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Determination of the emission rate and modeling of benzene dispersion due to surface evaporation from an oil pit

Abdolreza Karbasi[1], Shahrzad Khoramnezhadian[2], Saeed Reza Asemi Zavareh[2], Ghasem Pejman Sani[3,*]

[1] Department of Environmental Engineering, Faculty of Environment, Tehran University, Tehran, Iran
[2] Department of Environmental Engineering, Islamic Azad University, Damavand Branch, Damavand, Iran
[3] Department of Environmental Pollution, Islamic Azad University Damavand Branch, Damavand, Iran

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CORRESPONDING AUTHOR:

gpsani96@gmail.com
Tel: (+98 21) 76301225
Fax: (+98 21) 76301225

ABSTRACT:

Introduction: Air pollution is considered as one of the important challenges in oil fields and, determination of the emission levels and to identify the way of their dispersion is the first step to control and reduce the air pollutants more effective and efficient. The objective of this study was to determine the emission rate and analysis of VOCs due to surface evaporation from oil pit at one of the petroleum companies.

Materials and methods: This study was conducted in four seasons in 2017 on the Kharg island. The environmental benzene from the pit surface was measured then, dispersion method, analysis of the emissions of these pollutants was conducted using TANKS 4.0.9d and AERMOD dispersion model in an area of 10×۱۰ km^2 with a network spacing of 200.

Results: The maximum average concentration of airborne benzene at station A (0.53 ppm) and the station H (0.59 ppm) were obtained in the spring and, station M (0.72 ppm) and the station P (0.81 ppm) were obtained in the summer which are higher than the standard limit determined by DOE and EPA. The rate of emission from the oil pit was calculated as 0.0012 g / s. The motion of the pollutant plume is from the average hourly to yearly direction to the south and the results shows that the pollutant plume is moving in the direction of the wind, and because of low height of source of pollutants, the pollutant plume has remained in the oil pit area and has not moved.

Conclusion: In general, considering the estimation of predictions, the performance of the AERMOD dispersion model can be considered acceptable in predicting the concentration of benzene in the target area.

Introduction

Volatile organic compounds (VOCs) are the second most important factor in air pollution after suspended particles. These compounds are a large batch of gaseous hydrocarbons which are easily released in the atmosphere due to the high vapor pressure and their main characteristic is the high evaporation rate [1 - 3]. Among these

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compounds, benzene is a carcinogenic contaminant [4]. Therefore, exposure to benzene leads to serious consequences, including neurological diseases and cancers. The WHO guidelines allow an annual amount of atmospheric benzene at 0.5 ppm [5]. Oil industry activities are the most important source of benzene in the environment. In this industry, the release of benzene vapors from various sources, such as crude oil and petroleum products storage tanks, chemical storage tanks, chemical injection pumps and crude oil transfer pumps, leakage from the mechanical seals and flanges, open evacuation pits, emergency oil drainage ponds, oil drainage, outlet outdoor channels and industrial wastewater management etc. [1]. One of the most important issues in determining the amount of pollutants is to measure their amount at specified intervals and at certain time intervals, which should be repeated periodically. But this was not possible due to various reasons and lack of equipment, time and financial constraints and in order to eliminate constraints, simulation software such as AERMOD and TANKS are used to estimate the amount of pollutants and their emission rate in different areas and the results are compared with measurable items and analyzed [6, 7]. A case study was conducted in Dar es Salaam, Tanzania, in 2004. In this study, using the TANKS 4.0, the release rate of VOCs of organic liquid storage reservoirs of 8 different companies was determined and the release method and then the risk analysis of these pollutants in the region were determined by using US EPA standard regulatory storage tanks emission model (TANKS 4.9b) [8]. A research conducted on air pollution from VOCs adjacent to an oil terminal was conducted at the University of Lithonia and the main focus was on the evaporation and release of VOCs around an oil terminal. Experimental and scale assessment data such as temperature, humidity, atmospheric pressure, wind speed and wind direction were considered in this study. Experimental results were compared to monitoring and measurement criteria and it was found that air humidity, wind and atmospheric pressure did not affect the concentration of VOCs in the environment [9]. The release of hydrocarbon vapors from the storage reservoirs of oil products at the Ofogh - Qeshm terminal was investigated. In this research, TANKS 4.0 was used to model the estimation of evaporation and this program has been designed to calculate the release of organic liquid storage tanks. In the research, using the software and entering data, the amount of annual and monthly evaporation of oil hydrocarbons from the surface of reservoirs was calculated. The results showed that about 1151 m^3 of hydrocarbons evaporated annually from the surface of the reservoirs of this terminal. In this research, the modeling of the dispersion was done by Screen 3. In total, emissions of hydrocarbons were reported even at the most critical point far below the clean air standards of the country [10]. The VOCs in air of the special economic petrochemical zone in two cold and hot seasons has been estimated. In this study, some of these pollutants, such as benzene, toluene, xylene, acetaldehyde and formaldehyde, have been investigated. Sampling of pollutants was carried out during two cold and hot seasons so that the effects of temperature change on the concentration of the pollutants could be determined. In total, comparing the results of two measured seasons, the highest concentration is associated with the summer season and the lowest concentration is related to the cold season. The average concentration of benzene and ortho-toluene and ethylbenzene in the hot season and in the cool season of toluene with ethylbenzene and with each other have a significant relationship [11]. The oil industry in Iran is a major producer and exporter of crude oil and increasing exports requires an assessment of the health and safety of workers in this industry [12]. Previous studies have shown that no comprehensive research has been conducted on the modeling of benzene release from an oil pit using the AERMOD model in a country like Iran. Therefore, this study aimed to investigate the benzene dispersion and to calculate its emission rate using TANKS 4.0.9, modeling the dispersion of these pollutants with the AERMOD dispersion model, and presenting emission reduction plans and air quality monitoring program.

Materials and methods

Study area

The research environment in this study is the Kharg Island. This island is located about 57 km northwest of Bushehr, 30 km from Bandar Rig and 35 km from Bandar Ganaveh. Its average altitude is about 3 m from the sea level and is geographically located at 29 ° 15 ‘ north latitude and 50 ° 20’ east longitude. The big oil pit of Kharg is located in one of the oil subsidiaries in the region. The oil pit is a natural valley that drains industrial waste water from factories that are largely due to oil waste. In this research, the dispersion of benzene with a range of 10 × 10 km^2 was carried out in four seasons of the year.

Sampling and analysis of air samples

An environmental measurement of benzene from 20 points around the oil pit and adjacent to the residential campus and repair workshops was carried out using the individual sampler pump model 222 manufactured by SKC, according to the NIOSH 1501 method. The LZB-3W router was used for calibration of the sampling pump. The sampling pump discharge rate was considered at 200 ml / min. After sampling, based on the recommendations and instructions presented in the NIOSH - 1501 method, the samples were kept at a temperature below 4 °C and analyzed in less than 30 days. The extraction was done by adding 1 cc of carbon sulfide for half an hour and the desired solution for determination of benzene concentration was analyzed by Varian CP-3800 gas chromatograph with FID detector [13].

Analysis and dispersion method of benzene

Modeling the outflow pollutants from the Kharg oil pit with the aim of controlling and reducing the air pollutants more efficiently and effectively, determining the amount of pollutant emissions and identifying the dispersion method of pollutants in this industrial zone are essential. The main problem of oil pit in the study area is the emission of hydrocarbons therefore, the amount and manner of release of this pollutant are modeled at 1, 3, 8, 12, 24 h, monthly and yearly average. In this study, the surface evaporation rate of oil pit was modeled using TANKS 4.0.9d and then, the oil pit surface has been studied using the AERMOD distribution model, as a tool for analyzing the release of VOCs and their dispersion. The TANKS 4.0.9d model has been designed by the US Environmental Protection Agency (USEPA) and the American Petroleum Institute (API) to estimate the emission rate of organic fluids from storage tanks. This software provides a report about the air emissions by entering information such as reservoir diameter and structure, chemical composition and liquid temperature inside it (and reservoir location), proximity to the city, ambient temperature, etc. This report includes estimates of monthly, annually or part annually emissions for each material stored in reservoirs or a combination of them. The AERMOD model has also been designed to estimate the concentration of emissions from near sources (< 50 km) and can be used to predict the concentration of different pollutants of spatial, linear and surface sources. This model is also used for large resources or for static and fixed resources. In addition to the AERMOD main processor, this model uses a meteorological pre-processor called AERMET and a geophysical pre-processor named AERMAP. The AERMET pre-processor is designed in such a way that, all available meteorological information can be defined in the form of the in-site file and used for processing [14 - 16].

The input data required for this model include surface parameters (precipitation, cloud coverage, atmospheric pressure, sea level pressure, Bowen ratio, Albedo coefficient and surface roughness length), and profile parameters (dew point temperature, temperature, direction and wind speed and moisture content) according to the meteorological information as well as the specifications of the source of contamination (emission rate and height of pollutant release and surface radius) according to the output of the TANKS. The AERMET preprocessor is designed to define all the available meteorological information in the form of in - site file in order to be used for processing. In this part of the modeling, the 3 h recording and quality controlled data for the 10 year period from 2004 to 2013 has been used by the country's meteorological station, the Kharg station and atmospheric data.

Meteorological data are removed from the range by AERMET values during the evaluation process. Unallowed and deleted values are reported in a file by the model. The values for the upper and lower limit of each parameter, and also the unregistered data, are defined by default for AERMET. But the user himself can also correct these default values through the input file that controls the preprocessor operation. The AERMET output file has been organized for the model input in which each section contains all meteorological information for a 24 h period. This period starts at 1 local time and ends at 24 local time. Also, this model requires three surface parameters from the studied area, i.e., the Bowen ratio (an indicator for determining the surface moisture, which is obtained from the thermal flux to heat flux ratio and its value ranges from about 0.1 to the surface water to 10 for the desert surface), the Albedo coefficient (a fraction of the solar radiation which, is emitted again into space without adsorption by the surface, and its value varies from 0.1 for forests with massive trees up to 0.9 for soft snow) and, surface roughness length (associated with wind flow and the height of surface barriers and in fact, the height at which the average horizontal speed of the wind reaches zero and this parameter changes from less than 0.001 m to a stagnant water surface to more than 1 m for the surface of forests and urban areas). In order to determine these values, it is necessary to divide the studied area into appropriate sectors according to the type of land use and its vegetation, in the clockwise direction and the values of these three parameters have been presented seasonally in Table 1. Pitt's central point coordinates in the modeling network are 433061.43 and 3232562.75. The location of other resources has been entered into the model according to the site center.

Modeling range

Square modeling dimensions are on the side of 10 km where, the center of the site is located in the center of the square. The modeling area is 100 km^2. According to articles published by the EPA, the study area has been meshed with the length of 500 m from the 50 m long modeling center, with a length of 1000 m from the modeling center with a distance of 100 m and a length of 5000 m from the center of the modeling with a distance of 200 m.

Table 1. Surface characteristics used in case study with seasonal variations

Season	Beginning of the sector (degree)	End of the se ctor (degree)	Application type and vegetation	Albedo coe fficient	Bowen ratio	Surface roughnes s (m)
Spring	0	150	the desert	0.3	5	0.3
Summer	0	150	the desert	0.28	10	0.3
Autumn	0	150	the desert	0.28	6	0.3
Winter	0	360	the desert	0.45	10	0.3

Pollution resources specifications

The model receives specific information for any type of pollutant. Oil pits were considered as surface pollutants in the AERMOD model. For modeling information such as pollutant emission rate per unit area, the height of pollutant release from the ground surface was introduced to the model.

Physical characteristics of the reservoirs

Considering that it is possible to simulate the oil pit in the TANKS4.0, a fixed vertical roof tank is used as a simulation of the conditions. These types of reservoirs consist of a fixed cylindrical shell with fixed roof that can be flat, dome, or conical-shaped. But the tank's axis is always perpendicular to its foundation. These reservoirs are either ventilated freely or with a pressure regulating valve and, if necessary, the heater may also be used at the end of these containers [17]. Two important types of release of tanks with vertical fixed roofs are storage losses and exploitation losses. As a result of changes in pressure and temperature, the volume of vapors inside the reservoir has increased and, they are discharged out of the pressure control valves, which are storage losses.

The type of chemical compounds stored in the reservoirs

In this section, the type of compounds constituting the fluid is determined. Most of the components are available in the software database by default. But the compounds not found in this list have been defined as hypothetical combinations for the program. For this purpose, after entering the name, critical temperature and pressure, also the centrifugal coefficient, the new compounds were added to the list of compounds and then used. The chemical composition considered in the oil pit is residuel oil No.6.

Meteorological information input to the software

Given that our country's climate conditions are not available in the software, therefore, the meteorological information related to the synoptic station has been calculated and using the Meteorological Information Edit menu, it is added to the database and then used. Entering the name of the city, the name of the province, the average daily temperature of the environment and the average atmospheric pressure for the entire statistical period, and also the average maximum daily temperature of the environment, the average minimum daily temperature of the environment, the average solar radiation coefficient and the mean wind speed for each 12 months of the year during the statistical period of the region has been registered in the local database and used. To calculate the solar radiation coefficient, the angle of the solar elevation is obtained for the length of the statistical period according to latitude and longitude. Given the height of the sun, the angle of the sun is determined. R_0 the solar radiation characteristic in the clear sky was calculated using the following formula:

$$R_0 = 990(SIN\theta) - 30 \quad (1)$$

By obtaining the solar radiation of clear sky and the cloudiness fraction N, the solar radiation characteristic was obtained from the following relation:

$$R = R_0(1 - 0.75N^{3.4}) \quad (2)$$

Results and discussion

Fig. 1 has presented the descriptive values of airborne benzene in separate sections at four stations. Airborne benzene at the Station A (0.53 ppm) and Station H (0.6 ppm) in spring and station M (0.72 ppm) and station P (0.81 ppm) during summer is higher than the standard recommended by the American Conference of Governmental Industrial Hygienists (ACGIH) and the EPA (0.5 ppm). Using Kruskal - Wallis statistical test, there was a significant difference between stations in different seasons ($P_{value} < 0.05$). The seasonal release of VOCs in the petroleum - based industries in 2012 and 2013 was conducted. In this study, about 346 air samples were collected in the winter of 2012 and 370 samples in summer of 2013 from 18 complexes located in an industrial zone. The average of most compounds in all of the complexes was estimated in summer higher than winter, which is consistent with the present study. Also, according to the results, the average benzene ratio showed the highest percentage (64 %) and the mean gasoline concentration ratio indicated the lowest (3 % to 7 %) in both winter and summer [18].

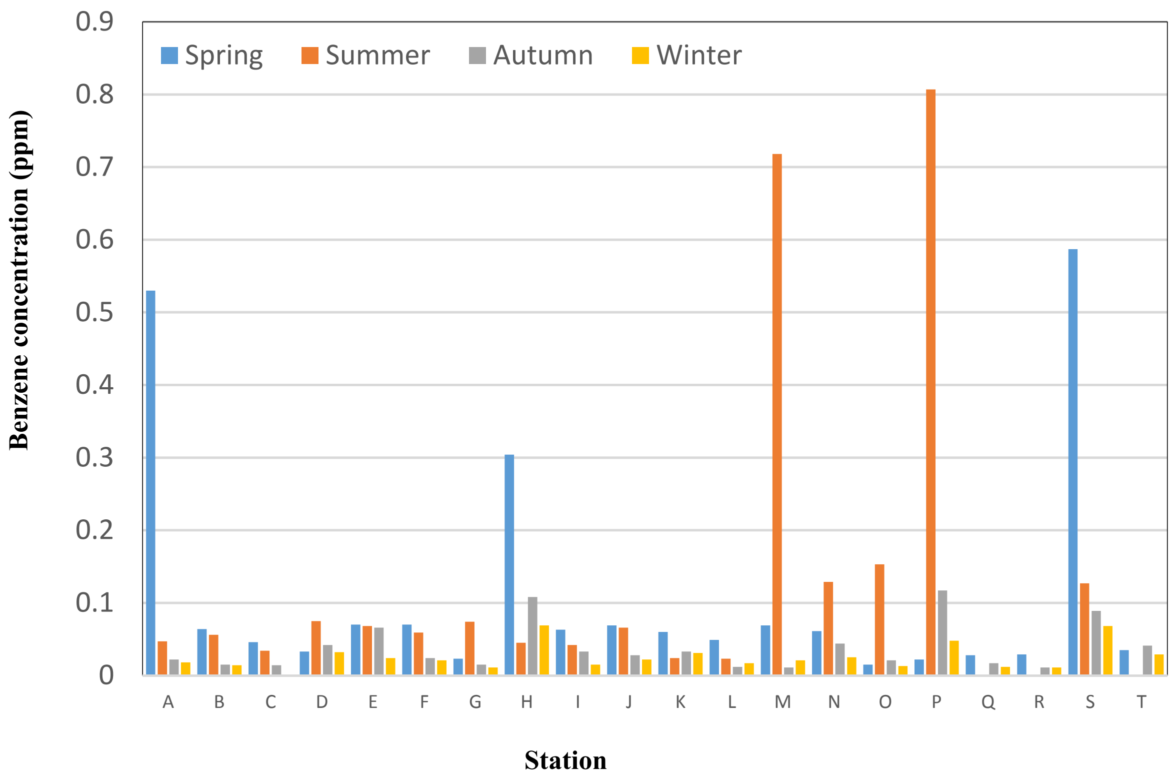

Fig. 1. The mean concentration of benzene at the studied stations

The solar energy can also cause concentration variation through oxidation and the amount of free radicals in the air in addition to the number of pollution sources. Therefore, considering this issue, the concentration of free radicals in the air can be considered as an indicator for the production of gases during measurement [19]. It was also found that the highest concentration of benzene was related to the stations of station A, H, M and P. These results are consistent with the wind direction in the region, which is often to the south and south - east (Fig. 2). Therefore, the high pro-

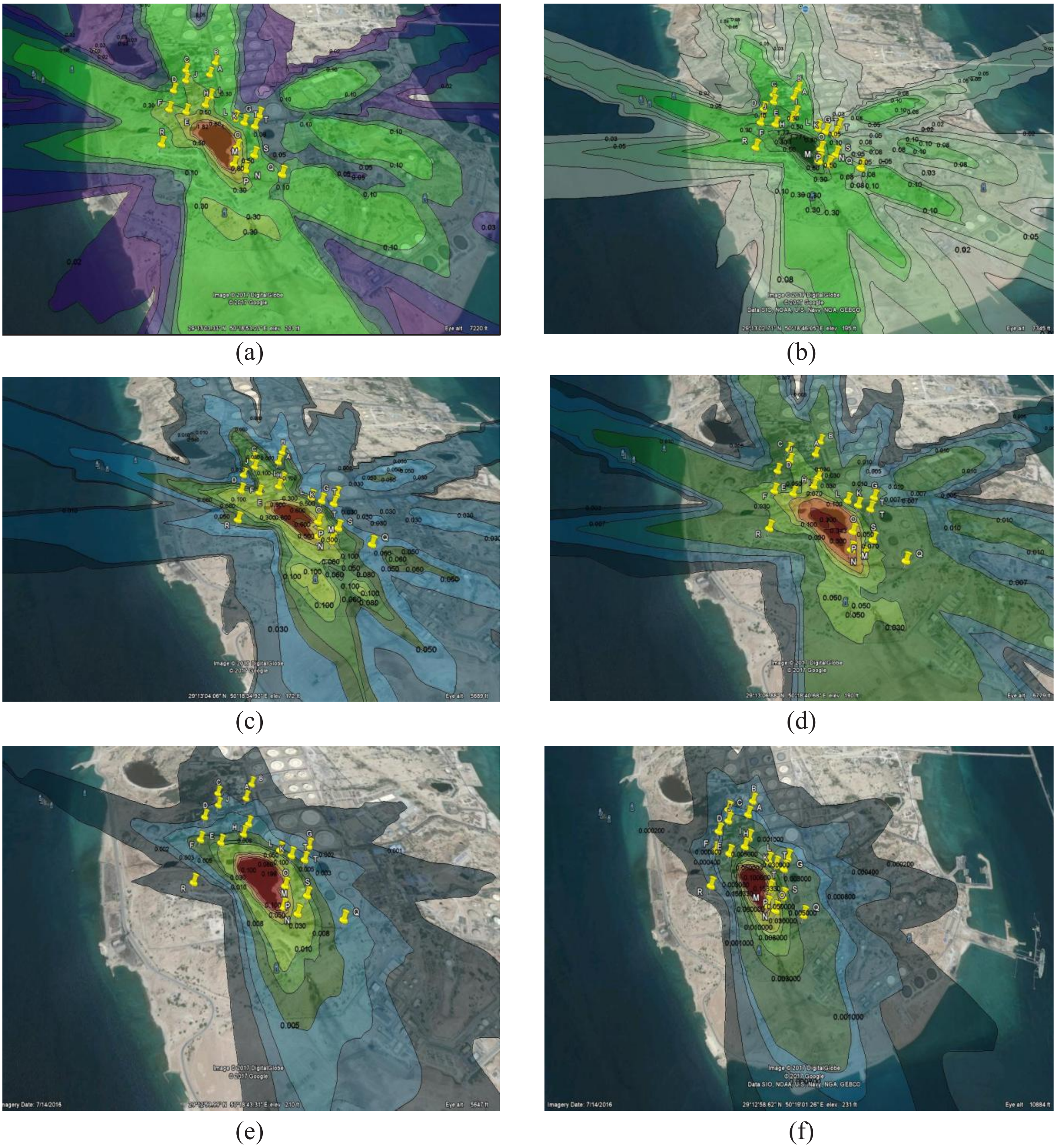

Fig. 2. Average 1 h (a), 3 h (b), 8 h (c), 24 h (d), monthly (e) and (f) annual dispersion of benzene

portion of benzene in these stations is due to the location of these complexes in the southern parts of the other sampling points. The results obtained from the software and the dispersion pattern of benzene showed that, pollutant plume has been scattered in the hourly average in all directions but due to the flatness of the area and the low height of pollutant source, the maximum pollutant is 121 m from the source in the direction of the northeast. The maximum monthly average is 90 m southwest of origin. In the mean annual, the maximum pollutant is 160 m from the origin of modeling in the direction of the south. The motion of the pollutant plume is from the average hourly to yearly direction to the south and, the results show that the pollutant plume moves in the wind direction (Fig. 2) and due to the lack of height of pollutant source, pollutant plume remains in the oil pit area.

The maximum concentration of benzene in the studied cases (Table 2) showed that, the maximum benzene concentration in the measured time intervals of one, two, and three hours is higher than the standard predicted by the EPA and the Iranian Department of Environment (DOE) (0.5 ppm).

The results of AERMOD modeling are in the form of hour, month or annual average. Given that the measured values at any time interval are not regularly measured, they will not be able to compare with the modeling results and the measured values are not an appropriate criteria for verifying the results of modeling. But in general, comparing the results of modeling and concentrations measured shows that, the average benzene concentration at south stations, including station A, H, M, and P, was higher than other points of measurement and higher than the standard predicted by the EPA and DOE. Also, the modeling results are closer to the measurement values in the 1 h average and as long as the time interval is larger, the modeling results are divergent from the measured values. The measured values are much higher than the modeling results in the monthly and annual average. The difference is due to the inability of the TANKS to simulate an oil pit in place of a tank with a fixed

Table 2. Maximum concentration of benzene in the studied area

Hourly average	Maximum concentration (ppm)	X (m)	Y (m)
1 h	0.57	433096	3232624
2 h	0.56	433096	3232624
3 h	0.55	433096	3232624
4 h	0.38	433096	3232624
6 h	0.34	433096	3232624
8 h	0.26	433096	3232624
12 h	0.18	433096	3232624
24 h	0.11	433196	5/3232424
monthly	0.06	433246	3232526.5
yearly	0.05	433196	3232424.5

Table 3. The measured and predicted benzene concentration in ppm

Measured benzene values (ppm)					Predicted benzene values (ppm)		
Station	Spring	Summer	Autumn	Winter	1 h	monthly	Annually
A	0.053	0.047	0.022	0.018	0.27	0.002	0.001
B	0.064	0.056	0.015	0.014	0.29	0.002	0.001
C	0.046	0.034	0.014	> 0.005	0.32	0.002	0.001
D	0.033	0.075	0.042	0.032	0.28	0.002	0.001
E	0.070	0.068	0.03	0.024	0.53	0.005	0.001
F	0.070	0.059	0.024	0.021	0.51	0.004	0.0005
G	0.23	0.074	0.015	0.011	0.5	0.003	0.002
H	0.587	0.127	0.089	0.068	0.52	0.02	0.05
J	0.304	0.045	0.108	0.068	0.51	0.005	0.004
K	0.063	0.042	0.033	0.015	0.47	0.005	0.004
L	0.069	0.066	0.028	0.022	0.48	0.002	0.0009
M	0.069	0.718	0.011	0.021	0.8	0.1	0.15
N	0.06	0.024	0.033	0.031	0.52	0.01	0.02
O	0.049	0.023	0.012	0.017	0.5	0.05	0.02
P	0.022	0.807	0.117	0.048	0.52	0.05	0.05
Q	0.061	0.129	0.044	0.025	0.61	0.06	0.11
R	0.015	0.153	0.021	0.013	0.83	0.16	0.05
S	0.028	< LOD	0.018	0.012	0.12	0.004	0.005
T	0.029	< LOD	0.011	0.011	0.28	0.002	0.0003
U	0.035	< LOD	0.041	0.029	0.42	0.004	0.1

vertical roof with limited depth and height. The emission rate of the oil pit of 0.0012 g / s has been calculated according to the software limitations for considering the Kharg oil pit.

According to the simulation results in Table 3, the 1 h values of benzene concentration are predicted and the concentration measured at the stations of the industrial waste water treatment plant unit, the output of the three factories and the southwest side of pit is higher. The modeling results at these points are maximum. Considering these factors, the model used has provided satisfactory logical results to predict the distribution of benzene concentration. Also, according to Table 3, it is seen that the trend of variations in the predicted 1 h concentration and the measured values in some places is also the same. Estimates show that model predictions are consistent with the measured results in oil pits and when the overall performance of the model was studied, all the calculated results indicate the success of the modeling. The results of the air quality study in the study area indicated that the AERMOD dispersion model is a suitable model for determining the average hourly benzene concentration from the point source emission sources. In general, considering the evaluation of predictions, the performance of AERMOD can be considered as an appropriate scientific tool for analyzing policy and control strategies to reduce pollutant concentrations.

Modeling the dispersion of carbon monoxide pollutant using the AERMOD model has been conducted in the South Pars Gas Refinery 4. It was concluded that the concentration of 1 h and 8 h of this pollutant is high compared to national and international standards. The authors concluded that the performance of the AERMOD software can be considered acceptable in predicting the pollutant concentration [14].

In another study on SO_2 modeling using the AERMOD model at Ramin Ahwaz power plant, the concentration of this pollutant in winter was lower than the standard concentration set by the DOE in all cases. The authors conclude that this model can be used as an effective model for determining the amount and dispersion of pollutants [20]. The release and modeled the dispersion of pollutants from VOCs from the surface evaporation of reservoirs located in Assaluyeh was investigated. In this case study, the emissions of VOCs due to surface evaporation from 16 reservoirs located in one of the South Pars gas field refineries containing 13 different types of organic liquid are determined by TANKS 4.0 and then, the dispersion of these pollutants using the AERMOD distribution model in an area of 15 × 15 km at network spacing of 150 m in 5 m of height (ground, 2, 10, 10, 30 m) and in the 12 month statistical period (2009) for time averages of 1, 3, 8 and 24 h and also 1 month and 1 year statistical periods and in the end, it was revealed that approximately 233 tons of VOCs pollutants were released from the reservoirs of this refinery. The share of reservoirs with an external floating roof with a release rate of approximately 47 tons / year is 20.08 percent and the share of reservoirs with a fixed vertical roof with an annual average of 186 tons, is 79.92 percent. Also, after simulating the dispersion of these pollutants in the area, it was found that the highest concentrations occur in all the mean time at a height of 20 m from the earth's surface and in the first half of 2009, the northwest wind (315 degrees), and in the second half of 2009, the southern eastern wind (135 degrees) further exacerbated the region [21].

Conclusion

In this study, benzene environmental measurements were studied in oil pit of Kharg island in 4 different seasons. Then, the release rate of benzene due to pit surface evaporation was simulated. The highest concentration of benzene was observed in summer. The study of air quality in the study area indicated that the AERMOD model is a suitable model for determining the average emission concentration. In general, considering the estimation of the predictions, the performance of the AERMOD software can be considered acceptable in predicting the benzene concentration. So that this model can be used as a suitable scientific tool for dispersion of the analysis of control and policy strategies for reducing and preventing air pollution. In addition, air pollution in the oil pit can be reduced by correct locating of industrial development, considering the prevailing wind direction in the region, and the use of standard equipment to prevent the release of benzene. Therefore, specific emission control policies are necessary to reduce pollution in contaminated areas.

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Competing interests

The authors declare that there are no competing interests.

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Ethical considerations

"Ethical issues (Including plagiarism, Informed Consent, misconduct, data fabrication and / or falsification, double publication and/or submission, redundancy, etc) have been completely observed by the authors."

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