Supplement of

An investigation on hygroscopic properties of 15 black carbon (BC) from different carbon sources: Roles of organic and inorganic components

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Table S1. Bulk elemental compositions determined by elemental analysis and surface elemental compositions determined by X-ray photoelectron spectroscopy (XPS) of different BC.

Samples	Bulk elemental compositions[a]						Surface elemental compositions[b]				
	C (wt%)	O (wt%)	H (wt%)	N (wt%)	S (wt%)	Ash (wt%)	C (wt%)	O (wt%)	N (wt%)	Si (wt%)	S (wt%)
Amaranth BC	32.08	21.92	2.56	2.63	0.781	40.03	75.21	18.98	4.28	1.05	0.47
Grass BC	58.8	23.18	3.92	2.285	0.156	11.66	76.02	19.53	2.67	1.42	0.37
Peanuts BC	49.31	18.35	3.81	1.485	0.874	26.17	71.44	22.52	3.4	2.25	0.4
Pea BC	64.065	22.19	4.13	1.35	0.454	7.809	74.75	20.56	2.62	1.75	0.32
Rice BC	54.87	17.4	3.35	0.76	0.315	23.31	73.39	20.37	1.57	4.67	ND[g]
Wheat BC	50.895	18.85	3.22	0.97	0.405	25.66	69.69	22.86	2.82	4.63	ND[g]
Millet BC	41.66	20.23	3.09	1.91	0.468	32.64	61.1	29.02	3.84	5.52	0.51
Corn BC	53.02	16	2.7	1.305	0.39	26.59	76.72	17.88	1.97	3.42	ND[g]
Sorghum BC	64.35	20.04	4.03	0.835	0.334	10.41	73.28	22.85	1.58	1.96	0.32
Bamboo BC	68.535	19.86	3.54	0.44	0.168	7.456	76.28	19.39	1.11	2.88	0.34
Red pine BC	69.985	25.52	3.8	0.22	0.077	0.395	82.9	16.43	ND[g]	0.66	ND[g]
Poplar BC	71.795	23.63	3.78	0.425	0.129	0.245	86.57	12.59	ND[g]	0.84	ND[g]
Diesel engine soot	36.94	20.05	2.64	3.31	1.652	35.41	78.95	18.72	1.07	1.26	ND[g]
Weifu diesel soot	76.455	18.51	2.19	0.385	0.979	1.482	78.07	21.15	ND[g]	0.78	ND[g]
Household soot	37.075	22.07	2.89	3.655	1.734	32.58	76.09	19.3	2.97	1.05	0.6

Table S2. Elemental composition, ash content, atomic ratio, and polarity index of alkali-extracted organic carbon (OC_{AE}) from three representative BC by elemental analysis.

Samples	Compositions (wt%)							
	C	O	H	N	S	H/C	(O+N)/C	Ash
Grass OC_{AE}	28.16	27.14	3.02	1.41	0.49	0.11	1.01	39.78
Wheat OC_{AE}	18.61	21.98	2.71	0.46	0.91	0.15	1.21	55.33
Household soot OC_{AE}	56.56	31.31	5.03	6.14	0.96	0.09	0.66	ND[a]

[a]Not detected.

Table S3. Ratio of the peak intensities of D band (1350 cm^{-1}) to G band (1582 cm^{-1}) of Raman spectra for different BC.

Samples	I_D/I_G[a]
Amaranth BC	1.01
Grass BC	0.88
Peanuts BC	0.89
Pea BC	0.85
Rice BC	1.09
Wheat BC	0.98
Millet BC	0.86
Corn BC	0.94
Sorghum BC	0.9
Bamboo BC	1.09
Red pine BC	0.59
Poplar BC	0.57
Diesel engine soot	1.04
Weifu diesel soot	1.12
Household soot	0.77

Table S4. Salinities of water extracts of different BC (BC to water ratio: 1/10, w/w).

Samples	Salinity (‰)
Amaranth BC	0.180
Grass BC	0.080
Peanuts BC	0.070
Pea BC	0.060
Rice BC	0.010
Wheat BC	0.097
Millet BC	0.133
Corn BC	0.030
Sorghum BC	0.080
Bamboo BC	0.010
Red pine BC	0.000
Poplar BC	0.010
Diesel engine soot	0.060
Weifu diesel soot	0.057
Household soot	0.217

Table S5. Mineral compositions of different BC measured by X-ray fluorescence spectroscopy (XRF).

Samples	Mineral compositions (%)																		
	SO_3	CaO	P_2O_5	Al_2O_3	ZnO	SiO_2	Fe_2O_3	Cl	K_2O	MgO	Na_2O	MnO	CuO	PbO	NiO	TiO_2	Cr_2O_3	BaO	Sr
Amaranth BC	0.500	1.070	0.240	0.480	0.002	1.730	0.210	0.170	2.140	1.060	0.130	0.006	0.001	ND[a]	ND[a]	0.022	ND[a]	0.007	0.002
Grass BC	0.092	0.290	0.360	0.037	0.001	1.500	0.014	0.140	2.380	ND[a]	ND[a]	0.004	ND[a]	ND[a]	ND[a]	ND[a]	0.001	ND[a]	ND[a]
Peanuts BC	0.120	0.510	0.320	0.310	0.002	1.920	0.073	0.038	1.260	0.360	0.058	0.015	ND[a]	ND[a]	ND[a]	0.011	ND[a]	ND[a]	ND[a]
Pea BC	0.120	0.480	0.120	0.016	N.D.	0.120	0.007	0.008	0.770	0.340	0.021	N.D.	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]
Rice BC	0.024	0.050	0.030	0.016	0.001	2.630	0.005	0.006	0.220	0.023	ND[a]	0.006	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]
Wheat BC	0.160	0.230	0.110	0.085	ND[a]	4.020	0.039	0.490	1.720	0.100	0.036	0.009	ND[a]	ND[a]	ND[a]	0.005	ND[a]	ND[a]	ND[a]
Millet BC	0.210	0.360	0.340	0.040	0.007	5.750	0.028	0.160	2.290	0.790	ND[a]	0.007	ND[a]	ND[a]	0.002	ND[a]	ND[a]	0.011	ND[a]
Corn BC	0.160	0.360	0.150	0.110	N.D.	3.740	0.038	0.260	1.200	0.190	0.027	0.022	ND[a]	0.004	ND[a]	0.007	ND[a]	ND[a]	ND[a]
Sorghum BC	0.190	0.710	0.059	0.047	0.002	0.500	0.025	0.300	1.820	0.320	0.037	0.002	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]
Bamboo BC	0.120	0.110	0.180	0.260	0.003	1.280	0.029	0.130	1.630	0.110	ND[a]	0.016	ND[a]	ND[a]	ND[a]	0.003	ND[a]	ND[a]	ND[a]
Red pine BC	0.010	0.110	0.008	0.006	N.D.	0.026	0.002	0.002	0.072	0.230	ND[a]	0.005	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]
Poplar BC	0.150	0.980	0.280	0.019	0.003	0.220	0.010	0.046	1.040	0.025	ND[a]	0.004	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]	ND[a]
Diesel engine soot	2.890	1.090	0.200	0.180	N.D.	0.900	3.090	0.100	0.067	0.100	0.054	0.013	ND[a]	ND[a]	0.013	0.019	0.010	0.013	ND[a]
Weifu soot	0.250	0.230	0.094	0.036	0.019	0.016	0.014	0.006	0.002	0.170	ND[a]	0.001	0.002	0.003	0.001	ND[a]	0.002	ND[a]	ND[a]
Household soot	1.530	1.730	0.130	0.740	0.005	2.550	0.230	2.070	0.960	0.240	0.055	0.037	ND[a]	ND[a]	ND[a]	0.051	ND[a]	0.012	0.002

[a]Not detected.

Table S6. Ionic constituents of different BC measured by ion chromatography.

Samples	Ionic contents (mg g^{-1})											
	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	COO^-	$C_2O_4^{2-}$	SO_4^{2-}	NO_3^-	PO_4^{3-}	F^-
Amaranth BC	1.6	0.77	9.52	7.64	0.86	13.46	ND[a]	ND[a]	14.9	0.17	1.94	0.09
Grass BC	1.71	2.37	6.4	0.72	2.57	8.43	0.79	0.54	2.27	0.22	2.7	0.1
Peanuts BC	0.4	0.36	5.79	0.41	0.43	0.69	0.04	2.07	3.72	0.05	0.67	0.34
Pea BC	2.03	2.26	4.74	0.78	1.43	0.28	0.06	2.69	5.56	0.1	0.45	0.15
Rice BC	0.57	2.69	0.13	0.11	0.26	0.06	0.01	1.06	0.63	0.01	0.16	0.01
Wheat BC	0.79	1.58	7.5	0.19	0.47	3.92	0.03	ND[a]	6.95	0.16	0.73	ND[a]
Millet BC	2.42	2.62	10.55	1.77	1.61	1.22	0.18	15.37	7.98	0.07	1.63	0.14
Corn BC	0.19	1.93	2.13	0.28	0.51	4.42	0.05	0.87	3.38	0.02	0.91	0.03
Sorghum BC	0.74	0.57	5.94	0.63	1	1.72	0.06	0.76	4.63	0.11	0.22	0.29
Bamboo BC	0.25	2.67	0.11	0.11	0.24	0.04	0.03	0.85	0.87	0.02	0.25	0.01
Red pine BC	0.15	1.28	0.15	0.05	0.13	0.05	0.02	N.D.	0.14	0.06	0.08	0.003
Poplar BC	0.1	0.55	0.42	0.17	0.38	0.28	0.01	0.07	0.38	0.01	0.24	0.5
Diesel engine soot	1.72	4.88	0.35	0.97	3.74	0.31	0.13	1.76	30.06	0.15	0.39	0.6
Weifu soot	1.15	0.13	0.07	0.29	3.65	0.19	ND[a]	ND[a]	23.51	0.2	5.41	0.42
Household soot	1.2	19.71	2.69	1.7	4.85	47.51	ND[a]	ND[a]	26.63	0.96	ND[a]	0.36

[a]Not detected.

Table S7. Accuracy (R^2 and P) values for regression on equilibrium water uptake against compositional and pore property parameters at different relative humidity (RH) levels.

Composition	23% RH		33% RH		43% RH		47% RH		75% RH		84% RH		94% RH	
	R^2	P	R^2	P	R^2	P	R^2	P	R^2	P	R^2	P	R^2	P
OC_{TGA}	0.32	0.028	0.36	0.0187	0.4	0.0113	0.47	0.0048	0.7	0.0001	0.82	$<$0.0001	0.52	0.0002
OC_{AE}	0.12	0.207	0.14	0.1652	0.17	0.1311	0.22	0.08	0.41	0.0097	0.64	0.0004	0.8	0.0001
EC	0.21	0.083	0.21	0.087	0.22	0.0798	0.25	0.06	0.39	0.0122	0.51	0.003	0.54	0.0019
Dissolved minerals	0.1	0.2471	0.11	0.23	0.12	0.2111	0.39	0.15	0.27	0.0468	0.45	0.0064	0.86	0.0001
NH_4^+	0.1	0.2548	0.13	0.19	0.16	0.1444	0.21	0.09	0.42	0.0092	0.6	0.0007	0.5	0.0034
Cl^-	0.04	0.4635	0.06	0.39	0.07	0.3305	0.11	0.23	0.24	0.0619	0.43	0.0076	0.7	0.0001
$C_2O_4^{2-}$	0.22	0.1745	0.21	0.1881	0.2	0.19	0.23	0.1566	0.29	0.1116	0.4	0.0516	0.23	0.1587
SO_4^{2-}	0.02	0.6518	0.01	0.7	0.006	0.7933	0.00003	0.95	0.06	0.395	0.15	0.1588	0.24	0.06
Total porosity	0.42	0.0095	0.4	0.01	0.39	0.0129	0.37	0.02	0.29	0.0368	0.22	0.0761	0.08	0.3
K^+	0.19	0.1088	0.16	0.139	0.14	0.1647	0.14	0.1764	0.09	0.2664	0.1	0.245	0.27	0.045

Table S8. Fitting parameters for water uptake kinetics of BC by pseudo-first order model at different relative humidity (RH) levels.

BC	33% RH				47% RH				94% RH			
	k_1[a] (10^{-5} s^{-1})	Q_e (cal)[b] (mg g^{-1})	Q_e (exp)[c] (mg g^{-1})	R^2	k_1[a] (10^{-5} s^{-1})	Q_e (cal)[b] (mg g^{-1})	Q_e (exp)[c] (mg g^{-1})	R^2	k_1[a] (10^{-5} s^{-1})	Q_e (cal)[b] (mg g^{-1})	Q_e (exp)[c] (mg g^{-1})	R^2
Amaranth BC	2±2	9±1	18±1	0.9879	2.01±0.03	22±2	30±1	0.9885	10±0	290±40	270±3	0.9993
Grass BC	3±2	10±2	16±3	0.9792	2±2	21±1	38±4	0.9359	0.9±0.1	120±10	180±6	0.9723
Peanuts BC	2±2	6±1	27±1	0.983	2±1	13±0	41±2	0.9154	10±0	100±10	140±4	0.9558
Pea BC	1±1	9±3	27±1	0.9979	2±3	15±2	45±6	0.9571	0.9±0.2	55±3	120±6	0.9258
Rice BC	0.4±0.1	5±2	21±4	0.9759	0.8±0.1	6±0	29±2	0.8805	1.4±0.1	25±0	55±3	0.8778
Wheat BC	1±1	9±2	21±5	0.948	3±1	15±4	40±4	0.9998	1.5±1.3	230±60	240±2	0.8017
Millet BC	2±1	13±0	38±5	0.9831	3±2	12±3	54±2	0.9804	1.3±0.6	96±7	190±7	0.8805
Corn BC	0.7±0.1	7±1	19±6	0.9042	3±1	15±2	28±0	0.9643	1.3±0.6	62±6	96±3	0.9663
Sorghum BC	1±1	12±1	30±13	0.9491	0.9±0.1	11±3	44±2	0.9709	1.3±0.6	130±12	190±10	0.9564
Bamboo BC	1±0	7±6	33±3	0.9159	0.6±0.4	10±2	50±8	0.9138	2±1	19±1	72±9	0.8948
Redpine BC	0.7±0.1	4.2±0.2	22±2	0.8172	5±3	20±6	39±1	0.8068	1±1	20±5	68±2	0.8252
Poplar BC	0.7±0.1	6.02±1.21	26±2	0.8745	2±1	8±2	33±2	0.8218	0.9±0.2	18±2	63±2	0.8771
Diesel engine soot	0.8±0.3	2.2±0.3	13±1	0.9932	0.8±0.4	8±3	30±7	0.9462	2±1	90±21	140±4	0.9686
Weifu diesel soot	0.4±0.1	14±5	21±14	0.8009	3±1	12±1	32±7	0.8926	6±1	31±7	69±6	0.9858
Household soot	1±0	2.12±0.01	9±1	0.8569	3±1	17±4	29±2	0.9524	1±1	410±40	420±20	0.9889

[a]Pseudo-first order rate constant. [b]Model calculated maximum sorbed concentration at equilibrium. [c]Measured maximum sorbed concentration at equilibrium.

Table S9. Fitting parameters for water uptake kinetics of BC by pseudo-second order model at different relative humidity (RH) levels.

BC	33% RH				47% RH				94% RH			
	k_2[a] (10^{-5} g mg^{-1}s^{-1})	Q_e (cal)[b] (mg g^{-1})	Q_e (exp)[c] (mg g^{-1})	R^2	k_2[a] (10^{-5} g mg^{-1}s^{-1})	Q_e (cal)[b] (mg g^{-1})	Q_e (exp)[c] (mg g^{-1})	R^2	k_2[a] (10^{-7} g mg^{-1}s^{-1})	Q_e (cal)[b] (mg g^{-1})	Q_e (exp)[c] (mg g^{-1})	R^2
Amaranth BC	0.5±0.1	18±1	18±1	0.9954	0.14±0.01	34±1	30±1	0.9963	0.31±0.02	330±11	260±4	0.9943
Grass BC	0.52±0.04	17±2	16±3	0.9963	0.20±0.03	40±3	38±4	0.9984	1.7±0.1	190±5	170±6	0.9975
Peanuts BC	2±1	28±0	27±1	0.995	0.6±0.2	41±2	41±2	0.9983	2.6±0.2	150±3	140±4	0.9974
Pea BC	0.8±0.2	28±1	27±1	0.9967	0.5±0.1	46±7	45±6	0.9987	5.3±0.4	120±5	120±6	0.9984
Rice BC	1.7±0.3	21±4	21±4	0.9955	5±2	28±2	29±2	0.9917	28±2	55±4	55±3	0.9985
Wheat BC	0.7±0.1	21±5	21±5	0.9945	1.2±0.1	37±9	40±4	0.9959	0.5±0.1	290±0	240±3	0.9889
Millet BC	0.6±0.2	39±5	38±5	0.9995	0.8±0.2	54±2	54±2	0.9997	3.1±0.2	190±7	180±7	0.9993
Corn BC	2±2	19±5	19±6	0.9986	0.36±0.01	29±1	28±0	0.9984	4.2±0.5	100±3	96±3	0.9959
Sorghum BC	0.34±0.03	29±6	30±13	0.9912	1.01±0.44	45±2	44±2	0.9986	1.7±0.1	210±11	190±10	0.9958
Bamboo BC	1.5±0.4	34±3	33±3	0.9978	4±2	50±8	50±8	0.9992	33±3	71±10	73±9	0.9961
Redpine BC	1.6±0.1	22±2	22±2	0.9938	0.43±0.01	40±1	39±1	0.9983	21±4	70±2	69±2	0.9991
Poplar BC	2±1	24±3	24±3	0.9996	0.55±0.04	33±2	32±2	0.9991	23±4	63±2	63±2	0.9985
Diesel engine soot	1.6±0.4	13±1	13±1	0.9931	0.6±0.1	30±4	30±4	0.9947	3.2±0.1	150±5	140±5	0.9987
Weifu soot	0.5±0.1	21±14	21±14	0.9706	0.9±0.3	31±7	32±7	0.9908	86±95	72±16	70±6	0.9954
Household soot	5±2	9±0	5±6	0.9948	0.20±0.03	32±2	28±2	0.9949	0.21±0.02	530±0	420±20	0.991

[a]Pseudo-second order rate constant. [b]Model calculated maximum sorbed concentration at equilibrium. [c]Measured maximum sorbed concentration at equilibrium.

Table S10. Accuracy (R^2 and P) values for regression on pseudo-second order rate constant against compositional and pore property parameters at different relative humidity (RH) levels.

Composition	33% RH		47% RH		94% RH	
	R^2	P	R^2	P	R^2	P
OC_{TGA}	0.47	0.0046	0.06	0.3845	0.28	0.0423
OC_{AE}	0.44	0.0070	0.10	0.2574	0.14	0.1672
EC	0.14	0.1700	0.05	0.4194	0.45	0.0061
Dissolved minerals	0.08	0.3100	0.19	0.1086	0.17	0.1302
NH_4^+	0.77	$<$0.0001	0.01	0.7118	0.06	0.3946
Cl^-	0.60	0.0007	0.08	0.3181	0.08	0.3118
$C_2O_4^{2-}$	0.16	0.2577	0.02	0.6964	0.07	0.4476
SO_4^{2-}	0.11	0.2286	0.10	0.2529	0.02	0.6618
Total porosity	0.03	0.5300	0.0001	0.9696	0.82	$<$0.0001
K^+	0.15	0.1509	0.13	0.1789	0.32	0.0266

II. Figures.

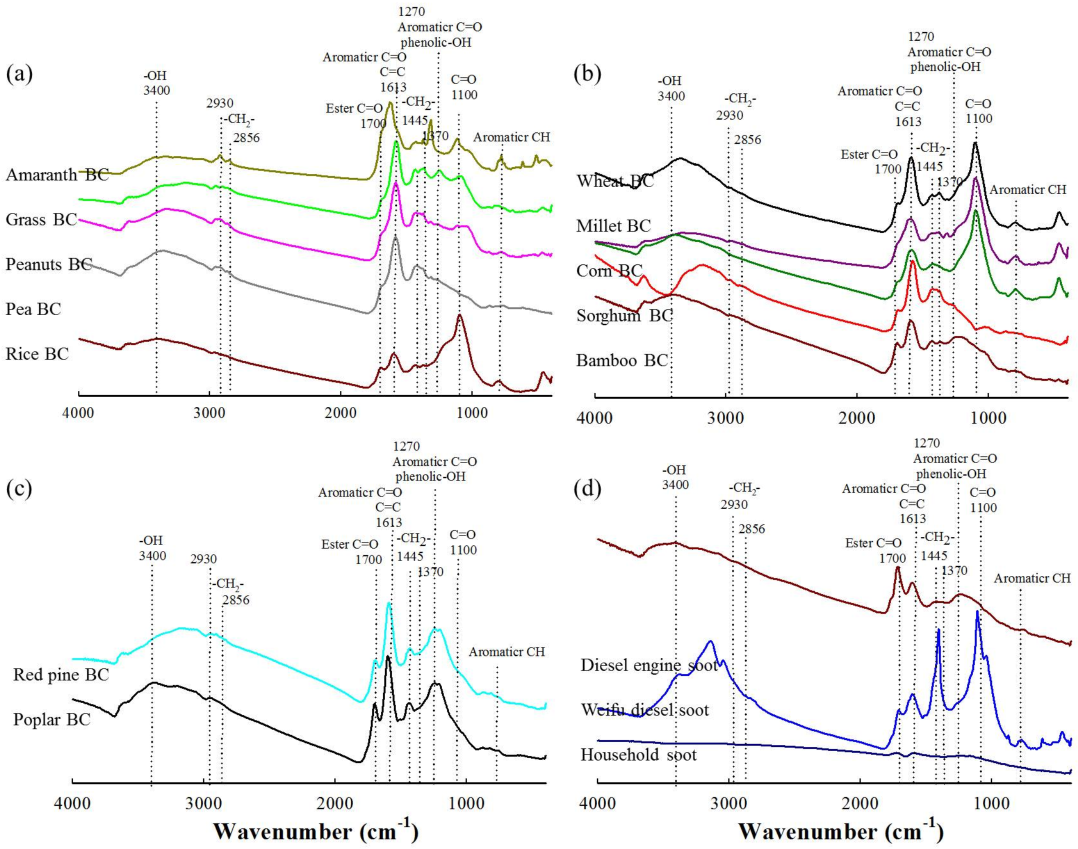

Figure S1. Fourier-transform infrared (FTIR) spectra of different BC. (a) Subgroup 1 of herbal BC. (b) Subgroup 2 of herbal BC. (c) Woody BC. (d) Soot.

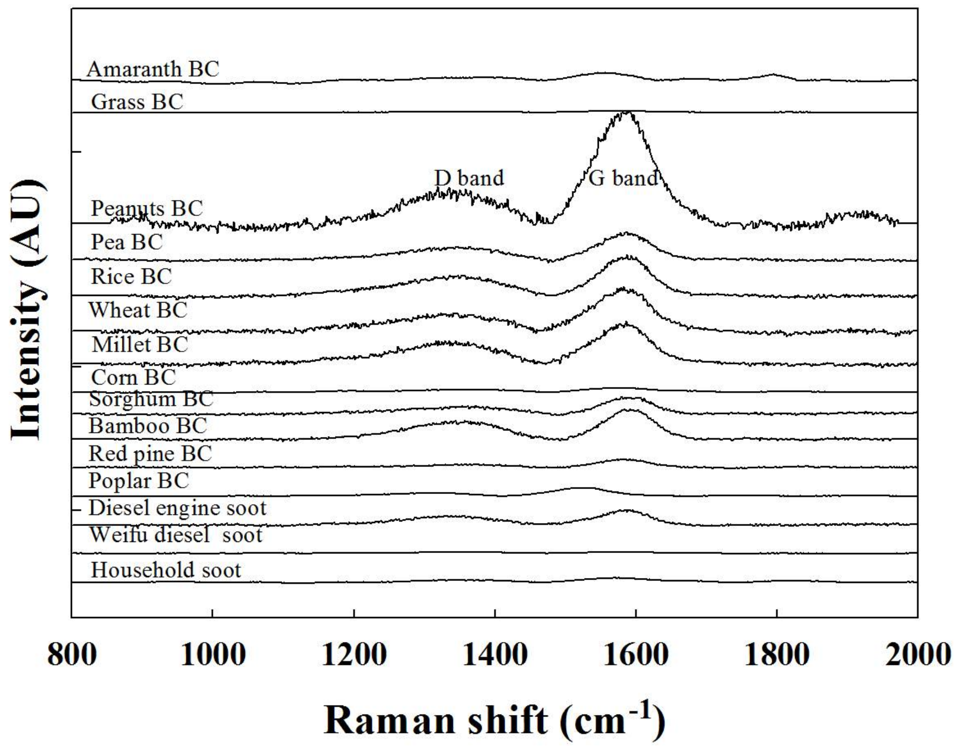

Figure S2. Raman spectra of different BC.

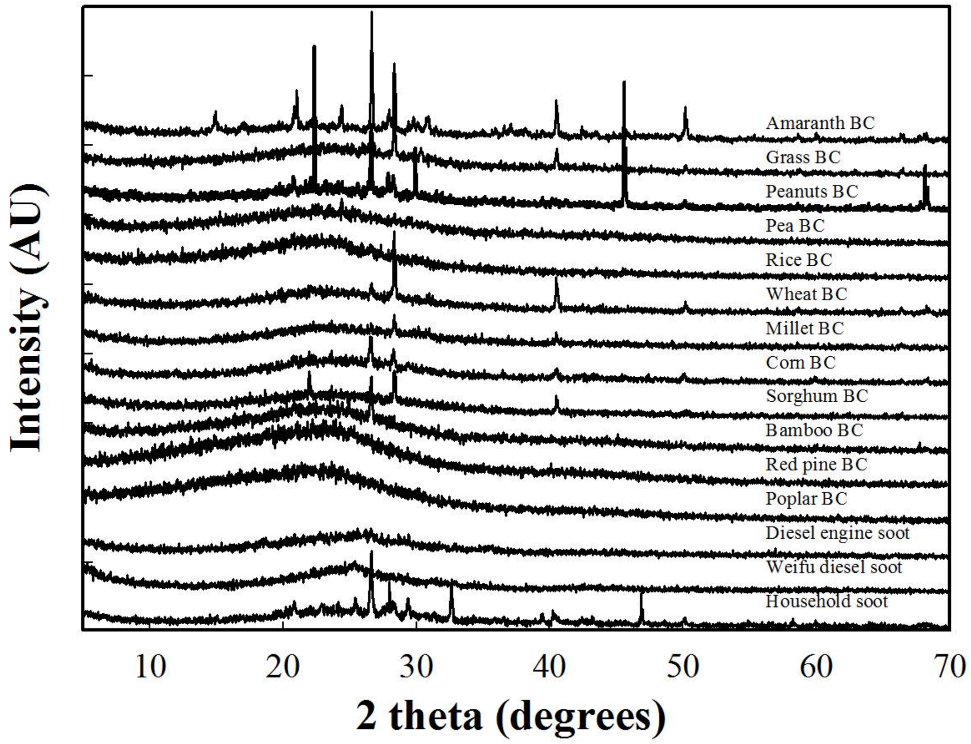

Figure S3. X-ray diffraction (XRD) profiles of different BC.

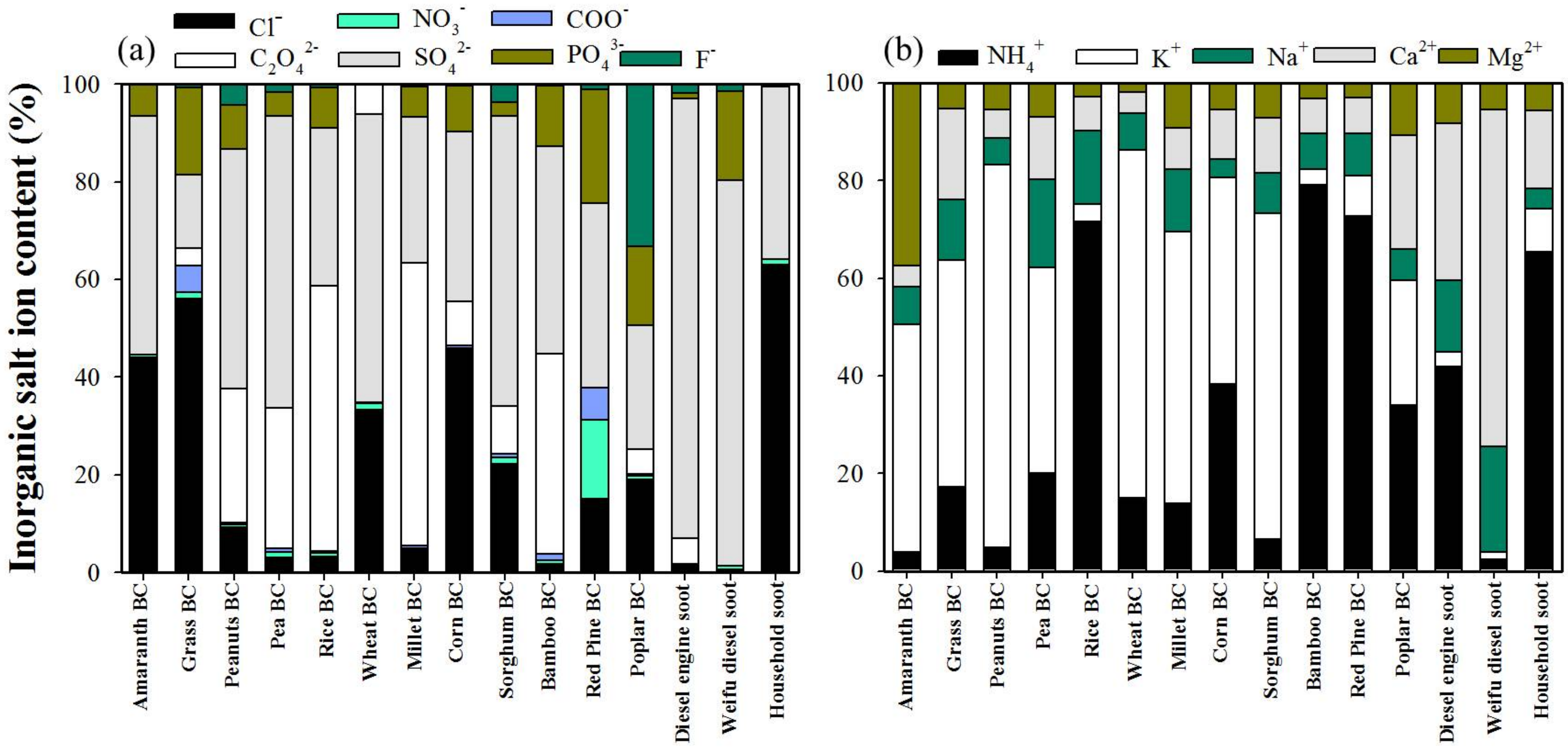

Figure S4. Compositional percentages of ionic constituents of different BC.

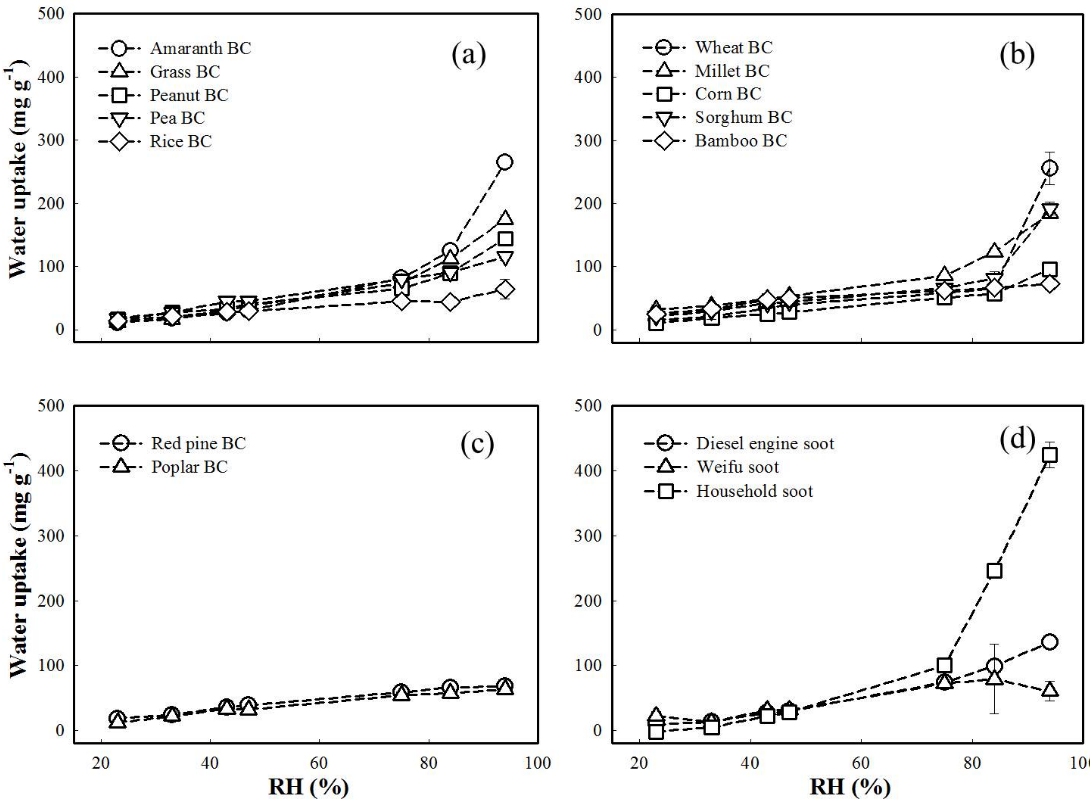

Figure S5. Sorption isotherms of water vapor plotted as equilibrium water uptake (mg g-1) vs. relative humidity (RH, %) obtained by saturated aqueous salt solutions for different BC.

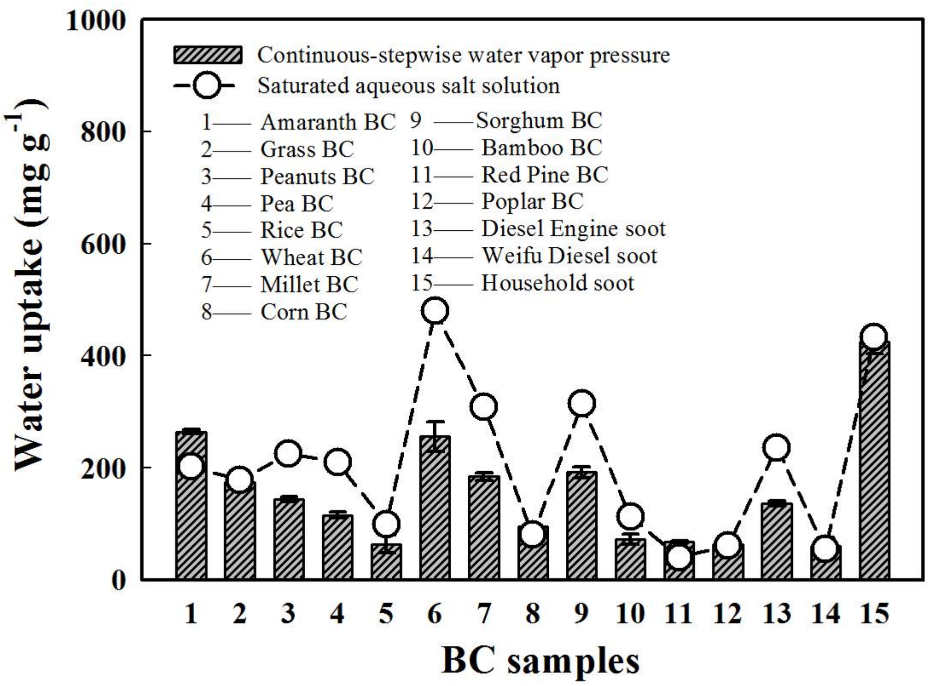

Figure S6. Comparison of equilibrium water uptake by BC at 94% relative humidity measured by two different gravimetric methods.

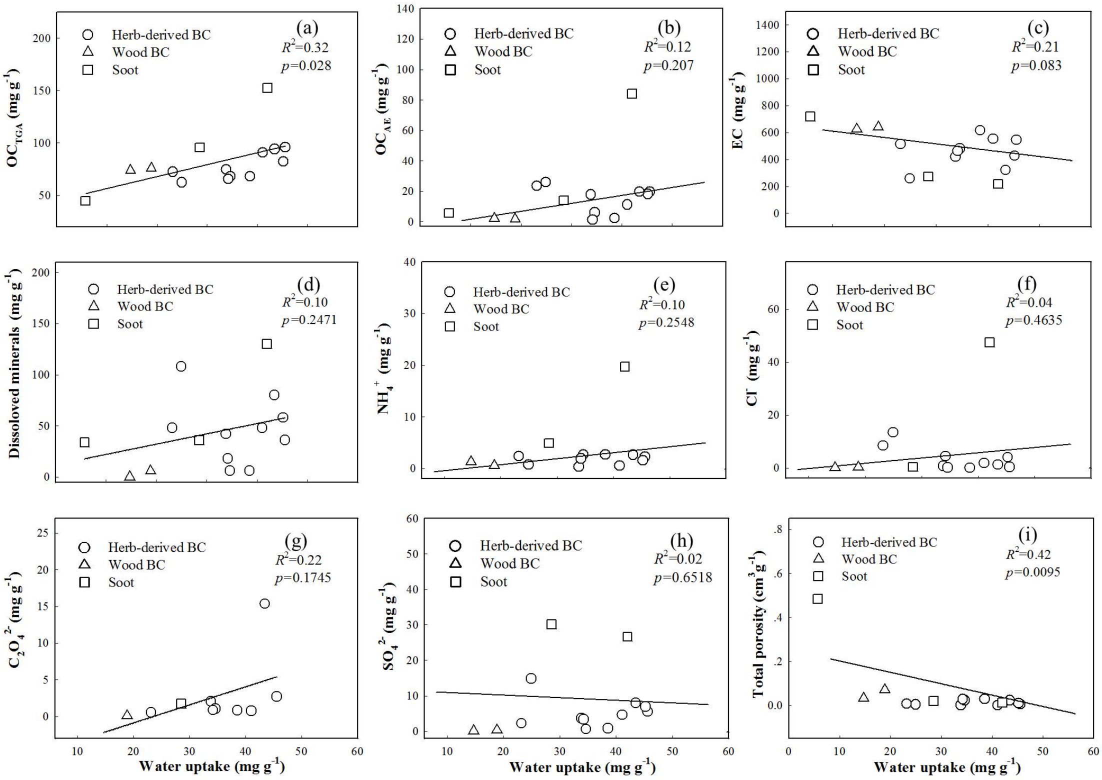

Figure S7. Relationships between equilibrium water uptake (mg g^{-1}) vs. compositional and pore property parameters for the BC pool at 23% relative humidity.

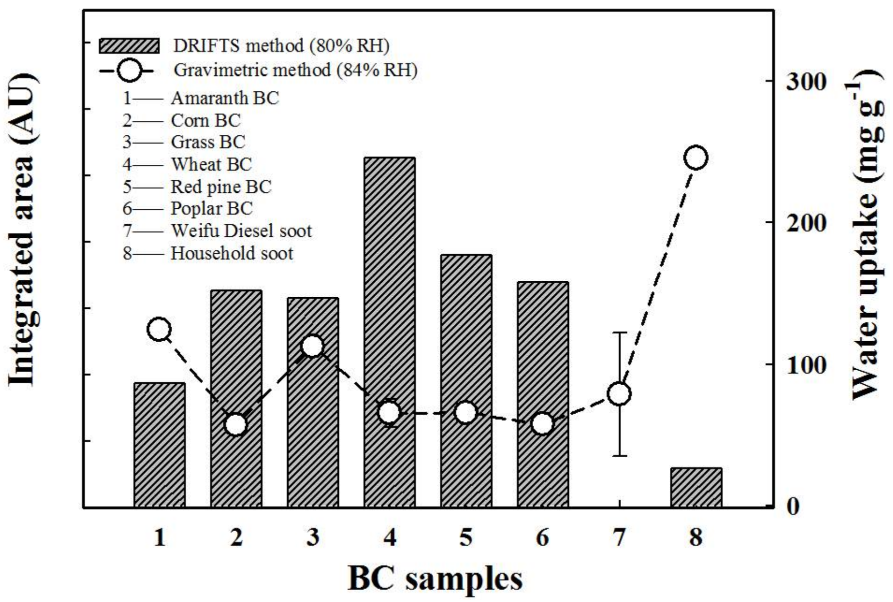

Figure S8. Comparison of equilibrium water uptake measured by gravimetric method and DRIFTS method for selected BC at high relative humidity.

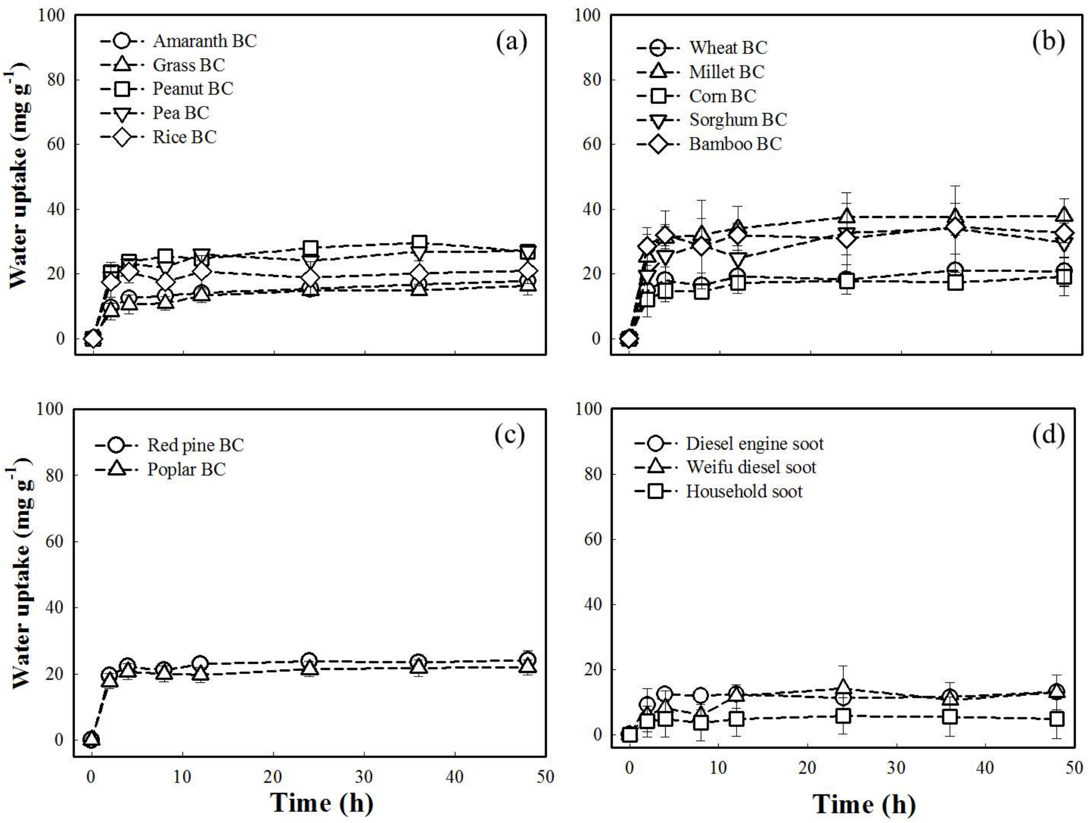

Figure S9. Sorption kinetics of water vapor plotted as water uptake (mg g^{-1}) vs. time (h) at 33% relative humidity.

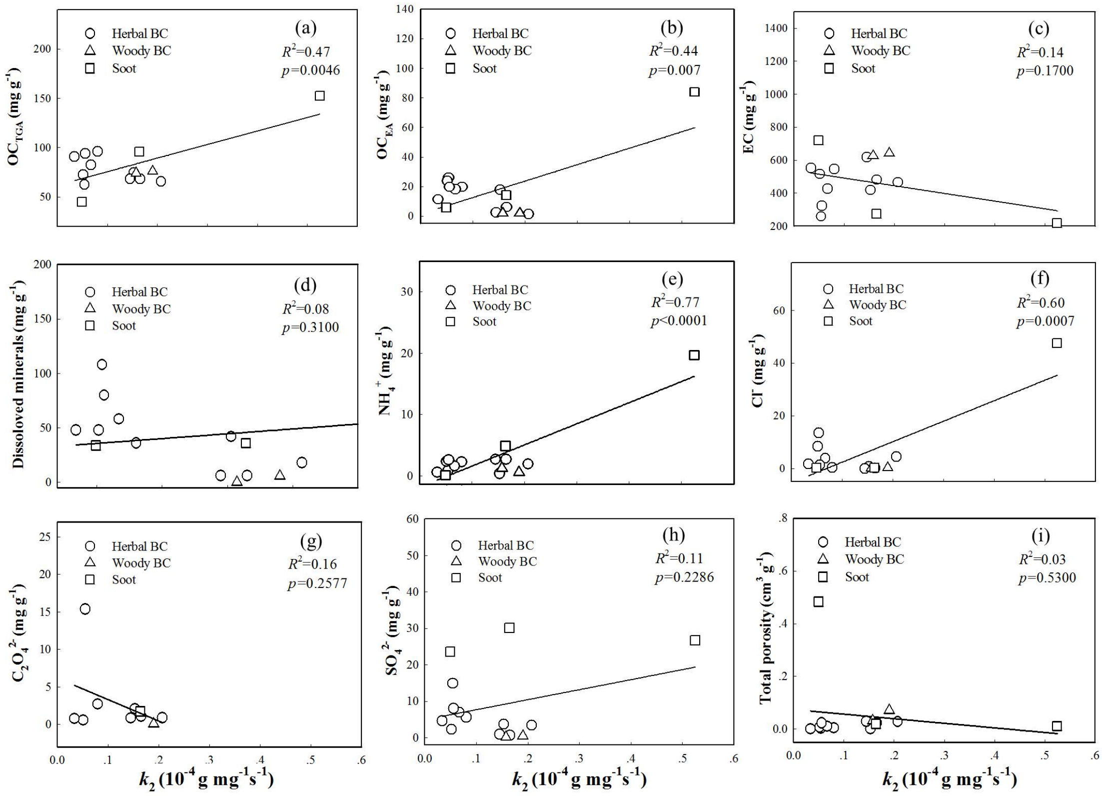

Figure S10. Relationships between pseudo-second water uptake rate constant (k_2) (g $mg^{-1}s^{-1}$) vs. compositional and pore property parameters for the BC pool at 33% relative humidity.