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Leachate Monitoring and an Assessment of Groundwater Pollution from the Tangier Landfill

Abstract: Leachate from public landfill in the city of Tangier, which is neither collected nor treated, could constitute a probable source of pollution of the groundwater table and of those of the Mlalah and Khandak Bou Hajjar wadis in the east, and the Mghogha and Ghir Boudra wadis to the south. This work aims to analyze the level of contamination at Tangier's municipal waste disposal. The leachate samples were collected and analyzed during the period from 2016 to 2019 and the physicochemical parameters (humidity, pH, organic matter, etc.) were determined on these samples as well as the contents of five heavy metals (lead, cadmium, iron, chrome and zinc). Analysis of the well water shows the presence of polluting elements in the leachate water and a high concentration of metals (especially iron) that exceeds standards. This makes the effluent extremely toxic and thus presents a permanent threat to the health of the local population and the surrounding environment.

Keywords: leachate, pollution, groundwater, physicochemical, heavy metals

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1. Introduction

Morocco's annual generation of solid household waste is estimated to be over 6 million tons or 0.76 kg per resident each day. In addition, 18,000 tons of home waste are produced daily, and 800,000 tons of industrial waste are dumped every year. The average ratios of waste generation per person differ in rural and urban locations, and are impacted by a number of factors, including the population's living standards, the area's uniqueness, and consumption habits. The weakest link in the waste management network is landfills or trash storage centers. However, Morocco, like other modern nations, is committed to environmental protection. Law No. 28-00 on waste management and disposal is a significant step forward towards this goal.

Because it is the most extensively utilized waste disposal technology worldwide, it is simple to apply and relatively inexpensive, a large volume of waste is still landfilled [1]. This poses a serious and long-term threat to the environment [2-7].

These are mostly unmanaged and open-air landfills in underdeveloped countries, where all forms of garbage (urban, industrial, hospital, and agricultural) are dumped in a raw and combined state. Different sorts of trash exist. In general, waste can be classified by its producer (household, industrial, or agricultural waste), the method of collection (municipal collection, voluntary contributions to garbage collection centers or selected sorting stations, etc.), or the future of the waste (landfill, incineration, recycling, etc.).

The dissolution of contaminating materials in the percolation fluids by means of physicochemical and biological processes produces liquid effluent rich in organic and mineral matter called leachate or often "discharge juice" as a result of the waste's fermentation and contact with rainwater.

Leachate is becoming better recognized as a significant source of groundwater pollution. It has a complex nature; containing high concentrations of conventional, unconventional, and hazardous chemicals such as BOD (biological oxygen demand), COD (chemical oxygen demand). Their estimations are useful in detecting toxic conditions and the presence of non-biodegradable substances [8], and so-called hazardous chemicals such as heavy metals and many chemical compounds that can seriously pollute the environment.

Due to the variety of garbage that the landfill receives, there is a risk of leachate toxicity. The infiltration of these pollutants into water tables or their flow into streams can lead to the insidious degradation of groundwater and surface water.

Several researchers have looked into the potential of landfill leachate to pollute groundwater. Since 1960, some authors [9] have demonstrated that landfill pollution of the water table is almost undetectable, while others [10] have demonstrated the presence of a real danger in a number of landfills studied in Wales and Canada. Landfill leachates cause enormous harm when they are released into the environment without proper treatment; they become a source of contamination for the surrounding ecosystem [11].

2. Study Area

The public dump of Tangier was put into operation in the early 1970s. It covers 30 ha and is located 5 km from the city center on the way to Tetouan. The location's latitude and longitude are $35^{\circ}44'35.32''N$ and $5^{\circ}45'17.39''S$ respectively. The site is part of the State's private domain. Charf Mghogha, Souani, Tangier Médina, and Beni Makada, as well as garbage from industrial zones, are served.

2.1. Hydrographic Network

The hydraulic network is composed of two wadis around the landfill: Mghogha and M'laleh (Fig. 1).

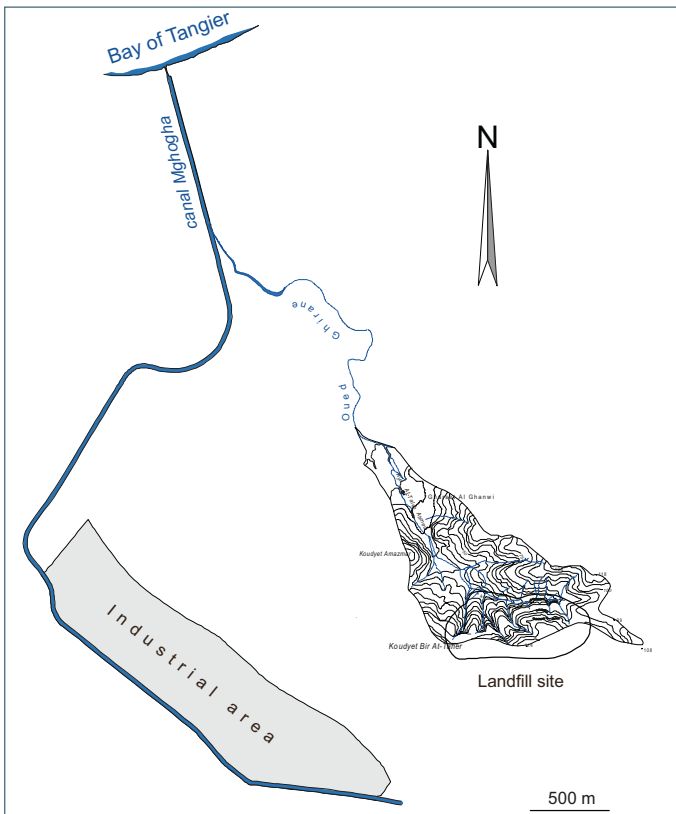


Fig. 1. Hydrographic network of the study area

Surface water and leachate flowing on the surface in the Tangier landfill area flow into Wadi Ayn At-Taleb Ahmed of Wadi Mghoga, which empties into the Bay of Tangier in the immediate vicinity of the Sanaa Beach residential district. Surface water and leachate can seep underground through faults. Since there is no significant

aquifer, the risk of groundwater pollution is low. However, small pockets of water which feed the wells around the landfill can be polluted. West of the landfill, approximately in the middle of the hillside, is a suspended water table. The limit of this is formed by an overlapping front with the Melloussa water table. In the southeast, a few sources of water supply limited fields. In addition, the valleys of Mghogha and the plain of Saha to the north-west of the landfill are cultivated on alluvial soils.

2.2. Field Geology

The landfill is located on the hills that form the eastern limit of the area where the Tanger unit outcrops. The unit is characterized by a vast marly schist or clayey schist complex of the Upper Cretaceous (Fig. 2).

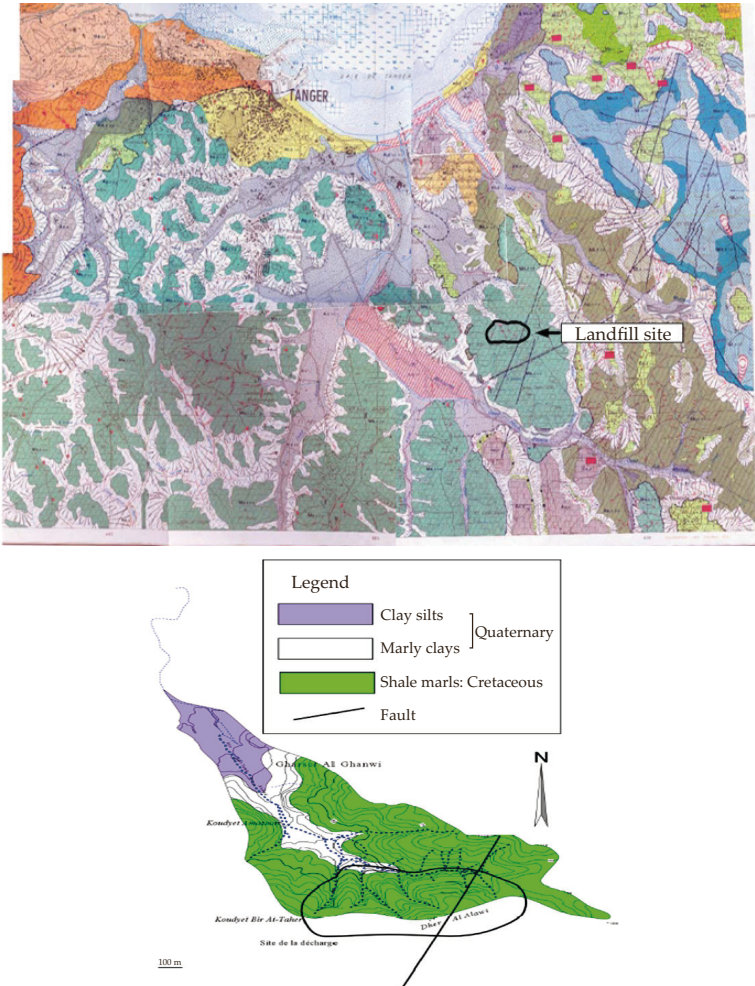


Fig. 2. Geological map of the study area

The hills are surrounded by the valleys of wadi Moghogha and Ghir Boudra to the south and those of wadis Mlaleh and Khandak Bou Hajjar to the east. The marl-schist formations which predominate in the facies of the site are characterized by their impermeability. The soil around the landfill is mostly clay. The geological map indicates two faults at the landfill site; at least one of these two crosses the landfill site.

2.3. Climate

By its proximity to the Strait of Gibraltar and its opening onto two maritime facades, Tangier has a subhumid climate with a mild tendency to the marine type.

2.4. Pluviometry

In the study area, rainfall is torrential and irregular (Fig. 3). Rainfall is high during the winter period and rare during the hot summer period.

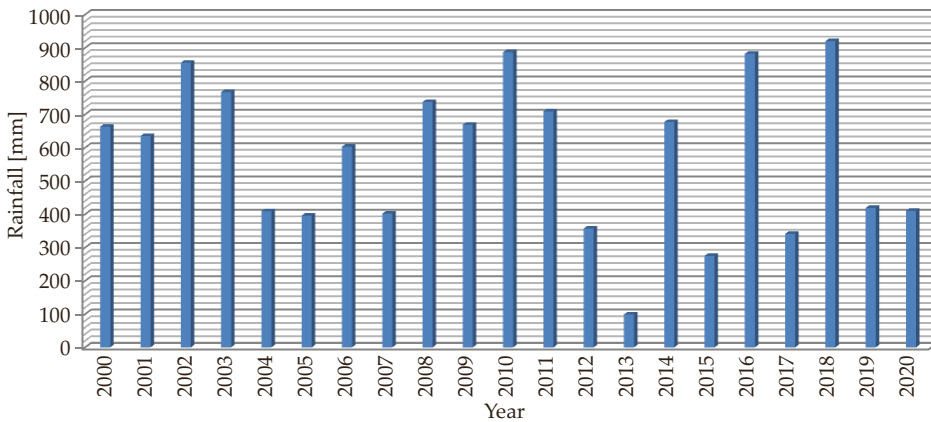


Fig. 3. Annual rainfall in Tangier over the period 2000–2020

The average inter-annual rainfall is 900 mm. Thanks to the variation in altitudes in the region; the rainfall is distributed between 1800 mm in the high reliefs and 600 mm in the plains [12]. The very high annual quantities of rainwater have an unfavorable effect on the formation of leachate. Due to the annual distribution of precipitation, significant amounts are to be expected between fall and spring.

3. Characterization of the Leachate from the Tangier Landfill

3.1. Sampling and Analysis of Leachate

To assess the risk of groundwater contamination by leachate from the public landfill in the city of Tangier, the leachate samples were collected and analyzed during the period from 2016 to 2019. Since the landfill is an undeveloped open-air landfill, it has neither a base coating nor a leachate collection and treatment system.

Thus, all the leachate generated ends up in the surrounding environment. Since the landfill is not equipped with any leachate collector, so we respected the leachate flow direction as much as possible (Fig. 4).

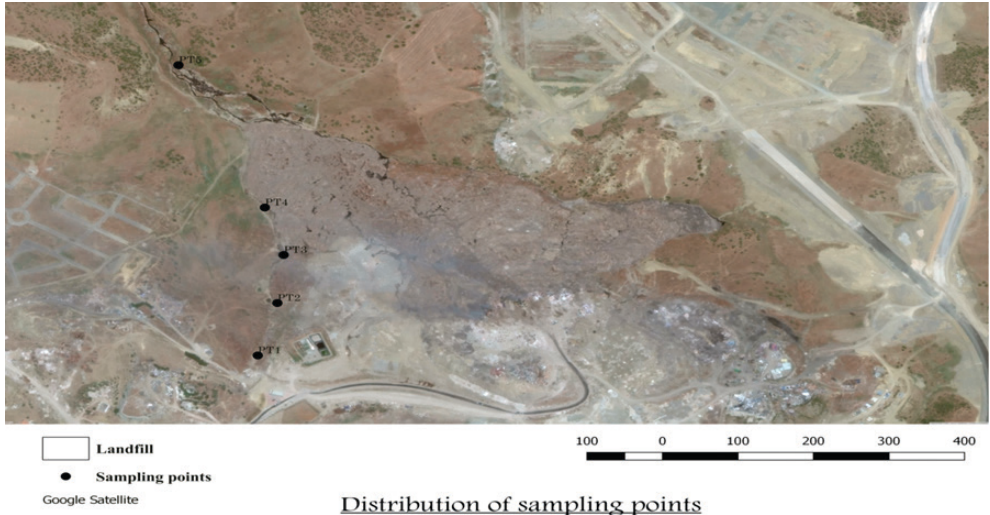


Fig. 4. Distribution of sampling points and leachate flow

Samples were taken in polyethylene bottles in the field. Before the bottles were filled, they were washed and then filled to the brim, with the cap screwed on to avoid any gas exchange with the atmosphere. Leachate samples were stored in a cooling device at 4°C during transport to the laboratory, with conductivity, pH, dissolved oxygen and temperature were measured in situ using a multiparameter device. HACH mark HQ40d to avoid the release of samples. The physical parameters tested include: color, conductivity and temperature. The chemical parameters were analyzed: pH and dissolved oxygen (DO). According to NM ISO 10523 and ISO 5814 respectively; nitrates, nitrites, chemical oxygen demand (COD), ammonium ions, sulfides and nitrogen were analyzed using the LCK, HACH LANGE rapid method.

3.2. Results and Discussion

The features of leachate are difficult to predict precisely because they change over time. Therefore, a physicochemical study was carried out during the period of 2016–2019 (Tab. 1, Figs. 5, 6).

pH

The pH in the leachate is considered to be one of the most important parameters affecting leachate concentrations in household and similar waste landfills.

The leachate from the Tangier landfill is basic. Recorded leachate pH values range from 7.97 to 8.58 (Fig. 5); similar results were found by [13]. The evolution of the pH during this monitoring shows an increase in pH as a function of the aging of the discharge. The pH values obtained in the leachate could be related to the low concentration of volatile organic compounds.

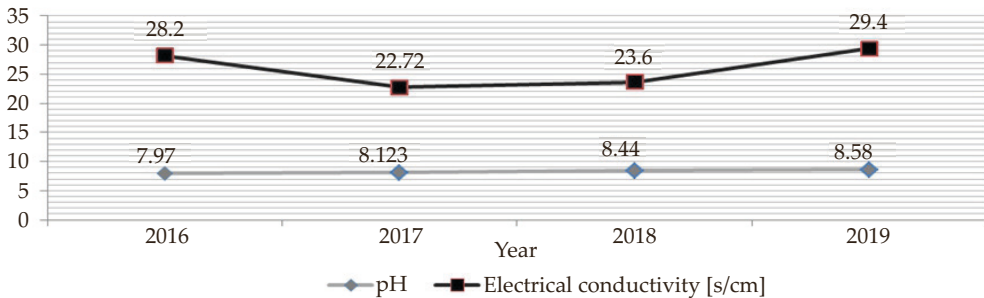


Fig. 5. Evolution of pH and electrical conductivity during the period 2016–2019

Electrical Conductivity

Electrical conductivity (EC) reflects the mineralization of the analyzed sample. It allows the evaluation of the overall mineralization and estimation of the total water-soluble salts [14]. The limit value for direct release into the receiving environment is 2700 $\mu\text{s}/\text{cm}$ [15].

The electrical conductivity values recorded during this monitoring fluctuate between a minimum of 22.72 s/cm recorded in 2017 and a maximum of 29.4 s/cm recorded in 2019 (Fig. 5). These leachates have high conductivity values, a characteristic common to all household refuse dumps [15–18]. The average value is 25.99 s/cm. The increasing evolution of EC over the next four years can be explained by the mineralizing activity of the bacteria.

Chemical Oxygen Demand

Chemical Oxygen Demand (COD) represents the amount of oxygen consumed by chemically oxidizable materials contained in water. It is representative of the majority of organic compounds and oxidizable mineral salts [19]. The recorded levels for O^2 are between 3515 mg/L and 1559 mg/L for the years 2016 and 2019 respectively (Fig. 6).

This content is ten times greater than the reference value (300 mg/L). The results obtained are higher than those reported by [20] in the Akouedo landfill in Abidjan (310–2495 mg/L), and by [21] in the Tiaret landfill in Algeria (1048 mg/L), and also by [22]. In the Agadir landfill (6220–7640 mg/L). However, they are much lower than those obtained by [1] in the Oujda landfill in Morocco (68,036 mg/L). Also, they are much lower than those of the Mohammedia landfill (51,456 mg/L) [23].

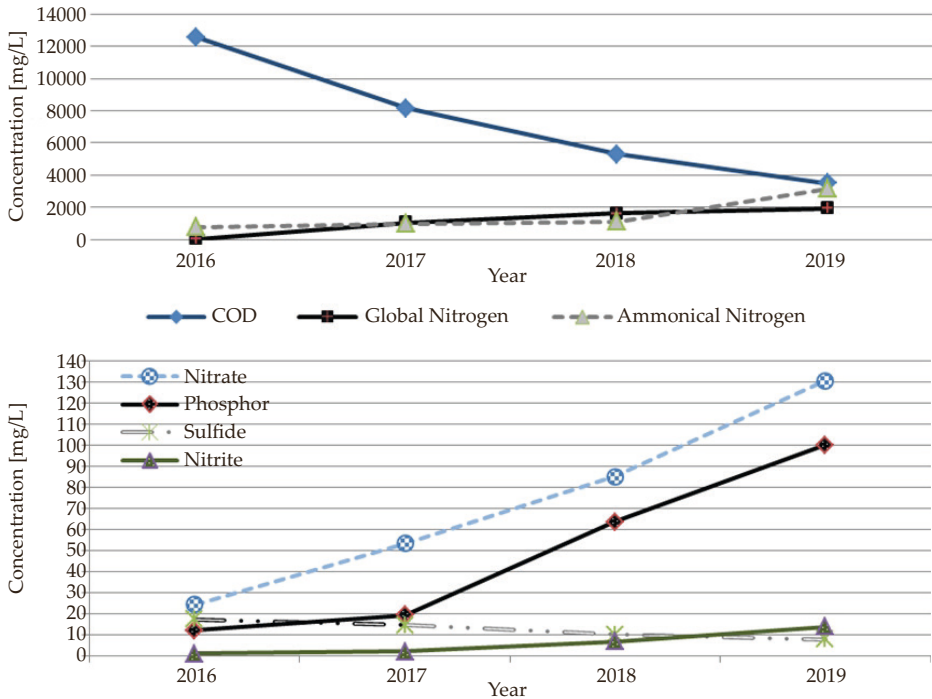


Fig. 6. Evolution of physicochemical parameters during the period 2016–2019

Table 1. Evolution of leachate characterization in the Tangier landfill during the period 2016–2019

Element	Concentration [mg/L]			
	2016	2017	2018	2019
Nitrate	24.34	53.14	85.08	130.6
Phosphorus	12.014	19.19	63.58	99.94
Sulfide	17.23	14.46	9.89	7.84
Nitrite	1.05	2.088	6.69	13.64
COD	12.559	8157.4	5297.6	3515.0
Global nitrogen	40.44	1060.8	1643.4	1957.0
Ammoniacal nitrogen	780.0	990.0	1119.4	3164.0

Nitrates

The average nitrate values presented on the histogram for the leachate from the Tangier landfill, the nitrate values evolve expansively from 2016 to 2019 and reached an extreme value of 130.6 mg/L in 2019 (Fig. 6). This evolution of nitrate is mainly due to a strong bacterial denitrification by a mineralization of the ammoniacal nitrogen in nitrates which confirms the studies carried out by [16], especially since the temperature conditions are very favorable for the development of this phenomenon and which is still proved by [16].

The low concentrations of nitrites recorded are probably due to the reduction of nitrates to nitrites, which can lead to unpleasant odors as reported by [24]. Nitrite values were relatively lower (1.5–13.5 mg/L) (Fig. 6) because they are generally not present in significant concentrations except in an oxygen-stressed environment, since nitrate is the most stable oxidation state.

Ammoniacal Nitrogen

Ammoniacal nitrogen constitutes one of the links in the complex nitrogen cycle in its primitive state. It is a gas which is soluble in water and this chemical element is the main reducing agent in landfill leachate and a significant long-term pollutant [15].

Ammoniacal nitrogen can have an environmental impact, and it is known to be one of the main toxins for living organisms. The ammonia concentrations observed ranged from 780 mg/L to 3164 mg/L with an average value of 990 mg/L (Fig. 6). At this concentration, methanogen is only slightly inhibited by ammonia, this chemical is the main reducing agent of landfill leachate and is a long-term pollutant [17] of mean values of concentrations of ammonia (600 mg/L) lower than those reported in the present study were obtained by [25].

Sulphides

Sulphate values of 98–374 mg/L and 22–650 mg/L have previously been recorded by other researchers [26]. Typical sulphate values of 300 mg/L and 20–50 mg/L have been recorded for new (less than years) and mature (more than 10 years) discharges respectively [27]. However, the measured sulphate values exceeded the permissible level (0.50 mg/L). The sulphate content of the leachate depends mainly on the decomposition of the organic matter present in the solid waste and this is expected to decrease with the age of the waste. This decrease is caused by the reduction of sulphate to sulphide coinciding with the onset of anaerobic conditions in the landfill [28]. Thus, the sulphate concentration in the leachate can also be used as an indicator of the stabilization of the waste in the landfill. The levels recorded fluctuate between 7.84–17.23 mg/L (Fig. 6) these are common values for old mature landfills. The low sulphide contents can be explained by the low reducing activity.

These ions are among the gases responsible for the bad odors given off by landfills [14]. For new and mature landfills, the sulfate values are 300 mg/L and 20–50 mg/L, respectively [27]. Other higher values (3056 mg/L and 1850 mg/L) have been published by [29].

Phosphorus

Phosphorus is an important indicator of water pollution because it is easily and quickly absorbed by biota and is therefore almost never found in high concentrations in unpolluted waters. For the years 2016–2019, phosphorus values ranged 12.014–99.94 mg/L (Fig 6). These high levels can be attributed to the organic load of the waste which contains phosphorus; this organic matter (mainly phospholipids and phosphoproteins), during its biodegradation release phosphorus and therefore increase the phosphate concentrations.

Total Kjeldahl Nitrogen (TKN)

Regardless of the importance of nitrogen in landfill emissions, nitrogen content has received significantly less attention in landfill studies than methanization of waste. No limitation has been imposed on the nitrogen content of MSWs from mechanical-biological pretreatments which are currently carried out with the aim of reducing methane emissions from MSW landfill [27]. On the other hand, in methanization studies, nitrogen concentrations are sometimes reported for the digestate, due to the possible inhibitory effects of ammonia and the effect of ammonia concentrations on the quality of the final product. The TKN content ranges from 40.5 mg/L to 1957 mg/L (Fig. 6).

Unfortunately, due to the wide range of leachate quality, projecting the leachate's characteristics over time has proven problematic. Although general quality trends are possible, these ranges remain crucial, and it is still impossible to anticipate when each phase begins and finishes. Current landfill research, such as the use of leachate recirculation, shows that it is possible to control waste decomposition and hence make the leachate characteristics more predictable.

The content of the solid waste, the interaction of the leachate with the environment, the age of the trash, sampling protocols, and the design and operation of the landfill are all factors that influence the quality of the leachate. To account for at least one of these factors that cause changes in leachate quality, data for each backfill was separated based on when (and if) each landfill switched from the acidogenic to the methanogenic phase.

The general trend between the composition of the leachate and the age of the landfill has been shown to decrease the toxicity of the leachate somewhat over time. Authors of [30] and [31] postulated that the waste stabilization rate depends on the movement of water through the backfill, which in turn is influenced by seepage. Therefore, leachate stabilization will not necessarily be directly related to chronological age. Natural leachate was very toxic within landfills and generally remained so even at the point of discharge from the stream, although it was somewhat attenuated over time due to the considerable dilution which allowed for surface drainage and groundwater.

Heavy Metals

Metal pollution from landfill waste is a long-term issue that poses several issues, particularly with its release. According to the literature, the metal levels measured

in the leachate are fairly low, and the bulk of heavy metals, primarily copper, nickel, lead, iron, zinc, and cadmium, stay trapped inside the waste pile. According to [32], more than 99.9% of heavy metals are still trapped in landfills after 30 years.

Heavy metals are found naturally in the environment and are also used in industry. They can, however, inflict more or less substantial harm to humans, wildlife, and vegetation at higher than typical quantities. To study a possible contamination of leachate by heavy metals, the concentrations of these elements were determined. Table 2 and Figure 7 show the distribution of the contents of the different elements. The iron concentration recorded a rate of 70 mg/L and 25 mg/L for the years 2016 and 2019 respectively. These numbers are quite far from the standards of discharge and thus irrigation. The lead concentrations mark numbers that fluctuate between 2.2 mg/L and 0.8 mg/L for the years 2016 and 2019. These concentrations do not exceed the irrigation standard which is 5 mg/L. Analysis of the metal composition of Zn recorded during the period 2016–2019 are 12 mg/L and 5 mg/L respectively; these high concentrations of Zn can be attributed to the disposal of large amounts of industrial waste in landfills. The chromium content in the leaching water from the Tangier landfill greatly exceeds the standards for both: discharge (0.2 mg/L) and irrigation (0.1 mg/L).

Table 2. The evolution of heavy metals in leachate from Tangier landfill

Element	Concentration [mg/L]			
	2016	2019	rejection standard	irrigation standard
Iron	72.5	23.0	2.5	5.0
Lead	2.19	0.73	1.0	5.0
Zinc	12.5	4.5	5.0	2.0
Chrome	0.895	0.06	0.25	0.12
Cadmium	0.129	0.02	0.2	0.015

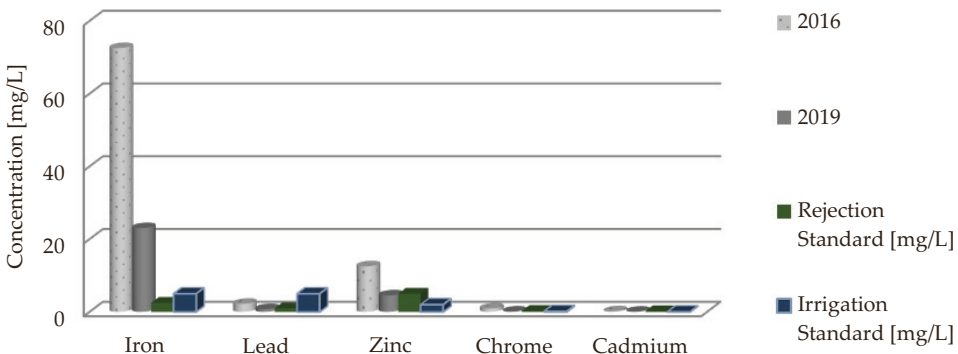


Fig. 7. The evolution of heavy metals in the Tangier landfill

We also noticed a very high concentration of this element, which could be linked to tannery waste, while chromium in the landfill could be linked to, among other things, pneumatic waste at the landfill. This chrome pollution comes from waste produced by industrial units without any prior treatment.

Cadmium has more or less low concentration (0.13 mg/L and 0.02 mg/L) and do not exceed discharge or irrigation standards. The cadmium content could be linked to the existence of an electric cell production plant in the industrial zone and/or to domestic discharges.

The presence of a reasonably substantial metallic load is revealed by monitoring metallic contamination (iron, zinc, chromium, lead, and cadmium) in the Tangier landfill leachate. The large variation in heavy metal contents may be associated with the high heterogeneity of the waste. According to the results, iron and zinc present large quantities with concentration ranges of 72.5 mg/L and 23 mg/L for 2016 and 2019 respectively. They are followed by zinc, with average concentrations of 12.54 mg/L and 4.5 mg/L, and then lead and chromium, with cadmium in last place with low concentrations of 0.129 mg/L and 0.02 mg/L for 2016 and 2019 respectively.

A concentration of iron above the permissible limit in water causes aesthetic problems related to taste, odor and color. The high iron concentration is confirmed by [33, 34] and this metal is mainly present in the metallic materials of the landfill.

The high zinc value can be attributed to the presence of fluorescent tubes, batteries and a variety of food waste as well as burning tires at the site. Zinc is a more leachable element in fresh waste [35, 36].

The low concentration of lead and cadmium in the landfill leachate can be attributed to the underground geology of the site which is made up of clay. These metals have the affinity to be absorbed by clay soils [37, 38]. The disposal of dry cell batteries and paint cans are possible sources of cadmium.

4. Assessment of the Impact of Leachate on Groundwater

There are seven wells around the landfill (Fig. 8, Tab. 3), with wells 3, 4 and 6 located near local residents, and wells 3 and 4 used especially as a source of water for livestock in the described region. Well 2 is dedicated to the local population for daily domestic use. Well 6 is located near a house and its inhabitants use it to obtain drinking water.

Well water analyzes show that there is contamination probably caused by pollutants coming from the landfill and seeping into the water table. This water table is mainly intended for agricultural activity wells 3, 4 and 6 are used to supply drinking water to the population in the vicinity of these wells.

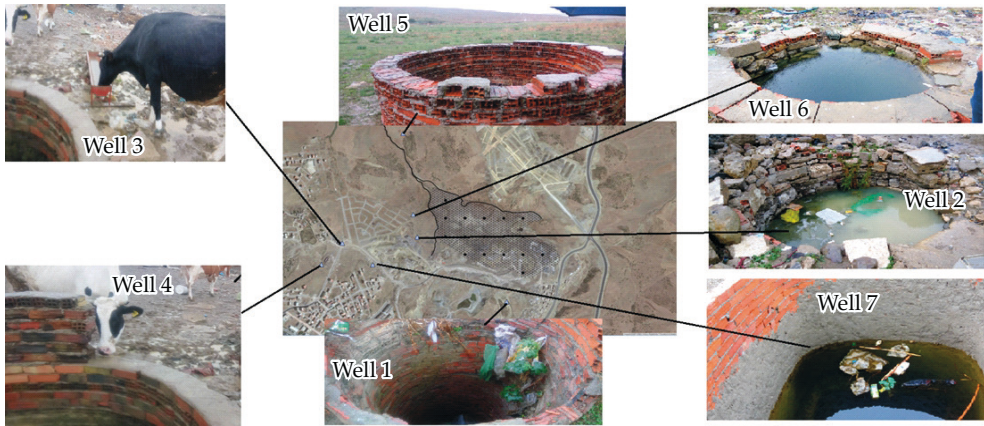


Fig. 8. Tangier landfill wells

Table 3. Discharge wells and their depths

No.	X	Y	Z	Depth [m]
Well 1	-5.76	35.74	53.46	1.65
Well 2	-5.76	35.75	22.08	3.6
Well 3	-5.76	35.75	44.11	1.4
Well 4	-5.77	35.74	47.77	-
Well 5	-5.77	35.74	38.79	-
Well 6	-5.77	35.74	51.11	1.32
Well 7	-5.76	35.74	79.28	2.87

4.1. Materials and Methods

Determination of Groundwater Flow Direction

Groundwater does not generally remain stationary, but moves or flows underground depending on the forces acting on it [39]. Piezometry is the depth measurement of the surface of the groundwater table. It is expressed either in relation to the ground in metres, or in relation to the zero altitude of the sea level in NGF (French General Levelling). The piezometric maps provide a better understanding of the hydrogeology and hydrology of the area.

In piezometric studies, the tools are varied and more precise than others, making it possible to make a precise study (measurement of latitudes, longitudes, altitudes and piezometric levels).

We can cite as material used for this fieldwork the following:

- a global positioning system GPS,
- a probe,
- a topographic map,
- computer equipment.

Reminder of the Characteristics to be analyzed

To study the quality of well water, a physicochemical analysis including pH, temperature, chloride and a characterization of the composition of metallic trace elements (iron, lead, nickel and cadmium) was carried out

4.2. Results

The piezometric levels generally follow the topography. The underground flow takes place from the high reliefs to the hollows.

Indeed, the general flow of the water table for this period in the same direction as the surface flow of water (southwest to northeast) and the curves remain parallel (Fig. 9).

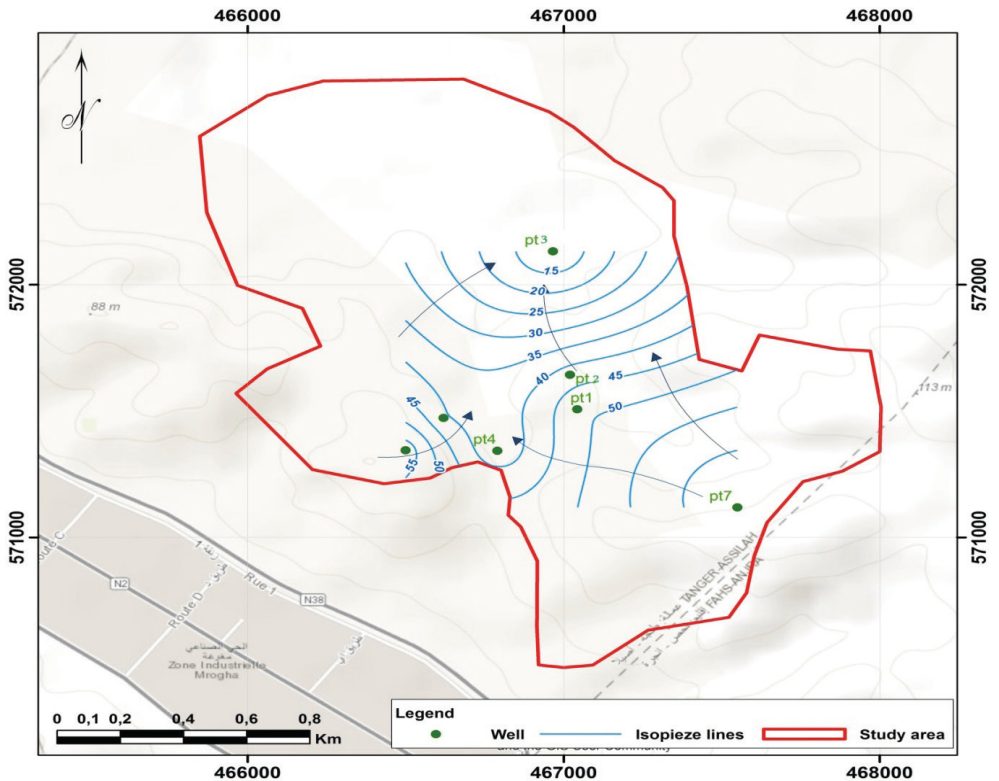


Fig. 9. Piezometric map of the study area

The piezometric lines are slightly tightened downstream of the zone; with a calculated hydraulic gradient of 0.7%. On the other hand, the hydraulic gradient calculated in the southern part remains large and is materialized by spaced isopiezium lines; it is equal to 1.68%.

The temperature ranges between 16.3°C and 20.1°C, pH shows that the water from the wells is slightly basic between 8.9 and 9.3 (Fig. 10), exceeds the standards intended for irrigation but not the standards of potable water.

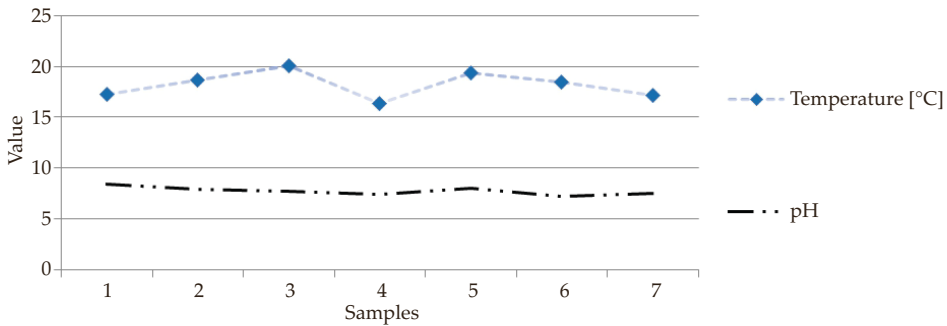


Fig. 10. Temperature and pH variation over time of the underground water from Tangier landfill

Chlorides are very concentrated in well water and exceed the standard set for irrigation water. These elements probably come from the infiltration of the leachate at depth, since the leachate water is very loaded with chlorides.

Water from wells 2, 3, and 6 are used as drinking water. Chlorides mark high levels which reach 1000 mg/L, 840 mg/L, and 779 mg/L in these wells 8 (Fig. 11), however, the standard for drinking water is 750 mg/L.

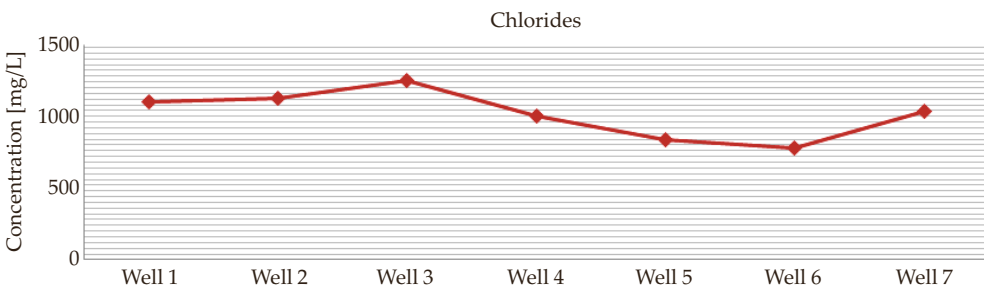


Fig. 11. Chloride levels in well water

The results obtained show that the chloride contents are very high (annual average = 1022 mg/L) and greatly exceed the standards of drinkability (250 mg/L) and the upper admissible limits (600 mg/L) set by the WHO. The chloride ion is a relatively

mobile element that readily migrates to the groundwater. It is unaffected by adsorption or ion exchange, does not interfere with acid-base or redox equilibrium, and is not maintained by soil clay-humic complexes. As a result, it is widely utilized as a reliable conservative tracer for demonstrating the impact of leachate on groundwater physicochemical quality [16, 40].

Groundwater pollution with heavy metals can occur as a result of natural sources or human activity; contamination from residential, municipal, commercial, industrial, and agricultural operations can all have an impact on groundwater quality [41].

Iron recorded high concentrations, the concentration of iron in water samples ranged 1.1–5.4 mg/L (Fig. 12). The presence of iron in water can cause groundwater to change color [42].

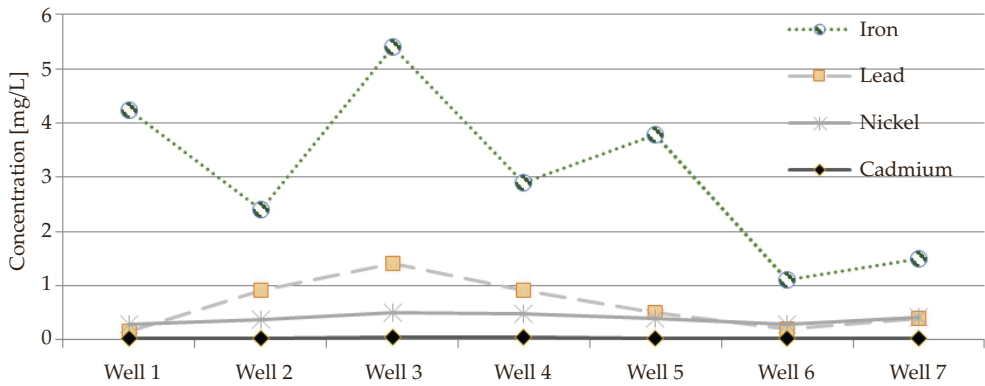


Fig. 12. Evolution of heavy metals from Tangier landfill groundwater

Nickel becomes more mobile and it can reach groundwater, its concentration fluctuates between 0.01–0.5 mg/L (Fig. 12). The fact that nickel is generally not found in groundwater or in very low quantities indicates that the presence of this metal is mainly related to human activities. Nickel is liable to cause corrosion in drinking water distribution circuits.

The cadmium content is of the order of 0.03 mg/L at the level of the wells, and this value greatly exceeds the cadmium standard for both irrigation water and potable water. When high levels of cadmium are encountered in groundwater, its origin must be sought in industrial effluent as cadmium can be washed away by rain from industrial sites.

Lead and cadmium remain somewhat constant but they increase in soil samples around these wells, a fact which could be explained by the fact that this element is retained at ground level.

The water sampled from the well located near the landfill site was found to be more contaminated than that from the well located further away. It obviously stems from the gravitational movement of the viscous fluid.

5. Conclusion

Leachate from municipal landfills represents a potential risk to the health of ecosystems in general and human populations in particular. From 2016 to 2019, this research focuses on the physicochemical evaluation of leachate and groundwater from the Tangier landfill (Northern Morocco). It made it possible to highlight the impact of the waste from the landfill and assess the degree of contamination on the environment.

The characterization of the leachate generated by the uncontrolled and open-air landfill of Tangier showed that it is old and stabilized leachate, carrying a significant mineral, organic and metallic pollutant load. These leachates with a high pollutant load pose a risk of contaminating the phreatic water table which circulates at shallow depths (about 20 m), under a moderately permeable substratum.

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