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Reuse of Treated Wastewater for Irrigation in Misurata, Libya: Pollution and Quality Assessment

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Abstract

This research focuses on evaluating the potential risks associated with the use of treated wastewater (TWW) for irrigation by drawing an analogy between the outcomes, international standards, and indices. Twelve TWW samples collected monthly for a duration of one year. The comprehensive pollution index (CPI) and organic pollution index (OPI) classified these waters as slightly and slightly to moderately polluted. Among the analyzed trace elements (Fe > Mn > Zn > Co > Cu > Cd > Pb > Cr), the Cd concentration was higher than the desired limit for irrigation water. Plotting Piper's diagram shows that most of the TWW samples are fall into the calcium bicarbonate zone. TWW samples are all within the permissible limits in terms of pH, EC, Sodium Adsorption Ratio (SAR), Total Dissolved Solids (TDS), Soluble Sodium Percentage (SSP), Magnesium Hazard (MH), and Residual Sodium Carbonate (RSC), while the integrated salinity and alkalinity effect revealed in the U.S. salinity diagram indicates that TWW samples drop at high salinity and low sodium exchange hazard (C3-S1). The use of the TWW from Misurata for irrigation ultimately calls for control, and surveillance action plans to avoid soil contamination, salination and/or sodification.

Keywords: *Misurata; Treated Wastewater; Irrigation; Health risks; Environmental risks.*

1. Introduction

Wastewater re-use is a key factor of sustainable water resource management, promoting the preservation of freshwater of high quality, reducing environmental pollution and the overall cost of production. Wastewater irrigation can have positive and negative repercussions on the environment [1], and the application of this category of water in agriculture can be beneficial for the environment, through careful planning and management. The wastewater is a significant source of organic constituents and nutrients valuable in preserving soil productivity [2], [3] but it may contain pathogens and adverse chemical components that pose environmental and human safety risks [4]. In this sense, a range of risk factors have arisen in reuse; some are short-term impacts (e.g., microbial pathogen) while others have more long-term impacts with ongoing wastewater use (e.g. soil Salinities) [5]. Libya is an arid nation with insufficient rainfall to meet various agricultural needs. Groundwater is a valued, naturalist source for the Libyans, as it is the most dependable water source for their domestic, industrial, and agricultural activities. Agriculture in Libya is the real water consumer, consuming over 85 percent of the water resources and based on groundwater. [6]. The increasing pressure on groundwater and soil resources along the country's Mediterranean coastline is bringing about saline intrusion into the coastal aquifer and degeneration of both resources [7]. In Misurata, there is a severe water scarcity like other Libyan coastal towns, which dependent on groundwater because of low rainfall rates and lack of surface water resources. Developing competition for rare water resources shows the importance of sustainable and efficient use of TWW as a key improvement in the water source. The city's sewage treatment facility installed in 1989 with an average capacity of 24,000 m^3/day through preliminary and biological treatment

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accompanied by disinfection units. The most influent is domestic wastewater with a small share of business wastes. Libya pays insufficient attention to the re-use of wastewater, except for a few schemes that use treated wastewater in agriculture, even though the country suffers from water scarcity. So far, the wastewater generated from the sewage treatment facility in Misurata City still not fully used. The water used does not exceed 5 percent of the TWW generated [8], to produce feed crops from a limited area of about 125 hectares since 1989. This research focused on the ecological and health risks inherent with the use of TWW derived from the wastewater treatment plant in Misurata and evaluating its adequacy for irrigation as non-conventional water resources. However, it is a first step towards offering TWW performance benchmarks for this fragile ecosystem in the Misurata region, which will form the basis for future water and soil resources conservation planning decisions.

2. Materials And Methods

A. Site Description

Misurata's wastewater processing facility established in 1989 at the Western-Central Coastal Region of Libya about 13 kilometers from the city center **(**Figure 0.1**)**.

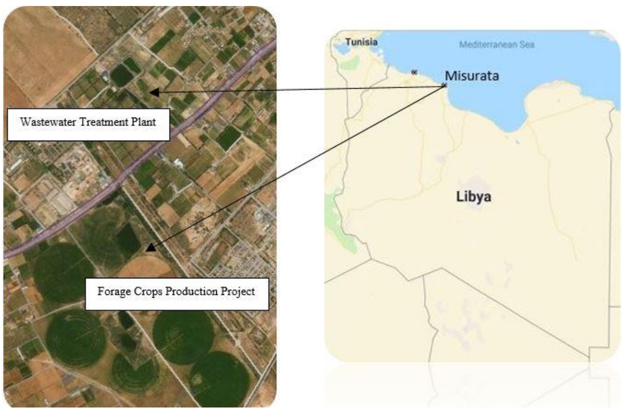

Figure 0.1 Location of Misurata's wastewater treatment plant and the nearby forage crops production project.

The Mediterranean climate is predominating in this region with some semi-desert elements.In this area, the Mediterranean climate prevails with some semi-desert elements with annual temperatures ranging from 13.51 °C to 28.22 °C and average annual precipitation of 246.9 mm/year, which classifies this region as an arid region. The air humidity is relatively constant and the evaporation rate is over 2,500 mm/year on the seacoast and increases as we move away from the coast (Figure 0.2).

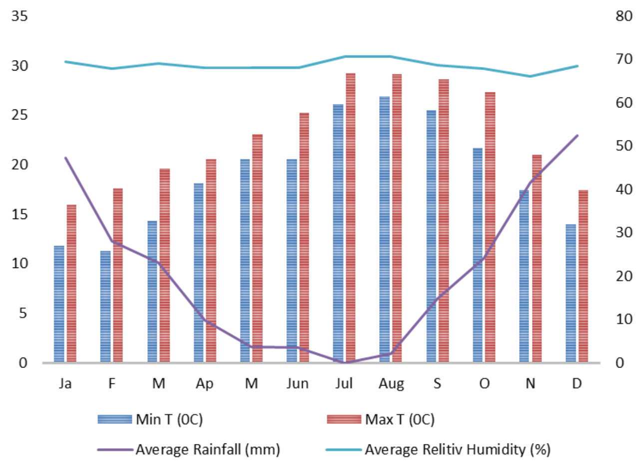

Figure 0.2 The mean climatic features of the study region over the period 2000 -2017 recorded at Misurata Metrological station.

B. Sampling and Analytical Procedures

TWW samples collected regularly from the processing facility effluent for a duration of one year (March-2017-February-2018). The collection, handling, and processing of

samples followed the approach proposed by the American Public Health Association [9]. The main Physico-chemical parameters analyzed in a quality-assured laboratory by the Misurata Iron and Steel Factory Laboratory. The estimated parameters are pH (Jenway 370 pH meter), electrical conductivity (470 Jenway EC meter), total dissolved solids (Oakton TDS meter), Chemical and Biological oxygen demand (HI98193 portable COD andBOD field devices), PO_4^{3-}, SO_4^{2-}, and NO_3^- (Hach DR/2800 spectrophotometer), Heavy metals, Na^+, and K^+ (180–30, HITACHI atomic absorption spectrophotometer), Cl^- (Mohr method Titrimetry), CO_3^{2-} and HCO_3^- (HCl titration using auto-titrator), Ca^{2+} and Mg^{2+} (standard EDTA using

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auto-titrator). Ca^{2+} and Mg^{2+} (standard EDTA using auto-titrator) (Table 0.1). A quality control process, including recalibration of the devices, triple sample analysis and retrieval testing of standard reference material, undertaken to control data quality. To verify the reliability of water analysis, charge balance error (CBE) was computed (Equ.1) TWW samples with lower anion concentrations (meq/L) show negative CBE while positive CBE results from high cation concentrations (meq/L). The approved water samples only those with less than ±5% CBE as per standard procedures.

Table 0.1 Statistical summary of the ionic composition of Misurata's TWW (n = 12).

Constituents	Units	Mean	Max	Min	SD
TDS	mg/L	689.85	851.10	512.32	93.02
EC	µS/cm	1297.04	1576.21	980.00	147.66
pH	-_	7.70	8.14	7.25	0.31
Ca^{2+}	mg/L	101.11	125.13	71.32	15.63
Mg^{2+}	mg/L	24.95	33.45	12.30	5.95
Na^{+}	mg/L	74.71	97.29	52.54	14.19
K^{+}	mg/L	24.54	40.26	10.91	9.29
HCO_3^-	mg/L	427.02	499.52	229.98	71.12
CO_3^{2-}	mg/L	1.60	3.70	0.25	1.29
$SO4^{2-}$	mg/L	104.19	152.15	48.38	30.01
Cl^-	mg/L	142.32	183.91	110.50	22.45
NO_3^-	mg/L	8.73	13.98	2.58	3.95
PO_4^{3-}	mg/L	1.12	1.53	0.73	0.27
BOD	mg/L	39.71	53.50	24.48	8.19
COD	mg/L	9.11	12.55	6.02	1.92
Fe	µg/L	137.37	193.66	88.56	29.06
Pb	µg/L	15.05	20.54	11.00	2.85
Cr	µg/L	2.58	3.28	2.00	0.50
Cd	µg/L	17.41	25.25	11.78	4.81
Mn	µg/L	63.34	97.00	35.32	19.23
Cu	µg/L	28.90	46.31	12.30	12.02
Zn	µg/L	57.80	75.19	34.58	11.04
CO	µg/L	47.10	66.15	26.88	13.40
SAR	meq/L	1.72	2.08	1.27	0.25
SSP	%	35.34	41.26	29.45	3.85
RSC	meq/L	-0.04	1.14	-1.52	0.96
MAR	%	28.69	39.10	22.14	4.45
Ca: Mg		2.56	3.52	1.56	0.54

$$CBE = \frac{(\sum cations \; \sum anions)}{(\sum cations + \sum anions)}) \times 100 \qquad (1)$$

C. *Irrigation Water Assessment Methods*

There are two kinds of hazards (Public Health hazards and Environmental hazards) engaged in using TWW.

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1) Health Hazards Assessment

This study adopted the pollution indices such as the OPI and the CPI to ascertain water quality for public health (Equ.2). The CPIdetermined using the assessed concentration [mg/L] of parameters for their allowable level for irrigation recommended by [10], [11].

$$CPI = \frac{1}{n}\sum_{i=1}^{n} PI \qquad (2)$$

Where:

$$n = parameters\ number$$

$$PI = \frac{concentration\ of\ individual\ parameter}{permissible\ concentration\ of\ parameter}$$

Water quality categories are:clean: (CPI = 0-0.2), sub clean: (CPI = 0.21–0.4), slightly polluted: (CPI = 0.41-1.0), moderately polluted: (CPI = 1.01-2.0), and severally polluted: (CPI = > 2.01) [12].The OPI is an extensive index of dissolved oxygen, dissolved inorganic phosphorus, COD, and, dissolved inorganic nitrogen to assess the organic matter in an aquatic environment. Depending on its benefit in monitoring organic contamination and water quality status, OPI extended to various water sources [13]–[15]. In the current study, OPI (Equ.3) evaluated using the assessed concentration of NO_3^-, PO_4, BOD, and COD for their allowable level for irrigation recommended by [10], [11].

$$OPI = \frac{NO3}{NO3} + \frac{PO4}{PO4} + \frac{BOD}{BOD} + \frac{COD}{COD} (1)$$

The water quality categories are: excellent (OPI less than zero), good (OPI = 0-1), contaminated: (OPI = 1-2), slightly polluted (OPI = 2-3), moderately polluted (OPI = 3-4), heavily polluted (OPI = 4-5).

2) Environmental Hazards Assessment

The experience of TWW's hydrochemical properties can help manage these waters. The outcomes of the laboratory study clarified using the FAO irrigation water classification [10],the US Salinity Laboratory classification[16]and various chemical standards and considerations for determining the adequacy of agricultural water quality. In addition, the present research uses Piper and USSL diagrams using Easy_Quim5.0_v2012 to comprehend the performance status of the TWW from Misurata. These diagrams may assist to determine the category, faces and cation-anion alternation of the irrigation water.

Salinity Hazards Assessment: Salinity is a frequent issue for farmers irrigating in arid environments. High salinity irrigation water is a plant toxic and poses a risk to salinity. Therefore, the salinity threat measured either by TDS or EC is the most important hydrochemical limit [17].

Sodium hazard Assessment: SARcommonly used as a standard to express the seriousness of the sodium which can indicate to what extent the irrigation water is prone to cation exchange reactions in the soil. Soils with a high sodium concentration are also sensitive to soil structure degradation relative to the concentration of calcium and magnesium. This causes a decrease in the soil's penetrability to water and air, which has a negative effect on soil ventilation and reduces the water available to the plant [18]. The expression of sodium risk performed by calculating the SAR (Equ.4) and SSP (Equ.5) in accordance with [16]. The concentrations displayed in meq/L.

$$SAR = \frac{Na}{(Ca + Mg/2)^{0.5}}$$ (Error! Bookma

$$SSP\% = \left(\frac{Na + K}{Na + K + Ca + Mg}\right) \times 100 \qquad (5)$$

Carbonate and bicarbonates hazard Assessment: Calculation of RSC values used to predict the critical effect of sodium on water quality [19], which is associated with $CaCO_3$ and

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$MgCO_3$ precipitation. The concentrations displayed in meq/L(Equ.6).

$$RSC = (CO3^2 + HCO3^-) - (Ca^2 + Mg^2) \qquad (6)$$

Magnesium hazard (MH) Assessment: MH is an important measurement tool for irrigation water quality [20]. The concentrations displayed in meq/L(Equ.7).

$$MAR = \left(\frac{Mg}{Ca + Mg}\right) \times 100 \qquad (7)$$

D. Statistical analysis

Statistical analysis performed using the IBM SPSS Statistics 23 Program for mean, maximum, minimum, and standard deviations.

3. Results And Discussion

A. Human Health Risks

Irrigation using wastewater could have beneficial and/or adverse effects on public health. The beneficial impacts are related to food safety, enhancing the quality of life by increased income in poor regions. While the negative effects result from the presence of pathogens and organic and/or inorganic toxic compounds. Thus, irrigation with TWW poses a range of potential hazards to social health through consumption or exposure to pathogenic microorganisms, heavy elements, and harmful organic compounds [21]. About chemicals in wastewater, heavy metals are the primary concern for health. Many of them have biological benefits in tiny amounts but dangerous at elevated exposure concentrations. Some heavy metals (Mn, Fe, Cu, Co, Ni, and Zn), which play a vital role in enzyme structure and other proteins, are essential to life in trace amounts.Other heavy metals (Cd, Sb, Cr, Pb, As, Co, Ag, Se, and Hg) are non-essential, they affect the metabolism of essential metals. Heavy metals, whether or not essential, cause serious harm to organisms at high concentrations [22].

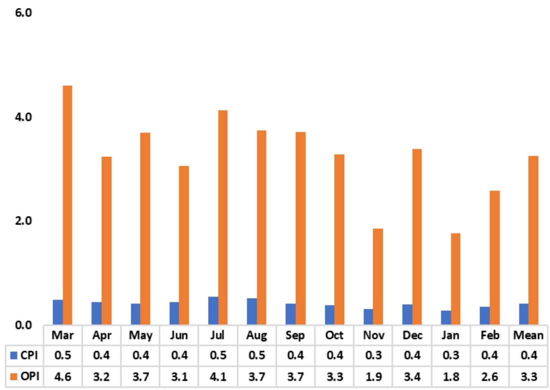

Figure 0.3 Monthly results of CPI and OPIindexes of the selected TWW quality parameters.

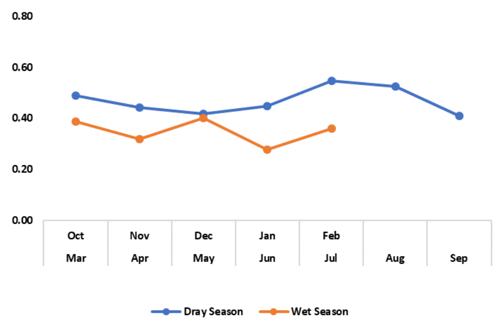

Figure 0.4 Seasonally variations of the CPI pollution index.

The specific impact, however, relies on the compound sort, its concentration, and the exposure path and length. The study did not cover Co, Zn, and Cu heavy metals, where crops are unlikely to consume enough of them to prove that they are hazardous to customers and toxic to crops before they reach humanly toxic levels [23]. Therefore, measured data of parameters (NO_3^-, PO_4, BOD, COD, Pb, Cr, Cd) have served to evaluate OPI and CPI for the assessment of human health hazards and wastewater pollution (Figure 0.3**)**.The CPI values found to be in the category of slightly polluted water quality (CPI range 0.41–1.0) during all sampling months. Nearly the same OPI average found in range 3 to 4 i.e. slightly to moderately polluted water quality during all sampling months.TWW quality is obviously much worse in the summer season (Jun to Sep). Pollution is more likely to happen in the dry season than in other

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seasons.Figure 0.4. shows the seasonally calculated CPI to verify the water quality change during the wet and dry season.The estimated CPI in the dry season is obviously higher than in the wet season. It shows that the wastewater network receives more amount of pollutes in the dry season and has less dilution especially due to rainwater.

B. Environmental Risk Assessment

The pH values ranged from 7.25 to 8.14 with an average value of 7.70, showing a slightly alkaline nature of TWW. Irrigation water with a pH outside the normal range (6.5 to 8.4) may induce a deficiency of nutrients or may involve toxic ions [10]. The average concentrations (mg/L) of cations were: Ca2+> Na+> Mg2+> K+, and the average anion rate was: HCO3->Cl->SO42->NO3->CO3-(Table All.(1.II cations and anion levels in all samples were within the adequate limits according to FAO irrigation guidelines.The average [10]water classification :concentrations (µg/L) of trace elements were <Fe>Mn>Zn>CoCu>Cd>Pb>Cr. Among those elements, Cd average concentration was higher than the desirable limit of 10 µg/L for irrigation water [10]. Cd is one of the most poisonous among all heavy metals, non-biodegradables and persistent in living organisms [24]. The major source of environmental cadmium emissions comes from manufacturing wastewater stream, such as electroplating, metal processing, battery, and circuit board industries [25]. Must caution in dumping these wastes and/or using more efficient treatment methods to remove toxic and dangerous components.

1) Salinity and Alkalinity Risks Assessment

The EC values of the tested samples ranged from 980,00 to 1576.21 µS/cm (average = 1297.04 µS/cm), while the TDS values ranged from 512.32 to 851.00 mg/L (average = 689,85 mg/L) which indicate a slightly to moderate limit (permissible) to irrigation of this TWW according to [10], [16]. Results also demonstrate that this water is viable for soils with minimal drainage, along with salinity regulation and salt-tolerant crop choice. However, irrigation waters of EC higher than 3000 µS/cm and TDS higher than 2000 mg/L, significantly influences physical and chemical soil properties [17], which could affect the suitability of a soil as a plant growth medium [10]. The validity assessment of specific water for irrigation should not rely only on salt concentration as a single criterion. Sometimes, it may still be appropriate to use water for irrigation even if it contains a high concentration of salt without causing adverse soil and plant damage. Regarding the use of waters with a high SAR, [10] and [26] have shown not to decrease soil permeability if these waters combined with high salinity. The effects of higher SAR increase with lower water salinity [26]. As a result, SAR waters considered, in combination with its EC, to provide a superior way of predicting the soil impact of water. USSL [16], used the integrated effect of water salinity and alkalinity hazard, as a foundation for assessing water quality for watering. The TWW EC and SAR plotted on the salinity diagram. Figure 0.5 shows that all the TWW samples come into the high - salinity class and low risk of exchangeable sodium (C3-S1). Water with high salinity (C3) is not suited for regular irrigation. Damage can occur to plants with a sensitive salinity tolerance crop. However, its successful use requires salt-tolerant plants, good soil drainage,excess irrigation, and/or regularuse of low salinity water [16].

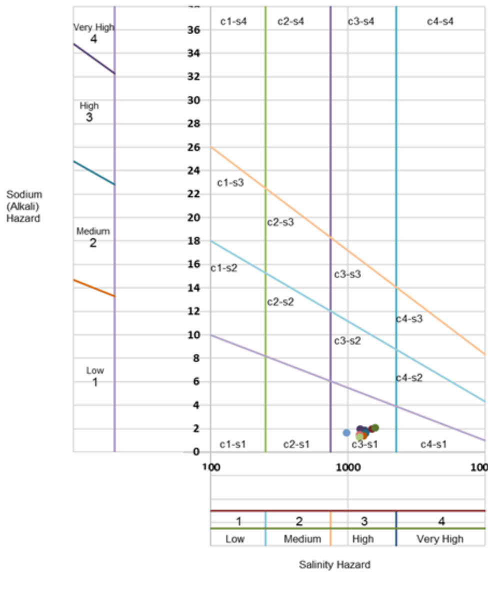

Figure 0.5 TWW suitability for irrigation in accordance with the USSL scheme.

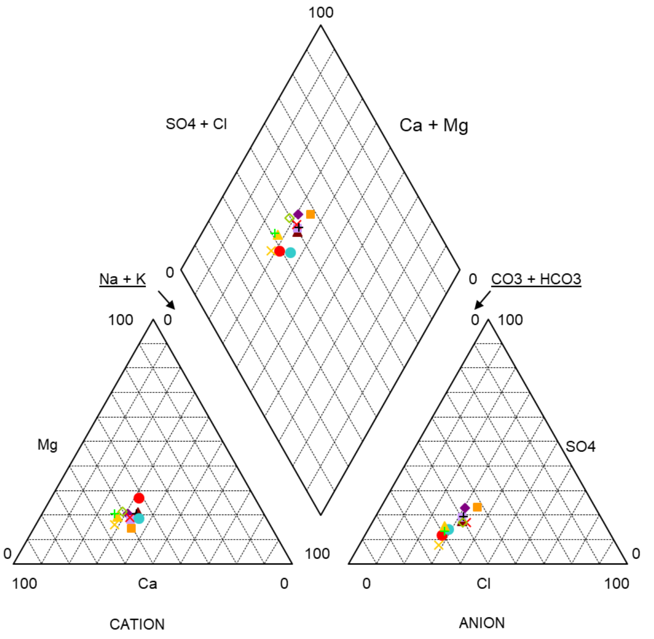

Figure 0.6 TWW type classification according to Piper (1944).

2) Sodium Hazard

Na^+concentration in the TWW samples varied from 52.54 to 97.29 mg/L (average = 74.71 mg/L), indicating a non- restriction (<100 mg/L) on the use of this TWW in irrigation [10]. The SARs in terms of sodium/calcium and magnesium ratio is a critical factor for the determination of the irrigation water suitability, as they are closely related to the percentage of sodium exchangeable in the soil than the percentage of sodium [16]. The SAR value of the TWW samples ranges between 1.27 and 2.08 meq/L with a mean of value = 1.72 meq/L. All the TWW samples fall within the water class grade "excellent. Irrigation waters with SAR < 10 are deemed to be of excellent quality, while waters with SAR values above 10 mainly in low terms of leaching, lead to problems on fine soils and plants sensitive to sodium, [16]. The calculated SSP values varied from 29.45 to 41.26% with an average of 35.34% (Table 0.1), intimating a low grade of restriction (<20%) on irrigation use of such TWW.SSP above 20 percent can lead to Na^+ buildups which will change the physical properties of the soil by replacing ions of Mg^{2+} and Ca^{2+}[16].Na^+substitution with $Ca^{2+}$and Mg^{2+}in soil complexes reduces permeability and leads to low soil drainage [10], [27], which could also influence sustainability in the long - term use of alkaline waters [28]. Salinity and sodium are stated to be the foremost water quality interests in irrigated areas receiving such water [29]. In low rainfall and high evaporation areas, irrigation with saline-sodic water can raise soil sodicity.

3) Carbonate and Bicarbonates Hazard

The RSC values of the tested samples ranged from -1.52 meq/L to 1.14 meq/L with an average RSC value of -0.04 meq/L **(**Table 0.1**).** About 45% of the samples reported negative values which demonstrated that the content of dissolved $Ca^{2+}$and $Mg^{2+}$was higher than the content of CO_3^- and HCO_3^-. All the TWW samples fall within the water class grade "safe" and are well suitable for irrigation. Irrigation waters with RSC less than 1.25 meq/L are deemed to be of the safe class grade, while waters with RSC values above 2.5 meq/L is unsuitable for irrigation [19]. High-water content of RSC increases the adsorption of sodium in the soil whereas excess carbonate in soil solution will enhance the precipitation of $Ca^{2+}$and Mg^{2+} as carbonates which increases sodium in solution and affects plant nutrient uptake. The high pH associated with the high level of carbonate makes irrigation with such water cause degradation of soil fertility because of sodium carbonate deposition [19].

4) Magnesium Hazard (MH)

Magnesium and calcium usually maintain their balance in most waters, they are both a major component of plant nutrients and are simultaneously associated with soil particle aggregation and friability, thus considered a key quality criterion [16]. High concentrations of Ca^{2+} and Mg^{2+} ions can increase soil pH and thus decrease the availability of phosphorus in the soil [27]. The calculated MH values ranged between 22.14% and 39.10% with a mean value of 28.69% **(**Table 0.1**)**. According to the MH class grade [20], the TWW samples are suitable for irrigation (MH<50%). The outcome supports that Ca: Mg molar proportion in almost all TWW samples was >1 (range 1,56 to 3,52, mean 2,56). [30], [31] early suggestedthat the water in Ca: Mg molar ratio <1, results in an increase of SAR values which adversely affect soil and plant production as the soils become salty and salty. The result suggests

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that calcium and magnesium are unlikely to precipitate in water irrigated soil by CO_3^- and HCO_3^- as $CaCO_3$ and $MgCO_3$which lead to reduced-sodium risk and related problems.

5) Chemical Classification and Hydrochemical Facies

Using the Pipers diagram helps to understand the hydrochemical issues associated with water resources. The diagram comprises three separate fields, two of which are triangular and one is a diamond. Anions stated in meq/l on the right triangle as the total anions %, while the cations in the left triangle [32]. Then each point in the upper field falls along a line parallel to the upper field margin and the point where the extension overlaps with the water character defined by the relationship between, Cl^-, CO_3^-+HCO_3^-, SO_4^{2-}, Ca^{2+}+Mg^{2+}, and Na^++K^+ ions. The trilinear diagram can show similarities and differences between water samples as the water of similar qualities plots together as clusters. TWW samples classification in the Piper chart showed that most of the TWW samples (about 83.3%) fall into the calcium bicarbonate zone of the diamond-framedfield, indicating a Prevailing water type of calcium bicarbonate. The plot showed Ca^{2+} plus Mg^{2+} alkaline earth are the dominant alkalis making up about 64% while Na^+ plus K^+ alkalis make up about 35.3%, and weak acids represented by CO_3^-+HCO_3^- exceed strong acids represented by Cl^- plus SO_4^{2-} which are constituting about 53% and 47% (Figure 0.6).Alkaline earth (Ca^{2+}+Mg^{2+}) and soft acids (CO_3^- + HCO_3^-) subjugate the hydrochemical properties of most TWW samples. For irrigation, the pH level of all TWW samples was lower than the prescribed limit of 8.4 [10]. High pH levels above 8.5 are frequently attributable to the high contents ofHCO_3^- and CO_3. The high carbonates and the bicarbonates make Ca^{2+} and Mg^{2+} ions insoluble, leaving sodium the predominant solution ion [33].

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