†**Electronic Supplementary Information**

Prediction of microalgae hydrothermal liquefaction products from feedstock biochemical composition

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Table of Abbreviations

%afdw	Percent ash free dry weight
%dw	Percent dry weight
BP	Boiling point
C14:0	Myristic acid; tetradecanoic acid
C16:0	Palmitic acid; hexadecanoic acid
C16:1	Palmitoleic acid; 9-hexadecenoic acid
C18:1	Oleic acid; 9-octadecenoic acid
C20:3n3	Eicosatrienoic acid
C20:3n5	Eicosapentaenoic acid
ECR	Energy consumption ratio
ER%	Energy recovery percentage
ESI	Electronic Supplementary Information
FA	Fatty acids
FAMEs	Fatty acid methyl ester(s)
FA model	Fatty acid model
HHV	Higher heating value
HTL	Hydrothermal liquefaction
LCA	Life cycle assessment
MUFAs	Mono-unsaturated fatty acid(s)
MW	Molecular weight
NER	Net energy return(s)
NL	Neutral lipid
NL/PL	Ratio of neutral lipids to polar lipids
PBR	Photobioreactor
PL	Polar lipid
PL/Prot	Ratio of polar lipids to proteins
PPM	Phototrophic Process Model
PUFAs	Poly-unsaturated fatty acid(s)
r^2	Coefficient of determination
SAFAs	Saturated fatty acid(s)
SA/MUFAs	Saturated and mono-unsaturated fatty acid(s)
SEC	Size exclusion chromatography
SimDist	Simulated Distillation
SPE	Solid phase extraction
TAGs	Triacylglyceride(s)
TEA	Techno-economic analysis
TKN	Total Kjeldahl nitrogen
TOC	Total organic carbon
TN	Total nitrogen
wt%	Percent weight

Specific to the ESI

DCM	Dichloromethane
DI	Deionized (water)

ESI-1. Photobioreactor operating conditions and biomass harvesting

The PBR was UV-sterilized prior to adding 3.5 L of autoclaved modified f2 growth media following the recipe of Guillard and Ryther,[1] with silica omitted and additional phosphate and nitrate added (0.03 g/L $NaH_2PO_4 \cdot H_2O$ and 0.5 g/L $NaNO_3$). Starter cultures of *Nannochloropsis oculata* (strain CCMP525) in L1-Si media obtained from the National Center for Marine Algae and Microbiota (East Boothbay, ME) were used to inoculate the PBRs immediately upon arrival of cultures. Interspaced blue and red LED lights mounted on a wall parallel to the reactor provided continuous illumination of 250 $\mu E/m^2 \cdot s$ at the surface of the reactor. Temperature was maintained at 18 °C with a circulating water bath. A diffuser at the bottom of the reactor provided 2 L/min of ambient air filtered through a HEPA filter for aeration and mixing. The culture was maintained at pH 7.8–8.2 with a pH controller and solenoid valve which delivered pure $CO_{2(g)}$ through the diffuser as necessary to reduce pH. Cells were grown in batch mode under axenic conditions and checked for contamination via light microscopy. To harvest biomass, a portion of the reactor suspension was removed, centrifuged at $6,300 \times g$ for 15 min (Sorvall RC 6+), and supernatant was decanted. To remove residual salts from the culture media, biomass pellets were twice rinsed with deionized (DI) water (>18 M$\Omega \cdot$cm) and centrifuged at $6,300 \times g$ (Eppendorf 5810R) for 15 min. To preserve the biochemical composition during storage prior to feedstock analysis and HTL processing, the wet harvested biomass was lyophilized (Model 77500 Freeze Dry System, FreeZone) and the resulting solid was ground and homogenized with a mortar and pestle and stored at 4 °C.

ESI-2. Sulfur analysis of batches

In order to supplement the assumption of negligible sulfur content in the *Nannochloropsis* batches based on previous reports from Rebolloso-Fuentes et al.[2] and Brown et al.[3] (who both reported ≤0.65 %dw and 0.5 %dw, respectively), additional analysis for sulfur content was conducted for four representative batches of *Nannochloropsis* – the batch purchased from Reed Mariculture (Batch 2), a low lipid batch (Batch 3), a mid lipid batch (Batch 4) and a high lipid batch (Batch 8) – via ICP-MS at the University of Illinois Microanalysis Laboratory (Urbana, IL) using a PerkinElmer SCIEX ELAN DRCe ICP-MS. The results are summarized below and support the assumption of negligible sulfur content in the representative batches.

Batch number	Lipid content (%dw)	Sulfur content (%dw)
2	23.0	0.11
3	30.7	0.10
4	46.8	0.07
8	58.7	0.04

ESI-3. Method of crude lipid fractionation via SPE

SPE fractionation was performed directly on crude lipids extracted via the Folch method.[4] The NL fraction is defined as the mix of compounds in the crude lipid extract with polarity less than or similar to triacylglycerides (TAGs),[5–7] and includes compounds such as sterols, waxes, and carotenoids in addition to the TAGs. The PL fraction is defined as the mix of compounds with polarity greater than TAGs, and includes phospholipids, sphingolipids, and glycophospholipids.[8,9] The SPE cartridge (Waters Sep-Pak cyanopropyl vac cartridges) was first conditioned with 10 mL of n-hexane and then loaded with approximately 30 mg crude lipids dissolved in 2 mL n-hexane. Elution with 8 mL of 9:1 hexane-diethyl ether provided the NL fraction, which appeared as a yellowish oil-like substance after solvent removal. Subsequent elution with 8 mL of 2:1 chloroform-methanol followed by 4 mL of methanol yielded the PL fraction, which was a deep-green substance after solvent removal, indicating the presence of pigments and dyes such as chlorophyll. It is noted here that efforts have been made to reduce (minimizing volume used) and reuse (recovering solvents when applicable) the use of undesirable solvents (as defined by Alfonsi et al.[10]) in order to adhere to the principles of green chemistry.

ESI-4. FAMEs analysis

Fatty acid profiles of the biomass were determined by in-situ direct transesterification (transesterification was conducted on dried biomass) fatty acid methyl ester (FAMEs) analysis according to Laurens et al,[11]

modified by replacing methyl tridecanoate (C13:0 methyl ester) with methyl tricosanoate (C23:0 methyl ester) as the internal standard. After transesterification and extraction with n-hexane, FAMEs samples were analyzed with a HP 5890 Series II gas chromatograph with a flame ionization detector (GC-FID) equipped with a Restek Stabilwax column (30 m × 0.25 mm × 0.25 μm). Helium (2.5 mL/min) served as the carrier gas and injector split flow was set at 50 mL/min. Oven temperature was held at 210 °C for 5 min, then increased to 250 °C at a rate of 20 °C/min and held for 12 min. The injector and detector were set to 250 °C. A 0.5 μL injection volume was used, and two injections were made for each sample. GC-FID response peaks were calibrated and quantified with F.A.M.E. mix analytical standard (Sigma-Aldrich #18919). Concentration was normalized to the recovery of the internal standard and reported as percent dry weight of biomass (%dw as FAMEs).

ESI-5. HTL and product recovery method

HTL of the harvested batches was conducted in duplicate using 316-stainless steel tube batch reactors. The method was adapted for 6 in. tubes (3/8 in. outer diameter, 0.049 in. wall thickness, and 5.93 mL working volume) plugged with Swagelok® stainless steel-316 port connectors on both ends. Deionized (DI) water was added to freeze-dried biomass to obtain an 80 wt% moisture slurry, approximately 4 g of which was loaded into the tube reactor under ambient air, which also served as initial reaction headspace gas. Reactors were placed in a preheated muffle furnace (Type 30400, Thermolyne) at 300 °C for 30 min, with an additional 5 min of reactor heat-up time. Reaction temperature was regulated at 300 °C by the muffle furnace, and reaction pressure is autogenous pressure (estimated as 8.5 MPa from saturated steam tables).[12,13] After reaction, the tubes were removed and quenched in a cold water bath, then transferred to a glass desiccator for 1 h at room temperature to allow equilibration prior to product recovery. The tube reactors were weighed to ensure that no mass was lost during the HTL reaction, and carefully opened to vent gas phase products generated from the reaction. Gas phase product yield was then determined by re-weighing the reactor. Reactor contents were poured out into a glass beaker, and 30 mL of dichloromethane (DCM) was added to completely extract any DCM-soluble products, which was classified as the biocrude phase. It is noted here that use of DCM for biocrude product recovery is an experimental step necessary for HTL conducted in small batch tube reactors; solvent-free biocrude separation after HTL has been demonstrated with continuous pilot-scale reactors.[14,15] The reactor was then rinsed with 30 mL of DI water to recover any residual aqueous phase product. Both DCM-dissolved biocrude and DI water were added to the same beaker. An equivalent amount of DCM and DI water ensured there were no experimental artifacts from the artificial partitioning of products with different volumes of aqueous and organic solvents. Finally, the reactors were scraped with glass Pasteur pipets to recover any solids stuck on the sidewalls. The tube reactor was dried at 65 °C for 1 h and weighed after cooling to room temperature to ensure minimal product remaining in the reactor (<2.5 %dw of loaded biomass observed for all experiments). The collected product mixture was filtered into a separatory funnel through 0.45 μm Teflon filter cartridges (Whatman) to isolate the DCM/DI water insoluble solid phase product. Cartridges were dried in a desiccator overnight and weighed to obtain the solid phase yield. The separatory funnel was shaken to thoroughly mix the biocrude and aqueous phases which were then allowed to separate. The DCM phase containing the biocrude was then collected and DCM was removed under a stream of N_2 at 50 °C for 2 h before weighing to obtain the biocrude yield. The aqueous phase was diluted to 50 mL DI water, and two separate 10 mL aliquots of the diluted sample were dried at 65 °C for 16 h before weighing the residual solids to determine the aqueous phase yield. Gravimetric mass yields of the four product phases are reported as %dw of the input feedstock.

ESI-6. ECR and ER% Calculations

The energy consumption ratio (ECR) was determined according to the following equation, as described previously:[16–18]

$$ECR = \frac{\Delta T\,[W_i C_{pw} + C_{pb}(1-W_i)](1-R_h)}{Y(1-W_i)(HHV_b)R_c} \qquad \textbf{(SIEQ. 1)}$$

where ΔT is the temperature increase to reach reaction conditions (i.e., 275 K for a 300 °C reaction, assuming 25 °C ambient temperature), W_i is the initial moisture content (0.8), C_{pw} is the specific heat of water (4.18 kJ/kg.K), C_{pb} is the specific heat of biomass (1.25 kJ/kg.K),[16,18] R_h is the heat recovery efficiency (assumed 0.5),

R_c is the combustion energy efficiency (assumed 0.7), Y is the measured biocrude yield (as ratio i.e. 0–1), and HHV_b is the higher heating value of the biocrude in kJ/kg as determined by the method of Dulong.[3,17]

The energy recovery percentage (ER%) was calculated according to the following equation:[12,17]

$$ER\% = \frac{(HHV_b)(Y)}{HHV_m} \times 100\% \qquad \textbf{(SIEQ. 2)}$$

where HHV_m is the higher heating value of the dry biomass in kJ/kg.

ESI-7. Demonstration of integrated modelling framework

Complete details on the calibration of the Phototrophic Process Model (PPM) are available in Guest et al.[19] The PPM describes the biomass productivity as two outputs, X_{CPO} for functional cell biomass, and X_{TAG} for storage products as TAG, with units of mg-VSS/L as a function of time. The calibration of the PPM regards functional FAs as part of functional cell biomass, and since it has been established in this study (in sections prior to **Section 4**) that any functional lipids that bear FAs would contribute those FAs to biocrude yield, the X_{CPO} parameter was simply adjusted by the fatty acid content of the lowest FA-containing biomass in this study (Batch 2, 13.6 %dw). This way, the two main outputs of the PPM are non-FA and FA concentrations which are easily linked to the FA model for downstream predictions as detailed in **Section 3.5.2** using **Eqs. 5**-7 in the main article. Since **Section 4** was meant as a demonstration, cultivation energy demand was estimated on a per-hour per-liter basis using arbitrary values of 0.072 $kJ.h^{-1}L^{-1}$ to represent an upper-bound high value, and 0.036 $kJ.h^{-1}L^{-1}$ to represent a lower-bound low value. Harvesting energy demand was estimated on a per liter basis for harvesting using information from Shoener et al,[20] with similar upper- and lower-bounds. "High" represents the sum of both upper-bound demands of cultivation and harvesting, and "Low" the sum of both lower-bound demands. Energy supply was estimated using the predicted yields and elemental compositions (provided by the FA model) to calculate HHV and then using the calculated ECR (i.e., the same steps undertaken in **Section 3.5.2** to obtain **Fig. 7E** in the main article) to account for energy demanded for the thermochemical processing of HTL.

Table S1 Microalgae composition and HTL data from literature

Species	Proximate analysis (%dw)[a]				HTL conditions[b]			Biocrude yield (%dw)[c]		Citation
	Lipids	Proteins	Carbs	Ash	Time (min)	Temp (°C)	Moist. (wt%)	Expt.	Pred.[d]	
300 °C, external literature data (▲ in Fig. 6; 22 points)										
Spirulina sp.	5	64	20	11	30	300	80	31	35	[16]Vardon et al. (2012) **Δ**
Spirulina platensis	5	60	18	8	60	300	90	33	33	[21]Biller et al. (2012) * ^
Chlorogloeopsis fritschii	6	46	41	8	60	300	90	36	33	Biller et al. (2012) **Δ** * ^
Spirulina platensis	11	49	31	7	60	300	80	29	37	[22]Jena et al. (2011) **Δ** #
Desmodesmus sp.	12	41	28	8	5	300	90	37	34	[23]Garcia Alba et al. (2012)[h] **Δ** # ^
Desmodesmus sp.	12	41	28	8	15	300	90	39	34	Garcia Alba et al. (2012)[h] # ^
Desmodesmus sp.	12	41	28	8	30	300	90	40	34	Garcia Alba et al. (2012)[h] # ^
Desmodesmus sp.	12	41	28	8	60	300	90	43	34	Garcia Alba et al. (2012)[h] # ^
Scenedesmus dimorphous	13	56	25	6	30	300	80	45	40	Vardon et al. (2012) **Δ**
Nannochloropsis sp.	14	52	27	6	30	300	75	41	40	[24]Li et al. (2014)[h] **Δ**
Nannochloropsis sp.	14	52	27	6	60	300	85	48	40	Li et al. (2014)[h]
Nannochloropsis sp.	14	52	27	6	90	300	80	36	40	Li et al. (2014)[h]
Mixed culture[g]	15	37	31[f]	17	30	300	80	38	35	[25]Zhou et al. (2013) **Δ**
Dunaliella tertiolecta	16	49	12	24	5	300	80	33	38	[26]Minowa et al. (1995) **Δ** * ^
Dunaliella tertiolecta	16	49	12	24	60	300	80	26	38	Minowa et al. (1995) * ^
Mixed culture[g]	20	66	9[f]	5	30	300	80	51	49	Zhou et al. (2013) **Δ**
Dunaliella tertiolecta	23	34	21	13	30	300	90	17	41	[27]Zou et al. (2010) $ **Δ**
Chlorella vulgaris	23	51	8	7	60	300	90	43	45	Biller et al. (2012) **Δ** * ^
Nannochloropsis sp.	28	52	12	3	60	300	75	32	51	[3]Brown et al. (2010) **Δ**
Chlorella sp.	60	9	26	5	30	300	75	63	66	Li et al. (2014)[h] **Δ**

Table S1 Cont.

Species	Proximate analysis (%dw)[a]				HTL conditions[b]			Biocrude yield (%dw)[c]		Citation
	Lipids	Proteins	Carbs	Ash	Time (min)	Temp (°C)	Moist. (wt%)	Expt.	Pred.[d]	
Chlorella sp.	60	9	26	5	60	300	85	61	66	Li et al. (2014)[h]
Chlorella sp.	60	9	26	5	90	300	80	66	66	Li et al. (2014)
Reaction network model calibration data (■ in Fig. 6; 22 points)										
Scenedesmus sp.	8	50	31	11	10	300	85	39	34	[28]Valdez et al. (2014) Δ
Scenedesmus sp.	8	50	31	11	20	300	85	38	34	Valdez et al. (2014)
Scenedesmus sp.	8	50	31	11	40	300	85	41	34	Valdez et al. (2014)
Scenedesmus sp.	8	50	31	11	60	300	85	34	34	Valdez et al. (2014)
Nannochloropsis sp.	9	56	32	3	10	300	95	42	38	[29]Valdez et al. (2013)[i] Δ
Nannochloropsis sp.	9	56	32	3	20	300	96	39	38	Valdez et al. (2013)
Nannochloropsis sp.	9	56	32	3	40	300	97	37	38	Valdez et al. (2013)
Nannochloropsis sp.	9	56	32	3	60	300	97	42	38	Valdez et al. (2013)
Nannochloropsis sp.	9	56	32	3	90	300	97	32	38	Valdez et al. (2013)
Nannochloropsis sp.	9	56	32	3	10	300	91	36	38	Valdez et al. (2013)
Nannochloropsis sp.	9	56	32	3	20	300	85	39	38	Valdez et al. (2014)
Nannochloropsis sp.	9	56	32	3	40	300	96	32	38	Valdez et al. (2013)
Nannochloropsis sp.	9	56	32	3	60	300	96	42	38	Valdez et al. (2013)
Nannochloropsis sp.	9	56	32	3	90	300	97	44	38	Valdez et al. (2013)
Nannochloropsis sp.	14	59	20	3	20	300	85	51	42	[12]Valdez et al. (2012) # Δ
Nannochloropsis sp.	14	59	20	3	40	300	85	47	42	Valdez et al. (2012) #
Nannochloropsis sp.	14	59	20	3	60	300	85	42	42	Valdez et al. (2012) #
Nannochloropsis sp.	14	59	20	3	90	300	85	42	42	Valdez et al. (2012) #
C. protothecoides	53	11	29	7	10	300	85	43	61	Valdez et al. (2014) Δ
C. protothecoides	53	11	29	7	20	300	85	30	61	Valdez et al. (2014)
C. protothecoides	53	11	29	7	40	300	85	46	61	Valdez et al. (2014)
C. protothecoides	53	11	29	7	60	300	85	46	61	Valdez et al. (2014)
Composition data in ternary plot, not used for validation										
Porphyridium cruentum	6	33	30	24	60	350	90	15	25	[17]Biller and Ross (2011) Δ * ^
Phaeodactylum purpureum	12	46	33[f]	10	5	250	94	22	36	[30]Lopez Barreiro et al. (2013) Δ ^
Scenedesmus dimorphous	16	38	14	12	60	350	90	24	34	Biller et al. (2012) Δ * ^
Scenedesmus obliquus	17	28	27[f]	28	5	250	94	13	33	Lopez Barreiro et al. (2013) Δ ^
Tetraselmis suecica	20	44	21[f]	16	5	250	94	25	41	Lopez Barreiro et al. (2013) Δ ^
Chlorella vulgaris	20	41	16[f]	22	5	250	84	26	40	Lopez Barreiro et al. (2013) Δ ^
Phaeodactylum tricornutum	22	38	16[f]	25	5	250	94	31	40	Lopez Barreiro et al. (2013) Δ ^
Scenesdesmus almeriensis	22	52	15[f]	12	5	250	94	31	45	Lopez Barreiro et al. (2013) Δ ^
Dunaliella tertiolecta	23	51	20[f]	6	5	250	94	42	47	Lopez Barreiro et al. (2013) Δ ^
Nannochloropsis oculata	24	42	6	26	60	350	90	26	41	Biller and Ross (2011) Δ * ^
Nannochloropsis gaditana	25	44	19[f]	12	5	250	94	30	46	Lopez Barreiro et al. (2013) Δ ^
Data from this study for calibration (● in Fig. 6; 9 points)										
Nannochloropsis oculata	23	58	13	6	30	300	80	51	49	
	31	51	12	6	30	300	80	54	53	
	32	50	11	6	30	300	80	54	54	
	31	50	13	6	30	300	80	54	53	
	47	28	22	3	30	300	80	64	61	
	49	32	17	2	30	300	80	64	64	
	51	26	19	4	30	300	80	61	64	
	56	23	18	2	30	300	80	67	67	
	59	17	22	2	30	300	80	68	68	

a Proximate data reproduced from source if reported as %dw; * denotes data reported as %afdw corrected to %dw using reported ash content; $ denotes data reported as % corrected to %dw using reported moisture content; # denotes %dw or %afdw was not explicitly stated and assumed to be %dw given that lipid + protein + carb + ash was typically ~100%; Δ denotes that the data set was used for the ternary plot (**Fig. 1** in main paper). **b** All experiments were conducted with water as reaction medium, without any catalysts, and a headspace of inert gases (N_2, He) or ambient air. **c** Yield data reproduced from source if reported as %dw; ^ denotes data reported as %afdw corrected to %dw using reported ash content. **d** Predicted using component additivity model (**Eq. 4** in main paper). **e** Refer to references at the end of the ESI for full citation. **f** Value was not reported in source and assumed using (100 %dw – lipid – protein – ash) since results were reported as %dw. **g** Cultivated from recycled HTL aqueous phase diluted in municipal wastewater. **h** carbs fraction is sum of crude fibre and non-fibrous carbohydrate fractions. **i** Composition results are from Valdez et al. (2014),[22] HTL results are from supplementary information of Valdez et al. (2013).[23]

Table S2 Proximate analysis of harvested batches[a] (wt%)

Batch	Moisture	Ash	Crude Proteins	Crude Lipids	Crude Carbs	Summation
1	1.0±0.3	5.5±0.1	69.9±0.3	n.d.	18.2±0.2	94.6±0.5
2	1.1±0.1	5.9±0.1	60.1±0.0	23.7±0.3	13.7±0.1	104.5±0.4
3	1.4±0.4	5.8±0.0	54.1±0.1	32.3±0.4	13.0±0.2	106.7±0.6
3b	0.5±0.3	6.1±0.2	52.3±0.0	33.5±0.9	11.8±0.2	104.2±1.0
3c	1.1±0.1	6.4±0.3	51.0±0.0	32.0±0.0	12.8±0.2	103.3±0.4
4	0.8±0.2	3.2±0.2	28.8±0.0	48.1±0.3	22.5±1.1	103.5±1.2
5	1.3±0.2	2.5±0.0	33.5±0.1	50.9±1.0	17.4±0.1	105.5±1.1
6	1.3±0.4	3.6±0.1	26.9±0.0	51.9±0.1	19.3±0.1	103.0±0.4
7	0.2±0.2	2.2±0.3	24.6±0.1	59.1±0.7	19.4±0.0	105.5±0.8
8	0.5±0.2	2.1±0.1	17.6±0.1	60.3±0.8	22.8±0.5	103.3±1.0

a All values reported as the mean of duplicate analysis, except for moisture and ash which was performed in triplicate. Batches 3b and 3c were cultivated and harvested under identical conditions as Batch 3.

Table S3 Replicate batch composition and corresponding HTL yields[a]

Batch	Proximate analysis			Elemental composition (%)					Lipid fractions[b]			HTL product yields			
	Lipids	Protein	Carbs	%C	%H	%N	%O	HHV (MJ/kg)	NL	PL	Total FAMEs	Biocrude oil	Aqueous phase	Gas phase	Filtered solids
3[c]	30.7	51.4	12.3	54.6	7.8	8.7	23.5	25.4	12.2	18.5	19.8	53.6	25.0	16.3	3.2
3b	32.3	50.4	11.4	54.1	7.9	8.4	23.9	25.2	17.4	14.9	21.0	53.7	31.1	12.0	3.2
3c	31.3	49.9	12.5	54.2	7.8	8.2	23.7	25.2	15.1	16.2	17.4	53.8	26.8	12.3	2.6

a All values (unless otherwise stated) reported in %dw as the mean of duplicate analysis. b NL = neutral lipids; PL = polar lipids. c From Table 1 in main article.

Table S4 Filtered solids product elemental analysis and biomass C/N distribution among HTL products[a]

Batch	Filtered solids[b]			Carbon distribution (%)					Nitrogen distribution[d] (%)			
	%C	%H	%N	Biocrude	Aqueous	Gas[c]	Solids	Total	Biocrude	Aqueous	Solids	Total
1	43.2	5.7	3.4	49.0 ± 0.8	33.6 ± 0.5	6.8	10.9	100.3 ± 2.6	27.0 ± 0.5	74.4 ± 1.0	3.5	104.9 ± 1.8
2	16.8	2.9	2.1	66.7 ± 1.2	22.2 ± 1.8	4.4	1.9	95.2 ± 0.6	32.5 ± 0.6	66.9 ± 1.6	1.3	100.7 ± 1.0
3	26.6	3.7	2.5	70.5 ± 1.3	21.5 ± 0.2	8.2	1.6	101.8 ± 0.9	34.1 ± 0.6	68.5 ± 0.9	0.9	103.5 ± 0.3
4	39.4	4.7	2.8	80.2 ± 0.1	11.9 ± 1.1	7.7	1.2	101.0 ± 0.9	47.6 ± 0.1	48.2 ± 1.3	1.2	97.0 ± 1.3
5	39.4	4.7	2.8	78.5 ± 0.1	11.7 ± 0.0	7.2	1.3	98.7 ± 0.1	43.9 ± 0.1	60.0 ± 7.4	1.1	104.8 ± 7.4
6	62.4	5.2	5.6	75.1 ± 1.0	10.8 ± 0.9	10.0	1.9	97.8 ± 2.2	48.9 ± 0.7	44.8 ± 2.3	2.4	96.1 ± 2.8
7	62.4	5.2	5.6	80.1 ± 1.0	10.1 ± 0.0	7.0	2.6	99.8 ± 1.1	45.3 ± 0.6	45.9 ± 3.0	3.8	95.0 ± 3.0
8	62.4	5.2	5.6	83.0 ± 0.1	9.6 ± 1.4	5.1	4.4	102.0 ± 1.1	48.8 ± 0.1	40.1 ± 0.8	8.8	97.6 ± 1.0

a Values reported as the mean of duplicate analysis with min/max values (±) shown only if > ±0.5%. b Composite solid phase samples were analysed for CHN content for Batches 4–5 and for Batches 6–8 due to the small yield of solid phase products from HTL of these batches (only 1.8–4.4 %dw). c Gas phase carbon distribution was estimated using the gravimetrically measured gas product yields (see **ESI-5**) and assuming the gas products to be 100% CO_2.[3,12] d No significant distribution of nitrogen because the headspace gas was assumed to be 100% CO_2.[3,12]

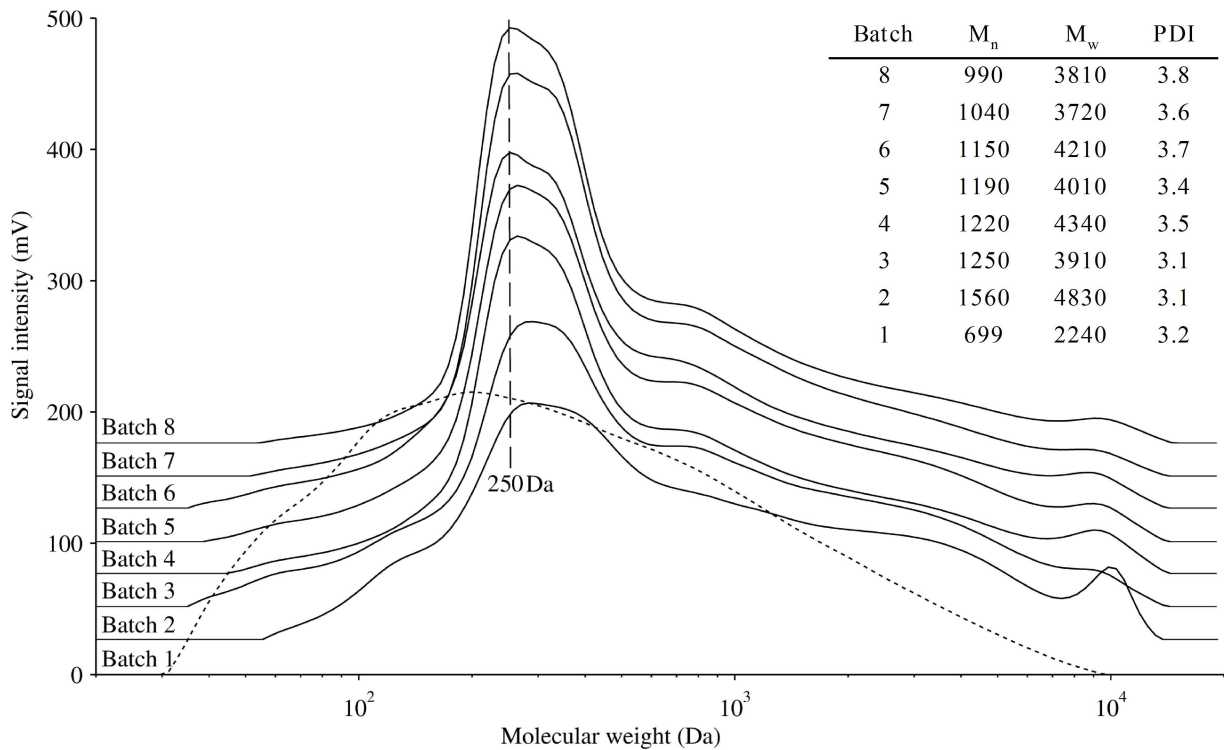**Table S5 HTL aqueous phase product analysis[a] (all units in mg/L)**

Batch	TOC[b]			NH$_3$			TKN[c]			NO$_3^-$/NO$_2^-$	% of TN[d] as NH$_3$	PO$_4^{3-}$		
1	38300	±	620	11200	±	390	20500	±	280	n.d	54.6	1800	±	10
2	29400	±	2400	7400	±	200	15900	±	390	n.d	46.5	140	±	4
3	28900	±	200	7300	±	130	14600	±	190	n.d	50.0	1900	±	40
4	17400	±	1600	3000	±	30	5500	±	150	n.d	54.5	520	±	50
5	17400	±	20	3700	±	20	7900	±	970	n.d	46.8	670	±	20
6	16100	±	1300	2800	±	200	4700	±	240	n.d	59.6	440	±	4
7	15900	±	60	2200	±	260	4500	±	290	n.d	48.9	630	±	40
8	14800	±	2100	1200	±	40	2800	±	60	n.d	42.9	270	±	30

a Concentrations based on water initially loaded with the biomass slurry prior to reaction. **b** Total organic carbon, equals total carbon assuming negligible inorganic carbon. **c** Total Kjeldahl nitrogen. **d** Total nitrogen, TN = TKN + NO$_3^-$ + NO$_2^-$ = TKN since NO$_3^-$/NO$_2^-$ is below detection limit (0.2 mg/L) for all batches.

Table S6 List of regression statistics generated by Microsoft Excel 2010 Analysis ToolPak

Summary	Multiple R	R^2	Adjusted R^2	Standard error	Observations
Regression Statistics	0.99962	0.99924	0.85616	0.01907	10

ANOVA	df	SS	MS	F	Significance F
Regression	3	3.33118	1.11039	3052.00	6.142E-10
Residual	7	0.00255	0.00036		
Total	10	3.33373			

Regression output	Coefficients	Standard error	t Stat	P-value	Lower 95%	Upper 95%
Lipids	0.97184	0.04449	21.84292	1.064E-07	0.86663	1.07705
Proteins	0.41936	0.02993	14.01345	2.232E-06	0.34859	0.49012
Carbohydrates	0.16903	0.14966	1.12947	0.29591	-0.18485	0.52292

Residual output	Predicted Y	Residuals	Cook's distance
1	0.48903	0.02392	0.1249
2	0.34606	-0.01437	12.4519
3	0.53486	0.00147	0.0005
4	0.54474	-0.00791	0.0238
5	0.53473	0.00278	0.0050
6	0.60999	0.02885	0.4592
7	0.63740	-0.00219	0.0089
8	0.63904	-0.02756	0.2277
9	0.67482	-0.00903	0.0394
10	0.67926	0.00415	0.0095

Batch	M_n	M_w	PDI
8	990	3810	3.8
7	1040	3720	3.6
6	1150	4210	3.7
5	1190	4010	3.4
4	1220	4340	3.5
3	1250	3910	3.1
2	1560	4830	3.1
1	699	2240	3.2

Fig. SI1 Molecular weight distribution of biocrude products derived from SEC analysis. Vertical dashed line indicates 250 Da. Defatted Batch 1 represented as dotted line to highlight the effect of extracting the lipids prior to HTL.
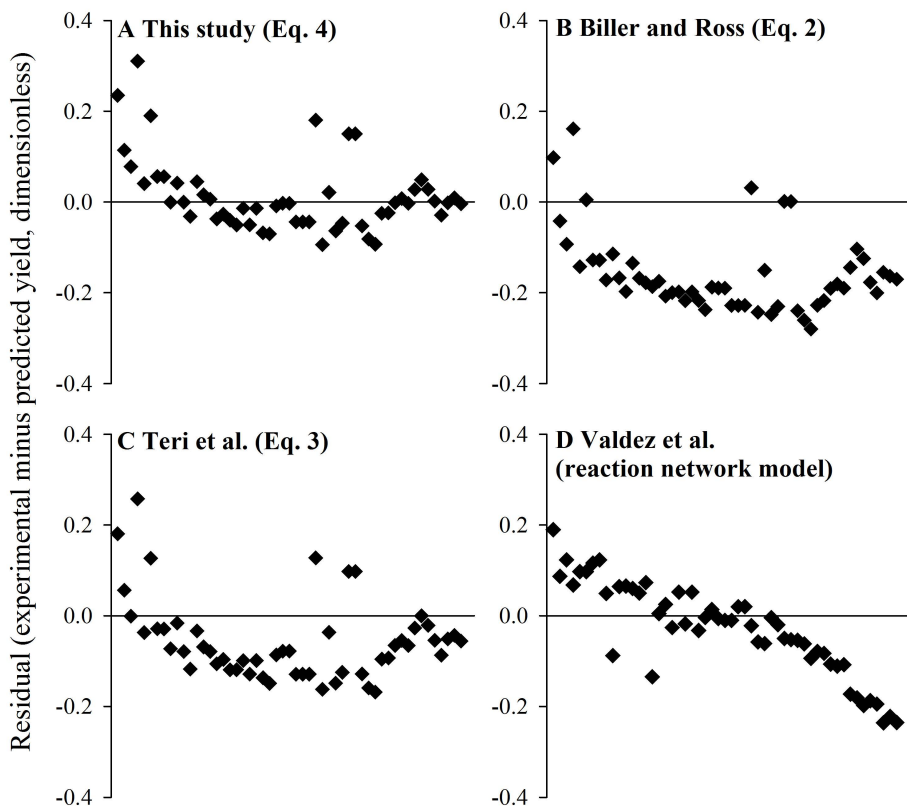
Fig. SI2 Plot of residuals for each predictive model. Points are arranged in ascending experimental biocrude yield (as %dw) from left (closer to y-axis) to right (further from y-axis).

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