



Removal of Arsenic Contamination from Gomti River Water by using Activated Charcoal Absorbent Integrated with Solar Distillation Unit

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Arsenic is a ubiquitous and short-term poisoning element, which affects living things. In this study, the removal of arsenic from Gomti river water, Lucknow, Uttar Pradesh integrated modified double slope solar still with activated adsorption bed has been applied. Arsenic contamination in Gomti river at five places were found in the river water in the range of 55.70 ppb to 681.60 ppb, which exceed the maximum permissible limit of 10 ppb as recommended by WHO. Maximum concentration of arsenic in water was found in Mohan Maekins (681.60 ppb). However, mean arsenic concentration in water followed the order: Gaughat (115.37 ppb) < Hanuman setu (297.80 ppb) < Barrage (302.62 ppb) < Kukrail (336.27 ppb) < Mohan Maekins (481.89 ppb). The amount of adsorbed arsenic was increased with increasing initial concentration of arsenic. The aim of this study is to develop a low-cost process for the removal of arsenic effectively including the production of potable water. After results, data were analyzed with ICP-MS method and also verified with Langmuir and Freundlich adsorption isotherm models.

Keywords: Arsenic, Gomti river, Solar distillation, Activated charcoal, Adsorption, Isotherms.

INTRODUCTION

In water, arsenic has been recongnized to have adverse human health effects and also known to cause cancer in humans with high risk. Both inorganic and organic species are present in the river water in which inorganic species are predominant [1]. The major soures of arsenic in river water are from widespread use in chemical & manufacturing industries (especially glass and electronic). The valence and species of inorganic arsenic are dependent on the oxidation-reduction condition, temperature and pH of the water. Arsenite, a reduced trivalent form, exists in four forms in aqueous solution, depending on pH value *i.e.* H_3AsO_3 , H_2AsO_3^- , HAsO_3^{2-} and AsO_3^{3-} . Arsenate, an oxidized pentavalent form, exists in also four forms in aqueous solution, depending on pH value, *i.e.* H_3AsO_4 , H_2AsO_4^- , HASO_4^{2-} and AsO_4^{3-} [2-4]. Most commonly, As^{3+} is found in river water and groundwaters (anaerobic conditions) while As^{5+} is found in surface water (aerobic conditions). However, this condition is not applicable for groundwater. Some groundwater contains As^{3+} , As^{5+} and the combination of both As^{3+} and As^{5+} .

All arsenic contaminations dissolved in water are toxic because the arsenic toxicity depends on its chemical changes. Inorganic arsenic contamination is commonly present in river water in more toxic form. Among different oxidation conditions due to ability to complex formation with certain coenzymes, inorganic As^{3+} is considered more toxic as compared to As^{5+} . In case of river water and seawater foods like fish & shellfish, often contains arsenic in significant amount but it is in mainly in organic forms which are approximately 1000 times less toxic compared to inorganic form [5-8]. But in case of animals and plants As^{3+} is more toxic than As^{5+} (~ 10 times) and methylated species (~ 70 times) [9].

Arsenic contaminations are present in river/drinking water in As^{3+} and As^{5+} oxidation states and order of toxicity varies as $\text{As}^{3+} > \text{As}^{5+} > \text{organic-As}$. Since As^{5+} is more efficiently removed as compared to As^{3+} , so pre-oxidation of As^{3+} to As^{5+} is mandatory for high arsenic contamination removal from river water as well drinking water supplies containing high concentration of As^{3+} [10,11]. In adsorption bed, activated charcoal has been recognized as a strong and most effective adsorbent for removal

of heavy metal like arsenic, impurities or hazardous materials such as aromatic compounds, *etc.* from water samples. Adsorption process depends largely upon surface area and pore volume. Activated charcoal is a highly porous material having surface area 300-2500 m²/g. It has large number of very fine pores (micropores) which gives large inner surface on the basis of its adsorption properties [12]. It's main application is removal of organic contaminations with relatively high molecular weight due to its more hydrophobic nature. Activated charcoal is less effectively applied for the removal of inorganic metallic ions (small size anions). However, this study is used in removal of inorganic arsenic from water, which exists more usually in anionic form [13].

Adsorption process involves two phases, first one is solid phase (adsorbent) and another is liquid phase containing various species to be adsorbed. The adsorption mechanism are affected with various parameters such as rate of adsorption, maximum adsorption capacity and suitable pH range, contact time to reach equilibrium, initial adsorbate concentration and dosage of adsorbent. Also the study of the kinetics and equilibrium isotherms, batch operation has been undertaken to investigate various related parameters fitting into appropriate models. Finally, these parameters were correlated to understand the adsorption mechanism for toxic form of arsenic, from the water on the surface of activated charcoal [14,15].

Arsenic contamination can be recovered from process streams as a commercial commodity, but the demand for arsenic, primarily for the production of pesticides and wood preservatives is limited. Various methods are available to treat arsenic contaminated river/waste water such as ion-exchange, adsorption onto activated charcoal, activated alumina, activated bauxite [16]. Other methods reported including biological processes (by algae aquatic plants), reverse osmosis, solvent extraction, precipitation and adsorption by metal hydroxide, precipitation as lead choloarsenate and solar distillation [17,18]. Among them, adsorption process (activated charcoal) with integrated modified double slope solar still (MDSSS) is considered to be a relatively simple, efficient and low-cost technology for removal of arsenic contamination and also the production of potable water. This technology is based on the principle of evaporation and the condensation of water vapour. This technology is used especially in rural areas of remote region because of low construction and operating cost with more durability [19-22].

In water purification, adsorption process is an efficient removal technique for the multiplicity of solute. In this case, molecules or ions are removed from the aqueous solution by adsorption onto solid surfaces which are characterized by active, energy-rich sites that are able to interact with solute in the adjacent aqueous phase due to their specific electronic and spatial properties. Typically, the surface is energetically heterogeneous [23]. Activated charcoal is highly porous, crude from of graphite having structure with broad range of pore sizes. It is produced by carbonization below 600 °C. They are activated by the treatment with oxidizing agents like steam, carbon dioxide or oxygenate elevated temperature or with chemical agents like ZnCl₂, H₃PO₄, H₂SO₄, K₂S, *etc.* Activated carbon have been extensively used for As³⁺ and As⁵⁺ adsorption from water [24-26].

Solar distillation can be an effective method of treating contaminated water in river water and specially crisis areas. The average yield of this conventional solar desalination technology was about 3.2 L/m²/day [27]. The potable water produced by this method is free from all chemicals including arsenic. This technology cannot produce enough drinking water at reasonable cost. So, this technology requires further development for cost effective (both quality and quantity) use in water supply in specially rural areas. In this study, we have tried to develop integrated model combined with adsorption-solar distillation system.

River Gomti is a tributary of river Ganga and a life-line of Lucknow city of India. Gomti river originates from natural impounding reservoir near madho taal (Phoolar Jheel), Pilibhit, India. It traverses a distance 900 kms (560 miles) through Uttar Pradesh province, before it joins Ganga river near Adiyar (Saidpur Kaithi) in the district of Ghazipur city. It is facing problems of pollution caused by heavy discharge of sewage, use of fertilizers in agriculture and industrial effluents. Before reaching to Lucknow city, it receives waste from various sugar, pulp & paper, textiles and distillery industries. After travelling about 240 kms, the river Gomti enters Lucknow, through which it meanders about 16 km. At the entrance point in Lucknow city *i.e.* at Gaughat 300 MLD of water is lifted from river for the city's water supply. At the downstream (near Gomti nagar), Gomti barrage impounds the river converting it into a lake.

Therefore, the object of this study is to investigate integrated modified double slope solar still with and without activated charcoal ability in arsenic removal from river water and also assess the effects of some parameters including effective temperature, contact time, initial concentration, pH, adsorption isotherms, reaction kinetics and thermodynamic studies on the absorption process.

EXPERIMENTAL

Sampling locations: For the analysis, the sample were collected in polypropylene container (10 L) from hand pumps, at five different locations (Gaughat, Mohan Meakins, Hanuman Setu, Kukrail and Barrage) of Lucknow city of India (26.8467° N, 80.9462° E) during period of April, 2019 to August, 2019. The arsenic contamination in water samples taken from Gomti river, were performed as per reported method [28]. The results are shown in Table-1.

Fabrication of solar distillation unit: This solar still (distillation unit) has been assembled with material fiber reinforced plastic (FRP). The total inner surface area of the still is 200 cm × 100 cm × 10 cm. For good absorptivity, black dye with resin was painted over the bottom surface [29]. For good performance of this still, it has been installed in the direction of east-west to collect more sun energy for the maximum absorption and gradually increasing the temperature of solar still. The depth of the solar still wall is 12 cm at east-west side ends location and 48 cm at the centre. The base surface and north surface wall are madeup of FRP material with 5 mm thickness in place of acrylic sheet. The top surface of still is covered with two transparent glasses with thickness 4 mm and for better absorptivity, two simple window glasses are used with dimen-

TABLE-1
ARSENIC CONCENTRATION IN WATER SAMPLES OBTAINED FROM FIVE DIFFERENT LOCATIONS OF LUCKNOW CITY, INDIA

Name of place	Sample number	Arsenic concentration (ppb)			Average conc. ⁿ (ppb)	Mean arsenic conc.* (ppb)
Gaughat	W1 (April)	125.0	95	105.2	108.40	115.37
	W2 (May)	142.10	130.28	108.07	155.86	
	W3 (June)	60.80	71.52	80.70	71.50	
	W4 (July)	177.90	189.10	190.10	185.70	
	W5 (August)	51.60	60.23	54.37	55.40	
Mohan Meakins	W6 (April)	560.50	577.23	580.1	572.61	481.89
	W7 (May)	680.45	657.90	706.75	681.70	
	W8 (June)	337.67	345.70	347.88	343.75	
	W9 (July)	480.13	487.50	489.16	485.72	
	W10 (August)	318.90	324.62	333.58	325.70	
Hanuman Setu	W11 (April)	340.51	351.47	332.82	341.60	297.80
	W12 (May)	180.41	178.60	204.24	187.75	
	W13 (June)	357.21	363.05	360.34	360.20	
	W14 (July)	197.40	203.70	204.00	201.70	
	W15 (August)	391.80	398.53	402.92	397.75	
Kukrail	W16 (April)	400.31	398.20	423.87	407.46	336.27
	W17 (May)	357.12	352.51	345.47	351.70	
	W18 (June)	250.40	249.58	264.25	254.75	
	W19 (July)	277.30	285.75	304.05	289.70	
	W20 (August)	378.00	380.25	375	377.75	
Barrage	W21 (April)	457.32	460.01	449.38	455.60	302.62
	W22 (May)	340.52	339.78	344.80	341.70	
	W23 (June)	260.00	257.89	267.42	261.77	
	W24 (July)	246.75	257.21	259.54	254.50	
	W25 (August)	201.70	197.50	201.39	199.53	

sions of 1.03 m × 1.06 m × 0.004 m designed over the walls surface of still with inclined angle at 15° on both sides using FRP frame. The adsorption bed (activated charcoal) output (filtrate) connected with one solar still input by using plastic pipe and the distilled water accumulated on both side walls of the still basin is continuously coming out through another plastic pipe and collected in a measuring jar.

The saline water as feed (inter) continuously to still for maintaining the water level from storage tank through a hole from the north wall of basin. The product water is received through a V-shaped drainage zone, which provided beneath the transparent glasses lower edge on both walls of still basin. The thermocouple device is used for measuring the temperature of the basin water surface, still water surface and condensate water temperature which are inserting in the side wall of still through a hole. To avoid the heat loss, we used an insulating material around the hole. A thermocouple is also used for measuring the atmospheric temperature. The flow rate and mass of water are maintained always as constant in still basin [30,31].

Methods for removal of arsenic: For analytical analysis, stock solution of As⁵⁺ (100 mg/L) was prepared by dissolving 410.62 mg sodium arsenate reagent in 1 L of distilled water and preserved with 0.5 % HNO₃. Similarly As³⁺ stock solution (3750 mg/L) was prepared by dissolving sodium arsenite (0.05 mol) in 1 L distilled water, which was preserved with 0.5 % HNO₃. Arsenic was analyzed with ICP-MS using an agilent 7500 ce spectrometer equipped with octopole system (ORS). Calibration was done using with external standards (2, 4, 20, 40, 80 and 100 µg/L), which were prepared with 1000 µg/L stock standard solution. Working with standard, blank solution were prepared with high purity nitric acid. The concentrations of sample were adjusted in the range of 5-100 µg/L. The experi-

mental data measurements were accepted as reasonable data in cases of less than 5% relative standard deviation (RSD). In order to amplify the consistency of results, the experiments were conducted in triplicate and also the mean value considered.

After analytical results, the average of at least triplicate measurements and the maximum expected error was ± 5 %. So, percentage (%) of sorption was calculated by using the following relationship:

$$\text{Sorption (\%)} = \frac{A_i - A_f}{A_f} \times 100 \quad (1)$$

where A_i and A_f are the initial concentration and final concentration of arsenic contamination. The amount of sorption per unit mass of sorbent (X/m) was calculated as follows:

$$\frac{X}{m} = \frac{A_i - A_f}{A_f} \times \frac{V}{m} \quad (2)$$

where, V is the volume of adsorbate in mL and m is the amount of adsorbent in g.

Error analysis: For experimental work, various instruments are used for measuring solar intensity, distillate collection and temperature, respectively. The instrument uncertainties are considered as they affect the desired accuracy. Uncertainties for the experimental instruments are given in Table-2. These instruments are used for measuring solar intensity, distillate collection and temperature. The minimum error in any instrument is equal to the ratio between its least count and minimum value of output measured.

RESULTS AND DISCUSSION

The performance of this integrated technology used to enhance the productivities of adsorption bed with integrated modifies double slope solar still. Performance of all the three techniques

TABLE-2
STANDARD UNCERTAINTY, ERROR AND MEASURING RANGE OF INSTRUMENTS

Instruments	Accuracy	Range	Errors (%)	Standard uncertainty
Pyranometer	± 1 W/m ²	0–5000 W/m ²	2.50	± 0.59 W/m ²
Calibrated flask	± 5 mL	0–1000 mL	5.00	± 5.79 mL
Thermocouple	± 1°C	0–100	0.25	± 0.59°C

were compared and found that the integrated system has given better performance for the removal of arsenic contamination as well as production of potable water.

Effect of pH: The adsorption of arsenic from river water and water purification using activated carbon as adsorbent in this study was found to be a highly pH dependent process. The pH of given solution, in fact determines the speciation of arsenic ions and also affects the surface charge of adsorbent. As per experimental results, adsorption behaviour of As³⁺ and As⁵⁺ ions were analyzed at different pH values, using 1.0 g of activated carbon and fixed amount of arsenic. The results obtained are shown in Table-3, which showed that adsorption or removal of As³⁺ ions increases with increasing the pH. Similarly, experiments were performed for As⁵⁺ and the results obtained are also shown in Table-3.

TABLE-3
INFLUENCE OF pH ON THE ADSORPTION OF As³⁺ AND As⁵⁺ ONTO ACTIVATED CHARCOAL BED

As ³⁺		As ⁵⁺	
pH	Sorption (%) (PAC)	pH	Sorption (%) (PAC)
3	12	4	77
8	57	7.5	72
11	79	11	64
12	93	–	–

Adsorbent = 1.0 g of powdered activated carbon (PAC)
Adsorbate = 10 mL of 25 ppm

It is observed that adsorption process of arsenic contamination by activated charcoal is dependent on the oxidation state and results were found within acceptable limit. But this integrated system is cost effective and provides better performance as compared to only adsorption process. As³⁺ and As⁵⁺ are more efficiently adsorbed by activated charcoal at basic and acidic conditions, respectively. In samples, As⁵⁺ exhibits anionic behaviour at a pH range 3-12, it is replaced by H₂AsO₄⁻, HAsO₄²⁻ and AsO₄³⁻. The removal of As⁵⁺ by activated charcoal is carried out *via* anion exchange mechanism as well as physio-chemical adsorption due to highly porous structure of activated charcoal.

Effect of concentration of adsorbate: The concentrations of As³⁺ and As⁵⁺ were varied from 10-150 ppm. A 10 mL of each concentration of arsenic was treated with the adsorbent at pH value of 12. The results are depicted in Table-4, which

showed that activated carbon is more efficient adsorbent at low concentration of arsenic. Decreasing in sorption percentage at higher concentrations might be due to the relatively smaller numbers of active sites available at higher arsenic concentration and with increasing of the initial concentration of arsenic total amount of arsenic removal (X/m) is increased.

TABLE-4
EFFECT OF INITIAL As(III) AND As(V) ION CONCENTRATION ON SORPTION USING ACTIVATED CARBON

Concentration (ppm)	10	20	50	100	150
Sorption (%), As ³⁺ (pH = 12)	95	92	85	75	66
Sorption (%), As ⁵⁺ (pH = 3)	84	75	60	45	30

The adsorption data for arsenic removal were also further analyzed by means of the Langmuir and Freundlich isotherm models (Table-5). The Langmuir and Freundlich equations can be represented as follows [32]:

$$\frac{1}{X} = \frac{1}{X_m} + \frac{1}{bC_e} = KC_e^{1/n} \tag{3}$$

or

$$\log \frac{X}{m} = \log K + \frac{1}{n} \log C_e \tag{4}$$

where, C_e is the equilibrium concentration of arsenic As⁵⁺ solution (mg/L), X is the amount sorbed by activated carbon (mg/g), X_m is the maximum amount sorbed, b & a are the Langmuir’s constants signifying energy of sorption; K and n are Freundlich’s constants indicating sorption capacity and intensity, respectively.

The analysis of experimental data obtained for As³⁺ sorption showed that it fits better with the linearized form of Langmuir equation (Fig. 1). In case of As⁵⁺ sorption, the data obtained showed that they fit better with the Freundlich adsorption isotherm equation (linearized form).

Conclusion

The removal of arsenic contamination from river water and the production of potable water with integrated adsorption-solar distillation revealed that the quality of water in Gomti river is deteriorating while entering into Lucknow city due to the discharge of huge quantity of sewage through different drains directly into Gomti river. Arsenic contamination of river

TABLE-5
EFFECT OF INITIAL CONCENTRATION OF As³⁺ ION ON ADSORPTION PARAMETERS USING FREUNDLICH AND LANGMUIR EQUATIONS

Conc. of As ³⁺ (ppm)	Ads (%)	X/m (mg/g)	C _e (ppm)	1/C _e	log X/m	log C _e	m/X
20	92	184.0	1/6	0.625	1.26	0.20	5.4
50	85	445.0	6	0.16	1.6	0.77	2.24
100	75	750.0	25	0.04	1.87	1.39	1.33
150	66	0.99	51	0.02	1.99	1.7	1.01

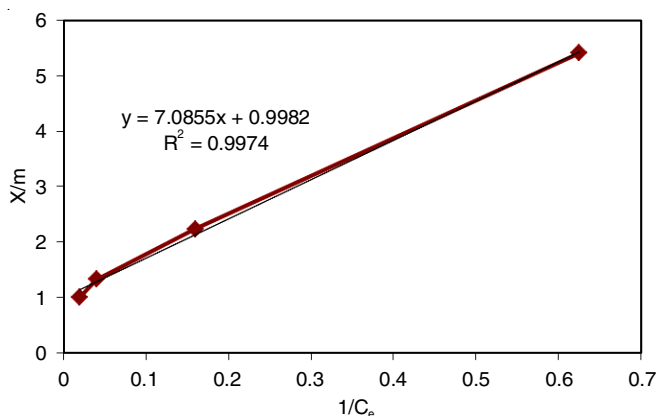


Fig. 1. Adsorption isotherm obtained from As(III) sorption by activated carbon using Langmuir equation (linearised form)

water was moderately polluted at the entrance of the city (Mohan Meakins) and pollution at this point is mainly found due to some industrial as well as domestic discharge into river. Concentration of total arsenic in examined drinking water samples was in the range of 55.70–681.60 ppb. Maximum concentration of arsenic in water was found in Mohan Meakins (681.60 ppb). However, mean arsenic concentration in water followed the order: Gaughat (115.37 ppb) < Hanuman setu (297.80 ppb) < Barrage (302.62 ppb) < Kukrail (336.27 ppb) < Mohan Meakins (481.89 ppb).

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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