Supporting Information

Life Cycle Assessment of Catechols from Lignin Depolymerization

Mahdokht Montazeri and Matthew J. Eckelman

Department of Civil & Environmental Engineering, Northeastern University, 360 Huntington Avenue, Boston, MA, 02115, USA

Corresponding Author: Mathew J. Eckelman, m.eckelman@neu.edu, +1 (617) 373-4256

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Part I: Methods

SI Text. Life Cycle Inventories

Bio-based Route Inventory. Detailed data about inventory set up and databases used for this study are included in the Supplemental Information (SI).

Table S1 lists all inventory inputs for production of 1 kg *tert*-butyl catechol from bio-based resources. Input parameters were based on experimental data[1] scaled for an industrial plant operating with a capacity of 75 ton lignin/day. ASPEN plus simulation output is 54 tons of TBC/day. Material inputs are linearly extrapolated from the experimental data source[1] and scaled as shown below for four different inputs of main steps:

Cultivation:

$$\begin{aligned}&\text{Cutivated amount (nuts + nutshells)}\\&= \frac{1\text{ g (nuts + nutshells)}}{0.7\text{ g nutshells}} * \frac{110\text{ g nutshells}}{13.62\text{ g crude lignin}} * \frac{8.39\text{ g crude lignin}}{8.077\text{ g purified lignin}}\\&* \frac{7.29\text{ g purified lignin}}{4.98\text{ g Et. Ac. soluble lignin}} * \frac{1\text{ ton (nuts + nutshells)}}{10^6\text{g (nuts + nutshells)}} * \frac{10^6\text{g Et. Ac. soluble lignin}}{1\text{ ton Et. Ac. soluble lignin}}\\&* \frac{75\text{ ton Et. Ac. soluble lignin}}{\text{day}} * \frac{\text{day}}{53.5\text{ ton TBC}} = 24.6\,\frac{\text{kg (nuts + nutshells)}}{\text{kg TBC}}\end{aligned}$$

Organosolv Extraction:

$$\begin{aligned}&\text{Methanol make} - \text{up flow}\\&= \frac{500\text{ ml MeOH}}{13.62\text{ g crude lignin}} * \frac{0.79\text{ g MeOH}}{1\text{ ml MeOH}} * \frac{3\text{ g MeOH used}}{100\text{ g MeOH}} * \frac{8.39\text{ g crude lignin}}{8.077\text{ g purified lignin}}\\&* \frac{7.29\text{ g purified lignin}}{4.98\text{ g Et. Ac. soluble lignin}} * \frac{10^6\text{g Et. Ac. soluble lignin}}{1\text{ ton Et. Ac. soluble lignin}} * \frac{1\ ton\ MeOH\ used}{10^6 g\ MeOH\ used}\\&* \frac{75\text{ ton Et. Ac. solube lignin}}{\text{day}} * \frac{\text{day}}{53.5\text{ ton TBC}} = 1.85\ \frac{\text{ton/kg MeOH}}{\text{ton/kg TBC}}\end{aligned}$$

Energy consumption for this process was sourced from ASPEN Plus simulations conducted for several extraction methods on softwood lignin as reported in Conde-Mejia et al.[2] and modified for our analysis:

$$\text{Heating energy} = \frac{7.78\ \text{MMBtu}}{\text{ton biomass (nutshells)}} * \frac{1055\ \text{MJ}}{1\ \text{MMBtu}} * \frac{1\ \text{ton nutshells}}{1000\ \text{kg nutshells}} * \frac{1\ \text{kg (nuts + nutshells)}}{0.7\ \text{kg nutshells}} * \frac{24.6\ \text{kg (nuts + nutshells)}}{\text{kg TBC}}$$

Lignin Purification:

$$\text{Dichloromethane make} - \text{up flow} = \frac{150\ \text{ml DCM}}{8.077\ \text{g purified lignin}} * \frac{1.322\ \text{g DCM}}{1\ \text{ml DCM}} * \frac{3\text{g DCM used}}{100\ \text{g DCM}} * \frac{7.29\ \text{g purified lignin}}{4.98\ \text{Et. Ac. soluble lignin}} * \frac{10^6\text{g Et. Ac. soluble lignin}}{1\ \text{ton Et. Ac. soluble lignin}} * \frac{1\ \text{ton DCM used}}{10^6\text{g DCM used}} * \frac{75\ \text{ton Et. Ac. soluble lignin}}{\text{day}} * \frac{\text{day}}{53.5\ \text{ton TBC}} = 1.5\ \frac{\text{ton/kg DCM}}{\text{ton/kg TBC}}$$

Lignin Depolymerization:

Sub-critical methanol is not reacting in this step since it is used as a solvent for lignin, so it can be recovered at rates exceeding 99%. Here we model 99% recovery of methanol, in contrast to 97% recovery rates for dichloromethane and xylene. A small amount of methanol is lost with the unreacted solubilized lignin:

$$\text{MeOH make} - \text{up flow} = \frac{30\ \text{ml MeOH}}{4.98\ \text{g Et. Ac. soluble lignin}} * \frac{0.79\ \text{g MeOH}}{1\ \text{ml MeOH}} * \frac{1\ \text{g MeOH used}}{100\ \text{g MeOH}} * \frac{10^6\text{g Et. Ac. soluble lignin}}{1\ \text{ton Et. Ac. soluble lignin}} * \frac{1\ \text{ton MeOH used}}{10^6\text{g MeOH used}} * \frac{75\ \text{ton Et. Ac. soluble lignin}}{\text{day}} * \frac{\text{day}}{53.5\ \text{ton TBC}} = 0.01\ \frac{\text{ton/ kg MeOH}}{\text{ton/ kg TBC}}$$

Energy consumption for nutshell preparation, lignin extraction and catalytic depolymerization were estimated from ecoinvent unit processes, literature[2] and ASPEN Plus simulations, respectively.

Catalyst Preparation:

Cu-PMO catalyst preparation was modeled separately based on experimental data by Hansen et al.[3] Unit processes for input metal salts aluminium nitrate, copper nitrate, and magnesium acetate were built based on industrial chemistry description in Ullman's Encyclopedia of Chemical Engineering[4] and Handbook of Inorganic Chemicals.[5] Energy consumption of the catalyst preparation process was estimated based on ASPEN Plus simulation.

For LCA modeling in SimaPro, all input parameters were chosen from existing unit processes in the ecoinvent 3.1. database, using life cycle inventory unit processes adjusted for the US energy system (US-EI database; Earthshift, Huntington, VT).

Table S1. Life cycle inventory for production of 1 kg TBC from bio-based resource (Organosolv extraction method)

Material/Assembly	**Total amount**	**Allocated amount**	**Unit**
Methanol, at plant/GLO	0.01	0.01	kg
Hydrogen, cracking, APME, at plant/RER	0.02	0.02	kg
Cu-PMO catalyst	0.56	0.56	kg
Ethyl acetate, at plant/RER	0.6	0.6	kg
Dichloromethane, at plant/RER	1.5	1.5	kg
Methanol, at plant/GLO	1.8	0.2	kg
Husked nuts harvesting, at farm/PH	24.5	2	kg
Nitrogen fertilizer, production mix, at plant, NREL/ US	0.61	0.05	kg
Proxy_Phosphorous Fertilizer (TSP as P2O5), at plant NREL /US	0.98	0.1	kg
Proxy_Potash Fertilizer (K2O), at plant NREL /US	0.32	0.03	kg
Processes	**Total amount**	**Allocated amount**	**Unit**
Electricity, production mix US/US	10	10	kWh
Heat, natural gas, at boiler modulating <100kW/RER	141.25	16.9	MJ
Cooling energy, natural gas, at cogen unit with absorption chiller 100 kW/CH	57	6.8	MJ
Wood chopping, mobile chopper, in forest/RER	17.21	2	kg

Table S2. Life cycle inventory for production of 1 kg Cu-PMO catalyst

Material/Assembly	**Total amount**	**Allocated amount**	**Unit**
Sodium carbonate from ammonium chloride production, at plant/GLO	0.7	7.8E-5	kg
Aluminium nitrate, Al(NO3)3.9H2O	0.07	7.8E-6	kg
Copper nitrate, Cu(NO3)2.2 H2O	0.03	3.4E-6	kg
Magnesium acetate, Mg(CH3COO)2.4 H2O	0.2	2.2E-6	kg
Tap water, at user/RER	89	9E-3	kg
Sodium hydroxide, 50% in H2O, production mix, at plant/RER	0.05	5.6E-6	kg
Processes	**Total amount**	**Allocated amount**	**Unit**
Electricity mix/US	21.8	2.44E-3	kWh

Fossil-based Route Inventory. Fossil-based production of TBC was modeled based on a two-step process with the life cycle inventory given in **Table 3**, and shown in equations 3 and 4 in the main text. The first step was based on the catalytic (SeO_2) hydroxylation of phenol with hydrogen peroxide.[4] The second step is butylation of catechol using triflouromethanesulfonic acid (TFMS) as a catalyst.[6] Input chemicals were scaled up based on equations 3 and 4 and their respective conversion yields of 100% and 35%, and scaled to 1 kg TBC as the target product. Energy inputs was estimated from ASPEN Plus simulations. TFMS was modeled as a new assembly[7] in the inventory (**Table S4**). Selenium dioxide was modeled as Se, adjusting for molecular weights.

Table S3. Life cycle inventory for production of 1 kg TBC from fossil-based resource

Material/Assembly	Total amount	Allocated amount	Unit
Phenol, at plant/RER	2.5	1.6	kg
Hydrogen peroxide, 50% in H2O, at plant/RER	0.9	0.6	kg
Isobutanol, at plant/RER	0.6	0.6	kg
Xylene, at plant/RER	1.25	1.25	kg
Trifluoromethane sulfonic acid	0.003	0.003	kg
Selenium, at plant/RER	0.0003	1.9E-4	kg
Processes	**Total amount**	**Allocated amount**	**Unit**
Electricity, medium voltage, at grid/US	2.95	2.4	kWh
Transport, freight, rail/RER	1	1	tkm
Transport, lorry >16t, fleet average/RER	0.2	0.2	tkm
Heat, natural gas, at boiler modulating <100kW/RER	2	2	MJ

Table S4. Life cycle inventory for production of 1 kg TFMS used in fossil-based route

Material/Assembly	Total amount	Allocated amount	Unit
Hydrogen fluoride, at plant/GLO	0.4	0.075	kg
Methanol, at plant/GLO	0.2	0.006	kg
Oxygen, liquid, at plant/RER	0.3	0.009	kg
Proxy_Sulfuric acid, at plant NREL /US	0.3	0.009	kg
Processes	**Total amount**	**Allocated amount**	**Unit**
Electricity, medium voltage, at grid/US	1	0.03	kWh
Transport, lorry >16t, fleet average/RER	0.2	0.006	tkm
Heat, natural gas, at boiler modulating <100kW/RER	2	0.06	MJ

Alternate Lignin Extraction. Here, we considered substitution of an alternate extraction method for organosolv extraction, the method in our base case scenario. This alternate method is based on a US patent[8] for separation of lignin. We assumed loblolly pine as an example of softwood that contains lignin fraction with approximately the same chemical structure as that found for candlenut shells. Ammonium hydroxide and sulfuric acid are used as solvents for extraction of lignin. Ammonium hydroxide volume to biomass weight ratio is 16:1 and the process can achieve 60% efficiency for lignin separation.

Input parameters were scaled up based on reported inputs for lab scale analysis and ASPEN Plus simulations:

<u>Cultivation:</u>

$$\begin{aligned}
&\text{Cutivated amount (nuts + nutshells)} \\
&\quad = \frac{1\text{ g (nuts + nutshells)}}{0.7\text{ g nutshells}} * \frac{110\text{ g milled shells}}{13.62\text{ g available lignin}} * \frac{13.62\text{ g available lignin}}{8.17\text{ g pure lignin}} \\
&\quad * \frac{10^6\text{g pure lignin}}{1\text{ ton pure lignin}} * \frac{1\text{ ton (nuts + nutshells)}}{10^6\text{g (nuts + nutshells)}} * \frac{75\text{ ton pure lignin}}{\text{day}} * \frac{\text{day}}{53.5\text{ ton TBC}} \\
&\quad = 26.22\,\frac{\text{kg (nuts + nutshells)}}{\text{kg TBC}}
\end{aligned}$$

<u>Solvent Extraction:</u>

$$\begin{aligned}
&\text{Ammonium Hydroxide make} - \text{up flow} \\
&\quad = \frac{800\text{ ml NH}_4\text{OH}}{50\text{ g nutshells}} * \frac{0.88\text{ kg NH}_4\text{OH}}{1000\text{ ml NH}_4\text{OH}} * \frac{3\text{ kg NH}_4\text{OH used}}{100\text{ kg NH}_4\text{OH}} * \frac{1000\text{ g nutshells}}{1\text{ kg nutshells}} \\
&\quad * \frac{0.7\text{ kg nutshells}}{1\text{ kg (nuts + nutshells)}} * \frac{26.22\text{ kg (nuts + nutshells)}}{\text{kg TBC}} = 7.7\ \frac{kg\ NH_4OH}{kg\ TBC}
\end{aligned}$$

Electricity consumption for this process is based on ASPEN plus simulation conducted in the same patent[8] for this method and we scaled based on our biomass flow (nutshell consumption):

$$\begin{aligned}
\text{Electricity} &= \frac{217\text{ kWh}}{1000\text{ kg nutshells}} * \frac{0.7\text{ kg nutshells}}{1\text{ kg (nuts + nutshells)}} * \frac{26.22\text{ kg (nuts + nutshells)}}{1\text{ kg TBC}} \\
&= 3.98\ \frac{\text{kWh}}{\text{kg TBC}}
\end{aligned}$$

Lignin depolymerization is assumed to proceed in an identical fashion as the base case, with output of 1 kg TBC. **Table S5** shows the inputs for bio-based route considering the alternate extraction method.

Table S5. Life cycle inventory for production of 1 kg TBC from bio-based resource (Solvent extraction method)

Material/Assembly	**Total amount**	**Allocated amount**	**Unit**
Methanol, at plant/GLO	0.01	0.01	kg
Hydrogen, cracking, APME, at plant/RER	0.02	0.02	kg
Cu-PMO catalyst	0.56	0.56	kg
Proxy_Sulfuric acid, at plant NREL /US	1.13	0.08	kg
Ammonia, liquid, at regional storehouse/RER	7.7	0.5	kg
Husked nuts harvesting, at farm/PH	26.22	1.3	kg
Nitrogen fertilizer, production mix, at plant, NREL/ US	0.65	0.03	kg
Proxy_Phosphorous Fertilizer (TSP as P2O5), at plant NREL /US	1.05	0.05	kg
Proxy_Potash Fertilizer (K2O), at plant NREL /US	0.34	0.02	kg
Processes	**Total amount**	**Allocated amount**	**Unit**
Electricity, production mix US/US	14	10.2	kWh
Wood chopping, mobile chopper, in forest/RER	18.45	0.9	kg

Alternate Lignin Source. This pathway is hypothetical, considering substitution of coconut shell lignin and solvent extraction for candlenut shell lignin and organosolv extraction method, respectively. Input chemicals were scaled based on original experimental data available for candlenut shell,[1] adjusting for lignin content of coconut shell (44%) and weight percent of nutshell (0.15% for coconut).[9]

Cultivation:

$$
\begin{aligned}
&\text{Cutivated amount (nuts + nutshells)} \\
&= \frac{1\text{ g (nuts + nutshells)}}{0.15\text{ g nutshells}} * \frac{110\text{ g nutshells}}{40.15\text{ g available lignin}} * \frac{40.15\text{ g available lignin}}{34.93\text{ g pure lignin}} \\
&* \frac{10^6\text{g pure lignin}}{1\text{ ton pure lignin}} * \frac{1\text{ ton (nuts + nutshells)}}{10^6\text{g (nuts + nutshells)}} * \frac{75\text{ ton pure lignin}}{\text{day}} * \frac{\text{day}}{53.5\text{ ton TBC}} \\
&= 29.4\,\frac{\text{kg (nuts + nutshells)}}{\text{kg TBC}}
\end{aligned}
$$

Solvent Extraction:

$$
\begin{aligned}
&\text{Ammonium Hydroxide make} - \text{up flow} \\
&= \frac{800\text{ ml NH}_4\text{OH}}{50\text{ g nutshells}} * \frac{0.88\text{ kg NH}_4\text{OH}}{1000\text{ ml NH}_4\text{OH}} * \frac{3\text{ kg NH}_4\text{OH used}}{100\text{ kg NH}_4\text{OH}} * \frac{1000\text{ g nutshells}}{1\text{ kg nutshells}} \\
&* \frac{0.15\text{ kg nutshells}}{1\text{ kg (nuts + nutshells)}} * \frac{29.4\text{ kg (nuts + nutshells)}}{\text{kg TBC}} = 1.86\,\frac{kg\ NH_4OH}{kg\ TBC}
\end{aligned}
$$

$$
\begin{aligned}
\text{Electricity} &= \frac{217\text{ kWh}}{1000\text{ kg nutshells}} * \frac{0.15\text{ kg nutshells}}{1\text{ kg (nuts + nutshells)}} * \frac{29.4\text{ kg (nuts + nutshells)}}{1\text{ kg TBC}} \\
&= 0.95\,\frac{\text{kWh}}{\text{kg TBC}}
\end{aligned}
$$

Table S6 shows the inventory for the alternate lignin source and process. As mentioned in the main text, while we still scale the output of the process to 1 kg of TBC, this method is hypothetical and the actual final products should be specified experimentally.

Table S6. Life cycle inventory for production of 1 kg TBC from bio-based resource (Coconut shells+ Solvent extraction method)

Material/Assembly	Total amount	Allocated amount	Unit
Methanol, at plant/GLO	0.01	0.01	kg
Hydrogen, cracking, APME, at plant/RER	0.02	0.02	kg
Cu-PMO catalyst	0.56	0.56	kg
Proxy_Sulfuric acid, at plant NREL /US	0.4	0.14	kg
Ammonia, liquid, at regional storehouse/RER	1.86	0.7	kg
Husked nuts harvesting, at farm/PH	29.4	1.6	kg
Nitrogen fertilizer, production mix, at plant, NREL/ US	0.7	0.04	kg
Proxy_Phosphorous Fertilizer (TSP as P2O5), at plant NREL /US	1.18	0.06	kg
Proxy_Potash Fertilizer (K2O), at plant NREL /US	0.4	0.02	kg
Processes	**Total amount**	**Allocated amount**	**Unit**
Electricity, production mix US/US	11	10.2	kWh
Wood chopping, mobile chopper, in forest/RER	4.41	1.6	kg

Waste Treatment Considerations. Waste management of various solvents used for both base case fossil-based and bio-based routes were considered as a separate analysis here. Dichloromethane, ethyl acetate and hydrogen peroxide were treated as hazardous wastes based on EPA Best Demonstrated Available Technology (BDAT)[10] for waste management of relevant group of chemicals. Table S7 and S8 show the chosen unit processes from eco-invent and the amount of corresponding solvent for landfill and incineration, respectively.

Table S7. Landfill waste treatment scenario for base case bio-based and fossil-based routes

Bio-based Route			
Treatment Process	**Treated solvent**	**Allocated amount**	**Unit**
Proxy_Disposal, n-butyl alcohol, to sanitary landfill NREL /US	Methanol	0.2	kg
Disposal, hazardous waste, 0% water, to underground deposit/DE	Dichloromethane	1.5	kg
Proxy_Disposal, formaldehyde, to unspecified treatment NREL /US	Ethyl acetate	0.6	kg
Fossil-based Route			
Treatment Process	**Treated solvent**	**Allocated amount**	**Unit**
Proxy_Disposal, light aromatic solvent naphtha, to sanitary landfill NREL /US	Phenol	1.6	kg
Disposal, hazardous waste, 0% water, to underground deposit/DE WITH US ELECTRICITY U	Hydrogen peroxide	0.6	kg
Proxy_Disposal, n-butyl alcohol, to sanitary landfill NREL /US	Isobutanol	0.6	kg
Proxy_Disposal, light aromatic solvent naphtha, to sanitary landfill NREL /US	Xylene	1.2	kg

Table S8. Incineration waste treatment scenario for base case bio-based and fossil-based routes

Bio-based Route			
Treatment Process	**Treated solvent**	**Allocated amount**	**Unit**
Disposal, solvents mixture, 16.5% water, to hazardous waste incineration/CH	Methanol	0.2	kg
Disposal, hazardous waste, 25% water, to hazardous waste incineration/CH	Dichloromethane	1.5	kg
Disposal, hazardous waste, 25% water, to hazardous waste incineration/CH	Ethyl acetate	0.6	kg
Fossil-based Route			
Treatment Process	**Treated solvent**	**Allocated amount**	**Unit**
Disposal, solvents mixture, 16.5% water, to hazardous waste incineration/CH	Phenol	1.6	kg
Disposal, hazardous waste, 25% water, to hazardous waste incineration/CH	Hydrogen peroxide	0.6	kg
Disposal, solvents mixture, 16.5% water, to hazardous waste incineration/CH	Isobutanol	0.6	kg
Disposal, solvents mixture, 16.5% water, to hazardous waste incineration/CH	Xylene	1.2	kg

Part II: Results

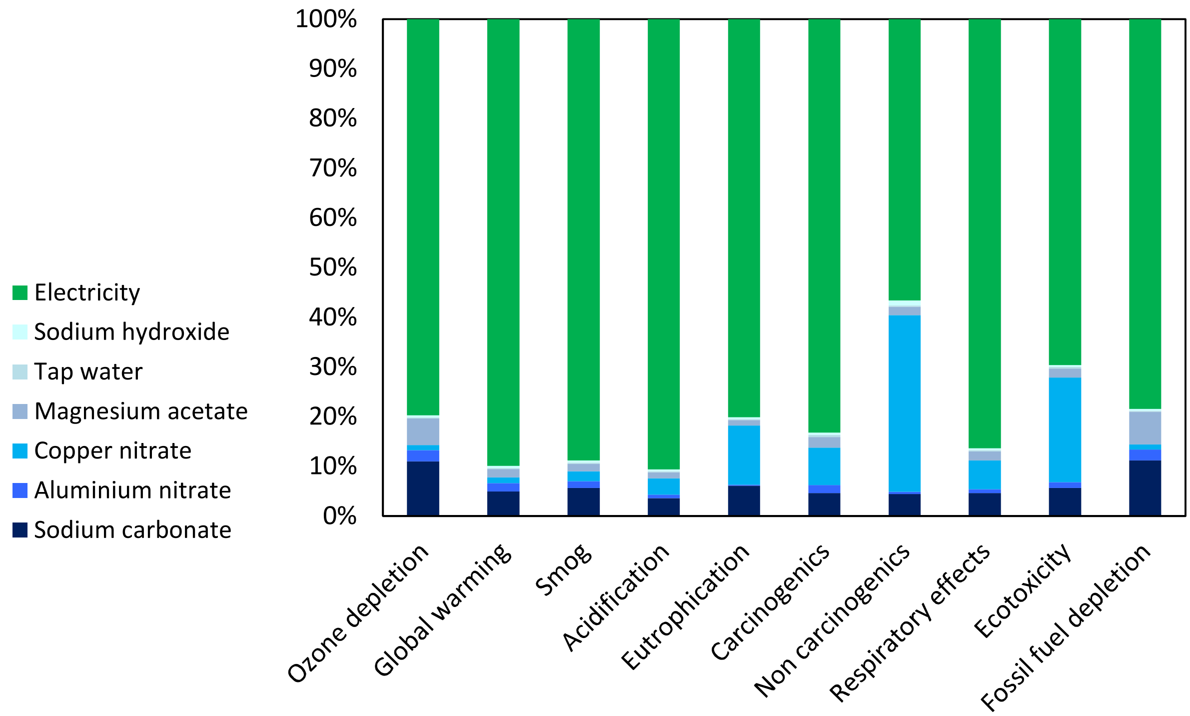

Figure S1. Results for process and material contribution in production of 1 kg Cu-PMO catalyst

Table S9. Relative results for residual solvents treatment methods

Impact Category	Ozone depletion	Global warming	Smog	Acidifi-cation	Eutrophi-cation	Carcino-genics	Non-carc-inogenics	Respiratory effects	Ecotoxicity	Fossil fuel depletion
Unit	**kg CFC-11 eq**	**kg CO_2 eq**	**kg O_3 eq**	**kg SO_2 eq**	**kg N eq**	**CTUh**	**CTUh**	**kg $PM_{2.5}$ eq**	**CTUe**	**MJ surplus**
Comparative Results	13,084%	-13%	65%	79%	35%	78%	144%	103%	18%	-54%
Fossil-based Route + incineration	7.72E-07	2.19E+01	6.59E-01	6.05E-02	4.29E-02	8.59E-07	5.77E-07	4.31E-03	2.23E+01	4.81E+01
Bio-based Route + incineration	1.02E-04	1.90E+01	1.09E+00	1.08E-01	5.79E-02	1.53E-06	1.41E-06	8.74E-03	2.64E+01	2.21E+01
Comparative Results	17,529%	-1%	75%	85%	54%	28%	184%	113%	-5%	-59%
Fossil-based Route + landfill	5.75E-07	1.37E+01	5.73E-01	5.42E-02	2.91E-02	6.27E-07	3.82E-07	3.93E-03	1.98E+01	4.60E+01
Bio-based Route + landfill	1.01E-04	1.35E+01	1.00E+00	1.00E-01	4.48E-02	8.00E-07	1.08E-06	8.36E-03	1.88E+01	1.91E+01

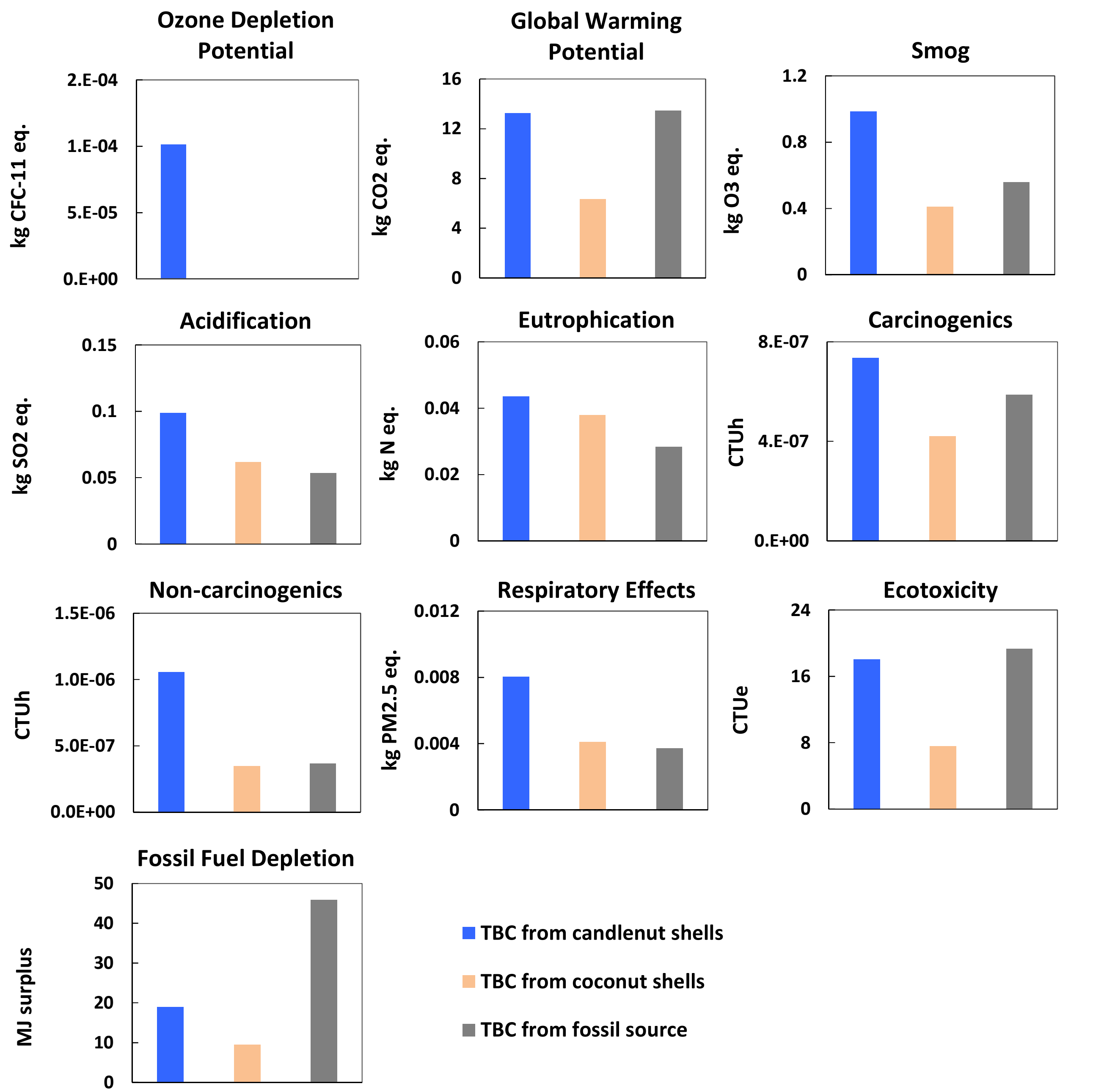

Figure S2. Total environmental impacts for 1 kg TBC from bio-based routes (candlenut shell and coconut shell) and fossil-based route (phenol)

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