Original Research

Simulating Ambient SO$_2$ Dispersion Patterns and Assessing their Health Risk in a Gas Refinery

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Abstract

The goal of this study was to investigate the ambient concentrations and dispersion patterns of SO$_2$ originating from a gas refinery located in Asaluyeh, Iran, to determine the refinery's contribution in emitting SO$_2$ in the region and also to assess SO$_2$-associated health risks in the study area. First, SO$_2$ emissions from the stacks and ambient SO$_2$ concentrations at 10 receptors in and around the refinery were measured from summer 2014 to spring 2015 using a Testo 350XL analyzer and a portable device (LSI-Lastem Babuc A). The amounts of SO$_2$ concentrations due to flaring were also calculated using the emission factors. Then ambient concentrations and dispersion patterns of SO$_2$ in the study area at 1-hr, 24-hr, and annual mean values were simulated on a scale of 10×10 km^2, using an AERMOD model. Moreover, a non-carcinogenic risk assessment was performed using a U.S. Environmental Protection Agency procedure. The results indicated that about 64% of ambient SO$_2$ concentrations were due to this refinery and the remaining concentrations were due to contributions from neighboring sources. The values of maximum simulated ambient SO$_2$ concentrations at average periods of 1-hr, 24-hr, and annual for the scale of 10×10 km^2 were 24,588, 1,366.1, and 498 μg/m^3, respectively, which were higher than the U.S. EPA standard limits. There was also a potential health risk for short-term exposure (HQ = 1.4), but in long-term exposure an acceptable level of concentration (HQ = 0.28) was created.

Introduction

The emission of pollutants in a gas refinery depends on some factors, including refinery age, location, capacity, specifications of the feedstock, types of products, and the complexity of the operating units. Air quality management strategy is of great importance in minimizing the effects of air pollution, especially in places with multiple pollution sources [1]. In general, refineries emit more than 100 types of atmospheric pollutants. These pollutants include dust, metals, carbon monoxide, sulfur dioxide, nitrogen oxides, methane, dioxin, hydrogen fluoride, chloride, benzene, and other harmful gasses [2].

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Some dispersion models such as AERMOD, CALPUFF, ISCST, CMAQ, and TAPM have been integrated with meteorological forecast models to simulate and identify the concentrations of various pollutants and ambient air quality. CALPUFF and AERMOD dispersion models simulate the concentrations of environmental pollutants as a function of the location of pollution sources, the sustainability of the emission, meteorological conditions, and topographical specifications. They can be used as appropriate tools to determine the temporal and spatial patterns of ground level concentrations and the dispersion patterns caused by different sources.

Sulfur dioxide is mainly emitted due to the use of fossil fuels in various industries, including most gas refineries. It is considered one of the most important pollutants and has harmful effects not only on human health, but also on the environment (e.g., acid deposition), animals, plants, and crops [3-5]. Meanwhile, some studies have been conducted to spatial-temporal patterns of SO_2 concentrations in previous publications over the past decade. Some research articles that have been recently published have used CALPUFF to simulate SO_2 [6-18].

The AMS/EPA regulatory model (AERMOD) was widely used by researchers in many studies, including dispersion modeling for various pollutants such as SO_2, NOx, $PM_{2.5}$, PM_{10}, mercury, etc. [19-26]. AERMOD has also been compared to other dispersion models, including CALPUFF and ADMS [27-31], and coupled with the WRF model to analyse sensitivity to metrological conditions associated with PM dispersion calculations [32-33]. Furthermore, AERMOD was used to evaluate the carcinogenic and non-carcinogenic SO_2 health risk associated with both short-term and long-term exposure to coal-fired power plant emissions [34], to assess epidemiological effects [35,36], to evaluate the health risk due to a sudden release of benzene in the vicinity of a highly populated urban area of Ulsan megacity in Korea [37], and to calculate the hazard index (HI) for a combined mixture of chemicals in Poland [38]. Regarding the health effects of pollutants emitted from coal, some studies have been conducted in countries such as Cuba [39] and Malaysia [40], based on the procedures proposed by U.S. EPA. A comparative study regarding the health effects of emissions due to vehicles and industrial emissions was conducted in China and Pakistan. In this study, ambient CO, NO_2, and SO_2 concentrations were measured and compared with the EPA, WHO, and national clean air standard limits in China and Pakistan [41]. Respiratory and cardiovascular mortality rates attributed to exposure to ambient SO_2 were evaluated in Ahvaz, Iran [42-43]. Moreover, chronic obstructive pulmonary disease (COPD) and acute myocardial infarction (MI) in Khorramabad, Iran, were estimated [44].

A much better air quality condition could happen under the hypothesis that the average meteorological data can be utilized in near future simulation using AERMOD [45]. In addition, performance results of the AERMOD model at different time scales were also evaluated. AERMOD performs better in predicting SO_2 concentrations when point and mobile sources were applied together as model inputs rather than using point or mobile emission sources individually. However, the performance of the AERMOD model on monthly average was much better than those on time scales of 1, 3, 8, and 24 hr [46]. The performances of AERMOD, ISCST-3, and CALPUFF for point, line, and area sources of NO_x and CO emissions were compared using statistical analysis in Körfez. The results showed that AERMOD predictions for NO_x emissions were lower than those predicted by ISCST-3 and CALPUFF models. However, CO concentrations simulated by AERMOD were among the concentration levels predicted by CALPUFF and ISCST-3 [47]. Evaluation of the performance of AERMOD results for simulating NO_2 ambient concentrations at South Pars Gas Field in Iran was performed by Jafarigol et al. 2016 [48].

The implementation of air quality standards created a requirement for SO_2 dispersion modeling in order to identify the unhealthy regions, to identify field measurements that could be made to meet the standard levels, and to estimate the economic impacts of SO_2 control measures. However, more studies were carried out for modeling spatial-temporal SO_2 using AERMOD. No research has been done on simulating ambient SO_2 concentrations emitted from stacks and flares in a gas refinery.

Since the feeds of South Pars gas refineries contain about 4,000 ppm of hydrogen sulfide, the potential emissions of various sulfur compounds are high. In the present study, SO_2 emissions in a gas refinery located in Asaluyeh was measured seasonally from summer 2014 to spring 2015. Thereafter, the concentration and dispersion patterns of SO_2 were simulated by AERMOD model and the results were compared with field measurements. Seasonal SO_2 dispersion patterns were displayed by contour plots, and non-carcinogenic health risks were analysed for the unhealthy zones in the study area.

Materials and Methods

Study Area

Asaluyeh Port is a sub city of Kangan in Bushehr Province, located between 52°36′27′′E and 27°28′34′′N in the northern margin of the Persian Gulf and Kangan City, south of Iran. The second gas refinery of South Pars is situated between 52°34' to 52°36' east longitude and 27°30' to 27°31' north latitude. It shares borders with the Zagros Mountains in the north, the Persian Gulf in the south, Shirino Village in the west, and Chah Mobarak in the east. This refnery is located in the vicinity of the 1st, 3rd, and 4th gas refineries.

Field Measurements

The second gas refinery was constructed in two phases to process 50 million m³ of natural gas, 80,000 barrels

Table 1. Point sources characteristics in the gas refinery.

Sources*	UTM Coordinates		Diameter	Height	Temperature	Exhaust Gas Velocity	Emission Rate
	East X (m)	North Y (m)	(m)	(m)	(ºC)	(m/s)	gr/s
BO-A	656755	3044611	3	42.7	160.5	6.1	0.49
BO-B	656738	3044601	3	42.7	154	5.2	0.36
BO-C	656722	3044585	3	42.7	159	5.2	0.39
BO-D	656706	3044576	3	42.7	154	5.1	0.35
GTG-A	656572	3044756	4	30	550	32	0.021
GTG-B	656588	3044741	4	30	550	32	0.029
GTG-C	656602	3044722	4	30	550	32	0.030
GTG-D	656616	3044705	4	30	550	32	0.038
X-A	656884	3044345	2.1	117	592	10	428.1
X-B	656786	3044473	2.1	117	500	10	428.1
GTC-A	657066	3044501	3.15	17.3	520	30	0.071
GTC-B	657080	3044482	3.15	17.3	520	30	0.064
GTC-C	657094	3044482	3.15	17.3	520	30	0.047
F-A**	657752	3044832	0.48	142.8	800	60	38.5
F-B**	657812	3045085	0.48	142.8	800	60	38.5

* BO: boiler , GTG: gas turbine generator, X: incinerator, GTC: gas turbine compressor, F: flare
**According to the EPA's Iowa procedure, the modified diameter and height were used for flares

of daily gas condensate, and 400 tons of sulfur element daily. Concentrations of sulfur compounds in the feed gas were 4,000 ppm. The main sources of SO$_2$ emissions in the gas refinery area and its surroundings were exhaust gasses from 19 point sources, including 13 stacks and six flares. Concentrations of SO$_2$ in the combustion gases, and velocity of the flue gas from active stacks in the refinery were measured directly by a portable emission analyzer (TESTO 350 XL) in four seasons (June 2014-May 2015) and three days in each season according to ASTM D6522-11 standard [49]. Emission rates of SO$_2$ resulting from the flares were also calculated using emission factors, and the flares were converted to equivalent point source based on the EPA's Iowa procedure. Then, SO$_2$ concentrations and dispersion patterns were simulated by AERMOD model. Since the exact contribution of each flare from total
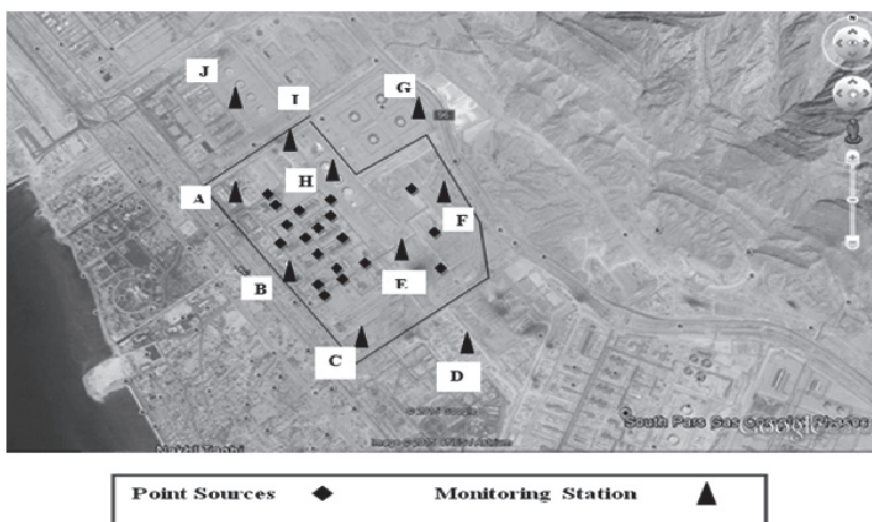

Fig. 1. Locations of 10 receptors and 19 point sources in the study area.

Table 2. Sampling dates and the coordinates of monitoring stations.

Monitoring Station	Coordinates (UTM)		Sampling Date
	X (m)	Y(m)	
A	656277	3045013	Summer 2014 (June) 22.06.2014 27.06.2014 30.06.2014 Autumn 2014 (Nov) 04.11.2014 12.11.2014 21.11.2014 Winter 2015 (Feb) 10.02.2015 15.02.2015 23.02.2015 Spring 2015 (May) 08.05.2015 14.05.2015 25.05.2015
B	656614	3044484	
C	657467	3044324	
D	657422	3044151	
E	657737	3044410	
F	657601	3044598	
G	657554	3044666	
H	657098	3045205	
I	656751	3045515	
J	656474	3045302	

flaring in the study area was not available, total flaring was considered as being two flares.

The ambient SO_2 concentrations over 24 h and 1.5 m above the ground level were measured in four seasons and three days in each season from June 2014 to May 2015 across 10 monitoring stations by applying a portable device (LSI-Lastem Babuc A) according to ASTM D3249-95 standard [50]. The monitoring stations were selected considering close distance to the emission sources, being in the direction of the prevailing wind, and being on the border of a neighborhood with other industries.

The characteristics of stacks and flares including heights, diameters, average exhaust gas velocities, emission rates, and their UTM coordinates were determined (Table 1). The locations of point sources and sampling stations are illustrated in Fig. 1, and the sampling dates and coordinates of monitoring stations in the study area are presented in Table 2. The current study considered the maximum SO_2 emissions for the continuous operation and constant emission rate with time. Total SO_2 emissions from 19 point sources was 935.09 gr/s.

Summer 2014
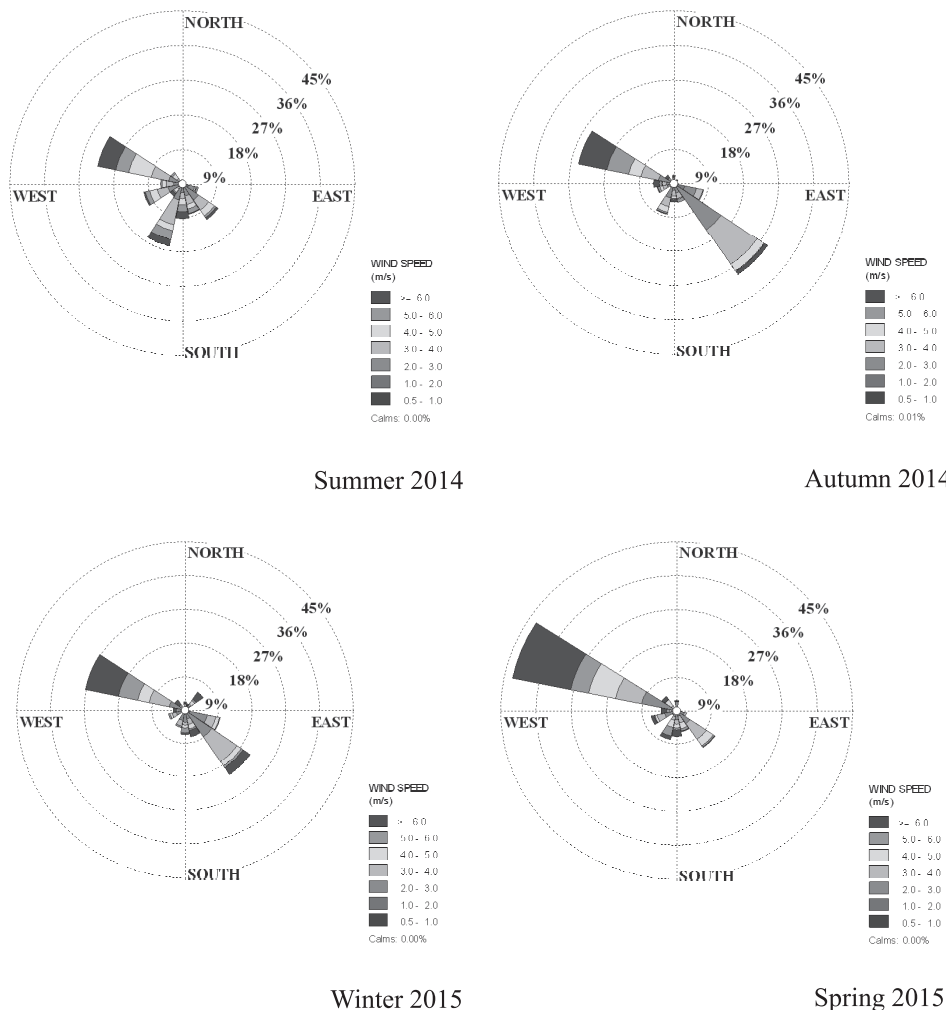

Autumn 2014

Winter 2015

Spring 2015

Fig. 2. Seasonal wind rose in the study area (summer 2014-spring 2015).

Technical Descriptions of AERMOD Model

In this study, AERMOD steady-state Gaussian model was applied to simulate SO$_2$ concentrations and dispersion maps due to the second gas refinery. It was assumed that the horizontal and vertical distributions of SO$_2$ concentrations in the sustainable boundary layer and the SO$_2$ distributions in the horizontal direction in the Gaussian convective boundary layer were similar [51].

The AERMOD model includes a main processing core to simulate the concentrations and dispersion patterns of the pollutants, and AERMET is responsible for providing and processing meteorological data [52]. In this study, the hourly meteorological data including wind speed and direction, atmospheric pressure at sea level, cloud cover, rainfall, ambient and dewpoint temperatures, and relative humidity were provided from Asaluyeh Airport Synoptic Station, which was the closest weather station to the refinery and was located 15 km southwest of the point of reference at 27°48' N and 52°62' E. Since the seasonal variations of the atmospheric parameters such as temperature, wind speed, wind direction, relative humidity, precipitation, and the height of the internal boundary layer in the study area all had a considerable effect on the pollutant concentrations and dispersion patterns, the simulation processes were carried out in four seasons and all upper and surface meteorological data in the modeling area were collected. The seasonal wind rose has been illustrated based on the available data of Asaluyeh Synoptic Station in Fig. 2. As shown, the prevailing wind direction in this station was from northwest (NW) to southeast (SE) in all seasons. The AERMET preprocessor was applied to simulate the meteorological conditions in the study area using the values of Bowen ratio, surface roughness lengths, and albedo parameters in proper sectors clockwise according to the types of the surrounding vegetation and land use. The values of the mentioned parameters for the present study are presented in Table 3. For analyzing the topographical effects (due to surface roughness, obstacles, and orography in the study area) on pollutant concentrations and dispersions, AERMAP was exe-cuted [52]. In this study, the required digital files were provided from Iran National Cartographic Center. The results of AERMET and AERMAP and all specifications of the emission sources and also UTM coordinates of the monitoring stations (receptors) were provided as input for execution of AERMOD. The receptors were introduced in the modeling domain (10 × 10 km²) with a grid spacing of 50 m, in X and Y directions. The stack of boiler A was considered as the reference point. It should be noted that the above-mentioned domain was also considered for assessing health effects due to SO$_2$ emissions on personnel working in the gas refinery, and the simulation was conducted at breathing height (1.5 m above ground level). Finally, the unhealthy zones were also determined.

Table 3. Surface characteristics based on annual climatological conditions in the study area.

Sector Number	Beginning Direction (degree)	Ending Direction (degree)	Albedo	Bowen Ratio	Roughness Lengh (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

Model Performance Evaluation

The performance of the AERMOD dispersion model has been evaluated using statistical parameters. In this study, the simulated 24-hr ambient concentrations of SO$_2$ were compared with the observed 24-hr data measured in the monitoring stations using the statistical methods proposed by the US EPA. The method included parameters such as correlation coefficient (CCOF), normalized mean bias (NMB), normalized mean error (NME), fractional bias (FB), and index of agreement (IOA), and are briefly defined below [46, 48]:

$$CCOF = \frac{\sum_{i=1}^{N}(M_i - \overline{M})(O - \overline{O})}{\left(\sum_{i=1}^{N}(M - \overline{M})^2 \sum_{i=1}^{N}(O_i - \overline{O})^2\right)^{1/2}} \quad (1)$$

$$NMB = \frac{\sum_{i=1}^{N}(M_i - O_i)}{\sum_{i=1}^{N} O_i} \times 100 \quad (2)$$

$$NME = \frac{\sum_{i=1}^{N}|M_i - O_i|}{\sum_{i=1}^{N} O_i} \times 100 \quad (3)$$

… where M_i is predicted values, O_i is measured concentrations, $\overline{M}$ is average predicted concentrations, $\overline{O}$ is average measured concentrations, and N is the total number of measurements. The variation ranges for CCOF, NMB, and NME were (-1 ~ + 1), (-1 ~ + ∞), and (0 ~ + ∞), respectively. The standard values defined by the EPA were NMB ≤ 15% and NME ≤ 30%.

$$FB = \frac{(\overline{O_1} - \overline{M_1})}{0.5(\overline{O_1} + \overline{M_1})} \quad (4)$$

The acceptable range for FB was +0.5 and -0.5.

$$IOA = 1 - \left[\frac{\sum_{1}^{N}(M - O)^2}{\sum_{1}^{n}(|M - \overline{O}| + |O - \overline{O}|)^2}\right] \quad (5)$$

The variation range for IOA was (0,1).

Health Risk Assessment (HRA)

Hazard Quotient (HQ) is applied for quantifying risk characterization. For assessing the non-carcinogenic SO$_2$ health risk, a comparison was made between chronic

Table 4. Default exposure parameters for evaluation of non-carcinogenic health risks [54].

Inhalation Rate (IR) (m³/d)	Exposure Frequency (EF) (d/yr)	Exposure Duration (ED) (yr)	Body Weight (BW) (kg)	Average Time (AT) (yr)
20	350	30	70	30

daily intake (CDI; mg kg⁻¹ d⁻¹) and the reference dose (RfD; mg kg⁻¹ d⁻¹; Eq. 6) [53].

$$HQ = \frac{CDI}{RfD} \qquad (6)$$

In the case of HQ ≤ 1, there were no health effects. However, it is important to note that HQ > 1 indicates that there was a potential risk for adverse health impacts [54]. Exposure depended on total daily intake (TDI) [mg kg⁻¹ d⁻¹] of each pollutant ingested by the recipient according to the following equation:

$$TDI = C \times \frac{IR \times EF \times ED}{BW \times AT} \qquad (7)$$

…where C is the SO_2 concentration in the receptor (mg kg⁻¹ or mg m⁻³). Other parameters have been described in Table 4. For non-carcinogenic chemicals, TDI = CDI.

In this study, the HQs for SO_2 ambient concentrations were calculated to determine short-term (1-hour) and long-term (annual) exposures for non-carcinogenic health risks.

Results and Discussion

Fig. 3 presents the mean 24-hr ambient concentrations of SO_2 measured in 10 receptors in different seasons from June 2014 to May 2015. For almost all receptors in summer 2014, the 24-hr measured ambient SO_2 concentrations were higher than the related EPA clean air quality standard limit. For other seasons the measured ambient concentrations in the monitoring stations were lower than the related standard limit, except for D in autumn 2014 and for J in winter 2015. However, in all seasons the predicted ambient SO_2 concentrations in all receptors were lower than the related standard except for G and J monitoring stations in summer 2014, due to the direction of predominant wind from NW to SE and due to the neighboring refinery. The average ambient SO_2 concentrations monitored in 10 receptors varied from 314 to 1,413 in summer 2014, from 89.4 to 366.4 in autumn 2014, from 104 to 379.6 in winter 2015, and from 78.5 to 257.9 in spring 2015. As a result, the highest measured ambient concentration was observed in summer and the lowest amount was observed in spring for a 24-hr average period. The predicted results also revealed that the variations of SO_2 concentrations were from 189 to 1,785 in summer 2014, from 61.8 to 187.8 in autumn
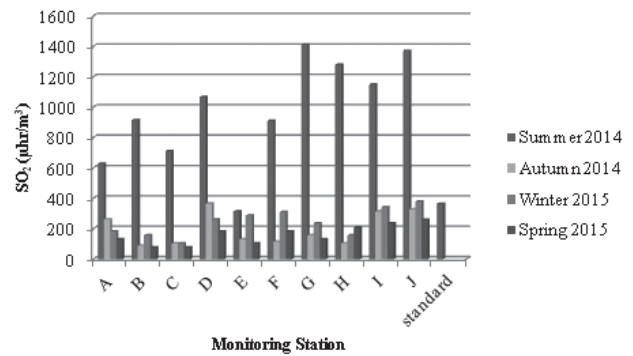

Fig. 3. Seasonal averages for 24-hr ambient SO_2 concentrations in 10 receptors.

2014, from 84.9 to 354.8 in winter 2015, and from 61.8 to 289.5 in spring 2015, which demonstrated the highest concentration in the summer and the lowest amount in the spring. The 24-hr average simulating results and observed data for ambient SO_2 concentrations in four seasons in the study area are illustrated in Fig. 4.

A comparison of the results showed that the simulated concentrations of SO_2 were less than the observed values in most of the receptors. The differences between the simulated and measured ambient SO_2 concentrations in G, D, and J monitoring stations (receptors) were higher than those in other receptors. These receptors were located in the boundary of the study area with other refineries, although some uncertainties regarding meteorological data or measurement methods could be reasons for the differences between the simulated and observed values. The simulated ambient SO_2 concentrations were compared with the related EPA clean air quality standards. The results indicated that the ambient SO_2 concentration levels in most parts of the modeling area were less than the standard levels (365) [55]. But according to the EPA
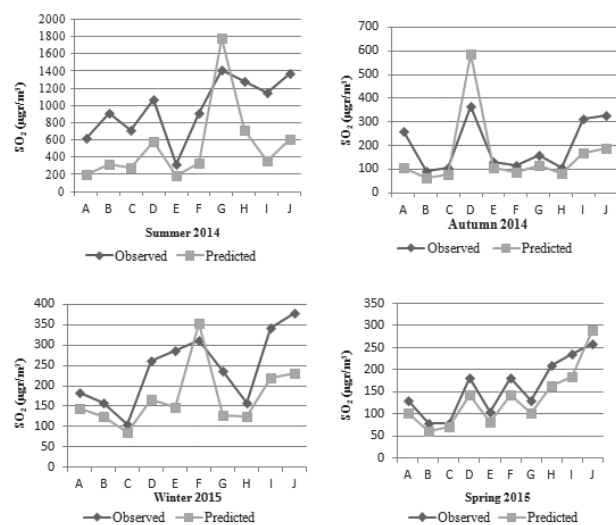

Fig. 4. Seasonal variations of observed and simulated mean 24-hr SO_2 concentrations for 10 receptors.

Table 5. Statistical analysis results.

Season	CCOF (0.1)	NMB ≤ %15	NME ≤ %30	IOA (0.1)	FB -0.5 <FB <0.5
Summer 2014	0.87	-23	8	0.57	-0.14
Autumn 2014	0.77	-18	8	0.78	-0.11
Winter 2015	0.93	-16	7	0.63	-0.11
Spring 2015	0.83	-22	4	0.87	-0.05

clean air quality standard, the number of exceedances per year is also a key index and this value is not to be exceeded more than once per year. As a result, ambient SO_2 concentrations in the modeling area were higher than the standard limits. The low height stacks of sulfur recovery units (about 26 m) located in the adjacent refinery with a considerable SO_2 emission (about 175 gr/s) had significant effects on the ambient SO_2 concentrations in the region. It also showed the contribution of emissions due to other neighboring industries. Since the main sources of SO_2 were the incinerator stacks of sulfur recovery units in this refinery, due to inadequate tail gas treatment unit (TGT), ambient SO_2 concentrations were much higher than the standard levels in several receptors. However, as shown in Fig. 4, variations of the distribution patterns of simulated SO_2 concentrations made by AERMOD were consistent with those of observed values for all receptors.

The values and locations (E, N) of maximum simulated ambient SO_2 concentrations at average periods of 1-hr and 24-hr for the scale of 10×10 km² were 24,588 µg/m³ and 1,366.1 µg/m³, respectively, at (657805, 3046961) in winter 2015. Furthermore, the annual
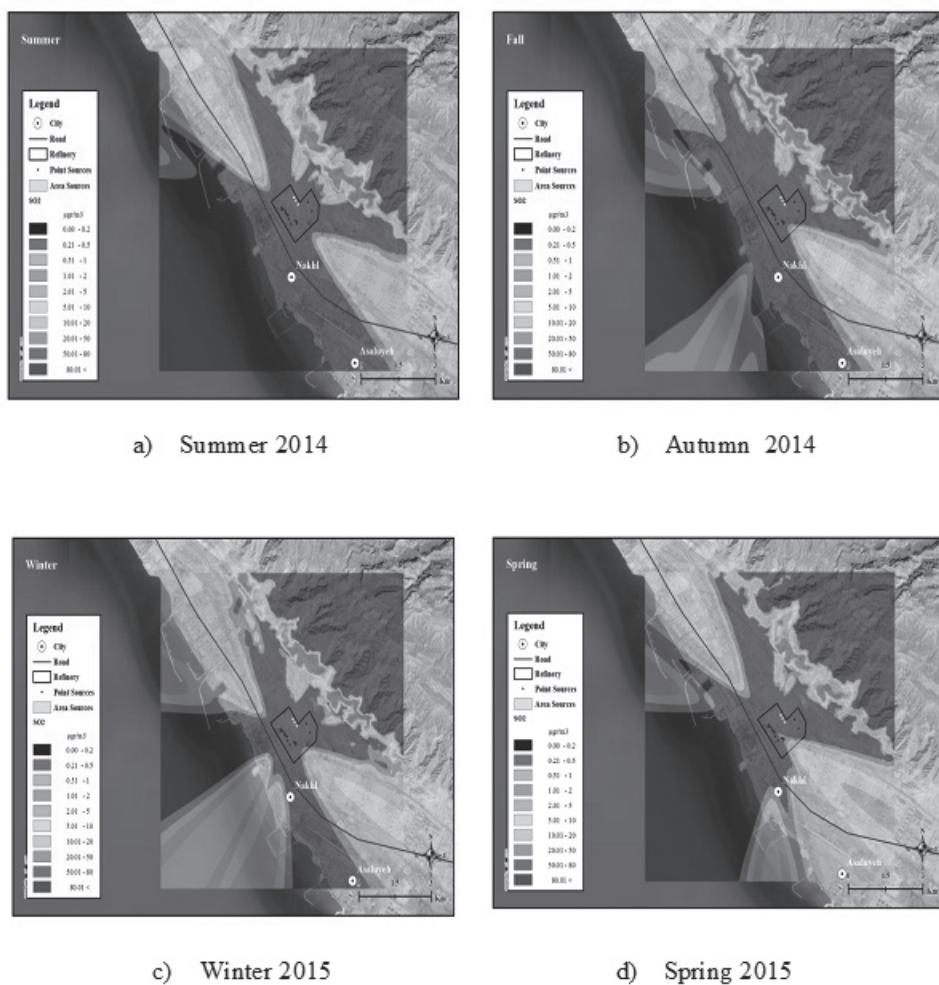

a) Summer 2014

b) Autumn 2014

c) Winter 2015

d) Spring 2015

Fig. 5. Simulated seasonal averages of SO_2 concentrations and distribution plots in June 2014-May 2015 in the study domain with a grid size of 50×50 m.

maximum simulated ambient SO_2 concentration was 498 μg/m³ at (658855, 3045561).

The ambient SO_2 concentrations and the seasonal distribution contour plots in four seasons are shown in Fig. 5. The concentration contour maps indicated that the areas with intensive color were more affected by SO_2 emissions, which were introduced as unhealthy areas. The areas with maximum SO_2 concentrations were observed in the right part of the modeling domain and were mainly affected by the high SO_2 emissions from incinerators and flares. The results revealed that 64% of the ambient SO_2 concentrations were due to point sources located inside the gas refinery, and the rest of the SO_2 concentrations indicated the contribution of mobile sources and other sources situated in the neighborhood of the gas refinery.

The 24-hr observed ambient SO_2 concentrations were compared with the simulated ones in 10 monitoring stations using statistical analysis (Table 5). Various factors may cause uncertainty in the simulating results: uncertainty of the observed data collected in monitoring stations, uncertainty in the meteorological data, and uncertainty in some model equations, etc. [56]. In this study, the contribution of SO_2 emissions due to other sources located in the neighborhood of the gas refinery, complex topography, and the geographical situation of the monitoring station might be the main reasons for the discrepancy between simulated and field measurement data. The values of correlation coefficients for ambient SO_2 measured and simulated concentrations were 0.87 in summer 2014, about 0.77 in autumn 2014, about 0.93 in winter 2015, and about 0.83 in spring 2015. The comparison indicated acceptable variations of concentrations created by the AERMOD model. Therefore, the AERMOD model can be used for simulating ambient SO_2 concentrations and dispersion maps with satisfactory accuracy. The HQs of SO_2 were also calculated to assess the short-term (1-hr) and long-term (annual) non-carcinogenic health risks. According to the HQ found for SO_2, a little potential for adverse health effects existed during short-term exposure to SO_2 as the HQ was more than one (HQ = 1.4), while long-term exposure indicated an acceptable level of SO_2 concentration with HQ less than one (HQ = 0.28). It should be noted that meteorological conditions had a considerable role for reducing the SO_2 health risk.

Conclusions

In the present study, for the second gas refinery located in South Pars Gas Complex, the ambient SO_2 concentrations and dispersion maps were simulated using the AERMOD model from June 2014 to May 2015. The field measurements data included SO_2 ambient concentrations in 10 monitoring stations in and around the study area. Moreover, the performance model was evaluated using statistical methods. The results obtained from the statistical analysis indicated that variations of

the distribution patterns for simulated SO_2 concentrations were consistent with the observed values. In this research, all flares and stacks of the second gas refinery were examined as the only SO_2 emission sources, and pollutants emitted from other industrial sources located in the neighborhood of the modeling area were not considered. The 24-hr average observed concentrations in the gas refinery were 1,023.1 in summer 2014, 222.9, 263.5 in winter 2015, and 159.2 in spring 2015. Simulation results indicated that the values of maximum ambient SO_2 concentrations at average periods of 1-hr, 24-hr, and annual for the scale of 10×10 km² were 24,588, 1,366.1, and 498 μg/m³, respectivel, which were higher than the EPA standard limits. A comparison made between the measured and simulated ambient SO_2 concentrations indicated that about 64% of the ambient SO_2 concentrations were due to the point sources located in this gas refinery and the rest of the SO_2 concentrations were due to the emissions produced from mobile and neighboring sources (other refineries). In general, the AERMOD model can be used for predicting ambient SO_2 concentrations and dispersion maps and for investigating control solutions with satisfactory accuracy.

Air pollution in the South Pars Gas Complex can be controlled through proper design of flares and stacks, installation of tail gas treatment units, using filtering at the stack burners for reducing pollution, and proper site location for flares and stacks considering prevailing wind direction in the industrial zone.

According to the HRA results, different health risks were recognized for long- and short-term exposure to SO_2 emissions. For non-carcinogenic human health risk, a potential for adverse health consequences existed during short-term exposure to SO_2 (HQ = 1.4), whereas long-term exposure indicated an acceptable ambient SO_2 concentration level (HQ = 0.28).

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