

Life Cycle Assessment Approaches of Plastic Recycling with Multiple Cycles: Mini Review

Yee Van Fan^{a,*}, Lidija Čuček^b, Jiří Jaromír Klemeš^a, Annamaria Vujanovic^b, Petar Sabev Varbanov^a

^aSustainable Process Integration Laboratory – SPIL, NETME Centre, Faculty of Mechanical Engineering, Brno University of Technology - VUT Brno, Technická 2896/2, 616 69 Brno, Czech Republic

^bFaculty of Chemistry and Chemical Engineering, University of Maribor, Smetanova 17, Maribor, Slovenia
 fan@fme.vutbr.cz

Circular Economy and Cradle to Cradle Design have been gaining significant attention in minimising the life cycle environmental footprint of a product or service. However, it remains challenging to model multiple cycles where the products could be finitely or infinitely recovered through upcycling or downcycling, depending on the quality changes, and used as a material in the different product life cycles. This study aims to summarise the Life Cycle Assessment (LCA) methods that consider the cascade utilisation and upcycling of a product which will also apply to the case of plastic recycling. The methodological challenges, particularly for secondary and tertiary plastic recycling, are highlighted. The mini-review is intended to identify the limitation of current assessment methods for the subsequent development of an improved LCA in future work.

1. Introduction

Plastic recycling has gained high research attention in supporting a sustainable and circular plastic value chain or cradle-to-cradle design. Life cycle assessment (LCA) is among the most applied approaches that could monitor, quantify, and evaluate a process or product's environmental performance for ensuing planning and decision making (Nyland et al., 2003). However, assessing the environmental performance of the plastics value chain with recycling requires a sequence of multiple re-loops creating cascading system and remains a challenge for the standard LCA. As introduced by Malabi Eberhardt et al. (2020), in general, loops in recycling can be categorised as open or closed, where closed refer to recycling into the same material or products and open often refers to downcycling into other materials or products. Recycling methods of plastics could be divided into four main categories, including primary, secondary, tertiary and quaternary recycling (Hopewell et al., 2009), as summarised in Figure 1. Primary and secondary recycling involves mechanical reprocessing. By comparison, primary plastic recycling incurs a lower cost and is suitable for plastic waste recycling with a low contaminant. It is usually referred to as closed-loop recycling, where the same product is produced.

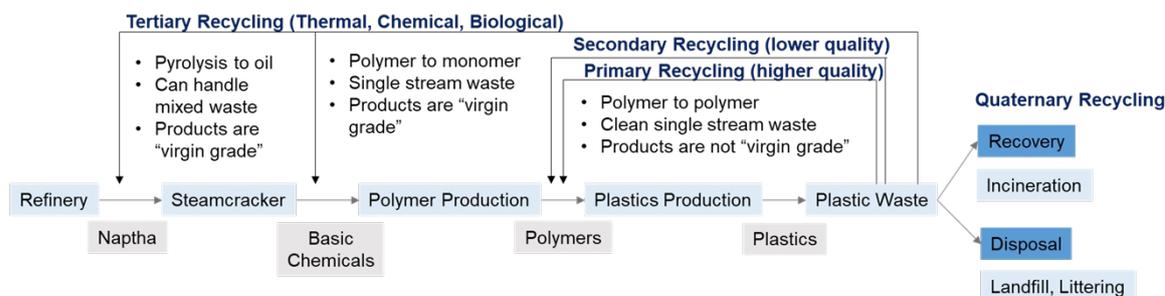


Figure 1: The possible plastics recycling pathway of plastic production, developed by the authors, adapted from Stark et al. (2021)

Secondary recycling refers to downgrading, undergoing sorting, cleaning, melting and reshaping. It has a limited number of cycles for recycling due to quality degradation (Schyns and Shaver, 2021). However, they accounted for most of the share of the recycling rate, owing to the simplicity and low cost. Tertiary recycling (Lee and Liew, 2021) is also known as chemical or feedstock recycling, where the depolymerisation process (or pyrolysis) is usually involved. The polymer is turned into a monomer or the oil (chemical building blocks) to reincarnate new plastics or make other synthetic chemicals. Tertiary recycling is claimed as an infinite system than a downward spiral system, but the infinite circular loop might not necessarily be environmentally sustainable as the process itself consumes resources and could produce a fair amount of footprints. The thermal degradation approach of tertiary recycling has been optimised for most of the common types of plastic waste (Lee and Liew, 2021), and recently biological tertiary recycling has also gained significant attention as a gentler degradation approach to the environment. Vollmer et al. (2020) suggested the CO_{2eq} performance of the end of life technologies for several plastic waste streams as incineration > incineration with energy recovery (quaternary) > landfill > pyrolysis (tertiary) > mechanical recycling > solvolysis (tertiary) > dissolution/ precipitation (tertiary). Polyethylene terephthalate (PET) is among the plastic types which are more mature for chemical recycling, where it can be broken down by glycolysis, methanolysis or hydrolysis (Lee and Liew, 2021).

The GHG of plastic waste ended in landfills is lower than incineration, as the embodied carbon is hardly released over time due to its properties. It highlights the need to assess the other environmental footprints rather than defining the environmental performance merely by climate change issues, especially in the case of plastic. Energy-intensive and price competitiveness with petrochemical feedstock remain the main challenges of tertiary plastic recycling. It should be aware that the different recycling pathways that are highlighted in this study, especially secondary, tertiary and quaternary, are not the ultimate answer to the plastic waste problem where different available solutions, including waste reduction, are required.

There have been vast and comprehensive reviews conducted summarising the recent advancement of plastic recycling. Vollmer et al. (2020) summarised the pathway of chemical recycling, including the emerging technologies to address problems encountered in conventional solvolysis and pyrolysis recycling. Thiounn and Smith (2020) focused on reviewing the chemical recycling for three major polymers (PET, PE and PP). In commenting on the future of plastic recycling, Garcia and Robertson (2017) highlight the importance of new low-energy catalysts and the development of decontamination techniques. Aside from the technical challenges and the economic feasibility, which have been developed reasonably in recent years, there is still a challenge in accounting for the environmental performance of recycling. However, a limited review study was performed to assess the different existing and improved LCA methods to evaluate the environmental sustainability of recycling processes. A fair environmental performance accounting is important to support appropriate plastic management decision-making. End-of-life allocation (Tanguay et al., 2021) is the most common approach in tackling the multiple cycle issue or circular and cascade use of resources, as reviewed intensively by Rehberger and Hiete (2020). Thoemann and Schumann et al. (2018) also reviewed the environmental impacts of cascade utilisation, focusing on the wood-based product. The LCA allocation approaches applied in Malabi Eberhardt et al. (2020) for building components are divided into cut-off, Circular Footprint Formula, 50:50 approach and linearly degressive approach. However, it remains a question of how to justify an appropriate allocation for plastic recycling cases, the main criterion of choice and the underlying meaning (Wilfart et al., 2021). Based on a general review by Jiang et al. (2022) with one subsection dedicated to LCA of plastic chemical recycling, mass and energy or no allocation are suggested as the common approaches. The authors (Jiang et al., 2020) highlighted the lack of a comprehensive LCA model in evaluating the overall sustainability of plastic recycling. This study reviews the LCA approaches proposed for modelling multiple cycles applicable to assessing the plastic products with the recycling stage. The challenges, limitations and way forward are discussed as the stepping stone for future work in developing a robust LCA model to assess the plastic value chain with multiple cycles under different scenarios. The mini-review is divided into two main sections, summarising the different LCA allocation approaches and system boundaries in assessing the environmental performance of plastic, followed by a conclusion section.

2. Life cycle assessment – General approach and system boundaries

Different LCA modelling approaches are applied based on the purpose of the assessment, application area (e.g. basis for policy decisions, strategic decisions in companies, external communication, numeric results), and the parties (e.g. plant manager, consumer, policymakers) who will use the results. Figure 2 shows the different system boundaries in assessing the environmental performance of plastics. The blue boxes show the typical or standard stages included in assessing the environmental performance of a product. In some cases, the end-of-life management (grey boxes), cradle to grave, is included, depending on the question to answer by performing the assessment. For example, if the main focus is to compare the different waste management options, a gate to grave boundaries is considered, where the upstream life cycle stages (production) phases are excluded. The

inclusion of avoided processes accounting (in orange boxes – system expansion (Höglmeier et al., 2014)) happens in consequential LCA but not attributional LCA (Ekvall et al., 2020), as the negative numbers would muddle the estimated share of the global emission that belongs to the product system (Ekvall et al., 2020). The most common approach that follows attributional LCA is the simple cut-off approach, and it is also known as the 100/0 method, where full responsibility (100) is accounted for. One example is by David et al. (2021), quantifying the environmental benefit of biocomposite packaging material, where the cut-off is at recycling, and the burden of recycling is allocated to the product of the first life (with no unburdening effect). However, the use of recovered material in the following life does not bear any environmental burden from a previous life. This approach is further discussed in Section 3, compared to the other allocation approach. The 100/0 and 0/100 cut-off methods could avoid double-counting but do not guarantee physically correct modelling at the product level.

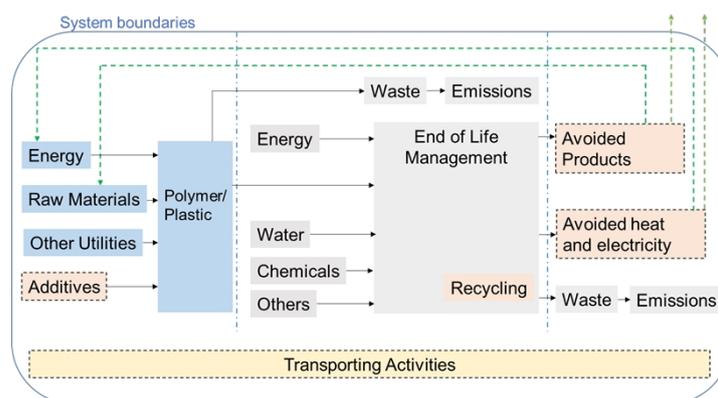


Figure 2: Different system boundaries in assessing the environmental performance of plastics. Blue boxes = the typical or basic systems boundary. Grey, yellow, and orange boxes are the stages/consumption which sometimes excluded from the assessment boundaries. The Figure originates from the authors.

One of the efforts to consider a wider system boundary (system expansion) is by Horodytska et al. (2020), where the avoided product or credit of the avoided impact (0/100 method) is considered. The LCA performance of downcycling, upcycling and incineration of printed plastic waste is performed where the avoided virgin plastics and energy are accounted for. The authors (Horodytska et al., 2020) highlighted that, in general, upcycling has a lower net environmental benefit than downcycling or incineration with energy recovery. However, they also highlighted the reason for this observation which is partially due to the current accounting method for the unburdening effect, limited by the baseline scenario applied, including the substitution rate that affects the avoided virgin plastic production. Schwarz et al. (2021) presented the LCA matrix results of 25 plastic polymers under different treatment approaches and highlighted that the environmental performance of recycling approaches depends on the type of polymer and did not always follow a certain pattern or recycling hierarchy and technology readiness level (TRL). As for most of the LCA and considering the objective of the assessment, collection and use phases are excluded. The results of Schwarz et al. (2021) are significantly different from the work of Horodytska et al. (2020), where the energy recovery (electricity, heat) is not better than upcycling or downcycling of PET (polymer pellet, monomer or feedstock). It highlights the discrepancy of LCA modelling approaches in assessing the effort toward a circular system, such as recycling, resulting in a different insight. Schwarz et al. (2021) applied recycling efficiency and quality factor to replace the virgin material in accounting for the recycling performance. It is helpful in assessing different recycling or end-of-life management of plastics; however, adaption (e.g. the cycles or rounds of recycling and hence the functional unit) is needed for comparison across different materials and for the recycling where the recovered products are applied in other systems. The extension of the system boundary, including the accounting of avoided impacts, often creates allocation problems. This is especially the case of open system recycling (upward green dotted lines in Figure 2), where the products or utilities are not consumed within the original system. One of the allocation challenges is what part of the avoided impacts is the consequences of the use of recycled materials and what is the share of the consequence of material recycling. The allocation of all savings to the use of recycled materials would not encourage recycling in the original system. Other than the allocation issue, there are also strong assumptions about the avoided scenarios, which are dynamic and that the avoided products are used to fulfilling the original demand than the increased consumption (Fan et al., 2020). For example, Horodytska et al. (2020) highlighted that incineration is surely better than recycling when the avoided accounting is assumed to replace energy from fossil fuels rather than the renewables. Ekvall et al. (2021) also highlighted the risk of the current framework of

Product Environmental footprints that creates an incorrect incentive for incineration and insufficient to guide recycling. The environmental impacts of a single-stage cascade, for example, quaternary recycling, are comparatively easier to quantify than multi-stage cascade, for example, primary and secondary recycling and even challenging for tertiary recycling with “infinite” recycling potential and could be used as materials for different processes and stakeholders. However, it is important to assess the environmental performance of multiple cascade or infinite recycling; as highlighted by Thonemann and Schumann (2018), the environmental benefit of cascading use has still to be approved by scientific facts.

3. Life cycle assessment – Allocation approaches

Allocation of burdens between different product systems in LCA is the most common means to account for the recycling stages. It has been discussed for many years, but there is no consensus on different approaches in the literature. The challenges are achieving a representative and fair burden and credit distribution of virgin material production and recycling, the physical reality and meaning, and avoiding double-counting of burden or over-crediting (Civancik-Uslu et al., 2019). Different modelling approaches provide a different degree of motivation to use recycled material and to recycle material, varying from application areas as mentioned in Section 2. It is unlikely that a single method for modelling recycling is adequate for all applications. Allocation is not preferred whenever possible but unavoidable in multiple cycle modelling where information on a single product is needed (e.g. ecolabelling - Civancik-Uslu et al., 2019). The allocation problems usually happen at virgin production, recycling and disposal stages. It is mainly to answer whether the environmental burden of virgin production, recycling and disposal stage should be assigned to the first cycles of product life or the subsequent cycles and in what proportion and the potential credit/unburdening effect. Other than the commonly applied simple cut-off and avoided burden (system expansion) method as discussed in Section 2, value corrected substitution (Koffler and Florin, 2013) is also introduced to address the modelling of downcycling or open-loop recycling (PE International, 2014), particularly for aluminium. However, they are not the only modelling or allocation approaches for multiple cycle assessment.

Figure 3 shows the characteristics of different modelling approaches summarised by Ekvall et al. (2020). There is a trade-off between each method. The Circular Footprint Formula using Factors A and B is among the current framework included in the EU guidelines in the framework of Product Environmental Footprint. However, it is not easy to apply and is less comprehensible. Ekvall et al. (2021) discuss the use of Factor B of the Circular Footprint Formula to minimise the incorrect incentives for incineration. Cut off Plus Credit (module D) and quality-adjusted 50/50 method (Figure 3) consider allocating credit from avoided virgin material production. Allocation at Point of Substitution method (Figure 3) involves allocation at the manufacturing and use stage. The other methods in blue boxes (8-10 in Figure 3) consider pricing justifying the allocation.

Method		A	B	C	D	E	F	G	H	I	J
1	Simple cut-off										
2	Cut-off with economic allocation										
3	Cut-off plus credit										
4	Allocation to virgin material use										
5	50/50 methods										
6	Quality-adjusted 50/50 method										
7	Circular Footprint Formula										
8	Market price-based allocation										
9	Market price-based substitution										
10	Price elasticity approaches										
11	Allocation at the point of substitution										

Criteria			
A	Easy to use	F	Explicit, justified and evaluated
B	Readily available data	G	Comprehensible
C	Generalisable result	H	Relevant to decision-makers
D	Reflects decisive characteristics	I	Legitimate
E	Life cycle scope	J	Reproducible

Figure 3: The allocation method for modelling recycling, developed by the authors, adapted from Ekvall et al. (2020). Green = criterion is fulfilled, yellow = criterion is partially fulfilled, red = criterion is not fulfilled

Figure 4 shows the allocation methods, which are comparatively easier to apply. Based on the simple cut-off (dark green dotted lines), the burdening impact of recycling is allocated to the second life where the recycled material is used in manufacturing. The final life (n^{th} life) bears the burdening impact of recycling and disposal. A similar allocation happens in the allocation to virgin material use method. However, the disposal stage in the n^{th} life is allocated to the 1st life, where the virgin plastics are produced and used in manufacturing. Cut off with economic allocation, on the other hand, have the recycling impacts share between the first and subsequent life. The following life, which uses the recycled material after the end of life of the first product that was produced from virgin material, shared most of the impact (75 %). The 50/50 method has all the burdening impact in

recycling stages, virgin plastic production, and disposal stages shared in a ratio of 50:50, as illustrated in yellow lines. Rehberger and Hiete (2020) classified the allocation approaches into cut-off, arbitrarily chosen, quantification base, and hybrid methods. Arbitrarily chosen is, for example, the 50/50 method, which is assigned randomly, compromising between simplicity and fairness. A quantification base is where the allocation is based on quality, price or other countable units. All the reported modelling for plastic summarised in the review of Rehberger and Hiete (2020) are for open-loop recycling (recycled into other products/material use) with the most commonly applied cut-off method, followed by system expansion and quantitative base allocation. In assessing different LCA allocations of the built environment (concrete, timber etc.), Malabi Eberhardt et al. (2020) suggest the developed linearly degressive approach is more promising than cut off, 50:50 and Circular Footprint Formula in incentivising Circular Economy. However, studies performed for material upcycling are limited, and the works on wood cascading are comparatively more than on plastics. Tanguay et al. (2021) suggest the quality integration in cascade would influence the results by up to 15 %, highlighting the need for adaptation for different materials.

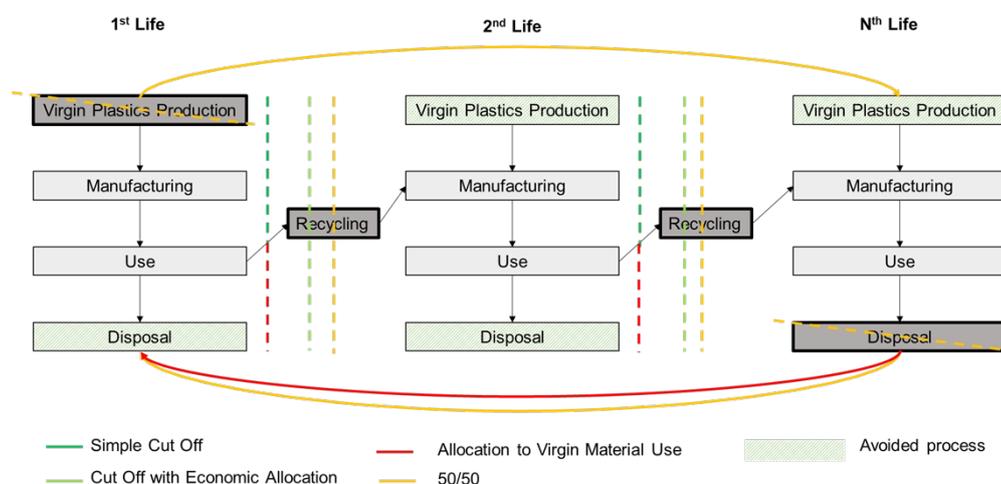


Figure 4: The illustration of the plastic and recycling process (closed or open-looped) and allocation methods, which are comparatively easier to use (Criteria A, Figure 3). The boxes in dark grey and thick outline are the stages where allocation usually occurs (e.g. to assign whether the burdening to the 1st life or the subsequent).

4. Conclusion

The summarised allocation methods should be, in general, applicable to different materials. However, a careful selection based on purpose and adaption is needed. This is especially the case for cradle-to-cradle, upcycling and open-loop recycling of plastics, which are less standardised than wood and aluminium LCA. Different materials have different properties, recycling tendencies, quality changes, environmental hotspots, market share and prices expected to affect the applicability of the allocation method. Plastic is claimed to have a tendency to reach "infinite" recycling through a closed loop, followed by an open loop, and chemical recycling continuously. However, the system is complicated and could involve different actors and technologies. It is important to assess which allocation methods could motivate various stakeholders at different material life cycle stages while reflecting the environmental performance. The full assumption of recycled material in open-loop utilisation has no environmental impacts as the burdening impact of recycling is entirely embraced by the first production could encourage the circularity use. However, not necessary for environmental mitigation. It could create a loophole for unsustainable businesses, especially if recycling has a higher impact than virgin production but is profitable. Future work will compare different methods applied to plastic, including the bioplastics, and cover a more comprehensive range of environmental impacts. Despite great policy attention, climate change is not the primary concern of plastic when ended in landfills. An inappropriate allocation and assessment would incentivise landfilling and incineration of plastics. The identified results could enable the development of mitigation strategies contributed by the plastic value chain and discover leverage that can turn the uncertain situation into favourable ones.

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References

- Civancik-Uslu D., Puig R., Voigt S., Walter D., Fullana-i-Palmer P., 2019, Improving the production chain with LCA and eco-design: application to cosmetic packaging. *Resources, Conservation and Recycling*, 151, 104475.
- David G., Croxatto Vega G., Sohn J., Nilsson A. E., Hélias A., Gontard N., Angellier-Coussy H., 2021, Using life cycle assessment to quantify the environmental benefit of upcycling vine shoots as fillers in biocomposite packaging materials. *The International Journal of Life Cycle Assessment*, 26(4), 738-752.
- Ekvall T., Albertsson G. S., Jelse K., 2020, Modeling recycling in life cycle assessment. IVL Swedish Environmental Research Institute and in RE: Source project database ISBN 978-91-7883-226-2, Gothenburg, Sweden.
- Ekvall T., Gottfridsson M., Nellström M., Nilsson J., Rydberg M., Rydberg T., 2021. Modelling incineration for more accurate comparisons to recycling in PEF and LCA. *Waste Management*, 136, 153-161.
- Fan Y.V., Tan R. R., Klemeš J. J., 2020, A system analysis tool for sustainable biomass utilisation considering the Emissions-Cost Nexus. *Energy Conversion and Management*, 210, 112701.
- Garcia J. M., Robertson M. L., 2017, The future of plastics recycling. *Science*, 358(6365), 870-872.
- Höglmeier K., Weber-Blaschke G., Richter K., 2014, Utilization of recovered wood in cascades versus utilization of primary wood—a comparison with life cycle assessment using system expansion. *The International Journal of Life Cycle Assessment*, 19(10), 1755-1766.
- Hopewell J., Dvorak R., Kosior E., 2009, *Philosophical Transactions of the Royal Society B: Biological Sciences*, 364(1526), 2115-2126.
- Horodytska O., Kiritsis D., Fullana A., 2020, Upcycling of printed plastic films: LCA analysis and effects on the circular economy. *Journal of Cleaner Production*, 268, 122138.
- Jiang J., Shi K., Zhang X., Yu K., Zhang H., He J., Ju T., Liu J., 2022, From plastic waste to wealth using chemical recycling: A review. *Journal of Environmental Chemical Engineering*, 10(1), 106867.
- Koffler C., Florin J., 2013. Tackling the downcycling issue—A revised approach to value-corrected substitution in life cycle assessment of aluminum (VCS 2.0). *Sustainability*, 5(11), 4546-4560.
- Lee A., Liew M. S., 2021, Tertiary recycling of plastics waste: an analysis of feedstock, chemical and biological degradation methods. *Journal of Material Cycles and Waste Management*, 23(1), 32-43.
- Malabi Eberhardt L. C., van Stijn A., Nygaard Rasmussen F., Birkved M., Birgisdottir H., 2020, Development of a life cycle assessment allocation approach for circular economy in the built environment. *Sustainability*, 12(22), 9579.
- Nyland C. A., Modahl I. S., Raadal H. L., Hanssen O. J., 2003, Application of LCA as a decision-making tool for waste management systems. *The International Journal of Life Cycle Assessment*, 8(6), 331-336.
- PE International, 2014. Best practice LCA: End of Life Modelling. <gabi.sphera.com/uploads/media/Webinar_End_of_Life_Oct2014.pdf> accessed 02.06.2022
- Rehberger M., Hiete M., 2020, Allocation of environmental impacts in circular and cascade use of resources— incentive-driven allocation as a prerequisite for cascade persistence. *Sustainability*, 12(11), 4366.
- Schwarz A. E., Ligthart T. N., Bizarro D. G., De Wild P., Vreugdenhil B., Van Harmelen T., 2021, Plastic recycling in a circular economy; determining environmental performance through an LCA matrix model approach. *Waste Management*, 121, 331-342.
- Schyns Z. O., Shaver M. P., 2021, Mechanical recycling of packaging plastics: A review. *Macromolecular Rapid Communications*, 42(3), 2000415.
- Stark, 2021. How Closing the Loop with Chemical Recycling: Potentials and Challenges, <www.process-worldwide.com/how-closing-the-loop-with-chemical-recycling-potentials-and-challenges-a-1036570/>, assessed 18.5.2022
- Tanguay X., Essoua Essoua G. G., Amor B., 2021. Attributional and consequential life cycle assessments in a circular economy with integration of a quality indicator: A case study of cascading wood products. *Journal of Industrial Ecology*, 25(6), 1462-1473.
- Thiounn T., Smith R. C., 2020, Advances and approaches for chemical recycling of plastic waste. *Journal of Polymer Science*, 58(10), 1347-1364.
- Thonemann N., Schumann M., 2018, Environmental impacts of wood-based products under consideration of cascade utilization: A systematic literature review. *Journal of Cleaner Production*, 172, 4181-4188.
- Vollmer I., Jenks M. J., Roelands M. C., White R. J., van Harmelen T., de Wild P., Weckhuysen B. M., 2020, Beyond mechanical recycling: Giving new life to plastic waste. *Angewandte Chemie International Edition*, 59(36), 15402-15423.
- Wilfart A., Gac A., Salaün Y., Aubin J., Espagnol S., 2021. Allocation in the LCA of meat products: is agreement possible?, *Cleaner Environmental Systems*, 2, 100028.