2014

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Research Journal in Engineering and Applied Sciences

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Article *in* Research Journal of Applied Sciences, Engineering and Technology · July 2014

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Rjeas Research Journal in Engineering and Applied Sciences 3(3) 167-174 Rjeas
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PLANT BIOMASSES FOR DEFLUORIDATION APPROPRIATENESS: UNLOCKING THEIR POTENTIALS

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ABSTRACT

Fluoride and fluorosis are endemic to many countries including Tanzania. Performances of different materials for use in water defluoridation systems have been reported. Some of these materials are; alum, oxides and hydroxide of metals, activated carbon, bone char and plant biomasses. This paper reviews and discusses the performances of selected defluoridation materials such as alum in Nalgonda technique, the oxides and hydroxides of metals (inorganic adsorbents) in ion exchange/adsorption, activated carbon, bone char and plant biomasses in ion exchange/adsorption. More discussion is on the strengths and limitations of these materials in removing fluoride from water. Furthermore, it describes a new approach that will likely enhance the fluoride removal capacity when plant biomasses are used, which involves special arrangement of different plant biomasses in a column. This promises to be of low cost and high performance and thus suitable for both urban and rural communities in developing countries.

KEYWORDS: defluoridation, fluorosis, activated carbon, bone char and plant biomas

INTRODUCTION

Water related problems continue to afflict the human population the world over (Goel and Kaur, 2012; Serena *et al*., 2012; Barrett, 2014). Most of these afflictions are associated with contaminants of water, which extends from microbial to chemical and being both natural and anthropogenic with their effects ranging from crippling to death (Schwarzenbach *et al*., 2006). The chemical contaminants include but not limited to mercury, arsenic, lead, cyanide, fluoride and sulphate ions.

Fluoride can also be found in soil, plants and animals in its different speciation (Fawell *et al*., 2006). Fluoride and fluoride related problems are endemic to a number of regions and Tanzania is not an exception, with about 30% of its water having fluoride levels above WHO guideline of 1.5mg/l. The presence of fluoride in water is a mixed blessing, with lower concentrations being beneficial to man but chronic exposure to higher concentrations causing fluorosis (Crinchton, 2008).

Thus, no wonder that both researchers and a number of world organisations are concerned about it (Fawell *et al*., 2006; Barrett, 2014). Several defluoridation materials and corresponding techniques, have been investigated worldwide for their fluoride removal performances, including but not limited to alum in Nalgonda technique, oxides and hydroxides of metals, activated carbon, bone char and plant biomass in adsorption and ion exchange (Fawell *et al*.,2006; Tomar and Kumar, 2013; Modi and Soni, 2013).

The contamination drinking water with fluoride poses a health risk to people in fluorotic areas. Fawell et al. (2006) showed that, over 70 million people are affected by fluorosis earth wide. The conventional defluoridation materials are expensive, non-sustainable or environmental unfriendly (Jamode et al., 2004). It is therefore important to investigate defluoridation by sustainable, cheap and environmental friendly materials such as plant biomasses.

Fairly, a good number of studies have been done to establish the fluoride removal capacities of different materials (Fawell *et al*., 2006; Loganathan *et al.,* 2013; Patil and Ingole, 2012; Bhatnagar *et al*., 2011; Tomar and Kumar, 2013). However, defluoridation by plant biomass has caught interest of researchers only recently, and reviews that include an in-depth description of fluoride removal capacities of plant biomasses are scanty. This paper therefore, reviews the fluoride removal characteristics of selected materials, and compares them with plant biomasses. It highlights research deficits in light of the available knowledge for the selected materials. Finally, it presents a new approach that involves special arrangement of plant biomasses in a column in light of their established optimal pH requirements and their respective influences on the treated water. This approach promises to do both cost lowering and fluoride removal enhancement.

Defluoridation by Coagulants/Precipitators

Coagulation or precipitation involves mainly the clotting or trapping and settling of the fluoride ion from water. This can be done by using either natural coagulants such as *Moringa* seeds extract which trap fluoride from water by using its long chain polymers (Vardhan and Karthkeyan, 2011) or synthetic coagulants such as salts of Mg^{2+}, Ca^{2+} or Al^{3+} (Shrivastava and Vani, 2009). At optimal dose, fluoride removal by *Moringa* seed extract is pH dependent, such that it is 75% at pH 3 and 89% at pH 6. (Vardhan and Karthkeyan, 2011). This implies that *Moringa* seed extract can be used at around neutral pH. Thus, pre- or post-treatment of water for pH regulation is not required. However, the disadvantage associated with this coagulant is that, the optimal dose is 1000mg/l of *Moringa* seed extract for water whose initial fluoride concentration is 5 mg/l (Vardhan and Karthkeyan, 2011). This therefore, requires a huge supply of *Moringa* seeds, and could lead into the production of huge amount of sludge.

The Magnesium, calcium and aluminium ions form fluoride compounds that are insoluble in water (Sharpe, 1992). Actually, these ions can bind preferentially with fluoride in solution (Shriver et al., 1994). These characteristics make these ions potential cationic coagulants. In the Nalgonda technique, alum and lime are added to effect precipitation of fluoride (Indian Standard [IS], 1989). The alum once added into water to be treated, hydrolyses to form polymeric compounds (polyhydroxo alumino complexes) which are responsible for trapping and settling fluoride ion from water (IS, 1989). The challenge with this technique is that, the use of alums can generate toxic fluoro-alumino complexes (Meenakishi and Maheshwari, 2006; Modi and Soni, 2013) which are accused of causing Alzheimer's disease (Modi and Soni, 2013). Experience has also shown that stirring speed affects the flock formation characteristics of the alums (Susheela, 1992), a phenomenon that would need either automated stirrer or informed operator (Modi and Soni, 2013). This is a challenge especially for point of use treatment in rural areas. The hydrolysis of alum is enhanced by alkaline medium (Susheela, 1992; Modi and Soni, 2013), therefore, lime is added to ensure optimal alkalinity for maximum hydrolysis and settling (Susheela, 1992; Modi and Soni, 2013). This means that the treated water will have elevated pH and may require post-treatment pH regulation before use. Due to the need for stoichiometric balance, coagulation or precipitation, regardless of the type of coagulant, would call for repeated pre-determination of the fluoride concentration in the water before treatment (Modi and Soni, 2013). Futhermore, production of sludge presents a disposal problem (Dahi *et al*., 1996; Mjengera and Mkongo, 2003 Tewari and Dubey, 2009; Shrivastava and Vani, 2009; Modi and Soni, 2013). This poses a challenge to users of the technique as the concentration of fluoride in water sources tend to vary seasonally. Moreover, the Al and Ca ion as used in the Nalgonda technique, cannot lower the level of fluoride in water to permissible levels set by WHO of 1.5mg/l, unless excessive amount of alum is used (Dahi *et al*., 1996).

Defluoridation by Oxides and Hydroxides of Metals

Inorganic adsorbents identified for removal of fluoride from water include exchange resins, inorganic oxides or hydroxides and natural rock adsorbents (Bhatnagar *et al*., 2011; Loganathan *et al*., 2013). The hydroxides of metals have shown selective affinity for fluoride ion in aqueous media (Shrivastava and Vani, 2009; Maliyekkal *et al*., 2010). These hydroxides remove fluoride from water by anion exchange process in which hydroxyl ion is exchanged for fluoride (Maliyekkal *et al* 2010). This will thus alter the pH of treated water and therefore may require post-treatment pH regulation of the treated water. One of the most studied inorganic adsorbents for fluoride removal is alumina. However, when alumina is saturated, the adsorbed fluoride is released back into the treated water (Veressinina *et al*., 2001; Renuka and Pushpanjali, 2013; Modi and Soni, 2013). Activated alumina works under specific pH range hence pre-and or post-treament pH regulation would be necessary (Renuka and Pushpanjali, 2013). The availability and cost of inorganic oxides and hydroxides makes these adsorbents not the sustainable means for fluoride removal from water (Loganathan *et al*., 2013) especially so in the developing countries. Natural rock materials such as pumice and bauxite have also shown good fluoride adsorption (Sajidu *et al*., 2008; Malakootian *et al*., 2011). Although they may be locally available, they require a rigorous processing before use to avoid contamination from the original rock (Sajidu *et al*., 2008). This calls for specialized machinery and personnel.

Defluoridation of Water By Activated Carbon

Recently, the term activated carbon or charcoal have been used to mean activated charcoal prepared from plant biomass. However, in a general sense, activated carbon can be taken to mean charcoal prepared from plant biomass, animal bones, petroleum residues, and coal (Patil and Ingole, 2012). The fluoride removal power of activated carbon is attributed mainly to the presence and distribution of the pores. This is because the adsorption of fluoride takes place in the pores that are charged. Nevertheless, some fluoride ions are adsorbed on the surface of the activated carbon where they form carbon-fluoride bond observable by FTIR (Hanumantharao et al., 2004). The fluoride removal capacity and optimal pH appears to be influenced by the plant type (Janaradhan *et al*., 2007; Chakrapani *et al*., 2010).

Although prepared at the same preparation conditions, groundnut shell and coconut shell activated carbon showed different fluoride removal capacity (Janaradhan *et al*., 2007). On the other hand, activated carbon from peels of three different plant species of citrus showed different fluoride removal capacity irrespective of the same preparation and operation conditions (Chakrapani *et al*., 2010). pH is yet another factor which seriously affect performance of activated carbon. Neem stem charcoal for example works best at pH 5 with fluoride removal capacity of up to 94% (Chakrabarty and Sarma, 2012). Tembhukar and Dongre (2006) observed a maximum removal of 85% at pH 2 for plant based activated carbon. Although, most activated plant charcoal has shown good fluoride removal capacity from aqueous solution, the optimum pH required is acidic (Tembhukar and Dongre, 2006; Vardhan and Karthkeyan, 2011; Chakrabarty and Sarma, 2012) thus, there is need for pre-treatment and post-treatment of water for pH regulation. This therefore, poses an additional challenge to users of activated charcoal especially in areas remote from trained personnel.

Defluoridation by Bone Char

Bone char is the charcoal prepared by heating animal bones at controlled temperature, heating duration and amount of oxygen. Bone char has been used as the defluoridation tool for several decades since 1940s (Fawell *et al*., 2006). Although bone char is old, its use in water treatment plants in developed countries is obsolete (Fawell *et al*., 2006). It has good defluoridation capacity even at neutral pH of up to 60% (Renuka and Pushpanjali, 2013). The fluoride removal capacity of bone char is attributed to the presence of hydroxyapatite in its structure (Fawell *et al*., 2006). When fluoride ion reacts with hydroxyapatite, fluorapatite and hydroxyl ions are formed (McCann, 1953; Kaseva, 2006) according to the reaction;

$$Ca_{10}(PO_4)_6(OH)_2 + 2F^- \rightarrow Ca_{10}(PO_4)_6F_2 + 2OH^- \qquad (1)$$

Defluoridation by use of bone char is currently the focus by the ministry of water in Tanzania through its research station called Ngurudoto Defluoridation Research Station (Mjengera and Mkongo, 2003). However, major limitations associated with the use of bone char are; bacteria harbouring characteristics, possible natural and religious objections (Renuka and Pushpanjali, 2013) and unavailability of commercially distributed products (Fawell *et al*., 2006). In as much as the defluoridation of water is cost effective when done at the point of use (Fawell *et al*., 2006) as this will prevent unnecessary misuse of treated water, limitations associated with bone char pose a threat to acceptability and sustainability of the material. Jacobsen and Dahi (1998) observed that, unavailability of commercially distributed bone char could be addressed by preparing bone char at village or household level. Charing at temperatures above 550^{o}C and allowing more oxygen lowers the fluoride adsorptive capacity of bone char (Fawell *et al*., 2006; Albertus *et al*., 2000; Puangpinyo and Osiriphan, 1997). On the other hand, bone char prepared at temperatures below 550^{o}C can produce odour, yellow colour and or offensive taste in the treated water (Puangpinyo and Osiriphan, 1997; Fawell *et al*., 2006; Jacobsen and Dahi, 1998). In fact, the optimal heating temperatures and duration in production of bone char is yet to be fixed (Fawell *et al*., 2006). This suggests that preparation of bone char at the household level or village level should be accompanied with training for those who will be involved. Likewise, since there is likelihood of the source water to be bacterial contaminated the bone char can serve as bacterial culturing medium, which will eventually make the treated water, require a serious disinfection.

Defluoridation by Plant Biomass

Plant biomass is the dry matter of the plant roots, stem or leaves. Essential preparation procedure for plant biomass involves washing, drying and grinding. The mechanism by which plant biomass removes fluoride ions from the water is not well established, though the affinity of plant biomass for fluoride ion is attributed to the presence of Ca, Mg, hydroxyl and amine groups in the biomass structure (Pandey *et al*., 2012; Vardhan and Karthkeyan, 2011; Bhatnagar *et al*., 2011; Harikumar *et al*., 2012). Interest in defluoridation by plant-based biomass is growing due to their lower cost and easy availability (Yadav *et al*., 2013). Ability of plants to survive in a wide range of environmental conditions coupled with cheap preparation procedures promises availability of this adsorbent even to people in rural areas. Different studies on defluoridation capacity of plant biomass reveal that these materials are potential for use in defluoridation of drinking water (Malde *et al*., 2006; Kumar *et al*., 2012; Balouch *et al*., 2013; Vardhan and Karthkeyan, 2011; Harikumar *et al*., 2012).

Different plant materials have different defluoridation capacity and operation conditions. For instance Malde *et al*. (2006) found that tea leaves could remove fluoride from aqueous 20 mg/l fluoride solution. This removal was possible irrespective of the fluoride content of the tea leaves biomass (Malde *et al*., 2006). Tamarind (*Tamarindus indica*) fruit cover powder treated with HCl, can remove up to 57.1% of fluoride from 3.5mg/l F natural water at the pH of 7.6 (Kumar *et al*., 2012). This fluoride removal capacity was enhanced by treating the biomass with HCl acid, which increased porosity of the biomass (Kumar *et al*., 2012). The defluoridation capacity of tamarind fruit cover biomass was impaired by the presence of carbonate and hydrogencarbonate ions more significantly than the presence of chloride and sulphate ions (Kumar *et al*., 2012). This implies that removal of fluoride from water by tamarind fruit cover biomass follows certain specific removal mechanisms. Balouch *et al*. (2013) reported the removal capacity of sawdust at 25°C to be maximal at neutral pH. The adsorption of fluoride by sawdust was observed to be exothermic and spontaneous (Balouch *et al*., 2013) which implies that the biomass has some special affinity for fluoride ion.

A comparative study of defluoridation capacities of different plant biomass (Table 1), in removing fluoride from a solution, whose fluoride content is 2mg/l, showed that, biomass from vetiver (*Vetiveria zizanoides*) roots, tamarind seed (*Tamarindus indica*), clove (*Eugenia carryophyllata*), neem (*Azardirachta indica*), acacia(*Acacia catechu willd*), nutmeg (*Myristica fragrans*) and coffee husk (*Coffea arabica*), could remove up to 80%, 75%, 70%, 52%, 47%, 45% and 38% of fluoride respectively, at neutral pH (Harikumar *et al*., 2012). This suggests that, not all plants biomasses have the same chemical structure and composition. Yadav *et al*. (2013) observed that sawdust of *Dalbergia sissoo* and wheat straw removed 49.8% and 40.2% of fluoride respectively from a solution of 5mgF/l at pH of 6. This removal was achieved at the dose of 4g of biomass per litre of water. Rice husks are yet another biomass that can remove up to 84% of fluoride from water (Vardhan and Karthkeyan, 2011). This removal was observed in both batch and column experiments. Pandey *et al*. (2013) observed that biomass of *Tinospora cordifolia* could remove up to 70% of fluoride from water whose fluoride content is 5mg/l at the dose of 7g/50ml of water at pH of 7. They also observed that biomass did not change the chemical nature of water. One thing that all plant biomasses have in common is that most of the plant biomasses work at around neutral pH, (Kumar *et al*., 2012; Balouch *et al*., 2013; Harikumar *et al*., 2012; Yadav *et al*., 2013; Renuka and Pushpanjali, 2013; Pandey *et al*., 2012) thus, there will be no need for pH regulation of water before or after treatment. Another thing, which is clear from these findings, is that different plant biomasses have different fluoride removal capacities (Harikumar *et al*., 2012) (see Table 1). These differences in fluoride removal capacity could be due to different chemical structures and composition of the different plant biomasses. However, to obtain facts about this, more studies on plant biomasses, as defluoridation means is required.

Table1: Some Plant Biomass and their Fluoride Removal Efficiencies

S/N	Biomass type	Optimal pH	Initial F^- concentration	Biomass dose (g/ml of water)	Removal efficiency	Reference(s)
1	*Tinospora cordifolia*	7	5mg/l	0.14g/ml	70%	Pandey *et al*., 2013
2	*Dalbergia sissoo*	6	5mg/l	0.004g/ml	49.8%	Yadav *et al*., 2013
3	Wheat straw	6	5mg/l	0.004g/ml	40.2%	Yadav *et al*., 2013
4	Tamarind fruit cover	6	10mg/l 2mg/l	0.7g/ml 1g/ml	>70% 75%	Kumar *et al*., 2012 Harikumar *et al*., 2012
5	Vetiveria zizanoides roots	neutral	2mg/l	1g/ml	80%	Harikumar *et al*., 2012
6	Neem	neutral	2mg/l	1g/ml	52%	Harikumar *et al*., 2012
7	Clove	neutral	2mg/l	1g/ml	70%	Harikumar *et al*., 2012
8	Acacia	neutral	2mg/l	1g/ml	47%	Harikumar *et al*., 2012
9	Rice husks	2-7	5mg/l	0.006g/ml	83-80%	Vardhan and Karthkeyan, 2011

The benefit of plant biomass as fluoride adsorbent extend from low production cost, simplicity of the preparation procedures and ultimate use to easy disposal as the material is biodegradable. Furthermore, most plant biomasses work best at around neutral pH, which means no need for pre- and or post-treatment pH regulation of the treated water.

The functional groups associated with fluoride removal capacities of plant biomasses are; Ca, Mg, -OH, and $-NH_2$ (Pandey *et al*., 2012; Vardhan and Karthkeyan, 2011; Bhatnagar *et al.*, 2011; Harikumar *et al*., 2012; Kumar *et al*., 2012). These groups could be responsible for van der Waals forces, hydrogen bonding, substitution, and columbic interactions between the biomass and the fluoride ion (Pandey *et al*., 2012; Loganathan *et al*., 2013; Harikumar *et al*., 2012) which in turn is responsible for biomass affinity for fluoride. However, synthetic ion exchangers that use OH and NH_2 as functional groups for exchange work optimally under acidic conditions (Sundaram and Meenakshi, 2009), contrary to the plant biomasses, most of which work optimally at around neutral pH (Harikumar *et al*., 2012; Pandey *et al*., 2012). The relationship between biomass functional groups and the fluoride removal capacity of plant biomass is worthy more study. Thus, the mechanisms by which functional groups are involved in fluoride adsorption needs to be extensively studied. However, the fluoride removal capacity of plant biomass is relatively lower when compared to other adsorbents such as bone char. Furthermore, some biomasses impart colour and or odour to the treated water and are prone to decomposition upon prolonged interaction with water. Nevertheless, colour and odour problems can be addressed by selecting appropriate plant materials and or washing the material prior to use. Some studies have revealed that, treatment of the plant biomass with acids increases their fluoride removal capacity (Kumar *et* *al*., 2012). Alternatively, a special arrangement of plant biomasses in the column, which exploits the variation of the optimal pH of the different plant biomasses, could enhance their removal capacities.

CONCLUSION AND FUTURE PERSPECTIVES

There are several materials used for defluoridation of water, some of which are; alum, alumina, activated carbon, bone char and plant biomass. All these materials present both advantages and disadvantages. The application of alum requires a large amount for defluoridation with consequent production of large volumes of sludge and alumina complexes that are accused of causing Alzheimer's disease. The alumina works best at narrow and acidic pH ranges, which will necessitate pH regulation of the treated water. Although, bone char has good fluoride removal capacity, its condition-sensitive preparation requirements demand that knowledgeable personnel are involved in its preparation. However, different plant biomasses influence differently on the pH of the treated water.

This can hinder wide application of the materials especially in remote areas. Therefore, plant biomasses remain to be more promising alternative defluoridation materials for further studies to unveil their potentials. Therefore, further studies are required to establish the fluoride removal mechanism of plant biomasses and capacity enhancement. One of the possible directions towards addressing the influence of plant biomasses on the pH of treated water is to couple different plant biomasses in column set up according to their influence. When adsorbents are arranged in the column, the adsorption capacity is enhanced (Fawell *et al*., 2006). This can be exploited to enhance the fluoride removal capacity by plant biomass. As proved by different studies, different biomasses have different optimal pH at which fluoride removal is maximal as shown in Table 1.

Preliminary results from the current work by authors to enhance the fluoride removal capacity of individual and combined plant biomasses has observed that; whereas some plant biomasses do not alter the pH of the treated water, others raise or lower the pH as shown in Fig.1. From Fig. 1, the Banana leaves works best at around pH 7.8, the sisal leaves at around 6.2 while the goose grass works best at around pH 5.4. This can be attributed to the affinity of the plant biomasses to ions other than fluoride (Pandey *et al*., 2012; Fallico *et al*., 2010; Schaeffer *et al*., 2012).

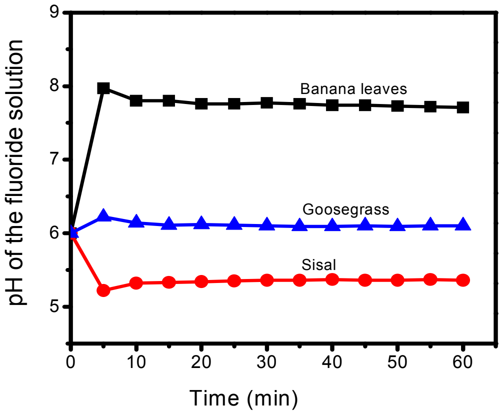

Figure 1: The graph showing the effect of different biomasses on pH of the treated water

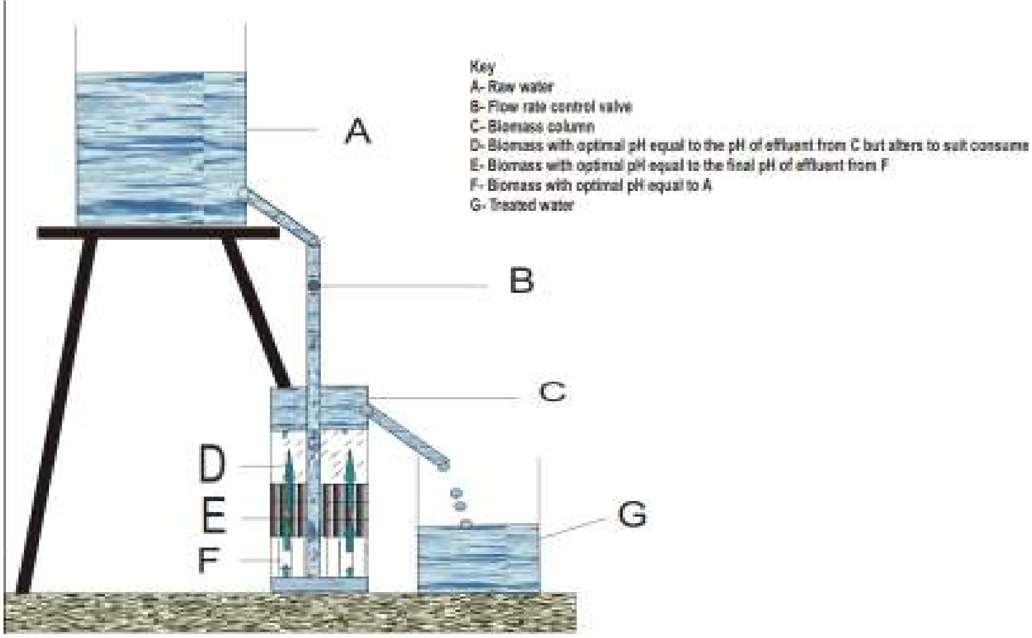

Fig. 2: Arrangement of biomasses in a column for defluoridation enhancement and pH regulation

Our current study combines different plant biomasses in a column to enhance fluoride removal capacity and maintain the neutral pH of the treated water as shown in Fig 2. As fluoride water traverses through the biomass column, its pH changes as depicted in fig 1. Since biomasses have certain pH at which fluoride removal is maximal, this change in pH in the column can somehow lower the performance of the column. Thus, column arrangement of biomasses as shown in fig. 1, which places different biomasses such that the next biomass in the column is one whose optimal pH equals the pH of the effluent of the previous could enable the column to perform at its best.

This special arrangement of plant biomasses in a column promises to enhance fluoride removal of the respective biomasses as it automatically provides an optimal condition for maximum performance of each individual biomass at the same time self-adjust the pH of the treated water. The new approach that involves differential packing of plant biomasses deserves more study in order to unlock its potential regarding fluoride removal, thereby providing cheap, sustainable and widely acceptable technology for defluoridation of water for both rural and urban communities in developing countries. However, the success of this approach is dependent on type and behaviour of the biomasses as some of the biomasses have water-soluble constituent that can render treated water toxic. Thus, database of appropriate biomasses is a prerequisite in making this approach a success.

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