Supplementary Material (SM) for Article

Enhancing the Fuel Properties of Spent Coffee Grounds through Hydrothermal Carbonization: Output Prediction and Post-Treatment Approaches

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(**a**) (**b**) (**c**)

Figure S1. Pressurized reactors used for performing HTC experiments:
(**a**) 1 L (model 4520, PARR, USA);
(**b**) 1.5 L (model BR-1000, Berghof, Germany);
(**c**) 18.75 L (model 4555, PARR, USA).

Figure S2. The intensive mixer type EL10 (EIRICH Machines, Inc., USA) was used for conducting the agglomeration experiments.

Table S1. Predicted and measured results of the outputs (incl. Solid yield - SY [wt%-db], Higher heating value - HHV [MJ/kg] and energy yield - EY [%]) of two groups of HTC set-up runs (DoE set-up and Random set-up).

HTC Runs	**SY [wt%-db]**			**HHV [MJ/kg]**				**EY [%]**				
	meas	**pred^{DoE}**	**pred$^{GP_SY-OP}$**	**meas**	**pred^{DoE}**	**pred$^{GP_HHV-EA}$**	**pred$^{GP_HHV-OP}$**	**meas**	**pred^{DoE}**	**pred$^{GP_EY-EA}$**	**pred$^{GP_EY-OP}$**	**pred$^{GP_EY-EA+OP}$**
SCG	-	-	-	22.54	-	-	-	-	-	-	-	-
DoE runs												
D_240_1_13%	59.87	55.94	54.83	31.11	31.06	32.54	27.16	82.63	77.47	78.31	66.05	65.36
D_160_5_6%	71.84	71.06	66.37	25.22	25.70	25.01	24.06	80.38	80.28	72.87	70.83	70.09
D_240_5_6%	51.74	52.26	47.16	32.97	32.68	34.74	27.93	75.66	75.64	71.92	58.42	57.81
D_160_1_6%	69.16	70.35	69.86	24.35	24.24	24.26	23.55	74.71	76.31	74.40	72.97	72.21
D_200_3_13%	66.24	65.67	63.63	29.11	28.31	29.78	25.67	85.53	81.75	83.19	72.45	71.70
D_160_5_13%	75.80	75.40	70.51	25.47	25.55	25.31	24.05	85.63	86.03	78.34	75.24	74.45
AAE [%]	-	1.96 %	6.13 %	-	1.08 %	2.35 %	8.89 %	-	2.24 %	5.20 %	14.08 %	14.98 %
ABE [%]	-	−1.05 %	−5.79 %	-	−0.34 %	1.74 %	−8.89 %	-	−1.37 %	−5.20 %	−14.08 %	−14.98 %
Random runs												
R_230_3_20%	61.91	63.12	60.92	30.70	30.76	35.70	27.07	84.30	85.96	95.47	73.15	72.39
R_250_3_20%	58.40	58.40	55.13	31.88	32.51	35.75	28.00	82.57	84.77	86.51	68.49	67.77
R_220_1_10%	58.79	58.77	59.24	29.03	29.39	26.22	26.25	75.71	76.15	68.17	68.99	68.27
R_240_1_10%	51.94	54.07	53.72	31.06	31.13	27.40	27.16	71.56	74.99	64.60	64.71	64.04
R_200_3_20%	70.21	70.11	67.76	28.13	28.15	28.73	25.67	87.61	87.62	85.45	77.15	76.34
R_220_3_20%	64.89	65.43	63.57	30.07	29.89	30.66	26.60	86.55	86.48	85.56	75.02	74.23
R_230_3_20%	62.73	63.11	58.05	31.03	30.76	31.32	27.07	86.36	85.95	79.81	69.71	68.98
AAE [%]	-	1.09 %	3.48 %	-	0.74 %	7.85 %	11.31 %	-	1.51 %	6.99 %	13.33 %	14.23 %
ABE [%]	-	1.04 %	−2.28 %	-	0.32 %	1.71 %	−11.31 %	-	1.35 %	−1.84 %	−13.33 %	−14.23 %
Both groups												
(DoE and Random)												
AAE [%]	-	1.49 %	4.71 %	-	0.89 %	5.32 %	10.19 %	-	1.85 %	6.16 %	13.67 %	14.58 %
ABE [%]	-	0.08 %	−3.90 %	-	0.02 %	1.73%	−10.19 %	-	0.10 %	−3.39 %	−13.67 %	−14.58 %

[1] meas = measured values; pred^{DoE} = predicted values from DoE model; pred$^{GP_SY-OP}$ = predicted solid yield from GP model based on HTC operating conditions; pred$^{GP_HHV-EA}$ = predicted HHV from GP model based on the elemental composition of the HC (or FS); pred$^{GP_HHV-OP}$ = predicted HHV from GP model based on measured HHV_0 - FS and HTC-operating conditions; pred$^{GP_EY-EA}$ = predicted energy yield from GP model based on the elemental composition of FS and HC (no measured HHV) and SY-OP; pred$^{GP_EY-OP}$ = predicted energy yield from GP model based on measured HHV_0 - FS value and HTC-operating conditions – using the HHV-OP and SY-OP correlations; pred$^{GP_EY-EA+OP}$ = predicted energy yield from GP model based on based on the HHV-EA for FS, HHV-OP for HC and SY-OP correlations; [2] AAE = average absolute error; ABE = average bias error.

Table S2. Comparision of measured values from the literature and this study to predicted values from DoE model .

HTC Operating Conditions			Measured Values			Predicted Values from DoE Model of this Study			References
T [°C]	*t* [h]	*%So* [wt%-db]	SY [wt%-db]	HHV [MJ/kg]	EY [%]	SY [wt%-db]	HHV [MJ/kg]	EY [%]	
240	1	12.98	59.87	31.11	82.63	55.94	31.06	77.47	This study
160	5	6.02	71.84	25.22	80.38	71.06	25.70	80.28	This study
240	5	6	51.74	32.97	75.66	52.26	32.68	75.64	This study
160	1	6.02	69.16	24.35	74.71	70.35	24.24	76.31	This study
200	3	12.96	66.24	29.11	85.53	65.67	28.31	81.75	This study
160	5	12.94	75.8	25.47	85.63	75.40	25.55	86.03	This study
230	3	20.11	61.91	30.7	84.30	63.12	30.76	85.96	This study
250	3	20.07	58.4	31.88	82.57	58.40	32.51	84.77	This study
220	1	10	58.79	29.03	75.71	58.77	29.39	76.15	This study
240	1	10	51.94	31.06	71.56	54.07	31.13	74.99	This study
200	3	20.02	70.21	28.13	87.61	70.11	28.15	87.62	This study
220	3	20.04	64.89	30.07	86.55	65.43	29.89	86.48	This study
230	3	20.1	62.73	31.03	86.36	63.11	30.76	85.95	This study
170	1	30	91	25	101.01	83.06	24.58	95.67	[16]
200	1	30	75	27	88.5	76.02	27.19	93.93	[16]
220	1	30	62	29	78.74	71.33	28.94	92.77	[16]
250	1	30	55	33	80.85	64.28	31.55	91.04	[16]
150	0.5	16.67	81.39	23.54	83.93	79.30	22.95	85.24	[17]
150	0.5	9.09	72	23.85	75.99	74.54	23.12	78.95	[17]
150	0.5	6.25	70	23.98	72.39	72.75	23.18	76.59	[17]
200	3	14.29	80	26	84	66.50	28.28	82.85	[27]
220	3	14.69	75	28	87	62.06	30.01	82.04	[27]
240	3	15.09	71	32.33	93	57.62	31.75	81.21	[27]
200	1	20	73.3	28.5	95.2	69.74	27.42	85.62	[56]
200	3	20	73.8	28.7	96.2	70.09	28.15	87.60	[56]
230	1	20	62.8	30.1	86.1	62.70	30.03	83.88	[56]
230	3	20	62.7	31.6	90.3	63.05	30.76	85.87	[56]
260	1	20	57	32.4	84.2	55.66	32.65	82.15	[56]
260	3	20	54.9	33.2	83	56.01	33.38	84.13	[56]

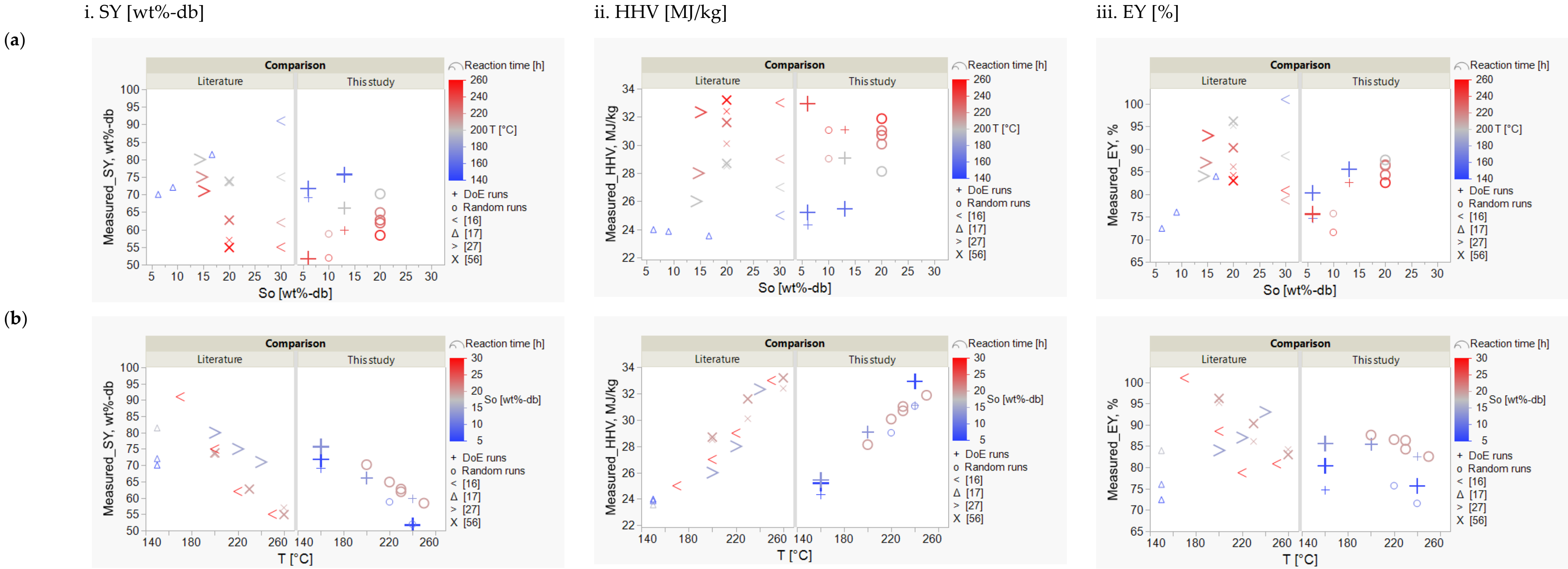
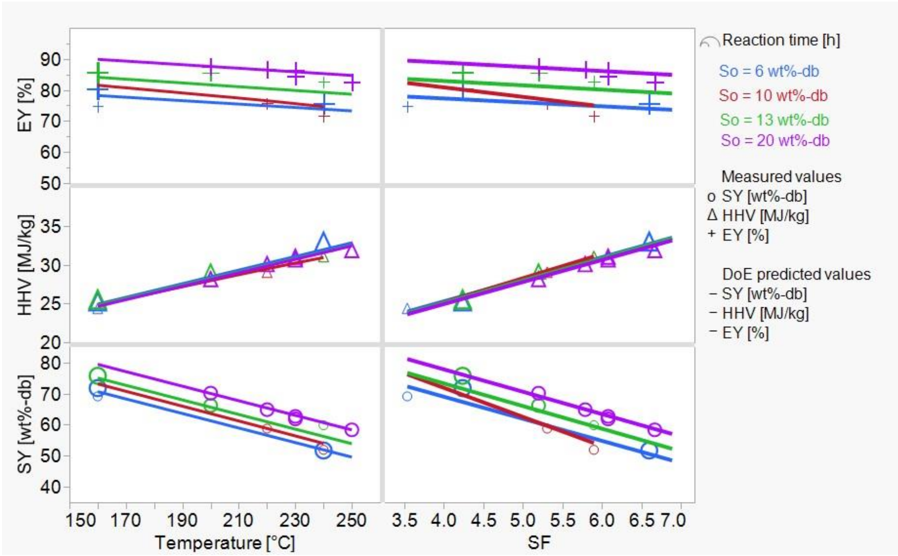


Figure S3. (**a**) Comparison of HTC outputs (i. SY [wt%-db], ii. HHV [MJ/kg], and iii. EY [%]) vs %*So* between literature and this study (**b**) Comparison of HTC outputs (i. SY [wt%-db], ii. HHV [MJ/kg], and iii. EY [%]) vs *T* between literature and this study.



(**a**) (**b**)

Figure S4. Comparison of measured and predicted (DoE model) values for the HTC outputs vs. (**a**) *T* and (b) severity factor (SF) for the three process parameters *T*, *t*, *%So*.

Table S3. Elemental analysis and atomatic ratios of hydrochars (HC) from both DoE and Random set-up runs.

SCG and its HC	C [wt%-db]	H [wt%-db]	N [wt%-db]	S [wt%-db]	O [wt%-db]	H/C [-]	O/C [-]
SCG	55.65 ± 0.035	6.27 ± 0.010	2.49 ± 0.007	0.17 ± 0.021	35.42 ± 0.078	1.35 ± 0.002	0.40 ± 0.001
From DoE runs							
HC_160_1_6%	57.98 ± 0.184	5.70 ± 0.078	2.19 ± 0.031	0.34 ± 0.019	33.78 ± 0.195	1.18 ± 0.017	0.44 ± 0.001
HC_160_5_6%	59.88 ± 0.049	5.89 ± 0.174	2.16 ± 0.129	0.23 ± 0.010	31.84 ± 0.168	1.19 ± 0.035	0.41 ± 0.003
HC_160_5_13%	58.54 ± 0.007	5.56 ± 0.133	2.38 ± 0.259	0.29 ± 0.006	33.24 ± 0.008	1.14 ± 0.027	0.43 ± 0.0001
HC_200_3_13%	69.09 ± 0.087	5.62 ± 0.030	2.85 ± 0.020	0.26 ± 0.005	22.18 ± 0.103	0.98 ± 0.005	0.24 ± 0.001
HC_240_1_13%	73.34 ± 0.166	5.55 ± 0.106	3.06 ± 0.035	0.27 ± 0.007	17.78 ± 0.088	0.91 ± 0.017	0.18 ± 0.001
HC_240_5_6%	77.42 ± 0.076	6.13 ± 0.005	3.10 ± 0.018	0.26 ± 0.010	13.09 ± 0.082	0.95 ± 0.038	0.13 ± 0.001
From Random runs							
R_230_3_20%	73.60 ± 0.099	5.49 ± 0.007	3.24 ± 0.034	0.24 ± 0.013	17.43 ± 0.120	0.89 ± 0.002	0.18 ± 0.001
R_250_3_20%	75.71 ± 0.148	5.95 ± 0.141	3.32 ± 0.046	0.21 ± 0.023	14.81 ± 0.006	0.94 ± 0.022	0.15 ± 0.001

R_220_1_10%	63.58 ± 0.014	6.97 ± 0.701	2.59 ± 0.010	0.00 ± 0.00	26.86 ± 0.014	1.32 ± 0.013	0.32 ± 0.0002
R_240_1_10%	66.11 ± 0.021	6.73 ± 0.014	2.98 ± 0.035	0.00 ± 0.00	24.17 ± 0.085	1.22 ± 0.003	0.27 ± 0.001
R_200_3_20%	68.95 ± 0.141	5.76 ± 0.075	3.03 ± 0.007	0.20 ± 0.010	20.82 ± 0.202	1.00 ± 0.013	0.27 ± 0.002
R_220_3_20%	72.42 ± 0.049	6.36 ± 0.136	3.14 ± 0.054	0.21 ± 0.013	16.31 ± 0.136	1.05 ± 0.023	0.22 ± 0.001
R_230_3_20%	73.76 ± 0.035	5.95 ± 0.102	3.22 ± 0.015	0.22 ± 0.010	15.49 ± 0.200	0.97 ± 0.017	0.19 ± 0.002

Table S4. The emission factor of trace elements of SCG and its HC from 6 DoE set-up runs and 5 Random set-up runs.

SCG and its HC	Cd [mg/MJ]	Cr [mg/MJ]	Cu [mg/MJ]	Ni [mg/MJ]	Pb [mg/MJ]	Zn [mg/MJ]
SCG	0.0009 ± 8.97*10^{-5}	n.d.	1.0051 ± 5.45*10^{-3}	0.0758 ± 8.32*10^{-4}	0.0199 ± 3.27*10^{-3}	0.9709 ± 3.02*10^{-2}
From DoE runs						
HC_160_1_6%	n.d.	0.0089 ± 6.75*10^{-3}	0.3224 ± 1.02*10^{-3}	0.0205 ± 2.46*10^{-2}	n.d.	0.1826 ± 1.59*10^{-3}
HC_160_5_6%	n.d.	n.d.	0.2857 ± 2.52*10^{-3}	0.0039 ± 2.54*10^{-4}	n.d.	0.1315 ± 1.48*10^{-3}
HC_160_5_13%	n.d.	n.d.	0.1361 ± 1.92*10^{-1}	0.0028 ± 3.95*10^{-3}	n.d.	0.1457 ± 9.67*10^{-3}
HC_200_3_13%	n.d.	0.0074 ± 5.65*10^{-3}	0.2698 ± 8.54*10^{-4}	0.0172 ± 2.06*10^{-2}	n.d.	0.1528 ± 1.33*10^{-3}
HC_240_1_13%	n.d.	n.d.	0.3228 ± 1.17*10^{-3}	0.0083 ± 2.61*10^{-3}	n.d.	0.1585 ± 6.53*10^{-5}
HC_240_5_6%	n.d.	n.d.	0.3965 ± 1.41*10^{-3}	0.0188 ± 2.94*10^{-3}	n.d.	0.1854 ± 6.40*10^{-4}
From Random runs						
R_230_3_20%	n.d.	0.0245 ± 7.80*10^{-4}	0.0978 ± 5.48*10^{-4}	0.0072 ± 5.75*10^{-4}	n.d.	0.0329 ± 1.43*10^{-4}
R_250_3_20%	n.d.	0.0621 ± 3.44*10^{-3}	0.1406 ± 8.16*10^{-4}	0.0322 ± 1.13*10^{-3}	n.d.	0.0518 ± 1.10*10^{-3}
R_200_3_20%	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
R_220_3_20%	n.d.	n.d.	0.0921 ± 2.27*10^{-3}	0.0132 ± 1.67*10^{-5}	0.0007 ± 8.10*10^{-4}	0.0080 ± 3.69*10^{-3}
R_230_3_20%	0.0001 ± 3.19*10^{-6}	0.0036 ± 2.12*10^{-5}	0.0988 ± 7.92*10^{-4}	0.0031 ± 5.96*10^{-6}	n.d.	0.0482 ± 3.32*10^{-3}

[1] detection limit of the analytical method for concentration of those elements in μg/L: Cd < 0.008; Cr < 0.02; Cu < 0.08; Ni < 0.04; Pb < 0.2; Zn < 0.01; [2] n.d. = not detected.

Table S5. Production of aromatic compounds in hydrochars (HC) and process water (PW) during HTC conversion using SCG.

SCG and its HC	HMF [mg/kg TS]	Furfural [mg/kg TS]	Phenol [mg/kg TS]	Cresol [mg/kg TS]	Catechol [mg/kg TS]	Guaiacol [mg/kg TS]
SCG	n.d.	243.47	n.d.	n.d.	n.d.	n.d.
HC_160_5_13%	4181.36	n.d.	n.d.	n.d.	1448.82	n.d.
HC_240_1_13%	133.78	916.92	2061.83	n.d.	1730.22	1114.95
HC_200_3_13%	1689.17	n.d.	915.52	n.d.	1009.02	380.35
HC_240_5_6%	n.d.	n.d.	3690.81	1032.01	1207.37	1007.04
HC_160_5_6%	2474.52	2193.23	n.d.	n.d.	1246.70	1092.62
HC_160_1_6%	409.31	n.d.	n.d.	n.d.	n.d.	n.d.
PW	**HMF [mg/L]**	**Furfural [mg/L]**	**Phenol [mg/L]**	**Cresol [mg/L]**	**Catechol [mg/L]**	**Guaiacol [mg/L]**
PW_160_5_13%	938.68	243.28	n.d.	n.d.	n.d.	n.d.
PW_240_1_13%	15.70	n.d.	241.66	n.d.	213.81	n.d.
PW_200_3_13%	376.26	49.14	101.87	n.d.	149.65	n.d.
PW_240_5_6%	n.d.	n.d.	286.80	n.d.	163.33	n.d.
PW_160_5_6%	489.54	253.55	n.d.	n.d.	n.d.	n.d.
PW_160_1_6%	80.24	94.70	n.d.	n.d.	n.d.	n.d.

[1] detection limit (mg/kg or mg/l) of the analytical instrument: HMF < 0.004; Furfural < 0.0124; Phenol < 0.079; Cresol < 0.058; Catechol < 0.027; Guaiacol < 0.008; [2] n.d. = not detected.

Table S6. Model and parameters from Borrero-López et al. (2018) were used in this study to predict the production of phenolic compounds from the HTC of SCG feedstock.

Aromatics Compounds	δ	B	κ
Guaiacol	0.014	2.87E-05	1.35
Phenol	-6.17E-04	2.27E-06	1.61
Cresol	1.65E-03	1.63E-13	3.58

$$C_{phenolic\ compound} = \delta + B \cdot e^{(\kappa(\log R_0))} \qquad (S1)$$

Table S7. Change in aromatic compounds of unwashed hydrochars (HC), washed hydrochars (wHC) from 6 DoE set-up runs.

HC and wHC	HMF [mg/kg TS]	Furfural [mg/kg TS]	Phenol [mg/kg TS]	Cresol [mg/kg TS]	Catechol [mg/kg TS]	Guaiacol [mg/kg TS]
HC_160_1_6%	409.31	n.d.	n.d.	n.d.	n.d.	n.d.
wHC1_160_1_6%	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
wHC2_160_1_6%	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
HC_160_5_6%	2474.52	2193.23	n.d.	n.d.	1246.70	1092.62
wHC1_160_5_6%	604.25	1225.67	n.d.	n.d.	n.d.	n.d.
wHC2_160_5_6%	233.73	1208.85	n.d.	n.d.	n.d.	n.d.
HC_160_5_13%	4181.36	n.d.	539.55	n.d.	1448.82	n.d.
wHC1_160_5_13%	1489.44	n.d.	579.71	n.d.	1139.96	n.d.
wHC2_160_5_13%	574.97	n.d.	n.d.	n.d.	1017.03	n.d.
HC_200_3_13%	1689.17	n.d.	915.51	n.d.	1009.02	380.35
wHC1_200_3_13%	705.65	n.d.	932.11	n.d.	962.10	380.42
wHC2_200_3_13%	392.28	n.d.	826.07	n.d.	962.27	334.36
HC_240_1_13%	133.78	916.92	2061.83	n.d.	1730.22	1114.95
wHC1_240_1_13%	115.48	1006.06	2464.96	n.d.	1092.44	1195.98
wHC2_240_1_13%	n.d.	n.d.	2425.52	1254.18	948.29	1147.40
HC_240_5_6%	n.d.	n.d.	3690.81	1032.01	1207.37	1007.04
wHC1_240_5_6%	n.d.	n.d.	2589.05	968.17	782.86	906.95
wHC2_240_5_6%	n.d.	n.d.	2484.17	1082.70	n.d.	1002.36

PW and WW	HMF [mg/L]	Furfural [mg/L]	Phenol [mg/L]	Cresol [mg/L]	Catechol [mg/L]	Guaiacol [mg/L]
PW_160_1_6%	80.24	94.70	n.d.	n.d.	n.d.	n.d.
WW1_160_1_6%	74.87	32.37	n.d.	n.d.	n.d.	n.d.
WW2_160_1_6%	n.d.	n.d.	n.d.	31.47	n.d.	n.d.

PW_160_5_6%	489.54	253.55	n.d.	n.d.	n.d.	n.d.
WW1_160_5_6%	96.01	34.21	n.d.	n.d.	n.d.	n.d.
WW2_160_5_6%	12.58	n.d.	n.d.	n.d.	n.d.	n.d.
PW_160_5_13%	938.68	243.28	n.d.	n.d.	n.d.	n.d.
WW1_160_5_13%	306.27	101.24	n.d.	n.d.	n.d.	n.d.
WW2_160_5_13%	88.75	n.d.	n.d.	n.d.	n.d.	n.d.
PW_200_3_13%	376.26	49.14	101.87	n.d.	149.65	n.d.
WW1_200_3_13%	115.02	32.14	61.65	n.d.	93.54	n.d.
WW2_200_3_13%	40.88	27.72	45.50	n.d.	74.16	n.d.
PW_240_1_13%	15.70	n.d.	241.66	n.d.	213.81	n.d.
WW1_240_1_13%	13.34	n.d.	116.64	n.d.	97.85	n.d.
WW2_240_1_13%	12.47	n.d.	68.67	32.57	69.32	n.d.
PW_240_5_6%	376.26	49.14	101.87	n.d.	149.65	n.d.
WW1_240_5_6%	115.02	32.14	61.65	n.d.	93.54	n.d.
WW2_240_5_6%	40.88	27.72	45.50	n.d.	74.16	n.d.

[1] detection limit (mg/kg or mg/l) of the analytical method: HMF < 0.004; Furfural < 0.0124; Phenol < 0.079; Cresol < 0.058; Catechol < 0.027; Guaiacol < 0.008; [2] n.d. = not detected.

Table S8. Elemental analysis, ash and HHV of unwashed hydrochars (HC) and washed hydrochars (wHC) from 6 DoE set-up runs.

HC and wHC	C [wt%-db]	H [wt%-db]	N [wt%-db]	S [wt%-db]	O [wt%-db]	Ash [wt%-db]	HHV [MJ/kg]
HC_160_1_6%	57.98 ± 0.184	5.70 ± 0.078	2.19 ± 0.031	0.34 ± 0.019	33.78 ± 0.382	0.30 ± 0.006	24.35 ± 0.039
wHC1_160_1_6%	58.30 ± 0.113	5.86 ± 0.013	2.00 ± 0.025	0.30 ± 0.019	33.54 ± 0.171	0.30 ± 0.011	24.63 ± 0.021
wHC2_160_1_6%	58.63 ± 0.163	5.80 ± 0.057	1.99 ± 0.016	0.28 ± 0.005	33.30 ± 0.163	0.29 ± 0.015	24.49 ± 0.077
HC_160_5_6%	59.88 ± 0.049	5.89 ± 0.174	2.16 ± 0.129	0.23 ± 0.010	31.84 ± 0.623	0.22 ± 0.002	25.22 ± 0.047
wHC1_160_5_6%	60.44 ± 0.085	6.21 ± 0.108	2.01 ± 0.001	0.22 ± 0.009	31.12 ± 0.013	0.22 ± 0.002	25.77 ± 0.017
wHC2_160_5_6%	60.73 ± 0.028	6.28 ± 0.191	2.02 ± 0.031	0.21 ± 0.008	30.76 ± 0.242	0.22 ± 0.002	25.76 ± 0.120
HC_160_5_13%	58.54 ± 0.007	5.56 ± 0.133	2.38 ± 0.259	0.29 ± 0.006	33.24 ± 2.249	0.30 ± 0.019	25.47 ± 0.068
wHC1_160_5_13%	60.58 ± 0.049	5.76 ± 0.062	2.14 ± 0.045	0.28 ± 0.014	31.24 ± 0.173	0.29 ± 0.003	25.96 ± 0.089
wHC2_160_5_13%	61.08 ± 0.035	5.84 ± 0.074	2.01 ± 0.026	0.26 ± 0.007	30.81 ± 0.131	0.29 ± 0.001	25.71 ± 0.062
HC_200_3_13%	69.09 ± 0.087	5.62 ± 0.030	2.85 ± 0.020	0.26 ± 0.005	22.18 ± 0.323	0.29 ± 0.009	29.11 ± 0.060
wHC1_200_3_13%	70.19 ± 0.028	5.47 ± 0.059	2.75 ± 0.017	0.27 ± 0.010	21.31 ± 0.349	0.30 ± 0.007	29.64 ± 0.024
wHC2_200_3_13%	70.52 ± 0.057	5.41 ± 0.101	2.70 ± 0.017	0.27 ± 0.009	21.10 ± 0.122	0.30 ± 0.002	29.70 ± 0.125
HC_240_1_13%	73.34 ± 0.166	5.55 ± 0.106	3.06 ± 0.035	0.27 ± 0.007	17.78 ± 0.088	0.29 ± 0.010	31.11 ± 0.050
wHC1_240_1_13%	74.37 ± 0.096	5.79 ± 0.116	3.01 ± 0.016	0.27 ± 0.013	16.55 ± 0.044	0.29 ± 0.002	31.66 ± 0.065
wHC2_240_1_13%	74.79 ± 0.046	5.60 ± 0.187	2.94 ± 0.037	0.27 ± 0.012	16.39 ± 0.217	0.30 ± 0.008	31.78 ± 0.031
HC_240_5_6%	77.42 ± 0.076	6.13 ± 0.005	3.10 ± 0.018	0.26 ± 0.010	13.09 ± 0.229	0.21 ± 0.004	32.97 ± 0.057
wHC1_240_5_6%	78.11 ± 0.028	6.57 ± 0.191	3.25 ± 0.021	0.27 ± 0.018	12.50 ± 0.178	0.29 ± 0.003	33.30 ± 0.063
wHC2_240_5_6%	78.15 ± 0.067	6.02 ± 0.127	3.02 ± 0.025	0.26 ± 0.012	12.55 ± 0.307	0.28 ± 0.002	33.36 ± 0.027

Table S9. Cumulative distribution Q_3 of final spherical pellets from both SCG and its hydrochars after the agglomeration.

Q3 [wt%-db]	Exp. 1 (SCG_M)	Exp. 2 (SCG_gSM)	Exp. 3 (SCG_HM)	Exp. 4 (wHC160_M)	Exp. 5 (wHC200_St)	Exp. 6 (Mix_HC 200_St)	Exp. 7 (wHC240_HM)
0 [μm]	0.03 ± 0.00	n.d.	0.00 ± 0.00	0.02 ± 0.01	4.48 ± 0.45	18.23 ± 0.04	0.07 ± 0.02
100 [μm]	0.08 ± 0.02	0.29 ± 0.01	0.00 ± 0.00	0.15 ± 0.03	0.00 ± 0.00	0.00 ± 0.00	0.18 ± 0.01
200 [μm]	1.71 ± 0.63	1.11 ± 0.40	2.58 ± 0.22	0.59 ± 0.00	0.00 ± 0.00	0.00 ± 0.00	0.45 ± 0.02
400 [μm]	3.32 ± 1.07	2.03 ± 0.78	4.79 ± 0.33	0.92 ± 0.01	0.00 ± 0.00	0.00 ± 0.00	0.55 ± 0.03
500 [μm]	19.11 ± 2.13	17.55 ± 4.69	38.79 ± 1.04	2.92 ± 0.19	0.00 ± 0.00	0.00 ± 0.00	1.24 ± 0.05
1000 [μm]	74.17 ± 0.44	67.52 ± 3.74	94.45 ± 0.18	21.48 ± 0.13	7.78 ± 0.23	61.08 ± 0.15	3.37 ± 0.06
2000 [μm]	99.66 ± 0.43	96.45 ± 1.13	100.00 ± 0.00	77.83 ± 0.55	27.03 ± 0.36	96.22 ± 0.21	15.96 ± 0.76
4000 [μm]	100.00 ± 0.00	100.00 ± 0.00	100.00 ± 0.00	100.00 ± 0.00	100.00 ± 0.00	100.00 ± 0.00	100.00 ± 0.00

[*] n.d. = not detected.

Table S10. Results of bulk density and mechanical stability agglomerated spherical pellets.

Pellets (Spherical and Cylindrical)	Bulk Density [kg/m³]	Mechanical Stability [wt%-db]
sp_ SCG_gSM	335.81 ± 2.29	98.04 ± 0.00
sp_ wHC160_M	262.50 ± 1.91	98.18 ± 0.20
sp_ Mix_HC 200_St	341.27 ± 1.00	76.59 ± 1.49
sp_ wHC240_HM	300.55 ± 0.45	96.49 ± 0.62
cp_SCG	675.43 ± 2.91	77.76 ± 0.94
cp_HC_220	490.89 ± 1.96	92.11 ± 0.16
cp_wHC_220	502.20 ± 2.10	94.00 ± 0.22
cp_Wood	659.72 ± 2.03	99.45 ± 0.05
DIN EN ISO 17225-8:2023	≥ 600	≥ 96.00

[1] sp = spherical pellets; [2] cp= cylindrical pellets with size of 8x10 mm (Diameter x Length) without additives; [3] DIN EN ISO 17225-8 :2023 = standard for cylindrical pellets from thermally treated non-woody biomass.