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A Comparative Study of Removal of As(III) using *Cascabela thevetia (kaner)* and *Tinospora cordifolia (giloy)*.

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Abstract

Bioremediation of arsenic by medicinal plants had been studied in the present paper. *Cascabela thevetia (Kaner)* having few anticancer properties and *Tinospora cordifolia (Giloy)* may adversely affect the health if grown on arsenic contaminated soil. Different initial concentrations of arsenic i.e. 5 ppm, and 2 ppm had been taken at different pH values of 2, 7, and 9 to find the optimum condition of maximum bioremediation using *C. thevetia* and *T. cordifolia.* The peaks in HPLC at 227 nm of the plants had shown the changes in peaks before and after treatment. The FTIR peaks also showed changes before and after adsorption. When an initial concentration of 5 ppm As(III) was treated with *T. cordifolia* at pH 7, the residual concentration was 0.15 ppm. Whereas the residual concentration with 2 ppm initial concentration of As(III) was 0.10 ppm at pH 7. The Maximum percentage removal using *C. thevetia* treated with the initial concentration of 5 ppm As(III) was 97% at pH 9. No considerable effect of pH had been observed on arsenic remediation. The experimental data best fitted the pseudo-second order reaction.

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Bioremediation;
FTIR; HPLC;
Remediation.

Introduction

Arsenic, a metalloid, has adverse health effects causing hyperpigmentation and skin cancer. The Gangetic plain in general and of Bhagalpur in particular is severely affected with arsenic contamination and several causes of arsenicosis have been detected in the plain along with prostate cancer, liver cancer, and other cancers.[1] The increase in cancer cases in the Gangetic plain has been linked to various environmental factors out of which inorganic arsenicals has emerged as the major causative factor. The inorganic arsenicals such as monomethyl arsenous acid, dimethyl arsenous acid hinder the generation of adenosine-5´triphosphate

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during oxidative phosphorylation. The binding of monomethylarsenous acid and dimethylarsenous acid takes place with cysteine residues in proteins. In addition, arsenic can inactivate around 200 enzymes.[2] One of the important reasons of escalating cancer patients in the Gangetic plain may be attributed to the large chunk of the population exposure to agrochemicals and consequent entry of heavy metals, pesticides, and arsenic into food chain.[3] Pesticides' exposure to the agro-based population causes long term health hazards due to non-biodegradibility.

The present work has been designed to study the uptake of arsenic by selected medicinal plants such as *T. cordifolia* and *C. thevetia*. The remediation potential of these medicinal plants has been investigated using different parameters. *T. cordifolia* has a large number of secondary metabolites which impart medicinal values to this plant.[4] Alkaloids, steroids, diterpenoids lactones, and glycosides are few worth mentioning chemical constituents of Giloy.[5] Owing to the presence of these compounds in the Giloy, it is used as antipyretic, antioxidant, antidiabetic, and hepato-protective. *Nerium oleander* (Kaner flower) contains gitoxigen neridiginoside, and adynerigenin. The flowers of *C. thevetia* may cause acute poisoning.[6] These medicinal uses of *C. thevetia* are in the treatment of amenorrhea, jaundice, migraines, and cancer.[7]

Arsenic uptake has been studied with the biomass of leaves and stem of *C. thevetia* and *T. cordifolia* with different initial concentrations at different pH values. Optimum condition of remediation has been set during batch experiments. The experimental data have been analysed to see the best fit of adsorption isotherms. Kinetic studies have also been done to explain the results. Percentage removal and residual concentrations are calculated as per the equations given below

$$q_t = (C_i - C_t) \cdot V / m \qquad \text{...(1)}$$

where, C_i represents the initial concentration and C_t the final concentration of As(III). V represents the volume of solution in litre, and m represents the mass of adsorbent in g.

$$\text{Percentage removal} = (C_i - C_t) / C_i \times 100 \qquad \text{...(2)}$$

Pseudo – first and second order models have been studied for removal of arsenic by *T. cordifolia* and *C. thevetia*. The removal efficiency of the adsorbent depends on several factors such as pH, adsorbent doses, initial concentration and contact time.[8] Phytoremediation of arsenic by *Withania somnifera* and *Aloe-vera* has also been reported.[9] *Cymbopogon flexuosus* (Lemon grass) has also been established as a potential remover of toxic heavy metals.[10] The Ganga as well as Koshi river plains are arsenic contaminated which has been established in various studies.[11,12] Medicinal as well as aquatic plants grow in a natural way in these river plains. Bentonite minerals are also found in abundance in Rajmahal hills of Jharkhand. Several natural adsorbents such as medicinal plants, aquatic plants, and bentonites have emerged as potential detoxicants of arsenic and chromium.[13-15]

Materials and Methods

C. thevetia and *T. cordifolia* have been collected from Acharya P. C. Ray Garden of University Department of Chemistry, T.M.B.U., Bhagalpur. The powdered biomass was stored in a bottle. Batch experiments have been done with fixed biomass of *T. cordifolia* and *C. thevetia* up to 15, 30, 45 and 60 minutes at pH 2, 7, and 9 to find the optimum adsorption condition. The pH has been changed to 2 and 9 by adding N/10 HCl and N/10 NaOH, respectively, in 100 ml As(III) solution and the pH was measured by pH meter Spectron model No. St -139. Similar experiments were repeated with different initial concentations of As (III) solution such as 2ppm, and 5 ppm. 1 g powdered biomass of *T. cordifolia* was added to 100 ml of 2 ppm and 5 ppm As(III) solutions at time intervals of 15, 30, 45 and 60 minutes, respectively and shaken on a magnetic stirrer at 220 rpm. A similar experiment was repeated with *C. thevetia*.

The residual concentrations obtained after treatment of As (III) solutions were estimated by Induced Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) analyzer Perkin Elmer Avio 560 Max (Model ULTIMA-2, Horiba Jobin YVON, France). HPLC of powdered mass of *C. thevetia* and were done and HPLC of the powdered mass after treatment with As (III) solution was fixed at 227 nm, because of the fact that several constituents absorbed light at this wavelength. The FTIR of

C. thevetia and *T. cordifolia* powders have been done before and after adsorption using KBr pellet. The model No of FTIR instrument is – Perkin Elmer Version 10.4.1, US.

Results

The maximum removal of arsenic with *T. cordifolia* takes place at a pH of 7 with initial concentrations of 2 ppm and 5 ppm arsenic solution as shown in Table 1. The maximum percentage removal for 2 ppm As(III) at pH 2 is 87.5% whereas at pH 7 it is 95%. For an initial concentration of 5 ppm, the maximum percentage removal is 98% at pH 7.

FTIR spectral peaks of *T. cordifolia* before and after adsorption indicated the adsorption of arsenic (Figure 5).

Table 1: Residual arsenic concentration after treatment with 1 g *T. cordifolia.*

S.N.	Initial concentration (in ppm)	Time (in minutes)	pH	Residual concentration (in ppm)	% Removal = $C_i - C_t/C_i \times 100$
1	2	15	2	0.25	87.5
2	2	30	2	0.25	87.5
3	2	45	2	0.25	87.5
4	2	60	2	0.25	87.5
5	2	15	7	0.15	92.5
6	2	30	7	0.10	95
7	2	45	7	0.10	95
8	2	60	7	0.10	95
9	2	15	9	0.25	87.5
10	2	30	9	0.25	87.5
11	2	45	9	0.25	87.5
12	2	60	9	0.25	87.5
13	5	15	2	0.30	94
14	5	30	2	0.30	94
15	5	45	2	0.40	92
16	5	60	2	0.40	92
17	5	15	7	0.15	97
18	5	30	7	0.15	97
19	5	45	7	0.25	95
20	5	60	7	0.10	98
21	5	15	9	0.35	93
22	5	30	9	0.40	92
23	5	45	9	0.50	90
24	5	60	9	0.30	94

Table 2: Residual arsenic concentration after treatment with 1g *C. thevetia.*

S.N.	Initial concentration (in ppm)	Time (in minutes)	pH	Residual concentration (in ppm)	% Removal = $C_i - C_t/C_i \times 100$
1	2	15	2	0.20	90
2	2	30	2	0.15	92.5
3	2	45	2	0.15	92.5
4	2	60	2	0.10	95

5	2	15	7	0.10	95
6	2	30	7	0.08	96
7	2	45	7	0.08	96
8	2	60	7	0.10	95
9	2	15	9	0.10	95
10	2	30	9	0.10	95
11	2	45	9	0.10	95
12	2	60	9	0.10	95
13	5	15	2	0.50	90
14	5	30	2	0.50	90
15	5	45	2	0.50	90
16	5	60	2	0.50	90
17	5	15	7	0.40	92
18	5	30	7	0.45	91
19	5	45	7	0.50	90
20	5	60	7	0.50	90
21	5	15	9	0.50	90
22	5	30	9	0.50	90
23	5	45	9	0.15	97
24	5	60	9	0.15	97

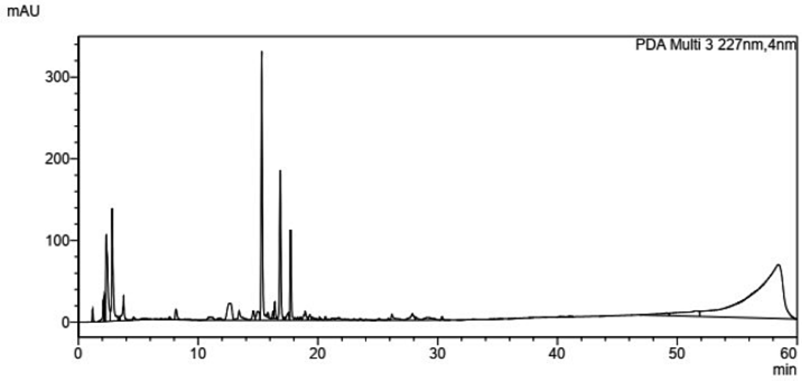

Fig. 1: HPLC results of untreated *T. cordifolia* at wavelength 227 nm.

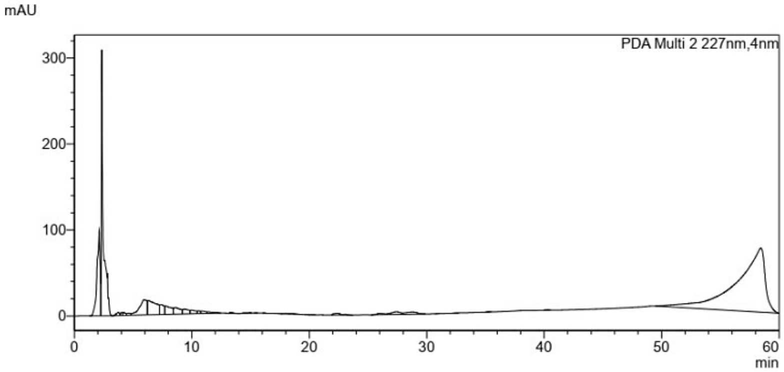

Fig. 2: HPLC results of treated *T. cordifolia* at wavelength 227 nm.

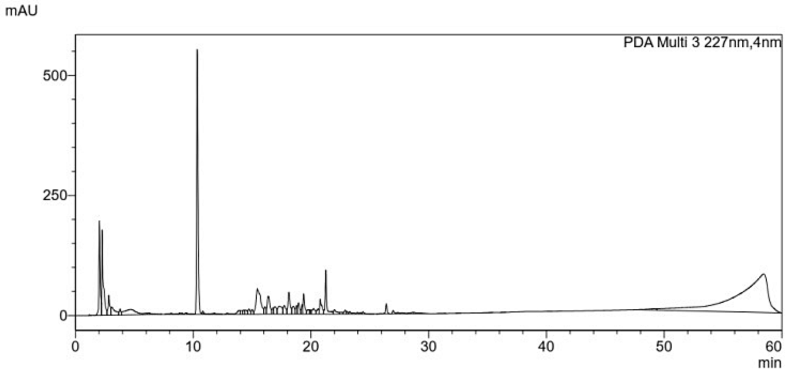

Fig. 3: HPLC results of untreated *C. thevetia* at wavelength 227 nm.

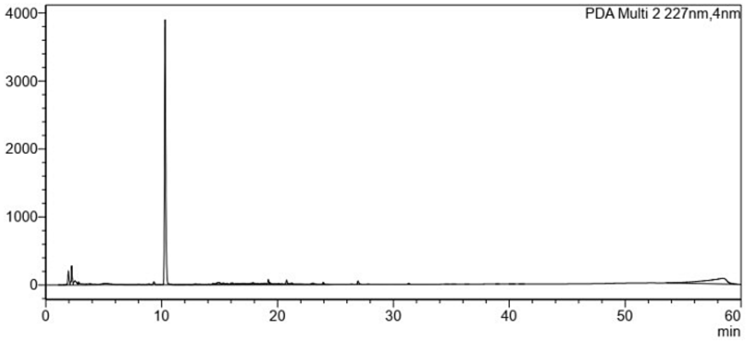

Fig. 4: HPLC results of treated *C. thevetia* at wavelength 227 nm

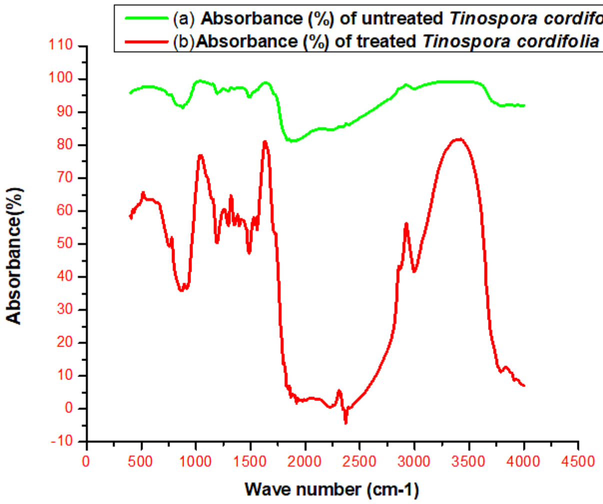

Fig. 5: FTIR spectra of *T. cordifolia* (a) untreated and (b) treated with 100 ml 2ppm As(III) solution for 30 minutes at pH 7.

Similarly, FTIR spectra of *C. thevetia* have also been done before and after adsorption of arsenic to see the change in peaks (Fig. 6).

Now the percentage removal of arsenic after treatment with 1 g powder of *T. cordifolia* is calculated by using equation (2) and Ci and Ct data taken from Table 1.

Similarly, percentage removal of arsenic by 1 g powder of *C. thevetia* was calculated and shown in Table 2.

In figure 7, the values of percentage removal of As(III) at pH 2 and 9 are the same as a result coincident lines are obtained in the graph.

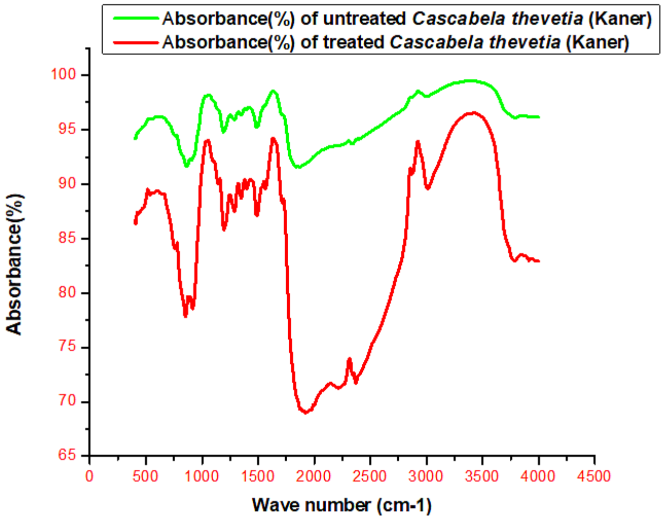

Fig. 6: FTIR spectra of *C. thevetia* (a) untreated and (b) treated with 100 ml 2ppm As(III) solution for 30 minutes at pH 7.

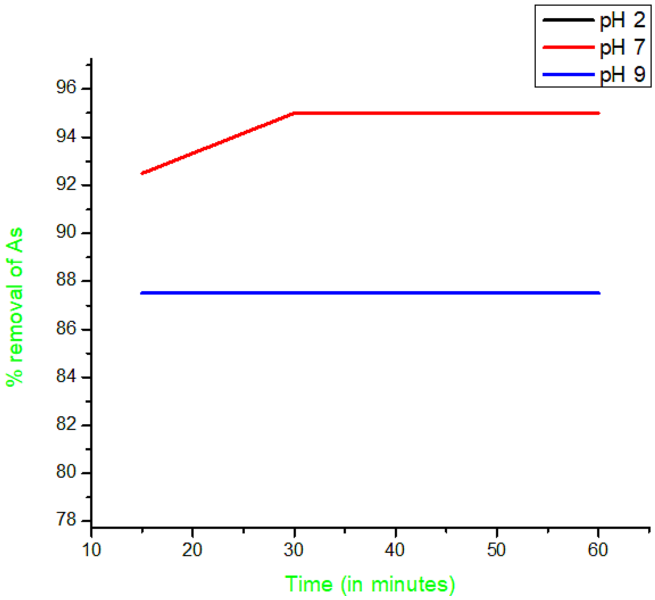

Fig. 7: Percentage removal of As by *Tinosora cordifo* (Giloy) at Ci 2 ppm and pH 2, 7 & 9

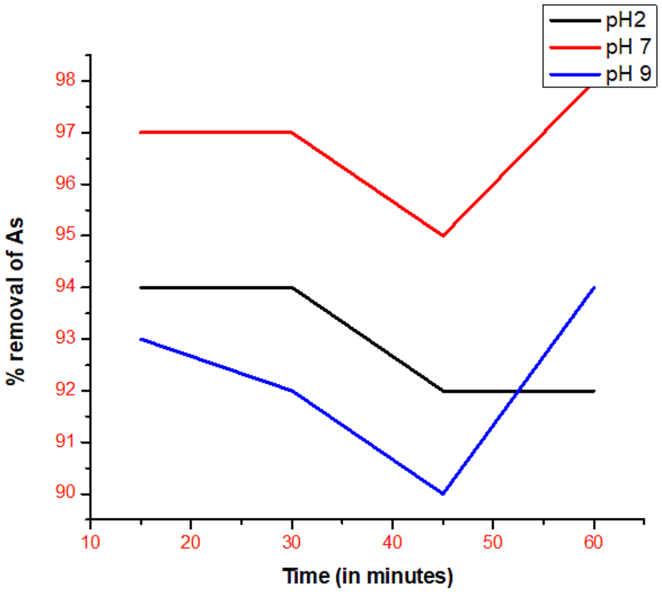

Fig. 8: Percentage removal of As by *Tinosora cordifo* (Giloy) at Ci 5 ppm and pH 2, 7 & 9

Figures 7 and 8 show percentage removal of As(III) by *T. cordifolia* for initial concentrations of 2 ppm and 5 ppm whereas figures 9 and 10 show percentage removal of As(III) by *C. thevetia* at pH 2, 7 and 9.

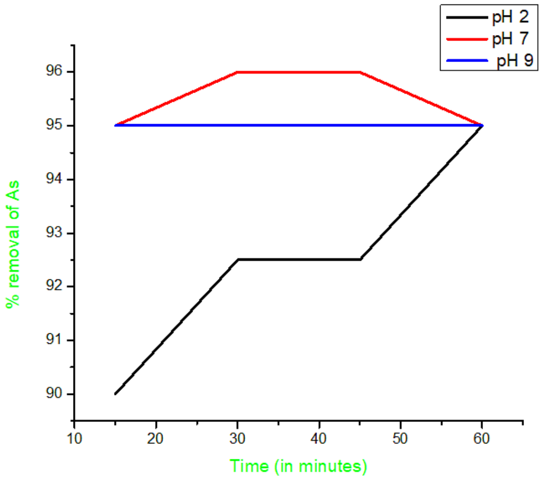

Fig. 9: Percentage removal of As by *Cascabela thevetia* (Kaner) at Ci 2 ppm and pH 2, 7 & 9

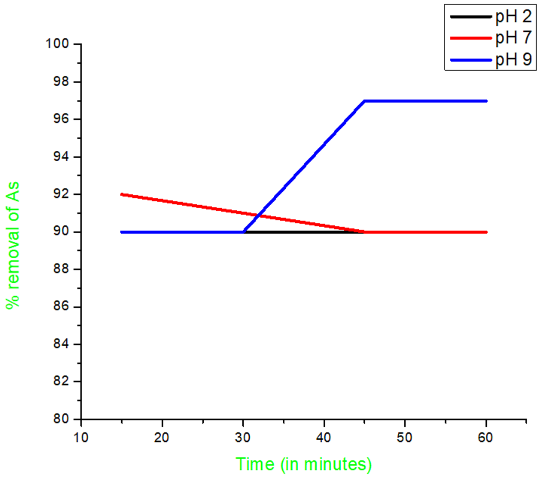

Fig. 10: Percentage removal of As by *Cascabela thevetia* (Kaner) at Ci 5 ppm and pH 2, 7 & 9

Kinetic adsorption data collected in Tables 3 and 4 for *T. cordifolia* and *C. thevetia* have been analysed for pseudo-first and second order kinetic model to see the mechanism of adsorption.

In figure 11, the values of log Ct are the same at pH 2 and 9. So, the lines coincide in the graph.

Table 3: Values of C_t, Q_t , C_t / Q_t , log Q_t and log C_t at different time intervals at pH 2, 7 and 9 for As(III) removal by 1g powder of *T. cordifolia.*

S.N.	Initial concen -tration (C_i) (in ppm)	Time (in minutes)	pH	Residual concen -tration (Ct) (in ppm)	As(III) adsor -bed (mg/L)	Q_t	C_t/Q_t	$\log C_t$	$\log Q_t$
1	2	15	2	0.25	1.75	0.175	1.429	-0.602	-0.757
2	2	30	2	0.25	1.75	0.175	1.429	-0.602	-0.757
3	2	45	2	0.25	1.75	0.175	1.429	-0.602	-0.757
4	2	60	2	0.25	1.75	0.175	1.429	-0.602	-0.757
5	2	15	7	0.15	1.85	0.185	0.811	-0.824	-0.733
6	2	30	7	0.10	1.90	0.190	0.526	-1	-0.721
7	2	45	7	0.10	1.90	0.190	0.526	-1	-0.721
8	2	60	7	0.10	1.90	0.190	0.526	-1	-0.721
9	2	15	9	0.25	1.75	0.175	1.429	-0.602	-0.757
10	2	30	9	0.25	1.75	0.175	1.429	-0.602	-0.757
11	2	45	9	0.25	1.75	0.175	1.429	-0.602	-0.757
12	2	60	9	0.25	1.75	0.175	1.429	-0.602	-0.757
13	5	15	2	0.30	4.70	0.470	0.638	-0.523	-0.328
14	5	30	2	0.30	4.70	0.470	0.638	-0.523	-0.328
15	5	45	2	0.40	4.60	0.460	0.870	-0.398	-0.337
16	5	60	2	0.40	4.60	0.460	0.870	-0.398	-0.337
17	5	15	7	0.15	4.85	0.485	0.309	- 0.824	-0.314
18	5	30	7	0.15	4.85	0.485	0.309	-0.824	-0.314
19	5	45	7	0.25	4.75	0.475	0.526	-0.602	-0.323
20	5	60	7	0.10	4.90	0.490	0.204	-1	-0.310
21	5	15	9	0.35	4.65	0.465	0.753	-0.456	-0.333
22	5	30	9	0.40	4.60	0.460	0.870	-0.398	-0.337
23	5	45	9	0.50	4.50	0.450	1.111	-0.301	-0.347
24	5	60	9	0.30	4.70	0.470	0.638	-0.523	-0.328

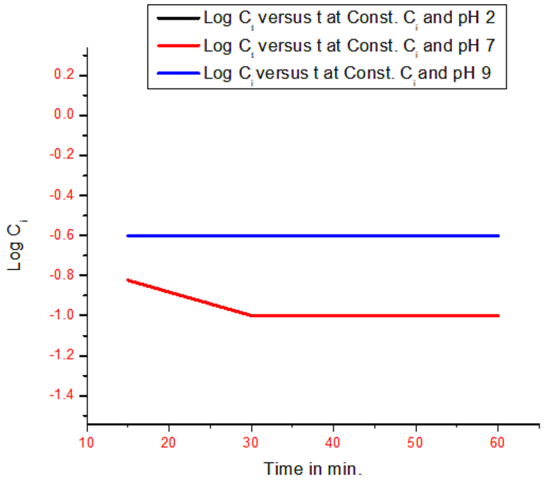

Fig. 11: Log C_i versus time(t) of *Tinosora cordifolia* at C_i=2ppm and pH 2, 7 & 9

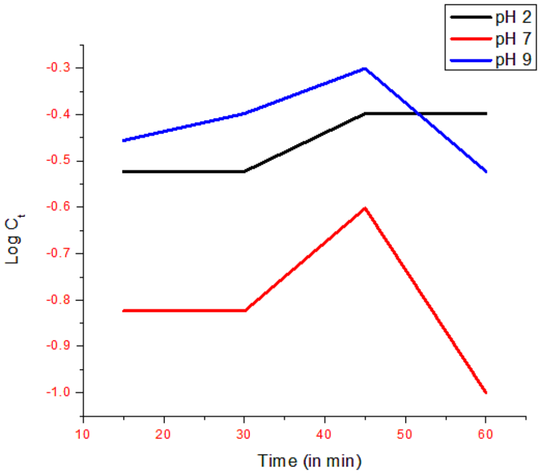

Fig. 12: Log C_i versus time(in min) at initial conc. 5 ppm and pH 2, 7 & 9

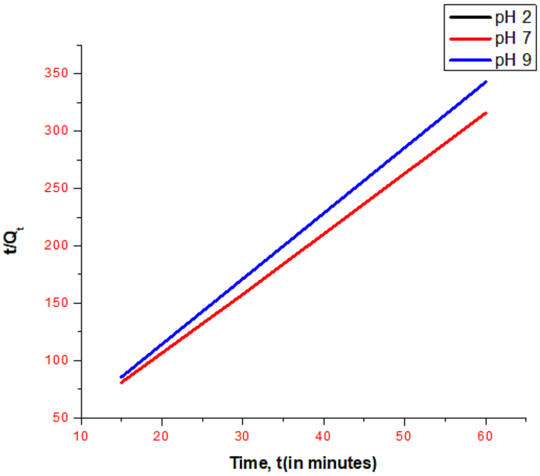

Fig. 13: t/Q$_t$ versus t of *Tinosora cordifolia* for C$_i$ 2 ppm and pH 2, 7 & 9

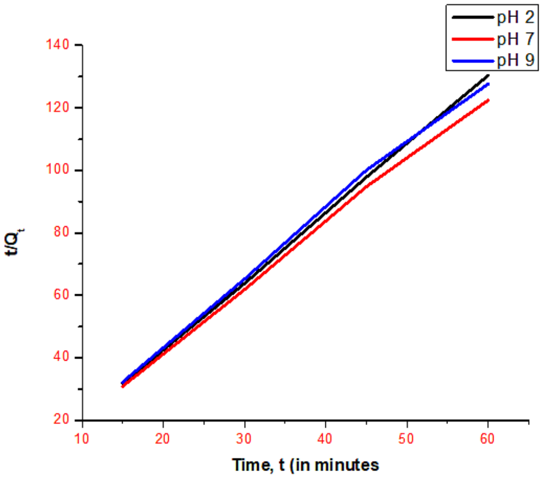

Fig. 14: t/Q$_t$ versus Time t of *Tinosora cordifolia* at C$_i$ 5 ppm and pH 2, 7 & 9

An extensive study of adsorption isotherms such as Langmuir, and Freundlich at initial concentrations of 2 and 5 ppm was done to arrive at a suitable mode of isotherm.

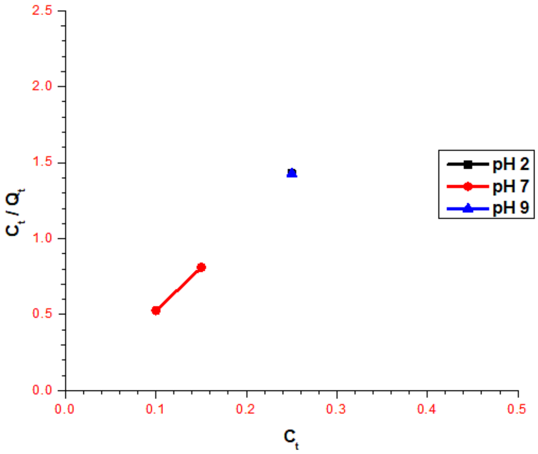

Fig. 15: Ct/Q$_t$ versus C$_t$ of *Tinosora cordifolia* for C$_i$ =2 ppm at pH 2, 7 & 9

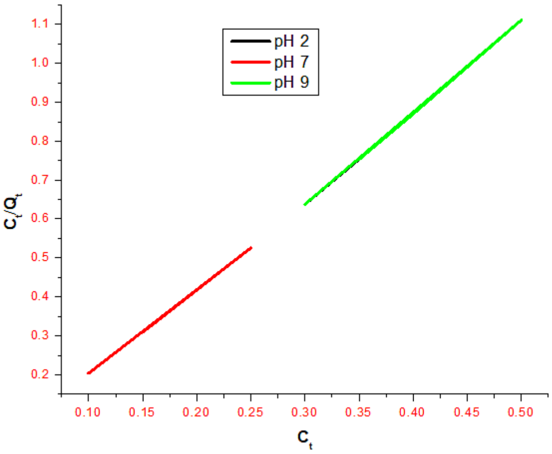

Fig. 16: C$_t$/Q$_t$ versus C$_t$ of *Tinosora cordifolia* for Ci =5 ppm As(III) at pH 2, 7 & 9

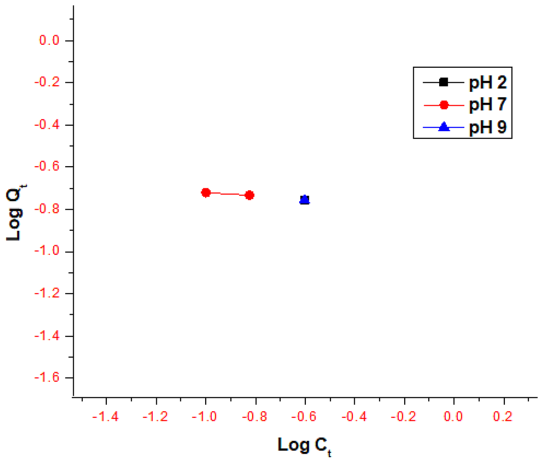

Fig. 17: Log Q_t versus C_t *Tinosora cordifolia* for Ci =2 ppm As(III) at pH 2, 7 & 9

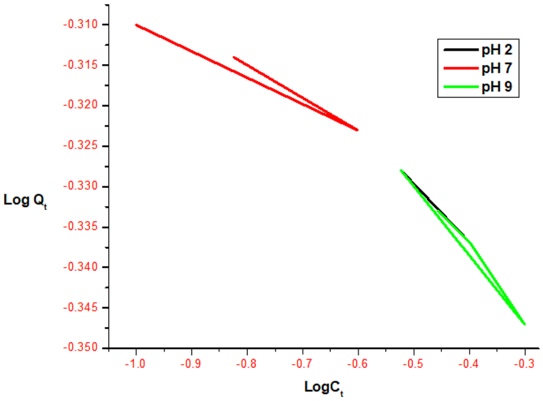

Fig. 18: Log Q_t versus C_t *Tinosora cordifolia* for Ci -5 ppm As(III) at pH 2, 7 & 9

Table 4: Values of Ct, Q t , C t / Q t , log Ct and log Q t at different time intervals at pH 2, 7 and 9 for As(III) removal by 1g powder of *C. thevetia*.

S.N.	Initial concen -tration (C_i) (in ppm)	Time (in minutes)	pH	Residual concen-tration (C_t) (in ppm)	As(III) adsor -bed (mg/L)	Q_t	C_t/Q_t	LogC_t	logQ_t
1	2	15	2	0.20	1.80	0.180	1.111	-0.699	-0.745
2	2	30	2	0.15	1.85	0.185	0.811	-0.854	-0.733
3	2	45	2	0.15	1.85	0.185	0.811	-0.852	-0.733
4	2	60	2	0.10	1.90	0.190	0.526	-1.000	-0.721
5	2	15	7	0.10	1.90	0.190	0.526	-1.000	-0.721
6	2	30	7	0.08	1.92	0.192	0.417	-1.10	-0.717
7	2	45	7	0.08	1.92	0.192	0.417	-1.10	-0.717
8	2	60	7	0.10	1.90	0.190	0.526	-1.000	-0.721
9	2	15	9	0.10	1.90	0.190	0.526	-1.000	-0.721
10	2	30	9	0.10	1.90	0.190	0.526	-1.000	-0.721
11		45	9	0.10	1.90	0.190	0.526	-1.000	-0.721
12	2	60	9	0.10	1.90	0.190	0.526	-1.000	-0.721
13	5	15	2	0.50	4.50	0.450	1.111	-0.301	-0.347
14	5	30	2	0.50	4.50	0.450	1.111	-0.301	-0.347
15	5	45	2	0.50	4.50	0.450	1.111	-0.301	-0.347
16	5	60	2	0.50	4.50	0.450	1.111	-0.301	-0.347
17	5	15	7	0.40	4.60	0.460	0.870	-0.398	-0.337
18	5	30	7	0.45	4.55	0.455	0.989	-0.347	-0.342
19	5	45	7	0.50	4.50	0.450	1.111	-0.301	-0.347
20	5	60	7	0.50	4.50	0.450	1.111	-0.301	0.347
21	5	15	9	0.50	4.50	0.450	1.111	-0.301	-0.347
22	5	30	9	0.50	4.50	0.450	1.111	-0.301	-0.347
23	5	45	9	0.15	4.85	0.485	0.309	-0.824	-0.314
24	5	60	9	0.15	4.85	0.485	0.309	-0.824	-0.314

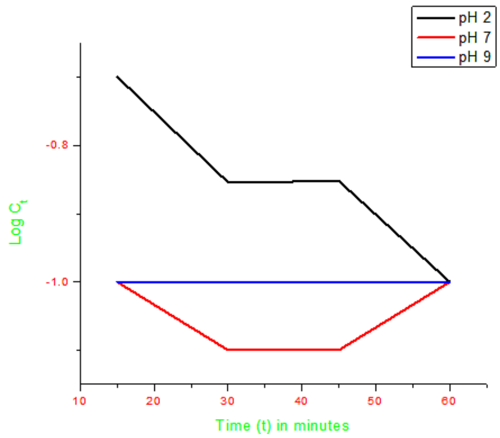

Fig. 19: Log C_i versus time(t) of *Cascabela thevetia* at initial con. 2 ppm and pH 2, 7 & 9

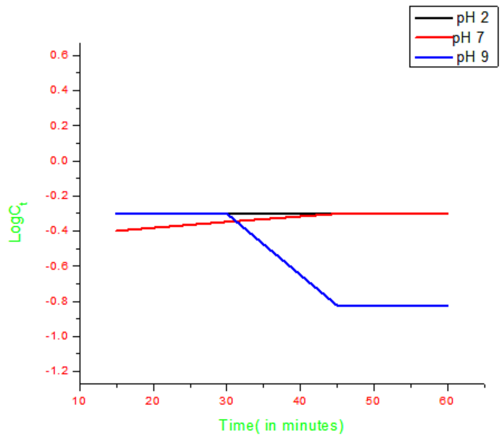

Fig. 20: Log C_i versus time(t) of *Cascabela thevetia* at initial con.5 ppm at pH 2, 7 & 9

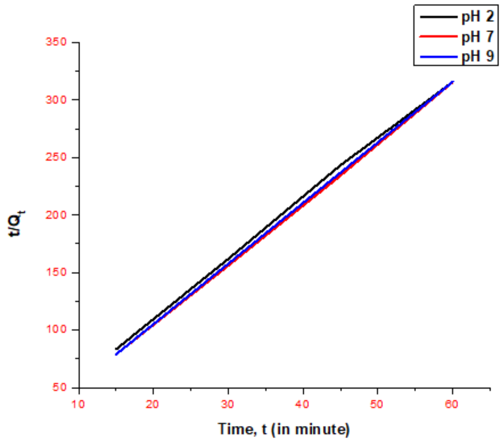

Fig. 21: t/Q_t versus t of *Cascabela thevetia* for Ci =2 ppm at pH 2, 7 & 9

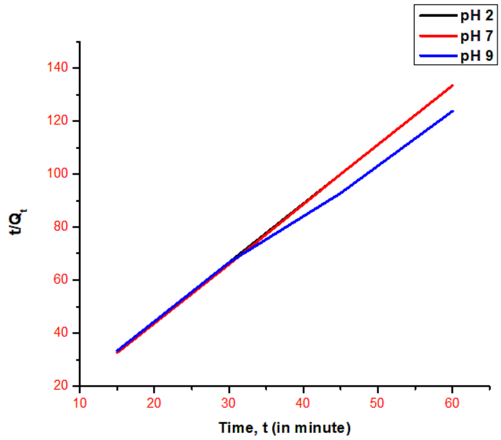

Fig. 22: t/Q_t versus t of *Cascabela thevetia* for Ci =5 ppm at pH 2, 7 & 9

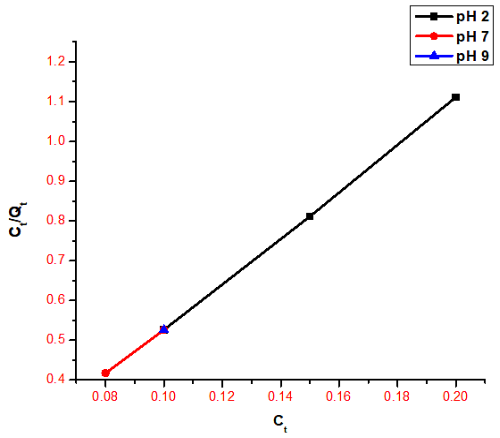

Fig. 23: C_t/Q_t versus C_t of *Cascabela thevetia* for Ci =2 ppm at pH 2, 7 & 9

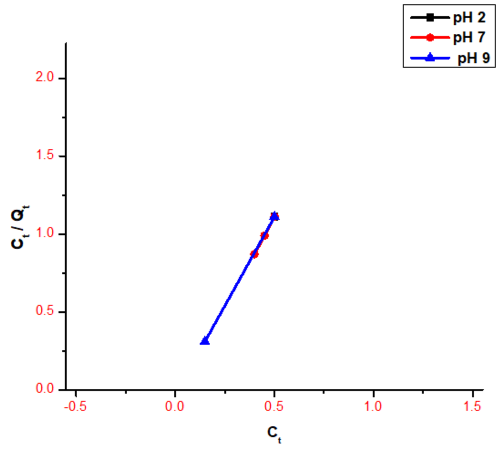

Fig. 24: C_t/Q_t versus C_t of *Cascabela thevetia* for Ci =5 ppm at pH 2, 7 & 9

Discussion

On one hand the residual concentration of arsenic by *T. cordifolia* is 0.10 ppm at pH 7, on the other the residual concentration of arsenic by *C. thevetia* is 0.10 ppm at pH 2 with an initial concentration of 2 ppm arsenic solution and maximum removal percentage is 95% (Table 2). With an initial concentration of 5 ppm, the maximum removal percentage is 97% at pH 9. HPLC has been utilized to observe the peaks before and after adsorption. The wavelength for UV detection is 227 nm clearly showing the peaks of the analyte.

HPLC of treated and untreated *T. cordifolia* and *C. thevetia* could be referred as an indication of adsorption clearly observed by change of peaks (Fig. 1, 2, 3 and 4) at wavelength 227 nm. HPLC has been utilized to observe the peaks before and after adsorption. The wavelength for UV detection is 227 nm which clearly shows the peaks of the analyte.

Fourier transmission infrared (FTIR) spectroscopy was done to explore the adsorption of As(III) by *T. cordifolia* and *C. thevetia*. The presence of -OH and –COOH groups, and lignin in the plant biomass can be explained as cause of adsorption of As(III) on the surface. The changes in shifts and intensity clearly indicate the adsorption on the surface.

Figures 11 and 12 show pseudo-first order kinetic model for an initial concentration of 2 ppm and 5 ppm respectively for *T. cordifolia* which clearly indicates that the experimental data is not fit for pseudo – first order kinetics.

Figure 13 and 14 represent pseudo – second order model of adsorption of As(III) by *T. cordifolia* for an initial concentration of 2 ppm and 5 ppm respectively at pH 2, 7 and 9. The linearity of the graph clearly showed the pseudo-second order reaction.

Figures 15 and 16 show Langmuir adsorption isotherm for *T. cordifolia* at initial concentration of 2 ppm and 5 ppm and pH 2, 7 and 9.

Figures 17 and 18 show Freundlich adsorption isotherm for *T. cordifolia*. Langmuir adsorption isotherm is a better fit for experimental data for *T. cordifolia*.

Figures 19 and 20 show pseudo first order kinetics for adsorption by *T. cordifolia* for initial concentrations of 2 ppm and 5 ppm at pH 2, 7 and 9.

Figures: 21 and 22 show pseudo second order model for adsorption by *C. thevetia* clearly indicating that this model is the fit for *C. thevetia*.

Figure 23 and 24: show the plot of C_t/Q_t versus C_t for adsorption of As(III) by *C. thevetia* for initial concentrations 2 and 5 ppm at pH 2, 7 and 9.

Comparison

T. cordifolia and *C. thevetia* have been widely used as a folk and ayurvedic medicine. The maximum removal efficiency of As(III) by these medicinal plants have been evaluated up to 97% which is very large in comparison to other adsorbents.

Conclusion

The present study revealed that the removal of As(III) solution was the best fit with pseudo second order model. Langmuir adsorption model suited the data which confirmed monolayer adsorption. The adsorption of As(III) by *T. cordifolia* and *C. thevetia* appears to be a complex process which may get affected by intra-particle diffusion, slow chemical diffusion, and chemisorption. If the medicinal plant is grown on arsenic contaminated soil, the medicinal values might get adversely affected. The soil growing the medicinal plants should be free from As(III) to get maximum benefits.

Thus, pseudo-second order reaction and Langmuir adsorption isotherms completely describe the adsorption mechanism and are the best fit for them.

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Conflict of Interest

The authors do not have any conflict of interest.

Data Availability Statement

The manuscript incorporates all datasets produced or examined throughout this research study.

Ethics Statement

This research did not involve human participants, animal subjects or any material that requires ethical approval.

Informal Consent Statement

This study did not involve human participants, and therefore informal consent was not required.

Author Contributions

- **Rajendra Kumar:** contributed to the methodology
- **Ashok Kumar Jha:** supervised and contributed to the analysis of data.
- **Raghbendra Thakur:** approved the daft.

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