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Investigation of the hydrochemical quality of surface waters of the Oued Inaouen watershed near the Driss 1er dam (Morocco) using multiv*ariate statistical methods and diagram analysis*

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Abstract. This study, conducted from 2021 to 2023, aimed to determine the physico-chemical quality of the surface waters of the Oued Inaouen watershed near the Idriss 1st dam, and to identify existing pollutants and their origins. The research included spatio-temporal monitoring of 26 surface water stations.

Results showed a spatio-temporal differentiation influenced by natural and human factors. The right bank exhibited significant mineralization, while the left bank had soft water due to Middle Atlas liasic limestones and dolomites. Multivariate statistical methods and diagrams (Piper, Schoeller, and Wilcox) were used for hydrochemical classifications and water quality assessment.

Over time, surface waters became increasingly enriched with BOD_5, COD, NO^{3-}, and NO2-, particularly downstream where more discharges occur. Pollution is linked to untreated household and industrial waste (notably Olive Mill Waste) and agricultural runoff from fertilizers, with greater enrichment during summer and autumn due to low upstream flows.

Key words. Inaouen watershed, Surface water, physico-chemical quality, Piper's diagram, Wilcox's diagram, Schoeler's diagram, water quality.

1. Introduction

The hydrochemical quality of surface waters, essential for human health, the environment and socio-economic development, is threatened by pollution of both anthropogenic and natural origin, particularly in the Oued Inaouen watershed in Morocco. Industrial discharges, domestic effluents and the use of pesticides and fertilizers degrade the water, affecting aquatic ecosystems and the supply of clean water [1], [2]. Accurate assessment is needed to identify pollution sources, understand contaminant dispersion, and implement effective management strategies. Multivariate statistical analyses and hydrochemical diagrams are advanced methods used to assess and guide water resource protection policies[3], [4].
In Morocco, numerous international and national studies have been carried out to assess the physico-chemical quality of watercourses in different regions. These include studies in the Tadla [5], Khenifra (Oued Oum Er-Rabi) [6], Khemissat [7], Meknes, Oujda [8], (Oued Zegzel) [9], [10] and Marrakech's El Haouz [11]. These studies have highlighted growing pollution risks and their impact on surface water quality.
The results of this work have revealed significant environmental problems, underlining the urgency of addressing water pollution issues. The degradation of surface water quality, due to various sources of pollution, threatens not only aquatic ecosystems, but also public health and the economic development of the regions concerned.
The waters of the Oued Inaouen watershed are no exception to this pollution phenomenon. An in-depth physico-chemical study is therefore necessary to complete existing knowledge on the sources of pollution in this region. To this end, several physico-chemical parameters were measured, such as conductivity, total hardness, calcium and sodium ions, etc. These data are crucial to understanding the sources of pollution. In addition, the impact of effluent discharges from the city of Taza and surrounding rural centers on water quality was analyzed using indicators such as five-day biological oxygen demand (BOD_5), chemical oxygen demand (COD), nitrates, and chlorides [12]. These indicators are essential for assessing the state of water pollution and determining the measures needed to protect and improve water quality[13].
Finally, based on the principle that chemical elements can serve as natural tracers, they are used to reveal the chemical history of waters. This approach provides a better understanding of pollution processes and environmental dynamics. The data obtained can thus serve as a basis for sustainable and efficient management of water resources, while also offering avenues for reducing pollution and restoring aquatic ecosystems.
The aim of this study, to be carried out from 2021 to 2023, is to assess the physico-chemical quality of surface waters in the Oued Inaouen watershed upstream of the Idriss 1er dam, and to identify pollutants and their origins. By delimiting 26 monitoring stations, variations influenced by natural and anthropogenic factors were observed: the right bank shows strong mineralization and the left bank soft water. Statistical methods and hydrochemical diagrams were used to classify and assess water quality. The study revealed a progressive increase in BOD_5, COD, NO^{3-} and NO^{2-} downstream, mainly due to untreated household and industrial discharges and agricultural leaching, exacerbated in summer and autumn by low flows.

2. Materials and methods

2.1. Study area

The Inaouen catchment covers an approximate area of 3396 km² (Figure1), which represents 8.3% of the Sebou basin. The Mouloya watershed borders the Inaouen catchment to the east, the upper Ouergha watershed to the northwest, and the upper Sebou watershed's sub-catchment to the south. In addition to these geographic characteristics, the Inaouen watershed has two different types of landforms.

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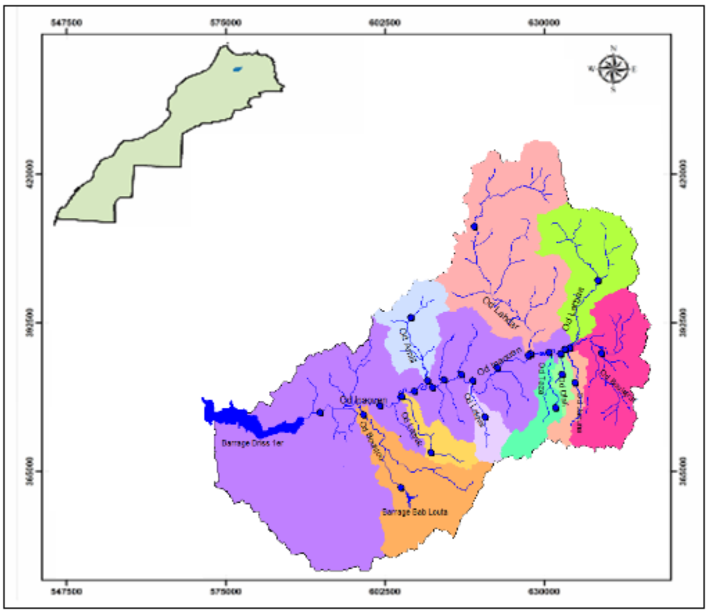

Fig1. Geographical location of the Inaouen watershed.

On the right bank of the river, the basin goes into the hills of the eastern pre-Rif, where water erosion is severe on unprotected land mostly made up of marl and marl-limestone rocks from the Cretaceous and late Tertiary periods [14]. On the left bank, the catchment area includes the northeastern Middle Atlas, characterized by rigid and permeable rocks like limestone and dolomite that are high in ammonite from the Lias period [15] (Figure 2). These geological features make runoff processes less effective, ultimately limiting water erosion and leading to steep landforms, with the highest point exceeding an altitude of 2000 meters. Uncontrolled human activity has significantly degraded the vegetation cover [16]. The region's climate is semi-arid, with hot summers and cold winters, and precipitation often occurs in the form of short-duration showers [17]. These rains enhance the speed of runoff, leading to flooding events [18]. The rivers have a torrential hydrological regime, characterized by sudden and abrupt floods in the fall and winter and low flows extending for several months [19].

The Taza region possesses significant water potential. Its geological structures have facilitated the formation of large aquifers in the highly karstified limestone and dolomite lithologies of the Lias period. Additionally, the karst springs sustain surface flow during periods of low flow. Ras El Mae, the most renowned spring in the region, provides drinking water [20].

2.2. Experimental protocol

Field trips organized by a certified expert from the National Office of Electricity and Drinking Water - Water Branch, are conducted to collect and analyze water samples. The sampling, transportation, and preservation of the water samples are carried out in accordance with the laboratory protocol for water quality control at ONEP [21], [22], [23], [24], [25], [26].

The hydrochemical analysis considered twenty parameters, of which five were measured in the field right after the device's calibration. We primarily conducted the work at the Laboratory of Natural Resources and Environment of the Polydisciplinary Faculty of Taza, utilizing four essential tools. We have used a volumetric device to identify multiple chemical parameters such as chlorides, calcium, and magnesium. Other instruments like molecular absorption spectrophotometry and flame spectrophotometry allowed us to determine nitrates, ammonium and sodium. We also used the OXITOP to figure out the BOD_5, and the COD was found by oxidizing materials with too much potassium dichromate at 148 °C with silver sulfate as a catalyst, mercury sulfate, [27].

3. Results and discussion

3.1. pH and electric conductivity

The pH values show that the water is slightly alkaline. This alkalinity is linked to the nature of the soils and rocks traversed, mainly formed of liasic dolomite. For the stations located on the Inaouen stream (O13, O14, O17 and O26), the alkalinization of their waters is the result of the use of CO_2 during photosynthesis, which is accompanied by the precipitation of insoluble carbonates [28].

A comparison of the values obtained in 2021-2022 and 2022-2023 revealed an alkalinization of the waters studied.

The electrical conductivity of water in the Inaouen watershed varies greatly between the left and right banks. On the right bank, the waters are characterized by high mineralization, with a maximum conductivity of 4059 µs/cm in 2021-2022 and 5077 µs/cm in 2022-2023. In contrast, the waters of the left bank tributaries show low mineralization, with a conductivity of 54.7 µs/cm in 2021-2022 and 94.9 µs/cm in 2022-2023. (Figure 2).

Water mineralization decreases during high-water periods. In contrast, the low-water period is characterized by an increase in conductivity. This may be due to the concentration of mineral salts in the water and the increase in alkalinity [29].

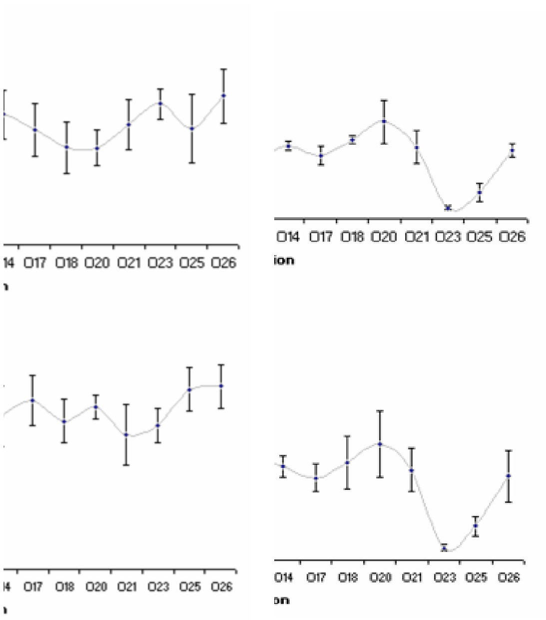

Fig 2. Spatial and temporal trends in the means and standard deviations of pH and electrical conductivity (µS/cm) of water analyzed over the 2021-2023 period.

Conductivity decreases progressively from upstream to downstream, and standard deviations are all the more significant the greater the initial mineralization. At station O1 in spring 2023, conductivity is 6690 µS /cm, which has only decreased at a distance of 45 km to 1976 µS /cm at station O17.

The map of the spatial distribution of conductivity (Figure 2) illustrates a significant variability in the chemical composition of the water, with values ranging from 54.7 to 6690µs/cm. This high level of mineralization is essentially linked to the nature of the terrain traversed.

3.2. Calcium and Magnesium

In terms of spatial evolution (Figure 3), water on the left bank is more concentrated in calcium than water on the right bank. However, maximum calcium concentrations on the right bank reached values of 138 mg/L and 77.6 mg/L in 2021-2022 and 2022-2023 respectively, whereas maximum concentrations on the left bank were 174.4 mg/L and 103 mg/L in 2021-2022 and 2022-2023 respectively. This variability in calcium levels is linked to the liasic carbonate formations of the Middle Atlas on the left bank and the Jurassic marl-limestone formations on the right bank.

In fact, calcium concentration is closely linked to the geological nature of the terrain through which the water flows. Calcium may be leached from salt layers (NaCl, Ca SO4), limestone and gypsum marl, or it may originate from the dissolution of organic matter in ditches, which favours the solubility of $CaCO_3$. A decrease in calcium ion content was noted from 2021-2022 to 2022-2023.

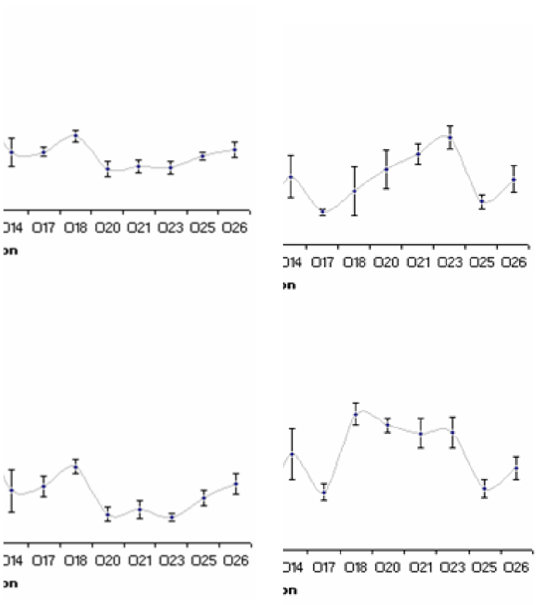

Fig 3. Spatial and temporal trends in mean values and standard deviations of calcium and magnesium levels (mg/L) in water analyzed over the period 2021 – 2023.

Mg^{2+} ion levels in Oued Inaouen watershed show irregular variations. Average values of 52.20 mg/L and 66.73 mg/L were recorded successively in the winters of 2021-2022 and 2022-2023 (Figure 3).

On the left bank of Oued Inaouen, calcium and magnesium dominate. This facies reflects the effect of the dissolution of Middle Atlas limestones and dolomites on the chemical composition of the water.

Mg^{2+} /Ca^{2+} ratio values fluctuate between 0.16 and 2.96, with an average of 1.75 at stations originating from the Middle Atlas. They illustrate the relative proportions of these two chemical elements in solution in the water and express the lithological nature of the watershed and aquifer terrain, composed mainly of dolomite. These values show the major role played by the dissolution of these rocks in supplying calcium and magnesium to surface waters, and the additional contribution of magnesium from soils and alterities.

3.3. Chloride and Sodium

The water at stations located on the left bank of the watershed (O8, O15, O16, O22 and O24) is characterized by the lowest concentrations of chloride ions, whereas maximum concentrations are observed at stations O1 and O12, as a result of leaching from the Jurassic marl-limestone formations upstream (in Prérif) and from the evaporite-rich South Rifain furrow downstream, as well as at stations O3, O5 and O7, O18 and O20 located respectively downstream of the town of Taza and the Oued Amlil. In addition to the geological nature of the terrain traversed, this evolution shows the contribution of anthropogenic inputs of urban or industrial origin to the enrichment of the waters studied in chloride ions.

In contrast to chloride ions, sodium concentration over the two study periods shows a slight increase in the first study period compared to the second at the majority of stations (Figure 4).

The values of chloride and sodium ion content recorded at the various stations sampled evolve in parallel with those of conductivity. In fact, these three parameters show a decreasing gradient from upstream to downstream.

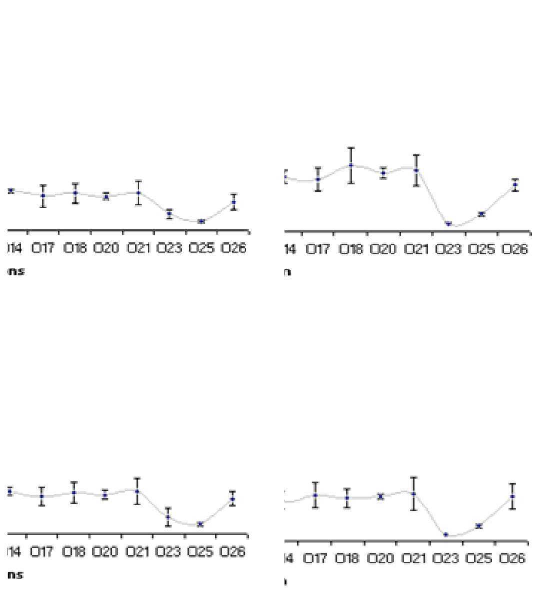

Fig 4. Spatial and temporal trends in average chloric and sodium levels (mg/L) in water analyzed over th 2021-2023 period.

3.4. Nitrogen Compounds

Trends in ammonium levels in the watershed (Figure 5) show significant fluctuations from station to station, and from year to year, with a clear increase in concentrations during 2021-2023.

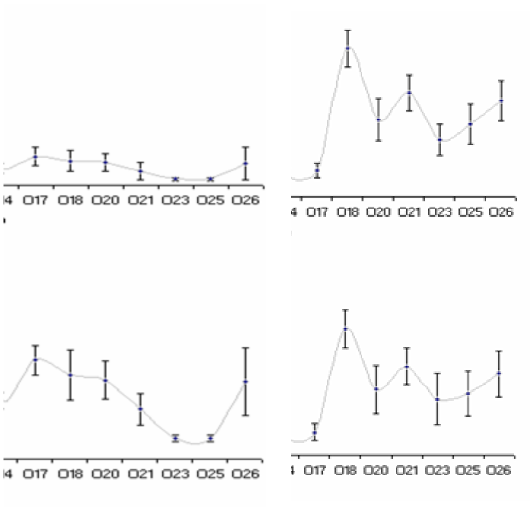

Fig 5. Spatial and temporal trends in average ammonium and nitrate levels (mg/L) in water analyzed over the 2021-2023 period.

Ammonium values in the wet season are significantly lower than in the dry season, particularly downstream of tributaries receiving wastewater, reflecting the effect of dilution and testifying to good oxygenation of the water, leading to nitrogen oxidation in winter. The relatively high levels recorded at stations O5 to O7 during the dry season reflect the incomplete degradation of organic matter.

Given their location upstream of the watercourses, stations O8, O15, O16, O22 and O24, which originate in the Middle Atlas, record low nitrate levels marked by small monthly variations. Downstream stations, mainly O5, O7, O9, O10 and O18, show irregular variations with significant monthly fluctuations.

Analysis of nitrate levels in the waters of the Inaouen catchment revealed an increasing gradient from upstream to downstream.

3.5. Biochemical oxygen demand and chemical oxygen demand

The values recorded (Figure 6) show an increase in the BOD_5 content of Oued Inaouen waters from upstream to downstream, particularly during dry periods. The increase in BOD_5 levels during dry periods can be explained by the establishment of conditions for the degradation of organic matter by microorganisms, whose activity intensifies as flow velocity decreases and water temperature rises. This activity, which consumes oxygen, is at the origin of water self-purification.

Comparison of analysis results for the 2022-2023 period with those for the 2021-2022 period showed a slight decline in BOD_5. The BOD_5 values observed show that Oued Inaouen cannot yet "defend itself" and ensure active self-purification, even at a distance of 30 km from the watercourses running through the town of Taza, and despite the dilution of its waters by tributaries of good to excellent quality.

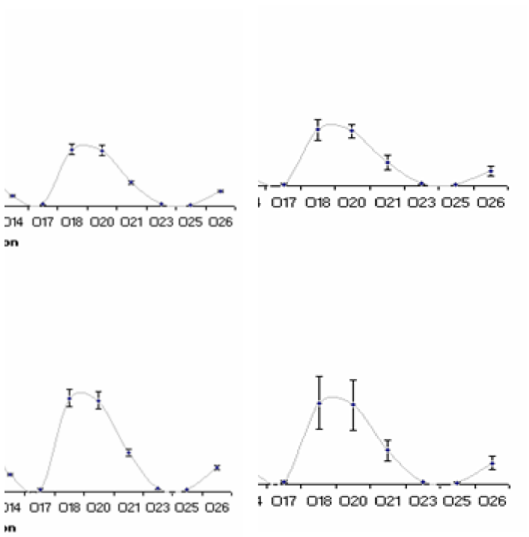

Fig 6. Spatial and temporal trends in average BOD_5 and COD (mg O_2 /L) of analyzed waters over the 2021-2023 period.

COD levels increase from 2021-2022 to 2022-2023, as well as from wet to dry seasons. Indeed, minimum values of around 2.31 and 2.60 mg O_2/L (Station O15) were recorded during the wet period, and maximum values of around 6541.43 and 7048.07 mg O_2/L (O10) in the dry period, in 2021-2022 and 2022-2023 respectively. The Moroccan standards grid classifies these waters as excellent (O2, O6, O8, O11, O15, O16, O22 and O23), average to poor quality (O1, O3, O4, O12 and O13, O14, O17, O21, O23, O25 and O26) and very poor quality (O5, O7, O9, O10, O18 and O20).

4. Water chemistry facies

The representation of physico-chemical data expressed in mg/L on the Piper diagram shows several chemical facies that depend on the geological nature of the water and on agricultural and industrial activities.

Plotting the 26 stations sampled in the Oued Inaouen watershed over the two years of the study on the Piper diagram (Figure 7); is a representation that allows chemical facies to be characterized, enabling a rapid approach to analytical results with a view to easily characterizing the waters, monitoring their evolution, as well as comparing them to other waters of similar physico-chemical composition.

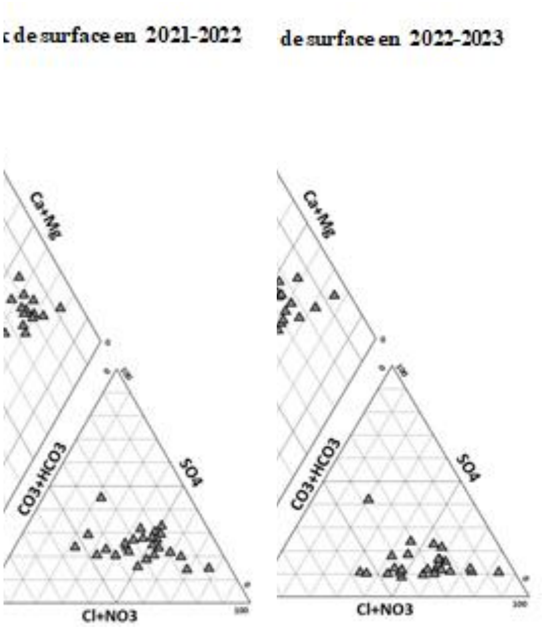

Fig 7. Diagramme de Piper des eaux de surface du bassin versant de l'Oued Inaouen durant la période 2021-2022 et 2022-2023

In this diagram (Figure 7), it can be seen that in the cation triangle, the points approach the magnesium-calcium pole, while in the anion plane they tend towards the chlorine-nitrate pole. Plotting the various surface water values on the Piper diagram shows that the waters of the Oued Inaouen watershed present 4 chemical facies: Chloride-Sodium, Bicarbonate-Calcium, Bicarbonate-Sodium and Bicarbonate-Magnesium.

5. Statistical analysis of physico-chemical data

In this study, principal component analysis (PCA) was carried out on the 16 sampling campaigns (12 in 2021-2022 and 4 in 2022-2023), with the aim of highlighting the various water points with high concentrations of chemical elements in relation to the basin as a whole. This gives us a preliminary idea of the elements and sites of pollution.

The results of this analysis are shown in figure 8. They highlight certain trends.

Data processing using principal component analysis was carried out using the 18 parameters as variables and the 26 stations analyzed in the Oued Inaouen watershed as individuals.

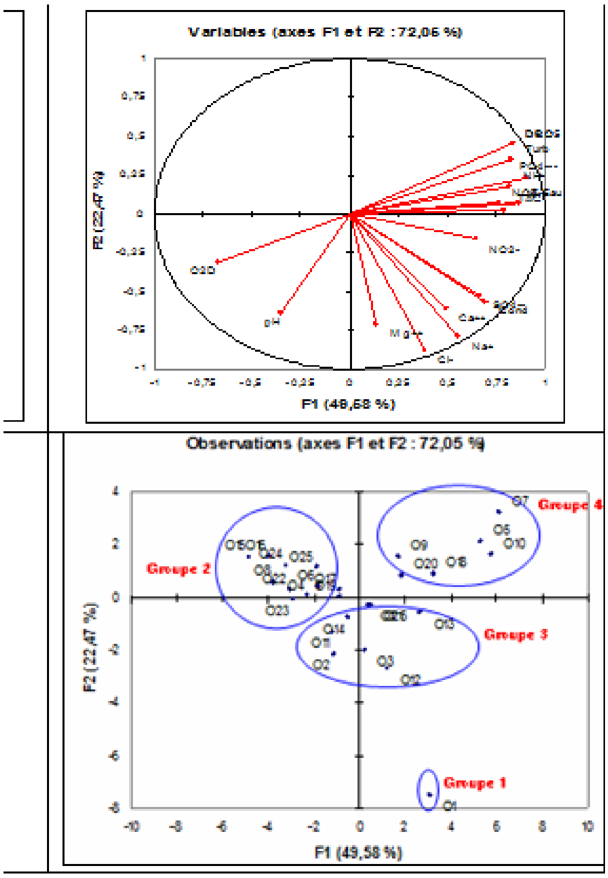

Fig 8. Projection of variables and representation of sources on the F1 and F2 factorial planes.

The correlation circle formed by axes F1 and F2, giving 72.05% of the total information, shows:

According to the F1 axis, well-oxygenated waters are opposed to waters polluted by organic matter, suspended matter, nitrates and nitrites.

The F2 axis contrasts highly mineralized waters, particularly rich in chloride, with waters polluted by ammonium ions. It thus defines a pollution gradient increasing from bottom to top, resulting in a decrease in oxygen and pH, and an increase in COD, BOD_5, SS, nitrates and nitrites. In fact, the spatial profiles of BOD_5, PO_4^{3-} and COD are opposite to that of dissolved oxygen.

The F1 axis reveals a group formed by sulfates, chlorides, nitrites, conductivity and sodium, which are positively correlated with the F1 axis. The F2 axis, on the other hand, differentiates 2 groups: One composed of turbidity, BOD5 and orthophosphates, which are positively correlated with the F2 axis, and another formed by pH and dissolved oxygen, which are negatively correlated with the F2 axis.

By projecting the individuals onto the F1 and F2 factorial planes, we were able to distinguish 4 different groups (Figure 8):

- Group 1: Contains a single point, station O1, with highly polluted water, 11% of which is of poor quality. This deterioration in water quality is due to the conductivity, which exceeds 4,000 µs/cm as the average value throughout the study period, resulting from the dissolution of gypsum (CaSO4) and halite (NaCl) present in the Jurassic marl-limestone formations. The dominant chemical facies at station O1 is chloride to sodium sulfate.

- Group 2: Includes stations O6, O8, O15, O16, O22, O23, O24 and O25, which are less polluted in terms of mineralization. The water from these stations comes from the Middle Atlas region, which is characterized by its excellent water quality and is far from urban areas. The water quality figures detailed below (Figure 3.21 to Figure 3.28) show the location of the freshwater zones that coincide with the water points sampled on the left bank of the basin, draining the carbonate liasic formations of the Middle Atlas.

Group 3: This group includes stations located downstream of Oued Inaouen tributaries, with good to average water quality, mainly due to the role of self-purification of these watercourses given the distance between towns and downstream (O2, O3, O12, O13, O14 and O26).

-Group 4: This group includes only stations with more contaminated water, with 55% of very poor quality. These stations are located in the center of the city of Taza and are characterized by high values of BOD_5, COD and SS, reflecting a high load of oxidizable and organic matter in the rivers that are the outlets for discharges from the city of Taza (O. Dfali, O. Taza, O. Jaouna and O. Larbaa) and from the urban center of Oued Amlil. Indeed, most of the discharges are domestic and/or industrial, and therefore rich in organic matter (mainly margines) responsible for the high TSS content. As a result, the water taken from these stations is of very poor quality.

The situation is very critical at stations O5, O7, O9, O10, O13, O18 and O20, which are constantly (all year round) subject to discharges of domestic wastewater and organic waste from olive oil mills during the olive-pressing season. The lowest dissolved oxygen values are recorded at stations O5, O7, O9, O10, O13, O18 and O20 (1.58 mg O2/L) during low-water periods (decrease in flow of Oued Inaouen tributaries), when the excessive input of organic matter leads to a significant drop in dissolved oxygen following its use in the biodegradation phenomenon. However, during rainy periods, a significant increase in dissolved oxygen levels is observed, thanks to water mixing.

Based on the results of water analyses carried out on the rivers that flow through the town of Taza, it can be concluded that the quality of these waters is very poor and therefore unsuitable for irrigating the region's vegetable crops.

The Wilcocx diagram (Figure 9) represents a classification of the waters; expressed as a ratio which writes the alkalizing power calculated by a formula called adsorbable sodium, "Sodium Absorption Ratio: SAR", as a function of electrical conductivity. For the same conductivity, the higher the ratio, the greater the risk.

$$SAR = \frac{Na^+}{\sqrt{(Ca^{2+} + Mg^{2+})/2}}$$

SAR and EC of effluent water receiving wastewater from the city of Taza, used for irrigation, are used in combination to assess the potential risk of soil salinization. To control the negative effects of irrigation water on soils and plants, the US Salinity Laboratory ,1954 established a quality scale for irrigation water based on its conductivity and sodium absorption rate [SAR]. In terms of conductivity, the water classes are: C1 (Conductivity< 250 µS/cm), C2 (250 µS/cm < Conductivity< 750 µS/cm), C3 (750 µS/cm <Conductivity< 2250 µS/cm) and C4 (2250 µS/cm < Conductivity< 5000 µS/cm). Thus, the waters are respectively low salinity, medium salinity, high salinity and very high salinity. In relation to SAR, the irrigation water classes are S1 (SAR< 10), S2 (10 <SAR <18), S3 (18 < SAR< 26) and S4 (SAR > 26). These different SARs indicate that the waters are respectively "excellent with low alkalinization hazards", "good with acceptable alkalinization hazards", "medium quality with significant alkalinization hazards" and "poor quality with very significant alkalinization hazards".

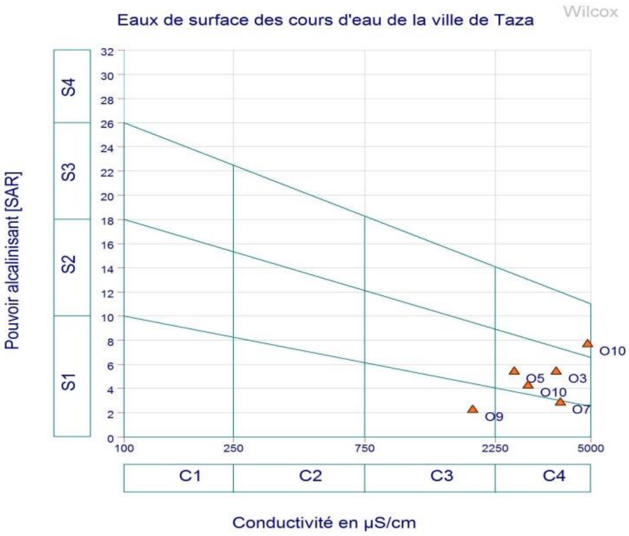

Fig 9. Wilcox diagram of sampled water points.

The results of analysis on the scale established by the US Salinity Laboratory, 1954, show that the effluents receiving wastewater from the town of Taza are of class C3S1 for the waters of Oued Taza, i.e. the irrigated wastewater belongs to class C3 of conductivities, which is the class of waters with high salinity, and to class S1 of excellent waters with low alkalinization hazards. Water downstream from Oued Bouljraf, Dfali, Jaouna and Oued Larbâa is classified in conductivity class C4 and in class S2 as good water with acceptable alkalinization hazards. For station O10 (Oued Larbâa after the Taza city dump), the water is of class C4S3, i.e. this wastewater belongs to class C4 of conductivities, which is the class of water with very high salinity, and to class S3 of average water with significant alkalization risk. (Figure 9).

6. Conclusion

Assessment of the physico-chemical quality of water in the Oued Inaouen watershed shows that the quality of these waters is influenced, on the one hand, by the nature of the lithological formations drained by these tributaries. Indeed, the stations on the right bank and those in the southern Rif mountains have high conductivity and concentrations of Na^+, Cl^- and SO_4^{2-} as a result of the dissolution of Jurassic marl-limestone formations. Whereas on the left bank of the Oued, high concentrations of bicarbonates, calcium and magnesium reflect the effect of Middle Atlas liasic limestone and dolomite.

On the other hand, anthropic action plays a major role in the degradation of water quality in the watershed through domestic, industrial (mainly Olive mill waste) and agricultural waste discharged without any prior treatment, particularly in the watercourses that flow through the town of Taza. During the dry season, this water can pose a risk of pollution transfer and contamination during agricultural use.

Spatial and temporal monitoring of several physico-chemical parameters has provided us with a picture of relatively intense pollution, resulting in a high mineral and organic load downstream of discharges in summer and autumn. This condition is accentuated in downstream stations that receive urban and industrial discharges. Dilution of Oued Inaouen waters during winter and spring floods. The dilution of Oued Inaouen water during the winter and spring floods, and by tributaries from the Middle Atlas, reduces the pollutant loads generated during the dry season. In addition, the role of self-purification improves the very poor to average quality of water entering the Idriss Iier dam, despite overly mineralized and polluted inputs from the city of Taza, urban and rural centers, and the right bank of the basin.

A study of the physico-chemical quality of the water in the Oued Inaouen watershed shows high values for certain pollution indicator parameters at certain stations, which adversely affects water quality. In order to better understand the problem of water pollution in this basin, and to get an idea of its biological quality, a bacteriological analysis of these waters proved useful.

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