

1 **Naturally derived carbon for *E. coli* and arsenic removal from water in rural**

2 **India**

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1 **Abstract**

2 Low cost solutions for water treatment in rural communities in India are key to improving
3 drinking water quality. Through a social enterprise, selected villagers from Khechare,
4 Maharashtra convert local wood and plant matter into a carbonaceous material (Village
5 carbon, "VC") which to date has been used for sale as deodorisers in urban areas. This work
6 demonstrates the effectiveness of VC material for bacterial filtering and arsenic adsorption.
7 BET surface area analysis determined that VC is mesoporous with a surface area of around
8 $95 \text{ m}^2 \text{ g}^{-1}$. Scanning electron microscopy revealed that much of the original fibrous features
9 and a wide range of elements from the local environment remain post carbonisation.
10 Bacterial filtering studies using *E. coli* as model bacteriological contaminant have been
11 demonstrated. Naturally relevant levels of bacteria (110 CFU / 100 mL *E. coli*) were removed
12 from water using a 15 g VC plug while >99-97% removal of highly contaminated (7×10^6 CFU
13 / 100 mL *E. coli*) were possible to be removed, demonstrating the capacity of the VC
14 material for bacteria filtering. Adsorption of As(V) and As(III) was achieved using VC, which
15 could reduce $200 \mu\text{g L}^{-1}$ As(III) to below $50 \mu\text{g L}^{-1}$ concentration, and $150 \mu\text{g L}^{-1}$ As(V) to 90
16 $\mu\text{g L}^{-1}$ concentration. The carbon generated from waste agricultural material used here is an
17 effective material for water purification on site. While its performance compared to other
18 literature carbonaceous materials may be slightly lower, it has great potential for use due to
19 its derivation from available waste products.

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21

22 **Keywords:** water disinfection; water treatment; heavy metals; developing countries; sus-
23 tainable development

1 1. Introduction

2 Clean drinking water is a basic human right although access to it is not widely established
3 especially for developing countries, where drinking water quality is low. India has a growing
4 water purification problem particularly in rural and peri-urban areas (Chaudhuri and Roy,
5 2017). As of the most recent census, around 830 million of India's 1.2 billion inhabitants live
6 in "rural" conditions (Anonymous, 2011). Water contamination issues in these areas can be
7 severe, particularly during the monsoon season when run-off is increased due to heavy
8 rainfall (Singh et al., 2004). People living in these conditions often suffer from waterborne
9 illnesses, such as diarrhoea, enteric fever, viral hepatitis, and cholera that can cause even
10 death (Mudur, 2003). Furthermore, excess amounts of heavy metals such as arsenic have
11 been detected in Indian groundwater (Ghosh et al., 2019), which can be consumed without
12 any prior treatment in many rural communities.

13 While effective in terms of removal of contaminants, large scale water treatment works are
14 unsuited for a widely dispersed population in remote locations and are currently prohibitively
15 costly to implement. Therefore, alternative efficient and sustainable technologies, such as
16 adsorption, photocatalysis, and solar thermal processes, are sought to provide clean
17 drinking water to such remote areas (De Vietro et al., 2019; Jun et al., 2019; Kumar et al.,
18 2019; Porley et al., 2020). Recent studies have shown that adsorption methods can
19 efficiently treat water using low-cost materials (Arias Arias et al., 2017; Kumar et al., 2019;
20 Tursi et al., 2018a; Tursi et al., 2018b; Tursi et al., 2019). Carbon is a well-known adsorbent
21 material (Smith and Rodrigues, 2015), with many studies detailing the effectiveness of
22 carbon upon removal of biological contaminants (Sharma et al., 2015) and toxic metals
23 (Kobya et al., 2020; Mohan et al., 2014) from water. One of the co-authors, Samuchit Enviro
24 Tech, has developed technology amenable to charring of loose biomass derived from
25 agricultural, forestry and garden wastes. This comprises a portable metallic kiln, based on
26 the Top Lit Up Draft (TLUD) principle.

1 The aim of this study is to take a new approach to water treatment by exploring whether the
2 carbon material (Samuchit Enviro Tech) generated from agricultural wastes by villagers in
3 Khechare (India) can be used as an effective material for water purification on site.
4 Experimental runs were carried out to remove pathogenic bacteria and heavy metals from
5 water.

6 **2. Materials and Methods**

7 **2.1 Materials**

8 The village carbon (VC) material is currently prepared on a developmental basis by selected
9 villagers in Khechare, Maharashtra, India. It is currently used for making an organic
10 deodoriser or air freshener for sale in urban areas, hence is a secondary livelihood option to
11 augment farming income. There is a hardware and labour element in preparing the VC,
12 however the raw material is free of cost. In our work, VC was ground using a mortar and
13 pestle for 5 min by hand before use. Sodium (meta)arsenite ($\geq 90\%$) and sodium arsenate
14 dibasic heptahydrate ($\geq 98\%$) were purchased from Sigma-Aldrich and used as received. The
15 *E. coli* strain used in bacteriological studies was the standard ATCC 23716 (American Type
16 Culture Collection, Rockville, MD, USA). A selective growth medium (Brilliance *E.*
17 *coli*/coliform agar (Oxoid) plates) was used in bacterial content determination experiments.

18

19 **2.2 Instrumentation**

20 BET surface area measurements were performed using a Quadrasorb Gas Sorption Surface
21 Area and Pore Size Analyzer. VC (0.8001 g) was outgassed under vacuum at 120 °C, leav-
22 ing 0.7557 g for N₂ uptake isotherm measurement. SEM image measurement and EDX
23 analysis was carried out using a Carl Zeiss SIGMA HD VP Field Emission SEM with Oxford
24 AZtec ED X-ray analysis system. ICP-OES measurements of arsenic were carried out using
25 a Perkin Elmer Optima 5300DV system.

26

1 **2.3 Methods**

2 *E.coli* filtration studies were carried out by packing VC (15 g) into a filter funnel fitted with a
3 filter paper (Whatman filter paper no. 1, 15 µm pore size), and then gently pouring 25 mL
4 samples of ultrapure, sterile water spiked with varying concentrations of *E. coli* to pass
5 through the apparatus under gravity. Repeat cycles were carried out in the same manner
6 using the same carbon filter set up. Bacterial content measurements were determined using
7 a serial dilution pour plate agar technique. Serial dilutions of the reaction solution were per-
8 formed in sterile aqueous solution and 200 µL of each dilution (including neat sample) were
9 pipetted and spread onto the selective growth agar plates. The plates were incubated at 37
10 °C for 20–24 h before viable colony counts were determined.

11 Arsenic adsorption studies were carried out initially using stocks of 4 ppm As(III) and As(V)
12 in ultrapure water. Various amounts of VC material was added to this stock and the resulting
13 suspension stirred overnight (at least for 16 h) before measurement. After determination of
14 the highest VC loading, pH adjustment of the stocks was achieved using aqueous 4 M solu-
15 tions of HCl and NaOH and the process repeated using the peak VC loading. Using the op-
16 timised loading and pH conditions, a final series of adsorption tests were carried out altering
17 the initial concentration of the As(V) and As(III) stocks by dilution of the original stocks to
18 give a range of different concentrations and repeating the adsorption process as before.

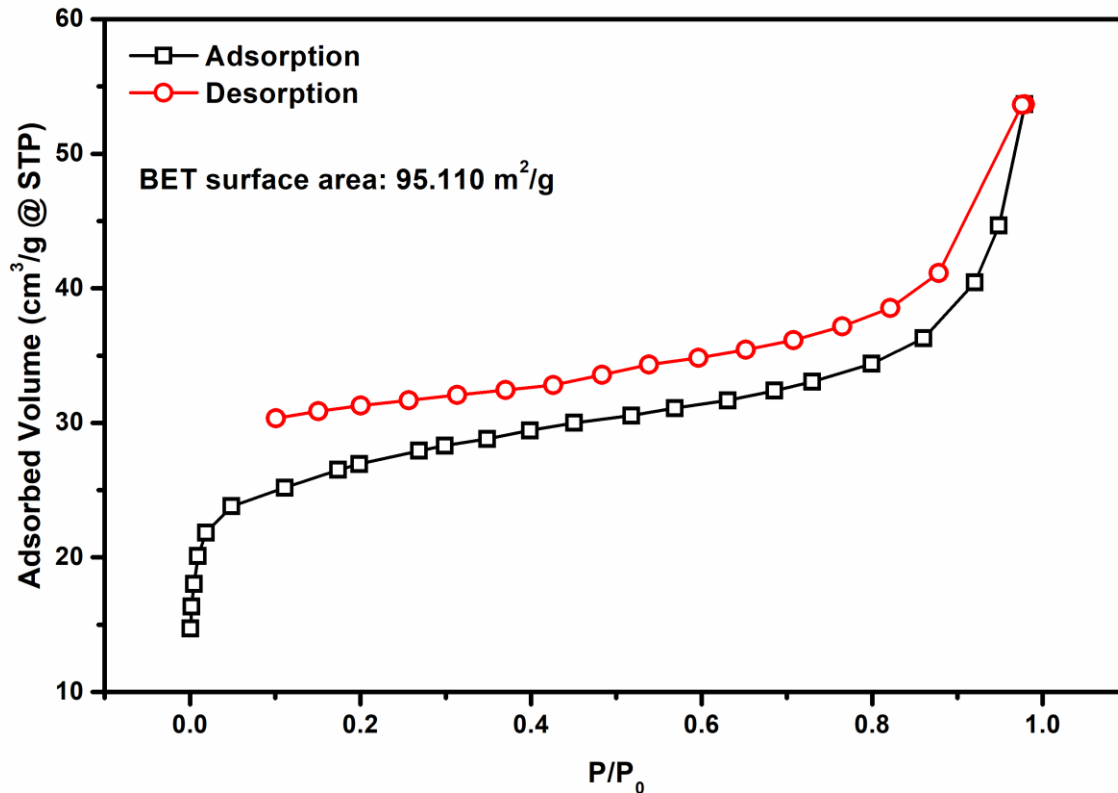
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21 **3. Results**

22 **3.1 Village Carbon Properties**

23 The surface area of VC was investigated by Brunauer-Emmett-Teller (BET) surface area,
24 displaying a type 4 N₂ uptake isotherm (Figure 1).



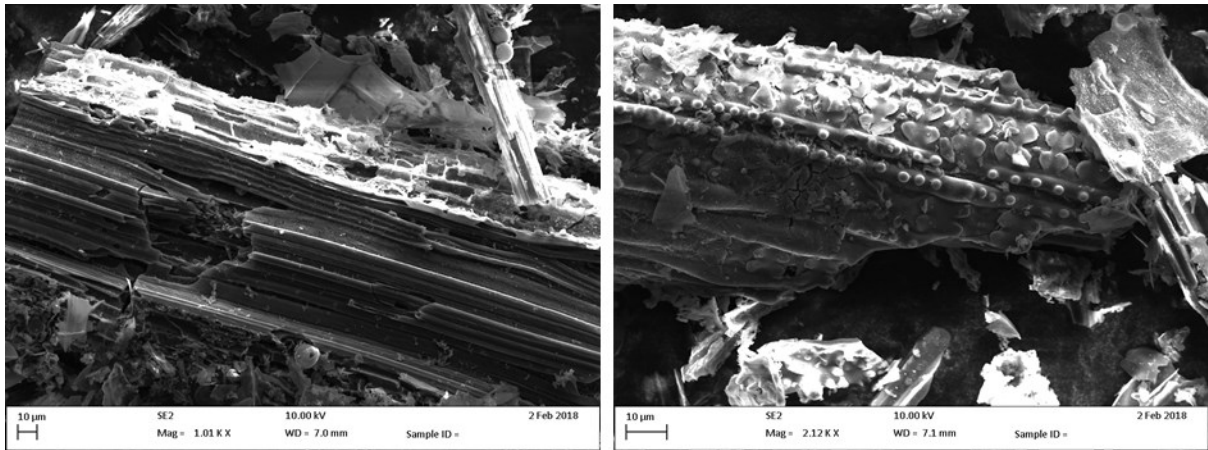
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2 Figure 1. N₂ uptake isotherm of the VC material showing a typical type 4 isotherm.

3

4 The type 4 isotherm indicates that the VC material has mesoporosity (Schneider, 1995),
 5 indicating pore sizes between 2 and 50 nm. According to this uptake isotherm the specific
 6 surface area of 95.11 g/m² can be calculated, which is within the range of values typically
 7 reported for natural carbon materials (Mohan and Pittman, 2007).

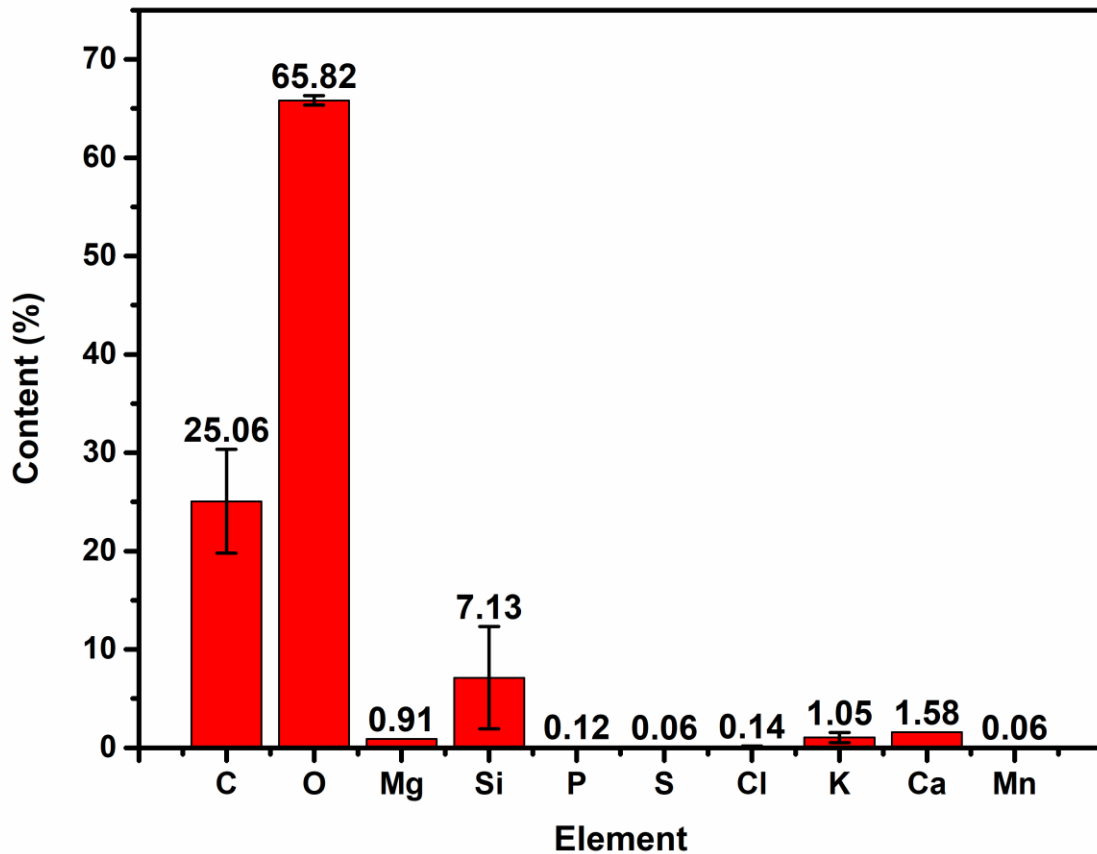
8 Scanning electron microscopy (SEM) images of the VC revealed that much of the structure
 9 of the plant matter from which the VC was derived remains after the carbonisation process
 10 (Figure 2). Many of the fibres and natural features of the source material can be seen in
 11 Figure 2.



2 Figure 2. Example SEM images of the VC material.

3

4 The biomass is not processed before charring, and is not densely packed or disturbed during
5 charring, which can explain why the char retains the shape and structure of the raw biomass.
6 The sample was found to be somewhat heterogeneous in nature, with a wide variety of
7 different particle shapes and sizes being observed, and many of the natural features such as
8 those in Figure 2 were found to be unique in the samples imaged. Additional SEM images of
9 the VC material are given in the supporting information (Figures S1- S4). In addition to
10 imaging, energy dispersive x-ray spectroscopy was carried out to identify the elements
11 present in the VC, which are summarised in Figure 3.



1

2

Figure 3. Elements found by EDX analysis of the VC material.

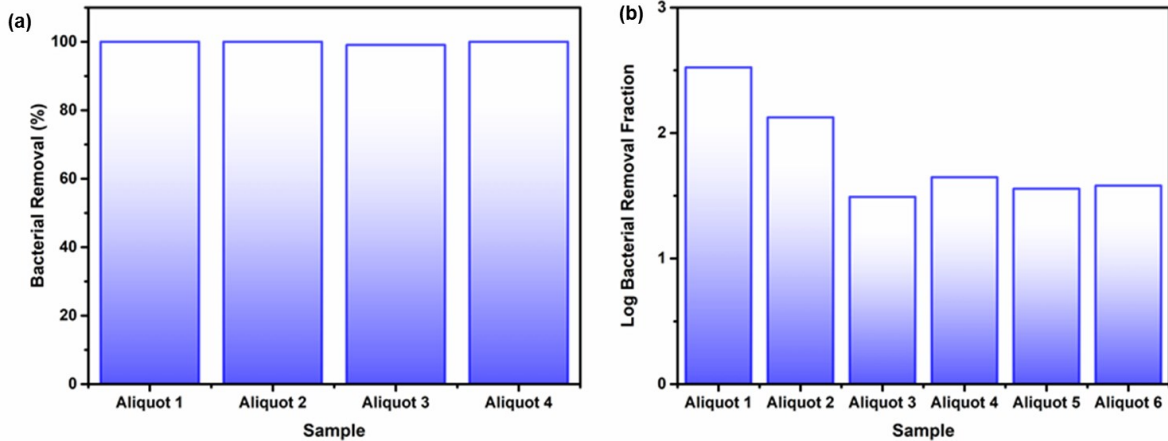
3 It was found within the sample of VC taken that different areas contained significantly
 4 different elements. The presence of a wide variety of elements is common in natural
 5 materials (Chia et al., 2012), and can be ascribed to the elements present in the
 6 environment from which they are derived. The only element other than carbon which was
 7 found consistently in high abundance in the samples was silicon. It has been noted
 8 previously that natural char material often contains a high silica content, due to concentration
 9 of Si in the feedstock during carbonisation due to losses of other elements (Lehmann J,
 10 2009; Tursi, 2019).

11 3.2 Bacterial Filtering

12 According to the WHO guidelines, no bacteria should be detectable in any 100 mL of
 13 drinking water sample (WHO, 2011b). It has been found that a typical drinking well in rural

1 India may contain between 1-100 colony forming units (CFU) of bacteria in a 100 mL sample
2 (Schriewer et al., 2015). *E. coli* is commonly used as a marker for bacterial content in water
3 samples (Edberg et al., 2000), and as such in this work the VC was tested against water
4 contaminated with *E. coli*. A 15 g plug of VC mounted upon a filter paper in a standard filter
5 funnel was able to provide 100% removal of samples of water containing 110 CFU / 100 mL
6 *E. coli* by simple passing of contaminated water samples through the filter under gravity
7 (Figure 4a). To probe the materials absorption capacity for bacteria, highly contaminated
8 water (7×10^6 CFU *E. coli* per 100 mL) was passed through the carbon plug. Images of
9 example agar plates demonstrating bacterial removal are given in the supporting information
10 (Figure S5 & S6). Despite the severe contamination level, the filter was still able to remove
11 >99 to 97% of the *E. coli* (Figure 4b). At no point during this test did the filter fail by
12 saturation, indicating the high capacity of the VC material for bacterial filtering. This suggests
13 that under regular (1-100 CFU/100 mL) operation that the VC material would be able to
14 handle a high quantity of water before failing due to saturation.

15 Considering domestic or village scale water treatment in developing world, where there is
16 usually abundant natural sunlight, appropriate disinfection methods include solar
17 disinfection, pasteurization, photocatalysis and filtration (Pichel et al., 2019; Porley et al.,
18 2020). All these technologies can effectively disinfect water although they may require higher
19 treatment times (> 5 h) than the proposed adsorption process (Domingos et al., 2019;
20 Monteagudo et al., 2017) (Porley et al., 2020).



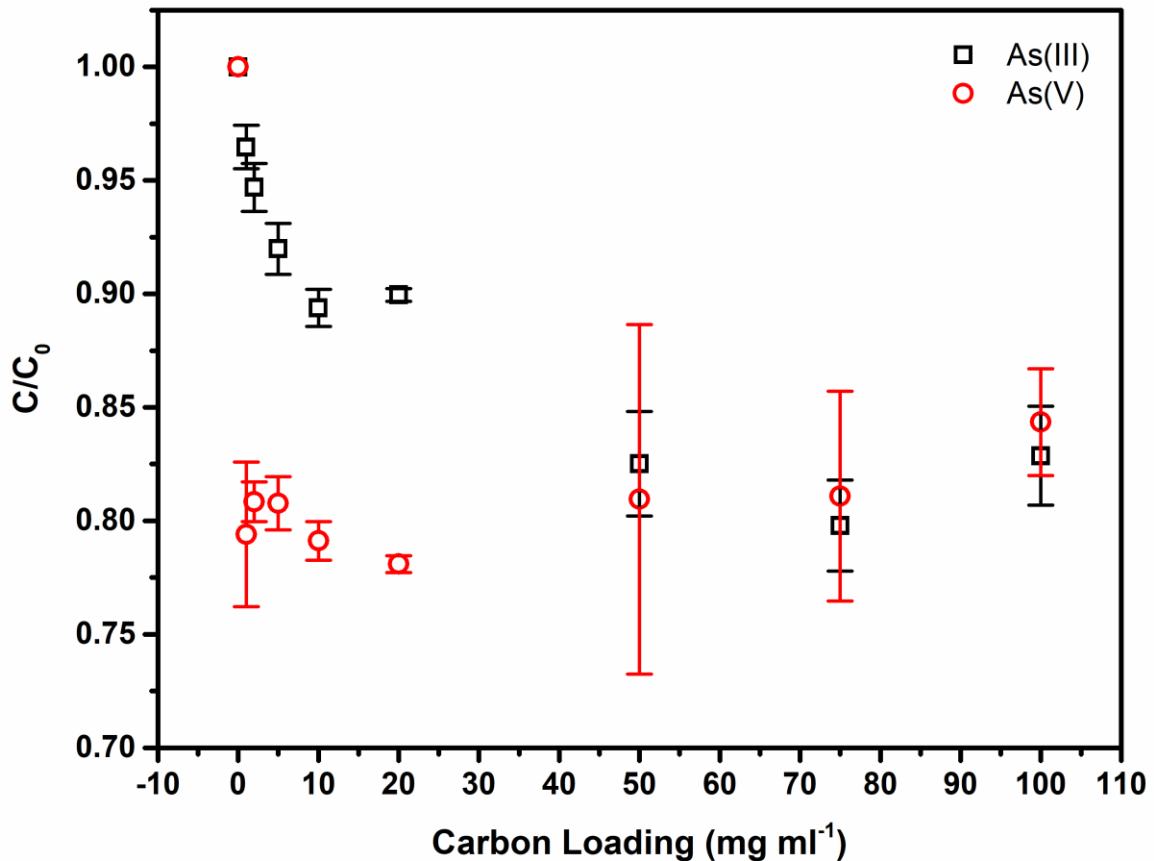
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2 Figure 4. (a) Percentage bacterial removal by the VC material using initial *E. coli*
 3 concentration 110 CFU/100 mL, and (b) Log Bacterial removal by the VC material using
 4 initial *E. coli* concentration 7×10^6 CFU/100 mL. In each case the aliquot represents a
 5 sequential 25 mL sample of contaminated water passed through the filter.

6

7 3.3 Arsenic adsorption

8 Arsenic contamination is a common issue in northern and eastern India and Bangladesh
 9 (Nordstrom DK, 2003). Naturally occurring arsenopyrite in the area enters ground water
 10 which may seep into borewells and other drinking water sources. The acceptable
 11 concentration of arsenic in drinking water according to the WHO is $10 \mu\text{g L}^{-1}$ (WHO, 2011a),
 12 while the Indian government accepts up to $50 \mu\text{g L}^{-1}$ where no better source exists (BIS,
 13 2012). Contamination of As in this area however in some extreme cases has been found to
 14 reach $1000\text{-}4000 \mu\text{g L}^{-1}$ ($1\text{-}4 \text{ mg L}^{-1}$) levels (Das et al., 1995), far beyond what is safe to
 15 consume. Various tests using the VC material were initially carried out on 4 mg L^{-1} solutions
 16 of sodium (meta)arsenite and sodium arsenate, the commonly occurring forms of As(III) and
 17 As(V) in water respectively. Firstly, the optimal loading of VC was investigated by
 18 suspending various amounts of VC in the above solutions and stirring overnight before
 19 measurement using inductively coupled plasma optical emission spectroscopy (ICP-OES)
 20 (Figure 5).



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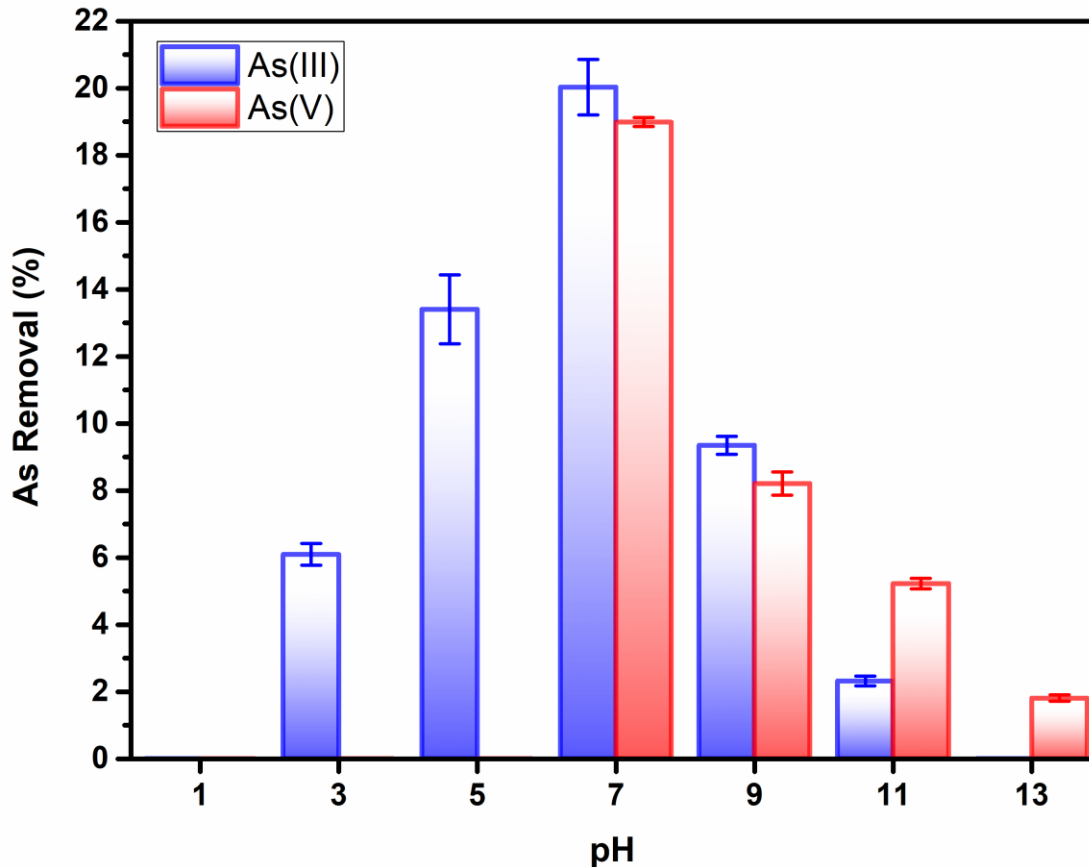
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Figure 5. Effect of VC loading on concentrations of both As(III) and As(V).

3

4 As shown in Figure 5 high loadings were required to achieve significant arsenic adsorption,
 5 likely due to the low surface area of the VC material. At very high VC loadings (>100 mg/mL)
 6 it was found that the adsorption failed to improve, which can be ascribed to poor mixing due
 7 to the As containing solutions becoming too thick with the addition of large quantities of VC.
 8 A loading of 50 mg/mL and 20 mg/mL was selected as optimal for further As(III) and As(V)
 9 tests respectively.

10 A common strategy to improve adsorption onto the surface of carbon materials is to alter the
 11 pH of the test solution by changing the surface charge of the carbon species (Al-Degs et al.,
 12 2000), and/or the chemical state of the arsenic species. Adopting this strategy allowed the
 13 optimal pH for the VC system to be found (Figure 6).



1

2 Figure 6. As(III) and As(V) removal percentage during adsorption by VC material at several
3 water pH values.

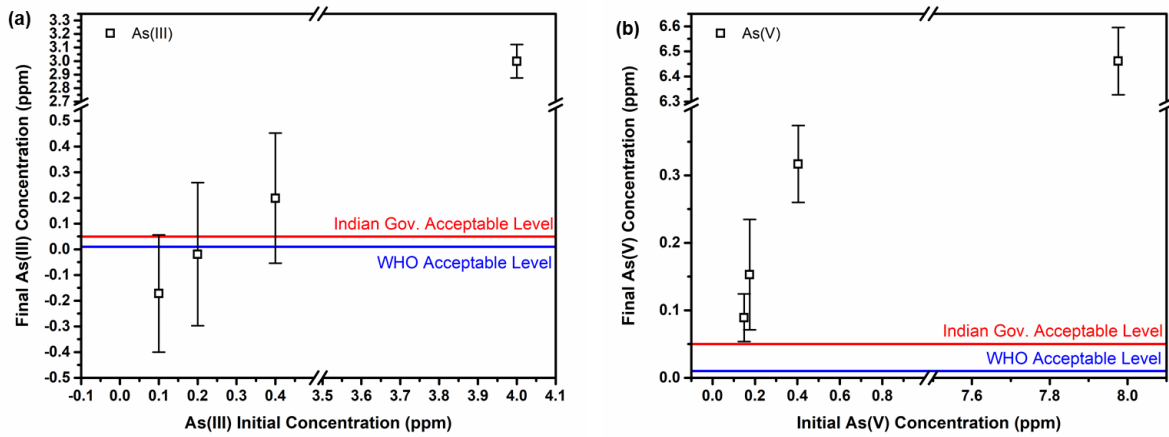
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5 It was found that at both low and high pHs the adsorption of both As(III) and As(V) was
6 significantly reduced, and that peak adsorption was in fact achieved at pH 7. While this
7 shows that unfortunately the adsorption cannot be improved in this way, it is advantageous
8 to not require addition of acids or bases which will require neutralisation pre-consumption as
9 well as constituting additional safety hazards for people dealing with them.

10 On the basis of the loading optimisation and subsequent confirmation of optimal conditions,
11 the adsorption capacity for As(III) and As(V) can be determined for the VC material to be 28
12 $\mu\text{g g}^{-1}$ and $76 \mu\text{g g}^{-1}$, respectively. These values are at the lower end of the range reported in
13 the literature for As adsorption on naturally derived carbon [8]. Based on this capacity for As

1 uptake, lower concentrations of the same As(III) and As(V) solutions were screened to
2 obtain the level of these species removable by the VC (Figure 7).

3



4

5 Figure 7. Removal of (a) As(III), and (b) As(V) in comparison to acceptable levels for
6 consumption.

7

8 Despite the higher capacity displayed for As(V) at higher concentrations, it was found that
9 the VC material failed to adsorb this form of arsenic to an acceptable level at lower
10 concentrations, while As(III) was readily adsorbed to below the level acceptable to the Indian
11 government when beginning with 100 and 200 $\mu\text{g L}^{-1}$ of As(III). This material therefore is
12 usable to remove low and intermediate levels of As(III) to an acceptable concentration, and
13 is able to reduce high concentrations significantly of both As(III) and As(V), but is unable to
14 sufficiently remove As(V) from solution.

15 Kumar et al. (2019) have very recently reviewed eco-friendly and simple to use technologies
16 for arsenic removal from water. They focused on low-cost equipment processes that are
17 applicable to small-scale household applications and to remote rural and peri-urban areas,
18 such as the Indian case study. Amongst several technologies, including oxidation, filtration,
19 and coagulation, it was concluded that adsorption technologies based on locally sourced

1 materials deserve further advancement to provide a sustainable technology for water
2 treatment in such regions. It was found that arsenic adsorption rates, in the presence of
3 locally sourced adsorbents, were moderate, which is consistent with the results of this study.
4 Nevertheless, efficiency was still better than other technologies, such as
5 coagulation/precipitation and lime softening, in which arsenic removal was poor, or such as
6 oxidation technologies with relatively slow adsorption rates (Kumar et al., 2019). On the
7 other hand, conventional filtration, micro-filtration, ultra-filtration, nano filtration, and reverse
8 osmosis achieved high removal rates only when initial arsenic concentration was low, while
9 Ar(III) removal was poor.

10

11 **4. Conclusions**

12 Carbon filtering and adsorption is a key technique which may be used in the purification of
13 water in rural communities in India. The aim of this study was to explore whether the carbon
14 material generated from agricultural wastes by villagers in Khechare(India) can be used as
15 an effective material for water purification on site. It was found that the VC material has good
16 removal capacity for *E. coli* with no failure due to saturation even when exposed to an
17 extremely high quantity of bacteria. It has also demonstrated some effectiveness for the
18 removal of both As(III) and As(V), which peaks at natural pH levels, however only As(III) was
19 able to be reduced to a level that could be considered safe for consumption. Carbon
20 materials derived from agricultural wastes (light bio-mass) are therefore promising for
21 simple, low-cost solutions to the problem of water purification in remote and rural areas.

22

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3 training from Samuchit Enviro Tech to set up the charring operation.

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