Assessment of Fluoride Concentration in Groundwater from the Districts of Mysore and Mandya in Karnataka, India

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Abstract: Background: Fluoride is one of the most common, highly reactive and non-biodegradable environmental pollutant. It is found in all natural waterways but groundwater is more contaminated when compared to other water ways. It is because of the geogenic source like rock and sediments rich in fluoride minerals contaminate groundwater. Another source of fluoride is from the phosphate fertilizers, industrial effluents and coal burning in thermal power plants. Fluoride from all these anthropogenic sources comes in contact with precipitation and then it seeps into the earth and contaminate groundwater. Increases in population in India increase demand for water, most of the population in urban and rural areas completely depend upon groundwater for drinking and cooking purpose. So it is very import to check the contaminants in water and remedial measures should be taken to remove the contaminants from the water. **Objective:** To determine the fluoride concentration in the groundwater of Mandya and Mysore district. **Methodology:** A total of about 120 water samples were collected from 40 villages of Mysore and Mandya district in 400ml pre-sterilized polyethylene bottles. The water sample was taken from the tap after five minutes of being flushed. Water flow was decreased, the sample was filled to the bottle's shoulder, and the cap was tightened to prevent leaks. The collected water sample was preserved and fluoride concentration was determined by using SPADNS method. **Results:** Out of 40 villages in Mysore and Mandya district, 15 villages have a greater than the allowed fluoride amount in drinking water, as per WHO guidelines. The remaining villages had fluoride levels below 1.5 mg/L, which is the recommended level of fluoride concentration. The fluoride concentrations ranges of all taluks as follows: KR Pete - 2.3±0.29 to 2.8±0.18ppm, Pandavapura - 0.1±0.34 to 2.8±0.63ppm, Malavalli - 0.1±0.29 – 0.5 ±0.44ppm, Srirangapatana 1.1±0.58 – 2.8±0.69ppm, Nagamangala 2.0±0.1 to 3.0±0.2ppm, Mysore 0 to 2.5±0.1ppm, Hunsur 0.5±0 – 1.29±0ppm, and KR Nagara 0.29±0.18 – 1.43±0.30 ppm. Out of 25 villages, 13 in Mandya district and 1 in Mysore district consists of the maximum fluoride content over the 1.5 mg/L allowable limit.

Keywords: Fluoride, Groundwater, environmental pollutant, anthropogenic source, Phosphate fertilizer, Industrial effluents, SPADNS method.

1. Introduction

Human existence is reliant on natural resources. Humanity cannot survive on our planet without clean water and pure air. River, lakes, pond and underground water are the primary water sources utilized in residential, commercial, and agricultural contexts (Sunitha, 2020). In many regions of the world, groundwater serves as the main source of fresh water (Kurwadkar et al., 2020). Elevated levels pollutants are added to groundwater by both natural and anthropogenic activity. Pollutants which are present in underground water is high for human and animals consumption. Fluoride is the primary inorganic contaminant detected in underground water (Kurwadkar et al., 2020). Fluoride contamination in groundwater is primarily due to geogenic processes, despite anthropogenic sources playing a significant role (Sivasankar et al., 2016).

Fluorine is found naturally in over 200 minerals, primarily as igneous or sedimentary rock deposits. It is also present as the fluoride ion, with different minerals having varying fluoride contents (Sivasankar et al., 2016). Fluoride is the primary contaminant from anthropogenic source can be traced back to fertilizer plants, aluminium smelters (Haidouti, 1991), and industrial processes like glassmaking, iron and steel production, cement manufacturing, ceramic fire, the burning of fuel, aluminium, iron industries, brick block and ceramic manufacture industries are major sources of fluorine released as gaseous and particulate species(John, 1990). Underground water may become more fluoridated due to the usage of phosphate-containing fertilizers and irrigation water in agricultural land, which pose a potential risk. Combustion of coal in thermal power plant releases high amount of ash partials each year, which seeps fluoride into groundwater. Groundwater, the largest freshwater resource in the world, is vital to global food security and human life since it may be used as drinking water ((Mukherjee & Singh, 2018). Groundwater has higher levels of fluoride than surface water because it has greater contact with fluoride bearing minerals during rock water interactions (Edmunds &Smedley, 2013). More than 100 countries have above permissible limit of fluoride concentration in underground water; 38 countries in Africa, 5 counties in South America, 3 countries in America, 28 countries in Asia, 3 countries in Australia and 2 countries in Europe shows high fluoride concentration in underground water (Shaji et al., 2024).

India's overpopulation has increased the country's need for groundwater, which has led to overexploitation, a decline in groundwater levels, and an increase in bore well depth. This could be the main reasons for high fluoride in ground water because as we dig deeper into the earth the rock which has fluoride elements comes in contact with the groundwater and contaminate it (Ayoob& Gupta, 2006). India is a highly populated country where fluoride poisoning of underground water becomes a serious concern, according to a study about 14% of the world's total geogenic fluoride found in India, suggesting that fluoride contamination in India has a geogenic origin (Mukherjee & Singh, 2018). The CGW Board investigation found that about 19 states of

India, 184 districts are have elevated level of fluoride concentration in underground water. According to UNICEF 1995 twenty of India's 17 states currently have high fluoride in them, and that number is expected to increase (Fawell, 2004). On the other hand, 60 to 70 million individuals are at risk of fluorosis in India. Exposed to high level of fluoride through water or food leads to several health complications, although it primarily results in skeletal and dental fluorosis. Fluorosis can appear in various degrees depending on the quantity and frequency of fluoride consumed (Kashyap et al., 2021). One of the main issues that is currently developing in the Indian state of Northern Karnataka is groundwater contamination from nitrate and fluoride. The hamlets of Jidga, Kamanalli, Padsawli, Sakkarga, and Nagelagaon are the most severely affected by high concentration of fluoride in groundwater (Rizvi et al., 2022). Data from the CGB indicates that elevated fluoride content affects 14 districts in the state of Karnataka (Dharumarajan et al., 2018)

In Mysore and Mandya district majority of the peoples depend on groundwater for drinking and cooking purpose. Majority of the study reported that the fluoride concentration in drinking water above permissible limit i.e. >1.5ppm causes severe health effect mainly dental and skeletal fluorosis which leads to socio economic burden to the country growth. Hence the present study was undertaken to known the fluoride concentration in Mysore and Mandya district of Karnataka state, India. This information would be very helpful in implementation of water ATM in the villages having high fluoride concentration in drinking water and to create awareness about health consequences due to excess of fluoride intake in drinking water.

2.Materials and Methods

2.1. Collection, preservation, and determination of fluoride level in underground water samples

Study area

The study was carried out in two districts, namely Mandya and Mysore. The Mandya district is located between 12° 13′ and 13° 04′ North Latitude and between 76° 19′ and 77° 20′ East Longitude. It receives 700 mm of rain on average each year. The district experiences mild summer temperatures (up to 35°C) and mild winter temperatures (down to 20°C). The district has a total geographical area of 4, 98,244 hectares, of which 2, 53,067 hectares are planted. Over half of the district's land area is used for agriculture. Mysuru is located in Karnataka's southern region. It is located at Latitude-12^0.3′, Longitude76^0.6′. Mysuru District occupies an area of approximately 6,854 square kilometres. The district has a minimum of 11°C and a maximum 38°C temperature. The average humidity of the district is 85 (max) and 60 (min). The average annual rainfall is 739mm. Study was conducted in 5 taluks of Mandya districts which include Srirangapatana, Pandavapura, Malavalli, Krishnarajapete and Nagamangala and 3 taluks of Mysuru district which consist of Krishnaraja Nagara and Hunsur. 5 villages from each taluk were randomly selected for the study (Figure 1). From each village, 3 samples of drinking water from bore wells belonging to different locations were collected and used for fluoride analysis.

2.2. Collection and preservation of water samples.

A total of about 120 water samples were collected from 40 villages of Mysore and Mandya district in 400ml pre-sterilized polyethylene bottles. The water sample was taken from the tap after five minutes of being flushed. Water flow was decreased, the sample was filled to the bottle's shoulder, and the cap was tightened to prevent leaks. The collected water sample was preserved and fluoride concentration was determined by using SPADNS method (APHA, 1992).

2.3. Analysis of fluoride in water samples by SPADNS method

Principe: The reaction depends on the reaction between the compounds in the SPADNS reagent and the fluoride ion present in the water sample. A zirconium-dye lake and fluoride in the water react to produce a complex compound. Some of the dye lake is dissociated by fluoride reaction, leaving behind the dye and a colourless complex anion (Figure 2). The colour generated lightens with increasing fluoride concentration.

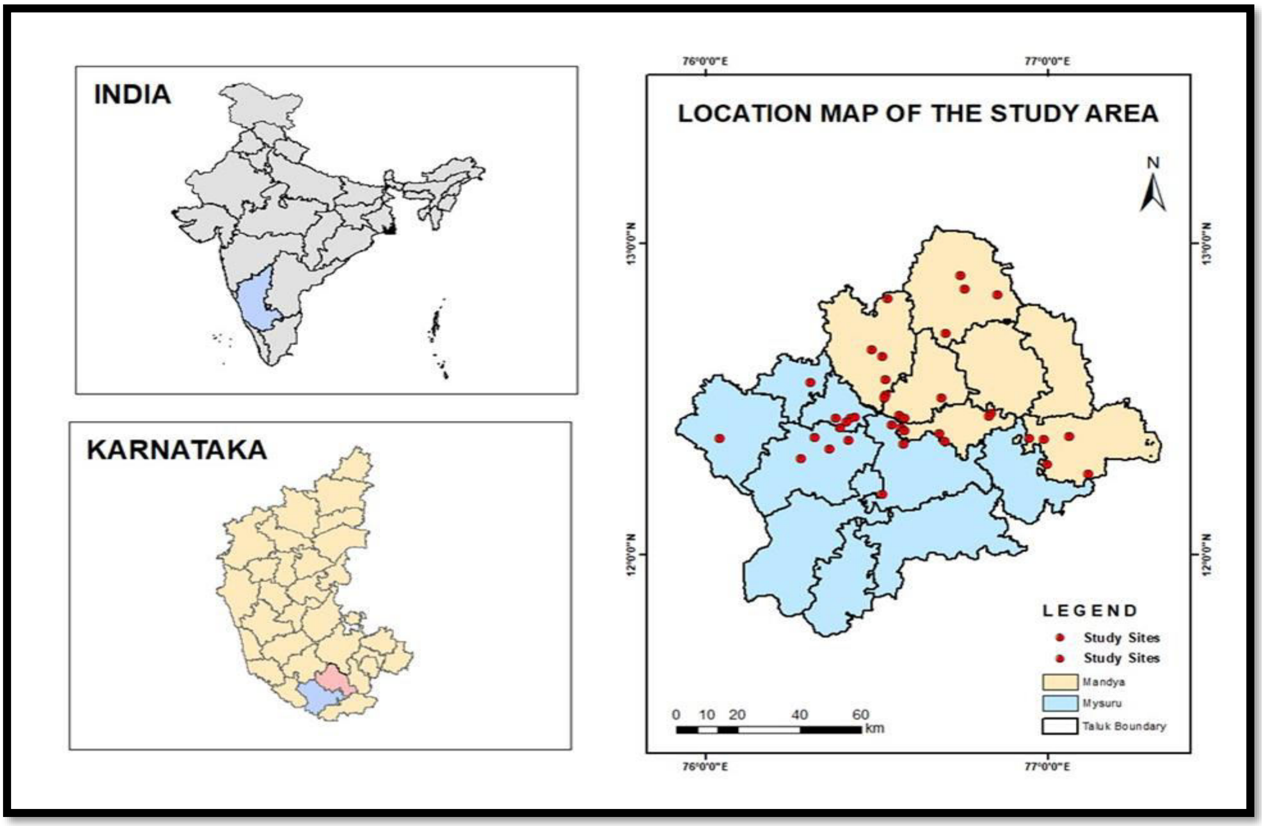


Figure 1. Location map of the study area

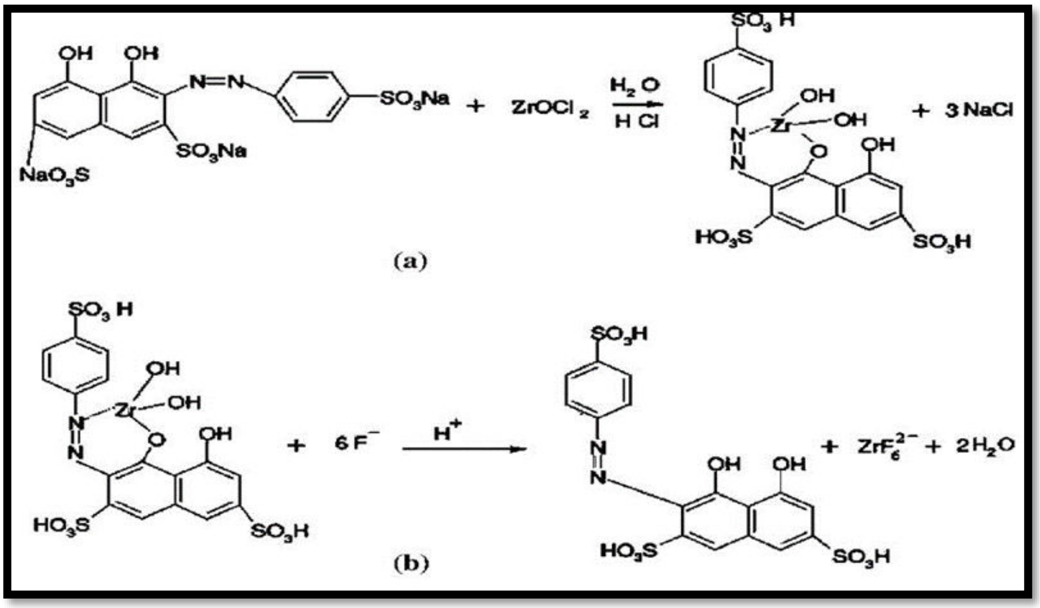


Figure 2. Reactions involved in the SPADNS method

a. SPADNS-Zr OCl 2 complex formation b. the complex's reaction when exposed to fluoride ions in water sample (source: (Ghosh et al., 2013)).

2.4. Reagents required:

(a) Zirconyl-acid reagent: A reagent for zirconyl-acid was prepared by dissolving 133.0 mg of Zirconyl dichloride oxide in 25 ml of clean water and then it was mixed with 500 ml of distilled water and 350 ml of strong Hydrochloric acid.

(b) Solution for SPADNS: A red solution was prepared by dissolving 0.958g of SPADNS in water and diluting the mixture up to 500 mL then the solution was stored in an amber bottle.

(c) Zirconyl-acid reagent and SPADNS solution were combined in equal parts to create a red solution, named acid Zirconyl-SPADNS reagent.

(d) Calibration solution: 100 millilitres of water that was distilled were used to dilute 10 millilitres of the SPADNS solution. 7 mL of pure water were used to create 10 mL of strong Hydrochloric acid. 10 ml of the acid solution were combined with the previously dilute SPADNS solution. The resulting mixture was then used to adjust the reference point, or zero, of the spectrophotometer.

(e) The calibration curve's standard fluoride solutions were all made in polyethylene flasks and kept in plastic bottles since glass is harmed by fluoride ions.

(f) Stock solution: Distilled water was used to dissolve 221.0 mg of anhydrous sodium fluoride and dilute it to 1,000 mL (100 mg/L).

(g) Standard fluoride solution: Distilled water (10 mg/L) was used to dilute 100 mL of stock to 1 liter.

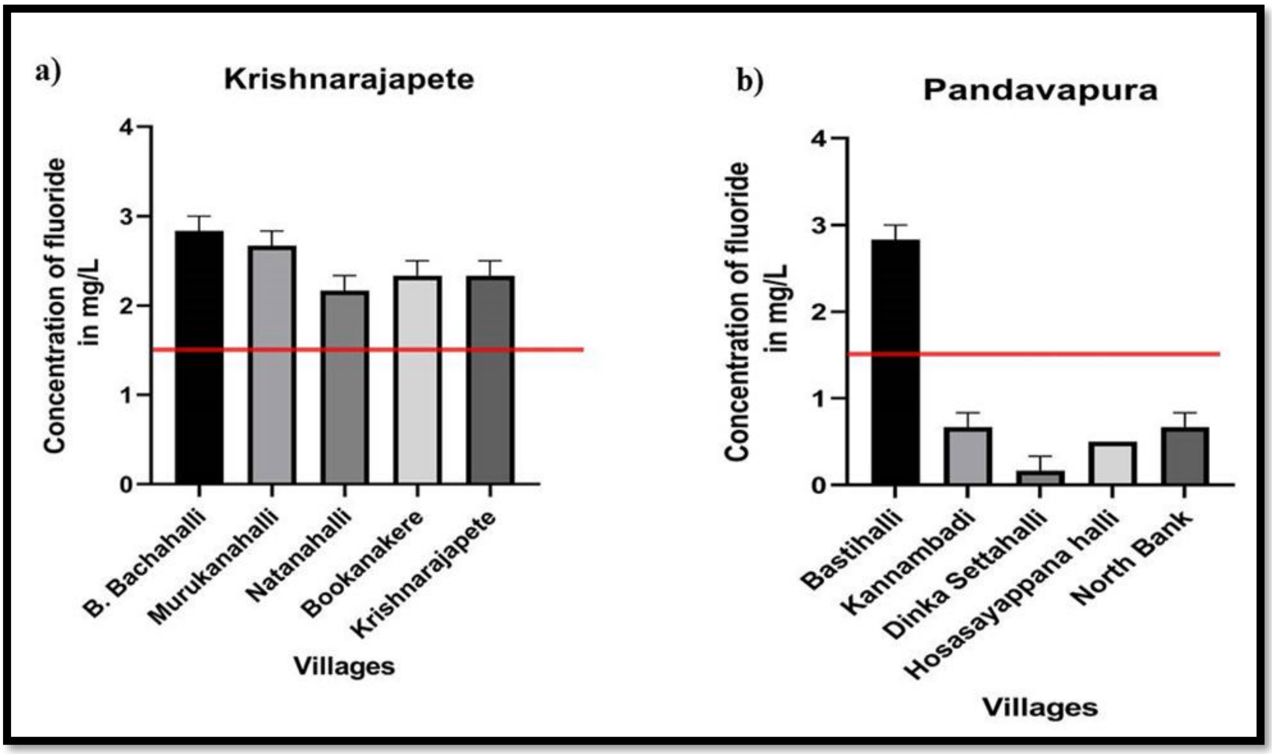
(h) Standard solutions: The standard fluoride solution was diluted to a volume of 50 ml using distilled water to create a range of concentrations: 0.5, 1, 1.5, 2, 2.5, and 3 ppm. To create a coloured solution for the photometric measurements, 10 mL of a

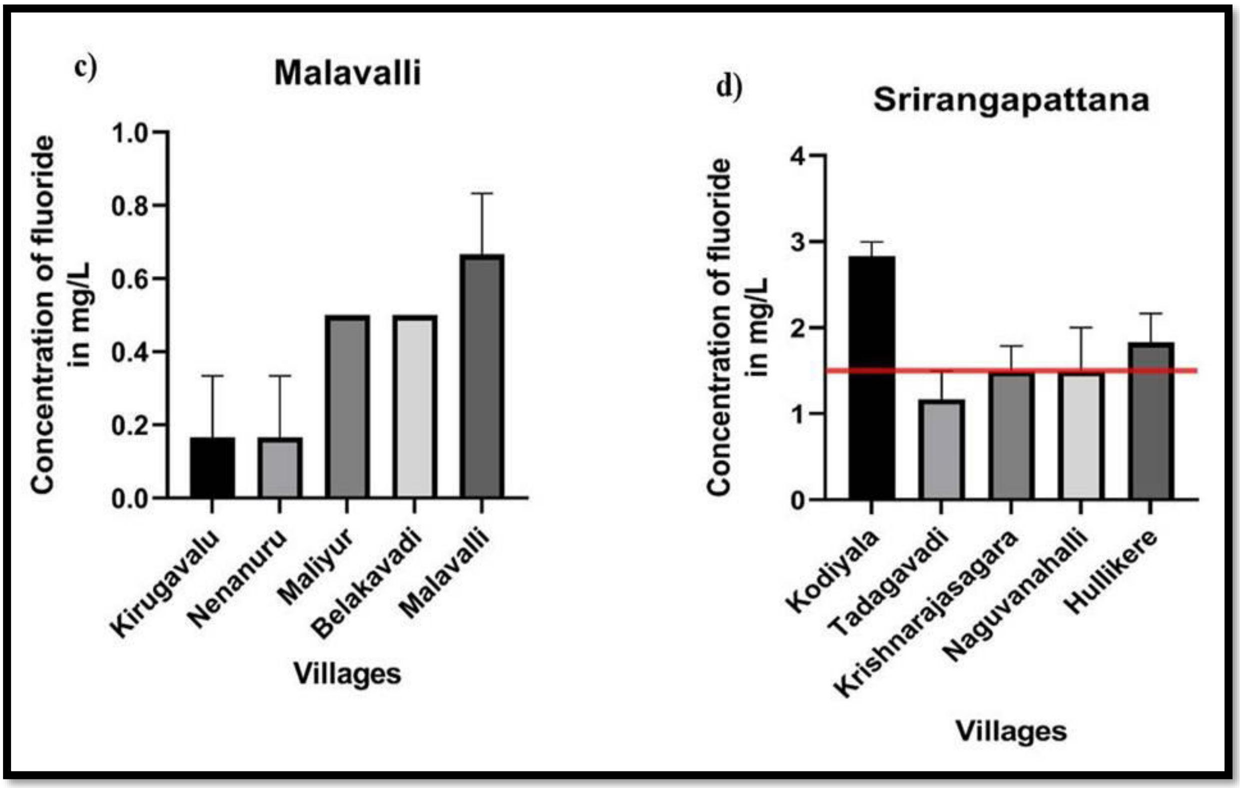
mixed acid-Zirconyl-SPADNS solution was added to each standard solution and thoroughly mixed.
ii. Sample pre-treatment: To remove any leftover chlorine from the water sample, one drop (0.05 mL) of $NaAsO_2$ solution was added. This solution is made by dissolving 5.0 grams of $NaAsO_2$ and adding distilled water to get a 1,000 ml solution.

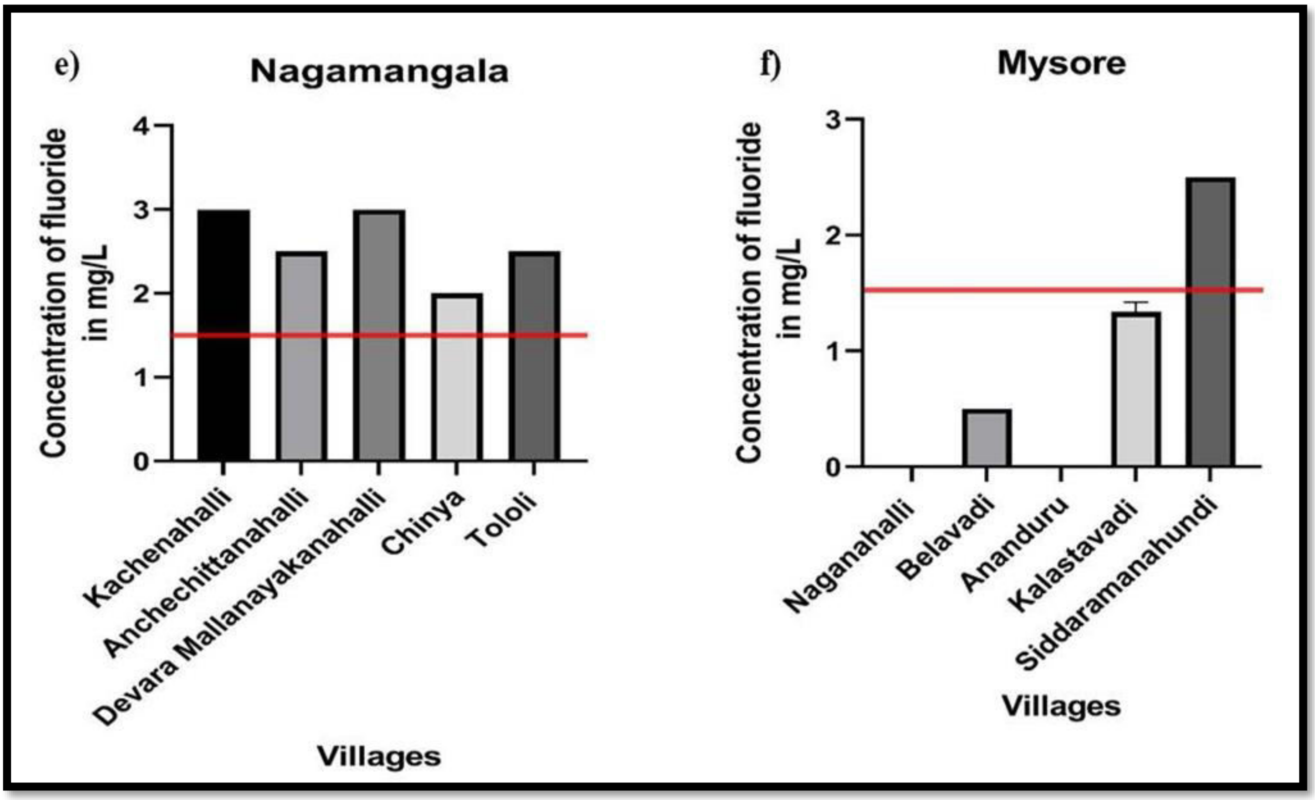
2.5. Procedure:
Fluoride concentrations were determined by the analytical SPADNS method (APHA, 1992). Microprocessor Photo colorimeter model-1312 was used to estimate fluoride concentration. An ion of fluoride and a lake of red zirconium dye are the two components of the SPADNS colorimetric technique. Zirconium hexafluoride is a colourless ion that is created when fluoride combines with Dye Lake. As seen below, the rise in the fluoride concentration decreases the intensity of the Dye colour. The standard fluoride graph was generated by taking sodium fluoride in the concentration range of 0 to 3 ppm. The fluoride concentration was determined by taking 25 ml of each standard solution and test sample in a separate clean 50ml conical flask. The contents of each flask were then thoroughly shaken after 5 ml of acid Zirconyl SPADNS solution was added. At 570 nm, the absorbance was determined using a photo colorimeter. The blank solution was the one without sodium fluoride. The absorbance of all standard and test samples was recorded. The concentration of standard solutions was plotted on the X-axis, and absorbance was plotted on the Y-axis, to create the standard graph. The amount of fluoride present in the test samples was calculated using the standard graph (APHA, 1992).

2.6. Statistical Analysis: Statistical analysis was performed in Graph Pad Prism 9.5.1 (Graph Pad Software, La Jolla, CA, USA) .Results was expressed in mean and ±SEM.

3. Results
Out of 40 villages in Mysore and Mandya district, 15 villages have a greater than the allowed fluoride amount in drinking water, as per WHO guidelines. The remaining villages had fluoride levels below 1.5 mg/L, which is the recommended level of fluoride concentration. The fluoride concentrations ranges of all taluks as follows: KR Pete - 2.3±0.29 to 2.8±0.18, Pandavapura - 0.1±0.34 to 2.8±0.63, Malavalli - 0.1±0.29 – 0.5 ±0.44, Srirangapatana 1.1±0.58 – 2.8±0.69, Nagamangala 2.0±0.1 to 3.0±0.2, Mysore 0 to 2.5±0.1, Hunsur 0.5±0 – 1.29±0, and KR Nagara 0.29±0.18 – 1.43±0.30 PPM. Out of 25 villages, 13 in Mandya district and 1 in Mysore district consists of the maximum fluoride content over the 1.5 mg/L allowable limit (Figure 3).


a)
Krishnarajapete
Concentration of fluoride in mg/L
0
1
2
3
4
B. Bachahalli
Murukanahalli
Natanahalli
Bookanakere
Krishnarajapete
Villages
b)
Pandavapura
Concentration of fluoride in mg/L
0
1
2
3
4
Bastihalli
Kannambadi
Dinka Settahalli
Hosasayappana halli
North Bank
Villages


c)
Malavalli
Concentration of fluoride in mg/L
0.0
0.2
0.4
0.6
0.8
1.0
Kirugavalu
Nenanuru
Maliyur
Belakavadi
Malavalli
Villages
d)
Srirangapattana
Concentration of fluoride in mg/L
0
1
2
3
4
Kodiyala
Tadagavadi
Krishnarajasagara
Naguvanahalli
Hullikere
Villages


e)
Nagamangala
Concentration of fluoride in mg/L
0
1
2
3
4
Kachenahalli
Anchechittanahalli
Devara Mallanayakanahalli
Chinya
Tololi
Villages
f)
Mysore
Concentration of fluoride in mg/L
0
1
2
3
Naganahalli
Belavadi
Ananduru
Kalastavadi
Siddaramanahundi
Villages

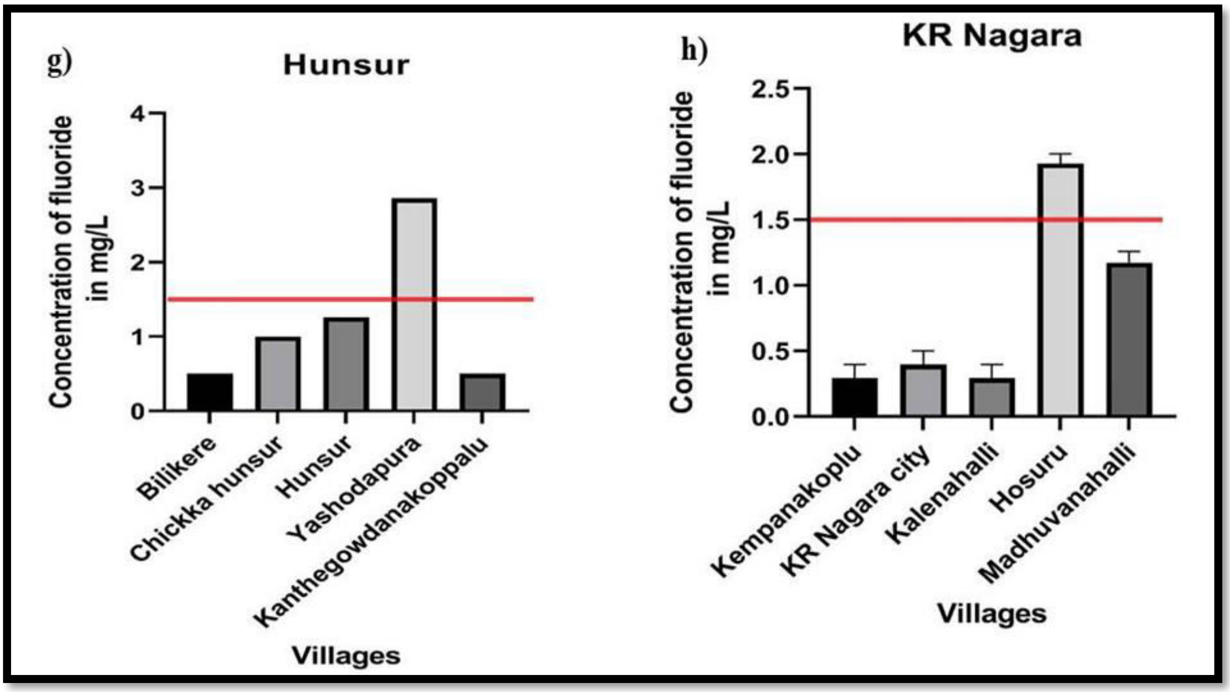

g)
Hunsur
Concentration of fluoride in mg/L
0
1
2
3
4
Bilikere
Chickka hunsur
Hunsur
Yashodapura
Kanthegowdanakoppalu
Villages
h)
KR Nagara
Concentration of fluoride in mg/L
0.0
0.5
1.0
1.5
2.0
2.5
Kempanakoplu
KR Nagara city
Kalenahalli
Hosuru
Madhuvanahalli
Villages

Figure 3. Fluoride concentration of groundwater samples at different taluks of Mandya and Mysore district.

a-e. The drinking water in 14 villages of Mandya district had fluoride concentrations more than 1.5 parts per million. f- h. 3 villages of Mysore district had a high fluoride concentration. The error bar represents mean ±SEM. The red line shows the amount of fluoride above the permissible limit of 1.5 parts per million in drinking water in the districts of Mandya and Mysore.

4. Discussion

Fluorine is a reactive element that is abundant in nature and is necessary for the development of human bones. It is frequently found in water. Minerals that contain fluorides, such as fluorite, amphiboles, micas, apatite, and biotite, leak fluorides into groundwater. Groundwater's alkaline Fluoride-rich dissolved minerals, calcium, bicarbonate, and pH are indicators of fluoride content. Elevated fluoride levels can also result from human actions such as burning coal, using phosphate fertilizers, and producing effluents (Kom et al., 2023; Chen et al., 2020).

Groundwater occurs in worn, fractured zones of orthoquartzites, sandstones, granites, gneisses, basalts, limestone, and shale under the water table and in semi-restricted to confined settings. India's main litho-unit is the Deccan traps and basalts. Most of the basaltic lava flows are level with a slight vertical dip. The Central Ground Water Board states that the differing water-bearing characteristics of several lava flow regulate the occurrence of groundwater in them. The climate of Mandya district is essentially semiarid, with extremely hot summers. Because there wasn't enough rain, the frequency of drought frequently occurs in this region (Gowda et al., 2015).

Fluoride concentration may vary during rainy and dry seasons. In comparison to the rainy season, the dry season has a higher fluoride level in groundwater (Dibal et al., 2017). Recent guidelines from WHO recommend the determination of fluorine and arsenic content in drinking water as these are predominantly causing negative impact on health in most of the countries (Jha&Tripathi, 2021). The present study region's fluoride contents varied from 0.1 to 3 mg/L. BIS 1992 states that the permissible amount of fluoride in drinking water is 1.2ppm. Higher than these threshold concentrations will cause fluorosis. Nevertheless, a fluoride shortage (<0.6 mg/L) may lead to tooth caries (Sajil Kumar et al., 2014). In this study, water samples from 120 bore wells were collected from different regions of the Mysore and Mandya districts. Among these, 40 water samples show a higher fluoride concentration above the permissible limit of WHO. Fluoride concentrations in water from several sources that were adjacent to one another in the same hamlet varied during this investigation. Out of 25 villages surveyed, 13 villages in Mandya district and 1 village in Mysore district has the highest fluoride content exceeding the 1.5 mg/L allowable limit. Similar findings were documented by El-Nadeef and Honkala (1998) at Nigeria and Gopala Krishnan et al. (1999) at Kerala. Madhusudhan et al. (2024) at Mandya.

The state of India was effectively mapped in terms of geographical distribution and fluoride, and there was ample evidence of both major and very significant variations in the "between-zone" and "between-district within zone" components of variation. ICMR suggests the drinking water fluoride level should be 1ppm; however the Bureau of Indian Standards (BIS) recommends 1.5 ppm for drinking water. Similarly, the Government of India's Committee on Public Health Engineering states that the amount of fluoride in drinking water should be 1.0 ppm. On the other hand, the World Health Organization (WHO) suggests that the fluoride content of drinking water should range from 0.8 to 1.5 parts per million (Petersen & Lennon, 2004;Chowdhury et al., 2016).

Additionally, our study showed that consuming water from villages which is having above 1.5 ppm of fluoride water is unfit for drinking purpose especially in the summer when human consumption is high. The northern districts of Karnataka have comparatively higher temperatures and significantly higher fluoride concentrations. There is a, partial correlation analysis verified the existence of weak but statistically significant and very significant negative relationships associated with the fluoride concentrations in drinking water and the average district temperature and the average district total precipitation, respectively The rainfall and water fluoride content partial correlation was shown to be negatively correlated (Chowdhury et al., 2016). In our study was limited to determination of fluoride content in Mysore and Mandya district and we did not focus on seasonal variation of fluoride concentration in groundwater.

In Mysore and Mandya district peoples affected by dental fluorosis, they calculated the average fluoride ion content in district drinking water sources, which ranged from 0.1 to 3 ppm. Actually, Chitradurga, Tumkur, Mysore, Bijapur, Gulbarga Chikkamangalore, Mandya, and Bangalore rural Gadag, Bellary, Belgaum, Raichur, and Bijapur Mangalore and Shimogga, Kolar are among the Karnataka districts impacted by fluorosis, according to UNICEF (Chowdhury et al., 2016). This information is broad, yet it has a high correlation with what we found here.

Fluoride contamination of drinking water and soil has been addressed through numerous corrective actions, most of which are expensive and out of reach for the underprivileged. Even though the government installed water ATMs, individuals do not want to drink the water, and those who are impoverished find it to be somewhat pricey. Fluoride-sensitive areas have notably lower levels of awareness regarding the health consequences associated with fluoride use. Future research should focus on raising public knowledge of the dangers that using contaminated water poses to one's health. Additionally, to identify a cheap and easy method for eliminating fluoride and other harmful substances from drinking water.

5. Conclusion

In this study we found more than 50% of water samples collected contaminated by fluoride. Most of the villages had a very higher content of fluoride in groundwater. Fluoride in lower concentration i.e. less than 0.5 ppm help full in protecting the teeth from dental cavities but when it exceeds 1.5ppm it causes several health complication mainly dental and skeletal fluorosis. Majority of the peoples residing in the villages use the same groundwater for drinking and cooking purpose. Most of the peoples residing in these fluoride rich regions are suffering from dental and skeletal fluorosis and many other health complication. Fluoride contamination of drinking water and soil has been addressed through numerous corrective actions, most of which are expensive and out of reach for the underprivileged. Even though the government installed water ATMs, individuals do not want to drink the water, and those who are impoverished find it to be somewhat pricey. Fluoride-sensitive areas have notably lower levels of awareness regarding the health consequences associated with fluoride use. Future research should focus on raising public knowledge of the dangers that using contaminated water poses to one's health. Additionally, to identify a cheap and easy method for eliminating fluoride and other harmful substances from drinking water.

Conflicts of Interest: There is no conflict of interest between the authors

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