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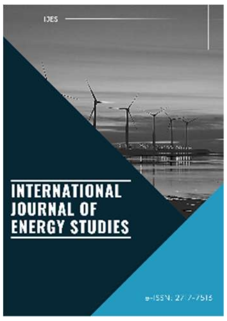

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A systematic study on adsorptive removal of arsenic using low cost adsorbents with or without the presence of NOM

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Highlights

- Steel slag showed higher adsorption capacity for arsenic removal.
- The results showed that the arsenic adsorption data were well fitted by modified Freundlich isotherm.
- Either with or without the presence of NOM, the arsenic adsorption reached steady-state within 24 h.
- Natural organic matter has a significant adverse effect on arsenic adsorption by natural/waste materials.

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ABSTRACT

Arsenic is a toxic and bio-accumulating element in water bodies, especially in drinking water sources.One of the methods used to remove arsenic from drinking water is adsorption. NOM (natural organic matter), which is ubiquitous in natural aquatic environments, can inhibit arsenic adsorption by competing with arsenic for adsorbent surface sites. The main objectives of this study were to investigate the competitive adsorption between NOM and arsenic using low-cost adsorbents. For this purpose, batch adsorption experiments were carried out in synthetic and natural water using pumice, steel slag and red mud as low cost adsorbents.The highest arsenic removal efficiencies obtained with steel slag, pumice and red mud were 95, 95 and 50 %, respectively without the presence of NOM. The results showed that the presence of NOM caused a significant reduction in arsenic adsorption for all tested adsorbents and 20-30% less arsenic removal efficiency was obtained in natural water compared to the removal efficiencies obtained in synthetic water. The maximum adsorption capacities obtained in synthetic water with all tested adsorbents vary between 1.6 -3.05 mgAs/g, while the maximum adsorption capacities obtained in natural water vary between 0.9-1.2 mgAs/g. All these findings indicate that NOM can hinder the uptake of arsenic by all tested adsorbents. Moreover, the arsenic adsorption in natural water were modelled by Langmuir, Freundlich and modified Freundlich. The results showed that the arsenic adsorption data were well fitted by modified Freundlich isotherm with R^2 values between 0.87 and 0.90.

Keywords: Pumice, Red mud, Steel slag, Arsenic, Adsorption, Background effect, NOM

1. INTRODUCTION

Arsenic is a metalloid that exists in many parts of the world such as soil, sediments, water. Arsenic exists in 4 oxidation states: -3, +3, 0 and +5 in nature. The most common constituents of arsenic found in nature are arsenite and arsenate [1]. It is known that arsenic is a carcinogen that causes various health effects on humans and other living organisms. Human exposure to arsenic is usually through the consumption of arsenic-contaminated water [2]. The World Health Organization (WHO) and the United States Environmental Protection Agency (USEPA) have recommended a maximum pollutant level (MCL) of 10µg/L for arsenic in drinking water [3], [4]. Also, the maximum amount of arsenic specified in the regulation on 'Guidelines for Drinking Waters' by The Turkish Ministry of Health is 10µg/L in Turkey [5]. Arsenic pollution in drinking water is a global problem and high arsenic concentrations have been detected in surface and ground waters in many regions in Turkey [6].

Many extensive studies have been carried out to remove arsenic from waters. The technologies that are currently in used include ion exchange, oxidation, adsorption, coagulation, chemical precipitation, and membrane filtration. Among these available techniques, adsorption has a wider applicability in water treatment because it is a simple, easily accessible and economical technique and has proven to have high efficiency in arsenic removal [7]. Ligand exchange, chemisorption, anion exchange and electrostatic interaction are main mechanisms for arsenic removal using adsorbents. Numerous adsorbents have been used to arsenic removal including biosorbents, iron oxides, activated carbons, resins, agricultural and industrial wastes [8]. It is extremely important that adsorbents should be economical and have high efficiency [9] and the research continues for adsorbents that provide benefits for the sustainable environment. Until now, various natural/waste materials such as clay [10], zeolite [2], pumice [11], fly ash [12] red mud [13] have been used as promising adsorbents. The efficacy of these materials in removing arsenic has been linked to metal-binding functional groups such as carboxyl and hydroxyl [14]. In addition, some studies have applied various pretreatments to the materials to increase the adsorption capacity of these materials. For example; Zhang et al [15], provided the modification of red mud using 0.5M $FeCl_3$ $6H_2O$ and stated that the modified red mud has higher adsorption capacity. Similarly, Kırkan and Bekaroğlu [11] and Far et al., [16] coated the pumice with iron oxide, [17] Lekic et al. treated the slag with sulfuric acid and they obtained the effective adsorbents in arsenic removal.

Pumice is a natural and low cost material that is widely used in water treatment thanks to its porous structure. Steel slag and red mud particles are industrial by products formed during alumina and steel production, respectively. Since these particles contain components such as iron oxide and calcium oxide, they can be used as an adsorbents in water treatment.

Natural organic matter is a complex mixture of organic compounds that are naturally present in raw water and cause a number of problems in drinking water quality and water treatment operations [18]. Several studies have shown that NOM is a component that controls the adsorption of other ions in water to the adsorbent surface [19] , [20], [21]. Moreover, NOM also affects arsenic adsorption in water depending on factors such as surface chemistry and pore structure of adsorbents. The studies conducted by Weng et al. [22] and Otero-Fariña et al. [21] have also shown that NOM inhibits arsenic adsorption. NOM is likely to affect the adsorption of arsenic-contaminated waters, since natural organic substances are commonly found in natural waters [23]. In this context, the purpose of this study was to reveal the competitive effect of NOM on arsenic removal by steel slag, pumice and red mud particles. For this purpose, batch adsorption tests were carried out using steel slag, pumice and red mud in water with NOM (natural water) and without NOM (arsenic solution) and it was determined how the NOM affects arsenic adsorption.

2. MATERIALS AND METHODS

2.1. Materials

Two different water sources were used in this study, including synthetic arsenic solution and real water. Arsenic stock solution (1000 mg/L) was prepared by dissolving a known amount of $HAsNa_2O_4.7H_2O$ in distilled and deionized water (DDW). It was then diluted with DDW to an initial concentration of 400 µg As/L. This concentration was chosen based on the fact that drinking water sources in Turkey contain between 10-900 µg L-1 of arsenic [24]. In addition to the adsorption experiments carried out with synthetic solution, adsorption experiments were also carried out in natural raw water to understand the competitive effect of NOM on arsenic removal. For this purpose, water samples taken from Lake Eğirdir were used. Water quality parameters such as pH, conductivity, DOC of Lake Eğirdir were carried out in accordance with the Standard Methods for Examination of Water and Wastewater, and some parameters are presented in Table 1 [25]. Lake Eğirdir has a dissolved organic concentration (DOC) of 3 mg/L with low arsenic concentration. For this reason, in order to compare natural water experimental results with synthetic solution test results, the prepared arsenic stock solution was spiked to have an initial arsenic concentration of 400 µg/L.

Table 1. Water quality parameters for the Egirdir Lake

Parameters	**Unit**	
pH		8.9
Conductivity	µS/cm	412
Total Alkalinity (as $CaCO_3$)	mg/L	235
Total Hardness (as $CaCO_3$)	mg/L	214
DOC	mg/L	3
$SUVA_{280}$	L/mg DOC.m	0.7
Arsenic	µg As/L	[a]

[a]:Below detection limit

As adsorbent, natural and waste materials were used in arsenic removal. Pumice was chosen as the natural material, red mud and steel slag were chosen as the waste material. Pumice (Isp), steel slag (SS) and red mud (RM) samples were obtained from Isparta BIMS Building Elements Industry, Eregli Iron and Steel Production Facility and Etibank-Seydisehir Aluminum Production Facility, respectively and used without any modification (Figure 1). The particle sizes and some physicochemical properties of the tested particles are shown in Table 2 [6]. The detailed information about the preparation and properties of the particles can be found in [19].

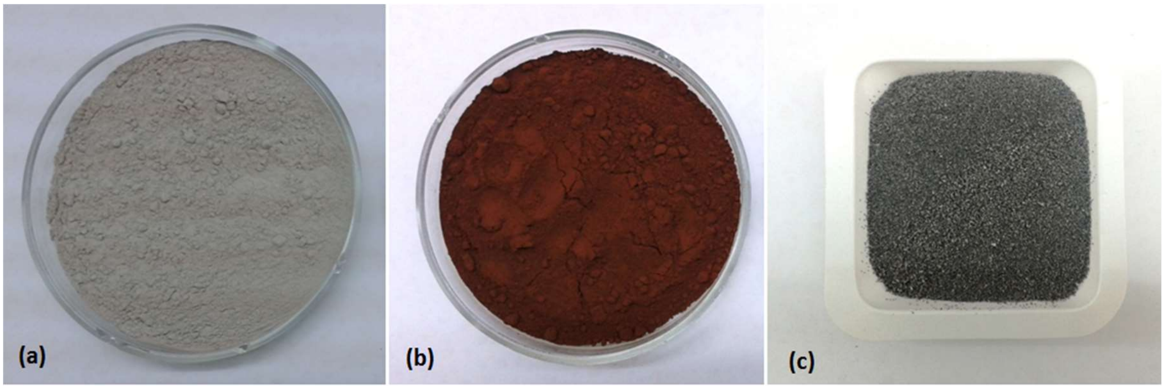

Figure 1. (a) Pumice (<63 µm) ; (b) Red Mud (<250 µm) ; (c) Steel Slag particles(<250 µm)

Table 2. The physicochemical Characteristics and important components of the tested particles

Adsorbents	BET Surface area (m^2 /g)	Particle Size (µm)	pHpzc	Fe_2O_3 (%)	CaO (%)	Al_2O_3 (%)
SS	<2	<250	11.4	26.7	40.96	2.58
Isp	14	<63	9	3.28	3.91	17.30
RM	10	<250	9.9	34.8	4.07	22.42

Abbreviations: SS, Steel Slag (<250 µm);Isp, Isparta Pumice (<63µm); RM, Red Mud (250 µm).

2.2. Analytic Methods

Arsenic was analyzed using inductively coupled plasma optical emission spectroscopy (ICP-OES) according to Standard Methods 3120B. The ICP-OES has a detection limit of 10 µg/ L for arsenic.

2.3. Experimental Procedure

Batch adsorption experiments were carried out at constant temperature (20°C) with the following experimental procedure. (1) The pH of the natural or synthetic water samples was adjusted to 7.0 using 1 M HCl (Merck, ACS reagent 37 %,) and/or 1 M NaOH (Merck, Reagent grade ≥98%). (2) A known amount of adsorbent and 100 mL of arsenic solution/natural water were added to amber bottles with a volume of 130 mL. (3) The samples were shaken at 150 rpm in rotary shaker for a known constant time. (4) At the end of the specified time, the samples were filtered to remove particles before arsenic measurement (5) The filtered samples were analyzed for arsenic. The adsorbed arsenic amount per unit adsorbent, q, (mg/g) was calculated as follows:

$$q = \frac{C_0 - C}{m} V \tag{1}$$

where C_0 is the initial arsenic concentration (mg/L); C is the residual arsenic concentration in equilibrium (mg/L); V is the volume of solution (L); m is the mass of adsorbent in the solution (g).

The isotherm experiments were tested at varying time intervals and varying adsorbent doses. Initial kinetic experiments indicated that 24 h was sufficient to reach adsorption equilibrium. For this reason, 24 h of mixing was selected for all batch experiments. After the kinetic tests, the adsorption isotherm tests were carried out at different adsorbent doses (0.05; 0.25; 0.5; 1; 2; 4; 6; 8 and 10 g/L) during the 24 hour equilibrium period.

3. RESULTS AND DISCUSSION

Firstly, isotherm tests were performed in synthetic arsenic solution (without NOM) at different adsorbent doses ranging from 0.05 to 10 g/L. Figure 2 shows the arsenic removal efficiencies obtained in synthetic water with the tested adsorbents. All tested adsorbents provided higher arsenic removal at higher doses. SS was the most effective adsorbent in arsenic removal among all tested adsorbents. It is seen that the steel slag contains components such as 26.7 % Fe_2O_3 and 40.96% CaO (Table 2). For this reason, these components can be held responsible for obtaining high arsenic removal efficiencies with SS particles. The highest arsenic removal efficiencies obtained with steel slag, pumice and red mud were 95, 95 and 50 %, respectively.

As can be seen from Table 2, all tested particles contain a high proportion of iron oxide. The adsorption of arsenic by adsorbents containing iron oxides is generally based on ligand exchange reactions [15, 26]. Arsenic adsorption occurs by the displacement of Ca/Fe coordinated surface hydroxyl groups in adsorbents with arsenic ions.

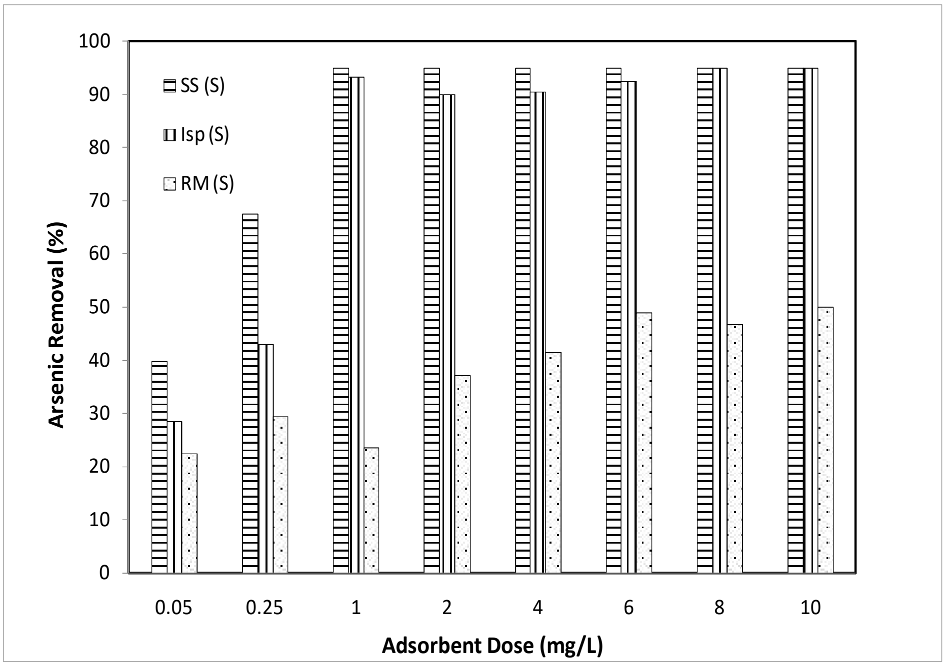

Figure 2. Arsenic removal efficiencies in synthetic arsenic solution by the tested particles. (Codes of Adsorbents: SS, Steel Slag (<250 µm); Isp, Isparta Pumice (<63µm; RM, Red Mud (250 µm))

After synthetic arsenic solution tests, natural water tests were performed to evaluate the effects of NOM on arsenic adsorption. The arsenic removal efficiencies obtained in natural water and synthetic arsenic solution are given comparatively in Figure 3. Similar to synthetic arsenic solution tests, steel slag and pumice particles were effective in removing arsenic in natural water. At 4 g/L dose, 55% and 90.5% arsenic removal effiencies were obtained by Isp and SS, respectively in natural water. In natural water, maximum arsenic removal efficiencies of 72.3, 71.8, 37.3% were obtained with Isp, SS and RM particles, respectively (at a dose of 10 g/L adsorbent). However, the removal efficiencies obtained in natural water were lower than those founded in synthetic solution. The removal efficiencies obtained at high doses in natural water were achieved at lower doses in synthetic water experiments. For example, 67.5% arsenic removal was achieved at 0.25 gr/L with SS, while 93.25 % removal efficiency was achieved at 1 g/L with Isp in arsenic solution. This decrease in arsenic removal in natural water is due to the presence of other ions present in the water, especially NOM. As a general trend, it can be said that the presence of NOM in natural water reduces the adsorption of arsenic with all particles by 20-25%.

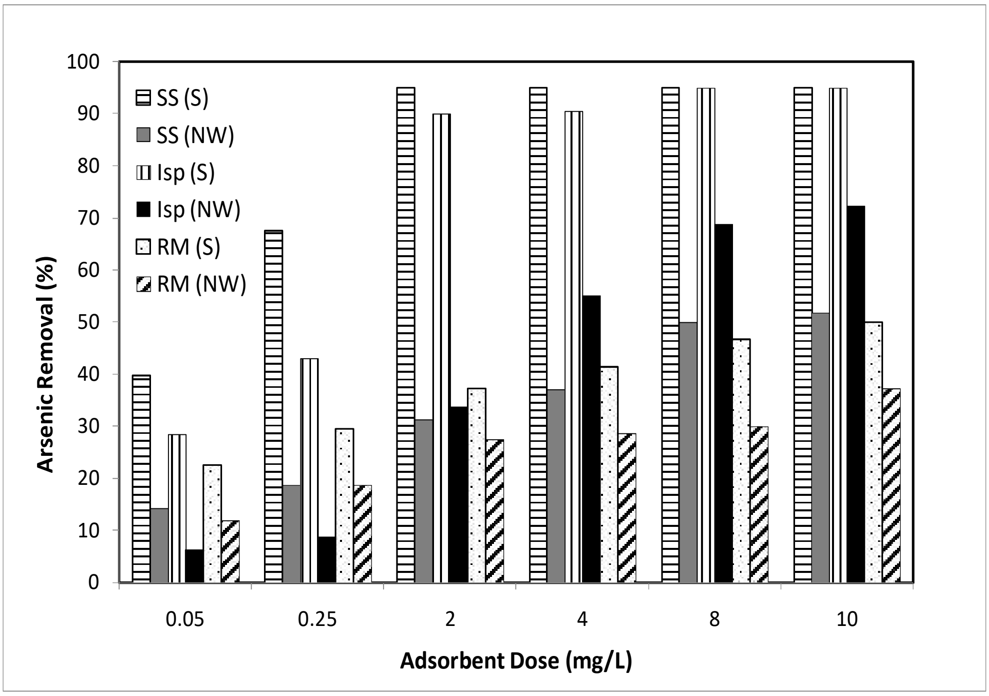

Figure 3. Comparison of the removal efficiencies obtained in synthetic water and natural water by the tested particles. (Codes of Adsorbents: SS, Steel Slag (<250 µm); Isp, Isparta Pumice (<63µm); RM, Red Mud (250 µm), S: Synthetic arsenic solution, NW: natural water).

Figure 4 shows the adsorption capacities calculated for all adsorbents tested in natural water and synthetic water. A similar trend has been observed in adsorption capacities to arsenic removal efficiencies. The adsorption capacities obtained in natural water are lower than the adsorption capacities obtained in synthetic water. For example, the maximum adsorption capacity obtained in natural water by Isp was reduced to 1.2 mgAs/g while the maximum adsorption capacity obtained in synthetic arsenic solution was 2.192 mgAs/g. Similarly, 3.058 mgAs/g adsorption capacity was obtained in natural water by SS particles, while a maximum adsorption capacity of 1.14 mgAs/g in synthetic water was achieved. Similar results were also observed in studies conducted by Weng et al, [22], Redman et al [23] and Liu et al [27]. For example, Liu et al [27], stated that the amount of arsenic adsorbed decreased significantly in the presence of 8 mg/L NOM. Similarly, Weng et al [22], reported that the adsorption of humic acids (HA) and (fulvic acids (FA) with goethite leads to desorption of arsenate due to the competition of arsenate with HA and FA.

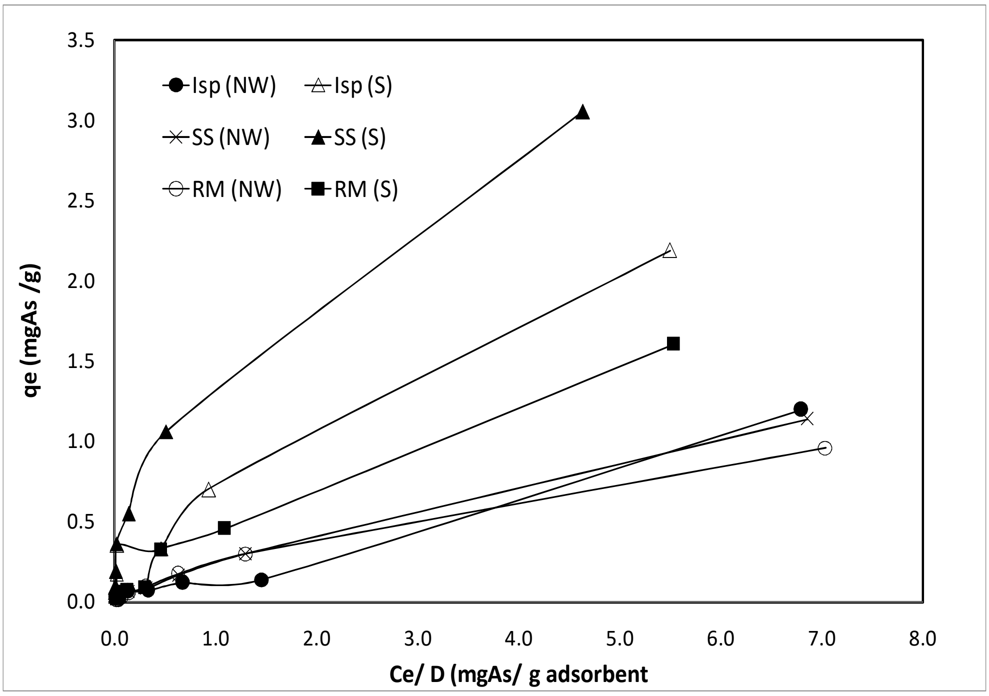

Figure 4. Arsenic adsorption capacities for the tested adsorbents in synthetic arsenic solution and natural water (SS, Steel slag (<250 µm); Isp, Isparta Pumice (<63µm); RM, red mud (250 µm), S: Synthetic water, NW: natural water).

In previous studies, the capacities obtained with various adsorbents used for arsenic removal from natural waters and aqueous solutions are listed in Table 3. The adsorption capacities of adsorbents vary according to the experimental conditions. However, it is possible to say that the Steel Slag used in this study is as effective as commercial activated carbons in arsenic adsorption (Table 3). Moreover, steel slag is more economical than commercial activated carbons in terms of the cost of adsorbent preparation and usage. The average commercial price of steel slag is about 3$/ton, while the prices of commercial activated carbons are 1000-2200$/ton [28, 29].

Table 3. Previous experimental results for arsenic onto some different adsorbents

Adsorbent	Maximum Adsorption Capacity	Water Sample Type	Reference
	mg/g		
Iron coated pumice	1.01	Synthetic	[16]
Iron Coated Pumice	1.92	Synthetic	[11]
Blast Furnace Slag	0.0476	Synthetic	[17]
Pisolite	1.4	River Water	[30]
Modified sepiolite	0.0906	Synthetic	[31]
Commercial Activated Carbon	2.72	Synthetic	[32]
Steel Slag	1.1	Lake Water	This study
Steel Slag	3.05	Synthetic	This study

In terms of modeling the arsenic isotherm data, Langmuir, Freundlich, Modified Freundlich (dose normalized) were used. Experimental data were consistent with modified Freundlich model. The Freundlich isotherm was developed for heterogeneous systems and its modified form (dose normalized) is expressed by the following equation.

$$q_e = K_f \left(\frac{C_e}{D_0}\right)^n \tag{2}$$

Where q_e represents the amount of As adsorbed per amount of adsorbent at the equilibrium (mg/g), Ce represents the equilibrium concentration (mg/L), D_0 (mg/L) represents adsorbent dosage and Kf (mgAs/g) and n (L/mg) are are the constants related to the adsorption capacity and intensity of adsorption, respectively. The K_f and n values as well as the R^2 values for the modified Freundlich model are given in Table 4. K_F values calculated according to the model are between 0.13 and 0.264, and the highest K_f value was obtained for SS particle. n values calculated for adsorbents vary between 0.31 and 0.783.

Table 4. Modified Freundlich isotherm model parameters for arsenic removal in natural water

	Modified Freundlich (Dose Normalized)		
Adsorbent Type	**K_f (mgAs/g)**	**n (L/mg)**	**R^2**
SS (NW)	0.264	0.664	0.886
RM (NW)	0.231	0.783	0.874
Isp (NW)	0.13	0.31	0.907

Abbreviations: SS, Steel Slag (<250 μm);Isp, Isparta Pumice (<63μm); RM, Red Mud (250 μm); NW, Natural Water

4. CONCLUSION

This study was conducted to examine the effect of the presence of NOM on the adsorption of arsenic by pumice, steel slag and red mud. For this purpose, batch experiments were carried out to determine the adsorption of arsenic in waters with (natural water) or without NOM (synthetic water). With the tested adsorbents, removal efficiencies of 50-95% were obtained from synthetic water and the highest adsorption capacities were observed by steel slag. The arsenic adsorption with and without NOM were modelled by Langmuir and modified Frendlich isotherms models , and modified Frendlich isotherms model gave a better fit for all tested waters. It has been found that NOM in natural water hindered the arsenic adsorption for all tested adsorbents. Due to the presence of NOM in natural waters, the amount of arsenic adsorbed decreased by 20-30%. This phenomenon occurred due to the NOM competing with arsenic for being adsorbed on to the pumice, steel slag and red mud. Despite the competitive effect of NOM, the results showed that natural/waste materials or by-products are promising adsorbents in removing arsenic and the potential use of natural/waste materials or by-products in arsenic removal that have a positive effect on the environment with the concept of sustainability.

NOMENCLATURE

WHO:World Health Organization
USEPA: United States Environmental Protection Agency
MCL: Maximum pollutant level
NOM: Natural organic matter
DDW:Distilled and deionized water

DOC: Dissolved organic concentration

ICP-OES: Inductively coupled plasma optical emission spectroscopy

HA:Humic Acid

FA:Fulvic Acid

SS:Steel Slag

RM:Red Mud

Isp:Isparta Pumice

NW: Natural water

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DECLARATION OF ETHICAL STANDARDS

The authors of the paper submitted declare that nothing which is necessary for achieving the paper requires ethical committee and/or legal-special permissions.

CONTRIBUTION OF THE AUTHORS

Seda Tözüm Akgül : Analyzed the results of the experiment and wrote the manuscript.

İbrahim Yıldıran Performed the experiments.

Turgut Erdem: Performed the experiments.

Nuray Ateş: Analyzed the results of the experiment.

Şule Kaplan Bekaroğlu: Analyzed the results of the experiment.

CONFLICT OF INTEREST

There is no conflict of interest in this study.

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