Supplementary Material

Biofilters and bioretention systems: The role of biochar in blue green city concept for stormwater management

K.S.D. Premarathna[1], Jayanta Kumar Biswas[2], Manish Kumar[3], Sunita Varjani[4], Bede Mickan[5], Pau Loke Show[6], John Lau Sie Yon[7], Luís A. B. Novo[8], Meththika Vithanage[1,3,5*]

[1]*Ecosphere Resilience Research Center, University of Sri Jayewardenepura, Nugegoda, Sri Lanka*

[2]*Enviromicrobiology, Ecotoxicology and Ecotechnology Research Laboratory (3E-MicroToxTech Lab), Department of Ecological Studies, and International Centre for Ecological Engineering, University of Kalyani, Kalyani, Nadia- 741235, West Bengal, India*

[3]*Sustainability Cluster, School of Engineering, University of Petroleum and Energy Studies, Dehradun, Uttarakhand 248007, India*

[4]*Gujarat Pollution Control Board, Gandhinagar 382010, Gujarat, India*

[5]*The UWA Institute of Agriculture, University of Western Australia, Perth WA6009, Australia*

[6]*Department of Chemical and Environmental Engineering, University of Nottingham, 43500 Semenyih, Selangor Darul Ehsan, Malaysia*

[7]*Department of Chemical and Energy Engineering, Curtin University, 98009 Miri Sarawak, Malaysia*

[8] *Scotland's Rural College (SRUC), West Mains Road, The King's Buildings, EH9 3JG Edinburgh, United Kingdom*

*Corresponding Author: meththika@sjp.ac.lk

Table S1: Summary of usage of pristine biochar for the removal of contaminants from stormwater

Feedstock	Temperature/ °C	Target contaminant	Observations	Inference	Reference
Wood	900 – 1000	Total ammonia nitrogen (TAN), NO_3^--N and NO_2^--N , dissolved organic carbon (DOC), *E. coli*	After 4 days of recirculation TAN concentration in effluents from sand, biochar (900 -1000 °C), and biochar (550 °C) reduced by 48%, 65%, and 67% respectively	Higher CEC and specific surface area	Rahman et al. [1]
			After 4 days of recirculation NO_x concentration in effluents of sand, biochar (900 -1000 °C), and biochar (550 °C) increased by 261%, 253%, and 84% respectively	Nitrification in each column and the simultaneous nitrification-denitrification occurring in biochar (550 °C) amended columns	
	550		*E coli* concentration in effluents from biochar (900 – 1000 °C) is below the detection limit and it is higher in biochar (550°C) than that of sand	High surface area of biochar	
Pine wood (PW)	-	TorC	TorCs removal of all biochar amended columns more than 99% (Effluent concentration less than 0.1 μg/l)	Small particle size of biochar provide more surface area for adsorption of TorC	Ulrich et al. [2]

Wood dust	300, 500, 700	BPA	Fixed bed columns containing biochar produced at 700 °C removed BPA more efficiently than biochar non amended columns and other biochar amended columns	High surface area and pore volume of biochar	Lu and Chen [3]
			Adsorption efficiency decreased at high humic acid concentrations	Humic acid compete with BPA for pores and block biochar' pores and reduce number of effective sites available for BPA to bind	
			Highest adsorption observed at neutral pH	At high pH's promote dissociation of carboxylic groups in biochar surface and dissociate carboxyl groups in BPA and make it hydrophilic so hydrophobic interactions reduced	
Reef soil	550	P, NH_4-N, NO_3^--N, total nitrogen (TN_b)	Peak flow rate and cumulative runoff volume decreased in biochar amended strips	High water retention ability of biochar	Imhoff et al. [4]
			No significant difference in concentrations of any pollutant (P, NH_4-N, NO_3-N or TN_b) in surface water samples from biochar amended and un-amended		
PW	-				

			Only NO_3-N or TN_b concentrations of subsurface water was lower in samples below the biochar amended soil		
Softwood with bark	815 – 1315	*E. coli*	In all the biochar amended columns removal of *E. coli* was higher than the sand only column Remobilization of deposited *E. Coli* during intermittent flow was lower in biochar amended columns	Overall increase of attraction forces (hydrophobic and steric interactions) Rough surface and irregular shape of biochar promote bacterial attachment via straining	Mohanty and Boehm [5]
Wood chips (commercially available)	350	*E. coli*	Removal efficiency 3 times higher than sand	Overall increase of attachment sites Increase of overall attractive forces between bacteria surface and grain surface	Mohanty et al. [6]
	700		Reduced the mobilization during the intermittent flow	Hydrophobic forces increased water holding capacity or decreased intrusion of air during gravitational drainage	
A blended mix of wood species 60% Monterey Pine, 20% Eucalyptus,	180 – 395	*E. coli*	Biochar-modified sand biofilters show enhanced *E. coli* removal compared to sand under all experimental conditions Retention of *P. aeruginosa* by biochar-	Higher specific surface area, surface roughness and hydrophobicity of biochar	Afrooz and Boehm [7]

10% Bay Laurel, 10% mixed hardwood and softwood			amended sand column is higher than the pure sand columns Injecting *E. coli* loaded stormwater to the biofilm-coated biochar-amended sand columns not showed observable ATP densities in the columns	 Contribution of ATP from *E. coli* retention is insignificant compared to the ATP from the *P. aeruginosa* biofilm	
Silvergrass (*Miscanthus* sp.)	-	P, *E. coli*	Average removal of *E. coli* by biochar amended columns higher than that of sand column and increased with the experiment time Removal of P higher in biochar amended columns than that of sand columns	The increased biological activity and the additional degradation at the deeper zones of filter High adsorption capacity of biochar due to porous structure	Kaetzl et al. [8]
A blended mix of wood species 60% Monterey Pine, 20% Eucalyptus, 10% Bay Laurel, 10% mixed hardwood and softwood	394	*E. coli*	Biochar-amended biofilters showed enhancement in *E. coli* removal during the first 31 weeks of conditioning over sand biofilters and media type did not influence *E. coli* removal during the last 30 weeks of conditioning	High organic carbon content promotes hydrophobic interactions between bacteria and biochar surface Provide additional attachment sites to bacteria and virus due to its high surface area	Kranner et al. [9]
Bamboo chips	600	NH_4^+-N, NO_3^--N, NO_2^--N or TN	Removal of large molecule organic contaminants increased with the addition of	Presence of considerable number of surface functional groups and large	Pan et al. [10]

			biochar	surface area of biochar	
			Biochar amended biofilters increased the removal efficiencies of NH_4^+- N, TN, NO_2^-–N	Good pore structure provide adequate surface area for the growth and reproduction of microorganisms and enhance the number of denitrifying microorganisms Biochar act as a supplementary carbon source for microorganisms involved in denitrification	
			NO_3^--N was accumulated in the effluent	High concentration of dissolved oxygen due to addition of biochar provided aerobic environment to facilitate the growth of nitrifying bacteria who enhanced the conversion of NO_2^--N to NO_3^--N	
Wood chips	700 – 1000	Dissolved organic nitrogen (DON), TAN, NO_x	Column contained the highest amount of biochar (BC–50%) showed higher TAN removal efficiency than column with low biochar amount (BC-20%)	Increase of surface charge availability for adsorption NH_4^+ due to the high amount of biochar in the column, Ca^{2+}, Mg^{2+}, Al^{3+}, Fe^{2+} available in the dairy runoff compete with NH_4^+ for adsorption sites of BC-20%	Rahman et al. [11]

			Higher DON removal by BC-50% than BC-20% Greater TN adsorption by BC-50% than BC-20%	Greater ammonification and nitrification-denitrification due to high adsorption capacity Enhanced adsorption, ammonification, nitrification and denitrification	
Poultry litter (PL)	300, 400, 500	NH_4^+ - N	Water retention capacity of PL-300 and SYP biochar is high relative to sand filter NH_4^+ - N adsorption capacity of PL-400 and PL-500 is higher than that of SYP biochar	Additional pour volume of biochar High CEC of PL biochar	Tian et al. [12]
Southern yellow pine (SYP)	550				
PL	400, 500	NH_4^+	PL biochar adsorbed more NH_4^+ than that of HW biochar PL-400 biochar sorbed more NH_4^+ than the PL-500 biochar Column with 10% biochar removed more than 90% of NH_4^+ than the sand only column Cation exchange is the dominated adsorption mechanism	High CEC of biochar produced at low pyrolysis temperatures	Tian et al. [13]
Hardwood pellets (HW)					

PW	>1100 >600	TorCs	TorC adsorption capacity of sand columns amended with PW-1100 biochar higher than that of PW-600 biochar	High surface area and pore volume	Ulrich et al. [14]
Spent coffee ground (SCG)	400	Caffeine (CAF), Atrazine (ATR), Diuron(DIU), Fipronil (FIP), Pentachlorophenol (PCP)	SCG adsorbed 15 – 25 % of CAF, ATR, and PCP and 60 – 90 % of DIU and FIP The removal efficiencies TOrCs decreased when exposed to mixed contaminants, except the highly hydrophobic TOrC.	The competition between multiple contaminants for binding sites reduce the removal efficiency	Redden [15]
Wood waste	700	Cu, Cd, Zn, Ni	Overall removal efficiency of metals Cu – 80 – 100% Cd – 41.1 – 100% Ni – 44.4 – 84% Zn – 51.6 – 100%	High BET surface area and pore volume facilitated adsorption The pH of outflow was slightly higher than the stormwater which reduce solubility of metals and enhance removal efficiency	Sun et al. [16]

Table S2: Summary of usage of modified biochar for the removal of contaminants from stormwater

Feedstock	Modifier	Temperature/ °C	Target contaminant	Observations	Inference	Reference
Forestry wood waste	H_2SO_4 H_3PO_4	*700*	*E. coli*	Removal percentage of E Coli by raw biochar was 96.6% and H_2SO_4 modification exceeded the removal efficiency of raw biochar slightly and minimized remobilization of bacteria H_3PO_4 and KOH modification has little influence on removal efficiency of *E. coli*	High surface area, porous structure and, surface characteristics of the biochar High surface hydrophobicity of biochar Due to presence of more oxygen	Lau et al. [17]

	KOH Amino			*E. coli* removal efficiency of amino modified biochar was 92.1% lower than other biochar yet higher than the sand only column	containing functional groups surface become polar and prevent hydrophobic bacteria	
PW	nZVI	600	As, Cd, Cu, Pb, Zn	As, Cd, Cu, Pb and Zn removal efficiency of homogeneous mixture of biochar 95%, 93%, 99%, 98% and 95% respectively As, Cd, Cu, Pb and Zn removal efficiency of biochar layered sand 96%, 99%, 100%, 100% and 99% respectively As, Cd, Cu, Pb and Zn removal efficiency of nZVI modified biochar layered sand 98%, 90%, 99%, 99% and 94% respectively	High surface area provide more sites for adsorption of Cu Complexation with active surface functional groups, cation exchange and precipitation As adsorption via surface complexation with hydroxide groups and intraparticle diffusion onto nZVI biochar complexes Cd adsorption by biochar mainly via cation exchange, surface complexation, precipitation and electrostatic interactions	Hasan et al. [18]
PW	nZVI	600	Cu, Cd	Percentage removal of Cu by sand, sand-	Metals interacted with biochar via	Hasan et al.

				biochar and sand-nZVI biochar was over 99% Percentage removal of Cd and Zn by sand-nZVI biochar was significantly high compared to pure sand column and raw biochar amended columns.	chemical reduction and surface complexation with functional groups of biochar and the iron oxide C-O and COOH groups on biochar transformed to C-O-Fe producing adsorption sites for metals	[19]
Wood waste	700	H_2SO_4	Cu, Cd, Zn, Ni	Only effective for Cu removal, efficiencies were 67.9 -97.9% and 99.6 – 100%	Increased BET surface area and total pore volume due to H_2SO_4 modification Low pH of outflow reduce removal efficiency of metals	Sun et al. [16]
Oak trees	285	Al	As	Adsorption capacity of As by Al impregnated biochar was significantly higher than that of pristine biochar. Adsorption of As increased with the increase of impregnated amount of Al	Available adsorption sites for As increased with the increase of Al content As adsorption occurred via interactions between $Al(OH)_3$ on biochar surface and arsenate in water. Main mechanisms involved are electrostatic attractions and ligand exchange	Liu et al. [20]

SCG	400	KOH	CAF, ATR, PCP, DIU, and FIP	Removed all contaminants present in the individual contaminant matrix Removed all TOrCs completely without exhibiting any preference for highly hydrophobic in the mixed contaminant matrix	High surface area, high porosity, and strong surface charge improve the adsorption of TOrCs	Redden [15]

References

1. M. Y. A. Rahman, M. H. Nachabe and S. J. Ergas, *Bioresource Technology*, 2020, **310**, 123428.
2. B. A. Ulrich, M. Loehnert and C. P. Higgins, *Environmental Science: Water Research & Technology*, 2017, **3**, 726-734.
3. L. Lu and B. Chen, *Environmental Pollution*, 2018, **243**, 1539-1549.
4. P. T. Imhoff, Culver, T. B. & Chiu, P. C., *Removing Nitrate from Stormwater with Biochar Amendment to Roadway Soils*, 2019.
5. S. K. Mohanty and A. B. Boehm, *Environmental Science & Technology*, 2014, **48**, 11535-11542.
6. S. K. Mohanty, K. B. Cantrell, K. L. Nelson and A. B. Boehm, *Water research*, 2014, **61**, 288-296.
7. A. R. M. N. Afrooz and A. B. Boehm, *PLoS One*, 2016, **11**, e0167489.
8. K. Kaetzl, M. Lübken, E. Nettmann, S. Krimmler and M. Wichern, *Scientific Reports*, 2020, **10**, 1229.
9. B. P. Kranner, A. Afrooz, N. J. M. Fitzgerald and A. B. Boehm, *PLoS One*, 2019, **14**, e0222719.
10. X. Pan, M. Chen, F. Wang and Q. Li, *Waste Management & Research*, 2020, **38**, 1176 – 1184.
11. M. Y. A. Rahman, R. Cooper, N. Truong, S. J. Ergas and M. H. Nachabe, *Chemosphere*, 2021, **283**, 130978.
12. J. Tian, S. Yi, P. T. Imhoff, P. C. Chiu, M.-X. Guo, J. A. Maresca, V. M. Beneski and S. H. Cooksey, presented in part at the World Environmental and Water Resources Congress 2014.
13. J. Tian, V. Miller, P. Chiu, J. Maresca, M. Guo and P. Imhoff, *The Science of the total environment*, 2016, **553**, 596-606.

14. B. A. Ulrich, E. A. Im, D. Werner and C. P. Higgins, *Environmental Science & Technology*, 2015, **49**, 6222-6230.

15. N. Redden, Master of Science in Civil Engineering, University of Washington, 2021.

16. Y. Sun, S. S. Chen, A. Y. T. Lau, D. C. W. Tsang, S. K. Mohanty, A. Bhatnagar, J. Rinklebe, K.-Y. A. Lin and Y. S. Ok, *Journal of Hazardous Materials*, 2020, **383**, 121243.

17. A. Y. T. Lau, D. C. W. Tsang, N. J. D. Graham, Y. S. Ok, X. Yang and X.-d. Li, *Chemosphere*, 2017, **169**, 89-98.

18. M. S. Hasan, R. Vasquez and M. Geza, *Processes*, 2021, **9**, 860.

19. M. S. Hasan, M. Geza, R. Vasquez, G. Chilkoor and V. Gadhamshetty, *Water, Air, & Soil Pollution*, 2020, **231**, 220.

20. Q. Liu, L. Wu, M. Gorring and Y. Deng, *Journal of Environmental Engineering*, 2019, **145**, 04019008.