



# Use of bauxite from active Iranian mines for the removal of fluoride from drinking water

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## Abstract

**Background:** Fluoride plays an important role in bone and dentin mineralization; however, excess fluoride intake is harmful to mankind.

**Methods:** This study evaluated the performance of bauxite from active Iranian mines in removing fluoride from drinking water. The effects of pH, contact time, adsorbent dose, and fluoride concentration on defluoridation and removal efficiency were determined. Kinetics and adsorption isotherms were studied. Fluoride levels were measured using SPADNS. Data analysis was performed using SPSS16.

**Results:** Bauxite from the Jajarm mine had the lowest adsorbency (20 g/L) and required the shortest contact time (90 minutes) to reach equilibrium compared with the ore from bauxite mines evaluated in another study which had greater efficiency rates in removing fluoride from drinking water (58.15%). The fluoride removal efficiency rates of the other bauxite mines were as follows: Mendon > Sadrabad > Khidabas > Khezri > Shahbalaghi > Tash > Biglar. Bauxite from Shomal-e Yazd, Hasanabad, and Shahid Nilchian mines could not achieve the required efficiency to remove fluoride from drinking water without initial preparation and modification. The removal efficiency rates of actual samples were much lower than the synthetic samples because of confounding factors.

**Conclusion:** As a result of the low cost and abundant availability of bauxite and the fact that its use does not require a particular expertise or sophisticated technology, the removal efficiency of this adsorbent can be increased to desirable levels through the use of corrective methods such as heating, acidifying, particle crushing, or the mixing of two or more removal systems.

**Keywords:** Fluoride, Iran, drinking water, kinetics

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## Introduction

Fluoride occurs naturally in mineral deposits and natural water systems (1). Since groundwater is a major source of human exposure to fluoride (2,3), determining the amount of fluoride present in drinking water is an important factor in human health. The treatment of groundwater resources is often the only disinfection performed on drinking water; fluoridation and defluoridation rarely occur (4).

Fluoride has been classified as a water pollutant by the World Health Organization (WHO). The presence of fluoride in drinking water, dependent upon concentration (5) and duration of continuous absorption (2), can be either helpful or harmful to humans. Maintaining a fluoride concentration of about 1 mg/L can prevent skeletal and dental problems, especially in children (2,6). However, the intake of excessive fluoride can cause serious concerns for public health (7).

Fluorosis is one of the most important public health issues

around the world (5). Dental and skeletal fluorosis is irreversible, and there is no treatment for it yet. The only remedy is prevention, which is possible by maintaining safe limits of fluoride intake. Following the WHO guidelines, many countries have reported 1.5 mg/L as the maximum contaminant level. Many epidemiologists have indicated in their reports, however, that there is the possibility of adverse effects with long-term exposure to fluoride, even where consumption is at the permissible limit (8,9). Global estimates suggest that over 70 million people suffer from fluorosis (10). Fluoride concentrations in the drinking water of large parts of the world are higher than allowed by the WHO guidelines (11). It is estimated that more than 200 million people worldwide use drinking water that has a fluoride concentration higher than set standards (2). The presence of high levels of fluoride in groundwater is a global problem and is found in various locations in Africa, Asia, and America (9).



Mesdaghinia et al studied the monitoring of fluoride levels in the groundwater of Iran (4). Their study showed that in various regions of Iran, especially Bushehr province and parts of Hormozgan, Khorasan-e Razavi, Fars, and Yazd provinces, fluoride levels in groundwater were higher than standard levels (4). A study conducted in 2012 by Haghighat et al showed that fluoride concentrations in the drinking water of Dashtestan and Borazjan from Bushehr port functions, Hormozgan (Bandar Abbas, Bandare Lenghe, Qeshm), Yazd (Ardakan), Hamedan, Tehran, Kerman (Shahrabak, Kuhbanan), Khorasan-e Razavi, South Khorasan, Semnan, Zabol, and Zahedan were higher than set standards (11).

The effects of excess fluoride on teeth and bones have been documented (12). Research on the removal of fluoride from water in order to prevent dental and skeletal fluorosis is improving around the world.

Methods such as coagulation, chemical precipitation, surface adsorption, electrochemical and membrane processes, and ion exchange are used to remove excess fluoride from water (13). The high costs and complexity of application of many of these methods prohibit their use in developing countries and low-income areas (14). There is a growing tendency to use local materials and low-cost methods for the removal of fluoride from water (15). Removing fluoride from water by surface adsorption is known as the ideal method in small communities. In recent years, the use of low-cost adsorbents to remove fluoride from water has been investigated. Hydroxyapatite, calcite, fluor spar and quartz (16), fly ash (17), red mud (18), and bentonite (19) are some such adsorbents. Bauxite is a native and available mineral consisting of aluminum hydroxides with minor and variable amounts of silica, iron oxide, titanium oxide, and alumina silicate (20). Many studies have investigated using bauxite to remove pollutants from water, such as Bhakat et al in India (21), Baral et al in India (22), and Debasish et al in Korea (23) which used bauxite to remove arsenic and chromium (VI) and chromium, respectively. In Tanzania, Thole used bauxite to remove fluoride from water (24). Sajidu et al In 2008 in India, used raw bauxite for the removal of fluoride from water (25). Atasoy et al in Turkey (26), Lavecchia et al in Italy (20), and Malakootian et al in Iran used bauxite to remove fluoride from water (3). Some areas of Iran have high fluoride concentrations; however, prior to this study, the efficiency of bauxite from Iranian mines in fluoride removal had not been studied. Thus, the current study evaluated the efficiency of activated bauxite from ore mined in Iran in removing fluoride from aqueous solutions.

## Methods

This experimental study was conducted on synthetic and actual samples in the Environmental Health Engineering Research Center, Kerman University of Medical Sciences, from May to November of 2013. Actual samples were

collected from drinking water from the Kuhbanan region in northwestern Kerman province ( $31^{\circ}24'37''\text{N}$ ,  $56^{\circ}16'57''\text{E}$ ) (Figure 1).

Previous studies reported that fluoride concentrations in the water resources of this region ranged from 2.3-5.4 mg/L (27). First, the chemical quality of actual samples was determined. Bauxite, used as adsorbent in this study, was prepared from bauxite mines that were active in Iran until 2011 (Jajarm mine located in north Khorasan; Shomal-e Yazd and Sadrabad mines located in Yazd; Shahblaghi mine located in Damavand; Hassanabad and Biglar mines located in Qazvin; Mendon mine located in Kohgelooye and Bovair Ahmad; Shahid Nilchian mine in Chaharmahal and Bakhtiari; Khezri and Khidbas mines in South Khorasan; and Tash mine in Semnan province). Initially, stones were crushed, ground, and sieved to obtain  $<250\ \mu\text{m}$  size (mesh 60). The resultant bauxite powder was washed with deionized water, dried in an oven at  $200^{\circ}\text{C}$  in an open-air chamber muffle furnace for 2 hours, and cooled at room temperature. Samples were stored in a desiccator. X-ray fraction (XRF) and x-ray diffraction (XRD) were used to determine the chemical composition and the phases in the bauxite powder samples. XRD and XRF analyses were done using the Philips X'PERT device in Iran Mineral Processing Research Center. Stock solution was prepared by dissolving 0.221 mg sodium fluoride (NaF) in 1000 mL deionized water ( $1\ \text{mL}=100\ \mu\text{g}/\text{F}$ ), and experimental solutions of the required concentrations were prepared from this solution. Fluoride was measured using the SPADNS method and spectrophotometry (wavelength = 570 nm). All analysis methods were taken from the "Standard Methods for Examination of Water and Wastewater", 20th edition (28). Absorption tests were used to determine equilibrium time, adsorption kinetics, and optimal conditions (pH, contact time, fluoride concentration, and amount of adsorbent), and the adsorption isotherm models were studied. Adsorption experiments were used to determine equilibrium time, synthetic adsorption, and optimum conditions and to study the adsorption isotherm models. The operational parameters were pH values (3–34); initial



**Figure 1.** Geographical map of Kuhbanan region in northwestern Kerman province ( $31^{\circ}24'37''\text{N}$ ,  $56^{\circ}16'57''\text{E}$ ).

fluoride concentrations (2, 4, 6 mg/L); dosages of bauxite (2, 5, 34, 15, 20, 25, 30 g/L); contact times (5-240 minutes). Tests were then performed under optimal conditions on actual samples, and removal efficiency was measured. The pH value of the samples was set to 0.1N by H<sub>2</sub>SO<sub>4</sub> and NaOH. The pH was measured by a microprocessor pH meter (model 211 Hanna). Deionized water was used in the preparation of all solutions and in all analyses. Removal efficiency (E%) and adsorption capacity (q<sub>e</sub> mg/L) were calculated using equations 1 and 2:

$$E = [(C_i - C_e) / C_i] \times 100 \quad (1)$$

$$q_e = [(C_i - C_e)V] / m \quad (2)$$

where C<sub>i</sub> and C<sub>e</sub> are the initial and final concentrations of fluoride in solution (mg/L), respectively, V is the volume of solution (L), and m is mass of the adsorbent (g).

Langmuir and Freundlich isotherm models were investigated to study the adsorption isotherms. Data analysis was performed using SPSS software version 16.

## Results

Composite sampling was done 3 times from the Kuhbanan water resource. Analysis of the samples was expressed as the average of three replicates. The commonly occurring cations and anions present in the Kuhbanan drinking water sample were found in mg/L as NO<sub>3</sub><sup>-</sup> (5.6), SO<sub>4</sub><sup>2-</sup> (139), Cl<sup>-</sup> (126), Total solid (711), Ca<sup>2+</sup> (83), K<sup>+</sup> (1.9), Mg (49), Na<sup>+</sup> (95), F<sup>-</sup> (2.3–2.7), Fe<sup>3+</sup> (<0.05), Al<sup>3+</sup> (<0.01), pH (6.9–7.66), and temperature (25–27°C). Sajidu et al examined the effects of bauxite on water quality after fluoride removal in the villages of southern Malawi in order to measure the amount of fluoride and fluoride removal by bauxite. In this study, a slight increase in aluminium and silicon contents and the pH of the water after bauxite was used; however, the amounts were less than the WHO standards (15). On this basis, there was no need to measure Al and Fe contents.

The names of each mine and the chemical analyses of ore from each using XRF and XRD are presented in Table 1.

The greatest constituents in all mines were Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, and TiO<sub>2</sub>. The main phases of most mines were diaspore, anatase and boehmite.

The maximum fluoride removal efficiency rate achieved using bauxite from the studied mines and the optimal amount of bauxite, contact time, and pH for both the synthetic and actual water samples from Kuhbanan are given in Table 2.

The results regarding the pseudo first order and pseudo second order kinetics for initial fluoride concentrations (2, 4, 6 mg) are presented in Table 3.

The Langmuir and Freundlich isotherm parameters for the adsorption of fluoride using 5 g of activated bauxite from ore mined in Iran, pH = 7, and initial fluoride concentrations of 2, 4, and 6 mg/L are given in Table 4.

## Discussion

The results showed that the ore from all active bauxite mines in Iran (with the exception of Nilchian, Hasanabad, and Shomal-e Yazd mines) had the required qualities for fluoride removal from drinking water samples and could achieve much lower concentrations than the WHO guidelines (from 2.74 mg to less than 1.5 mg) without any preparation or modification. The bauxite particles from Jajarm, Tash, Mendon, and Sadrabad mines proved more efficient than that from other mines in adsorbing fluoride from drinking water. The differences in performance are associated with several factors, such as the composition of the adsorbent.

Table 1 shows that the ore from all studied bauxite mines have aluminium, silicon, titanium, and iron oxide (the four main elements of bauxite). The fluoride removal efficiency is correlated with the percentage of Al<sub>2</sub>O<sub>3</sub> and Fe<sub>2</sub>O<sub>3</sub> compounds. Percentages of silicon oxide (SiO<sub>2</sub>) and titanium oxide (TiO<sub>2</sub>) in each of the 11 studied mines were higher than the percentages reported in other studies. Gomoro et al in Ethiopia used 3 lateritic soils including red gullale soil (RGS), yellow gullale soil (YGS), and ambo soil

**Table 1.** Chemical analysis of Iranian active bauxite mines by XRF and XRD

Name of mine	XRD pattern of the bauxite ore mines	Percentage of chemical compounds of Bauxite by XRF							
	The main minerals of bauxite	P <sub>2</sub> O <sub>5</sub>	K <sub>2</sub> O	MgO	CaO	Fe <sub>2</sub> O <sub>3</sub>	Al <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	SiO <sub>2</sub>
Tash	Diaspore Kaolinite Chamosite	0.05	0.52	0.48	0.66	20.60	49.31	3.98	10.39
Nilchian	Diaspor-Boehemite-Anatase-Kaolinit	0.05	0.12	0.13	0.42	1.07	53.90	2.78	30.51
Mendon	Boehemite-Hematite-Kailinite	0.06	0.05	0.17	0.29	19.51	53.64	2.48	12.43
Jajarm	Diaspore- Anatase -Chamosite	0.09	0.22	0.69	1.53	21.06	51.05	5.56	9.82
Biglar	Dioaspore-Anatase-Hematite-Chlorite	0.16	0.65	0.12	0.44	38.50	24.96	6.18	11.87
Khidbas	Boehemite-Hematite-Kaolinite	0.07	0.25	0.33	0.44	20.97	40.04	2.43	24.00
Sadrabad	Diaspore-Hematite-Cholorite-Anatas	0.10	0.03	2.35	0.52	24.77	45.77	5.87	10.20
Hassanabad	Diaspore-Anatase	0.12	0.99	0.04	0.45	1.89	59.90	11.60	9.31
Shahbalaghy	Kaolinite-Chamosite-Anatase	0.09	<0.03	0.67	0.29	39.80	21.85	3.79	16.97
Kaezri	Boehemite-Kaolinite	0.08	0.20	0.19	0.30	17.67	44.36	3.76	13.28
Shomal-e-Yazd	Diaspore-Hematite-Dolomite-Quartz	0.09	1.00	3.35	6.62	19.93	34.38	4.74	17.27

**Table 2.** Maximum efficiency of fluoride removal by using bauxite of studied mines with optimal amounts of bauxite, contact time and pH for synthetic and water samples of Kuhbanan

Entry	Name of mine	Contact time (min)	pH	Fluoride removal from Kuhbanan drinking water (2.74 mg/L)	Efficiency of fluoride removal from synthetic samples (2 mg/L)	Adsorbent dose (g)
1	Tash	120	7	61.67	70.39	25
2	Nilchian	180	6	30.19	60.16	35
3	Mendon	120	7	56.64	85.75	20
4	Jajarm	90	7	58.15	93.12	20
5	Biglar	180	6	49.31	66.47	25
6	Khidbas	180	6	52.69	80.41	30
7	Sadrabad	180	7	53.21	84.69	25
8	Hassanabad	180	6	36.05	62.72	30
9	Shahbalaghy	180	6	46.05	77.17	25
10	Kaezri	180	7	50.87	82.91	35
11	Shomal-e-Yazd	180	7	36.68	64.74	30

**Table 3.** The results of investigation of pseudo first and pseudo second order of kinetics for initial fluoride concentrations (2, 4, 6 mg)

Entry	Name of mine	Fluoride concentrations mg/L	First class synthetic			Second class synthetic		
			$K_1$	$q_e$	$R^2$	$K_2$	$q_e$	$R^2$
1	Tash	2	0.023	0.562	0.976	0.148	0.303	0.985
		4	0.023	0.707	0.956	0.062	0.501	0.967
		6	0.016	0.664	0.959	0.086	0.494	0.979
2	Nilchian	2	0.013	0.540	0.975	0.292	0.053	0.911
		4	0.018	0.652	0.998	0.419	0.047	0.933
		6	0.018	0.648	0.950	0.466	0.049	0.933
3	Mendon	2	0.023	0.553	0.942	0.218	0.347	0.991
		4	0.027	0.762	0.988	0.116	0.613	0.988
		6	0.034	0.810	0.940	0.156	0.667	0.989
4	Jajarm	2	0.018	0.567	0.958	0.138	0.339	0.984
		4	0.032	0.748	0.982	0.090	0.543	0.983
		6	0.018	0.816	0.872	0.025	0.754	0.906
5	Biglar	2	0.009	0.484	0.950	0.089	0.254	0.929
		4	0.011	0.634	0.979	0.043	0.447	0.919
		6	0.006	0.710	0.984	0.027	0.580	0.887
6	Khidbas	2	0.016	0.549	0.962	0.063	0.320	0.940
		4	0.013	0.752	0.973	0.025	0.683	0.842
		6	0.013	0.816	0.968	0.024	0.803	0.866
7	Sadrabad	2	0.009	0.538	0.936	0.078	0.331	0.939
		4	0.011	0.692	0.975	0.043	0.559	0.948
		6	0.013	0.800	0.987	0.035	0.734	0.954
8	Hassanabad	2	0.009	0.478	0.975	0.063	0.248	0.883
		4	0.009	0.648	0.985	0.025	0.484	0.851
		6	0.006	0.653	0.965	0.029	0.501	0.878
9	Shahbalaghy	2	0.011	0.502	0.886	0.222	0.296	0.991
		4	0.011	0.653	0.909	0.105	0.563	0.990
		6	0.020	0.669	0.949	0.114	0.656	0.996
10	Khezri	2	0.011	0.521	0.977	0.062	0.292	0.940
		4	0.009	0.661	0.982	0.034	0.509	0.928
		6	0.009	0.714	0.950	0.029	0.623	0.932
11	Shomal-e-Yazd	2	0.004	0.511	0.780	0.146	0.223	0.949
		4	0.004	0.644	0.784	0.154	0.352	0.989
		6	0.004	0.747	0.888	0.042	0.494	0.891

**Table 4.** Langmuir and Freundlich isotherm (5 g/L bauxite, pH = 7, initial fluoride concentrations 2, 4 and 6 mg)

Entry	Name of mine	Freundlich isotherm			Langmuir isotherm		
		n	$k_f$	R <sup>2</sup>	$q_m$	$k_l$	R <sup>2</sup>
1	Tash	3.484	0.616	0.884	0.504	2.319	0.991
2	Nilchian	2.906	0.540	0.980	0.470	1.090	0.999
3	Mendon	3.225	0.732	0.908	0.712	2.900	0.997
4	Jajarm	2.801	0.677	0.994	0.718	1.500	0.998
5	Biglar	1.766	0.526	0.981	0.750	0.438	0.995
6	Khidbas	1.795	0.623	0.915	0.899	0.645	0.955
7	Sadrabad	1.992	0.513	0.998	0.581	0.603	0.982
8	Hassanabad	1.992	0.513	0.917	0.581	0.603	0.968
9	Shahbalaghy	1.988	0.664	0.873	0.859	0.926	0.959
10	Kaezri	1.751	0.563	0.977	0.841	0.476	0.992
11	Shomal-e-Yazd	2.016	0.521	0.993	0.663	0.485	0.964

(AS) to remove fluoride from drinking water (9). Under oxidative conditions and as a result of the weathering of rocks, this soil loses silica. The main component of this soil depends on lateritic phase. The last step is the formation of bauxite. Maximum fluoride removal using 2 g/L of RGS soil was 32.32%. The findings of a more complex fluoride formation with RGS soil could be due to the presence of high levels of alumina ( $Al_2O_3$ ) (9).

In a study conducted in Tanzania in 2009, Peter used two types of kaolinite and bauxite-rich soil to remove fluoride (29). His results showed that soil rich in bauxite had a higher adsorption capacity than soil rich in kaolinite. He identified the chemical and mineral composition of soils as reasons for different removal rates. Soil chemical analyses revealed that soil rich in bauxite had a large amount of aluminum oxide compared to soil rich in kaolinite. Peter introduced aluminum oxide as a good adsorbent for fluoride removal due to the Al and F ions reaction (29). XRF analyses of ore from all bauxite mines achieved results similar to those of Altundoğan and Tümen who used bauxite in phosphate removal (30), Ayoob et al who used arsenic in bauxite removal (31), Sujana et al (8), Sajidu et al (25), and Malakootian et al (32) who used bauxite in fluoride removal. However, there are differences in the percentages of major bauxite compositions among all studies. Gomoro et al indicated that changes in the various formation stages of bauxite are reasons for different percentages of bauxite ore compounds in various locations (9). According to the results of the presented XRD patterns, all mines included the same minerals, such as bauxite, diaspore ( $AlOOH$ ), anatase ( $TiO_2$ ), and quartz ( $SiO_2$ ). Diaspore is from the aluminum hydroxide family, and this can be the main factor in removing fluoride with bauxite. High amounts of anatase and kaolinite in some mines can cause decreasing fluoride removal efficiency. Peter attributed the low fluoride removal efficiency using kaolinite to the high content of  $SiO_2$  (50.60%) (29). Gomoro et al found that anatase and quartz rarely have a good adsorption capacity for fluoride removal (9). With the ore from all bauxite mines in Iran, with a fixed

amount of adsorbent (5 g/L), initial concentrations of fluoride (2, 4, and 6 mg/L), and over time, the removal efficiency increased to the maximum extent. However, it did not increase for a longer time. After equilibrium time, the repulsive forces between the pollutant on the adsorbent and contaminants in the solution prevented contact between the pollutants and the adsorbent. The shortest time to reach equilibrium was 90 minutes, achieved using bauxite from the Jajarm mine; the longest time to reach equilibrium was 180 minutes, achieved using bauxite from Khidbas, Khezri, Sadrabad, Shomal-e-Yazd, Biglar, Hasanabad, Nilchian, Mendon, Tash, and Shahbalaghy mines. The equilibrium time for bauxite from Tash mine was 120 minutes. In all cases, equilibrium time was independent of the initial concentration of fluoride. Peter used soils rich in bauxite and kaolinite to remove fluoride from water with high concentrations of fluoride. He achieved a range of 90 to 180 minutes as the maximum time to achieve efficiency of fluoride removal by both soils. Peter expressed that saturation of the ion-exchange positions explained achieving equilibrium over time (29). Sujana et al used bauxite to remove fluoride. Equilibrium time was calculated at 120 minutes for concentrations of 4 and 12 mg-F (8). Atasoy et al used raw and modified bauxite in fluoride removal and reached equilibrium after 3 hr. They found that the removal rate was fast at first, because all the absorption positions were empty and free fluoride ions were available (26). Evaluations of the effect of time on removal efficiency in the mentioned studies revealed results similar to those of the present study.

One factor that has a significant impact on the adsorption process is pH. Basically, bauxite is a mixture of iron, aluminum, silica, titanium oxide, and hydroxide. When bauxite is hydrated at neutral pH, a net positive or negative charge will be formed which is able to adsorb the fluoride anion. According to the results for bauxite from all 11 mines, the highest fluoride removal efficiency was observed at neutral pH, and the lowest fluoride removal efficiency was observed at acidic and alkaline pHs. The reduction in removal efficiency at pH <5 could

be attributed to the formation of poor hydrofluoric acid (HF). The reduction in efficiency in alkaline pHs could be due to competition between hydroxide ions ( $\text{OH}^-$ ) and fluoride ions ( $\text{F}^-$ ) in the absorption process. Shrivastava and Vani reviewed fluoride removal methods and found that the maximum fluoride absorption by alumina takes place in pH 5-7. They justified reduction efficiency at pHs less than 5 and greater than 7 with these two reasons: 1) At pH <5, activated alumina is insoluble in water, and that leads to the loss of surface absorption; and 2) at pH >7, silicate and hydroxide have a stronger bond with the positions of absorption toward fluoride ions (33). These results concur with those of Sujana et al (pH = 6.4) (8), Malakootian et al (pH = 7) (34-36), and Sajidu et al (pH = 7) (15). At acidic and alkaline pHs, they showed the same trend.

Initial fluoride concentration is the most effective factor for fluoride ion transfer from the liquid phase to a solid phase adsorbent (37). The results showed that removal efficiency rates are higher in low concentrations of fluoride. By using fixed amounts of all bauxite samples, increasing the fluoride concentration from 2 to 6 mg/L increased the absorption capacity, but decreased the removal efficiency. A study by Atasoy et al showed that increasing the fluoride concentration from 1 to 10 mg/L reduced the removal efficiency from 66% to 36% (26). Lv et al used double-modified hydroxides with polycarbonate, aluminum, and manganese in fluoride removal. By increasing the fluoride concentration from 20 to 200 mg/L (in a fixed amount of adsorbent), the residual fluoride was changed from 0.4 to 50 mg/L. This result showed that the fluoride removal efficiency increased when the initial concentration of fluoride was decreased. The results of the mentioned studies were consistent with the results of the current study (38).

The results showed that by increasing amount of bauxite in all studied samples, the removal efficiency was increased, but the absorption capacity was decreased. Efficiency was almost constant with the addition of higher absorption. The maximum adsorption efficiency was 46.58%, obtained using 2 g/L of bauxite from the Sadrabad mine in 6 mg/L of fluoride concentration. In similar conditions, when bauxite was increased to 25 g/L, the adsorption efficiency was 75.31%. Increasing the amount of adsorbent made more active positions available for fluoride ions, but the removal efficiency did not show any dramatic increase for values higher than 25 gr/L; in this amount only a limited amount of free fluoride ions remained. A study was conducted in Iran in 2011 by Malakootian et al to remove fluoride using regenerated spent bleaching earth. In this study, increasing the amount of adsorbent from 10 to 20 g/L (concentration of fluoride = 2.5 mg/L) increased removal efficiency from 66.41% to 74.01% (34). In India, Mohapatra et al used mineral oxides to remove fluoride. Similar behavior was observed with increased amounts of

adsorbent (39).

In this study, fluoride removal using bauxite from the Sadrabad, Shahid Nilchian, Biglar, Khidbas, Hassanabad, and Khezri mines and using pseudo first order kinetics with a correlation coefficient greater than 0.950 showed better results than pseudo second order kinetics. The results of this study were consistent with those of other similar studies, such as Mohapatra et al (39), Das et al (40), and Sujana et al (8).

Fluoride removal using bauxite from the Shomal-e-Yazd, Tash, Mendon, Jajarm, and Shahbalaghi mines and using pseudo second order kinetics with a correlation coefficient greater than 0.984 showed better results than the pseudo first order kinetics. Thole (24) and Tor et al (41) found similar results. Analysis of the data regarding fluoride absorption on bauxite particles by isotherm showed that using bauxite from the Jajarm, Khidbas, Khezri, Shahid Nilchian, Mendon, Hassanabad, Biglar, Shahbalaghi, and Tash mines with a correlation coefficient greater than 0.955, fluoride absorption obeyed the Langmuir adsorption isotherm. Sujana et al, Malakootian et al, and Das et al used bauxite as a fluoride adsorbent, and their experimental data was better described by the Langmuir adsorption model (8,34,40). These mentioned studies are consistent with this study.

Fluoride absorption by bauxite from the Sadrabad and Shomal-e Yazd mines with a correlation coefficient greater than 0.993 obeyed the Freundlich adsorption isotherm. Lavecchia et al (20) and Wang et al (42) obtained similar results. In this study, 20 g/L of bauxite from the Jajarm and Mendon mines, 25 g/L of bauxite from the Sadrabad, Biglar, Shahbalaghi and Tash mines, 30 g/L of bauxite from the Hassanabad, Khidbas, and Shomal-e Yazd mines, and 35 g/L of bauxite from the Khezri and Shahid Nilchian mines was added to the drinking water of Kuhbanan as the actual sample. Under these conditions and with no change to the pH of the water, fluoride concentrations changed from 2.74 mg/L to 1.15, 1.14, 1.28, 1.42, 1.48, 1.48, 1.75, 1.30, 1.74, 1.35, and 1.91 mg/L, respectively. Generally, under the same conditions, the removal efficiency is less in actual samples than in synthetic samples. The presence of interfering ions in the actual samples (e.g., nitrate, sulfate, chloride) compete with fluoride to adsorb the iron and aluminum of the bauxite, resulting in a decrease in the removal efficiency of the actual sample. Sujana et al used bauxite to remove fluoride from groundwater (8). At pH = 6, increasing the sulfate, nitrate, and phosphate concentrations (0-20 mg/L) in solution caused a reduction in fluoride removal, and increasing the number of carbonate ions had a negligible effect on absorption (43). The absorption rate was lower in actual samples with normal pH values than in synthetic samples. In addition to interfering anions, water pH can cause a reduction. Das et al reported water pH as a reason for the reduction in absorption efficiency using activated titanium rich

bauxite in the groundwater samples (40). According to the results, bauxite from the mines of Jajarm, Mendon, Sadrabad, Khezri, Khidbas, Shahbalaghi, Tash, and Biglar performed as needed to reach the WHO guidelines (<1.5 mg/L) to remove fluoride from actual (Kuhbanan drinking water with 2.74 mg/L fluoride concentration) and synthetic samples (2, 4, and 6 mg/L concentrations of fluoride) with no initial preparation and as a raw material; however, the residual concentration of fluoride in the drinking water samples after using bauxite from the Hassanabad, Shahid Nilchian, and Shomal-e Yazd mines was greater than the WHO guidelines (>1.5 mg/L). The highest removal efficiency obtained using bauxite was seen at the Jajarm mine and equaled 93.12% for synthetic samples and 58.15% for actual samples from Kuhbanan. The use of the following methods is suggested to increase the fluoride removal efficiency, it is suggested that raw bauxite be improved by milling it to nanometer-sized particles, a high sorbent be used in actual samples, or the contact time with adsorbent be increased, and bauxite be modified using acid or heat.

### Conclusion

Bauxite from the Jajarm mine had a higher removal efficiency (58.15%) than the other studied mines. The removal efficiency rates of the other mines were as follows: Jajarm > Mendon > Sadrabad > Khidbas > Khezri > Shahbolaghi > Tash > Biglar. Bauxite from the Shomal-e Yazd, Hasanabad, and Shahid Nilchian mines were not sufficiently efficient without initial preparation and modification. Although the removal efficiency in actual samples is much lower than that in synthetic samples, its low cost, abundant availability, and no need for expertise and sophisticated technology are beneficial. The removal efficiency of this adsorbent can be increased to the desirable level using corrective methods such as heating, acidifying, particle crushing, and the mixing of two or more removal systems.

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### Ethical issues

The author hereby certifies that all data collected during the study is as stated in the manuscript, and no data from the study has been or will be published separately elsewhere.

### Competing interests

The author declares that he has no competing interests.

### Authors' contributions

The author contributed and is involved in the suggestion of the problem, design of experiments, data collection, and article approval.

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