Removal of Fluoride from Drinking Water by Utilizing Modified Bagasse Sugarcane as Low-Cost Adsorbents for Bilaspur City, Chhattisgarh

D. Verma¹, J. Supe¹ *, S. Verma² *, and R. R. Singh¹

¹Department of Civil Engineering, Rungta College of Engineering and Technology, Bhilai 490023, India
²Department of Civil Engineering, MATS University Aarang, Raipur 493441, India

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ABSTRACT. Recently, there has been a noticeable increase in fluoride contamination in drinking water, leading to evident harmful effects in various locations. Tackling the challenge of reducing fluoride levels in potable water has become a significant concern for drinking water supply agencies. In our current study, we focused on creating and characterizing different adsorbents, namely sugarcane bagasse, coconut husk, and rice husk, for their ability to effectively remove fluoride. These adsorbents are not only cost-effective and readily available but also straightforward to prepare. We conducted thorough characterizations of all three adsorbents to assess their performance under various conditions. This included investigating the impact of pH on removal efficiency, evaluating removal efficiency with varying doses of adsorbent, analyzing removal efficiency across different temperatures, studying the influence of the initial concentration of fluoride on the total amount of adsorbent per unit mass, and examining the impact of altering initial fluoride concentrations while maintaining a constant temperature of 30 ± 2 °C. Our findings revealed noteworthy insights into the effects of these parameters on removal efficiency; fluoride contamination in the groundwater of Bilaspur, Chhattisgarh, originates not only from anthropogenic sources but also from geogenic sources. The efficiency of contamination reduction rises as the pH increases, reaching a plateau after a certain point. When employing an adsorbent dose of 0.5 g/100 mL of modified bagasse sugarcane (MBS) and a contact time of 75 minutes, along with a temperature of 323 K, the highest efficiency is observed in the MBC. Subsequently, the efficiency is lower in aluminum-hydroxide-coated rice husk and then in modified coconut husk. Analysis indicates that as the initial fluoride concentration increases, the removal efficiency diminishes due to the capacity limitations of the adsorbent materials.

Keywords: fluoride removal, low-cost adsorbents, Bilaspur city, Chhattisgarh, water treatment

1. Introduction

Increased levels of fluoride in drinking water have become a prominent worldwide issue due to their harmful impacts on human well-being. Dental and skeletal fluorosis, a condition characterized by the deterioration of teeth and bones, can result from the excessive consumption of fluoride, primarily through water sources (Zou and Ashley, 2014; Ahmad et al., 2022; Surati et al., 2022). In response to these health issues, the World Health Organization (WHO) has set a suggested maximum threshold of 1.5 mg/L for the fluoride content in drinking water. Nonetheless, numerous areas globally experience water sources that surpass this limit due to natural geological processes or human-induced actions, thus putting communities at risk of potential health problems (Connor, 2015; Tzanakakis et al., 2020). In addressing this concern, a range of strategies have been devised to mitigate elevated fluoride levels in water sources. Among these strategies, one particularly promising method involves the application of adsorption techniques (Rashed, 2013; Rathi and Kumar, 2021). These techniques utilize substances called adsorbents to attract and attach fluoride ions to water, consequently lowering their concentration to acceptable levels. The utilization of adsorption presents numerous benefits, including operational simplicity, affordability, and the possibility of employing readily accessible resources (Choksi et al., 2015; Crini et al., 2018; Aragaw and Bogale, 2021; Achour et al., 2022; Ngeno et al., 2022; Patel and Mehta, 2022; Perez-Botella et al., 2022; Patel et al., 2023; Umrigar et al., 2023). This investigation centers on the effective use of cost-effective materials as adsorbents for tackling fluoride contamination.

Approximately 85% of India’s population relies on groundwater for their drinking water needs, as indicated by Iyer et al. (2014) and Carrad et al. (2019). This makes the quality of water a crucial issue that requires dedicated attention (Yadav et al., 2015; Mehta et al., 2018; Yadav et al., 2018; Achour et al., 2022; Zolghadr et al., 2023). Although there was a prevailing belief that water in India was generally safe for consumption and was commonly used without comprehensive risk assessment, numerous studies have contradicted this notion. These studies have emphasized that water pollution is a significant problem, resulting from both natural factors and human activities (Shekhar and...
Fluoride is a member of the halogen group and holds the distinction of being the lightest and most electronegative element within the periodic table. According to Suxena and Ahmed (2001) and Ali et al. (2016), fluoride is notably abundant in the earth’s crust at approximately 625 mg/kg. In aqueous solutions, fluoride manifests as a negatively charged fluoride ion (F\(^-\)), as pointed out by Ali et al. (2016). The prevalence of elevated F\(^-\) levels in groundwater primarily stems from the existence of diverse fluoride-containing minerals such as fluorite, cryolite, topaz, apatite, amphiboles, micas, sellaite, villamite, and specific clays, as outlined in the works of Daneshpoooy et al. (2018) and Yousefi et al. (2018).

Several methods for removing fluoride, such as precipitation/coagulation, adsorption, reverse osmosis, ion exchange, and electro-dialysis, are available, but their operation typically requires skilled personnel and involves substantial capital and running expenses. This renders these techniques impractical for underdeveloped communities from both a technical and financial perspective (Grimm et al., 1998). For instance, in developing nations, methods like reverse osmosis and ion exchange are seldom used due to their significant upfront and operational expenses. Electro-dialysis, a process using an electric potential gradient to transfer fluoride ions through a semi-permeable membrane, also faces challenges due to its high initial costs and susceptibility to interference from other ions (Grimm et al., 1998). Hence, there is a necessity to explore alternative methods for removing fluoride that are both user-friendly and economically feasible for underprivileged communities (Demelash et al., 2019).

The elimination of fluoride from water using activated carbon primarily occurs through various mechanisms, predominantly adsorption. The effectiveness of the fluoride removal process relies significantly on the properties of the activated carbon (Saidi et al., 2019). Activated carbon obtained from agricultural by-products has proven to be relatively cost-effective, largely due to the abundance of these by-products from agricultural processing. Sugarcane bagasse, a residual product of sugarcane processing, is generated in significant quantities, often creating challenges in disposal. Therefore, utilizing bagasse for manufacturing activated carbon could aid in reducing the volume of bagasse requiring disposal (Hesas et al., 2013). This research assessed the application of activated carbon derived from sugarcane bagasse (SBAC) in the removal of fluoride from artificially created laboratory water and natural water containing high fluoride levels.

Low calcium levels and increased bicarbonate alkalinity are factors affecting elevated fluoride levels in groundwater (Bulusu and Pathak, 1980; USGS,1985). Low mineral content, high pH levels, and a notable presence of silica are typical indicators of groundwater with elevated fluoride levels. The inherent fluoride concentration in groundwater is subject to a multitude of factors, encompassing geological, chemical, and physical attributes of the aquifer, soil composition, rock permeability, temperature, chemical interactions, and the depth of the well. Due to the numerous variables involved, fluoride concentrations in groundwater can vary widely, ranging from less than 1.0 mg/L to over 35.0 mg/L (Ozsvath, 2009). The United States Public Health Service (USPHS) (U.S. Department of Health and Human Services Federal Panel on Community Water Fluoridation, 2015) has established specific permissible fluoride concentration ranges for drinking water in different communities, taking into account variations in air temperature, which in turn affect water consumption and fluoride intake. This information is presented in Table S1 which is found in the Supplementary Material.

The effective execution of the present study signifies a significant opportunity for addressing fluoride contamination in Bilaspur, Chhattisgarh. Through the utilization of affordable materials that are readily accessible locally, communities grappling with fluoride-related challenges can secure improved access to safe and purified drinking water. Furthermore, this study makes a valuable contribution to the wider realm of environmental remediation, highlighting the importance of tailored approaches to intricate water quality problems. The present study seeks to achieve its main objective by identifying, developing, and assessing affordable adsorbent materials that can efficiently eliminate fluoride from Bilaspur, Chhattisgarh. The study is designed to tackle the subsequent key aspects: (1) the development of an economical and efficient modified adsorbent using readily accessible resources like rice husk ash (RHA), sugarcane bagasse, and coconut husk; (2) investigation into the efficacy of the cost-effective adsorbent in eliminating fluoride.

In the following parts of this article, we explore the methodology, conduct experimental procedures, analyze results, and engage in discussions regarding the utilization of inexpensive materials for adsorption in fluoride removal.

1.1. Present Scenario of Fluoride Concentration

1.1.1. Global Scenario

The global geological characteristics of various regions have an impact on the distribution of elevated fluoride in water. Particularly in midlatitude areas, the earth’s crust contains a total of 85 million tonnes of fluoride deposits (Teotia and Teotia, 1994; Sahu et al., 2018). Regions such as Africa, China, the Middle East, and parts of southern Asia (like India and Sri Lanka) have significant occurrences of groundwater with high fluoride content. Fluoride belts, as identified by the WHO, encompass extensive regions ranging from Eritrea to Malawi, continuing through Syria, Turkey, Afghanistan, India, and China. These geographical areas exhibit comparable fluoride concentration patterns, and similar fluoride belts can also be found in places like the Americas, Kenya, Iraq, Japan, and Iran, as noted in the study by Sahu et al. (2018). Fluorosis symptoms become apparent in communities living in regions where the fluoride concentration is elevated.
content in groundwater ranges from 1.5 to 10 mg/L. However, recommends a fluoride concentration of 1.5 mg/L in drinking water, but its implementation varies due to factors like water consumption patterns, climate, and dietary habits (Mullen, 2005; Dharmshaktu, 2013). The severity of fluoride-related issues varies based on the environmental context of a geographical area. endemic fluoride-related issues in their drinking water or intake of water affect 200 million people in 29 countries worldwide.

1.1.2. Indian Scenario

Elevated concentrations of fluoride have been documented in specific regions: for instance, Elementa in Kenya recorded 1,640 mg/L, Nakuru Lake in Kenya measured 2,800 mg/L, Ethiopia showed 177 mg/L, and India exhibited 69.7 mg/L (Nair and Manji, 1982; Haimanot et al., 1987; Kloos and Haimanot, 1999; Varughese et al., 2009; Dharmshaktu, 2013). When it comes to the bioaccumulation of fluoride in fruits and vegetables, typical levels range from 0.1 to 0.4 mg/kg, contributing to everyday exposure. However, higher concentrations have been detected in specific foods such as rice and barley (2 ~ 8 mg/kg), fish protein (370 mg/kg), pulses (around 13 mg/kg), fish (2 ~ 5 mg/kg), and radish (63 mg/kg) (Mumtaz et al., 2015; Bhattacharya et al., 2017) (see Table 1).

Table 1. Fluoride Concentration Ranges and Impacted Districts across Various Indian States

<table>
<thead>
<tr>
<th>Indian States</th>
<th>Range of Fluoride (mg/L)</th>
<th>Number of Affected Districts</th>
</tr>
</thead>
<tbody>
<tr>
<td>Andhra Pradesh</td>
<td>0.12 ~ 22.00</td>
<td>20</td>
</tr>
<tr>
<td>Assam</td>
<td>0.21 ~ 23.00</td>
<td>5</td>
</tr>
<tr>
<td>Bihar</td>
<td>0.61 ~ 0.81</td>
<td>9</td>
</tr>
<tr>
<td>Chhattisgarh</td>
<td>0.32 ~ 0.52</td>
<td>12</td>
</tr>
<tr>
<td>Delhi</td>
<td>0.42 ~ 3.00</td>
<td>6</td>
</tr>
<tr>
<td>Gujarat</td>
<td>1.60 ~ 32.00</td>
<td>18</td>
</tr>
<tr>
<td>Haryana</td>
<td>0.18 ~ 47.00</td>
<td>14</td>
</tr>
<tr>
<td>Jammu and Kashmir</td>
<td>0.06 ~ 4.22</td>
<td>3</td>
</tr>
<tr>
<td>Jharkhand</td>
<td>0.20 ~ 4.60</td>
<td>6</td>
</tr>
<tr>
<td>Karnataka</td>
<td>0.22 ~ 19.00</td>
<td>21</td>
</tr>
<tr>
<td>Madhya Pradesh</td>
<td>0.07 ~ 4.40</td>
<td>19</td>
</tr>
<tr>
<td>Maharashtra</td>
<td>0.10 ~ 10.40</td>
<td>8</td>
</tr>
<tr>
<td>Orissa</td>
<td>0.62 ~ 5.80</td>
<td>11</td>
</tr>
<tr>
<td>Punjab</td>
<td>0.45 ~ 6.40</td>
<td>12</td>
</tr>
<tr>
<td>Rajasthan</td>
<td>0.24 ~ 70.00</td>
<td>31</td>
</tr>
<tr>
<td>Tamil Nadu</td>
<td>1.51 ~ 5.40</td>
<td>16</td>
</tr>
<tr>
<td>Uttar Pradesh</td>
<td>0.11 ~ 2.40</td>
<td>1</td>
</tr>
<tr>
<td>Uttar Pradesh</td>
<td>0.14 ~ 16.40</td>
<td>11</td>
</tr>
<tr>
<td>West Bengal</td>
<td>1.58 ~ 13.70</td>
<td>10</td>
</tr>
</tbody>
</table>

India, the world’s second-largest country by population, stands as the seventh-largest in terms of landmass. Predictions indicate that by 2020, water usage will rise by 20 ~ 40%, coinciding with a population growth of approximately 1.55 billion. With only 4% of the world’s water resources, India accommodates a significant 16% of the global population (Susheela, 1999; Carrard et al., 2019; Verma et al., 2022a, 2022c, 2022d). In a study by Ayoob and Gupta (2006), the first documented case of fluoride presence in drinking water in India was identified in 1937, specifically in the Nellore district of Andhra Pradesh. Before the 1930s, symptoms of fluorosis were only reported in four states. However, by the years 1986, 1992, 2002, 2013, and as of today, the issue has expanded to affect 13, 15, 17, 18, and 19 states in India, respectively (Kumar and Kumar, 2015; Kisku and Sahu, 2020; Yadav et al., 2023). An analysis of the affected regions reveals that states such as Andhra Pradesh, Assam, Gujarat, Rajasthan, Tamil Nadu, Chhattisgarh, Uttar Pradesh, and Bihar are among the most severely impacted areas in India. Even Bihar, while not as severely affected as these states, still falls under the category of moderate fluoride contamination. Approximately 30 ~ 50% of Bihar’s districts show signs of fluorosis. According to research by Grover et al. (2003) and Yasmin et al. (2011), the affected districts in Bihar are Kaimur, Rohtas, Aurangabad, Gaya, Nalanda, Nawada, Shaikhpura, Munger, Jamui, Banka, and Bhagalpur.

According to Susheela (2001), the most extreme endemic regions for chronic fluorosis are Rajasthan, Gujarat, and Andhra Pradesh. In 9 out of 19 Indian states, including Andhra-Pradesh, Rajasthan, Madhya Pradesh, Haryana, Maharashtra, Assam, Delhi, Gujarat, Karnataka, and West Bengal (Dharmshaktu, 2013), the concentration of fluoride in groundwater exceeded 10 mg/L. According to a study conducted by Sahu et al. (2018), the fluoride levels in the groundwater in the Raebareli district of Uttar Pradesh were higher than 10 mg/L. Additionally, varying concentrations of fluoride, ranging from below 10 mg/L to above 5 mg/L, were detected in the groundwater of Bihar, Chhattisgarh, Odisha, Punjab, and Tamil Nadu. In 2002, Susheela pointed out that around 66 million individuals residing in 250 districts throughout India faced the threat of endemic fluorosis. Among them, 25 million individuals, primarily under the age of 18, suffered from dental fluorosis. Teotia and Teotia (1994) noted a fluoride deposition of 12 million tonnes in India’s earth’s crust. According to a 2009 report from the Ministry of Environment, Forestry, and Climate Change of the Indian Government, fluorosis affected 65 million people across 19 states in India, including 6 million children. This situation raises significant concerns for public health and well-being. During the 11th Five-Year Plan, the Ministry of Health and Family Welfare earmarked financial resources in 2008 ~ 2009 for the National Programme for Prevention and Control of Fluorosis (NPPCF) to tackle this problem. This initiative targeted 100 districts out of the total 230 districts with endemic fluorosis in India (Dharmshaktu, 2013).

In recent years, various approaches have been formulated to tackle the rising levels of fluoride found in water supplies. These methods encompass precipitation, ion exchange, electrocoagulation, and adsorption (Ahmad et al., 2022). Among these alternatives, adsorption has emerged as a particularly encouraging and versatile approach. This technique revolves around the adherence of fluoride ions to the surface of a solid substance referred to as an adsorbent (Yadav et al., 2018; Ahmad et al., 2022). The selection of the adsorbent material plays a pivotal role in dictating the effectiveness and economic viability of the procedure.
1.1.3. Effects on Human Health Due to Fluoride Concentration

The effect of fluoride on human health varies based on the length and intensity of exposure (Everett, 2011). Elevated fluoride levels can lead to dental issues such as discolored teeth, ranging from blackened and mottled to chalky-white, and, in some cases, the emergence of yellow to dark brown lines on teeth (Susheela and Jethanandani, 1996; Han et al., 2021). Additionally, excessive fluoride can cause the teeth to become rough. These manifestations collectively define dental fluorosis and are primarily associated with excessive fluoride exposure during early childhood (under 10 years of age) (Kloos and Zein, 1993; Everett, 2011; Ramesh et al., 2014; Mallishevy et al., 2020; Yadav et al., 2023). Initial signs of skeletal fluorosis, like intense discomfort in the joints, spine, and hips, become evident alongside the augmentation of bone mass and density. As the condition progresses to its more severe neurological phases, there might be spinal distortion, significant joint issues, total paralysis, muscle atrophy, and heightened spinal cord density (Boyle and Chagnon, 1995; Shahab et al., 2017).

Inhaling hydrogen fluoride (HF) leads to respiratory discomfort like coughing and choking, inducing significant irritation and potentially causing severe burns within the respiratory tract (Lee and Jeong, 2021). Direct contact with HF, whether in liquid or vapor form, with the eyes can result in prolonged exposure, possibly leading to enduring visual impairments. The WHO noted in 2002 that individuals exposed to high levels of HF are more likely to develop conditions like asthma or chronic lung diseases. When the concentration of fluoride surpasses the natural saturation levels (typically ranging from 0.02 to 0.83% within the hard tissues), it leads to increased absorption of fluoride (Maheshwari, 2006; Johnston and Strobel, 2020; Han et al., 2021). This excessive absorption results in an overflow of fluoride into the soft tissues, giving rise to various complications such as urinary, gastric, skin, muscular, and neurological issues. These complications can be compounded by concurrent infections and might even lead to fatalities. The processes of fluoride storage, distribution, and elimination within the human body are illustrated in Figure S1 which is found in the supplementary file (Kanduti et al., 2016; Shahab et al., 2017).

In summary, eliminating fluoride from water sources emerges as a vital undertaking to guarantee the universal availability of safe drinking water (Mohapatra et al., 2009). The use of affordable substances as adsorbents offers a practical resolution that merges fluoride reduction with the responsible repurposing of waste (Abdul-Rahman and Wright, 2014; Gai et al., 2021). This study adds to the expanding knowledge in water treatment and highlights the significance of factoring in economic, ecological, and technological aspects when crafting successful approaches for fluoride removal. In the face of global water quality challenges, the exploration of inventive and achievable remedies becomes crucial, and this research takes a leading role in such initiatives.

Fluorine, being a highly electronegative element, exhibits a strong attraction to positively charged ions like calcium. This property is important in terms of how it affects mineralized tissues like bones and teeth, which have a lot of calcium and are therefore good places for fluoride to build up in the form of calcium-fluorapatite crystals. Tooth enamel, for instance, primarily consists of crystalline hydroxyapatite.

It is crucial to emphasize that various factors, including fluoride intake from alternative sources, levels of physical activity, and dietary preferences, contribute to the extent of fluorosis. Refer to Table S2 for a concise overview of diverse fluorosis manifestations arising from excessive fluoride ingestion, based on the studies conducted by Murray and Bennett (1973) as well as Chaturvedi et al. (1990).

1.2. Adsorbent Concepts and Mechanisms

Adsorption is the process of using non-living biological materials to absorb organic and inorganic substances from an aqueous solution. The substances can be soluble or insoluble. It is important to distinguish adsorption from bioaccumulation, which involves the active and metabolic accumulation of metals and other substances by living organisms (Gadd, 2009). Adsorption is remarkably effective compared to conventional biotreatment methods (Fomina and Gadd, 2014), the significant reduction of pollutant ion concentrations, and in certain instances, their complete elimination, can be accomplished by employing cost-effective adsorbent materials (Moubarak and Grimí, 2015). The effectiveness of adsorption processes is notably influenced by various factors such as the dosage of the adsorbent, the initial pollutant concentration, the pH level of the solution, temperature, contact duration, and the size of the sorbent particles.

In general, increasing the amount of biosorbent used makes adsorption more effective because it exposes more active sites and gives pollutant ions more space to bind to (Homagai et al., 2010). Conversely, as the initial concentration of pollutant ions increases, the adsorption efficiency typically decreases.

1.2.1. Composition of Sugarcane Bagasse and Its Potential as an Adsorbent

Sugarcane bagasse is the residual fibrous material remaining once the juice has been extracted from sugarcane stalks. It is notable for being one of the most plentiful agro-industrial residues, characterized by its high content of lignocellulosic components (Paixão et al., 2014). Comprising cellulose, hemicelluloses, lignin, ash, and a small number of extractives, sugarcane bagasse possesses a wealth of valuable constituents (refer to Figure S2). Because lignocellulosic matter has many different functional groups, it has a strong pull–on pollutant ion (Okoro and Okoro, 2011; Pérez-Botella et al., 2022). Adsorbents derived from sugarcane bagasse consist of various large molecules, including humic and fulvic substances, lignin, cellulose, hemicelluloses, and proteins. These molecules exhibit diverse functional groups, such as –OH, –COOH, –NH₂, –CONH₂, –SH₂, and –OCH₃, which serve as sites for adsorption. Boni et al. (2016) found that sugarcane bagasse is very good at removing pollutants. This is because it has different binding sites, which Yu et al. (2012) had already pointed out, and a high silica content of 10.3%. Ngah and Hanafah (2008) pointed out that the biological polymers in sugarcane bagasse, such as
cellulose and lignin, give the developed adsorbent materials more good qualities.

1.2.2. Adsorbents Derived from Sugarcane Bagasse

Adsorbents originating from sugarcane bagasse are developed from the residual byproduct of sugarcane processing, specifically the fibrous material that remains after extracting sugar juice from sugarcane. This sustainable and renewable resource can be harnessed to manufacture diverse high-value products, including adsorbents. These adsorbents have garnered significant interest due to their capacity to effectively eliminate impurities and pollutants from liquids, notably water, and other fluid streams. Certainly, here are some essential details regarding adsorbents sourced from sugarcane bagasse:

- Sugarcane bagasse-based adsorbents offer sustainability as they harness the abundant and renewable resource of sugarcane waste, effectively turning it into an eco-friendly material, thus minimizing waste and promoting environmental responsibility.
- Sugarcane bagasse can undergo various techniques for the creation of adsorbents, such as chemical alteration, physical activation, and thermal processing. These methods serve to improve the adsorption characteristics of materials derived from bagasse.
- Adsorbents made from sugarcane bagasse can be made more effective by increasing their surface area and porosity. This is usually done through processes like activation, which creates more adsorption sites.
- These materials are efficient at adsorbing various contaminants from aqueous solutions, including heavy metals, dyes, organic pollutants, and nutrients like phosphates and nitrates.
- Utilizing sugarcane bagasse as a raw material offers a cost-effective advantage, especially in regions with significant sugarcane production, making the production of adsorbents economically feasible.
- These adsorbents find frequent application in environmental remediation initiatives aimed at purifying polluted water sources, effectively removing toxins and pollutants, and thus elevating water quality.
- Adsorbents derived from sugarcane bagasse are inherently biodegradable, thus mitigating their environmental footprint during disposal. These versatile adsorbents are deployed across various sectors, including wastewater treatment, industrial effluent management, and agricultural endeavors, where they play a crucial role in mitigating nutrient runoff from fields.

The choice of suitable adsorbents is impacted by several elements, including surface area, porosity, chemical composition, and accessibility (Crini et al., 2019; Pourhakkak et al., 2021). A variety of economical adsorbents have been investigated by scientists, including activated alumina, bone char, chitosan, zeolites, and different agricultural waste materials (Rezende et al., 2011; Sabzehmeidani et al., 2021). The fundamental adsorption mechanism centers around surface interactions, during which fluoride ions stick to active sites on the surface of the adsorbent (Ahmad et al., 2022; Turki et al., 2023). Several factors, such as pH, duration of contact, initial fluoride concentration, and the amount of adsorbent used, influence this process (Mohapatra et al., 2009; de Moraes Rocha et al., 2015; Yadav et al., 2018). The efficacy of affordable adsorbents for eliminating fluoride has been thoroughly researched and recorded. These substances have displayed encouraging outcomes, frequently on par with or surpassing traditional adsorbents (Turki et al., 2023). Furthermore, their adaptability and widespread availability underscore their suitability for extensive water treatment usage (Yadav et al., 2018). Nonetheless, there are ongoing research and development efforts aimed at tackling challenges like restoring adsorbent capacity, ensuring prolonged effectiveness, and transitioning from small-scale laboratory testing to industrial-level implementation (Ngeno et al., 2022).

2. Materials and Methods

2.1. Study Area

Bilaspur City, located in Bilaspur district, serves as the administrative center and is the second-largest city in Chhattisgarh state. Positioned along the banks of the Arpa River, it has attracted numerous companies, both large and small, to establish their manufacturing and production facilities in and around the city. However, the extensive industrialization in Bilaspur has led to continuous pollution of the air, water, and soil in the region. Therefore, it is imperative to assess the level of pollutants present in the local water sources. The increasing values of parameters raise significant concerns for public health, especially when people consume water from these bore wells without prior treatment. Bilaspur predominantly experiences its annual precipitation during the monsoon season, with an average yearly rainfall of approximately 1,300 millimeters (51 inches). Precipitation plays a crucial role in sustaining the agricultural sector, enabling the growth of various crops like rice, wheat, and pulses (Verma et al., 2021; Sahu et al., 2022a; Verma et al., 2022b, 2022f). However, an excess of rainfall leads to flooding, causing challenges for city infrastructure and residences (Sahu et al., 2022b, 2022c; Verma et al., 2022e). Additionally, Bilaspur is characterized by a tropical wet and dry climate, falling into the Aw category according to the Köppen climate classification (Mishra and Ramgopal, 2015). This implies that the city goes through well-defined periods of wet and dry weather. Warm summers, a monsoon season, and mild winters are characteristics of the climate in Bilaspur. Consequently, Bilaspur City has seen significant growth in its population over the years. As per the latest update in September 2021, the city’s population was estimated to be over 400,000 people. With its relatively compact geographical area, Bilaspur boasts a noteworthy population density, surpassing 2,000 people per square kilometer. This high population density reflects the city’s status as a thriving economic and cultural hub in the Chhattisgarh region. In addition, Figure 1 depicts the index map of the study area.

The main motive behind the motivation or the selection of Bilaspur City as a case study for the present study, according to a Central Ground Water Board (CGWB) report from March 2019.
groundwater in the Bilaspur region of Chhattisgarh contains fluoride levels exceeding 1.5 mg/L. Therefore, in short, modified bagasse sugarcane (MBS) is used as an inexpensive adsorbent to remove fluoride from drinking water. This is done for several reasons, such as health concerns, following rules, caring about the environment, having limited funds, making good use of resources, and the constant search for new and eco-friendly ways to treat water.

2.2. Data Used
Accurate and representative water sample collection in any urban area like Bilaspur City demands meticulous preparation and strict adherence to established procedures. Water sampling plays a pivotal role in multiple facts, encompassing the evaluation of water quality, environmental surveillance, and safeguarding public health. Below are some details regarding the data type and its source.

The samples were taken from the Arpa River Basin in Bilaspur City, Chhattisgarh. Subsequently, we have also collected the data based on random sampling techniques from different houses in different locations of the city.

3. Methodology
In the present study, utilizing the modified bagasse, a by-product of sugarcane processing, is a viable approach for addressing the removal of fluoride from water, which is crucial for averting health problems linked to excessive fluoride consumption. Various techniques and methods can be employed to achieve this objective which are summarized in the following sub-sections.

3.1. Physical Modification
Fluoride can be effectively eliminated from water or other solutions using a range of physical modification techniques. These methods encompass processes such as cutting, grinding, ball milling, heating, and boiling. They serve the purpose of either causing the precipitation of fluoride ions or promoting the adsorption of fluoride onto particular materials. Below, we detail how these physical modification approaches can be employed for fluoride removal:

(a) Machining processes such as cutting and grinding are employed to generate fine metal particles or powder from materials like aluminum, calcium, or magnesium. When these metals come into contact with fluoride ions in water, they undergo a chemical reaction leading to the formation of insoluble metal fluoride precipitates. The finely fragmented metal can exhibit an increased surface area, thereby promoting the reaction.

(b) Ball milling is an effective method for enhancing the surface area and reactivity of substances. It can be employed to finely grind and activate materials such as calcium hydroxide (lime) or calcium oxide (quicklime). Subsequently, these activated materials can be introduced into water to engage with fluoride ions and induce the precipitation of calcium fluoride.

(c) Heating can facilitate the removal of fluoride ions by encouraging the creation of metal fluoride compounds that are insoluble. As an illustration, when water containing fluoride ions is heated with calcium chloride, calcium fluoride will precipitate.

(d) Boiling water in the presence of a calcium-based precipitant such as calcium hydroxide or calcium carbonate can facilitate the removal of fluoride. The application of heat promotes a reaction that leads to the formation of calcium fluoride precipitates.

3.2. Chemical Modification
This method can be employed in various ways, including chemical treatment and surface modification techniques which are presented below:

(a) When calcium-containing substances like calcium chloride or calcium hydroxide are added to water, insoluble calcium fluoride (CaF₂) precipitates can form. These can be removed by settling or filtration. Adding aluminum-containing compounds like aluminum sulphate or alum to water can also cause aluminum fluoride (AlF₃) to form, which can be separated from the water using the right techniques.

(b) When amino, hydroxyl, or carboxyl units are added to the surface of nanoparticles during their creation, they can be used as adsorbents to remove fluoride ions. These specialized functional groups exhibit a high affinity for binding to fluoride ions. By designing membranes with surface modifications, it becomes possible to allow the selective passage of water while...
effectively trapping fluoride ions. These surface alterations may involve the incorporation of specific molecules or groups designed to attract and capture fluoride ions.

3.3. Biological Modification

Utilizing microorganisms for biological modification to eliminate fluoride from water holds significant potential as an eco-friendly and effective approach to address the prevalent problems of fluorosis and groundwater pollution. Microorganisms like bacteria and fungi have a natural ability to engage with different substances in their surroundings, including their capacity to adsorb, absorb, or enzymatically break down fluoride ions. Researchers have been investigating methods to improve these microorganisms' capacity to remove fluoride through biological modification techniques. Moreover, the application of biofilms, which are communities of microorganisms adhering to surfaces, can improve the process of fluoride removal. Biofilms possess a substantial surface area and can be tailored to include particular microorganisms capable of adsorbing fluoride. These biofilm-based setups can be incorporated into water treatment plants or employed within localized water purification units serving communities.

Figure 2. Plots of the fluoride removal efficiency versus contact time due to (a) RHA, (b) modified coconut husk, and (c) MBS, respectively.

In summary, the application of biological modification with microorganisms shows significant potential for tackling the issue of excessive fluoride. Leveraging the inherent capabilities of these microorganisms and improving their fluoride-removal capacities through genetic engineering or optimizing their growth conditions offers a promising avenue for addressing fluoride contamination sustainably and economically. As ongoing research in this area progresses, we can anticipate the prospect of providing safer and purer water sources for communities impacted by fluorosis.

4. Results and Discussion

4.1. Effect of Contact Time

The fluoride solution was diluted to a known concentration, and then 250 mL of this solution was utilized for experimentation. Various doses ranging from 0.1 to 0.7 g/100 mL of RHA, coconut husk, and sugarcane bagasse were applied to assess their impact on fluoride removal under pH = 6 conditions with a contact time of 60 minutes. The results indicated that as the dosage increased, fluoride removal also increased, but beyond a certain point, in the case of aluminum-hydroxide rice husk, removal efficiency began to slightly decrease, as illustrated in Figure 2a. The highest removal efficiency, ranging from 75 to 89%, was achieved at a dosage of 0.7 g/100 mL.

4.2. Effect of Adsorbent Dose

The effectiveness of fluoride removal using modified coconut husk exhibited varying results, with removal efficiency ranging from 61 to 75%. Initially, the removal efficiency increased, then slightly decreased at doses of 0.1 and 0.3 g/100 mL. However, at a dose of 0.5 g/100 mL, the removal efficiency consistently increased from 15 to 75%, as depicted in Figure 2b. Regarding MBS, at doses of 0.1, 0.3, 0.5, and 0.7 g/100 mL and a contact time of 75 minutes, the percentage removal efficiency exhibited continuous improvement until it reached a plateau after 75 minutes. The highest percentage removal efficiency ranged from 48% (at 0.1 g/100 mL) to 78% (at 0.7 g/100 mL). This can be attributed to the presence of an optimal dose that provides a higher availability of adsorption sites (refer to Figures 2b and 2c).
4.3. Effect of pH
The influence of pH on the adsorption of fluoride from a known concentration fluoride solution was investigated using an adsorbent dose of 0.5 g/100 mL and a contact time of 75 minutes. The pH was varied from 2 to 14 in the experiment. At lower pH values, the surface of the adsorbent became positively charged, which facilitated the adsorption of fluoride ions through anion exchange. The removal efficiency exhibited its lowest value of 30% at pH = 5 and reached its highest efficiency of 53% at pH = 13 when using MBS as the adsorbent. Similarly, when using aluminum hydroxide (AH) coated RHA as the adsorbent, the removal efficiency varied from 63 to 92%. It exhibited a continuous increase from pH = 2 to pH = 7, followed by a decrease in efficiency within the pH range of 7 ~ 9. Subsequently, it increased again within the pH range of 9 ~ 13. Likewise, for modified coconut husk, the removal efficiency ranged from 22 to 79%. It demonstrated a consistent increase from pH = 2 to pH = 7, a decrease in efficiency within the pH range of 7 ~ 9, and another increase within the pH range of 9 to 11 (see Figure 3).

4.4. Effect of Initial Concentration
The solution was diluted to create a range of known concentrations, spanning from 2 to 10 mg/L, with each concentration placed in 300 mL bottles. These bottles were then placed inside an incubator shaker to ensure continuous mixing. Notably, Figure 4a illustrates that as the initial fluoride concentration increased, the removal efficiency decreased. This decline occurred because the adsorbent material’s capacity was rapidly exhausted due to the higher initial fluoride concentrations. This was primarily because, for a fixed amount of adsorbent, the available adsorption sites became limited and saturated at higher initial concentrations. Additionally, it was observed that the total amount of adsorbate adsorbed per unit mass of adsorbent increased as the initial concentration of adsorbate was altered within the range of 2 ~ 10 mg/L at various temperatures ranging from 293 to 323 K, as depicted in Figures 4a and 4b.

4.5. Effect of Temperature
A known concentration of fluoride solution was utilized in an experiment where a constant adsorbent dose of 0.5 g/100 mL was applied. The investigation focused on assessing the impact of temperature variation in the range of 283 ~ 333 K, with measurements taken every 15 minutes. The results revealed a notable trend: As the temperature increased, the percentage of fluoride removed from the solution also increased. At the temperature of 323 K, the maximum removal efficiency was observed, reaching 78% for MBS, 78% for AH coated RHA, and 49% for magnetic catonic hydrogel (MCH), as illustrated in Figure 4c. In conclusion, the present article indicates that utilizing MBS as an adsorbent can be a cost-efficient and efficient method for eliminating fluoride from drinking water. Enhancing the process’s effectiveness can be achieved through optimizing factors such as contact time, adsorbent quantity, pH levels, initial concentration, and temperature. Grasping these influences is essential when developing practical and economical systems for eliminating fluoride in regions where high fluoride concentrations in drinking water are a health concern.

4.6. Mitigation Measures in Fluoride Affected Area
An awareness initiative has been orchestrated in districts grappling with arsenic, fluoride, and iron contamination, aiming to achieve the following objectives. The communities residing in impacted regions are cognizant of the presence of arsenic and fluoride in hand pump water, which can lead to severe health complications. The residents in the impacted regions have a clear comprehension of the significance associated with a hand pump spout colored in red and blue. Developing a conventional water treatment facility to provide piped water supply to multiple villages in areas impacted by arsenic and fluoride contamination. A water distribution system sourced from the most secure underground water reservoir. Engage international organizations and non-governmental organizations (NGOs) specializing in water quality and public health to provide technical expertise and financial support. Allocate resources to ongoing research efforts aimed at gaining a deeper understanding of local geological and hydrological factors contributing to fluoride contamination. Extend dental care and rehabilitation services to individuals affected by dental issues related to excess fluoride exposure. Disseminate and advocate for the adoption of household water filtration systems capable of effectively removing surplus fluoride from drinking water. Implement water treatment infrastructure, such as deflourination plants, to eliminate excessive fluoride from the public drinking water supply. Encourage the utilization of alternative water sources that naturally contain lower levels of fluoride.
5. Conclusions

This study presents preliminary findings on fluoride contamination and fluorosis in Bilaspur, Chhattisgarh. Individuals living below the poverty line face a high risk of fluorosis. The presence of fluoride in Bilaspur’s groundwater is not attributed to human activities since there are no industries in the vicinity. Instead, it originates from natural geological sources. Due to the area’s propensity for Naxalite insurgencies, the residents of this fluoride-endemic region in Bilaspur endure unfortunate circumstances. The initial solute concentration is one of many factors that affect fluoride ion removal efficiency. When the contact time and the amount of adsorbent used increase, the percentage of fluoride ion removal efficiency also increases, whereas it decreases when these variables decrease. Regarding pH levels, the lowest removal efficiency is observed at pH = 5, where it stands at 30%, while the highest efficiency is at pH = 13, reaching 53% for MBS. Similarly, for AH coated RHA, the removal efficiency varies from 63 to 92%. In terms of temperature, both MBS and AH coated RHA show the highest efficiency, followed by modified coconut husk. As the initial concentration of fluoride increases, the effectiveness of adsorbent materials in removing it decreases. This decline occurs because the adsorbent’s capacity becomes saturated at higher initial fluoride concentrations. During a sequence of experiments where we manipulated the initial fluoride concentration within the range of 2 ~ 10 mg/L and conducted these experiments at various temperatures spanning from 293 to 323 K, we noted that the quantity of adsorbate adsorbed per unit mass of adsorbent exhibited an upward trend with increasing initial concentrations. To address the issue of fluoride contamination in Bilaspur’s widespread areas, it is imperative to monitor water samples regularly and assess the health status of the local villagers.

This information will enable us to effectively communicate the severity of the problem to the government, urging them to install defluorination plants. Additionally, it is essential to raise awareness among the villagers about the causes and consequences of fluorosis and promote solutions like rainwater harvesting for access to safe drinking water.

Eliminating fluoride from drinking water can yield both positive and negative outcomes. On the positive side, this action can decrease the likelihood of dental fluorosis, a condition stemming from excessive fluoride consumption, which causes discoloration or staining of teeth. Additionally, it grants individuals greater control over their fluoride intake, especially for those who already have access to fluoride from sources such as toothpaste or dental procedures. Conversely, the absence of fluoride in drinking water might amplify the prevalence of tooth decay, particularly in areas where access to dental care or other fluoride sources is limited. Fluoride plays a crucial role in cavity prevention, notably supporting children’s dental health.

The removal of fluoride could potentially raise healthcare expenses due to an increase in dental treatments for preventable cavities, disproportionately affecting lower-income communities. Finding a delicate balance is crucial to mitigating risks to dental health while considering personal choice and alternative fluoride sources.

6. Limitations of the Present Study

Similar to any water treatment technique, it comes with its own set of constraints and difficulties. Here are some of the constraints linked to this approach.

The availability of bagasse sugarcane as an adsorbent material hinges on the sugarcane industry. In areas with limited or seasonal sugarcane cultivation, ensuring a steady provision of bagasse for water treatment purposes can be a formidable task. Furthermore, the sustainability of employing bagasse as an adsorbent may come into question due to its potential competition with other valuable applications, like bioenergy production.

Improving the ability to absorb and selectivity of bagasse-based adsorbents often requires chemical changes, which make the treatment process more complicated. Additionally, the management of the chemicals employed in this process is crucial to mitigating potential environmental repercussions.

In summary, utilizing MBS as an economical adsorbent for fluoride removal in drinking water shows promise, but it is important to acknowledge its drawbacks. These problems include limited adsorption capacity, sensitivity to pH levels, limited reusability, competition with other anions, slow adsorption kinetics, limited availability, and the need for pre-treatment. Despite these challenges, bagasse-based adsorbents can still contribute significantly to fluoride removal, particularly in areas where cost-effective solutions are crucial. Nevertheless, gaining a comprehensive understanding of these limitations and how to manage them is vital to ensuring the effectiveness and sustainability of this approach to providing safe drinking water. Researchers and practitioners must continue their efforts to enhance the efficiency of MBS and address its limitations, making it a more viable solution for fluoride removal in various contexts.

7. Future Scope

The present study not only provides an effective way to remove fluoride but also holds promise for various future applications and enhancements. Future research can concentrate on enhancing the modification methods used on bagasse sugarcane to further boost its adsorption capacity. This may encompass chemical alterations, nano-structuring, or integrating additional materials to heighten its effectiveness in removing fluoride. The advancement of more effective and specific adsorbents has the potential to result in significant advancements in water treatment.

Gaining a more profound insight into the kinetics and mechanisms underlying fluoride adsorption onto MBS is imperative. Subsequent research endeavors can explore the intricate microscopic processes that occur during adsorption, thus facilitating enhanced control and optimization of the removal process. Although laboratory experiments have demonstrated potential, the ability to scale up this technology remains a hurdle. Future investigations should emphasize the development of viable and expandable systems suitable for real-world implementation, particularly in areas afflicted by significant fluoride contamination. This might entail initiating pilot initiatives and partnering with local communities.

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In the overall scenario, the future potential of employing MBS as a cost-effective adsorbent for eliminating fluoride from drinking water appears highly encouraging. This method not only tackles the pressing problem of fluoride contamination but also harmonizes with economic, environmental, and sustainability objectives. Through ongoing research and innovation, this approach could potentially transform water treatment techniques, especially in areas heavily affected by fluoride contamination.

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References


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