Dispersion of NO\textsubscript{2} pollutant in a gas refinery with AERMOD model: A case study in the Middle East

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\textbf{ABSTRACT}

\textbf{Introduction:} Air pollution from industrial sources is a growing problem increasing the amount of air pollution by emitting various gaseous pollutants such as Nitrogen Oxides (NO\textsubscript{x}). This study analyzed Nitrogen dioxide (NO\textsubscript{2}) emissions using American Meteorological Society/Environmental Protection Agency Regulatory Model (AERMOD) from the stacks and flares of a gas refinery in the Middle East.

\textbf{Materials and methods:} The NO\textsubscript{2} emissions were measured from the stacks and flare of the refinery (231 samples). The distribution of emissions was investigated over a statistical period of 1 year for an average time of 1 h using the AERMOD dispersion model in an area of 25×25 km\textsuperscript{2}. The predicted concentrations were compared with national and international standards and are plotted for the desired zones.

\textbf{Results:} Comparison of simulation results with national and international clean air standards showed that NO\textsubscript{2} emission modeled in all periods of 4 seasons is higher than the standard. Examination of NO\textsubscript{2} emission and distribution maps also showed that the maximum concentration of NO\textsubscript{2} pollutants occurred in the central parts and the area close to the refinery. The highest maximum concentration of 1-h NO\textsubscript{2} was 3744.3716 μg/m\textsuperscript{3} in summer in the west and south of the refinery. Validation results also showed a high correlation between the predicted and actual results.

\textbf{Conclusion:} The power of resources in emission and distribution, topographic conditions, and meteorological characteristics of the region are three important and influential factors in the distribution of NO\textsubscript{2} pollutants. So pollution reduction strategies are needed due to the different types of use, surrounding residential areas, personnel, and people involved in the gas refining company.

\textbf{Keywords:}
Air pollution modeling; AERMOD; Nitrogen dioxide (NO\textsubscript{2})

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\textbf{Introduction}

Emissions of greenhouse gases from industrial facilities are one of the most significant environmental problems in many countries of the world such as refineries. These industrial facilities emit many Criteria pollutants, including nitric oxide, carbon monoxide, and sulfur dioxide [1, 2]. Combustion of fossil fuels releases Sulfur dioxide (SO\textsubscript{2}), Nitrogen Oxides (NO\textsubscript{x}), Carbon monoxide (CO), and Particulate Matter (PMs) into the environment [3-5], and the emission
of pollutants periodically or continuously from point and non-point sources harm air quality [6]. NO\textsubscript{x} gases are usually released through fuel combustion sources in the form of Nitric Oxide (NO) and smaller quantities such as NO\textsubscript{2} gas [7]. The amount of NO\textsubscript{x} emissions from the stacks of a gas refinery generally increases with raising in combustion temperature, the main component of which is NO, and it oxidizes to NO\textsubscript{2} at ambient temperature in the presence of oxygen [8]. Nitrogen compounds are chemically different, but the most important form from a health perspective is NO\textsubscript{2}, which has different effects on human health [9]. Nitrogen dioxide (NO\textsubscript{2}) is a gas with oxidizing properties that can reduce air quality in many urban and industrial areas [10]. The effects of this gas include hospitalization, upper and lower respiratory diseases, bronchitis, chronic cough, and increased neonatal mortality. In addition to its health effects on humans, NO\textsubscript{2} can absorb sunlight and reduce vision [9].

Air quality modeling is one of the methods that can provide air pollution information with a relatively simple approach [11]. It is possible to understand the distribution of pollutants emitted from airborne emission sources using numerical simulation [12]. Their applications include assessing air quality, determining the share of emissions from various sources, obtaining permits to release gases and dust in the air, and identifying critical measures to reduce emissions [13]. Also, the dispersion model results can be used to manage emission sources to reduce their emission [14]. AERMOD is one of the most widely used software for modeling the dispersion of air pollutants [15]. AERMOD is a Gaussian column model that can evaluate the concentration of pollutants from different sources and predict the dispersion of pollutants for short distances (up to 50 km) [11]. AERMOD consists of three preprocessors. AERMOD consists of three preprocessors including, a Meteorological preprocessor (AERMET), a geological preprocessor (AERMAP), and the main processor (AERMOD) [6]. AERMAP is a geological preprocessor for topographic analysis of the area, and AERMET is a meteorological preprocessor that requires three surface features of the area, including the albedo, Bowen ratio, and surface roughness length [11]. Detailed studies have demonstrated modeling of NO\textsubscript{2} emissions using AERMOD and the health effects of exposure to this pollutant [16-19].

Due to establishing a considerable volume of oil and gas industries in the Middle East, the need for a comprehensive environmental study is more than ever. Hence, the implementation of dispersion modeling helps the authorities to align their technical knowledge about the emission of pollutants with regulatory requirements, health standards, and environmental regulations. Previous studies show that comprehensive and planned research on NO\textsubscript{2} emission modeling using the AERMOD dispersion model for the studied refinery has not yet been conducted. Therefore, assessing NO\textsubscript{2} concentrations in ambient air with advanced AERMOD modeling using the AERMOD dispersion model for the studied refinery has not yet been conducted. Therefore, assessing NO\textsubscript{2} concentrations in ambient air with advanced AERMOD modeling systems to control and reduce air pollutants to support future health and ecosystem assessment studies in the region will be very important. In this study, after measuring NO\textsubscript{2} gas output from 16 stacks, modeling of NO\textsubscript{2} gas dispersion in 21 separate receptors and 441 regular Cartesian receptors was performed using the AERMOD 8.9.0 model, which is approved and proposed by Environmental Protection Agency (EPA). The values obtained from the implementation of the model were compared with national and international standards. This study was performed to predict NO\textsubscript{2} concentration levels in the range of 2500 km\textsuperscript{2} for 1-h averages based on the maximum NO\textsubscript{2} gas concentration in four seasons using AERMOD 8.9.0.

Materials and methods

This descriptive-analytical study was conducted in the Gas Refining company over one year, from
January 2020 to December. The Gas Refinery Company has been exploited for gas purification of gas fields with an approximate recoverable reserve. It is designed to refine part of the produced sour gas. This refinery has several rows of gas sweetening units and supplies about 25-30% of the country's gas consumption. The different stages of measuring the exhaust gases from the stacks of the gas refinery and the different stages of modeling with AERMOD 8.9.0 are described in detail in the following paragraphs.

**NO₂ gas output from fixed source stacks**

NO₂ emissions, velocity, and flow rate of exhaust gas from the refinery's fixed source (boiler, compressor, incinerator, turbine, furnace 8100) stacks were measured by Gas Analyzer TESTO 350 in four seasons (spring, summer, autumn, winter). The standard type of device operation was determined based on EPA-CTM030-41, ASTM D 6522-00 [20]. The total number of gas stacks is 25, and the total number of flares is three. Measurement days are determined based on the maximum activity of the refinery in each season, and March, June, September, and November were considered the months for measuring exhaust gas and ambient air.

**NO₂ gas at the breathing height and different distances from emission sources**

The Aeroqual S500 (Air Quality and Dust Monitoring) ambient air monitoring device was used to measure the concentration of NO₂ in ambient air [21]. To validate and determine the accuracy of the model and compare the results of the model implementation with the measured values, the ambient air monitoring device Aeroqual S500 has been used to measure the concentration of NO₂ in the ambient air. For this purpose, 21 separate receivers were considered in the vicinity of the refinery. The number of stacks in service and the number of measurements of NO₂ gas from the stacks outlet as well as the number of environmental measurements of NO₂ gas at different intervals (21 separate receivers) in each season is shown in Table 1.

| Seasons | The month of sampling | Number of stacks in service | Number of NO₂ gas measurements output from the stacks | Environmental measurements of NO₂ |
|---------|-----------------------|----------------------------|------------------------------------------------------|---------------------------------|
| winter  | March                 | 16                         | 48                                                   | -                               |
| Spring  | June                  | 12                         | 36                                                   | 21                              |
| Summer  | September             | 14                         | 42                                                   | 21                              |
| Fall    | November              | 14                         | 42                                                   | 21                              |
| Total   | -                     | -                          | 168                                                  | 63                              |
Separate and network receptors

Regular Cartesian receptor tools and discrete receptor tools were used to determine the concentration of pollutants at different distances from the refinery in AERMOD. The study area with an area of 25×25 km², 441 network receptors in Cartesian coordinates, and a specified network distance (2400 m) in each of the two directions X and Y are defined, so the NO₂ gas distribution is evident in the whole modeling range.

Meteorological and topographic information

Meteorological information of the nearest meteorological station to the gas refinery, located 20 km, was used. Meteorological information, including rainfall, temperature, pressure, wind direction, wind speed, cloud cover, and humidity of the synoptic weather station in the study area, were provided by the General Meteorological office of the country and the province. This information was processed using Excel, and the hourly data was collected and converted into an acceptable SAMSON (Scalable Active Memory Server On a Network) format and then loaded into the AERMET module. The wind rose of the region was also prepared with wind speed and direction information using WRPLOT. The output of the AERMET processor is two files, including a file containing high atmospheric meteorological information with a PFL extension and a meteorological data file related to the surface with an SFC extension. In this study, the information from the Middle East Mapping Organization has been used as input data for AERMAP preprocessor. A Digital Elevation Model (DEM) with a 10-m pixel size was prepared using ARCMAP 10.3 and processed in a dedicated topographic data processing module (AERMAP) for final execution in AERMOD. Modeling has been done for receptors located on the ground and receptors located at the height of 1.5 m above the ground (breathing height) in the area of 2500 km² around the refinery.

Validation

To validate, the predicted data and the real data were compared and evaluated using Spearman’s correlation coefficient test in the three seasons of summer, spring, and autumn.

Results and discussion

Seasonal simulation of 1-h NO₂ concentration with AERMOD

The distribution of 1-h NO₂ concentration in winter is shown in Fig. 1a, the maximum value of which is 1721.799 μg/m³, which occurred in the center and south of the refinery. Fig. 1b shows the distribution of 1-h NO₂ concentration in spring with a maximum value of 3467.449 μg/m³, which occurred in the south and southwest directions of the refinery. Fig. 1c shows the distribution of 1-h NO₂ concentration in summer with a maximum value of 3744.3716 μg/m³, which occurred in the west and south of the refinery. Fig. 1d also shows the distribution of 1-h NO₂ concentration in autumn, with a maximum value of 2740.429 μg/m³ occurring in the center and area around the refinery. In general, the highest maximum concentration of 1-h NO₂ gas occurred in summer. These studies show that AERMOD with long-term information from pollution sources can provide helpful information for emission control to improve air quality. The study in modeling the dispersion of SO₂, NO₂, and CO pollutants from gas turbine power plant emissions in Makassar utilizing AERMOD showed that the distribution of SO₂, NO₂, CO, and particulate matters in the gas has a direct relation with prevailing wind direction, which agrees with the results of the present study [22]. The maximum concentration of NO₂ in 1h is equal to 3744.4 μg/m³, which occurred in summer. According to Iran and EPA clean air standards, the maximum NO₂ emissions in 1-h intervals are 200 and 100 μg/m³, respectively [23, 24]. A comparison of the simulation results and the clean air standard of Iran and the EPA standard demonstrated that the amount of NO₂ emission in all modeled periods in 4 seasons is higher than the standard.
Fig. 1. One-h dispersion of NO$_2$ concentration over four seasons (Fig. 1a. Winter, Fig. 1b. Spring, Fig. 1c. Summer, Fig. 1d. Autumn)
Fig. 1. One-h dispersion of NO$_2$ concentration over four seasons (Fig. 1a. Winter, Fig. 1b. Spring, Fig. 1c. Summer. Fig. 1d. Autumn)
Concentration changes of $\text{NO}_2$ from the refinery to the last residential area upstream and downstream

Fig. 2a shows the changes in the 1-h $\text{NO}_2$ concentration from the refinery (zero point of the horizontal axis) to the village (20 km upstream of the refinery). According to this diagram, the maximum and minimum concentrations of $\text{NO}_2$ for 1-h are related to autumn and spring, respectively, and the concentration of pollutants up to a distance of 13 km from the refinery is above the standard and after this distance is below the standard. Fig. 2b also shows the trend of changes in the 1-h $\text{NO}_2$ concentration from the refinery (zero point of the horizontal axis) to the village (24 km downstream of the refinery). In this diagram, the highest and lowest 1-h $\text{NO}_2$ concentrations are related to autumn and spring, respectively, and the concentration of pollutants up to a distance of 14 km from the refinery is above the standard, and after this, the distance is below the standard.

![Plot File of 1st High 1-Hr Values for Source Group: ALL (NO$_2$)](image)

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*Fig. 2. 1-h NO$_2$ concentration changes from refinery to last upstream (a) and downstream (b) residential area and comparison with clean air standards*
Based on the presented results, the concentration decreases with increasing distance from the source. The maximum concentration of NO\textsubscript{2} occurred near the emission source (gas refinery) in the west and southwest foothills of the mountains due to the high altitude and the lack of suitable conditions for pollutants' escape. In addition, NO\textsubscript{2} concentration changes from the refinery to the village at a distance of 20 km were considered the last residential upstream area and from the refinery to the village at 24 km was the last residential downstream area. The concentration changes from the refinery to the studied villages are decreasing, but sometimes it has several increasing peaks along the route. The power of resources in emission and dispersion, topographic conditions, and meteorological characteristics of the region are three important and influential factors in the dispersion of NO\textsubscript{2} pollutants.

In a study land use plots of the domain of study in Edmonton showed that there were not many varieties in the height of the land. Because of the relatively flat topography of Edmonton, the geographic features and metropolitan structures were inadequate to go about as hindrances, reducing the effect of the direction of wind stream on NO\textsubscript{2} plume dispersion. Due to the differences in the topography and meteorological states of Yarmouth and Newfoundland, despite the two districts being beachfront locales, NO\textsubscript{2} plumes were dispersed into the land in Yarmouth, but onto the water in Newfoundland [25].

**Meteorological results**

The wind rose in the study area is shown in Fig. 3a. Wind directions include 25% from the southwest, 17% from the south, 10% from the northeast, and 8% from the northwest. Due to the highest percentage of winds, the region's wind rose blows from the southwest. The percentage and speed of winds are also shown in Fig. 3b. 44.7% of the blown winds have a speed of (0.5-2.10 m/s), 4.4% of the total-blown winds are gentle winds, and 1.5% of the blown winds have a speed of 11.10≤ m/s. In general, the average wind speed in the study area is 4.8 m/s, which is a significant factor in the distribution of NO\textsubscript{2} pollutants.
The study was conducted in 2019 under the title of AERMOD program to assess the dispersion of formaldehyde emissions from chipboard production in Belarus. The maximum hourly concentration of formaldehyde was calculated at a distance of 3 km from the emission sources. The relationship between the measured and simulated concentrations was illustrated. In a study, the change in NO\textsubscript{2} concentration was influenced by the amount of emission and then meteorological changes [26]. The uncertain sources of the results related to the lack of available meteorological information are described, which indicates the importance of entering accurate meteorological information to achieve reliable results. This issue was also considered in the present study, and 261191 1-h meteorological data, including seven parameters (temperature, pressure, humidity, precipitation, cloud cover, wind speed, and wind direction), were used [27].

**Comparison of predicted results and actual measurement results**

To validate and determine the verification of the model, the model results were compared with the values measured by the Aeroqual S500 device and were evaluated using Spearman correlation coefficient test. The correlation coefficients between actual results and results modeled in for summer, autumn, and spring were 48%, 71%, and 56%, respectively (Tables 2, 3, and 4). The Spearman correlation results illustrated a significant medium positive relationship between X and Y, $(r (19) = 0.49, p = .02)$ (Table 2.). The validation results also showed that the correlation coefficient for summer, autumn, and spring was 48%, 71%, and 56%, respectively. According to the results of previous studies, AERMOD predictions showed a high correlation with the results [19].
The Spearman correlation results showed that there is a significant large positive relationship between X and Y, \((r (19) = .713, p < 0.0001)\) (Table 3).

Table 2. Statistical analysis of predicted data and actual summer data

| Parameter                              | Value   |
|----------------------------------------|---------|
| Spearman's rank correlation coefficient (\(r_s\)) | 0.4898  |
| p                                      | 0.02    |
| Covariance                             | 18.35   |
| Sample size (n)                        | 21      |
| Statistic                              | 2.4491  |

Table 3. Statistical analysis of predicted data and actual data of the autumn season

| Parameter                              | Value   |
|----------------------------------------|---------|
| Spearman's rank correlation coefficient (\(r_s\)) | 0.713   |
| p                                      | 0.000286|
| Covariance                             | 27.0375 |
| Sample size (n)                        | 21      |
| Statistic                              | 4.4323  |

Table 4. Statistical analysis of predicted data and actual spring data

| Parameter                              | Value   |
|----------------------------------------|---------|
| Spearman's rank correlation coefficient (\(r_s\)) | 0.5669  |
| p                                      | 0.007   |
| Covariance                             | 20.875  |
| Sample size (n)                        | 21      |
| Statistic                              | 2.9997  |

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The Spearman correlation results demonstrated that there is a significant large positive relationship between X and Y, (r (19) = 0.567, p = 0.007). (Table 4).

According to Iran clean air standard, the maximum emission of NO$_2$ in 1 h intervals is 200 µg/m$^3$. By comparing the amount of NO$_2$ emitted from the refinery, it is evident that the maximum amount of NO$_2$e/h in all measured seasons is higher than the standard. The results of this study agree with a study in 2018, the title Modeling atmospheric pollutants from thermal power plants with AERMOD that can affect the health of local populations and animals and plants [28]. In the part of CO and NO$_2$ pollutants at a distance of 25 km, the amount of emissions reaches close to zero, which is consistent with the results of a study that analyzed the distribution of CO, NO$_2$, SO$_2$ gases in Masjed Soleiman Oil and Gas Exploitation unit No. 9 utilizing AERMOD at a distance of 25 km [29].

**Conclusion**

Our investigation focused on measuring and modeling NO$_2$ in one of the largest Gas Refineries in the Middle East. We modeled the NO$_2$ dispersion using AERMOD in this Gas Refinery for the first time, and considering the dispersion maps of NO$_2$, it can be concluded that the maximum concentration of NO$_2$ pollutants occurred in the central parts and the area close to the refinery. The maximum concentration of 1-h NO$_2$ was in the summer which was equal to 3744.4 µ/m$^3$. Due to the type of use and surrounding residential areas, personnel and people involved in the gas refining company need solutions to reduce NO$_2$. The results of this study will be important in developing and improving human health risk assessment studies for future exposure to air pollutants (NO$_2$).

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**Authors’ contributions**

All the authors contributed to the design, review, and revision of the study, and approved the final.

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**Competing interests**

The authors declare no conflicts of interest.

**Ethical considerations**

Ethical considerations (including plagiarism, informed consent, misconduct, data fabrication or falsification, double publication, and submission) have been completely observed by the authors.
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