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Fluoride health effects and defluoridation using adsorption: A critical review

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Abstract
Fluoride is a monotonic ion and an essential mineral for body. The use of Fluoride include cavity prevention, prevent from tooth decay, biochemical reagent. The consumption limit is set to be maximum 1.5 mg/l by WHO. The long term exposure to excess fluoride has several negative impact on human health, water bodies, environment and agricultural fields. The source of fluoridation may include the natural occurring minerals, coal mines or the anthropogenic action. Several materials (Biomass, rice husk ash, bone char, shale, and low grade coal) and methods (Adsorption, membrane separation, and column studies) have been in use for defluoridation of water. Among the above mentioned methods adsorption is proved to be economical, easy and efficient for fluoride removal for domestic and industrial help.

Keywords: Fluoride, health impact, defluoridation technique, adsorption and adsorbents

Introduction
Water is a natural essential need of human as well as environment. The most important thing is it should be safe and portable for drinking purpose. Natural deep ground water is safe but the presence of some hazardous substance (fluoride, arsenic, nitrate, and lead) makes it unsuitable for daily consumption. The permissible limits for these substances are 1.5 mg/L, 10 µg/L, 50 mg/L, and 10 µg/L respectively for fluoride, arsenic, nitrate, and lead (WHO, 2004) [83]. Most recently the fluoride contamination in water has raised a concern worldwide. Fluoride is an abundant trace element which is found with an average concentration of 625 mg/kg of fluorine in the earth's crust (Gupta and Ayoob, 2006) [5]. The natural mineral that contains fluorine is granite, basalt, and shale. The other sources may include vast industrialization, effluent discharge, and waste water. The long term exposure to fluoride in excess amount leads to adverse effects to human being. The most common health impacts include different type of fluorosis (skeleton, dental), infertility, brain damage, stones and thyroid disorder. High fluoride concentrations in drinking water and associated fluorosis issues were reported from African countries such as South Africa, Kenya, Ghana, Sudan and Tanzania (Ayoob, 2008) [6]. More than 27 countries including India are suffering from the excess fluoride contamination in water. 62 million people including 6 million children in the country in 17 states are affected with dental, skeletal and non-skeletal fluorosis (Patil and Ingole, 2014) [62].

Fluoride Metabolism
Fluoride enters to the human body either in food or through the respiration process (Cagetti, 2013) [18]. About 90% of fluoride is absorbed in the gastrointestinal tract after consumption (around 25% is consumed in the stomach and up to 77% in some part of the small intestine). The remaining 10% is excreted in feces (Buzalaf and Whitford, 2011) [17]. The absorbed fluoride is transported in to the blood stream, distributed to organism. Fluorine is also capable of crossing our blood-brain barrier, which causes biochemical and functional changes in our nerve system. Fluoride is basically obtained in two forms (Topical and Systemic).Topical fluoride is most useful to strengthen the hard tissue of body i.e. teeth, bone. Systemic fluoride is indigested to our body in order to protect and develop those hard tissue. About 99% of all retained fluoride is contained in mineralized tissues, other 1% is found in soft tissue (Buzalaf and Whitford, 2011) [17]. The remains are absorbed in kidney and that is the main reason kidney plays a very important role in balancing the fluoride concentration in our body.

Fluoride health impact

Fluoride is present in the environment occurring in water, soil, and air. The only effective use of fluoride for human is dental and bone care, otherwise the intakes in improper quantity may lead to severe health risk. The most chances of exposure to fluoride is through drinking water as our water intake in minimum 2-4 liter/day. Some organization has set the permissible limit for fluoride in drinking water as shown in table 1.

Table 1: Permissible Limit of Fluoride Concentration In Drinking Water.

S. N	Organization	Permissible limit of fluoride (mg/L)
1	World Health Organization (WHO)	1.5
2	Bureau of Indian Standards (BIS)	1.0
3	Indian Council of Medical Research (ICMR)	1.0
4	Indian Standard Institute (ISI)	0.6-1.2
5	Central Pollution Control Board (CPCB)	1.5

The use of fluoride more than the recommended limits is highly toxic. Maithani (1998) [48] reported the case of crippling and mottling in the western area of Sirohi district, Rajasthan. The fluoride amount was found to be as high as 16 mg/L in their drinking water. Not only in India have other nations also raised a concern about high fluoridation. A study was done in by taking consecutive 15 years data showed the effect on children in age group of 6-11, the fluoride content was 4.3 mg/L and the analysis showed decrease in their IQ, poor visibility. The inner organ of our body are also effected like swelling of the mitochondria, chromatin clumping, granular endoplasmic reticulum, damage to the nuclear membrane (Valdez-Jiménez, 2011) [78]. Not only among the normal people, has it had also dangerous effect on the pregnant woman consuming it. Total 213 pairs of mother-children were studied in a region having fluoride concentration more than standard value, the result was behavioral changes, mental illness in child in their growing age. Kidney plays an important role to balance the fluoride as it subsequently gathers the residue to be discharged from our body system. So directly or indirectly the amount of fluoride intake in our body affects the function of kidney and liver. Ludlow (2007) [47] gave a report on absorption of fluoride in our body, it states that consuming water with fluoride concentration 3.5 to 4.9 mg/L in an area lead to higher risk of kidney stone formation among the people. A recent analysis was done in Argentina by Pollo (2019) [64] to estimate the fluoride content and its harmful effect. The result was shocking as the daily consumption of fluoride in that area was more than 14 mg/L. that leads to severe abnormalities among people. The fluoride was directly effecting their brain tissues spreading about 21.2% of the population in Decantation ponds, Argentina. A brief summary of some research are mentioned below in table 2.

Table 2: Fluoride Health Effect Summary of Different Areas.

S.N	Author (year)	Area	Fluoride Concentration (mg/l)	Health effect
1	Freni (1994) [30]	US	3	Decrease in TFR (total fertility rate)
2	Apambire (1997) [4]	Bolgatanga and Bongo Districts	0.11 to 4.60	Dental and skeletal fluorosis
3	Nayak (2005)	Sahibganj district, Jharkhand	0.331–10.36	Joint pain, deformity of the limbs and spine, abnormal tooth enamel, ligamentous calcifications
4	Rocha-Amador (2007) [68]	---	>1.5	Lower the IQ level in children
5	Xiong (2007) [84]	---	2	Increased serum lactic dehydrogenase (LDH), urine N-acetyl-beta-glucosaminidase (NAG), and urine gamma-glutamyl Trans' peptidase (gamma-GT) in children, damage to liver and kidney
6	Villa (2010) [80]	---	2	Fluorosis
7	Brindha (2011) [16]	Nalgonda, Andhra Pradesh	0.1 to 8.8	Fluorosis
8	Peckham (2014) [63]	Some parts of Delhi	4.37	Clinical hypothyroidism. Skeletal fluorosis, chronic metabolic bone disease
9	Dey (2015) [26]	---	>1	Effect diabetic patient, lower IQ, permanent damage to the brain, and neurotoxicity
10	Mondal (2016) [54]	Birbhum district, West Bengal	0.33 to 18.08	Dental and skeletal fluorosis, lower IQ
11	Choubisa (2016) [22]	India	1.89-2.14	Dental fluorosis, maladies, non-skeletal fluorosis, impaired endocrine, neurological disorders, and reproductive functions, renal effects, genotoxic effects, teratogenic effects, apoptosis, excitotoxicity in man.
12	Malin (2018) [50]	Some parts of Canada	> 1.5	Effect on thyroid glands
13	Bashash (2018) [9]	---	> 1.5	child's hyperactivity disorder
14	Dhar (2019) [28]	---	> 1.5	Abdominal pain, diarrhea, and vomiting, excess salivation, osteosclerosis, osteoporosis, osteomalacia, exostosis formation, calcification of ligaments, and dental fluorosis.
15	Ghadherpoori (2019) [34]	Mashhad city, Iran	< 1.5	No health effect

Defluoridation Techniques

A large part of the world's population directly depends on the ground water for drinking purpose; this is also associated with a risk of contagious disease to them. Fluoride contamination in water is a global problem now, the only way to reduce the risk is defluoridation of water. This is a technique where we remove the fluoride from water using various technique and materials as adsorbent. So many techniques are in use like adsorption, membrane separation, precipitation methods, ion-exchange, nano filtration, electro Coagulation, and column studies. This paper reviews the adsorption methods using various type of adsorbent.

The adsorption is economical in terms of availability of adsorbent material; either we can use the natural materials, synthesized new material or a byproduct of mine waste. The leaf from some tree; pipal (Ficusreligiosa), khair (Acacia catechu wild), and neem (Azadirachtaindica) were chosen as adsorbent and it was 90% efficient for defluoridation (Jamode, 2004) [40]. Again with the leaf of Neem (Azadirachtaindica) and Kikar (Acacia arabica) the efficiency of removal was high followed by Freundlich, Langergren absorption equation of isotherm (Kumar, 2007) [32]. A modified rice husk ash (RHA) coated with aluminum hydroxide was taken for the test, which successfully removed fluoride by 9-10 mg/g. These mechanism best fitted to pseudo-second-order kinetic model (r^2 = 0.9990), which is highly efficient (Ganvir, 2011) [31]. The methodologies include adsorbent preparation, synthesis of the adsorbent and characterization of material is done to know the surface mechanism. X-ray diffraction (XRD) analysis, thermo gravimetric analysis (TGA), differential thermal analysis (DTA), FTIR spectroscopy and XPS analysis are some general methods of characterization.

Adsorption analysis is done in a batch process with varying the parameters like pH, temperature, contact, time, amount of adsorbent, particle size. Table 3 shows various researches that conducted adsorption with different type's adsorbent.

Table 3: Researches on adsorption method for fluoride removal from Year 2004-2019.

S. N.	Author	Material used	Isotherm	Kinetics	Capacity (mg/g)
1.	Dey (2004) [27]	Hydrous ferric oxide (HFO)	Freundlich model	First-order Lagergren Equation	_
2.	Maliyekkal (2008) [51]	Magnesia-amended activated alumina granules.	Langmuir, Radke-prausmitz, Redlich–Peterson, Sips, Toth, Frendlich	Pseudo-first-order, Pseudo- second-order, Intraparticle diffusion model	10.12
3.	Deng (2011) [24]	Mn–Ce oxide	Langmuir	Pseudo-second-order model	137.5
4.	Chen (2012) [21]	Fe—Ti oxide nano-adsorbent	Langmuir	_	47.0
5	Chai (2013) [20]	Sulfate-doped Fe_3O_4/Al_2O_3 nanoparticles	one site and two-site Langmuir models	Pseudo-first-order, pseudo second-order and Elovich models	70.4
5.	Jahin (2014) [39]	Nano scale zero valent iron (nZVI)	Langmuir and Freunlich	_	18.91
6.	Goswami and Purkait (2014) [35]	Schwertmannite $(Sh,Fe_8O_8(OH)_6SO_4)$	Langmuir and Temkin	Pseudo-second-order	17.24
7.	Yin (2015) [87]	Natural calcium-rich attapulgite (NCAP)	Langmuir and Freundlich	Pseudo-first-order, second order, Intraparticle diffusion models	_
8	Biswas (2017) [14]	Dry biomass (DBM)	Langmuir model, Freundlich model, Temkin model and Dubinin-Radushkevich model	Lagergren first-order, Ho pseudo second order and Moris-Weber	34.36
9.	Mondal (2017) [55]	Natural banana peel (NBP) dust	Langmuir, Freundlich, D-R and Temkin	Pseudo-first, pseudo-second, intra-particles diffusion and Bahangam models	1.212
10.	Nunes-Pereira (2018) [59]	Montmorillonite (MMT), zeolites (NaY), bayerite (BAY), hydroxyapatite (CaHAp).	_	_	13
11.	Nagaraj (2018) [57]	Nanocomposites of hydroxyapatite (mHAp)	Freundlich, Langmuir and Dubinin–Radushkevich (D–R)	Pseudo-first Order, pseudo-second order and Lagergren model	8.36
12.	Sadhasivam (2019) [69]	Cao and SiO_2 nano particle	N_2 sorption	_	_
13.	Bai (2019) [7]	Nial-LMO	Quasi-first-order, quasi-second-order	_	49.28
14.	Shahid (2019) [71]	Bone char (BC)	Langmuir and Freundlich	Pseudo-first-order, second order	10.56
15.	Yu (2019) [88]	One-dimensional ZrO_2 mesoporous	Freundlich	Pseudo-second order	297.7

Conclusion

As the two sides of a coin, Fluoride has its own benefit and harm depending up on the concentration we consume. The necessity of fluoride in our life cannot be ignored but the severe health risk associated with high amount of consumption needs some special attention. The risks include fluorosis of teeth and bone, lower IQ, behavioral changes, mottling, crippling, hyperactivity disorder, kidney stone, liver failure, thyroid gland function, visual loss, osteosclerosis, and joint pain. The only solution to this worldwide problem is fluoride removal. Various techniques have been in practice now days but Adsorption method stands strong among them because of high efficiency in removal and providing very high quality water in a specific time constraint. But still, selection of best material, technique is important to enhance the water quality.

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