

**RISK ASSESSMENT OF FLUORIDE IN DRINKING WATER AND
FOOD CHAIN AND PERFORMANCE STUDY OF
DEFLUORIDATION UNIT**

**A thesis submitted towards partial fulfillment of
the requirements for the degree of**

**Master of Engineering in
Water Resources and Hydraulic Engineering**

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ABSTRACTS

Ground water is the major source for various purpose in the world. Presence of low or high concentration of certain ions is a major issue as they make the ground water unsuitable for various purposes. Fluoride is one such ion that causes health problems in people living in more than 25 nations in the world. Fluoride concentration of at least 0.6 mg/l is required for human consumption as it helps to develop stronger teeth and bones. Consumption of water with fluoride concentration above 1.5 mg/l results in acute to chronic dental fluorosis where the tooth becomes colored from yellow to brown. Skeletal fluorosis which causes weakness and bending of the bones also results due to long term consumption of water containing high fluoride. Presence of low or high concentration of fluoride in groundwater is because of natural or anthropogenic causes or a combination of both. Natural sources are associated to the geological conditions of an area. Several rocks have fluoride bearing minerals like apatite, fluorite, biotite and hornblende. But not only in tea there are so many food items that consume fluoride. Tea when mixed with water it has a high concentration of fluoride. Apart from tea in Hazmola, Hazmi Guli, Black Salt it also consume fluoride. Shrimp, blue crabs, soda, raisins also have a certain amount of water. Fluoride in limited amount is considered to be useful, but presence of high concentration of fluoride leads to a severe damage to human body. In this paper we have discussed about lots of defluoridation method. Nalgonda, Reverse osmosis, Nano Technology, Electrocoagulation, adsorption. Among them here we choose a method named as adsorption method. Fluoride contamination in drinking water due to natural and anthropogenic activities has been recognized as one of the major problems worldwide imposing a serious threat to human health. Among several treatment technologies applied for fluoride removal, adsorption process has been explored widely and offers satisfactory results especially with mineral-based and/or surface modified adsorbents. In this review, an extensive list of various adsorbents from literature has been compiled and their adsorption capacities under various conditions (pH, initial fluoride concentration, temperature, contact time, adsorbent surface charge, etc.) for fluoride removal as available in the literature are presented along with highlighting and discussing the key advancement on the preparation of novel adsorbents tested so far for fluoride removal. It is evident from the literature survey that various adsorbents have shown good potential for the removal of fluoride. However, still there is a need to find out the practical utility of such developed adsorbents on a commercial scale, leading to the improvement of pollution control. This study showed that Fluoride is not only available in tea. It is also available in different types of food samples. The aim of this research work is to assess the concentration of fluoride in water & food product sample and develop a sustainable treatment methodology and evaluate its efficacy. The study results shows that the percentage of fluoride removal by adsorption process using activated alumina is 76.13 % which can used as sustainable fluoride removal methodology.

Key Words: Fluoride, Fluorosis, Food, Adsorption, Fluoride removal technology, Health impact, Water quality.

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CHAPTER-1

INTRODUCTION

Chapter 1

1. Introduction:

1.1 General information on Fluoride:

Fluoride contamination in drinking water sources due to geogenic causes has been recognized as one of the major problems worldwide. Anthropogenic discharges can also lead to increased levels of fluoride in the environment. Fluorine, a fairly common element of the earth's crust, is present in the form of fluorides in a number of minerals and in many rocks. Fluorine is the 24th most abundant element in the universe (4×10^{-5} %), and thus relatively rare. However, in Earth's crust it is enriched and is the 13th most abundant element by weight percent (0.054%) or 625 mg/kg, just ahead of carbon (0.02%) and exists in trace amounts in almost all groundwater throughout the world. The elements to which fluorine has a high affinity, Si, Al, Ca, and Mg are also common, both in Earth's crust and the universe (28.2/0.07%, 8.1/0.005%, 4.1/0.007%, and 2.3/0.06%, respectively). This is why fluorine is likely to be found tightly bound within stable substances outside of liquid conditions (Budisa, N. et al., 2014). Fluorine is the 24th most abundant element in the universe (4×10^{-5} %), and thus relatively rare. However, in Earth's crust it is enriched and is the 13th most abundant element by weight percent (0.054%) or 625 mg/kg, just ahead of carbon (0.02%) and exists in trace amounts in almost all ground waters throughout the world. The elements to which fluorine has a high affinity, Si, Al, Ca, and Mg are also common, both in Earth's crust and the universe (28.2/0.07%, 8.1/0.005%, 4.1/0.007%, and 2.3/0.06%, respectively). This is why fluorine is likely to be found tightly bound within stable substances outside of liquid conditions (Budisa, N. et al., 2014).

1.2. Significance of Fluoride in drinking water:

Drinking water is often the main source of fluoride intake by humans, especially in areas where fluoride concentrations in groundwater and/or surface water are high. It is estimated that more than 200 million people worldwide rely on drinking water with fluoride concentrations that exceed the present WHO guideline of 1.5 mg/l. In some area foodstuffs and/or indoor air pollution due to the burning of coal may make significant contributions to the daily intake of fluoride. Excess fluoride intake causes different types of fluorosis, primarily dental and skeletal fluorosis, depending on the level and period of exposure. Fluorosis, associated with elevated fluoride concentrations in drinking water, has been reported in various countries around the world such as India, China, Tanzania, Mexico, Argentina, and South Africa, among others (Amini, Manouchehr et al., 2008).

Fluorine is essential for mineralization of bones & formation of dental enamels. 96% of fluoride of body found in bones & teeth. Normally small amount of fluoride is required (0.5 to 0.8 mg/l) in drinking water. Prolonged ingestion of fluoride through drinking water in excess of the daily requirement is associated with dental and skeletal Fluorosis. On the other hand, inadequate intake of fluoride in drinking water is associated with dental caries. World Health Organization (WHO) has set the upper limit of fluoride concentration in drinking water at 1.5 mg/l, while the Bureau of Indian Standards has laid down Indian standards as 0.6 to 1.2 mg/l as maximum permissible limit of fluoride with further remarks as "lesser the better". Intake of fluoride higher than the optimum level is the main reason for dental and skeletal fluorosis.

1.3 Health impact of Fluoride:

At concentrations above 1.5 mg/L (79 $\mu\text{mole/L}$) however, fluoride is dangerous to human health, leading to dental and skeletal fluorosis, a disease that can cause mottling of the teeth, calcification of ligaments, crippling bone deformities, and many other physiological disorders that can, ultimately, lead to death. over 10mg/l to develop, skeletal fluorosis is more severe than its dental counterpart, characterized by deformation of the bone structure can be detected early on radiologically. It is now well established that fluoride in drinking water can cause 'non-ulcer dyspeptic' complaints in human subjects. The main complaints are nausea, loss of appetite, pain in the stomach, gas formation and bloated feeling, constipation followed by intermittent diarrhea and headache. Fluoride has also been linked to cancer, decreased cognitive ability, lower Intelligence Quotient (IQ), and developmental issues in children.

Fluorosis has significant economic impacts in the developing world. In addition to fluorosis removing people from the workforce, water supply programs have thrown away significant finances while providing costly boreholes that become useless upon the discovery of the toxic levels of fluoride that they contain.



Fig-1: Fluorosis is caused due to contamination of drinking water with high fluoride content (Source: Siasat.com)

1.4 Global contamination scenario of Fluoride:

The problem of excessive fluoride in drinking water has engulfed many parts of the world, and today many millions of people rely on groundwater with concentrations above the World Health Organization (WHO) guideline value. There are more than 20 developed and developing nations in which fluorosis is endemic. Fluorosis is an endemic which is important public health problem in 20 more countries, including India, which lies in the geographical fluoride belt that extends from Turkey to China and Japan through Iraq, Iran and Afghanistan. Of the 85 million tons of fluoride deposits on the earth's crust, 12 million are found in India. Hence it is natural that fluoride contamination is widespread, intensive and alarming in India. Endemic fluorosis resulting from high fluoride concentration in groundwater is prevalent in India since 1937. Countries like Iraq, Turkey, Syria, Jordan, Palestine, Morocco, Algeria, Egypt has higher fluoride concentration.

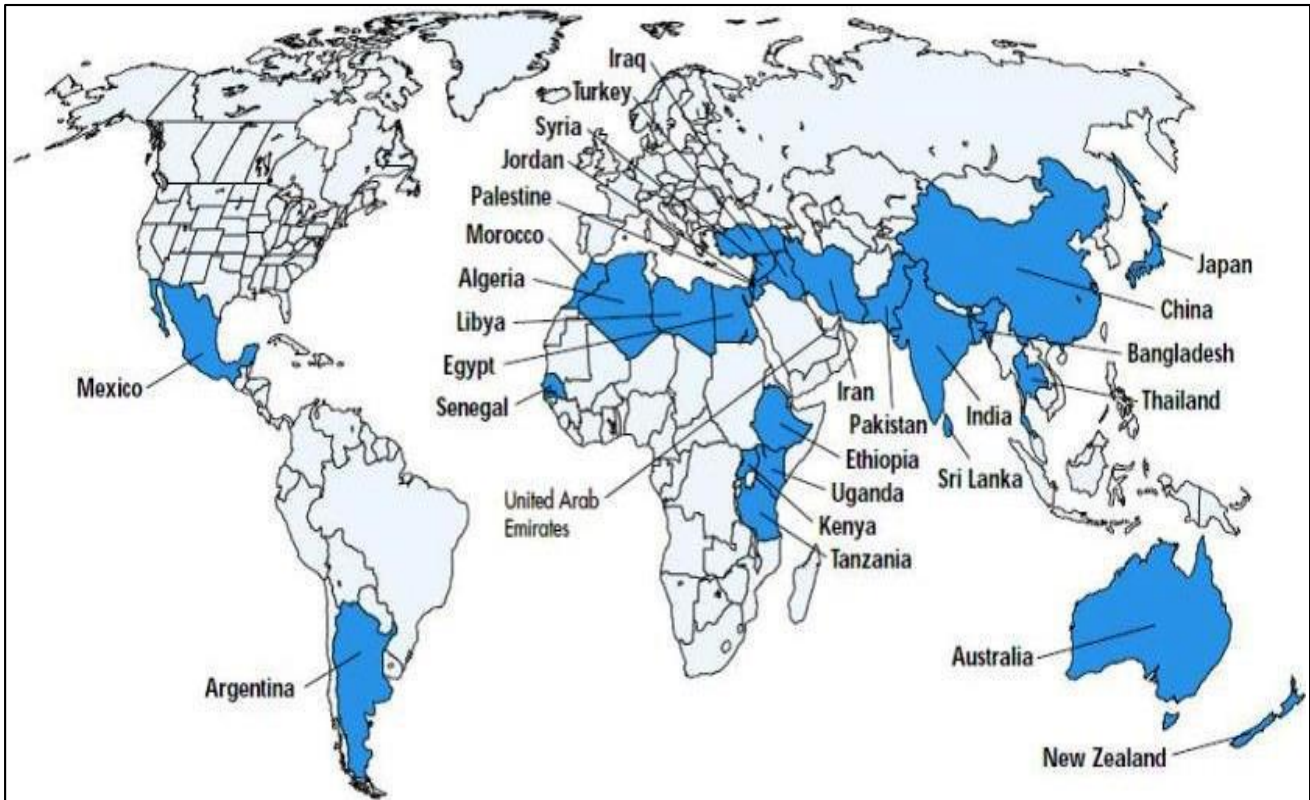


Fig: 2- Countries with endemic fluorosis due to excess fluoride in drinking water sources.
(Source: Slideshare.net)

1.5 Indian scenario of Fluoride contamination: In India fluorosis was first detected in teeth of cattle at Nellore district of Andhra Pradesh in 1937. Since then, considerable work has been done in different parts of India to explore the fluoride-laden water sources and their impacts. Fluorosis is an endemic disease prevalent in 20 states of Indian Republic. The problem of excessive fluoride is more severe, particularly in arid parts of the country.

- 70-100% districts are affected in Andhra Pradesh, Gujarat and Rajasthan.
- 40-70% districts are affected in Bihar, National Capital Territory of Delhi, Haryana, Jharkhand, Karnataka, Maharashtra, Madhya Pradesh, Odisha, Tamil Nadu and Uttar Pradesh
- 10-40% districts are affected in Assam, Jammu & Kashmir, Kerala, Chattisgarh and West Bengal [30].
- In India, endemic fluorosis is thought to affect 65 million people.

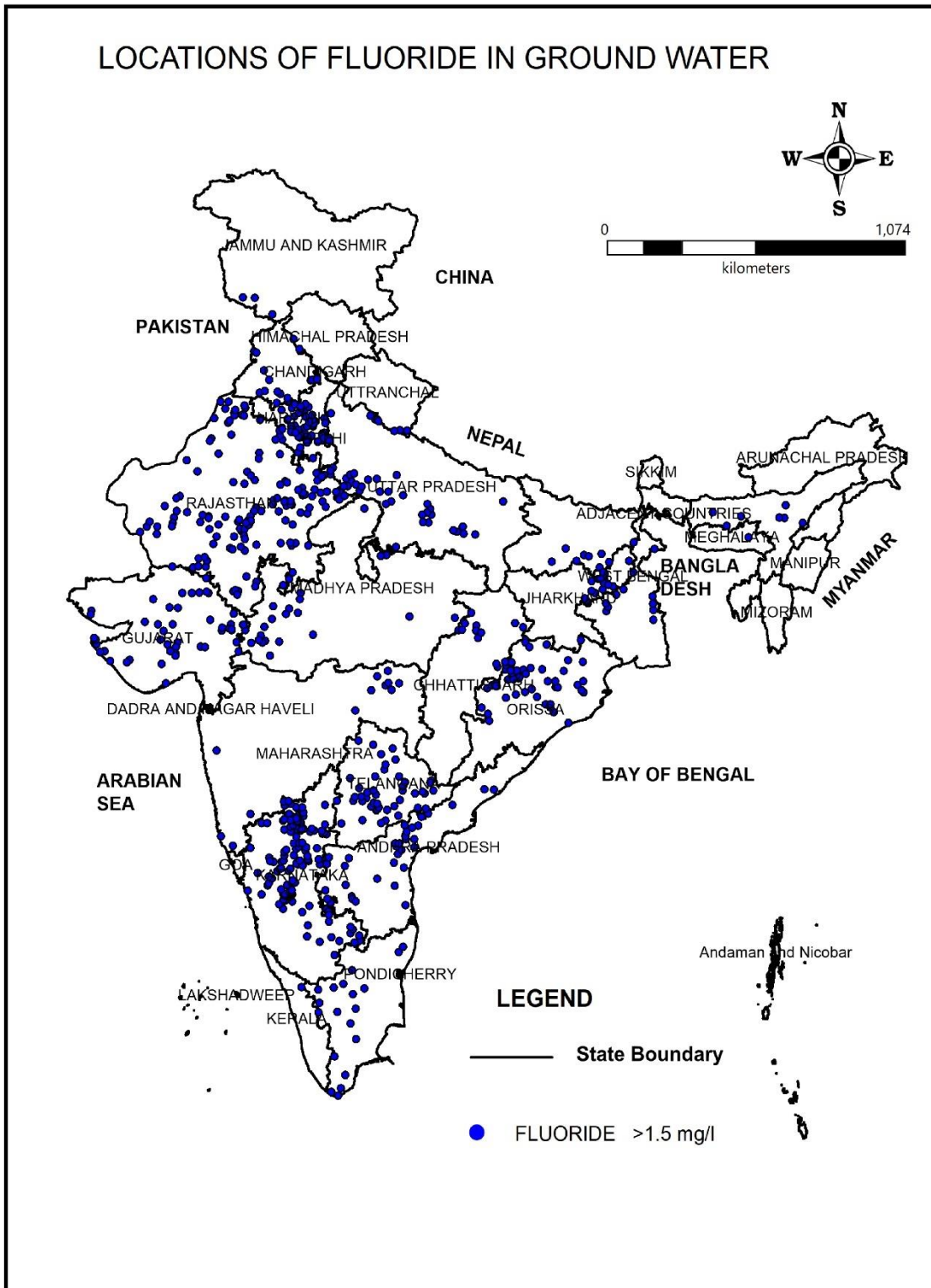


Fig-3: Figure showing fluoride affected areas in India (Source: ScienceDirect.com)

1.6 West Bengal scenario:

According to Central Ground Water Board, the available data suggest that 23 States in India (CGWB, March 2015) are affected by fluorosis endemic (occurrence of Fluoride in ground water >1.5 mg/l). About 62 million people, including 6 million children are at risk in India suffering from dental, skeletal and/or non-skeletal fluorosis (Gitte S. Vilasrao et al., 2015).

And in West Bengal, districts like Bankura, Bardhaman, Birbhum, DakshinDinajpur, Uttar Dinajpur, Malda, Puruliya are all have fluoride at higher concentration (>1.5 mg/l).

And in Bankura there are 9 blocks which are fluoride affected just like Chhatna, Barjora, Gangajalghati, Saltora, Raipur, Taldangra, Indpur, Hirbandh.

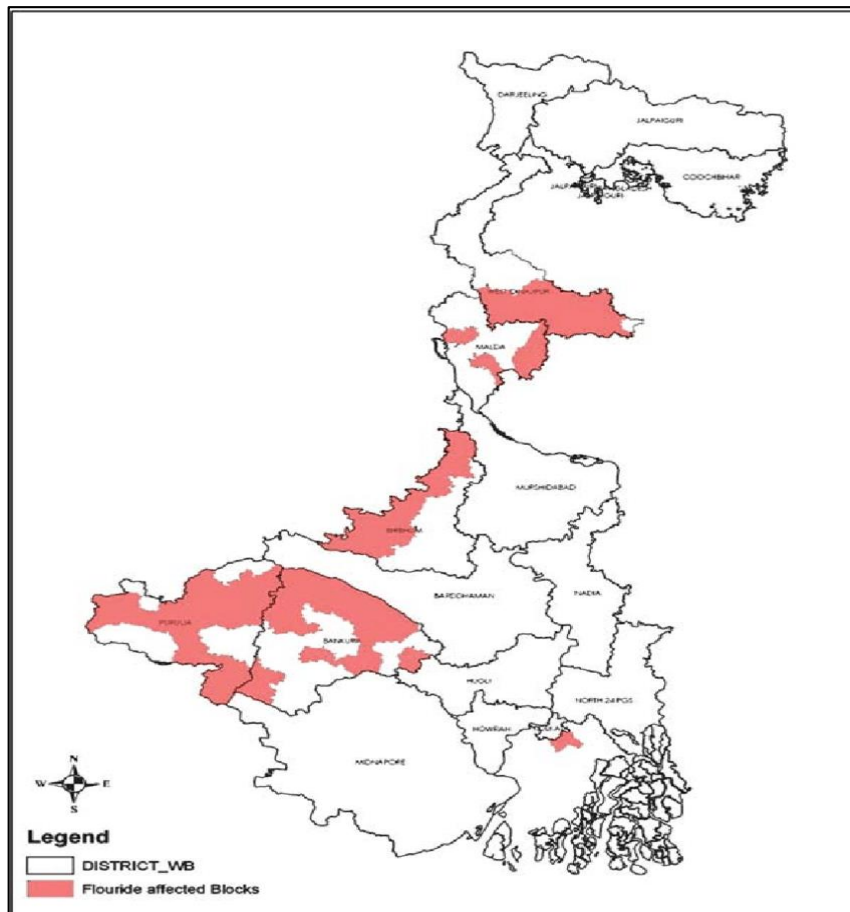


Fig-4: Figure Showing Fluoride affected areas in West Bengal (Source: Researchgate.net)

1.7 Research Gap:

Fluoride is not only present in water. It can be present in sea food. Grapes, Raisins and wine Grapes in all the forms contain fluoride. Potatoes, Baked Potatoes are a good source for fluoride. All vegetation contains some fluoride which is absorbed from soil and water. And tea in particular can contain high fluoride concentrations. So, we can say that in addition to drinking water, traditional habits of tea consumption can cause an additional intake of fluoride in the population. In Sri-Lanka, a total number of 39 locally blended black tea samples were collected from a village where chronic kidney disease with undetermined origin (CKDu) is prevalent. An estimate of daily total average fluoride intake via tea consumption per person is 2.68 mg per day. So, in this paper we have collected tea samples and we examine the sample, and get the result.

1.8. Objective:

The main objective of the research study is to estimate the fluoride concentration in drinking water and in different food items and also development of sustainable fluoride removal methodology.

- To quantification content of fluoride in food items just like tea, hazmola, black salt, dalmut etc.
- To quantification content of fluoride in water sample just like tap well, hand tube well, well
- To analyze the efficiency of the filter. How much it can remove fluoride from filter.

1.9. Proposed Outcome:

- Quantify content of fluoride in various food items.
- Quantify content of fluoride of in food in water sample.
- To analyze the efficiency of the filter.

1.10 Thesis Outline: Fluoride in food, fluoride in water, development of a filter is done in this research work. This research work consists of nine chapters. Chapter 2 includes of literature review, chapter 3 consists of fluoride in food, chapter 4 includes health in food, chapter 5 includes of study area, chapter 6 is discussed about methodology, chapter 7, 8, 9 are discussed about materials and methods, and results, and conclusion.

CHAPTER 2

LIRERATURE REVIEW

2. Literature Review:

International:

Y.S.Solanki, et al. (2022) has studied on Fluoride occurrences, health problems, detection, and remediation methods for drinking water. Fluoride contamination has become a considerable threat to our society worldwide. Fluoride in drinking water is primarily due to rich fluoride soil, volcanic activity, forage, grasses and grains, and anthropogenic reasons. World Health Organization has regulated the upper limit for fluoride in drinking water to be 1.5 mg/L while different countries have set their standards according to their circumstances. Excess amounts of fluoride ions in drinking water can cause dental fluorosis, skeletal fluorosis, arthritis, bone damage, osteoporosis, muscular damage, fatigue, joint-related problems, and chronic issues. In extreme conditions, it could adversely damage the heart, arteries, kidney, liver, endocrine glands, neuron system, and several other delicate parts of a living organism, briefed in the present article. Moreover, a comprehensive scenario for the situations in countries like, China, Canada, Mexico, United States, Yemen, Pakistan, Saudi Arabia, South Korea, Sri Lanka, Indonesia, Iran, Turkey, Australia, and India affected with high fluoride levels in ground water has been described. To analyse the presence of fluoride molecule, out of different detections methods, ion selective and colorimetric method has been adopted for real situation in the field of water application. Also, different methods to remove fluoride from water like reverse osmosis, nano filtration, adsorption, ion-exchange, and precipitation/coagulation with their removal mechanism were highlighted in the review.

R.Chandrajith, et al. (2021) has studied on exposure assessment of fluoride intake through commercially available Black Tea (*Camellia Sinesis L.*) in Sri Lanka. Fluoride is a beneficial trace element for human health effects. In Sri Lanka, dry zone regions can have excessive levels of fluoride can cause dental and skeletal problems. Addition to drinking water, traditional habits of tea consumption can cause an additional intake of fluoride in the population. This article showed elevated levels of F⁻ in tea infusions. Such conditions are present in ground waters in the Girandurukotte region in Sri Lanka and likely in other regions of the dry zone. But not only by drinking water, by drinking tea we also intake fluoride. A total number of 39 locally blended black tea samples were collected from a village where chronic kidney disease with undetermined origin is prevalent. The mean fluoride content was more in loose tea than in packed tea samples, and then in unblended tea. An estimate of the daily total average fluoride intake via tea consumption is 2.68 mg per day.

Teshome.L.Yami, et al. (2018) has discovered a research paper on performance enhancement of nalgonda technique and pilot testing electrolytic defluoridation system for removing fluoride from drinking water in East Africa. High fluoride concentrations in groundwater pose a health risk to people living in the Rift valley of Ethiopia and beyond. The Nalgonda and electrolytic defluoridation (EDF) fluoride treatment systems were developed and adapted in India for fluoride removal. A recent study evaluated twenty Nalgonda techniques that were implemented in the Rift valley of Ethiopia. A number of these systems were found to be non-functional or had never been utilized. The purpose of this study is to evaluate the performance of the Nalgonda technique and seek ways to enhance the fluoride uptake capacities. This study has shown that the performance of the Nalgonda system was significantly enhanced by adding aluminum hydro(oxide)(AO) and cow bone char powder into the existing Nalgonda systems; the initial fluoride concentration of 9.3 mg/L was lowered to 2.5 mg/L on average. The EDF system proved to be effective at removing the excess fluoride concentration in drinking water in the Rift Valley of Ethiopia; the initial fluoride concentration of 7.9mg/L was lowered to 2.8 mg/L meeting the USEPA standard fluoride level of 4 mg/L. The pilot study showed Aluminum leaching into the treated water. Thus,

further optimization of the electrode size, electrolysis time, and voltage/current used during the electrolysis process is needed to meet the WHO target treatment goal of 1.5 mg/L fluoride level and eliminate aluminum leaching as well.

M.T.Samadi, et al. (2014) has studied on the removal of fluoride ions by ion exchange resin. The extent of the problem of the excessive fluoride in drinking water remains an important health issue. In the present work, strong-base anion resins were investigated to assess their sportive removal capacity of fluoride from water. It was found that the removal of fluoride was high at natural pH (7) and was improved for increasing contact time and adsorbent dosage. Removal of fluoride by a strong-base anionres in was investigated. The considered adsorbent showed high fluoride removal efficiency. Three isotherm models and four kinetic models were tested. The adsorption process was found to follow a Freundlich isotherm model. Maximum capacity was obtained (13.7 mg/g) and was compared to other sorbents given in the available literature. Pseudo-first order kinetic model appeared relevant to describe the fluoride adsorption onto the resin; the calculated q_e values were close to the experimental ones. It was also observed that Film diffusion model was the rate-controlling step. Response surface method and 3D counter plots were used for the determination of the optimal conditions for fluoride removal in the considered concentration range. To approach real conditions, the effect of coexisting ions was investigated showing a slightly reverse effect of hardness and chloride ions while a higher reverse effect for nitrate. Contrarily, fluoride removal was found to be not affected by the presence of sulfate.

M.E.Kaseva, (2006) studied on optimization of regenerated bone char for fluoride removal in drinking water, which is a case study of Tanzania. In Tanzania where more than 30% of water of all water sources have fluoride concentration above 1.50 mg/l which is recommended by WHO. In this study, regeneration temperature, regeneration duration, contact time, regenerated bone char dosage and particle size were investigated. Results indicate that the highest fluoride removal and adsorption capacity were 70.64%, for a sample with bone char material that was regenerated at 500°C. In this study the optimum burning duration was found to be 120 min, which resulted in residual fluoride that varied from a maximum value of 17.43 mg/l for a 2 min contact time to a minimum value of 8.53 mg/l for a contact time of 180 min. In This study further we can say that the smallest size of regenerated bone char media (0.5– 1.0 mm diameter) had the highest defluoridation capacity, with residual fluoride which varied from 17.82 mg/l at 2 min contact time to 11.26 mg/l at 120 min contact time. In terms of dosage of the regenerated bone char media it was established that the optimum dosage was 25 g of bone char media with a grain size of 0.50–1.0 mm. Column filter experiments indicated that regenerated bone media is capable of removing fluoride from drinking water to meet both WHO and Tanzania recommended values.

C.Y. Hu, et al.(2003) has discovered a paper on effects of co-existing anions on fluoride removal in electrocoagulation (EC) process using aluminum electrodes. Batch experiments with bipolar aluminum electrodes and potentiodynamic polarization tests with monopolar systems were conducted to investigate the effects of the type and concentration of co-existing anions on defluoridation in electrocoagulation (EC) process. The results demonstrate that the type of the dominant anion directs the reaction. The defluoridation efficiency was almost 100% and most of the fluoride removal reaction occurred on the surface of the anode in the solution without the co-existing anions, due to the electro-condensation effect. In the solutions with co-existing anions, most of the defluoridation took place in bulk solution. The existence of sulfate ions inhibits the localized corrosion of aluminum electrodes, leading to lower defluoridation efficiency because of lower current efficiency. The presence of chloride

or nitrate ions prevented the inhibition of sulfate ions, and the chloride ions were more efficient. Different corrosion types occurred in different anion-containing solutions and the form of corrosion affected the kinetic over-potential. The bypass flow causes the decrease of current efficiency and the proportion of the bypass flow of current increased due to a rise of the kinetic over potential and the conductivity of the solution.

M.K.Malde, et al. (1996) has studied on fluoride content in selected food items from five areas in east Africa. Dental fluorosis is widespread in the eastern part of Africa. Drinking water has traditionally been considered the main reason for the development of fluorosis, but food items may also be a contributor in areas with high concentrations of fluoride in the soil. The purpose of this study was to assess the fluoride content of fish and staple food items commonly consumed by children. Sampling was undertaken during November and December 1993 and March 1994 from the following areas: Awassa and Zwai, Ethiopia; Mwanza and Dar es Salaam, Tanzania; and Bujumbura, Burundi. The fluoride concentration was analyzed by the use of a fluoride selective electrode. Whole fish and bone samples had a high fluoride concentration while fillet samples generally were low in fluoride. The highest fluoride concentrations in fish were found in marine species that are eaten whole. The fluoride concentrations in the vegetables varied, ranging from 0.3 mg/kg dry wt in maize to 7.7 mg F/kg dry wt in spinach. Since a typical East African meal often has starchy food as the central component, a high fluoride content in cereals and legumes maybe of greater concern than food made from whole fish. It is difficult to assess the amount of fluoride absorbed as fluoride interacts with other elements in the diet, which will most likely reduce the absorption.

Table 1: Table showing International papers regarding fluoride removal and adsorbent

Sl No.	Author	Study Area	Focus of the work	Remarks
1	Y.S.Solanki, M.Agarwal, A.B.Gupta, S.Gupta, P.Shukla(2022)	China, Canada, Mexico, Pakistan, United States, India,	To analyze the presence of fluoride molecules, out of different detection methods.	Out of different detections methods, ion selective and coliometric method has been adopted for real situation in the field of water application. Also, different methods to remove fluoride from water with their removal mechanism were highlighted in the review.
2	R.Chandajith, S.Bhagya, S.Diyabalanage, S.Wimalasiri, M.A.B.Ranatunga , J.A.C.Barth. (2021))	SriLanka	To find out, apart from drinking water, tea consumption can also cause an additional intake of fluoride in population.	The mean fluoride content was 2.68±1.03 mg/L in loose tea, 1.87±0.57mg/L in packed tea samples, and 1.14±0.55 mg/L in unblended tea.
3	T.L.Yami, J.F.Chamberlain, F.Z.Beshah D.A.Sabtani(2018)	East Africa	To evaluate the performance of the Nalgonda technique and seek ways to enhance the fluoride uptake capacities.	The raw water fluoride concentration of 7.9 mg/L considered in the pilot testing of the EDF system was reduced to 2.8 mg/L at an electrolysis time of 4 h.
4	M.T.Samadi, M.Zarabbi,	Iran	In the present work, strong-base anion resins were	It was found that the removal of fluoride was high at

	M.N.Sepher, S.M.Ramhormozi , S.Azizan, A.Amrane (2014)		investigated to assess their sorptive removal capacity of fluoride from water.	natural pH (7) and was improved for increasing contact time and adsorbent dosage. The final fluoride concentration was 2.3 mg/L, showing a decrease in removal efficiency which however remained at an acceptable level
5	M.E.Kaseva (2006)	Tanzania	To know how much fluoride removal capacity by bone char.	In this study the optimum burning duration was found to be 120 min, which resulted in residual fluoride that, varied from a maximum value of 17.43 mg/l for a 2 min contact time to a minimum value of 8.53 mg/l for a contact time of 180 min
6	C.Y.Hu, S.L. Lo, W.H.Kuan, Y.D.Lee (2003)	Taiwan	This work employs an anodic surfactant, sodium dodecyl sulfate (SDS), to improve the flotation performance of the electrocoagulation flotation (ECF) process to treated fluoride containing semiconductor wastewater following calcium precipitation.	The results demonstrate that the type of the dominant anion directs the ECdefluoridation reaction. The defluoridation efficiency was almost 100% and most of the fluoride removal reaction occurred on the surface of the anode in the solution without the co-existing anions, due to the electro-condensation effect.
7	M.K.Malde, A.Maage, E.Macha, K.Julshamn, K.Bjorvatn (1996)	East Africa	The purpose of this study was to access the fluoride content of fish and staple food items commonly consumed by children.	The fluoride concentrations in the vegetables varied, ranging from 0.3 mg/kg dry wt in maize to 7.7 mg F/kg dry wt in spinach. Since a typical East African meal often has starchy food as the central component, a high fluoride content in cereals and legumes may be of greater concern than food made from whole fish.

National senerio:

J.Soni, et al. (2020) has studied on impact and removal of techniques of fluoride from the drinking water. Fluorine which is estimated to be the 13th-most abundant element in the earth's crust and is widely dispersed in nature in the form of fluorides. It is found naturally in soil, water, and foods. High-level exposure to fluoride can lead to Fluoride poisoning. Fluoride contamination in drinking water due to natural and anthropogenic activities has been accepted as one of the major problems worldwide imposing a serious threat to human health. The present review emphasizes on efficacy of different methods for the removal of fluoride from water. This review has endeavoured to cover an extensive variety of procedures which have been utilized so far for the removal of fluoride from the drinking water. A deep insight of the survey of literature for defluorination techniques during last twenty years reveals that each of the discussed techniques can remove fluoride under specified conditions. The fluoride removal efficiency varies according to many site specific chemical, geographical and economic conditions, so actual applications may vary from the generalizations made. Any particular process, which is suitable at a particular region may not meet the requirements at some other place. Therefore, any technology should be tested using the actual water to be treated before implementation in the field.

A.Mittal, et al. (2016) has studied on application of Egg Shell and Egg Shell Membrane as adsorbents. Worldwide consumption of hen eggs is quite high, which results into availability of large amount of discarded egg wastes, particularly egg shell (ES) and egg shell membrane (ESM). Disposal of ES and ESM is always a problem particularly to the authorities of food, bakery and poultry units. In recent years, attempts have been made to utilize ES and ESM for the production of biodiesel and collagen but results are not much economically viable. In the hunt of waste materials as potential scavenger of hazardous chemicals, ES and ESM have emerged as non-toxic, versatile and efficient adsorbents. Last 10 years have witnessed a systematic growth in the use of ES and ESM as adsorbents for the removal of variety of organic as well as inorganic hazardous chemicals, particularly from wastewater. Literature survey reveals that ES and ESM in their natural as well as chemically modified forms have provided excellent results for the removal of various classes of dyes, oxalic acid, phenol, pesticides, humic acid, pharmaceuticals, surfactants, PAHs, heavy-, precious- and light- metals, actinides, fluorides, etc. In recent years powdered ESM have been modified to nano-particles and used as adsorbent in various interesting applications. Reports are also available on the use of nanostructured material CHAP, derived from ES, for the removal of cadmium and lead ions from waste water. Present review article is an attempt to summarize the research carried out on the above-mentioned applications of ES and ESM.

K.Das, et al. (2016) has studied on delineation of groundwater quality in the presence of fluoride in selected villages of Simlapal block, Bankura district, West Bengal, India. Fluoride contamination and other physicochemical parameters in groundwater of Simlapal block of Bankura district were investigated. A total of 50 deep tube well (DTW) samples were collected from 18 villages of Simlapal. The higher concentration of iron values was recorded, 9.40 mg/L with an average value of 2.11 mg/L. Drastically, 58 % of water samples exceed the permissible limit of 1.5 mg/L (Guidelines for drinking water quality, World Health Organization, Geneva, 2004). Spatial distribution of F⁻ (Guidelines for drinking water quality, World Health Organization, Geneva, 2004) concentration classes is represented by the simple Inverse Distance Weighting (IDW) interpolation method. The affinity between the Phand F⁻ in groundwater suggests the dissolution of fluoride bearing minerals in groundwater. F1 values have high loading (32.71%), second component (F2) exhibits 19.98 %, third component (F3) shows 12.74 %, and fourth component (F4) shows only 8.72 % of the total variability to justify the significant correlation between chemical constituents. In terms of quality of water, sodium adsorption rate (SAR) showed that all the samples are from the group of excellent to good. With respect to permeability, sodium percentages show that 18 % are permissible but doubtful and 2 % are unsuitable for irrigation purposes.

Sanghratna S. Waghmare, et al. (2015) has studied on fluoride removal from water by various techniques which depicts the fluoride removal from drinking water can be accomplished by different methods, for example, coagulation-precipitation, membrane separation process, ion exchange, adsorption techniques and so on. Among these procedures, membrane and ion exchange processes are not extremely regular because of its high establishment and support price. Other two techniques are extremely regular in India. Nalgonda procedure is one of the well-known strategies generally utilized for defluoridation of water as a part of developing nations, for example, India, Kenya, Senegal and Tanzania. Among different techniques utilized for defluoridation of water, the adsorption procedure is broadly utilized and offers acceptable results and is by all accounts more appealing technique for the removal of fluoride regarding expense, straightforwardness of outline and operation. It is apparent from the literature study that different methods have indicated novel potential for

the removal of fluoride. This review has endeavored to cover an extensive variety of procedures which have been utilized so far for the removal of fluoride from the drinking water and industrial wastewater. It was reasoned that coagulation strategies have by and large been discovered compelling in defluoridation, however they are unsuccessful in conveying fluoride to fancied concentration levels. The quest for option and appropriate fluoride removal methods thusly still stays of curiosity.

M.Suneetha, et al. (2015) studied on defluoridation techniques. Fluoride is frequently depicted as a 'double-edged sword' as an insufficient intake is related to dental caries and too much ingestion leads to dental and skeletal fluorosis which has no treatment. Only Prevention is by using defluoridation techniques. This review article is aimed at providing specific information on attempts made by various researchers in the field of fluoride removal from polluted waters. Lots of de-fluoridation techniques are named as, precipitation, ion-exchange, reverse osmosis, electro dialysis, donnan dialysis, nanofiltration, electro coagulation and adsorption etc. And among these techniques, adsorption process is economical, efficient, ease of operation and produces high-quality water. Studies on fluoride removal from waste waters using various adsorbents such as alumina/aluminium based materials, lime, clays and soils, calcium based minerals, bone, bone char, synthetic compounds and carbon based materials are reviewed. Out of all the methods, the adsorption technique is inexpensive, efficient, ease of operation and produces high-quality water. Thus, we can conclude that the studies in the field of fluoride remediation have been reviewed effectively based on adsorption and the sorbents derived from natural sources is increasingly probed and due to the encouraging results, much research is endeavored to be devoted as these methods are based on eco-friendly and easily available adsorbents

T.Gill, et al. (2014) studied on conventional fluoride removal technique in urban areas. This study was carried out to evaluate the feasibility of conventional rural based defluoridation technique Nalgonda Technique, Activated Alumina in urban areas. Nalgonda technique is being used extensively due to ease in construction of the reactor, operation and maintenance. This technique is very effective even when the fluoride concentration is above 20 mg/L. However, generation of acid or alkali water, residual aluminium, soluble aluminium fluoride complexes and fluoride contaminated sludge limits its practical applications. Activated Alumina though showed effective removal of fluoride, slow rate of adsorption, pH adjustment, and accumulation of bacteria in the long run inhibits its commercial application. Also sludge generation is one of its drawbacks. Nalgonda process did not achieve a great degree of success in field application as it removes only (18-33)% in the form of precipitates and converts a greater portion of ionic fluoride (67-82)% into soluble aluminium fluoride complex ion and therefore this process is erroneous. Defluoridation using Activated Alumina has been one of the widely used adsorption/ ion exchange methods water and many reports are available on large scale installation for townships, requiring superposition and skilled personnel. The quality of treated water from such facilities was assured. This process is highly selective and is pH specific with low adsorption capacity, poor physical integrity, requires acidification, and pretreatment and its effectiveness for fluoride removal reduces after each generation.

J.Shen, et al. (2014) researched a paper on removal of fluoride by reverse osmosis and nano filtration. Inorganic contamination in drinking water, such as fluoride has been recognized as a worldwide problem imposing a serious threat to human health. Among several treatment technologies applied for fluoride removal, nanofiltration (NF) and reverse osmosis (RO) have been studied extensively and proven to offer satisfactory results with high selectivity. In this

review, a comprehensive summary and critical analysis of previous NF and RO applications on fluoride removal is presented. Removal of fluoride by NF/RO from water has been reported to be successful. (Ayoob et al.,2008). Dolar et al. (2011) published fluoride rejections of >96% with RO, >90% with tight NF and >50%with loose NF membranes. A study compared fluoride rejections of three NF membranes with different configuration and found that the double pass with TR60 or NF270 membranes is comparable to the simple pass with NF90 which is similar to RO membranes functional groups. The influence of operating conditions (pressure, cross flow velocity), water quality (concentration, solution pH), solute–solute interactions, membrane characteristics and membrane fouling on fluoride and uranium retention is critically reviewed.

A.Bhatnagar, et al. (2011) has studied fluoride removal from water by adsorption has been explored widely and offers satisfactory results especially with mineral-based and/or surface modified adsorbents. In this review, an extensive list of various adsorbents from literature has been compiled and their adsorption capacities under various conditions (pH, initial fluoride concentration, temperature, contact time, adsorbent surface charge, etc.) for fluoride removal as available in the literature are presented along with highlighting and discussing the key advancement on the preparation of novel adsorbents tested so far for fluoride removal. This review has attempted to cover a wide range of adsorbents which have been used so far for the removal of fluoride from the water and wastewater. Based on the literature reviewed, the adsorbents are activated alumina, rare earth oxide base materials, carbon based adsorbents, biosorbents, red mud, slag, sludge etc.

M.M.Emamjomeh, et al. (2008) has studied on fluoride removal by a continuous flow reactor electro coagulation process which is an electrochemical technique, in which variety of unwanted dissolved particles and suspended matter can effectively remove from an aqueous solution by electrolysis. Continuous flow experiments with monopolar aluminium electrodes for fluoride removal were undertaken to investigate the effects of the different parameters such as: current density, flow rate, initial pH, and initial fluoride concentration. The highest treatment efficiency was obtained for the largest current and the removal efficiency was found to be dependent on the current density, the flow rate and the initial fluoride concentration when the final pH ranged between 6 and 8. The fluoride removal efficiency is increased to 99% at 50 A/m² when the flow rate and initial fluoride concentration are, respectively, kept at 150 mL/min and 10 mg/L. However, in order to avoid excessive energy consumption, it is advisable to limit the current rate when a desirable effluent fluoride concentration range is achieved.

M.Sarkar, et al. (2006) has studied on use of laterite soil of fluoride from contaminated water which is now days a modern solution for fluoride removal. In this process the laterite soil was collected and then severally water washed with distilled water. The column efficiency was tested from the depth-service model. Adsorptive removal of fluoride from water using laterite is influenced by several operational parameters: initial fluoride concentration, agitation time and speed, dose and particular size of laterite, solution pH, and temperature. The bed depth–service time model is constructed to predict fluoride removal corresponding to a certain bed height. The use of laterite Adsorptive removal of fluoride from water using laterite is influenced by several operational parameters: initial fluoride concentration, agitation time and speed, dose and particle size of laterite, solution pH, and temperature. The percent decrease in fluoride concentration was evaluated at the optimized experimental condition. It is found that fluoride removal is favoured at lower concentration, smaller particle size of laterite, increased agitation time and speed, increased dose of laterite,

and lower temperature. The optimum pH for maximum solute removal, as an adsorbent for fluoride removal is found to be cost effective and hence can be considered as an alternative for defluoridation.

Meenakshi, et al. (2006) has discovered a paper on fluoride removal on drinking water. Excessive fluoride concentrations have been reported in ground waters of more than 20 developed and developing countries including India where 19 states are facing acute fluorosis problems. Various technologies are being used to remove fluoride from water but still the problem has not been rooted out. In this paper, a broad overview of the available technologies for fluoride removal and advantages and limitations of each one have been presented based on literature survey and the experiments conducted in the laboratory with several processes. It has been concluded that the selection of treatment process should be site specific as per local needs and prevailing conditions as each technology has some limitations and no one process can serve the purpose in diverse conditions. The literature survey and the laboratory experiments have indicated that each of the discussed techniques can remove fluoride under specified conditions. The fluoride removal efficiency varies according to many site-specific chemical, geographical and economic conditions, so actual applications may vary from the generalizations made. Any particular process, which is suitable at a particular region may not meet the requirements at some other place. Therefore, any technology should be tested using the actual water to be treated before implementation in the field.

N.Roy (2003) has studied on fluoride and environment. The presence of fluoride in exceeding limits and its related problems of drinking water prevailing in many parts of India is well documented. Fluoride in drinking water is known for both beneficial and detrimental effects on health. Many solutions to these problems were also suggested. Fluoride from water or wastewater can be removed by an ion exchange/adsorption process or by a coagulation, precipitation process. The ion exchange/adsorption can be applied to either concentrated or diluted solutions and they are capable of achieving complete removal under proper conditions. The method suitable for a given situation needs to be judiciously selected considering the various aspects. The paper presents the current information on fluoride in environment and its effects on human health and available methods of defluoridation in detail. Certain under-exploited but abundantly available materials like rare earth materials have indicated excellent potential for fluoride uptake. Attempts may be made to develop cost effective, simple to use technologies based on this material.

Table-2: Table showing national papers regarding fluoride removal and adsorbent

Sl No.	Author	Study Area	Focus of the work	Remarks
1	J.Soni, N.Bansal, M.Gupta(2020)	India (Kashmir, Punjab, Uttar Pradesh, Rajasthan, Gujrat)	This present review emphasizes on efficacy of different methods for the removal fluoride from water.	The fluoride removal efficiency varies according to many site specific chemical, geographical and economic conditions, so actual applications may vary from the generalizations made.

2	A.Mittal, M.Teotia, R.K.Soni, J.Mondal (2016)	India	To evaluate the performance of egg shell and egg shell membrane as adsorptions of various pollutants.	Bird eggs have interesting anatomy and their waste materials, ES and ESM can be utilized for various purposes, including adsorption of various pollutants. The literature review presented above clearly indicates that the adsorption abilities of ES and ESM have been smartly exploited by various researchers.
3	K.Das, U.Dey, N.K.Mondal (2016)	Simlatal block, (Bankura district)	In this article Fluoride contamination and other physicochemical parameters in groundwater of Simlatal block of Bankura district were investigated.	The higher concentration of iron values was recorded, 9.40 mg/L with an average value of 2.11 mg/L. Drastically, 58 % of water samples exceed the permissible limit of 1.5 mg/L.
4	M.Suneetha, B.S.Sundar (2015)	India	This review article is aimed at providing specific information on attempts made by various researchers in the field of fluoride removal from polluted waters.	Studies on fluoride removal from waste waters using various adsorbents such as alumina/aluminium based materials, lime, clays and soils, calcium based minerals, bone, bone char, synthetic compounds and carbon based materials are reviewed.
5	S.S.Waghmare, T.Arfin (2015)	India	This paper depicts the fluoride removal from drinking water can be accomplished by different methods.	The advantages and disadvantages of every treatment procedure has been explained.
6	J.Shen, A.Schafer (2014)	World wide	A comprehensive summary and critical analysis of previous NF and RO applications on fluoride removal is presented.	Removal of fluoride by NF/RO from water has been reported to published fluoride rejections of >96% with RO, >90% with tight NF and >50% with loose NF membranes.
7	T.Gill, S.Tiwari, P.A.Kumar(2014)	Urban areas in India.	To remove fluoride from water in urban areas in India.	Nalgonda process removes only a smaller portion of fluoride (18-33%) and Activated Alumina has been one of the widely used adsorption method.

8	A.Bhatnagar, E.Kumar, M.Sillanpaa (2011)	India	In this review, an extensive list of various adsorbents from literature has been compiled and their adsorption capacities under various conditions.	A large number of effective, low cost adsorbents such as activated alumina, amorphous alumina, calcite, rare earth oxides, bleaching powder, fly ash, lime stone, clay minerals and many others are used. This process is highly efficient.
9	M.M.Emanjomeh, M.Sivakumar(2008)	India, China, Australia	To remove fluoride from water and give safe water.	The highest treatment efficiency was obtained for the largest current and the removal efficiency was found to be dependent on the current density, the flow rate and the initial fluoride concentration when the final pH ranged between 6 and 8.
10	M.sarkar, A.Banerjee, P.P.Pramanick, A.R.Sarkar (2006)	Bankura district, West Bengal, India.	To know the effects of different operational variables on the mechanistic function of laterite in removal of fluoride has been investigated.	It is found that fluoride removal is favoured at lower concentration, smaller particle size of laterite, increased agitation time and speed, increased dose of laterite, and lower temperature.
11	S.Meenakshi, R.C.Maheswari (2006)	New Delhi, India	To know which removal process is useful in which condition and their efficiency.	The fluoride removal efficiency varies according to many site-specific chemical, geographical and economic conditions, so actual applications may vary from the generalizations made.
12	N.Roy (2003)	India	The paper presents the current information on fluoride in environment and its effects on human health and available methods of defluoridation in detail	The ion exchange/adsorption can be applied to either concentrated or diluted solutions and they are capable of achieving complete removal under proper conditions. The method suitable for a given situation needs to be judiciously selected considering the various aspects.

CHAPTER 3

FLUORIDE IN FOOD

Chapter 3

3.1 Fluoride in Food

Fluoride in Food: Dentists love fluoride as it can strengthen and re-mineralize damaged enamel, making it more resistant to decay. And this is the reason fluoride is added to public drinking water and toothpaste and mouthwash. Contrary to what you might hear, fluoride is a mineral that occurs in nature in many different places, including our food items. Here are a few fluoride-rich foods you now have a new excuse to enjoy.

Spinach: Popeye’s favourite superfood, spinach is packed with all kinds of great vitamins and minerals, and fluoride is among them. Spinach contains 0.07 mg of fluoride per cup. It is also a great source of Vitamin C, which can strengthen the immune system and help your body fight infections.

Grapes, Raisins, and Wine: Grapes are also a natural source of fluoride, but not in very high concentrations compared to raisins, which have more than 20 times as much fluoride as raw grapes. Wine is also a good source of fluoride, particularly white wine, which contains nearly twice as much as red wine.

Black Tea: You get your daily caffeine hit from black tea, you’re also adding to your fluoride intake. Some people believe that the high level of fluoride in tea simply comes from preparing it with fluoridated tap water. In fact, the tea leaves themselves contain the mineral. Brewing your black tea with tap water just increases the fluoride content a little bit more. But be warned, black tea will stain your teeth, even quicker than black coffee!

Potatoes: Potatoes are also a decent source, particularly russet potatoes, which contain 0.49 ppm of fluoride. However, this fluoride content depends on how the potatoes are prepared: baked or baked potatoes and hash browns contain twice as much fluoride as french fries.

Please note, even if you eat a lot of the foods mentioned above, the best ways to get the fluoride you need to keep your teeth strong are drinking tap water and using fluoride toothpaste

Age	Male	Female	Pregnancy	Lactation
Birth to 6 months	0.01 mg	0.01 mg		
7–12 months	0.5 mg	0.5 mg		
1–3 years	0.7 mg	0.7 mg		
4–8 years	1 mg	1 mg		
9–13 years	2 mg	2 mg		
14–18 years	3 mg	3 mg	3 mg	3 mg
19+ years	4 mg	3 mg	3 mg	3 mg

Table-4 : Fluoride Content of Selected Foods	
Food	Milligrams per Serving
Tea, black, brewed, 1 cup	0.07 to 1.5
Coffee, brewed, 1 cup	0.22
Shrimp, canned, 3 ounces	0.17
Bottled water with added fluoride, 1 cup	≤0.17
Raisins, ¼ cup	0.08
Oatmeal, cooked, ½ cup	0.08
Grapefruit juice, ¾ cup	0.08
Potatoes, russet, baked, 1 medium	0.08
Rice, cooked, ½ cup	0.04
Cottage cheese, ½ cup	0.04
Pork chop, baked, 3 ounces	0.03
Yogurt, plain, low-fat, 1 cup	0.03
Lamb chop, cooked, 3 ounces	0.03
Tortilla, flour, 1 tortilla, approx. 10” diameter	0.02
Corn, canned, ½ cup	0.02
Beef, cooked, 3 ounces	0.02
Tuna, light, canned in water, 3 ounces	0.02
Cheese, cheddar, 1½ ounces	0.01
Bread, white or whole wheat, 1 slice	0.01
Asparagus, cooked, 4 spears	0.01
Chicken, cooked, 3 ounces	0.01
Milk, fat-free or 1%, 1 cup	0.01
Apple, raw, with skin, 1 medium	0.01
Avocado, raw, ½ cup sliced	0.01
Macaroni, plain, cooked, ½ cup	0.00
Tomato, raw, 1 medium	0.00
Bananas, 1 medium	0.00
Egg, cooked, 1 large	0.00
Carrots, raw, 1 medium	0.00
Peanut butter, 1 tbsp	0.00

3.2 Sources of Fluoride

Food

Brewed tea typically contains higher levels of fluoride than most foods, depending on the type of tea and its source, because tea plants take up fluoride from soil. Fluoride levels can range from 0.3 to 6.5 mg/L (0.07 to 1.5 mg/cup) in brewed tea made with distilled water

Fluoride concentrations in breast milk are so low that they cannot always be detected; when these levels can be measured, they range from less than 0.002 to 0.01 mg/L, even when mothers live in communities with fluoridated water. Fluoride concentrations in cow's milk are also very low, ranging from 0.007 to 0.086 mg/L. Fluoride levels in infant formulas in the United States vary, depending on the type of formula and the fluoride content of the water used to prepare the formula. The typical fluoride concentration is less than 0.2 mg/L in milk-based infant formula and 0.2 to 0.3 mg/L in soy-based infant formula (not including contributions from tap water used to prepare the formula).

Only trace amounts of fluoride are naturally present in most foods, and most foods not prepared with fluoridated water provide less than 0.05 mg/100 g.

A variety of types of foods and their fluoride levels per serving are listed in Table 2.

The element named as fluorine, a pale greenish yellow gas, condenses to liquid at 120°C and freezes at minus 250°C. It is very active and, because of its strong tendency to combine with other elements, rarely occurs in gaseous form. Fluoride ion is widespread in nature it is estimated to be thirteenth in abundance among the elements of the earth.

3.3 Fluoride Ion in Food From:

Fluoride is a mineral well known for strengthening teeth and helping prevent dental cavities. Since the amount of fluoride in foods can vary, it is added to public water supplies in some countries, including the US.

According to the National Library of Medicine (NLM), the amount of fluoride you need depends on your body mass (weight). Children typically need 0.5mg (500µg) per day, while adults need 3-4mg (3000-4000µg) of fluoride each day. For the adequate intake (AI) calculations below 4mg (4000µg) was used.

Since consuming too much fluoride can have ill health effects, the NLM sets the upper limit at 10mg, or 10,000µg. This is just 2.5 times the adequate intake.

Fluoride tends to accumulate in a mixture of both healthy and unhealthy foods including tea, coffee, shellfish, grapes (raisins, wine, grape juice), artificial sweeteners, sodas, potatoes, flavored popsicles, baby foods, broths, stews, and hot cereals made with tap water. If fluoride fortified water is used to make these foods and drinks, they will contain even more fluoride.

Further sugary or carbonated drinks can lead to more tooth decay, wiping out any good effect from the fluoride.

For all these fluoride foods and drinks, if fluoride fortified water was used in their creation, they will have even more fluoride. Below are the top 10 sources of fluoride, for more, see the extended list of fluoride rich foods and drinks.

Black Tea:

The black tea is brewed with often water. The amount of fluoride will depend on the levels in the water used to prepare the tea. Even commercial teas sold in bottles and cans may contain some kind of fluoride.

Fluoride per cup gives 883 μg and fluoride per 100 g gives 372.9 μg , and fluoride per 200 calories 74580 μg .



Figure-5: Black Tea, Source-: www.myfooddata.com

Fruit Juice: fruit juice which is high in sugar, which can harm teeth. Fluoride per cup gives 349.1 μg , fluoride per 100 gm gives 138 μg and fluoride



Figure-6: Fruit Juice, Source-: www.myfooddata.com

Sodas: Here also the amount of fluoride will depend on how much water we use to prepare it. Sugary sodas are not healthy food and are best avoided. Further, the acidic carbonation in soda can cause tooth decay. Fluoride per 12oz Can give 299.8 μg and fluoride per 100 gm gives you 80.6 μg and fluoride per 200 calories give 335.8 μg of fluoride.



Figure-7: Soda, Source-: www.myfooddata.com

Blue Crabs: Fluoride uptake rates were studied in exoskeleton, gill, hepatopancreas,

and muscle tissues. Concentration of this ion in crabs from natural, unpolluted water (0.5–1.5 ppm F⁻) averaged 298 ppm in the exoskeleton, 253 ppm in the gills, 22 ppm in the hepatopancreas, and 10 ppm in the muscles on a dry weight basis. Crabs were maintained, and tissue analyses were made, for up to 90 days at 20 and 100 ppm, 22 days at 200 ppm, and 70 days at 400 ppm fluoride. The tissues accumulated significant amounts of fluoride at all experimental water fluoride levels.



Figure-8: Shrimp, Source- www.myfooddata.com

Shrimp: Shrimps have the lowest fluoride content in their outer shell since it acts as an effective biosorbent. There are lots of sea foods and fishes which contain fluoride, among them shrimp is one. Shrimp contains 0.17 mg per serving fluoride.



Figure9-:Blue Crabs, Source- www.myfooddata.com

Coffee: The amount of fluoride the coffee contains will depend on how much water we used to prepare it. Coffee generally contains 0.1-0.58 mg/L of fluoride in infusion solution and 0.15-0.56 mg/L in instant pack. There are few publications on the amount of fluoride in alcoholic beverages, but Polish studies that 0.093 mg/L of fluoride is contained in 10-20% alcohol and 0.056 mg/L in (20-40)%



Figure-10: Coffee, Source: www.myfooddata.com

Raisins: Raisins are very high in sugar and they can get stuck to between teeth. As such, they can increase the risk of teeth decay. Fluoride per Oz can give up to 60.8 μg , fluoride per 100 gm give 233.9 μg and Fluoride per 200 calories give up to 156.5 μg of fluoride.



Figure -11: Raisins, Source: www.myfooddata.com

CHAPTER-4

HEALTH

Chapter-4

4.1 Impact of excess fluoride on health

As a whole fluoride is one of the most beneficial micro nutrients for our body but in excess it can harm us in many ways. According to recommendation of World Health Organization total daily fluoride exposure would be approximately 0.6 mg/adult/day in an area in which no fluoride is added to the drinking-water and 2 mg per adult per day in a fluoridated area (Sananda et al., 2015). The most important toxic effect of fluoride on human beings is skeletal fluorosis, which is endemic in areas with soils and water containing high fluoride concentrations. The sources of fluoride that contribute to the total human intake vary geographically between endemic fluorosis areas, but the symptoms are generally similar. They range from skeletal histological changes, through increases in bone density, bone morphometric changes, and exostoses to crippling skeletal fluorosis. This condition is usually restricted to tropical and subtropical areas, and is frequently complicated by factors such as calcium deficiency or malnutrition (IPCS et al., 1984).

In non-endemic areas, skeletal fluorosis has occurred as a result of industrial exposure. This condition, whether of endemic or industrial origin, is normally reversible by reducing fluoride intake (IPCS et al., 1984).

In endemic fluorosis areas, developing teeth exhibit changes ranging from superficial enamel mottling to severe hypoplasia of the enamel and dentine (IPCS et al., 1984). Patients with kidney dysfunction may be particularly susceptible to fluoride toxicity. (IPCS et al., 1984). Acute toxicity usually occurs as a result of accidental or suicidal ingestion of fluoride, and it results in gastrointestinal effects, severe hypocalcaemia, nephrotoxicity, and shock. Inhalation of high concentrations of fluorine, hydrogen fluoride, and other gaseous fluorides may result in severe respiratory irritation and delayed pulmonary oedema. Exposure of the skin to gaseous fluorine results in thermal burns, while hydrogen fluoride causes burns and deep necrosis (IPCS et al., 1984). A special case of acute toxicity is the reversible water-losing nephritis caused by metabolic liberation of fluoride ions from fluoride-containing anaesthetics gases (IPCS et al., 1984).

Table- 5. Adverse effects of fluoride on physiological health (Sananda et al., 2015)

Fluoride concentrations	Exposure	Physiological effects
Greater than 1 ppm	Chronic	Impaired glucose metabolism in both animals and humans.
Greater than 4 ppm	Chronic	Skeletal fluorosis, dental fluorosis hypo-mineralization of tooth enamel.
Sub-acute exposure to fluoride at a dose of 20 ppm/day (orally to rat)	Chronic	Induces thyroidal dysfunction including suppressed synthetic machinery of the thyroid gland to produce thyroid hormones, mainly T3 and T4. Alteration of Na ⁺ -K ⁺ -ATPase, thyroid peroxidase, and 5,5'- deiodinase.
100-200 ppm	Chronic	Changes on the thyroid hormone status, the histopathology of discrete brain regions, the acetylcholine esterase activity and the learning and memory abilities in multi-generation rats.
High fluoride	Chronic	Increases risk of Alzheimer's disease.

High fluoride	Chronic	Increased levels of FSH and LH, Decreased estrogen levels, Decreased testosterone levels and changes in its conversion into its potent metabolites, Reduced thyroid hormones, Disturbed androgen to estrogen ratios (A/E) and estrogen receptor to androgen receptor ratios (ER/AR).
Greater than 1,000-1,500 ppm (in toothpastes)	Chronic	Enamel fluorosis of the front permanent incisors.
1.5 mg/l or above (through water)	Daily intake	Produces insulin resistance.

4.2 Caustic effects of Fluorine and Hydrogen Fluoride

Gaseous fluorides may cause considerable damage to the skin and respiratory tract. Permissible occupational levels in the USA for hydrogen fluoride and fluorine are 2.5 mg/m³ and 2.0 mg/m³, respectively (ACGIH, 1983-84), (IPCS et al., 1984).

Continued inhalation of hydrogen fluoride or fluorine at high levels results in coughing, choking, and chills, lasting 1-2 h after exposure; in the next one or two days, fever, coughing, chest tightness, rales, and cyanosis may develop, indicating delayed pulmonary oedema (Dreisbach, 1971), (IPCS et al., 1984).

4.3 Chronic toxicity

Elevated intake of fluoride over prolonged periods of time may result in skeletal fluorosis, i.e., an accumulation of fluoride in the skeletal tissues associated with pathological bone formation (IPCS et al. 1984)



Figure-12: Skeletal fluorosis (Source: The Telegraph, 2019, Nov 7. UK)

4.4 Endemic skeletal Fluorosis

On the basis of more recent balance studies on patients with endemic fluorosis, which showed an average daily fluoride intake of 9.88 mg, Jolly (1976) suggested that a daily intake exceeding 8 mg in adults would be harmful. In tropical areas with endemic fluorosis, high fluoride levels in the drinking-water seem to constitute an important factor in a multi factorial causation (Reddy, 1979), (IPCS et al., 1984). In non-tropical countries, no cases of skeletal fluorosis with clinical

signs and symptoms have been detected in relation to drinking-water containing fluoride levels of less than 4 mg/l (Victoria Committee, 1980), (IPCS et al., 1984). The earliest reports of skeletal fluorosis in developed countries came from industries where exposure of workers to an intake of 40-80 mg per day for periods exceeding 4 years resulted in severe skeletal changes. Such occupational fluorosis has been reported from industries with old or outmoded control technology. Simultaneously with the reports from industry, skeletal fluorosis was diagnosed in several areas where there was excessive fluoride in soil, water, dust, or vegetable matter (IPCS et al., 1984).



Figure-13: Skeletal fluorosis of types genuvalgum, genu varum, flexion, and rotational

Where industrial exposure is concerned, variability of occupational exposure and the difficulty of assessing the amount of fluoride absorbed and retained, has made it difficult to establish satisfactory dose-response relationships.

In addition to monitoring air concentrations, urinary fluoride concentrations are used as a means of indicating individual exposure. Fluorosis is unlikely to develop when preshift urine fluoride concentrations are consistently below about 4 mg/l. Endemic fluorosis involving severe debilitation of a substantial proportion of the population remains a serious problem in areas of several developing countries. It is difficult to define the exposure that results in these effects.

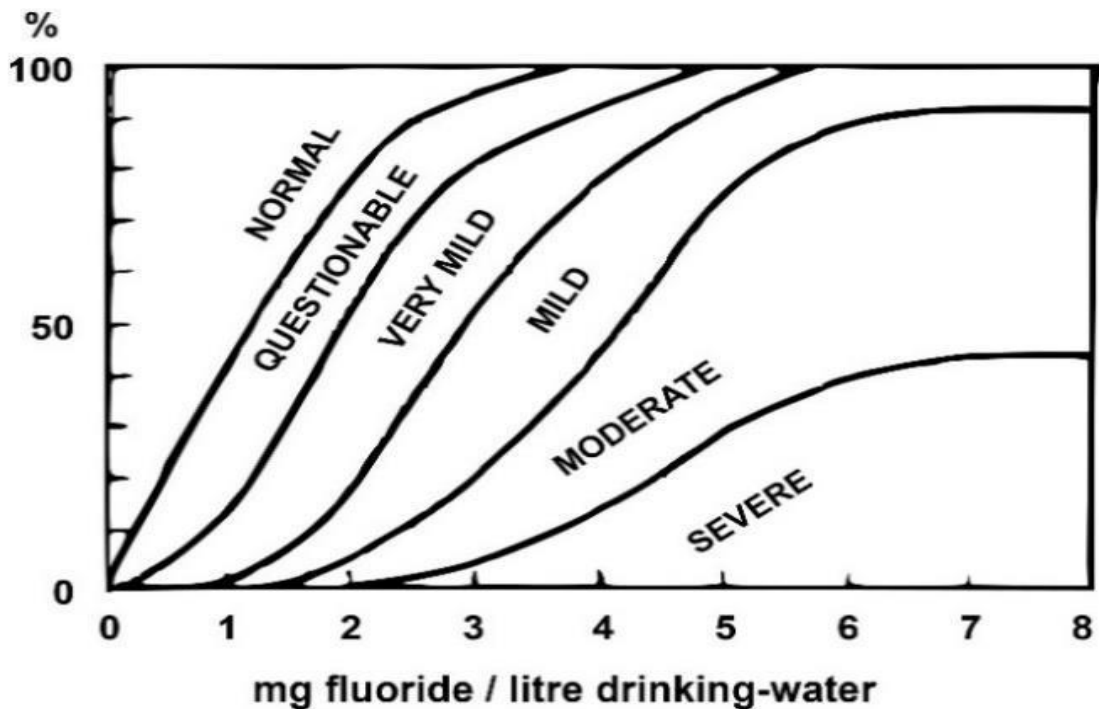


Figure-14: Distribution of dental fluorosis at different levels of fluoride in the drinking-water(Dean,1942)

4.5 Dental fluorosis

Dental fluorosis is hypo-mineralization of tooth enamel caused by intake of too much fluoride during enamel formation. It appears as a range of visual changes in enamel resulting degrees of intrinsic tooth discoloration. The severity of the condition is dependent on the dose, duration and age of the individual. In the mildest form (most common form) there are faint white lines or specks. Slightly more severe cases appear as white mottled patches, while severe fluorosis is characterized by brown discoloration and brittle, pitted and rough enamel (Sananda et al., 2015).

Excessive fluoride exposure during the period of tooth development may result in defective tooth formation. The earliest changes may resemble or be identical to abnormalities caused by other factors and this makes differential diagnosis difficult. The changes are rarely considered aesthetically objectionable. Depending on the fluoride intake from other sources, and the amount of drinking-water consumed, even these early changes occur in only a small proportion of a population that is using optimal levels of water fluoridation (IPCS et al., 1984). However, with increasing fluoride exposure, dental fluorosis becomes more prevalent and severe and may pose a public health problem (IPCS et al., 1984).



Figure -15: Categories of enamel fluorosis according to Dean's index(Source: Browne,D.,2012)

4.6 Effects on kidneys

No cases of renal signs or symptoms are mentioned in connection with prolonged intake of fluoride in the treatment of osteoporosis and otospongiosis (Causse et al, 1980; Schamschula, 1981; Dixon, 1983), although a thorough examination of kidney function may not have been carried out. No indications of increased frequency of kidney diseases or disturbed kidney functions have been recognized in areas with water fluoride concentrations of 8 mg/l (Leone et al., 1954, 1955), 2.0-5.6 mg/l (McClure, 1946; Geever et al., 1958) and 1.0 mg/l (Summens&Keitzer, 1975), (IPCS et al., 1984).

Although there are no reports of fluoride-induced chronic renal disorders in healthy individuals, several studies have dealt with the possible influence of fluoride on people with manifest kidney diseases. In patients with kidney failure, fluoride excretion is decreased, and the ionic plasma fluoride concentration is higher than the normal (Juncos &Donadio, 1972; Berman &Taves, 1973; Hanhijärvi, 1974). The capacity of the skeleton to store fluoride may provide a sufficient safety margin (Hodge & Smith, 1954; Hodge &Taves, 1970). On the other hand, it seems also plausible that an increased plasma fluoride concentration may result from fluoride liberation from the bone resorption processes involved in certain kidney diseases. Patients with diabetes insipidus may absorb excess amounts of fluoride because of the large quantities of fluids ingested (IPCS et al., 1984). Adverse effects like increased hepatic cell size, nephrosis, myocardial mineralization and degeneration of the semi niferous tubules in testis have been reported (IPCS, 2002) by WHO. The majority of fluoride is excreted via the kidneys (USNRC, 1993) therefore it is logical that those with impaired renal function might be at greater risk of fluoride toxicity (Table-6) (Sananda et al., 2015). Exposures to high levels of fluoride occur in connection with the use of fluorine-containing aesthetic agents, in particular methoxy flurane. These exposures have given rise to water-losing nephritis. A number of other toxic effects and specific health problems have been suggested and studied during recent years (IPCS et al., 1984).

4.7 Carcinogenicity

Excess cancer rates have been documented in various occupational groups exposed to fluorides. Thus, fluorspar miners (de Villiers &Windish, 1964) and aluminium production workers (Gibbs & Horowitz, 1979; Milham, 1979; Andersen et al., 1982) have been subject to lung cancer more frequently than expected (IPCS et al., 1984). Cancer mortality rates in areas with different amounts of fluoride naturally present in the drinking water have been compared in a considerable number of epidemiological studies. These studies have been carefully reviewed and evaluated by IARC (1982) with the following conclusions: “When proper account was taken of the differences among population units in demographic composition, and in some cases also in their degree of industrialization and other social factors, none of the studies provided any evidence that an increased level of fluoride in water was associated with an increase in cancer mortality.” Thus, “variations geographically and in time in the fluoride content of water supplies provide no evidence of an association between fluoride ingestion and mortality from cancer in humans” (IPCS et al., 1984)

4.8 Teratogenicity

The results of a recent study suggest that fluoride may indeed exert effects on fetal growth: babies, whose mothers had received fluoride tablets during pregnancy, were somewhat heavier and slightly longer at birth, and prematurity was much less frequent, compared with control groups (Glenn et al., 1982), (IPCS et al., 1984). Yet, no considerable evidence has been presented indicating that fluoride exposure does represent any teratogenic hazard (IPCS et al., 1984).

4.9 Effects on mortality patterns

Limited evidence is available concerning the possible effects of occupational fluoride exposures on mortality patterns (IPCS et al., 1984).

4.10 Allergy, hypersensitivity, and dermatological reactions

In 1971, the American Academy of Allergy examined the literature on alleged allergic reactions to fluoride: (Feltman, 1956; Feitman&Kosel, 1961; Burgstahler, 1965; Waldbott, 1965; Shea et al., 1967). The conclusions of the Executive Committee were (Austen et al., 1971): "There is no evidence of allergy or intolerance to fluorides as used in the fluoridation of community water supplies" (IPCS et al., 1984).

Since 1971, only in a few reports in the allergy literature have allergic reactions been suspected to be connected with fluoride exposure (IPCS et al., 1984).

In some occupational environments, aluminium pot room workers frequently complain about dyspnoea, chest tightness, and wheezing. The asthmatic response could be potentiated by beta—blockade with propranolol, and abolished by atropine (Saric et al., 1979). Increased bronchialexcitability, as shown by the metacholine inhalation test, can be induced by aluminium compounds including aluminium fluoride (Simonsson et al., 1977). These studies therefore suggest that respiratory exposure to irritants in the potroom atmosphere, including fluorides, may cause a non—specific hypersensitivity reaction that resembles bronchial asthma (IPCS et al., 1984).

Skin telangiectases were found in an increased number on the upper chest, back, and shoulders in 401 of aluminium reduction workers in a comprehensive, cross—sectional study (Theriault et al., 1980). These skin changes were not related to any excess of associated diseases, but the occurrence of large telangiectases was closely related to the length of exposure, and almost all workers with high exposures for more than ten years had telangiectases. However, the role of fluorides alone cannot be evaluated (IPCS et al., 1984)

4.11 Biochemical effects

Studies in human beings have shown minimal increases in urinary cyclic adenosine monophosphate excretion and unchanged plasma levels following an oral intake of about 7 mg fluoride, which resulted in peak plasma fluoride levels of about 0.3 mg/l (Mörnstad& van Dijken, 1982), (IPCS et al., 1984).

In the mineralization of bones and teeth, the proteoglycans and their constituent glycosaminoglycans may play an important role, and they form an integral part of the organic matrix of these tissues. Fluoride-induced changes in the formation of these compounds could be part of a common mechanism for the skeletal and dental effects of fluoride (IPCS et al., 1984).

CHAPTER-5

STUDY AREA

Chapter 5

5.0 Study Area

The study area lies in Bankura district of West Bengal, within the co-ordinates of latitude 23°25'37.3356' 'N and longitude 87°17'13.2648". Barjora is a Town and Subdivision in Bankura District of West Bengal. According to census 2011 information the sub-district code of Barjora Block (CD) is 02363. Total area of barjora subdivision is 388 km² including 375.13 km² rural area and 12.41 km² urban area. Barjora subdivision has a population of 2,02,049 peoples, out of which urban population is 25,786 while rural population is 1,76,263. Barjora has a population density of 521.4 inhabitants per square kilometre. There are about 44,421 houses in the sub-district, including 5,757 urban houses and 38,664 rural houses.

When it comes to literacy, 63.57% population of barjora subdivision is literate, out of which 71.84% males and 54.84% females are literate. There are about 196 villages in barjora subdivision.

5.1 Area overview: The map alongside shows the Bankura Sadar subdivision of Bankura district. Physiographically, this area is part of the Bankura Uplands in the west gradually merging with the Bankura-Bishnupur Rarh Plains in the north-east. The western portions are characterised by undulating terrain with many hills and ridges. The area is having a gradual descent from the Chota Nagpur Plateau. The soil is laterite red and hard beds are covered with scrub jungle and sal wood. Gradually it gives way to just uneven rolling lands but the soil continues to be lateritic. There are coal mines in the northern part, along the Damodar River. It is a predominantly rural area with 89% of the population living in rural areas and only 11% living in the urban areas.



Fig16:- A clear map of Barjora

5.2 Demographics: According to the 2011 Census of India, Barjora had a total population of 14,012 of which 7,203 (51%) were males and 6,809 (49%) were females. Population below 6 years was 1,269. The total number of literates in Barjora was 10,535 (82.67% of the population over 6 years).

As of 2001 India census, Barjora had a population of 11,509. Males constitute 52% of the population and females 48%. Barjora has an average literacy rate of 73%, higher than the national average of

59.5%; 57% of the literates are males and 43% are females. 9% of the population is under 6 years of age.

5.3 Infrastructure: According to the District Census Handbook 2011, Bankura, Barjora covered an area of 7.25 km². Among the civic amenities, the protected water supply involved tap water from treated sources, covered wells. It had 2,840 domestic electric connections, 406 road lighting points. Among the medical facilities it had 1 hospital, 1 dispensary/ health centre, 1 veterinary hospital. Among the educational facilities it had were 5 primary schools, 2 middle schools, 2 secondary schools, 2 senior secondary schools, 1 general degree college. It had 1 recognised shorthand, typewriting and vocational training institution, 2 non-formal education centres (SarvaShikshaAbhiyan), 1 special school for disabled. Among the social cultural and recreational facilities, it had 1 auditorium/ community hall, 1 public library, 1 reading room. It had the branch offices of 2 nationalised banks and 1 cooperative bank.

5.4 Transport: State Highway 9 connects Durgapur with Bankura via Barjora. The Barjora – Maliara-Durlavpur Road originates from 36 km point of the State Highway 9 (Bankura-Durgapur) and terminates on the National Highway 14, which is adjacent to Mejia Thermal Power Station.

5.5 Education: Barjora College was established in 1985 with the active support of Aswini Kumar Raj, Lalbehari Bhattacharya, Radhakanta Mondal, Manik Chandra Mukherjee and others. It is affiliated to the Bankura University and offers honours courses in English, Bengali, Sanskrit, history, political science, philosophy, economics and geography.

Barjora High School is a Bengali-medium coeducational institution established in 1962. It has facilities for teaching from class V to class XII. The school has 15 computers and a library with 2,300 books.

Barjora Girls High School is a Bengali-medium girls only institution established in 1973. It has facilities for teaching from class V to class XII. The school has 11 computers and a library with 2,000 books.

5.6 Healthcare: Barjora Rural Hospital, with 30 beds, is the major government medical facility in the Barjora CD block. There are primary health centres at Beliatore (with 10 beds), Chhandar (with 4 beds), Godardihi (Jagannathpur) (with 4 beds) and Pakhanna (with 10 beds).^{[16][17]}

The 300-bedded Sperscialty Hospital at Barjora was inaugurated in 2015.

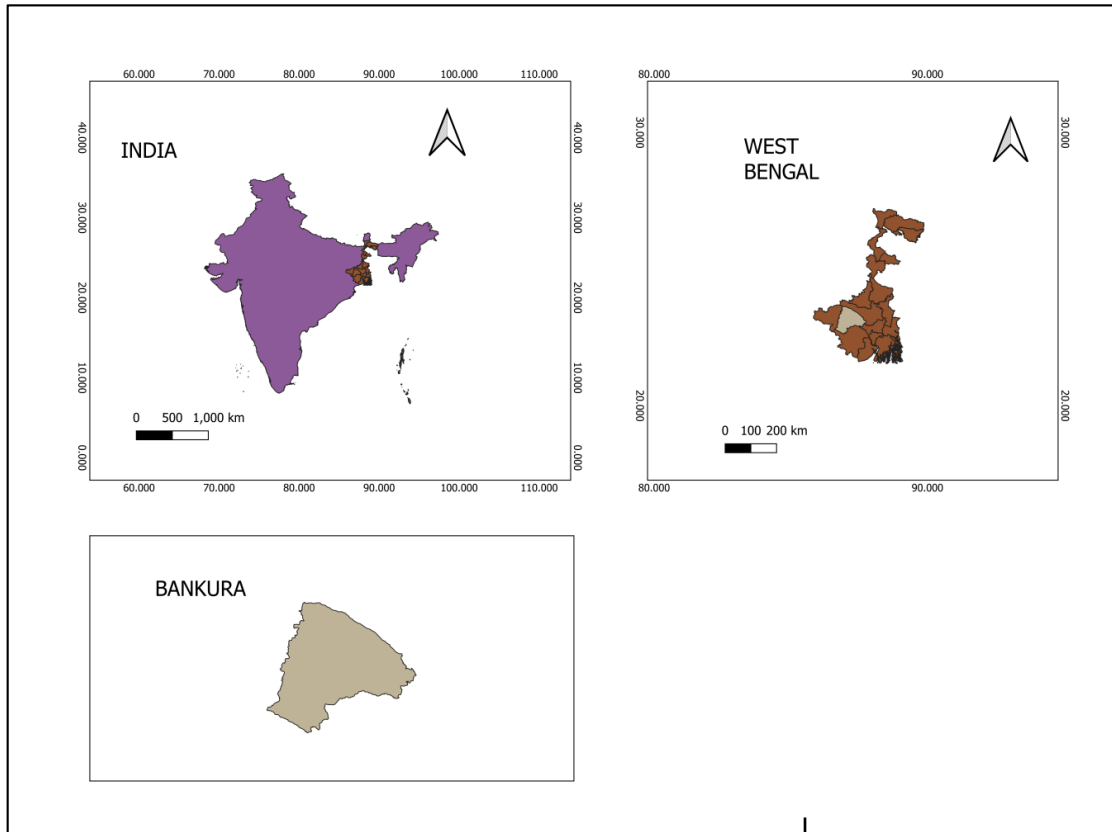


Fig-17: Location map of our Study Area

OUR STUDY AREA- BARJORA
(MARKED IN GREY COLOUR)

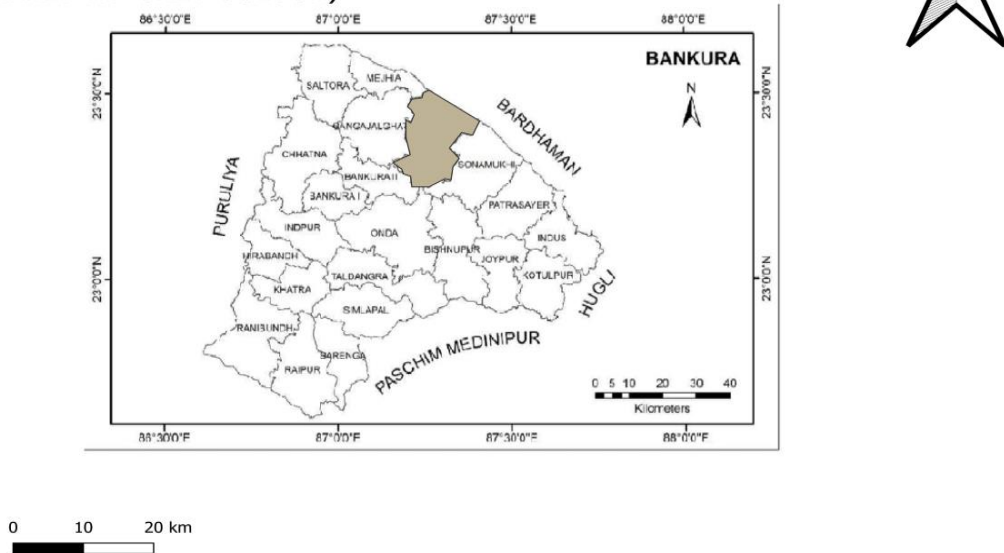


Fig-18: A map of Barjora

CHAPTER-6

METHODOLOGY

Chapter 6

6. Methodology:

In this chapter, methodologies to achieve various activities of the study objectives are discussed. The various activities carried out in the study is shown through the flow chart in Fig. 19

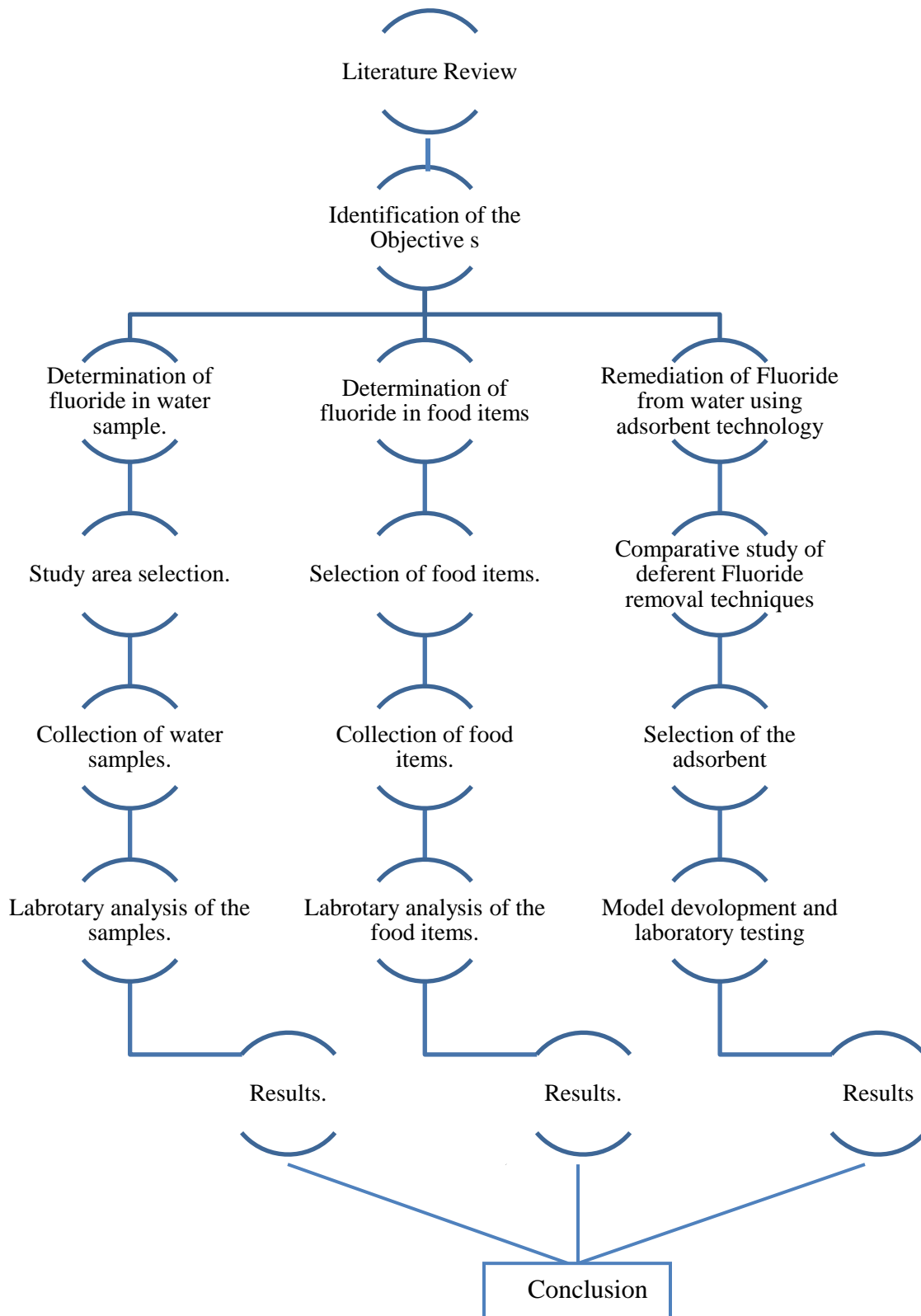


Fig-19: A brief methodology of flowchart

6.1. Determination of fluoride in water samples.

6.1.1. Selection of the study area: In Bankura there are some places which is fluoride affected. Among them we choose Barjora. Barjora is a locality in the Barjora CD block in the BankuraSadar subdivision of the Bankura district in the state of West Bengal, India.

6.1.2. Collection of the water sample: Wear gloves and eye protection when collecting samples. Rinse the bottle and cap three times with sample water and fill the bottle to within one to two inches from the top. Plastic bottles are used. Water is collected from tap well, deep tube well, well. We have collected water in 9 water sample bottles.

Table-6: Showing classification of various types of water collection method

Sl No.	Classification	Latitude	Longitude
1	Time Call	N 23 ⁰ 25'42.96"	E 87 ⁰ 17'0.564"
2	Hand Tubewell	N 23 ⁰ 25'31.374"	E 87 ⁰ 16'48.63"
3	Hand Tubewell	N 23 ⁰ 25'27.138"	E 87 ⁰ 16'40.488"
4	Well	N 23 ⁰ 25'23.49"	E 87 ⁰ 16'35.412"
5	Time Call	N 23 ⁰ 25'43.344"	E 87 ⁰ 17'2.142"
6	Time Call	N 23 ⁰ 25'43.248"	E 87 ⁰ 17'2.508"
7	Hand Tubewell	N 23 ⁰ 25'39.702"	E 87 ⁰ 17'10.02"
8	Time Call	N 23 ⁰ 25'28.686"	E 87 ⁰ 16'56.148"
9	Well	N 23 ⁰ 25'28.362"	E 87 ⁰ 16'56.286"



Fig-20: Collecting water samples from hand tube well, time well

6.1.3. Laboratory Analysis:

- Start Program **190 Fluoride**.
- Use a pipet to add 10.0 mL of deionized water to a sample cell.
- Use a pipet to add 10.0 mL of sample to a sample cell.
- Use a pipet to add 2.0 mL of SPANDS Reagent Solution into each sample cell
- Swirl to mix.
- Start the instrument timer. A 1-minute reaction time starts.
- When the timer expires, clean the blank sample cell then insert the blank into the cell holder.
- Now we push zero. The display shows 0.00 mg/L F⁻.
- Clean the prepared sample cell
- Insert the prepared sample into the cell holder.
- We push **READ** button. Results show in mg/L F⁻.



Fig-21: Images of pipet and 10 ml sample tube which are used in laboratory

6.1.4. Result: The permissible limit of fluoride is 1.5 mg/L. Maximum result gives the value around (0.30-1.17) mg/L. Only one value (2.85 mg/L) exceeds permissible limit.

6.2. Determination of fluoride in food samples.

6.2.1. Selection of food items: We know that fluoride is present in water. But not only water, we can get fluoride by intake of Tea, Hazmola, Kalonamak, HazmiGuli etc. So here we select food items like Tea, HazmiGuli, Kalonamak, Hazmola.

6.2.2. Collection of food items: 7-8 types of tea samples are collected from a stationary shop. Tea samples are of various type just like Assam tea, Darjeeling Tea, Red Label tea, Tata Gold tea. We also buy HazmiGuli, Kalonamak, Hazmola



Fig-22: Food items such like Hazmi Goli, Black Salt, various Tea samples

6.2.3. Laboratory Analysis of the food items:

- Start Program **190 Fluoride**.
- Use a pipet to add 10.0 mL of deionized water to a sample cell.
- Use a pipet to add 10.0 mL of sample to a sample cell.
- Use a pipet to add 2.0 mL of SPANDS Reagent Solution into each sample cell
- Swirl to mix.
- Start the instrument timer. A 1-minute reaction time starts.
- When the timer expires, clean the blank sample cell then insert the blank into the cell holder.
- Now we push zero. The display shows 0.00 mg/L F⁻.
- Clean the prepared sample cell
- Insert the prepared sample into the cell holder.
- We push **READ** button, and Results show in mg/L F⁻.



Fig-23: Weighing of tea sample and preparation of it

6.2.4. Result: We got the result. The result has come out in mg per gm. Means it give us an idea about how much fluoride we can intake after taking how much amount of food items we intake.

6.3. Remediation of Fluoride from water using adsorbent technology

6.3.1. Comparative study of deferent Fluoride removal techniques: Here first we have done a comparative study of effectiveness of fluoride remove. There are lots of process, among them which is suitable in our work.

6.3.2. Selection of the process: We have found that among all the process adsorption is the most useful method in our work. So, we choose adsorption process and, in this process, fluoride from water.

6.3.3. Model development: And finally, we develop the filter. We use sand, gravel, and activated alumina as the media. The height of the filter is 53 cm. Bottom of the filter is gravel, and it is up to 5 cm and next is followed by coarse sand. It is up to 10 cm. and media activated alumina is poured into 20 cm.

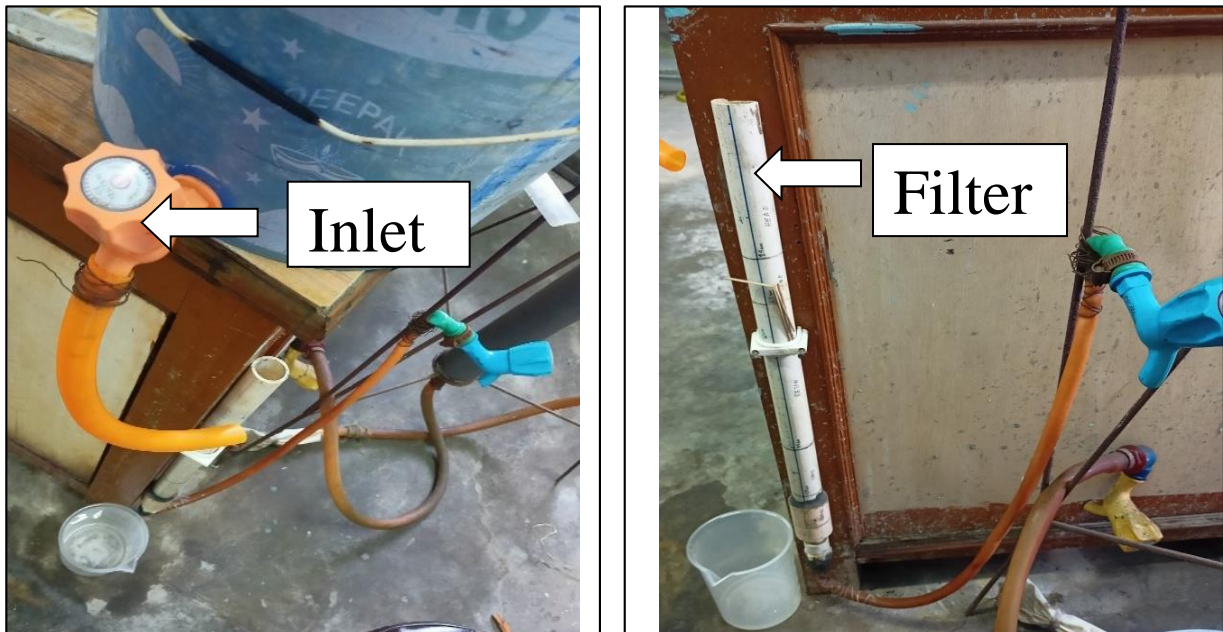


Fig-24: Images of Inlet and filter

6.3.4. Laboratory testing and result:

- We take the filter water out.
- And we check the result hour by hour.
- And we get our desired result. Start Program **190 Fluoride**.
- Use a pipet to add 10.0 mL of deionized water to a sample cell.
- Use a pipet to add 10.0 mL of sample to a sample cell.
- Use a pipet to add 2.0 mL of SPANDS Reagent Solution into each sample cell
- Swirl to mix.
- Start the instrument timer. A 1-minute reaction time starts.
- When the timer expires, clean the blank sample cell then insert the blank into the cell holder.
- Now we push zero. The display shows 0.00 mg/L F⁻.
- Clean the prepared sample cell
- Insert the prepared sample into the cell holder.
- We push **READ** button. Results show in mg/L F⁻.

CHAPTER 7

MATERIAL & METHODS

Chapter-7

7. MATERIAL & METHODS:

7.1. Material used as adsorbent

Activated Alumina: Activated alumina is manufactured from aluminium hydroxide by dehydroxylating it in a way that produces a highly porous material; this material can have a surface area significantly over 200 m²/g. The compound is used as a desiccant (to keep things dry by adsorbing water from the air) and as a filter of fluoride, arsenic and selenium in drinking water. It is made of aluminium oxide (alumina; Al₂O₃). It has a very high surface-area-to-weight ratio, due to the many "tunnel like" pores that it has. Activated alumina in its phase composition can be represented only by metastable forms (gamma-Al₂O₃ etc.). Corundum (alpha-Al₂O₃), the only stable form of aluminum oxide, does not have such a chemically active surface and is not used as a sorbent.



Fig-25: Images of Activated Alumina

Activated Alumina as adsorbent: Activated alumina is also widely used to remove fluoride from drinking water. In the US, there are widespread programs to fluoridate drinking water. However, in certain regions, such as the Rajasthan region of India, there is enough fluoride in the water to cause fluorosis. A study from the Harvard school of Public Health found exposure to high levels of fluoride as a child correlated with lower IQ.

Activated alumina filters can easily reduce fluoride levels from 10 ppm to less than 1 ppm. The amount of fluoride leached from the water being filtered depends on how long the water is actually touching the alumina filter media. Basically, the more alumina in the filter, the less fluoride will be in the final, filtered water. Lower temperature water, and lower pH water (acidic water) are filtered more effectively too. Ideal pH for treatment is 5.5, which allows for up to a 95% removal rate.

As per researches conducted by V.K.Chhabra, Chief Chemist (retd.) P.H.E.D. Rajasthan, activated alumina, when used as a fluoride filter, under field conditions can best be regenerated by a solution of lye (sodium hydroxide; NaOH), sulfuric acid (H₂SO₄).

7.2. Method:

7.2.1. Preparation of the filter

- Analytical techniques a method to determine a chemical or physical property of a chemical substance, chemical element, or mixture. There are a wide variety of techniques used for analysis, from simple weighing to advanced techniques using highly specialized instrumentation.
- Here first we arrange a bucket. The bucket consists of 12 litres of water. We prepare stock solution and add to it.
- The bucket is attached with a tap opening. And in the mouth of tap there is a pipe, which is connected to the filter.
- Then we adjust the filtration rate. The dia of the filter is 2.6 cm. We get the area of the filter as 0.00053 m². We adjust the filtration rate, we can see that in 4 min 16 ml of water is coming to the filter, and we set the filtration rate as 450 lt/hr/m².

- So, the water is coming into the filter. The filter is 53 cm. at the bottom most part of the filter is gravel of 5 cm, coarse sand is followed by gravel, it is up to 10 cm. Our media is activated alumina, it is up to 20 cm.
- The filter is also connected with a tap that is called outlet. We can open the tap and collect the water, we collect the water and do the test hour by hour.

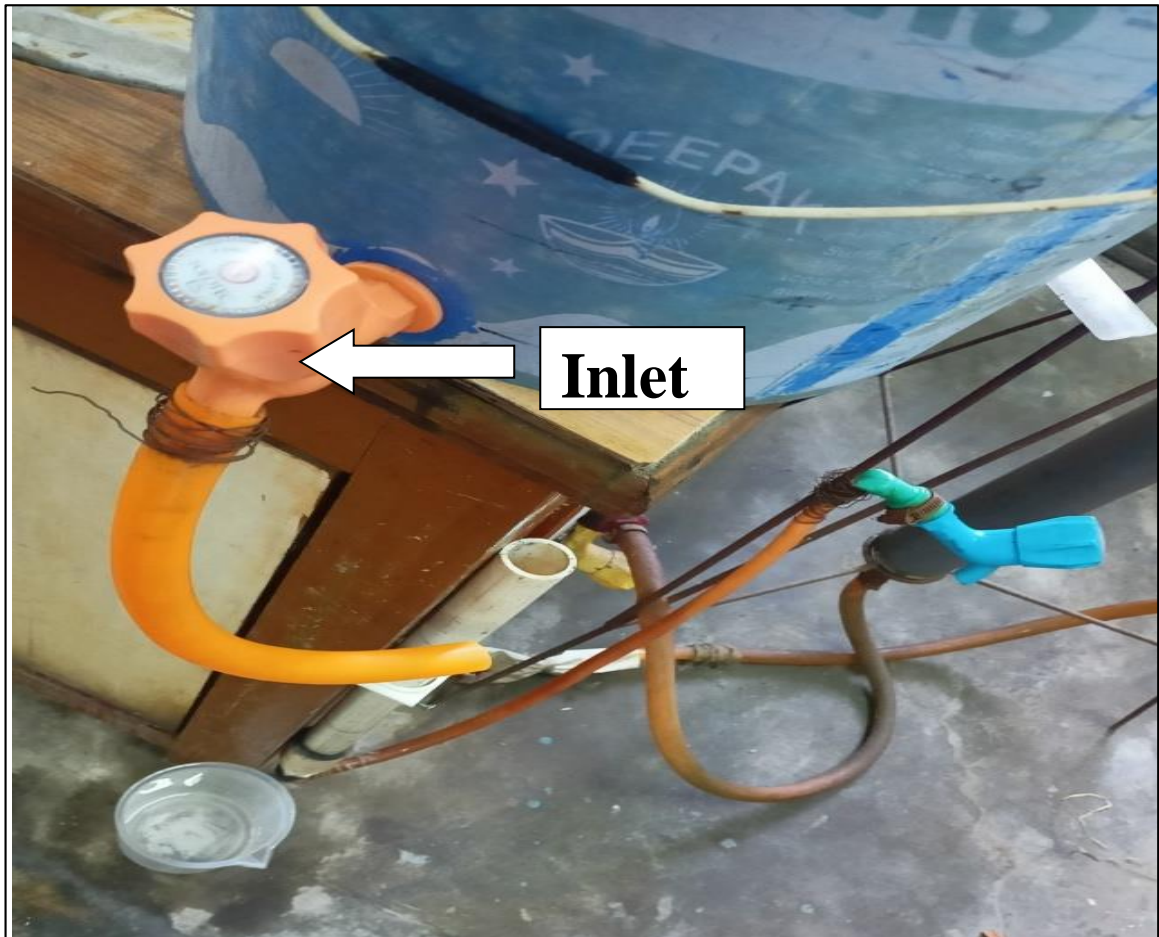


Fig-26: A detailed picture of Inlet

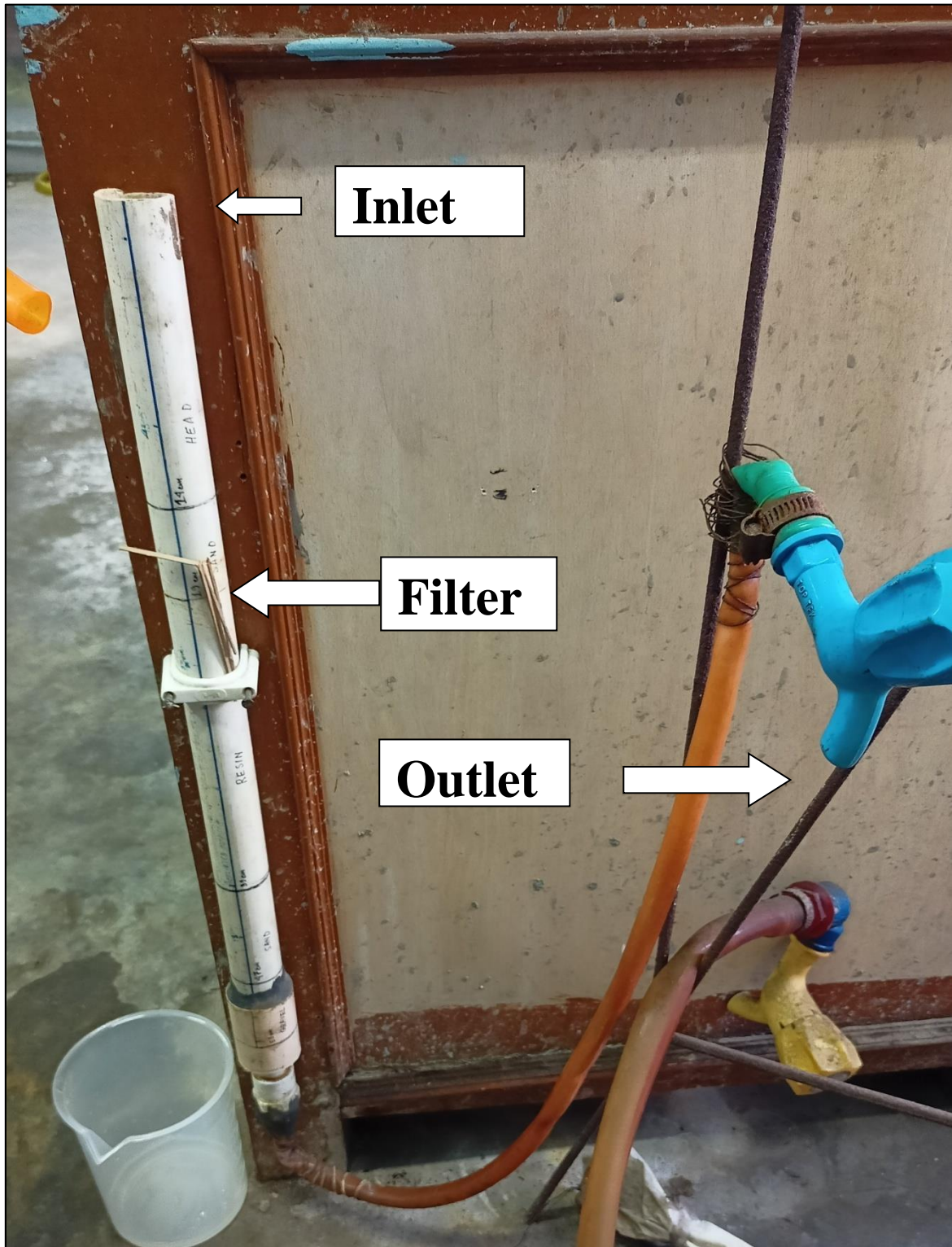


Fig-27: A detailed picture of Filter

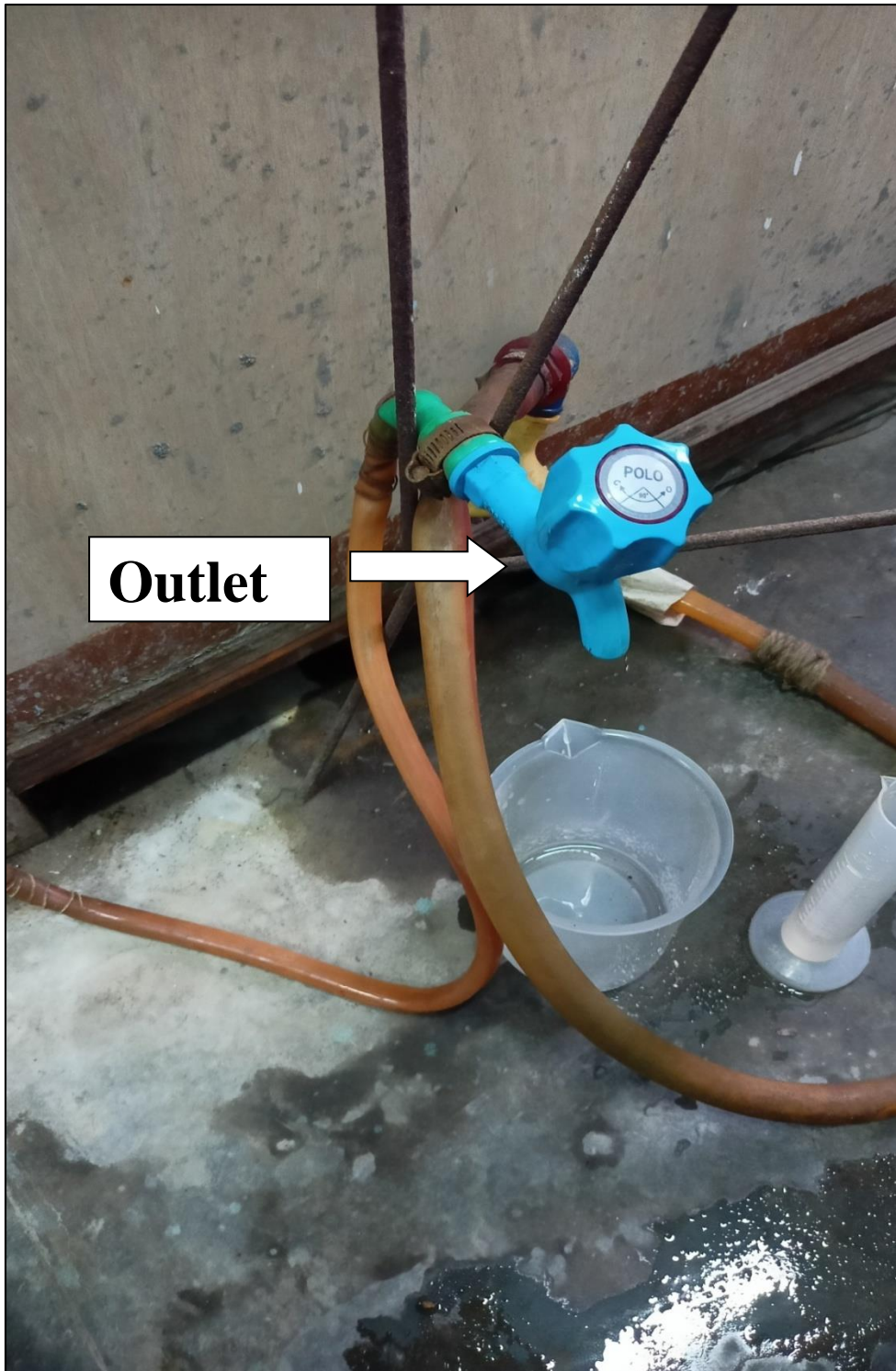


Fig-28: A detailed picture of Outlet

7.2.2. Determination of Fluoride removal efficiency of the filter:

$$\eta\% = \frac{X_o - X_I}{X_o} \times 100 \quad (1)$$

Where X_o refers as the output value and X_I refers as the input value. And the efficiency is expressed as percentage value.

CHAPTER-8

RESULT & DISCUSSION

8. Result & Discussion: We have done three types of fluoride test. One test is done with sample water which is collected from Barjora. Second test is about fluoride in different food items. Third is a model development and fluoride removal efficiency of the filter.

8.1 Water Sample: Water sample is collected from Barjora. Wear gloves and eye protection when collecting samples. Rinse the bottle and cap three times with sample water and fill the bottle to within one to two inches from the top. Plastic bottles are used. Water is collected from tap well, deep tube well, well. We have collected water in nine water sample bottles.

Table-7: Showing result of fluoride in water sample

Sl no	Classification	Latitude	Longitude	Fluoride Present (mg/L)
1	Time Call	N 23 ⁰ 25'42.96"	E 87 ⁰ 17'0.564"	0.75
2	Hand Tubewell (30-32 ft)	N 23 ⁰ 25'31.374"	E 87 ⁰ 16'48.63"	1.17
3	Hand Tubewell	N 23 ⁰ 25'27.138"	E 87 ⁰ 16'40.488"	0.84
4	Well (42 ft)	N 23 ⁰ 25'23.49"	E 87 ⁰ 16'35.412"	0.91
5	Time Call	N 23 ⁰ 25'43.344"	E 87 ⁰ 17'2.142"	0.58
6	Time Call	N 23 ⁰ 25'43.248"	E 87 ⁰ 17'2.508"	0.66
7	Hand Tubewell	N 23 ⁰ 25'39.702"	E 87 ⁰ 17'10.02"	0.17
8	Time Call	N 23 ⁰ 25'28.686"	E 87 ⁰ 16'56.148"	2.85
9	Well	N 23 ⁰ 25'28.362"	E 87 ⁰ 16'56.286"	0.30

We all know that in general permissible limit of fluoride is 1.5 mg/L. So in maximum cases the result is safe, but one result has come out as 2.85, which is more than its permissible limit. So we should avoid that source.

8.2 Food Sample:-We know that fluoride is present in water. But not only water, we can get fluoride by intake of Tea, Hazmola, Black Salt, Hazmi Guli etc. So here we select food items like Tea, Hazmi Guli, Kalonamak, Hazmola.

7-8 types of tea samples are collected from a stationary shop. Tea samples are of various type just like Assam tea, Darjeeling Tea, Red Label tea, Tata Gold tea. We also buy Hazmi Guli, Kalonamak, Hazmola.

Table-8: Table showing fluoride content in food item

Sample	Amount Taken (gm)	Fluoride (mg/L)	Fluoride (in 100ml) (mg) in 5 g	Fluoride/gm (mg)	Fluoride/kg (mg)
Assam Tea (Rs 200/Kg)	5	20.8	2.08	0.416	416
Assam Tea (Rs 300/Kg)	5	18.1	1.81	0.362	362
Assam Tea (Rs 500/kg)	5	17.4	1.74	0.348	348

Darjeeling tea (Rs 600/kg)	5	21.8	2.18	0.436	436
Darjeeling tea (Rs 800/- per kg)	5	17.6	1.76	0.352	352
Red Label Tea (Rs 10/-)	5	22.9	2.29	0.458	458
Tata Gold (Rs 10/-)	5	22.1	2.21	0.442	442
Black salt	5	2.15	0.215	0.043	43
Hazmola	5	5.70	0.57	0.114	114
Hazmiguli	5	8.85	0.885	0.177	177

Assam Tea which is 200 Rs/kg, contains 0.416 mg of fluoride per gm. Another Assam tea which is 300 Rs/kg, contains 0.362 mg of fluoride; for Assam tea of Rs 500 Rs/kg we intake 0.348 mg of fluoride, Darjeeling tea which is Rs 600/kg contains 0.436 mg of fluoride, 1 gm of red label tea contains 0.458 mg of fluoride, 1 gm of Tata Gold tea contains 0.458 mg of fluoride, 1 gm of black salt contains 0.043 mg of fluoride, 1 gm of Hazmola contains 0.114 mg of fluoride, 1 gm of Hazmiguli contains 0.177 mg of fluoride.

So we are aware about permissible limit of fluoride. One example may be given as such that, 1000gm of hazmola contain 114 mg of fluoride. So we can't take 1000 gm of hazmola in a day which is also a quite impossible thing to intake. So we shall be aware about our food consumption.

8.3 Fluoride removal efficiency of filter:-Here first we arrange a bucket. The bucket consists of 12 litres of water. We prepare stock solution and add to it.

- The bucket is attached with a tap opening. And in the mouth of tap there is a pipe, which is connected to the filter.
- Then we adjust the filtration rate. The dia of the filter is 2.6 cm. We get the area of the filter as 0.00053 m^2 . We adjust the filtration rate, we can see that in 4 min 16 ml of water is coming to the filter, and we set the filtration rate as 450 lt/hr/m^2 .
- So, the water is coming into the filter. The filter is 53 cm. at the bottom most part of the filter is gravel of 5 cm, coarse sand is followed by gravel, it is up to 10 cm. Our media is activated alumina, it is up to 20 cm.
- The filter is also connected with a tap that is called outlet. We can open the tap and collect the water, we collect the water and do the test hour by hour.

Table-9: Showing fluoride removal efficiency

Sl no	Water Consumption	cumulative consumption of water	Fluoride Influent	Fluoride Effluent	Fluoride Removal
1	1	1	6.5	0	100
2	1.5	2.5	6.5	0	100
3	1.5	4	6.5	0	100
4	1.5	5.5	6.5	0	100
5	1.5	7	6.5	0.018	99.72
6	1	8	6.5	0.027	99.58
7	1.5	9.5	6.5	0.043	99.34
8	1	10.5	6.5	0.098	98.49
9	1.5	12	6.5	0.156	97.6
10	1.5	13.5	6.5	0.472	92.74
11	1.5	15	6.5	0.851	86.9
12	1	16	6.5	1.097	83.1
13	1.5	17.5	6.5	1.296	80.06
14	1.5	19	6.5	1.535	76.38

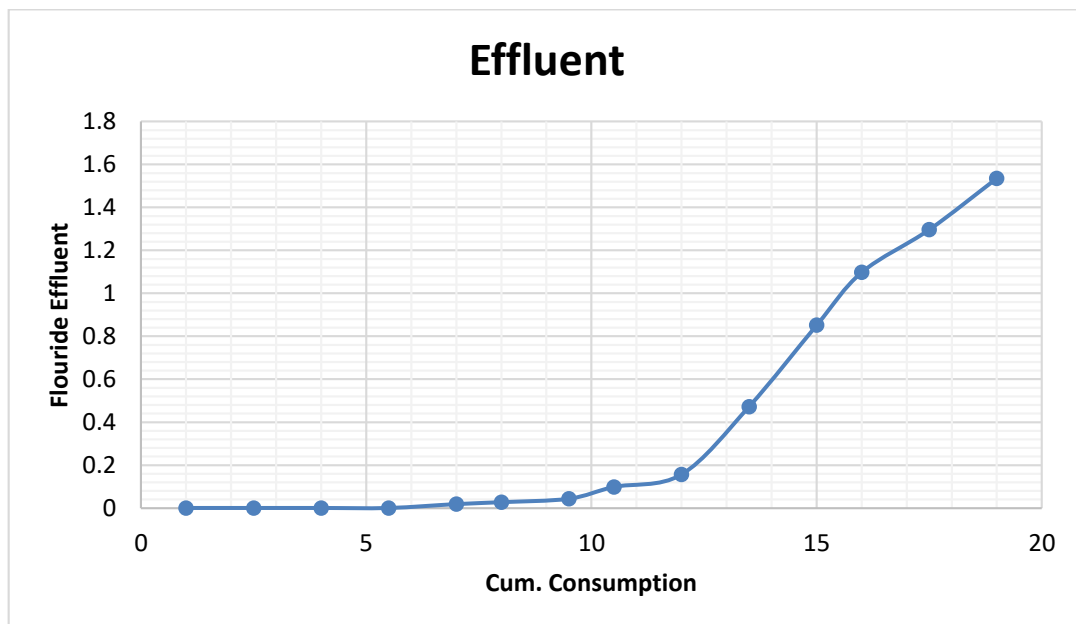


Fig-29: figure showing a graph between fluoride effluent and cumulative consumption

In this graph we can notice the effluent fluoride is zero at first. That means when we just run the filter in that particular time no effluent fluoride is recorded. But when we run the filter for some time effluent fluoride is noted. No effluent fluoride till five to six liter of water. But when the filter is continuing

running its process effluent fluoride is recorded. We can see from the graph that at near about twenty liter of water, fluoride crosses its permissible limit. Up to fifteen liter of water it shows 0.851 mg/L. which is below desirable limit. But in the sixteen liter it crosses 1 mg/L. It is not desirable limit.

If we have to know the removal efficiency, we know that (input-output)/input gives the removal efficiency.

Removal Efficiency is expressed as
$$\eta\% = \frac{X_o - X_I}{X_o} \times 100 \quad (1)$$

$$\eta\% = \frac{6.5 - 1.535}{6.5} \times 100 = 76.138\%$$

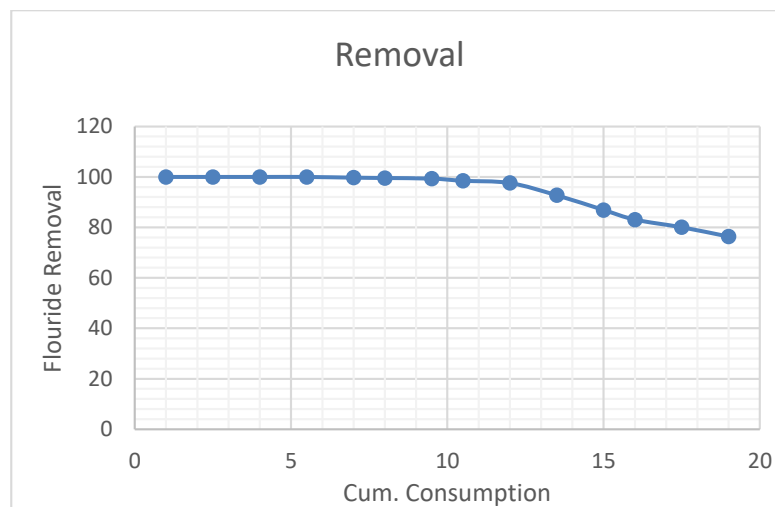


Fig-30: figure showing a graph between fluoride removal and cumulative consumption of water

From here we can say that our influent fluoride is 6.5 mg/L. And our effluent capacity is 1.535 mg/L. So if we come to know that how much fluoride has been removed we have to subtract influent fluoride from effluent fluoride. So we get the value as (6.5-1.535) = 4.965 mg/L. This value comes after when we run 19 litre of water in filter. We have taken 100 gm of Activated Alumina. And we have to know the removal capacity of Activated Alumina in mg/g. 100 gm of Activated Alumina is used in (4.965 mg/L × 19L) = 94.335 mg of fluoride removing. So we can get the value (94.335/100) = 0.943 mg/g activated alumina fluoride adsorption capacity.

Adsorption Capacity of Activated Alumina: Effluent fluoride = 6.5 mg/L
Influent fluoride = 1.535 mg/L

Amount of fluoride adsorb by Activated alumina is = (Effluent – Influent)
(6.5-1.535) = 4.965 mg/L

Amount of fluoride in mg adsorb by Activated Alumina = 4.965 mg/L * 19L
= 94.335 mg.

We have taken 100 gm of Activated Alumina.

Adsorption Capacity of Activated Alumina = 94.335/100
= 0.94335 mg/g.

So we can say that our adsorption capacity can be expressed as 943.33mg/Kg.

CHAPTER 9

CONCLUSION

Chapter-9:

Conclusion:-

The purpose of the present study was to determine the fluoride concentrations in tea as the potential dietary exposure to fluoride from tea and associated health risks. This study showed that Fluoride is not only available in tea. It is also available in different types of food samples. Among them most popular is tea. Consumption of fluoride in different types of tea samples very widely speeded. Presence of fluoride is also depends on the branding & costing of tea samples. Based on the fluoride levels in tea from study results suggest that the general population in the study area is at risk of excessive intake of fluoride and that fluoridation of drinking water further contributes to the risk of fluoride intoxication. The findings of this study also show that for the general population who consume tea daily, fluoride exposure can significantly exceed the levels that can cause thyroid dysfunction in persons with either low or adequate iodine intake. Fluoride can cause cardiovascular system dysfunctions in humans mainly presenting as arrhythmias. Decisions about water fluoridation should include the total amount of fluoride intake from all sources including foods, beverages and medications such that cumulative effects and the risk of chronic fluoride intoxication are reduced. Thus, the effect of fluoridation of drinking water has resulted in compounding safety factors associated with uncontrolled exposure and contributed to excessive accumulation of fluoride in the general population and needs sustainable remedial measures. In the present study efficacy of cost effective defluorination also evaluated. Among them adsorption is one of most used method. In this adsorption method, we can use lots of adsorbents. Among them Activated Alumina showed higher capacity for removal of adsorbent for fluoride ions. When fluoride concentration are high and if low alumina dosage are used. And in this paper we have discussed about cumulative water and fluoride removal efficiency. From all the experimental results and data, it can be concluded that the percentage of fluoride removal by adsorption process using activated alumina was found to be best at an efficiency rate of 76.13%. Thus, it can be concluded that the removal efficiency was maximum in case of activated alumina fluoride removal adsorption process followed by other conventional and non conventional techniques studied from literature.

Limitation :

The objective of this study was to determine the fluoride content in tea infusions using an established and recognized methodology. The most widely used method of fluoride quantification has involved potentiometry employing the fluoride ion selective electrode. It is noted that our results are similar to those reported in published literature However, the possible interference of aluminium (Al) with fluoride measurement using ISE procedures was not examined in present study.

Future Recommendation:

In future studies can emphasis on Al content. Which was not determined in all tea products. Further studies are warranted to determine the concentration and dietary intake of Al and other metal contaminants from tea and corresponding health impact. More advance instrument like Ion chromatograph (IC) can also very useful for estimate the trace concentrations in food products. Finally, detailed studies on fluoride concentration in food chain will also create a greater impact.

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