

In-Situ and Ex-Situ Management and Mitigation Techniques of Fluoride in Drinking Water: A Systematic Review from Purulia District of West Bengal, India

Dr.Sumana Bhattacharjee

Assistant Professor, Department of Geography (Jogesh Chandra Chaudhuri College), University of Calcutta

To Cite this Article

Dr.Sumana Bhattacharjee, "In-Situ and Ex-Situ Management and Mitigation Techniques of Fluoride in Drinking Water: A Systematic Review from Purulia District of West Bengal, India", *International Journal for Modern Trends in Science and Technology*, Vol. 04, Issue 08, August 2018, pp.-83-88.

ABSTRACT

Fluoride is a colourless micro-nutrient which is very much essential for the strong teeth and bones. Beyond the prescribed range (0.6–1.5 mg/l) of fluoride in drinking water is harmful or hazardous for human health. Granite gneiss and pegmatite are the natural sources of fluoride minerals in this North Singhbhum Craton. Purulia district which is the extended part of Chhota Nagpur Gneissic complex hosts such fluoride bearing rocks and minerals. Fluorosis associated health hazards now become an emerging problem in India as well as in World. The main objective of this scientific paper is to review and explain the in- situ and ex- situ fluoride management techniques in drinking water. To combat fluorosis cost-effective, eco-friendly and easily accessible scientific techniques and suitable planning must be executed in different blocks of Purulia according to the preliminary intensity of fluoride, its origin and socio-economic condition of rural people. Scientific perception and rational strategies can help to overcome the devastating problem of fluoride contamination in Purulia.

Keywords: Fluoride; In-situ and ex-situ management techniques; Chhota Nagpur Gneissic complex

Copyright © 2018 International Journal for Modern Trends in Science and Technology
All rights reserved.

I. INTRODUCTION

Fluorosis related health hazards now become an emerging problem in India as well as in World. In 1984, World Health Organization estimated that more than 260 million people are consuming fluoride contaminated groundwater where the concentration level is above 1 mg/l. The residents of approximately 29 countries have been suffered due to fluoride contamination in drinking water (Bhattacharya and Chakrabarti, 2011). Fluorosis is a very common disease in U.S.A, Morocco, Algeria, Libya, Turkey, Australia, Canada, Japan, Sri Lanka etc. (Amini et al., 2008). The excessive

concentration of fluoride in groundwater has been identified in different pockets of India. Nearly 65 million people in India are facing the tremendous problem of dental and skeletal fluorosis (Susheela, 1999). Most of the Indians depend on groundwater for drinking purpose since historical time (Phansalkar et al., 2005). But in recent decades, the quality of groundwater is being degraded due to several natural and anthropogenic processes (Jalali, 2005; Srinivasamoorthy et al., 2011). So it is necessary to improve the quality of ground water for the sake of mankind. Among the seven fluorides affected districts of West Bengal, Purulia holds a remarkable position in this context. The high level of Fluoride contamination imposes deadly impact

in different areas of Purulia. The suitable management techniques must be implemented to reduce the fluoride intensity and negative impacts of fluorosis hazards. Generally, identification of substitute sources of purified drinking water, implementation of different defluoridation techniques, upliftment of nutritional scenario of vulnerable people are some basic preventive measures which have been adopted to check the devastating impacts of fluorosis in various regions of developing countries like India (Bhattacharya and Samal, 2018). The remedial methods are broadly classified into two categories – i) In-situ Method and ii) Ex-situ Method. In-situ method helps to reduce the intensity of fluoride in groundwater directly through artificial recharge system (Brindha and Elango, 2011). Proper application of different types of In-situ methods can provide fluoride-free drinking water to the local people of different blocks in Purulia district. But the effectiveness of these methods is varied in different regions (Bhattacharya and Chakrabarti, 2011). Ex-situ method can remove fluoride ions from water applying through different techniques of defluoridation (Brindha and Elango, 2011). There are different types of defluoridation techniques like physical, chemical and biological technique (Annadurai et al., 2014). The defluoridation techniques include different processes like Adsorption (Raichur and Basu, 2001), Precipitation-Coagulation (Saha, 1993; Reardon and Wang, 2000), Ion-exchange (Singh et al., 1999), Electrodialysis (Hichour et al., 1999; Hichour et al., 2000; Adhikari et al., 1989), Electrolytic Defluoridation (Mameri et al., 2001), Membrane Separation (Amer et al., 2001; Dieye et al., 1998). The most effective processes of defluoridation are Precipitation and Adsorption process (Hussain et al., 2012). The defluoridation techniques play a significant role to mitigate the problem of extreme fluoride concentration in drinking water where of the In-situ methods are not reliable with respect to above mentioned techniques (Meenakshi and Maheshwari, 2006). Nalgonda method is based on the principle of precipitation process which is mostly used in India to remove excessive fluoride from the drinking water (Hussain et al., 2012). Adsorption process is also very popular for its cost-effectiveness and immense user-friendliness (Annadurai et al., 2014). Beside this, Biosorption is an innovative method to purify water with the help of biotic ingredients (Annadurai et al., 2014). Chitosan, chitin, fungal and algal biomass are very useful

biosorbents which act as fluoride remover. In addition with In-situ and Ex-situ method, community involvement, development of nutritional status, development of awareness and expansion of health and effective education are highly crucial to avert the curse of fluoride contamination in Purulia (Hussain et al., 2012; Sreedevi et al., 2006; Meenakshi and Maheshwari, 2006; Bhattacharya and Chakrabarti, 2011).

II. IN-SITU AND EX-SITU METHODS

2.1. In-Situ Method

In-situ methods include different substitute sources of water like rainwater, surface water etc. which ensure the supply of fluoride free drinking water to the local people (Meenakshi and Maheshwari, 2006). In this context, construction of check dams can play a significant role to reduce the amount of fluoride in groundwater (Bhagavan and Raghu, 2005). Proper hydro-geological and geomorphological investigations are required for the construction of dams /check dams. So, proper site selection is the utmost important issue to construct dams / check dams. Another technique is to provide fluoride safe surface water that is rainwater harvesting (Hussain et al., 2012). Construction of recharge pits and percolation tanks can be useful for proper functioning of rainwater harvesting technique and not only that, percolation of rain water through the present wells can help to diminish the level of fluoride in ground water (Brindha and Elango, 2011). Adequate municipal water supply should be ensured in fluoride affected regions (Ravindra and Garg, 2006). Furthermore, different projects to provide piped water from lake, river, large tube well, and large bore tube wells with fluoride reduction units and construct deep tube wells should be given importance to increase the availability of fluoride free water to the local people of Purulia (Bhattacharya and Chakrabarti, 2011).

The main demerit of In-situ method is the high primary cost of development of piped water supply system (Hussain et al., 2012). Not only that, use of unpurified surface water for drinking purpose may be harmful for human health (Meenakshi and Maheshwari, 2006). Although rainwater harvesting is a very cost-effective and simplified technique (Meenakshi and Maheshwari, 2006) but scanty and uneven rainfall in different pockets of Purulia may reduce the usefulness of this technique.

2.2. Ex-Situ Method

Several Ex-situ methods can be applied to remove excessive fluoride from water. Ex-situ methods are

mainly based on scientific principles and various chemical ingredients. These defluoridation processes like precipitation, Adsorption, Ion-exchange, Membrane process are very effective to prevent the deadly problem of fluoride contamination in different parts of Rarh Bengal.

2.2.1. Adsorption

In adsorption process, excessive fluoride is captivated upon the matrix during the flow of water passes within a contact bed (Brindha and Elango, 2011). Various adsorbent are used in this process like activated carbon, activated alumina, activated saw dust, fly ash, calcite, activated alumina coated silica gel, bone charcoal, carbion, serpentine, groundnut shell, coconut shell carbon, rice and coffee husk, tricalcium phosphate, defluoron-1, defluoron-2, activated soil sorbent etc. (Kariyanna, 1987; Barbier⁰ and Mazounie, 1984; Muthukumaran et al., 1995; Rongshu et al., 1995; Min et al., 1999; Wang and Reardon, 2001; Nava et al., 2003; Padmavathy et al., 2003; Thergaonkar and Nawalakhe, 1971). But activated alumina and activated carbon are the mostly used adsorbents (Chidambaram et al., 2003; Chauhan et al., 2007). The efficacy of adsorption process is determined by hardness, pH, nature of adsorbent, preliminary amount of fluoride, duration of contact (Meenakshi and Maheswari, 2006; Brindha and Elango, 2011). In this process, pH level must be remain within 5-6 because when pH level decreases below 5, activated alumina is disappeared due to acidic effect whereas when pH level increases above 7, silicate and hydroxide play active role to replace the fluoride upon activated alumina (Bishop and Sansoucy, 1978). Activated carbon acts as a very good absorbent but it becomes active when pH level is 3 or less than 3 (Mckee and Johnston, 1999; Meenakshi and Maheswari, 2006). The main limitations of adsorption process are weak integration capacity, regular pre-treatment, gradual decrease of efficiency after regeneration, poor adsorption capability etc. In spite of these demerits, this method is able to dilute fluoride upto 90% and this method can be implemented in different villages because of its low cost (Meenakshi and Maheswari, 2006).

2.2.2. Precipitation-Coagulation

In precipitation-coagulation process, lime and alum are most commonly used ingredients which help to precipitate excessive fluoride (Meenakshi and Maheswari, 2006). At the first phase, lime plays a crucial role in precipitation process and at the second phase, alum is mixed to occur coagulation and reaction between alum and

fluoride removes excess fluoride concentration (Meenakshi and Maheswari, 2006). Nalgonda technique is considered as the most efficient and accessible technique of defluoridation and it can be widely used in different villages of India (Meenakshi and Maheswari, 2006; Hussain et al., 2012). National Environmental Engineering Research Institute had propounded this technique in India (Brindha and Elango, 2011).

Although it is a low cost technique, but the major constraints of this method are creation of huge amount of sludge and its scientific disposal, ignorance about the actual dose of chemicals, inefficacy to remove fluoride during the presence of huge total dissolved solid and excessive hardness in water (Apparao et al., 1990). Nalgonda technique is only used when the concentration of fluoride in water is less than 10 mg/litre (Bhattacharya and Samal, 2018). It is remarkable that Nalgonda technique has the capacity to remove only 18%-33% fluoride from water by precipitation and 67%-87% ionic fluoride is converted into aluminium complex (Hussain et al., 2012). There is a high possibility of contamination due to the existence of metallic ions specially aluminium (Hussain et al., 2012). Aluminium contamination may be occurred due to the boiling of defluoridated water in aluminium utensils (Bhattacharya and Samal, 2018). To avoid this problem, now the use of floral residue like coconut shell carbon, chemically activated carbon, fishbone charcoal, burnt clay, natural zeolites, bone media, clay etc. are gradually become popular (Arulanantham et al., 1992; Muthukumaran et al., 1995; Killedar and Bhargava, 1993; Kartikeyan et al., 1999; Shrivastava and Deshmukh, 1994; Heidweiller, 1990; Hussain et al., 2012).

2.2.3. Ion-Exchange

In this process, water flows within a column which sustains ion exchange resin and the calcium ions within the resin are displaced by fluoride ions (Brindha and Elango, 2011). When every part of the resin is filled up with fluoride ions, the resin is backwashed with water which is enriched by sodium chloride (Meenakshi and Maheswari, 2006). The fluoride ions are now displaced by chloride ions and the mechanism is reactivated (Brindha and Elango, 2011). Actually the electronegativity of fluoride ions replaces the chloride ions from the resin (Meenakshi and Maheswari, 2006).

Due to the reactivation of resin, the amount of fluoride rich wate is increased. Not only that, the

existence of phosphate, carbonate and sulfate decrease the capability of this process. Although it is a very high –cost technique, it can diminish fluoride upto 90%-95%. The colour and taste of the water remain same after processing (Meenakshi and Maheswari, 2006).

2.2.4. Membrane Process

Reverse osmosis membrane process is considered to be one of the most effective technique to provide purified drinking water at household level and this process is able to overcome the limitation of other techniques (Brindha and Elango, 2011; Meenakshi and Maheswari, 2006). Reverse Osmosis is a physical mechanism where pressure is applied to channelize the flow of water within a semi permeable membrane for separating the pollutants. The capacity of this process is determined by the temperature and pressure condition, nature of raw water, intensive observation and assessment. Reverse Osmosis can not only remove fluoride but this process is capable to separate all the ions and pollutants. Fluoride can be removed upto 98% with the help of this technique. Furthermore, Reverse Osmosis can disinfect water without the help of any chemicals. In recent decades, the efficiency of this technique is highly increased and it becomes the best way to produce safe drinking water (Meenakshi and Maheswari, 2006). The process is more expensive and it separates all the necessary ions which are helpful for the growth and development of human body (Meenakshi and Maheswari, 2006; Brindha and Elango, 2011).

Electrodialysis is another method under membrane process. The proper activation of this highly expensive technique is depending on modern and vast infrastructure (Brindha and Elango, 2011).

2.3.Upliftment of Nutritional Status

Except In-situ and Ex-situ method, development of balanced diet structure is highly essential to control the spread of fluorosis. Calcium and phosphorous are two important ingredients which play an active role to check the growth of fluoride content in human body (WHO, 1984; Deshmukh and Chakravarti, 1995). So, the fluorosis affected people of Purulia district should intake calcium and phosphorous rich food to combat against this deadly disease. The local administrative body should take necessary action to uplift the nutritional condition of the poor villagers. Vitamin C is also a necessary element to prevent the terrible

impact of fluorosis (Rajiv Gandhi National Drinking Water Mission, 1993).

2.4. Community based fluoride management programme and expansion of awareness

Active participation of local people in community based management programme can facilitate to control the fluoride related problem (Hussain et al., 2012). Development of awareness and expansion of health education can motivate the local people to join in this programme. They must be aware about the fatal impact of fluoride rich water. The local administrative body should play active role and adopt necessary action to make this programme successful.

2.5.Biosorption

Biosorption process is one of the biotic defluoridation process which is not only a low-cost technique but also a renewable and eco-friendly process. This process mainly includes chitin and chitosan, but now fungal and algal biomass is also applied as biosorbents. Water purification with the help of biotic elements is now become popular worldwide.

2.6. Other Techniques

Optimum use of fertilizers and practice of organic farming should be implemented in fluoride affected regions of Purulia. Afforestation programme must be increased to check the rate of evapotranspiration in semi-arid region like Purulia. Vegetal cover can check the precipitation of fluoride rich salt on unsaturated zone which may be mixed with groundwater due to rainfall (Brindha and Elango, 2011).

III.CONCLUSION

Worldwide fluoride contamination becomes a severe problem and it is highly necessary to control this massive problem immediately. The poor villagers of different blocks in Purulia are victimized due to dental and skeletal/non-skeletal fluorosis. The local people from every sphere of society should come forward to prevent fluoride contamination. To combat fluorosis cost-effective, eco-friendly and easily accessible scientific techniques and suitable planning must be executed in different blocks of Purulia according to the preliminary intensity of fluoride, its origin and socio-economic condition of rural people (Brindha and Elango, 2011). Fluoride vulnerability map and hazard zonation map should be analysed to adopt appropriate remedial actions. Different NGO, local administrative body, educated people must come forward collectively to mitigate the hazard due to fluoride contamination. Scientific perception and

rational strategies can help to overcome the devastating problem of fluoride contamination in Purulia.

REFERENCES

- [1] Adhikary, S. K., Tipnis, U. K., Harkare, W. P., &Govindan, K. P. (1989). Defluoridation during desalination of brackish water by electrodialysis. *Desalination*, 71(3), 301-312.
- [2] Amini, M., Mueller, K., Abbaspour, K. C., Rosenberg, T., Afyuni, M., Møller, K. N., ...& Johnson, C. A. (2008). Statistical modeling of global geogenic fluoride contamination in groundwaters. *Environmental science & technology*, 42(10), 3662-3668.
- [3] Amor, Z., Bariou, B., Mameri, N., Taky, M., Nicolas, S., &Elmidaoui, A. (2001). Fluoride removal from brackish water by electrodialysis. *Desalination*, 133(3), 215-223.
- [4] Annadurai, S. T., Rengasamy, J. K., Sundaram, R., &Munusamy, A. P. (2014). Incidence and effects of fluoride in Indian natural ecosystem: a review. *AdvApplSci Res*, 5(2), 173-185.
- [5] Arulanantham, A., Ramakrishna, T. V., &Balasubramanian, N. (1992). Studies on fluoride removal by coconut shell carbon. *Indian Journal of Environmental Protection*, 12, 531-531.
- [6] Apparao, B. V., Meenakshi, S., &Karthikayan, G. (1990). Nalgonda technique of defluoridation of water. *Indian J. Environ. Protect*, 10(4), 292-298.
- [7] Barbier, J. P., &Mazounie, P. (1984). Methods of reducing high fluoride content in drinking water. Fluoride removal methods—filtration through activated alumina: a recommended technique. *Water Supply*, 2, 3-4.
- [8] Bhattacharya, H. N., &Chakrabarti, S. (2011). Incidence of fluoride in the groundwater of Purulia District, West Bengal: a geo-environmental appraisal. *Current Science*, 101(2), 152-155
- [9] Bhagavan, S. V. B. K., & Raghu, V. (2005). Utility of check dams in dilution of fluoride concentration in ground water and the resultant analysis of blood serum and urine of villagers, Anantapur District, Andhra Pradesh, India. *Environmental Geochemistry and Health*, 27(1), 97-108
- [10] Bhattacharya, P., and Samal, A.C., (2018). Fluoride contamination in groundwater, soil and cultivated foodstuffs of India and its associated health risks : A review. *Research Journal of Recent Sciences*, 7(4), 36-47.
- [11] Bishop, P. L., &Sansoucy, G. (1978). Fluoride removal from drinking water by fluidized activated alumina adsorption. *Journal-American Water Works Association*, 70(10), 554-559.
- [12] Brindha, K., &Elango, L. (2011). Fluoride in groundwater: causes, implications and mitigation measures. *Fluoride properties, applications and environmental management*, 1, 111-136.
- [13] Chauhan, V. S., Dwivedi, P. K., &Iyengar, L. (2007). Investigations on activated alumina based domestic defluoridation units. *Journal of Hazardous materials*, 139(1), 103-107.
- [14] Chidambaram, S., Ramanathan, A. L., &Vasudevan, S. (2003). Fluoride removal studies in water using natural materials. *Water SA*, 29(3), 339-344.
- [15] Deshmukh, A. N., &Chakravarti, P. K. (1995). Hydrochemical and hydrological impact of natural aquifer recharge of selected fluorosis endemic areas of Chandrapur district. *Gondwana Geol. Mag*, 9, 169-184.
- [16] Diaz-Nava, C., Solache-Rios, M., &Olguin, M. T. (2003). Sorption of fluoride ions from aqueous solutions and well drinking water by thermally treated hydrotalcite. *Separation science and technology*, 38(1), 131-147.
- [17] Dieye, A., Larchet, C., Auclair, B., & Mar-Diop, C. (1998). Elimination des fluorures par la dialyse ioniquecroisée. *European Polymer Journal*, 34(1), 67-75.
- [18] Heidweiller, V.M.L., (1990). Fluoride removal methods. In J. E. Frencken (Ed), *Proc. Symposium on endemic fluorosis in developing countries: causes, effects and possible solutions*, Chapter 6, 51-85.
- [19] Hichour, M., Persin, F., Sandeaux, J., Molenat, J., &Gavach, C. (1999). Water defluoridation by Donnan dialysis and electrodialysis. *Revue des Sciences de l'Eau/Journal of Water Science*, 12(4), 671-686.
- [20] Hichour, M., Persin, F., Sandeaux, J., &Gavach, C. (1999). Fluoride removal from waters by Donnan dialysis. *Separation and Purification Technology*, 18(1), 1-11.
- [21] Hussain, I., Arif, M., &Hussain, J. (2012). Fluoride contamination in drinking water in rural habitations of Central Rajasthan, India. *Environmental monitoring and assessment*, 184(8), 5151-5158.
- [22] Jalali, M. (2005). Nitrates leaching from agricultural land in Hamadan, western Iran. *Agriculture, Ecosystems & Environment*, 110(3-4), 210-218
- [23] Kariyanna, H. (1987). Geological and geochemical environment and causes of fluorosis—possible treatment—a review. In *Proceedings seminar on role of earth sciences in environment, Bombay* (pp. 113-122).
- [24] Karthikeyan, G., MuthulakshmiAndal, N., &SaravanaSundar, G. (1999). Defluoridation property of burnt clay. *JOURNAL-INDIAN WATERWORKS ASSOCIATION*, 31, 291-292.
- [25] Killedar, D. J., &Bhargava, D. S. (1993). Effects of stirring rate and temperature on fluoride removal by fishbone charcoal. *Indian Journal of Environmental Health*, 35(2), 81-87.
- [26] Maheshwari, R. C. (2006). Fluoride in drinking water and its removal. *Journal of Hazardous materials*, 137(1), 456-463.
- [27] Mameri, N., Lounici, H., Belhocine, D., Grib, H., Piron, D. L., &Yahiat, Y. (2001). Defluoridation of Sahara water by small plant electrocoagulation using bipolar aluminium electrodes. *Separation and Purification Technology*, 24(1-2), 113-119.
- [28] McKee, R., & Johnston, W. S. (1999). Removal of fluorides from drinking water using low-cost adsorbent. *Ind. J. Environ. Health*, 41(1), 53-58.
- [29] Muthukumaran, K., Balasubramanian, N., & Ramakrishna, T. V. (1995). Removal of fluoride by chemically activated carbon. *Indian Journal of Environmental Protection*, 12(1), 514-517.
- [30] Padmavathy, S., Amali, J., Raja, R. E., Nagarajan, P., &Kavitha, B. (2003). A study of fluoride level in potable water of Salem district and an attempt for defluoridation with lignite. *INDIAN JOURNAL OF ENVIRONMENTAL PROTECTION*, 23, 1244-1247.
- [31] Phansalkar, S. J., Kher, V., &Deshpande, P. (2005). Expanding rings of dryness: water imports from hinterlands to cities and the rising demands of mega-cities. *IWMI-Tata annual partner's meet, Anand*.
- [32] Prevention and Control of Fluorosis in India, Rajiv Gandhi National Drinking Water Mission, Manual, 1993.
- [33] Ravindra, K., &Garg, V. K. (2006). Distribution of fluoride in groundwater and its suitability assessment for drinking purpose. *International Journal of Environmental Health Research*, 16(2), 163-166.

- [34] Raichur, A. M., & Basu, M. J. (2001). Adsorption of fluoride onto mixed rare earth oxides. *Separation and Purification Technology*, 24(1-2), 121-127.
- [35] Reardon, E. J., & Wang, Y. (2000). A limestone reactor for fluoride removal from wastewaters. *Environmental Science & Technology*, 34(15), 3247-3253.
- [36] Rongshu, W., Haiming, L., Ping, N., & Ying, W. (1995). Study of a new adsorbent for fluoride removal from waters. *Water Quality Research Journal*, 30(1), 81-88.
- [37] Saha, S. (1993). Treatment of aqueous effluent for fluoride removal. *Water Research*, 27(8), 1347-1350.
- [38] Singh, G., Kumar, B., Sen, P. K., & Majumdar, J. (1999). Removal of fluoride from spent pot liner leachate using ion exchange. *Water Environment Research*, 71(1), 36-42.
- [39] Sreedevi, P. D., Ahmed, S., Madé, B., Ledoux, E., & Gandolfi, J. M. (2006). Association of hydrogeological factors in temporal variations of fluoride concentration in a crystalline aquifer in India. *Environmental Geology*, 50(1), 1-11.
- [40] Srinivasamoorthy, K., Nanthakumar, C., Vasanthavigar, M., Vijayaraghavan, K., Rajivgandhi, R., Chidambaram, S., ... & Vasudevan, S. (2011). Groundwater quality assessment from a hard rock terrain, Salem district of Tamilnadu, India. *Arabian Journal of Geosciences*, 4(1-2), 91-102.
- [41] Shrivastava, P. K., & Deshmukh, A. (1994). Defluoridation of water with natural zeolite. *Journal of the Institute of Public Health Engineers (India)*, 14, 11-14.
- [42] Susheela, A. K. (1999). Fluorosis management programme in India. *CurrSci*, 77(10), 1250-1256.
- [43] Thergaonkar, V. P., & Nawalakhe, W. G. (1971). Activated magnesite for fluoride removal. *Ind. J. Environ. Health*, 16, 241-243.
- [44] Wang, Y., & Reardon, E. J. (2001). Activation and regeneration of a soil sorbent for defluoridation of drinking water. *Applied Geochemistry*, 16(5), 531-539.
- [45] WHO, (1984). Guidelines for Drinking Water Quality. In: *Health Criteria and Other Supporting Information*, second ed., vol. 2. World Health Organization, Geneva.
- [46] Yang, M., Hashimoto, T., Hoshi, N., & Myoga, H. (1999). Fluoride removal in a fixed bed packed with granular calcite. *Water Research*, 33(16), 3395-3402.