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ORIGINAL ARTICLE

Potentiality of a fruit peel (banana peel) toward abatement of fluoride from synthetic and underground water samples collected from fluoride affected villages of Birbhum district

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Abstract

Contamination of underground water with fluoride (F) is a tremendous health hazard. Excessive F (> 1.5 mg/L) in drinking water can cause both dental and skeletal fluorosis. A fixed-bed column experiments were carried out with the operating variables such as different initial F concentrations, bed depths, pH and flow rates. Results revealed that the breakthrough time and exhaustion time decrease with increasing flow rate, decreasing bed depth and increasing influent fluoride concentration. The optimized conditions are: 10 mg/L initial fluoride concentration; flow rate 3.4 mL/min, bed depth 3.5 and pH 5. The bed depth service time model and the Thomas model were applied to the experimental results. Both the models were in good agreement with the experimental data for all the process parameters studied except flow rate, indicating that the models were appropriate for removal of F by natural banana peel dust in fix-bed design. Moreover, column adsorption was reversible and the regeneration was accomplished by pumping of 0.1 M NaOH through the loaded banana peel dust column. On the other hand, field water sample analysis data revealed that 86.5% fluoride can be removed under such optimized conditions. From the experimental results, it may be inferred that natural banana peel dust is an effective adsorbent for defluoridation of water.

Keywords Banana peel · Fluoride · Column study · Dental and skeletal fluorosis · Regeneration

Introduction

Due to rapid industrialization, huge quantity of pollutants discharges in to the environment. Fluorine is one such pollutant that threatens living organisms, in particular humans (Koilraj and Kannan 2013). Fluoride is a strongly electronegative element, and its presence in drinking water is essential for human health (Chen et al. 2012). However, excess consumption of fluoride can lead to both dental and skeletal fluorosis (Kierdorf et al. 2016; Death et al. 2015). Previous literature (Mohan et al. 2012) demonstrated that more than 200 million people worldwide are affected by fluoride-contaminated drinking water with very high concentration. As per the guideline adopted by WHO (2011) and Bureau of Indian standard (BIS 1991), the concentration of fluoride in drinking water should not exceed beyond 1.5 mg/L. Many technologies such as precipitation, ion exchange, electrolysis have been developed. However, all the above techniques are not cost-effective. These are high maintenance charge and not easy to regenerate. But there is another technique, called adsorption which is an important cost-effective technique widely used in most of the developing country for removal of fluoride from drinking water (Mondal and Roy 2015).

Pertinent literature highlighted that many cost-effective adsorbents such as sugarcane bagasse, tea waste ash, aluminum impregnated potato plant ash and coconut fiber ash, lemon leaf dust, banana peel dust, rice husk carbon, tea ash, silica gel, egg shell dust, calcareous soil, coconut fiber, banana peel have been used for removal of fluoride from contaminated drinking water (Ghosh et al. 2016; Mondal et al. 2015; Bhaumok et al. 2013; Mondal et al. 2012a, b, c, d; Bhaumik et al. 2012; Bhaumik and Mondal 2014a, b). But none of the above adsorbents showed excellent removal performance along with high adsorption capacity. Therefore, it is prime important to synthesize an efficient and cost-effective adsorbent for decontamination of fluoride from the aqueous medium (Chen et al. 2012).

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Adsorption technique normality processed through batch and column process (Tor et al. 2009). But in batch process, all the operating variables remain constant while one variable changes at a time, and the process is costly and time-consuming. Therefore, column separation process may be the suitable one for defluoridation of domestic as well as community level. In our previous work, we have used the banana peel for removal of fluoride through batch process followed by response surface optimization techniques. Very recently, we have tried with another variety of banana peel for the same purposes. But, we cannot achieve to reach the desired level.

Bananas are a popular fruit consumed worldwide with an early production of over 145 million tonnes in 2011. Once the peel is removed, the fruit can be raw or cooked and peel is generally discarded. On average, banana peel contains 6–9% dry matter of protein and 20–30% fiber. Normally, the ripe banana peels contains 30% free sugar and 15% more starch than green banana peels. Moreover, banana peel has many biomolecules such as lignin, cellulose and hemicellulose with variety of active functional groups (carboxyl, hydroxyl, amine, etc.) which suppose to bind the pollutants (Deshmukh et al. 2017; Kumar et al. 2011).

The present work is aimed at synthesizing an efficient and economical adsorbent using banana peel toward removal of fluoride from water through column study. Finally, the prepared adsorbent was used for removal of fluoride from field samples which was collected from the fluoride affected villages of Birbhum district, West Bengal.

Materials and methods

Preparation of banana peel dust

The raw waste banana peels (*Musa paradisiaca*) were collected from nearby fruits stall of Burdwan town, West Bengal. After collection, peels were thoroughly washed with distilled water followed by washing with de-ionized water and dried in hot air oven at 50 °C for 12 h. The dried peels were cut into small pieces and again dried in hot air oven maintaining 60 °C temperatures for 24 h. The dried banana peels were ground using kitchen grinder to get the desired particle size (200 μm), the powder form of banana peel was stored in airtight container, the physico-chemical analysis was done, and the results are depicted in Table 1.

Point of zero charge (pH_{ZPC})

Zero point charge of the banana peel was determined by following the solid addition method (Mondal 2010). Initially, a series of conical flasks were taken with 50 mL of 0.1 (M) KNO_3 solutions in each flask along with 0.1 g banana peel.

Table 1 Physical characteristics of banana peel

Parameter	Values
pH	6.59
Conductivity (mS/cm)	190
Specific gravity	2.66
Bulk density (g/cc)	0.384
Particle density (g/cc)	0.138
Porosity (%)	178.26
Moisture content (%)	2.98
P^{H}_{zpc}	5.63
Dry matter	89.73
Crude fiber	10.33
Ash	17.94
BET surface area	23.983 m^2/g

The pH of the 0.1 (M) KNO_3 solutions was adjusted by using 0.05 (N) HNO_3 and 0.1 (N) KOH solutions. After thoroughly shaking (24 h), the pH of the final solutions was recorded and a graph was constructed by pH versus ΔpH.

Column study

Thomas model

The Thomas model can be used to predict the breakthrough curve and the maximum adsorption capacity of the banana peel in a fixed-bed operation. The linearized form of Thomas model can be presented in Eq. (1):

$$\ln\left(\frac{C_0}{C-1}\right) = \frac{K_{Th}q_0X}{Q} - \frac{K_{Th}C_0t}{Q} \quad (1)$$

where X is the amount of sorbent taken in the column (g) and Q is the effluent flow rate (mL/min). The kinetic coefficient K_{Th} and the adsorption capacity of the bed q_0 for the defluoridation process can be calculated by plotting $\ln(C_0/C-1)$ versus t.

Bed depth service time model (BDST)

It is familiar that BDST model provide a simplest approach and suitable prediction of absorber performance (Al-Degs et al. 2009). BDST model basically provides a relationship between column bed depth, X and service time t. However, this particular model also focused on the estimation of characteristic parameters such as maximum adsorption capacity (N_0) kinetic constant (K). Moreover, this model considers one valuable assumption that is the adsorption rate is proportional to residual capacity of the sorbent and the concentration of the sorbing species. The relationship of service time with process conditions and operating parameters is given as Eq. (2):

$$\ln\left(\left[\frac{C_0}{C_b}-1\right]\right)=\ln\left(\exp\left(\frac{KXN_0}{v}\right)-1\right)-KC_0t \quad (2)$$

A linear relationship between bed depth and service time may be given by Eq. 3 (Al-Degs et al. 2009)

$$t=\frac{N_0X}{vC_0}-\frac{1}{KC_0}\ln\left(\frac{C_0}{C_b}-1\right) \quad (3)$$

where C_0 is the initial concentration of solute (mg/L), C_b the desired concentration of solute at breakthrough (mg/L), K the adsorption rate constant (L/mg/h), N_0 the adsorption capacity (mg/L), X the bed depth of column (cm), v the linear flow velocity of feed to bed (cm/h), and t the service time of column under above conditions (hrs). Equation (4) can be rewritten in the form of a straight line.

$$T = aX - b \quad (4)$$

where 'a' is slope of BDST line ($a=\frac{N_0}{vC_0}$) and the intercept of this equation represents as Eq. (5):

$$b=\frac{1}{KC_0}\ln\left(\frac{C_0}{C_b}-1\right) \quad (5)$$

Thus, N_0 and K can be evaluated from slope (a) and the intercept (b) of the plot of t versus X, respectively.

Column regeneration study

In order to activate the exhausted column, regeneration experiments were conducted by using 0.1 (M) NaOH solutions. The desorption experiments were conducted with the flow rate 2 mL/min. After passing 100 mL of 0.1 (M) NaOH, the entire column bed was washed with double distilled water. Finally, the regenerated column was then used for the next cycle of column adsorption.

The desorption efficiency E_D was calculated by the following Eq. (6) (Zhu et al. 2007)

$$E_D=\frac{M_D\times 100}{[(q_E^m+M_A)]} \quad (6)$$

where M_D is desorption fluoride amount (mg) eluted by desorption solution (0.3 M NaOH). q_E and m are the capacity at exhaustion point (L) and adsorbent dose, respectively, and M_A is the fluoride amount (mg) retained in the column.

Analytical procedure

Effluent solution was collected at regular interval, and their concentrations were measured (Thermo Orion, 4 star); pH of the experimental solution was measured by using pH meter (Systronic 206). The breakthrough curves were constructed by Origin software (Version 6.0).

Dental and skeletal fluorosis study

Primary school children aged between 5–10 years were selected from fluoride affected villages of Birbhum district, West Bengal. Dental fluorosis was examined by a trained examiner who held a master degree in dentistry with the help of Dean index (Dean 1934). Skeletal fluorosis was assessed by clinical symptoms and physical exercise (Susheela and Bhatnagar 2002; Shashi et al. 2008).

Results and discussion

Adsorbent characterization

Preparation of banana peel dust was subjected to pH_{ZPC} and SEM study for understanding the zero point charge and surface morphology (HITACHI-S-530, Scanning Electron Microscope and ELKO Engineering, accelerating voltage of 20.0 kV) and surface chemistry of the banana peel was assessed by FTIR (BRUKER, Tensor 27). The zero point charge of the banana peel was recorded as 5.63 (Fig. 1), and from the scanning micrograph it is clear that the surface of the banana peel dust is absolutely rough in nature and huge heterogeneity (Fig. 2a). However, after F removal, the surface of banana peel is smooth with less degree of heterogeneity (Fig. 2b). The Fourier transformer infrared spectroscopy clearly revealed that it has sharp peaks at 3340, 2890 and 1741 cm^{-1} which corresponds the functional groups hydroxyl (–OH), alkyl C–H, and carboxyl (–COOH), respectively (Fig. 3). Therefore, it is expected that the functional groups such as –OH and –COOH may interact with fluoride during adsorption.

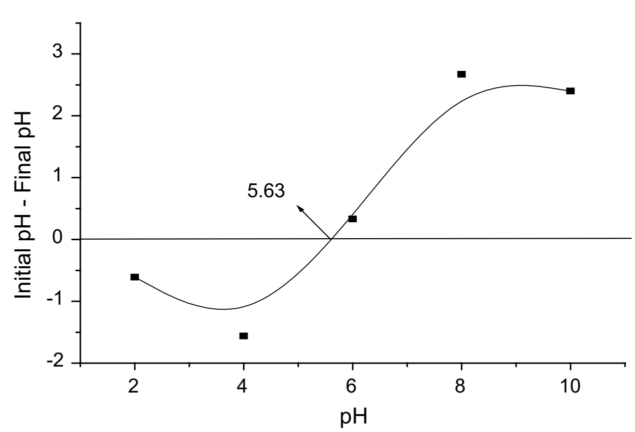


Fig. 1 ZPC of Banana peel dust

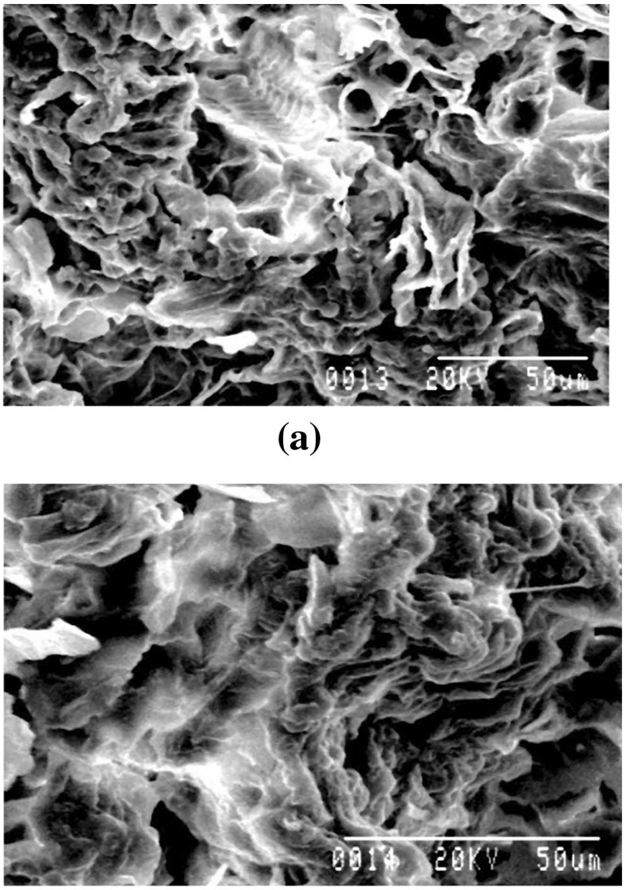


Fig. 2 Scanning electron microscopy of banana peel: **a** before passing fluoride solution (×1500 magnification), **b** after passing fluoride solution (×1500 magnification)

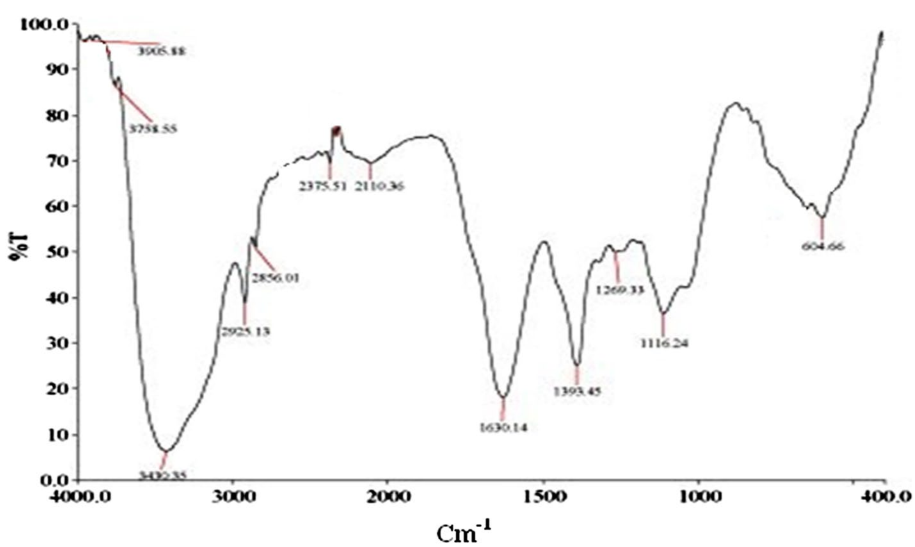
The effect of initial concentration of solution on breakthrough curve

Three different initial concentrations (1.5, 10, 35 mg/L) were used in this study. The values were chosen because they were considered to fairly represent fluoride concentration range in most groundwater. The bed mass and flow velocity of the present experiment have been taken as 2 g and 3.4 mL/min, respectively. The breakthrough curves (BTCs) obtained from the experimental investigation of the effect of the initial concentration on fluoride removal are depicted in Fig. 4. From Fig. 4 it is clear that higher the fed concentration, the steeper is the breakthrough curve and smaller is the breakthrough time. The adsorbent exhaustion rates (AER) (bed volume (BV) in parenthesis) were computed and found to be 0.046(3.31), 0.023(6.16) and 0.009(15.39) g/L for the initial concentrations of 35, 10 and 1.5 mg/L, respectively (Table 2). As the concentration increase, the driving force for adsorption increases and the active sites are consumed faster. The results also highlighted that both saturation rate and breakthrough time change with changing influent concentration (Goel et al. 2005). This is because, at higher concentration, the active sites of the adsorbent get saturated very quickly (Mondal et al. 2013).

The effect of flow rate on breakthrough curve

Steeper breakthrough curve was obtained at higher flow rates (Fig. 5). Further it was studied that the steepness of the breakthrough curve reduces with decreasing flow rates i.e., increase in breakthrough time. The breaks through curves appeared steeper at higher flow rates may be due to faster movement of adsorption zone along the bed aiding its quick saturation (Sulaiman et al. 2009; Tor et al. 2009). Again the variation in the slope of the breakthrough curve

Fig. 3 FTIR spectrum of banana peel dust (BPD)



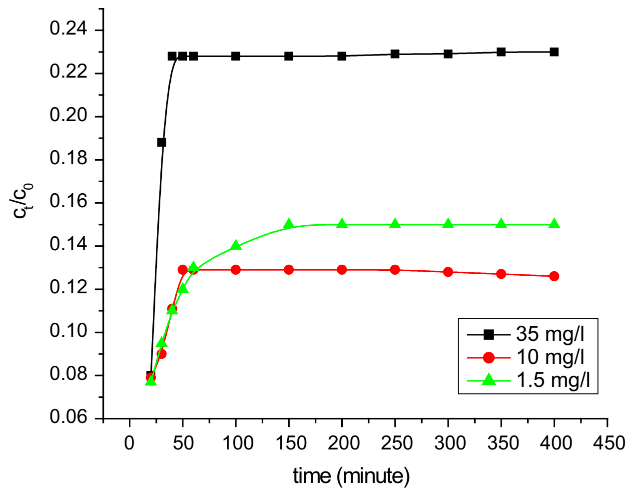


Fig. 4 Breakthrough curve of the effect of different influent concentration on fluoride adsorption onto banana peel ($v = 3.4$ ml/min, $Z = 0.9$ cm, pH = 5)

and adsorption capacity may be explained on the basis of mass transfer fundamentals. The reason is that at higher flow rate the rate of mass transfer increases, i.e., the amount of fluoride adsorbed onto unit bed height (mass transfer zone) increased with increasing flow rate leading to faster saturation at higher flow rate (Talnikar et al. 2004). The increase in adsorption capacity (both up to breakthrough and exhaust) of banana peel was more pronounced at the flow rate of 1.7 mL/min, whereas the effect is almost negligible at the flow rate of 8.0 mL/min. The rate constant, K_{Th}, increased with increasing the flow rate which indicates that the mass transport resistance decreases (Tor et al. 2009). The different bed volumes (BV) of water processed at breakthrough curve were found to be 6.02, 30.08, and 176.93 L for 1.7, 3.4, and 8.0 mL/min, respectively (Table 2). The increase in volume processed with an increased in velocity can be attributed to reduced influence of external mass transfer resistance (Ghribi and Chlendi 2011). The AER values for this respective flow rate are 0.029, 0.006 and 0.001 g/mL, respectively.

The effect of pH on breakthrough curve

The breakthrough curves for different pH are shown in Fig. 6. The pH of the aqueous solution has been recognized as one of the most important factors influencing adsorption kinetics due to its direct influence on the surface properties of the adsorbent (Memon et al. 2008). As shown in Fig. 6, where the value of pH was 5, the value of C_t/C_0 reached 0.042 in 30 min. But the breakthrough curve of the pH 7.0 and 9.0 was not more than 0.045 and 0.109 in the different time periods, respectively. With a decrease in pH in the influent, the breakthrough curves shifted from left to right, which indicates higher removal of fluoride. It would spend more time reaching the saturation, and the efficiency of biosorption was much higher (Ma et al. 2008). The F ion binding could be attributed to several mechanisms such as ion exchange, complexation, electrostatic attraction and precipitation. For banana peel, ion exchange has been considered as a main mechanism responsible for pollutant sequestering (Sheng et al. 2004). The result suggests that with the decrease in pH, under experimental condition, the adsorption capacities increase (Table 2). So the removal of fluoride from aqueous solution was more efficient at lower initial pH value.

Table 2 Summary of pertinent results at breakthrough point of banana peel

Parameters	Service time (min)t_b	Adsorption capacity (mg/g) q_b	Bed volume processed (BV)	AER (g/ml)
Adsorbent mass (g)				
2	40	0.405	12.03	0.015
4	100	0.429	15.92	0.012
8	250	0.449	19.34	0.009
pH				
5.0	30	0.479	9.02	0.020
7.0	60	0.477	18.05	0.010
9.0	300	0.441	90.23	0.002
Different flow (ml/min)				
1.7	40	0.495	6.02	0.029
3.4	100	0.477	30.08	0.006
8.0	250	0.466	176.93	0.001
Initial concentration (mg/L)				
35.0	30	0.014	3.31	0.046
10.0	60	0.109	6.16	0.023
1.5	150	0.419	15.39	0.009

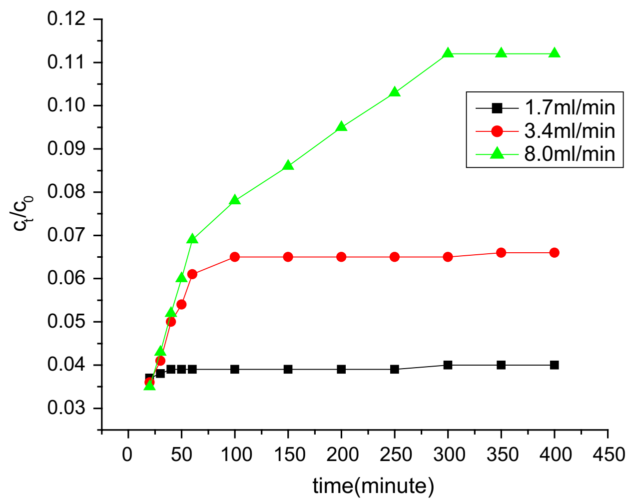


Fig. 5 Breakthrough curve of the effect of different flow rate on fluoride adsorption onto banana peel dust ($C_0 = 10.0$ mg/L, $Z = 0.9$ cm, pH = 5)

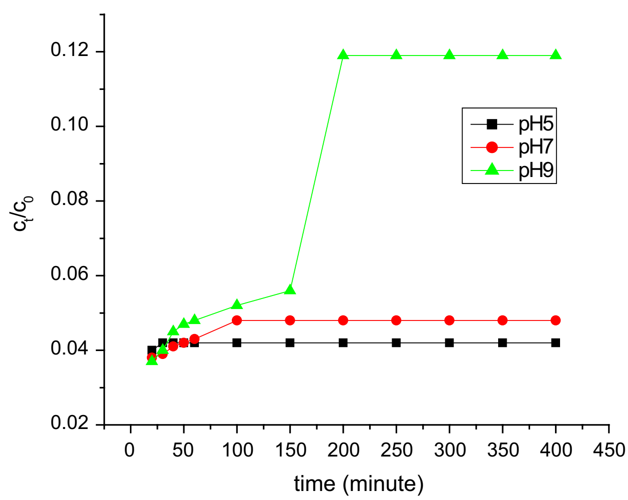


Fig. 6 Breakthrough curve of the effect of different pH on fluoride adsorption onto banana peel dust ($v = 3.4$ ml/min, $C_0 = 10.0$ mg/L, $Z = 2$ g)

The AER for different pH are 0.0196, 0.098, 0.020 g/mL, respectively. There are many reasons which may be attributed toward the fluoride adsorption behavior of the sorbent relative to solution pH. At lower pH, the surface of banana peel may get positively charged, which enhances the negatively charged fluoride ion through electrostatic force of attraction (Han et al. 2007). Almost similar results reported by Paudyal et al. (2013) for removal of F using a fixed-bed column packed with Zr (IV) loaded dried orange juice residue.

The effect of different bed depths on the breakthrough curve

Keeping the initial concentration of fluoride, flow rate and pH of the solution constant and with varying bed depth, the number of active sites available for sorption and the contact time of solute with the adsorbent were determined. Figure 7 shows the BTs obtained upon varying the quantity of adsorption media between 2, 4 and 8 g. From the BTCs, the t_b and q_b were determined and are summarized in Table 2. From the bed depth study, it was found that as the bed height increases, adsorbate had more time to contact with banana peel dust that resulted in higher removal efficiency of fluoride ion in column (Fig. 7). So, the higher bed depth results in a decrease in the solute concentration in the effluent at the same time. The slope of breakthrough curve decreased with increasing bed height, which resulted in a broadened mass transfer zone. High uptake was observed with highest bed depth 3.7 cm height due to an increase in the surface area of biosorbent, which provided more binding sites for the sorption (Vadivelan and Kumar 2005). Almost similar results were reported by Paudyal et al. (2013) and Mondal et al. (2013) for F removal by dried orange juice residue and sugarcane charcoal. The different bed volumes (BV) of water processed at breakthrough curve were found to be 12.03, 15.92, 19.34 L, and the AER value was found to be 0.0145, 0.0117, 0.0094 g/mL for 2, 4 and 8 g of banana peel dust adsorbent, respectively.

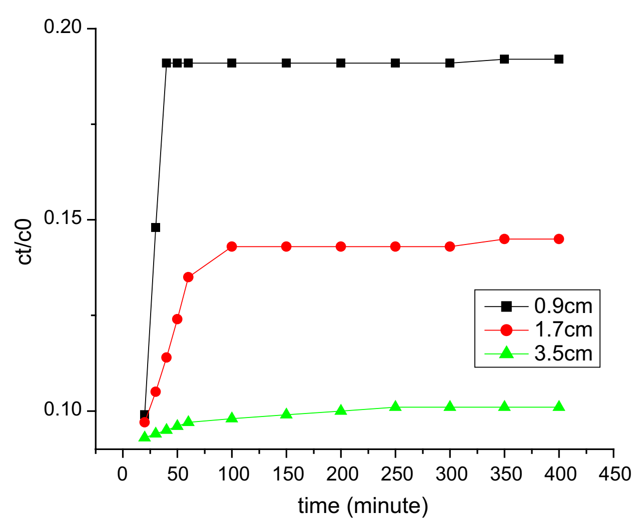


Fig. 7 Breakthrough curve of the effect of different bed depth on fluoride adsorption onto banana peel dust ($v = 3.4$ ml/min, $C_0 = 10.0$ mg/L, pH = 5)

Table 3 Calculated constants of Thomas model at different conditions using linear regression analysis

pH	Z (cm)	v (ml/min)	C_0 (mg/L)	K_{th} (ml/min/mg)	q_0 (mg/g)	R^2
5	3.5	3.4	10	55.12	98.26	0.999
7	3.5	3.4	10	55.12	98.26	0.999
9	3.5	3.4	10	55.12	98.26	0.999
5	0.9	3.4	10	7.80	3.76	0.996
5	1.7	3.4	10	7.80	1.88	0.996
5	3.5	3.4	10	7.80	0.94	0.996
5	3.5	1.7	10	12.61	29.81	0.775
5	3.5	3.4	10	25.22	59.61	0.775
5	3.5	8.0	10	59.34	140.27	0.775
5	3.5	1.7	1.5	43.58	8.15	0.996
5	3.5	1.7	10	6.54	8.15	0.996
5	3.5	1.7	35	1.87	8.15	0.996

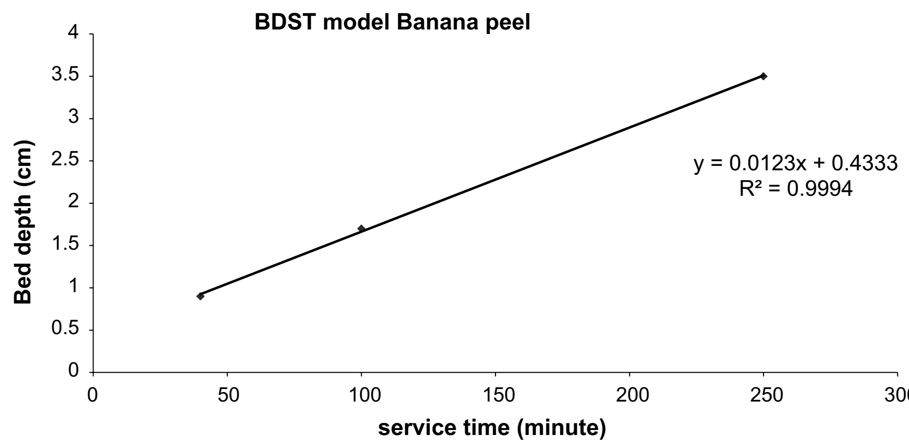


Fig. 8 BDST model at breakthrough curve in fixed-bed column for F adsorption by banana peel

Table 4 Calculated constants of BDST model for the adsorption of fluoride by banana peel dust using linear regression analysis

C_t/C_0	a (min/cm)	b (min)	K_a (l/mg/min)	N_0 (mg/l)	R^2
0.0101	0.0123	0.4333	0.005	0.4182	0.9994

$C_0 = 10.0$ mg/L, $v = 3.4$ ml/min

Table 5 Column regeneration parameters for NBP column

Regeneration cycle	Percentage of desorption Q_E (mg/g) (%)	E_D (%)
1	84	2.091
2	72	1.321
3	54	0.694

Adsorption operation constant–initial fluoride concentration: 8 ml/min; $Z = 0.9$ cm; pH = 5; $C_0 = 10$ mg/L. Desorption condition—desorption solution 0.1 M NaOH, solution flow rate 2 ml/min

Modeling of breakthrough curve

The entire experimental column study has been fitted with Thomas model, results revealed that all the operating variables are well acquainted with Thomas model, and goodness of the fit of the model (R^2) is very high except variation of flow rate (Table 3). A plot of service time (t) versus bed depth (X) at a flow rate of 3.4 mL/min is shown in Fig. 8. The equation of the linear relationship was obtained with R^2 above 0.999. This indicated the validity of the BDST model for present column system. The calculated adsorption capacity (N_0) and the rate constant (K) are 0.418 mg/L and 5×10^{-3} L/mg/min, respectively (Table 4). The advantage of the BDST model is that any experimental test can be reliably scaled up to other flow rates without further experimental runs.

Regeneration study

After regeneration of the exhausted column, it has been found that desorption gradually decreases from first to second and second to third cycles. The calculated column desorption parameters are listed in Table 5. From Table 5, it is clear that E_D value decreases with consecutive cycles of desorption (Tor et al. 2009). This is probably due to the loss of adsorption capability of banana peel dust column.

Defluoridation from field sample

Forty three (43) tube well water samples were collected from the severally fluoride affected villages of Birbhum district, West Bengal. The children aged between 5 and 10 years were selected for understanding the status of dental and skeletal fluorosis (Fig. 9). From the data (data not shown), it is clear that about 63% boys and 48% girls were severally affected by dental fluorosis (Fig. 9). However, 18% boys and 12% of girls were affected by skeletal fluorosis (Fig. 9). The physico-chemical analysis of the field samples is presented in Table 6. From Table 6 it is found that the highest fluoride level in the affected areas is 9.89 mg/L. Moreover, for field sample defluoridation study, highest fluoride containing tube well water (9.89 mg/L) was selected and it was recorded that about 86.5% removal can be achieved through standardized column study ($v = 3.4$ mL/min, $C_0 = 9.89$, pH = 5, $Z = 3.7$ cm).

Table 6 Physico-chemical characteristics of ground water

Parameters	Values
Conductivity (μS/cm)	113 ± 2.07
Hardness (mg/L, $CaCO_3$)	178 ± 3.11
Chlorine (mg/L)	12.3 ± 0.71
Nitrate (mg/L)	9.3 ± 0.06
Sulfate (mg/L)	0.81 ± 0.013
Fluoride (mg/L)	9.89 ± 0.33
pH	7.9

Mean ± SD

Conclusions

Present finding highlights the removal of fluoride from both synthetic and field samples using banana peel dust through column study. The surface morphology of the banana peel dust was assessed by SEM study, and the active functional group was evaluated by FTIR study (Table 7). Fluoride adsorption results highlighted that the adsorption capacity of banana peel increased with decreasing medium pH. Column study revealed the best fluoride removal conditions as: 10 mg/L initial fluoride concentration; flow rate 3.4 ml/min, bed depth 3.5 and pH 5. The study results were best fitted with both Thomas and BDST models. Moreover, Thomas equation indicated that the increase in bed height caused an increase in the mass transport resistance and axial dispersion, which was confirmed from the K_{Th} values. The exhausted banana peel dust can be regenerated by 0.1 M NaOH and reused with minimal loss of efficiency for three adsorption–desorption cycles. Finally, best optimized conditions were applied for defluoridation of the field sample, and 86.5% removal was achieved. Therefore, it can be conclude that the use of banana peel as an adsorbent for fluoride removal is potentially cost-effective and may provide an alternative method for fluoride removal from contaminated water.

Fig. 9 Children affected by skletal and dental fluorosis in the study area

Table 7 Comparative evaluation of adsorption capacities of various biosorbents for fluoride removal

Adsorbent	pH	Adsorption capacity (mg/g)	References
1. Acid and alkali treated *Tamarindous indica* fruit shall	2–10	–	Ramanjaneyulu et al. (2013)
2. Acid treated Banana peel	2–12	1.34	Mohammad and Majumder (2014)
3. Untreated Tamarind fruit corner	1–8	4.14	Sivasankar et al. (2012)
4. Mousabi (*Citrus limetta*) peel	7.0	1.915	Singh and Majumder (2015)
5. Ground nut (*Arachishypogaea*)	7.0	1.15	Singh and Majumder (2015)
6. Untreated sweet lemon peel	2–12	0.744	Mohammad and Majumder (2014)
7. Untreated Banana peel (BPD-1)	2–10	17.43	Bhaumik and Mondal (2014b)
8. Algal Biosorbent *Spirogyra* sp.	2.0	1.272	Mohan et al. (2007)
9. Leaf biomass powder sample	2.0		Jamode et al.(2004)
10. Treated coffee husk	2.0	0.416	Getachew et al. (2015)
11. Guava seeds	–	116.5	Sánchez-Sánchez et al. (2013)
12. Banana peel	5.0	8.15	Present work

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Compliance with ethical standards

Conflict of interest Authors declare that they have no conflict of interest for publishing the present manuscript.

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