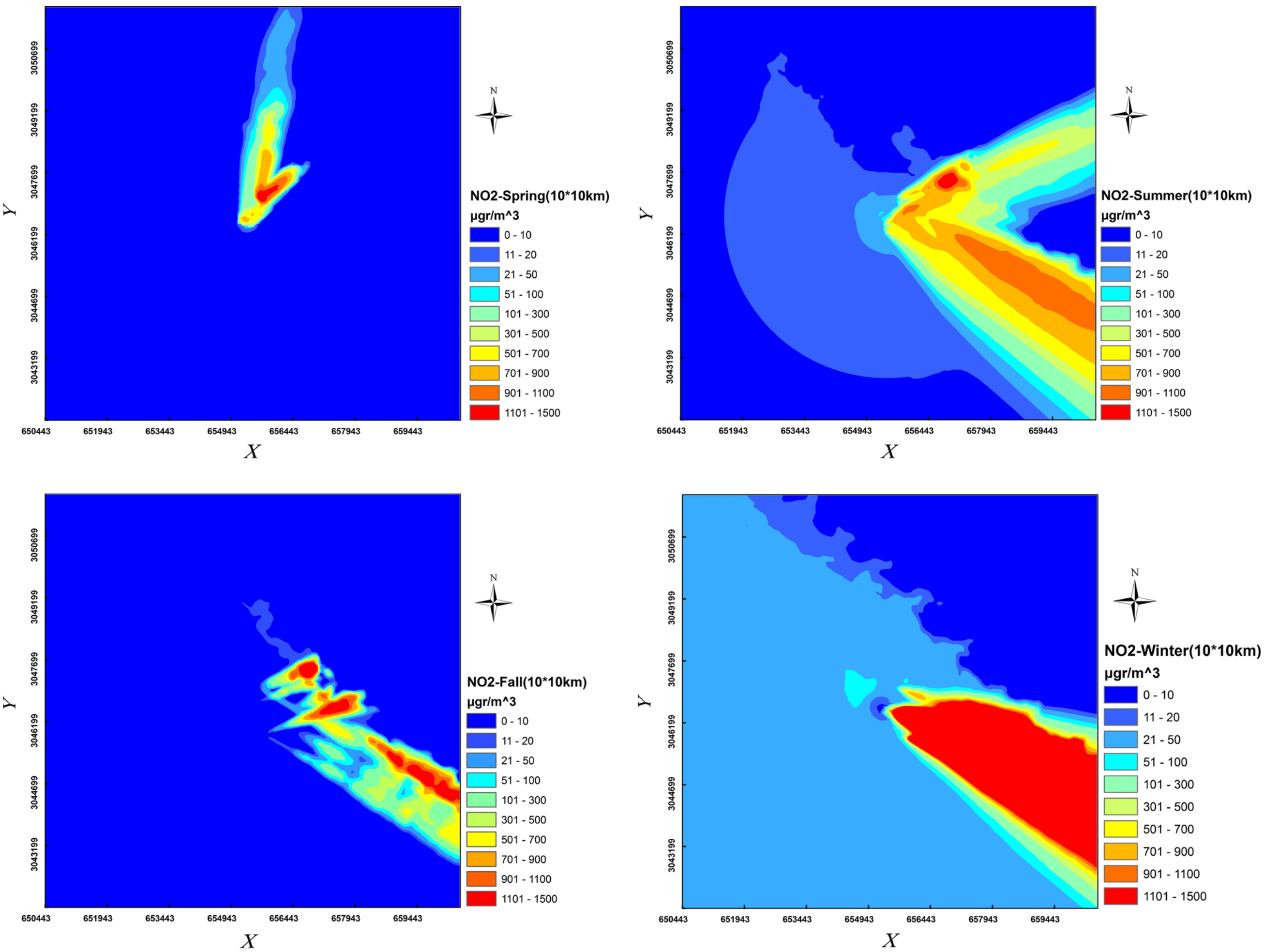


Fig. 5 Seasonal simulation results of NO_2 distribution in 2013 for 10 × 10 km^2 domain ($\mu g/m^3$). **a** Spring average NO_2 (10 × 10 km), **b** summer average NO_2 (10 × 10 km), **c** fall average NO_2 (10 × 10 km), **d** winter average NO_2 (10 × 10 km)

Table 5 The statistical analysis of the predicted and observed ambient concentrations of NO_2

Statistical parameter	Spring 2013	Summer 2013	Fall 2013	Winter 2013
CCOF	0.67	0.82	0.86	0.72
NMB %	−20	−20	−14	−12
NME %	37	42	33	45

measurements has been done by the use of statistical parameters proposed by US EPA. These parameters include:

Correlation Coefficient (CCOF): According to Eq. (1), parameter CCOF indicates the relationship between the predicted and the observed values, and the value of correlation close to 1 indicates perfect correlation between the observed and the predicted values that means a proper model performance.

$$\mathrm{CCOF} = \frac{\sum_{i=1}^{N}\left(X_i - \overline{X}\right)\left(Y_i - \overline{Y}\right)}{\left(\sum_{i=1}^{N}\left(X_i - \overline{X}\right)^2 \sum_{i=1}^{N}\left(Y_i - \overline{Y}\right)^2\right)^{1/2}} \quad (1)$$

In which, X_i: predicted values, Y_i: observed values (monitoring), $\overline{X}$: average of predicted values, $\overline{Y}$: average of observed values, N: the total measured number and its extent of the variation is (−1 to 1).

Normalized mean bias (NMB) and normalized mean error (NME): NMB and NME parameters are used to evaluate the performance of the model for modeling of pollutants, and standard values (2) defined by US EPA (2003) are NMB $\leq$ 15 % and NME $\leq$ 30 % (Eqs. 2, 3).

$$NMB = \frac{\sum_{i=1}^{N}(X_i - Y_i)}{\sum_{i=1}^{N} Y_i} \times 100 \tag{2}$$

Variation extent of NMB is (-1 to $+\infty$).

$$NME = \frac{\sum_{i=1}^{N}|X_i - Y_i|}{\sum_{i=1}^{N} Y_i} \times 100 \tag{3}$$

Variation extent of NME is (0 to $+\infty$).

Results and discussion

Seasonal variations of hourly ambient concentrations of NO_2 in the nine receptors in 2013 are shown in Fig. 3. Almost in all seasons except spring the measured and predicted ambient NO_2 concentrations were high at C, D and E receptors due to the prevailing wind direction which is almost from the northwest to the southeast. Wind rose analysis was completed using WRPLOT View and by incorporating the SURFACE and PROFILE files from the AERMET metrological preprocessor (Software 2011). The average ambient concentrations monitored in 1 h for NO_2 in the nine receptors are varied from 92 to 623 μg/m^3 in spring, from 183 to 672 μg/m^3 in summer, from 42 to 925 μg/m^3 in fall and from 92 to 952 μg/m^3 in winter. The results show the lowest ambient concentrations in spring and the highest amounts in winter. The simulation results also indicate that NO_2 concentrations are varied from 42 to 780 μg/m^3 in spring, from 52 to 1095 μg/m^3 in summer, from 12 to 1279 μg/m^3 in fall and from 25 to 1460 μg/m^3 in winter, which indicate the lowest concentrations in spring and the highest amounts in winter. The average of the hourly measured and predicted ambient concentrations of NO_2 for each season in the gas refinery is shown in Fig. 3. For the most receptors, the predicted amounts of NO_2 ambient concentrations are less than the measured one, except for the amounts of the highest predicted concentrations which are greater than the similar measured concentrations. This may be due to the contribution of the pollutants resulted from other nearby refineries or emissions from transportation and flares that have not been considered in this study.

The comparison of the predicted hourly ambient concentrations of NO_2 with the hourly US EPA air quality standards represents that the NO_2 concentrations in most parts of the study area are above the desired standards. Maximum distribution of daily NO_2 concentrations has occurred in fall and winter, respectively. Since the main source of NO_2 is power plants and heating installations, increased pollutant concentrations are due to the increased activity of these utilities and also the atmospheric stability conditions in cold seasons (Fig. 4).

In order to compare the NO_2 emissions, their image distribution in 2013 is shown in Fig. 5 for the domain of 10 × 10 km^2. Seasonal distribution results of the pollutants show that the areas with high color intensity (more than standard concentration levels) have been more affected by NO_2 and these are referred as unhealthy regions. The areas with the maximum NO_2 pollution severity are also in the middle of modeling region which are due to the presence of the pollution sources with high emissions in this region. In addition, emissions are focused in the southeast part of the study area which is significantly affected by prevailing wind direction and air turbulence (northwest to southeast).

Comparison of the predicted concentrations in ground level with the field measurement results in the nine monitoring stations in 2013 has been done through statistical analysis. Statistical parameters including CCOF, NMB and NME are given in Table 5. It should be noted that several factors create uncertainty in air quality modeling projects: the uncertainty of the model equations; the uncertainty in the model input data (e.g., meteorological data); and uncertainty in the measurement and monitoring of model input data, etc. (Dresser and Huizer 2011). In this study, contribution of the pollutants resulted from other industries located in the neighborhood of the study area might be a reason for disagreement between the observed and predicted values. The correlation coefficient values for NO_2 are 0.67 in spring, 0.82 in summer, 0.86 in fall and 0.72 in winter. The correlation index showed a realistic variability pattern of concentrations made by AERMOD. Considering these factors, AERMOD model has shown satisfactory and reasonable results for predicting ambient concentrations of NO_2.

Conclusion

In this study, comparison between the predicted and measured ambient concentrations of NO_2 has been presented in the fourth gas refinery in Asaluyeh for the year 2013. The evaluation data included results of the field measurements from nine monitoring stations (receptors) in the area of the gas refinery.

When the overall performance of the model has been examined, all results calculated through the statistical parameters indicate the successful modeling, and the predicted NO_2 concentrations agreed well with the measured data. In this simulation, all stacks of the forth gas refinery were considered as the only sources of NO_2 emissions, and emissions emitted from other neighboring refineries and industrial sources located beside the study area have not been considered. This is the main reason for the small difference between the predicted and the field measurements results.

For more accurate estimation of NO_2 emissions, it is recommended to compare the results of this model with the results of other models like California Puff Dispersion Model (CALPUFF). In general, the performance of AERMOD model can be considered acceptable in predicting pollutants' concentrations, and the AERMOD dispersion model can be used as an appropriate scientific tool for analyzing control strategies and making proper policies to reduce and prevent air pollution. In addition, air pollution in Asaluyeh can be reduced through careful design of stacks, the use of filters to reduce pollution at the stack burners, proper locating of industrial development in the region according to the prevailing wind direction in the region.

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