

### Supporting information

#### **Closing the loop for minimizing generation of liquid waste: Recycling phosphorus species from solid residual in activation with H<sub>3</sub>PO<sub>4</sub> for next-round activating biomass**

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**Table S1** The relative abundance of light organics in the bio-oil produced from the pyrolysis of poplar wood at different temperatures

Compound	Relative abundance (%)			
	300°C	500°C	700°C	
<b>Acids</b>	Acetic acid	95.9	237.8	194.9
<b>Ketones</b>	1-hydroxy-2-Propanone	28.9	93.2	73.4
	1-(acetyloxy)-2-Butanone	14.3	0	0
	1-(acetyloxy)-2-Propanone	9.6	22.3	19.5
	1,2-Cyclopentanedione	10.3	0	31.6
	2-Cyclopenten-1-one, 2-hydroxy-3-methyl-	15.5	42.43	35.6
	2-Cyclopenten-1-one, 3-ethyl-2-hydroxy-	7.3	12.7	0
	Levogluosenone	11.5	35.5	35.7
	1-(4-hydroxy-3-methoxyphenyl)-2-Propanone	8.7	9.5	14.5
	3',5'-Dimethoxyacetophenone	0	0	16.7
	4-Hydroxy-2-methylacetophenone	0	14.3	15.9
	1-(4-hydroxy-3,5-dimethoxyphenyl)-Ethanone	5.1	0	9.1
	4-Heptanone	0	0	29.1
	Apocynin	0	0	5.0
	<b>Aldehydes</b>	Furfural	0	61.7
	2-ethyl-Butanal	0	0	20.4
<b>Alcohols</b>	2-Furanmethanol, tetrahydro-	22.9	26.3	0
	Cyclopropyl carbinol	58.1	80.5	97.5
	Butanol	0	0	131.2
<b>Esters</b>	Butyrolactone	8.6	28.8	23.8
	Hexanoic acid, 2-propenyl ester	13.3	0	0
	Propanoic acid, 2-oxo-, ethyl ester	0	0	29.1
<b>Ethers</b>	2,5-diethoxytetrahydro-Furan	23.2	7.4	0
	1,2,3-trimethoxy-5-methyl-Benzene	23.1	40.3	45.6
<b>Phenols</b>	Phenol	82.3	0	0
	2-methyl-Phenol	0	9.7	12.5
	p-Cresol	0	15.0	17.8
	2-methoxy-Phenol	22.1	56.0	51.5
	Creosol	7.8	42.3	39.7
	4-ethyl-2-methoxy-Phenol	8.3	24.4	26.6
	2-Methoxy-4-vinylphenol	2.7	0	0
	2,6-dimethoxy-Phenol	54.6	111.6	123.9
	2-methoxy-3-(2-propenyl)-Phenol	2.4	0	0
	3,5-Dimethoxy-4-hydroxytoluene	20.9	54.8	60.6
	3-methoxy-1,2-Benzenediol	0	24.6	27.1
	2,6-dimethoxy-4-(2-propenyl)-Phenol	0	7.0	9.8
	2-methoxy-5-(1-propenyl)-,	0	6.9	5.5

(E)-Phenol			
2-methoxy-4-(1-propenyl)- (Z)-Phenol	9.0	19.2	21.4
(E)-2,6-Dimethoxy-4-(prop-1- en-1-yl)phenol	28.0	30.2	28.4

**Table S2** The relative abundance of light organics in the bio-oil produced from the activation of poplar wood with fresh H<sub>3</sub>PO<sub>4</sub> and recycled P-containing species

Compound		Relative abundance (%)					
		H <sub>3</sub> PO <sub>4</sub> activation			Recycled P species		
		300°C	500°C	700°C	300°C	500°C	700°C
<b>Acids</b>	Acetic acid	31.6	57.0	40.4	14.8	27.0	19.7
<b>Alcohols</b>	1-Butanol	11.3	0	0	2.3	0	0
<b>Esters</b>	Ethyl levulinate	54.2	91.4	37.2	14.8	20.3	14.4
	Butanedioic acid, dimethyl ester	0	6.5	0	0	2.8	0
	2(3H)-Furanone, 5-(acetyloxy)dihydro-5-methyl-	0	0	7.5	0	0	0
	Diethyl phosphite	0	0	34.5	0	0	0

**Table S3** Average structural parameters of ACs from the X-ray diffraction profiles

Samples	Average structural parameters				
	$2\theta^a$ (°)	$B_{002}^b$ (°)	$d_{002}^c$ (nm)	$L_c^d$ (nm)	$N^e$
<b>Biochar-500</b>	23.43	7.30	0.3794	1.0993	3.8979
<b>Biochar-700</b>	23.35	7.03	0.3806	1.1417	3.9998
<b>AC-I-300</b>	23.03	8.03	0.3858	0.9984	3.5877
<b>AC-I-500</b>	24.59	5.47	0.3618	1.4703	5.0641
<b>AC-I-700</b>	23.63	7.56	0.3762	1.0619	3.8224
<b>AC-II-300</b>	22.97	8.62	0.3869	0.9300	3.4040
<b>AC-II-500</b>	24.50	6.45	0.3631	1.2459	4.4317
<b>AC-II-700</b>	23.36	7.85	0.3806	1.0220	3.6855

<sup>a</sup> $2\theta$ : diffraction angle;

<sup>b</sup> $B_{002}$ : half height width of the (002) peak;

<sup>c</sup> $d_{002}$ : interlayer spacing of biochar;

<sup>d</sup> $L_c$ : crystalline height of biochar;

<sup>e</sup> $N$  = crystalline stacks of AC.

**Table S4** Environmental impacts, energy consumption, and resource depletion for production of the ACs from activation of poplar with fresh H<sub>3</sub>PO<sub>4</sub> and recycled P-containing species

Factors	Fresh H <sub>3</sub> PO <sub>4</sub>			Recycled P species		
	300°C	500°C	700°C	300°C	500°C	700°C
<b>Acidification (kg SO<sub>2</sub> eq)</b>	7.56E+04	1.18E+05	1.60E+05	5.83E+04	9.16E+04	1.07E+05
<b>Carcinogenics (CTUh)</b>	5.04E+01	7.84E+01	1.06E+02	3.89E+01	6.11E+01	7.14E+01
<b>Ecotoxicity (CTUe)</b>	7.38E+08	1.15E+09	1.56E+09	5.69E+08	8.93E+08	1.04E+09
<b>Eutrophication (kg N eq)</b>	5.95E+04	9.13E+04	1.23E+05	4.53E+04	7.06E+04	8.22E+04
<b>Fossil fuel depletion (MJ surplus)</b>	1.32E+07	2.06E+07	2.79E+07	1.02E+07	1.60E+07	1.87E+07
<b>Global warming (kg CO<sub>2</sub> eq)</b>	1.82E+07	2.83E+07	3.84E+07	1.40E+07	2.20E+07	2.57E+07
<b>Non carcinogenics (CTUh)</b>	1.82E+01	2.84E+01	3.85E+01	1.41E+01	2.21E+01	2.58E+01
<b>Ozone depletion (kg CFC-11 eq)</b>	1.10E+00	1.71E+00	2.32E+00	8.50E-01	1.33E+00	1.56E+00
<b>Respiratory effects (kg PM<sub>2.5</sub> eq)</b>	2.63E+04	4.09E+04	5.55E+04	2.03E+04	3.19E+04	3.72E+04
<b>Smog (kg O<sub>3</sub> eq)</b>	1.07E+06	1.66E+06	2.26E+06	8.26E+05	1.30E+06	1.51E+06

**Table S5** Input and output of production of AC using fresh H<sub>3</sub>PO<sub>4</sub> as the activator, based on 2 tons of poplar

Input/Output	Fresh H <sub>3</sub> PO <sub>4</sub>			Unit
	300°C	500°C	700°C	
Quantity				
<b>Step 1: Transportation</b>				
<b>Input</b>				
Feedstock	2000.0	2000.0	2000.0	kg
transportation	2.0	2.0	2.0	tkm
<b>Output</b>				
Feedstock after transportation	1980.0	1980.0	1980.0	kg
<b>Step 2: Pretreatment</b>				
<b>Input</b>				
Feedstock after transportation	1980.0	1980.0	1980.0	kg
Water	5940.0	5940.0	5940.0	kg
Electricity	2851.2	2851.2	2851.2	MJ
<b>Output</b>				
Feedstock after pretreatment	1960.2	1960.2	1960.2	kg
Waste water	5940.0	5940.0	5940.0	kg
<b>Step 3: Impregnation</b>				
<b>Input</b>				
Feedstock after pretreatment	1960.2	1960.2	1960.2	kg
H <sub>3</sub> PO <sub>4</sub>	3920.4	3920.4	3920.4	kg
Water	17641.8	17641.8	17641.8	kg
Electricity	2822.7	2822.7	2822.7	MJ
<b>Output</b>				

<b>Feedstock after impregnation</b>	5880.6	5880.6	5880.6	kg
<b>Waste water</b>	17641.8	17641.8	17641.8	kg

#### Step 4: Activation

Input				
<b>Feedstock after impregnation</b>	5880.6	5880.6	5880.6	kg
<b>Natural gas</b>	1475.1	2294.6	3114.1	m <sup>3</sup>
<b>Electricity</b>	31.8	38.8	45.9	MJ
Output				
<b>Mixture</b>	4839.7	4287.0	2740.4	kg
<b>Bio-oil</b>	623.3	1311.4	2458.1	kg
<b>H<sub>2</sub></b>	0	2.9	42.3	kg
<b>CO</b>	88.2	58.8	241.1	kg
<b>CO<sub>2</sub></b>	329.3	217.6	376.4	kg
<b>CH<sub>4</sub></b>	0	5.3	17.1	kg

#### Step 5: Post-treatment

Input				
<b>Mixture</b>	4839.7	4287.0	2740.4	kg
<b>Water</b>	14519.2	12860.9	8221.1	kg
<b>Electricity</b>	5207.9	4411.9	3996.9	MJ
Output				
<b>AC</b>	3616.6	3063.8	2775.6	kg
<b>Waste water</b>	14519.2	12860.9	8221.1	kg

**Table S6** Input and output of production of AC using recycled P-containing species as the activator, based on 1 ton of poplar

Input/Output	Recycled P species			Unit
	300°C	500°C	700°C	
	Quantity			
<b>Step 1: Transportation</b>				
<b>Input</b>				
Feedstock	1000.0	1000.0	1000.0	kg
transportation	1.0	1.0	1.0	tkm
<b>Output</b>				
Feedstock after transportation	990.0	990.0	990.0	kg
<b>Step 2: Pretreatment</b>				
<b>Input</b>				
Feedstock after transportation	990.0	990.0	990.0	kg
Water	2970.0	2970.0	2970.0	kg
Electricity	1425.6	1425.6	1425.6	MJ
<b>Output</b>				
Feedstock after pretreatment	980.1	980.1	980.1	kg
Waste water	2970.0	2970.0	2970.0	kg
<b>Step 3: Impregnation</b>				
<b>Input</b>				
Feedstock after pretreatment	1960.2	1960.2	1960.2	kg
H <sub>3</sub> PO <sub>4</sub>	1960.2	1960.2	1960.2	kg
Water	8820.9	8820.9	8820.9	kg
Electricity	1411.3	1411.3	1411.3	MJ
<b>Output</b>				

<b>Feedstock after impregnation</b>	2940.3	2940.3	2940.3	kg
<b>Waste water</b>	8820.9	8820.9	8820.9	kg

#### Step 4: Activation

<b>Input</b>				
<b>Feedstock after impregnation</b>	2940.3	2940.3	2940.3	kg
<b>Natural gas</b>	737.5	1147.3	1557.0	m <sup>3</sup>
<b>Electricity</b>	15.9	19.4	22.9	MJ
<b>Output</b>				
<b>Mixture</b>	2419.9	2143.5	1370.2	kg
<b>Bio-oil</b>	311.7	655.7	1229.0	kg
<b>H<sub>2</sub></b>	0	1.5	21.2	kg
<b>CO</b>	44.1	29.4	120.6	kg
<b>CO<sub>2</sub></b>	164.7	108.8	188.2	kg
<b>CH<sub>4</sub></b>	0	2.6	8.5	kg

#### Step 5: Post-treatment

<b>Input</b>				
<b>Mixture</b>	2419.9	2143.5	1370.2	kg
<b>Water</b>	7259.6	6430.4	4110.5	kg
<b>Electricity</b>	2603.9	2205.9	1998.5	MJ
<b>Output</b>				
<b>AC</b>	1808.3	1531.9	1387.8	kg
<b>Washing filtrate</b>	7259.6	6430.4	4110.5	kg

#### Step 6: Transportation

<b>Input</b>				
<b>Feedstock</b>	1000.0	1000.0	1000.0	kg

<b>transportation</b>	1.0	1.0	1.0	tkm
<b>Output</b>				
<b>Feedstock after transportation</b>	990.0	990.0	990.0	kg

#### Step 7: Pretreatment

<b>Input</b>				
<b>Feedstock after transportation</b>	990.0	990.0	990.0	kg
<b>Water</b>	2970.0	2970.0	2970.0	kg
<b>Electricity</b>	1425.6	1425.6	1425.6	MJ
<b>Output</b>				
<b>Feedstock after pretreatment</b>	980.1	980.1	980.1	kg
<b>Waste water</b>	2970.0	2970.0	2970.0	kg

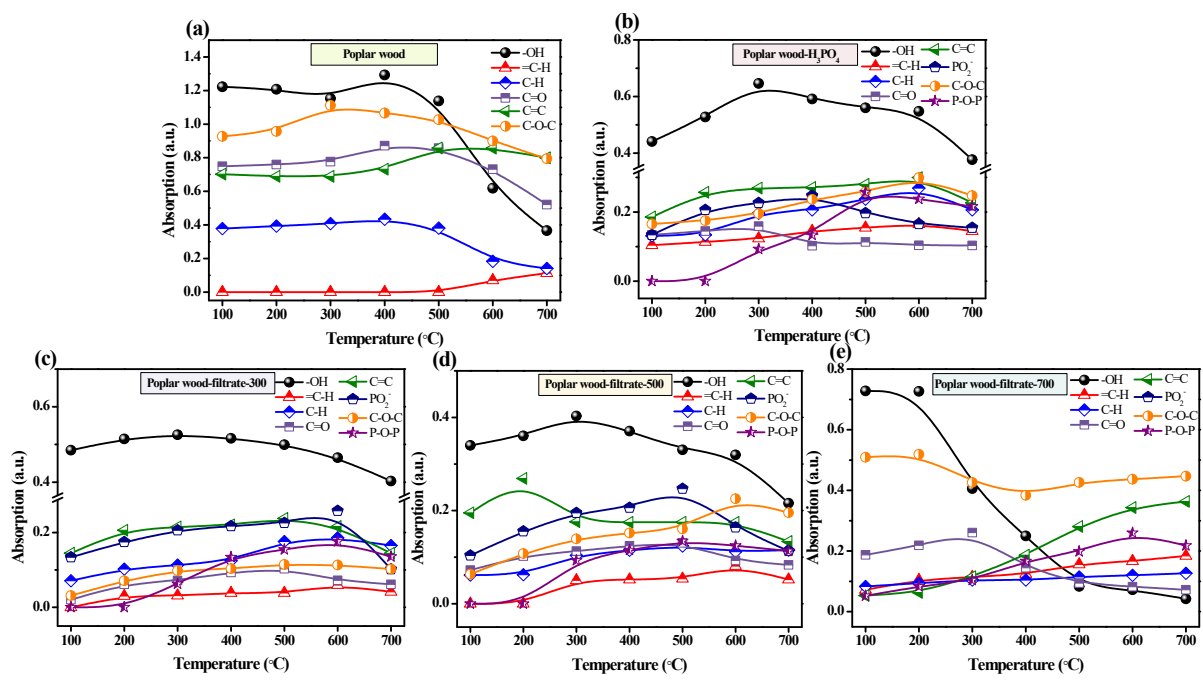
#### Step 8: Impregnation

<b>Input</b>				
<b>Feedstock after pretreatment</b>	1960.2	1960.2	1960.2	kg
<b>Washing filtrate</b>	7259.6	6430.4	4110.5	kg
<b>Electricity</b>	1411.3	1411.3	1411.3	MJ
<b>Output</b>				
<b>Feedstock after impregnation</b>	1597.6	1641.7	1004.6	kg

#### Step 9: Activation

<b>Input</b>				
<b>Feedstock after impregnation</b>	1597.6	1641.7	1004.6	kg
<b>Natural gas</b>	400.7	640.6	532.0	m <sup>3</sup>
<b>Electricity</b>	8.6	10.8	7.8	MJ
<b>Output</b>				
<b>Mixture</b>	1263.7	1170.5	381.7	kg

<b>Bio-oil</b>	156.6	170.7	500.3	kg
<b>H<sub>2</sub></b>	0	6.2	6.6	kg
<b>CO</b>	52.7	64.0	55.3	kg
<b>CO<sub>2</sub></b>	124.6	221.6	59.3	kg
<b>CH<sub>4</sub></b>	0.3	2.3	1.4	kg
<b>Step 10: Post-treatment</b>				
<b>Input</b>				
<b>Mixture</b>	1263.7	1170.5	381.7	kg
<b>Water</b>	3791.0	3511.5	1145.2	kg
<b>Electricity</b>	1035.2	924.3	441.2	MJ
<b>Output</b>				
<b>AC</b>	718.9	641.9	306.4	kg
<b>Wastewater</b>	3791.0	3511.5	1145.2	kg



**Figure S1** Evolution of functionalities in *in-situ* DRIFTS spectra of the pyrolysis and activation of poplar with various activators. (a) poplar pyrolysis; (b) poplar activation with fresh H<sub>3</sub>PO<sub>4</sub>; (c) poplar activation with filtrate-300; (d) poplar activation with filtrate-500; (e) poplar activation with filtrate-700.