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RESEARCH ARTICLE

ARSENIC MITIGATION APPROACH IN SOIL BY SOME INDIGENOUS SOURCES OF BIOCHAR OF DIFFERENT PYROLYSIS TEMPERATURE

[1]Priyanka Dey Suchi, [2]Nishat Ferdousi, [2]Nadia Noor, [3]Imamul Huq, S. M., [1]Badhan Saha and [1,] *Mohammad Moniruzzaman

[1]Soil and environment section, BCSIR Laboratories, Dhaka, Bangladesh Council of Scientific and Industrial Research, Bangladesh
[2]Department of soil, water and environment, University of Dhaka, Bangladesh
[3]University of Barisal, Bangladesh

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ABSTRACT

Water-soluble inorganic pollutants may constitute an environmental toxicity problem if their movement through soils and potential transfer to plants or groundwater is not arrested. Biochar has recently been used to sequester carbon and remediate soil with both heavy metal and organic pollutants. The characteristics of biochar are influenced mainly by the preparation temperature and biomass sources. Biochars were produced from three different sources (viz., cow dung, poultry manure, and sewage sludge) at two different temperatures (low temperature - +250ºC and high temperature - +450º C). Different physical (e.g. surface area, SEM, EDX), chemical (CEC, organic carbon, N, P, K, S, As) and physicochemical (pH) properties of the prepared biochars were measured. Two sets of experiment (Arsenic treated and non-treated soils) were done. The capability of biochars produced at different temperature to abate arsenic (As) accumulation in plants (*Ipomoea aquatica*) was carried out in pot culture experiment. Biochars were applied at 5 t/ha to soil spiked with 1 mg/L of As solution (80% arsenite + 20% arsenate). Plants were grown for 45 days after germination. Incubation study was also done to see the sorption of water soluble arsenic (As) by biochars with time. To alleviate arsenic accumulation in plant the efficacy of biochar was in order of $S_{bch}>P_{bcl}>C_{bcl}$ for arsenic non-treated soil and in arsenic treated soil the order was in $S_{bch}>P_{bch}>C_{bcl}$.

INTRODUCTION

Arsenic poisoning can be minimized through biological immobilization and stabilization using a range of organic compounds, such as biochar, which is a form of environmental black carbon, produced by using the pyrolysis of C-based biomass (Verheijen *et al.,* 2010). Application of biochar to soil has been considered as to having great potential to enhance long-term carbon sequestration because most carbon in biochar has an aromatic structure and is very recalcitrant in the environment (Lehman, 2007). Typically biochar has a high pH value and cation exchange capacity, and can enhance soil productivity (Jeffery *et al.,* 2011; Kookana *et al.,* 2011). A number of studies have also demonstrated that biochar has a high capacity to absorb pollutants in soils (Beesley*et al.,* 2011; Yuan and Xu, 2011). However the effect of biochar on arsenic (As) mitigation is not revealed yet. It becomes relevant to find out its effect on As alleviation and pyrolysis temperature of biochar preparation modify its characteristics. Antal and Gronli (2003) and Uchimiya*et al.* (2011) found that pyrolysis temperature can affect the presence of surface functional groups on biochars and thus control their heavy metal sequestration ability in soils. Recent studies have also suggested that conversion/production methods can also play an important role in controlling biochar properties (Libra *et al.,* 2011). The biochar derived from relatively low-temperature pyrolysis is characterized by a high content of volatile matter that contains easily decomposable substrates, which can support plant growth (Robertson *et al.,* 2012; Mukherjee and Zimmerman, 2013). In contrast, the structure of biochar derived from high temperature pyrolysis is characterized by a large surface area and aromatic-carbon content, which may increase the adsorption capacity (a desirable property for bioremediation) as well as the recalcitrant character (for carbon sequestration) (Lehmann, 2007). The type of feedstock material is another important factor that determines the final application of the biochar and its effect on soil, because its properties are affected by the nature of the original material (Lehman *et al.,* 2006). Decrease of atomic ratios H/C and O/C resulted from removing H- and O-containing functional groups with increasing temperature will produce high aromaticity and low polarity biochars (Ahmad *et al.,* 2013)**.** The diverse range of biochar applications depends on its different properties, which are governed by the pyrolysis conditions (heating temperature and duration) and the original feedstock (Enders *et*

***Corresponding author:* Mohammad Moniruzzaman**
Soil and environment section, BCSIR Laboratories, Dhaka, Bangladesh Council of Scientific and Industrial Research, Bangladesh

al., 2012). The objective of this work was to find out the difference in various characteristics of biochar produced from three different sources of biomass at two different temperatures (high and low) and their impact on the phytoavailability of arsenic (As) in soil.

MATERIALS AND METHODS

Biochar and soil sample Preparation

Biochar was produced from biomass sources both at low (+250ºC) and high temperature (+450ºC) pyrolysis process. The low temperature pyrolysis process was carried out following the method as described in Mahmud *et al.* (2014). The high temperature pyrolysis was carried out in an aluminum pot containing the biomass and covered with an aluminum lid and was heated by gas burner. In both cases, oxygen was not allowed to enter into the pot. A portion of produced biochar samples were then sieved through a 0.25 mm sieve for various chemical and physicochemical analyses. Soil sample was collected from the agricultural field of Manikgonj Sadar Upazila in Manikganj district (23°51.884' N and 90°06.219' E), Bangladesh. It is young Brahmaputra floodplain soil belonging to the Melandaha series. According to the USDA soil taxonomy the soil is a typicendoaquepts and according to the FAO-UNESCO legend it is a Gleysol. The bulk of the soil samples (0-15 cm) were collected by composite soil sampling method (USDA, 1951) and processed (Huq and Didar, 2005).

Pot culture experiment

Kalmi plant (*Ipomoea aquatica*) was used as a study plant in pot culture experiment using the following 7 treatments including control arranged in a completely randomized design (Table 3). Urea, Muriate of potash, TSP (Triple Super Phosphate) and Gypsum ($CaSO_4$) fertilizer were applied at the rate of 0.12 t/ha, 0.05 t/ha, 0.04 t/ha and 0.008 t/ha respectively to the soil. The application rate of biochar was 5 t/ha in the soil. After the germination of the Kalmi seeds, arsenic dose (80% arsenite + 20% arsenate) at a rate of 1mg/Kg was applied through irrigation water. The experiment was carried out for arsenic treated and non-treated soil.

Incubation Study

An *in vitro* incubation study was also conducted to see the sorption and desorption of arsenic at field condition for 0, 15, 30 and 45 days of incubation period and the experimental setup was similar to that of the pot culture experiment.

Background Analysis

Various properties i.e. pH, CEC (Cation Exchange Capacity), O.C. (Organic Carbon), Nutrient Status (N,P,K,S) of soil and biochar were analyzed according to Huq and Alam (2005). Arsenic analysis was accomplished according to the method described in Mahmud *et al.,* 2014.

Physical Properties

a. **Surface properties:** The surface area, total pore volume, mean pore diameter of biochar was measured by BET plot method (Table 2).

b. **Morphology:** Surface morphology of biochar was measured by SEM (Scanning Electron Microscope) and elemental content (Carbon and Arsenic) by EDX (Energy Dispersive X-ray Spectroscopy)

RESULTS AND DISCUSSION

Basic soil characteristics (Table 1) such as pH, CEC, O.C., texture, Arsenic, available N, P, Kand Swere analyzed. Basic biochar characteristics (Table 1) were analyzed to see the effect of two pyrolytic temperature on the properties of different sources. Higher temperature increased the CEC of biochar except for cowdung. pH, carbon content, phosphorus and potassium content enhanced with the increasing temperature. Biochars produced at higher pyrolysis temperatures have higher CEC, higher pH (Bagreev*et al.,* 2001; Novak *et al.,* 2009a). Nitrogen content (38.27%) was higher in cow dung biochar produced at high temperature and it was lower for other two sources of biochar compared to low temperature biochars. The content of nitrogen decreased with the increasing temperature (Lei and Zhang, 2013).Arsenic content was highest in S_{bcl} (6.82 ppm) and lowest in C_{bch} (1.22 ppm). Higher temperature increased the arsenic content of biochar than lower temperature.

Surface properties: The original biomass structure strongly influences the final biochar structure and its eventual physical characteristics. Most typically pyrolysis reactions lead to a number of structural and physical changes. The most commonly observed feature is pore structure. High pyrolysis temperature leads to greater specific surface area and aromaticity of biochar than low temperature does (Ahmad *et al.,* 2012). The surface area of biochar (Table 2) was measured by BET plot method. C_{bcl}(12.335 $m^2 g^{-1}$) and S_{bcl} (6.4659 $m^2 g^{-1}$) possess higher surface area than biochar produced at high temperature. For P_{bc} (3.6042 m^2g^{-1}) surface area was higher at high temperature. Total pore volume was highest C_{bcl} (3.8888 cm^3g^1) and lowest for C_{bch} (1.5490 cm^3g^1). Mean pore diameter was higher for high temperature biochar.

SEM (Scanning Electron Microscope): Scanning electron microscopy (SEM) is a microscopic technique to determine image for macroporosity and physical morphology of solid substances (Angin, 2013). The figure 1 shows the micrographs (X5000) of C_{bch}, C_{bcl}; P_{bch}, P_{bcl}; S_{bch}, S_{bcl}. Surface morphology was well defined for high temperature biochar. Sharp edges and plate like structure were observed in biochar produced at high temperature. Low temperature biochar were found mostly in cloded state.

EDX: EDX spectrumrepresents the content of carbon and arsenic content in saturated and unsaturated condition. Different sources of biochar were saturated with arsenic solution (1ppm) for 15 days to see the sorption capacity of arsenic for two temperatures.

Saturated condition: In saturated condition (fig. 2), sorption of arsenic was higher in C_{bch} (As=11.2 and C= 88.2 in weight %,) and it was lower in P_{bch} (As=2.9 and C= 97.1 in weight %) and sorption capacity was almost same for S_{bch}(As= 9.2, C= 90.8 in weight %) and S_{bcl}(As=9.5, C= 90.5 in weight %).

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

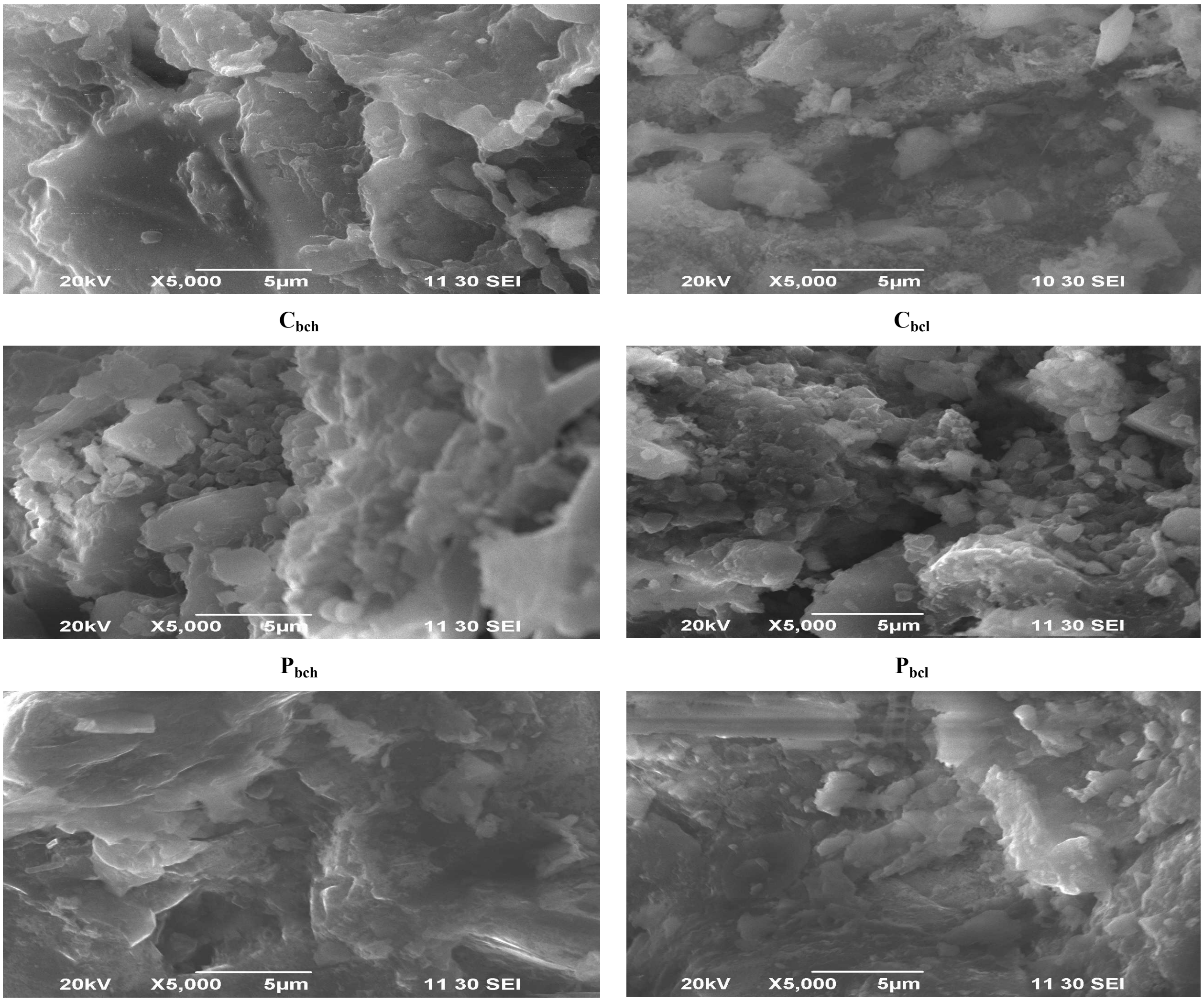
Properties	Soil	C_{bcl}	P_{bcl}	S_{bcl}	C_{bch}	P_{bch}	S_{bch}
pH	6.61	8.13	7.64	6.46	8.13	8.26	7.09
CEC(me/100g)	0.23	39.8	16.9	6.35	26.43	18.7	15.5
Textural Class	Silt loam	_	_	_	_	_	_
Moisture content (%)	21.54	_	_	_	_	_	_
Organic carbon (%)	1.3	92	94	89	94	96	92
N (%)	0.097	8.47	41.49	18.59	38.27	17.04	12.26
P (ppm)	3.23	1.08	0.71	0.21	2.36	1.92	0.68
K (me/100g)	0.11	1.48	1.48	2.69	39.96	33.52	47.18
S (ppm)	10.17	11.4	15.0	81.07	1.35	0.76	5.62
Arsenic (ppm)	BDL*	0.93	1.35	6.82	1.22	1.61	1.90

C_{bcl}, P_{bcl}, S_{bcl}= cow dung, poultry manure and sewage sludge biochar respectively produced at low temperature; C_{bh}, P_{bh}, S_{bh}= cow dung, poultry manure and sewage sludge biochar respectively produced at high temperature

Table 2. Surface Area, pore volume and mean pore diameter of Biochar produced at low and high temperature

Sample	Surface Area (m^2g^{-1})	Total Pore Volume (cm^3g^{-1})	Mean Pore Diameter (nm)
C_{bcl}	12.355	3.8888	12.590
P_{bcl}	2.8330	2.5350	35.792
S_{bcl}	6.4659	4.5399	28.085
C_{bch}	1.7151	1.5490	36.127
P_{bch}	3.6042	3.1211	34.638
S_{bch}	5.1371	4.5124	62.596

C_{bcl}, P_{bcl}, S_{bcl}= cow dung, poultry manure and sewage sludge biochar respectively produced at low temperature; C_{bh}, P_{bh}, S_{bh}= cow dung, poultry manure and sewage sludge biochar respectively produced at high temperature

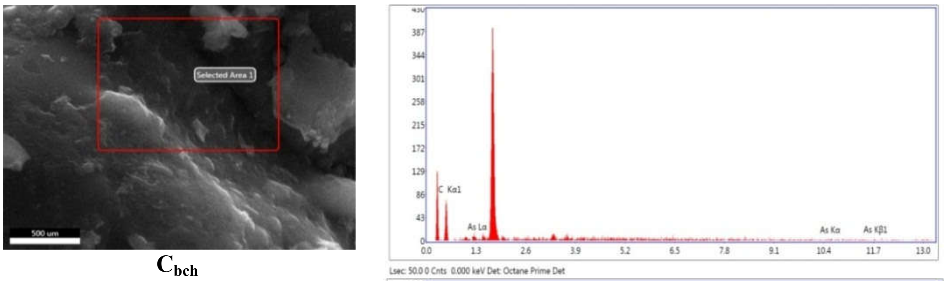


C_{bch} C_{bcl}

P_{bch} P_{bcl}

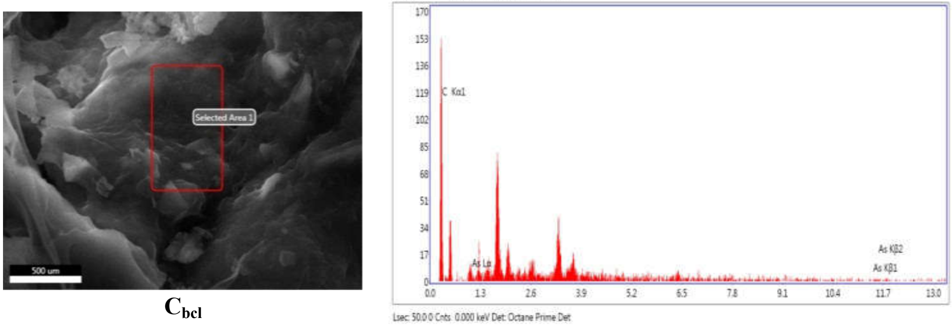
S_{bch} S_{bcl}

Figure 1. Photographic images of biochars from SEM



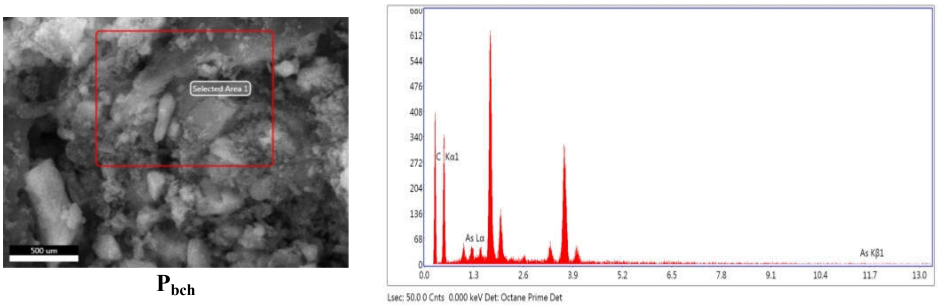
C_{bch}

%	C	As
weight	88.2	11.2
Atomic	98.0	2



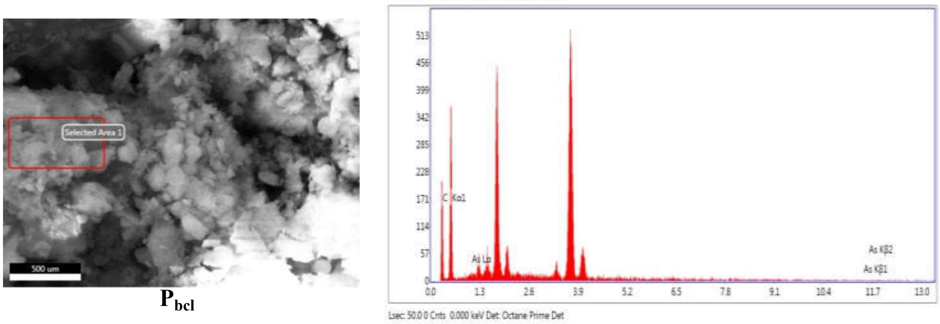
C_{bcl}

%	C	As
weight	96.4	3.6
Atomic	99.4	0.6



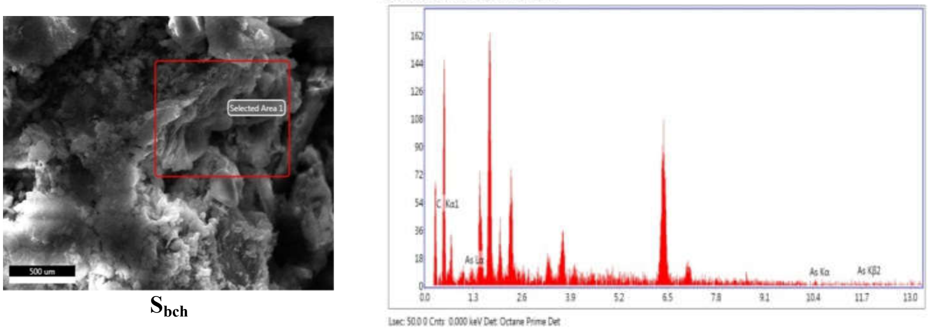
P_{bch}

%	C	As
weight	97.1	2.9
Atomic	99.5	0.5



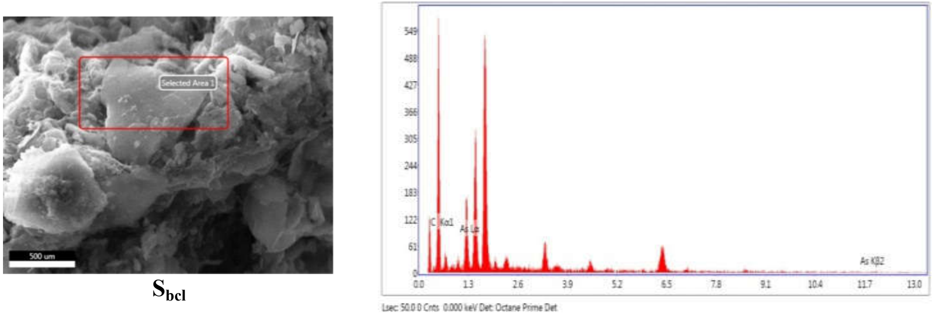
P_{bcl}

%	C	As
weight	94.3	5.7
Atomic	99	1



S_{bch}

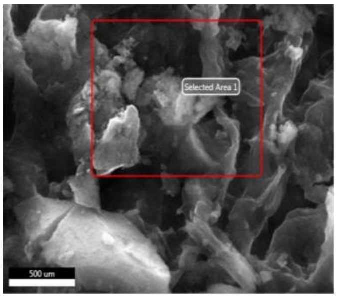
%	C	As
weight	90.8	9.2
Atomic	98.4	1.6



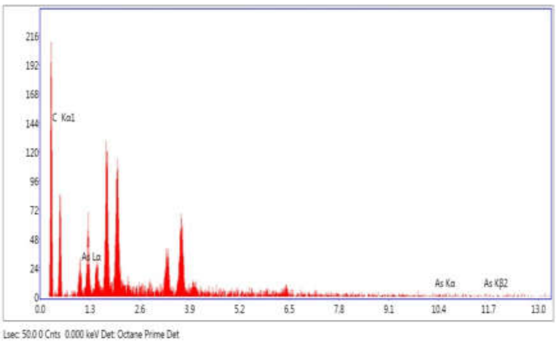
S_{bcl}

%	C	As
weight	90.5	9.5
Atomic	98.3	1.7

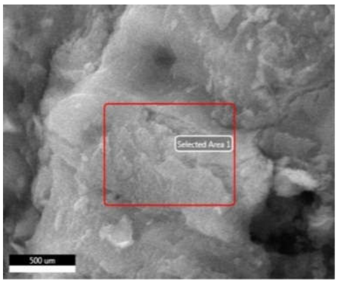
Figure 2. Photographic images and content (C, As) of biochars in saturated condition from EDX



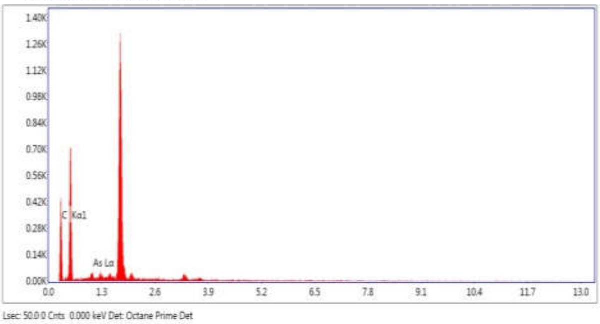
C_{bch}



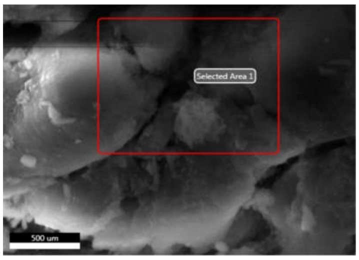
%	C	As
weight	95.9	4.1
Atomic	99.3	0.7



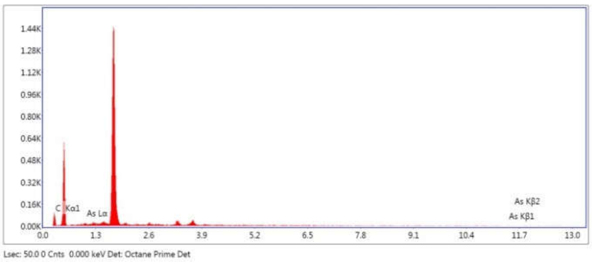
C_{bcl}



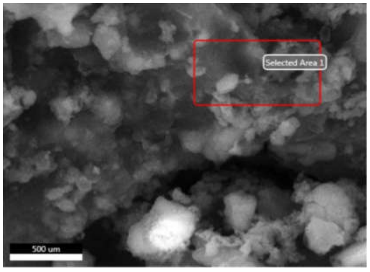
%	C	As
weight	99	1
Atomic	99.8	0.2



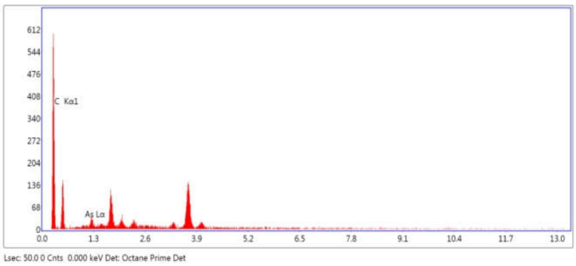
P_{bch}



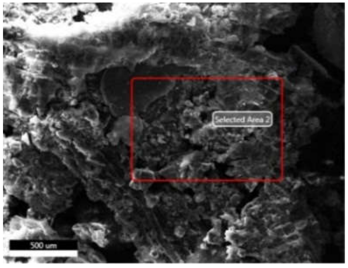
%	C	As
weight	90.6	9.4
Atomic	98.4	1.6



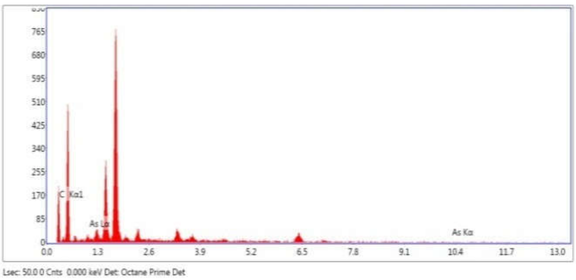
P_{bcl}



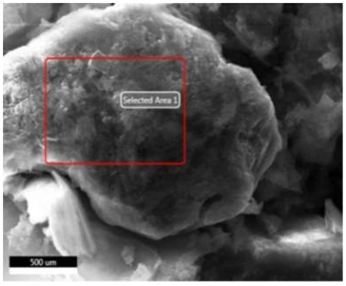
%	C	As
weight	98.2	1.8
Atomic	99.7	0.3



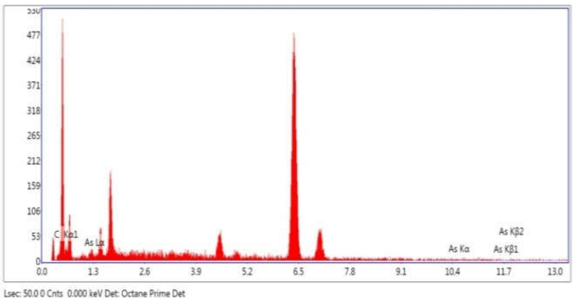
S_{bch}



%	C	As
weight	96.9	3.1
Atomic	99.5	0.5



S_{bcl}



%	C	As
weight	82.5	17.5
Atomic	96.7	3.3

Figure 3. Photographic images and content (C, As) of biocharsin unsaturated condition from SEM

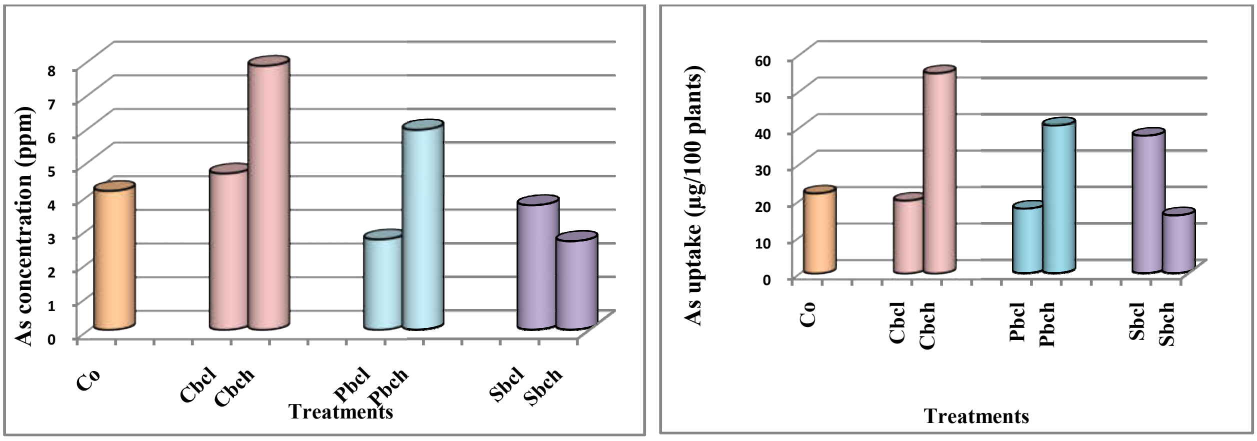


Figure 4. Arsenic accumulation in Kalmi plant for As non-treated soil

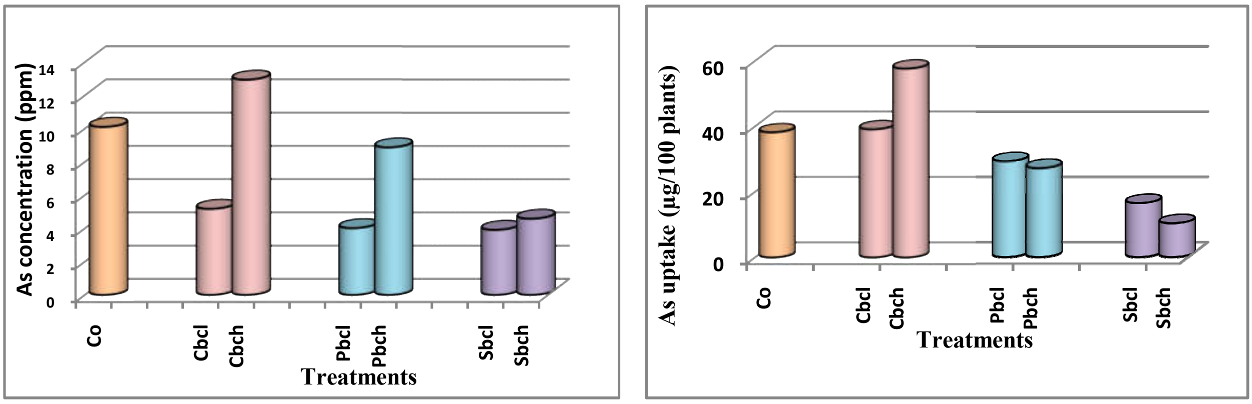
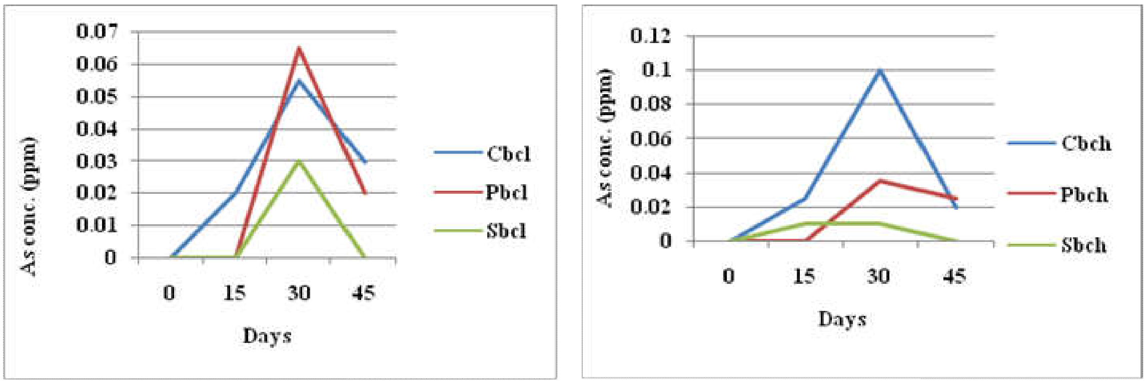


Figure 5. Arsenic accumulation in Kalmi plant for Astreated soil

Arsenic non-treated soil



Arsenic treated soil

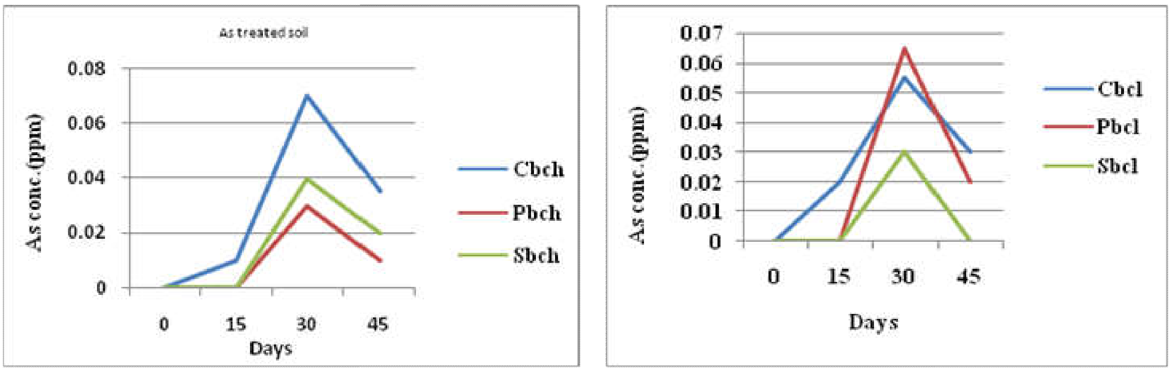


Figure 6. As availability in non- treated and treated soil at different incubation days

Unsaturated Condition: In unsaturated condition S_{bcl} (17.5 in % weight) had the highest arsenic content and lowest carbon content (82.5 in % weight) among all the biochars (Fig. 3). For other two sources of biochar i.e. poultry manure (9.4 in % weight) and cow dung (4.1 in %weight), arsenic content was higher for high temperature than low temperature biochar.

Plant Growth

Fresh and dry matter production: Different biochars have different effect on the growth of kalmi plant (Table 3). In arsenic non-treated soil, high temperature biochars were more effective on the growth of plant than low temperature biocharexcept P_{bch}whereas in arsenic treated soil, low temperature biochars were more effective. Among all the biochar treatments S_{bch}wassupposed to be the best treatment for As non-treated soil and in As treated soil, C_{bcl} appeared to be the best treatment. Blackwell *et al.* (2009)and Lehman *et al.* (2003) mentioned that biochar can be used as an amendment to improve soil quality and crop production in a variety of soils.

Table 3. Fresh and dry weight production of Kalmi plant (g/100 Plants)

Treatments	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_{bcl}	80.67	4.3	112.2	7.56
P_{bcl}	111.53	6.82	96.0	7.3
S_{bcl}	99.3	6.03	103.1	6.50
C_{bch}	99.5	7.3	107.23	7.22
P_{bch}	96.78	5.53	95.87	5.93
S_{bch}	113.31	10.2	99.2	5.22

C_{bcl}, P_{bcl}, S_{bcl}= cow dung, poultry manure and sewage sludge biochar respectively produced at low temperature; C_{bh}, P_{bh}, S_{bh}= cow dung, poultry manure and sewage sludge biochar respectively produced at high temperature

ANOVA was done and found that for arsenic non-treated soil, the effect of low temperature biochars was not significant in the production of Kalmi but the treatments (high temperature biochar) effect was significant on fresh weight (P=0.018) production and non-significant for dry weight production. In arsenic treated soil, the effect of high temperature biochars was not significant on the growth of plant whereas the treatments (low temperature biochar) effect was significant on dry weight (P=0.003) production and non-significant for fresh weight (P=0.545) production.

Accumulation of arsenic in plant (As non-treated soil): Between two temperature (low and high) biochars, low temperature biochar seemed to be the better treatment to diminish the arsenic concentration in plant except sewage sludge (fig. 4). Among high temperature biochars, sewage sludge was the best treatment to decrease the concentration of arsenic in plant.

Accumulation of arsenic in plant (As treated soil): The high temperature biochar increased the concentration of arsenic (fig. 5) but it decreased the uptake of arsenic in plant except for cow dung biochar. Though low temperature biochars decreased the concentration of arsenic plant it increased the uptake of arsenic except cow dung biochar.

Incubation Study: Soil was incubated for 1 and half month to observe the release pattern of arsenic with increasing time. The concentration of arsenic recorded at 0,15,30 and 45 days.

Arsenic non-treated and arsenic treated soil: Arsenic concentration was BDL (below detection limit) from 0 to 15 days in poultry manure and sewage sludge biochar treated soil and lower concentration of arsenic in cow dung biochar treated soil. From 15-30 days the availability of arsenic increased and the concentration was at peak at 30 days. The release of arsenic started to decrease when goes from 30 to 45 days. In arsenic non-treated soil S_{bch} and in treated soil P_{bch} and S_{bcl} showed the lowest availability. Namgay*et al.* (2010) reported that the concentrations of extractable As increased with biochar application rate. The increasing and decreasing arsenic content of plants in biochar treated soil could be due to several reasons. From the background analysis it was observed that the concentration of As in high temperature biochar materials was higher than low temperature biochar except sewage sludge. The surface area (table 2) of biochar may have also played a vital role in the release of As in soil. The feedstock material determines the peak temperatures at which the micropores are opened up within the biochar(Uchimiya*et al.,* 2010). This was reported by James *et al.*2005, where heating at 820 °C resulted in reduction of micropores and surface area of wood when compared to that at 700 °C. This indicates that there may be a peak temperature to open all pores (micro-, meso-, macro-) for each feedstock material; exceeding such temperatures may further encourage reduction in distribution of micropores. In As non-treated soil, cow dung and poultry manure biochar produced from low temperature decreased the accumulation but sewage sludge increased the accumulation in plant.

On the other hand, sewage sludge produced from high temperature abated the accumulation. In case of As treated soil, high temperature biochar increased the concentration of As in plant but except cow dung biochar others reduced the uptake in plant. It could be possible that biochar trapped arsenic on its surface. As pyrolysis temperatures increase, volatile compounds in the biochar matrix are lost, surface area and ash increase, but surface functional groups that can provide exchange capacity decrease (Guo and Rockstraw, 2007). The pH condition could be another factor for the increasing arsenic content in the plants.High temperature biochar possess high pH than low temperature and also biomass (Table 1). Chan and Xu (2009)in their work showed that the increased pH in the biochar treated soil increased arsenic (As) solubility by creating an alkaline condition. Biochars have a pH greater than 8 (Yin and Xu, 2009) and it might increase the solubility of arsenic in soil by creating an alkaline condition. O'Neill (1990); Fitz and Wenzel (2002) also reported an increase in arsenic solubility with more alkaline pH at soil pH> 7. Beesley and Marmiroli (2011) did a soil elute test , during the experiment they have found that the solubility of arsenic (As) increased slightly when the soil eluate passed through the biochar but that concentrations were very low from the biochar alone . Indeed they have found no significant correlation between pH of the eluate and As concentrations in the eluate.The CEC of biochar could be another reason to retain As (Table 1). The higher pyrolysis temperature decreased CEC of cowdungbiochar. Recent literature has shown that natural long-term oxidation of biochar in the soil increases the amount of negative charges on the biochar surface (Cheng *et al.,* 2008). The study suggests that sewage sludge biochar produced from high temperature is suitable for arsenic mitigation in soil and among low temperature biochars, poultry manure and cow dung biochar both are effective. This study also suggests that source and pyrolysis temperature are the important factor to determine its efficacy. So, the effect of biochar on mitigating arsenic highly depends on the sources and pyrolysis temperature as these two factors determine its behavior.

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