TRACKING CHEMICAL ADDITIVE RELEASES IN THE PLASTICS END-OF-LIFE MANAGEMENT STAGE TO CLOSE THE LOOP

John D. Chea[1, 2], Matthew Conway[1], Austin Lehr[1], Gerardo J. Ruiz-Mercado[2], and
Kirti M. Yenkie[1*]
[1.] Department of Chemical Engineering, Rowan University, Glassboro, NJ, 08028,
USA.
[2.] Office of Research & Development, US Environmental Protection Agency,
Cincinnati, OH, 45268, USA.

Abstract

The continuous increase in US plastic production directly shifts plastic waste toward incineration and landfilling. As a result, research efforts have been made to improve the plastic recycling infrastructure to achieve a circular economy. However, chemical additives within plastics can be released throughout all stages of a plastic life cycle, potentially restricting plastics recycling and reuse at their end-of-life (EoL) stage. Therefore, this work examines the EoL treatment of plastics by tracking and estimating potential releases of chemical additives and emissions during mechanical recycling, incineration, and landfilling activities. The 2018 US municipal solid waste data was used as the basis for all calculations. Additionally, a case study analysis was performed to determine the effect of altering the recycling efficiency on chemical releases, environmental impact, and energy footprint. A python-based graphic user interface (GUI) was created to model the chemical additive releases, greenhouse gas emissions, and energy footprint. This tool can streamline the risk estimation process, allowing decision-makers to adjust parameters and run sensitivity analyses of the existing EoL management processes. The transformation of the US plastic linear economy to circular can thus be supported with the aid of the potential hazards and risks identified with this work.

Keywords

Material Flow Analysis, Plastic Recycling, Sensitivity Analysis

Introduction

Plastics have established their use as a key material in applications ranging from insulation to storage, transportation, and packaging because of their low cost, adaptability, durability, and low weight (Nielsen et al., 2020). However, the current approaches to managing plastics in the end-of-life stage (EoL) are not sustainable and frequently cause the release of hazardous chemical compounds into the environment (Hahladakis et al., 2018). The chemical additives inside plastics are free to migrate because the polymer chains and the additives are not chemically linked. This migration represents the movement of chemical additives through the polymer matrix due to external driving forces. The occurrence frequency depends on temperature, polymer molecular weight, chemical additive molecular weight, compatibility, and solubility

---

[*] Corresponding author.

Email address: yenkie@rowan.edu

(Crompton, 2007). This phenomenon has been a concern during the plastics ‘Use stage’. The driving forces behind chemical additive migration remain active for a long time after disposal.

Furthermore, demand for plastics will continue to rise, with production expected to double between 2019 and 2040 (Nielsen et al., 2020). Without a significant change in current plastics production, usage, and EoL, the ocean is expected to contain more plastics pieces than the amount of fish by 2050, with the plastics industry alone consuming 20% of the total oil produced and 15% of the annual carbon budget, which corresponds to the maximum allowable CO_2 emission that can be emitted to create a minimal increase in global temperature. Over time, plastics in the environment and consumer goods may produce microplastics and nanoplastics that could end up in the digestive tracts of many species (Sridharan et al., 2022). As a result, the existing EoL management activities must be modified to prevent or minimize plastic waste and toxic chemical releases to create a circular economy considering the growing reliance on plastics and the potential for additive migration.

The three main stages of the plastics life cycle are (i) production, (ii) use, and (iii) end of life. Acquisition of raw materials, monomer synthesis, and polymerization are the first steps in the “production stage,” followed by compounding chemical additives to produce plastics for consumer use. Up to 99 percent of the raw materials used to make plastic worldwide are compounds derived from petroleum. (Nielsen et al., 2020). The remaining 1% of production, or about 4 million tons/yr (3.6 billion kg/yr), comes from bio-based and biodegradable feedstocks. These numbers highlight our dependence on non-renewable resources, many of which are hard to recover and may linger in the environment for many years. The “use stage” generates plastic waste processed through the EoL stages. Consumers frequently utilize plastic bags, bottles, cups, containers, and packaging, and these items are regularly discovered in municipal solid waste (MSW). The following four EoL activities determine the fate of plastics: (1) recovery, (2) incineration, (3) landfilling, and (4) littering. Chemical additive releases are anticipated at every stage of the plastics life cycle (Hahladakis et al., 2018).

This work analyzes the existing management of plastic wastes using a material flow analysis to identify potential releases and exposure scenarios creating causes for concerns to the environment, human health, and safety. Thus, a Python-based modeling tool was created to decrease the complexity of the material flow analysis calculations for decision-making and estimate chemical additive releases, greenhouse gas emissions, and energy footprint.

Materials and Methods

All data used for the material flow analysis were estimated using publicly available data, UN trade information, and key research articles (Hahladakis et al., 2018; Horodytska et al., 2020; Jambeck et al., 2015; US EPA, 2020; van Velzen et al., 2017). The calculations in this study prioritized EoL activities of plastic management, such as collection, sorting, mechanical recycling, incineration, and landfilling. General data were defined for each significant EoL pathway, including the number of businesses, facilities, and employees. The EoL routes naturally differ, and these distinctions call for different investigations. The quality of recycled goods is ultimately impacted by problems with plastics, foreign substances, chemical additive contamination, and material deterioration, which can expose facility personnel to toxic substances.

All calculations for the material flow and sensitivity analyses were completed in Microsoft Excel. We then transformed the spreadsheet into a Python-based tool, utilizing a graphical user interface from the Tkinter package for easy navigation.

Results & Discussions

A generic EoL plastics composition was estimated using MSW data from the United States in 2018. Over 35.7 million tons (32.4 billion kg) of EoL plastic were reportedly produced in the US in 2018, according to the US EPA. The municipal plastic waste is made up of 24.1% low-density polyethylene (LDPE), 22.8% polypropylene (PP), 6.3% polystyrene (PS), 11.7% other plastics, and 14.7% polyethylene terephthalate (PET), 17.7% high-density polyethylene (HDPE), 2.4% polyvinyl chloride (PVC), and 14.7%. The plastic waste categories coincide with polymer resin identification codes 1 through 7, with the addition of polylactic acid (PLA). Most of the recycling efforts have been allocated toward recovering PET, HDPE, LDPE, and a select group of uncategorized plastics. Approximately 8.4% of the waste plastics, or up to 3 million tons (2.7 billion kg), were recycled; the remaining 75.8% were landfilled, and the residual 15.8% were incinerated. (US EPA, 2020). Of the amount reported recycled, 4.5% were exported overseas, and 3.9% were recycled domestically, totaling 8.4%. Therefore, almost half of reported recycled plastic is exported overseas and is subjected to a variety of EoL treatments that are not necessarily effective. Therefore, a large accumulation of US EoL plastic in the foreign environment can be observed. Similar EoL statistics were reported by the UN, which found that 9% of worldwide plastic wastes were recycled, 79% were landfilled, and 12%

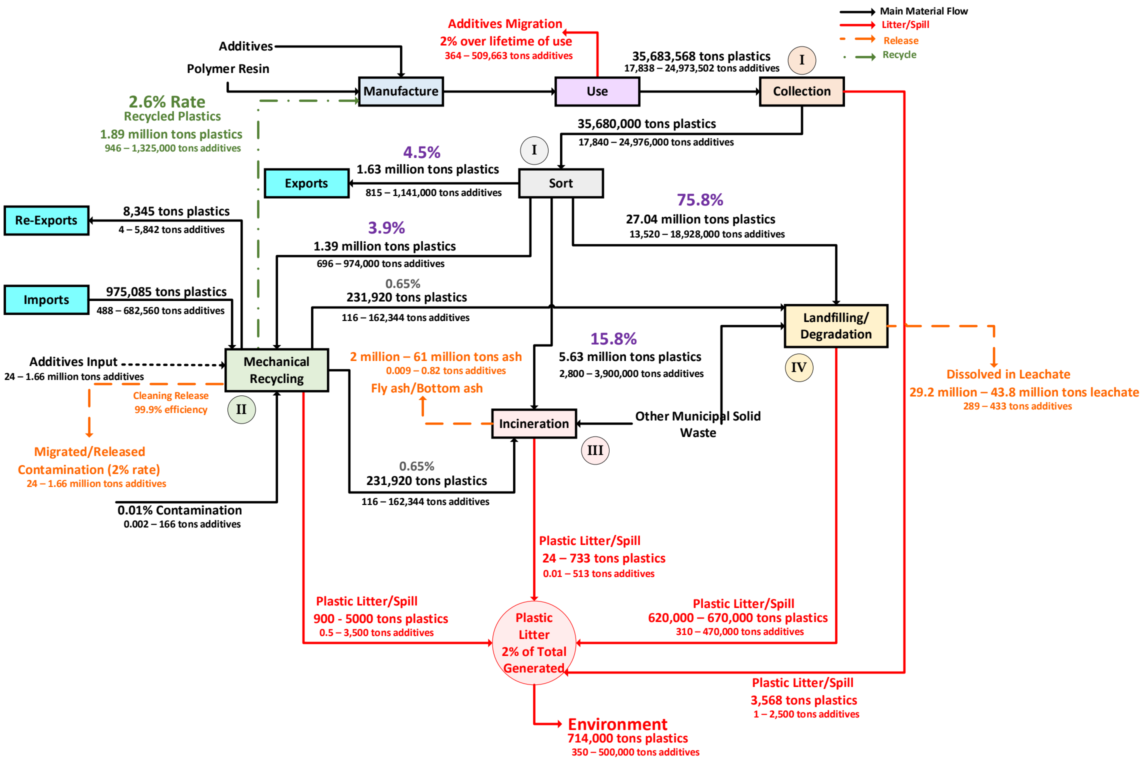


Figure 1. Material flow analysis of the plastic and additive mass flow using the 2018 municipal solid waste data as a basis.

were incinerated. These statistics suggest enhancing the existing infrastructure for plastic waste processing to reduce hazardous exposure and excess environmental accumulation of toxic chemicals. The material flow analysis completed in this research is illustrated in Figure 1.

Consumer plastics are produced using a combination of polymer resins and chemical additives to acquire the necessary properties for specific applications. The chemical additives may include, but are not limited to plasticizers (10–70 wt.%), flame retardants (3–25 wt.%), antioxidants (0.05–3 wt.%), UV stabilizers (0.05–3 wt.%), heat stabilizers (0.05–3 wt.%), slip agents (0.1–3 wt.%), lubricants (0.1–3 wt.%), antistatics (0.1–1 wt.%), curing agents (0.1–2 wt.%), blowing agents (0.5–20.5 wt.%), biocides (0.001–1 wt.%), colorants (0.25–5 wt.%), pigments (0.001–10 wt.%), fillers (0–50 wt.%), and reinforcements (15–30 wt.%) (Hahladakis et al., 2018).

Chemical additive migration to consumer products remains challenging because up to 2% of chemical additives can be released during the use stage (Crompton, 2007). This migration rate, although low, can cause bioaccumulation of toxins and unwanted consequences to the user's health. Particularly, chemicals added to the plastic may seep into the surrounding water and be consumed by marine life. Aquatic life impacted by chemical additive bioaccumulation may eventually return to humans as food.

Chemical additives are commonly used during recycling to maintain stability, compatibility, and properties. However, during reprocessing, the additives can easily migrate out of the polymer matrix, resulting in zones within the polymer matrix with an uneven distribution of chemical additives. The inclusion of additives to aid in material processing reduces the quality of recycled plastics even further because these chemicals are mixed with the existing plastic network. Excess accumulation of toxic chemicals in recycled materials may increase the consumer's risk and exposure. In addition, some additives may not be suitable for health-sensitive applications, reducing the theoretical applicability of recycled plastics overall.

Case Study: EoL Plastic Management in 2018 and Hypothetical Enhancement to a Maximum Technical Feasibility Point

The existing EoL plastic management method was analyzed. We calculated the effects of the existing practices and their hypothetical efficiencies on chemical additive releases, GHG emissions, and energy footprints (Devasahayam et al., 2019; Jeswani et al., 2021). Plastic recovery from the collection, sorting, and mechanical recycling stages can theoretically achieve maximum recovery of 72 % (Brouwer et al., 2020). As a result, the recovery rate of plastics sent for recycling could theoretically range from 0% to 72%. Regardless of the increase in recycling efficiency, the value of plastic waste exported remained constant at 4.5% because imports and export values depend on the year rather than the material flow rate (Zhao et al., 2021). Incineration and landfilling were chosen as the secondary methods for processing non-recyclable plastic and were kept at a constant ratio of 17.2:82.8, corresponding to the ratio reported in 2018. Figure 2 shows that increasing the mechanical recycling rate increases the total chemical additive release, greenhouse gas emissions, and energy footprint.

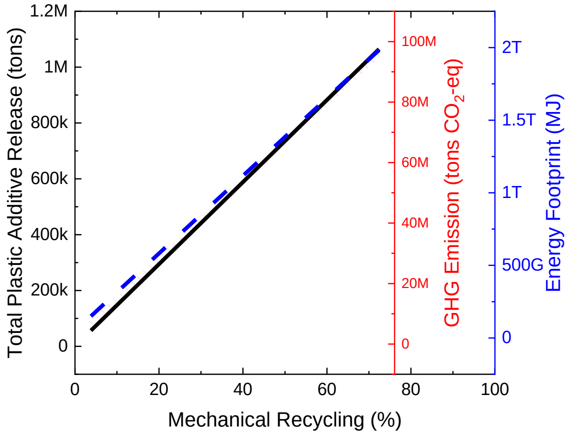


Figure 2. The effects of increasing the plastic recycling rate to the maximum technical feasibility limit

The chemical additive release is directly proportional to chemical additive contamination in recycled plastics. Unrecyclable plastics are sent to incineration and landfilling, respectively, creating opportunities for harmful gas-phase emissions and plastic mass accumulation. Chemical additives may slowly leach into the surrounding environment as leachate over time. Mechanical recycling is expected to release more chemical additives within a given time than other plastic waste processing methods. This statement, however, should not overshadow the importance of increasing the rate of plastic recycling. The successful recovery of EoL plastic reduces the potential for plastic release and accumulation in the environment. Chemical additives would be accumulated within recycled plastics in a more controlled environment. Additional processing could be used to remove harmful chemicals from recycled plastics. In contrast, non-recycled plastics that accumulate in the environment can uncontrollably release harmful chemical additives into the ecosystem.

Python-Based EoL Plastic Tracking

The case study analysis performed in this work was initially completed in Excel and later transformed into a python-based tool. This tool can simulate the effects of altering key parameters (recycling, incineration, landfilling rate, MSW composition, import and export rate, and efficiency at various stages) on chemical releases, greenhouse gas emissions, and energy footprint. Figure 3 displays the major operations contained within the python-based tool. A graphic user interface (GUI) was created through the Tkinter package, allowing all users to input specific information. There are algorithms implemented to check and correct proportion errors related to instances where the total percentage must sum to 100%.

The data used in this work is specifically from 2018. However, this tool also contained 2016 and 2017 data, allowing users to generate a custom dataset representative of a given year. Figure 4 displays a screenshot of the plastic EoL estimation tool input tab, in which the users may use default values reported by the US EPA or enter custom data to simulate a specific waste plastic waste stream.

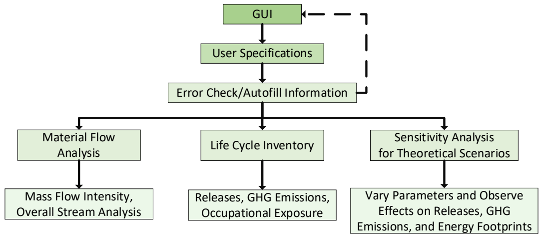
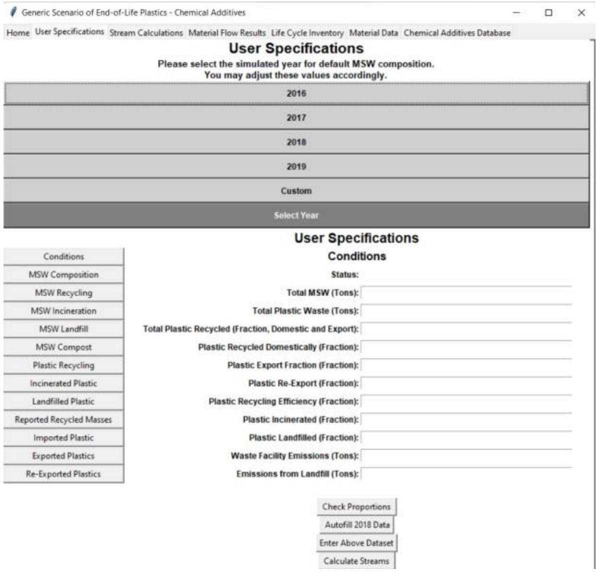


Figure 3. The main features of the python-based chemical release tracker

Three major results can be obtained from this each iteration of specification submission. A material flow analysis, similar to Figure 1, is shown and updated according to the values inputted by the users. Relevant plots and mass flow intensity diagrams are created to illustrate the state of plastic waste management and potential chemical additive releases in the EoL stages. Life cycle inventory presents the second major result, which contains information regarding plastics and chemical additives releases, greenhouse gas emissions, and occupational exposure. This tool can also perform sensitivity analysis by varying specific parameters to the maximum technical feasibility limit as part of the third major result. Furthermore, custom scenarios regarding the usage of chemical recycling, in addition



User Specifications

Please select the simulated year for default MSW composition.
You may adjust these values accordingly.

User Specifications

Conditions

Status:
Total MSW (Tons):
Total Plastic Waste (Tons):
Total Plastic Recycled (Fraction, Domestic and Export):
Plastic Recycled Domestically (Fraction):
Plastic Export Fraction (Fraction):
Plastic Re-Export (Fraction):
Plastic Recycling Efficiency (Fraction):
Plastic Incinerated (Fraction):
Plastic Landfilled (Fraction):
Waste Facility Emissions (Tons):
Emissions from Landfill (Tons):

Figure 4. A preview of the plastics EoL release estimation tool

to the conventional MSW plastics EoL processes, were added to assist with comparative studies.

The development of this python-based tool thus ensured that releases and harmful effects from the current EoL plastic stage are estimated with the necessary plots and valuable information for preliminary studies. Diagrams and plots like Figure 1 and Figure 2 are available as results and can change according to the parameters inputted by the users. We compiled data from multiple sources and stored them in this tool for quicker and more convenient estimation. Additionally, this tool is modular, and can create new scenarios to predict the outcome of various methods and identify concerning steps in the EoL stages that demand immediate attention.

Remarks on Achieving a Circular Economy

A circular economy can be achieved if plastic manufacturers design plastics with considerations for recycling and potential releases. The existing EoL waste management technique should also consider minimizing incineration and landfilling and maximizing the recycling content. This suggestion is difficult to achieve because of difficulties with technological efficiencies, operation costs, incentives, and legislative support (Barra and Leonard, 2018; Hahladakis et al., 2018). Figure 1 demonstrates how the existing EoL waste management created many opportunities for unintentional releases, chemical migration, and exposures. Recycling efficiency has been one of the largest bottlenecks that prevent a circular economy from being achieved. Incineration and landfilling can release fewer chemical additives to the environment at the expense of higher greenhouse gas emissions and land areas, respectively. Chemical additive releases with conventional recycling are controllable because additional processing can be applied to the recycled plastics before the recycled materials undergo a new life cycle. One solution to this problem is implementing a chemical additive removal stage after mechanical recycling to eliminate the primary contamination source. This method may employ solvent extraction and dissolution-precipitation to separate the polymer from the chemical additives that are loosely held in the polymer matrix. The solvent used in this process can be recycled and reused. In addition to mechanical recycling, upcycling methods such as chemical recycling can be considered to recover the original monomer and other pyrolysis-derived products such as aromatics, fuels, and waxes (Gracida-Alvarez et al., 2019). Incentives and policies can be put in place to reduce chemical release. Manufacturers should design plastics with consideration for potential toxic chemical additives released during use and EoL stages and design plastics to be biodegradable or degradable in the presence of a specific substance without adverse health and environmental consequences. The consequences of these methods can be more apparent with the aids from the software tool generated from this work.

Conclusions

Chemical additives are constantly being released into the environment throughout the plastic EoL stages. The intensity of these releases is greatly determined by the earlier plastic management stages, including manufacturing, collection, and sorting. Given that conventional EoL plastic recycling techniques are not optimized to separate chemical additive components from plastics, unwanted contamination and degradation may be introduced to various recycled plastics, increasing toxic substance concentration. As a result, conventional mechanical recycling should be combined with chemical recycling and chemical additive extraction to processing plastic in the EoL stage while minimizing releases efficiently. In plastic EoL management, separating plastics and additives without compromising the original product's structural and mechanical properties can make plastic recycling safer and more sustainable. The Python-based GUI tool developed, although at its preliminary stage, can provide a quick overview to process engineers and policymakers of the releases and exposure during the plastics end-of-life waste management stages. Data on chemical releases and plastic EoL management are typically scattered throughout multiple sources, which increases the difficulty of estimating emissions and releases. This tool compiled the relevant data necessary for modeling the EoL stages of plastics waste management and potential chemical additive releases to create test scenarios, in which various parameters can thus be adjusted to predict changes to emissions, releases, and energy footprints. Although recycling appears to be a straightforward solution to maintaining a circular economy, the releases highlighted in this work and the python-based tool can identify non-intuitive release problems that would otherwise be ignored. This contribution could help achieve the United Nations Sustainable Development Goals by ensuring sustainable consumption and production patterns with plastic waste.

Acknowledgments

This research was supported in part by an appointment to the US Environmental Protection Agency (EPA) Research Participation Program administered by the Oak Ridge Institute for Science and Education (ORISE) through an interagency agreement between the US Department of Energy (DOE) and the US Environmental Protection Agency. ORISE is managed by ORAU under DOE contract number DE-SC0014664. All opinions expressed in this paper are the author's and do not necessarily reflect the policies and views of US EPA, DOE, or ORAU/ORISE.

Disclaimer

The views expressed in this article are those of the authors and do not necessarily represent the views or policies of the EPA. Any mention of trade names, products, or services does not imply an endorsement by the US Government or the EPA. The EPA does not endorse any commercial products, services, or enterprises.

References

Barra, R., Leonard, S.A., 2018. Plastics and the circular economy. Scientific and Technical Advisory Panel to the Global Environment Facility.

Brouwer, M.T., Thoden van Velzen, E.U., Ragaert, K., ten Klooster, R., 2020. Technical Limits in Circularity for Plastic Packages. Sustainability 12, 10021. https://doi.org/10.3390/su122310021

Crompton, T.R., 2007. Additive migration from plastics into food: a guide for analytical chemists, New ed. ed. Smithers Rapra Technology, Shawbury.

Devasahayam, S., Bhaskar Raju, G., Mustansar Hussain, C., 2019. Utilization and recycling of end of life plastics for sustainable and clean industrial processes including the iron and steel industry. Materials Science for Energy Technologies 2, 634–646. https://doi.org/10.1016/j.mset.2019.08.002

Gracida-Alvarez, U.R., Winjobi, O., Sacramento-Rivero, J.C., Shonnard, D.R., 2019. System Analyses of High-Value Chemicals and Fuels from a Waste High-Density Polyethylene Refinery. Part 1: Conceptual Design and Techno-Economic Assessment. ACS Sustainable Chem. Eng. 7, 18254–18266. https://doi.org/10.1021/acssuschemeng.9b04763

Hahladakis, J.N., Velis, C.A., Weber, R., Iacovidou, E., Purnell, P., 2018. An overview of chemical additives present in plastics: Migration, release, fate and environmental impact during their use, disposal and recycling. Journal of Hazardous Materials 344, 179–199. https://doi.org/10.1016/j.jhazmat.2017.10.014

Horodytska, O., Cabanes, A., Fullana, A., 2020. Non-intentionally added substances (NIAS) in recycled plastics. Chemosphere 251, 126373. https://doi.org/10.1016/j.chemosphere.2020.126373

Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law, KL, 2015. Plastic waste inputs from land into the ocean. Science 347, 768–771. https://doi.org/10.1126/science.1260352

Jeswani, H., Krüger, C., Russ, M., Horlacher, M., Antony, F., Hann, S., Azapagic, A., 2021. Life cycle environmental impacts of chemical recycling via pyrolysis of mixed plastic waste in comparison with mechanical recycling and energy recovery. Science of The Total Environment 769, 144483. https://doi.org/10.1016/j.scitotenv.2020.144483

Nielsen, T.D., Hasselbalch, J., Holmberg, K., Stripple, J., 2020. Politics and the plastic crisis: A review throughout the plastic life cycle. WIREs Energy Environ. 9. https://doi.org/10.1002/wene.360

Sridharan, S., Kumar, M., Saha, M., Kirkham, M.B., Singh, L., Bolan, N.S., 2022. The polymers and their additives in particulate plastics: What makes them hazardous to the fauna? Science of The Total Environment 824, 153828. https://doi.org/10.1016/j.scitotenv.2022.153828

US EPA, 2020. National Overview: Facts and Figures on Materials, Wastes and Recycling [WWW Document]. United States Environmental Protection Agency. URL https://www.epa.gov/facts-and-figures-about-materials-waste-and-recycling/national-overview-facts-and-figures-materials

van Velzen, E.U.T., Jansen, M., Brouwer, M.T., Feil, A., Molenveld, K., Pretz, Th., 2017. Efficiency of recycling post-consumer plastic packages. Presented at the PROCEEDINGS OF PPS-32: The 32nd International Conference of the Polymer Processing Society - Conference Papers, Lyon, France, p. 170002. https://doi.org/10.1063/1.5016785

Zhao, C., Liu, M., Du, H., Gong, Y., 2021. The Evolutionary Trend and Impact of Global Plastic Waste Trade Network. Sustainability 13, 3662. https://doi.org/10.3390/su13073662