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Trace Elements Analysis in Different Water Categories

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Abstract: Water quality is a permanent problem that continues to arise worldwide. The measurement and monitoring of impurities in water becomes paramount. An informative analysis of water chemistry to determine the content of regulated and unregulated metals is becoming increasingly necessary. The concentrations of 5 trace elements (Al, Fe, Cu, Ni and Zn) in drinking water, in surface waters of Beni Mellal city (Morocco) as well as in samples of bottled water from different units of 17 Moroccan and foreign commercial brands have been determined by Inductive Coupling Atomic Emission Spectrometer (ICP-AES) and compared to local, European and international *standards*. This study provides a general indication of the categories of water samples whose the concentration of its trace elements have reached or exceeded water quality standards. The data presented in this work will be useful from the health point of view since the trace elements are important for the good functioning of the biological system in particular the enzymatic system by facilitating the attraction and the ionic bonds of the molecules of substrate with the enzymes and their conversion into specific end products. It was found from the preliminary results of this study that these interesting elements can vary considerably depending on the region, the treatment organization and the origin of the water. However, water, whatever its nature, must comply with regulatory standards at all times.

1. Introduction

Water is a real source of minerals. In its natural course, in contact with the soil and rocks, it is loaded with mineral salts and trace elements, thus presenting different contents of these components depending on the region. The mineral content of drinking water, whether from the tap or in bottles (natural spring water, mineral water or treated water), is set by standards so that this water benefits from a real mineral balance. Trace elements such as copper, iron, manganese, molybdenum, nickel, selenium and zinc can be classified as essential elements (Hossain *et al.,* 2018; Ma *et al.,* 2020; Mickael *et al.,* 2020) for humans while others such as silver, mercury, arsenic, cadmium, chromium and lead are toxic (Guardia *et al.,* 2012; Marcelo *et al.,* 2020).

In the literature, there are researchers who classify Ni as an essential trace element (Mickael *et al.,* 2020; Prashanth *et al.,* 2015) and others who consider it a probable essential trace element according to the biological classification proposed by researcher Frieden (Prashanth *et al.,* 2015; Frieden, 1985).

According to a categorical classification, other researchers call it an additional trace element, whose role is unclear but may be essential. The elements belonging to this group are cadmium, nickel, silica, tin, vanadium and aluminium (Prashanth *et al.,* 2015). Essential trace elements have a dual role in body physiologies and the proper functioning of the biological system, and as their name suggests, they are essential in low concentrations but toxic in high quantities (Mickael *et al.,* 2020; *WHO,*1993). In recent years many researchers have focused on the study of the mineral quality of water intended for human consumption (Alaqarbeh *et al.,* 2022; Mambou *et al.,* 2021; Daniela *et al.,* 2020; Jie *et al.,* 2017; Magesh *et al.,* 2017) as there has been an increase in global public health problems associated with the contamination of drinking water by major chemical elements and trace elements (Pepaj *et al.,* 2017).

Indeed, the analysis of the chemical composition of water is the only guarantee of the excess or deficit of these minerals. This is the aim of the present study which aims to determine the concentrations of five trace elements (Al, Fe, Cu, Ni and Zn) in drinking water, in surface waters of the city of Beni Mellal, Morocco as well as in bottled water samples from different units of 17 Moroccan and foreign commercial brands. In order to determine these inorganic elements in the samples several analytical techniques are currently used such as flame atomic absorption spectroscopy (FAAS) (Kazantzi *et al.,* 2019; El Ammari *et al.,* 2015; Haggar *et al.* 2018), inductively coupled plasma optical emission spectrometry (ICP-OES), inductively coupled plasma mass spectrometry (ICP-MS) (Manousi *et al.* ,2020) and inductively coupled atomic emission spectrometry (ICP-AES) (Ioanna *et al.,* 2022; Manousi *et al.,* 2020). This study used the latter technique to analyse the water samples and ensure compliance of their trace element contents with local, European and international standards.

2. Methodology

2.1 Study matrix

Five categories of waters were analyzed, namely: bottled Moroccan mineral waters designated by MMW_1, MMW_2, MMW_3, MMW_4, bottled foreign mineral waters designated by FMW_1, FMW_2, FMW_3, FMW_4, FMW_5, FMW_6, FMW_7, FMW_8, bottled table waters designated by : TW_1, TW_2, TW_3 and tap water TW, bottled natural spring water MSW and FSW (Moroccan and foreign respectively); and finally, surface water from two areas of the city of Beni Mellal: *Ain Asserdoune* and *Bouykoub* designated as SW_1 and SW_2 respectively.

The tap water and surface water were placed in PET (polyethylene) bottles that were carefully washed beforehand with a slightly acidified nitric acid solution and rinsed several times with distilled water (AFNOR,1972; Herzog *et al.,* 2020). The samples were treated in the field with pure nitric acid (HNO_3 ultra) and then transported in a cooler to the laboratory.

2.2 Experiments

In the present study, the selected trace elements: Al, Cu, Fe, Ni and Zn, were analysed by the technique of inductively coupled plasma atomic emission spectrometry ICP-AES. It is based on the coupling of a plasma torch and a Horiba Jobin Yvon Ultima 2 atomic emission spectrometer. It is particularly sensitive since the sample, placed in solution, is nebulised in an Argon plasma of up to 8000°C, which allows very effective excitation of the atoms. The spectrometer used is equipped with a thermo-regulated optical system comprising two back-to-back gratings of 4343 rpm and 2400 rpm, thus covering the spectral range of 120-800 nm with a nitrogen scan. The detection system is composed of two photomultipliers.

2.3 *Principal component analysis*

Principal Component Analysis (PCA) is an unsupervised multivariate analysis technique used to reduce the size of data dimensions without losing information about the samples. This allows the data to be analyzed and examined in a more conceptual way (Mahmoudi *et al.,* 2021; Karamizadeh *et al.,* 2020; Hassan *et al.,* 2021). The PCA is based on the transformation of a group of original quantitative variables into a new variable space formed of the linear combinations of the initial variables thus reducing the space (Alami *et al.,* 2021). A spectrum of 1000 wavelengths can be represented by a point in the new space. These combinations are called principal components PCs, that must be interpreted independently of each other, because they contain a part of the variance that must not be expressed in any other principal component (Hassan *et al.,* 2021). Le Logiciel utilisé pour la réalisation de l'ACP est Unscrambler X.

3. Results and Discussion

Figure 1 shows the distribution of essential trace element levels in surface water in two areas of the city of Beni Mellal, namely *Ain Asserdoune* (SW_1) and *Bouyakoub* (SW_2). We then notice that the *Ain Asserdoune* surface water has lower Al, Fe and Zn contents than those of the *Bouyakoub* surface water, on the other hand the Cu and Ni contents are higher. the *Bouyakoub* surface water, on the other hand the Cu and Ni contents are higher.

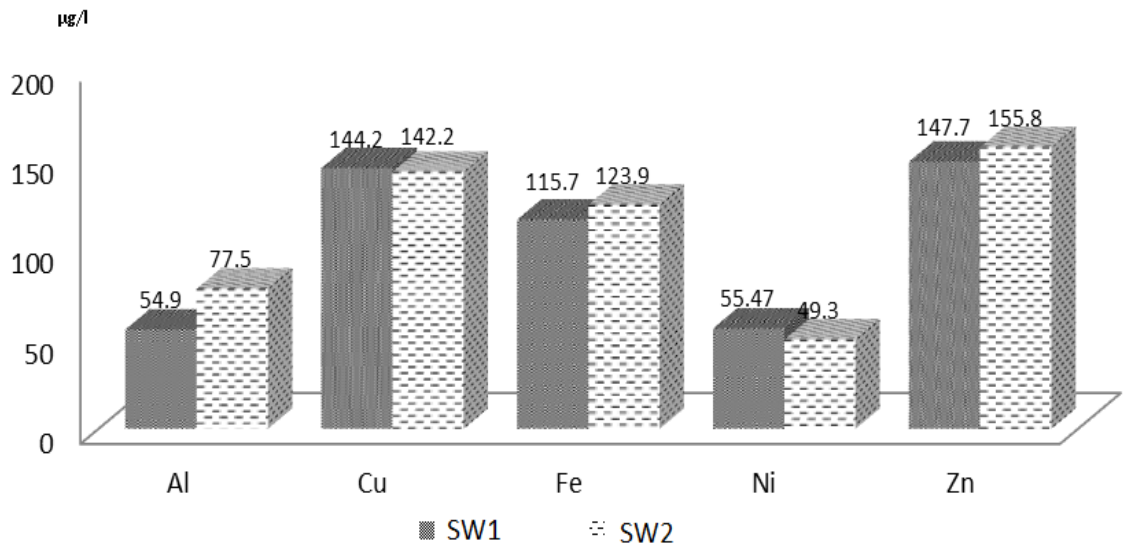

Figure 1. Trace element levels in surface waters of *Ain Asserdoune* (SW_1) and *Bouyakoub*(SW_2).

Figure 2 illustrates the distribution of trace elements namely: Al, Cu, Fe, Ni and Zn in bottled Moroccan table water as well as in the tap water of the city of Beni Mellal. According to this figure, we notice that the difference is very clear in the case of aluminum and iron from one water to another. Moreover, the table water TW_2 has the highest content of Al followed by TW_3, while the lowest content belongs to the tap water TW. The highest value of Iron is recorded in the tap water TW followed by the table water TW_2 while the lowest value is attributed to the table water TW_1.

Regarding Copper, Nickel and Zinc, the difference is not great and does not exceed 5.36%, 5.14% and 7.98% respectively. The highest content of Cu is recorded in the table water TW_1, followed by the two table waters TW_3 and TW_2 and finally the tap water TW. In the case of Ni the highest value is recorded at TW_2 followed by ER, TW_3 and TW_1 respectively. Finally, the table water with the highest Zn concentration is TW_3 followed by TW_2 and then TW tap water, the lowest value is obtained at TW_1.

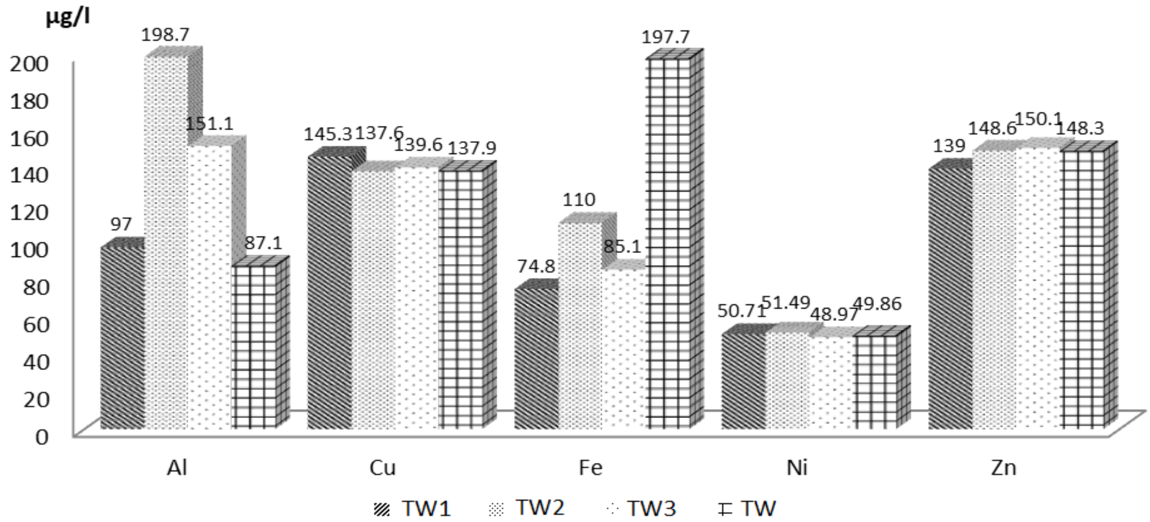

Figure 2. Trace element content in Moroccan bottled table waters (TW_1, TW_2 and TW_3) and in tap water (TW).

Figure 3 illustrates the distribution of essential trace elements in three types of foreign bottled waters, namely sparkling water, natural mineral water and natural spring water. Overall, it can be seen that the Copper and Zinc contents are indifferent when passing from one water to another since the difference is of the order of 7.4% and 6.48% respectively. On the other hand, Nickel presents a difference of the order of 17.71%, thus creating a difference between the waters, the highest content is attributed to foreign mineral water FMW_3 and the smallest content is attributed to foreign mineral water FMW_8.

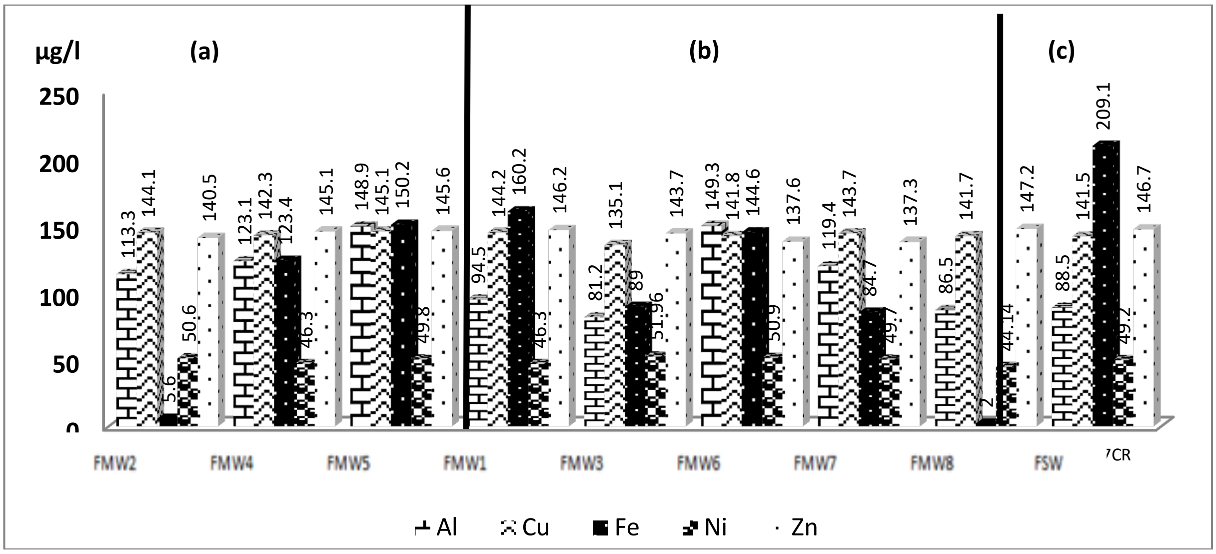

Figure 3. Trace element levels in foreign bottled waters [(a) carbonated waters (b) natural mineral waters and (c) natural spring water].

In the case of Aluminum and Iron, the difference is obvious when oscillating between foreign bottled waters, it is of the order of 83.86% and 79.10% respectively. In category (a) it can be seen that both in the case of Copper and in that of Nickel and Zn, the contents are almost the same for the three foreign mineral waters bottled FMW_2*, FMW_4* and FMW_5* with differences around 1.96%; 9.28% and 3.62% respectively. On the other hand, Iron and Aluminum have very large differences, thus reflecting a large gap between the waters of this category. As for category b, we also note that both in the case of copper and in that of Zn, the contents are almost the same when passing from one water to another with differences of the order of 6, 73% and 6.48% and respectively. Nickel is relatively different since we found 17.71%, on the other hand aluminum and iron present large differences between the waters (83.86% and 79.10% respectively) of this category. Moreover, the bottled foreign mineral water FMW_1 has the highest Fe content while the foreign mineral water FMW_8 has the lowest. As for Aluminum, the highest value is recorded at the level of mineral water FMW_6 and the lowest value is attributed to foreign bottled water FMW_3.

The sample of foreign natural spring water FSW representing category (c), presents contents of Al, Cu, Zn, Ni similar to category (b) except for the case of Fe where it exceeds them.

Concerning the samples of Moroccan bottled mineral waters represented in **Figure 4**, the Cu and Zn contents are very close since the difference is of the order of 2.4% and 8.5% respectively. Nickel is relatively different since there is a difference of 19%, on the other hand aluminum and iron present large differences between the waters of this category (173.76% and 578.53% respectively). Moreover, Moroccan bottled mineral water MMW_3 has the highest Al content while Moroccan bottled natural spring water MSW has the lowest. As for Fe, the highest content is recorded at the level of MSW water and the lowest content is attributed to Moroccan mineral water MMW_2.

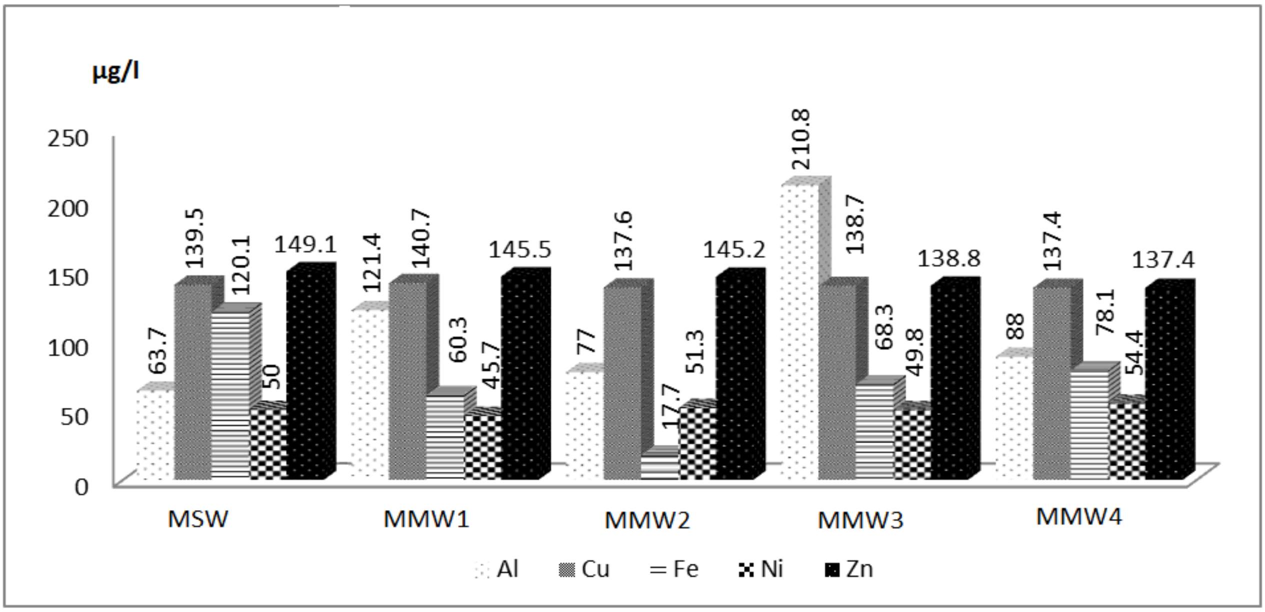

Figure 4. Contents of trace elements in Moroccan natural bottled waters (MSW: spring water and MMW: mineral waters).

Table 1. Drinking water (NM,NE et WHO) standards

	NM	NE	WHO
Al	200 µg/l	200 µg/l	200 µg/l
Cu	2000 µg/l	2000 µg/l	2000 µg/l
Fe	300 µg/l	300 µg/l*	200 µg/l
Ni	50 µg/l	20 µg/l	70 µg/l
Zn	3000	3000 µg/l	3000 µg/l

* No guidelines, Desired: 300 µg/l

Aluminum is the most abundant element in the earth's crust, it is found in mineral rocks and clays, but it is found in trace amounts in natural waters. In humans, the ingestion of large amounts of aluminum causes reports of gastrointestinal disturbances, skin rash, muscle pain, ulceration of the lips and mouth (Clayton, 1989) also aluminum can cause neurotoxicity that is associated with impaired blood-brain barrier function (ATSDR, 2015). Thus, in our study, the concentration of aluminum in all the water samples was lower than the standard limits of the WHO, NM and NE which are of the order of 200 µg/l (**Table 1**), except for the two water samples: Moroccan bottled mineral water: MMW_3 and table water TW_2 which have an aluminum content of around 210 µg/l and 199 µg/l respectively (**Figure 5**). The Al contents detected in the present study are higher than those in Iran whose content is around 115.42 µg/l (Ghaffari *et al.,* 2021) on the other hand they are lower than those in Turkey which are between 210 and 272 µg/l (Yilmaz *et al.,* 2021) and those in Malaysia which are around 330 µg/l (Ahmad, 2012).

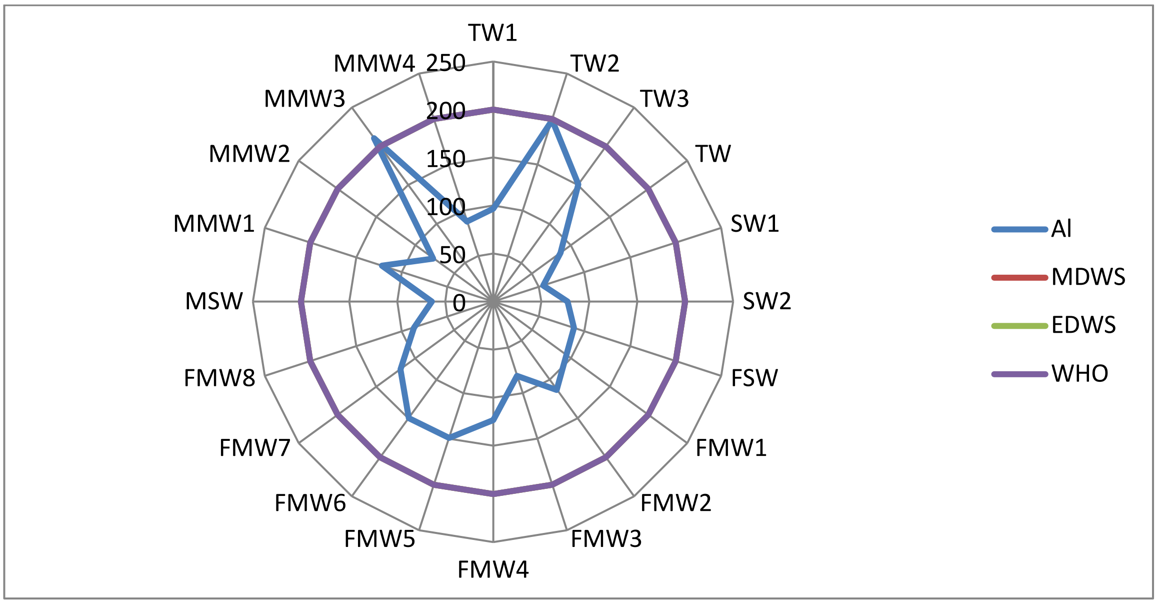

Figure 5. Distribution of Al content compared to NM, EU and WHO standards, in all the water samples studied).

The highest Cu content was detected in sample TW_1 with an order of 145.3 µg/l while the lowest Cu content was detected in sample FMW_3 with an order of 135.1µg/l as shown in **Figure 6**. The present study indicated that the overall level of Cu in the samples studied was below the WHO standard allowable levels, NM and NE which are in the order of 2000 µg/l.

Copper is the third most widely used metal in the world (Fjallborg *et al.,* 2003) and is an essential component of many enzymes. It is an essential trace element entering into the composition of many enzymes and proteins (ceruloplasmin, certain cytochrome oxidases, etc.). Its presence in trace amounts is essential for hemoglobin formation, wound healing and immune function (Sunde, 2012) but at the same time it is a potential hazard causing various health problems when people are exposed to it levels above the permissible value. Short periods of exposure to or use of water whose copper level exceeds the maximum allowable value may cause gastrointestinal disturbances, including nausea and vomiting, while long-term exposure causes liver or kidney damage (Sunde, 2012). At high doses, copper becomes toxic in animals (>15 mg Cu/kg ingested in sheep (Soli, 1980; Maiorkka *et al.,* 1998) and in humans (>10 mg/d) (Abdelmajeed *et al.,* 1990). The Cu levels detected in the present study are slightly lower than those in Bangladesh, whose content is around 160 µg/l (Rafiul *et al.,* 2020), and higher than those in Turkey, whose values are between 9 and 26 µg/l (Yilmaz *et al.,* 2021).

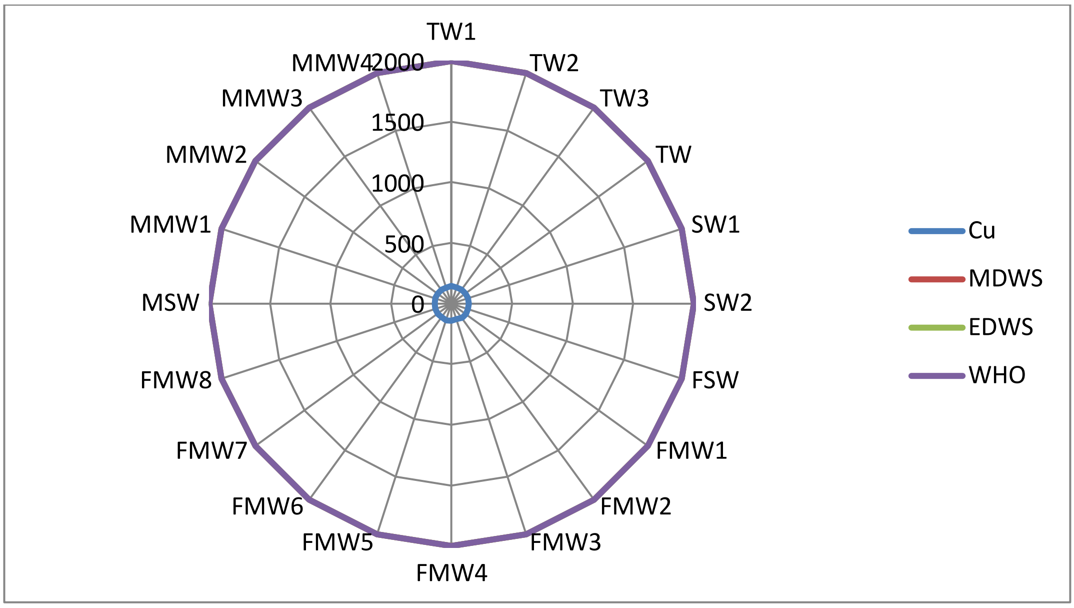

Figure 6. Distribution of Cu contents compared to standards: NM, EU and WHO.

The Fe content, the concentrations of all the water samples analyzed were below the acceptable standard limits which are of the order of 300 µg/l, according to the NM and the NE, and 200 µg/l according to the WHO at apart from two samples with high Fe contents in the order of 210 µg/l and 198 µg/l respectively, characterizing the foreign surface water samples FSW and tap water TW respectively, while the lower content of Fe was detected in the FMW_8 extraneous mineral water sample with a value of around 2µg/l as shown in **Figure 7**. Iron is ubiquitous in all freshwater environments and often reaches much higher concentrations in water. water than trace metals. It is an essential element for human nutrition and plays an important role as a constituent of enzymes such as cytochromes and catalase, and oxygen-carrying proteins such as hemoglobin and myoglobin (Sheldon *et al.,* 2016). Iron deficiency leads to hyochromic macrobiotic anemia, one of the most common health problems in the world. Prolonged consumption of drinking water with a high concentration of iron can lead to liver disease (hemosiderosis).

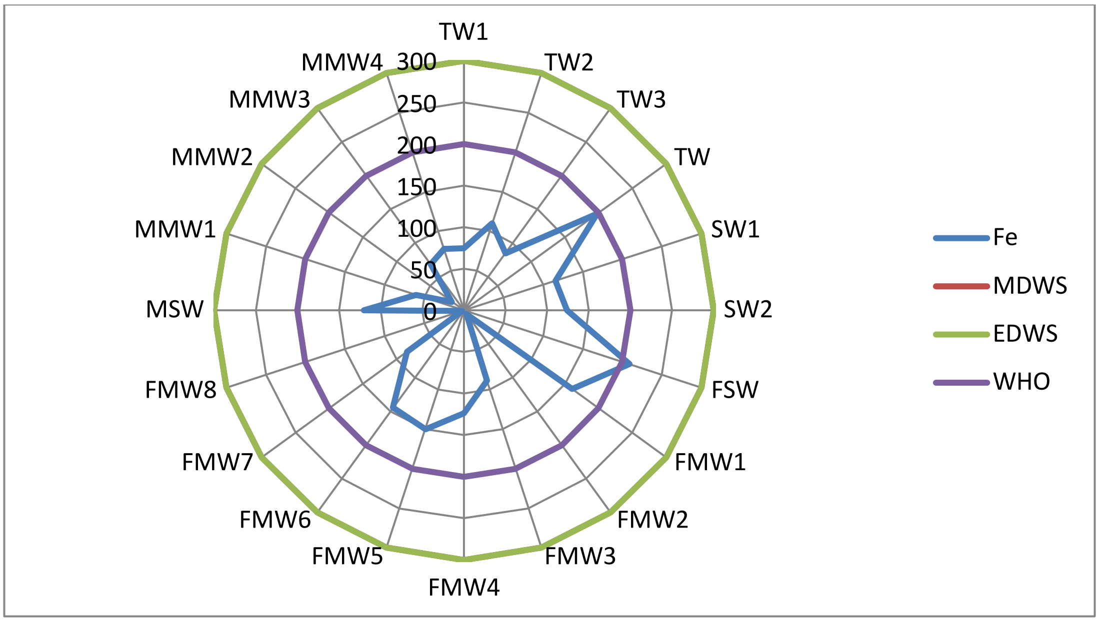

Figure 7. Distribution of Fe contents compared to NM, EU and WHO standards in all water samples.

The Fe contents in the present work are lower than those reported in different countries in Ethiopia the concentrations of Fe in surface water, is ranged from 310 to 730 µg/l and 520 to 810 µg/l in the dry season and wet season, respectively (Berhe, 2020); in Turkey the levels fluctuate between 216 and 331 µg/l (Yilmaz *et al.,* 2021); Pakistan recorded a content of 399 µg/l in these waters (Abeer, 2020) and finally, in Bangladesh the iron content is between 30 and 970 µg/l (Rafiul *et al.,* 2020). The Nickel content (**Figure 8**) in all the water samples analyzed was below the acceptable standard limit given by the WHO, which is around 70 µg/l. The highest value detected is at the level of the two samples of surface water SW1 and Moroccan mineral water MMW_4 with Ni contents of around 55.5 µg/l and 54.5 µg/l, but these two values as well as most of the samples are limit-limits with the Moroccan standard which is estimated at 50 µg/l. On the other hand, for the European standard, we note that the content in all the samples exceeds the standard estimated at 20 µg/l. In general, the presence of nickel in groundwater or surface waters can be due to rock weathering and human activities, such as the burning of fossil fuels, smelting and the electroplating industry. Certainly, it has been shown that nickel is not an essential nutrient for humans (Prashanth *et al.,* 2015), but it can serve as a cofactor or structural component of specific metalloenzymes with a variety of physiological functions. Nickel facilitates the absorption or metabolism of ferric iron. In small quantities, Ni is essential, entering into the composition of many enzymes. But if it exceeds the toxicity threshold, it can manifest itself by a dermatitis called Ni scabies which is allergic eczema, irritation of the respiratory tract with sometimes asthma or bronchitis or even bronchial cancers. The comparison of these results with those reported in other countries shows that the Nickel contents of the water samples studied are much lower than in the south-east of Nigeria 1260 µg/l (Obasi *et al.,* 2020) and also in the north-south of Nigeria 590 µg/l (Maigari *et al.,* 2016).

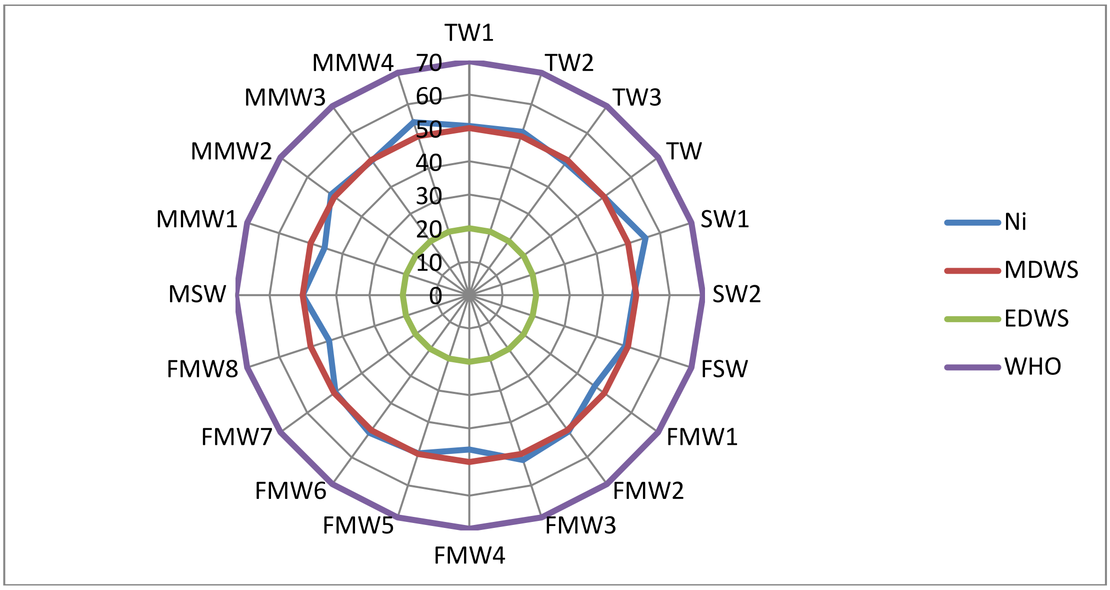

Figure 8. Distribution of Ni contents in relation to standards: NM, EU and WHO.

In **Figure 9**, all the water samples analyzed show zinc concentrations well below the admissible limits, which are around 3000 µg/l; the highest concentration was observed in surface water SW2 with a content of 155.8 µg/l while the lowest concentration was found in the foreign mineral water sample FMW7 with a content of 137.3 µg/l. Zinc is an essential trace element that can be used as a dietary supplement in medicine (Reza *et al.,* 2021), it acts as a catalyst for enzymatic activity in the human body. It is needed to support the immune system, protein synthesis and reproductive health (Yilmaz *et al.,* 2021), (Ahmad,2012). Zinc can intervene in certain metallo-enzymes (carbonic anhydrase, alcohol dehydrogenase, carboxypeptidase), it takes part in the exchanges oxygen-carbon dioxide by the red corpuscles. The intake of this trace element significantly reduces the occurrence of certain infections (mainly pneumonia and diarrhoea) in children in developing countries (Fjallborg *et al.,* 2003), which can translate into a gain in life expectancy (Sune, 2012). On the other hand, if zinc is present in water at high levels and with an accumulation effect, it can cause anemia, damage to the pancreas and kidneys, disturb protein metabolism and cause arteriosclerosis. It can also cause nausea, disorders of the gastrointestinal system, complications in the respiratory system, skin conditions (Soli, 1980) without neglecting the fact that it is also carcinogenic for humans (Maiorka *et al.,* 1998).

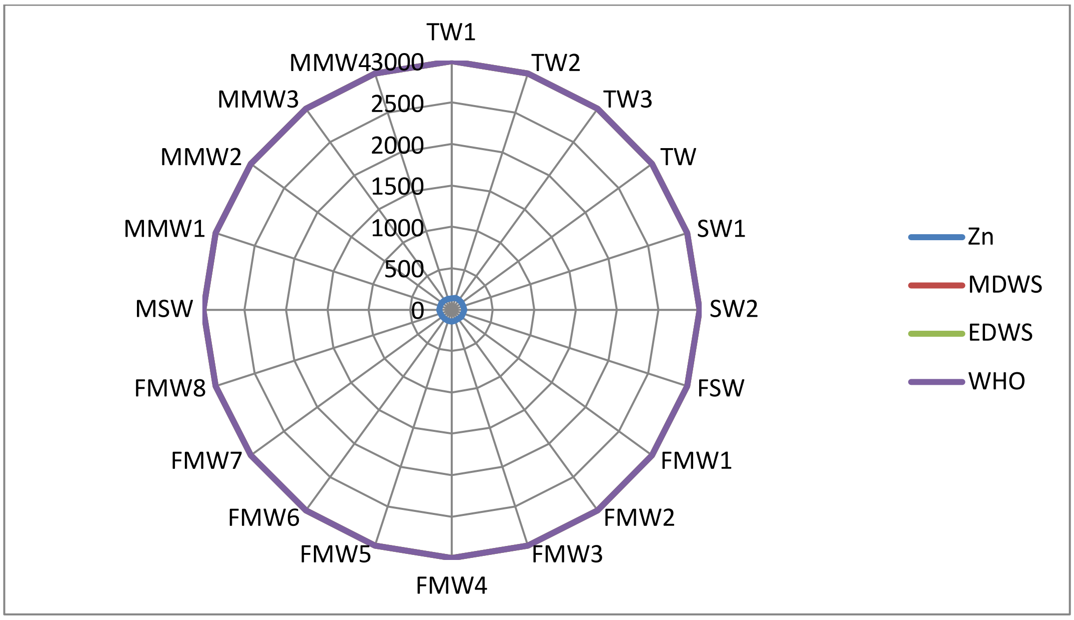

Figure 9. Distribution of Zn contents compared to NM, EU and WHO standards.

The zinc levels in the waters studied in this work are very close to those of Bangladesh (120 µg/l) (Rafiul *et al.,* 2020), lower than those of Pakistan (222 µg/l) (Abeer *et al.,* 2020) and Turkey (202 µg/l) (Yelmaz *et al.,* 2021); on the other hand, they are higher than those of the waters of Iran (14.59 µg/l).

Figure 10 below reports the results of the PCA in terms of the factorial map of the waters studied superimposed on the loadings of the contents of chemical elements (As, Cd, Cr and Pb) in the factorial plane PC1-PC2:

- The optimization of the representation of the main component planes from 5 to 2 dimensions allowed us to obtain 98% of the reliable information identified between the main components PC1 and PC2 with a very good projection of the point clouds of the samples analyzed.

- The analysis of the distribution of the samples belonging to the five classes of water according to their concentrations in trace elements shows the existence of two relatively clear classes groups of individuals distributed in such a way as:

  - ✓ 50% of the samples have a globally close projection reflecting the similarity of their concentrations by type of chemical element.
  - ✓ 50% of the samples have projections spaced apart and grouped by hen of 2 to 3 samples, reflecting their specificities in terms of trace element content.

- ➢ The distribution of point clouds shows us the weak relation of influence of the category of water on the concentration of trace elements of the samples studied.
- ➢ The category of bottled foreign mineral spring waters holds 30% samples with extreme specific concentrations, followed equally by the categories of Moroccan table waters 10% and Moroccan mineral waters 10%.
- ➢ Each category of water has samples that distinguish it from the other main components by the extreme concentration values they hold, namely:
  - ✓ Sample N°19 (Bottled Moroccan Mineral Water) has a high (Al) content.
  - ✓ Sample N°1 (Moroccan table water) has a high concentration of (Cu).
  - ✓ Sample N°7 (Foreign natural spring water) has a high content of (Fe)
  - ✓ In the two samples N°2 (Moroccan table water) and N°19 (Moroccan bottled mineral water) the disparity of the values of the contents of chemical elements is linked to aluminum (Al). They respectively represent the highest increasing gradient in this trace element while the concentrations for the other elements (Cu, Fe, Ni and Zn) remain comparable to the other samples.
  - ✓ The three samples N°7 and N°8 (foreign bottled mineral waters) – N°4 (Moroccan table water) have a distribution that is different from the cloud of points since they represent the increasing gradient with a high iron content (Fe).
  - ✓ Also the three samples N°18 (Moroccan bottled mineral water) – N°9 and N°15 (foreign bottled mineral waters) represent the increasing gradient of the lowest iron (Fe) contents, hence their distinctive grouping

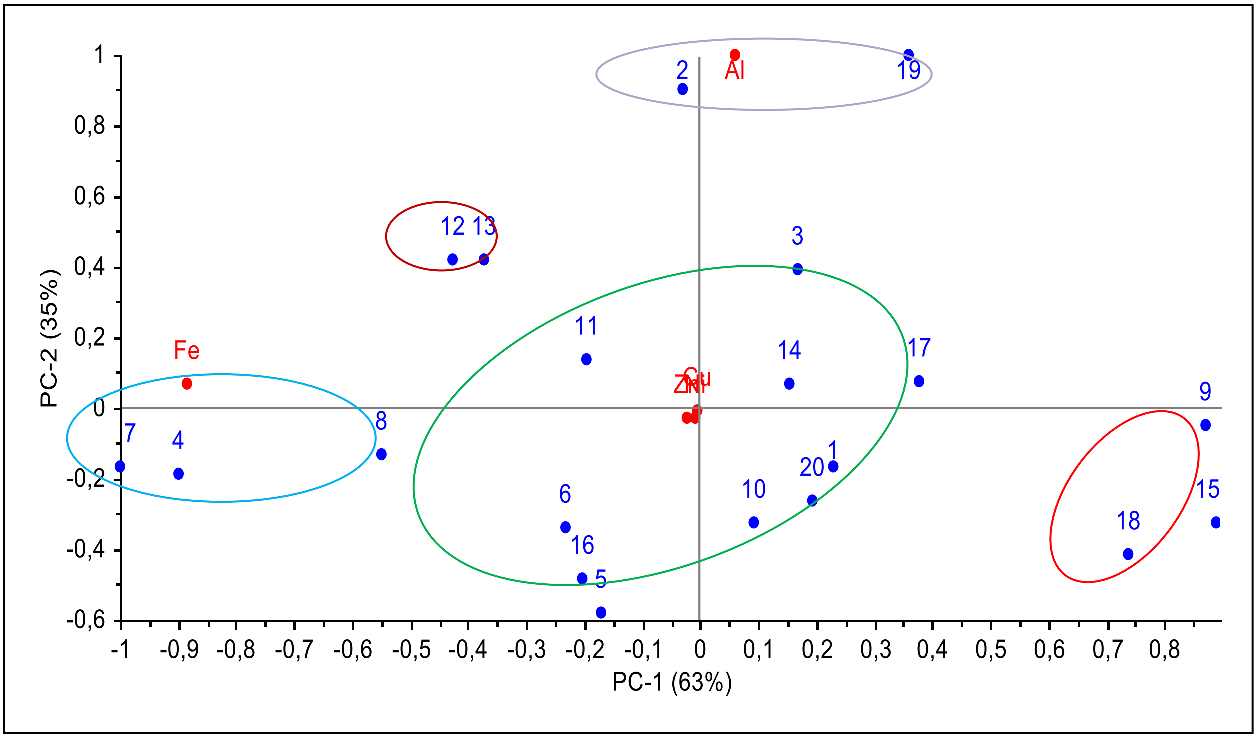

Figure 10. Bi-plot by PCA of waters in the PC1-PC2 principal component plane (table waters: 1,2, 3 and 4; surface waters: 5 and 6; foreign bottled waters: 7,8,9,10,11 ,12,13,14 and 15; Moroccan bottled waters: 16,17,18,19 and 20).

➢ The graph of the correlation circles comprising 98% of the information generated between the main components PC1 and PC2, helps us to better understand the possible correlations existing between the contents of chemical elements (Al, Fe, Cu, Zn and Ni) (**Figure 11**).

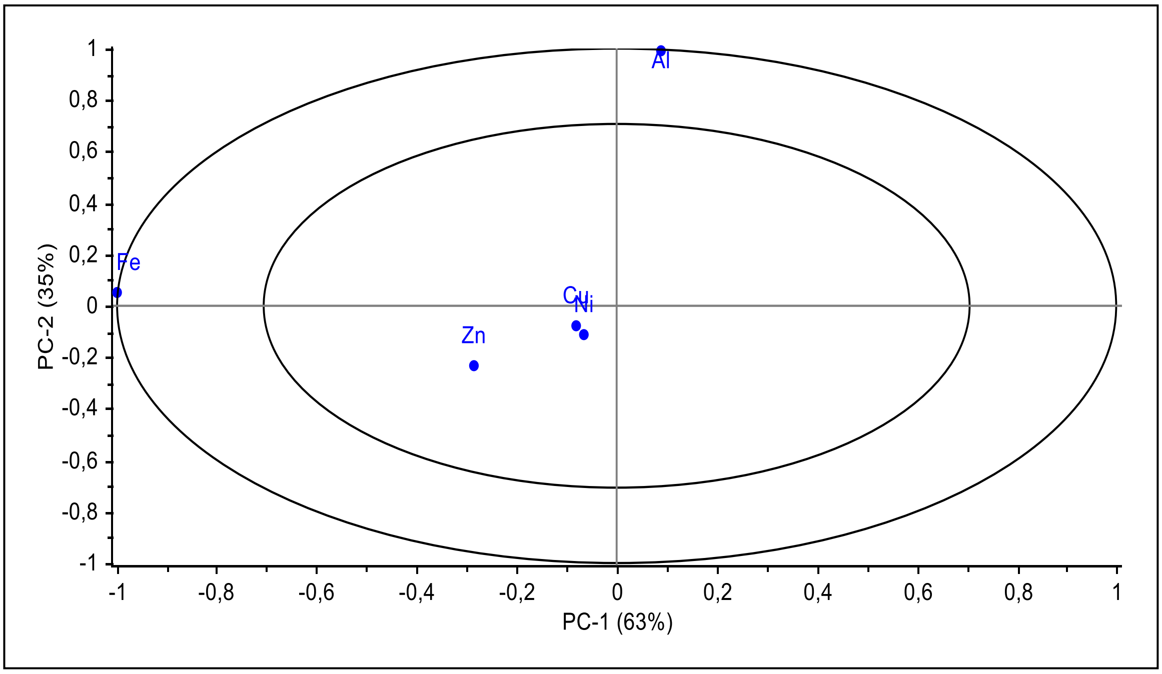

Figure 11. Circles of correlations, between the contents of chemical elements (Al, Fe, Cu, Zn and Ni) in the plane of the main components PC1-PC2.

➢ The distribution of the variables Cu and Ni relatively are very far from the center and which are not close to the two correlation circles Cu and Ni in the center outside the correlation circles as well as their projections can be significant with respect to the axes of the principal components, shows both the absence of specific correlation between them and with the principal components PC1 and PC2. Indeed, the concentrations of the samples specific to each of the elements Zn, Cu and Fe are similar whatever the category of water.

➢ We also note that the variables Al and Fe are linked by a specific correlation since they are arranged on the same circle of correlation. In fact, in 75% of cases there is an inversely proportional correlation between these elements, while in 25% of cases their levels remain globally close, especially for the category of water from foreign natural springs.

➢ We also identify underlying phenomena respectively between the variables Fe, Al and the main components PC1 and PC2, namely that:

✓ The PC1 axis is strongly correlated with the element (Fe) given the good superposition of the latter with the axis, in the same way we advance the same conclusion for the PC2 axis and the element (Al).

✓ All the samples that contribute strongly and negatively with the PC1 axis have a high Fe content and vice versa in the opposite direction.

✓ All the samples that contribute strongly and positively with the PC2 axis have a high Al content and vice versa in the opposite direction.

Conclusion

Trace elements play a considerable role in health. It is essential to provide it in quantity; this contribution is made through food. From the results obtained, it can be concluded that the levels of trace elements in the water samples were classified within the authorized limits set by the directives of the standards, namely the Moroccan, European and WHO standards. The work showed that there is an overlap of all the categories of water studied, which means that the category, the nature of the water and its geographical origin do not represent a significant effect on the distribution of trace elements. Surface waters relatively rich in trace elements such as Iron or Zinc need further work to be able to confirm their possible healing properties in cases of illness such as anemia for iron (Ech 4,7,8) and pathologies linked to a lack of zinc such as congenital malformation or failure of the immune system (Ech 6.8). The fact that the surface waters studied in Morocco present standards respecting the requirements of their potability challenges both manufacturers and decision-makers to promote projects for the marketing of such waters after conditioning by bottling.

Declaration of Competing Interests

The authors declare that they have no competing interests.

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