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Assessment of industrial and urban atmospheric pollution from Liquefied Natural Gas (LNG) plant GL1K Skikda Algeria

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Article Info	ABSTRACT
Article type: Research Article	The atmospheric pollution is caused by industrial activities has been recognized as an important factor affecting the air quality. Liquefied natural gas (LNG) plant emit plenty pollutants such as methane (CH_4), sulfur dioxide (SO ₂), nitrogen monoxides (NO), Nitrogen
Article history:	dioxide (NO_2) and carbon monoxide (CO). The Skikda LNG plant is the major provider of LNG in East of Algeria and it contributes to national economy This study seeks to assess the
Revised: 29 March 2024 Accepted: 28 April 2024	concentration of gases: CH_4 , SO_2 , NO, NO_2 and CO generated by LNG plant of Skikda in Algeria GL1K.Six regions have been considered in this study. Atmospheric pollutants were analysed in each site and compared to the Algerian permissible limits. Results indicate that
Keywords: Air pollution Environmental risks Petrochemical plants Emissions	the mean concentration of CH ⁴ in LNG plant was 2775.88 mg/Nm3and significantly higher than Algerian permissible limits at 150 mg/Nm ³ . However, the average concentration of CO detected in LNG plant was 200.01 mg/Nm ³ and exceed the limit which is 150 mg/Nm ³ . The measurement of SO ₂ , NO, NO ₂ has shown an acceptable mean concentrationin all sites which were within the Algerian permissible standards. This study highlights that LNG plants constitute a major source of air pollution, presenting substantial environmental hazards and detrimentally affecting human well-being. Consequently, it is crucial for authorities to implement significant initiatives, including waste minimization, robust recycling practices, and the integration of continuous emissions monitoring systems to protect the environment from the detrimental impacts of air pollution in petrochemical plants.

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INTRODUCTION

In recent environmental studies, scientists disclosed that air pollution is an acute worldwide concern. Air pollution can be defined as an atmospheric condition in which certain elements are present in higher quantities than natural levels. This latter affect people plants, as well as certain materials tremendously (Isaksen et al., 2009). These ambient elements are constituted of gas carbon dioxide, methane, propane, helium, and particulate matters (aerosols, fumes, smoke, dust, radioactive materials). Such components are essentially harmless since they exist in the atmosphere in low concentration (Rao, 2007).

Nonetheless, air pollution has been attributed to various natural and anthropic factors. These factors range from the stratospheric-tropospheric exchanges, the altitude of a location.

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In addition to the photochemical reactions that occur with volatile organic compounds (VOCs) in the presence of sunlight (Rabideau et al., 2012, Lin et al., 2021, Thambiran et al., 2010). Environmental pollution from substances such as: Sulfur dioxide (SO₂), Carbon Monoxide (CO), Nitrogen Monoxide (NO), Carbon Dioxide (CO₂), Nitrogen dioxide (NO₂), and methane (CH₄) is an alarming issue, particularly in densely populated, industrialized regions of nations with petrochemical activity like petroleum refining and natural gas liquefaction (Keawboonchu et al., 2023, Etemadfard et al., 2021).

Air pollution indeed poses a perilousrisk for human beings in relation to their health and the quality of their life (Ozcan et al., 2023). Accordingly, among the most pressing problems that result from constant exposure to high volumes of pollutants; respiratory problems, exacerbation of people's cardiovascular diseases, and even early deaths can be noted (Esposito et al., 2014).

In this regard, many studies reported that air pollution is causing a major health impact in urban cities of the world (Ilyas et al., 2010, Cassidy et al., 2014, Fridell et al., 2014, Kumar, 2016).

Essentially, natural gas is a hydrocarbon gas and non-renewable fossil fuel that materializes below the Earth's surface. Natural gas provides fuel and energy services for heating, cooking, energy service which makes it both a source of heat and power. Moreover, it is a fundamental raw material in many industrial processes. Natural gas can also be used as fuel for motor vehicles and in the manufacture of plastics. Additionally, natural gas does assuredly contribute to climate change (Mathur et al., 2022, Siddiqi, 2002).

Algeria is one of the largest worldwide exporters of liquefied natural gas. Skikda LNG (GL1K) plants produces and sells Liquefied Natural Gas (LNG) and its products. The company SONATRACH undertakes activities necessary to liquefy, store, transport and market Skikda's natural gas. The company also delivers LNG to international customers. Substantially, the company operates a three-train liquefaction plant with a capacity of 10.4 million tonnes per annum (mtpa), near the port in east of Algeria to facilitate expedition.

According to previous studies, Skikda is a metropolis in Algeria witch suffers from the harmful effects caused by different types of pollution released by urban and industrials sources (Rouaïnia et al., 2012, Benaïssaet al., 2020, Khelfaoui et al., 2022, Benaïssa et al., 2022). The major source of atmospheric pollution at Skikda is petroleum refining and liquefaction of natural gas. However, human and industrial activities have resulted in serious pollution in this area (Nafissa et al., 2005).

However, our study is the first concerning evaluation of air pollution and dispersion from Liquefied Natural Gas complex of Skikda one of the large petrochemical areas of North-East of Algeria: In second part of this paper, we focus on the diffusion of atmospheric pollutants from LNG plant and in several urban cities nearly petrochemical zone. This work was carried out to estimate the atmospheric pollutants: CH_4 , SO_2 , CO, NO_2 and NO concentrations in Skikda region, one of the very active, and populated urban-industrial areas of Algeria (East of Algeria, Fig. 1).

MATERIALS AND METHODS

Description of liquefaction natural gas LNG plant

Skikda city, this study's area, is located in north eastern coast of Algeria with an area of 4.026 km². Skikda encompasses the largest petrochemical platform in North Africa with a 1.200 ha of numerous oil complexes. The majority of these complexes reject atmospheric pollutants in air which affect the ecosystem. Fig. 1 depicts a map of the area of the study sites, the Skikda LNG plant is highlighted.

The petrochemical complex LNG is a natural gas liquefaction plant and it is situated to about four kilometres to the east of Skikda. It is implanted in the Industrial zone of Skikda located in



Fig. 1. Location map of the study area.

the Gulf of Stora. The LNG complex is supplied with natural gas from the Hassi R'Mel deposit by a pipeline with a length of 580 km and 40 inches in diameter throughout five compressor stations along the pipeline (SONATRACH., 2010).

Sampling was conducted from June 2019 to May 2020 on 36 sampling sites, including 360 air samples. In Skikda, the air quality is checked via fixed and mobile stations. The stations possess equipment that vary between measuring instruments and analyzers of the pollutants such as Methane (CH₄), Sulfur dioxide (SO₂), Nitrogen oxides (NO_x), and Carbon monoxide (CO). They also contain devices for meteorological measurements. The cloud concentrations have been measured by three portable sensors of type BM25, which cloud has allowed us to calculate the concentration of cloud dispersion that is composed of many partial gases by itself. As a matter of a fact this system operates coherently with the ability to read the air quality daily in the areas most affected by this phenomenon, like the big cities and industrial zones.

Description of portable sensor

Essentially, the portable sensor BM25 gas detector detects up to 5 gases simultaneously. It is the most popular and widespread of the area monitors. We can mainly find it on public works sites in standard four-gas configurations (explosive gases, SO_2 , CO, and NO_x). It is equipped with a high-intensity 360° flashlight and a powerful 103 dB screamer.

Statistical analysis

Based on the 360 observations, the mean, minimum and maximum values and standard deviations were computed and recorded. Package for Social Science (SPSS) version 21 was used for computing all statistical analyses. One-Way Analysis of Variance (ANOVA) was used to test the differences of atmospheric pollutants concentrations between areas. The difference by regions was considered to be significant when at a probability threshold (p) is less than

5% (p<0.05). The tests were used to compare means of concentrations of different atmospheric pollutants with Algerian permissible limits.

Principal component analysis (PCA) is a multivariate analysis used in environmental research. PCA has been applied to represent the association between different gases in different areas, and to identify different pollutant sources. Hierarchical Clustering (HC) is a traditional method of dimension reduction (Douabul et al., 2013).

RESULTS AND DISCUSSION

The measured concentration of atmospheric pollutants in the six sites revealed the presence of atmospheric pollutants, represented in boxplot form in Fig. 2. The statistical description of



Fig. 2. Box plots of hazardous gas concentration (mg/Nm³) for air in study area.

all atmospheric pollutants, including CH4, NO, NO2, and CO and the analysis of variance by one-way ANOVA are given in Table 2.

Fig. 3 shows the mean concentration levels of CH_4 in the six sites of Skikda, with a total sample population of 360. The maximum concentration of CH_4 found in S4 (LNG plant) ranged

Sites	stations	Location
Site 4	P1-P10	LNG plant
Site 5	P11-P19	HamroucheHamoudi*
Site 3	P14-P21	Arbi Ben M'hidi*
Site 1	P22-P26	Filfila*
Site 2	P27-P31	Alaoui Taghane*
Sites 6	P32-P36	Ilot des chèvres*

Table 1. Detailed location of sampling stations

Table 2. Comparison of atmospheric pollutant	s (mg/Nm ³) in the air of LNG plant in Skikda.
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Gases Site		Mean \pm SD (mg/Nm ³)	Min (mg/Nm ³)	Max (mg/Nm ³)	LMP **	One way ANOVA	
	(ing i vii		(((mg/Nm ³)	F	р
CH ₄	Site 1	$800.64 \pm 64.11*$	500.70	1300.48			
	Site 2	$413.84 \pm 42.81 *$	110.17	900.25			
	Site 3	$310.52 \pm 29.3*$	98.91	499.76	150	55 15	0.0001
	Site 4	$2775.88 \pm 49.65*$	74	4200.98	150	55.15	0.0001
	Site 5	$367.21 \pm 42.97*$	111.03	925.43			
	Site 6	$1780.41 \pm 600.93*$	100.20	10000			
CO	Site 1	$315.49 \pm 28.93 *$	99.20	500.94			
	Site 2	$304.91 \pm 33.56*$	100.20	500.94			
	Site 3	$301.60 \pm 4.26*$	200.11	500.01	150	22.80	0.0001
	Site 4	$3313.64 \pm 683.1*$	250.01	10000	150	33.00	0.0001
	Site 5	$380.55 \pm 44.30*$	100.26	900.25			
	Site 6	$414.02 \pm 43.01 *$	100.20	906.23			
NO_2	Site 1	$5.06\pm0.37\texttt{*}$	3.01	56			
	Site 2	$5.68\pm0.66\texttt{*}$	1.2	12.75			
	Site 3	$3.60 \pm 0.3*$	1.20	5.94	200	15 21	0.0001
	Site 4	$31.38 \pm 2.56*$	1.98	98.85	300	13.21	0.0001
	Site 5	4.20 ± 0.49 *	1.20	9.25			
	Site 6	$19.17 \pm 4.86*$	2.01	89			
NO	Site 1	$7.08\pm0.46\texttt{*}$	4.58	12.75			
	Site 2	$8.44\pm0.58\texttt{*}$	5.40	13.50			
	Site 3	$3.24\pm0.30\texttt{*}$	1.20	5.94	200	7 73	0.0001
	Site 4	$39.28 \pm 8.33*$	2.50	100	200	1.23	0.0001
	Site 5	$6.88 \pm 2.3*$	3.16	336			
	Site 6	$24.59 \pm 6.51 *$	1.20	100			
SO_2	Site 1	$3.37 \pm 0.29*$	1.20	5.94			
	Site 2	$7.64 \pm 2.25*$	1.20	69			
	Site 3	$3.21 \pm 0.29*$	1.20	5.69	300	1 32	0.26
	Site 4	$31.94\pm6.84\texttt{*}$	2.01	100	500	1.32	0.20
	Site 5	7.67 ± 0.55 *	5.59	12.75			
	Site 6	19.17 ± 6.79	5.01	618			

SD: refer to Standard Deviation of gases concentration

F: refer to Friedman values of one away ANOVA

p: refer to significant of ANOVA

* Significant differences from Algerian permissible limits (P<0.05 of t-Student test) are marked with asterisks

**LMP: Official Journal of the Algerian Republic, July, 2003. No. 03of 10 of 19 July 2003(Official Journal of the Algerian Republic.2003).

from 74 mg/Nm³ to 4200.98 mg/Nm3 with a mean of 2775.88 mg/Nm³ and a standard deviation of 49.65 mg/Nm³. On the other hand, the lowest concentration of CH_4 was detected in S3 compared to other sites. The level of CH_4 in the six sites was significantly different and higher than the permissible limit of 150 mg/Nm³. However, the one-way ANOVA analysis revealed a significant difference in CH_4 concentration among the different sites (ANOVA, F=55.15, p<0.05) because of the activities related to natural gas liquefaction and refining of petroleum.

Table 2 shows that there were significant differences between CO levels in the six areas. Fig. 4 shows the mean concentration of CO in the six studied sites. The highest mean concentration of CO was detected in S4, representing the LNG plant (3313.64 mg/Nm^3). CO atmospheric pollution is the outcome of the activities related to flue gases following petroleum refining activities, natural gas liquefaction, and automobile escape gases. The lowest mean concentration of CO was found in S6 with 301.60 mg/Nm³. As shown in Table 2, the test of CO demonstrated that the mean concentrations in the studied sites were remarkably different and high compared to the limit threshold value. A comparison of CO concentrations between the studied areas showed significant differences (Fig. 4) (ANOVA, F=33.80, p<0.05).



Fig. 3. Concentration of CH₄ (mg/Nm³)



Fig. 4. Concentration of CO (mg/Nm³).

Fig. 5 presents the variation of the mean concentration of NO₂ between sites. The mean maximum and minimum concentrations of NO₂ were 31.38 mg/Nm³ and 3.60 in S4 and S3, respectively. The NO₂ levels in the six studied areas were lower than the permissible limits, and the ANOVA test revealed significant differences between the studied areas (ANOVA, F=15.21, p<0.05).

The measured concentrations of NO in atmospheric samples of the studied sites are shown in Fig. 6. The maximum and minimum mean concentrations were 39.28 mg/Nm³ and 3.24 mg/Nm³, respectively, detected in S4 and S3. The one-way ANOVA analysis revealed a significant difference in NO concentration among the different sites (ANOVA, F=7.23, p<0.05).

Fig. 7 shows the concentration level of SO_2 in the six study areas in Skikda, with a sample population of 60. The maximum mean concentration of SO_2 was 31.94 mg/Nm³ found in S4 (LNG plant). However, the lowest mean concentration was found in S3 with 3.21 mg/Nm³ compared to other sites. As shown in Table 2, the mean concentrations in the regions studied do not exceed the limits allowed for SO_2 . For these reasons, there were no differences between the means of SO_2 (ANOVA, F=1.32, p>0.05) in the six areas.



Fig. 5. Concentration of NO_2 (mg/Nm³).



Fig. 6. Concentration of NO (mg/Nm³).



Fig. 7. Concentration of SO₂ (mg/Nm³).

This study also shows that the mean concentrations of gas for the six sites types S1, S2, S3, S4, S5, and S6 follow the order:

Classification by site S1: CH₄>CO >NO>NO₂>SO₂ S2: CH₄>CO>NO>SO₂>NO₂ S3: CH₄>CO>NO>SO₂>NO>SO₂ S4: CH₄>CO>NO>SO₂>NO₂ S5: CO>CH₄>SO₂>NO>NO₂ S6: CH₄>CO>NO>SO₂>NO₂ Trends in gas concentrations in the different sites were in the following order: CH₄: S₄>S₆>S₁>S₂>S₂>S₃ CO: S₄>S₆>S₂>S₁>S₂>S₃ NO₂: S₄>S₆>S₂>S₁>S₅>S₃ NO: S₄>S₆>S₂>S₁>S₅>S₃

 $SO_2: S_4 > S_5 > S_5 > S_2 > S_1 > S_3$

Table 2 shows that the air collected behind the LNG plant contains the following air pollutants: CH_4 , CO, NO, NO₂ and SO₂. However, differences in atmospheric pollutant levels among the sites can be attributed to various petroleum plant activities. Moreover, results revealed that the samples contained hazardous levels of atmospheric pollutants, especially the constituents of natural gas, which include CH_4 . The composition of natural gas can explain the presence of CH_4 . As a result, CH_4 contributes to global warming potential concerning the previous estimate. It absorbs radiation at a shorter wavelength in the lower atmosphere. CH_4 is a greenhouse gas that contributes to Climate Change (Etminan et al., 2016, Jackson et al., 2020).

This study is similar to those of (Ghasemzade et al., 2017) The study found that CH_4 is a substantial problem, particularly in highly urbanized, industrialized regions of nations with petrochemical activity like petroleum refining and natural gas liquefaction (Likus-Cieślik et al., 2020).

Whereas the presence of the pollutants, CO, can be explained by emissions from industrial petrochemical activities due to flare releases and gas leaks at the natural gas liquefaction complex, emissions from fossil-fuel-powered engines and Produced gas in landfills (Ghasemzade et al., 2017).Carbon monoxide (CO) is a pollutant that can adversely affect human health as it can reduce the delivery of oxygen to the body's organs and tissues (Jalali et al., 2019).

On the other hand, the lowest values are found far away from roads and in proximity to

residential areas and green spaces. Furthermore, results pointed out that the samples contained hazardous levels of SO_2 , especially near the LNG plant of Skikda. The presence of SO_2 can be explained by the pretreatment of natural gas petroleum fuels, which consists of desulfurization as flared gases and a significant role in energy production, especially crude oil production (Salmabadi et al., 2019).

According to the results obtained and compared to the Algerian standard, the NO₂ and NO concentrations do not exceed Algerian limits. NO₂ is produced from anthropogenic emissions, industrial burning of fossil fuels: oil, gas, and coal; vehicle exhaust, electricity and natural sources (soils through the decomposition process of nitrates and lightning).

In addition, results suggest that the natural gas liquefaction activities at the Skikda LNG plant could harm the health of sensitive individuals working in the plant's operational areas and those living nearby. Several studies conducted that NOx can cause human health impacts (Afghan et al., 2020).

Based on the concentration of atmospheric pollutants in all of this study's samples, the trends gases concentrations in the different sites were in the following order: $CH_4 > CO > NO > NO_2 > SO_2$, which is in accordance with the overall trend in the literature of Liquefied natural gas (LNG) plants. Overall, all measurements showcased the necessity to take action to reduce the emission of pollutants and gases emitted from LNG plants.

Subsequently, people living near roads or industrial areas are endangeredon a daily basis to several harmful emissions that may affect their health. Thus, a proper strategy should be applied to reduce air pollution in this area.

ACP

Multivariate statistical methods such as principal component analysis and cluster analysis were widely applied to study differences and correlations between atmospheric pollutants concentrations to find the distributions of pollutants in explored areas. They are efficient in representing air pollutants.

The correlation matrix is helpful because it can point out associations between variables that can show the overall coherence of the dataset and identify the influence factors, which help identify the sources of different elements. Table 3 presents the correlation matrix of the five gas variables in the atmosphere. Only those with correlation values greater than 0.50 are taken into account. However, the gas correlation matrices show that different gas pollutants do not have positive correlations.

Principal Component Analysis (PCA) has been implemented to study differences and correlations between the air gas concentrations to find the distribution of the atmospheric pollutants in explored areas. According to the results, waste gas concentrations in different regions can be arranged into two principal components, described as 25.70 % and 22.57% of the variability observed in gas levels produced in various study areas. It can be observed from Fig. 8 (a) that the correlation between CO and NO was significant. These results imply that CO, CH_4 and NO may originate from a similar pollution source: the LNG plant. These results could prove that the complexity of natural gas liquefaction is a significant source of NO, CO and CH_4 pollution.

	СО	NO	NO ₂	SO ₂	CH ₄
СО	1,000	-,006	-,023	-,011	-,171
NO	-0,006	1,000	-,036	-,023	-,102
NO ₂	-0,023	-0,036	1,000	,138	-,214
SO ₂	-0,011	-0,023	0,138	1,000	,014
CH4	-0,171	-0,102	-0,214	0,014	1,000

Table 3. Correlation matrix calculated using Pearson coefficients between pollutants.

In addition, the principal component analysis and factor analysis were performed for sites on 365 observation points. PCA better explains the possible groups/sources that influence air systems (Table 4, Fig. 8 (b)).

From table 4, the site correlation matrices show that different preferment sites pairs have strong positive correlations between S2 and all other sites, including S2-S4 (r = 0,841), S2-S5 (r = 0,669), S3–S4(r = 0,780), S3–S5 (r = 0,660), S1–S5(r = 0,732), S1-S2 (r = 0,639) and S1-S3 (r = 0,671). S2 is strongly and positively correlated with S3 (r = 0,956). The results obtained show that gas in the atmosphere have a common source.

From the loading plot in Fig. 8(b), it was observed that the concentration of waste gas in S4 can be correlated more easily. The F1 axis described 63.94 % is linked to S4, representing the LNG Skikda plant, the most polluted site. The F1 axis is related to S4, S2, and S3 on the right side. While the F2 axis described 18.58 % and is associated with S6, S1 and S5.

Comparing between the two plots in Fig. 8assisted to identify the variations between sites and their different atmospheric pollutants. Looking at the two plots in Fig8 (a), a distinctive variety among regions was observed, and the various pollutants introduced in them were also seen.

Hierarchical Cluster Analysis

The cluster analysis has successfully classified hazardous and contaminated sites by identifying the spatial similarity between the studied sites according to atmospheric pollutants. It identifies polluted areas. This study used cluster analysis to visualize air pollutant clustering.



Fig. 8. (a): Loading plot of variables, (b): Score plot of samples sites.

Table 4. Correlation matrix calculated using Pearson coefficients between sites

	S1	S2	S3	S4	S 5	S6
S1	1,000	,639	,671	,320	,732	,422
S2	0,639	1,000	,956	,841	,669	,229
S3	0,671	0,956	1,000	,780	,660	,251
S4	0,320	0,841	0,780	1,000	,479	,065
S5	0,732	0,669	0,660	0,479	1,000	,279
S6	0,422	0,229	0,251	0,065	0,279	1,000



Fig. 9. Dendrogram of Hierarchical Cluster Analysis of sampling stations from area

Fig. 9 shows the results. The Dendrogram of 6 sampling sites for atmospheric pollutants is divided into three major clusters in Fig. 9.

Cluster 1 has four sites: S1, S2, S3 and S5. These sites are the regions around the petrochemical complex where atmospheric pollution spreads; even inhabitants in the Hamrouche Hamoudisites constitute a risk for the population. Whereas cluster 2 consists of site 6, close to the industrial area, generating significant pollution. However, cluster 3 consists only of S4. This point of Natural gas liquefaction complex LNG plant is the primary source of air pollution. These findings show that the spatial groupings produced from the multivariate analysis are the same.

CONCLUSION

In this study, air quality near the LNG Skikda plant in northeast Algeria was assessed by determining hazardous air pollutants and revealing their sources. The results show that the concentration of hazardous air pollutants: CH_4 and CO are present in most samples from the natural gas liquefaction activities at the Skikda LNG plant. PCA and HCA analysis identified that the LNG plant is a significant anthropogenic source of air pollution in Skikda. The air analysis clearly demonstrates that hazardous air pollutants released from petrochemical plants pose a high contamination risk to the air and to the people who live nearby these LNG plants. To address this issue effectively, crucial steps include waste minimization, implementing robust recycling practices, adopting continuous emissions monitoring systems, ensuring regular equipment maintenance, optimizing processes, using cleaner fuels, and creating green spaces around the petrochemical plant. These elements are integral components of a comprehensive strategy aimed at minimizing air pollution, promoting sustainable practices, and ensuring a healthier, cleaner environment for both the community and the ecosystem at large.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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