Treatability Study of Arsenic, Fluoride and Nitrate from Drinking Water by Adsorption Process

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Summary: Natural contamination of nitrate, fluoride, arsenic and dissolved salts in ground water sources is the main health menace at present in different parts of Pakistan. The metalloids especially arsenic, fluoride and nitrate pose severe health hazards to human being. The present research work investigated the removal techniques for arsenic, fluoride and nitrate from drinking water by adsorption process. Ion exchange resins, activated carbon and activated alumina were used for removal of selected contaminants. These adsorbents were evaluated by comparing their removal efficiency as well as requisite operator skills. The result of activated alumina was found good as compared to activated carbon, mix bed resins and ion exchange resins (IRA-400) for maximum removal of arsenic, nitrate and fluoride. The removal efficiency of arsenic, fluoride and nitrate were found 96%, 99%, 98% respectively in case of activated alumina. The advantage of adsorption process is easy to use and relatively cheaper as compared to other treatment methodologies.

Key words: arsenic; fluoride; nitrate; activated carbon; activated alumina; adsorption.

Introduction

Most commonly pollutants found in drinking water are arsenic, fluoride, chromium, nitrate as well as some organic pollutants. Arsenic is a famous toxic metal mostly present as oxyanion compounds in groundwater [1]. The guideline of World Health Organization (WHO) indicates maximum provision of arsenic in drinking water as 10 µg/L but this limit is not followed by most of developing countries. They are still struggling to keep up with the previous WHO guideline value of 50 µg/L [2].

Due to anthropogenic and natural activities, arsenic compounds are well-known contaminants in the hydrosphere. Higher concentration of arsenic in drinking water is serious problems for human health and can also induce carcinogenity. The population in under-developing countries like Bangladesh, Pakistan, Western Bengal, Vietnam and Chile are seriously affected by arsenic toxicity [3, 4].

Fluoride can cause significant impact on human health and other living organisms when its higher concentration discharged into water bodies. Fluorine is the top member of halogen family that does not occur in the element state due to its high reactivity [5]. On contrary presence of fluoride in drinking water in acceptable concentrations is an essential constituent for human health, especially in children below 8 years [6]. The excessive intake of fluoride usually has adverse effect on body metabolism [7] and leads to dental and skeletal fluorosis, lesions of endocrine glands, thyroid and liver [8]. The maximum provision established by World Health Organization (WHO) for fluoride in drinking water is 1.5 mg/L [9].

The higher concentration of nitrate in drinking water can cause adverse health effects. The nitrate in groundwater used for drinking, especially in rural areas is becoming serious problem due to its harmful effects. An excessive level of nitrates causes serious illness, sometimes death. Main causes of toxicity of nitrate in drinking water include shortness of breast, blueness of the skin and can cause potential formation of carcinogenic nitrosamines especially in infants [10, 11].

Several methods, such as reverse osmosis, ion exchange, adsorption, coagulation, precipitation, and electro coagulation have been used for the removal of excess fluoride, nitrate and arsenic from drinking water. Among these methods, adsorption is the most extensively used and is a promising technique for the removal of water contaminants [12-14].

Numerous materials such as activated carbon, metal hydrides and synthetic resins are commonly used in adsorption process for purification of water and wastewater in different industries. The most common material used for arsenic removal is activated carbon [15, 16]. The use of ion exchange resins are also commonly reported for removal of various contaminants from water, particularly dissolved solids [17]. The ions held electrostatically in ion exchange process on the surface of a solid

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phase and similar charged are exchanged of similar charge in a solution. It is a reversible interchange where there is no permanent change in the structure of the solid. Ion exchange is used generally in drinking water treatment for softening. It efficiently removes calcium, magnesium etc. as well as nitrate and arsenic from municipal water [18].

In under developed and developing countries of the world, the most communicable diseases are water borne due to drinking of unsafe water. In Pakistan, large numbers of people drink polluted water. About 50 percent diseases are waterborne that cost millions of dollar each year. The aim of this study is to evaluate the adsorption capacity of different filtering media and selection of most suitable treatment method for arsenic, fluoride and nitrate removal from drinking water.

Results and Discussion

The present study is focused, especially on ease of treatment method and comparison of removal efficiency of selected contaminants. The summery of methods used for nitrate removal is shown in Table-1. The removal efficiency with activated alumina was found to be maximum up-to 98 percent and it required low operator skills. Mix bed resins also showed excellent removal efficiency of 97 percent but it demands high operator skills. In case of activated carbon, removal of nitrate was found 94 percent. Activated carbon is commonly known as universal adsorbent for the removal of various types of water pollutants, especially organic pollutants. Although in case of anionic pollutant, it shows poor adsorption. In literature only fewer studies are available that report the adsorption of nitrate by activated carbon [19].

For fluoride removal, the ion exchange process is not found to be efficient in comparison of activated alumina shown in Fig. 1. Activated alumina efficiently absorbs the fluoride concentration up-to 99 percent as compared to the other adsorbing materials used in these batch experiments as shown in Table-2. Activated alumina is porous, granular materials that contain excellent ion exchange properties for removal of pollutant from water bodies [20]. The removal efficiency of fluoride and nitrate were found maximum in case of activated alumina and mixed bed resin as shown in Fig. 1. The activated alumina showed optimum removal at pH (5.5 to 6.5), thus water source required to pretreatment with hydrochloric acid [21]. In previous studies the result showed that a significant decrease in sorption capacity when deviating from neutral pH values. At pH 5–6, maximum fluoride removal was occurred. From the zeta potential measurement, it was achieved that fluoride adsorbed onto Al2O3 by replacing hydroxyl ions from positively charged surfaces and through hydrogen bond [22, 23].

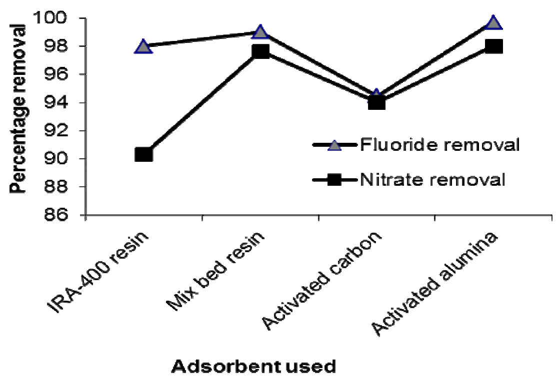

Fig. 1: Fluoride and nitrate removal by using different adsorbing media.

Arsenic removal was also found to be higher in case of activated alumina as illustrated in Fig. 2. In addition, it required low operator skill so it seems quite suitable for arsenic removal. The removal efficiency was found as 92 percent in case of ion exchange resins as well as with activated carbon using as filtration media (Table-3) whereas removal efficiency was found 94 percent in mix bed resin. The removal efficiency of activated alumina was found maximum when compared to Ion exchange (IRA-400) resin, Activated carbon and mixed bed resin. Moreover, it also required low operation skill. The removal of arsenic, fluoride and nitrate were found 96%, 99%, 98% respectively, that showed higher efficiency as compared other adsorbents used in experiment.

Table-1: Summary of methods used for nitrate removal.

Technology	Water loss (%)	Removal Efficiency (Re)			Avg. Re ± S.D	Observation and interference	Operator skill
		X_1	X_2	X_3			
Ion exchange (IRA-400) resin	1-3	88.7	90.2	90.7	90±1.04	Maximum efficiency at pH 6.5-9.0, however, decreased at high pH	High
Activated alumina	1-2	98.6	97.5	98.1	98±0.55	Maximum efficiency was found at pH 5.5-8.3 but decreased at high pH	Low
Activated carbon	1-2	93.2	94.6	93.8	94±0.70	Silica must less than 30 %	Medium
Mixed bed resin	1-3	96.4	97.1	97.3	97±0.47	The optimum pH was found in the range of 6.5-9.0	High

Table-2: Summary of methods used for fluoride removal.

Technology	Water loss (%)	Removal Efficiency (Re)			Avg. Re ± S.D	Observation and interference	Operator skill
		X_1	X_2	X_3			
Ion exchange (IRA-400) resin	1-3	84.7	85.1	85.3	85±0.30	Maximum efficiency was found at pH 6.5-9.0 but decreased at high pH	High
Activated alumina	1-2	99.2	99	98.2	99±0.20	The optimum removal was noted at pH 5.5-8.3	Low
Activated carbon	1-2	90.4	89.6	90.1	90±0.40	Silica not affected on process efficiency. It is economical for TDS 3000-5000 mg/L	Medium
Mixed bed resin	1-3	88.7	88.6	89.3	89±0.31	Maximum efficiency was found at pH range 6.5-9.0	High

Table-3: Summary of methods used for arsenic removal.

Technology	Water loss (%)	Removal Efficiency (Re)			Avg. Re ± S.D	Observation and interference	Operator skill
		X_1	X_2	X_3			
Ion exchange (IRA-400) resin	1-3	92.5	92.2	91.7	92±.40	Pilot scale is used in industrialized as well as household system. Adsorption capacity is very high but needs continuously regeneration that increases its cost. The regeneration process produces arsenic rich brine which is difficult to dispose.	High
Activated alumina	1-2	95.6	96.1	96.4	96±.40	Used in developing countries as household and industrial system, documentation required for long term of performance of regenerated media	Low
Activated carbon	1-2	91.6	92.2	92.6	92±.50	Silica not affected on process efficiency. It is economical for TDS 3000-5000 mg/L	Medium
Mixed bed resin	1-3	94.4	94.0	93.7	94±.30	Very effective for laboratories to obtained high efficiency but need continuously regeneration that increases its cost. Arsenic rich solution obtained in the form of regeneration waste	High

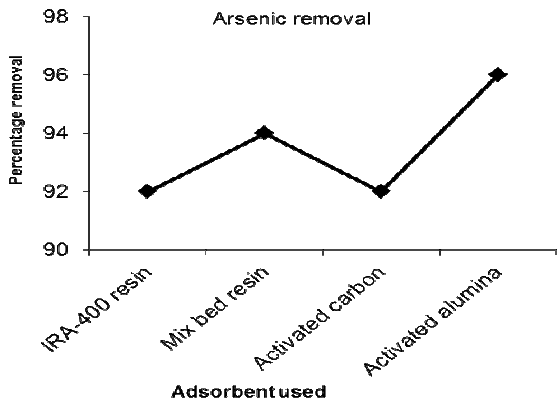

Fig. 2: Arsenic removal by using different adsorbing media.

Experimental

Study was carried out at Center for Environmental Protection Studies, PCSIR (Pakistan Council of Scientific and Industrial Research) Labs Complex Lahore by preparing samples in the laboratory. Samples solution contained 100 mg/L fluoride, 100 mg/L nitrate and 50 ug/L arsenic. All the solutions were prepared in double distilled water having conductivity less than 1 µS/cm. Four glass columns having one-inch internal diameter were used in the experiment. The first and second columns contained anion exchange resin (IRA-400) and mixed bed ion exchange resins (i.e cation and anion exchange in the form of sodium and chloride) respectively. These resins can be regenerated or recharged when exhausted due to continuous adsorption by specific regenerates [24].

Regeneration of resins was carried out by 10 % aqueous sodium chloride solution. The third column contained activated alumina. Its activation was done by 0.1 N HCl whereas it was regenerated by 0.1 N Sodium Hydroxide. The fourth column contained activated carbon. The activation of this column was done with 1 N HCl then washed with plenty of double distilled water until it neutralized. The operative condition of experiments is shown in Table-4.

Table-4: Operation conditions of different filtering media.

Parameters	Ion exchange resins	Activated carbon	Activated alumina
Effective depth(m)	0.5	0.5	0.5
Effective size(mm)	0.4-0.6	2.0-2.5	2.0-2.5
Bed mass(g)	100	150	150
Flow rate (ml min^{-1})	10	10	10
pH	6.5± 0.15	7.0± 0.2	7.5± 0.2
Temperature (°C)	23.0 ± 2	22.0 ± 2	22.0 ± 2

Preparation of Reagents

Stock solution of arsenic was prepared by dissolving arsenic salt As_2O_3 in double distilled water. Arsenic solution used in the experiments was prepared by diluting stock solution up-to desired concentration with double distilled water, pH value of the arsenic solutions was adjusted by adding 0.1 M hydrochloric acid or 0.1 M sodium hydroxide. The stock fluoride solution was prepared by NaF whereas nitrate solution was prepared by KNO_3. All chemicals used in these experiments were of analytical grade whereas all the working solutions were prepared in freshly prepared double distilled water.

Analysis of water Quality

The changes in water quality were measured before and after treatment with adsorbents. The concentration of arsenic was determined by Inductive couple plasma (PerkinElmer optimum-5300). The concentration of fluoride and nitrate were determined by using ion chromatograph Shimadzu C-R4A Chromatopac. The removal efficiency was calculated by using following formula:

$$R_e = \frac{C_i - C_f}{C_i} \times 100$$

where,

R_e Removal efficiency,
C_i Initial concentration
C_f Final concentration

Conclusion

The result of present study reveals that activated alumina is best adsorbent for arsenic, nitrate and fluoride as compared to resin and activated carbon. Moreover, it required low operator skill and the optimum removal was found at pH 5.5-8.3. During process, the loss of water was only 1-2 percent. The main advantages of adsorption method are easy to use and relatively cheaper as compared to other treatment methodologies due to regeneration of adsorbing material so that these could be used for more than one time.

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