

Evaluation of Fluoride Contents of Kirkuk City's Drinking Water and Its Source: Lesser Zab River and Its Effect on Human Health

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Abstract—In this study, forty samples had been collected from water of Lesser Zab River and drinking water to determine fluoride concentration and show the impact of fluoride on general health of society of Kirkuk city. Estimation of fluoride concentration and determination of its proportion in water samples were performed attentively using a fluoride ion selective electrode. The fluoride concentrations in the Lesser Zab River samples were between 0.0265 ppm and 0.0863 ppm with an average of 0.0451 ppm, whereas the average fluoride concentration in drinking water samples was 0.102 ppm and ranged from 0.010 to 0.289 ppm. A comparison between results obtained with World Health Organization (WHO) show a low concentration of fluoride in the samples of the study. Thus, for health concerns we should increase the concentration of this ion in water of Kirkuk city at least to about (1.0 ppm) and this will take place after fluorination process.

Keywords—Fluoride concentration, Lesser Zab River, drinking water, health society, Kirkuk city.

I. INTRODUCTION

FLUORINE is the lightest member of the halogen group. It is the most electronegative of all chemical elements and is common element that does not occur in the elemental state in nature because of its high reactivity [1]. Fluorine in the environment is found as fluorides which represent about 0.06–0.09% of the earth's crust. [2]. Reference [3] explains that the fluorine exists in rocks, soil, water and the biological chains of animal and plant lives. It is considered as an essential micronutrient for both humans and animals. And it is also considered as a contaminant in many industrial activities prone areas such as semiconductors and phosphate fertilizers [4].

Concentration of fluoride in the environments varies according to the nature of the rocks and the occurrence of fluoride bearing minerals. Seawater typically contains about 1 mg l⁻¹ while rivers and lakes generally exhibit concentrations of less than 0.5 mg l⁻¹. In waters, concentrations are limited by fluorite solubility, so that in the presence of 40 mg l⁻¹ calcium it should be limited to 3.1 mg l⁻¹ [1]. It is the absence of calcium in solution which allows higher concentrations to be stable [5]. High fluoride concentrations therefore are expected

in ground waters from calcium poor aquifers and from areas where fluoride bearing minerals are common. On the other hand, the concentrations of fluoride may also increase in ground waters in which cation exchange of sodium for calcium occurs [5].

Fluoride in accurate quantity is an essential component for formation of dental enamel and normal mineralization of bones [6]. However, fluorosis caused by excessive intake of F⁻ has been a considerable health problem worldwide, which is afflicting millions of people in many areas of the world, for example East Africa [7]-[9], India [10], [11], and northern China [12]-[17]. The acceptable maximum concentration of F in drinking water is 1.5 mg l⁻¹ [18] and recommended safe limit is 1.0 mg l⁻¹ by the WHO [19]. Due to the threat to human health, many studies have been conducted on water fluoride such as fluoride source [4], [3], [20] and its removal methods [21], [15], [22]. Moreover, some researches about the high F content of tea products confirmed that fluorosis disease had a close relationship with the drinking and eating habits of inhabitants [13], [14], [23], [24]. However, little attention has been paid on the spatial distribution of soil fluoride concentrations on the regional scale.

Generally, fluoride is a typical lithophile element under terrestrial conditions. The bulk of the element is found in the constituents of silicate rocks, where the complex fluorophosphates like apatite (Ca₅(PO₄)₃F), seems to be one of the major fluoride minerals. Fluoride in the ground/surface water derives from the weathering and subsequent leaching of fluoride bearing minerals in rocks and soils like Fluorspar [CaF₂], Cryolite [Na₃AlF₆] and Fluorapatite [Ca₅(PO₄)₃F] and clay minerals which are the major sources of F⁻ in the groundwater [25]-[27]. In the study area, the clay fraction is dominantly controlled by Palygorskite and kaolinite, with only small amounts of chlorite and smectite [28].

The present study was conducted to: (i) Estimate water quality by determined physical and chemical properties of waters and estimate the upper baseline concentration of fluoride in surface water of Lesser Zab River and drinking water in Kirkuk City; (ii) Evaluate controlled factors that decided the spatial distribution of water fluoride concentration using a comparison with the permissible values given by the WHO; and (iii) Clarify their health effective on society.

II. GEOLOGICAL AND HYDROLOGICAL OUT LOOK

The studied area is situated in Kirkuk city and covers an area of 20000 km² between longitudes 44° 22' 12" to 44° 24'

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36' E and latitudes 35° 22' 12' to 35° 30' 36' N (Fig. 1). The area in general is a flat plain, but it also includes some undulating land containing relief due to many seasonal streams. The area is covered by recent sediments and sedimentary rocks. The geological formations outcrop has a large extension ranging in age from Miocene (Fatha formation) to Quaternary sediments. The common rocks are limestone and clastics with the presence of gypsum rocks. [29].

The study area lying within the Foothill Zone consists mainly of Upper Miocene and Pliocene coarse detrital sediments which are gently folded along the NW–SE axis parallel to the structural trend of the Zagros Mountains [30]. These detrital sediments expose at the distance of 5 km along the northeastern part of the studied area. The Lesser Zab River

(LZR) is located in the northeastern part of Kirkuk city, which represents the main source of surface drinking waters in Kirkuk. The LZR passes through a variety of geological formations aged from Jurassic (Qulqula Series) to Pliocene (Mukdadiyah and Bi-Hassan Formations). The Khasa Chay River drains through the Kirkuk city, which is a seasonal river originated in the hilly area on the north of city. This river is considered as an important surface water recharging the deep water resources in the plain, while the shallowest depth wells were drilled at the terraces and the alluvium plain of this stream. The water bearing formation are thick beds of Bai Hassan formations (mainly consists of gravely and pebbly silt, clay and sand partially act as cementing material) while the old and recent alluvium is composed of thick layers of gravel, sand and strips of clay are varying vertically and laterally.

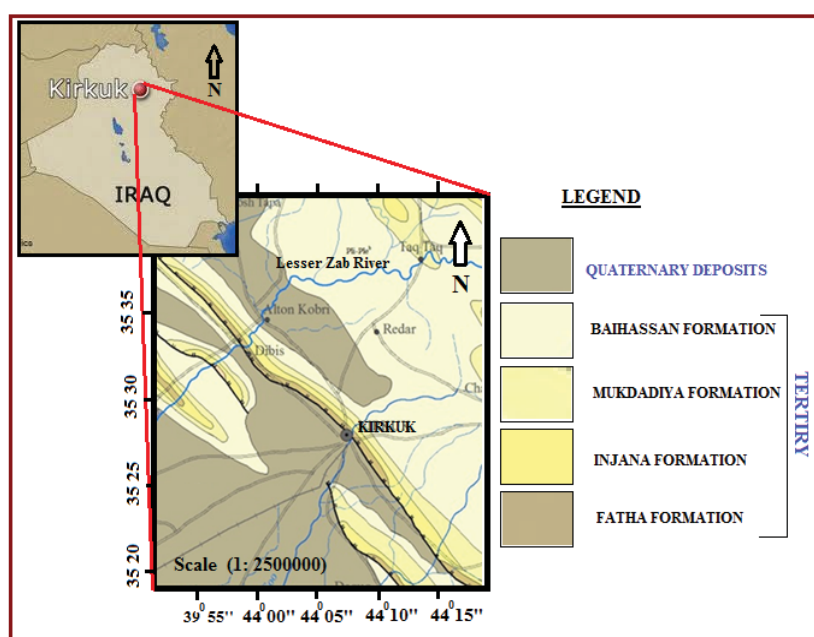


Fig. 1 Geological map of the studied area modified from the [31]

III. MATERIALS AND METHODOLOGY

Forty surface water samples were collected from Lesser Zab River at an average density of 1 sample per km² and the rest were taken from drinking water of different parts of Kirkuk province (Fig. 2). Before collecting water samples, the drinking water was allowed to flow for about 15 minutes to ensure that pipes were cleaned. Samples were refrigerated at 4 °C from the time of collection until completion of all analysis and water treatment devices of any kind were avoided. [32].

Physical properties of water samples including temperature, pH, TDS, tur and conductivity were determined on site at the time of sample collection. Temperature and pH was determined using pH meter (Model 720, WTW). Conductivity and TDS were measured using Conductivity and TDS meter (Model HI99300, HANNA). Turbidity was measured using Turbidity meter (Model TB 210 IR, Lovibond).

Fluoride concentration in water samples was determined with a combination fluoride selective electrode (ISE-BNC)

against silver/silver chloride reference, after adding total ionic strength adjustment buffer (27503-13 F), coupled with anion analyzer (WTW, model inoLab pH/ION 735, GmbH Company). Owing to its simplicity and short analysis time, the fluoride ion selective electrode is widely used for the determination of F⁻. Before determining the fluoride contents, equal volume quantities of total Ionic Strength Adjustment Buffer (TISAB) were mixed with all samples. The TISAB contains an acetic acid/acetate buffer that fixes the pH of the solution at about 5. At this pH, the formation of hydrogen fluoride (HF) and the concentration of (OH⁻) are negligible. Other anions detectable by the electrode are insignificant. It also contains NaCl to create a high and constant ionic strength and a complexing agent that removes cations that could interfere by forming complexes with fluoride. Fluoride standards ranging from 0.020-0.500 mg L⁻¹ fluoride were used to calibrate the measurement.

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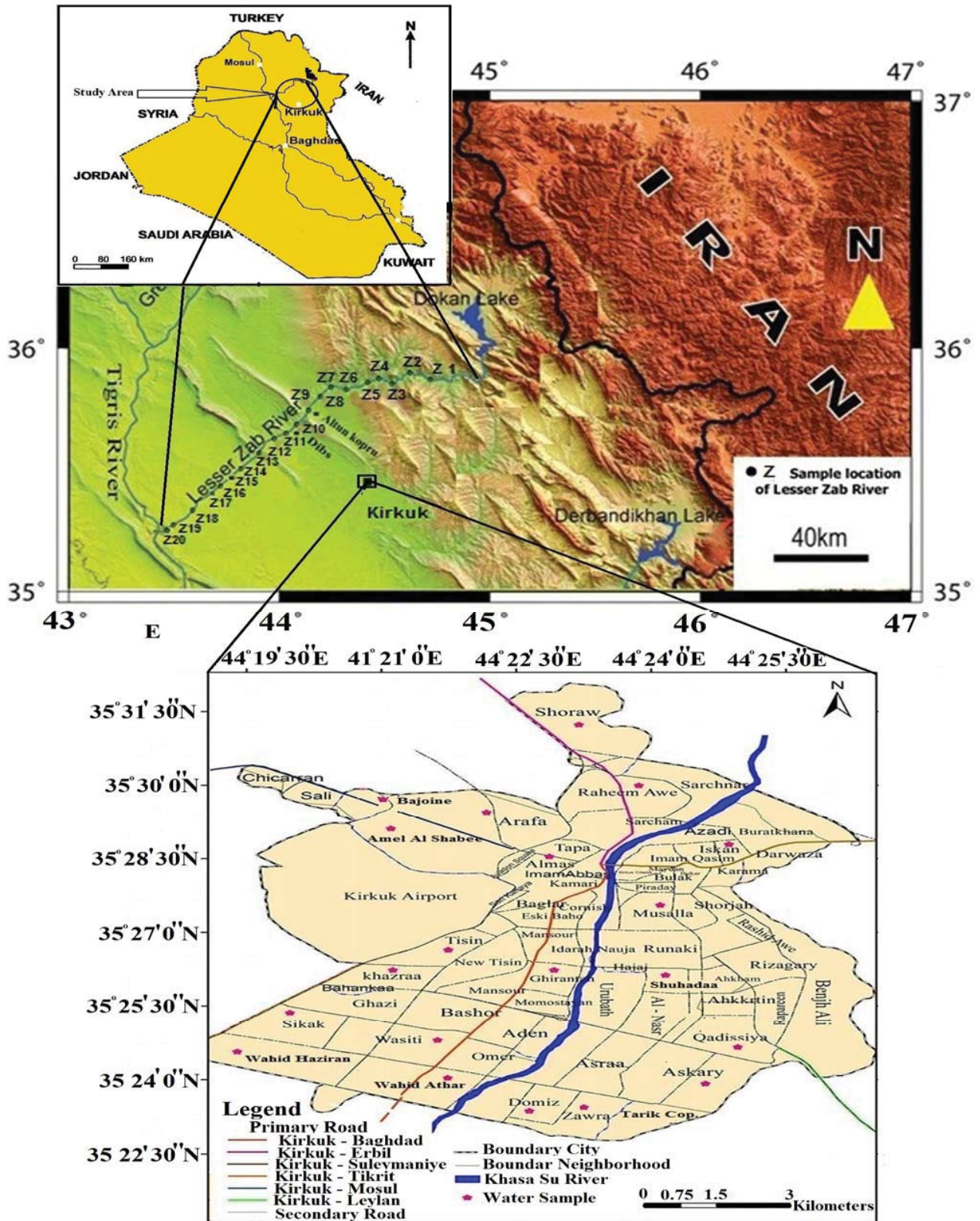


Fig. 2 Locational map of the studied samples

IV. RESULT AND DISCUSSION

The physical and chemical proprieties of waters that were taken from LZR are presented in Table I. The pH value of aquatic system is an important indicator of the water quality and the extent pollution in the water shed areas [33], [34]. The pH of river water that was not affected by contamination typically ranges between (6.5) and (8.0); and for fish growth and survival, the pH should remain in the range of 6.5-9.0 [1]. The pH values for the LZR samples fall between (7.41) and (8.64) and the average value was (8.03), Table I, whereas the pH values for drinking waters fall between (7.7) and (8.14) and the average value was (7.91), Table II, which indicates that waters are slightly alkaline in nature and all the water samples are within permissible limit prescribed by WHO [35], [36].

The ability of water to conduct an electrical current was measured to determination of salinity. Salinity is expressed in two different ways, either as electrical conductivity (EC) or as total dissolved salts (TDS). The EC values ranged from 242 to 402 $\mu\text{s}/\text{cm}$ with an average value of 311.25 $\mu\text{s}/\text{cm}$, the values of TDS varied from 124 to 182 ppm and the average value was 142 ppm as shown in Table I. Meanwhile, the WHO and the European standards recommended value of EC is 250 $\mu\text{s}/\text{cm}$, and all the results are under the acceptable Iraqi limits (EC = 400 $\mu\text{s}/\text{cm}$) [37]. In general, EC and TDS displayed the same trends along the LZR, whereas the variation in EC and TDS values are related with the rate of river flows, weathering and dissolve different rocks that were exposed in catchment area [38].

The values of conductivity varied from 280 to 621 $\mu\text{s}/\text{cm}$, TDS values ranged from 140 to 311 ppm with an average value of 187.25 ppm shown in Table II. The most desirable limit of TDS in drinking water is 500 as per WHO standard, and all the samples are under this limit. Thus the conductivity showing positive relationship with dissolved ion concentrations may be explained by the increased of Lesser Zab River (source of drinking water in Kirkuk city) discharge rates, which lead to decreases of dissolved ion concentrations in the water, and also the reference [39] explains that the physical and chemical properties of water depend entirely on the tap water pipeline system and their difference and qualities.

Turbidity refers to water clarity. The greater the amount of suspended solids in the water, consequently the turbidity values will be higher. Turbidity values ranged from a minimum of 1.21 to a maximum of 41.10 with an average value of 8.84 (NTU) as shown in Table II. According to drinking water standard (WHO) and Canadian water Quality Guidelines for the Protection of Aquatic Life (CCME) (5.0 NTU) [40], turbidity in drinking water was not acceptable, whereas the high values of turbidity in some region results from increment concentrations of suspended materials in this region.

The concentration of fluoride in the LZR samples ranged from 0.0265 to 0.0863 ppm with an average level 0.0451 ppm, Table III. The highest fluoride concentration was recorded in to the junction point of LZR with Tigris River (Fig. 2). That is

due to water contents and presence rates of fluoride concentration in to the soil and sediments nearest the river edge. While the concentration of fluoride in Kirkuk's drinking water was 0.010-0.289 ppm with an average 0.102 ppm (Table III). These low levels showed similar trend with studies performed outside Iraq for unpolluted drinking water, which were usually considered to be insufficient to prevent caries by WHO [41]-[47].

TABLE I
 PHYSICAL PARAMETERS OF THE LZR WATERS

No.	Sample location	pH	EC (μs)	TDS (ppm)
1	Z1	7.92	262	125
2	Z2	7.82	255	124
3	Z3	8.04	389	126
4	Z4	8.31	242	130
5	Z5	7.58	388	158
6	Z6	7.76	402	182
7	Z7	8.13	245	131
8	Z8	7.95	275	145
9	Z9	7.41	244	166
10	Z10	7.92	312	138
11	Z11	8.05	372	129
12	Z12	7.89	342	132
13	Z13	8.24	299	154
14	Z14	8.02	395	125
15	Z15	8.56	289	149
16	Z16	7.62	310	126
17	Z17	8.42	285	133
18	Z18	7.63	277	127
19	Z19	8.64	315	178
20	Z20	8.62	327	162
	Minimum	7.41	242.00	124.00
	Maximum	8.64	402.00	182.00
	Average	8.03	311.25	142.00

A comparison between results obtained with the desirable and maximum permissible limit for fluoride in drinking water determined by WHO and Europeans' Directive (1.5 mg L^{-1}) or by Bureau of Iraqi Standards (1 mg L^{-1}) [48], [49], shows that the fluoride levels of Kirkuk City's drinking water and may be that of whole Iraq are rather low. This is in agreement with that recorded in Babil City, Iraq [50].

The fluoride concentrations in the above investigated waters were lower than what is required for the human health according to global specifications (1.0 ppm). If there is not enough fluoride content within the water, this may result in tooth decay and dental caries [51]. However, if there are high concentrations of fluoride within the water, this may result in dental and skeletal fluorosis [51]. The severity depends upon the amount ingested and duration of intake. Dental fluorosis is a condition where excessive fluoride can cause yellowing of teeth, while spots and pitting or mottling of enamel. Consequently, the teeth become unsightly. A bone disease exclusively caused by excessive consumption of fluoride is called skeletal fluorosis, which depending on the degree of fluorosis can cause increase in bone mass, stiffness in joints and osteoporosis [51].

TABLE II
PHYSICAL PARAMETERS OF DRINKING WATERS

No.	Location	pH	EC (μ s)	TDS (ppm)	Turb (NTU)
1	Domiz	7.93	347	173	8.09
2	Askary	7.76	360	179	7.78
3	Qadissiya	7.82	502	248	6.73
4	Shuhadaa	8.08	296	148	4.91
5	Musalla	8.14	402	201	7.79
6	Iskan	7.82	377	188	7.88
7	Shoraw	7.81	413	208	15.7
8	Almas	7.82	375	188	5.1
9	Arafa	7.99	362	181	2.76
10	Tisin	8.02	339	169	3.47
11	Khazraa	7.71	359	179	4.9
12	Wasiti	8.04	320	159	1.21
13	Wahid Athar	7.9	310	155	11.1
14	Ghirantah	8.05	334	167	8.18
15	Wahid Haziran	7.7	417	208	7.22
16	Amel Alshabee	7.91	362	181	15.4
17	Sikak	7.88	621	311	2.98
18	Raheem Awe	8.05	280	140	7.14
19	Zawra	7.8	370	185	7.34
20	Bajoine	7.88	354	177	41.1
	Minimum	7.7	280	140	1.21
	Maximum	8.14	621	311	41.1
	Average	7.91	375.00	187.25	8.84

TABLE III
FLUORIDE CONTENT IN LZR AND DRINKING WATER SAMPLES

Sample No.	River Water	F ⁻ (ppm)	Drinking Water	F ⁻ (ppm)
1	Z1	0.0332	Domiz	0.118
2	Z2	0.0311	Askary	0.05
3	Z3	0.0286	Qadissiya	0.091
4	Z4	0.0387	Shuhadaa	0.01
5	Z5	0.0434	Musalla	0.106
6	Z6	0.0461	Iskan	0.088
7	Z7	0.0463	Shoraw	0.01
8	Z8	0.0525	Almas	0.091
9	Z9	0.0482	Arafa	0.09
10	Z10	0.0375	Tisin	0.06
11	Z11	0.0321	Khazraa	0.04
12	Z12	0.0362	Wasiti	0.087
13	Z13	0.0265	Wahid Athar	0.096
14	Z14	0.0468	Ghirantah	0.13
15	Z15	0.0345	Wahid Haziran	0.129
16	Z16	0.0519	Amel Alshabee	0.289
17	Z17	0.0501	Sikak	0.197
18	Z18	0.0561	Raheem Awe	0.128
19	Z19	0.0863	Zawra	0.116
20	Z20	0.0763	Bajoine	0.114
	Min.	0.0265	Minimum	0.01
	Max.	0.0863	Maximum	0.289
	Average	0.0451	Average	0.102

At drinking water concentrations 0.9–1.2 mg/L, fluoride may give rise to mild dental fluorosis. Values of 1.5–2 mg/L of fluoride in drinking water gives rise to higher chance of dental fluorosis, while values exceeding 2 mg/L may have very high chance of dental and skeletal fluorosis [52]. Total fluoride intakes above 6 mg/day have been shown to increase

the effects on the skeletal, while fluoride intakes above 14 mg/day pose serious threats of severe skeletal effects [52]. On the basis of these directives and according to [53], we recommended that Iraq's drinking water be fluoridated to the required level of (1.0 ppm). According to climatic conditions that prevail in Iraq, the optimal fluoride concentration varies between 0.5 and 1.0 mgL⁻¹ being generally recommended [50].

V. CONCLUSION

1. The pH values for the LZR indicate that water is slightly alkaline in nature and all the water samples are within permissible limit prescribed by WHO.
2. The results of EC and TDS are under the acceptable Iraqi limits. Thus, EC and TDS display the same trends along the LZR, whereas the variation in EC and TDS values are related to the rates of river flow, weathering and different rock type's dissolving in the catchment area.
3. LZR is the main source of drinking water of Kirkuk city, Iraq. After evaluating the fluoride concentration by ionic selective electrode, it is found that the level of fluoride in both LZR and drinking waters is far below the upper level recommended by WHO and by Bureau of Iraqi Standards and global dental Protect organizations. It is recommended that Kirkuk City's drinking water be fluoridated to the global standard to prevent tooth decay. Fluoride also strengthens bones hence reducing spinal bone fracturing ratio and osteoporosis in old women. Fluoride is also known to increase children's Intelligence Quotient.
4. The result of the present study as well as other available data from water quality should be taken into account when developing strategies for safe drinking water supplies.

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