Supplementary Information

Quantifying the Environmental Benefits of a Solvent-Based Separation Process for Multilayer Plastic Films

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1 Process description

1.1 STRAP-A

In STRAP-A, the polymer PE is first selectively dissolved by toluene (4 hours, 110 °C), a solvent that only solubilizes the targeted resin while leaving other resins in solid phase. The dissolved PE layer (with minor EVA content) is separated from the multilayer film A1 through mechanical filtration followed by the addition of acetone to facilitate precipitation of the PE resin. The PE layer is then sent to another round of filtration and drying processes to be separated from solvents. The toluene and acetone mixture is reused in the process after being recycled via distillation. In the last step, the isolated PE layer is sent to the extruder to be restructured in pellet form and finally sent to the storage tank. The solid residual from the first filtration step, namely the EVOH and PET layers, went through similar set of procedures. The EVOH layer is selectively dissolved by DMSO (30 mins, 95 °C), a solvent solubilizing the targeted EVOH resin while leaving PET insoluble, then it is separated through filtration and precipitated by the addition of water in a precipitation vessel. EVOH is sent to the filter and dryer to be separated from the 60% DMSO/40% water (v/v) solvent/antisolvent mixture, then extruded to its final shape to be stored in the storage tank. The DMSO and water mixture in this step is distilled to be reused in the process. The remaining PET layer is extruded and stored as well.

1.2 STRAP-B

Similarly, STRAP-B (Figure SI 2) is developed for recycling the same multilayer film (A1) in which temperature-driven precipitation strategy is adopted and the EVA component is further extracted

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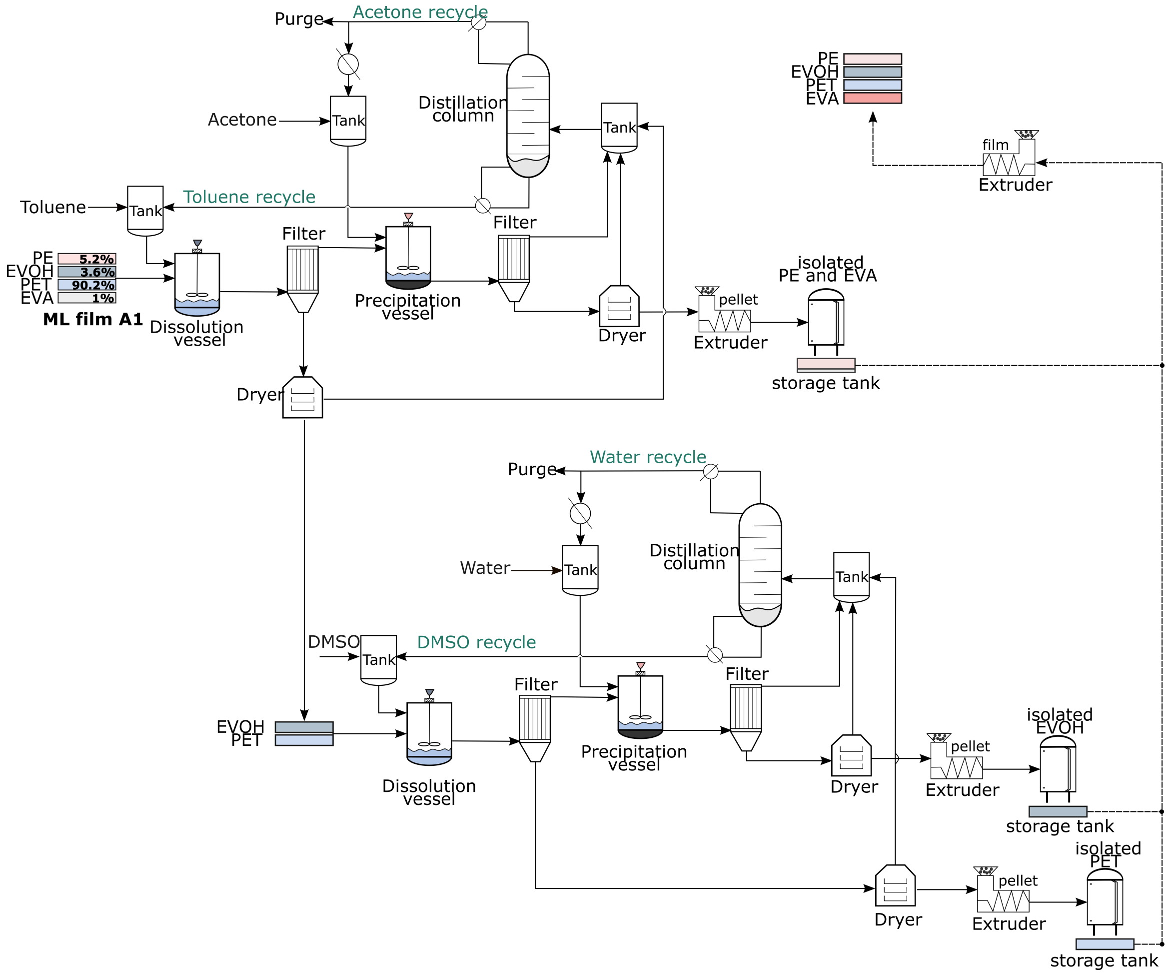

Figure SI 1: Process flow diagram to recycle multilayer film A1 composed of polymer resins PE, EVOH, PET, and EVA through STRAP-A, including inputs to the process (orange) and the solvent recycle streams (green). Each polymer component is recovered in the pellet form, which is then sent to the film extruder to produce recycled multilayer film A1 (grey box).

from the PE resin. In this strategy, instead of adding an antisolvent, the temperature of the polymer-solvent solution is lowered to the point that the targeted polymer becomes insoluble, thus enabling polymer precipitation and isolation of the polymer. PE and EVA components are selectively dissolved in toluene (4 hours, 110 °C) and separated from EVOH and PET via mechanical filtration. The temperature of the toluene mixture, containing the dissolved polymers PE and EVA, is reduced from 110 °C to 35 °C in order to enable PE precipitation, while the remaining EVA component is recovered by the addition of acetone which acts as an antisolvent. After filtration and drying, both polymers are sent to the extruder and storage tank. The acetone/toluene mixture is separated by distillation and then recycled back into the process. For the EVOH and PET layers, EVOH is selectively dissolved in a 60% DMSO/40% water (v/v) solution (30 mins, 95 °C), then cooled down to 35 °C to achieve precipitation. Precipitated EVOH is stored after going through the filtration, drying and extrusion processes while the mixture is directly recycled back into the process. After EVOH separation, the remaining PET component in the dissolution vessel is extruded and stored after filtration and drying.

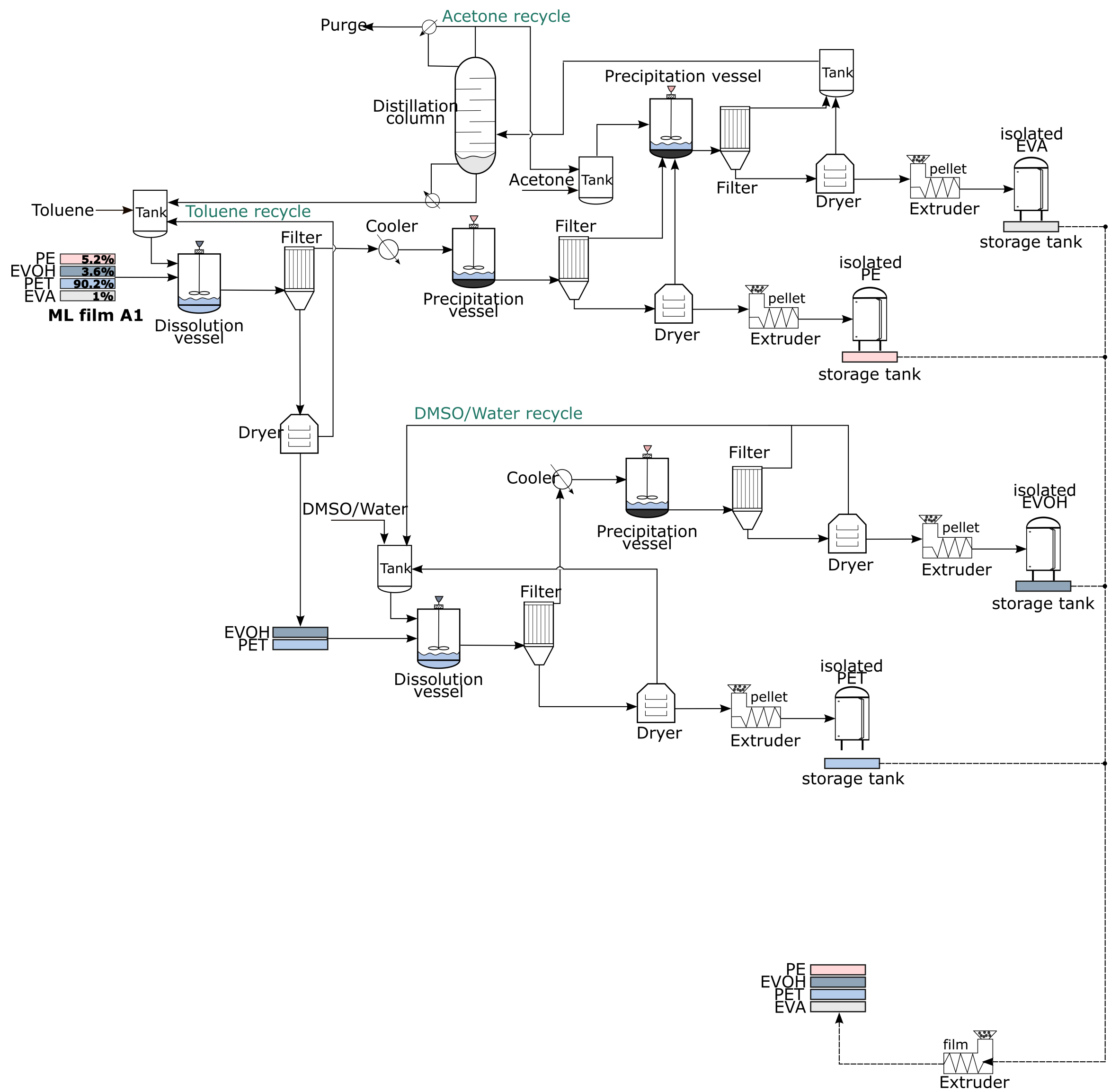

Figure SI 2: Process flow diagram to recycle multilayer film A1 composed of polymer resins PE, EVOH, PET, and EVA through STRAP-B, including inputs to the process (orange) and the solvent recycle streams (green). Each polymer component is recovered in the pellet form, which is then sent to the film extruder to produce recycled multilayer film A1 (grey box).

2 Inventory analysis and impact assessment

Environmental profiles of the polymers -PET, PE, EVOH and EVA- are taken from the Environmental Footprint (EF) database v.2.[1] The data is used to estimate the life cycle impacts of the manufacturing of the virgin materials, providing a basis for quantifying the avoided impacts.

The only polymer that is not available in the EF database is glycol- modified polyethylene terephthalate (PETG). Impacts related to PETG is estimated based on the reaction stoichiometry by considering the impacts of raw materials (PET, purified terephthalic acid (PTA), monoethylene glycol (MEG), diethylene glycol (DEG) and water) used to produce PETG and assuming similar reaction conditions to PET production.[2]

Table SI 1: Environmental impacts related to manufacture of polymers used in this study. Database name and the region information are included. The impact categories are normalized for 1 kg of material production and reported using Environmental Footprint method[3]

Material	Resource use, fossils (MJ)	Climate change (kg CO2eq.)	Water use (m3)	Ecotoxicity, freshwater (CTUe)	Human toxicity, cancer (CTUh)	Human toxicity, non-cancer (CTUh)	Database	Region
PET	58.25	2.21	0.55	0.48	1.6×10^{-8}	1.5×10^{-7}	EF 2.0	EU
PE	74.58	2.07	0.14	0.63	2.7×10^{-8}	1.5×10^{-7}	EF 2.0	EU
EVOH	161.78	7.30	1.33	1.07	2.5×10^{-8}	4.2×10^{-7}	EF 2.0	EU
EVA	70.88	2.22	0.99	0.91	2.4×10^{-8}	9.4×10^{-8}	EF 2.0	GLO
PETG	59.77	2.11	1.23	0.98	2.9×10^{-8}	1.4×10^{-7}	Stoichiometry	EU (RER)

Data for the STRAP inputs for the process utilities (steam, electricity and cooling water), as well as the transportation of commodities and polymer film extrusion process are taken from the EF database v.2[1] (see Table SI 2). Solvents (and/or antisolvents) used in STRAP process such as acetone, toluene, DMSO, water, DMF, THF, 1-propanol are taken from the ecoinvent 3.6 cut-off by classification database.[4]

Table SI 2: Environmental impacts related to STRAP process inputs involving process utility (steam/electricity/cooling water), material transportation and polymer extrusion. Database name and the region information are included. The impact categories are normalized for the corresponding unit and reported using Environmental Footprint method[3]

Item	Unit	Resource use, fossils (MJ/unit)	Climate change (kg CO2/unit)	Water use (m3/unit)	Ecotoxicity, freshwater (CTUe)	Human toxicity, cancer (CTUh)	Human toxicity, non-cancer (CTUh)	Database	Region
Steam — mix	MJ	1.09	0.07	1.4×10^{-3}	1.2×10^{-3}	2.2×10^{-11}	1.1×10^{-9}	EF 2.0	EU
Electricity	MJ	2.02	0.12	1.6×10^{-2}	4.7×10^{-3}	9.4×10^{-11}	2.5×10^{-9}	EF 2.0	EU
Cooling Water	m3	5.09	0.33	4.3×10^{-2}	4.3×10^{-4}	3.6×10^{-11}	1.4×10^{-10}	EF 2.0	EU
Extrusion	kg	4.39	0.26	3.5×10^{-2}	1.2×10^{-2}	2.8×10^{-10}	6.5×10^{-9}	EF 2.0	EU
Transportation	1 t*km	1.04	0.08	2.9×10^{-3}	1.3×10^{-2}	4.2×10^{-10}	7.1×10^{-9}	EF 2.0	EU

2.1 Regional data variation: toxicity values

During the data inventory building phase, environmental profiles of the process input materials and utilities are gathered for both US (denoted as RNA in the database: North America region) and EU (denoted as RER) regions. Regarding the toxicities, we observed major variations between locations for the ecotoxicity and human toxicity (cancer/non-cancer) impact categories, especially for the process utilities sourced from natural gas.

The difference between toxicity values based on the utility source and region are plotted on a log scale in Fig. SI 3. The electricity mix is modeled based on Table SI 3 for US and EU regions, whereas for steam the following mix is used: EU (12.5% hard coal, 75% natural gas, 12.5% biomass[5]) and US (27% hard coal, 73% natural gas[6]). We noticed that EU data results in considerably lower toxicities in all categories, with an order of magnitude of 3. Given that the polymer database region is EU, modeling STRAP process based on US regional data for the process utilities and inputs resulted in an overestimation of the toxicity values for the STRAP process, while having lower toxicities for the activities related to the manufacturing of virgin polymer resins modeled by European market data.

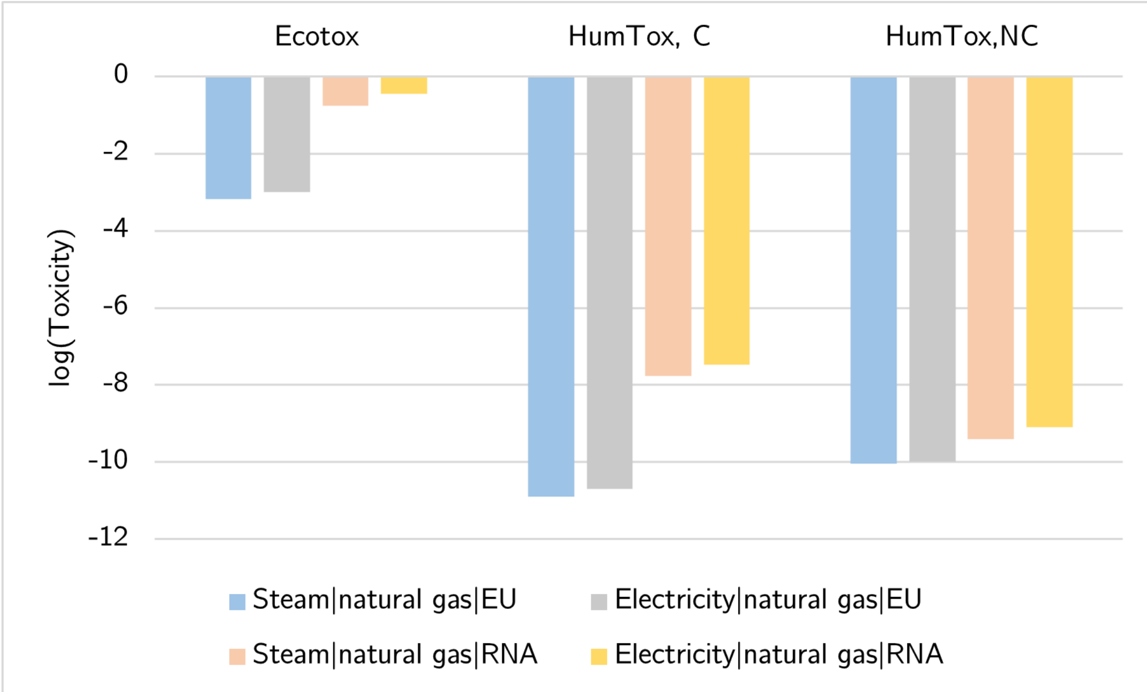

Figure SI 3: Regional toxicity values for steam and electricity production from natural gas. Toxicity categories: Ecotox (Ecotoxicity, freshwater), HumTox, C (Human toxicity, cancer), HumTox, NC (Human toxicity, non-cancer). Regions: EU and RNA.

Figures SI 4 and SI 5 suggest that freshwater ecotoxicity values are considerably higher for the RNA based data and worse than the virgin resin production in EU regions. However, toxicity profiles are lower for the STRAP process if the utility impacts are modeled from EU sources and compared with the virgin material production toxicities that are sourced from the same region, i.e. EU. Based on the regional inventory analysis, we have modeled the production of virgin polymer resins and STRAP process inputs (raw materials and utilities) based on the EU region to ensure consistent system boundaries, thus providing a fair basis for the comparison of two perspectives.

Table SI 3: Country electricity mix for different regions[7] (Note: remaining % comes from photovoltaic, solarthermal and geothermal resources).

Region/Source	Nuclear	Coal	Natural Gas	Heavy fuel oil	Biomass	Biogas	Waste	Hydro	Wind
EU-27	27.69 %	26.6 %	21.25 %	2.24 %	2.24 %	1.25 %	1.17 %	10.25 %	5.47 %
US	19.11 %	39.71 %	26.93 %	0.86 %	1.06 %	0.3 %	0.46 %	6.74 %	3.95 %

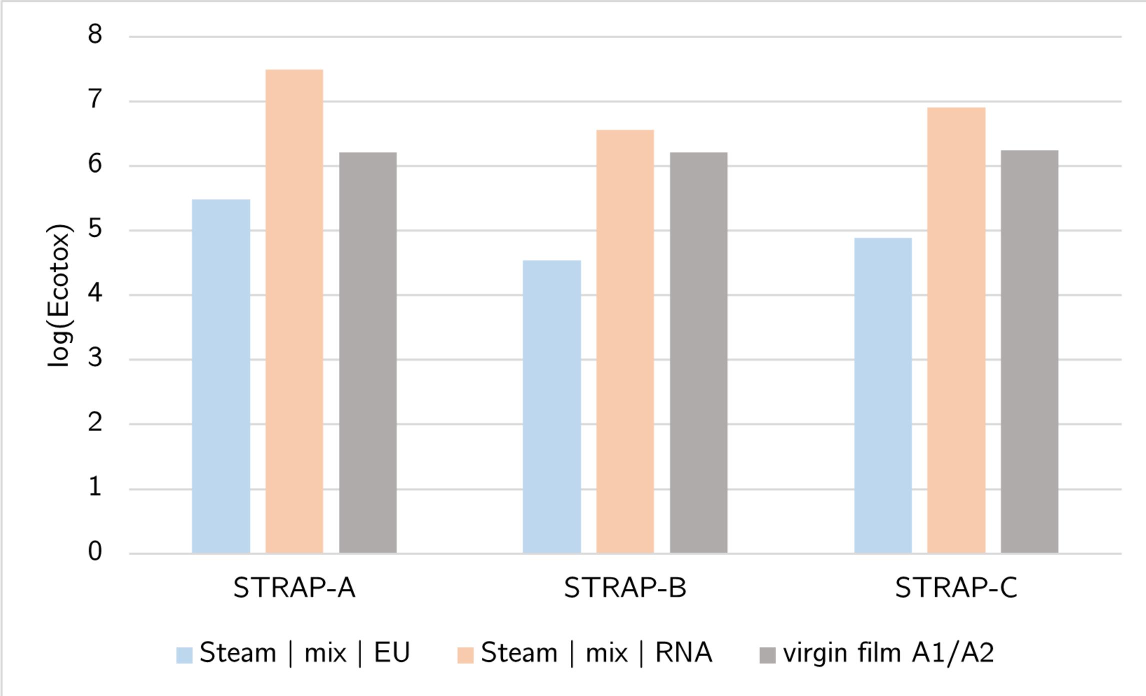

Figure SI 4: Ecotoxicity values for steam usage in STRAP processes based on EU and RNA regions compared to the virgin film production: film A1 (STRAP-A and STRAP-B) and film A2 (STRAP-C) | Region: EU.

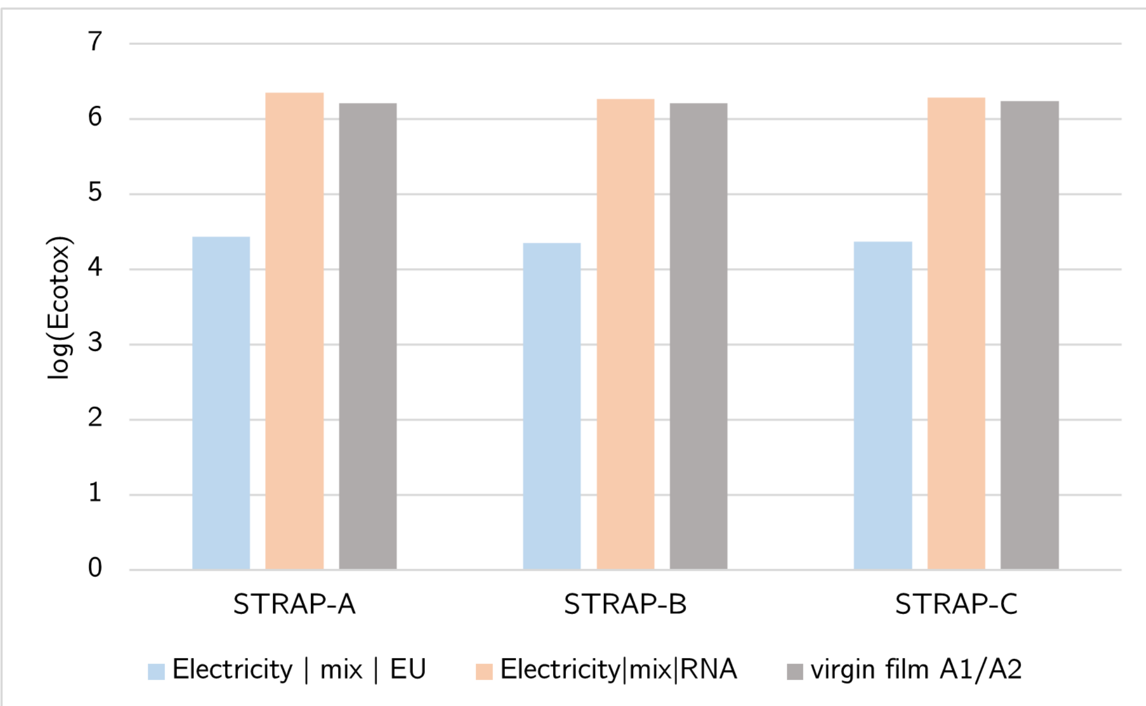

Figure SI 5: Ecotoxicity values for the electricity usage in STRAP processes based on EU and RNA regions compared to the virgin film production: film A1 (STRAP-A and STRAP-B) and film A2 (STRAP-C) | Region: EU.

3 Energy use

We also report the benefits of the STRAP process based on the avoided impacts (net savings) achieved by the recovery of resins and production of virgin-quality ML film from PIW (Eq. 3.1). The difference between the impact of producing the film via the virgin production process and the STRAP process is considered as the avoided impact. This avoided impact value represents the savings in energy and feedstock that would be required to manufacture the virgin material. Therefore, a positive avoided impact indicates that the STRAP process is more efficient than the virgin manufacturing method. While a negative avoided impact means that replacing the virgin method with the STRAP process would worsen the environmental impacts of ML film production.

$$I_{i,\ avoided} = I_{i,\ virgin} - I_{i,\ STRAP} \tag{3.1}$$

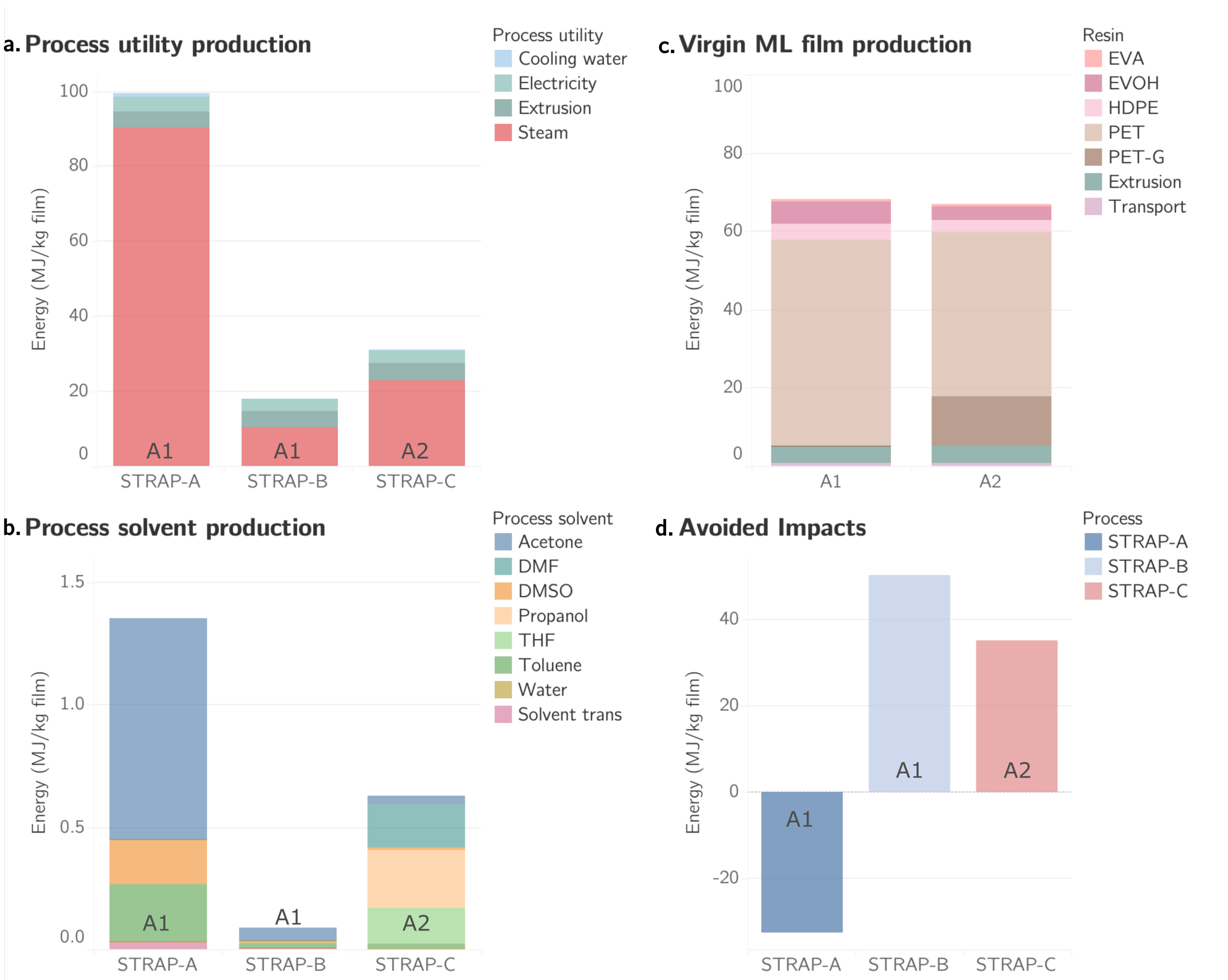

Figure SI 6: Energy use (MJ) of STRAP process in terms of utility and raw materials for the production of 1 kg of film A1 (STRAP-A and STRAP-B) and 1 kg of film A2 (STRAP-C), their constituent virgin polymer production and avoided impacts via STRAP recovery technology.

4 Climate change impact

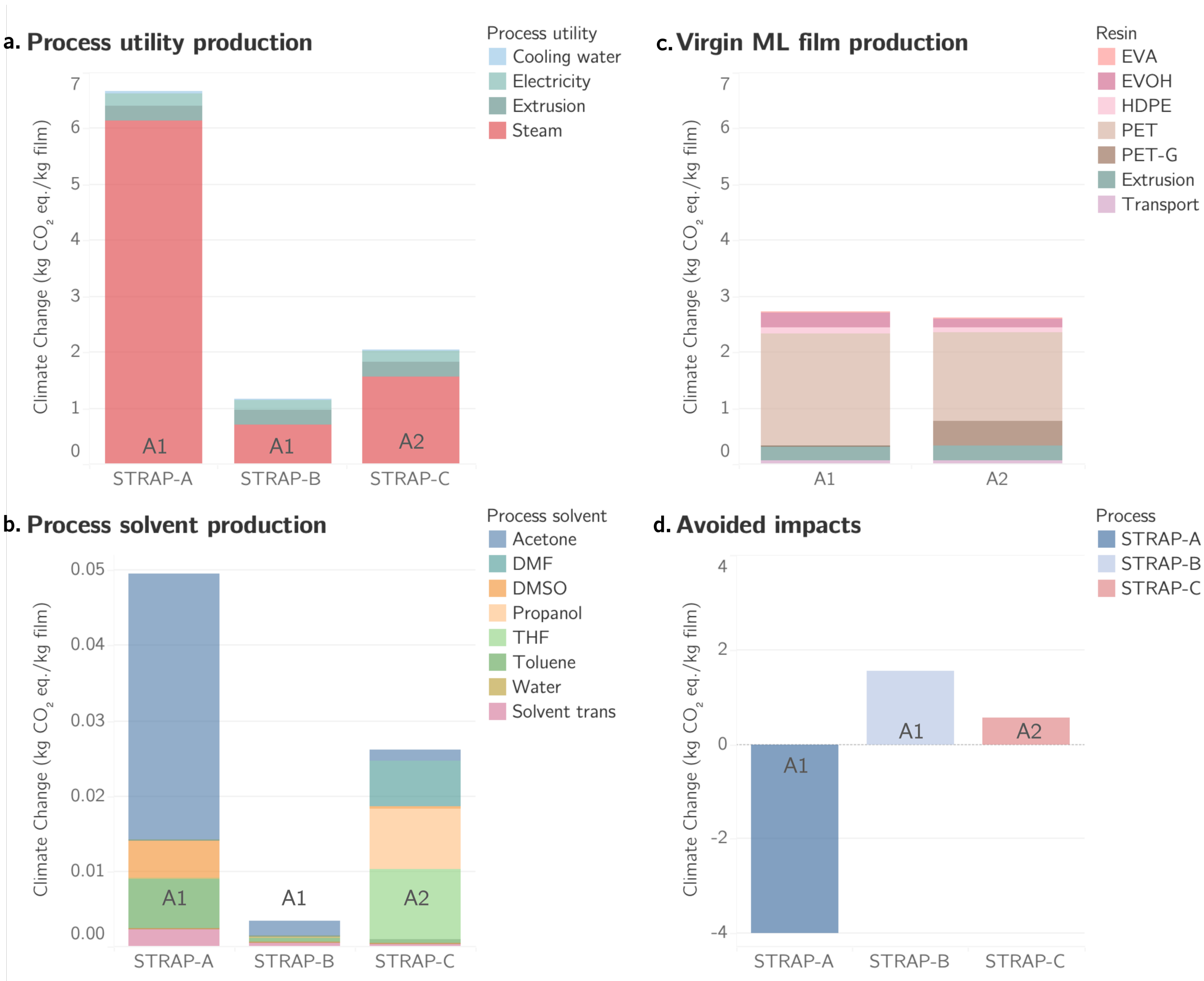

Figure SI 7: Climate change impact (kg CO_2 eq.) of STRAP process in terms of utility and raw materials for the production of 1 kg of film A1 (STRAP-A and STRAP-B) and 1 kg of film A2 (STRAP-C), their constituent virgin polymer production and avoided impacts via STRAP recovery technology.

5 Water usage

Table SI 4: Resin-based water usage for 1 kg film A1 and A2 via virgin material production or STRAP technology

	Film A1		Film A2		STRAP-A		STRAP-B		STRAP-C	
Resin	Water usage (kg/kg film)	%	Water usage (kg/kg film)	%	Water usage (kg/kg film A1)	%	Water usage (kg/kg film A1)	%	Water usage (kg/kg film A2)	%
PET	631.1	90.2	502.4	61.6	62.8	4.2	56	14.8	61.7	10.7
PE	9.4	1.3	7.3	0.9	305.1	20.5	36.2	9.6	28.0	4.9
EVOH	49.2	7.0	29.1	3.6	1123.2	75.3	32.3	8.6	25.4	4.4
EVA	10.3	1.5	10.6	1.3	-	-	253.2	67.0	201.2	35.0
PETG	-	-	266.1	32.6	-	-	-	-	257.8	44.9
Total	700.0	100	815.5	100	1491.1	100	377.7	100	574.1	100

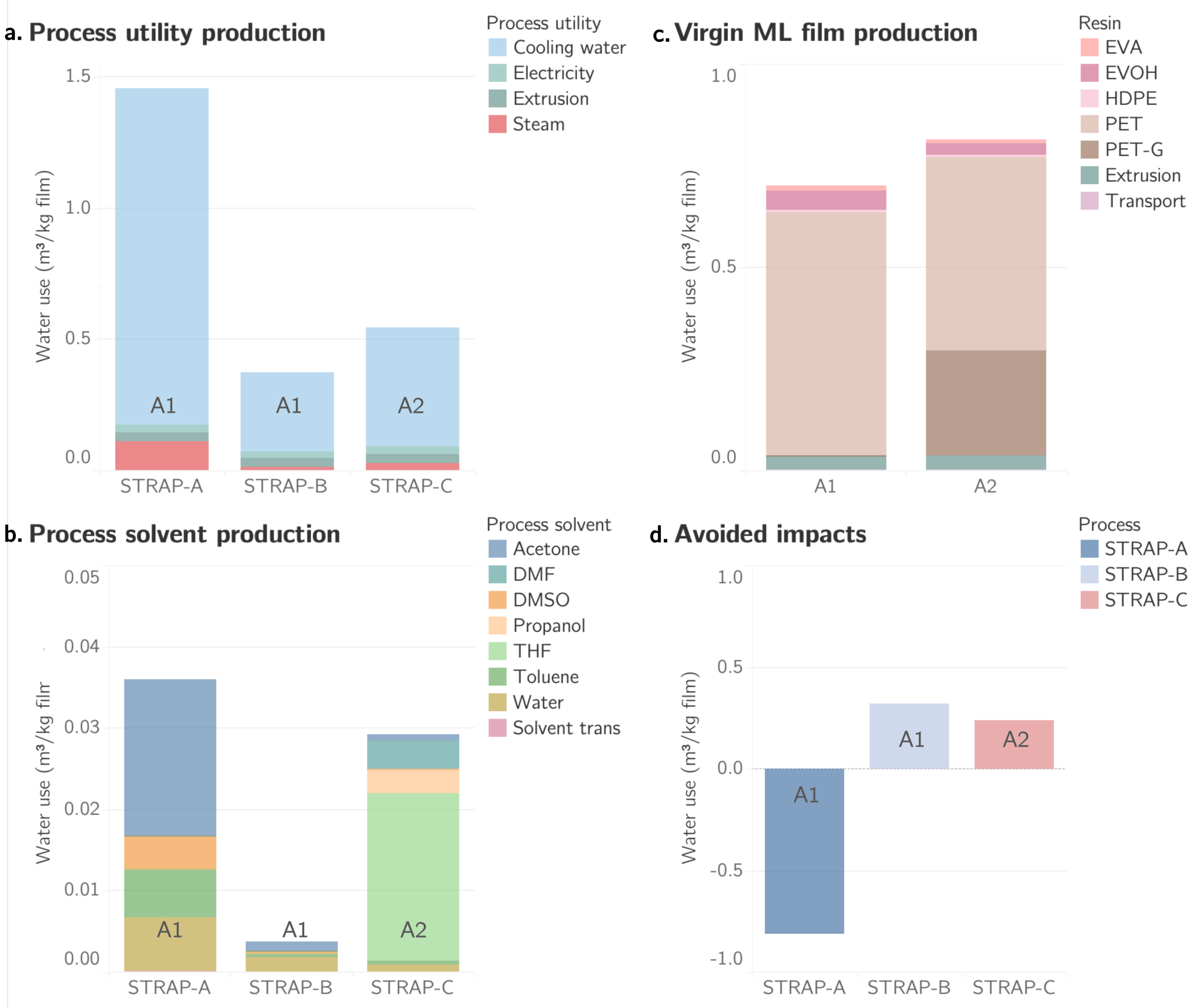

Figure SI 8: Water usage factor (m^3) of STRAP process in terms of utility and raw materials for the production of 1 kg of film A1 (STRAP-A and STRAP-B) and 1 kg of film A2 (STRAP-C), their constituent virgin polymer production and avoided impacts via STRAP recovery technology.

6 Toxicity

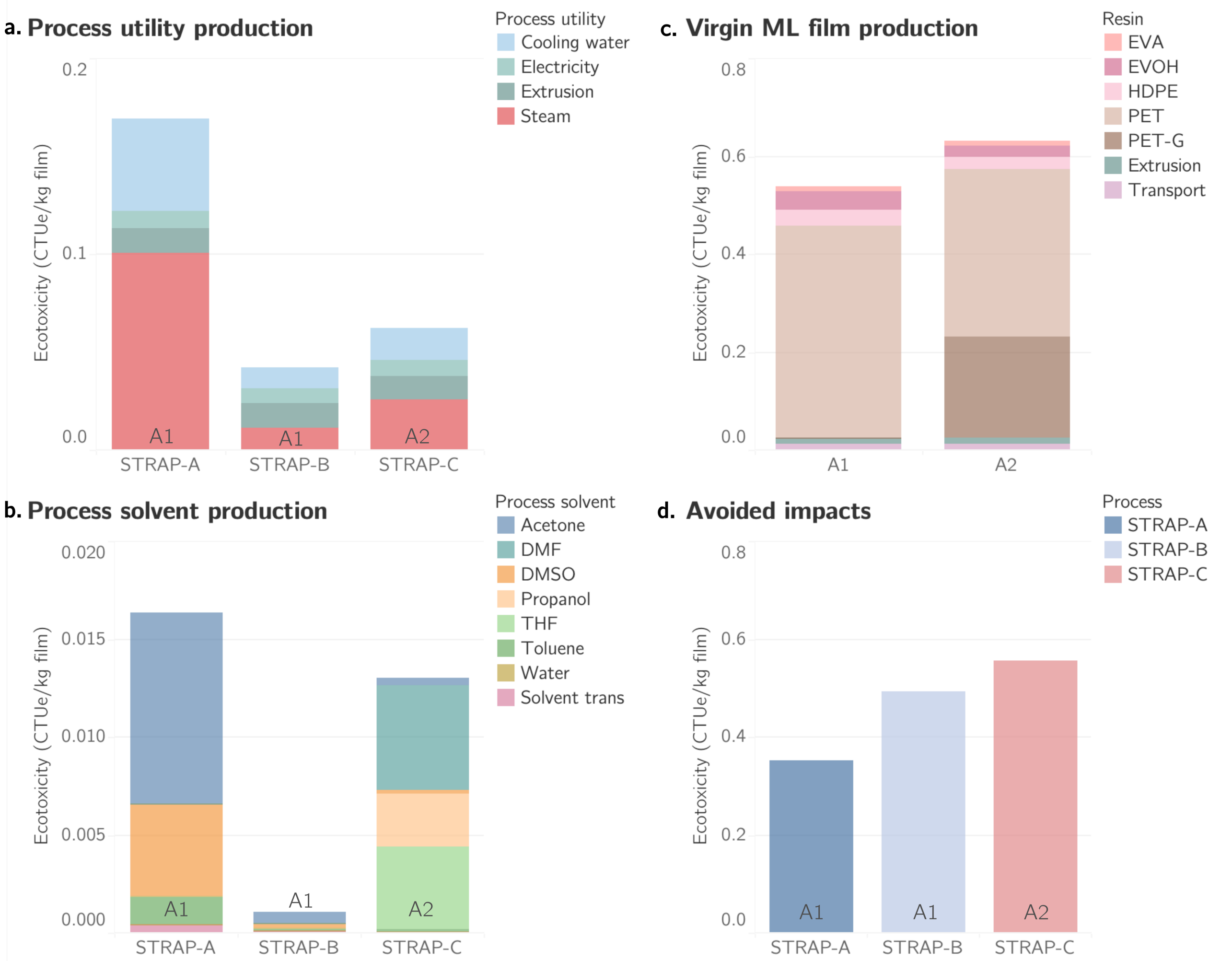

Figure SI 9: Freshwater ecotoxicity impacts (CTUe) related to the production of the a. process utilities and b. process solvents used in STRAP recovery technology. Impacts are estimated based on the STRAP process inputs required for the production of 1 kg of film A1 (STRAP-A and STRAP-B) and 1 kg of film A2 (STRAP-C). c. Freshwater ecotoxicity impacts (CTUe) related to the virgin multilayer films of equivalent amount. Impacts are estimated considering the individual impacts of virgin polymer resins (0.902 kg PET, 0.052 kg PE, 0.036 kg EVOH and 0.010 kg EVA for manufacturing 1 kg of film A1 and 0.718 kg PET, 0.040 kg PE, 0.022 kg EVOH, 0.010 kg EVA, and 0.210 kg PETG for manufacturing 1 kg of film A2), their transportation and extrusion. d. Avoided impacts of multilayer films with respect to ecotoxicity (CTUe) by replacing the virgin manufacturing method with the STRAP recovery technology.

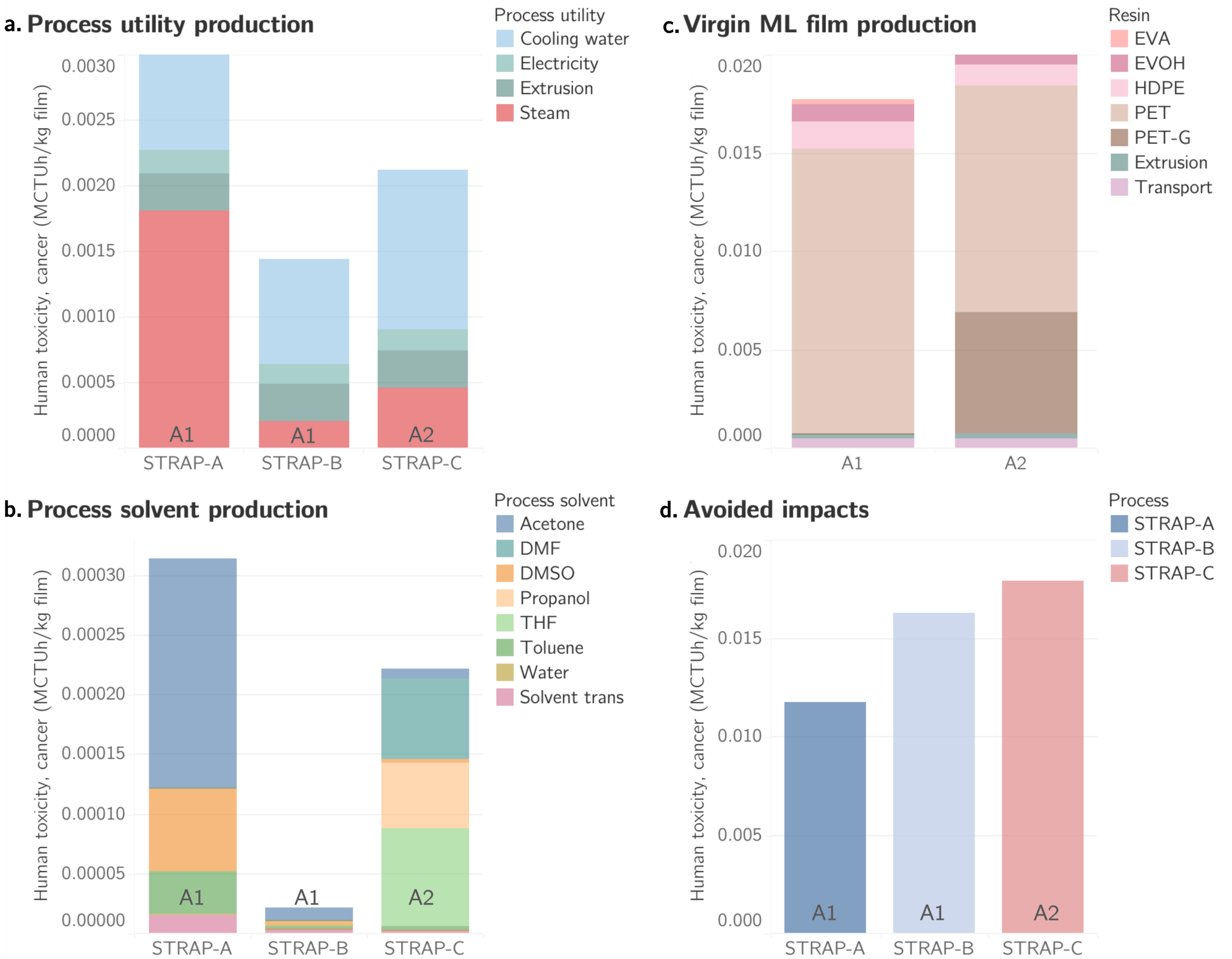

Figure SI 10: Human toxicity, cancer impacts (expressed in millions CTUh) related to the production of the a. process utilities and b. process solvents used in STRAP recovery technology. Impacts are estimated based on the STRAP process inputs required for the production of 1 kg of film A1 (STRAP-A and STRAP-B) and 1 kg of film A2 (STRAP-C). c. Human toxicity, cancer impacts (MCTUh) related to the virgin multilayer films of equivalent amount. Impacts are estimated considering the individual impacts of virgin polymer resins (0.902 kg PET, 0.052 kg PE, 0.036 kg EVOH and 0.010 kg EVA for manufacturing 1 kg of film A1 and 0.718 kg PET, 0.040 kg PE, 0.022 kg EVOH, 0.010 kg EVA, and 0.210 kg PETG for manufacturing 1 kg of film A2), their transportation and extrusion. d. Avoided impacts of multilayer films with respect to human toxicity, cancer (MCTUh) by replacing the virgin manufacturing method with the STRAP recovery technology.

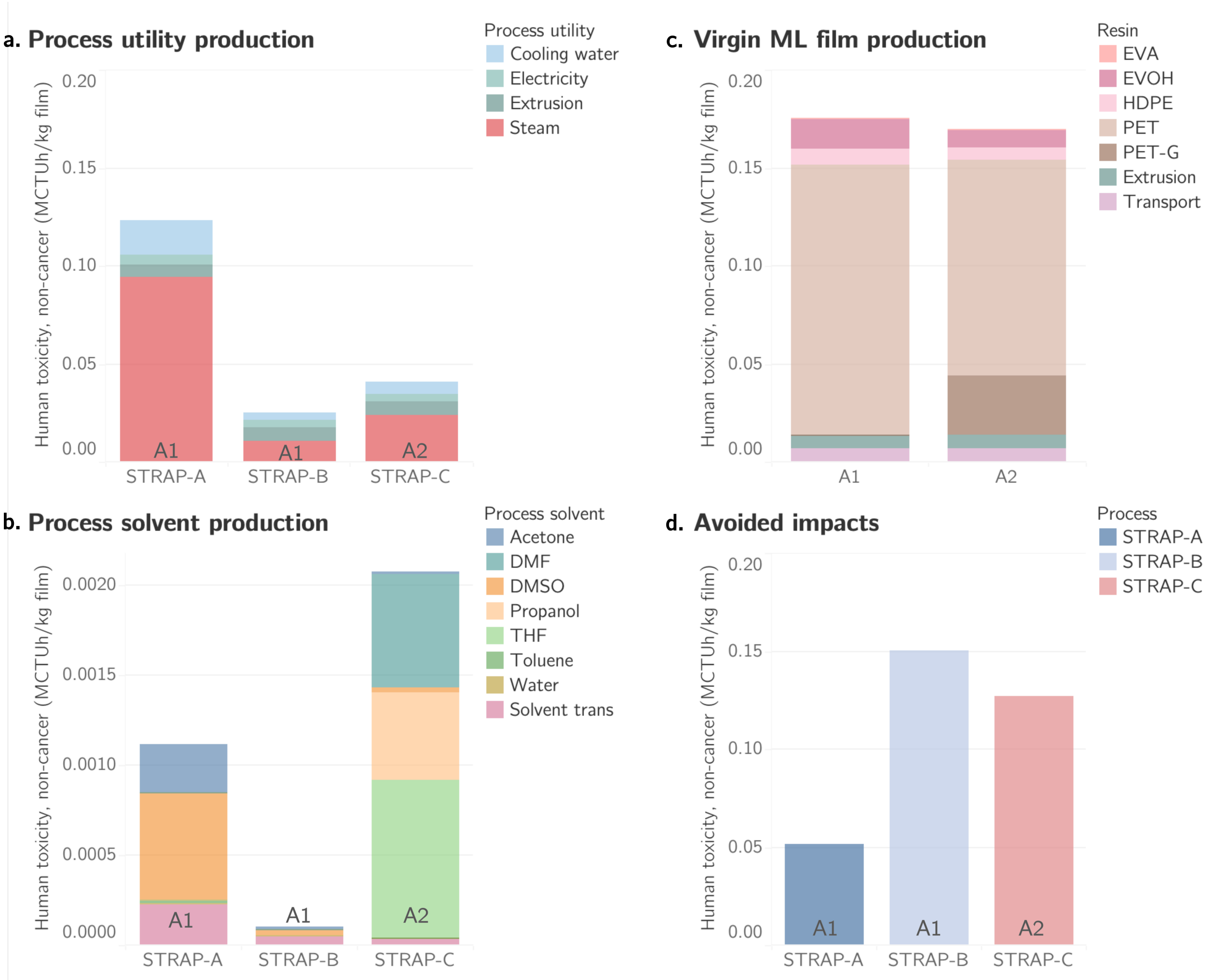

Figure SI 11: Human toxicity, non-cancer impacts (expressed in millions CTUh) related to the production of the a. process utilities and b. process solvents used in STRAP recovery technology. Impacts are estimated based on the STRAP process inputs required for the production of 1 kg of film A1 (STRAP-A and STRAP-B) and 1 kg of film A2 (STRAP-C). c. Human toxicity, non-cancer impacts (MCTUh) related to the virgin multilayer films of equivalent amount. Impacts are estimated considering the individual impacts of virgin polymer resins (0.902 kg PET, 0.052 kg PE, 0.036 kg EVOH and 0.010 kg EVA for manufacturing 1 kg of film A1 and 0.718 kg PET, 0.040 kg PE, 0.022 kg EVOH, 0.010 kg EVA, and 0.210 kg PETG for manufacturing 1 kg of film A2), their transportation and extrusion. d. Avoided impacts of multilayer films with respect to human toxicity, non-cancer (MCTUh) by replacing the virgin manufacturing method with the STRAP recovery technology.

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