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POLLUTION ESTIMATES OF DISCHARGE WATER FROM A NATURAL GAS LIQUEFACTION COMPLEX

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Abstract- The purpose of this research is to make a spatio-temporal monitoring of the pollution of industrial effluents generated by a natural gas liquefaction complex. We targeted 04 main discharge sites during a period of 06 months from February to July 2018. The statistical results of physicochemical analyses of the parameters temperature (*T*), Hydrogen Potential (*pH*), suspended solids (*SS*), chemical oxygen demand (*COD*) and five-day biochemical oxygen demand (BOD_5) showed a significant increase of temperature and *pH* (35.80 °C - 10.8) at site E1, BOD_5 and *SS* at site E2 (30.41 mg/L - 45.39 mg/L) and *COD* (135.11 mg/L) at site E3. Metallic analysis of the elements cadmium (Cd), copper (Cu), chromium (Cr), nickel (Ni), iron (Fe), manganese (Mn), lead (Pb) and mercury (Hg) recorded an increase in Fe during the months of February and march in site E1 (3.73 mg/L - 0.78 mg/L). Ni and Mn mean values increased during April at all four sites.

Keywords: Natural Gas Liquefaction Complex, Water Pollution, Heavy Metals, Physicochemical Parameters, Wastewater, PCA.

1. INTRODUCTION

When water is used in industrial processes, it becomes depleted or enriched with all kinds of substances, or changes in temperature, and the resulting pollution ends up in the aquatic environment. The nature and composition of the discharges are different from one site to another and are responsible for the negative impacts on the natural environment, which requires a specific analysis for each type of pollution. The international scientific community has announced that anthropogenic activities have, or will have, consequences on the functioning of all ecosystems on the planet [1]. Uncontrolled urbanization, tourism and industrialization around coastal areas have led to an alarming level of pollution of aquatic environments due to anthropogenic inputs. Moreover, industry is the human activity that generates the most wastewater [2] and considerable loads of pollutants [3]. It is a major interface between man and the natural environment; it requires resources to design goods or services that meet human needs and discharges waste and effluents [4]. The large industrial complexes are all located on the Algerian coast, on 1622 km of seafront [5]. Algerian industry is dominated by the petrochemical, chemical and steel industries, focus on coastal areas; more than 50% of Algerian industrial complex are registered.

Coastal zones occupy a considerable area along the Algerian coast and are exposed to pollution due to population growth, increasing urbanization and industrialization. Sewage contaminated with organic matter, suspended solids, detergents and lubricants is one of the main reasons for the degradation of Algeria's coastal ecosystems [6]. More than 90 million cubic meters of a wastewater are discharged each year on the coast. Pesticides, fertilizers and solvents are discharged directly into the environment when used by humans and indirectly as industrial waste from various activities such as mining, industrial manufacturing, incineration, fuel consumption or accidental discharges [7]. Living organisms are exposed in the marine environment to a complex mixture of chemicals and transformation products responsible for multiple damages at the organism, population and ecosystem levels [8]. The latter, with a high enrichment factor and a slow rate of elimination, are an early indicator of the disturbance induced by environmental contamination in populations [9].

The presence of organic substances comes more precisely from the presence of oils and/or the use of solvents and degreasers; the latter can accumulate in living organisms and present biomagnification phenomena in food webs [10]. The imbalance created between metals in sediments and surface waters is disturbed, which may be a determining factor in the intensification of water contamination [11]. The uncontrolled discharge of liquid effluents generated by industrial complexes is due to the poor treatment of wastewater treatment plants, which are generally non-operational or absent; this will lead to water pollution, a real danger for marine flora and fauna and for human health. Industrial effluents are the major actors of the ecological imbalance at the level of coastal ecosystems.

The composition and concentration of industrial effluents are extremely variable according to the type of industry. The contamination of these waters is strongly influenced by the site environment. Industrial effluents are the major cause of environmental pollution [12] and contain toxic organic and mineral substances within certain limits and cause ecological imbalance of the receiving environment and poor biological treatment [13].

The impact of industrial effluents on the environment is currently a clear reality and a serious threat to the quality of these waters [14]. The multiple anthropogenic activities cause discharges of chemical substances of various types and origins, whether diffuse such as urban and rural run-off, atmospheric fallout or collected such as industrial effluents. Water, a precious commodity that is indispensable for the operation of factories, is undergoing degradation of various origins (industrial, domestic or agricultural). It is subject to difficult situations leading to the disruption of all ecosystems, especially the marine system. Pollution of the sea by effluents is clearly visible in the industrial zone, which is a site for the production of petrochemicals (ammonia, urea, and methanol) and the processing of petroleum products (oil refining, natural gas liquefaction).

This study is based on analyses of the physicochemical (*Temperature*, *pH*, *SS*, BOD_5, *COD*) and metallic (Cadmium, Copper, Chromium, Nickel, Iron, Manganese, Lead and Mercury) parameters of the wastewater from an industrial complex. The complex discharges a significant quantity of water used in its natural gas liquefaction process: desalination of seawater to produce fresh water and cooling of certain gases (propane). The aim of this research is to highlight the level of pollution generated by the various activities of the natural gas liquefaction complex through the analysis of various wastewater characterization parameters. A six-month follow-up of the liquid discharges from this industrial complex was carried out to better identify the nuisances generated.

2. MATERIALS AND METHODS

2.1. Selection of Sample Stations

Four sample stations were chosen in the natural gas liquefaction complex to investigate the pollution of its discharge waters (Figure 1, Table 1).

Figure 1. Location of sample sites

Table 1. Description and location of sample sites

Site	Description	Location
E1	Boiler drain	35°48'30°N 0°15'28°W
E2	Discharge pipe for oily water from natural gas liquefaction trains	35°48'40°N 0°15'26°W
E3	Loading bay	35°48'48°N 0°15'14°W
E4	General discharge channel	35°48'43°N 0°15'47°W

This complex was chosen because several sources of pollution were identified. The important pollution discharges in the port of Arzew are urban and industrial. In the marine environment, manganese (Mn), zinc (Zn), mercury (Hg), copper (Cu), chromium (Cr), nickel (Ni), aluminum (Al), lead (Pb), cadmium (Cd) and arsenic (As) are the heavy metals that have received the most research attention due to their toxicity to organisms and risks to human health [15].

These metals (Cd, Cu, Cr, Ni, Fe, Mn, Pb and Hg) were chosen for this study due several reasons. The analysis of metals was realized by the Perkin-Elmer flame atomic absorption spectrophotometer (AAS) in the laboratory of the RAFFINERIE of the industrial zone of Arzew (RA1/z) which has specific lamps for these metals. All these metals were selected because they are the most represented and most problematic in the environment, for their ability to concentrate in the body of the organism and to propagate along the trophic chain, and for their potential toxicity (even at very low concentrations) to ecosystems and human health, which is a global concern. They are measurable at trace levels, their assays are easy to perform, and levels measured in tissues are correlated with exposure [16]. Pollution by metals and hydrocarbons presents definite dangers for the Mediterranean in the short and long term.

2.2. Sampling

The samples were collected over a period of 6 months, from February to July 2018, at a rate of 3 samples per month. The samples were collected between 9H00 and 12H00 am, taken between 30 and 50 cm deep [17]. The samples were collected in polyethylene bottles, previously rinsed 3 times with the distilled water to be analyzed. They were then stored at 4 °C until transported to the laboratory, where they were analyzed within 24 hours [18].

The methods of analysis were those recommended by the French and European standard (ISO 5667-3) and the guide to the documentation booklet (T-90-523-2) [18] to avoid any contamination. Two milliliters of nitric acid were added to each sample to avoid any precipitation and or metal absorption on the walls of the bottles. For each sample the temperature and pH were measured in situ using a Multi Parameter Instrument (MPI). Table 2 summarizes the methods used to analyze the physicochemical parameters.

Table 2. Method of analysis of the physicochemical parameters studied

Physico chemical parameter	Analysis method	Analysis instrument	Limit values
pH	pH-Meter	Multi Parameter Instrument (Hanna)	6.5-8.5
T	Thermometer	Multi Parameter Instrument (Hanna)	30 °C
SS	Effluent filtration	Electronic scale BS-1105	40 mg/L
BOD_5	Dilution	DBO Meter	40 mg/L
COD	Spectrometry	Pre-measured tube	130 mg/L

2.3. Statistical Analysis

Significant differences were established according to Tukey's test ($p < 0.05$) for the various comparison between the variations in the monthly means of the different parameters studied. Statistical analysis was performed using STATISTICA software (version 12). The PEARSON Correlation Matrix is used for determination of the correlation coefficients between the heavy metals analyzed in the discharge water of the various sampling points and the physicochemical parameters. The Principal Component Analysis (PCA) method was used.

3. RESULTS AND DISCUSSION

The estimation of industrial pollution is a complex and challenging problem that requires the determination and testing of different parameters to characterize the level of pollution in effluents in a comprehensive and relevant manner. For most sampling series, the physicochemical and metallic parameters of the discharge water vary significantly from one site to another.

3.1. Monthly Variation in Physicochemical Contamination of Wastewater

Figures 2-6 show the results of the monthly variations in the physicochemical parameters of the discharge water taken from the four sites (E1, E2, E3 and E4) of the industrial complex.

3.1.1. The Variation of the Temperature and *pH*

The temperature as well as *pH* values are high, exceeding the national standard [19]. The values of the monthly averages of the temperature of industrial discharge water vary from 30.33 °C in February to 45.66 °C in April, in site E1, and from 18.7 °C to 34.23 °C in sites E2, E3 and E4 (Figure 2). The minimum values are observed in March at both sites E3 and E4, while the maximum values are recorded in April at site E1 and in June at sites E2 and E3.

The minimum temperature recorded is due to the gradual cooling in contact with the air temperature, with the industrial effluents being discharged into an open-air system. The maximum temperature recorded can be explained by the influence of the process on the temperature increase [20]. Higher water temperature increases primary production and the risk of dystrophic crises, which influences the quantity of dissolved dioxygen in the water [17]. The water temperature varies according to the outside air temperature, insolation and the time of sampling. Temperature increases can kill some species, but can also promote the development of other organisms, thus causing ecological imbalance.

The *pH* values of the various recorded sites range from 6.9 to 10.03 (Figure 3). On the other hand, there is a significant increase in the *pH* at site E1 during all the months (from 9.73 to 10.26) and this comes back to the boiler service water, which must have a basic *pH* to avoid clogging of the tubes and perforation of the enclosure. *pH* conditions a large number of physicochemical equilibria and depends on multiple factors such as temperature and water origin.

The *pH* value reflects the stability of the equilibrium established by the various forms of a weak acid (H_2CO_3) and is associated with the system developed by the anions HCO_3^- (bicarbonate) and CO_3^{2-} (carbonate) [21]. Fluctuation in effluent *pH* is explained by chemical consumption at the site level. Wastewater sometimes has a high *pH* due to the use of caustic soda for washing equipment or preparing raw materials. The quantity and quality of the discharged water is also highly variable due to batch washing and rinsing operations. High or low *pH* levels in water affect aquatic life and alter the toxicity of other pollutants in another form.

3.1.2. Analysis of Suspended Solids

National and European standards set a limit value for *SS* in industrial liquid effluents a concentration of 40 mg/L for old installations, if applicable. The results of the physico-chemical analyses recorded a large pollution by suspended solids (*SS*) at sites E2 and E3 with a peak of 51.66 mg/L during the months of February and April (Figure 4). These results confirm the maximum values of *SS* recorded in the effluent from the Skikda industrial complex [22]. This increase in suspended solids is probably due to: rainwater that has run off on oily areas, water leaks from trains that run on oily areas, effluents related to the maintenance operations of facilities, effluents from boiler conservation, effluents from canteens (water from washing floors and appliances, vegetable washing, defrosting cold rooms, dishwashing water, emptying of cooking appliances), sanitary discharges, washbasins, showers in administrative buildings, in adding spills resulting from accidents during production operations [23].

All these discharges are mixed with industrial discharges in the same channel (E2), which then passes without prior treatment into the channel (E3). The presence of these *SS* in the discharges can considerably compromise the functioning of the sewage treatment system. Furthermore, it can generate nuisances such as mud deposits and fouling of the receiving water beds. *SS* have harmful effects when they have a high level on the physico-chemical characteristics of water, this can lead to a reduction in water transparency, changes in turbidity and reduced light penetration, but they can be considered an important source of sustenance for fauna and flora when they meet the standard values. Suspended solids can cause sludge deposits which interfere with the maintenance of natural biological buildings and fouling of aquatic receiving waters which impede the biodegradation of micropollutants [22].

3.1.3. Analysis of BOD_5 and *COD*

The main effect of the release of biodegradable organic matter into the natural environment is the resulting consumption of oxygen. Low BOD_5 values may be the result of intensive chemical treatment, inhibiting any biological activity [24]. The *BOD* measurement is widely used for monitoring wastewater treatment plant discharges, as it gives an approximation of the biodegradable organic matter load. The *BOD* is attached directly to the DO, when the concentration of *BOD* is high, it is found that the reduction of available oxygen is fatal; it increases the mortality of aquatic species. The highest values of biochemical oxygen demand (BOD_5) were recorded at the E2 site with a value of 33.66 mg/L during the dry season (Figure 5), this may be due to the creation of conditions for the degradation of organic matter by microorganisms whose activity increases with warming water. These results confirm the results found by Mohamed [22] for BOD_5 values (28.2 mg/L) in the discharge water from the industrial complex in Skikda (LNG), and largely meet Algerian standards [19].

The chemical oxygen demand (*COD*) of the discharge water samples generally ranged from 25.66 mg/L to 309.66 mg/L over the study period. The *COD* value is an important indication with which to characterize the overall pollution of water by organic compounds. The *COD* measurement is an estimate of the biodegradable and non-biodegradable organic matter present in wastewater in colloidal or *SS* form. The values measured reached a concentration of 309.66 mg/L at site E2 during the month of April and a value of 242.33 mg/L in the month of February at site E4, far exceeding the Algerian standard [19] limited to 130 mg/L for old installations (Figure 6). The increase in *COD* can have an adverse effect on the quality of seawater and this affects aquatic life and fish [25]. Industrial effluent from the complex could be hazardous to the living environment if the level of organic matter is too high and irregular (E4). According to Sankpal [20], the average *COD* value was 1825 mg/L, which is above the limit allowed by (WHO) [26]. Negative impacts are observed on the marine environment (E4) such as deoxygenation of the environment caused by the input of organic matter as well as chemical pollution related to mineral and organic micropollutants.

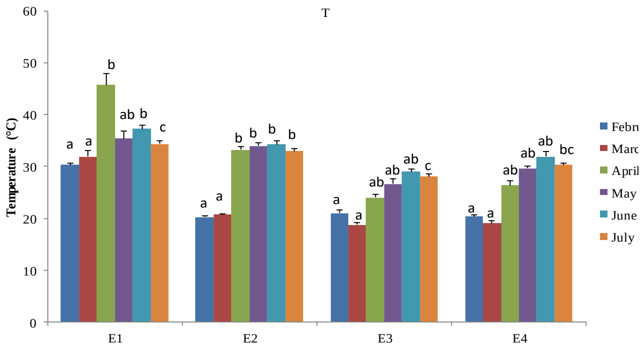

Figure 2. Monthly variations of the temperature at sites (E1, E2, E3, E4)

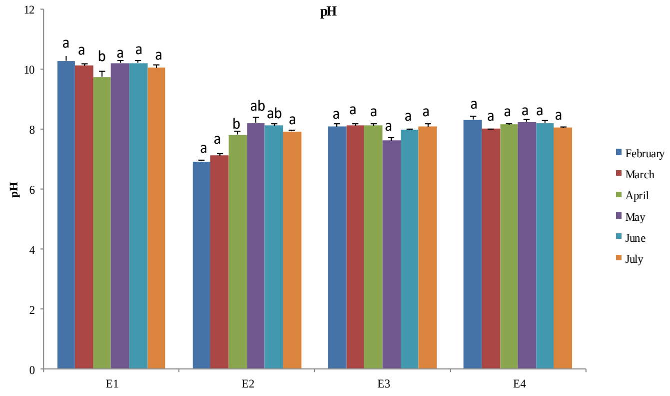

Figure 3. Monthly variations of *pH* at sites (E1, E2, E3, E4)

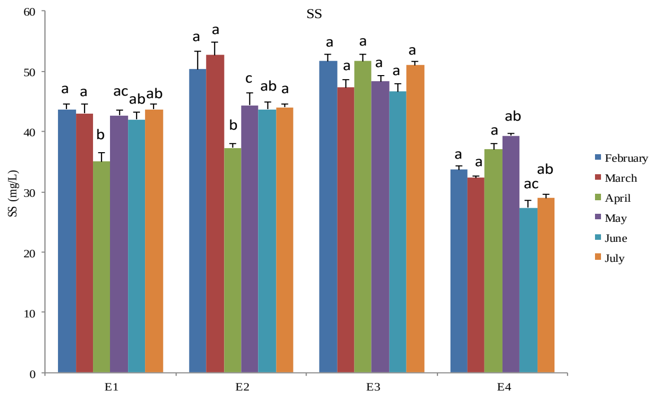

Figure 4. Monthly variations of suspended solids at sites (E1, E2, E3, E4)

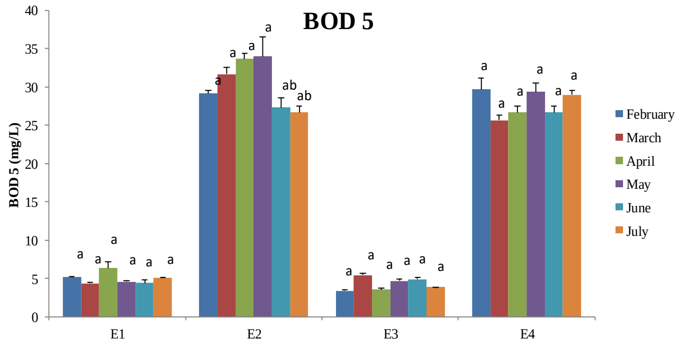

Figure 5. Monthly variations of the BOD_5 at sites (E1, E2, E3, E4)

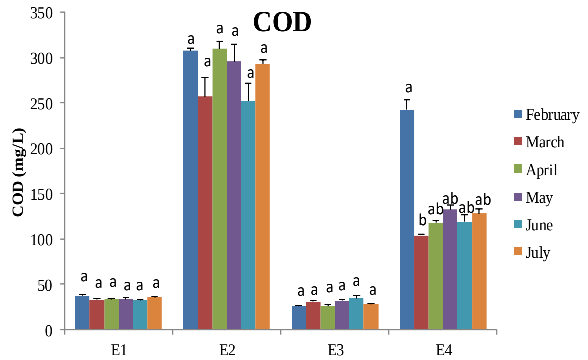

Figure 6. Monthly variations of the *COD* at sites (E1, E2, E3, E4)

3.2. Monthly Variation in Metallic Contamination of Wastewater

Heavy metals have the capacity to concentrate along the food chain and bind to various organs in the human body [15]. It is therefore necessary to reduce their concentrations in industrial effluents to acceptable levels compared to Algerian standards. The results of the different parameters are represented by the mean ± standard error (mean ± *SE*). In this study, the metallic analysis of the discharge water from the industrial complex focused on estimating the concentrations of 08 metals: Cd, Cu, Cr, Ni, Fe, Mn, Pb and Hg. Figure 7 shows the results of the six-month analysis averages.

The concentration of Iron in the analyzed wastewater ranged from 3.733 mg/L at site E1 during the month of February to a minimum of 0.001 mg/L at site E3 during the month of March. These recorded concentrations confirm the maximum iron values (26.47 mg/L) obtained by Mohamed [22]. The high iron concentration is due to maintenance work carried out before the boiler was commissioned, corrosion of the plant's pipes, soil leaching and the age of the complex, which dates back to 1981. As a result, there may be a risk of toxicity of this pollutant to aquatic fauna and flora.

The Manganese values recorded in the month of April considerably exceed the Algerian norm [19] for the four sites (E1, E2, E3 and E4), they vary between 1.5 mg/L and 1.646 mg/L. Manganese is an element that has the particularity of being present in several oxidation degrees from -3 to +7, which allows the existence of numerous inorganic and organometallic compounds that are produced either from ores or from metallic manganese [27]. The main inorganic compounds are manganese chloride, manganese sulphate, manganese tetroxide, manganese dioxide and potassium or sodium permanganate. Industrially, manganese is indispensable to the steel industry, mainly in the form of ferroalloys. As an alloying additive, manganese gives steel increased hardness and improves many of its mechanical properties. Non-ferrous alloys such as aluminum and some copper alloys also benefit from the addition of manganese to improve their properties [27]. In the workplace, manganese can be emitted in the form of powders, dusts or fumes, during machining, grinding or polishing operations, which emit particles, and during welding or cutting operations, which emit manganese fumes [27].

It was also noted that during the same month there was a significant increase in copper 0.463 mg/L. Although these results do not surpass the limit values set by Executive Decree 06-141 [19] (3 mg/L as limit value and 5 mg/L as extreme tolerance for old installations in operation). These values are explained by the corrosion of the installations. Nickel values recorded range from 0.001 mg/L (minimum value) to 0.851 mg/L (maximum value) with a mean value of 0.086 mg/L. Only site (E1) recorded an exceedance of the standard in April. This concentration is synonymous with the aggressiveness of seawater on the condenser alloys, especially at the start of the process.

Mercury values showed negligible variation over the study period; they ranged from 0.001 mg/L to 0.008 mg/L with a mean value of 0.003 mg/L. The values of mercury, chromium, lead and cadmium at the various sites are at trace levels. These values are in conformity with the directives of (Executive Decree, No. 06-141) [19]. The metals are classified in descending order of specific toxicity as follows: Hg < Cr < Pb < Cd < Cu < Ni < Fe < Mn.

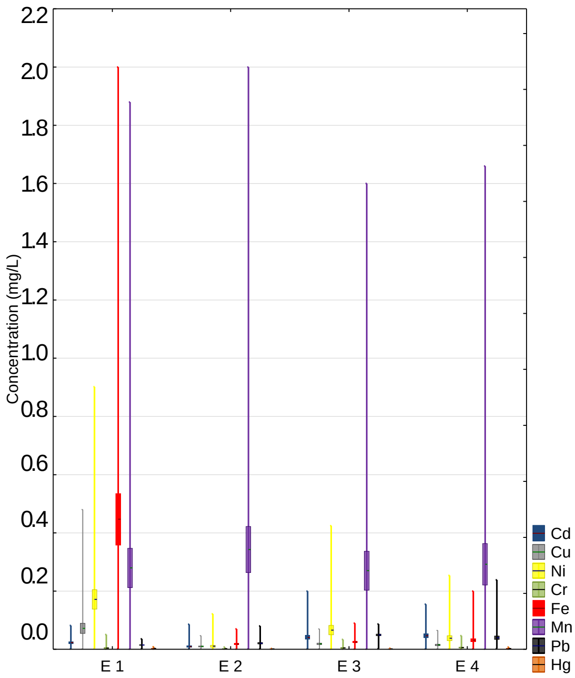

Figure 7. Box plot of six-month analysis averages of heavy metals at sites (E1, E2, E3, E4), (mean ± *SE*) (*n*=3)

3.3. PEARSON Correlation Matrix

Table 3 shows the correlation coefficients between the metals analyzed in the effluents of the different sampling points and their relation to the different physico-chemical parameters. The PEARSON matrix correlation showed high correlations between Cu and *pH*, as well as between Ni and temperature, *pH*, BOD_5, *COD* and Cd, showing a resemblance in their geo-chemical origin. In addition, some positive significant correlations between several pairs of metals have been found: Strong correlations were found in Fe with Cu and Cr concentrations (r = 0.77 and 0.29 respectively) at the significance level 0.01. Furthermore, moderate correlations were observed in Mn with Cd and Ni (r = 0.55 and 0.59 respectively), Pb with Cr (r = 0.39). Hg with Ni (r = 0.36), Ni with Cd (r = 0.36) at the significance level 0.05. This indication shows a high concentration of a metal at a local site, which is probably due to contamination. However, it does not necessarily mean that metal values are high elsewhere. It also reflects different sources of bio-geochemical behavior [28].

Table 3. Correlation matrix between metals and physicochemical parameters analyzed, the bold entries highlight the statistically significant correlations, ** ($p < 0.01$), * ($p < 0.05$)

	T	*pH*	*SS*	*BOD*$_5$	*DCO*	Cd	Cu	Ni	Cr	Fe	Mn	Pb	Hg
T	1.00												
pH	0.59**	1.00											
SS	-0.29*	-0.17	1.00										
BOD$_5$	-0.14	-0.55*	-0.40*	1.00									
COD	-0.08	-0.54*	-0.08	0.88**	1.00								
Cd	0.04	-0.04	0.04	-0.07	-0.21	1.00							
Cu	0.07	0.32*	0.00	-0.20	-0.18	-0.13	1.00						
Ni	0.40*	0.32*	-0.09	-0.27*	-0.27*	0.36*	-0.07	1.00					
Cr	0.00	0.00	-0.10	-0.02	-0.09	-0.07	0.42*	-0.15	1.00				
Fe	0.06	0.43*	0.04	-0.23*	-0.20	-0.11	0.77**	-0.08	0.29**	1.00			
Mn	0.23	-0.03	-0.15	0.06	0.05	0.55*	-0.08	0.59*	-0.10	-0.10	1.00		
Pb	-0.10	-0.22	-0.08	-0.05	-0.15	0.13	-0.06	-0.02	0.39*	-0.14	0.06	1.00	
Hg	0.33*	0.27*	-0.35*	-0.08	-0.23*	0.12	-0.07	0.36*	-0.09	-0.08	0.19	-0.17	1.00

3.4. Principal Component Analysis (PCA)

To further investigate the results obtained in our study, a correlation principal component analysis (PCA) was performed by applying as variables: average concentrations of *T*, *pH*, *SS*, *BOD*$_5$, *COD*, Cd, Cu, Cr, Ni, Fe, Mn, Pb and Hg of the sampled industrial wastewater, in order to detect any different level of contamination between the sites in the study area. Three principal components with eigenvalues >1 was extracted, representing 67.3% of the total variance. The first principal component (PC1) accounted for 31.8% and was mainly associated (factor loadings >0.7) with pH, while the second principal component (PC2) was clearly associated with *BOD*$_5$ and *COD* accounting for 23.4% of the data variability (Figure 8).

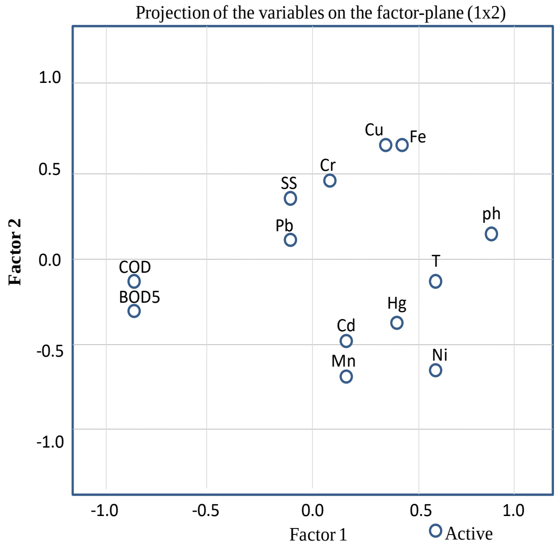

Figure 8. PCA representing the inter-site correlation according to the studied parameters

4. CONCLUSION

This study presented an ecological approach to the quality of wastewater effluent, in which the public authorities set an example by taking the necessary steps to protect the environment properly during the design, installation and operation of the various industrial complexes.

The monitoring of the results of six months of sampling revealed a slight metal contamination that exceeds the thresholds recommended by national and European legislation. In addition, the results of the physicochemical analyses recorded a high level of pollution by suspended solids. The two parameters of temperature and *pH* recorded high values that exceed the national standard.

The main cause of these high concentrations is the lack of prior treatment of wastewater and the absence or malfunctioning of treatment plants. Thus, it should help to raise awareness and take urgent and effective decisions to save coastal waters through the strict application of Executive Decree No. 07-300 of 27 September 2007 setting the terms of application of the additional tax on industrial wastewater and the possibility of increasing this tax to encourage industrial units to install an autonomous pre-treatment system.

Consequently, the preservation of aquatic ecosystems becomes imperative in the face of their degradation and requires the establishment of wastewater stations and the creation of regulated discharges for the cities of western Algeria. To this end, it is recommended that treatment plants be installed, regularly monitored and properly maintained to protect the Algerian coastline in general and the various industrial complexes in particular. The renovation of the damaged equipment of the complexes' industrial waste treatment plants is necessary and useful to reconcile industrial development with the protection of the marine environment, which is a common heritage to be preserved within the framework of sustainable development.

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