Electronic Supplementary Information

Life cycle assessment of enzymatic poly(ethylene terephthalate) recycling

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Contents

Abbreviations

EG	Ethylene glycol
LCA	Life cycle assessment
LPG	Liquified petroleum gas
MRF	Materials recovery facility
MSP	Minimum selling price
PET	Poly(ethylene terephthalate)
TPA	Terephthalic acid

Supporting Figures

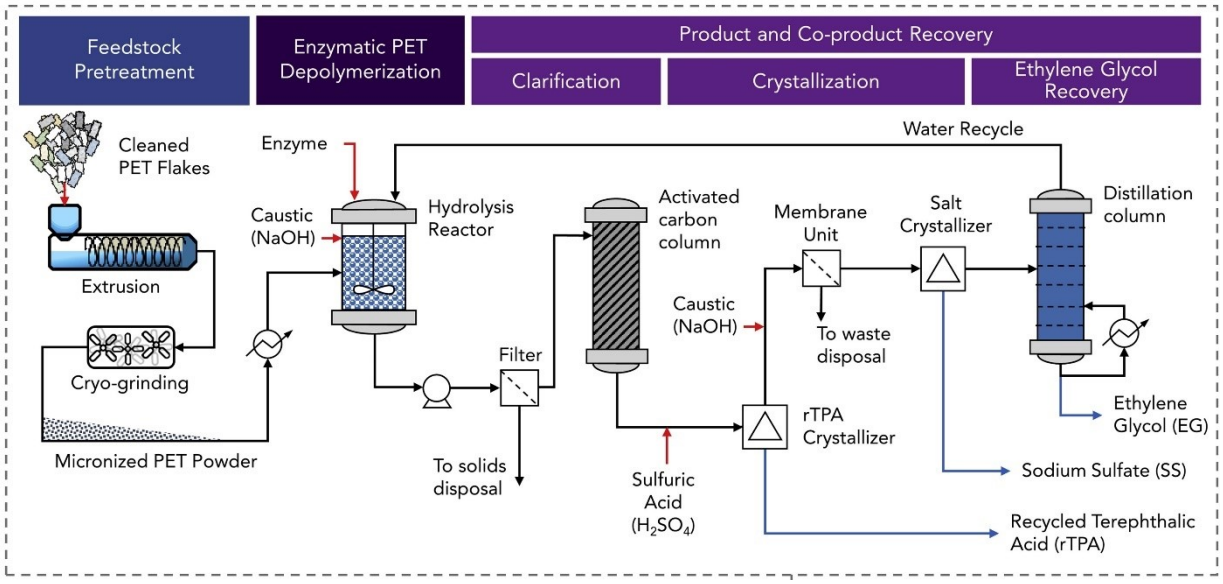

Figure S1. Simplified process flow diagram of the "base case" enzymatic PET recycling process, including PET pre-treatment from post-consumer flakes to smaller amorphized particles, depolymerization to rEG and rTPA, and monomer recovery via downstream processing. Material inputs are shown in red; intermediate, recycle and waste streams are shown in black; and product and co-product streams are shown in blue. Adapted from Singh *et al*.[1]

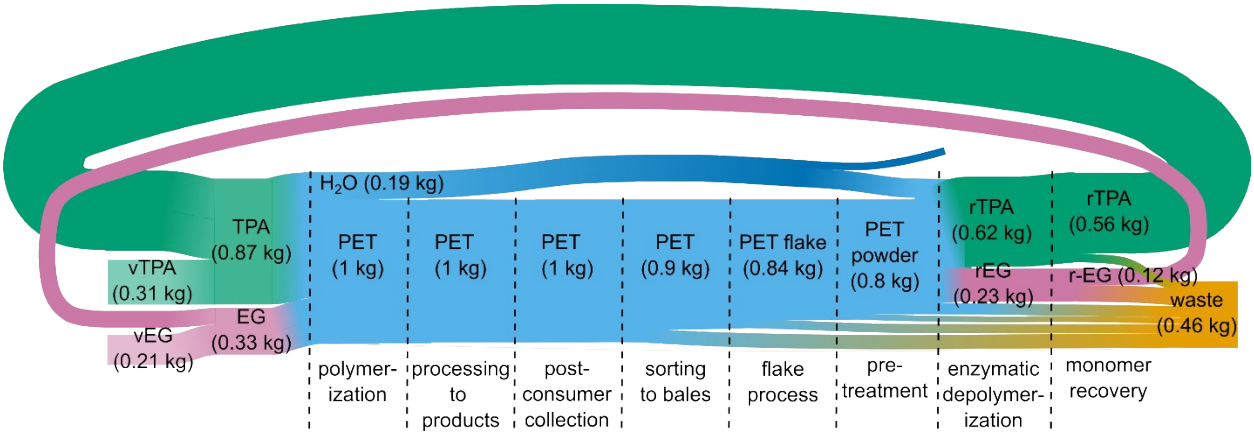

Figure S2. Sankey diagram depicting the terephthalic acid (TPA), ethylene glycol (EG), and poly(ethylene terephthalate) (PET) flows throughout the enzymatic recycling life cycle.

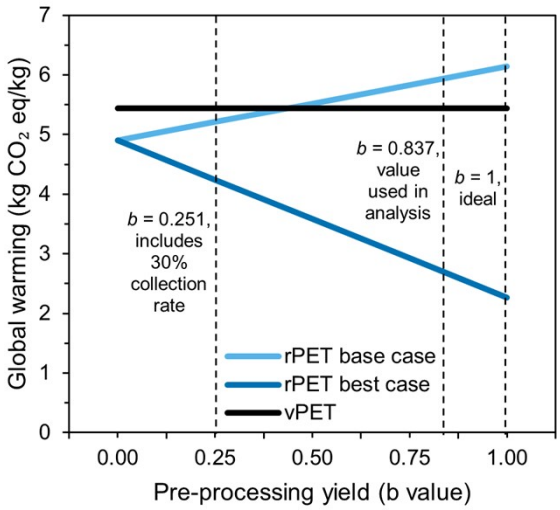

Figure S3. Effect of different pre-processing (collection, sorting and shredding into flake) yield (*b* value) assumptions on the calculated environmental impacts (showcased here as global warming impact) of enzymatically recycled rPET by the system expansion approach. Three cases are highlighted with the vertical dashed lines: *b* = 0.251 (30% collection yield, 90% sorting yield, 93% flake yield) for a more realistic scenario, *b* = 0.837 (100% collection yield, 90% sorting yield, 93% flake yield) for the value used in this analysis, and *b* = 1 (100% collection yield, 100% sorting yield, 100% flake yield) for an ideal scenario. When the impacts of enzymatic recycling are higher than virgin production (base case), their values increase with higher *b* values and thus higher lifetimes. When the impacts of enzymatic recycling are lower than virgin production (best case), their values decrease with higher *b* values and thus higher lifetimes.

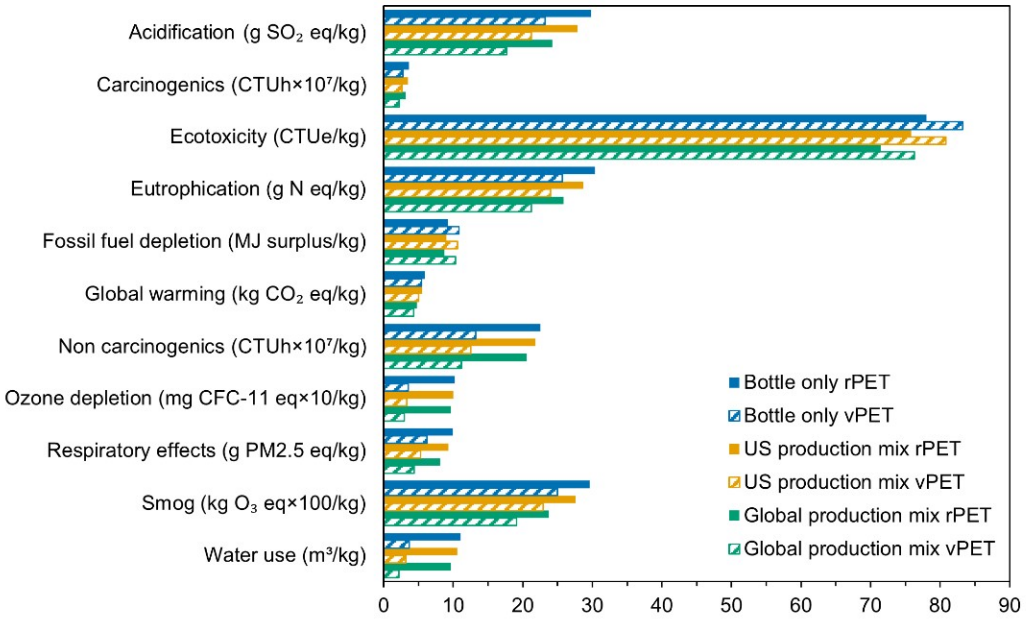

Figure S4. Effect of different PET processing assumptions on the life cycle impacts of enzymatically recycled rPET and virgin vPET, including 100% stretch blow molding to bottles (bottle only); a combination of 13% film, 71% stretch blow molding, and 16% fiber production (US production mix);[2] and a combination of 3% film, 29% stretch blow molding, 53% fiber and 15% chip production (global production mix).[3] The overall trends remain consistent, with impacts decreasing slightly for the US and global production mixes due to their lower use of stretch blow molding (which is the most environmentally impactful processing type).

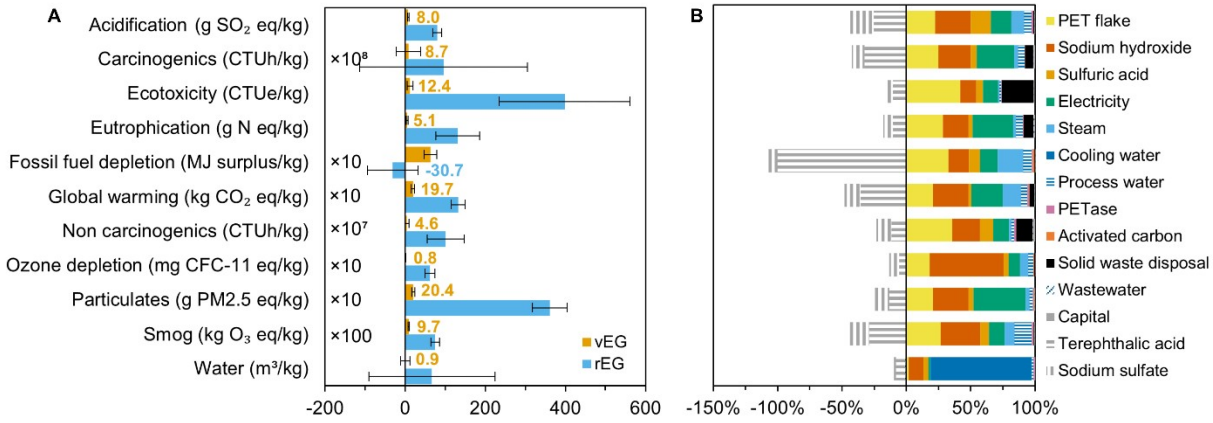

Figure S5. **(A)** Life cycle impacts of enzymatically recycled rEG (blue) and virgin vEG (orange). **(B)** Contribution of different process components to the environmental impacts of rEG.

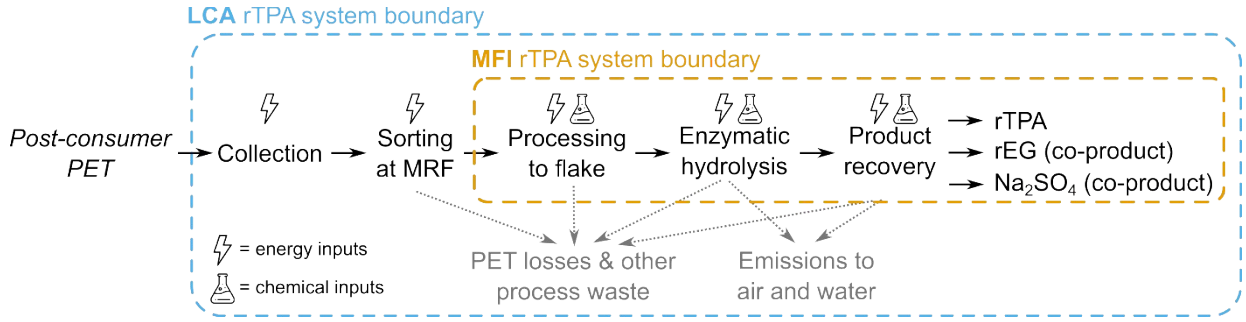

Figure S6. Comparison of the system boundaries used for assessing the impacts of enzymatically recycled rTPA by the previously utilized Materials Flows through Industry (MFI) tool[1] and the current life cycle assessment (LCA). The MFI analysis included shredding of post-consumer PET to flake and all enzymatic hydrolysis and product recovery steps (with U.S.-specific background data). In contrast, the LCA incorporates all of these stages *as well as* post-consumer PET collection and sorting at a Materials Recovery Facility (MRF) and environmental impacts associated with the disposal of process wastes and emission of chemicals to air and water.

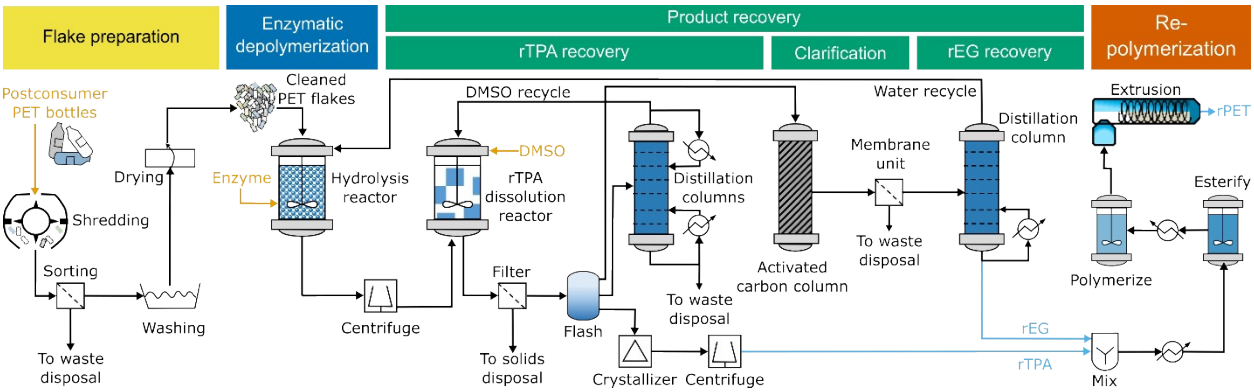

Figure S7. Simplified process flow diagram of the enzymatic PET recycling process using the "moist-solid" system reported by Kaabel *et al.*[4] Post-consumer PET is first shredded into flakes, sorted to remove contaminants, washed, and dried (93% PET yield). The PET flakes are then directly fed into an aqueous hydrolysis reactor (40% solid loading) containing a hydrolase enzyme (30 mg/g PET) at pH 7 and 55°C (90% depolymerization yield). The resulting solids, which are a mixture of precipitated rTPA and unreacted PET, are separated from solution by centrifugation. rTPA is selectively solubilized from this solids mixture in dimethyl sulfoxide (DMSO) at 80°C and 20% loading, flash concentrated, and recovered by crystallization (90% yield). The aqueous phase containing rEG is clarified to remove the enzymes and other contaminants, and then distilled to yield pure rEG (73% yield). The monomers can then be sent to repolymerization into rPET. Inputs are shown in orange and outputs in blue.

Supporting Tables

Table S1. Life cycle inventory of the collection of waste PET for sorting or disposal. Reproduced from a CalRecycle 2011 report.[5]

Category	Item	Quantity	Unit
Product	PET, on tipping floor	1.00	kg
Used products	Baling wire	1.64E−3	kg
	Diesel	5.33E−3	L
	Electricity	0.056	kWh
	Lubricating oil	2.68E−7	kg
	LPG	1.03E−5	L
	Petrol	0.034	L
	Transport, heavy truck	0.119	t*km
	Transport, truck	0.026	t*km
	Water	0.037	kg

Table S2. Life cycle inventory of the sorting of PET at a materials recovery facility. Reproduced from a Franklin Associates 2018 report.[6]

Category	Item	Quantity	Unit
Product	PET bale	1.0	kg
Used products	Diesel	2.0E−3	L
	Electricity	0.014	kWh
	LPG	3.3E−3	L
	Natural gas	3.3E-6	m3
	PET, on tipping floor	1.1	kg
Infrastructure	Sorting facility	4.0E−10	p
Produced waste	Disposal, solid waste, for sanitary landfill	0.1	kg

Table S3. Life cycle inventory of the mechanical preparation of a sorted post-consumer PET bale into flakes. Adapted from a Franklin Associates 2018 report.[6]

Category	Item	Quantity	Unit
Product	PET flake, postconsumer	1.0	kg
Used products	Acid[a]	1.0E−3	kg
	Chemicals with aluminum compounds[b]	6.8E−4	kg
	Defoamants[c]	3.1E−3	kg
	Diesel	6.6E−4	L
	Electricity	0.34	kWh
	Ferric chloride	6.8E−5	kg
	Hydrogen peroxide, 35%	5.4E−6	kg
	LPG	1.1E−3	L
	Natural gas	0.067	m3
	Other confidential chemicals[d]	1.8E−5	kg
	PET bale, from MRF	1.075	kg
	Propane	3.1E−3	L
	Salt[e]	4.8E−4	kg
	Sodium hydroxide, 50%	9.5E−3	kg
	Transport, truck, diesel (for chemicals)	3.3E−3	t*km
	Transport, truck, diesel (for resin)	0.70	t*km
	Washing agent[f]	2.7E−3	kg
	Wastewater treatment polymer[g]	9.9E−5	kg
	Water	0.88	L
Infrastructure	Plastic processing factory	4E−10	p
Emission, air	Particulates, unspecified	7.4E−6	kg
Emission, water	Biological oxygen demand	1.8E−3	kg
	Chemical oxygen demand	1.6E−3	kg
	Dissolved solids, unspecified	3.6E−5	kg
	Suspended solids, unspecified	7.8E−4	kg
Produced waste	Disposal, solid waste (incoming contaminants), to sanitary landfill	0.075	kg
	Disposal, solid waste (process), to sanitary landfill	0.011	kg

[a] assumed to be sulfuric acid
[b] assumed to be aluminum sulfate
[c] assumed to be a silicone product
[d] not included in LCA due to lack of information
[e] assumed to be sodium sulfate
[f] assumed to be soap
[g] assumed to be polyethylene oxide

Table S4. Life cycle inventory of the enzymatic production of terephthalic acid (rTPA) and ethylene glycol (rEG) from post-consumer PET. Included steps are post-consumer PET collection, sorting at a materials recovery facility, mechanical preparation into flakes, feedstock pre-treatment, enzymatic depolymerization and downstream processing (the original production of PET is not considered). Adapted from Singh, et al.[1]

		Per kg terephthalic acid		*Per kg ethylene glycol*	
Category	**Item**	**Quantity**	**Unit**	**Quantity**	**Unit**
By-products	Ethylene glycol	0.2075	kg	n.a.	--
	Sodium sulfate	0.7017	kg	3.381	kg
	Terephthalic acid	n.a.	--	4.818	kg
Used products	Activated carbon	8.109E−3	kg	0.0391	kg
	Electricity	1.756	kWh	8.460	kWh
	Enzyme	7.096E-3	kg	0.0342	kg
	PET flake	1.584	kg	7.631	kg
	Sodium hydroxide, 50%	1.107	kg	5.333	kg
	Steam	4.028	kg	19.41	kg
	Sulfuric acid	0.6847	kg	3.299	kg
	Water, cooling	294.6	kg	1420	kg
	Water, purified	5.748	kg	27.70	kg
Infrastructure	Chemical factory, organics	4E−10	p	4E−10	p
Emission, air	Ethylene glycol	2.238E−6	kg	1.078E−5	kg
Emission, water	Ethylene glycol	0.1685	kg	0.8120	kg
	Sulfuric acid	7.12E−23	kg	3.43E−22	kg
Produced waste	Disposal, solid waste, incineration w/ energy recovery	0.0451	kg	0.2173	kg
	Disposal, solid waste, sanitary landfill	0.1802	kg	0.8681	kg
	Disposal, waste plastic mixture, sanitary landfill	0.0792	kg	0.3816	kg
	Wastewater	5.33E−3	m3	0.0255	m3

Table S5. Life cycle inventory of enzymatically recycled rPET. Included steps are virgin PET production, processing into bottles, sorting at a materials recovery facility, mechanical preparation into flakes, feedstock pre-treatment, enzymatic depolymerization, downstream processing, and re-polymerization of the resulting monomers back into PET.

Category	**Item**	**Quantity**	**Unit**
Product	PET, enzymatically recycled	1.0	kg
Used products	Ethylene glycol	0.143	kg
	PET flake	1.374	kg
	PET repolymerization[a]	1.0	kg
	PET, virgin	1.57	kg
	Purified terephthalic acid, enzymatically produced	0.865	kg
	Stretch blow molding	1.0	kg
	Transport, truck	0.165	t*km
	Transport, truck	0.216	t*km

[a] adapted from the ecoinvent PET global inventory by excluding virgin ethylene glycol and terephthalic acid.

Table S6. Life cycle inventory for PET hydrolase enzyme production. It combines a prepared inventory (labelled with superscript a)[1] and assumptions from a cellulase inventory (labelled with superscript b).[7]

Category	Item	Quantity	Unit
Product	Enzyme[a]	1.0000	kg
Used products	Ammonia, liquid[a]	0.1875	kg
	Ammonium sulfate[a]	0.0417	kg
	Corn oil[a]	0.0313	kg
	Corn steep liquor[a]	0.2813	kg
	Glucose[a]	3.9375	kg
	Hard coal[b]	0.0217	kg
	Magnesium sulfate[a]	0.0417	kg
	Sodium sulfate[a]	0.0417	kg
	Water, purified[b]	0.2085	kg
Infrastructure	Transformation, from pasture and meadow[b]	0.7226	m^2
	Transformation, to industrial area[b]	0.7226	m^2
Emission, air	Carbon dioxide[b]	4.0890	kg
	Ethene[b]	2.0039E−3	kg
	Sulfur dioxide[b]	0.0153	kg
Emission, soil	Phosphate[b]	9.3660E−3	kg

Table S7. Life cycle impacts of enzymatically recycled rTPA and rPET and virgin vTPA and vPET, as calculated by the ReCiPe 2016 Midpoint (I) method.

Impact category	**Unit** (per kg product)	**Base case rTPA**	**vTPA**	**Base case rPET** (lifetime normalized)	**vPET**
Global warming	kg CO_2 eq	5.26	2.55	7.32	6.81
Stratospheric ozone depletion	kg CFC-11 eq	2.63E-6	2.28E-7	2.33E-6	1.12E-6
Ionizing radiation	kBq Co-60 eq	0.541	0.0689	0.690	0.44
Ozone formation, human health	kg NO_x eq	0.00904	0.00363	0.0123	0.0106
Fine particulate matter formation	kg PM2.5 eq	0.00716	0.000913	0.00820	0.00489
Ozone formation, terrestrial ecosystem	kg NO_x eq	0.00918	0.00386	0.0126	0.011
Terrestrial acidification	kg SO_2 eq	0.0195	0.00618	0.0251	0.0193
Freshwater eutrophication	kg P eq	0.0032	0.000413	0.0035	0.002
Terrestrial ecotoxicity	kg 1,4-DCB e	0.00175	0.00045	0.00178	0.00119
Freshwater ecotoxicity	kg 1,4-DCB e	0.343	0.0302	0.282	0.184
Marine ecotoxicity	kg 1,4-DBC e	0.107	0.0103	0.090	0.067
Human carcinogenic toxicity	kg 1,4-DBC e	0.002	0.000523	0.002	0.00129
Human non-carcinogenic toxicity	kg 1,4-DBC e	2.02	0.175	1.66	1.1
Land use	m^2a crop eq	0.0294	0.0267	0.153	0.167
Mineral resource scarcity	kg Cu eq	0.0118	0.00246	0.0123	0.00776
Fossil resource scarcity	kg oil eq	1.2	1.15	2.00	2.09
Water consumption	m^3	0.36	0.0261	0.281	0.0992

Table S8. Scoring rubric for the pedigree approach for estimating uncertainty. All data were given reliability, completeness, temporal correlation, geographical correlation, and further technological correlation scores according to expert judgement. The sum of these scores were then assigned a certain standard deviation: scores of 5-9 were assigned σ = ±5%, 10 to 14 σ = ±10%, 15 to 19 σ = ±15%, 20 to 24 σ = ±20%, and 25 σ = ±25%.

	Score								
Category	**1**	**2**	**3**	**4**	**5**	**Score for enzymatic materials & energy**	**Justification**	**Score for enzymatic emissions**	**Justification**
Reliability	Verified data based on measurements	Verified data partly based on assumptions of non-verified data based on measurements	Non-verified data partly based on qualified estimates	Qualified estimate (by industrial expert)	Non-qualified estimate	4	Inventory estimated with Aspen Plus by a qualified expert[1]	4	Inventory estimated with Aspen Plus by a qualified expert[1]
Completeness	Representative data from all sites relevant for the market considered over an adequate period to even out normal fluctuations	Representative data from >50% of the sites relevant for the market considered over an adequate period to even out normal fluctuations	Representative data from only some sites (<<50%) relevant for the market considered OR >50% of sites but from shorter periods	Representative data from only one site relevant for the market considered OR some sites but from shorter periods	Representativeness unknown or data from a small number of sites AND from shorter periods	4	Short-term experiments from several research groups	5	Emissions estimates based only Aspen Plus modelling
Temporal correlation	Less than 3 years of difference to our reference year	Less than 6 years of difference to our reference year	Less than 10 years of difference to our reference year	Less than 15 years of difference to our reference year	Age of data unknown or more than 15 years of difference to our reference year	1	Data collected within last 3 years[1,8,9]	5	Estimates are not based on hands-on data and therefore age is unknown
Geographical correlation	Data from area under study	Average data from larger area than area under study, or from similar area	Data from smaller area than area under study, or from similar area	Data from area with slightly similar production conditions	Data from unknown OR distinctly different area	3	Data from U.S.-based NREL, which is smaller than the assessed U.S. area	3	Data from U.S.-based NREL, which is smaller than the assessed U.S. area
Further technological correlation	Data from enterprises, processes and materials under study (identical technology)	Data from processes and materials under study but from different enterprises	Data on related processes or materials but same technology, OR data from processes and materials under study but from different technology	Data on related processes or materials but different technology, OR data on laboratory scale processes and same technology	Data on related processes or materials but on laboratory scale of different technology	4	Data from laboratory-scale enzymatic PET recycling	4	Data from laboratory-scale enzymatic PET recycling
					Total Assigned σ	**16 ±15%**		**21 ±20%**	

Table S9. Life cycle assessment results for all terephthalic acid production cases. n.m. = not measured

	Natural environment										*Natural resources*				*Human health*							
	Acidification kg SO_2 eq kg^{-1}		**Ecotoxicity** CTUe kg^{-1}		**Eutroph-ication** kg N eq kg^{-1}		**Global warming** kg CO_2 eq kg^{-1}		**Ozone depletion** kg CFC-11 eq ($\times10^{-6}$) kg^{-1}		**Fossil fuel depletion** MJ surplus kg^{-1}		**Water use** m^3 kg^{-1}		**Carcinogencs** CTUh ($\times10^{-7}$) kg^{-1}		**Non-carcin-ogenics** CTUh ($\times10^{-6}$) kg^{-1}		**Particulates exposure** kg PM2.5 eq ($\times10^{-3}$) kg^{-1}		**Smog** kg O_3 eq kg^{-1}	
Case	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ
Virgin	0.007	0.001	10.3	5.78	0.004	0.002	1.88	0.21	0.083	0.024	7.04	1.07	1.01	7.2	1.12	1.10	0.311	0.306	1.33	0.181	0.081	0.010
Base case	0.023	0.003	93.5	34.9	0.030	0.011	4.27	0.30	1.36	0.278	5.13	0.84	14.7	68.7	3.09	4.52	2.45	2.10	8.52	0.935	0.220	0.022
Ammonia	0.017	0.003	73.4	66.4	0.023	0.009	3.16	0.35	0.674	0.224	5.28	0.88	16.2	58.0	1.88	3.74	1.61	0.90	6.18	0.871	0.146	0.023
“Best” case	0.008	0.001	30.4	16.0	0.007	0.003	0.90	0.19	0.785	0.161	0.83	0.59	7.19	21.9	0.969	1.56	0.980	0.676	1.60	0.312	0.078	0.012
Depolymerization – 80%	0.025	n.m.	105	n.m.	0.034	n.m.	4.80	n.m.	1.44	n.m.	5.89	n.m.	16.3	n.m.	3.47	n.m.	2.70	n.m.	9.41	n.m.	0.244	n.m.
Depolymerization – 99%	0.021	n.m.	85.3	n.m.	0.028	n.m.	3.88	n.m.	1.30	n.m.	4.57	n.m.	13.5	n.m.	2.85	n.m.	2.26	n.m.	7.86	n.m.	0.202	n.m.
EG recovery – 40%	0.023	n.m.	94.0	n.m.	0.031	n.m.	4.35	n.m.	1.36	n.m.	5.39	n.m.	14.7	n.m.	3.13	n.m.	2.46	n.m.	8.60	n.m.	0.224	n.m.
EG recovery – 65%	0.022	n.m.	92.6	n.m.	0.030	n.m.	4.14	n.m.	1.35	n.m.	4.73	n.m.	14.6	n.m.	3.03	n.m.	2.42	n.m.	8.39	n.m.	0.214	n.m.
EG selective membrane	0.021	0.003	88.2	32.2	0.029	0.012	3.77	0.31	1.32	0.285	3.83	0.95	13.4	67.9	2.89	5.70	2.31	1.68	8.10	0.922	0.199	0.023
Enzyme load – 1	0.023	n.m.	93.2	n.m.	0.030	n.m.	4.21	n.m.	1.36	n.m.	5.10	n.m.	14.6	n.m.	3.07	n.m.	2.41	n.m.	8.45	n.m..	0.218	n.m.
Enzyme load – 10	0.023	n.m.	93.8	n.m.	0.031	n.m.	4.33	n.m.	1.36	n.m.	5.16	n.m.	14.8	n.m.	3.11	n.m.	2.48	n.m.	8.59	n.m.	0.222	n.m.
Moist-solid	0.011	0.002	80.2	43.5	0.014	0.004	2.20	0.38	0.398	0.125	2.35	0.70	9.28	17.5	1.48	1.68	1.84	0.954	2.80	0.340	0.100	0.014
Moist-solid + renewables	0.011	0.001	79.7	36.0	0.014	0.004	2.13	0.38	0.390	0.178	2.30	0.68	9.27	15.4	1.43	0.676	1.82	1.06	2.56	0.302	0.098	0.011
No EG recovery	0.022	0.003	96.0	35.5	0.032	0.012	4.06	0.29	1.31	0.295	5.28	0.75	6.40	72.9	3.24	5.20	2.53	1.48	8.76	0.940	0.236	0.026
No EG/Na_2SO_4 recovery	0.029	0.003	254	130	0.045	0.016	6.05	0.54	1.45	0.282	5.93	0.76	6.83	79.5	5.08	15.5	4.93	2.56	9.84	1.01	0.292	0.027
No mechanical pre-treat	0.019	0.002	83.8	34.0	0.022	0.007	3.18	0.28	1.24	0.251	4.32	0.76	14.5	40.7	2.24	4.22	2.16	1.42	5.12	0.531	0.192	0.022
PET content – 91%	0.024	n.m.	100	n.m.	0.032	n.m.	4.46	n.m.	1.39	n.m.	5.40	n.m.	15.2	n.m.	3.20	n.m.	2.58	n.m.	8.84	n.m.	0.228	n.m.
PET content – 99%	0.022	n.m.	87.4	n.m.	0.028	n.m.	4.09	n.m.	1.33	n.m.	4.88	n.m.	14.3	n.m.	2.98	n.m.	2.32	n.m.	8.22	n.m.	0.212	n.m.
pH – 6	0.023	n.m.	93.4	n.m.	0.030	n.m.	4.26	n.m.	1.36	n.m.	5.13	n.m.	14.7	n.m.	3.09	n.m.	2.44	n.m.	8.51	n.m.	0.220	n.m.
pH – 10	0.023	n.m.	93.5	n.m.	0.030	n.m.	4.27	n.m.	1.36	n.m.	5.13	n.m.	14.7	n.m.	3.09	n.m.	2.45	n.m.	8.52	n.m.	0.220	n.m.
Plant size – 50 tpd	0.025	n.m.	94.5	n.m.	0.031	n.m.	4.90	n.m.	1.44	n.m.	6.29	n.m.	21.6	n.m.	3.20	n.m.	2.49	n.m.	8.83	n.m.	0.240	n.m.
Plant size – 300 tpd	0.023	n.m.	93.4	n.m.	0.030	n.m.	4.26	n.m.	1.36	n.m.	5.12	n.m.	14.7	n.m.	3.08	n.m.	2.44	n.m.	8.50	n.m.	0.220	n.m.
Renewable electricity	0.017	0.002	80.4	31.4	0.017	0.007	2.68	0.27	1.19	0.247	3.91	0.75	14.5	46.8	2.02	2.34	2.04	1.10	3.31	0.469	0.182	0.022
Residence time – 10 h	0.023	n.m.	93.5	n.m.	0.030	n.m.	4.27	n.m.	1.36	n.m.	5.13	n.m.	14.7	n.m.	3.09	n.m.	2.45	n.m.	8.52	n.m.	0.220	n.m.
Residence time – 240 h	0.023	n.m.	93.5	n.m.	0.030	n.m.	4.27	n.m.	1.33	n.m.	5.13	n.m.	14.7	n.m.	3.09	n.m.	2.45	n.m.	8.52	n.m.	0.220	n.m.
Solids load – 10	0.028	n.m.	114	n.m.	0.034	n.m.	5.43	n.m.	1.51	n.m.	6.78	n.m.	21.9	n.m.	3.59	n.m.	2.86	n.m.	9.45	n.m.	0.271	n.m.
Solids load – 20	0.020	n.m.	83.4	n.m.	0.028	n.m.	3.69	n.m.	1.28	n.m.	4.31	n.m.	11.0	n.m.	2.84	n.m.	2.24	n.m.	8.05	n.m.	0.194	n.m.
Sorting yield – 69%	0.023	n.m.	132	n.m.	0.034	n.m.	4.46	n.m.	1.38	n.m.	5.28	n.m.	14.7	n.m.	3.18	n.m.	2.99	n.m.	8.60	n.m.	0.223	n.m.
Sorting yield – 89%	0.023	n.m.	69.7	n.m.	0.028	n.m.	4.15	n.m.	1.35	n.m.	5.03	n.m.	14.6	n.m.	3.03	n.m.	2.11	n.m.	8.46	n.m.	0.218	n.m.
Temperature – 40°C	0.021	n.m.	93.1	n.m.	0.030	n.m.	4.23	n.m.	1.18	n.m.	4.58	n.m.	14.7	n.m.	3.08	n.m.	2.43	n.m.	8.24	n.m.	0.224	n.m.
Temperature – 80°C	0.021	n.m.	93.1	n.m.	0.030	n.m.	4.23	n.m.	1.18	n.m.	4.58	n.m.	14.7	n.m.	3.08	n.m.	2.43	n.m.	8.24	n.m.	0.224	n.m.
TPA recovery – 80%	0.026	n.m.	105	n.m.	0.034	n.m.	4.79	n.m.	1.53	n.m.	5.76	n.m.	16.5	n.m.	3.45	n.m.	2.73	n.m.	9.56	n.m.	0.247	n.m.
TPA recovery – 98%	0.022	n.m.	88.8	n.m.	0.029	n.m.	4.04	n.m.	1.29	n.m.	4.86	n.m.	13.9	n.m.	2.94	n.m.	2.33	n.m.	8.08	n.m.	0.209	n.m.
Water recycle	0.021	0.002	90.1	33.3	0.028	0.011	3.84	0.29	1.30	0.276	4.21	0.78	14.0	64.6	2.87	4.32	2.32	1.29	8.09	0.950	0.181	0.018

Table S10. Life cycle assessment results for all ethylene glycol production cases.

	Natural environment										*Natural resources*				*Human health*							
	Acidification kg SO_2 eq kg^{-1}		**Ecotoxicity** CTUe kg^{-1}		**Eutrophication** kg N eq kg^{-1}		**Global warming** kg CO_2 eq kg^{-1}		**Ozone depletion** kg CFC-11 eq ($\times10^{-6}$) kg^{-1}		**Fossil fuel depletion** MJ surplus kg^{-1}		**Water use** m^3 kg^{-1}		**Carcinogencs** CTUh ($\times10^{-7}$) kg^{-1}		**Non-carcinogenics** CTUh ($\times10^{-6}$) kg^{-1}		**Particulates exposure** kg PM2.5 eq ($\times10^{-3}$) kg^{-1}		**Smog** kg O_3 eq kg^{-1}	
Case	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ
Virgin	0.008	0.001	12.4	7.63	0.005	0.003	1.97	0.381	0.082	0.026	6.32	1.61	1.03	22.7	0.871	0.304	0.456	0.535	2.04	0.388	0.097	0.017
Base case	0.080	0.012	398	163	0.131	0.055	13.2	1.69	6.20	1.27	−3.07	6.31	66.7	315	9.62	20.9	10.1	4.62	36.1	4.30	0.748	0.108

Table S11. Contribution of all process components to the life cycle impacts of enzymatically recycled terephthalic acid.

	Natural environment					*Natural resources*		*Human health*			
Component	**Acidification** %	**Ecotoxicity** %	**Eutrophication** %	**Global warming** %	**Ozone depletion** %	**Fossil fuel depletion** %	**Water use** %	**Carcinogenics** %	**Non-carcinogenics** %	**Particulates exposure** %	**Smog** %
Activated carbon	0.29	0.04	0.09	0.39	0.79	1.47	0.0	0.09	0.07	0.16	0.26
Sulfuric acid	15.3	5.05	3.73	2.07	3.86	8.15	4.07	4.37	9.82	3.66	6.82
Sodium hydroxide	27.0	12.2	21.4	27.6	57.3	16.1	11.3	24.1	20.6	27.4	30.3
Cooling water	0.03	0.04	0.03	0.04	0.01	0.03	78.2	0.14	0.04	0.03	0.05
Process water	5.40	1.49	5.92	4.90	3.67	6.76	1.49	4.55	2.11	2.51	13.4
Steam	9.66	1.01	2.25	14.1	6.24	19.84	0.25	3.01	1.58	3.02	7.74
Capital	1.91	3.77	2.61	1.21	0.31	0.68	0.11	5.25	5.92	1.34	1.53
Enzyme	1.35	0.34	0.63	1.32	0.19	0.45	0.89	0.52	1.40	0.77	0.94
PET flake	22.2	40.5	31.2	20.4	18.0	32.5	1.81	23.9	33.8	20.5	26.5
Electricity	15.9	11.3	34.2	24.1	9.14	13.8	1.38	27.8	12.0	40.1	11.6
Sodium sulfate	−17.7	−5.28	−7.79	−12.1	−7.71	−8.98	−2.21	−9.40	−11.2	−10.8	−15.4
Ethylene glycol	−5.57	−2.54	−3.48	−7.69	−1.14	−18.6	−1.38	−5.01	−3.31	−4.22	−7.08
Solid waste	0.81	24.0	0.16	3.88	0.54	0.25	0.13	6.05	12.1	0.47	0.84
Wastewater	0.09	0.07	0.55	0.06	0.02	0.04	−1.35	0.25	0.56	0.05	0.09

Table S12. Life cycle inventory of the complete cradle-to-grave life cycle of virgin PET. Included steps are PET production, processing into bottles, waste collection, and disposal by landfill (80%) and incineration (20%).

Category	Item	Quantity	Unit
Product	Poly(ethylene terephthalate), full life cycle	1.00	kg
Used products	Poly(ethylene terephthalate), virgin resin	1.00	kg
	Stretch blow moulding	1.00	kg
	Poly(ethylene terephthalate), collection	1.00	kg
Produced waste	Disposal, PET, to sanitary landfill	0.800	kg
	Disposal, PET, to incineration w/ energy recovery	0.200	kg

Table S13. Raw data for the life cycle assessment of all PET production and recycling cases (no systems expansion).

	Natural environment										*Natural resources*				*Human health*							
	Acidification		**Ecotoxicity**		**Eutroph-ication**		**Global warming**		**Ozone depletion**		**Fossil fuel depletion**		**Water use**		**Carcinogencs**		**Non-carcin-ogenics**		**Particulates exposure**		**Smog**	
	kg SO_2 eq kg^{-1}		CTUe kg^{-1}		kg N eq kg^{-1}		kg CO_2 eq kg^{-1}		kg CFC-11 eq ($\times10^{-6}$) kg^{-1}		MJ surplus kg^{-1}		m^3 kg^{-1}		CTUh ($\times10^{-7}$) kg^{-1}		CTUh ($\times10^{-6}$) kg^{-1}		kg PM2.5 eq ($\times10^{-3}$) kg^{-1}		kg O_3 eq kg^{-1}	
Case	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ
Virgin	0.023	0.003	83.2	41.4	0.026	0.009	5.44	0.52	0.353	0.091	10.8	1.66	3.68	66.3	2.78	3.46	1.32	0.810	6.21	0.808	0.250	0.026
Virgin (no disposal)	0.023	n.m.	32.8	n.m.	0.016	n.m.	4.90	n.m.	0.314	n.m.	10.5	n.m.	3.61	n.m.	2.70	n.m.	1.10	n.m.	6.05	n.m.	0.241	n.m.
Base case	0.034	0.003	103	38.9	0.039	0.017	6.51	0.45	1.41	0.312	8.52	0.89	15.2	106	4.20	4.12	2.89	2.00	12.1	1.29	0.327	0.027
Ammonia	0.029	0.003	86.1	30.5	0.032	0.015	5.56	0.43	0.822	0.280	8.65	0.93	16.6	77.6	3.15	3.00	2.17	1.22	10.1	1.08	0.263	0.025
“Best” case	0.013	0.002	42.2	19.1	0.011	0.006	1.88	0.17	0.802	0.174	3.62	0.42	8.52	45.4	1.74	3.28	1.36	1.03	3.02	0.376	0.122	0.012
Depolymerization – 80%	0.036	n.m.	113	n.m.	0.041	n.m.	6.97	n.m.	1.49	n.m.	9.18	n.m.	16.6	n.m.	4.48	n.m.	3.12	n.m.	12.9	n.m.	0.348	n.m.
Depolymerization – 99%	0.032	n.m.	96.3	n.m.	0.036	n.m.	6.17	n.m.	1.36	n.m.	8.04	n.m.	14.2	n.m.	3.99	n.m.	2.73	n.m.	11.6	n.m.	0.311	n.m.
EG recovery – 40%	0.034	n.m.	104	n.m.	0.039	n.m.	6.58	n.m.	1.42	n.m.	8.75	n.m.	15.3	n.m.	4.23	n.m.	2.91	n.m.	12.2	n.m.	0.330	n.m.
EG recovery – 65%	0.033	n.m.	103	n.m.	0.038	n.m.	6.40	n.m.	1.41	n.m.	8.18	n.m.	15.2	n.m.	4.23	n.m.	2.87	n.m.	12.0	n.m.	0.322	n.m.
EG selective membrane	0.032	0.003	98.9	37.4	0.038	0.015	6.08	0.45	1.39	0.313	7.40	0.80	14.2	101	4.03	5.86	2.78	2.16	11.8	1.26	0.309	0.028
Enzyme load – 1	0.034	n.m.	103	n.m.	0.038	n.m.	6.46	n.m.	1.41	n.m.	8.50	n.m.	15.2	n.m.	4.19	n.m.	2.86	n.m.	12.1	n.m.	0.325	n.m.
Enzyme load – 10	0.034	n.m.	104	n.m.	0.039	n.m.	6.57	n.m.	1.42	n.m.	8.55	n.m.	15.4	n.m.	4.21	n.m.	2.93	n.m.	12.2	n.m.	0.329	n.m.
Moist-solid	0.024	0.003	91.6	35.3	0.025	0.010	4.72	0.45	0.581	0.188	6.10	0.58	10.6	58.9	2.80	3.28	2.36	1.10	7.18	0.822	0.222	0.020
Moist-solid + renewables	0.013	0.002	79.4	31.0	0.012	0.004	2.36	0.33	0.408	0.140	4.15	0.41	10.2	34.2	1.80	0.94	1.96	1.09	2.15	2.31	0.122	0.011
No EG recovery	0.033	0.003	106	39.6	0.040	0.017	6.33	0.45	1.37	0.313	8.65	1.00	8.10	105	4.33	5.68	2.96	3.14	12.4	1.25	0.341	0.030
No EG/Na_2SO_4 recovery	0.039	0.004	242	131	0.051	0.019	8.05	0.65	1.50	0.343	9.21	1.03	8.47	110	5.92	8.04	5.04	2.92	13.3	1.37	0.389	0.034
No mechanical pre-treat	0.030	0.003	95.0	38.0	0.031	0.012	5.57	0.43	1.31	0.258	7.81	0.81	15.1	84.7	3.46	5.22	2.64	1.58	9.20	1.03	0.303	0.028
PET content – 91%	0.035	n.m.	109	n.m.	0.040	n.m.	6.68	n.m.	1.44	n.m.	8.76	n.m.	15.7	n.m.	4.30	n.m.	3.01	n.m.	12.4	n.m.	0.334	n.m.
PET content – 99%	0.033	n.m.	98.2	n.m.	0.037	n.m.	6.36	n.m.	1.39	n.m.	8.30	n.m.	15.0	n.m.	4.11	n.m.	2.78	n.m.	11.9	n.m.	0.320	n.m.
pH – 6	0.034	n.m.	103	n.m.	0.039	n.m.	6.51	n.m.	1.41	n.m.	8.52	n.m.	15.2	n.m.	4.20	n.m.	2.89	n.m.	12.1	n.m.	0.327	n.m.
pH – 10	0.034	n.m.	103	n.m.	0.039	n.m.	6.51	n.m.	1.41	n.m.	8.52	n.m.	15.2	n.m.	4.20	n.m.	2.89	n.m.	12.1	n.m.	0.327	n.m.
Plant size – 50 tpd	0.036	n.m.	104	n.m.	0.039	n.m.	7.06	n.m.	1.48	n.m.	9.52	n.m.	21.3	n.m.	4.29	n.m.	2.93	n.m.	12.4	n.m.	0.344	n.m.
Plant size – 300 tpd	0.034	n.m.	103	n.m.	0.039	n.m.	6.50	n.m.	1.41	n.m.	8.51	n.m.	15.2	n.m.	4.19	n.m.	2.89	n.m.	12.1	n.m.	0.327	n.m.
Renewable electricity	0.020	0.002	85.6	35.0	0.020	0.006	3.42	0.30	1.16	0.258	6.30	0.72	14.8	66.4	2.65	1.84	2.30	1.72	0.451	0.513	0.212	0.021
Residence time – 10 h	0.034	n.m.	103	n.m.	0.039	n.m.	6.51	n.m.	1.41	n.m.	8.52	n.m.	15.2	n.m.	4.20	n.m.	2.89	n.m.	12.1	n.m.	0.327	n.m.
Residence time – 240 h	0.035	n.m.	104	n.m.	0.039	n.m.	6.54	n.m.	1.53	n.m.	8.90	n.m.	15.20	n.m.	4.21	n.m.	2.90	n.m.	12.3	n.m.	0.324	n.m.
Solids load – 10	0.038	n.m.	122	n.m.	0.042	n.m.	7.52	n.m.	1.55	n.m.	9.95	n.m.	21.5	n.m.	4.63	n.m.	3.25	n.m.	13.0	n.m.	0.371	n.m.
Solids load – 20	0.032	n.m.	94.7	n.m.	0.037	n.m.	6.01	n.m.	1.35	n.m.	7.81	n.m.	12.10	n.m.	3.99	n.m.	2.72	n.m.	11.7	n.m.	0.305	n.m.
Sorting yield – 69%	0.034	n.m.	137	n.m.	0.042	n.m.	6.68	n.m.	1.43	n.m.	8.66	n.m.	15.3	n.m.	4.28	n.m.	3.37	n.m.	12.2	n.m.	0.330	n.m.
Sorting yield – 89%	0.034	n.m.	82.6	n.m.	0.036	n.m.	6.41	n.m.	1.40	n.m.	8.43	n.m.	15.2	n.m.	4.15	n.m.	2.60	n.m.	12.1	n.m.	0.325	n.m.
Temperature – 40°C	0.035	n.m.	104	n.m.	0.039	n.m.	6.54	n.m.	1.53	n.m.	8.90	n.m.	15.2	n.m.	4.21	n.m.	2.90	n.m.	12.3	n.m.	0.324	n.m.
Temperature – 80°C	0.035	n.m.	104	n.m.	0.039	n.m.	6.54	n.m.	1.53	n.m.	8.90	n.m.	15.2	n.m.	4.21	n.m.	2.90	n.m.	12.3	n.m.	0.324	n.m.
TPA recovery – 80%	0.036	n.m.	113	n.m.	0.042	n.m.	6.96	n.m.	1.56	n.m.	9.07	n.m.	16.8	n.m.	4.51	n.m.	3.14	n.m.	13.1	n.m.	0.350	n.m.
TPA recovery – 98%	0.033	n.m.	99.3	n.m.	0.037	n.m.	6.32	n.m.	1.35	n.m.	8.29	n.m.	14.6	n.m.	4.07	n.m.	2.79	n.m.	11.8	n.m.	0.317	n.m.
Water recycle	0.032	0.003	101	46.8	0.037	0.015	6.14	0.11	1.36	0.299	7.73	0.77	14.7	97.2	4.00	5.66	2.78	2.18	11.8	1.26	0.294	0.024

Table S14. Life cycle assessment results of all PET recycling cases after adjustment by the system expansion technique.

	Natural environment										*Natural resources*				*Human health*							
	Acidification kg SO_2 eq kg^{-1}		**Ecotoxicity** CTUe kg^{-1}		**Eutroph-ication** kg N eq kg^{-1}		**Global warming** kg CO_2 eq kg^{-1}		**Ozone depletion** kg CFC-11 eq ($\times10^{-6}$) kg^{-1}		**Fossil fuel depletion** MJ surplus kg^{-1}		**Water use** m^3 kg^{-1}		**Carcinogencs** CTUh ($\times10^{-7}$) kg^{-1}		**Non-carcin-ogenics** CTUh ($\times10^{-6}$) kg^{-1}		**Particulates exposure** kg PM2.5 eq ($\times10^{-3}$) kg^{-1}		**Smog** kg O_3 eq kg^{-1}	
Case	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ	Mean	±σ
Base case	0.030	0.003	78.0	38.9	0.030	0.017	5.94	0.45	1.02	0.312	9.23	0.89	11.1	106	3.67	4.12	2.25	2.00	9.95	1.29	0.296	0.027
Ammonia	0.026	0.003	67.1	30.5	0.026	0.015	5.32	0.43	0.641	0.280	9.31	0.93	12.0	77.6	2.99	3.00	1.79	1.22	8.66	1.08	0.255	0.025
“Best” case	0.015	0.002	39.9	19.1	0.012	0.006	2.63	0.17	0.681	0.174	5.32	0.42	7.30	45.4	1.98	3.28	1.30	1.03	3.77	0.376	0.151	0.012
Depolymerization – 80%	0.030	n.m.	78.7	n.m.	0.030	n.m.	6.08	n.m.	0.987	n.m.	9.74	n.m.	11.1	n.m.	3.72	n.m.	2.26	n.m.	9.97	n.m.	0.302	n.m.
Depolymerization – 99%	0.029	n.m.	77.8	n.m.	0.030	n.m.	5.80	n.m.	1.05	n.m.	8.76	n.m.	11.1	n.m.	3.61	n.m.	2.25	n.m.	9.98	n.m.	0.291	n.m.
EG recovery – 40%	0.030	n.m.	78.7	n.m.	0.031	n.m.	5.98	n.m.	1.03	n.m.	9.37	n.m.	11.1	n.m.	3.69	n.m.	2.27	n.m.	10.0	n.m.	0.298	n.m.
EG recovery – 65%	0.030	n.m.	78.0	n.m.	0.030	n.m.	5.87	n.m.	1.02	n.m.	9.01	n.m.	11.1	n.m.	3.63	n.m.	2.24	n.m.	9.88	n.m.	0.293	n.m.
EG selective membrane	0.029	0.003	74.8	37.4	0.030	0.015	5.65	0.45	0.997	0.313	8.53	0.80	10.3	101	3.54	5.86	2.17	2.16	9.70	1.26	0.284	0.028
Enzyme load – 1	0.030	n.m.	78.0	n.m.	0.030	n.m.	5.90	n.m.	1.02	n.m.	9.21	n.m.	11.1	n.m.	3.66	n.m.	2.23	n.m.	9.95	n.m.	0.295	n.m.
Enzyme load – 10	0.030	n.m.	78.7	n.m.	0.031	n.m.	5.98	n.m.	1.03	n.m.	9.24	n.m.	11.2	n.m.	3.67	n.m.	2.28	n.m.	10.0	n.m.	0.298	n.m.
Moist-solid	0.023	0.003	70.7	35.3	0.021	0.010	4.78	0.45	0.486	0.188	7.67	0.58	8.11	58.9	2.76	3.28	1.91	1.10	6.78	0.822	0.229	0.020
Moist-solid + renewables	0.016	0.002	62.8	31.0	0.014	0.004	3.26	0.33	0.375	0.140	6.41	0.41	7.85	34.2	2.12	0.94	1.65	1.09	3.54	2.31	0.164	0.011
No EG recovery	0.030	0.003	79.9	39.6	0.031	0.017	5.82	0.45	0.994	0.313	9.31	1.00	6.50	105	3.75	5.68	2.30	3.14	10.1	1.25	0.305	0.030
No EG/Na_2SO_4 recovery	0.033	0.004	167.5	131	0.038	0.019	6.93	0.65	1.08	0.343	9.67	1.03	6.74	110	4.77	8.04	3.64	2.92	10.7	1.37	0.336	0.034
No mechanical pre-treat	0.028	0.003	72.9	38.0	0.025	0.012	5.33	0.43	0.955	0.258	8.77	0.81	11.0	84.7	3.19	5.22	2.09	1.58	8.08	1.03	0.281	0.028
PET content – 91%	0.030	n.m.	79.8	n.m.	0.031	n.m.	6.00	n.m.	1.01	n.m.	9.43	n.m.	11.1	n.m.	3.69	n.m.	2.28	n.m.	9.97	n.m.	0.298	n.m.
PET content – 99%	0.030	n.m.	76.7	n.m.	0.030	n.m.	5.88	n.m.	1.04	n.m.	9.02	n.m.	11.3	n.m.	3.65	n.m.	2.23	n.m.	9.98	n.m.	0.294	n.m.
pH – 6	0.030	n.m.	78.0	n.m.	0.030	n.m.	5.94	n.m.	1.02	n.m.	9.23	n.m.	11.1	n.m.	3.67	n.m.	2.25	n.m.	9.95	n.m.	0.296	n.m.
pH – 10	0.030	n.m.	78.0	n.m.	0.030	n.m.	5.94	n.m.	1.02	n.m.	9.23	n.m.	11.1	n.m.	3.67	n.m.	2.25	n.m.	9.95	n.m.	0.296	n.m.
Plant size – 50 tpd	0.031	n.m.	78.6	n.m.	0.031	n.m.	6.29	n.m.	1.06	n.m.	9.87	n.m.	15.0	n.m.	3.72	n.m.	2.28	n.m.	10.1	n.m.	0.307	n.m.
Plant size – 300 tpd	0.030	n.m.	78.0	n.m.	0.030	n.m.	5.93	n.m.	1.02	n.m.	9.22	n.m.	11.1	n.m.	3.66	n.m.	2.25	n.m.	9.95	n.m.	0.296	n.m.
Renewable electricity	0.021	0.002	66.8	35.0	0.018	0.006	3.95	0.30	0.859	0.258	7.80	0.72	10.8	66.4	2.67	1.84	1.87	1.72	5.06	0.513	0.222	0.021
Residence time – 10 h	0.030	n.m.	78.0	n.m.	0.030	n.m.	5.94	n.m.	1.02	n.m.	9.23	n.m.	11.1	n.m.	3.67	n.m.	2.25	n.m.	9.95	n.m.	0.296	n.m.
Residence time – 240 h	0.031	n.m.	78.7	n.m.	0.030	n.m.	5.96	n.m.	1.10	n.m.	9.47	n.m.	11.1	n.m.	3.67	n.m.	2.26	n.m.	10.1	n.m.	0.294	n.m.
Solids load – 10	0.033	n.m.	90.2	n.m.	0.033	n.m.	6.59	n.m.	1.11	n.m.	10.2	n.m.	15.1	n.m.	3.94	n.m.	2.48	n.m.	10.5	n.m.	0.325	n.m.
Solids load – 20	0.028	n.m.	72.7	n.m.	0.029	n.m.	5.61	n.m.	0.981	n.m.	8.77	n.m.	9.08	n.m.	3.53	n.m.	2.14	n.m.	9.69	n.m.	0.282	n.m.
Sorting yield – 69%	0.029	n.m.	88.0	n.m.	0.030	n.m.	5.84	n.m.	0.905	n.m.	9.53	n.m.	9.80	n.m.	3.54	n.m.	2.30	n.m.	9.31	n.m.	0.288	n.m.
Sorting yield – 89%	0.031	n.m.	70.7	n.m.	0.031	n.m.	6.05	n.m.	1.14	n.m.	8.92	n.m.	12.4	n.m.	3.80	n.m.	2.24	n.m.	10.7	n.m.	0.305	n.m.
Temperature – 40°C	0.031	n.m.	78.7	n.m.	0.030	n.m.	5.96	n.m.	1.10	n.m.	9.47	n.m.	11.1	n.m.	3.67	n.m.	2.26	n.m.	10.1	n.m.	0.294	n.m.
Temperature – 80°C	0.031	n.m.	78.7	n.m.	0.030	n.m.	5.96	n.m.	1.10	n.m.	9.47	n.m.	11.1	n.m.	3.67	n.m.	2.26	n.m.	10.1	n.m.	0.294	n.m.
TPA recovery – 80%	0.030	n.m.	78.7	n.m.	0.031	n.m.	6.08	n.m.	1.03	n.m.	9.68	n.m.	11.1	n.m.	3.74	n.m.	2.27	n.m.	10.1	n.m.	0.303	n.m.
TPA recovery – 98%	0.030	n.m.	79.2	n.m.	0.031	n.m.	5.89	n.m.	1.04	n.m.	8.96	n.m.	11.3	n.m.	3.66	n.m.	2.28	n.m.	10.1	n.m.	0.294	n.m.
Water recycle	0.028	0.003	76.2	46.8	0.029	0.015	5.69	0.11	0.980	0.299	8.74	0.77	10.7	97.2	3.53	5.66	2.17	2.18	9.71	1.26	0.275	0.024

Table S15. Yields associated with (a) the mass ratio of rTPA and rEG recovered from the enzymatic depolymerization process (beginning from the clean PET flake); (b) the recovery rate of PET flake from all pre-treatment (collection, sorting, and shredding to flake) steps; and (c) the mass ratio of enzymatically produced rTPA and rEG contained in the secondary produced rPET. These values were used to calculate the aggregate number of product lifetimes (L) for enzymatically recycled rPET, as described in the methods section.

Case	a	b	c	L
Base case	0.677	0.837	0.879	2.81
Ammonia	0.677	0.837	0.879	2.81
“Best” case	0.869	0.837	0.966	4.04
Depolymerization – 80%	0.601	0.837	0.879	2.34
Depolymerization – 99%	0.744	0.837	0.879	3.43
EG recovery – 40%	0.653	0.837	0.849	2.81
EG recovery – 65%	0.711	0.837	0.925	2.81
EG selective membrane	0.758	0.837	0.999	2.74
Enzyme load – 1	0.677	0.837	0.879	2.81
Enzyme load – 10	0.677	0.837	0.879	2.81
Moist-solid	0.730	0.837	0.949	2.81
Moist-solid + renewables	0.730	0.837	0.949	2.81
No EG recovery	0.560	0.837	0.728	2.81
No EG/Na_2SO_4 recovery	0.560	0.837	0.728	2.81
No mechanical pre-treatment	0.677	0.837	0.879	2.81
PET content – 91%	0.648	0.837	0.879	2.61
PET content – 99%	0.705	0.837	0.879	3.04
pH – 6	0.677	0.837	0.879	2.81
pH – 10	0.677	0.837	0.879	2.81
Plant size – 50 tpd	0.677	0.837	0.879	2.81
Plant size – 300 tpd	0.677	0.837	0.879	2.81
Renewable electricity	0.677	0.837	0.879	2.81
Residence time – 10 h	0.677	0.837	0.879	2.81
Residence time – 240 h	0.677	0.837	0.879	2.81
Solids load – 10	0.677	0.837	0.879	2.81
Solids load – 20	0.677	0.837	0.879	2.81
Sorting yield – 69%	0.677	0.690	0.879	2.12
Sorting yield – 99%	0.677	0.990	0.879	4.20
Temperature – 40°C	0.677	0.837	0.879	2.81
Temperature – 80°C	0.677	0.837	0.879	2.81
TPA recovery – 80%	0.614	0.837	0.8982	2.34
TPA recovery – 98%	0.726	0.837	0.8715	3.31
Water recycling	0.708	0.837	0.931	2.75

Table S16. Contribution of all process components to the life cycle impacts of enzymatically recycled rPET.

	Natural environment					*Natural resources*		*Human health*			
Component	**Acidification** %	**Ecotoxicity** %	**Eutrophication** %	**Global warming** %	**Ozone depletion** %	**Fossil fuel depletion** %	**Water use** %	**Carcinogenics** %	**Non-carcinogenics** %	**Particulates exposure** %	**Smog** %
PET flake	12.5	29.5	17.5	10.3	14.7	14.0	1.42	13.2	24.2	11.6	14.3
Ethylene glycol	2.47	1.46	1.55	3.06	0.74	6.33	0.86	2.19	1.87	1.89	3.02
rPET repolymerization	3.94	5.28	4.84	2.60	3.53	3.82	0.44	5.88	7.34	4.48	2.18
Transportation	0.34	0.28	0.13	0.68	0.96	0.95	0.03	0.33	0.41	0.24	0.47
rPET processing	20.2	9.42	17.4	20.3	8.77	9.36	12.4	15.5	9.98	21.8	20.1
Enzymatic depolymerization	33.5	39.1	40.1	33.7	60.3	25.0	73.2	36.6	38.9	38.5	31.0
Original PET	27.1	14.9	18.4	29.4	10.9	40.5	11.6	26.2	17.4	21.6	29.0

Table S17. Comparison of Global Warming Potential as calculated by SimaPro (ecoinvent data, TRACI method) and the Materials Flows through Industry tool (MFI).

		Global warming (kg CO_2 eq $unit^{-1}$)	
Item	**Unit**	**ecoinvent**	**MFI**
Electricity	kWh	0.654	0.578
Ethylene glycol	kg	1.97	0.937
PET	kg	3.03	3.40
Post-consumer PET flake	kg	0.686	0.376
Sodium hydroxide	kg	2.64	1.31
Sodium sulfate	kg	0.918	0.086
Steam	kg	0.185	0.238
Sulfuric acid	kg	0.161	-0.013
Terephthalic acid	kg	1.88	3.27
Water, cooling	kg	7.76E−6	1.59E−4
Water, process	kg	0.0453	2.01E−4

Table S18. Values utilized for all sensitivity cases. Adapted from Singh, et al.[1]

	Optimistic case	**Base case**	**Pessimistic case**
Depolymerization extent (%)	99	90	80
Enzyme loading (mg_{enzyme} $g_{feedstock}^{-1}$)	10	5	1
Ethylene glycol recovery (%)	65	50	40
Incoming flake PET content (%)	99	95	91
Plant size (tpd)	300	150	50
Reactor pH	10	8	6
Reactor retention time (h)	240	96	10
Reactor temperature (°C)	80	60	40
Solid loading (%)	20	15	10
Sorting phase yield (%)	69	84	99
Terephthalic acid recovery (%)	98	90	80
Other cases			
Ammonia instead of NaOH			
“Best” case			
Moist-solid			
Moist-solid + renewable electricity			
No ethylene glycol recovery			
No ethylene glycol or sodium sulfate recovery			
No mechanical pre-treatment			
No pH control			
Renewable electricity use			
Water recycling			

Table S19. Combined life cycle impact scores for all terephthalic acid production and recycling cases. Scores were calculated by first normalizing all impact categories against the corresponding base case impact categories. The normalized impacts were then subsequently summed to obtain the natural environment (acidification, ecotoxicity, eutrophication, global warming and ozone depletion), natural resources (fossil fuel depletion and water use) and human health (carcinogenics, non-carcinogenics, fine particulates exposure, and smog) scores. n.m. = not measured.

Case	Natural environment score	Natural resources score	Human health score	Total score	±σ
Virgin	1.05	1.44	1.02	3.51	1.63
Base case	5.00	2.00	4.00	11.0	1.56
Ammonia	3.50	2.13	2.65	8.29	1.74
“Best” case	1.70	0.65	1.26	3.61	1.73
Depolymerization – 80%	5.52	2.26	4.42	12.2	n.m.
Depolymerization – 99%	4.62	1.81	3.69	10.1	n.m.
EG recovery – 40%	5.04	2.05	4.04	11.1	n.m.
EG recovery – 65%	4.92	1.92	3.93	10.8	n.m.
EG selective membrane	4.67	1.66	3.73	10.1	1.71
Enzyme load – 1	4.96	1.99	3.96	10.9	n.m.
Enzyme load – 10	5.04	2.01	4.04	11.1	n.m.
Moist-solid	2.63	1.09	2.01	5.73	1.44
Moist-solid + renewables	2.57	1.08	1.95	5.60	1.37
No EG recovery	4.96	1.46	4.18	10.6	1.88
No EG/Na_2SO_4 recovery	7.93	1.62	6.14	15.7	2.29
No mechanical pre-treatment	4.09	1.83	3.08	9.00	1.60
PET content – 91%	5.24	2.09	4.16	11.5	n.m.
PET content – 99%	4.77	1.92	3.84	10.5	n.m.
pH – 6	5.00	2.00	3.99	11.0	n.m.
pH – 10	5.00	2.00	4.00	11.0	n.m.
Plant size – 50 tpd	5.34	2.70	4.18	12.2	n.m.
Plant size – 300 tpd	4.99	2.00	3.99	11.0	n.m.
Renewable electricity	3.67	1.75	2.70	8.12	1.44
Residence time – 10 h	5.00	2.00	4.00	11.0	n.m.
Residence time – 240 h	4.98	2.00	4.00	11.0	n.m.
Solids load – 10	5.95	2.81	4.67	13.4	n.m.
Solids load – 20	4.52	1.59	3.66	9.77	n.m.
Sorting yield – 69%	5.61	2.03	4.27	11.9	n.m.
Sorting yield – 99%	4.62	1.97	2.93	9.52	n.m.
Temperature – 40°C	5.00	2.00	4.00	11.0	n.m.
Temperature – 80°C	5.00	2.00	4.00	11.0	n.m.
TPA recovery – 80%	5.61	2.25	4.48	12.3	n.m.
TPA recovery – 98%	4.74	1.89	3.80	10.4	n.m.
Water recycle	4.65	1.77	3.65	10.1	1.54

Table S20. Combined life cycle impact scores for all poly(ethylene terephthalate) production and recycling cases. Scores were calculated by first normalizing all impact categories against the corresponding base case impact categories. The normalized impacts were then subsequently summed to obtain the natural environment (acidification, ecotoxicity, eutrophication, global warming and ozone depletion), natural resources (fossil fuel depletion and water use) and human health (carcinogenics, non-carcinogenics, fine particulates exposure, and smog) scores.

Case	Natural environment score	Natural resources score	Human health score	Total score	±σ
Virgin	3.95	1.50	2.81	8.26	2.46
Base case	5.00	2.00	4.00	11.0	1.98
Ammonia	4.13	2.09	3.34	9.56	1.83
“Best” case	2.53	1.24	2.00	5.77	1.90
Depolymerization – 80%	5.02	2.05	4.04	11.1	n.m.
Depolymerization – 99%	4.99	1.95	3.97	10.9	n.m.
EG recovery – 40%	5.03	2.02	4.02	11.1	n.m.
EG recovery – 65%	4.97	1.98	3.97	10.9	n.m.
EG selective membrane	4.82	1.86	3.86	10.5	2.08
Enzyme load – 1	4.99	2.00	3.99	11.0	n.m.
Enzyme load – 10	5.03	2.01	4.02	11.1	n.m.
Moist-solid	3.68	1.56	3.06	8.30	1.84
Moist-solid + renewables	2.72	1.40	2.22	6.35	1.74
No EG recovery	4.99	1.60	4.09	10.7	2.47
No EG/Na_2SO_4 recovery	6.75	1.66	5.13	13.5	2.46
No mechanical pre-treatment	4.53	1.94	3.56	10.0	1.94
PET content – 91%	5.04	2.02	4.03	11.1	n.m.
PET content – 99%	4.97	1.99	3.98	10.9	n.m.
pH – 6	5.00	2.00	4.00	11.0	n.m.
pH – 10	5.00	2.00	4.00	11.0	n.m.
Plant size – 50 tpd	5.17	2.42	4.08	11.7	n.m.
Plant size – 300 tpd	5.00	2.00	4.00	11.0	n.m.
Renewable electricity	3.67	1.82	2.82	8.31	1.73
Residence time – 10 h	5.00	2.00	4.00	11.0	n.m.
Residence time – 240 h	5.12	2.03	4.01	11.2	n.m.
Solids load – 10	5.52	2.47	4.33	12.3	n.m.
Solids load – 20	4.76	1.77	3.84	10.4	n.m.
Sorting yield – 69%	4.93	1.92	3.89	10.8	n.m.
Sorting yield – 99%	5.11	2.09	4.13	11.3	n.m.
Temperature – 40°C	5.00	2.00	4.00	11.0	n.m.
Temperature – 80°C	5.00	2.00	4.00	11.0	n.m.
TPA recovery – 80%	5.07	2.06	4.06	11.2	n.m.
TPA recovery – 98%	5.03	1.99	4.01	11.0	n.m.
Water recycle	4.81	1.91	3.83	10.6	2.05

Table S21. Life cycle inventory of the moist-solid enzymatic production of terephthalic acid from post-consumer poly(ethylene terephthalate).

Category	Item	Quantity	Unit
By-products	Ethylene glycol	0.3026	kg
Used products	Activated carbon	8.109E−3	kg
	Dimethyl sulfoxide (DMSO)	0.0467	kg
	Electricity	0.1192	kWh
	Enzyme	0.0496	kg
	PET flake	1.584	kg
	Steam	3.546	kg
	Water, cooling	199.8	kg
	Water, purified	0.3566	kg
Infrastructure	Chemical factory, organics	4E−10	p
Emission, water	Ethylene glycol	0.107	kg
	DMSO	0.0330	kg
Produced waste	Disposal, solid waste, incineration w/ energy recovery	0.0740	kg
	Disposal, solid waste, sanitary landfill	0.2960	kg
	Wastewater	2.8E−4	m^3

Table S22. Contribution of all process components to the life cycle impacts of terephthalic acid produced by "moist-solid" enzymatic hydrolysis.

	Natural environment					*Natural resources*		*Human health*			
Component	**Acidification** %	**Ecotoxicity** %	**Eutrophication** %	**Global warming** %	**Ozone depletion** %	**Fossil fuel depletion** %	**Water use** %	**Carcinogenics** %	**Non-carcinogenics** %	**Particulates exposure** %	**Smog** %
Activated carbon	0.62	0.05	0.17	0.73	2.79	2.44	0.00	0.19	0.10	0.46	0.57
Dimethyl sulfoxide	2.92	0.65	1.34	2.01	2.86	5.71	0.44	1.81	1.11	1.97	2.39
Cooling water	0.05	0.03	0.04	0.06	0.03	0.04	85.42	0.19	0.04	0.06	0.07
Process water	0.72	0.11	0.71	0.58	0.80	0.70	0.15	0.59	0.19	0.46	1.83
Steam	18.33	1.07	3.81	23.49	19.37	28.96	0.36	5.50	2.01	7.80	14.95
Capital	4.11	4.56	5.02	2.29	1.09	1.13	0.18	10.89	8.55	3.93	3.36
Enzyme	20.38	2.89	8.47	17.56	4.68	5.18	10.06	7.52	14.12	15.78	14.40
PET flake	47.99	48.97	60.07	38.81	63.40	53.89	2.92	49.51	48.92	60.16	58.18
Electricity	2.09	0.83	4.00	2.78	1.96	1.39	0.13	3.51	1.05	7.15	1.54
Ethylene glycol	−17.52	−4.47	−9.76	−21.29	−5.86	−44.85	−3.24	−15.14	−6.97	−18.06	−22.67
Solid waste	2.78	40.66	16.31	11.68	3.02	0.56	0.33	20.27	23.78	2.21	2.71
Wastewater	0.01	0.00	0.06	0.01	0.00	0.00	−0.11	0.03	0.04	0.01	0.01

Table S23. Annual operating cost summary for the “moist-solid” case study.

Operating Parameters				
Metric/Parameter	**Value**			**Units**
Operating Hours/Yr	7,884			hrs/yr
On Stream Factor	90			%
Feedstock Contaminants	5.0			wt %
Feedstock PET Tonnes/yr	46,811			Dry U.S. Tons/yr
Total TPA Rate (tonnes/yr)	31,088			Tonnes/yr
Tonnes TPA / tonne PET feed	0.66			
PET Flake feed mass flow	6,250			kg/hr
TPA Production Rate (kg/hr)	3,943			kg/hr
Variable Operating Costs				
Process Hierarchy/Section	**Raw Material/Utility**		**Mass Flow, kg/hr**	**MM$/yr (2016)**
Raw Materials				
A100: Pretreatment	PET Flake feedstock		6,250	32.59
A200: Depolymerization	Caustic (as pure), neutralize		0	0.00
	Water Makeup		1,406	0.00
	Enzyme Consumption		203	24.01
A300: Clarification	Ultrafilter Replacement		-	0.14
	Activated carbon replacement		-	0.25
A400: TPA Purification	Dimethyl Sulfoxide (DMSO) Make-up		184	5.37
A600: OSBL Utilities	Steam, 125C/34 psig		158	0.02
	Steam, 175C/114 psig		9,020	1.03
	Steam, 250C/560 psig		4,802	0.55
	Cooling Water		787,739	0.24
	Chiller Water, 40F		0	0.00
	Grid Electricity		470	0.18
	Subtotal			**64.39**
Waste Streams				
A300: Clarify	Disposal of ash and solids		1,257	0.34
A400: TPA Purification	Disposal of brine, Organic waste		1,316	0.11
	Subtotal			**0.45**
Co-Products and Credits				
A500: EG Recovery	EG Co-product		1,193	9.03
	Subtotal			**9.03**
	Total Variable Operating Costs			**55.81**
Fixed Operating Costs				
Position				**MM$/yr (2016)**
Labor & Supervision				
Total Salaries (Managers, Supervisors, Engineers, Technicians, Secretaries)				1.27
Labor Burden (90% of Total Salaries)				1.14
Other Overhead				
Maintenance	3.0%	of ISBL		0.44
Property Insur. & Tax	0.7%	of FCI		0.43
Total Fixed Operating Costs				**3.28**
				MM$/yr (2016)
Total Operating Costs				**59.08**

Table S24. Minimum selling price (MSP) for the moist-solid case study, broken down by process section. "Other operational" cost category includes cooling water, waste treatment, and fixed costs.

Process Area	Cost Category ($/kg TPA)								
	Feedstock	Enzyme	Electricity	DMSO	Steam	Capital Charge	Other Operational	Co-product	Total
Feedstock & Handling	1.05	0.00	0.00	0.00	0.00	0.11	0.02	0.00	1.18
Depolymerization	0.00	0.77	0.00	0.00	0.00	0.03	0.02	0.00	0.82
Clarification	0.00	0.00	0.00	0.00	0.00	0.04	0.04	0.00	0.08
TPA Purification	0.00	0.00	0.00	0.17	0.01	0.04	0.03	0.00	0.25
EG Recovery	0.00	0.00	0.00	0.00	0.04	0.01	0.03	-0.29	-0.22
OSBL	0.00	0.00	0.00	0.00	0.00	0.06	0.00	0.00	0.06
Total	1.05	0.77	0.01	0.17	0.05	0.28	0.14	-0.29	2.18

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