



# **Arsenic Mitigation Approach in Soil by Some Indigenous Sources of Biochar Made at Low Pyrolysis Temperature**

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## **Authors' contributions**

*This work was carried out in collaboration between both authors. Author SMIH designed the study, and guided the author NF who performed the laboratory analysis, wrote the protocol and wrote the first draft of the manuscript. Author SMIH revised the manuscript before submission. Both the authors read and approved the final manuscript.*

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## **ABSTRACT**

Biochar is being reported now a-days to potentially reduce the bioavailability of arsenic (As). A pot culture and an *in vitro* incubation study was conducted to evaluate the efficacy of biochar produced from different sources (*viz.*, cow dung, poultry manure and sewage sludge) at low temperature (250°C) on the phytoavailability of As. An experimental setup containing two sets of treatments (As and no-As) were followed. Biochar was applied at the rate of 5t/ha to the soils that received As treatment at a rate of 1mg/L As (80% arsenite and 20% arsenate). After 45 days of growth of Kalmi (*Ipomoea aquatica*), plant and soil samples were analyzed. Biochar insignificantly increased plant growth. Biochar was observed to be more effective in alleviating As accumulation in plant than the biomass. The incubation study was done for 0, 15, 30 and 45 days to diagnose the temporal sorption of water soluble As by biochars. All the biochar materials reduced As availability than their corresponding biomass materials. Measurement of different physical (e.g. surface area, morphology, elemental composition), chemical (CEC, nutrient content, As) and physicochemical

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(pH) properties of the biochar showed that through pyrolysis process nitrogen(N), sulfur (S), phosphorus (P) content and pH of the biomass decreased and on the other hand potassium (K), As content and CEC increased. Slow pyrolysis process and variation in biomass material influenced the properties and behavior of biochar. Among the biochars, sewage sludge biochar showed the maximum and cowdung showed the minimum sorption capacity for As. Due to the threat of As to health and environment, this topic requires more consideration. Moreover, covering all the above issues, this study identifies research gaps in the use of biochar as an adsorbent for As and proposes potential areas for future application of biochar.

*Keywords: As; phytoavailability; biochar; feedstock; pyrolysis; temperature.*

## 1. INTRODUCTION

Massive health hazard and a large-scale damage of socio-economic structure due to environmental pollution caused by metals is one of the talks in the present era [1]. The volume of toxicity per year from these inorganic materials released into water and soil has been estimated to be higher than the volume of toxicity accounted for both organic and radioactive sources together. In addition, a fair share of these inorganic substances is responsible for soil contamination. The adverse consequences on environment of substances like As from omnipotent sources such as, volcanoes, forest wild fires, erosion from mineral deposits and so on are at the center of concern today [2].

Arsenic itself occupies approximately 1.8mg/kg in the earth's crust; however, this amount increases due to anthropogenic pollution [3]. Approximately, 80% of the mean amount of As reaches to soil due to anthropogenic activities and the plants receive it via root uptake and also through foliar adsorption. As a result, As enters into the food chain. Bio-concentration of As occurs particularly in algae and lower invertebrates [4]. Arsenic gets released into water bodies in small quantities through these types of biological sources (biogenic). A recent empirical study found that an episode of extensive As contamination of groundwater in Bangladesh and the Indian state of West Bengal is of geological origin, transported by rivers from sedimentary rocks in the Himalayas over thousands of years, rather than anthropogenic sources [5]. Elevated concentrations of As in ground water have been reported in many parts of the world, especially Argentina, Chile, Mexico, China, Hungary, West Bengal (India), Bangladesh, Vietnam, and the USA [6]. Interestingly, every year, some new locations are found with high level of As concentrations [1].

Since overwhelming spread of As pollution is less likely to get reduced, decontamination of soil

is not a viable option. Therefore, effective methods need to be introduced to prevent As being released into the soil. In that case, several methods, such as, application of zinc fertilizer [7], application of balanced fertilizer [8], management of water regime [9], use of organic matter [10], phytoremediation through fern [11] have been applied.

Furthermore, retention of static As concentration in soil is possible by increasing surface area, forming complex with soil or by using other commercial adsorbents like activated carbon (C), titanium, agricultural wastes (rice husk), iron oxide granules that are not harmful for soil or plant [12,13]. But all these materials are either speciation specific or pollutant specific, not suitable for simultaneous removal of As from soil solution [14]. Among various adsorbents used for decontamination of soil, activated C is one of the most effective one. It covers a wide spectrum of applications in reducing phytotoxicity of many herbicide residues and other chemicals in agricultural soils due to its hydrophobicity, high specific surface area and microporous structure. However, soil remediation through activated C is highly expensive. Biochar has recently drawn attention as an alternative to activated C. Biochar is an organic charcoal material which is cost effective and environmentally feasible and derived by the pyrolysis of certain feedstock of agricultural biomass in absence of oxygen (O). Absence of O in the system controls the burning that results in capturing much more of the natural C from the biomaterial [14].

In the past decade, biochar has been experimented extensively in solving various agricultural and environmental problems such as immobilization of contaminants, improvement of soil health and C sequestration in relation to climate change [15,16,17]. Biochar can be produced from plant remaining that are often burnt in fields or buried in landfill and have a triple line benefit namely economic, reduction of

polluted soil and water and production of renewable energy [18]. Biochar has a well-developed pore structure, large specific surface area, abundant O containing functional groups and excellent adsorption performance.

Biochar immobilizes heavy metals in soil by electrostatic attraction, ion exchange and surface complexation with functional groups [19,20]. Many studies have reported an excellent ability of biochar to remove heavy metals, organic and inorganic pollutants from aqueous solutions [21,22]. Biochar has also evidenced to reduce the concentration of plant extractable As in soil [23]. Most of the chemical interactions between biochar and the environment are directly related to its surface chemistry. When biomass materials are pyrolyzed into char, they develop very different surface chemistry. Most of the functional groups like hydroxyls (-OH), carboxylic acids (COOH) and small alkyl chains such as methyl groups (-CH<sub>3</sub>) (containing O, H and N) volatilize off, leaving aromatic C surfaces behind. These surfaces are reduced, non-polar, and hydrophobic. When the surface gets exposed to air over time, the C oxidizes, creating new O-containing aromatic functional groups such as hydroxyls (-OH), carbonyls (-C=O) and carboxylic acids (-COOH), and making the surface polar. These functional groups are similar to those found on soil organic matter and are critical for biochar-soil interactions in similar ways. These functional groups can receive or donate a proton (H<sup>+</sup>) depending on the pH. At a higher pH, the carboxylic acids (-COOH) and some of the hydroxyls (-OH) give up protons and become negatively charged (-COO<sup>-</sup> and -O<sup>-</sup>, respectively). At low pH environments, these same groups can accept a proton. In this way, biochar acts as a weak acid and partially buffers the pH of the system. The ash fraction of the feedstock affects pH separately and may override any effect of the carbon fraction, especially with high-ash, alkaline chars. The negatively charged surface functional groups can attract positively charged cations and thus contribute significantly to the soil's cation exchange capacity (CEC). In cases of metal toxicity due to low soil pH, biochar can help in two ways: raising the pH, which makes plant-toxic metals like aluminum (Al<sup>3+</sup>) less soluble, and adsorbing the positively charged metal ions, which removes them from the solution. Finally, the hydrophobic and hydrophilic regions of the biochar surface can serve as adsorbents for non-polar and polar organic molecules in the environment. On the downside, these same

surfaces might also adsorb a pesticide and reduce its effectiveness [24]. Due to its versatile functionality as surface adsorbent, the demand of research on biochar is increasing day by day.

The effect of microorganisms on biochar is difficult to determine, as biochar is quite recalcitrant to microbial attack due to its long half-life [25]. Due to the increasing temperature of pyrolysis process, the C fraction in char develop new aromatic C-C bonds. The "orderliness" of the aromatic structures also increases with increasing temperature, forming gradually larger sheets of interconnected aromatic rings. The C in dense aromatic structures are more resistant to oxidation and few microorganisms have enzymes capable of breaking down such bonds [24]. Again, according to studies, a large part of biochar is mineralized over a short time-scale and a small part remains in a very stable, high aromatic form, displaying greater C-14 age than the oldest soil organic matter fraction. Microorganisms are able to change the amount and properties of biochar in soil, which may have significant influence on nutrient cycles and nutrient availability to plants. Pore geometry and size distribution of biochar has been found to promote growth and activity of certain microorganisms. Biochar provides suitable habitat for large and diverse group of soil microorganisms [25]. On the other hand, studies on the effects of biochar on soil organisms showed that, biochar exerted negative effect on soil microbial population. It reduced the growth of *Bradyrhizobium*, sulphate reducing bacteria and iron oxidizing bacteria [26]. An incubation study was conducted to observe the effect of biochar and biomass on the abundance of *Bacillus badius*, *Bacillus krulwichiae*, *Bacillus siralis*, *Bacillus sylvestris*, *Bacillus flexus*, *Aneurinibacillus aneurinilyticus* and *Bacillus thuringiensis*. Here, biochar treated soil showed the disappearance of all the biocterial isolates except *Bacillus badius*, which was the most tolerant one [27].

The physicochemical properties including pore structure, surface area and adsorption properties of biochar are very complex and depend on the production temperature and feedstock composition and residence time [19,28]. Biochar prepared at low temperature generally has lower pH, higher water holding capacity, lower specific surface area, more surface functional groups (i.e., carboxylic, phenolic, hydroxylic etc.) and higher CEC [29,30]. As pyrolysis temperature increases, the degree of carbonization of the

feedstock increases as indicated by the increased aromatic C content and decreased hydrogen content, lower hydrophobicity and decreased polar functional groups in the resulting biochar [31].

Different feedstocks have different ratios and forms of organic biomass building blocks (cellulose, hemicelluloses, lignin, extractives etc.) each of which reacts by different pyrolysis mechanisms. Different feedstocks also have different mineral compositions that catalyze some pyrolysis reactions and dictate the mineral composition of the resulting chars [32].

A systematic study should be done for investigating the effect of different feedstock materials on the surface characteristics as well as the chemical and the physicochemical properties of biochar. In the recent years, many studies have been conducted on the efficacy of biochar in ameliorating various agricultural and environmental problems [33,15,17]. The excellent ability of biochar to immobilize heavy metals and improvement of soil health has been reported by many study findings [19]. Only a few studies are focused on As immobilization in soil and at the same time, there has not been a center of attention to understand the As alleviation in soil by different biochar materials produced from various pyrolysis temperatures and feedstock material. The objective of our study was to find out the differences in various characteristics of biochar (surface area, pore volume, pH, CEC, nutrient content etc.) resulting from the difference in biomass sources (*i.e.*, cowdung, poultry manure, sewage sludge) and their impact on the phytoavailability of As in soil.

## 2. MATERIALS AND METHODS

### 2.1 Biochar Preparation

The different sources of biomass/feedstock materials (*i.e.*, cowdung, poultry manure and sewage sludge) were collected locally for biochar preparation. The biomass samples were air dried and ground. Biochar was produced at low (+250°C) temperature pyrolysis process. An earthen pot containing the biomass and covered with an earthen lid was heated on an open fire. During this process, air was not allowed to enter into the pot and the temperature of the pot was maintained. It took about 45 minutes to turn all the biomass into biochar [34]. In this slow pyrolysis process, the total biomass gets converted into about 30% of water, 35% of

syngas and 35% of char [35]. Both biomass and biochar samples were screened through 5mm sieve for pot experiment and 0.25 mm sieve for various chemical and physicochemical analysis.

**Chart 1. The biomass and biochar samples were labeled as follows**

Biomass	Biochar
Cowdung biomass=C <sub>bm</sub>	Cowdung biochar=C <sub>bc</sub>
Poultry biomass=P <sub>bm</sub>	Poultry biochar=P <sub>bc</sub>
Sewage sludge biomass= S <sub>bm</sub>	Sewage sludge biochar= S <sub>bc</sub>

### 2.2 Soil Sample Preparation

Soil sample was collected from a depth of 0-15cm by composite soil sampling method [36] from Manikgonj Sadar Upazila in Manikganj district (23°51.884'N and 90°06.219'E), Bangladesh. It is a Young Brahmaputra Floodplain soil belonging to the Melandaha series. According to the USDA soil taxonomy, the soil is a typic endoaquepts belonging to the order Inceptisols. According to the FAO-UNESCO legend, it is Gleysol [37]. The Soil was air dried, cleared off the debris and crushed to make the bigger clods smaller. The crushed soil samples were screened through 5mm sieve. The sieved samples were then mixed thoroughly for making a composite sample. Soil sample was used for pot experiment and incubation study.

### 2.3 Background Analysis of Soil and Biochar Samples

The texture, pH, CEC and N of the soil and biochar samples were determined using Marshall's triangle by hydrometer method, glass electrode method, ammonium acetate extraction method and Kjeldahl's method respectively [36]. For the total elemental analysis of P, K and S; the soil and the biochar samples were digested with aqua regia. The total K and S was determined by flame photometer. The total P content was determined using spectrophotometer. The As content of the soil (both initial and after harvest and also of the incubated soil), biochar and plant samples were determined by 'Hydride Generation Atomic Absorption Spectrophotometry (HGAAS)' [38]. All the statistical analysis was done using Minitab (version 16 package).

### 2.3.1 Surface properties

The surface area, total pore volume, mean pore diameter of biochar were measured by BET plot method by BELSORP mini-II. It is a compact, precision instrument for measuring surface area and pore size distribution by the volumetric gas adsorption technique.

### 2.3.2 Morphology

Surface morphology of biochar was measured by SEM (Scanning Electron Microscope) and elemental content (C and As) by EDX (Energy Dispersive X-ray Spectroscopy). Different sources of biochars were saturated with As solution (1 ppm) for 15 days. Then SEM-EDX was done for both As treated and As non-treated biochar samples.

### 2.4 Pot Culture Experiment

Kalmi plant (*Ipomoea aquatica*) was used as a study plant in pot culture experiment using 7 treatments including control (Table 4) arranged in a completely randomized design. Plastic pots without hole at the bottom were filled with 1kg soil per pot. According to the design setup, soil of each pot was mixed with different biochar samples at a rate of 5 t/ha i.e. 2gm/kg. The pots were kept at ambient condition for 15 days for the biochar to be stabilized when the seeds were sown (10 seeds per pot). After the germination, 7 seedlings were kept in each pot and allowed to grow. After three days of germination, Arsenic (80% arsenite + 20% arsenate) at a rate of 1mg per liter was applied every day at a rate of 100ml per day as irrigation water. The plants were irrigated with 100ml of irrigation water every day. But during the rainy days, no irrigation was done. The plants were uprooted after 45 days of growth. The plants were dried and processed [36]. Soil samples from each pot was also collected after harvest and processed as described above.

### 2.5 Incubation Study

An *in vitro* incubation study was also conducted to see the sorption and release of As. For incubation, 500g sized plastic pots were filled with 450 g soil sample. Then the soils were incubated at field moisture condition for 0, 15, 30 and 45 days. The field moisture condition was maintained by visual observation. The experimental setup was similar to that of the pot culture experiment. After incubation, the soil from each of the pot was collected for further analysis.

## 3. RESULTS AND DISCUSSION

### 3.1 Results

#### 3.1.1 Background analysis soil, biomass and biochar

Table 1 shows the laboratory analysis of soil, biochar and biomass for pH, CEC, texture, As, N, P, K and S contents. Analysis of soil sample showed that the soil had lower CEC, N and K content but higher P content than biomass and biochar. As content of soil was below detection limit (BDL). Biochar was observed with lower N, P, S content and pH (except  $C_{bc}$ ), but higher CEC, K and As content than their corresponding biomass materials.

#### 3.1.2 Surface properties

The test results of surface properties examination are shown in Table 2.  $C_{bc}$  showed highest surface area ( $12.355 \text{ m}^2\text{g}^{-1}$ ) and lowest pore diameter (12.590 mm).  $P_{bc}$  showed the lowest surface area ( $2.8330 \text{ m}^2\text{g}^{-1}$ ) but highest pore diameter (35.792 mm). On the other hand,  $S_{bc}$  showed the highest pore volume ( $4.5399 \text{ cm}^3\text{g}^{-1}$ ) and  $P_{bc}$  showed the lowest pore volume ( $2.5350 \text{ cm}^3\text{g}^{-1}$ ).

**Table 1. Some physical, chemical and physicochemical properties of the soil and biochar**

Properties	Soil	$C_{bm}$	$C_{bc}$	$P_{bm}$	$P_{bc}$	$S_{bm}$	$S_{bc}$
pH	6.61	7.45	8.13	7.65	7.64	6.52	6.46
CEC (me/100g)	0.23	5.9	39.8	4.59	16.9	1.87	6.35
Textural class	Silt loam		-		-		-
Moisture content (%)	21.54		-		-		-
N (%)	0.097	93.33	8.47	106.03	41.49	79.98	18.59
P (ppm)	3.23	1.37	1.08	8.49	0.71	5.99	0.21
K (me/100g)	0.11	0.66	1.48	0.93	1.48	0.21	2.69
S (ppm)	10.17	41.07	11.4	20	15.0	123.57	
As (ppm)	BDL*	0.86	0.93	1.11	1.35	3.52	6.82

**Table 2. Surface area, pore volume and mean pore diameter of biochar**

Sample	Surface area ( $\text{m}^2\text{g}^{-1}$ )	Total pore volume ( $\text{cm}^3\text{g}^{-1}$ )	Mean pore diameter (mm)
C <sub>bc</sub>	12.355	3.8888	12.590
P <sub>bc</sub>	2.8330	2.5350	35.792
S <sub>bc</sub>	6.4659	4.5399	28.085

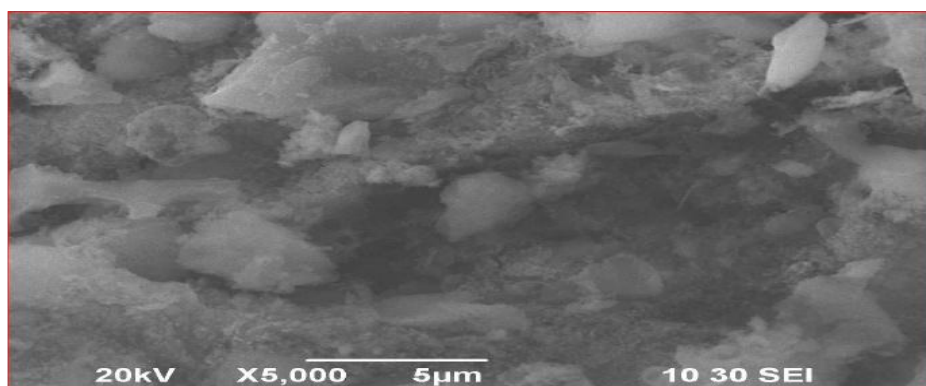
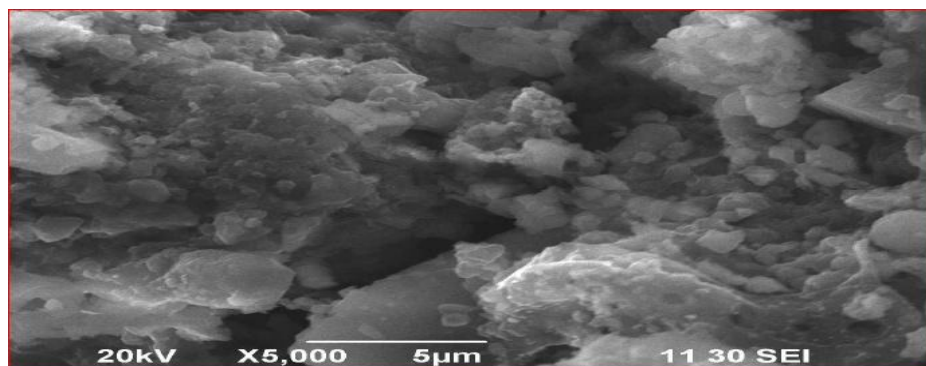
From SEM images (Figs. 1-3) it can be observed that, the C<sub>bc</sub> and S<sub>bc</sub> are mostly of clodded structure. On the other hand, P<sub>bc</sub> is observed to have more sharp and porous structure.

SEM-EDX analysis of the biochar samples (Table 3) showed that, in both saturated and unsaturated condition, As sorption was highest for S<sub>bc</sub> (in saturation 9.5% and in instauration 17.5% by weight) and lowest for C<sub>bc</sub> (in saturation 3.6% and in instauration 1% by weight). A correlation coefficient analysis between C and As content of the biochar materials (of both weight and atomic percentages) showed that, there is a perfect negative correlation (-1) between C and As content of each of the three biochars. This value indicates that, the higher is the As

content of the char, the lower is the C content and vice versa

### 3.1.3 Plant growth

Different biochar and biomass treatments showed different effects on plant growth (Table 4). In As non-treated soil, biomass showed a higher efficacy in plant growth than their biochar derivatives except P<sub>bc</sub>. Again, in As-treated soil, biomass materials showed higher efficacy in plant growth than their corresponding biochar materials except C<sub>bc</sub>. According to ANOVA test results, there is no significant effect of the biochar and biomass treatments on fresh and dry weight production of Kalmi in As non-treated soil and in As-treated soil, effect of the treatments was significant for dry weight at 1% level of significance but not significant for fresh weight.

**Fig. 1. SEM image of C<sub>bc</sub>****Fig. 2. SEM image of P<sub>bc</sub>**

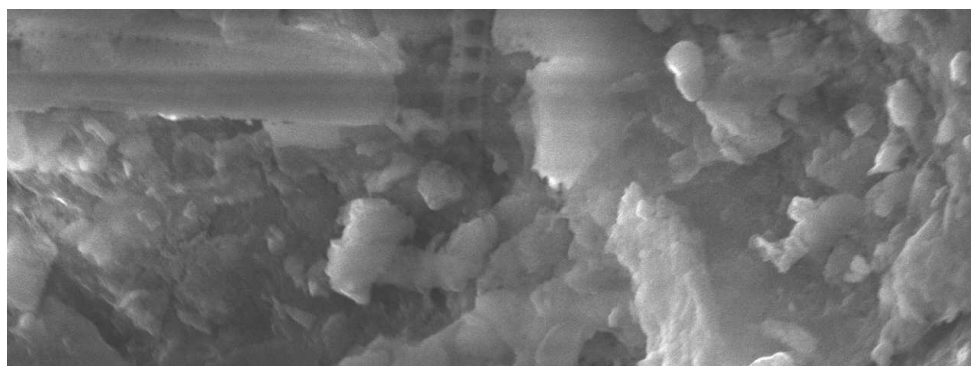


Fig. 3. SEM image of S<sub>bc</sub>

Table 3. SEM-EDX

Saturated				Unsaturated			
		C%	As%			C%	As%
C <sub>bc</sub>	Weight	96.4	3.6	C <sub>bc</sub>	Weight	99	1
	Atomic	99.4	0.6		Atomic	99.8	0.2
P <sub>bc</sub>	Weight	94.3	5.7	P <sub>bc</sub>	Weight	98.2	1.8
	Atomic	99	1		Atomic	99.7	0.3
S <sub>bc</sub>	Weight	90.5	9.5	S <sub>bc</sub>	Weight	82.5	17.5
	Atomic	98.3	1.7		Atomic	96.7	3.3

Table 4. Fresh and dry weight production of Kalmi plant (g/100plant)

Sample	As non-treated soil		As treated soil	
	Fresh weight	Dry weight	Fresh weight	Dry weight
Co	101.4	5.32	98.5	6.75
C <sub>bm</sub>	113.92	6.2	108.6	7.66
P <sub>bm</sub>	100.9	5.4	114.7	7.1
S <sub>bm</sub>	104.5	6.33	104.2	6.77
C <sub>bc</sub>	80.67	4.3	112.2	7.56
P <sub>bc</sub>	111.53	6.58	96.0	7.3
S <sub>bc</sub>	99.3	10.2	103.1	4.22

\*Co =Control soil

### 3.1.4 As accumulation in plant

The concentration and uptake of As by Kalmi plant showed that, in absence of As treatment (Figs. 4 and 5), biomass treatments decreased As concentration and uptake in plant with an exception to C<sub>bm</sub>. In presence of As treatment, biochar reduced As concentration and uptake in plant except S<sub>bc</sub>.

In absence of As treatment, among the biomass treated soils, maximum concentration (4.58 ppm) and uptake (28.4 µg/100 plants) of As was found in plant treated with C<sub>bm</sub> while the minimum As concentration (1.08 ppm) and uptake (6.8 µg/100 plants) was observed in plants treated with S<sub>bm</sub>. In presence of As treatment, maximum As concentration (7.99 ppm) uptake (99.2 µg/100

plants) was observed in plants treated with C<sub>bm</sub> and plants treated with S<sub>bm</sub> showed minimum As concentration (1.24 ppm) and uptake (31.07 µg/100 plants).

Among the biochar treatments, in absence of As, maximum As concentration (4.64 ppm) was observed for C<sub>bc</sub> treatments, maximum uptake (37.74 µg/100 plants) was observed for S<sub>bc</sub> treatment. Minimum As concentration (2.69 ppm) and uptake (29.35 µg/100 plants) were observed for P<sub>bc</sub> treated soil. In presence of As treatment, C<sub>bc</sub> treatment showed the maximum As concentration (5.18 ppm) and uptake (39.16 µg/100 plants). On the other hand, S<sub>bc</sub> treatment showed the minimum As concentration (3.92 ppm) and P<sub>bc</sub> showed the minimum uptake (29.35 µg/100 plants).

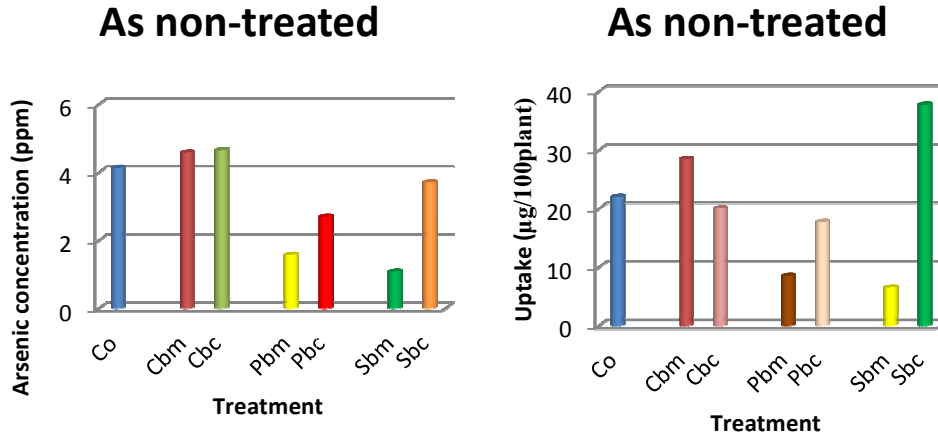


Fig. 4. As accumulation in both biomass and biochar treated kalmi plant in absence of As treatment

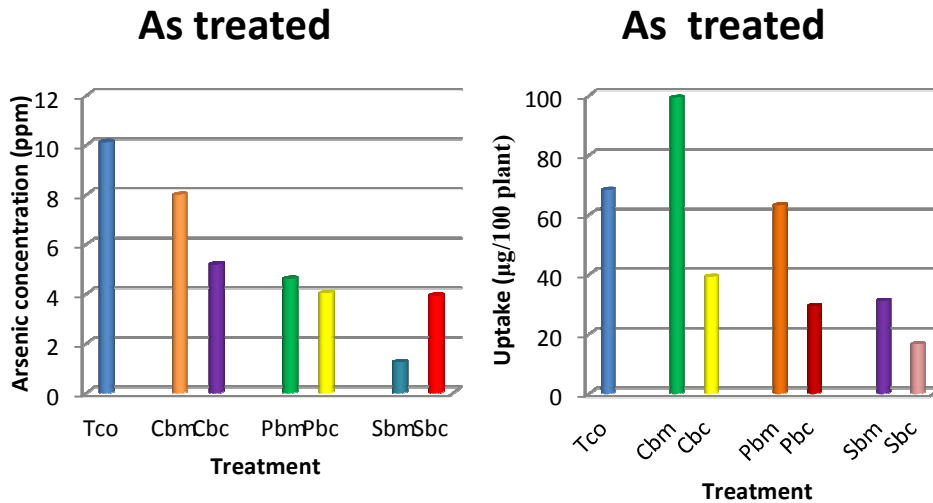


Fig. 5. As accumulation in both biomass and biochar treated plant in presence of As treatment

ANOVA test result shows that, both biochar and biomass treatment showed significant effects on As concentration (1% level) in presence and absence of As treatment. The effect was also significant on As uptake in As treated (1% level) and non-treated (0.1% level) soil.

The balance sheet (Table 5) shows that all the pots initially contained As ranging from 1.14 to 7.39 mg/100 plants. Some of this As is taken up by the Kalmi plants. So, the excess amount of As should remain in the after harvest soil. But the data indicates some missing value in the experimental pots and it is more in the biochar and biomass treated soils than the control soils. The missing

amount of As is maximum (almost 99.9%) in biochar treated soil and is minimum (69%) in control soil.

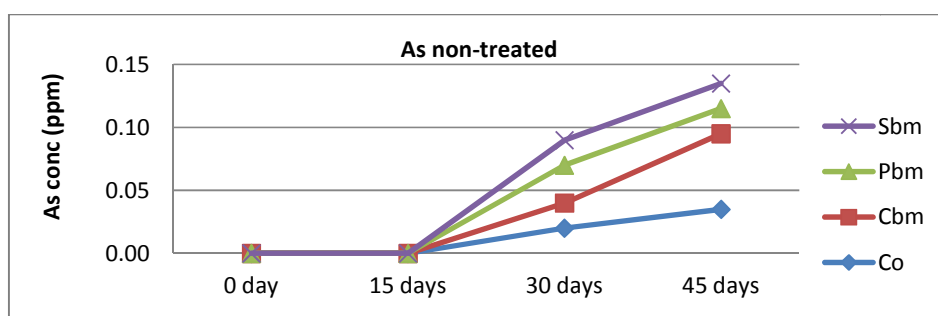
### 3.1.5 Incubation study

In absence of As treatment, As content was below detection level (BDL) for, control, biomass and biochar treatments (Figs. 6 and 7). At 15 days of incubation, As content slightly increased for biochar treatments. At 30 days, As content increased both in biochar and biomass treated soils, but the increase was higher in biochar treated soils. At 45 days, As content in all biochar treated soil decreased but fluctuated in biomass treated soil.

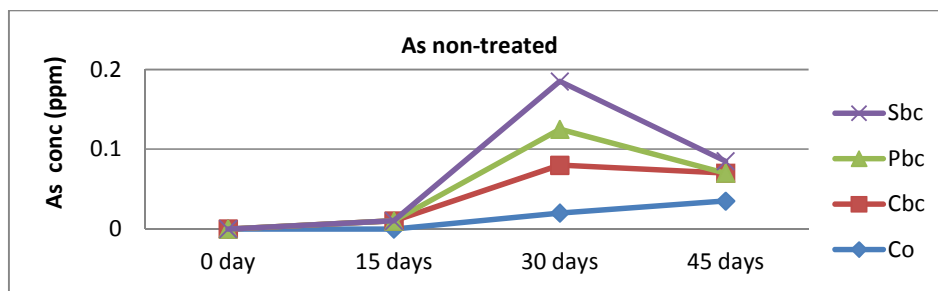


**Table 5. Balance sheet for as (g/100 plant) in different experimental pot**

As	Treatment						
	Co-As	C <sub>bm</sub> -As	P <sub>bm</sub> -As	S <sub>bm</sub> -As	C <sub>bc</sub> -As	P <sub>bc</sub> -As	S <sub>bc</sub> -As
Initial content in the soil	0	0	0	0	0	0	0
Amount added through irrigation	1.14	1.46	1.38	1.34	0.69	1.85	0.57
From biomass/biochar source	0	0.86	1.11	3.52	0.93	1.35	6.82
Total As content in the pot (a)	1.14	2.32	2.49	4.86	1.62	3.2	7.39
Removed through plant uptake (b)	0.0034	0.0042	0.0018	0.0004	0.0005	0.0042	0.0004
Content in soil after harvest b+c=d	0.35	0.17	0.23	0.13	0.003	0	0
Percent (%) not accounted	69	92	91	97	99.8	99.9	99.9



**Fig. 6. As availability in biomass treated soil at different days of incubation in absence of As treatment**



**Fig. 7. As availability in biochar treated soil at different days of incubation in absence of As treatment**

In presence of As treatment (Figs. 8 and 9), As content slightly increased from BDL for biochar treatment and for C<sub>bm</sub> at 15 days. For biomass treated soil, increase in As content continued upto 45 days of incubation except C<sub>bm</sub>. In case of biochar treated soils, As content increased at 30 days but decreased sharply at 45 days of incubation. The rate of increase and decrease was higher for S<sub>bc</sub> and poultry P<sub>bc</sub> than C<sub>bc</sub> and control.

From the data analysis of incubation it can be observed that, in absence of As treatment, As availability was higher in biochar treated soil than

their corresponding biomass treated soil (except P<sub>bm</sub> and P<sub>bc</sub>). A supportive trend was followed in case of plant As content, i.e. both plant As concentration and uptake was higher in biochar treated soil than their corresponding biomass treated ones (except C<sub>bc</sub>). In presence of As treatment, lower As availability was observed in biochar treated soils than their corresponding biomass treated soils in incubation (except P<sub>bm</sub> and P<sub>bc</sub>). Similarly, plant As concentration and uptake was also lower in biochar treated soil than the biomass treated soils (except plant As concentration in S<sub>bm</sub> and S<sub>bc</sub>) in presence of As treatment.

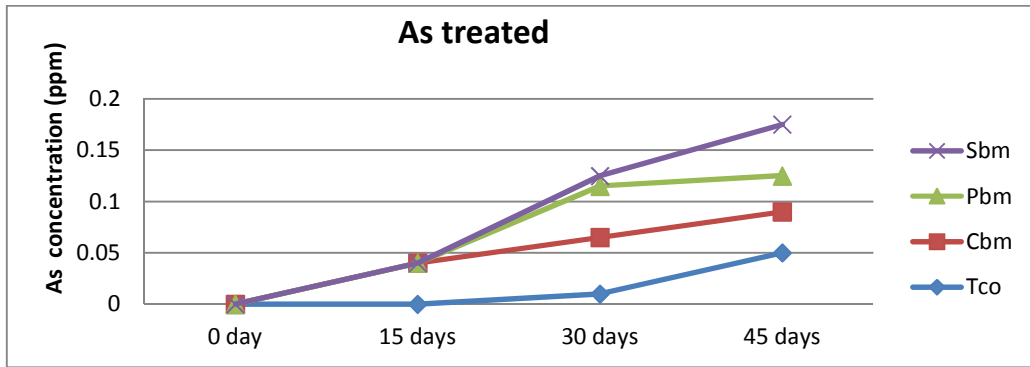


Fig. 8. As availability in biomass treated soil at different days of incubation in presence of As treatment

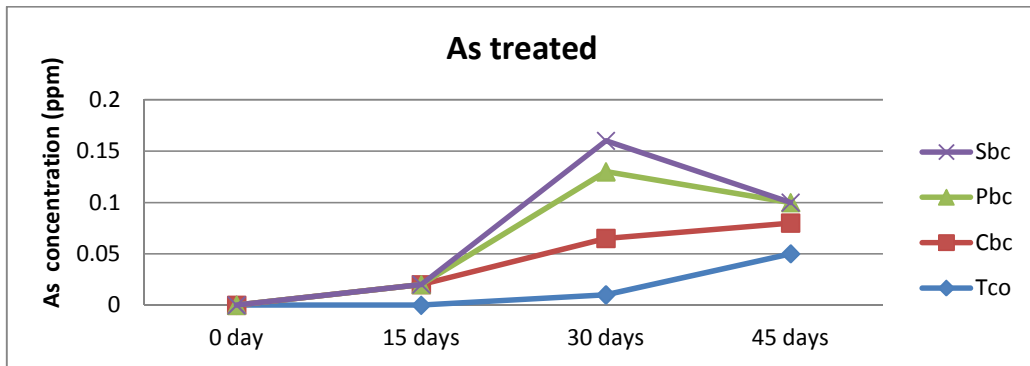


Fig. 4. As availability in biochar treated soil at different days of incubation in presence of As treatment

Table 6. As availability in soil and plant

Without as							
Incubation period	0 day	15 days	30 days	45 days	Plant As	Plant as concentration	Plant as Uptake
C <sub>bm</sub>	BDL	BDL	0.02	0.06	C <sub>bm</sub>	4.58	28.4
C <sub>bc</sub>	BDL	0.01	0.06	0.035	C <sub>bc</sub>	4.65	20
P <sub>bm</sub>	BDL	BDL	0.02	0.06	P <sub>bm</sub>	1.57	8.48
P <sub>bc</sub>	BDL	0.0	0.045	0.0	P <sub>bc</sub>	2.69	17.7
S <sub>bm</sub>	BDL	BDL	0.02	0.02	S <sub>bm</sub>	1.08	6.84
S <sub>bc</sub>	BDL	0.0	0.06	0.015	S <sub>bc</sub>	3.7	37.74
With As							
Incubation period	0 day	15 days	30 days	45 days	Plant As	Plant as Concentration	Plant as Uptake
C <sub>bm</sub>	BDL	0.04	0.055	0.04	C <sub>bm</sub>	7.99	99.2
C <sub>bc</sub>	BDL	0.02	0.055	0.03	C <sub>bc</sub>	5.18	31.96
P <sub>bm</sub>	BDL	BDL	0.05	0.035	P <sub>bm</sub>	4.6	63.05
P <sub>bc</sub>	BDL	0.0	0.065	0.02	P <sub>bc</sub>	4.02	29.35
S <sub>bm</sub>	BDL	BDL	0.01	0.05	S <sub>bm</sub>	1.24	31.07
S <sub>bc</sub>	BDL	0.0	0.03	BDL	S <sub>bc</sub>	3.92	16.54

### 3.2 Discussion

Three different biomass sources were pyrolyzed at fire wood temperature. This low temperature pyrolysis increased K, As content but reduced N, P, S contents in resulting biochar. The same biomass pyrolyzed at higher temperature (-+450°C) was diagnosed with nearly similar elemental composition except higher P content than low temperature chars [39]. Generally, biochar materials are reported to contain high concentration of P [40,41] as charring enhances P availability from biomass. The combustion causes disproportionate volatilization of C which leads to cleavage of organic P bonds and yields biochar with high soluble P salts [42]. But reduced P content in low temperature biochar may be because of the lower P solubility in charred mass compared to uncharred material due to bonding of P with multivalent metal cations in char [43,44]. P content in biochar is also a function of biomass type and pyrolysis condition [16]. Increase in K content may be due to the high temperature required for the release of K from biomass [45]. Again when heat passes through the biomass, a higher diffusional resistance is produced by the still intact inorganic matrix during devolatilization [46]. Increase in As content may be due to the concentration of non-volatile As during the loss of volatiles from feedstock during pyrolysis [47] as heavy metals like As (that are intrinsic to source material) are neither created nor destroyed [48,49]. With increasing temperature, N containing structures in biomass, i.e., amino sugars, amino acids and amines are transformed into heterocyclic N aromatic structures [50,51]. S present in the biomass form gaseous product with increasing pyrolysing temperature [19]. These may be the reasons behind the lower S and N content in biochar.

According to the surface area analysis,  $C_{bc}$  and  $S_{bc}$  having higher surface area and pore volume was observed to have lower pore diameter on the other hand,  $P_{bc}$  with lowest surface area was observed with largest pore diameter. Increased number of pores in biochar causes the thinning and collision of pore walls which develops new pores too small to be measured by surface area analysis [52]. As a consequence, significant reduction in micropore volume and an increase in macro porosity takes place. These findings were also evident in SEM image observations.  $P_{bc}$  were observed with larger, visible pores and sharp structures on the other hand,  $C_{bc}$  and  $S_{bc}$  was diagnosed with no visible pores

and clodded structures that implies higher surface area.

Biochars derived at high temperature from the same biomass were observed to have decreased surface area than those of the low temperature biochars. The high temperature biochars also possessed sharper structures [39]. Studies have shown that surface area and pores of biochar may be higher in low temperature biochar due to the longer retention time required for slow pyrolysis process [20].

CEC of biochar increased with increasing pyrolysis temperature and pH decreased in low temperature then again increased in high temperature except  $C_{bc}$ . The pyrolysis of biomass leads to the oxygenation of biochar surface [53]. This oxygenation results in the formation of O containing functional groups (e.g., carboxyl, hydroxyl, phenol groups) over the vast internal surface area of the biochar [54,55,56,29]. These functional groups give rise to a considerable negative charge and a high CEC as well as low pH. At high pyrolysis temperature, the amount of carboxyl groups in the produced biochar get reduced and/or become deprotonated to the conjugate bases resulting in more alkaline pH of biochar. On the other hand, CEC of biochars first increases and then decreases with increasing pyrolysis temperatures [30,55,57,58] with a peak CEC of up to 45  $\text{cmol}_c \text{ kg}^{-1}$  generally occurring between 250 and 350°C, depending on the source material. The lower CEC observed after higher temperature pyrolysis is concurrent with a lower O:C ratio and a decrease in the abundance of oxygenated (acid) functional groups [53,55,57,35,59]. But the higher CEC of the high temperature biochars in our study may be due to the difference in property of biomass sources [60]. Here,  $C_{bc}$  showed different behavior than other biochars i.e, alkaline pH in low temperature and reduced CEC in high temperature. Along with the dependence of pH and CEC on biomass property [61], other reasons may act behind high CEC of biochar. Firstly, the increased surface area after pyrolysis and secondly the increased charge density on biochar surface [62]. The higher CEC of  $C_{bc}$  may have a significant relation with its larger surface area than other biomass materials.

According to our plant As accumulation study, in absence of As treatment, the plant grown in biomass treated soil showed lower As accumulation than plant grown in biochar treated soil. This may be due to the lower As content in

biomass than their corresponding biochar. Again in presence of As treatment, As accumulation was lower in biochar treated plants. According to our incubation study, biochar treated soil showed reduced As availability than biomass treated soil in presence of As treatment. Balance sheet shows a higher missing percentages of As in biochar treated soils. Biochars are able to complex metal ions on their surfaces and therefore, reduce their bioavailability [63]. Metal sorption on biochar surface is generally governed by several mechanisms *i.e.*, electrostatic interaction, surface complexation with functional groups, chemical reduction and ion exchange [19].

Between, two temperature biochars, low temperature ones seemed to be better treatment in reducing As accumulation in plant [39]. The lower pH of the low temperature biochar may act as a reason behind the increased sorption of As. The pH demonstrates the H<sup>+</sup> ions of specific functional groups on the biochar surface and varies the form of As in the solution., As sorption is comparatively higher at lower pH (2-6.8) [64]. As at lower pH, biochar surface remains positively charged and gives a strong electrostatic attraction between surface groups and anion species in the solution which enhance As adsorption process. At higher pH(>6.8), adsorption gets decreased, because the OH<sup>-</sup> ions at alkaline condition can compete with As (V) anion for the active site resulting in the blocking of As adsorption on biochar surface. This is because, As(V) exist in the aqueous solution in the form of H<sub>3</sub>AsO<sub>4</sub>, H<sub>2</sub>AsO<sub>4</sub><sup>-</sup>, HAsO<sub>4</sub><sup>2-</sup>, AsO<sub>4</sub><sup>3-</sup>, and especially H<sub>2</sub>AsO<sub>4</sub><sup>-</sup> at the pH range of 2.0–6.0 [65].

Among the low temperature biochar treatments, S<sub>bc</sub> showed highest and C<sub>bc</sub> showed lowest efficacy in reducing As accumulation in plant. The More alkaline pH of C<sub>bc</sub> and more acidic pH of S<sub>bc</sub> may be the reasons behind the variation in their efficacy in reducing As accumulation. This finding relates with our SEM-EDX analysis, both in As saturated and unsaturated condition, S<sub>bc</sub> showed highest As sorption and C<sub>bc</sub> showed lowest As sorption.

Between high temperature and low temperature biochar originated from same biomass, low temperature biochars showed higher As sorption (except C<sub>bc</sub>) in saturated condition. The reason behind this may be the lower pH of low temperature biochar (except C<sub>bc</sub>). In unsaturated condition, low temperature biochar showed lower

As sorption (except S<sub>bc</sub>) [39]. This may be due to the higher As content of high temperature biochar than low temperature biochar (except S<sub>bc</sub>).

According to our incubation study, in P<sub>bc</sub> and S<sub>bc</sub> treated soil, As concentration was 0 upto 15 days of incubation which increased at 30 days then again decreased at 45 days of incubation. On the other hand, an increasing trend of As content was observed in C<sub>bc</sub> treated soil throughout the incubation period. This also evidences that, As sorption capacity of C<sub>bc</sub> is lower than S<sub>bc</sub> and P<sub>bc</sub>.

According to previous study findings, higher surface area of biochar may enable diffusion of As into the biochar pores thereby providing more metal(loid) active sites to bind metal ions on their surface [66]. But this concept contradicts with our findings, as C<sub>bc</sub> having maximum surface area showed lowest As sorption. Large BET surface area of biochar always does not guarantee good adsorption for compounds having large molecular dimension. Adsorption capacity is dependent on the size of biochars mesopores (1.7-50 nm). In a study conducted to observe the efficacy of biochar in removing herbicide, an adsorption gap was found between two biochars with similar surface area but different porosity. As a large portion of biochar surface area consists of mesopores most of which may be too small to be accessible for the nano-scale herbicide molecules. It was hypothesized from this study that the mesopores of biochars may have played critical role on adsorption of herbicides [67]. According to our findings, C<sub>bc</sub> contained the lower pore diameter than S<sub>bc</sub> and P<sub>bc</sub>. This may be another reason behind the higher adsorption capacity of S<sub>bc</sub> and P<sub>bc</sub>. than C<sub>bc</sub>.

#### 4. CONCLUSION

It could be concluded that among low temperature biochar materials, S<sub>bc</sub> and P<sub>bc</sub> showed positive impact in reducing As accumulation in plant. This study also suggested that, low pyrolysis temperature and the different source materials regulate the properties of biochar as well as the effects of biochar in remediating As contamination. However, more research is required in future to fully identify the As-biochar interaction in soil along with the behavior of biochar developed from slow pyrolysis condition and various feedstock materials to develop more reliable and

sustainable biochar for application on an industrial scale.

## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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