Influence of biochar addition on gas permeability in unsaturated soil

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Biochar (BC) has been proposed as an alternative soil amendment in landfill cover. Its effects on soil hydraulic properties (e.g. soil water retention) have been explored to some extent, while its influence on landfill gas flow characteristics is not clear yet. The objective of this study is to investigate the water retention and gas permeability (k_g) of BC-amended soil and its relationship with soil water content in unsaturated state. In total, three BC contents (0, 5 and 10%) were considered in clay–sand mixtures. Soil suction, water content and gas pressure were measured continuously using a new in-house developed apparatus. Test results show that the presence of BC increased soil water content and the increased amount was almost the same over a wide range of suction values (up to 3 MPa). k_g increased linearly with soil suction in semi-log scale, regardless of the BC content. The addition of 5 and 10% of BC could decrease k_g by up to 50 and 65%, respectively, compared with that of the bare soil. k_g decreased with water content for both soil and BC -amended soils. Moreover, the decrease rate of k_g with water content became smaller when the BC percentage was higher.

KEYWORDS: partial saturation; permeability; suction

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NOTATION

A	soil cross-sectional area
i	hydraulic gradient
k_g	gas permeability
L	length of soil sample
q	flow rate
γ	specific weight of flow medium
Δh	head difference
Δp	pressure difference
θ	soil water content
ψ	soil suction

INTRODUCTION

Biochar (BC) is produced from plant-based biomass under high temperature with little or no available oxygen. It has been applied as soil amendment because it can increase soil fertility and carbon (C) storage (Lehmann & Joseph, 2009). Recently, BC has been proposed as a promising final landfill cover material, as it can promote soil physical and hydraulic properties, such as soil bulk density, soil water retention (SWR), water permeability and so on (Wong *et al.*, 2015; Ni *et al.*, 2018; Pardo *et al.*, 2018). By adding BC, SWR ability increased (Lei & Zhang, 2013; Ni *et al.*, 2018) and water infiltration/permeability reduced in sandy soils (Blanco-Canqui, 2017; Gopal *et al.*, 2019). However, BC may have

no effect on soil water capacity (Ouyang *et al.*, 2013; Jeffery *et al.*, 2015) and increase water infiltration/permeability in clay soils (Blanco-Canqui, 2017).

In addition to soil hydraulic properties, gas emission must be considered for final landfill covers. BC application can reduce methane emission from landfills by enhancing methane oxidation (Reddy *et al.*, 2014). It could also absorb odorous landfill gas, such as hydrogen sulfide (Xu *et al.*, 2014). However, those processes are highly dependent on gas permeability of BC -amended soils.

BC application may increase the gas permeability of agricultural sandy loam due to high porous structure (Sun *et al.*, 2013). Unlike agricultural soil, which has low bulk density for crop growth, soil in landfill final cover is highly compacted at around 90% degree of compaction (DOC). Compacting at high DOC could minimise water infiltration and reduce slope instability (GCO, 2000). Soil pore structure changes when subjected to compaction (Bronick & Lal, 2005). Moreover, the organic particles of BC enhance the particle bonding between large particles and promote soil aggregation especially in coarse-grained soils (Liu *et al.*, 2012). All the aforementioned changes in soil structure would affect soil pore-size distributions and hence possible changes in gas migrations in soil (Deepagoda *et al.*, 2011). Therefore, it is necessary to understand the gas flow mechanisms throughout the unsaturated range in soils with BC amendment when considering its use as landfill cover materials.

This study aims to investigate the gas permeability of BC-amended silty sand in unsaturated state. The gas permeability (k_g) was determined using an in-house-developed apparatus, with soil suction (ψ) and soil water content (θ) measured continuously. The microstructure of BC and surface functional groups were analysed to facilitate the interpretation of measured SWR and k_g results. BC was produced from water hyacinth (WH) – world's most invasive weed species (Malik *et al.*, 2007), which requires constant management for removal. This conversion of waste ligno-cellulose material to BC further helps in the carbon sequestration process aiding the environment (Mašek, 2013).

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Table 1. Basic and index properties of soil and BC

Soil properties	Standard	Soil	BC
Particle-size distribution	ASTM D 422 (ASTM, 2007a)		
Coarse sand (2–4.75 mm)		23.11	0.00
Medium sand (0.425–2 mm)		37.28	000
Fine sand (0.075–0.425 mm)		20.84	30.48
Silt (<0.075 mm)		17	68.52
Clay (<0.002 mm)		1.77	1
Atterberg limits	ASTM D 4318 (ASTM, 2017)		
Liquid limit (LL)		42.4	ND
Plastic limit (PL)		26.0	ND
Plastic index (PI)		16.4	ND
MDD: kN/m ³	ASTM D 698 (ASTM, 2007b)	15.6	ND
OMC: %	ASTM D 698 (ASTM, 2007b)	16.5	ND
Specific gravity	ASTM D 854 (ASTM, 2014)	2.46	0.8
pH	ASTM D 4972 (ASTM, 2018)	6.7	9.25

Note: ND, not determined.

TEST PLAN, SET-UP AND INSTRUMENTATION

Three test series (bare soil – BS; BC-amended soil at 5% and 10% – BAS5% and BAS10%) were conducted by compacting soil where ψ , θ and k_g were simultaneously measured. The soil is categorised as SC (sand clay mixture) according to the Unified Soil Classification System (USCS; ASTM, 2010) and its geotechnical properties are tabulated in Table 1. The WH feedstock (Fig. 1) was taken from the same water body to minimise genetic variations and dried stems were fragmented to equal length (5 cm) for ease of pyrolysis. The pyrolysis temperature and conditions (Table 2) were determined by conducting thermo-gravimetric analysis (TGA) on the dried feedstock. The largest weight loss was observed at around 350–400°C. The in-house produced BC properties are shown in Table 2.

Figure 2 shows the diagrammatic representation of the gas permeability column set-up. The cylindrical column (20 cm in diameter and 25 cm in height) consisted of two chambers. The lower chamber (5 cm in height) measured the gas discharge and corresponding pressure, and the upper chamber was filled with the compacted soil specimen. The upper chamber was provided with a porous base plate with mesh size of 75 μ m pores, over which a linen membrane was attached. The set-up was connected with flow meter and digital pressure sensor to measure the gas flow rate and pressure, respectively. The range and sensitivity of the flow meter and pressure gauge are given in Fig. 2. The compaction state was chosen as per 0.8 maximum dry density (MDD) and corresponding water content along the compaction curve for each individual soil type (Fig. 3). The wet side of optimum moisture content (OMC) was selected for ease of compaction and static compaction was applied after mixing as per Bordoloi *et al.* (2018a). The sensors (ψ by MPS-6) and (θ by EC-5) from Meter/Decagon group (DDI, 2016) were installed at the soil surface. MPS-6 is used as it can measure suction from near-field capacity (10 kPa) to past wilting point (3000 kPa).

According to Darcy's law, flow velocity is expressed as

$$v = \frac{q}{A} = k_g i \quad (1)$$

where k_g is the gas permeability, i is the hydraulic gradient, q is the flow rate and A is the soil cross-sectional area. $i = \Delta h/L$, where Δh is the head difference and L is the length of the soil. Therefore

$$\Delta q = k_g \frac{\Delta p}{\gamma L} A \text{ or } k_g = \frac{\Delta q \gamma}{\Delta p A} L \quad (2)$$

where Δp is the pressure difference and γ is the specific weight of the flow medium.

Darcy's law is used to calculate the advective flow velocity for any fluid (gas or liquid) in response to a pressure gradient (Hubbert, 1957). For the permeability experiments, the applied pressure was only slightly higher than the atmospheric pressure. Thus, gas dissolution can be reasonably ignored when the gas passed through the sample. The set-up was initially calibrated to take account of the resistance to gas flow due to the porous base plate. The gas selected was compressed carbon dioxide. The soil sample was initially ponded until the suction value reached about 10 kPa, after which the sample was dried in a temperature-controlled room (24°C and 50% RH). The drying was done until each sample reached post-wilting point of 3000 kPa. The k_g was measured after every 7–10 d, depending on the BC content.

RESULTS AND DISCUSSION

Soil compaction curves

Figure 3 shows the compaction curves for soils with BC addition. The MDD $\rho_{\max}$ and corresponding optimum water content w_{opt} for the bare soil were 1715 kg/m³ and 18%, respectively. By adding 5% BC, $\rho_{\max}$ decreased by 10%, while w_{opt} increased by 11%. Further increase of BC addition from 5 to 10% had minor effects on $\rho_{\max}$ and w_{opt} (<2%). Ni *et al.* (2018) observed that 10% of BC addition decreased $\rho_{\max}$ by only 8%, while w_{opt} increased significantly by 42%. The decreased MDD was mainly due to the lower specific gravity of BC particles (Liu *et al.*, 2016) and the smaller compressibility of the amended soils at a given compaction energy (Reddy *et al.*, 2014). The higher optimum water content could be caused by the highly porous structure (Fig. 1(b)) and larger surface area of BC particles (Guo *et al.*, 2014).

SWR curves

Figure 4 shows the water-retention curves (WRCs) for BS, BAS5% and BAS10%. Volumetric water content (VWC) decreased with suction, regardless of the BC content. At small suction range (<100 kPa), the retained water content increased by 6.5 and 10.5% for BAS5% and BAS10%, respectively, as compared with bare soil. The increased magnitude is much larger than the results shown by Ni *et al.* (2018), who found that 10% peanut shell BC addition could only increase retained VWC by 6%. After it dried up to 3 MPa, VWCs in BAS5% and BAS10% increased by the

Fig. 1. Schematics of (a) conversion of weed to BC, (b) WH BC morphology at various magnifications and (c) Fourier transformation infrared spectroscopy of WH BC

same magnitude with respect to the bare soil. The WRCs of both bare soil and BAS had the same shapes and were almost parallel throughout the suction regime. This indicates that BC could consistently increase SWR abilities over a wide range of suction values (up to 3 MPa). This can be attributed to the intra-pores within the BC (Fig. 1(b)) and occupancy of inter-pore space by BC particles. Moreover, the clay particles could also fill the BC intra-pores and hence further decrease the diameter of pore throats of BC (Wong *et al.*, 2015). For (DOC > 80%), it is reported that the

retained water in BC-amended soil could increase by more than 40%, due to the embedded effects of clay particles in BC pores (Wong *et al.*, 2015).

Furthermore, as per the Fourier transformation infrared spectroscopy (FTIR), the three major surface functional groups (i.e. -OH, -COOH, -CO) shown in Fig. 1(c) are hydrophilic. When the contact angle of water is <90°, the BC surface is hydrophilic. Those hydrophilic micropores (0.2–30 µm) in BC can improve water retention of sandy soils (Hardie *et al.*, 2014). On the contrary, when BC is

Table 2. Production conditions, elemental composition and other chemical properties

Feedstock	WH stem sourced from deep or lake, India
Pyrolysis process	Slow pyrolysis
Pyrolysis temperature: °C	350–400
Elemental analysis	
C: %	53.39
O: %	42.80
H: %	1.99
N: %	1.82
Ash content: %	39
CEC: cmol/kg	21.95

hydrophobic (contact angle $>90^\circ$), BC may have no effect on water retention of sandy soil (Jeffery *et al.*, 2015).

Gas permeability

Figure 5(a) shows the k_g variations with suction of soils with different BC content. It clearly shows that k_g increased linearly with soil suction in semi-log scale, regardless of the BC content. High suction means large number of pores filled with air and hence, more available flow channels for gas migration. When suction was low (i.e. ~ 20 kPa), there was no difference in k_g . However, the difference became larger with an increase in suction of soils with BC amendment. The difference in k_g increased with an increase in BC content. For higher suction (i.e. 2 MPa), k_g of BAS5% and BAS10% were

Fig. 2. Experimental set-up for measuring gas permeability

Fig. 3. Compaction curves for soils with different BC content

Fig. 4. SWR curves for soils with different BC content. Each soil case was best fitted by using van Genuchten model (van Genuchten, 1980). In the model, a is related to the air entry value; n and m control the shape of the fitted curve

smaller than that of bare soil by 50 and 65%, respectively. Similar behaviour was also observed for BC-amended clay soil at 90% DOC (Wong *et al.*, 2015).

Increased high water content due to BC (Fig. 4) may slow down the gaseous transport processes in soil as gas diffusion in water is much slower than that in the air (Cabral *et al.*, 2004). However, at low suction (~ 20 kPa), even though there was 40% difference in VWC, BC had minor effects on k_g . This is probably because there was no significant difference in air pore connectivity of BAS with different BC content when suction was low. On the contrary, when suction becomes higher, water in large pores would dissipate first without any significant changes in small pores or intra-pores retention (shown in Fig. 1(b)). The additional water in intra-pores of BC, would then cause reduction in air conductive channels for gas migration, causing the decrease in k_g in BAS. This phenomenon is greatly enhanced with higher BC content. In addition, BC can form aggregates with clay particles (Moldrup *et al.*, 2001) and act as inter-pore-filling materials. This would also provide resistance to the gas flow. However, it should be noted that when the DOC is lower ($<85\%$), BC may either have no effects on k_g in clay or increase the gas flow in agricultural

Fig. 5. Relationship between gas permeability and (a) soil suction and (b) VWC for soils with different BC content. Best-fitting equations and coefficient of best-fitting R^2 are shown for each soil case

sandy loam (Sun *et al.*, 2013; Wong *et al.*, 2015). In addition, Bordoloi *et al.* (2018b) showed that BC could reduce soil shrinkage limit. In their study, the addition of 10% BC was found to reduce desiccation cracks by more than 50% compared to bare soil. This can be a reason that might cause the reduction of gas flow paths and hence k_g in BAS.

Figure 5(b) shows the correlations between k_g and VWC. Data from both bare and BC-amended soils was superimposed for comparison. Each test series has a unique curve that is characterised by a decrease of k_g at higher levels of VWC. Moreover, the decrease rate of k_g with VWC became smaller when the BC percentage was higher. At higher BC percentage, k_g changed within a narrower range and the average k_g values were much lower. By comparing Figs 4 and 5(b), it can be inferred that the decrease of k_g with VWC is likely because the pores and gas pathway within the soil are gradually clogged with water when VWC increases. The reduced decrease rate of k_g with VWC in BAS can be explained in context of pore-size distribution. The amount of micropores is expected to increase with gradual increment of finer BC particles (Wong *et al.*, 2015). According to the capillary model, k_g is positively correlated with soil porosity and pore size (Ohji, 2013). Hence, the increased micropores of BAS (i.e. smaller pore size) with respect to bare soil could reduce the k_g even at low water content.

CONCLUSIONS

This study explored the effects of BC addition (0, 5 and 10%) on water retention and gas permeability of clay–sand mixture. Gas permeability (k_g) was interpreted together with SWR characteristics and microstructure of BC. The test results show that with 5 and 10% BC addition, water-retention capacities increased almost constantly by 6.5 and 10.5%, respectively, over a wide range of suction up to 3 MPa. k_g decreased by up to 50 and 65% in soils with 5 and 10% BC, respectively. The decreased amount became smaller at lower suction. For each BC-amended soil, there existed a unique relationship between k_g and soil VWC. k_g decreased with an increase in water content. The decrease rate was observed to reduce when the soil had a higher percentage of BC addition.

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