A Quantitative Determination at the Health Risk Assessment of Fluoride Content: Turkey, Ceyhan Basin Case Study

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Abstract: - The increase in fluoride content in surface water causes negative effects on health. For this reason, it is important to routinely perform health risk analyses and update fluoride data. This study aims to compare the fluoride content in some surface waters used for drinking and irrigation purposes in the Ceyhan basin, located in the Mediterranean region of Türkiye, and to assess the potential health risk of fluoride intake in age groups (men, women, children, and infants). Quantitative determination of fluoride content in water was determined on a spectrophotometer following the SPADNS colorimetric method. Health risk assessments of age groups were calculated for each region. In general, fluoride concentrations were found to be between 0.04 and 1.91 mg/l, with an average of 0.715 mg/l. The mean estimated daily intake (EDI) was calculated for the four age groups as 0.033, 0.038, 0.039, and 0.269 for men, women, children, and infants, respectively. While the non-carcinogenic health risk value for infants is greater than 1, this value poses a moderate risk for children. It may be important in the future to take various precautions against the consumption of water specified in these age groups with high metabolic rates.

Key-Words: - Age groups, fluoride, health risk, quantitative, spectrophotometer, water.

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1 Introduction

Good-quality drinking water is among the indispensable and primary needs of public health all over the world, [1]. The quality of water in terms of quantity and characteristics affects the general structure of the environment and human health, [2]. On the other hand, changing climatic conditions, increasing human population and activities, and environmental pollution factors cause changes in the quality of water, which is important for all organisms, [3]. Considering public health and livestock, it is critically important to determine the fluoride levels in surface waters and to keep the data up-to-date, [4]. It has been reported that the areas where natural fluoride poisonings are encountered in Türkiye are mostly areas with volcanic land structures and areas containing fluoride reserves, [5]. It is known that chronic fluoride poisoning, known as fluorosis endemic disease, occurs when fluoride exceeds the required limits in the organism, [6]. Fluoride concentrations in unpolluted freshwater have been reported to typically range between 0.01 and 0.3 mg/l, [7]. However, the fluoride concentration in drinking water in Türkiye was reported to be approximately between 0.9 and 1.7, [8]. It is known that fluoride has an extremely important and interesting function in the biological systems of organisms, [9]. However its long-term excess fluoride calcifies in teeth and bone tissue and affects the regulations of these tissues and structural areas such as the thyroid gland, [10]. Nevertheless, tooth mottling and skeletal fluorosis occur as a result of increased fluoride intake in arid and semi-arid continental areas. This is due to the fact that the amount of fluoride intake naturally increases due to excessive water consumption in hot places, while this amount decreases in cold places, [11]. As such, it is advisable to exercise caution when consuming it and to consider the fluoride content of the water, [12].

Although there are still open-ended questions that need to be investigated in studies on the geochemistry of fluoride in inland waters, especially in public health, it is important to keep the data up-to-date on this subject, [13]. Within the boundaries of Osmaniye province, tributaries of the Ceyhan River provide drinking and irrigation water to people living in densely populated areas like Osmaniye city center, Kadirli, Sumbas, and Düziçi, [14]. Nonetheless, the presence of long heat waves in recent years indicates that the wetlands in the region may face the problem of periodic drought, [15].

In general, exposure to pollutants for public health occurs through the ingestion, inhalation, and surface contact of chemicals in water. In this regard, sequential models such as the definition of the substance that poses a risk, the dose-response status of the chemical, exposure assessment, and the category of the observed risk have been deemed appropriate in public health risk assessment, [16]. The main aim of this study is to determine the fluoride content in the surface water of the Ceyhan basin, located within the borders of Osmaniye province of the Mediterranean region, and to evaluate the health risk for various age groups according to the water resources of districts.

2 Materials and Methods

2.1 Study Area

Osmaniye province, which is located in the easternmost part of Çukurova in the Mediterranean region of Türkiye, has a surface area of 3,767 square kilometers. Overall, 42% of its land is forested, 39% is planted agricultural land, 17% is unsuitable land for agriculture, and 2% consists of other lands.

The province is 121 m above sea level and 20 km away from the Mediterranean Sea. The rock structure of the Osmaniye region reflects the distinctive characteristics of the Cambrian-Tertiary range of the Taurus Mountains, making it a geologically exceptionally unique place. Carbonate and ophiolite formations from Mesozoic units, as well as coral limestone, sandstone, and siltstone shales from Paleozoic units, are also observed, [17]. This region has a Mediterranean climate with hot, semi-arid summers and mild, rainy winters, [18].

In midwinter, the temperature ranged from -5 to 29 °C, while in the summer season, it ranged from 11 to 44 °C, [19]. As of the end of the pandemic period, the total population in the 7 districts of Osmaniye was determined to be 553,012 people. Kadirli, Düziçi, Sumbas, and Osmaniye city centers are the most densely populated areas, [20].

A series of water samples were taken to determine the fluoride levels in water sources in Osmaniye province (Figure 1, Appendix). The samples were taken from Hamus's stream in the center of Osmaniye, Savrun stream in Kadirli district, Sumbas stream in Sumbas district, and Karasu stream in Düziçi district. GPS-data software (Examobile, Poland) was used to determine the coordinates and altitudes of the sampling areas. Water quality parameters such as temperature (T, °C), pH, and electrical conductivity (EC, µS/cm) of water sources were measured with a probe meter (EcoSense, YSI, USA). Some geographical features and water quality parameters of the sampling areas are given in Table 1 (Appendix).

2.2 Collection and Analysis of Samples

A total of 60 fluoride analyses were performed at 15 sampling points along each selected stream in the spring season. As a precaution against surface water contaminants, samples were taken from 30 to 50 cm below the shore, [21]. Water samples were preserved in clean 100 ml polyethylene bottles according to the 1060 C Sample Preservation and Storage Method protocol. Collected water samples from the sampling sites were filtered with a syringe filter (0.45 µm, Isolab, Germany) and samples were acidified using ultra-purified nitric acid to pH<2 (HNO_3, purity 67-70%, Thermo Fisher Scientific Inc., USA). According to the American Public Health Association (APHA) guide for sample collection, the samples were brought to the commercial research laboratory to be analyzed in the cold and dark chain as soon as possible and stored in a +4 °C refrigerator, [22]. Spectrophotometry analysis has become a useful and easy technique widely used in the fields of biomedical and food industries for the identification, quantification, wavelength scanning, kinetic models, and enzyme activity of compounds in various samples, [23]. Water samples were taken from sampling points and were analyzed using a Cary 60 UV–VIS Spectrophotometer (Agilent Technologies, CA, USA). Fluoride determination in water samples was determined by the SPADNS colorimetric method, [24]. After the reaction of Zirconium with fluoride, measurement was performed at the maximum absorbance wavelength of 570 nm, depending on the observed color change.

2.3 Health Risk Assessment

The probability of negative effects that potential environmental contaminants and other stressors may have on different human groups, such as locals, employees, and tourists, is the broadest definition of human health risk assessment, [31]. Various health risk assessment calculations have been developed to estimate the potential risk of contamination from contaminated water from oral intake or ingestion, inhalation, and dermal contact, [32]. A detailed description of sex- and age-group-specific health risk parameters in various references used in health risk assessment is shown in Table 2 (Appendix). The hazard coefficient (HQ) was computed by accounting for the estimated daily intake dose (EDI) in order to estimate the non-carcinogenic health risk of the fluoride effect in the surface waters for various age groups (men, women, children, and infants).

Thus, daily fluoride exposure (also known as chronic daily intake, or CDI) from consuming water was determined using Equation 1. While the estimation of the daily intake rate of fluoride is shown by EDI; C indicates the fluoride concentration in the water, and IR is the ingestion rate per unit of time. The exposure frequency of fluoride is denoted by EF. It shows the annual exposure duration of fluoride for ED. ABW indicates the average body weight of a person, and AET stands for the average exposure time. Equation 2 was used to calculate the hazard quotient of fluoride from drinking potentially highly fluoridated spring water. In general, when the reference dose is exceeded, a hazard quotient situation in which toxic effects emerge is likely to occur. The reference dosage of fluoride is indicated by the abbreviation R*f*D. And each chemical R*f*D value is specific. When examining fluoride contents in surface water and groundwater, the generally recommended HQ limit is 1 [33]. If the results of hazard coefficient calculations are determined to be greater than 1 (i.e., HQ > 1), it is stated that the determined age categories may be at a significant health risk due to fluoride pollution.

$$EDI = (C \times IR \times EF \times ED) / (ABW \times AET) \quad (1)$$

$$HQ = EDI / RfD \quad (2)$$

2.4 Data Analysis and Statistics

The analysis of variance (ANOVA), Student-Newman-Keuls (S-N-K) multiple comparisons, and descriptive statistics in JASP software (V 0.17.2.1, JASP Project, University of Amsterdam, Amsterdam, The Netherlands) were applied to evaluate the characteristics of the sampling areas and statistical differences between regions, [34].

Data storage, grouping, and calculation of the health risk assessment of the results were provided via the Microsoft Excel software (Microsoft, USA).

3 Results and Discussion

The average (mean), minimum (min), and maximum (max) levels of the surface water changes obtained for the fluoride parameters in the sampling areas are shown in Appendix in Table 3 and Figure 2.

The order of the average, minimum, and maximum fluoride level values of the surface waters in the basin, ranging from high to low, was determined as Karasu > Savrun > Hamus > Sumbas for the mean values, Karasu > Hamus > Sumbas > Savrun for the minimum values, and Savrun > Karasu > Sumbas > Hamus for the maximum values.

The minimum and maximum fluoride levels detected in the regions were found to be 0.04 and 1.91 mg/l, respectively, in the Savrun stream. Fluoride levels in the samples were evaluated by comparing them with national legislation values such as surface water pollution regulation and water pollution regulation.

It is seen in Figure 2 that the maximum fluoride concentration exceeded 1, which is the limit value of AA-EQS (the Annual Average-Environmental Quality Standard) and MAX-EQS (Maximum-Environmental Quality Standard) of Turkish Surface Water Quality Regulation, [35]. A study conducted in the surface waters of Gökdere valley within the borders of Gümüşhane province in Türkiye found that the maximum fluoride content in surface waters was between 0.19-9.87 mg/l, [36].

Another study examined the spring and surface waters in Kahramanmaraş province (Türkiye) and found that the total fluoride content was 3.79–5.03 mg/l, [39]. This situation has been explained as a kind of natural pollution situation arising from the elements that the waters add to their composition as a result of their interaction with the rocks in the natural process. The maximum fluoride levels in the Hamus and Sumbas streams were 0.86 and 0.97 mg/l, respectively, and did not surpass the limit value according to both the AA-EQS and the MAX-EQS. Additionally, it is acknowledged that all regions had class I water quality due to the fact that the limit values' results for fluoride concentration were ≤1000, [40]. However the analysis of variance showed significant differences in fluoride concentration comparisons obtained from surface waters in different locations ($P \leq 0.05$). These differences in fluoride content in the Savrun and Karasu streams varied statistically from the fluoride content measured in the Hamus and Sumbas streams ($P \leq 0.05$). Fluoride levels are generally found in low concentrations in waters, and itheir presence in aquatic environments increases or decreases depending on geogenic factors or anthropogenic activities, [41].

The estimated daily intake dose (EDI) of fluoride in different locations was categorized for different age groups, and EDI values in all age groups were found in a similar regional order as Karasu>Savrun>Hamus>Sumbas (Table 4, Appendix). The average, minimum, and maximum

EDI values for men were found to be 0.033, 0.006, and 0.063, respectively. These values were 0.038, 0.007, and 0.071 in women. In addition, the EDI value was calculated as 0.039, 0.007, and 0.074 in children and 0.269, 0.051, and 0.509 in infants (Table 4, Appendix). The non-carcinogenic health risks of fluoride for men, women, children, and infants in the sampled regions, especially in the Savrun stream, approached the hazard quotient value of 1 for men, women, and children's groups.

However, in infants, this value remained above the 1 scale in this study (Figure 3, Appendix). In a study conducted in the semi-arid region of southwestern Iran, the estimated HQ value of fluoride intake was calculated for infants (0.01-0.24), children (0.04-1.83), youth (0.07–1.10), and adults (0.05-0.94). The maximum HQ value for children and adolescents was found to be >1, [42]. The results observed in this study were similar to our study at some points. Considering the health risks associated with fluoride concentration, especially for infants and secondarily for children, it can be considered that there is a potential risk for dental and skeletal fluorosis through drinking water intake. In Tianjin, China, the average, minimum, and maximum fluoride concentrations in drinking water were found to be 0.99 mg/l and 0.01-6.30 mg/l, respectively. According to this study, children between the ages of one and four are particularly sensitive to fluoride exposure within this specified range of fluoride concentrations, [43].

In order to prevent fluorosis in the younger generation, various measures must be taken to reduce the intake of fluoride contents in drinking water, [44]. It has been stated that in cooler regions, drinking water consumption will be lower, and more fluoride intake in drinking water may be required in another study. The average fluoride content in spring and groundwater during the hot and cold seasons in West Azerbaijan and Iran was found to be 0.01-3 and 0.01-4 mg/l, respectively, [45].

The classification of the natural fluoride content in drinking water is made possible by the minimum and maximum temperature conditions in annual temperature fluctuations; in other words, temperature changes provide us with preliminary information about the water's drinkability, [46].

According to WHO guidelines, it has been reported that the average fluoride concentration in drinking water in the temperature range of 12.2-14.6 °C is appropriate to be in the range of 0.8-1.5 mg/l, [47]. This is not fully compatible with the temperature and fluoride concentration ranges measured in our study areas (Table 1, Table 3 in Appendix). There may be serious risks associated with the drinkability of water when temperature changes occur in the sampling areas because of the fluoride level equivalency in the temperature scale developed by the Turkish Standards Institute. As a result, different action plans may need to be supplied. Therefore, due to the effect of temperature on water consumption, the fluoride standard in the region should be regulated based on the current maximum annual temperature.

The fluoride content observed in all regions in this study was evaluated according to the regulation values of various organizations and did not exceed the maximum permissible value limits of WHO and Turkish standards (1.5 and 2.4 mg/l), respectively [48], [49]. However, the quantitative results of fluoride determined in this study do not show a serious risk in various international criteria, and unfortunately, this cannot be said for the EDI and HQ results of children and infants. It is recommended to use bottled water with specially regulated and controlled fluoride content for children and infants, [50]. The increasing fluoride concentration in the environment is not only caused by drinking water but also due to the increasing use of fluoride-containing drugs in our lives in recent years. Accordingly, fluoride overload can occur. Those most vulnerable to fluoride toxicity are infants, children, those who consume a lot of water, the elderly, those who have developed fluoride intolerance, and those with kidney failure. In addition, in the presence of aluminum, which can be found in trace amounts in water, fluoride can cause abnormalities in G proteins in the organism. Therefore, in the field of modern biomedicine, awareness of the health risks of increasing fluoride and aluminum ion loads can be created, and possible patient burdens can be prevented, [51]. Additionally, care should be taken in fluoride intake from various sources other than water through food. Quantitative measurements of the transfer of fluoride content from water to animals and then to humans should also be considered. Water with high fluoride content should be cleaned with simple and low-cost purification technologies such as sand water filtration and ultraviolet (UV) disinfection methods before being offered to animals, [52].

4 Conclusion

The fluoride level in some surface waters of the Ceyhan basin within the borders of Osmaniye province was examined and evaluated according to public health criteria in different age categories in this study. The fluoride levels, water quality standards, and criteria of the examined waters had

Class I water quality, but that does not imply that the non-carcinogenic health risk assessment would not pose a risk of fluoride in the age categories. The fluoride content in some water points considered in the water resources in the province and districts slightly exceeded the standard limit values in this study. The non-carcinogenic health risk of fluoride for adults has not been determined, but fluoride status may always pose a risk under changing climatic conditions in the region with options other than waterborne fluoride intake (through food, inhalation, skin contact, etc.). The non-carcinogenic health risk value of fluoride for 1-year-old infants was higher than 1 (HQ > 1). Because of the metabolic mobility of their body structures, it may also be thought that children may be a secondary age group that can be impacted by non-carcinogenic fluoride exposure. The stimulatory or beneficial effect of a subinhibitory concentration of a toxic substance is defined as hormesis. Similarly, considering that the hormesis status of fluoride has not yet been fully determined and that most of the findings from in vitro studies have only been observed in the millimolar range, there are still many important issues to be investigated in the field of modern medicine. Although this preliminary research has created a clear data source on fluoride distribution in water resources in the Ceyhan basin of Osmaniye province, more clinical studies are needed to determine the effects of the determined limits for different age categories in the long term.

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Contribution of Individual Authors to the Creation of a Scientific Article (Ghostwriting Policy)

The author contributed in the present research, at all stages from the formulation of the problem to the final findings and solution.

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The author declares that no funding was received for conducting this study.

Conflict of Interest

The author declares no conflict of interest.

APPENDIX

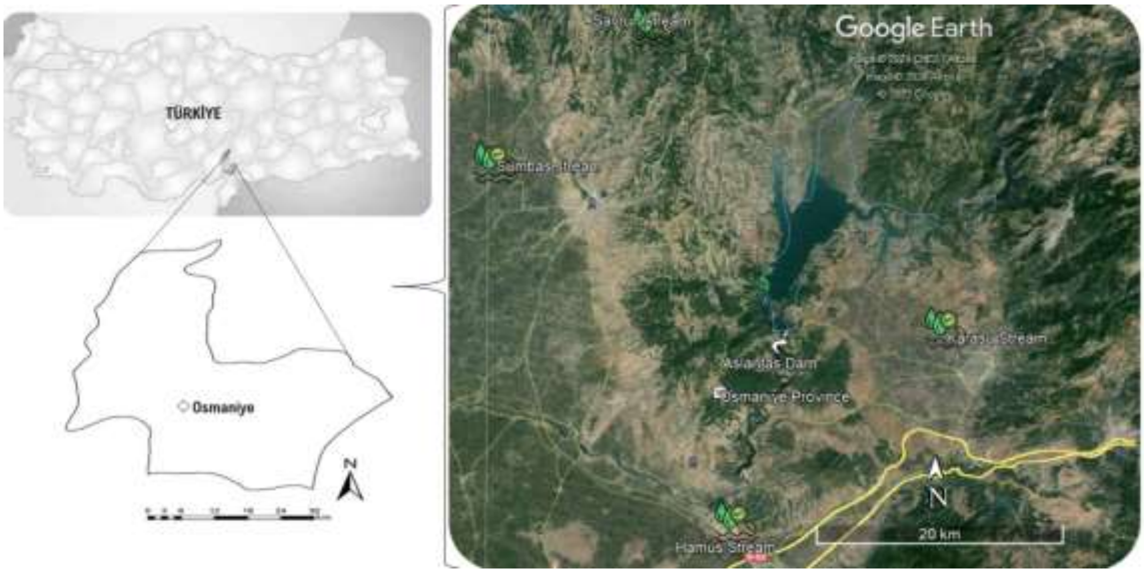

Fig. 1: The map of fluoride content monitoring in locations

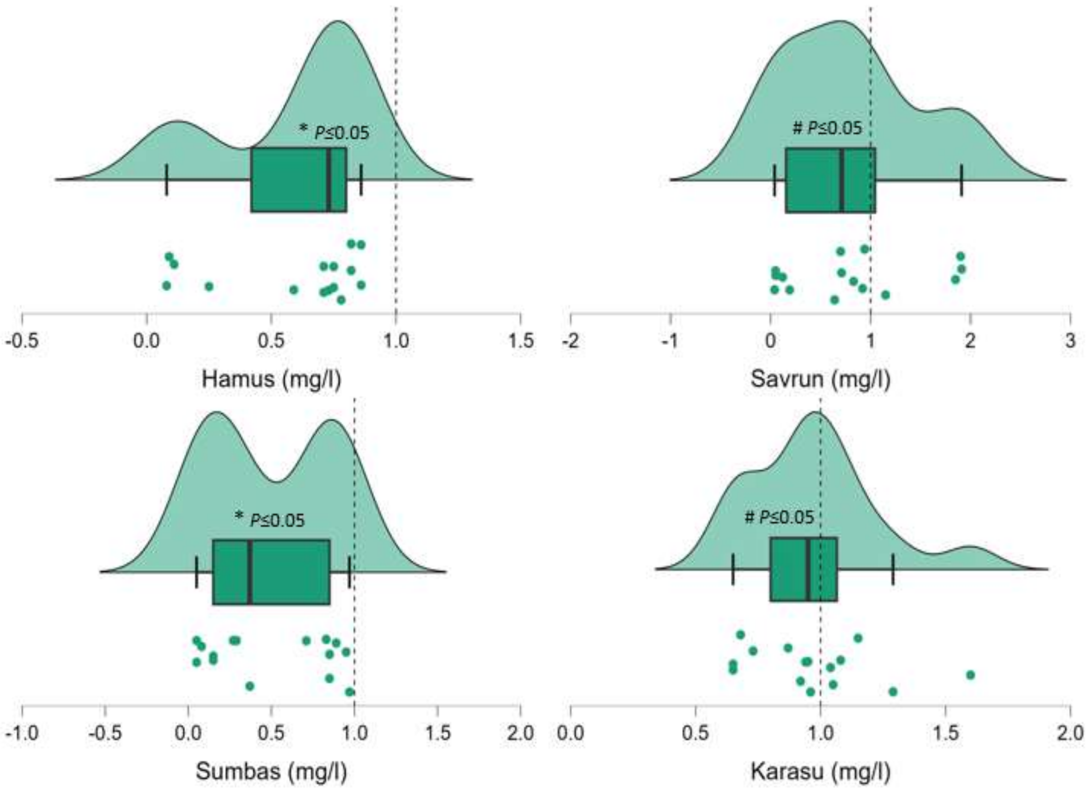

Fig. 2: Raincloud difference plots of the fluoride level in locations (The asterisk (*) and hashtag (#) indicate differences at the $P \leq 0.05$ level between groups)

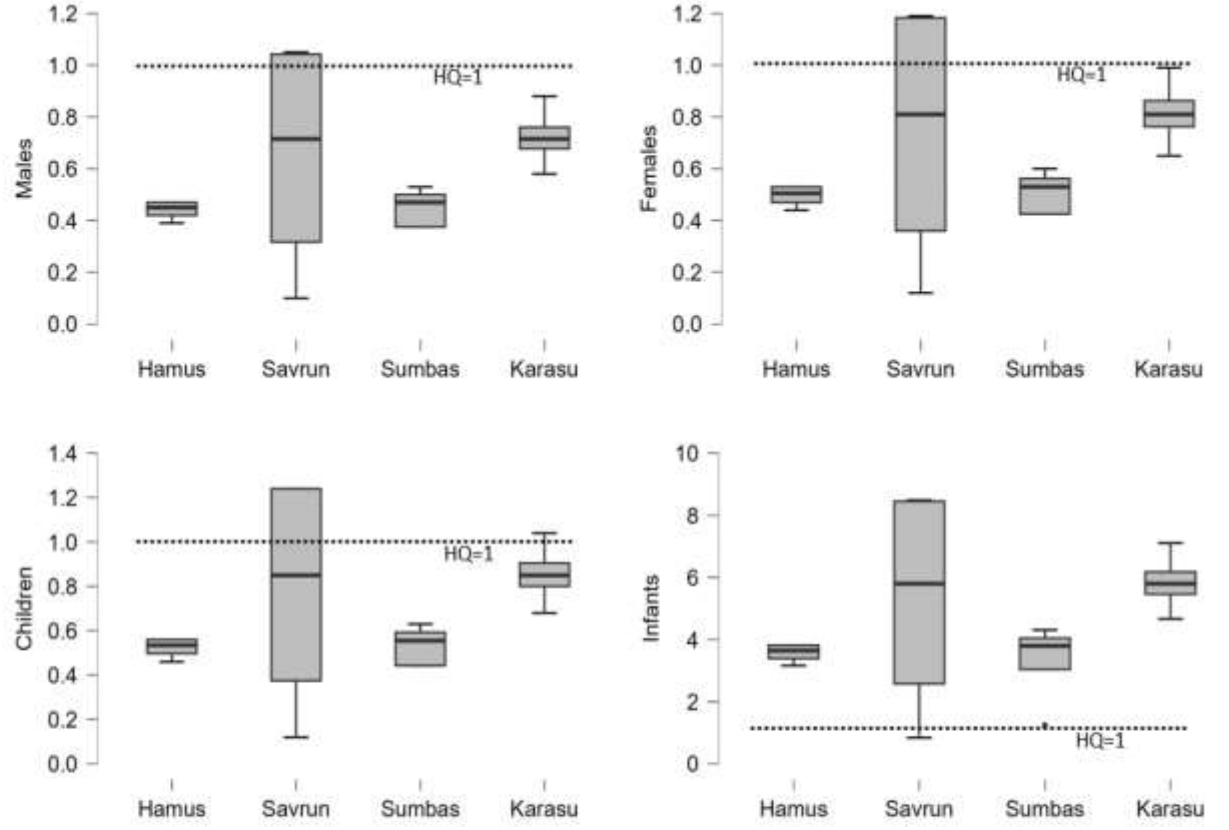

Fig. 3: Boxplots showing the results of the non-carcinogenic health risks of fluoride for men, women, children, and infants in the sampled regions

Table 1. Some characteristics of water quality sampling locations

Altitude (m)	pH	EC (µS/cm)	T (°C)
65	8.65±0.2	410.81±2.51	15.51±0.9
189	8.10±0.3	377.03±3.21	13.09±0.8
57	8.06±0.1	369.25±3.10	14.35±0.8
245	8.55±0.1	541.53±4.24	12.51±0.9

Table 2. Health risk parameters specific to gender and age groups in various references

Parameters	Units	Males	Females	Children	Infants	References
Ingestion rate (IR)	L/day	2.5	2.5	0.85	0.8	[25], [26]
Exposure frequency (EF)	Days/year	365	365	365	365	
Exposure duration (ED)	Years	64	67	12	1	[27]
Average body weight (ABW)	Kg	76	67	20	3	[28]
Average exposure time (AET)	Days	23360	24455	4380	365	[29]
Reference dose (R*f*D)	mg/kg/day	0.06	0.06	0.06	0.06	[30]
The concentration of element in surface water (C)	mg/l	Fluoride 0.04 to 1.91 mg/l				In this study

Table 3. Descriptive statistics parameters of the fluoride level measured in locations

Locations	Mean±SEM	Min-Max	Coefficient of variation	95% CI for Mean Difference		*P*
				Lower	Upper	
Hamus	0.59±0.07	0.08-0.86	0.50	0.44	0.75	<0.001
Savrun	0.80±0.17	0.04-1.91	0.84	0.46	1.14	<0.001
Sumbas	0.50±0.09	0.05-0.97	0.74	0.31	0.68	<0.001
Karasu	0.97±0.06	0.65-1.60	0.26	0.84	1.10	<0.001

Table 4. Estimated daily intake of fluoride content for surface water among age

Location	EDI (mg/kg/day)			
	Male	Female	Children	Infants
Hamus	0.026	0.030	0.031	0.214
Savrun	0.039	0.044	0.046	0.314
Sumbas	0.024	0.028	0.029	0.197
Karasu	0.043	0.049	0.051	0.351
Average	0.033	0.038	0.039	0.269
Min- Max	0.006-0.063	0.007-0.071	0.007-0.074	0.051-0.509
RDI	0.034[a]	0.034[a]	0.084[a]	0.03-0.07[b]

(RDI: Recommended daily intake. a) [37]; b) [38]