



# Treatment of Arsenic Contaminated Groundwater using *Arachis hypogaea*

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## Abstract

Arsenic has a detrimental effect on both the environment and humans. In many parts of the world, arsenic has been detected in drinking water sources above acceptable levels. Considering the fact that millions of people are at high risk of arsenic poisoning via water intake, arsenic remediation from drinking water has become a global issue of serious concern. Chemical coagulants are already being used for treatment but their enormous cost, human and environmental issues associated with their use have led to searching for alternatives like natural coagulants (plant-based). In this study, laboratory scale studies using jar test experiments were performed on synthetic arsenite contaminated groundwater to analyse the efficiency of *Arachis hypogaea* (groundnut seed) on the treatment of arsenic contaminated water. Experiments were carried out using synthetic arsenite contaminated borehole water with pH kept constant at 7.30. Results indicated that groundnut seed extract was able to significantly reduce arsenite in water either as primary coagulant or as coagulant aid (using alum as primary coagulant). As primary coagulant, above 90% arsenite reduction was achieved and as a coagulant aid about 99.97% arsenite reduction was achieved. The pH of the treated water was within neutral (basic) due to the buffering capacity of the seed extract. Therefore, results from this study revealed that groundnut seed has potential for use as primary coagulant or as coagulant aid (with alum as primary coagulant) for the treatment of arsenic contaminated groundwater.

**Keywords:** *Arachis hypogaea*, Arsenic, Arsenite, Coagulation, Contamination, Groundnut seed, Natural coagulants, Seed extract, Treatment, Water source.

## 1.0 INTRODUCTION

Water is imperative for all known forms of life even though it provide no calories or organic nutrients. It is vital for sustainable development and maintenance of the ecosystems [1]. Drinking water supply is a challenging situation in most part of the world, particularly in developing countries like Nigeria. Even though Nigeria is divided into three parts by rivers and about 10 out of the 36 states (excluding the Federal Capital Territory) in the country are named after water bodies [2], yet the problem of portable water supply seems to be exaggerating every day. In Nigeria, fresh water supply for both domestic and drinking purposes comes from three main sources namely: surface water (river, stream, pond, lake etc.), groundwater (borehole, hand dug well etc.) and rainwater [2-5]. Due to

anthropogenic activities (as a result of increased population) among other causes, these water sources are often polluted particularly the surface water source (due to its exposed nature). Thus, groundwater source is majorly used (for both domestic and drinking purposes) in most part of Nigeria with little or no treatment as it seems to contain lower levels of impurities [6]. The discharge of wastes (including municipal waste materials) into the environment is the major cause of water pollution as these wastes are washed into the aquatic ecosystem through soil erosion [2]. Further, the discharge of these waste into the environment can also lead to the released of toxic substances such as heavy metals into the environment as the waste are being decomposed by microorganisms through oxidative process [7].

Heavy metal pollution such as Mercury and Arsenic is one of the essential water contaminants associated with severe health impacts [2]. Arsenic (As) is a trace element which is highly toxic and carcinogenic and

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it constitutes approximately 0.00005% of the Earth's crust. It occurs naturally as the 20<sup>th</sup> abundant element in the Earth's crust, 14<sup>th</sup> in the seawater and 12<sup>th</sup> in human body [8, 9]. It is found at variable concentrations in the atmosphere, soils and rocks, natural waters [2]. Arsenic exist in both organic and inorganic form. In water it exists in inorganic form and it is occurring abundantly as arsenite ( $\text{As}^{\text{III}}$ ) and arsenate ( $\text{As}^{\text{V}}$ ) and arsenite is 60 times more toxic and mobile than arsenate [9]. Accumulation of arsenic in the human body mainly occurs through intake of arsenic contaminated drinking water and food (such as rice and vegetables) [9, 10]. Arsenic has a detrimental effect on both environment and humans. Its health's effects include skin lesion, liver damage, kidney and brain cancers and stomach poisoning etc. Both the World Health Organization (WHO) and the United States Environmental Protection Agency (USEPA) have decreased As threshold level for drinking water from  $50 \mu\text{g L}^{-1}$  to  $10 \mu\text{g L}^{-1}$  in 2001 [11, 12] due to its serious health effects. The presences of Arsenic (above permissible levels) has been detected in water (both groundwater and surface water) in many parts of the world including Bangladesh, India, China, Taiwan, USA, Canada, Brazil, Vietnam, Indonesia, Hungary, Mexico, and Pakistan [9], South Eastern Europe [13], Asia, North, and South America [14] and Nigeria [2, 15]. Considering the fact that millions of people are at high risk of arsenic poisoning via water intake, arsenic remediation from drinking water has become a global issue of serious concern.

In Nigeria, arsenic has been detected in drinking water sources (both surface water and groundwater) above acceptable levels of 0.01mg/l in the Northern region with a concentration of 0.02 to 0.80mg/l [2]. Such Northern regions with high arsenic contamination include Biu Volcanic North-Eastern Province of Nigeria [16], Kano [17], Zaria and environs [18], Kaduna [3, 4], Borno [19], and Sokoto [20] and also, it has been detected in high concentration in some places in south western Nigeria, such places include Osun and Ogun States [21-23]. Although, arsenic contamination is not too severe in the south-south region of Nigeria, however some levels have been detected in some areas and it's below the permissible limit of 0.01mg/l [2] for drinking water. Such areas include Bayelsa and Abia States [24-26]. Despite abundant water resources, Nigerians still do not have portable water and water supply is capital intensive (considering all the process it takes including treatment before getting to the consumer) such that without adequate funding, there is little the utility companies can do. Therefore, most of the populace rely heavily on untreated water sources, hence there is need for the development of cost effective material

which uses simple technique for the removal of arsenic from contaminated water.

Although conventional methods such as ion exchange, precipitation, membrane filtration, reverse osmosis, electro-dialysis [27, 28] and sorption [29, 30] have been applied for the removal of As from As contaminated water and these methods have been found successful, but the high chemical, energy requirements, complex techniques and very high construction, operation and maintenance cost are the major constraints in their applicability in water treatment processes [31], thus there is need to consider applying coagulation technique which is a relatively simple, efficient and low-cost arsenic removal technique and most especially convenient for application in the rural areas. In water treatment, coagulation is use for removing colloidal particles and other contaminants from water. Coagulants (either natural or chemical) are used to achieve this aim, due to their ability to agglomerate fine colloidal particles in water into larger ones (flocs) which settle easily and thus facilitate their removal [32]. Coagulants are classified as primary coagulants and coagulant aid. Primary coagulants neutralise the electrical charge of particles in water which result in particles clumping together while coagulant aids add density and toughness to slow-settling flocs so that they do not break up during the mixing and settling process. Removal of As from As contaminated water using both aluminium-based and metal-based coagulants such as alum and ferric chloride have been well documented [33]. But the relatively high cost and chemical nature of these coagulants and in some cases, high sludge production limit the widespread use of these chemical coagulants, hence it has become necessary to transform scientific knowledge on the use of locally available natural materials such as *Arachis hypogaea* (groundnut seed) as potential coagulant for the removal of arsenic from water. A lot of natural materials including agricultural and food-industry solid wastes (such as hazelnut and coconut shell, rice husk, pecan nut shell, jackfruit peel, sugarcane bagasse, water chestnut shell, and peels of various fruits) have been applied as biosorbents for the removal of As from water and they have been found successful in removing As from water.

*Arachis hypogaea* (Groundnut) is a major crop grown in the arid and semi-arid zone of Nigeria. The crop is mainly grown in the Northern part of Nigeria, as over 85% of the groundnuts produced in the country are accounted for by Kano, Kaduna, Taraba, Bauchi, Borno and Adamawa State [34]. It is either grown for its nut, oil or vegetative residue (haulms). Almost every part of groundnut including the seed is edible. *Arachis hypogaea* (Groundnut seed) contain high quality edible oil (50%),

easily digestible protein (25%) and carbohydrate (20%) [35]. Groundnut seeds are valuable in traditional medicine due to their anti-inflammatory and antibacterial properties, while the extracted oil is usually the basis of ointments [36] and also used for cooking. Studies have demonstrated that groundnut seed is very effective in treating turbid water as well as water contaminated with heavy metals [37, 38]. The heavy metals studied include Chromium, Lead, Zinc, Cadmium and Copper, but there are no studies regarding their effectiveness in treating arsenic contaminated water. Therefore, this study aimed at using coagulation method to evaluate the efficiency of groundnut seed for the treatment of arsenic contaminated groundwater. Groundnut seeds are readily available, cheap, safe, eco-friendly and non-toxic.

## 2.0 MATERIALS AND METHODS

The groundnut seeds used for this study were purchased from a local market (New Benin) in Benin City, Edo State, Nigeria. They were identified and authenticated by a Botanist in the Department of Plant Biology and Biotechnology, University of Benin, Benin City, Nigeria. The raw water was obtained from a borehole at Ugbowo, Benin City, Edo State, Nigeria. Experiments were carried out at Quality Analytical Laboratory, Evbumore Quarters, Benin City and Civil Engineering Laboratory, University of Benin, Benin City, both in Edo State, Nigeria.

### 2.1 Preparation of the Seed Extract

Fresh skin groundnut seed (see Plate 1) were oven dried at 150 °C for about 10 minutes. They were removed from the oven and allowed to cool, then they were sorted to remove bad ones. The groundnut seed were skinned manually by hand peeling to obtain the seed kernels. About 30 g of the seed kernels were pulverized mechanically to fine powder using a stainless grain laboratory pulveriser and sieved to make it of appropriate size of about 425 µm.



Plate 1: Skin Groundnut Seed

The groundnut seed powder was defatted using ethanol (96 %). About 20 g of the seed powder was packed in a thimble (made of cheese cloth) and placed inside the

Soxhlet extractor which is connected to a flask and a condenser (setting up the Soxhlet extraction apparatus). About 300 to 350 ml of ethanol (96 %) was used to extract oil from the seed powder in the column; the apparatus was left running for about 7 to 8 hours and switched off when the extraction was completed. The cake was placed in the oven at 105°C for about 8 to 10 hours to evaporate the ethanol. The defatted seed powder was then extracted with 1 M sodium chloride solution: about 1 g of the defatted seed powder was added to 1 M sodium chloride solution. The suspension was mixed using a magnetic stirrer for 10 minutes, it was then filtered using Whatman Number 1 equivalent filter paper. The filtrate (resulting to 1 % w/v solution) was used as the seed extract. Fresh solution of the seed extract was prepared at every day of use. The extract is a by-product of oil production. It is noteworthy that oil can also be removed by other means such as pressing.

### 2.2 Preparation of Aluminium Sulphate (Alum) Solution

Alum solution was prepared to give 1% w/v solution, 1 g of alum powder was weighed and added to 100 ml of distilled water, then the solution was mixed using a magnetic stirrer for 10 minutes in order for the alum powder to be completely soluble in the water.

### 2.3 Preparation of Standard 1000ppb Arsenic (Arsenite) Solution

Arsenic trioxide ( $As_2O_3$ ) was pre-dried in an oven at about 105 °C. About 0.132g of the pre-dried Arsenic trioxide was dissolved in 10 ml of 10% sodium hydroxide solution. It was neutralized with 1N sulphuric acid solution and 20 ml was added in excess, then it was diluted to 1L volume with distilled water and mixed properly. About 10 ml of this solution was then pipetted in a 1L volumetric flask and about 20 ml of 1N sulphuric acid was added, then it was diluted to 1L volume distilled water and mixed.

### 2.4 Preparation of the Synthetic Water Sample

About 10 mg/l of arsenite was added to 5 L of borehole water. The water sample was mixed with a magnetic stirrer for about 20 minutes to achieve uniform and homogenous sample. The recommended pH range for efficient coagulation is usually in the range of 6.0 to 8.5 [39, 40]. Hence, the pH of the water sample was kept constant at 7.30 using 0.1N hydrochloric acid (HCl). The characteristics of the raw borehole water and groundnut seed powder used for the study are presented in Table 1 and 2 respectively.

**Table 1:** Characteristics of the Borehole Water used for the Study

Parameter	Value
pH	5.0
Turbidity (NTU)	ND
Total Dissolved Solids, TDS (mg/l)	33
Temperature (°C)	29.2
Electrical Conductivity, EC (µs/cm)	62
Arsenic (mg/l)	ND

ND: Not Detected

**Table 2:** Proximate Analysis of the Groundnut Seed Powder used for the Study

Parameter	Groundnut Seed (Dry Basis)
Moisture (%)	1.74
Crude Protein (%)	39.33
Crude Fat (%)	48.25
Crude Fibre (%)	4.02
Ash Content (%)	3.95
Carbohydrate (%)	5.45

### 2.5 Coagulation/Flocculation Experiment (Jar Test Operations)

Standard jar test apparatus (PB- 700) with six stirrers, consisting of rotation regulator of mixing rods was used to carry out the coagulation test on the water sample using various coagulants (Groundnut seed extract only, alum only and combination of Groundnut seed extract and alum in different proportions). Six 1 litre beakers were used to analyse the effect of coagulants (Groundnut seed extract only and alum only) dosage on coagulation of the water sample. The water sample (400 ml) was filled into six beakers (1000 ml) out of which five were dosed with different volumes of the coagulant ranging from 5 to 25 ml, while one was left without coagulant to serve as control. The apparatus was coupled properly by putting the stirrers in the jar test kits and lowering them into each beaker, thus ensuring that the beakers were well centred, before the apparatus was turned on. The experimental conditions for coagulation/flocculation with the coagulants were: 2 minutes of rapid mixing (150 rpm) followed by 25 minutes of slow mixing (50 rpm). Then, the treated water was allowed to settle for 90 minutes and the supernatant samples were withdrawn using ex-20 °C Citoglas pipette (10 ml) from 2 cm below the liquid level for analyses. The samples were analyzed for Arsenite and pH. Then, the optimum dosage obtained for the seed extract was combined with that of alum in different proportions (with alum added first before the seed extract) and used separately to treat the water sample, maintaining the same

mentioned experimental conditions. The arsenite reduction efficiency was calculated using the following equation [41, 42];

$$\text{Arsenite Reduction Efficiency}(\%) = \frac{As_0 - As_1}{As_0} \times 100 \quad (1)$$

Where;  $As_0$  = Initial Arsenite and  $As_1$  = Final Arsenite

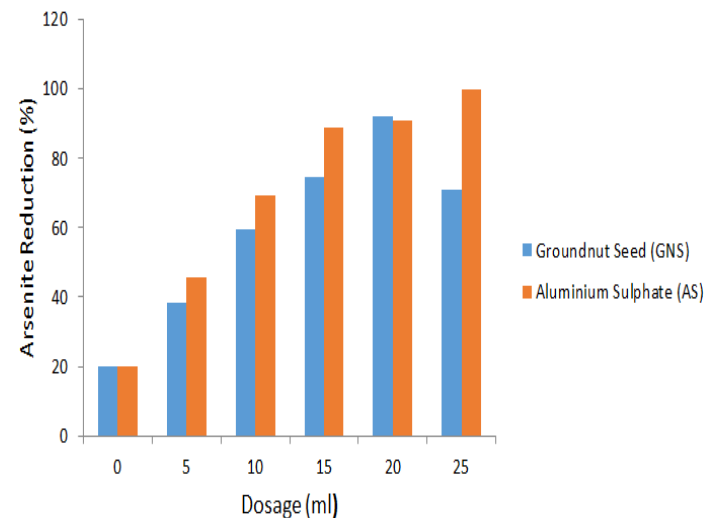
### 2.6 Analytical Methods

All analytical methods including those used for characterizing the raw water and the groundnut seed powder correspond to standard methods [43-46]. Arsenite was measured using a Spectrophotometer (A 500) and the pH was measured using Hanna pH meter.

### 3.0 RESULTS AND DISCUSSION

Figure 1 shows arsenite reduction efficiency of coagulants (groundnut seed extracts, alum) while Table 3 shows the effect of combination of groundnut seed extract and alum on arsenite reduction.

Results from Figure 1 indicated that the seed extract demonstrated appreciable efficiency in improving the quality of water in terms of arsenite reduction compared to alum. Optimal arsenite reduction efficiencies with initial arsenite values of 10 mg/l are: for Groundnut Seed (GNS) extract: 91.90% and for Alum (AS): 99.88%. These correspond to dosages of 20 ml for Groundnut Seed (GNS) extract and 25 ml for Alum. Previous studies have shown that extraction of soluble proteins from plant seeds using sodium chloride solution can greatly improve

**Figure 1:** Arsenite Reduction Efficiency of Coagulants (Groundnut Seed Extract and Alum)

**Table 3:** Effect of Combination of Groundnut Seed Extract and Alum on Arsenic Reduction

Coagulant	Proportion (%) Alum-Seed Extract	Dosage (ml) Alum- Seed Extract	Arsenite Reduction (mg/l)
Alum and Groundnut Seed (GNS)	00:100	0, 20	0.81
	20: 80	5, 16	0.56
	40: 60	10, 12	0.12
	<b>60: 40</b>	<b>15, 8</b>	<b>0.003</b>
	80: 20	20, 4	0.47
	100: 0	25,0	0.34

coagulation process due to the intense force exerted by the salt in breaking the plant cells or tissues [42, 47]. This may be explained by the salting-in effect of proteins at higher ionic strength [47-50]. The disparity between the coagulating activities of the seed extract and alum (in terms of arsenite reduction) may be attributed to the mixture of organic compounds present in the seed extract compared to the pure compound contained in alum (standard coagulant). It was observe that for the seed extract, beyond the optimum dosage, there was slight decrease in arsenite reduction efficiency. This phenomenon is due to over dosing of coagulants (seed extract) which saturates the surface of colloids and leads to insufficient sites for the formation of polymer bridge [51]. This phenomenon is suggesting that the groundnut seed extract may contain cationic polymers. Since, arsenite [As(III)] is a negatively-charged anion [52, 53], it may have sorbs to these cationic polymers or flocs which settle easily and thus facilitate their removal. Studies have noted the removal of heavy metals (Cu, Pb, Cd, Cr and Zn) from groundwater (boreholes and wells) using *Moringa* seed and peanut [37, 38]. Results also revealed that pH of the water remain largely unaffected after treatment with the seed extract as values were more or less constant between 7.15 and 7.28. This may be due to the buffering capacity of the seed extract [42].

Although optimum arsenite reduction value (using seed extract) was above the World Health Organization (WHO) acceptable limit of 0.01mg/l [54], however the value obtained is still of significant quantum and previous study has established that once arsenic reduction efficiency above 90 % is achieved (such as those obtained in this study) with coagulation, then sedimentation and filtration processes can be used to remove arsenic particulate [33], but filtration was not a part of this study. Hence the seed extract can help rural households of poor developing countries to produce drinking water, although seed extract dosage need to be carefully controlled so as to reach optimum destabilization. Groundnut is widely grown, thus the seeds will be readily available and will help to improve the life of rural people.

Results from Table 3 indicated that the combination of groundnut seed extract and alum resulted in significant arsenite reduction of treated water much lower than when alum was used alone. Optimum dose obtained was as follows: alum-15 ml and Groundnut seed extract-8 ml and the corresponding arsenite reduction was 0.003mg/l. This value is well below the WHO acceptable limit of 0.01mg/l [54], hence groundnut seed extract work better as coagulant aid (using alum as primary coagulant) to remove arsenite from water. Studies have noted the use of *Moringa oleifera* seed as a coagulant aid with alum as primary coagulant for the removal of heavy metal from water [55]. Results further suggested that the combination use of alum and groundnut seed extracts (60 and 40 %) can result to 40 % savings in alum which in turn minimizes its toxicological effect and the cost of water treatment.

#### 4.0 CONCLUSION AND RECOMMENDATION

In this study, it was revealed that *Arachis hypogaea* (groundnut seed) extract has demonstrated considerable capability in reducing arsenite in groundwater (borehole water) either as primary coagulant or coagulant aid (using alum as primary coagulant). Groundnut seed extract may have potentials for use as coagulant aid with alum as primary coagulant for treatment of arsenic contaminated groundwater. This study has revealed that Groundnut seed is suitable for the treatment of arsenic contaminated groundwater. The seeds are readily available, cheap and eco-friendly because of their biodegradable nature. Although, as primary coagulant, groundnut seed extract was not able to reduce arsenite below the WHO acceptable limit of 0.01mg/l, but above 90% arsenite reduction was achieved. Hence, it is recommended that coagulant proteins should be purified from groundnut seed extract using a simple method in order to obtain a pure compound which can be used to achieve arsenite reduction below the WHO acceptable limit of 0.01mg/l.

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