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Environmental Impacts from Organic and Plastic Waste Management

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By

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Committee in charge:

Professor Robert Harley, Chair
Dr. Corinne Scown
Professor Marta Gonzalez

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Abstract

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Most municipal solid waste in the United States is currently landfilled despite growing concerns of waste accumulation and associated greenhouse gas emissions. In response, significant efforts have been made to implement and pursue zero-waste goals and develop circular economy strategies to recover value from waste products. My research aims to inform decision-makers on the environmental tradeoffs between waste management strategies by evaluating the emissions implications of various novel and existing technology options for organic and plastic waste, two major components of municipal solid waste streams. This dissertation includes three studies: a life-cycle assessment of organic waste management strategies, a comprehensive and quantitative review of emissions from composting organic waste, and a life-cycle assessment of plastic waste management strategies. All of this work provides evidence of the benefits from landfill diversion and suggests more sustainable solutions for both organic and plastic waste management. A more detailed summary of these studies and their results are provided below.

Life-Cycle Greenhouse Gas Emissions and Human Health Tradeoffs of Organic Waste Management Strategies

Waste-to-energy systems can play an important role in diverting organic waste from landfills. However, real-world waste management can differ from idealized practices, and emissions driven by microbial communities and complex chemical processes are poorly understood. This study presents a comprehensive life-cycle assessment, using reported and measured data, of competing management alternatives for organic municipal solid waste including landfilling, composting, dry anaerobic digestion (AD) for the production of renewable natural gas (RNG), and dry AD with electricity generation. Landfilling is the most greenhouse gas (GHG)-intensive option, emitting nearly 400 kg CO_{2e} per tonne of organic waste. Composting raw organics resulted in the lowest GHG emissions, at -41 kg CO_{2e} per tonne of waste, while upgrading biogas to RNG after dry AD resulted in -36 to -2 kg CO_{2e} per tonne. Monetizing the results based on social costs of carbon and other air pollutant emissions highlights the importance of ground-level NH_3 emissions from composting nitrogen-rich organic waste or post-AD solids. However, better characterization of

material-specific NH_3 emissions from landfills and land-application of digestate is essential to fully understand the tradeoffs between alternatives.

Greenhouse Gas and Air Pollutant Emissions from Composting

Composting can divert organic waste from landfills, reduce landfill methane emissions, and recycle nutrients back to soils. However, the composting process is also a source of greenhouse gas and air pollutant emissions. Researchers, regulators, and policy decision-makers all rely on emissions estimates to develop local emissions inventories and weigh competing waste diversion options, yet reported emission factors are difficult to interpret and highly variable. This study reviews the impacts of waste characteristics, pretreatment processes, and composting conditions on CO_2, CH_4, N_2O, NH_3 and VOC emissions by critically reviewing and analyzing 38 emission factors from 46 studies. The values reported to-date suggest that CH_4 is the single largest contributor to 100-year global warming potential (GWP_{100}) for yard waste composting, comprising approximately 80% of total GWP_{100}. For nitrogen-rich wastes including manure, mixed municipal organic waste, and wastewater treatment sludge, N_2O is the largest contributor to GWP_{100}, accounting for half to as much as 90% of the total GWP_{100}. If waste is anaerobically digested prior to composting, N_2O, NH_3 and VOC emissions tend to decrease relative to composting the untreated waste. Effective pile management and aeration are key to minimizing CH_4 emissions. Increasing aeration of piles, while useful for minimizing CH_4 emissions, can drive increases in NH_3 emissions in some cases.

Complementary Roles for Mechanical and Solvent-Based Recycling in Low-Carbon, Circular Polypropylene

Plastic recycling presents a vexing challenge. Mechanical recycling offers substantial GHG emissions savings relative to virgin plastic production but suffers from degraded aesthetic and mechanical properties. Polypropylene, one of the most widely used and lowest-cost plastics, is particularly susceptible to declining properties, performance, and aesthetics across a succession of mechanical recycles. Advanced processes, such as solvent-assisted recycling, promise near-virgin quality outputs at a greater energy and emissions foot-print. Mechanical and advanced recycling are often presented as competing options, but real-world plastic waste streams are likely to require preprocessing regardless of whether they are routed to an advanced process. This study quantifies the life-cycle GHG implications of multiple recycling strategies and proposes a system in which mechanical and solvent-assisted recycling can be leveraged together to boost recycling rates and satisfy demand for a wider range of product applications. Polypropylene can be recovered from mixed-plastic bales produced at material recovery facilities and processed through mechanical recycling, with a varying fraction sent for further upgrading via solvent-assisted recycling to produce material approved for food packaging and other higher-quality applications. The resulting mechanically recycled rigid polypropylene reduces life-cycle greenhouse gas emissions by 80% relative to the same quantity of virgin material, while the upgraded higher-quality material achieves GHG savings of 30%.

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Chapter 1

Introduction

On federal, state, and local levels, U.S. policymakers are setting zero-waste goals to divert waste from landfills and reduce greenhouse gas (GHG) emissions. Despite these efforts, landfilling is still the predominant end-of-life (EOL) pathway for municipal solid waste (MSW) accounting for 50% of MSW generated in 2018.[1] Waste packed into landfills create anaerobic environments where microbial degradation of organic wastes produces methane, a potent GHG. Today, landfills are still the third largest contributor to national methane emissions.[2] While there is generally consensus regarding the negative externalities of landfilling, the relative benefits of alternative waste management processes and EOL pathways are uncertain. The most effective landfill diversion strategy is reducing waste generation, but developing infrastructure to divert generated MSW from landfills to other EOL pathways is also a key component of zero-waste strategies.[3]

Sustainable alternatives to landfilling include composting for organic waste and recycling for plastics, paper, glass and metals. Unlike landfilling, these alternative technologies are material specific and require sorting of the MSW stream. MSW is composed of a variety of materials including paper, food waste, yard trimmings, plastics, metals, glass, and other non-hazardous materials that are disposed of by consumers, households, and businesses (Figure 1.1). Further development of sustainable EOL pathways must therefore be supplemented with adequate sorting infrastructure. Optimizing zero-waste strategies to reduce environmental impact requires system-wide understanding of the emissions impacts of implementing or scaling up landfilling alternatives and the associated infrastructure needed to recover and sort relevant waste materials.

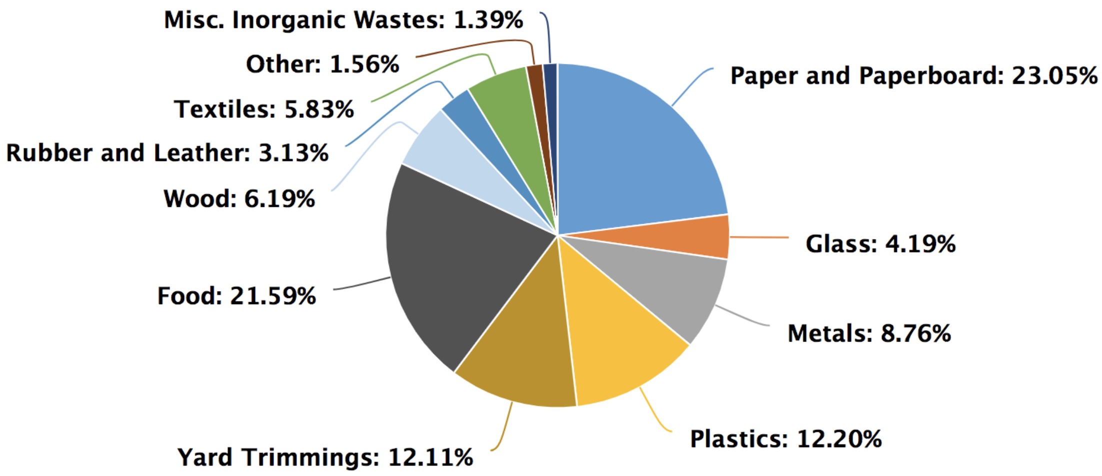

Figure 1.1 Composition of municipal solid waste stream
This chart shows MSW generation by material type in the United States in 2018. Source: EPA (2018)[1]

The research in this dissertation explores the management of two distinct, but commingled components of the MSW stream: organic and plastic waste. While these two waste streams require separation for effective EOL management, these carbon-based materials are still connected in a sustainable, circular waste system. Plastic waste persistently contaminates organic waste management processes and vice versa. The development of bio-based and compostable plastic further connects these two waste streams. Some bioplastics can be produced from processes that use organic waste as a feedstock. Other bioplastics are designed to be biodegradable and are meant to be treated alongside other organic waste. As the circular economy and bio-based industries continue to develop, comprehensive understanding of various EOL pathways and how they interact with each other from a system-wide perspective will be imperative to designing sustainable waste management systems.

1.1 Background on Organic Waste

The organic waste component of MSW includes paper and paperboard, food waste, yard trimmings, and wood; these materials combined make up the majority of the MSW waste stream (Figure 1). Because organic waste drives the GHG impact of landfills and is generated in relatively high volumes, diverting this component of the MSW stream from landfills is particularly important. Alternatives to landfilling for organic waste include composting and anaerobic digestion (AD). Paper and paperboard are organic materials and technically can be composted; however, paper recycling is an effective, profitable process with a well-established industry so there is little reason to expand paper composting. In fact, paper and paperboard explain 67% of recycling in the U.S.[4] More novel research is needed for non-paper, organic waste that is more typically composted. For the purposes of this dissertation, “organic waste” will mainly refer to non-paper organic waste.

While both alternatives to landfilling involve the microbial decomposition of waste, composting happens in aerobic environments and AD involves controlled anaerobic conditions. AD produces biogas via similar processes that produce methane in a landfill, but in a controlled environment where biogas is recovered for energy production and gaseous leaks to the atmosphere are reduced. In an AD system, organic waste is fed into closed digesters attached to a biogas recovery system. Biogas can then be used to generate electricity, upgraded into renewable natural gas (RNG), or converted into bioproducts. The remaining biosolids in the digester is a nutrient-rich byproduct of AD that can be composted or directly applied to land. Chapter 2 of this dissertation discusses how organic waste management strategies can incorporate AD and evaluates the system-wide emission implications of AD systems relative to landfilling and composting.

Composting, in comparison to AD, is the more conventional alternative to landfilling. Even still, only 8% of total MSW generated in 2018 was composted, corresponding to about 23% of total organics and 37% of non-paper organics.[4] During composting, organics undergo microbial decomposition in an aerobic environment, primarily emitting biogenic carbon dioxide (CO_2) which is considered to be climate neutral. The composting process also generates CH_4 and N_2O among other air pollutants, but composting is still considered favorable to landfilling with respect to GHG mitigation.[9] Composting has a secondary GHG reduction benefit which comes from producing compost, a nutrient-rich soil amendment. Compost applied to land is beneficial to plant growth and can offset the need for synthetic fertilizers which are energy- and emissions-intensive to produce.[10,11] Chapter 3 of this dissertation investigates variability in the direct emissions from composting, providing a comprehensive review of relevant literature and a quantitative guide to selecting emission factors for waste management research.

1.2 Background on Plastic Waste

Plastic waste holds promise for GHG emissions reduction because of its potential for circularity. There are no substantial, direct GHG benefits from diverting plastic waste from landfills; synthetic polymers are durable, do not easily degrade, and accumulate rather than contributing to landfill GHG emissions.[12] Promising, sustainable alternatives to landfilling plastic waste include mechanical recycling and solvent-assisted recycling. These EOL pathways have non-zero GHG footprints from either direct emissions and/or from energy and materials consumption.[13–15] When only considering waste management, landfill diversion of plastic waste to alternative EOL pathways does not seem to hold any GHG reduction benefits. However, when considering an expanded system boundary including plastic supply chains, the recycling pathways become more attractive EOL options for plastic waste. Recycling contributes to plastic circularity and can offset demand for virgin polymer production which is fossil-based and emissions-intensive (Figure 1.2). In 2015, plastic production accounted for 4.5% of global GHG emissions.[16] GHG emissions associated with plastic EOL pale in comparison. Though the offset benefit can be substantial because of a variety of limitations and obstacles, recycling rates are very low; in the U.S., only about 6% of plastics are recycled.[17]

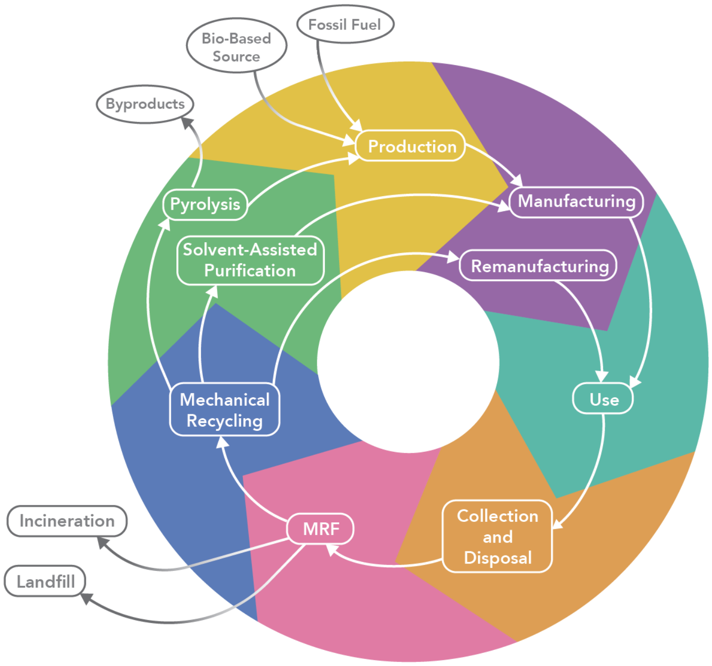

Figure 1.2 Plastic circularity

This diagram is generalizable for most common thermoplastics with the exception of polystyrene which can probably avoid mechanical recycling before solvent-assisted purification. The circular path including pyrolysis depicts chemical or feedstock recycling. The circular path including solvent-assisted purification depicts solvent-assisted recycling.

The contribution to plastic circularity and potential emission reduction of recycling varies based on purity of sorted plastic waste, type of recycling process and quality of the recycled products. Before any efficient recycling process, plastic waste is sorted from other recyclable components of the MSW stream.[18] Sorting first occurs at material recovery facilities (MRFs) where recyclable waste is separated by general material type (e.g. plastics, fibers, metals, glass). Where there is demand for recycling feedstock, some MRFs also separate plastics by polymer type using optical sorters and near infrared technology.

Mechanical recycling involves grinding and re-extruding sorted plastic waste to produce recyclate for remanufacturing.[19] During this process, long polymer chains are broken and thermal stress can be introduced. Because of such physical degradation and difficult-to-separate impurities in plastic waste streams, the material produced, called recyclate, is usually of lower quality than virgin polymer material.[20] Recyclate can be blended with virgin material to create a stronger or higher quality product, but realistically, mechanically recycled material can only be used for a fraction of virgin polymer applications and products.

Solvent-assisted recycling is emerging as a strategy for producing high quality recyclates which are similar in physicochemical properties to virgin polymers.[21] Examples of solvent-assisted recycling processes are dissolution and depolymerization. On their own, these processes remove impurities and recover polymers without breaking polymer chains or causing thermal stress [22]. However, they do not necessarily avoid losses associated with the mechanical recycling EOL pathway. Current scientific literature implies that grinding and extrusion during mechanical recycling is avoided with solvent-based recovery, but industry interviews with plastic sorting and recycling facilities have revealed that in real-life settings, these processes are typically a part of pre-treatment before chemical or solvent-based recycling. Because of this disconnect between science and industry, solvent-based recycling has not been accurately assessed for its environmental and circularity impacts. Chapter 4 of this dissertation aims to resolve this disconnect and offers a rigorous assessment of solvent-based recycling using polypropylene as an example.

1.3 Broader Research Objective

Broad Research Questions:

- What are the system-wide emissions implications of implementing circular waste management strategies?
- What are effective strategies for reducing environmental impacts from organic and plastic waste management?

Effective, sustainable MSW management is a complex, evolving problem involving many industries and decision-making bodies. The research presented in this dissertation aims to inform effective policy making with robust life-cycle assessments of existing waste infrastructure and current or novel technology options for both organic and plastic waste.

I am particularly focused on these two components of the MSW stream because diverting these materials from landfills is essential for reaching zero-waste goals and can reduce GHG emissions from the waste sector, either directly or indirectly. Diverting organic waste from landfills has obvious GHG benefits because organic material drives methane emissions from landfills. Plastic wastes, on the other hand, are relatively stable in landfills and do not contribute significantly to associated GHG emissions. However, diverting plastic waste to recycling can have upstream GHG benefits by increasing circularity and reducing emissions-intensive, virgin resin production. This dissertation examines technology options for both types of waste, quantifying emissions from stand-alone processes and system-scale strategies.

1.4 Approach and Methodology

I approach my work with a system-wide perspective, looking to understand waste life-cycles and identify key processes that drive cradle-to-grave impacts. The primary research tool I employ to conduct my studies is life-cycle assessment (LCA). LCA is a widely recognized, standardized methodology that involves four main phases: goal and scope definition, life cycle inventory, impact assessment and interpretation.[23] I use an input-output physical units-based hybrid LCA model that integrates the best available estimates from scientific literature and empirical data from industry partners to assess GHG emissions (CO_2, CH_4 and N_2O) and other air pollution emissions with human health impacts. These other pollutant types include nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOC), sulfur dioxide (SO_2), carbon monoxide (CO), ammonia (NH_3), and particulate matter ($PM_{2.5}$). The research in this dissertation uses LCA to evaluate the environmental impact of different management strategies for organic and plastic waste. The scope and system boundaries are determined for each study separately. When relevant, I use an expanded system boundary to credit co-products with offset credits. Mass and energy balances needed for life-cycle inventories are taken from literature, offered by industry partners or generated from process modeling. Final impact assessment is produced by the hybrid model and interpretation varies from study to study. Further details are provided for each LCA study in Chapters 2 and 4.

References

(1) EPA. *National Overview: Facts and Figures on Materials, Wastes and Recycling*; U.S. Environmental Protection Agency, 2018.

(2) EPA. *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2020*; U.S. Environmental Protection Agency, 2022.

(3) IPCC. *Climate Change 2014: Mitigation of Climate Change. Working Group III Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*; Cambridge University Press, 2014.

(4) EPA. *Advancing Sustainable Materials Management: 2018 Facts and Figures Sheet*; U.S. Environmental Protection Agency, 2020.

(5) Ward, A. J.; Hobbs, P. J.; Holliman, P. J.; Jones, D. L. Optimisation of the anaerobic digestion of agricultural resources. *Bioresour. Technol.* **2008**, *99*, 7928–7940.

(6) Chatterjee, B.; Mazumder, D. Anaerobic digestion for the stabilization of the organic fraction of municipal solid waste: A review. *Environ. Rev.* **2016**, *24*, 426–459.

(7) De Baere, L. Anaerobic digestion of solid waste: state-of-the-art. *Water science and technology* **2000**, *41*, 283–290.

(8) Clarke, W. P. The uptake of anaerobic digestion for the organic fraction of municipal solid waste - Push versus pull factors. *Bioresour. Technol.* **2018**, *249*, 1040–1043.

(9) Lou, X. F.; Nair, J. The impact of landfilling and composting on greenhouse gas emissions--a review. *Bioresour. Technol.* **2009**, *100*, 3792–3798.

(10) Boldrin, A.; Andersen, J. K.; Møller, J.; Christensen, T. H.; Favoino, E. Composting and compost utilization: accounting of greenhouse gases and global warming contributions. *Waste Manag. Res.* **2009**, *27*, 800–812.

(11) Favoino, E.; Hogg, D. The potential role of compost in reducing greenhouse gases. *Waste Manag. Res.* **2008**, *26*, 61–69.

(12) Geyer, R.; Jambeck, J. R.; Law, K. L. Production, use, and fate of all plastics ever made. *Sci. Adv.* **2017**, *3*, e1700782.

(13) Lazarevic, D.; Aoustin, E.; Buclet, N.; Brandt, N. Plastic waste management in the context of a European recycling society: Comparing results and uncertainties in a life cycle perspective. *Resources, Conservation and Recycling* **2010**, *55*, 246–259.

(14) Hou, P.; Xu, Y.; Taiebat, M.; Lastoskie, C.; Miller, S. A.; Xu, M. Life cycle assessment of end-of-life treatments for plastic film waste. *J. Clean. Prod.* **2018**, *201*, 1052–1060.

(15) Moretti, C.; Hamelin, L.; Jakobsen, L. G.; Junginger, M. H.; Steingrimsdottir, M. M.; Høibye, L.; Shen, L. Cradle-to-grave life cycle assessment of single-use cups made from PLA, PP and PET. *Resources, Conservation and Recycling* **2021**, *169*, 105508.

(16) Cabernard, L.; Pfister, S.; Oberschelp, C.; Hellweg, S. Growing environmental footprint of plastics driven by coal combustion. *Nat. Sustain.* **2021**.

(17) Di, J.; Reck, B. K.; Miatto, A.; Graedel, T. E. United States plastics: Large flows, short lifetimes, and negligible recycling. *Resources, Conservation and Recycling* **2021**, *167*,

105440.
(18) Pressley, P. N.; Levis, J. W.; Damgaard, A.; Barlaz, M. A.; DeCarolis, J. F. Analysis of material recovery facilities for use in life-cycle assessment. *Waste Manag.* **2015**, *35*, 307–317.
(19) Larrain, M.; Van Passel, S.; Thomassen, G.; Van Gorp, B.; Nhu, T. T.; Huysveld, S.; Van Geem, K. M.; De Meester, S.; Billen, P. Techno-economic assessment of mechanical recycling of challenging post-consumer plastic packaging waste. *Resources, Conservation and Recycling* **2021**, *170*, 105607.
(20) Ragaert, K.; Delva, L.; Van Geem, K. Mechanical and chemical recycling of solid plastic waste. *Waste Manag.* **2017**, *69*, 24–58.
(21) Zhao, Y.-B.; Lv, X.-D.; Ni, H.-G. Solvent-based separation and recycling of waste plastics: A review. *Chemosphere* **2018**, *209*, 707–720.
(22) Collias, D. I.; James, M. I.; Layman, J. M. Chapter 3. Solvent-Based Recycling. In *Circular economy of polymers: Topics in recycling technologies*; American Chemical Society: Washington, DC, 2021; pp. 33–59.
(23) ISO. *Environmental management — Life cycle assessment — Requirements and guidelines (ISO Standard No. 14044)*; International Organization for Standardization, 2006.

Chapter 2

Life-Cycle Greenhouse Gas Emissions and Human Health Tradeoffs of Organic Waste Management Strategies

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2.1 Introduction

Local and state governments are pursuing ambitious "zero waste" policies with the goal of reducing methane emissions to the atmosphere and minimizing the quantity of waste sent to landfills. For example, California's strategy for reducing short-lived climate pollutant emissions (Senate Bill 1383) includes a goal to reduce the fraction of organic waste sent to landfills by 75% in 2025 relative to 2014 levels.[1] The highest-emitting point sources of methane in California are a subset of the state's landfills.[2] Dedicated facilities capable of processing mixed solid organic waste streams will be critical to meeting ambitious organics diversion and renewable energy goals.[3,4] Organic waste anaerobic digestion projects can also earn valuable credits for producing low-carbon fuel when biogas is sold for use in transportation applications. As of 2019, the only net negative carbon-intensity fuel pathways approved as part of the California Low Carbon Fuel Standard (LCFS) are based on landfill gas utilization, anaerobic digestion (AD) of manure, and AD of mixed organic solid waste.[5] Previous literature, as reviewed by Morris et al.[6], overwhelmingly agree that the GHG footprint of landfilling organic waste is higher relative to composting or waste-to-energy by as much as a factor of nine, even when landfill gas is captured and utilized.[7,8] However, there is less consensus around the GHG footprints of specific waste-to-energy and composting options, and limited research is available on non-GHG emissions.

Cities hoping to reduce landfill disposal of organic waste must weigh a complex set of competing options across a range of environmental metrics including GHG emissions, air quality and human health burdens, public nuisances such as odor impacts, and environmental justice implications. In this study, we conduct a rigorous life-cycle assessment (LCA) that integrates the best available estimates across the scientific literature and newly-collected empirical data to explore the climate and human health tradeoffs between landfilling, composting, and dry AD of mixed municipal organic waste. Our choice to focus on dry AD (solids loading 22-40% versus < 16% for wet AD[9]) for waste-to-energy stems from its usefulness in processing solid organic waste streams, particularly those with appreciable inorganic contamination, in dedicated facilities and its potential to reduce costs.[10–13] This study also explores variations in the management of solid digestate (residual solids remaining after AD), including landfilling, raw digestate application to land, and composting, including estimated net GHG impacts and fertilizer offset credits after the material is applied to working lands. By establishing a system boundary that extends from waste collection through application of residual solids/compost to soils, this study provides a comprehensive analysis of life-cycle GHG emissions (carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O)), air pollutant emissions (nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOC), sulfur dioxide (SO_2), carbon monoxide (CO), ammonia (NH_3), and particulate matter ($PM_{2.5}$)), and monetized climate and human health damages associated with organic waste management options.

2.2 Methods and Data

Clearly defined and sufficiently expansive system boundaries are essential to understanding the tradeoffs between different organic waste management and utilization strategies, along with input data that is as robust and representative as possible. Trucking distances, landfill emissions, composting emissions, and net emissions post-land application are all closely tied to the specifics of a location, waste composition, and detailed management strategy. Attempting to quantify a broadly applicable set of average values is of limited usefulness. We have chosen to begin with an existing set of operations in San Jose, California. Specific mass and energy balances, emission rates, and transportation distances are tied to a dry AD facility built and operated by Zero Waste Energy Development Company (ZWEDC), referred to simply as ZWEDC in the following sections (see Figure A2 for an aerial photo). In addition to the ZWEDC case, we evaluate alternative management options for the same material as variations on this scenario (see Figure 2.1). In the existing ZWEDC operations, mixed municipal organic waste (largely dominated by food waste and food-soiled paper) is sent to a dry AD facility and raw biogas is combusted to generate electricity for on-site use and export to the grid. The solid digestate is sent to a composting facility before it is ultimately applied to land as finished compost (Figure A7 shows detailed ZWEDC operations). For the purposes of this study, we will refer to the ZWEDC waste stream as "mixed organics", which are approximated as food waste. The additional hypothetical alternatives include: landfilling all mixed organics, composting all mixed organics, variations on the ZWEDC configuration in which digestate is either directly land-applied or landfilled, dry AD with biogas upgrading for pipeline injection to offset natural gas, and dry AD with biogas upgrading to fuel an otherwise diesel-powered truck fleet. Key details of these scenarios are discussed in following subsections.

To compare these scenarios on a common basis, we express all emissions in term of one wet tonne of mixed organic waste processed. The question we seek to answer is: given a unit mass of organic waste, what management strategy results in the most favorable net GHG and human health impacts? The results are dependent on the waste composition, and for this analysis, the mixed commercial organics processed at ZWEDC are approximated as food waste. Visual inspection at ZWEDC indicated that the organics received by ZWEDC are, in large part, food and food-soiled paper products (Figure A1), although the exact composition varies day-to-day and is not characterized on a regular basis. For the landfilling and composting scenarios, as well as for hypothetical variations on ZWEDC operations such as biogas upgrading to RNG, best-available literature and industry values form the basis of our analysis. We expect these results to be generalizable in the U.S. national and international context for similar waste mixtures and technologies, with the exception of possible variations in composting and land application emissions. Landfill emissions will also be higher in states and countries that do not tightly regulate fugitive emissions.

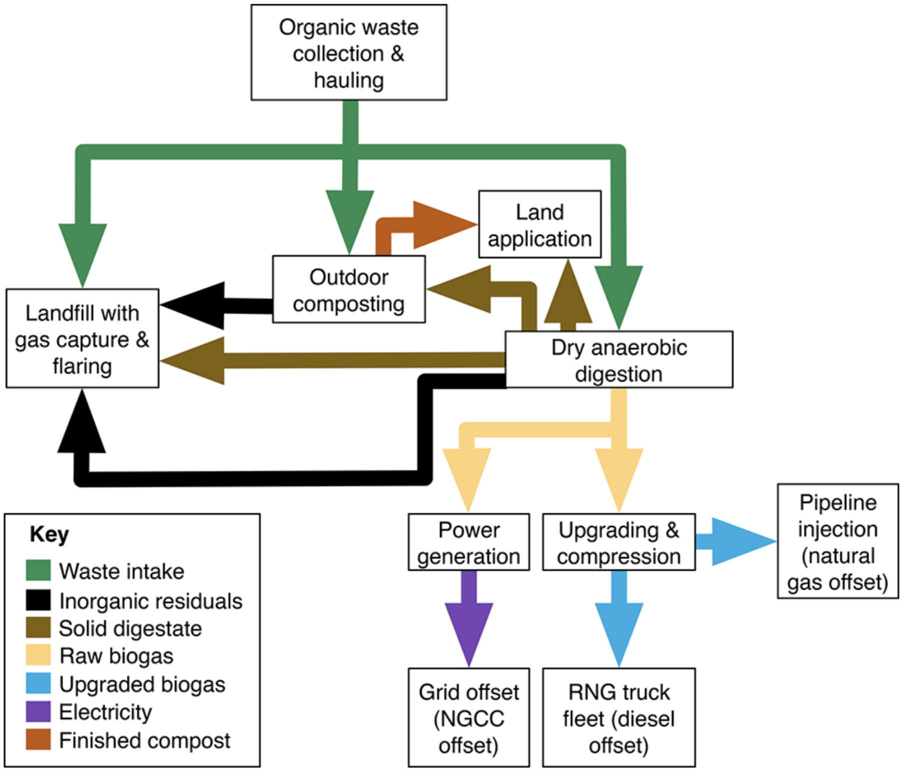

Figure 2.1 System boundary for life-cycle assessment

2.2.1 Landfilling Organic Waste

The most common basis for comparison in organic waste management is landfilling. This reflects "business as usual" practices for 76% of food waste and other collected MSW organics across the U.S.[14] In the specific ZWEDC case, waste would be transported for disposal at the Newby Island Landfill. Large commercial waste streams (e.g., grocery stores and company cafeterias) are hauled directly, whereas municipal streams are sent first to processing facilities for initial sorting. In places like California where there is a marketable need for isolated organic waste streams, sorting/processing facilities may conduct sorting for organics in addition to plastic or paper sorting for recycling. Emissions sources in this scenario include diesel trucks hauling waste from commercial facilities and waste sorting/processing facilities to the landfill, fugitive emissions from waste decomposition in the landfill not captured by the gas capture system, and emissions from the landfill gas flare. We account only for emissions that occur within 100 years of disposal. As mentioned above, we approximate digested organics at ZWEDC as food waste. Fugitive landfill gas emissions are based on food waste-specific data in the literature.[15,16] Food waste decays relatively quickly and to some extent before individual landfill cells can be capped and connected to the gas capture system, so a significant proportion of total methane emitted over their lifetime is emitted to the atmosphere.[15] The emissions of NO_x, NH_3, SO_2, CO, NMVOCs, and $PM_{2.5}$ emissions from the landfill operation and flaring are estimated using data from Ecoinvent (Table A1). We do not account for landfills' potential to sequester biogenic carbon, because the global warming potential offset is fairly small compared to methane emitted and uncertainty surrounding the fugitive methane emissions and related sequestration offset is captured in our sensitivity analysis.[15]

2.2.2 Composting Organic Waste

The most conventional alternative to landfilling is composting of raw organic waste. In the U.S., 61% of yard trimmings are composted and only 5% of mixed organics/food waste is composted.[14] Composting can be a useful alternative for diverting either raw organic waste or further processing solid digestate to make it more suitable for land application. That said, even well-managed compost can release NH_3, N_2O, CH_4, SO_2, CO and odor. These emissions are not well-studied across a range of starting materials, management techniques, and local climates.[17–20] In the raw organics composting scenario, we model direct transportation of all raw organic waste to the Z-Best composting facility near the City of Gilroy (approximately 70 km from ZWEDC), which is an outdoor composting operation capable of handling up to 1,200 tonnes of organic waste per day. This longer driving distance will likely be representative of large-scale composting options for cities across the U.S., given odor and emissions concerns associated with such operations. In the scenario where all organic waste is shipped directly to Z-Best for composting rather than ZWEDC for digestion, we assume it is bagged and composted for 14 weeks as per typical practice at Z-Best. We assume finished compost is applied to cropland as a soil amendment and partial fertilizer replacement.[21–23] This compost ultimately displaces the need for urea fertilizer (46% nitrogen by mass), and the offset credit is calculated on the basis of nitrogen, using an assumption of 1.7% nitrogen content in the compost.[24,25] The life-cycle of urea production is modeled assuming electricity, transportation (truck and rail), and natural gas production in the United States (see Table A2).

2.2.3 Dry Anaerobic Digestion of Organics with On-Site Electricity Generation

Dry AD for conversion of solid organic waste to biogas is under-studied relative to wet AD, whereas the life-cycle impacts of wet AD systems used to process the municipal organic waste, manure, and biosolids have been explored in numerous papers.[26–30] To populate our model, we were able to obtain operating data over multiple years from the ZWEDC dry AD facility in San Jose, CA. Detailed operations are laid out in Figure A7. The facility is designed to accept approximately 81,650 tonnes (90,000 short tons) of waste annually. Waste intake at ZWEDC is dominated by mixed organics including food and food-soiled paper products, often accompanied by a substantial quantity of inorganic contamination that must be separated and landfilled. Our model relies on delivery logs that include the origin of each truckload of waste, some of which is delivered from waste sorting/processing facilities while other loads are hauled directly from commercial sources including grocery stores and office parks. Assumptions for the origins and driving distances of inbound waste, based on these logs, are described in the SI.

At the ZWEDC facility, sorted organic waste is dewatered using an extruder and loaded into one of 16 digester bays for a typical residence time of 21 days. Produced biogas is first sent to storage bladders located on the facility roof, which provide storage for a few hours' worth of biogas production. In overpressure events, raw biogas can be vented from these bladders. Stored biogas is then treated to reduce H_2S concentrations using an iron sponge and fed to an on-site combined heat and power (CHP) facility comprised of two 800 kW generators, for a combined nameplate capacity of 1.6 MW. Approximately 30% of the biogas is flared due to gas storage limitations as well as the nature of batch digestion, which produces low-methane content (referred to as lean) gas at the start and end of each cycle that cannot be sent to CHP units (see Figure A7). Daily electricity consumption at ZWEDC averages 3,700 kWh/day (translating to an average load of 156 kW), including operation of the extruder, lighting, and fans. We assume net electricity exports offset generation from natural gas combined cycle (NGCC) power plants which often satisfy the marginal demand on California's grid.[31] The solid digestate generated at ZWEDC (4,040 tonnes per month on average, as shown in Figure A4) is aerated in four in-vessel composting tunnels on-site for 4-5 days before being sent to the Z-Best composting facility (72 km from ZWEDC).

After being trucked down to the Z-Best facility, solid digestate from ZWEDC is placed into commercial composting bags that are approximately 100 m × 6 m × 3 m when filled. Each encased windrow is filled with approximately 635 tonnes of material and undergoes a 14-week composting cycle, during which piles are force aerated but not turned. The finished compost is ultimately sold for agricultural and landscaping applications. Emission rates of CO_2, CH_4, N_2O, and NH_3 were determined from in-situ measurements at the Z-Best facility. As described in Kirchstetter et al.[32], concentrations of emitted gas were measured from nine windrows that captured different stages in the 14-week composting cycle. Bag samples were collected at ~35 locations across each windrow pile surface and later analyzed in the laboratory with three cavity ring-down spectrometers (Los Gatos Research, models 915-0011, N2OCM-919, and 915-0039; San Jose, CA). While bag samples were drawn, the aeration flow into each windrow was continuously measured using pairs of integrating pitot tubes (Dwyer Instruments, series PAFS-1005; Michigan City, IN). Emission rates with units of pollutant mass emitted per mass of digestate composted over the 14-week cycle were determined from the windrow-average emitted concentrations, average aeration flow, average mass of digestate per windrow, and average mass of digestate trucked from ZWEDC to Z-Best.

2.2.4 Solid Digestate Landfilling and Land Application

An alternative to the current ZWEDC operations, as described above, is a system in which all on-site operations are identical, but solid digestate is not sent to a composting facility. The first option is to landfill the digestate. At landfills, digestate can be handled as traditional waste or possibly used as alternative daily cover (ADC) to control insects, rodents, odors, and fire. In both cases the same material is being placed in the landfill (and ultimately covered as more waste is placed in the landfill), hence we do not expect that the use of digestate as ADC would result in substantial differences in the GHG footprint or other emissions relative to traditional landfilling. For this case, we modeled outbound trucking of raw digestate to the Newby Island Landfill nearby, which captures and flares its landfill gas. Emissions associated with the landfilling of digestate, or using digestate as ADC, are highly uncertain and empirical data in the literature is inadequate although it is intuitive that the fugitive methane emissions will be reduced for waste that has undergone AD. Thus, we scale the emission factors for landfilled digestate based on the volatile solids content reduction that occurs during AD. For food waste, AD reduces volatile solids content by about 80%.[33] Another alternative fate for raw digestate is direct land application. In this case, we assume the raw digestate can offset the use of inorganic fertilizers like urea (as with compost), but achieves negligible net long-term carbon sequestration.[34–36] We conservatively use the same nitrogen content of 1.7% for dried digestate as food waste-derived compost because the range of digestate nitrogen contents reported in the literature is comparable to that of compost. Although uncertain, the urea offsets are a relatively small contributor to the overall results, as discussed in the Results section. Another factor incorporated in our analysis are fugitive CH_4 and N_2O emissions from land application of biosolids, estimated at 65 g CO_{2e} per tonne of dry digestate.[37,38] Because of nutrient runoff concerns, land application of digestate only occurs for half of the year, with digestate being sent to landfills during the winter rainy season.[39,40]

2.2.5 Dry Anaerobic Digestion of Organics and Renewable Natural Gas Use for On-Site Truck Fleet

Understanding the tradeoffs between on-site combustion versus RNG applications is important for owners and operators of anaerobic digestion facilities, particularly when building new facilities or expanding existing ones. Without additional cleaning (removal of H_2S and water) and upgrading (where CO_2 is removed to increase the heating value), raw biogas cannot be compressed for use in pipelines or vehicles. This means raw biogas must either be flared or combusted for on-site heat and electricity generation, as is the case at ZWEDC. In the RNG for trucks scenario, we explore a hypothetical alternative scenario in which ZWEDC utilizes its biogas to fuel a retrofitted fleet of trucks rather than combusting it for electricity generation. Conversion of biogas to RNG is energy-intensive and reported mass/energy balances vary across the literature. Removing moisture, particles, contaminants and other gases (such as CO_2, O_2, N_2, H_2S and VOCs) increases the biogas methane content to 90% or more, depending on the upgrading technology. Commonly used biogas upgrading technologies include water scrubbing, pressure swing adsorption, and membrane separation. Some studies estimate membrane separation energy requirements around 0.3 kWh/m^3 [41,42] but the energy demand estimates can be as high as 0.5 kWh/m^3.[43] Pressure swing adsorption and water scrubbing require around 0.2 kWh/m^3 and 0.27 kWh/m^3, respectively.[43] We use an approximate value of 0.32 kWh/m^3 with a 0.6% loss factor and methane content of upgraded biogas of 96%. Because the biogas is being compressed and thus longer-term storage (beyond a few hours' worth of production) is more feasible, we conservatively approximate that venting events can be cut by 50% relative to the base case and flaring is also reduced by 50%. Flaring is not reduced by more than half because some rich gas will still be required as supplemental fuel when lean gas is flared. We assume produced RNG displaces diesel use in trucks that would be fueled on-site (Figure 2.1).

2.2.6 Dry Anaerobic Digestion of Organics and Renewable Natural Gas Pipeline Injection

Upgraded biogas with methane content more than 96% can also be used as renewable pipeline-injected natural gas. The upgrading process and associated energy demand is identical to the case described above for on-site RNG use in trucks. However, the offset credit is different because we assume the RNG displaces fossil natural gas (as opposed to offsetting diesel in the on-site truck fleet scenario) in unspecified end-uses and that the facility transports biogas via an interconnecting pipeline to an existing commercial pipeline located one mile away. In other words, end-use emissions are assumed to remain unchanged relative to a base case in which fossil natural gas is used. Emissions associated with the construction of the one-mile pipeline interconnection are assumed negligible when amortized over its lifetime, and thus are excluded.

2.2.7 Life-Cycle Emissions Inventory

The life-cycle inventory includes the following emissions: CO_2, CH_4, N_2O, NO_x, NH_3, NMVOC, SO_2, CO, and $PM_{2.5}$. These are all evaluated across a common functional unit of one wet tonne of organic waste processed (Figure 2.1). To construct a life-cycle inventory for each scenario, we collected direct mass and energy flow data, using as much measured and facility-logged data as possible from the ZWEDC facility's four years of operation. This is particularly important given the lack of data on dry AD and solid digestate composting in the existing literature, as well as the gap between best practices in an idealized scenario and what is typical at organic waste management facilities that handle highly contaminated waste streams. Through a collaboration with the ZWEDC facility owners and operators, we accessed inbound and outbound logs, including organics by type, residuals (trash for landfilling), and solid digestate. The facility also provided total biogas production, biogas flared, and electricity production; venting frequency and duration at the storage bladders were measured by the co-authors on-site.[32] As described in Kirchstetter et al.[32], venting volume of biogas released to the atmosphere was determined with measurements of CO_2 (LI-COR, model LI-820; Lincoln, NE), gas temperature (Onset, HOBO model UX120 with Type T thermocouple; Bourne, MA), and gas velocity (The Energy Conservatory, model DG-700; Minneapolis, MN) within the pressure relief valve chimney for one of the two ZWEDC biogas storage bladders. Emission factors for digestate composting, biogas flaring, and biogas venting are all based on measured values at Z-Best and ZWEDC. Values that could not be or were not directly measured are assembled from literature sources, including peer-reviewed articles, GREET, and the Ecoinvent database (Table A1).

Direct mass and energy flows from the waste sources to final product(s) were incorporated into a physical units-based input-output life-cycle inventory model, Agile-Cradle-to-Grave (Agile-C2G), which has been documented extensively in previous literature.[44–47] This model was used to calculate indirect emissions associated with electricity generation, fertilizer production, diesel fuel production, and other minor material/energy inputs. California-based sources were considered wherever appropriate. To account for net CO_2, CH_4, and N_2O emissions after land application of composted organics, raw digestate, and composted digestate, we use GHG emission and sequestration factors documented in Breunig et al.[44] Details are also provided in Table A1. Other non-GHG air pollutant emission factors during the post-land-application phase are assumed to be negligible relative to the emissions during waste management, AD, and composting.

To capture parameter uncertainty, we established probability distributions for key parameters based on previous literature and used these in a Monte Carlo analysis (see Table A3). The model was run for 10,000 trials drawing from these distributions to develop the box and whisker plots shown in the results. Although the distributions were developed based on wide-ranging literature values from both in and outside California, the expected values (denoted by black dots in Figure 2.2 and Figure A8) indicate values specific to the ZWEDC case study. At times, the specific study result may lie beyond the upper or lower quartile because the measured values at ZWEDC or Z-Best are not in the middle of the ranges published in previous literature. This text will focus its discussion on the expected-value results for ZWEDC/Z-Best.

2.2.8 Social Cost and Public Health Damage Cost

Although monetized externality estimates are an imperfect measure of environmental impacts, converting GHG emissions and air pollutant impacts into social costs is useful. First, these estimates provide a means of comparing different inventory metrics based on their relative importance to one another. Second, monetizing human health damages allows for differentiation between emissions that occur within or outside densely populated areas and thus the expected impact on the population. Third, the dollar values provide some guidance as to what governments may wish to pay in order to avoid undesirable externalities. To account for the human health damages associated with air pollutant emissions, we compare two common integrated assessment models: Air Pollution Emission Experiments and Policy analysis (APEEP, specifically version 3, hereafter referred to as AP3) and Estimating Air Pollution Social Impact Using Regression (EASIUR).[48–50] Multipliers to convert emissions to social costs are provided in Table A4 of the SI. In these cases, we include only pollutants that occur locally, either at the ZWEDC facility, Z-Best compost facility, or nearby transportation routes, assuming ground-level emissions values. The damage factor most difficult to refine on a scientific basis is the social cost per tonne of CO_{2e} emitted, and the cost of carbon used in regulations can be highly politicized. We use a relatively conservative social cost of carbon of $42 per tonne CO_{2e}, which was established by the Interagency Working Group on the Social Cost of Greenhouse Gases for use in regulatory analyses.[51]

2.3 Results

The results of our analysis are presented in three sections. First, we show life-cycle GHG emissions results, followed by results for all air pollutants (NO_x, NH_3, NMVOC, SO_2, CO and $PM_{2.5}$). Last, we convert these life-cycle inventory results into monetized damages using the multipliers discussed in the Methods section and provided in Table A4.

2.3.1 Life-Cycle Greenhouse Gas Inventory

The life-cycle GHG results (see Figure 2.2), which include non-biogenic CO_2 as well as all CH_4 and N_2O emissions, normalized using 100-year global warming potentials (298 and 25, respectively), indicate that landfilling organic waste is the most GHG-intensive option on a per-tonne basis, with a GHG footprint of almost 400 kg CO_{2e} per tonne of organic waste. Any option for diverting organic waste, particularly higher-moisture material such as food waste that releases substantial fugitive methane, provides GHG benefits. The footprint will be roughly doubled if organics are sent to a landfill without a functioning gas capture system in place. The next most GHG-intensive options are the dry AD configurations in which some or all of the solid digestate must be landfilled. If all digestate is landfilled, the GHG footprint is 40 kg CO_{2e} per tonne of organic waste. As mentioned in the Methods section, solid digestate can only be land applied for a portion of the year in California because of water quality/runoff concerns during the rainy season, so the land application scenario still results in large landfill emissions. Thus, the land application scenario reduces, but does not eliminate, landfill methane emissions, resulting in a net GHG footprint of 27 kg CO_{2e} per tonne of organic waste. Each of these scenarios is dominated by landfill methane emissions. Some facilities may choose to avoid this seasonal limitation by trucking digestate long distances to locations that do not regulate digestate land application in the winter. In that case, the avoided landfill GHG emissions are likely to be larger than the increased trucking emissions. However, depending on the local climate where digestate is land-applied, there may be other concerns such as increased nitrogen runoff and N_2O emissions.[44]

The GHG footprints of composting raw organics and the three dry AD scenarios that do not require any landfilling of solid digestate all have much lower GHG footprints than scenarios that involve landfilling. The scenario that combines dry AD, electricity generation, and composting digestate (ZWEDC current operations) results in a net GHG footprint of 9 kg CO_{2e} per tonne of organic waste. The composting scenario and the two AD with RNG scenarios all resulted in net negative GHG emissions. These results are reflective of the specific conditions defined in the model and cannot be directly applied to future conditions. Offsets and negative emissions are dependent on the avoidance of current emission-intensive processes (e.g. carbon-intensive electricity generation, fertilizer use). The factors driving the differences between these three net negative scenarios, such as the net soil carbon impacts of compost application, are nuanced and uncertain. This finding is consistent with previous literature, as shown in the meta-analysis by Morris et al.[6] Composting results in the lowest GHG footprint, totaling -41 kg CO_{2e} per tonne of organic waste. A large GHG sequestration credit and a more limited fertilizer offset credit are both based on expected benefits from land application of the compost. If biogas is upgraded to RNG and used to offset diesel fuel use in a fleet of new or retrofitted trucks, the net GHG footprint is -36 kg CO_{2e} per tonne of organic waste (in this scenario, digestate is sent to be composted). This demonstrates that offsetting diesel can avoid a larger quantity of fossil CO_{2e} emissions than offsetting NGCC electricity, as is assumed in the biogas-to-electricity scenarios. Upgrading biogas to RNG and injecting it into the pipeline for use in place of fossil natural gas results in reduced GHG mitigation (-2 kg CO_{2e} per tonne of organic waste) relative to the scenario in which RNG offsets diesel use.

A point of confusion may be the fact that cleaning up biogas and injecting it into the pipeline to be combusted in place of fossil natural gas (at a power plant or otherwise) is preferable to combusting raw biogas on-site for electricity and heat. The process of cleaning and upgrading biogas does, after all, involve energy inputs and methane losses. ZWEDC operates two 800 kW engines at approximately 40% efficiency, not accounting for rich biogas that must be flared or vented when units are down for maintenance or are otherwise not able to utilize all available biogas. Aside from heat losses during electricity generation, 30% of rich biogas is flared at ZWEDC and a negligible fraction is vented. By comparison, NGCC plants are able to use waste heat in a secondary steam cycle to generate additional electricity, resulting in an average NGCC plant efficiency across California of 47%.[52] We also assume that, once the facility invests in a gas cleanup/upgrading system and pressurized storage, flaring and venting will be cut in half, resulting in only a 15% loss. Thus, even after accounting for beneficial waste heat recovery for use in the digesters, the choice to clean and upgrade the biogas for use as a drop-in replacement for natural gas results in greater GHG reductions. If instead power exports to the electricity grid displace the average California grid mix, given its high share of renewable energy, the disparity is likely to become more pronounced. Furthermore, if pipeline-injected RNG is used for vehicles in place of diesel, the GHG advantage will grow. In short, the benefits of biogas upgrading to

RNG are likely to be greater, no matter the end use of the RNG, relative to using biogas for on-site, small-scale electricity generation and export to the electricity grid in California or any other location in which the grid is relatively clean.

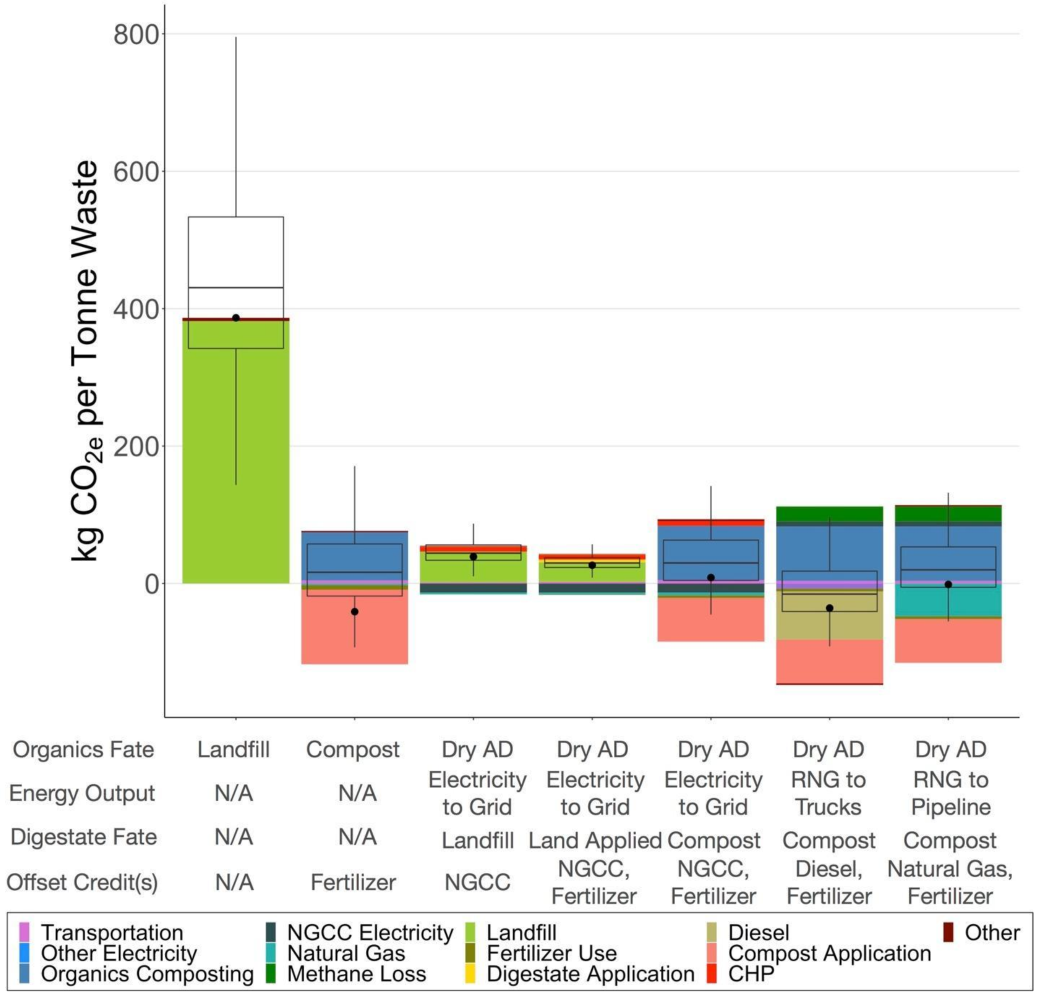

Figure 2.2 Life-cycle greenhouse gas emissions of different organic waste management options
Contributors totaling less than 1% are categorized as “Other”. “Other Electricity” category refers to avoided electricity consumption from reduced urea fertilizer consumption

2.3.2 Life-Cycle Air Pollutant Emissions Inventory

The life-cycle air pollutant emissions vary dramatically across the different scenarios, as shown in Figure A8. $PM_{2.5}$ is recognized as the air pollutant primarily responsible for human health damages.[53] $PM_{2.5}$ can be emitted directly (primary $PM_{2.5}$) or formed in the atmosphere as the product of chemical reactions of precursors including NO_X, SO_2, VOCs, and NH_3 (referred to as secondary $PM_{2.5}$). Landfills are estimated to release the greatest primary $PM_{2.5}$ per tonne of organic waste across all options, and these emissions are dominated by the on-site flaring of landfill gas. Flares generally do not have emissions control technology and, given varying methane concentrations and imperfect mixing, they tend to emit more PM than biogas-fired power generators. The two dry AD cases in which some or all solid digestate must be landfilled are the next highest-emitting options in terms of primary $PM_{2.5}$. In contrast, the dry AD case in which RNG is used in place of diesel fuel for trucks is a net-negative because of the avoided $PM_{2.5}$ associated with operating diesel trucks (and the relatively negligible $PM_{2.5}$ emissions from RNG trucks). We do not account for potential non-combustion sources of $PM_{2.5}$ because they are expected to emit particles larger than 2.5 μm in diameter, such as dust.

Nitrogen oxides (NO_x), accounted for as mass of NO_2, and SO_2 are respiratory irritants for humans and precursors to secondary PM and ozone. Both pollutants are the product of combustion. Flares at landfills and AD facilities are the dominant source of SO_2. Flaring also emits NO_x, and because flares do not have NO_x emissions controls, the emissions per unit of fuel input are higher than for biogas and natural gas combustion in power generators, as reflected in Figure A8. Composting can result in net-negative NO_x and SO_2 emissions because direct emissions are negligible and applying finished compost to soil can reduce the need for nitrogenous fertilizer, which is energy- and emissions-intensive to produce. In the case where biogas is cleaned and upgraded to RNG, the distinction between RNG used to offset diesel in a fleet of trucks versus use in pipelines to offset fossil natural gas is critical. NO_x and SO_2 emissions are net negative if RNG is used in vehicles because of the avoided tailpipe emissions from diesel combustion. If RNG is used to offset fossil natural gas (through pipeline injection), net emissions are positive but still lower than most other scenarios.

NMVOC and NH_3 emissions are both challenging to quantify because of limited data. However, the life-cycle emissions for both are likely dominated by emissions from composting operations. NH_3 is produced during microbial decomposition, which occurs in both the digesters and during the composting process, as a way to discard excess nitrogen not required as a nutrient for the microbes. Thus, NH_3 is present in rich and lean biogas at the facility as well, but is largely removed by the acid scrubber or oxidized to NO_x through combustion. In the case of NMVOCs, small negative values are owed to the fact that offsetting fossil natural gas use reduces fugitive emissions (a small fraction of which are non-methane compounds such as ethane and propane). CO emissions are also dominated by composting operations, although incomplete combustion during flaring and biogas-fired electricity generation also contribute to the total emissions.
To compare the social cost of primary and secondary $PM_{2.5}$ exposure and GHG emissions across waste management scenarios, we used two different integrated assessment models, APEEP and EASIUR, in combination with a \$42 per tonne CO_{2e} social cost for GHG emissions (see Figure 2.3). The results indicate that the social cost of landfilling wet organic waste is approximately \$25-40 per tonne (this does not include odor externalities or non-emissions related costs such as impacts on local property values). Because GHG-related damages make up the largest fraction of the overall monetized damages for landfilling, this value will change depending on the assumed social cost of carbon. For comparison, the median landfill tipping fee in California, as of 2015, was \$45 per tonne while countries that landfill very little of their waste, including Germany and Sweden, have tipping fees around \$200 per tonne,[54] suggesting that incorporating even a fraction of the estimated social cost into tipping fees could greatly encourage diversion from landfills.

Both AP3 and EASIUR indicate that NH_3 emissions are the dominant contributor to social costs in every case where some or all organic material is composted. This is because NH_3 emissions per tonne of organic waste processed are at least two orders of magnitude greater than any other non-GHG pollutant in each scenario that includes composting (see Figure A8). NH_3 plays an important role in the formation of secondary $PM_{2.5}$ by reacting with nitric acid (HNO_3) and sulfuric acid (H_2SO_4), resulting from NO_x and SO_x emissions, to form ammonium nitrate (NH_4NO_3) and ammonium sulfate ($(NH_4)_2SO_4$) aerosols. However, AP3 and EASIUR disagree in some cases by more than a factor of three, with AP3 estimating greater NH_3-related damages than EASIUR. An additional caveat is that there is very little known about NH_3 emissions from landfills before cells are capped off and gas capture/flaring systems are in place, particularly for specific waste types, such as nitrogen-rich food wastes. Similarly, very little is known about NH_3 emissions from land application of raw or composted digestate. Section A6 of Appendix A provides further details on the challenges of modeling secondary PM formation from NH_3 emissions, particularly in California. Despite these challenges, both EASIUR and AP3, when combined with the social cost of GHG emissions, indicate that composting has a greater social cost than landfilling and that the four scenarios including composting had the highest costs. Both assessments also predict that landfilling and land applying digestate are the least damaging options among all scenarios considered in this study. However, the models yield slightly more contrasting estimates of the relative health impacts of the ZWEDC and two RNG scenarios versus landfilling: AP3 indicates that landfilling is the preferred option cutting emissions nearly in half relative to the other scenarios while EASIUR indicates that landfilling is the least damaging of these scenarios but only 10% lower in cost than the RNG Onsite scenario.

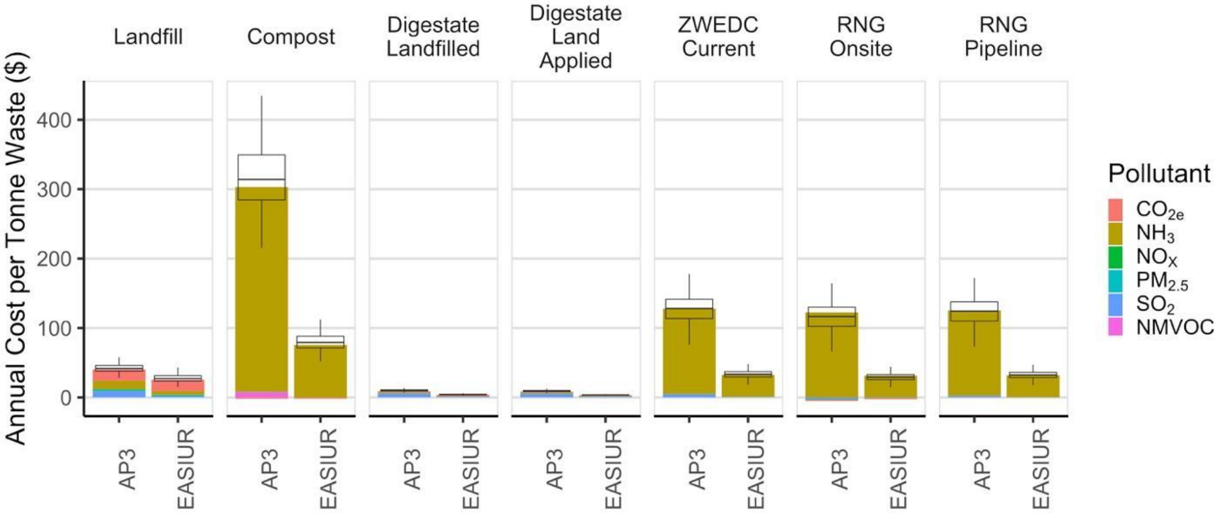

Figure 2.3 Life-cycle social costs of different organic waste management options

These results are presented for two different reduced-form public health cost tools (EASIUR and AP3), using shortened titles for scenarios outlined previously.

2.4 Discussion

This study reveals the complexity of estimating environmental tradeoffs in organic waste management systems, and the difficulty of making broadly applicable recommendations for how organic waste should be handled. Previous literature has indicated consistently that landfilling is the least attractive option, even in more tightly regulated states like California that require efficient gas-capture systems.[6] Our GHG emissions results reinforce this conclusion. Fugitive methane emissions are the key driver in the GHG footprint of organic waste, and any scenario in which organics are landfilled will result in higher GHG emissions. The offset credits for electricity, RNG, and finished compost are also important for determining both net GHG emissions and other air pollutant emissions. If, for example, compost application does not cause a net reduction in nitrogenous fertilizer use, the net negative values for NMVOCs, NO_x, SO_2, and GHGs will be eliminated. The question of whether RNG offsets diesel or fossil natural gas will have a substantial impact on net NO_x emissions. However, on a social cost basis, none of these changes to the assumptions would alter the basic conclusions.

Our results suggest that NH_3 emissions resulting from composting nitrogen-rich waste may outweigh any other air pollutant or GHG-related social costs, yet NH_3 emissions are not well documented for organic waste management systems, even relative to other air pollutants such as VOCs. At the very least, these results warrant further study to determine how NH_3 emissions and human health damages will vary based on waste composition, composting practices, and local meteorology. The results also call into question the wisdom of making waste management decisions based solely on GHG emissions, given the potential for unintended human health consequences. If NH_3 emissions are confirmed to be a driving factor in social costs of organic waste management options and are indeed greater on average at composting sites relative to landfills, the larger question is how and to what degree those emissions can be reduced. Because large composting windrows are not well-mixed controlled environments, some pockets of excess nitrogen are inevitable, particularly when nitrogen-rich food waste or digestate serves as the input. However, maximizing microbial activity and thus increasing demand for nitrogen through improved monitoring and control of pH, temperature, and aeration level during the composting process can reduce NH_3 emissions.[55] Another alternative for minimizing total social cost is to locate large composting operations in sparsely populated areas, although this may result in environmental justice/inequity issues if rural populations are socioeconomically disadvantaged relative to urban populations. Further empirical research, exploring a range of material types and composting practices, will be essential to better understanding which options for diverting organic waste from landfills provides the greatest public good.

References

(1) Lara, R. *Short-Lived Climate Pollutants (SLCP): Organic Waste Methane Emissions Reductions*; Sacramento, CA, 2017.

(2) Duren, R. M.; Thorpe, A. K.; Foster, K. T.; Rafiq, T.; Hopkins, F. M.; Yadav, V.; Bue, B. D.; Thompson, D. R.; Conley, S.; Colombi, N. K.; et al. California's methane super-emitters. *Nature* **2019**, *575*, 180–184.

(3) Satchwell, A. J.; Scown, C. D.; Smith, S. J.; Amirebrahimi, J.; Jin, L.; Kirchstetter, T. W.; Brown, N. J.; Preble, C. V. Accelerating the deployment of anaerobic digestion to meet zero waste goals. *Environ. Sci. Technol.* **2018**, *52*, 13663–13669.

(4) Breunig, H. M.; Jin, L.; Robinson, A.; Scown, C. D. Bioenergy Potential from Food Waste in California. *Environ. Sci. Technol.* **2017**, *51*, 1120–1128.

(5) California Air Resources Board. Pathway Certified Carbon Intensities https://ww3.arb.ca.gov/fuels/lcfs/fuelpathways/pathwaytable.htm (accessed Oct 16, 2019).

(6) Morris, J.; Scott Matthews, H.; Morawski, C. Review and meta-analysis of 82 studies on end-of-life management methods for source separated organics. *Waste Manag.* **2013**, *33*, 545–551.
(7) Milutinovic, B.; Stefanovic, G.; Ekic, P. S.; Mijailovic, I.; Tomic, M. Environmental assessment of waste management scenarios with energy recovery using life cycle assessment and multi-criteria analysis. *Energy* **2017**, *137*, 917–926.
(8) Escamilla-Alvarado, C.; Poggi-Varaldo, H. M.; Ponce-Noyola, M. T. Bioenergy and bioproducts from municipal organic waste as alternative to landfilling: a comparative life cycle assessment with prospective application to Mexico. *Environ. Sci. Pollut. Res. Int.* **2017**, *24*, 25602–25617.
(9) Ward, A. J.; Hobbs, P. J.; Holliman, P. J.; Jones, D. L. Optimisation of the anaerobic digestion of agricultural resources. *Bioresour. Technol.* **2008**, *99*, 7928–7940.
(10) De Baere, L. Anaerobic digestion of solid waste: state-of-the-art. *Water science and technology* **2000**, *41*, 283–290.
(11) Chatterjee, B.; Mazumder, D. Anaerobic digestion for the stabilization of the organic fraction of municipal solid waste: A review. *Environ. Rev.* **2016**, *24*, 426–459.
(12) Li, Y.; Park, S. Y.; Zhu, J. Solid-state anaerobic digestion for methane production from organic waste. *Renewable and Sustainable Energy Reviews* **2011**, *15*, 821–826.
(13) Clarke, W. P. The uptake of anaerobic digestion for the organic fraction of municipal solid waste - Push versus pull factors. *Bioresour. Technol.* **2018**, *249*, 1040–1043.
(14) U.S. Environmental Protection Agency. *Advancing Sustainable Materials Management: 2015 Fact Sheet*; EPA530-F-18-004; U.S. Environmental Protection Agency, 2018.
(15) USEPA. Waste Reduction Model (WARM), 2018.
(16) Behera, S. K.; Park, J. M.; Kim, K. H.; Park, H.-S. Methane production from food waste leachate in laboratory-scale simulated landfill. *Waste Manag.* **2010**, *30*, 1502–1508.
(17) Amlinger, F.; Peyr, S.; Cuhls, C. Green house gas emissions from composting and mechanical biological treatment. *Waste Manag. Res.* **2008**, *26*, 47–60.
(18) Boldrin, A.; Andersen, J. K.; Møller, J.; Christensen, T. H.; Favoino, E. Composting and compost utilization: accounting of greenhouse gases and global warming contributions. *Waste Manag. Res.* **2009**, *27*, 800–812.
(19) Saer, A.; Lansing, S.; Davitt, N. H.; Graves, R. E. Life cycle assessment of a food waste composting system: environmental impact hotspots. *J. Clean. Prod.* **2013**, *52*, 234–244.
(20) CEPA. *Method for estimating greenhouse gas emission reductions from diversion of organic waste from landfills to compost facilities*; California Air Resources Board, 2017.
(21) Favoino, E.; Hogg, D. The potential role of compost in reducing greenhouse gases. *Waste Manag. Res.* **2008**, *26*, 61–69.
(22) Martínez-Blanco, J.; Lazcano, C.; Christensen, T. H.; Muñoz, P.; Rieradevall, J.; Møller, J.; Antón, A.; Boldrin, A. Compost benefits for agriculture evaluated by life cycle assessment. A review. *Agron. Sustain. Dev.* **2013**, *33*, 721–732.
(23) Martínez-Blanco, J.; Muñoz, P.; Antón, A.; Rieradevall, J. Life cycle assessment of the use of compost from municipal organic waste for fertilization of tomato crops. *Resour. Conserv. Recycl.* **2009**, *53*, 340–351.
(24) Sullivan, D. M.; Cogger, C. G.; Bary, A. I. *Fertilizing with Biosolids*; Oregon State University Extension, 2015.
(25) Torres-Climent, A.; Martin-Mata, J.; Marhuenda-Egea, F.; Moral, R.; Barber, X.; Perez-Murcia, M. D.; Paredes, C. Composting of the Solid Phase of Digestate from

Biogas Production: Optimization of the Moisture, C/N Ratio, and pH Conditions. *Commun. Soil Sci. Plant Anal.* **2015**, *46*, 197–207.

(26) Scipioni, A.; Mazzi, A.; Niero, M.; Boatto, T. LCA to choose among alternative design solutions: the case study of a new Italian incineration line. *Waste Manag.* **2009**, *29*, 2462–2474.

(27) Möller, K.; Müller, T. Effects of anaerobic digestion on digestate nutrient availability and crop growth: A review. *Eng. Life Sci.* **2012**, *12*, 242–257.

(28) Turconi, R.; Butera, S.; Boldrin, A.; Grosso, M.; Rigamonti, L.; Astrup, T. Life cycle assessment of waste incineration in Denmark and Italy using two LCA models. *Waste Manag. Res.* **2011**, *29*, 78–90.

(29) Tonini, D.; Martinez-Sanchez, V.; Astrup, T. F. Material resources, energy, and nutrient recovery from waste: are waste refineries the solution for the future? *Environ. Sci. Technol.* **2013**, *47*, 8962–8969.

(30) Boesch, M. E.; Vadenbo, C.; Saner, D.; Huter, C.; Hellweg, S. An LCA model for waste incineration enhanced with new technologies for metal recovery and application to the case of Switzerland. *Waste Manag.* **2014**, *34*, 378–389.

(31) Denholm, P. L.; Margolis, R. M. *The Potential for Energy Storage to Provide Peaking Capacity in California under Increased Penetration of Solar Photovoltaics: Report Summary*; National Renewable Energy Laboratory (NREL): Golden, CO (United States), 2018.

(32) Kirchstetter, T. W.; Scown, C. D.; Satchwell, A.; Jin, L.; Smith, S. J.; Preble, C. V.; Amirebrahimi, J.; Sohn, M. D.; Brown, N. J. *Enabling Anaerobic Digestion Deployment for Municipal Solid Waste-to-Energy*; California Energy Commission, 2020.

(33) Chen, X.; Romano, R.; Zhang, R. Anaerobic digestion of food wastes for biogas production. *International Journal of Agricultural and Biological Engineering* **2010**, *3*.

(34) Kibler, K. M.; Reinhart, D.; Hawkins, C.; Motlagh, A. M.; Wright, J. Food waste and the food-energy-water nexus: A review of food waste management alternatives. *Waste Manag.* **2018**, *74*, 52–62.

(35) Levis, J. W.; Barlaz, M. A. What is the most environmentally beneficial way to treat commercial food waste? *Environ. Sci. Technol.* **2011**, *45*, 7438–7444.

(36) Mu, D.; Horowitz, N.; Casey, M.; Jones, K. Environmental and economic analysis of an in-vessel food waste composting system at Kean University in the U.S. *Waste Manag.* **2017**, *59*, 476–486.

(37) Holly, M. A.; Larson, R. A.; Powell, J. M.; Ruark, M. D.; Aguirre-Villegas, H. Greenhouse gas and ammonia emissions from digested and separated dairy manure during storage and after land application. *Agriculture, Ecosystems & Environment* **2017**, *239*, 410–419.

(38) ECN.TNO. Phyllis2 - Database for (treated) biomass, algae, feedstocks for biogas production and biochar https://phyllis.nl/ (accessed Nov 26, 2019).

(39) California Association of Sanitation, A. *An Evaluation of the Sustainability of Biosolids Use as Landfill Burial or Beneficial Cover Material*; CALIFORNIA ASSOCIATION of SANITATION AGENCIES, 2017; p. 4.

(40) State Water Resources Control Board. *General Waste Discharge Requirements for the Discharge of Biosolids to Land for Use as a Soil Amendment in Agricultural, Silvicultural, Horticultural, and Land Reclamation Activities*; STATE WATER RESOURCES CONTROL BOARD, 2018.

(41) Makaruk, A.; Miltner, M.; Harasek, M. Membrane biogas upgrading processes for the production of natural gas substitute. *Separation and Purification Technology* **2010**, *74*, 83–92.
(42) Pöschl, M.; Ward, S.; Owende, P. Evaluation of energy efficiency of various biogas production and utilization pathways. *Appl. Energy* **2010**, *87*, 3305–3321.
(43) Pertl, A.; Mostbauer, P.; Obersteiner, G. Climate balance of biogas upgrading systems. *Waste Manag.* **2010**, *30*, 92–99.
(44) Breunig, H. M.; Amirebrahimi, J.; Smith, S.; Scown, C. D. Role of Digestate and Biochar in Carbon-Negative Bioenergy. *Environ. Sci. Technol.* **2019**.
(45) Neupane, B.; Konda, N. V. S. N. M.; Singh, S.; Simmons, B. A.; Scown, C. D. Life-Cycle Greenhouse Gas and Water Intensity of Cellulosic Biofuel Production Using Cholinium Lysinate Ionic Liquid Pretreatment. *ACS Sustain. Chem. Eng.* **2017**, *5*, 10176–10185.
(46) Baral, N.; Kavvada, O.; Perez, D. M.; Mukhopadhyay, A.; Lee, T. S.; Simmons, B.; Scown, C. D. Greenhouse Gas Footprint, Water-Intensity, and, Production Cost of Bio-Based Isopentenol as a Renewable Transportation Fuel. *ACS Sustain Chem Eng* **2019**.
(47) Scown, C. D.; Gokhale, A. A.; Willems, P. A.; Horvath, A.; McKone, T. E. Role of lignin in reducing life-cycle carbon emissions, water use, and cost for United States cellulosic biofuels. *Environ. Sci. Technol.* **2014**, *48*, 8446–8455.
(48) Muller, N. Z.; Mendelsohn, R. Measuring the damages of air pollution in the United States. *J. Environ. Econ. Manage.* **2007**, *54*, 1–14.
(49) Heo, J.; Adams, P. J.; Gao, H. O. Public health costs of primary PM2.5 and inorganic PM2.5 precursor emissions in the united states. *Environ. Sci. Technol.* **2016**, *50*, 6061–6070.
(50) Heo, J.; Adams, P. J.; Gao, H. O. Reduced-form modeling of public health impacts of inorganic PM2.5 and precursor emissions. *Atmos. Environ.* **2016**, *137*, 80–89.
(51) Interagency Working Group on the Social Cost of Greenhouse Gases. *Technical Update of the Social Cost of Carbon for Regulatory Impact Analysis*; U.S. White House: Washington, DC, 2016.
(52) Nyberg, M. *Thermal Efficiency of Natural Gas-Fired Generation in California: 2016 Update*; CEC-200-2017-003; California Energy Commission: Sacramento, CA, 2017.
(53) Cohon, J. L.; Cropper, M. L.; Cullen, M. R.; English, M. R.; Field, C. B.; Greenbaum, D. S.; Hammitt, J. K.; Henderson, R. F.; Kling, C. L.; Krupnick, A. J.; et al. *Hidden costs of energy: unpriced consequences of energy production and use*; National Academies Press: Washington, D.C., 2010.
(54) CalRecycle. *Landfill Tipping Fees in California*; California Department of Resources Recycling and Recovery (CalRecycle), 2015.
(55) Jiang, T.; Schuchardt, F.; Li, G.; Guo, R.; Zhao, Y. Effect of C/N ratio, aeration rate and moisture content on ammonia and greenhouse gas emission during the composting. *Journal of Environmental Sciences* **2011**, *23*, 1754–1760.

Chapter 3

Greenhouse Gas and Air Pollutant Emissions from Composting

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3.1 Importance of Composting Emissions

Composting is an essential part of any strategy to divert organic waste and reduce fugitive methane (CH_4) emissions from landfills.[1,2] In the United States (U.S.), 6-9% of total municipal solid waste is currently composted, although as much as 34% could be composted if all food and yard waste were diverted from landfills.[3,4] Composting can treat organic waste directly or treat solids remaining after organic waste has undergone anaerobic digestion (AD), ultimately reducing the total mass of waste through aerobic biochemical decomposition and yielding soil amendments for agricultural or landscaping applications. Most reported values for mass loss during composting on a dry basis fall in the range of 10–60%.[5–8] The motivations for composting are: (1) avoidance of fugitive CH_4 emissions associated with the anaerobic decomposition that occurs in solid waste landfills and manure storage lagoons, (2) the diversion of organic waste from landfills, and (3) generation of compost that is free of harmful pathogens and ready for use in agricultural applications or for erosion control.[9,10] Although avoiding CH_4 emissions from landfills is one of the motivations for composting organic waste, the composting process itself emits greenhouse gases (GHGs) and air pollutants, and these emissions are still not well understood.[11]

Gaseous emissions from the composting of organic waste have impacts on both climate change and air quality. The GHG emissions are directly relevant to policy. For example, life-cycle GHG emissions from bioenergy production routes—some of which incorporate composting of residual solids—must be thoroughly documented in the U.S., as they are tied to the Renewable Fuel Standard Renewable Identification Numbers (RINs) and California's Low Carbon Fuel Standard (LCFS) Carbon Intensity (CI) scores, both of which carry substantial monetary value.[12]

Non-GHG air pollutant emissions from composting facilities affect local and regional air quality and, as a result, human health in surrounding communities.[13] Emissions of ammonia (NH_3) and volatile organic compounds (VOCs) from composting are of particular concern because they are precursors of secondary fine particulate matter ($PM_{2.5}$), which is the primary driver of air pollution-related health impacts.[14,15] A detailed description of NH_3 emissions and $PM_{2.5}$ formation is provided in Appendix B. VOCs are also precursors to tropospheric ozone formation which impacts human health and sensitive vegetation and ecosystems.[14] Lastly, VOCs and NH_3 have low odor detection thresholds and can cause a public nuisance for surrounding communities. Odorous pollutants can impact permitting for new facilities, particularly in non-attainment areas in the U.S. where ambient air pollutant concentrations exceed the National Ambient Air Quality Standards. Our review covers NH_3 and VOC emissions but does not include other odorous compounds such as non-carbon containing volatile sulfur compounds (e.g. hydrogen sulfide). Because of the additional environmental and human health impacts from non-GHG emissions, it is essential to balance ambitious landfill diversion goals with local air quality and odor concerns associated with operating composting facilities.[16]

Despite the importance of GHG and air pollutant emissions from composting, available data can be difficult to interpret and use for policy implementation. The California Air Resources Board released a recommended methodology for estimating composting emissions in 2015, but the method was only applicable to mixtures with at least 85% green waste and a maximum of 15% food waste, biosolids, or manure.[17] The degree to which specific composting practices and incoming waste composition affect emissions per unit of composted material is not well understood. Furthermore, researchers incorporating composting emissions into life-cycle assessments (LCAs) are often not experts in different measurement techniques and the degree to which measurement methods affect the accuracy of empirical data. This knowledge gap makes prioritization of emissions mitigation strategies and scenario planning for zero-waste policies challenging.

The purpose of this review is to evaluate data available in the scientific literature on air emissions from composting operations, discuss the merits and tradeoffs of measurement strategies employed in past studies, and provide guidance for researchers and decision-makers who seek to integrate composting emission factors into policy and environmental impact studies. Regarding GHGs, we mainly focus on CH_4 and nitrous oxide (N_2O) emissions because they are the primary drivers of net climate forcing impacts from composting.[11,18] Although contemporary carbon emitted as CO_2 during composting is not thought to have a net climate impact, we also include limited data on CO_2 emissions results, reported separately from CH_4 and N_2O, in Appendix B (Figure B1). This review also includes data on NH_3 and VOCs because of their importance for air quality and air pollution-related human health impacts.[15] Empirical emission values collected from the literature were differentiated based on the type of material being composted, measurement methods used in the study, and the management strategies employed during the composting process, with the goal of developing more representative and material-specific recommendations for composting emission factor ranges.

Prior reviews have explored some dimensions of this topic but fall short of providing recommended ranges for emission factors that can be used in future LCAs and policy-making. For example, Amlinger et al. (2008) primarily focused on their own measured results for CH_4, N_2O, and NH_3, but included a review of prior results to inform the development of a helpful, mostly qualitative table summarizing the effects of different compost management strategies on emissions and the mechanisms behind those effects.[19] Brown et al. (2008) reviewed a broader set of literature values on GHG emissions associated with different alternatives for disposing of/treating organic waste, including landfilling and anaerobic digestion, in comparison to composting.[20] Lou and Nair (2009) compared GHG emissions from composting and landfilling organic waste, concluding that landfilling results in higher GHG emissions as compared to composting, a conclusion that reflects broad consensus in the research community.[21] Pardo et al. (2015) conducted a meta analysis of 50 studies to establish the relative impacts of different management strategies, such as forced aeration versus turning and the addition of bulking agents.[22] Pardo et al. (2015) considered the same raw feedstocks as those included in this review and focused on the relative impact of different operational practices and conditions, but did not establish emission factors per tonne of waste composted. Bong et al. (2017) and Sayara and Sánchez (2021) provide more qualitative reviews of composting GHG emissions and discuss GHG mitigation strategies.[23,24] Sayara and Sánchez (2021) summarize research regarding the impact of composting practices and feedstock characteristics on emissions, while Bong et al. (2017) focuses more specifically on the variability of scope definition and inventory analysis in published LCAs of composting. Neither review provides recommended emission factors.

Although this review focuses on gaseous emissions during the composting process itself, excluding truck transport and combustion of fuels to operate equipment, composting emission factors are more meaningful in a broader context, where each end-to-end process for managing organic waste can be compared. There are two main competing routes of relevance: (1) composting followed by land application of finished compost, and (2) landfilling untreated organic waste. The use phase for finished compost is essential to include in life-cycle emissions inventories; applying compost can reduce the need for synthetic fertilizers and, in some cases, increase the net primary productivity on degraded lands.[25] The comparison between composting emissions and landfilling organic waste is another important topic, and this has been explored more thoroughly in prior reviews, although gaps in empirical data remain.[21,26] In Appendix B, we provide an overview of the state of knowledge related to how compost application and landfilling organic waste affect net GHG emissions.

3.2 The Role of Composting in Organic Waste Management

Organic wastes that can be composted include the entire organic fraction of municipal solid waste (OFMSW, which includes a variety of organic waste types), food waste, yard waste, sewage sludge, manure, and digestates (residual solids remaining after AD). The most commonly composted material is source-separated yard waste. According to the U.S. Environmental Protection Agency (EPA), approximately 0.4% of food waste and 63% of yard waste are currently composted in the U.S. While technically compostable, paper waste is more commonly recycled unless it is soiled or otherwise unsuitable,[27] so we have excluded it from this review. Solid digestate can be directly applied to agricultural land as a fertilizer amendment, but there are typically seasonal limitations on this practice due to nutrient runoff concerns in some states, so AD facilities may send digestate to composting facilities during part or all of the year.[15,28–31]

The wastes processed at composting facilities vary in moisture content, carbon-to-nitrogen ratio (C:N), pH, volatile solids (VS) content, and other characteristics that lead to varying rates of aerobic decomposition and emissions to the atmosphere. VS refers to the part of compostable materials that is combusted at 550 ºC in the presence of air after 2 hours, and can be a proxy for the fraction of biodegradable material.[32] The composting process itself involves a diverse microbial community, in which the relative abundance and activity level of different microbes shift over time. Because levels of aeration and the composition of organic matter will vary, there is also heterogeneity across a given pile or windrow. The multi-stage composting process begins with the mesophilic phase, in which mesophilic microbes break down easily degradable compounds until the generated waste heat increases the temperature to 40 ºC, which inhibits their growth.[33] It is during the mesophilic phase that nitrifying and denitrifying bacteria produce N_2O.[33] Above 40 ºC, the thermophilic microbes begin to dominate and the increased activity of methanogens results in greater CH_4 emissions.[33] At this point, reaching temperatures above 55 ºC is desirable because this kills most human and plant pathogens; however, aeration is necessary to prevent the pile from exceeding 65 ºC, the threshold where most microbes are killed and the rate of decomposition decreases.[34] After the thermophilic phase, the compost cools and undergoes a curing and maturation stage, during which slow decomposition continues as mesophilic microbes become dominant again.

Composting operations are designed to facilitate this natural process, and practices at different facilities are distinguished by the manner in which material is stored and aerated, either in windrows or vessels and with manual, passive, or forced aeration. With in-vessel composting, material is contained in a series of containers or concrete bunkers, in which the temperature and air flow are controlled. This approach requires less land area than windrow composting and can be more efficient with proper management, but is a more expensive method.[35] In industrial-scale windrow composting operations, material is placed in rows of long and narrow piles called windrows. These windrows can either be left uncovered or can be enclosed by plastic sheeting or within bags. The dimensions of these piles are typically 2–6 meters (m) wide and 1–3 m in height, which is large enough to maintain thermophilic composting conditions while also ensuring adequate aeration.[36,37] There are several methods of aeration used to ensure the aerobic conditions required for composting. One method is to periodically turn uncovered compost piles manually or mechanically. Alternatively, static piles, either uncovered or enclosed by plastic sheeting, are aerated by natural, passive, or forced means. Natural aeration strictly relies on diffusion for air flow through the pile, but this approach can be inhibited by high moisture content material that reduces air space and increases the likelihood of conditions in the pile becoming anaerobic.[38] Passively aerated piles include perforated pipes to promote air circulation that is driven by thermal gradients. Forced aeration similarly uses perforated pipes but includes a positively or negatively pressurized pump to either push or pull air through the composting pile on prescribed cycles to control temperature and optimize the composting process. In negative aeration, the air drawn from piles may be treated with a biofilter to control odor and VOCs.[39,40] Naturally and passively aerated piles compost at a slower rate, whereas the controlled forced aeration or turning of piles result in shorter composting cycles.[41]

3.3 Understanding and Measuring Emissions from Composting

3.3.1 Overview of Key Emission Sources

By mass, CO_2 is the dominant compound emitted to the atmosphere during composting operations.[33] During each stage of composting, some carbon present in the organic material is oxidized to CO_2. Because this CO_2 production is a natural part of organic decomposition and the carbon present in most compost feedstocks is biogenic (part of the contemporary carbon cycle, in contrast to fossil carbon), these emissions are considered to be climate-neutral.[33,42,43] Emissions of other air quality and climate-relevant pollutants vary, depending on factors like oxygen availability, temperature, and moisture content. Under anaerobic conditions, decomposition occurs more slowly and methanogens create greater quantities of CH_4, while emitted CO_2 decreases. Localized areas of anaerobic decomposition in composting operations are inevitable, but turning and aeration can minimize CH_4 emissions. Methanotrophs play an important role in consuming CH_4 that may be produced in localized anaerobic regions of the pile or windrow; one study suggested that 46–98% of CH_4 produced during composting operations is consumed by methanotrophs before it can escape to the atmosphere.[44] Carbon monoxide (CO) formation is well documented, but the mechanisms are still not fully understood by the scientific community.[45] CO in composting environments can be formed through thermochemical processes, stimulated by heat and ultraviolet radiation, and CO can also be produced and consumed by microbes.[45–47]

Nitrogen cycling in composting operations involves numerous direct and indirect processes, but an understanding of the basic mechanisms is important, given the relevance of N_2O and NH_3 emissions to the climate and human health. Biological removal of nitrogen involves nitrification and denitrification, and ultimately results in N_2O emissions.[48] NH_3 is produced as microbes consume peptides and amino acids present in protein-rich waste. Nitrification is a two-step process in which microbes oxidize NH_3 to nitrite (NO_2^-) and subsequently oxidize NO_2^- to nitrate (NO_3^-). A fraction of the NO_2^- formed will be converted to nitric oxide (NO) and eventually N_2O by ammonia oxidizing bacteria, rather than forming NO_3^-. During denitrification, microbes anaerobically convert NO_3^- back to NO_2^-, then to NO, and ultimately to N_2O, most (but not all) of which is ultimately converted to nitrogen gas (N_2). NH_3 can also be directly emitted to the atmosphere, particularly from well-aerated piles where it escapes before microbes are able to oxidize it. NH_3 emissions from compost increase with increasing aeration, lower C:N ratios, higher temperatures, and higher pH.[49] The conditions for reducing NH_3 volatilization, unfortunately, can be counter to the optimal microbial conditions for fast and efficient composting.[50]

3.3.2 Emissions Measurement Methods

Many methods are used to measure emission rates from composting, and each has advantages and disadvantages to consider when interpreting and using the empirical data. Emissions can be characterized in controlled laboratory experiments or with in-situ field measurements. Sampling can be conducted continuously in the field with pollutant analyzers or intermittently by collecting discrete samples of emitted gas into canisters or bags that are later analyzed in the laboratory. Pollutant concentrations can be measured at a single point or integrated across the composting pile. The tradeoff in temporal and spatial resolution between these sampling approaches depends on the sampling conditions and objectives of the study.

Laboratory experiments have been used to approximate the composting process under controlled conditions in reactors that are typically ~10–200 L in volume.[51–64] Lab experiments allow for a better understanding and characterization of how specific environmental conditions, such as temperature, pH, moisture content, and material, affect pollutant emissions than can often be attained with field measurements. However, the smaller lab-scale and experimental conditions may not be representative of the real-world. These emission factors should be used with caution, or ideally validated against field measurements for similar materials and conditions.

Measurements can be made in the field as relatively controlled experiments of pilot- or full-scale test windrows that are maintained separately from normal operations.[19,42,65–70] In-situ sampling of full-scale commercial windrows operating under normal composting conditions is also common.[11,71–75] Ideally, field measurements of emissions would be fully integrated over the windrow or pile surface, over the full duration of the composting cycle, and without disrupting normal composting conditions. This ideal measurement approach is not practical under many sampling scenarios, however, given researcher resources and environmental/operational conditions. As such, many sampling methods have been used for field measurements, including but not limited to: flux chambers, gas probes, wind tunnels, open emission chambers, tracer releases, inverse dispersion analysis, micrometeorological mass balance, and high-density spot sampling. Each approach has its limitations, as described below. These emission measurements can also be accompanied by intermittent measurements and/or continuous monitoring of conditions in the windrow, which is important for developing a deeper understanding of the mechanisms driving emissions over time and space. Detailed descriptions of each measurement method and their impacts on reported emission factors are included in Appendix B.

3.3.3 Characterization of Composting Emissions Studies in Current Literature

We conducted a survey of peer-reviewed studies that report CO_2, CH_4, N_2O, NH_3, and/or VOC emissions from composting, keeping track of feedstock type and composting conditions. Where possible, we converted reported emission factors to units of kilograms of pollutant emitted per kilogram of wet (sometimes referred to as fresh or green) feedstock material composted. Composting emission factors are most commonly reported in terms of wet material because this is practical for commercial operations and general material flow tracking. However, these values should be converted to a per-dry-mass basis for use in carbon or nitrogen balance modeling since water makes up a significant portion of composting feedstocks (36–85%, Table B1). We do not provide emission factors on a dry basis because several studies did not provide sufficient data on moisture content to calculate these conversions.

Studies that did not give enough information to calculate reasonable emission factors and secondary sources that did not provide original data were excluded from our review. In one case, we excluded 6 measured emission factors from further analysis because the authors acknowledged that two of their small-scale measurement methods, a static flux chamber method and a funnel method, significantly underestimated GHG emissions.[76] In total, 388 emission factors from 46 studies reporting emission measurements were considered in the survey, corresponding to 140 composting scenarios (Table B1).

A majority of currently available research on composting emissions is focused on manure composting. Therefore, manure composting comprises most of the emissions observations across all pollutant types (Figure 3.1 and Table B1). The literature survey does include an extensive accounting of available literature on GHG composting emissions from food waste, OFMSW, yard waste, and anaerobically digested materials. Emissions from the composting of solid digestate are particularly under-studied and, given the importance of these emissions for regulatory decision-making in waste-to-energy pathways, this topic requires further research. Of the collected data, most emission factors are associated with forced aeration (Figure 3.1). This is not necessarily the most common industry practice but is more easily replicated in lab-based studies (Figure 3.1). Most commercial composting operations involve outdoor windrows that can be turned or forcibly aerated, are not equipped with effective emission control systems, and allow all fugitive emissions to be released to the atmosphere. Alternatively, in-vessel or fully enclosed composting facilities can more easily be equipped with scrubbers and biofilters to reduce atmospheric emissions.[77]

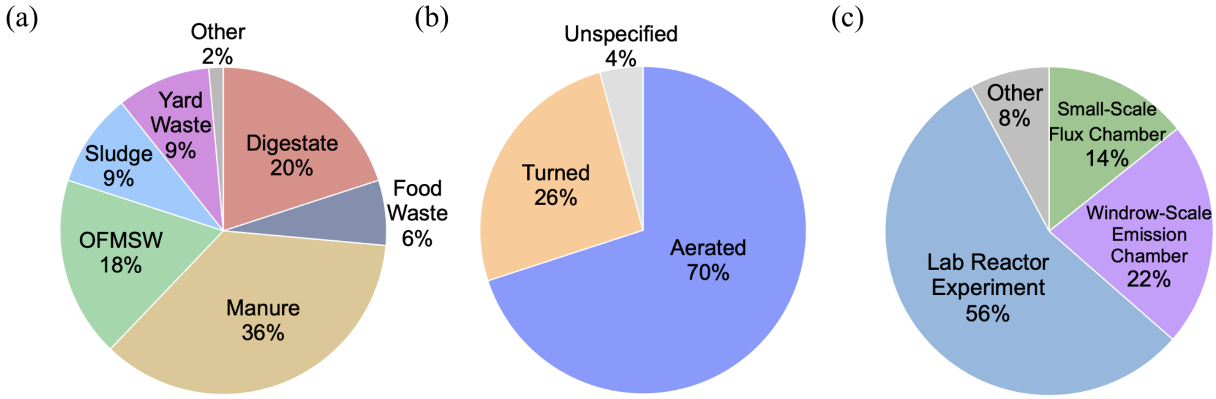

Figure 3.1 Breakdown of the 140 composting scenarios and study methods associated with the reported emission factors collected for analysis
Emission factors are categorized by: (a) composted material, (b) aeration method, (c) measurement method.

3.4 Composting Emission Factors by Source Material and Management Practices

3.4.1 Greenhouse Gas Emissions

Establishing definitive, broadly-applicable GHG emission factors for composting organic wastes is difficult because emissions vary due to a number of factors beyond feedstock (waste) type. These include: local climatic conditions at the composting site; composting method and duration; aeration method and frequency; use of bulking agent intended to provide structure to piles/windrows and facilitate aeration; and the feedstocks' VS content, C:N ratio, moisture content, and pH. In this section, we differentiate previously published emission factors based on source material and management practices to elucidate the impact of these variables on GHG emissions. Our discussion of GHG emissions from composting is focused on CH_4 and N_2O, as these gases are most likely to drive net changes in radiative forcing from composting operations. Biogenic CO_2, by contrast, is not included in our GHG footprint calculations because it is part of the contemporary carbon cycle and will be re-sequestered during plant regrowth.[20,33,42,43,78] However, depending on how a particular researcher or practitioner chooses to account for carbon flows, it may be important to account for CO_2. Further information and emission factor distributions for CO_2 are discussed in Appendix B.

3.4.1.1 Variation by Feedstock Type

GHG emission factors by feedstock type are presented in Figure 3.2 and Table 3.1. Figure 3.2 shows the distributions of CH_4 and N_2O emission factors by feedstock type (manure, OFMSW, sludge, and yard waste) and for digestate. The distribution for digestate includes data across all original feedstocks to allow for a general comparison to raw material composting; the effect of AD as a pre-treatment to composting is discussed in more detail in Section 4.1.3. We grouped together studies examining OFMSW, household waste, kitchen waste, and food waste because of ambiguous distinctions between these feedstocks. If yard waste is collected separately and paper/paperboard is recycled, the remaining OFMSW will be primarily composed of food, food-soiled paper products, and other paper products that cannot be recycled.[15] However, in that case, a composter processing this high-moisture food waste-dominated material will likely need to add a bulking agent, such as wood chips, sawdust, dry leaves, shredded paper/cardboard, or other materials that are very similar to yard waste and/or paper and paperboard. Therefore, the final material that is composted in all of these studies is likely to be similar regardless of whether yard waste and/or paper/paperboard in the original waste stream are diverted for other uses.

Table 3.1 provides mean and median emission factors by feedstock type, which can be useful for researchers and LCA practitioners who must approximate composting emissions as part of their analyses of waste management or waste-to-energy systems. It is important to note that assembling results from all prior field- or lab-based research may not provide a representative sample of real-world composting operations. For example, the majority of emissions data for composting manure came from studies examining either beef cattle, dairy cattle, or swine manure. In almost all studies considered here, composting operations for yard waste involved open, turned windrows. Most surveyed studies of sludge composting emissions were lab-based, involving closed reactors and forced aeration, and only examined wastewater treatment sludge.

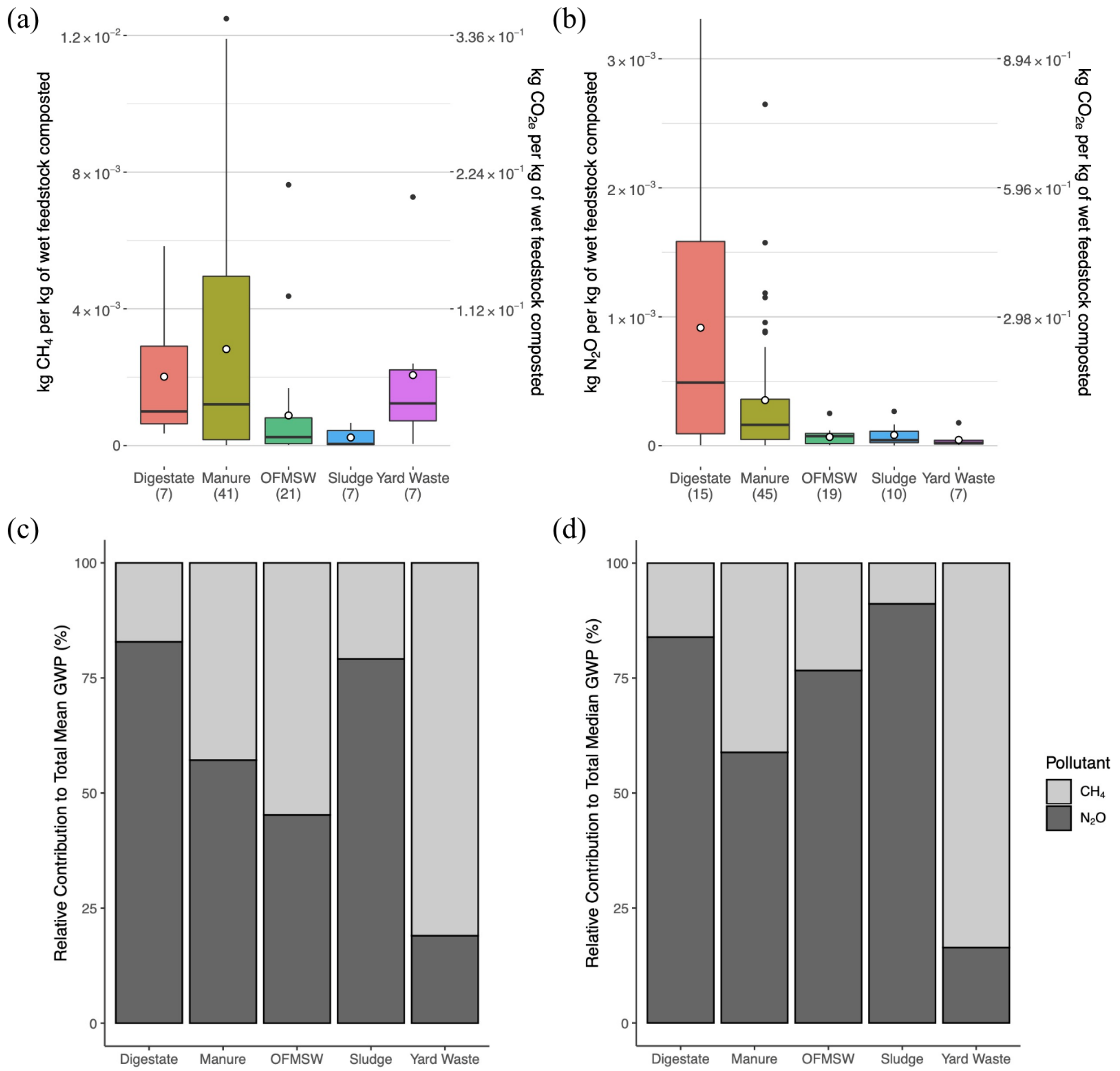

Figure 3.2 Distributions of (a) CH_4 and (b) N_2O emission factors for composting reported in the literature and relative contribution to total GWP_{100} based on (c) mean values and (d) median values

The sample size (n) of data points contributing to each boxplot is indicated in the x-axis labels for figures (a) and (b); the first value refers to the sample size of CH_4 emission factors and the second value refers to that of N_2O emission factors. Figures (a) and (b) have two y-axes: the left indicates the per-tonne mass of the specified pollutant emitted and the right shows the CO_2-equivalent emission factor, so that CH_4 and N_2O emissions can be compared with respect to GWP_{100}. The mean values for the boxplot data are indicated by the open point symbols, while outliers are shown as closed circles.

Table 3.1 Summary of GHG emission factor data for composting raw materials

Feedstock	Pollutant	Emission Factor				Sample Size
		Mean	Median	Mean	Median	
		kg pollutant/kg of wet feedstock		kg CO_{2e}/kg of wet feedstock		
Manure	CH_4	2.82×10^{-3}	1.21×10^{-3}	7.90×10^{-2}	3.39×10^{-2}	41
	N_2O	3.54×10^{-4}	1.62×10^{-4}	1.05×10^{-1}	4.83×10^{-2}	45
	CO_2	1.40×10^{-1}	1.47×10^{-1}	1.40×10^{-1}	1.47×10^{-1}	30
OFMSW	CH_4	8.79×10^{-4}	2.43×10^{-4}	2.46×10^{-2}	6.80×10^{-3}	21
	N_2O	6.80×10^{-5}	7.50×10^{-5}	2.03×10^{-2}	2.24×10^{-2}	19
	CO_2	5.63×10^{-2}	4.30×10^{-2}	5.63×10^{-2}	4.30×10^{-2}	3
Sludge	CH_4	2.34×10^{-4}	4.50×10^{-5}	6.55×10^{-3}	1.26×10^{-3}	7
	N_2O	8.36×10^{-5}	4.36×10^{-5}	2.49×10^{-2}	1.3×10^{-2}	7
	CO_2	1.75×10^{-2}	1.75×10^{-2}	1.75×10^{-2}	1.75×10^{-2}	2
Yard Waste	CH_4	2.06×10^{-3}	1.23×10^{-3}	5.77×10^{-2}	3.44×10^{-2}	7
	N_2O	4.54×10^{-5}	2.27×10^{-5}	1.35×10^{-2}	6.76×10^{-3}	7
	CO_2	1.71×10^{-1}	1.56×10^{-1}	1.71×10^{-1}	1.56×10^{-1}	4

Digestate is excluded in this table because of variation in the original raw feedstock materials.

Based on 100-year global warming potential (GWP_{100}) values for N_2O and CH_4, (GWP_{100} equal to 298 and 28, respectively), the median emission values for sludge, digestate, and OFMSW suggest that N_2O is the largest contributor to total CO_2-equivalent (CO_{2e}) emissions, while CH_4 emissions are higher on a CO_{2e} basis for yard waste (Figure 3.2). Manure composting resulted in the highest total GWP_{100}, with a roughly even split between CH_4 and N_2O on a CO_{2e} basis (Figure 3.2, Table 3.1). Pardo et al. (2015) similarly found that composting manure resulted in the highest CH_4 and N_2O emissions when comparing across different feedstocks.[22]

3.4.1.2 Impact of Feedstock Characteristics

In addition to the type of feedstock (e.g., manure, food waste), measurable characteristics including moisture content, VS content, and pH play a role in determining emissions. We attempted linear regressions using the ordinary least squares method and more robust regressions using M-estimation to assess the relationship between each feedstock characteristic listed in Table B1 (% bulking agent, VS content, C:N ratio, moisture content and pH) and each emission factor. The observed relationships were not statistically significant, even when controlling for feedstock type or measurement methods. However, a few general trends emerged, consistently offering residual standard error less than 0.01 with varying degrees of freedom. The data collected in the literature survey suggests a positive correlation between moisture content and CH_4 emissions, and this holds true when controlling for feedstock type, which is supported by results from Pardo et al. (2015).[22] There is limited data that may suggest a negative correlation between moisture content and N_2O emissions ($n = 84$) and positive correlation between VS content and N_2O emissions ($n = 22$), but further study is required to support any definitive conclusions. The collected data does not support a correlation between C:N ratio and GHG emissions and the impact of pH on overall emissions is likely negligible. Contrary to the results of our literature survey and analysis, Jiang et al. (2011) found in a lab-based study of swine manure composting that moisture content did not significantly impact CH_4 emissions, C:N ratio was negatively correlated with CH_4 emissions, and neither moisture content nor C:N ratio had an impact on N_2O emissions.[53]

3.4.1.3 Impact of Anaerobic Digestion Prior to Composting

The literature on GHG emissions from composting of digestate is limited, making it difficult to draw conclusions about the impact of AD as a strategy for pretreating organic waste prior to composting. However, even with the limited data available, there are some basic relationships that can be used to approximate differences in composting emissions between post-AD material and untreated material.

Li et al. (2018) is one of the only studies that directly compares emissions from the composting of post-AD digestate to the same undigested material as a control.[63] In this lab-based experimental study, Li et al. composted raw, untreated feedstock—a mixture of manure and agricultural residues—as well as feedstock that first underwent AD for varied digestion times. Corn stover was added to ensure a similar bulk density across all samples during composting. CH_4 emissions during composting increased relative to the non-digested control treatment when the feedstock material underwent AD for only 15 days, but decreased when the digestion time was 30 or 45 days. Without additional data on the microbiomes and volatile solids content in these composting experiments, it is only possible to speculate as to why shorter AD residence times caused elevated CH_4 emissions during subsequent composting. It is possible that insufficient residence times during AD may allow digestate to be "seeded" with methanogens.[63] It is also possible that insufficient AD residence times result in higher concentrations of intermediate products from the hydrolysis, acidogenesis, and acetogenesis stages of AD in the final digestate. Additional studies and data would be required to support the development of feedstock-specific composting emission factors for post-AD materials.

Li et al. (2018) offers more conclusive results regarding the impact of AD on N_2O emissions from composting, which can be the primary contributor to total GWP_{100} from composting (Figure 3.2).[63] Piles pretreated with AD had 57–81% lower N_2O emissions relative to the non-digested control. Longer digestion times resulted in further reductions in N_2O emissions. Li et al. reported an average VS reduction during AD of 61% (individual VS reduction data for each batch treated with AD was not reported). For perspective, the mean N_2O emission factor reported by Li et al.

for post-AD manure is 69% lower than the mean N_2O emission factor for composting untreated manure in Table 3.1.

In addition to Li et al. (2018), seven other studies measured GHG emissions from composting digested materials, but did not include controls (identical untreated materials).[11,54,61,62,64,71,77,79] For instance, Colón et al. (2012) included a comparison of in-vessel composting with and without AD pretreatment, finding that N_2O emissions were 53% lower and CH_4 emissions were ~7 times higher for OFMSW treated with AD relative to raw OFMSW.[77] These results support the assertion that AD can reduce N_2O emissions from composting, but it is important to note that Colón et al. observed real-world operations at facilities with similar but not identical OFMSW feedstocks. Maulini-Duran et al. (2013) compared two different types of wastewater treatment sludge: (1) sludge sent directly from a wastewater treatment facility to composting without undergoing AD; and (2) sludge at a separate facility, treated with AD and subsequently sent to composting.[54] They found that CH_4 and N_2O emissions were respectively 60 and >100 times higher for the post-AD material. However, there was not a proper control in this study because the source material originated from entirely different facilities. The N_2O emission factors reported by Maulini-Duran et al. for composting post-AD sludge aligned better with N_2O emissions reported by several other studies for composting raw, untreated sludge.[19,60]

Preble et al. (2020) measured emissions at a commercial-scale composting facility that processed digestate remaining after dry (high-solids) AD of OFMSW and calculated GHG emission factors per unit of incoming material.[11] This study did not include a control comparison to untreated OFMSW. However, it is notable that their reported N_2O emission factor is approximately 80% lower than mean and median N_2O emission factors for composting untreated OFMSW shown in Table 3.1. Conversely, Preble et al. report a CH_4 emission factor that is ~5 times higher than the mean value and ~18 times higher than the median value for untreated OFMSW (Table 3.1). Although Preble et al. did not directly measure VS reduction during the dry AD process, EPA WARM uses a VS reduction of 75% during AD of municipal food waste, a reasonable proxy for OFMSW.[80]

Like Preble et al. (2020), Beylot et al. (2015) studied the emissions from composting post-AD OFMSW and observed N_2O emissions that were 75% lower than what is reported by Preble et al. and per-tonne CH_4 emissions that were ~30% higher.[79] Zeng et al. (2016) conducted a series of lab-based trials to assess nitrogen emissions from composting digested OFMSW under a variety of conditions, including varied bulking agents, feedstock mixing ratios, and initial moisture content, and found N_2O emissions ranging from 5.6×10^{-4} to 3.3×10^{-3} kg per tonne of wet feedstock.[61] This range is higher than what has been reported from field measurements of both pre- and post-AD OFMSW composting. While lab-based experiments can be useful for comparing a range of materials and conditions under controlled conditions, we advise against relying on these values to represent commercial composting conditions.

Based on the limited data available on emissions from pre- and post-AD organic waste, the question is whether there is a defensible method for approximating differences in composting emissions in the absence of reliable measured data. The studies reviewed here suggest that treating waste with AD, thereby lowering its VS content, can subsequently reduce N_2O emissions during composting relative to the alternative approach of sending untreated material straight to composting without AD. For researchers and practitioners who must approximate emission factors for composting digestate, it may be appropriate to select a measured emission factor for composting raw materials and apply a reduction factor equivalent to the estimated VS reduction. For example, by applying the lowest observed reduction in N_2O emissions (57%) from directly comparable emission measurements Li et al. (2018) to the mean values in Table 3.1, we estimate that composting digested OFMSW emits 3.9×10^{-5} kg of N_2O per wet tonne and digested manure emits 2.0×10^{-4} kg of N_2O per wet tonne.[63] In practice, longer AD residence times and greater reductions in VS may lead to further reductions in N_2O emissions during composting.

Unfortunately, approximating differences in CH_4 emissions may be more challenging than estimating N_2O. Li et al. (2018) found that changes in CH_4 emissions were dependent on AD residence time, with shorter residence times translating to elevated CH_4 emissions. The increase in CH_4 emissions when comparing the data for digested OFMSW from Preble et al. (2020) and Beylot et al. (2015) to mean or median values reported in Table 3.1 may be driven by management practices and/or the fact that the material was anaerobically digested. Highly degradable feedstocks, like manure, OFMSW, and digestate, can create oxygen-depleted zones in compost piles that are compacted and/or not sufficiently aerated, thereby increasing CH_4 production.[51,54] Because of variability in pile management and lack of detailed reporting on these practices, it is likely safest to assume that AD has no effect on CH_4 emissions during composting, provided AD residence times are not below industry standard practices.

3.4.1.4 Impact of Composting Methods

As noted previously, different methods for managing compost piles are likely to impact emissions, particularly if some are more effective than others at maintaining aerobic conditions. With regards to composting methods, our analysis focuses on how turning or forced aeration impacts GHG emissions. Using OFMSW as an example, Figure 3.3 demonstrates the differences in distributions of both CH_4 and N_2O emission factors when grouping by aeration method. The median CH_4 emission factor was ~1.5 times higher when the primary aeration method was turning versus forced aeration, and the mean value was nearly 4 times higher. This is supported by a meta-analysis from Pardo et al. (2015), which found turning to be associated with higher GHG emissions.[22] An important caveat is that this trend may be related to the relatively high number of lab-based studies among those involving forced aeration. Because it is easier to control conditions and maintain proper aeration in laboratory settings that often use enclosed compost reactors, these results may not accurately reflect emissions in industrial scale composting.

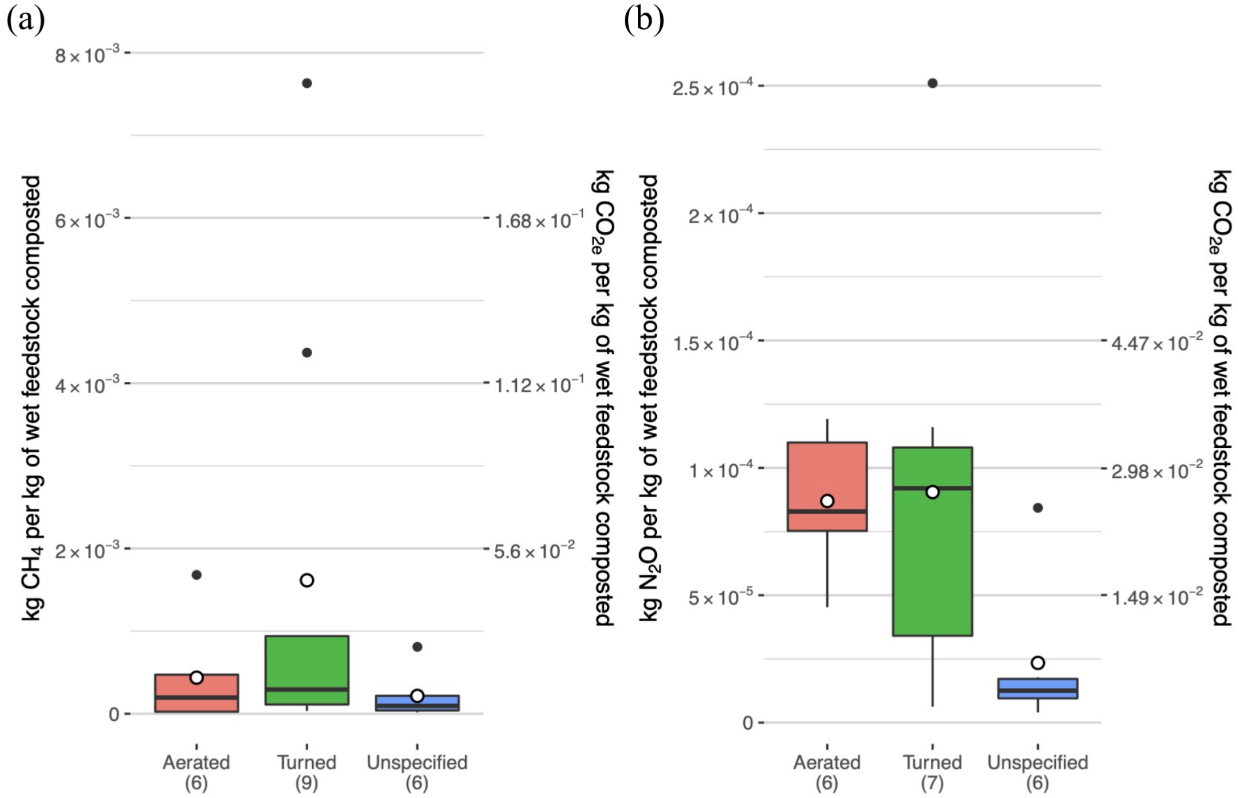

Figure 3.3 Distributions of (a) CH_4 and (b) N_2O emission factors for OFMSW composting based on aeration method

The sample size (n) of data points contributing to each boxplot is indicated in the x-axis labels. Each figure has two y-axes: the left indicates the per-tonne mass of the specified pollutant emitted and the right shows the CO_2-equivalent emission factor. The mean values for the boxplot data are indicated by the open point symbols, while outliers are shown as closed circles.

As would be expected, several studies have confirmed through measurements that CH_4 emissions decrease with higher aeration rates, but these studies were less consistent in their findings regarding the impact of aeration on N_2O emissions.[53,58,60,81] Unlike the CH_4 emission factor distributions, the N_2O emission factor distributions do not diverge significantly based on aeration methods (Figure 3.3). For instance, the average N_2O emission factor for composting with turning is only 4% greater than that for composting with forced aeration. There is not sufficient evidence to suggest that aeration method has a significant impact on N_2O emissions from composting.

3.4.2 Ammonia Emissions

Table 3.2 presents the feedstock-specific mean and median NH_3 emission factors from our literature survey. Boxplot visualizations of the NH_3 data are provided in Appendix B (Figure B2). As is the case with GHG emissions, the highest average NH_3 emission factor is associated with manure, followed by OFMSW. Composting yard waste emits the least NH_3. As discussed earlier, NH_3 is a product of microbial decomposition of proteins in the composted waste, and a fraction of that nitrogen will ultimately be emitted as N_2O. Elevated NH_3 and N_2O emissions can simply indicate that a protein-rich feedstock is being decomposed through the nitrification and denitrification processes, although these emissions can also be sensitive to compost management methods.[60]

Table 3.2 Summary of NH_3 and VOC emission factor data for composting raw materials and digestate

Feedstock	NH_3 Emission Factors (kg NH_3/kg of wet feedstock)			VOC Emission Factors (kg VOC/kg of wet feedstock)		
	Mean	Median	Sample Size	Mean	Median	Sample Size
Manure	2.04×10^{-3}	1.64×10^{-3}	44	6.06×10^{-5}	6.06×10^{-5}	2
OFMSW	1.03×10^{-3}	2.79×10^{-4}	29	1.71×10^{-3}	3.60×10^{-4}	13
Sludge	7.70×10^{-4}	3.27×10^{-4}	13	1.77×10^{-4}	1.80×10^{-4}	3
Yard Waste	8.91×10^{-5}	2.50×10^{-5}	5	5.23×10^{-4}	4.62×10^{-4}	4
Digestate	5.50×10^{-4}	6.22×10^{-5}	25	1.16×10^{-4}	3.72×10^{-5}	11

According to [82], maintaining aerobic conditions is imperative for controlling NH_3 and other odorous emissions because many of these compounds are produced from anaerobic processes.[82] Bulking agents can increase porosity to facilitate better aeration; Zhang et al. (2021) found that composting kitchen waste emitted 62% more NH_3 than composting a mixture of 85% kitchen and 15% garden waste.[83] Shao et al. (2014) observed a similar effect on NH_3 emissions as the bulking agent-to-substrate ratio increased.[84] In terms of operational methods, there appear to be tradeoffs in NH_3 emission rates when forced aeration is used to maintain aerobic conditions. Several studies have found that intermittent aeration at lower rates reduced NH_3 emissions during swine manure composting.[53,60,81] This is generally supported by other experimental studies that have observed increases in NH_3 emissions when forced aeration increases.[49,53,60] In addition to managing aeration, composters can also use microbial inoculation to control NH_3 emissions.[82,85] Chen et al. (2022) measured a ~20% reduction in NH_3 emissions when composting sewage sludge with a compound bacterial consortium relative to the control.[85]

Unlike for N_2O, there is not consistent evidence to suggest whether treating waste with AD prior to composting increases or decreases NH_3 emissions. A study by Smet et al. (1999) measured odors from OFMSW composting, AD of OFMSW and digested OFMSW composting and found that AD pretreatment reduced NH_3 emissions by 73%.[86] Even when including emissions during AD, composting raw OFMSW still emitted 72% more NH_3 than combined AD and composting. Maulini-Duran et al. (2013) observed a 98% decrease in NH_3 emissions when comparing raw sludge composting to digested sludge composting; however, as noted earlier, this study did not include a proper experimental control, as the material came from two entirely different facilities.[54] Rincón et al. (2019) compared emissions from 15 different feedstocks, including 5 different digestates, and found that on average and a wet mass basis, NH_3 emissions were 87%

lower for digested materials compared to raw feedstocks. [87] Like Maulini-Duran et al, Rincón et al. did not include proper experimental controls since the feedstocks all came from different sites and the digestates were not derived from the same material in the raw feedstocks. In contrast, Colón et al. (2012) found that NH_3 emissions from OFMSW composting roughly doubled with AD pretreatment.[77] Similarly, Li et al. (2018) observed an increase of up to 40% in NH_3 emissions from manure composting when materials first underwent AD.[63] The mean NH_3 emission factor for composting post-AD materials (including OFMSW, manure, and sludge) is lower than that for raw OFMSW or manure, but on the same order of magnitude as the mean value for composting sludge (Table 3.2). Generally, composting untreated yard waste appears to emit less NH_3 than composting digestates, but there is no available emissions data on composting digested yard waste.

3.4.3 VOC Emissions

Of all the compounds discussed in this review, VOCs are the least commonly reported and, although individual compounds may have differing effects on local odor concerns and air quality, VOC emissions are typically summed and reported as a total mass. VOCs include ketones, alcohols, terpenes and other carbon compounds that can participate in atmospheric reactions with the exception of carbon monoxide, carbon dioxide, carbonic acid, metallic carbides or carbonates, and ammonium carbonate as defined by the EPA.[88] CH_4 is a VOC, although it is often reported separately because of its relevance for climate forcing; remaining VOCs are reported as non-methane VOCs. All of the values reported in this section and in Table 3.2 exclude CH_4. The surveyed literature includes a total of 33 VOC emission factors, 11 of which are for digestate composting. The summary data broken down by feedstock type is provided in Table 3.2, but the sample sizes are limited (n<5 for all raw feedstocks except OFMSW) so those emission factors should be used with caution. Further research is required to establish accurate distributions of these feedstock-specific emission factors. Without controlling for feedstock type, the mean emission factor is 8.14×10^{-4} and the median is 1.06×10^{-3} kg VOC per kg of wet feedstock for composting.[11,54,72,77,84,86,87,89,90] If pressed to assume a non-zero value, researchers and practitioners may choose to use a median or mean value that excludes digestate. For non-digestate feedstocks (including sludge, OFMSW, and yard waste), the mean emission factor is 1.18×10^{-3} and the median is 2.1×10^{-4} kg VOC per kg of wet feedstock (n = 21).

All of the surveyed VOC emission factors for digestate composting are at least an order of magnitude lower than the average for non-digestate composting, suggesting AD may reduce VOC emissions from composting. Most of the surveyed literature provides evidence to support this conclusion with the exception of Colón et al. (2012) which measured higher VOC emissions from composting post-AD OFMSW compared to raw OFMSW.[77] Smet et al. (1999) compared the VOC emissions from composting and AD of OFMSW, as they did with NH_3, and the results show a 99% reduction in VOC emissions from composting when AD pretreatment was used. Expanding the system boundary, the combined AD and composting scenario had 63% fewer VOC emissions than direct OFMSW composting. [86] Maulini-Duran et al. (2013) observed a decrease in VOC emissions from composting post-AD sludge relative to sludge that was not treated with AD.[54] More recently, Rincón et al. (2019) compared odorous emissions from each raw feedstock type and digestate type with the exception of digested yard waste for which they had no data.[87] On average, VOC emissions from digestate composting are 94% lower than those from raw material composting. When controlling for feedstock type, the VOC emission factors for composting digested materials are consistently lower than their raw counterparts, but as stated before, Rincón et al did not include ideal experimental controls.[87] Beyond AD, other technology options exist to specifically target and reduce VOC emissions from composting; these include but are not limited to pretreatment techniques, incineration, biotrickling filters, bioscrubber technology and membrane bioreactors.[82,91]

3.5 Conclusions

Properly accounting for composting emissions, and for organic waste management-related emissions in general, in an LCA can be exceptionally challenging. There are still large gaps in the empirical data available for the range of materials that can be composted and the key greenhouse gases and air pollutants. More fundamentally, there is a limited scientific understanding of the complex microbial communities that break down plant matter, and emissions estimates are likely to evolve as scientists gain an improved understanding of the complex chemical and biological mechanisms at work in these environments. However, by analyzing data reported across the literature and disaggregating emission factors based on pile management strategies and starting material, basic patterns emerge that can inform best-estimates for use in future analyses.

Our findings suggest that N_2O is typically the dominant contributor to the GWP_{100} of direct emissions from composting operations in properly aerated piles/windrows, assuming biogenic CO_2 emissions do not have a net climate impact. When controlling for feedstock type, N_2O accounts for 45–79% based on mean values and 59–91% based on median values of total GHG emissions on a GWP_{100} basis. Yard waste is a notable exception where GHG emissions are dominated by CH_4 (80% of GWP_{100} based on mean values or 83% based on median values), likely because of its high C:N ratio compared to other waste types such as food waste and manure. Among observed feedstock types, N_2O emissions appear to be highest for manure and lowest for yard waste and may be influenced by initial VS content. N_2O emissions seem to be impacted by whether the incoming material was previously processed in an AD facility, and this paper outlines a suggested method for adjusting the estimated N_2O emission factor based on VS reductions during AD. Conversely, CH_4 appears to be primarily related to whether the pile or windrow is adequately aerated. The impact of VS reduction, through AD or otherwise, prior to composting did not appear to have a substantial impact on CH_4 emissions, although direct comparisons in the empirical data is extremely limited and warrants further study. Assuming CH_4 emissions are heavily influenced by pile management, reducing GHG emissions from well-managed, properly aerated compost piles may require more focus on the composition and quality of feedstock materials to reduce N_2O emissions.

Regarding NH_3 and VOCs, the available data suggests that treating waste with AD prior to composting may reduce these emissions but more measurements are required to definitively support this conclusion. This uncertainty is echoed by inconsistent results and disagreement in current scientific literature, emphasizing the need for further research in this area. Reducing these emissions are a key part of improving air quality in local and surrounding communities not only because of odor concerns, but because both NH_3 and VOC contribute to atmospheric formation of $PM_{2.5}$, which has significant human health impacts. Therefore, though AD does not have a clear benefit with respect to limiting GHG emissions from composting, it can still play a role in effective organic waste management because of its potential to reduce other harmful emissions.

References

(1) Slater, R. A.; Frederickson, J. Composting municipal waste in the UK: some lessons from Europe. *Resources, Conservation and Recycling* **2001**, *32*, 359–374.

(2) Hottle, T. A.; Bilec, M. M.; Brown, N. R.; Landis, A. E. Toward zero waste: composting and recycling for sustainable venue based events. *Waste Manag.* **2015**, *38*, 86–94.

(3) Staley, B. F.; Kantner, D. L. *Municipal Solid Waste Management in the U.S.: 2010 & 2013*; Environmental Research and Education Foundation, 2016.

(4) EPA. *Advancing Sustainable Materials Management: 2018 Fact Sheet*; U.S. Environmental Protection Agency, 2020.

(5) Breitenbeck, G. A.; Schellinger, D. Calculating the reduction in material mass and volume during composting. *Compost Sci. Util.* **2004**, *12*, 365–371.

(6) Larney, F. J.; Buckley, K. E. Dry matter mass balance estimates for composted feedlot manure. *Compost Sci. Util.* **2007**, *15*, 222–227.

(7) Andersen, J. K.; Boldrin, A.; Christensen, T. H.; Scheutz, C. Mass balances and life-cycle inventory for a garden waste windrow composting plant (Aarhus, Denmark). *Waste Management & Research* **2010**, *28*, 1010–1020.

(8) Tiquia, S.; Richard, T.; Honeyman, M. Carbon, nutrient, and mass loss during composting. *Nutrient Cycling in Agroecosystems* **2002**, *62*, 15–24.

(9) Brown, S. Greenhouse gas accounting for landfill diversion of food scraps and yard waste. *Compost Sci. Util.* **2016**, *24*, 11–19.

(10) Martínez-Blanco, J.; Lazcano, C.; Christensen, T. H.; Muñoz, P.; Rieradevall, J.; Møller, J.; Antón, A.; Boldrin, A. Compost benefits for agriculture evaluated by life cycle assessment. A review. *Agronomy Sust. Developm.* **2013**, *33*, 721–732.

(11) Preble, C. V.; Chen, S. S.; Hotchi, T.; Sohn, M. D.; Maddalena, R. L.; Russell, M. L.; Brown, N. J.; Scown, C. D.; Kirchstetter, T. W. Air pollutant emission rates for dry anaerobic digestion and composting of organic municipal solid waste. *Environ. Sci. Technol.* **2020**, *54*, 16097–16107.

(12) Bracmort, K. Low Carbon Fuel Standard: In Brief (No. R46835). *Congressional Research Service* **2021**.

(13) Domingo, J. L.; Nadal, M. Domestic waste composting facilities: a review of human health risks. *Environ. Int.* **2009**, *35*, 382–389.

(14) Thakrar, S. K.; Goodkind, A. L.; Tessum, C. W.; Marshall, J. D.; Hill, J. D. Life cycle air quality impacts on human health from potential switchgrass production in the United States. *Biomass and Bioenergy* **2017**.

(15) Nordahl, S. L.; Devkota, J. P.; Amirebrahimi, J.; Smith, S. J.; Breunig, H. M.; Preble, C. V.; Satchwell, A. J.; Jin, L.; Brown, N. J.; Kirchstetter, T. W.; Scown, C.D. Life-cycle greenhouse gas emissions and human health trade-offs of organic waste management strategies. *Environ. Sci. Technol.* **2020**, *54*, 9200–9209.

(16) Abbs, A.; Reul-Chen, C. *Composting in California: Addressing Air Quality Permitting and Regulatory Issues for Expanding Infrastructure*; Discussion Paper; California Air

Resources Board, 2018.

(17) CARB. *ARB Emissions Inventory Methodology for Composting Facilities*; California Air Resources Board, 2015.

(18) Boldrin, A.; Andersen, J. K.; Møller, J.; Christensen, T. H.; Favoino, E. Composting and compost utilization: accounting of greenhouse gases and global warming contributions. *Waste Manag. Res.* **2009**, *27*, 800–812.

(19) Amlinger, F.; Peyr, S.; Cuhls, C. Greenhouse gas emissions from composting and mechanical biological treatment. *Waste Manag. Res.* **2008**, *26*, 47–60.

(20) Brown, S.; Kruger, C.; Subler, S. Greenhouse gas balance for composting operations. *J. Environ. Qual.* **2008**, *37*, 1396–1410.

(21) Lou, X. F.; Nair, J. The impact of landfilling and composting on greenhouse gas emissions--a review. *Bioresour. Technol.* **2009**, *100*, 3792–3798.

(22) Pardo, G.; Moral, R.; Aguilera, E.; Del Prado, A. Gaseous emissions from management of solid waste: a systematic review. *Glob. Chang. Biol.* **2015**, *21*, 1313–1327.

(23) Bong, C. P. C.; Lim, L. Y.; Ho, W. S.; Lim, J. S.; Klemeš, J. J.; Towprayoon, S.; Ho, C. S.; Lee, C. T. A review on the global warming potential of cleaner composting and mitigation strategies. *J. Clean. Prod.* **2017**, *146*, 149–157.

(24) Sayara, T.; Sánchez, A. Gaseous emissions from the composting process: controlling parameters and strategies of mitigation. *Processes* **2021**, *9*, 1844.

(25) Breunig, H. M.; Amirebrahimi, J.; Smith, S.; Scown, C. D. Role of digestate and biochar in carbon-negative bioenergy. *Environ. Sci. Technol.* **2019**, *53*, 12989–12998.

(26) Morris, J.; Scott Matthews, H.; Morawski, C. Review and meta-analysis of 82 studies on end-of-life management methods for source separated organics. *Waste Manag.* **2013**, *33*, 545–551.

(27) EPA. Paper and Paperboard: Material-Specific Data, *Waste and Recycling* https://www.epa.gov/facts-and-figures-about-materials-waste-and-recycling/paper-and-paperboard-material-specific-data (accessed May 26, 2020).

(28) Logan, M.; Visvanathan, C. Management strategies for anaerobic digestate of organic fraction of municipal solid waste: Current status and future prospects. *Waste Manag. Res.* **2019**, *37*, 27–39.

(29) Kong, D.; Huitric, R.; Iacoboni, M.; Chan, G. *Evaluation of Green Waste Management Impacts on GHG Emissions Alternative Daily Cover Compared with Composting*; Solid Waste Management Department, Los Angeles County Sanitation Districts, 2009.

(30) van Haaren, R.; Themelis, N. J.; Barlaz, M. LCA comparison of windrow composting of yard wastes with use as alternative daily cover (ADC). *Waste Manag.* **2010**, *30*, 2649–2656.

(31) Smith, S. J.; Satchwell, A. J.; Kirchstetter, T. W.; Scown, C. D. The implications of facility design and enabling policies on the economics of dry anaerobic digestion. *Waste Manag.* **2021**, *128*, 122–131.

(32) U.S. Environmental Protection Agency. *METHOD 1684: Total, Fixed, and Volatile Solids in Water, Solids, and Biosolids*; U.S. Environmental Protection Agency: Washington DC, 2001.

(33) Hellmann, B.; Zelles, L.; Palojarvi, A.; Bai, Q. Emission of climate-relevant trace gases and succession of microbial communities during open-windrow composting. *Appl. Environ. Microbiol.* **1997**, *63*, 1011–1018.

(34) Hassen, A.; Belguith, K.; Jedidi, N.; Cherif, A.; Cherif, M.; Boudabous, A. Microbial

characterization during composting of municipal solid waste. *Bioresour. Technol.* **2001**, *80*, 217–225.

(35) Lim, L. Y.; Bong, C. P. C.; Lee, C. T.; Klemes, J. J.; Sarmidi, M. R.; Lim, J. S. Review on the current composting practices and the potential of improvement using two-stage composting. *Chemical Engineering Transactions* **2017**, *61*, 1051–1056.

(36) Hay, J. C.; Kuchenrither, R. D. Fundamentals and application of windrow composting. *J. Environ. Eng.* **1990**, *116*, 746–763.

(37) Kuchenrither, R. D.; Martin, W. J.; Smith, D. G.; Williams, D. W. Design and operation of an aerated windrow composting facility. *J Water Pollut Control Fed* **1985**, *57*, 213–219.

(38) Fernandes, L.; Sartaj, M. Comparative study of static pile composting using natural, forced and passive aeration methods. *Compost Sci. Util.* **1997**, *5*, 65–77.

(39) Sironi, S.; Botta, D. Biofilter efficiency in odor abatement at composting plants. *Compost science & utilization* **2001**, *9*, 149–155.

(40) Park, K. J.; Choi, M. H.; Hong, J. H. Control of composting odor using biofiltration. *Compost Sci. Util.* **2002**, *10*, 356–362.

(41) Szanto, G. L.; Hamelers, H. V. M.; Rulkens, W. H.; Veeken, A. H. M. NH_3, N_2O and CH_4 emissions during passively aerated composting of straw-rich pig manure. *Bioresour. Technol.* **2007**, *98*, 2659–2670.

(42) Vergara, S. E.; Silver, W. L. Greenhouse gas emissions from windrow composting of organic wastes: Patterns and emissions factors. *Environmental Research Letters* **2019**, *14*, 124027.

(43) Bogner, J.; Ahmed, M. A.; Diaz, C.; Faaij, A.; Gao, Q.; Hashimoto, S.; Mareckova, K.; Pipatti, R.; Zhang, T. Chapter 10: Waste Management. In *Climate Change 2007: Mitigation. Contribution of Working Group III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*; Metz, B.; Davidson, O. R.; Bosch, P. R.; Dave, R.; Meyer, L. A., Eds.; Cambridge University Press: Cambridge, United Kingdom and New York, NY, USA, 2007.

(44) Jäckel, U.; Thummes, K.; Kämpfer, P. Thermophilic methane production and oxidation in compost. *FEMS Microbiol. Ecol.* **2005**, *52*, 175–184.

(45) Stegenta-Dąbrowska, S.; Drabczyński, G.; Sobieraj, K.; Koziel, J. A.; Białowiec, A. The biotic and abiotic carbon monoxide formation during aerobic co-digestion of dairy cattle manure with green waste and sawdust. *Front. Bioeng. Biotechnol.* **2019**, *7*, 283.

(46) Westlake, D. W.; Roxburgh, J. M.; Talbot, G. Microbial production of carbon monoxide from flavonoids. *Nature* **1961**, *189*, 510–511.

(47) Conrad, R.; Seiler, W. Role of microorganisms in the consumption and production of atmospheric carbon monoxide by soil. *Appl. Environ. Microbiol.* **1980**, *40*, 437–445.

(48) Habart, J.; Tlustoš, P.; Hanč, A.; Švehla, P.; Váňa, J.; Tluka, P.; Jelínek, F. The role of aeration intensity, temperature regimes and composting mixture on gaseous emission during composting. *Compost Sci. Util.* **2010**, *18*, 194–200.

(49) de Guardia, A.; Petiot, C.; Rogeau, D.; Druilhe, C. Influence of aeration rate on nitrogen dynamics during composting. *Waste Manag.* **2008**, *28*, 575–587.

(50) Koenig, R. T.; Palmer, M. D.; Miner, F. D.; Miller, B. E.; Harrison, J. D. Chemical amendments and process controls to reduce ammonia volatilization during in-house composting. *Compost Sci. Util.* **2005**, *13*, 141–149.

(51) He, Y.; Inamori, Y.; Mizuochi, M.; Kong, H.; Iwami, N.; Sun, T. Measurements of N2O

and CH4 from the aerated composting of food waste. *Sci. Total Environ.* **2000**, *254*, 65–74.

(52) Pagans, E.; Barrena, R.; Font, X.; Sánchez, A. Ammonia emissions from the composting of different organic wastes. Dependency on process temperature. *Chemosphere* **2006**, *62*, 1534–1542.

(53) Jiang, T.; Schuchardt, F.; Li, G.; Guo, R.; Zhao, Y. Effect of C/N ratio, aeration rate and moisture content on ammonia and greenhouse gas emission during the composting. *Journal of Environmental Sciences* **2011**, *23*, 1754–1760.

(54) Maulini-Duran, C.; Artola, A.; Font, X.; Sánchez, A. A systematic study of the gaseous emissions from biosolids composting: raw sludge versus anaerobically digested sludge. *Bioresour. Technol.* **2013**, *147*, 43–51.

(55) Yang, F.; Li, G. X.; Yang, Q. Y.; Luo, W. H. Effect of bulking agents on maturity and gaseous emissions during kitchen waste composting. *Chemosphere* **2013**, *93*, 1393–1399.

(56) Chowdhury, M. A.; de Neergaard, A.; Jensen, L. S. Potential of aeration flow rate and bio-char addition to reduce greenhouse gas and ammonia emissions during manure composting. *Chemosphere* **2014**, *97*, 16–25.

(57) Luo, W. H.; Yuan, J.; Luo, Y. M.; Li, G. X.; Nghiem, L. D.; Price, W. E. Effects of mixing and covering with mature compost on gaseous emissions during composting. *Chemosphere* **2014**, *117*, 14–19.

(58) Puyuelo, B.; Gea, T.; Sánchez, A. GHG emissions during the high-rate production of compost using standard and advanced aeration strategies. *Chemosphere* **2014**, *109*, 64–70.

(59) Sun, X.; Lu, P.; Jiang, T.; Schuchardt, F.; Li, G. Influence of bulking agents on CH_4, N_2O, and NH_3 emissions during rapid composting of pig manure from the Chinese Ganqinfen system. *J. Zhejiang Univ. Sci. B* **2014**, *15*, 353–364.

(60) Yuan, J.; Chadwick, D.; Zhang, D.; Li, G.; Chen, S.; Luo, W.; Du, L.; He, S.; Peng, S. Effects of aeration rate on maturity and gaseous emissions during sewage sludge composting. *Waste Manag.* **2016**, *56*, 403–410.

(61) Zeng, Y.; De Guardia, A.; Dabert, P. Improving composting as a post-treatment of anaerobic digestate. *Bioresour. Technol.* **2016**, *201*, 293–303.

(62) Wang, T.-T.; Wang, S.-P.; Zhong, X.-Z.; Sun, Z.-Y.; Huang, Y.-L.; Tan, L.; Tang, Y.-Q.; Kida, K. Converting digested residue eluted from dry anaerobic digestion of distilled grain waste into value-added fertilizer by aerobic composting. *J. Clean. Prod.* **2017**, *166*, 530–536.

(63) Li, Y.; Luo, W.; Lu, J.; Zhang, X.; Li, S.; Wu, Y.; Li, G. Effects of digestion time in anaerobic digestion on subsequent digestate composting. *Bioresour. Technol.* **2018**, *267*, 117–125.

(64) Wang, K.; Wu, Y.; Li, W.; Wu, C.; Chen, Z. Insight into effects of mature compost recycling on N2O emission and denitrification genes in sludge composting. *Bioresour. Technol.* **2018**, *251*, 320–326.

(65) Amon; Amon, Th.; Boxberger; Alt, Ch. Emissions of NH_3, N_2O and CH_4 from dairy cows housed in a farmyard manure tying stall (housing, manure storage, manure spreading). *Nutrient Cycling in Agroecosystems* **2001**.

(66) Hao, X.; Chang, C.; Larney, F. J. Carbon, nitrogen balances and greenhouse gas emission during cattle feedlot manure composting. *J. Environ. Qual.* **2004**, *33*, 37–44.

(67) Paillat, J.-M.; Robin, P.; Hassouna, M.; Leterme, P. Predicting ammonia and carbon dioxide emissions from carbon and nitrogen biodegradability during animal waste composting. *Atmos. Environ.* **2005**, *39*, 6833–6842.

(68) Ahn, H. K.; Mulbry, W.; White, J. W.; Kondrad, S. L. Pile mixing increases greenhouse gas emissions during composting of dairy manure. *Bioresour. Technol.* **2011**, *102*, 2904–2909.

(69) Maeda, K.; Hanajima, D.; Morioka, R.; Toyoda, S.; Yoshida, N.; Osada, T. Mitigation of greenhouse gas emission from the cattle manure composting process by use of a bulking agent. *Soil Sci. Plant Nutr.* **2013**, *59*, 96–106.

(70) Chen, R.; Wang, Y.; Wang, W.; Wei, S.; Jing, Z.; Lin, X. N_2O emissions and nitrogen transformation during windrow composting of dairy manure. *J. Environ. Manage.* **2015**, *160*, 121–127.

(71) Czepiel, P.; Douglas, E.; Harriss, R.; Crill, P. Measurements of N_2O from Composted Organic Wastes. *Environ. Sci. Technol.* **1996**, *30*, 2519–2525.

(72) Cadena, E.; Colón, J.; Sánchez, A.; Font, X.; Artola, A. A methodology to determine gaseous emissions in a composting plant. *Waste Manag.* **2009**, *29*, 2799–2807.

(73) Kumar, A.; Alaimo, C. P.; Horowitz, R.; Mitloehner, F. M.; Kleeman, M. J.; Green, P. G. Volatile organic compound emissions from green waste composting: Characterization and ozone formation. *Atmos. Environ.* **2011**, *45*, 1841–1848.

(74) Hrad, M.; Binner, E.; Piringer, M.; Huber-Humer, M. Quantification of methane emissions from full-scale open windrow composting of biowaste using an inverse dispersion technique. *Waste Manag.* **2014**, *34*, 2445–2453.

(75) Zhu-Barker, X.; Bailey, S. K.; Paw U, K. T.; Burger, M.; Horwath, W. R. Greenhouse gas emissions from green waste composting windrow. *Waste Manag.* **2017**, *59*, 70–79.

(76) Andersen, J. K.; Boldrin, A.; Samuelsson, J.; Christensen, T. H.; Scheutz, C. Quantification of greenhouse gas emissions from windrow composting of garden waste. *J. Environ. Qual.* **2010**, *39*, 713–724.

(77) Colón, J.; Cadena, E.; Pognani, M.; Barrena, R.; Sánchez, A.; Font, X.; Artola, A. Determination of the energy and environmental burdens associated with the biological treatment of source-separated Municipal Solid Wastes. *Energy Environ. Sci.* **2012**, *5*, 5731–5741.

(78) Pippati, R.; Alves, J. W. S.; Gao, Q.; Cabrera, L.; Mareckova, K.; Oonk, H.; Scheehle, E.; Sharma, C.; Smith, A.; Svardal, P.; Yamada, M. Chapter 4: Biological Treatment of Solid Waste. In *2006 IPCC Guidelines for National Greenhouse Gas Inventories*; IPCC, 2006; Vol. 5.

(79) Beylot, A.; Vaxelaire, S.; Zdanevitch, I.; Auvinet, N.; Villeneuve, J. Life Cycle Assessment of mechanical biological pre-treatment of Municipal Solid Waste: a case study. *Waste Manag.* **2015**, *39*, 287–294.

(80) EPA. Waste Reduction Model (WARM). **2018**.

(81) Jiang, T.; Li, G.; Tang, Q.; Ma, X.; Wang, G.; Schuchardt, F. Effects of aeration method and aeration rate on greenhouse gas emissions during composting of pig feces in pilot scale. *J. Environ. Sci. (China)* **2015**, *31*, 124–132.

(82) Andraskar, J.; Yadav, S.; Kapley, A. Challenges and control strategies of odor emission from composting operation. *Appl. Biochem. Biotechnol.* **2021**, *193*, 2331–2356.

(83) Zhang, B.; Fan, F.; Guo, C.; Yu, M.; Zhao, M.; Song, Y.; Li, Y. Evaluation of maturity and odor emissions in the process of combined composting of kitchen waste and garden

waste. *Appl. Sci.* **2021**, *11*, 5500.

(84) Shao, L.-M.; Zhang, C.-Y.; Wu, D.; Lü, F.; Li, T.-S.; He, P.-J. Effects of bulking agent addition on odorous compounds emissions during composting of OFMSW. *Waste Manag.* **2014**, *34*, 1381–1390.

(85) Chen, L.; Li, W.; Zhao, Y.; Zhou, Y.; Zhang, S.; Meng, L. Effects of compound bacterial agent on gaseous emissions and compost maturity during sewage sludge composting. *J. Clean. Prod.* **2022**, *366*, 133015.

(86) Smet, E.; Van Langenhove, H.; De Bo, I. The emission of volatile compounds during the aerobic and the combined anaerobic/aerobic composting of biowaste. *Atmos. Environ.* **1999**, *33*, 1295–1303.

(87) Rincón, C. A.; De Guardia, A.; Couvert, A.; Le Roux, S.; Soutrel, I.; Daumoin, M.; Benoist, J. C. Chemical and odor characterization of gas emissions released during composting of solid wastes and digestates. *J. Environ. Manage.* **2019**, *233*, 39–53.

(88) EPA. What is the definition of VOC? https://www.epa.gov/air-emissions-inventories/what-definition-voc (accessed Nov 10, 2022).

(89) Büyüksönmez, F. Full-scale VOC emissions from green and food waste windrow composting. *Compost Sci. Util.* **2012**, *20*, 57–62.

(90) Rincón, C. A.; De Guardia, A.; Couvert, A.; Soutrel, I.; Guezel, S.; Le Serrec, C. Odor generation patterns during different operational composting stages of anaerobically digested sewage sludge. *Waste Manag.* **2019**, *95*, 661–673.

(91) Dhamodharan, K.; Varma, V. S.; Veluchamy, C.; Pugazhendhi, A.; Rajendran, K. Emission of volatile organic compounds from composting: A review on assessment, treatment and perspectives. *Sci. Total Environ.* **2019**, *695*, 133725.

Chapter 4

Complementary Roles for Mechanical and Solvent-Based Recycling in Low-Carbon, Circular Polypropylene

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4.1 Significance

Polypropylene is a relatively low-cost polymer with useful material properties, making it one of the most widely produced plastics. Unfortunately, repeated mechanical recycling of polypropylene degrades its properties, performance, and aesthetics, so recycling infrastructure for polypropylene is underdeveloped and it often ends up in landfills. Solvent- assisted recycling processes like dissolution have emerged, offering near virgin- quality recycled polypropylene and the promise of greater circularity. To clarify the sustainability of circular polypropylene, we offer a detailed life- cycle evaluation of mechanical recycling, dissolution- based recycling, and virgin polypropylene production. We find that while dissolution- based recycling offers modest greenhouse gas savings relative to virgin polypropylene, it serves as an important upgrading step to broaden markets served by recycled polypropylene and displace demand for virgin resin.

4.2 Introduction

Despite setting ambitious goals, most countries have struggled to reduce plastic waste accumulation, even in the face of growing evidence of its serious ecosystem, human health, and climate implications.[1–3] These struggles have caused some to question the very premise that plastics recycling is a viable solution.[4–6] In the U.S., less than 6% of all plastics are recycled.[7] Market forces, inadequate collection and sorting infrastructure, and an inability to produce virgin-quality recyclate have all played their roles in limiting recycling rates. Plastic recycling rates are higher in the European Union (averaging ~30%) due to stronger policies and higher tipping fees,[8] but many of the same limitations stand in the way of further improvements.[9] Some plastics are more easily recycled than others. Polyethylene terephthalate (PET) is arguably the easiest; it makes up just 10% of total U.S. plastic production and yet 18% is collected for recycling.[10–12] Polypropylene (PP), in contrast, makes up 14% of total polymer production and is recycled at a rate of less than 1%.[7,13] The low recycling rate for PP is driven by the same forces that hinder much of the plastic recycling industry: mechanical recycling results in an aesthetically and mechanically inferior product, making it difficult to compete with low-cost virgin material.[14] The remaining question is whether, and how, greater circularity is achievable without driving up energy use and greenhouse gas (GHG) emissions.

Advanced recycling processes can create higher quality products, although often with higher energy use and emissions.[15,16] For example, pyrolyzing PP produces a diverse array of useful hydrocarbons, however, few of these get funneled back into virgin resin production and do so with low yields and high CO_2 emissions.[15,17] Solvent-based processes have emerged as a strategy to more directly displace virgin plastic production with recycled material. Although solvent-assisted recycling processes can include either dissolution or depolymerization to monomers (also referred to as chemolysis), options for PP are currently limited to dissolution. Depolymerization to monomers (chemolysys) reverses a condensation reaction, which is not a viable option for addition polymers such as PP, PE, and polyvinyl chloride.[18] This paper focuses on solvent-assisted dissolution of PP to produce polymer chains. This direct polymer-to-polymer recycling through dissolution and precipitation offers high yields and near-virgin quality with a reduced energy penalty relative to pyrolysis.[15,19,20] We use process simulation and life-cycle assessment to show that, rather than treating mechanical and solvent-assisted PP recycling as competing options,[21] enabling plastic circularity and driving down life-cycle GHG emissions will require both processes to be scaled in tandem. Maximizing mechanical recycling will minimize

GHG emissions and produce low-grade recyclates for a market that has not yet been saturated. Developing solvent-assisted recycling processes as an optional upgrading step can provide higher-quality recyclates for a wider array of applications, including food packaging, while still achieving GHG reductions relative to virgin PP.

Our work indicates that a reframing of polymer recycling more broadly is necessary to develop realistic strategies for converting mixed plastic waste streams to recyclates that satisfy the diverse needs of the market, both for PP and potentially for other under-recycled polymers. By applying rigorous process simulation and life-cycle assessment informed by industry experts and real-world practices along the entire waste supply chain, our life-cycle energy and GHG emissions results provide the most industrially-relevant insights to-date on how conventional and advanced recycling techniques can be leveraged to minimize GHG emissions and maximize waste diversion for one of the most commonly landfilled polymers on the market today.

4.3 Results

4.3.1 Impact of Sorting Constraints and Contamination

Sorting constraints and realistic contamination levels are overlooked in much of the advanced recycling literature.[15,22] While the energy footprint of physical sorting processes is modest, melt filtration and other steps required to remove contaminants downstream can substantially impact energy use and yields. Hence, the design of recycling processes is dependent on sorting practices at material recovery facilities (MRFs). Most MRFs in the U.S. remove PET (#1) and high density polyethylene (HDPE, #2) separately, then produce mixed plastic waste bales that include PP (#5) and other plastic waste, known as #3-7 bales. The composition of these bales varies considerably by individual MRF; for our analysis, we assume PP (#5) makes up 59% of our input bale (Figure C1).

Upon arrival at recycling facilities, additional sorting is required to improve PP purity prior to dissolution or other advanced recycling processes. Solvent-based recycling is tolerant to contamination by other plastics, but increased plastic contamination in the incoming stream makes solvent selection and separation more challenging, so single-polymer feedstocks are preferable.[19] Additives and dyes are also of minimal concern for solvent-based recyclers seeing as dissolution is capable of separating these impurities. The additional sorting or "preprocessing" necessary for PP dissolution includes the same steps that are used prior to mechanical PP recycling. This means mechanical recyclers could preprocess a larger quantity of material to produce clean PP flakes and then choose, based on market conditions, what fraction to extrude on-site versus export to solvent-assisted recycling facilities. Like mechanical recycling, dissolution-based recycling includes an extrusion step. However, at dissolution-based facilities, solvents are added to reduce shearing during this step and enable the eventual removal of shorter-chain polymers.[23]

4.3.2 Life-Cycle Greenhouse Gas Impacts of PP Production and Recycling

The goal of this life-cycle assessment is to compare cradle-to-gate GHG emissions from dissolution-based recycling of rigid PP waste with conventional mechanical recycling and petroleum-derived virgin PP production in the United States. In all scenarios, the functional unit is defined as 1 tonne of PP resin produced, although, as shown in Figure 4.1, recyclate quality varies by process. We use facility-scale industry data in the open literature to assemble mass and energy balances for conventional mechanical PP recycling and virgin PP production. Our model of the PP dissolution process is based on pilot-scale operations documented in industry reports (additional detail in the Methods section and Table C2 in Appendix C), although our results are not specific to any individual site. The boundary of the analysis begins with raw material extraction in the case of virgin production and with plastic waste sorting for the recycling scenarios. Both recycling processes begin with mixed #3-7 bales.[24,25] Our analysis ends with the production of PP polymer resin that is ready for manufacturing, with the acknowledgement that the output from mechanical recycling cannot be used for all applications of virgin or solvent-assisted recycled PP. Further details on each recycling scenario are included in the Methods section and virgin PP production is described in Section C3.1 of Appendix C.

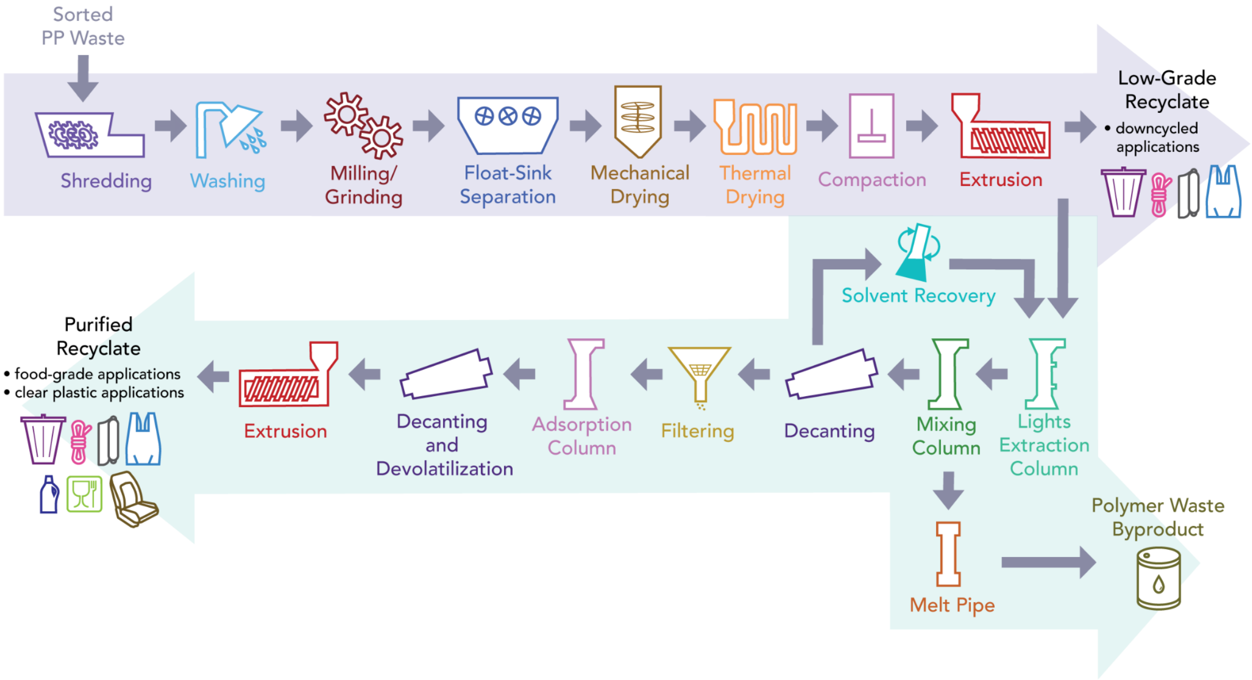

Figure 4.1 Polypropylene recycling with solvent-assisted upgrading
This flow diagram depicts both mechanical and solvent-assisted recycling. The processes highlighted by the purple arrow and ending with low-grade recyclate describe traditional mechanical recycling. The processes highlighted by the green arrows show how solvent-assisted upgrading produces purified recyclate from low-grade recyclate. The upgrading process also produces a waste polymer byproduct stream that can be used at petrochemical refineries. This diagram is adapted from information in academic literature and industry reports [17,22,26,27].

Our results demonstrate that mechanical recycling is 70% less GHG-intensive than solvent-assisted recycling and 80% less GHG-intensive than virgin production on a per-tonne output basis (Figure 4.2). However, mechanically recycled PP will be unsuitable for some applications and/or require blending with virgin material (see Figure C2 and Section C3 in Appendix C for further discussion). The solvent-assisted recycling scenario includes all upstream transportation, sorting, grinding, and extrusion impacts associated with mechanical recycling, with additional emissions associated with the dissolution process itself. Although the solvent-assisted process results in life-cycle GHG emissions nearly triple that of the mechanical recycling footprint, it still represents a 30% savings relative to virgin PP production. The box and whisker plots in Figure 4.2 represent the Monte Carlo simulation results for the recycling scenarios based on parameter probability distributions with asymmetrical triangular distributions using mode, minimum, and maximum values from both literature and our process modeling in SuperPro (Table C5). The probability distributions capture variation in pretreatment energy use, process energy use, process yields, and transportation requirements. The box and whisker plot for mechanical recycling is more offset from the bar graph than may be expected because the bar graph reflects average facility-scale operational data from Franklin Associates (2018)[28] while the probability distributions informing the Monte Carlo simulation are derived from our SuperPro results (Table C5). Even after incorporating uncertainty, mechanical recycling remains substantially less emissions-intensive than solvent-assisted recycling. That said, the potential for process optimization in commercial-scale PP dissolution recycling is not captured by our analysis. Instead, we base our solvent-assisted recycling scenario on information and data reflecting recent, real-world operations still at pilot-scale to provide a conservative estimate of the associated GHG footprint. In contrast, mechanical recycling and virgin production are both mature processes, unlikely to change appreciably in the next decade. The range of life-cycle GHG emissions from virgin PP production is shown with the box-and-whisker plot for that scenario in Figure 4.2.

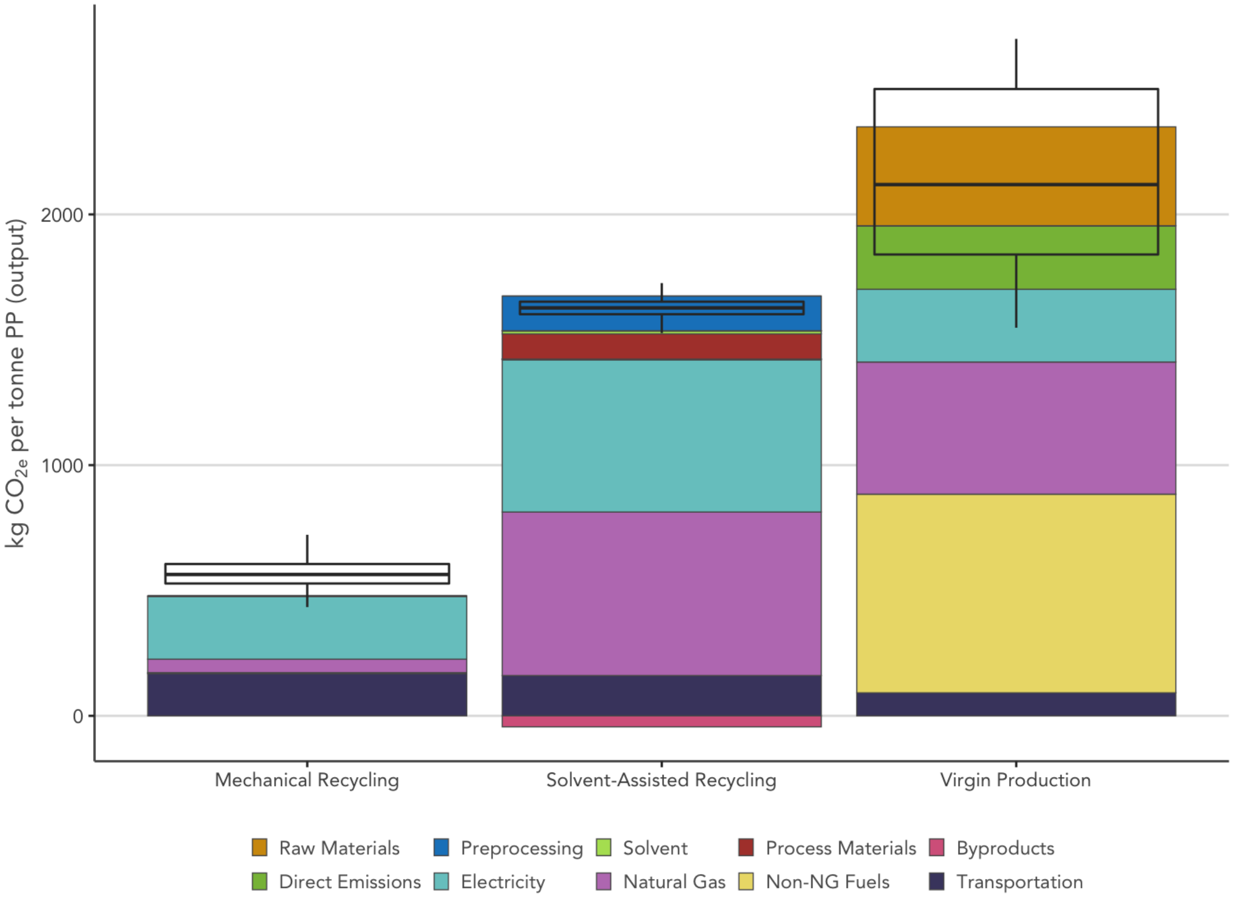

Figure 4.2 Life-cycle greenhouse gas impacts from virgin production and recycling
“Solvent-Assisted Recycling” refers to mechanical recycling with solvent-assisted upgrading. In the case of solvent-assisted recycling, “process” refers to dissolution and extrusion while “preprocessing” includes shredding, washing, grinding, float-sink separation and drying. Process energy consumption is broken down by electricity and thermal energy (from natural gas). GHG impacts from preprocessing energy consumption, both electrical and thermal, is included in the blue area labeled “Preprocessing” along with impacts from cleaning agents. The assumed grid mix is the U.S. average for the recycling cases and the TRE NERC region for virgin production. The box and whisker plots for both recycling scenarios show the Monte Carlo results from a sensitivity analysis varying model parameters using probability distributions based on SuperPro results and literature values. Rather than a parallel sensitivity analysis, the box and whisker plot for virgin production depicts the distribution of estimated life-cycle GHG impacts for this mature process from literature.

The largest contributors to life-cycle GHG emissions for mechanical recycling are electricity consumption and transportation, which suggests that future grid decarbonization and electrification of freight trucks could increase the GHG benefits of recycling relative to virgin PP production. Approximately 45% of the life-cycle GHG emissions associated with solvent-assisted recycling are attributable to electricity consumption during both preprocessing and recycling. Another 40% of these emissions come from natural gas (including upstream and combustion emissions). Electricity consumption for virgin production is about 61% lower than that for solvent-assisted recycling. Impacts from virgin production are instead dominated by the consumption and combustion of petroleum-based fuels which are responsible for nearly 73% of life-cycle GHG emissions. While the results in Figure 4.2 represent a snapshot of how each recycling process and virgin production compare given the current average U.S. energy mix and incoming bale composition, it is important to note that both of these variables are likely to change by location and over time.

4.3.3 Greenhouse Gas Impact from Energy Consumption during Recycling

The GHG impacts from PP production and recycling are primarily driven by energy-consuming processes and, particularly for mechanical recycling, the breakdown of primary fuels vs. electricity can vary (Figure 4.2, Figure 4.3). To better understand how and why the energy use and GHG emissions vary depending on equipment choices and the incoming waste's form factor, we use process modeling to estimate impacts by unit process. Energy-related GHG impacts from operational data (shown in Figure 4.2) are presented in Figure 4.3 alongside modeling results for three mechanical recycling scenarios from SuperPro Designer; the corresponding energy consumption data is provided in Appendix C (Figure C3, Table C1). The first and second scenarios depict mechanical recycling of rigid PP. The first scenario uses electric heating for extrusion while the second uses natural-gas driven heating. The third scenario reflects film PP recycling using electric heating. Because dissolution is a relatively new commercial process, we only have data for a single process configuration and thus do not conduct a similar unit-process-level analysis for solvent-assisted recycling. However, because dissolution requires similar preprocessing and extrusion steps, the results in Figure 4.3 have implications for dissolution recycling as well.

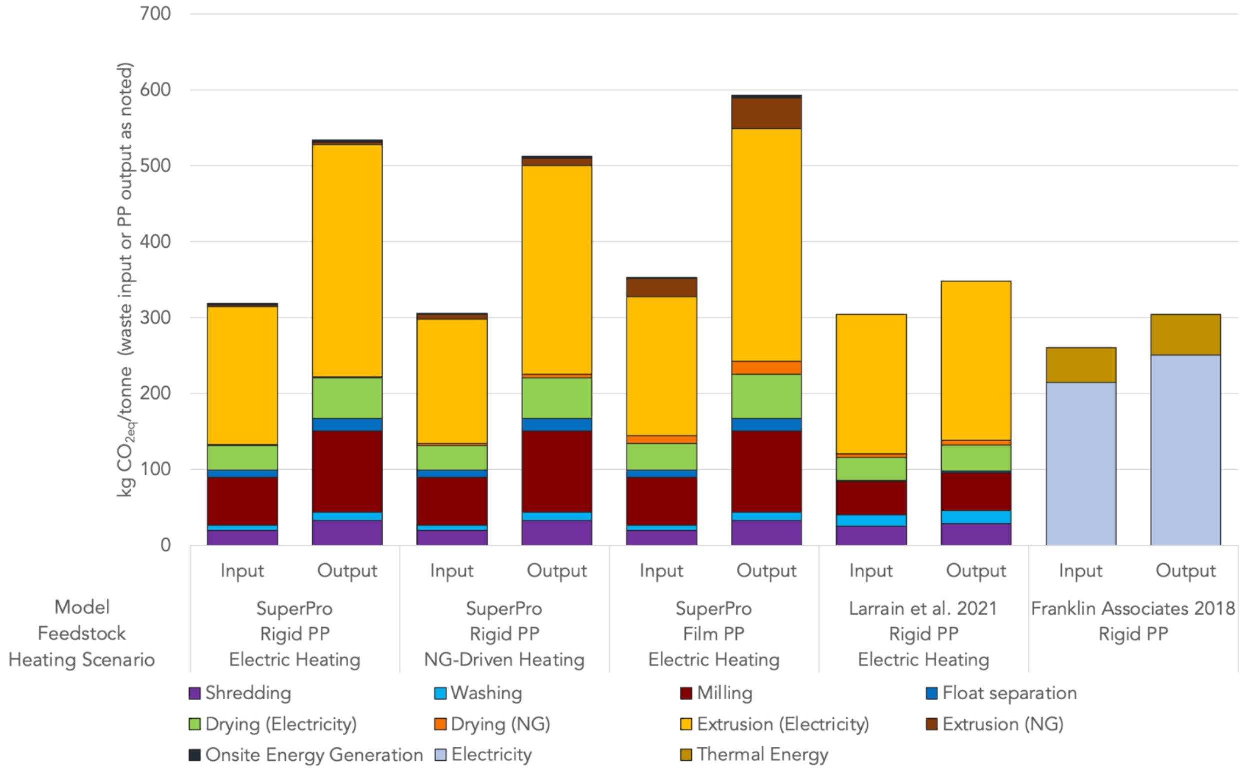

Figure 4.3 Energy-related greenhouse gas emissions by mechanical recycling unit process
The first eight bars (associated with 4 scenarios) show modeling results for facility-scale mechanical recycling operations broken down by unit process from SuperPro Designer and Larrain et al. (2021). The last two bars depicted in this figure reflect the same operational data that was used for Figure 4.2. There is insufficient information to break down this data by unit process so only GHG impact from total electrical and thermal energy consumption is reported.

The SuperPro modeling results for mechanical recycling shown in Figure 4.3 highlight the importance of incoming waste stream purity and suggest that the GHG footprint of mechanical PP recycling in the U.S. may be higher than previously reported values from countries that recover PP separately. A prior study by Larrain et al. (2021) modeled mechanical recycling by unit process for a number of polymer types;[26] their results for rigid PP are plotted alongside our results in Figure 4.3. On a per-tonne *input* basis, our results are similar to those of Larrain et al. and to aggregated average operational data reported by Franklin Associates (2018), but on a per-tonne recyclate *output* basis, our results indicate higher energy needs and GHG emissions for mechanical recycling of rigid PP. For the most part, this can be explained by differences in input composition and therefore, final yield. While Larrain et al. modeled recycling of PP bottle bale, which is over 90% PP and available from sorting facilities in Europe, we model the recycling of a typical mixed #3-7 bale in the U.S., which averages 59% PP. Franklin Associates (2018) does not report the incoming stream composition for their data, but their reported PP yield as a fraction of total incoming material (~85%) indicates that their incoming mix is likely more similar to that of Larrain (>90% PP) than a mixed #3–7 bale.

There are two important distinctions for mechanical recycling: film plastic vs. rigid and electric vs. natural gas-driven heating for extrusion. Recycling PP film requires more thermal energy relative to rigid PP because of the additional energy needed to dry the film plastic after washing and float-sink separation, given its higher surface area-to-volume ratio.

In the case of rigid PP recycling with electric heating, electricity makes up almost 98% of energy consumption while thermal energy from natural gas supplies the remaining 2%. Even when heat for extrusion is supplied by steam generated with natural gas (referred to as natural gas-driven heating), this thermal energy only contributes 8% of the total energy needs with the rest supplied by electricity. Operational data from Franklin Associates reflects the highest contribution from thermal energy to total energy-related GHG impact, close to 19% (Figure 4.3). Our results from SuperPro modeling, along with those from Larrain et al., indicate even fewer GHG impacts from thermal energy. In all rigid PP scenarios, only 1–3% of energy-related GHG impacts are attributable to natural gas or other fuel consumption. In the case of film PP, up to 11%. The dominance of electricity in the overall energy requirements and subsequent emissions impacts for mechanical recycling suggests that the trend toward a decarbonized grid will reduce its GHG footprint over time.

Given the maturity of all the unit processes involved in mechanical recycling, any potential for future energy savings is likely to be small. Extrusion is the most energy intensive, contributing 56-60% to total energy-related GHG impacts. After extrusion, milling and drying are the next most energy intensive processes. In the case of film PP, additional energy, particularly thermal energy for drying and extrusion, is required for mechanical recycling. Compared to the scenario with rigid PP and electric heating, drying and extruding film PP uses about 15% more electricity and almost 13 times as much energy from natural gas on a per tonne input basis.

4.3.4 Forecasting Polypropylene Production and Recycling Emission Factors

Because electricity is a large contributor to life-cycle GHG emissions in all scenarios, the assumption regarding grid mixes and electricity sources has a substantial impact on final results. The results for PP recycling presented in Figure 4.2 reflect the U.S. average grid mix for the recycling processes. We use the Texas Reliability Entity (TRE) North American Electric Reliability Corporation (NERC) region's grid mix for virgin production because petrochemical production is concentrated in that region. Figure 4.4 demonstrates how different and changing grid mixes impact the life-cycle GHG emissions from virgin production, mechanical recycling, and solvent-assisted recycling. We plot results using assumptions based on the average U.S. grid mix and that of California (CA), a state whose grid mix is rapidly decarbonizing.[29] Only the electricity directly consumed by production and recycling facilities is varied (upstream electricity use is not). Forecasts for future carbon intensities of electricity are based on two types of U.S. electricity sector scenarios from the National Renewable Energy Laboratory's Cambium datasets: one standard "mid-case" with average costs assuming current policies and no nascent technology, and one "high renewables" case with low costs for renewable energy assuming nascent technology integration and 95%-decarbonization-by-2050 policy.[30] As the carbon intensity of electricity decreases, the life-cycle GHG impact from PP recycling similarly decreases. In the plotted Cambium cases, GHG emissions from solvent-assisted recycling decrease by 26–40% in 2050 relative to 2022. Virgin production, on the other hand, is less electricity-intensive and the life-cycle GHG emissions from this scenario will not change considerably even if electricity is entirely decarbonized.

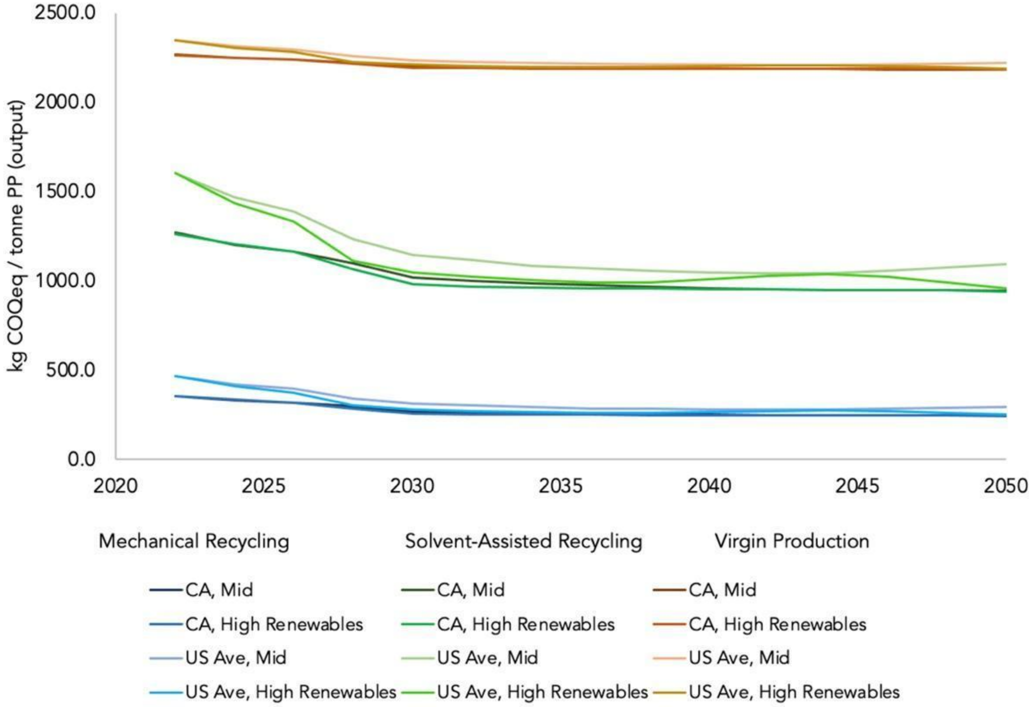

Figure 4.4 Emission factors forecast for polypropylene production and recycling
Sensitivity around electricity emission factor; using Cambium forecasts of grid carbon intensity for CA and the U.S. average for a mid-case scenario (no nascent technology and current policies) and a high renewables case (low renewable energy cost, includes nascent technologies and decarbonization policy with goal of 95% GHG reduction by 2050). For this figure, we assume that electricity for each scenario is coming from the same source and grid mix (i.e. electricity source is not differentiated for virgin production as it was for Figure 4.2). Upstream electricity demand is not updated in these forecasts but contributes less than 5% to the overall GHG footprint.

4.4 Discussion

4.4.1 Enabling Solvent-Assisted PP Recycling

To facilitate the effective integration of solvent-assisted PP recycling in the broader system, new infrastructure plans should consider appropriate feedstock availability and market demand for recycled materials. While PP waste is generated by nearly every community and therefore should be obtainable in most places, high purity, sorted PP waste bales are not widely available from local waste sorting facilities and MRFs. The PP dissolution process does not require pure or contamination-free inputs,[19] but input composition substantially affects the yield of recycled PP output, so sourcing high purity feedstocks will be important for recyclers.

Since pretreatment for dissolution is essentially the same as mechanical recycling, mechanical recyclate is a potentially suitable feedstock for solvent-based reprocessing. It is important to note that PP material undergoes more quality loss during extrusion at a mechanical recycling facility than it does during solvent-assisted extrusion where solvents lower melt viscosity and reduce shearing.[23] Additionally, extrusion is likely to occur on the front-end of any solvent-based process, regardless of previous extrusion, for size control and to enable a continuous process. To avoid needless extrusion and wasted energy, solvent-based recyclers should ideally source non-extruded excess material (flake) from mechanical recyclers. Beyond upgrading surplus recyclate from mechanical recyclers, solvent-based processes can serve as a recycling solution for hard-to-recycle materials (e.g. multilayer plastic packaging) that are not viable feedstocks for mechanical treatment.[31]

4.4.2 Greenhouse Gas Footprint and Other Environmental Impacts

Solvent-assisted recycling is already less GHG-intensive than virgin PP production, by about 30%, and is expected to become increasingly beneficial over time as the electricity generation mix continues to decarbonize. By 2050, dissolution-based PP recycling is expected to emit up to 40% less GHG emissions relative to virgin production based on expected average reductions in the carbon-intensity of the U.S. grid. Mechanical recycling will continue to outperform solvent-assisted recycling on a GHG basis. Furthermore, the market for mechanically recycled PP is underdeveloped and far from reaching its technically-feasible maximum, even after accounting for the range of PP product specifications. Increasing mechanical recycling will continue to yield GHG emission reductions. However, developing the infrastructure necessary to enable advanced processes such as solvent-assisted upgrading will be important for moving beyond a sole focus on downcycling.

This study focuses on the GHG emissions and energy use tied to PP recycling, but scaling up solvent-assisted PP recycling may also lead to other, non-GHG environmental benefits. Most obviously, scaling up any type of recycling process allows for more waste diversion from landfills. Apart from landfill diversion and GHG/energy benefits, which can be provided by mechanical recycling, solvent-assisted recycling, as a more circular technology, can offer virgin production offset credits across metrics. For instance, by enabling circularity and reducing virgin production, enzymatic recycling of PET can reduce smog formation, eutrophication, acidification, ecotoxicity, and human health impacts related to air quality.[32] Using the same logic, we expect some additional sustainability benefits from solvent-assisted PP recycling and reduced virgin PP production beyond reducing GHG emissions and energy use. However, we do not have sufficient data to confidently analyze life-cycle emissions of non-GHG pollutants for the PP dissolution process. Additionally, several non-GHG environmental impacts (e.g. air quality and associated human health impacts) can be very location specific and this analysis is not tied to a particular site. Further research and emissions-related process data is required to confidently quantify other potential benefits.

4.5 Conclusions

As more companies and industry groups pledge to reduce reliance on virgin material, the gap between available recycled material and the required mechanical and aesthetic characteristics required will only become more obvious. Comparing advanced and mechanical recycling processes as competing options suggests a false choice; both are needed to process post-consumer plastics into recycled materials capable of meeting the wide range of quality materials demanded for modern manufacturing. This is particularly true for PP, where solvent-assisted recycling opens up the possibility of using recyclates in food contact materials and other applications that are still solely reliant on virgin material, while still reducing GHG emissions relative to virgin PP production. Furthermore, both mechanical and solvent-assisted recycling will become less carbon-intensive relative to virgin PP as the U.S. electricity mix becomes cleaner and more reliant on renewable energy.

A key barrier to enabling lower-cost, lower-GHG recycling of all types is the level of contamination in plastic waste bales. Pre-processing steps, from milling to drying to extrusion all contribute to higher emissions and lower recycled plastic yields when incoming bales are highly contaminated. As novel recycling processes are developed and tested, incoming material must represent the full range of possible contaminants in real-world waste streams and future energy and mass balances should reflect realistic industry practices. To reduce the burden of additional pre-processing costs and energy penalties, countries seeking to increase recycling rates can invest in waste collection and sorting infrastructure needed to reduce contamination in waste plastic bales and enable the next generation of recycling facilities.

4.6 Methods

Across all recycling scenarios, we assume the initial waste input to the main recycling processes is a mixed #3–7 bale (Figure C1) from a MRF to represent realistic, current conditions. Because the energy footprint of MRFs is relatively small (about 5-8 kWh of electricity per tonne of waste throughput),[33] we assume the MRF energy and GHG emissions attributable to #3–7 bales, which only make up 3.7% of MRF throughput,[10] are negligible in comparison to the more substantial energy and emissions associated with mechanical recycling and upgrading. Section C3.2 of Appendix C includes further discussion on plastic waste sorting. Curbside waste collection is not included in the scope of our analysis; we assume collection and transportation to a waste processing facility occurs regardless of if and how PP is recycled. Because the impact of capital goods is uncertain and variable, we assume the impact to be negligible and exclude them from our analysis.[34,35]

4.6.1 Mechanical Recycling

Mechanical recycling typically involves shredding, washing, milling or grinding, float-sink separation, drying, and extrusion.[26] The process flow highlighted by the purple arrow in Figure 4.1 describes conventional PP mechanical recycling. This process applies to most mechanical recycling of thermoplastics, although individual facilities may vary. Plastic waste entering a recycling facility is first shredded and washed to remove organic and water-soluble impurities. It is then milled for further size reduction before passing through a float-sink separation tank where polymer pieces are separated by density. The target polymer material is mechanically and thermally dried before extrusion, at which point material is heated and forced through a screw extruder. This is typically followed by a pelletizer or some other equipment to cut and shape the extruder output. Extrusion may include melt filtration to remove any remaining contaminants before the recyclate is ready for remanufacturing. There are material losses during mechanical recycling and residual waste polymer can be landfilled or incinerated for energy recovery. This will vary facility-to-facility, but for our analysis, we assume mechanical recyclers do not have on-site incineration and instead send residuals to landfilling. Because plastics take a long time to degrade in landfills, we assume this has a negligible impact on GHG emissions. The output from traditional mechanical recycling is lower grade material that cannot be used for all applications of virgin PP Some studies account for this imperfect substitution between mechanically recycled and virgin PP by using a substitution factor, but we do not apply such a factor because these values are uncertain and product-specific (Figure C2). Detailed discussion on the limitations of mechanical recyclate and on the uncertainty of substitution factors is presented in Section C3.3 of Appendix C.

In addition to using facility-level data on mechanical recycling from literature,[28] we also model shredding, washing, milling, float-sink separation, drying and extrusion in SuperPro Designer to understand the key drivers of energy use and trade-offs for different process configurations. Two studies were ultimately used as benchmarks for mechanical recycling: a report by Franklin and Associates and a study by Larrain et al. (2021).[26,28] Franklin and Associates provide the average material energy balance data from three real-life PP reclaimer facilities, but do not include breakdowns for energy consumption by unit process [28]. Larrain et al. (2021) used a physical based input-output process model to conduct a rigorous techno-economic assessment of mechanical recycling of PP along with polyethylene, polystyrene, and mixed polyolefins. They modeled the same unit processes although in slightly different configurations than what is included here (e.g. Larrain et al. model an additional milling step after thermal drying) and provide energy data by unit process.[26] Across these studies, it is clear extrusion is the main driver of total energy consumption. To capture potential variations in energy inputs, we model two distinct cases for rigid PP extrusion: electric heating and natural gas (NG)-driven steam heating. Both are viable technology options, although electric heating is more common in the industry.[36,37] Conversely, some unit operations consume such a small amount of energy that we have chosen to exclude them. Pelletization (cutting extruder output) is expected to have a negligible impact on the facility's energy demand (<3% of extrusion impact) and the specifics depend on the desired form factor for remanufacturing, so it is excluded from the analysis.[38] Similarly, we assume negligible energy impacts from compaction, which is not technically essential for recycling and depends on facility-specific configurations. If metal contamination is a concern, some facilities may also choose to include a magnet and/or eddy current separator on the front-end of their process. The addition of metal-removing equipment has an energy cost of less than 2 kWh per tonne input,[37] less than any other unit process modeled for mechanical recycling (Table C1). Because we assume low metal contamination (2%) in our initial recycling input, we do not include dedicated metals removal and instead assume these contaminants are removed during float-sink separation.

4.6.2 Solvent-Assisted Upgrading

Dissolution uses solvents to dissolve plastic waste and separate polymer chains from additives, dyes, and other impurities without involving the physical degradation of the original molecules. Selecting solvents depends on the target polymer being recycled and supercritical butane has proved to be an effective solvent for PP dissolution.[15,39,40] Another recent study models dissolution recycling of PP using xylene as a potential solvent and found the life-cycle GHG impact to be 2.2 kg of $CO_{2eq.}$ per kg of recyclate produced, almost 40% higher than our result.[21] The process modeled for our study uses supercritical butane and involves a series of columns for extraction, mixing, filtering and adsorption, and then concludes with decanting and extrusion (Figure 4.1).[27] After being dissolved, the PP solution is purified in the columns, where other contaminants are removed before being precipitated and extruded.[15,27,40] The final output from dissolution is near-virgin quality recycled material. Publicly reported yields for PP recycling via dissolution vary based on original product forms, but are as high as 99% for cups and containers in laboratory settings and as low as 32% for carpet fibers in pilot-scale facility testing.[27,41] Additionally, the dissolution process generates a secondary waste stream of polymer byproduct that can be used as a general hydrocarbon feedstock for the petrochemical industry (Figure 4.1). Unlike mechanical recycling, solvent-based treatments of plastic materials, in isolation, do not noticeably impact the rheological, thermal, or mechanical properties of the polymer.[19,42] Because virgin production and solvent-based recycling produce PP of similar quality, these two production pathways can be directly compared.[42,43]

Important considerations for any advanced recycling process are whether contaminants are allowable and how material must be preprocessed. Early-stage tests are often done carefully-chosen input materials and thus preprocessing requirements are minimal. Real-world facilities must be capable of handling a wide variety of contaminants that may include chlorinated compounds, metals, and flame retardants. Commercial-scale solvent-based recycling requires preprocessing of plastic waste beyond the basic sorting which occurs at an MRF.[27,44,45] As depicted by Figure 4.1, pretreatment to PP dissolution involves washing, grinding, float-sink separation, drying, and extrusion. In other words, solvent-based PP recycling (and likely other advanced recycling processes) occurs after the material is subjected to a series of preprocessing steps that closely resemble the entire mechanical recycling process. Industry interviews with plastic sorting and recycling facilities, along with publicly available reports and data, have confirmed that in commercial operations, extrusion is often used to filter out remaining impurities (through melt-filtration), enable continuous process flows, and to improve subsequent process efficiency.[27,46,47] This is not only true in the case of solvent-based processes, but also for chemical recycling processes like pyrolysis.[23,46,47] Systems already equipped to deal with solvents are particularly attractive hosts for extruders since the addition of solvents during extrusion can lower the melt viscosity, enabling better melt filtration and reducing material degradation.[23] Because of this contrast between what preprocessing is required in a controlled laboratory setting and what is practical in commercial operations, some early-stage studies, including techno-economic analyses and life-cycle assessments, have likely underestimated the energy footprint of advanced recycling by omitting some or all of the preprocessing steps included in our study.[43,48] Rather than framing solvent-based recycling technologies as alternatives to mechanical recycling, they may be more accurately characterized as upgrading options that produce higher-value recycled material.

4.6.3 Life-Cycle Assessment

To conduct the life-cycle assessment, we collected direct mass and energy flow data for each PP-producing/recycling process from process simulation models developed as part of this study and from literature sources. Those mass and energy flows then served as inputs to a physical units-based input-output life-cycle inventory model, Agile-Cradle-to-Grave (Agile-C2G).[49] The combined material and energy balance data for propylene production and conversion to polypropylene comes from literature.[50] For the mechanical recycling scenario, we used data from literature that reflects average material and energy balance data from three real-life PP reclaimer facilities.[28] For the solvent-assisted recycling scenario, we used SuperPro Designer to model mechanical pretreatment processes and used publicly available information supplemented with data from proprietary sources to model facility-scale PP dissolution.[26,27] Because this scenario includes polymer byproducts, which can be used as a hydrocarbon feedstock for other petrochemical processing, we employ system expansion in our analysis and conservatively assume crude oil production is offset by this byproduct stream based on an equivalent higher heating value. In practice, the use of the byproduct stream is uncertain and may end up being landfilled, providing no offset credits. However, changing this assumption does not have a significant impact on results as the credit provided by the byproduct stream is relatively small (Figure 4.2). Film PP is excluded from the life-cycle GHG analysis because film plastics are not currently practical to recover at most MRFs and no data was available on solvent-assisted recycling of PP films. However, we did include a PP film scenario for mechanical recycling to provide a sense for how the energy balance differs relative to rigid PP. We used current and projected grid electricity carbon intensity factors from Cambium, which provides access to annual average emission factors for the National Renewable Energy Laboratory (NREL) Standard Scenarios.[30] We assume the U.S. average grid mix for recycling facilities and the grid mix for the North American Electric Reliability Corporation (NERC) region containing Texas, where petroleum refining and petrochemical production is concentrated, the Texas Reliability Entity (TRE) region. Other relevant emission factors and input-output data are assembled from literature sources, including peer-reviewed articles, GREET, and the Ecoinvent database (Table C4 and Table C5).

To capture recycling process variation, we established probability distributions for key parameters, including efficiencies and energy consumptions, based on previous literature and used these in a Monte Carlo analysis (Table C5). The model was run for 10,000 trials drawing from these distributions to develop the box and whisker plots shown in the results for the recycling scenarios. We do not conduct a similar sensitivity analysis for virgin PP production because the technology is comparatively mature and the values provided here reflect the industry average. Instead, we reviewed recently published estimates for life-cycle GHG impacts of virgin production and presented the distribution of estimates as a box and whisker plot for the virgin PP production scenario (Table C6).

References

(1) Zheng, J.; Suh, S. Strategies to reduce the global carbon footprint of plastics. *Nat. Clim. Chang.* **2019**, *9*, 374–378.

(2) Shen, M.; Huang, W.; Chen, M.; Song, B.; Zeng, G.; Zhang, Y. (Micro)plastic crisis: Un-ignorable contribution to global greenhouse gas emissions and climate change. *J. Clean. Prod.* **2020**, *254*, 120138.

(3) Ahmad, M.; Li, J.-L.; Wang, P.-D.; Hozzein, W. N.; Li, W.-J. Environmental perspectives of microplastic pollution in the aquatic environment: a review. *Mar. Life Sci. Technol.* **2020**.

(4) Enck, J.; Dell, J. Plastic recycling doesn't work and will never work. *The Atlantic*, 2022.

(5) Lam, M. "You can't recycle your way out": California's plastic problem and what we can do about it https://www.kqed.org/news/11901288/you-cant-recycle-your-way-out-californias-plastic-problem-and-what-to-do-about-it (accessed Oct 4, 2022).

(6) Miltimore, J. America finally admits recycling doesn't work

https://catalyst.independent.org/2021/11/23/recycling-doesnt-work/ (accessed Oct 4, 2022).

(7) Di, J.; Reck, B. K.; Miatto, A.; Graedel, T. E. United States plastics: Large flows, short lifetimes, and negligible recycling. *Resources, Conservation and Recycling* **2021**, *167*, 105440.

(8) Ackerman, J.; Levin, D. B. Rethinking plastic recycling: A comparison between North America and Europe. *J. Environ. Manage.* **2023**, *340*, 117859.

(9) De Tandt, E.; Demuytere, C.; Van Asbroeck, E.; Moerman, H.; Mys, N.; Vyncke, G.; Delva, L.; Vermeulen, A.; Ragaert, P.; De Meester, S.; et al. A recycler's perspective on the implications of REACH and food contact material (FCM) regulations for the mechanical recycling of FCM plastics. *Waste Manag.* **2021**, *119*, 315–329.

(10) Smith, R. L.; Takkellapati, S.; Riegerix, R. C. Recycling of plastics in the United States: plastic material flows and polyethylene terephthalate (PET) recycling processes. *ACS Sustain. Chem. Eng.* **2022**, *10*, 2084–2096.

(11) Heller, M. C.; Mazor, M. H.; Keoleian, G. A. Plastics in the US: toward a material flow characterization of production, markets and end of life. *Environmental Research Letters* **2020**, *15*, 094034.

(12) EPA. *Advancing Sustainable Materials Management: 2018 Facts and Figures Sheet*; U.S. Environmental Protection Agency, 2020.

(13) ACC. *Resin review: the annual statistical report of the North American plastics industry*; American Chemistry Council, 2020.

(14) Eriksen, M. K.; Christiansen, J. D.; Daugaard, A. E.; Astrup, T. F. Closing the loop for PET, PE and PP waste from households: Influence of material properties and product design for plastic recycling. *Waste Manag.* **2019**, *96*, 75–85.

(15) Vollmer, I.; Jenks, M. J. F.; Roelands, M. C. P.; White, R. J.; van Harmelen, T.; de Wild, P.; van der Laan, G. P.; Meirer, F.; Keurentjes, J. T. F.; Weckhuysen, B. M. Beyond mechanical recycling: giving new life to plastic waste. *Angew. Chem. Int. Ed* **2020**, *59*, 15402–15423.

(16) Miao, Y.; von Jouanne, A.; Yokochi, A. Current technologies in depolymerization process and the road ahead. *Polymers (Basel)* **2021**, *13*.

(17) Ragaert, K.; Delva, L.; Van Geem, K. Mechanical and chemical recycling of solid plastic waste. *Waste Manag.* **2017**, *69*, 24–58.

(18) Dolan, M.; Vandebeek, J. *Advances in Depolymerization Technologies for Recycling*; NexantECA, 2021.

(19) Li, H.; Aguirre-Villegas, H. A.; Allen, R. D.; Bai, X.; Benson, C. H.; Beckham, G. T.; Bradshaw, S. L.; Brown, J. L.; Brown, R. C.; Sanchez Castillo, M. A.; et al. Expanding plastics recycling technologies: chemical aspects, technology status and challenges. **2022**.

(20) Collias, D. I.; James, M. I.; Layman, J. M. Chapter 3. Solvent-Based Recycling. In *Circular economy of polymers: Topics in recycling technologies*; American Chemical Society: Washington, DC, 2021; pp. 33–59.

(21) Uekert, T.; Singh, A.; DesVeaux, J. S.; Ghosh, T.; Bhatt, A.; Yadav, G.; Afzal, S.; Walzberg, J.; Knauer, K. M.; Nicholson, S. R.; et al. Technical, economic, and environmental comparison of closed-loop recycling technologies for common plastics. *ACS Sustain. Chem. Eng.* **2023**, *11*, 965–978.

(22) Meys, R.; Frick, F.; Westhues, S.; Sternberg, A.; Klankermayer, J.; Bardow, A. Towards a circular economy for plastic packaging wastes – the environmental potential of chemical

recycling. *Resources, Conservation and Recycling* **2020**, *162*, 105010.
(23) Zolghadr, A.; Foroozandehfar, A.; Kulas, D. G.; Shonnard, D. Study of the viscosity and thermal characteristics of polyolefins/solvent mixtures: applications for plastic pyrolysis. *ACS Omega* **2021**, *6*, 32832–32840.
(24) G.A.A. Inc. *2016-2017 Database on material recovery facilities and mixed waste processing facilities in the U.S., with updates*; Governmental Advisory Associates, Inc., 2019.
(25) Ma, J.; Tominac, P. A.; Aguirre-Villegas, H. A.; Olafasakin, O. O.; Wright, M. M.; Benson, C. H.; Huber, G. W.; Zavala, V. M. Economic evaluation of infrastructures for thermochemical upcycling of post-consumer plastic waste. *Green Chem.* **2022**.
(26) Larrain, M.; Van Passel, S.; Thomassen, G.; Van Gorp, B.; Nhu, T. T.; Huysveld, S.; Van Geem, K. M.; De Meester, S.; Billen, P. Techno-economic assessment of mechanical recycling of challenging post-consumer plastic packaging waste. *Resources, Conservation and Recycling* **2021**, *170*, 105607.
(27) Leidos. *PureCycle Ohio Phase II Facility Independent Engineer's Report*; U.S. Securities and Exchange Commission, 2020.
(28) Franklin Associates. *Life cycle impacts for postconsumer recycled resins: PET, HDPE and PP*; The Association of Plastic Recyclers, 2018.
(29) Mazmanian, D. A.; Jurewitz, J. L.; Nelson, H. T. State leadership in U.S. climate change and energy policy: the California experience. *The Journal of Environment & Development* **2020**, *29*, 51–74.
(30) Gagnon, P.; Frazier, W.; Cole, W.; Hale, E. *Cambium documentation: version 2021*; National Renewable Energy Laboratory (NREL): Golden, CO (United States), 2021.
(31) Walker, T. W.; Frelka, N.; Shen, Z.; Chew, A. K.; Banick, J.; Grey, S.; Kim, M. S.; Dumesic, J. A.; Van Lehn, R. C.; Huber, G. W. Recycling of multilayer plastic packaging materials by solvent-targeted recovery and precipitation. *Sci. Adv.* **2020**, *6*.
(32) Singh, A.; Rorrer, N. A.; Nicholson, S. R.; Erickson, E.; DesVeaux, J. S.; Avelino, A. F. T.; Lamers, P.; Bhatt, A.; Zhang, Y.; Avery, G.; et al. Techno-economic, life-cycle, and socioeconomic impact analysis of enzymatic recycling of poly(ethylene terephthalate). *Joule* **2021**.
(33) Pressley, P. N.; Levis, J. W.; Damgaard, A.; Barlaz, M. A.; DeCarolis, J. F. Analysis of material recovery facilities for use in life-cycle assessment. *Waste Manag.* **2015**, *35*, 307–317.
(34) Astrup, T. F.; Tonini, D.; Turconi, R.; Boldrin, A. Life cycle assessment of thermal waste-to-energy technologies: review and recommendations. *Waste Manag.* **2015**, *37*, 104–115.
(35) Brogaard, L. K.; Riber, C.; Christensen, T. H. Quantifying capital goods for waste incineration. *Waste Manag.* **2013**, *33*, 1390–1396.
(36) Rauwendaal, Chris. *Polymer extrusion*; 4th ed.; Hanser Gardner Publications: Cincinnati, OH, 2001.
(37) Brooks, T. Bulk Handling Systems: MRF Discussion , 2022.
(38) Martín-Lara, M. A.; Moreno, J. A.; Garcia-Garcia, G.; Arjandas, S.; Calero, M. Life cycle assessment of mechanical recycling of post-consumer polyethylene flexible films based on a real case in Spain. *J. Clean. Prod.* **2022**, *365*, 132625.
(39) Alex Tullo. Europe is implementing a tax on plastic. *C&EN Global Enterp.* **2021**, *99*, 34–34.

(40) Layman, J. M.; Gunnerson, M.; Bond, E. B.; Nelter, A. E.; Hosmer, J. E. Articles of reclaimed polypropylene compositions. WO2017003802A1, May 1, 2017.

(41) Achilias, D. S.; Giannoulis, A.; Papageorgiou, G. Z. Recycling of polymers from plastic packaging materials using the dissolution–reprecipitation technique. *Polym. Bull.* **2009**, *63*, 449–465.

(42) Poulakis, J. G.; Papaspyrides, C. D. Recycling of polypropylene by the dissolution/reprecipitation technique: I. A model study. *Resources, Conservation and Recycling* **1997**, *20*, 31–41.

(43) Zhao, Y.-B.; Lv, X.-D.; Ni, H.-G. Solvent-based separation and recycling of waste plastics: A review. *Chemosphere* **2018**, *209*, 707–720.

(44) Tsampanakis, I.; Orbaek White, A. The mechanics of forming ideal polymer-solvent combinations for open-loop chemical recycling of solvents and plastics. *Polymers (Basel)* **2021**, *14*.

(45) Triebert, D.; Hanel, H.; Bundt, M.; Wohnig, K. Solvent-Based Recycling. In *Circular economy of polymers: topics in recycling technologies*; Collias, D. I.; James, M. I.; Layman, J. M., Eds.; ACS Symposium Series; American Chemical Society: Washington, DC, 2021; Vol. 1391, pp. 33–59.

(46) Pohjakallio, M.; Vuorinen, T.; Oasmaa, A. Chemical routes for recycling—dissolving, catalytic, and thermochemical technologies. In *Plastic waste and recycling*; Letcher, T. M., Ed.; Elsevier, 2020; pp. 359–384.

(47) Thiounn, T.; Smith, R. C. Advances and approaches for chemical recycling of plastic waste. *J. Polym. Sci.* **2020**, *58*, 1347–1364.

(48) Schwarz, A. E.; Ligthart, T. N.; Godoi Bizarro, D.; De Wild, P.; Vreugdenhil, B.; van Harmelen, T. Plastic recycling in a circular economy; determining environmental performance through an LCA matrix model approach. *Waste Manag.* **2021**, *121*, 331–342.

(49) Nordahl, S. L.; Devkota, J. P.; Amirebrahimi, J.; Smith, S. J.; Breunig, H. M.; Preble, C. V.; Satchwell, A. J.; Jin, L.; Brown, N. J.; Kirchstetter, T. W.; et al. Life-cycle greenhouse gas emissions and human health trade-offs of organic waste management strategies. *Environ. Sci. Technol.* **2020**, *54*, 9200–9209.

(50) Franklin Associates. *Cradle-to-grave life cycle inventory of nine plastic resins and four polyurethane precursors*; Plastics Division of the American Chemistry Council, 2011.

Chapter 5

Conclusions

5.1 Summary of Major Findings

Landfill diversion and sustainable waste management begin with effective municipal solid waste (MSW) sorting. Because more sustainable alternatives to landfilling are material-specific, MSW must be separated into distinct waste streams to maintain the effectiveness and efficiency of downstream waste treatment processes. Organic and plastic wastes, major contributors to the overall composition of MSW, are two such materials that require dedicated processing to improve environmental performance. Proper management of these waste types outside of a landfill can result in significant environmental benefits: avoided waste accumulation, greenhouse gas (GHG) emissions reduction, renewable energy production, and improved circularity.

5.1.1 Organic Waste Management

For organic waste, the most industry relevant alternatives to landfilling are composting and anaerobic digestion (AD). Chapter 2 clearly demonstrates that either of these alternatives, regardless of specific scenario configurations, provides significant GHG savings (over 50% reductions) relative to landfilling. Composting and AD both emit fewer GHGs than landfilling but from a life-cycle perspective, these processes are even more favorable because they can generate other offsets and avoid emissions outside of the system boundary. For composting, this means offsetting synthetic fertilizer use and application. Because AD systems also generate digestate, biosolid material that can be composted or applied to land, they similarly offset synthetic fertilizers. However, the more significant offset for AD systems comes from energy generation either in the form of electricity or renewable natural gas (RNG). Chapter 2 offers life-cycle results for a variety of organic waste management scenarios assuming California conditions and finds that the least GHG-intensive options for treating organic waste are direct composting and AD with digestate composting and RNG production to offset diesel emissions. On a per-tonne organic waste managed basis, both of these scenarios have net-negative GHG emissions. In areas where electricity is more carbon-intensive than California, the scenario with AD and electricity production may also have a net-negative GHG footprint.

The environmental favorability between organic waste management options becomes less obvious when taking into consideration life-cycle air quality and associated human health impacts. In addition to GHG footprints, Chapter 2 offers life-cycle assessments (LCAs) with respect to NO_x, NH_3, SO_2, VOCs, and $PM_{2.5}$ emissions. Life-cycle air pollutant emissions are aggregated and presented in terms of human health damages or social cost. These results show that while GHGs are the primary emissions issue associated with landfilling, NH_3 emissions are the largest contributor to total social costs, including the social cost of carbon emissions, from any organic waste management system involving composting. This is to say, from a social cost and human health perspective, composting may not be the most environmentally favorable option for organic waste even relative to landfilling. This finding brings attention to the importance of air quality impacts from organic waste management which have been understudied relative to GHG impacts.

Even though composting plays a key role in strategies to reduce GHG emissions from organic waste and contributes to air pollution, the emissions from composting are not particularly well-understood. Composting emissions are driven by microbial processes that are nuanced and complex. Many studies in current literature measure the emissions from composting different types of organic waste feedstocks under varied conditions, but there is no consensus for predicting the emissions impact from a particular composting operation. Chapter 3 provides a comprehensive and quantitative review of existing CH_4, N_2O, biogenic CO_2, NH_3 and VOC emission factors disaggregated by feedstock type. With regards to GHG impact, the data shows that for most organic wastes, N_2O is the dominant contributor to the 100-year global warming potential of direct emissions from composting assuming properly aerated operations. While CH_4 emissions can be managed with increased aeration, these N_2O emissions may be harder to control. One possible solution may be AD treatment prior to composting. Chapter 3 shows evidence of consistent reductions to N_2O emissions from composting when the starting material undergoes AD prior to composting. This chapter also discusses NH_3 and VOC emissions from composting, but found little evidence to suggest that any of the observed operational practices, including aeration and AD pretreatment, can consistently reduce these emissions.

5.1.2 Plastic Waste Management

In the case of plastic wastes, the most sustainable alternative to landfilling is recycling. There are several forms of recycling but there are two main categories for polymer-to-polymer recycling: mechanical and solvent-assisted. Mechanical recycling cleans, shreds, and re-extrudes plastic waste to create recyclate that is typically of lower quality than virgin resin, but can still be used for a number of plastic applications. Solvent-assisted recycling uses depolymerization or dissolution to produce near-virgin quality recyclate. Chapter 4 provides a detailed LCA of both recycling options for polypropylene (PP), one of the most widely produced and lowest-cost plastics. In the case of PP, solvent-assisted recycling implies dissolution-based recycling. Findings show mechanical recycling is 80% less GHG intensive than virgin production while solvent-assisted recycling achieves 30% GHG savings. This suggests that until the technically feasible market limit for lower-grade mechanical recycling is met, expanding mechanical recycling will continue to provide substantial GHG benefits. Solvent-assisted or dissolution-based recycling is an effective GHG solution for upgrading surplus mechanical recyclate to near-virgin quality PP recyclate to accommodate a wider array of plastic applications. Furthermore, both recycling technologies are electricity-intensive and their life-cycle GHG impact is dependent on the carbon intensity of electricity production. While virgin PP production is not expected to become less emissions-intensive overtime, impacts from recycling should decrease with grid decarbonization. While this particular study focuses on PP, the broader conclusions regarding the roles of mechanical and solvent-assisted recycling are likely generalizable to most thermoplastics.

5.2 Limitations

The most obvious limitation of the research in this dissertation is a limitation of the methodology used. LCA is a common approach used to quantify the environmental benefits and tradeoffs of waste management strategies. If done well, LCA can inform the development and implementation of sustainable processes, guiding decision-makers toward approaches that achieve the greatest environmental benefits. However, LCAs are not verifiable and can be skewed by lack of reliable data. The LCAs presented in this dissertation use Monte Carlo simulations to provide sensitivity analysis of results, but inherent uncertainty in underlying data collected from literature makes it hard to accurately quantify error bars that reflect real-world variation of environmental impacts for a given system.

Another limitation of the work presented here has to do with environmental metrics considered. While gaseous emissions are the most often discussed environmental impacts from organic and plastic waste management, they are not the only negative environmental impacts from these systems. For instance, organic waste systems can have liquid emissions and significant eutrophication impacts while plastic waste systems can lead to microplastic pollution. Expanding the list of environmental metrics considered would provide a more comprehensive understanding of life-cycle environmental impacts from these systems.

5.3 Policy Implications

LCA studies have obvious policy implications as they compare the environmental impacts of various sustainable strategies and identify the least impactful option for decision-makers. With regards to organic waste, the research in this dissertation recommends management with either direct composting or AD with digestate composting and RNG production to reduce GHG emissions. In population-dense areas, where human exposure to air pollution is high, it may be more appropriate to avoid composting and associated NH_3 emissions by treating organic waste with AD and either landfilling or land applying digestate. While still avoiding landfilling, this strategy reduces the burden from air pollution and associated social costs. With regards to plastic waste, this dissertation recommends developing better infrastructure for plastic waste sorting and recovery, maximizing mechanical recycling whenever possible, and using solvent-assisted recycling to upgrade any surplus or hard-to-recycle plastic wastes. By enabling more circularity with this strategy, GHG emissions and fossil fuel use from plastic supply chains can be reduced.

Not only do the results presented in this dissertation suggest solutions for waste management policy, but they also have implications for bioenergy, bioproduct and novel plastic industries. The findings on organic waste show that current GHG-saving alternatives to landfilling still have substantial environmental impacts and suggest that developing the use of organic waste as feedstocks to bioenergy or bioproduct systems may prove beneficial if they can avoid composting. The findings on plastic waste show the importance of sorting and separation. This has implications for the development of new plastics, particularly compostable plastics that may be hard to selectively separate and route for composting. Without considering end-of-life management, designers of new plastics may end up contaminating existing organic and plastic waste systems. By highlighting the various obstacles to effective end-of-life management for organic and plastic wastes, the research in this dissertation can inform sustainable materials design and production.

5.4 Recommendations for Future Research

Waste management is an evolving industry and potential solutions are constantly being proposed. As novel materials and waste treatment processes are developed, life-cycle research must also be conducted to understand how these technologies operate in real world conditions at commercial scales. Therefore, there are constantly new opportunities for LCA research. However, it is imperative that future LCA research in this area is informed by operational data, industry activity, and/or rigorous process modeling rather than being solely based on lab-scale work. In this section, I will discuss specific examples for future research endeavors related to plastic and organic waste management.

With regards to research on organic waste, I recommend future research further investigate the environmental impacts of composting. As shown in Chapter 2, composting appears to be key to carbon-saving solutions for organic waste management. However, as shown in Chapter 3, there is substantial variability and uncertainty associated with gaseous emissions. There is also uncertainty associated with the soil amendment and carbon-saving benefits of compost application. I recommend future research assesses the current state of literature on composting benefits and provide quantitative recommendations for how to credit composting more accurately in LCAs. Future LCA research should explore specific options for designing composting operations that are location and feedstock specific. Because composting and associated ammonia emissions have high social costs, it is imperative that future LCA work considers air pollution with human health impacts in addition to GHGs. GHG and ammonia emissions from composting present continued opportunities for future experimental and measurement studies. While there are many studies in this research area, there are still no accurate, mechanistic models for predicting emissions from composting piles. Any research that could contribute to building such a model would be incredibly valuable.

There is also a particular need for research on the impact of plastics on composting. For instance, future work could explore plastic contamination in composting operations. There are few available estimates for how much non-degradable plastic waste ends up in composting operations, how it affects the quality and use of compost, and to what extent microplastics are a concern. Similarly, there are few studies characterizing the breakdown of compostable bioplastics in commercial-scale operations and the associated environmental impacts. In recent years, there have been continued developments in the design of biodegradable plastics, but the potential performance and system-wide impacts of newer plastics in the current waste management system have yet to be explored. Future LCA research should consider the environmental performance of compostable plastics in various existing and hypothetical future end-of-life pathways. While typical research of degradable plastics may look at degradation efficiency and compost quality, few studies have explored how these materials impact composting emissions. I recommend future research to identify promising developments in biodegradable plastic design and investigate the potential impact of these materials on existing organic waste management systems. This could include experimental and modeling work to understand how existing systems would respond to the expansion of biodegradable plastics production and associated change in composition of waste streams.

Multi-input, multi-output biorefineries offer another opportunity for future research that connects organic and plastic materials. Biorefineries can use organic waste feedstocks to create energy or other useful products like bioplastics. More LCA research is needed to understand the environmental tradeoffs between various biorefinery configurations under different conditions. In particular, little to no research has been conducted on biorefineries with respect to non-GHG air pollution. As identified by the LCA in Chapter 2, air pollution from systems using organic waste can have substantial human health costs and so understanding the air quality impacts of multi-input biorefineries is essential for equitable, sustainable development of the bioenergy and bioproduct industries. Contributing to this understanding is my next research goal. In my next study, I will provide a comprehensive LCA of multiple corn stover-to-ethanol biorefinery designs with respect to local social cost, air pollution with human health impacts, and GHG emissions.

Regarding conventional, non-degradable plastics, future research can build off of the work presented in Chapter 4 which provides a LCA of advanced, solvent-based polypropylene recycling. While research of new bioplastics is important for sustainable development, we still need robust understanding of how to best manage conventional materials, like petrol-based plastics, that still dominate waste streams. I recommend that future studies conduct similar work to Chapter 4 but looking at other types of plastic resins for which there are viable, advanced recycling pathways. For instance, I recommend future research focus on polystyrene (plastic resin code #6) which is rarely mechanically recycled, often landfilled, and could be relatively easily recycled via dissolution. Alternatively, future LCA work could investigate depolymerization as a recycling technology for viable plastic types (e.g. polyamides, polyurethanes, or polystyrene) and explore the circularity benefits relative to mechanical recycling. Besides evaluating the environmental implications of novel recycling options, future research could focus on plastic waste management infrastructure in the United States and use LCA in combination with techno-economic analysis to identify where and to what extent plastic sorting infrastructure is needed to enable advanced recycling. Achieving sustainable plastic circularity is no small feat and substantial research is still needed in this area to inform future progress.

Appendix A

Supplementary Information

Life-Cycle Greenhouse Gas Emissions and Human Health Tradeoffs of Organic Waste Management Strategies

A1 LCA Process Calculations and Assumptions

A1.1 Feedstock Characterization

The system boundary of our model is limited to waste that is anaerobically digested at ZWEDC. We approximate the organic waste materials relevant in this study as food waste, taking into consideration visual appearance, the origin of the waste (e.g. company cafeterias, local grocery stores, and restaurants), waste classification on delivery logs, and the impact of on-site sorting. The inorganic or non-digestable materials (such as large tree branches) present in the incoming hauls to ZWEDC are removed from the waste stream during sorting at ZWEDC and are not included in the scope of this study. The materials that are actually fed into the digesters, and are therefore included within the scope of this study, visually appeared to be mostly food waste during multiple visits to the facility (Figure A1 shows an example). Incoming waste to ZWEDC consists of commercial food waste from San Jose and Palo Alto (e.g. from grocery stores or cafeterias), a relatively small portion of green waste from Palo Alto (mostly yard trimmings – some of which is hard to digest and therefore sorted out and excluded from this study's scope), and sorted organics from waste processing/sorting facilities (mostly consisting of food waste as paper and non-compostable yard waste materials are sorted separately).

A1.2 Trucking

For all AD scenarios, based on ZWEDC delivery logs, we modeled monthly average totals of 5,142 tonnes of commercial waste from Republic (a major municipal waste collection company), including 891 tonnes of processed organics from the SMaRT Station (a waste processing/sorting facility in Sunnyvale, CA), 1,170 tonnes of commercial waste from locations around Palo Alto, 1,040 tonnes of yard waste from Palo Alto, and 35 tonnes from other sources (Figure A3). Aside from a small portion, which has sufficiently small particle sizes to enter the digesters, yard waste is sent directly to be composted after arriving at ZWEDC. The scope of our model is limited to waste that is received by ZWEDC and then fed into the digesters. Every month, the ZWEDC facility produces outbound loads averaging 1,840 tonnes of residuals consisting of 1,640 tonnes of trash to Newby Island Landfill, 184 tonnes of trash to the Marina landfill, and 12 tonnes of recyclables sent to the Newby Island Resource Recovery Park. The transportation emissions associated with these loads are excluded from our analysis.

We assume average distances traveled to the compost facility (Z-Best), the landfill (Newby Island Landfill) and a Gilroy farm (for land application) of 45 miles, 8 miles and 32 miles respectively. In the scenario involving land application of digestate, we assume digestate is landfilled in the winter from October to April and land applied as fertilizer at a farm otherwise. We assume all transportation is provided by a fleet of flatbed trucks.

A1.3 Landfilling

We use a food waste specific emission factor for landfilling "mixed organics" from Behera et al.[1] Because we do not have we do not have reliable emission factors for specifically landfilling digestate, we apply the same emission factor for landfilling to digestate but scaled down by the volatile solids (VS) reduction resulting from anaerobic digestion. The VS reduction was assumed to be about 80%.[2]

A1.4 Composting

We assume a mass reduction of 33% during composting. We assume that compost application offsets urea and that the total nitrogen in compost is 1.7 %,[3] while the total nitrogen in urea is 46% .[4] We used measured emission factors for composting digestate and used emission factors for "mixed organics" from literature for composting the original waste feedstocks (without digestion).

A1.5 Digestate Land Application

We assume an 80% mass reduction during dewatering of digestate before land application.[5] As with the compost scenarios, we assume digestate application offsets urea use.

A1.6 Electricity Consumption

We assume all direct electricity consumption by facilities would be provided by a natural gas combined cycle power plant. In the RNG scenarios, biogas must be upgraded to RNG so there is an additional energy cost. Upgrading raw biogas requires 0.32 kWh/m^3.[6]

A1.7 Electricity Generation

We assume electricity generation by facility offsets electricity provided by a natural gas combined cycle power plant.

A1.8 CHP Operation

We used facility data and measured emission factors to calculate emissions from this stage.

A1.9 RNG Offsets

In the scenario where biogas is upgraded for truck fleet fueling, we assume upgraded RNG offsets diesel combustion. In the scenario where biogas is upgraded for pipeline injection, assume RNG offsets natural gas combustion.

A1.10 Flaring

We have data for emissions from flaring from the facility which is included in all scenarios with anaerobic digestion. However, we assume that in scenarios where biogas is upgraded into RNG, that flaring would be reduced by 50%.

A1.11 Venting and Methane Loss

We use a total venting factor of 0.1% as a basic assumption. We further assume that most venting comes from the bladder so total venting is accounted for with two subfactors: biofilter venting factor of 0.075% and a bladder venting factor of 0.025%.

We apply these factors to facility measurements of biogas production. We assume the biogas feedstock composition is equivalent to venting emissions. Carbon dioxide emissions during venting are excluded because they are assumed to be biogenic. We assume that there is 50% less bladder venting in the RNG scenarios.

We assume a loss of 0.65% to the atmosphere during biogas upgrading for RNG scenarios.[7] We assume the methane content of upgraded RNG is 95%.[8]

A2 Facility Data

Provided/measured data for ZWEDC facility:

- Biogas consumption (daily CHP data) (Figure A5)
- Daily inbound and outbound tonnage (Figure A3, Figure A4)
- Inbound and outbound locations (Figure A3, Figure A4)
- Daily biogas flaring
- Electricity generated (Figure A6)

We use an average ratio between consumption and power production after April 2016 to estimate missing biogas consumption data prior. Data is available upon request to the corresponding author.

Figure A1. Image of feedstock to digesters at ZWEDC

Figure A2. Aerial view of ZWEDC facility (photo credit: Zero Waste Energy Development Company)

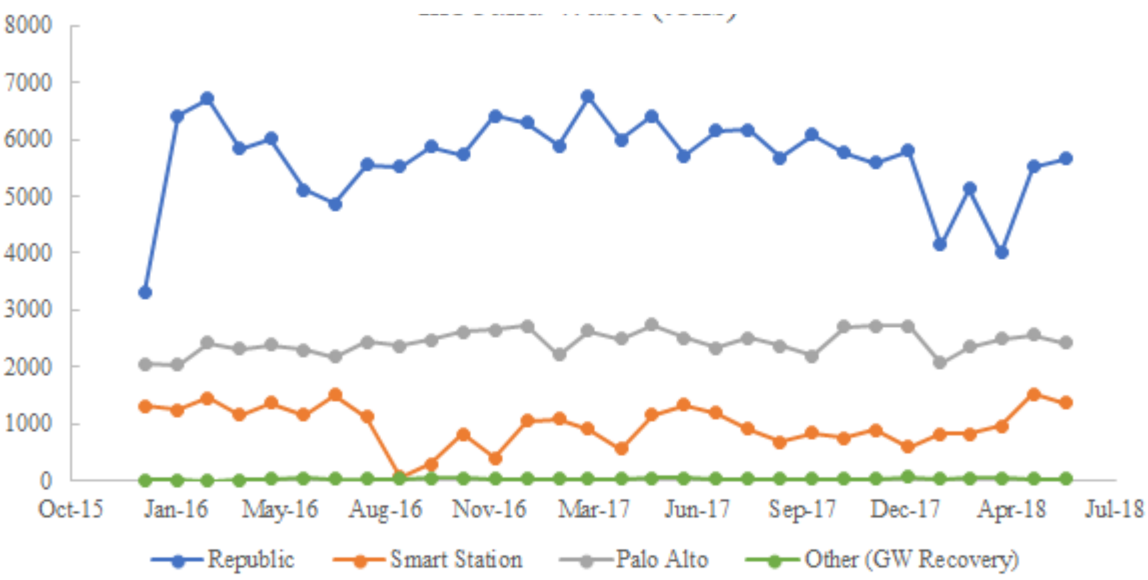

Figure A3. Inbound waste at ZWEDC facility from various sources

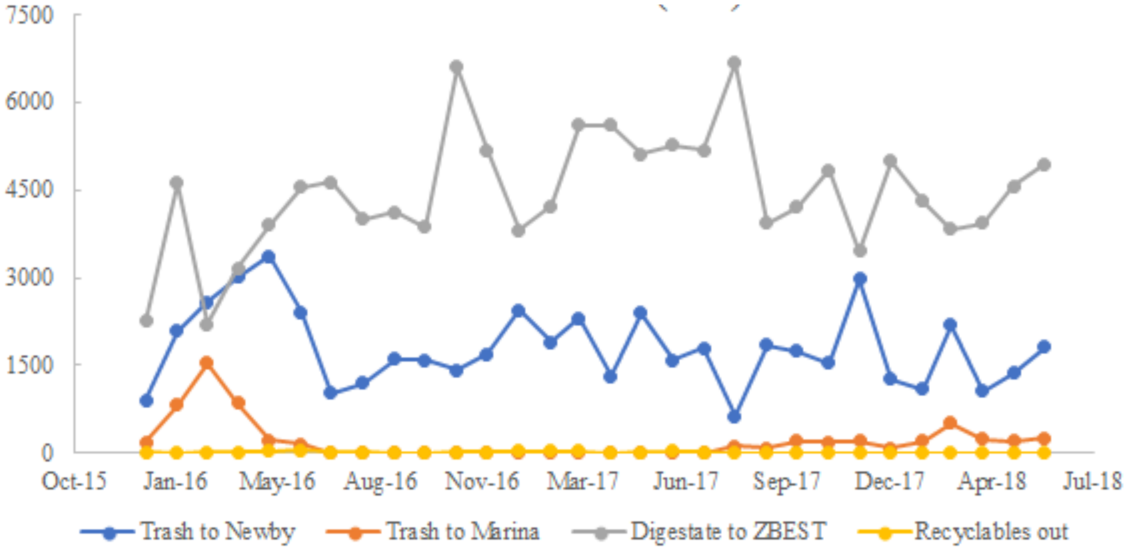

Figure A4. Outbound waste from ZWEDC facility in wet tonnes

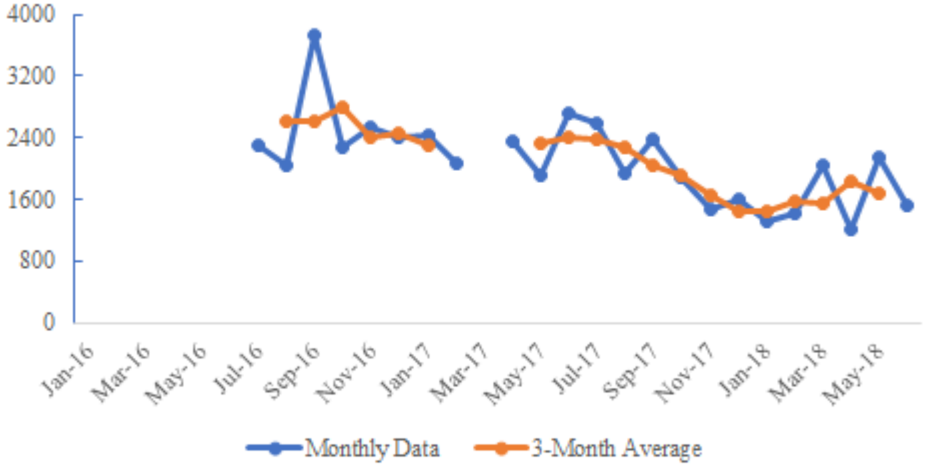

Figure A5. Biogas yield in cubic feet per wet tonne of waste in digesters

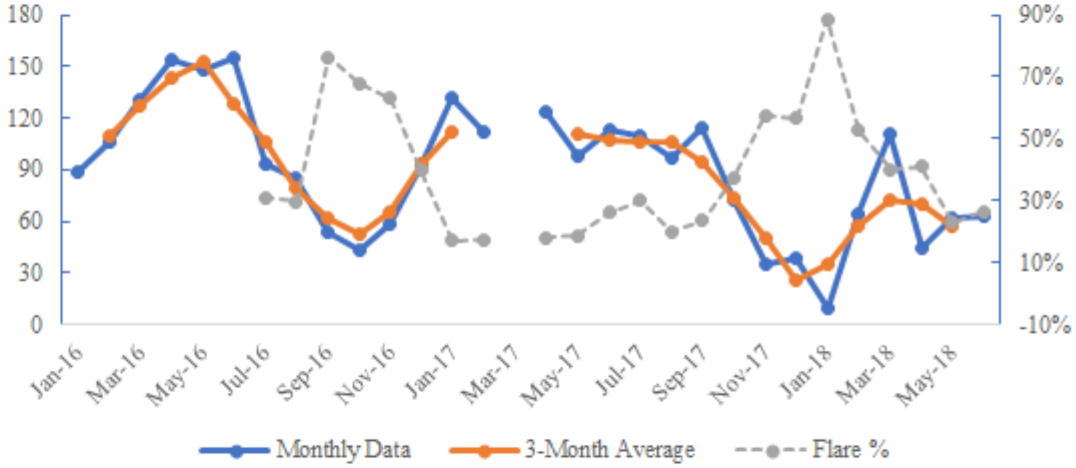

Figure A6. Electricity yield (kWh) per wet tonne of waste in digesters

A3 Input Output Data and Emission Factors

Figure A7 shows detailed operations at the ZWEDC facility.

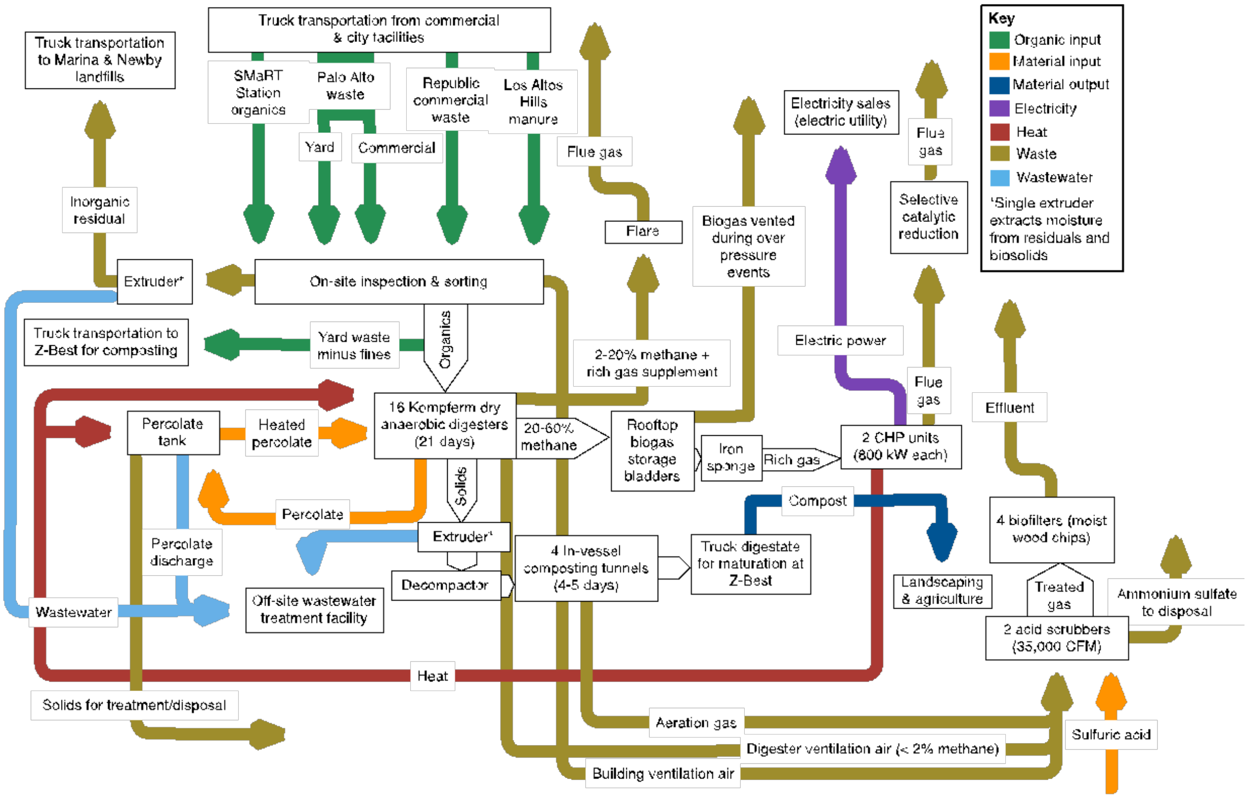

Figure A7. Zero Waste Energy Development Company (ZWEDC) dry anaerobic digestion process

Table A1 lists all of the emission factors used for each unit process/product involved in the scenario lifecycles.

Table A1. Emission factors (in kg of given emission type per unit listed in parameter name)

Parameters	CO_2	CH_4	N_2O	CO	NO_x	$PM_{2.5}$	SO_2	VOC	NH_3	Sources and Assumptions
atrazine.kg	8.6E+00	3.9E-05	3.2E-05	2.0E-03	2.9E-02	7.8E-04	2.3E-04	0.0E+00	0.0E+00	GREET [9]
atrazine_brazil.kg	8.6E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
insecticide.kg	1.2E+01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
insecticide_brazil.kg	1.2E+01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
cellulase.kg	1.1E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
alpha_amylase.kg	2.5E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
gluco_amylase.kg	6.3E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
lime.kg	9.4E-01	0.0E+00	6.0E-07	6.0E-05	2.3E-03	1.2E-05	5.6E-05	0.0E+00	0.0E+00	Assumed slaked lime at a biorefinery; Process emissions[10]; Combustion emissions calculated based on 80% heat conversion efficiency, assuming natural gas as a fuel and using energy consumption from Research Triangle Institute[11]
caco3.kg	1.6E-01	0.0E+00	1.1E-06	4.3E-05	6.5E-04	1.0E-05	2.9E-07	0.0E+00	0.0E+00	GREET [9]
h2so4.kg	0.0E+00	0.0E+00	4.6E-08	1.7E-06	5.0E-06	1.6E-07	2.0E-03	0.0E+00	0.0E+00	Assumed 99.7% conversion of sulfur; GREET [9]; [12]
hcl.kg	5.3E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	Based on natural gas consumption; [13]
naoh.kg	2.1E-01	2.0E-05	1.5E-05	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	[14]
csl.kg	1.1E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
glucose.kg	8.8E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
corn_starch.kg	2.0E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
k2o.kg	3.0E-01	0.0E+00	2.4E-06	9.3E-05	1.2E-03	2.0E-05	6.4E-07	0.0E+00	0.0E+00	GREET [9]
ammonia.kg	2.9E+00	0.0E+00	0.0E+00	7.9E-03	0.0E+00	0.0E+00	2.9E-05	0.0E+00	0.0E+00	Process emissions[12]; Emission factors[15]
n.kg	2.3E+00	0.0E+00	1.5E-02	1.5E-03	8.1E-03	2.3E-04	1.3E-05	0.0E+00	0.0E+00	GREET [9]
urea.kg	2.6E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
p2o5.kg	3.5E-01	0.0E+00	1.3E-06	5.5E-05	1.6E-03	2.3E-05	3.6E-07	0.0E+00	0.0E+00	GREET [9]
p.kg	1.2E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
nacl.kg	4.9E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
triethylaluminum.kg	4.8E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	Based on stoichiometry; Assumes aluminum is recycled
coal.MJ	1.9E-04	2.0E-10	5.1E-11	5.2E-06	1.0E-06	2.2E-08	8.1E-09	6.7E-06	0.0E+00	GREET [9]
diesel.MJ	5.1E-03	3.6E-06	2.4E-07	2.2E-06	4.1E-06	4.6E-07	1.6E-05	2.9E-06	0.0E+00	GREET [9]

rfo.MJ	7.2E-02	0.0E+00	1.4E-08	5.2E-07	2.1E-06	8.3E-08	3.7E-09	0.0E+00	0.0E+00	GREET [9]
refgas.MJ	7.1E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
ethylene.MJ	5.5E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	Assumed 10% loss by energy in production (crude) emitted; Used simple energy content-based allocation for ethylene (byproduct of cracking heavy molecules in petroleum refinery)
propene.MJ	7.1E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	Assumed 10% loss by energy in production (crude) emitted; Used simple energy content-based allocation for propene (byproduct of cracking heavy molecules in petroleum refinery)
acetone.kg	1.8E+00	1.7E-02	7.1E-09	1.9E-03	4.6E-03	1.5E-04	6.9E-03	3.5E-03	9.7E-09	Assume production via cumene process; [16]
crudeoil.MJ	6.1E-03	0.0E+00	1.4E-08	5.4E-07	1.6E-06	4.9E-08	3.9E-09	0.0E+00	0.0E+00	GREET [9]
electricity.US.kWh	4.5E-01	3.6E-05	5.9E-06	0.0E+00	3.3E-04	0.0E+00	3.6E-04	0.0E+00	0.0E+00	[17]
electricity.NGCC.kWh	1.8E-01	0.0E+00	0.0E+00	8.3E-05	9.9E-05	1.0E-06	4.0E-06	2.0E-06	0.0E+00	[18]
electricity.Coal.kWh	9.8E-01	1.1E-05	1.6E-05	0.0E+00	9.5E-04	0.0E+00	2.9E-03	0.0E+00	0.0E+00	[19]
electricity.WECC.kWh	4.3E-01	9.5E-06	5.8E-06	0.0E+00	5.1E-04	0.0E+00	3.5E-04	0.0E+00	0.0E+00	[19]
electricity.MRO.kWh	7.2E-01	1.3E-05	1.3E-05	0.0E+00	1.0E-03	0.0E+00	2.0E-03	0.0E+00	0.0E+00	[19]
electricity.TRE.kWh	5.2E-01	7.6E-06	5.9E-06	0.0E+00	3.3E-04	0.0E+00	1.0E-03	0.0E+00	0.0E+00	[19]
gasoline.MJ	7.5E-02	6.9E-06	1.6E-07	5.6E-06	1.7E-05	1.9E-07	5.7E-05	0.0E+00	0.0E+00	Assumed sufficiently similar to diesel; [20]
h2.kg	7.2E+00	6.0E-02	4.0E-05	6.0E-03	1.2E-02	2.0E-03	9.5E-03	0.0E+00	0.0E+00	
naturalgas.MJ	1.0E-03	1.2E-04	1.7E-08	6.6E-07	1.9E-06	6.0E-08	4.7E-09	6.1E-06	0.0E+00	[10]; [21]; GREET [9]
uranium.kg	7.1E+01	0.0E+00	1.2E-03	4.7E-02	1.3E-01	4.2E-03	3.3E-04	2.5E-02	0.0E+00	[22]; GREET [9]
steel_chinese.kg	4.0E-01	0.0E+00	5.9E-06	2.3E-04	1.1E-03	2.2E-05	1.3E-05	0.0E+00	0.0E+00	[23]; GREET [9]
flatbedtruck.mt_km	1.2E-01	0.0E+00	0.0E+00	8.6E-06	2.5E-05	2.8E-06	2.8E-06	0.0E+00	0.0E+00	[24]
tankertruck.mt_km	8.5E-02	0.0E+00	0.0E+00	0.0E+00	1.7E-05	1.9E-06	1.9E-06	0.0E+00	0.0E+00	[24]
gaspipeline.mt_km	0.0E+00	1.6E-03	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	Assume all natural gas travels 4000 km [25]; [10]
rail.mt_km	1.9E-02	0.0E+00	0.0E+00	0.0E+00	3.8E-06	4.3E-07	4.3E-07	0.0E+00	0.0E+00	[24]
barge.mt_km	2.2E-02	0.0E+00	0.0E+00	0.0E+00	4.6E-06	5.1E-07	5.1E-07	0.0E+00	0.0E+00	[24]
marinetanker.mt_km	6.9E-03	8.0E-08	0.0E+00	1.4E-06	1.4E-05	9.0E-07	4.5E-07	0.0E+00	0.0E+00	[24]
corn.bushel	6.7E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	GREET [9]
landfill_mixedorganics.wet_kg	0.0E+00	1.3E-02	0.0E+00	1.9E-05	1.4E-05	4.4E-06	4.5E-05	1.5E-07	4.7E-05	[1]; [26]

organics_composting_wet.kg	0.0E+00	1.5E-03	8.7E-05	2.7E-04	0.0E+00	0.0E+00	0.0E+00	5.9E-04	1.6E-03	GHGs [27]; ammonia [26]; carbon monoxide [28]; VOCs [29]
compost_application.kg	-1.5E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	[30]
biosolids_land_application_dry.kg	0	1.3E-04	2.1E-04	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	5.5E-05	[31]; Phyllis2 [32]; [26]
biogas_flare.m3	0.0E+00	0.0E+00	1.2E-06	2.5E-04	2.1E-04	5.5E-06	7.3E-04	5.0E-06	0.0E+00	Measured, reported in Kirchstetter et al.[33]
biogas_CHP.m3	0.0E+00	5.7E-03	-1.9E-06	5.5E-04	1.1E-04	5.5E-06	1.3E-04	4.6E-05	0.0E+00	Measured, reported in Kirchstetter et al.[33]
biogas_biofilter.m3	7.2E-01	3.3E-02	1.9E-04	2.0E-04	0.0E+00	0.0E+00	1.0E-04	0.0E+00	1.6E-03	Measured, reported in Kirchstetter et al.[33]
outdoor_compost.wet_kg	0.0E+00	4.5E-03	1.4E-05	1.1E-04	0.0E+00	0.0E+00	0.0E+00	2.2E-05	1.2E-03	Measured, reported in Kirchstetter et al.[33]
naturalgas_combust.MJ	5.0E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	Based on stoichiometry of methane combustion; Assumes perfect oxidation
diesel_combust.MJ	7.5E-02	0.0E+00	0.0E+00	2.1E-05	1.5E-05	4.6E-07	8.2E-06	2.9E-06	0.0E+00	GREET [9]

Table A2 lists all of the upstream/downstream impacts associated with each unit process/product involved. Each unit process/product is listed with a unit. The value indicates the amount of the reference product in its listed unit required to make 1 unit of the primary product.

Table A2. Input output matrix relationships

primary	Input Output References	value	Source
atrazine_brazil.kg	diesel.MJ	4.9E+01	GREET [9]
	rfo.MJ	4.9E+01	GREET [9]
	electricity.NGCC.kWh	7.6E+00	GREET [9]
	naturalgas.MJ	3.7E+01	GREET [9]
insecticide.kg	diesel.MJ	1.4E+02	GREET [9]
	electricity.US.kWh	1.2E+01	GREET [9]
	naturalgas.MJ	5.3E+01	GREET [9]
	tankertruck.mt_km	4.3E-01	GREET [9]
	rail.mt_km	1.3E+00	GREET [9]
insecticide_brazil.kg	diesel.MJ	1.4E+02	GREET [9]
	electricity.NGCC.kWh	1.2E+01	GREET [9]
	naturalgas.MJ	5.3E+01	GREET [9]
cellulase.kg	csl.kg	1.8E-01	GREET [9]
	glucose.kg	1.3E+00	GREET [9]
	ammonia.kg	6.0E-02	GREET [9]
	glycerin.kg	4.0E-01	GREET [9]
	nacl.kg	2.0E-01	GREET [9]
	electricity.MRO.kWh	1.1E+00	GREET [9]
	naturalgas.MJ	2.2E+00	GREET [9]
alpha_amylase.kg	glucose.kg	7.0E-04	GREET [9]
	electricity.MRO.kWh	1.9E+00	GREET [9]
	naturalgas.MJ	5.0E+00	GREET [9]
gluco_amylase.kg	glucose.kg	4.7E-01	GREET [9]
	corn_starch.kg	4.7E-01	GREET [9]
	electricity.MRO.kWh	8.9E+00	GREET [9]
	naturalgas.MJ	1.3E+01	GREET [9]
lime.kg	caco3.kg	1.4E+00	GREET [9]
	coal.MJ	4.0E+00	GREET [9]
	diesel.MJ	3.2E-01	GREET [9]
	electricity.US.kWh	8.2E-02	GREET [9]
	naturalgas.MJ	8.2E-01	GREET [9]
	tankertruck.mt_km	1.9E-01	Distance from Gabi [34]
caco3.kg	diesel.MJ	1.4E+00	GREET [9]
	electricity.US.kWh	6.5E-03	GREET [9]
	naturalgas.MJ	1.2E+00	GREET [9]
	flatbedtruck.mt_km	8.0E-02	GREET [9]
h2so4.kg	electricity.US.kWh	6.6E-02	GREET [9]
	tankertruck.mt_km	4.3E-01	GREET [9]
	rail.mt_km	1.3E+00	GREET [9]
hcl.kg	coal.MJ	9.2E-01	GREET [9]
	diesel.MJ	2.7E-02	GREET [9]
	rfo.MJ	2.7E-02	GREET [9]
	electricity.US.kWh	9.7E-01	GREET [9]
	gasoline.MJ	2.3E-03	GREET [9]
	naturalgas.MJ	8.8E+00	GREET [9]
naoh.kg	coal.MJ	3.8E-01	GREET [9]

	rfo.MJ	2.0E-01	GREET[9]
	electricity.US.kWh	1.8E+00	GREET[9]
	naturalgas.MJ	3.3E+00	GREET[9]
	tankertruck.mt_km	4.3E-01	GREET[9]
	rail.mt_km	1.3E+00	GREET[9]
csl.kg	electricity.MRO.kWh	6.5E-03	GREET[9]
	naturalgas.MJ	2.2E+00	GREET[9]
	corn.bushel	2.2E-01	GREET[9]
glucose.kg	naoh.kg	2.8E-06	GREET[9]
	corn_starch.kg	9.4E-01	GREET[9]
	naturalgas.MJ	1.7E-01	GREET[9]
corn_starch.kg	electricity.MRO.kWh	9.0E-02	GREET[9]
	naturalgas.MJ	4.0E+00	GREET[9]
	corn.bushel	6.9E-02	GREET[9]
k2o.kg	diesel.MJ	2.4E+00	GREET[9]
	naturalgas.MJ	2.7E+00	GREET[9]
	tankertruck.mt_km	4.3E-01	GREET[9]
	rail.mt_km	1.3E+00	GREET[9]
ammonia.kg	naturalgas.MJ	4.0E+01	GREET[9]
	tankertruck.mt_km	4.3E-01	GREET[9]
	rail.mt_km	1.3E+00	GREET[9]
n.kg	diesel.MJ	1.8E+00	GREET[9]
	naturalgas.MJ	4.4E+01	GREET[9]
	tankertruck.mt_km	4.3E-01	GREET[9]
	rail.mt_km	1.3E+00	GREET[9]
urea.kg	electricity.US.kWh	3.2E-01	GREET[9]
	naturalgas.MJ	8.1E+00	GREET[9]
	tankertruck.mt_km	4.3E-01	GREET[9]
	rail.mt_km	1.3E+00	GREET[9]
p2o5.kg	diesel.MJ	3.9E+00	GREET[9]
	naturalgas.MJ	1.5E+00	GREET[9]
	tankertruck.mt_km	4.3E-01	GREET[9]
	rail.mt_km	1.3E+00	GREET[9]
p.kg	electricity.NGCC.kWh	6.4E+00	GREET[9]
	naturalgas.MJ	2.3E+01	GREET[9]
nacl.kg	rfo.MJ	1.3E-01	GREET[9]
	electricity.US.kWh	7.1E-02	GREET[9]
	naturalgas.MJ	7.9E-01	GREET[9]
triethylaluminum.kg	ethylene.MJ	7.5E+01	Based on stoichiometry
	h2.kg	2.7E-02	Based on stoichiometry
	tankertruck.mt_km	4.3E-01	GREET[9]
	rail.mt_km	1.3E+00	GREET[9]
coal.MJ	diesel.MJ	2.3E-03	GREET[9]
	rfo.MJ	2.5E-04	GREET[9]
	electricity.US.kWh	2.1E-03	GREET[9]
	gasoline.MJ	1.9E-04	GREET[9]
	naturalgas.MJ	5.8E-05	GREET[9]
	flatbedtruck.mt_km	2.6E-03	GREET[9]
	rail.mt_km	5.3E-02	GREET[9]
	barge.mt_km	2.2E-02	GREET[9]
diesel.MJ	refgas.MJ	2.4E-02	GREET[9]
	crudeoil.MJ	1.0E+00	GREET[9]
	electricity.US.kWh	2.6E-03	GREET[9]

	naturalgas.MJ	2.4E-02	GREET [9]
	tankertruck.mt_km	3.5E-03	[20]
	liquidpipeline.mt_km	2.1E-02	[20]
rfo.MJ	refgas.MJ	1.6E-02	GREET [9]
	crudeoil.MJ	1.0E+00	-
	electricity.US.kWh	2.6E-03	GREET [9]
	naturalgas.MJ	1.6E-02	GREET [9]
	tankertruck.mt_km	3.5E-03	GREET [9]
	liquidpipeline.mt_km	2.1E-02	GREET [9]
refgas.MJ	crudeoil.MJ	1.0E+00	-
ethylene.MJ	diesel.MJ	6.2E-05	GREET [9]
	refgas.MJ	2.6E-02	GREET [9]
	electricity.TRE.kWh	3.1E-03	GREET [9]
	naturalgas.MJ	1.1E+00	GREET [9]
	tankertruck.mt_km	5.2E-04	GREET [9]
	liquidpipeline.mt_km	1.6E-03	GREET [9]
propene.MJ	crudeoil.MJ	1.0E+00	-
acetone.kg	h2so4.kg	4.9E-04	Ecoinvent [35]
	propene.MJ	1.5E+01	Ecoinvent [35]
	electricity.US.kWh	2.7E-01	Ecoinvent [35]
	gasoline.MJ	2.7E+01	Ecoinvent [35]
	naturalgas.MJ	1.9E+00	Ecoinvent [35]
	tankertruck.mt_km	4.3E-01	GREET [9]
	rail.mt_km	1.3E+00	GREET [9]
crudeoil.MJ	crudeoil.MJ	7.5E-02	GREET [9]
	electricity.US.kWh	1.5E-03	GREET [9]
	naturalgas.MJ	1.6E-02	GREET [9]
	tankertruck.mt_km	5.5E-03	GREET [9]
	liquidpipeline.mt_km	2.9E-02	GREET [9]
	rail.mt_km	2.4E-02	GREET [9]
	barge.mt_km	4.4E-03	GREET [9]
	marinetanker.mt_km	1.1E-01	GREET [9]
electricity.US.kWh	coal.MJ	5.0E+00	[17]
	diesel.MJ	6.7E-03	[17]
	rfo.MJ	3.0E-02	[17]
	electricity.US.kWh	6.5E-02	[17]
	naturalgas.MJ	1.9E+00	[17]
	uranium.kg	2.8E-07	[17]
electricity.NGCC.kWh	electricity.NGCC.kWh	6.5E-02	[17]
	naturalgas.MJ	7.2E+00	[17]
electricity.Coal.kWh	coal.MJ	1.1E+01	[17]
	electricity.Coal.kWh	6.5E-02	[17]
electricity.WECC.kWh	coal.MJ	3.3E+00	[17]
	diesel.MJ	9.4E-04	[17]
	electricity.WECC.kWh	8.2E-02	[17]
	naturalgas.MJ	2.6E+00	[17]
	uranium.kg	1.3E-07	[17]
electricity.MRO.kWh	coal.MJ	7.9E+00	[17]
	diesel.MJ	3.7E-03	[17]
	electricity.MRO.kWh	5.8E-02	[17]
	naturalgas.MJ	2.0E-01	[17]
	uranium.kg	1.9E-07	[17]
electricity.TRE.kWh	coal.MJ	3.7E+00	[17]

	diesel.MJ	1.9E-04	[17]
	electricity.TRE.kWh	8.0E-02	[17]
	naturalgas.MJ	3.9E+00	[17]
	uranium.kg	1.7E-07	[17]
gasoline.MJ	refgas.MJ	3.7E-02	GREET [9]
	crudeoil.MJ	1.0E+00	GREET [9]
	electricity.US.kWh	2.5E-03	GREET [9]
	naturalgas.MJ	3.7E-02	GREET [9]
	tankertruck.mt_km	3.5E-03	GREET [9]
	liquidpipeline.mt_km	2.1E-02	GREET [9]
h2.kg	electricity.US.kWh	2.7E-01	[25]
	naturalgas.MJ	1.4E+02	[25]
	gaspipeline.mt_km	1.2E+00	GREET [9]
naturalgas.MJ	naturalgas.MJ	2.0E-02	GREET [9]
	gaspipeline.mt_km	7.7E-02	GREET [9]
uranium.kg	electricity.WECC.kWh	1.0E+02	[23]
	electricity.MRO.kWh	4.4E+01	[23]
	naturalgas.MJ	1.4E+03	[22]
	flatbedtruck.mt_km	4.5E+00	GREET [9]
steel_chinese.kg	coal.MJ	6.9E-01	GREET [9]
	electricity.Coal.kWh	8.2E-01	GREET [9]
	electricity.Renewables.kWh	3.5E-01	GREET [9]
	naturalgas.MJ	6.8E+00	GREET [9]
	rail.mt_km	8.0E-01	GREET [9]
	barge.mt_km	1.0E+01	GREET [9]
flatbedtruck.mt_km	diesel.MJ	1.8E+00	Fuel economy from Strogen et al.[36]
	flatbedtruck.mt_km	2.5E-01	[20]; assumption of 25% empty miles
tankertruck.mt_km	diesel.MJ	1.2E+00	Fuel economy from Strogen et al.[36]
	tankertruck.mt_km	2.5E-01	[20]; assumption of 25% empty miles
rail.mt_km	diesel.MJ	2.7E-01	Fuel economy from Strogen et al.[36]
	rail.mt_km	2.5E-01	[20]; assumption of 25% empty miles
barge.mt_km	diesel.MJ	3.2E-01	Fuel economy from Strogen et al.[36]
	barge.mt_km	2.5E-01	Assume 25% empty miles
marinetanker.mt_km	rfo.MJ	1.0E-01	Average of crude tanker & product tanker energy intensities [36]
	marinetanker.mt_km	2.5E-01	Assume 25% empty miles
corn.bushel	atrazine.kg	3.5E-03	GREET [9]
	glyphosate.kg	3.5E-03	GREET [9]
	insecticide.kg	6.0E-05	GREET [9]
	caco3.kg	1.1E+00	GREET [9]
	k2o.kg	1.5E-01	GREET [9]
	n.kg	4.2E-01	GREET [9]
	p2o5.kg	1.5E-01	GREET [9]
	diesel.MJ	9.6E+00	Assumed all energy provided diesel; GREET [9]
	flatbedtruck.mt_km	2.0E+00	Assumed 50 miles; GREET [9]

compost_application.kg	n.kg	-1.0E-02	[30]
outdoor_compost.wet_kg	diesel.MJ	5.8E-02	[30]
	electricity.US.kWh	7.9E-03	[30]
	flatbedtruck.mt_km	5.0E-02	Assume 50 km
organics_composting_wet.kg	diesel.MJ	5.8E-02	[30]
	electricity.US.kWh	7.9E-03	[30]

Note: If any primary parameters are missing from the table above, there are no upstream/downstream impacts for that parameter recorded in the IO table.

A4 Sensitivity Analysis

Table A3. Monte Carlo simulation inputs

	Parameter	Model Value	Distribution Type	Distributional Parameters	Source/Assumption
CH_4	landfill_mixedorganics.wet_kg	2.70E-02	triangle	Min = 4.63E-03 Max = 2.70E-02	Maximum from WARM model [37] and minimum from Behera et al.[1]
	organics_composting_wet.kg	2.09E-03	triangle	Min = 1.42E-04 Max = 7.63E-03	Minimum and maximum [27]
	biosolids_land_application_dry.kg	1.32E-04	triangle	Min = 1.19E-04 Max = 1.66E-04	Distribution from non-combustion emissions; varied carbon content of dry digestate, Phyllis2 [32]
CO_2	outdoor_compost.wet_kg	4.46E-03	triangle	Min = 1.27E-03 Max = 1.22E-02	Distributional parameters from measured data
	gasoline.MJ	7.48E-02	triangle	Min = 7.28E-02 Max = 7.92E-02	Combustion emissions distribution [38]
	diesel_combust.MJ	7.53E-02	triangle	Min = 7.36E-02 Max = 8.01E-02	[38]
N_2O	organics_composting_wet.kg	8.70E-05	triangle	Min = 2.20E-05 Max = 3.00E-04	Minimum and maximum [27]
CO	electricity.NGCC.kWh	9.00E-05	uniform	+/- 20%	
	landfill_mixedorganics.wet_kg	1.87E-05	triangle	Min = 1.31E-05 Max = 2.75E-05	Minimum and maximum from Ecoinvent [35]
	organics_composting_wet.kg	5.22E-05	triangle	Min = 1.00E-06 Max = 1.11E-04	Minimum from Hellebrand and Schade[28] and maximum from measured data
	biogas_CHP.m3	5.51E-04	normal	SD = 4.28E-05	Distributional parameters from measured data
	diesel_combust.MJ	2.10E-05	uniform	+/- 20%	
NO_X	electricity.NGCC.kWh	1.08E-04	uniform	+/- 20%	
	landfill_mixedorganics.wet_kg	1.37E-05	triangle	Min = 2.39E-06 Max = 2.28E-05	Minimum and maximum from Ecoinvent [35]
	biogas_CHP.m3	1.07E-04	normal	SD = 1.03E-04	Measured, reported in Kirchstetter et al.[33]
	diesel_combust.MJ	1.49E-05	uniform	+/- 20%	
$PM_{2.5}$	electricity.NGCC.kWh	1.00E-06	uniform	+/- 20%	
	landfill_mixedorganics.wet_kg	4.44E-06	triangle	Min = 3.15E-06 Max = 6.70E-06	Minimum and maximum from Ecoinvent [35]
	biogas_flare.m3	5.46E-06	triangle	Min = 5.11E-06 Max = 1.10E-05	Measured, reported in Kirchstetter et al.[33]
	diesel_combust.MJ	4.60E-07	uniform	+/- 20%	
SO_2	electricity.NGCC.kWh	4.00E-06	uniform	+/- 20%	

	naturalgas.MJ	4.74E-09	uniform	+/- 20%	
	landfill_mixedorganics.wet_kg	4.54E-05	triangle	Min = 3.44E-05 Max = 4.96E-05	Minimum and maximum from Ecoinvent [35]
	biogas_CHP.m3	1.32E-04	triangle	Min = 1.50E-06 Max = 5.68E-04	Distributional parameters from measured data
	diesel_combust.MJ	8.19E-06	uniform	+/- 20%	
VOC	electricity.NGCC.kWh	2.00E-06	uniform	+/- 20%	
	naturalgas.MJ	6.10E-06	uniform	+/- 20%	
	landfill_mixedorganics.wet_kg	1.51E-07	triangle	Min = 1.51E-07 Max = 3.21E-07	Minimum and maximum from Ecoinvent [35]
	biogas_CHP.m3	4.55E-05	triangle	Min = 5.00E-06 Max = 4.00E-04	Measured, reported in Kirchstetter et al.[33]
NH_3	organics_composting_wet.kg	1.76E-03	triangle	Min = 1.14E-03 Max = 2.36E-03	Minimum and maximum [26]
	outdoor_compost.wet_kg	1.16E-03	triangle	Min = 6.70E-04 Max = 1.58E-03	Measured, reported in Kirchstetter et al.[33]

Note: All triangular distributions assume the mode (peak of distribution) to be the model value.

A5 Air Pollutant Inventory Results

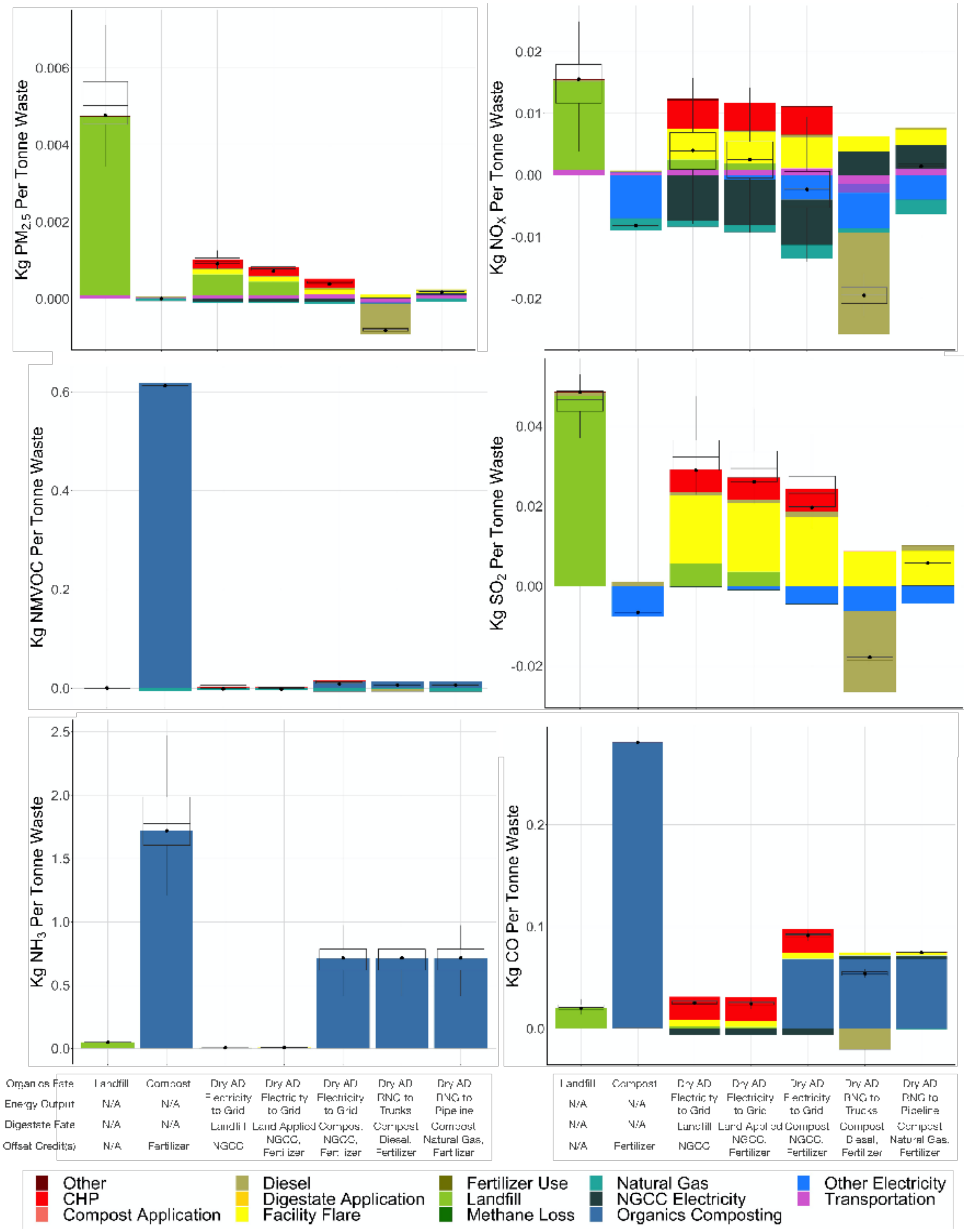

Figure A8. Life-cycle $PM_{2.5}$, NO_x, NMVOC, SO_2, NH_3, and CO emissions

Contributors totaling less than 1% are categorized as "Other". "Other Electricity" category refers to avoided electricity consumption from reduced urea fertilizer consumption.

A6 Social Costs

The recent inter-comparison of integrated assessment models by Gilmore et al.[39] confirmed the relatively high social cost of NH_3 in all three models evaluated, especially when compared to NO_x. Since the molecular weight of NO_x is nearly three times larger than NH_3 per unit mass, NH_3 will generate more NH_4NO_3 molecules than will NO_x. Also, NH_3 reacts faster than SO_2 and NO_x to form secondary $PM_{2.5}$,[40] such that the secondary $PM_{2.5}$ plume is smaller and more concentrated at ground level near people.

It is important to note the difficulty of accurately predicting secondary $PM_{2.5}$ formation in integrated assessment models; this is especially true for California. While sulfate formation is most important in the Eastern U.S., in California, secondary inorganic $PM_{2.5}$ is largely dominated by NH_4NO_3 due to use of low sulfur fuels in the power and transportation sectors.[41] As discussed in Heo, Adams and Gao[42], NH_4NO_3 is a more difficult component to model than others in $PM_{2.5}$ due to greater uncertainties in emissions and atmospheric processes. Furthermore, the impact of NH_3 on particle formation is dependent on the relative abundance of NH_3 versus HNO_3 in the atmosphere, so the effect is location dependent. Both EASIUR and AP3 have been calibrated using data in the Eastern U.S., where the meteorology and atmospheric chemistry are different from the Western U.S. As such, further modeling for specific locations, particularly in the Western U.S., is warranted before making definitive conclusions. Additionally, care should be taken when comparing studies using different APEEP versions, as substantial changes to the model have occurred, particularly with respect to damage multipliers for NH_3.

Table A4. Social cost multipliers

IAM	Location	Marginal Costs ($/tonne of emission)				
		$PM_{2.5}$	SO_2	NO_X	NH_3	VOC
EASIUR [40,42]	ZWEDC facility (San Jose, CA 95134)	390250	34475	19743	108800	-
EASIUR [40,42]	Z Best (Gilroy, CA 95020)	192500	32725	12823	43900	-
AP3 [43]	Santa Clara County (FIPS: 06085)	523842.5	238548.2	86300.09	322440.8	23635.8

References

(1) Behera, S. K.; Park, J. M.; Kim, K. H.; Park, H.-S. Methane production from food waste leachate in laboratory-scale simulated landfill. *Waste Manag.* **2010**, *30*, 1502–1508.

(2) Chen, X.; Romano, R.; Zhang, R. Anaerobic digestion of food wastes for biogas production. *International Journal of Agricultural and Biological Engineering* **2010**, *3*.

(3) Torres-Climent, A.; Martin-Mata, J.; Marhuenda-Egea, F.; Moral, R.; Barber, X.; Perez-Murcia, M. D.; Paredes, C. Composting of the Solid Phase of Digestate from Biogas Production: Optimization of the Moisture, C/N Ratio, and pH Conditions. *Commun. Soil Sci. Plant Anal.* **2015**, *46*, 197–207.

(4) Sullivan, D. M.; Cogger, C. G.; Bary, A. I. *Fertilizing with Biosolids*; Oregon State University Extension, 2015.

(5) Pell Frischmann Consultants Ltd. *Enhancement and treatment of digestates from anaerobic digestion*; WRAP, 2012.

(6) Masebinu, S. O.; Aboyade, A. O.; Muzenda, E. Economic Analysis of Biogas Upgrading and Utilization as Vehicular Fuel in South Africa. *Proceedings of the World Congress on Engineering and Computer Science* **2015**, *2*.

(7) Ullah Khan, I.; Hafiz Dzarfan Othman, M.; Hashim, H.; Matsuura, T.; Ismail, A. F.; Rezaei-DashtArzhandi, M.; Wan Azelee, I. Biogas as a renewable energy fuel – A review of biogas upgrading, utilisation and storage. *Energy Conversion and Management* **2017**, *150*, 277–294.

(8) Sun, Q.; Li, H.; Yan, J.; Liu, L.; Yu, Z.; Yu, X. Selection of appropriate biogas upgrading technology-a review of biogas cleaning, upgrading and utilisation. *Renewable and Sustainable Energy Reviews* **2015**, *51*, 521–532.

(9) Wang, M. *The Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) Model Version 1.5*; Center for Transportation Research, Argonne National Laboratory, 2008.

(10) EPA. *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990 - 2011*; 430-R-13001; U.S. Environmental Protection Agency, 2013.

(11) Research Triangle Institute. *Lime Production: Industry Profile*; EPA Contract 68-D-99-024; U.S. Environmental Protection Agency, 2000.

(12) EPA. Synthetic Ammonia, Chapter 8: Inorganic Chemical Industry. In *AP-42: Compilation of Air Emissions Factors*; U.S. Environmental Protection Agency, 1993.

(13) ecoinvent. *Ecoinvent v3.5*; 2018.

(14) Wicke, B.; Dornburg, V.; Junginger, M.; Faaij, A. Different palm oil production systems for energy purposes and their greenhouse gas implications. *Biomass and Bioenergy* **2008**, *32*, 1322–1337.

(15) EPA. *Technical Support Document for the Ammonia Production Sector: Proposed Rule for Mandatory Reporting of Greenhouse Gases*; U.S. Environmental Protection Agency, 2009.

(16) ecoinvent. *Ecoinvent v3.6*; 2019.

(17) eGRID2016. *Emissions & Generation Resource Integrated Database* ; U.S. Environmental Protection Agency, 2018.

(18) EIA. *Natural Gas 1998: Issues and Trends*; DOE/EIA-0560(98); Office of Oil and Gas, Energy Information Administration, U.S. Dept. of Energy: Washington, DC, 1999.

(19) eGRID2012. *Emissions & Generation Resource Integrated Database (Version 1)*; U.S. Environmental Protection Agency, 2012.

(20) Sheehan, J.; Camobreco, V.; Duffield, J.; Shapouri, H.; Graboski, M.; Tyson, K. S. *An*

overview of biodiesel and petroleum diesel life cycles; National Renewable Energy Laboratory (NREL), 1998.

(21) EIA. California Natural Gas Consumption by End Use https://www.eia.gov/dnav/ng/ng_cons_sum_dcu_SCA_a.htm (accessed Feb 1, 2019).

(22) Lenzen, M. Life cycle energy and greenhouse gas emissions of nuclear energy: A review. *Energy Conversion and Management* **2008**, *49*, 2178–2199.

(23) Scown, C. D.; Horvath, A.; McKone, T. E. Water footprint of U.S. transportation fuels. *Environ. Sci. Technol.* **2011**, *45*, 2541–2553.

(24) Cohon, J. L.; Cropper, M. L.; Cullen, M. R.; English, M. R.; Field, C. B.; Greenbaum, D. S.; Hammitt, J. K.; Henderson, R. F.; Kling, C. L.; Krupnick, A. J.; Lee, R.; Matthews, H.S.; McKone, T.E.; Metcalf, G.E.; Newell, R.G.; Reversz, R.L.; Wing, I.S.; Surles, T.G. *Hidden costs of energy: unpriced consequences of energy production and use*; National Academies Press: Washington, D.C., 2010.

(25) Spath, P.; Mann, M. *Life Cycle Assessment of Hydrogen Production via Natural Gas Steam Reforming*; NREL/TP-570-27637; National Renewable Energy Laboratory, 2001.

(26) Roe, S.; Spivey, M.; Lindquist, H.; Thesing, K.; Strait, R.; E.H. Pechan & Associates, Inc. *Estimating Ammonia Emissions from Anthropogenic Non-Agricultural Sources*; Emission Inventory Improvement Program, 2004.

(27) Amlinger, F.; Peyr, S.; Cuhls, C. Green house gas emissions from composting and mechanical biological treatment. *Waste Manag. Res.* **2008**, *26*, 47–60.

(28) Hellebrand, H. J.; Schade, G. W. Carbon monoxide from composting due to thermal oxidation of biomass. *J Environ Qual* **2008**, *37*, 592–598.

(29) Smet, E.; Van Langenhove, H.; De Bo, I. The emission of volatile compounds during the aerobic and the combined anaerobic/aerobic composting of biowaste. *Atmos. Environ.* **1999**, *33*, 1295–1303.

(30) CARB. *Method for Estimating Greenhouse Gas Emission Reductions from Diversion of Organic Waste from Landfills to Compost Facilities*; California Environmental Protection Agency, 2017.

(31) Holly, M. A.; Larson, R. A.; Powell, J. M.; Ruark, M. D.; Aguirre-Villegas, H. Greenhouse gas and ammonia emissions from digested and separated dairy manure during storage and after land application. *Agriculture, Ecosystems & Environment* **2017**, *239*, 410–419.

(32) ECN.TNO. Phyllis2 - Database for (treated) biomass, algae, feedstocks for biogas production and biochar https://phyllis.nl/ (accessed Nov 26, 2019).

(33) Kirchstetter, T. W.; Scown, C. D.; Satchwell, A.; Jin, L.; Smith, S. J.; Preble, C. V.; Amirebrahimi, J.; Sohn, M. D.; Brown, N. J.; Nordahl, S.; Devkota, J.; He, Y.; Ho, T.; Maddalena, R.; Tang, N.W.; Zhou, W.; Chen, S.S.; Hotchi, T. *Enabling Anaerobic Digestion Deployment for Municipal Solid Waste-to-Energy*; California Energy Commission, 2020.

(34) Thinkstep. *GaBi*; 2018.

(35) Wernet, G.; Bauer, C.; Steubing, B.; Reinhard, J.; Moreno-Ruiz, E.; Weidema, B. The ecoinvent database version 3 (part I): overview and methodology. *Int. J. Life Cycle Assess.* **2016**, *21*, 1218–1230.

(36) Strogen, B.; Horvath, A.; McKone, T. E. Fuel miles and the blend wall: costs and emissions from ethanol distribution in the United States. *Environ. Sci. Technol.* **2012**, *46*, 5285–5293.

(37) USEPA. Waste Reduction Model (WARM), 2018.

(38) Venkatesh, A.; Jaramillo, P.; Griffin, W. M.; Matthews, H. S. Uncertainty analysis of life

cycle greenhouse gas emissions from petroleum-based fuels and impacts on low carbon fuel policies. *Environ. Sci. Technol.* **2011**, *45*, 125–131.

(39) Gilmore, E. A.; Heo, J.; Muller, N. Z.; Tessum, C. W.; Hill, J. D.; Marshall, J. D.; Adams, P. J. An inter-comparison of the social costs of air quality from reduced-complexity models. *Environmental Research Letters* **2019**, *14*, 074016.

(40) Heo, J.; Adams, P. J.; Gao, H. O. Public health costs of primary PM2.5 and inorganic PM2.5 precursor emissions in the united states. *Environ. Sci. Technol.* **2016**, *50*, 6061–6070.

(41) Chow, J. C.; Chen, L. -W. A.; Watson, J. G.; Lowenthal, D. H.; Magliano, K. A.; Turkiewicz, K.; Lehrman, D. E. PM2.5 chemical composition and spatiotemporal variability during the California Regional PM10/PM2.5 Air Quality Study (CRPAQS). *J. Geophys. Res. Atmos.* **2006**.

(42) Heo, J.; Adams, P. J.; Gao, H. O. Reduced-form modeling of public health impacts of inorganic PM2.5 and precursor emissions. *Atmos. Environ.* **2016**, *137*, 80–89.

(43) Muller, N. Z.; Mendelsohn, R. Measuring the damages of air pollution in the United States. *J. Environ. Econ. Manage.* **2007**, *54*, 1–14.

Appendix B

Supplementary Information

Greenhouse Gas and Air Pollutant Emissions from Composting

B1 Relevance of Ammonia Emissions

In addition to being malodorous, NH_3 emissions are an important precursor to $PM_{2.5}$ formation, so it is not uncommon for studies to report NH_3 emissions alongside GHGs. Once in the atmosphere, NH_3 can react with nitric acid (HNO_3) to form particle-phase ammonium nitrate (NH_4NO_3) and/or sulfuric acid (H_2SO_4) to form particle-phase ammonium sulfate ($(NH_4)_2SO_4$). The precursors that NH_3 reacts with, H_2SO_4 and HNO_3, are formed in the atmosphere as a result of NO_x and SO_x emissions from power plants, motor vehicles, and other combustion activities. Alternatively, NH_3 may undergo wet or dry deposition, accumulating in nearby water bodies or on soil surfaces, where a portion of the nitrogen is later converted to N_2O.[1] Predicting the impact of NH_3 emissions on $PM_{2.5}$ concentrations has proved challenging and integrated assessment models vary in their predictions.[2] Nonetheless, Tschofen et al. (2019) found that NH_3 is responsible for the largest share of air quality-related monetized health damages from the agriculture sector.[3] A prior study found that NH_3 dominated the total social costs—including both climate change impacts and air quality-related human health damages—in any organic waste processing scenario that included composting.[4]

B2 Measurement Methods

B2.1 Flux Chambers

One commonly used approach to quantify composting emissions is to place static, open-bottomed chambers with small surface area footprints on the emitting surface. There are two common variants of these flux chambers, one in which swept air flow from over the emitting surface is sampled and a modified version that relies on diffusive transport to accumulate emitted gas into a headspace volume that is then sampled. In the first, ultrapure or "zero" air that is free of the air pollutants of interest is introduced at the inlet and gas samples are taken from the chamber outlet flow, either collected into a canister or bag for laboratory analysis or measured in situ.[5–7] This method follows the U.S EPA protocol for measuring gaseous emission rates from land surfaces.[8] A modified approach for non-aerated windrows where diffusion is the main emission mechanism outside of pile turning periods is to use closed/airtight or vented chambers, where the concentrations measured in the chamber headspace can be related to gas fluxes.[9–15] With flux chambers, the small surface area may not be representative of the entire emitting surface and measurements can be temporally constrained in resolution. The chamber can also introduce pressure and concentration gradients that impact emission fluxes from the windrow surface.

B2.2 Gas Probes

To collect cross-sectional gas samples from within the pore space of the composting pile rather than the emitting surface, probes are inserted into the windrow at varying depths.[9,11,13,15,16] The probes are flushed prior to collecting a sample to ensure that gas from the pore space itself is sampled rather than the probe's dead volume. From these measurements, it is possible to determine the spatial distributions of gas concentrations within the windrow, providing insight into composting dynamics like pockets of anaerobic activity with elevated CH_4 concentrations.

B2.3 Wind Tunnels

Wind tunnels have been used as an alternative to flux chambers when the high water content of sampled gas has been an issue.[7] These static, flow-through enclosures also have open bottoms and are inserted ~1 cm into the windrow surface. A fan introduces ambient air dilution to emitted gas, which is then collected into canisters or bags. These tunnels cover larger surface areas than flux chambers, the dilution offers more control of water content in samples, and the greater air exchange rate is more similar to ambient conditions. Similar to flux chambers, though, the small surface area sampled may not be representative of the entire windrow and measurements can be temporally constrained. The inlet and outlet air must also be simultaneously sampled to properly calculate emission flux, as ambient air with non-zero concentrations of the air pollutants of interest is used for dilution rather than zero air.

B2.4 Open Emission Chambers

To capture emissions from across the emitting surface, open chambers are built over the windrow, with air flowing in and out of the control volume either naturally or by a ventilation system.[17–21] The difference in measured concentrations in the incoming and outgoing air can be used to calculate the emissions inside the chamber from the composting pile. In theory, this sampling approach does not alter the conditions inside the chamber, and compost can be maintained in the usual manner, for example aeration with pile turning. If pollutant analyzers with a fast time response are used, this measurement can be temporally-resolved and show emissions changes over time. However, as it integrates emissions over the entire windrow, it will not discern spatial variability in emissions across the surface.

B2.5 Tracer Releases

If an inert gas is released at the emission source at a known rate, the downwind ratio of tracer gas to pollutant concentration can be measured to determine the pollutant emission rate.[13,22] These measurements are relatively simple to conduct, but the emission point may not be representative of the entire composting surface. It may also be difficult to isolate a specific windrow from facility-wide emissions, if there are multiple emission sources of the pollutant species of interest.

B2.6 Inverse Dispersion Analysis

For sources with known geometry, emission rates can be determined with a dispersion model that pairs measured downwind concentrations and local meteorology.[22] This micrometeorological analysis technique gives an integrated measure of emission flux, but may also be limited to facility-wide emission rates if windrows are in close proximity to each other or if there are other nearby emission sources of the pollutant of interest. While the measurements are relatively simple to conduct, the analysis relies on an accurate dispersion model.

B2.7 Micrometeorological Mass Balance

Another micrometeorological approach relies on mass balance, in which pollutant fluxes in and out of a control volume surrounding the emission source are determined from measured gas concentrations and wind dynamics.[23] This method captures the integrated emission rate from an isolated windrow or full-scale facility operations, and measurements can be made across the composting cycle to characterize emission rates as a function of time.

B2.8 High-Density Spot Sampling

At some facilities, the density of windrow placement and local environmental conditions preclude the use of open emission chambers or micrometeorological approaches to capture the emissions from individual composting windrows. Moreover, when spatial heterogeneity across the emitting surface is expected—such as with heterogeneous OFMSW or digestate feedstocks compared to more uniform materials like yard waste—small surface area footprint approaches like flux chambers and gas probes may not be sufficient to capture a representative sample of emissions. In these cases, a high-density spot sampling method can be employed instead.[24] With this technique, the characteristic emission rate for individual windrows is calculated from the measured forced aeration flow and numerous spot gas samples that are collected into bags from across the composting surface and later analyzed in the laboratory. Multiple windrows can be sampled in a given day to compare emission rates across the composting cycle. This approach is intensive before and after sampling, though, in terms of sample bag preparation and analysis. Measurements are intermittent across the composting cycle rather than continuous, producing snapshots of emissions at given points of time. This method is also limited to force-aerated windrows and cannot be applied to composting piles that are turned or naturally/passively aerated.

B2.9 Tradeoffs Among Common Measurement Methods

When selecting composting studies to draw from for use in a broader environmental analysis, one could reasonably ask whether particular measurement methods are superior and should be given priority. There is a clear tradeoff among the methods discussed here between specificity to the feedstock of interest and the degree to which the measurements accurately represent emissions in commercial composting conditions. In most cases, it is not realistic to run experiments in which large-scale windrows are composed of a single material, nor would a mixed windrow result in emissions equal to the sum of its parts in isolation. Generally, measurements taken in the field from actual composting operations are preferable to lab-based studies. However, lab-based measurements can be valuable, particularly when done in combination with field studies, by better characterizing the relative impact of specific changes to feedstock composition or composting conditions (e.g., moisture content, pH, etc.) on emissions.

Of the field-based measurement methods, there is no single approach that is obviously superior. Spot sampling methods, which include flux chambers, gas probes, wind tunnels, tracer releases, and high-density spot sampling, can provide some spatial resolution of emissions from a composting windrow. Sampling size and distribution across the windrow are important factors in calculating a total, cumulative emission factor from these methods. Therefore, one should be wary of emission factors from studies that employed one of these methods with a small number of samples or if the spatial distribution of sampling locations along the windrow or pile is limited. In addition to spatial distribution, the temporal distribution of measurements over the composting cycle is equally important. Commercial composting takes 3–6 months, and emissions will vary across the mesophilic, thermophilic, and maturation phases. If a study uses flux chambers, wind tunnels, tracer releases or high-density spot sampling, it is important that measurements were taken with some regularity over the entire composting cycle to determine a final emission factor.

Employing spot sampling measurements at multiple locations across a windrow for an entire multi-month composting cycle is labor-intensive and may not be practical in some cases. Other approaches, like open emission chambers, inverse dispersion analysis, and micrometeorological mass balances, can offer windrow-wide or facility-wide results but do not offer spatially resolved results. The downside to such approaches is that emissions cannot be easily connected back to the types of material being composted, the composting conditions at a large facility that accepts a range of wastes, or the composting dynamics within the windrow itself (e.g., nonuniform aeration that leads to pockets of anaerobic activity). Moreover, some of these results can be restricted by the detection and quantification limits of the pollutant analyzers used, such that emissions may be non-zero but not detectable downwind of the source. Ultimately, researchers and other practitioners may choose to draw emission factors from multiple studies and use a range or probability distribution when incorporating composting emissions into life-cycle assessments and other environmental impact studies. However, the information provided here may be useful in selecting the most rigorous and representative studies for a given application.

B2.10 Impact of Measurement Methods

Our analysis of the impact of measurement methods was inconclusive because there are not enough comparable studies to find meaningful results. The data collected for this systematic review does not provide sufficient evidence to suggest that any particular method consistently overestimates or underestimates emission measurements more than other methods. Researchers and practitioners should select emission factors based on their suitability for the specific analysis in which they are being used, and the quality of the associated study's measurement approach (e.g., sufficient temporal and spatial distribution of sampling in spot sample approaches). More research observing the same feedstocks and composting conditions with varied measurement techniques is required to better understand how these methods may skew results.

B3 Carbon Dioxide Emissions from Composting

Figure B1 shows the results for biogenic CO_2, excluding one major outlier from,[16] which observed an unusually high emission factor of 0.87 kg CO_2 per kg of green waste composted. During composting, a fraction of initial carbon in the feedstock is emitted, primarily as CO_2 and CH_4, and the remaining carbon is retained in the material and can contribute to soil organic carbon once applied to land.[23,25] If a composting operation is not well aerated, microbes will consume the organic material more slowly, leading to elevated CH_4 emissions and a reduction in CO_2 emissions. This negative correlation between CO_2 and CH_4 emissions has been measured by Jiang et al. (2011) and Chowdhury et al. (2014).[26,27] This finding is not consistent across the literature, however, with other studies reporting a positive correlation between CO_2 and CH_4 emissions.[19,20,24,28] Many of these studies provide limited data, with samples ranging from just 3 to 9 measurement pairs.[28,26] Therefore, we cannot recommend using measured CO_2 as a good predictor of the relative magnitude of CH_4 emissions (or any other pollutants) based on the data available.

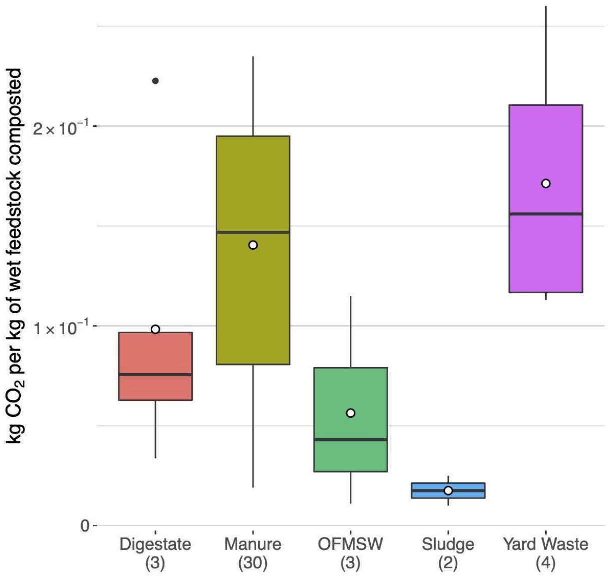

Figure B1. Distributions of CO_2 emission factors for composting reported in the literature
The sample size (n) of data points contributing to each boxplot is indicated in the x-axis labels. The mean values for the boxplot data are indicated by the open point symbols, while outliers are shown as closed circles.

B4 GHG Emissions Implications of Landfilling and Land Application

B4.1 Net Emissions and Offsets From Compost Application to Soils

Composting produces nutrient-rich material that can be beneficial to plant growth, contribute to pest and disease prevention, and offset the need for synthetic mineral fertilizers.[29] The benefits of compost application to soils include increased soil organic matter, stability, and water retention.[30] For this to hold true, the compost must be applied in an agricultural application, although some benefits may still be achieved in landscaping applications. Compost must also be applied at the agronomic rate, meaning the recommended application rate to achieve optimum plant growth. Assuming that all material sent to commercial composting operations will ultimately be used beneficially as compost in agricultural applications is likely too optimistic, and the market for compost will vary regionally. For example, a 2017 market analysis conducted by CalRecycle in California (U.S.) indicated that 22% of output from in-state composters went to landfills, mostly as alternative daily cover.[31] Researchers quantifying the life-cycle GHG benefits of waste diversion must be mindful of the fact that, if supply of finished compost exceeds demand, the marginal use-phase benefits of compost may be diminished or near-zero if it is either used for alternative daily cover or applied at levels exceeding the agronomic rate. An adjustment factor may be necessary to account for the fraction of finished compost not being used in an agronomic application.

If compost is applied to agricultural soils, there are two key drivers of net emissions/offsets: (1) increasing crop yields and thus reducing land and other inputs needed to produce the same amount of agricultural product; and (2) reducing the need for mineral fertilizer production and application. Pest prevention and associated reductions in pesticide use are a third contributor to offsets, but these impacts are complex, highly variable, and too ambiguous to be useful from an LCA perspective.[29]

Compost use as a soil amendment allows for the slow release of nitrogen on the scale of multiple years, as nitrogen in the compost is immobilized when it is taken up by microbes during the composting process and is only released as those microbes die and their cell walls lyse.[32] For instance, Sullivan et al. (2003) observed a continued increase in soil organic matter and crop yields for seven years after a one-time application of compost.[33] Sullivan et al. (2003) also found that composting feedstocks with higher nitrogen content, such as food waste, produced a soil amendment that resulted in greater crop yield benefits.

Offsetting nitrogen fertilizer use is particularly important with respect to net GHG emissions because fertilizer production (e.g., urea) is energy- and emissions-intensive to produce.[34] Compost is most commonly used in addition to fertilizers, as opposed to a full replacement.[35] Given the complementary nature of synthetic fertilizers with compost when used concurrently to increase agricultural yields, there may not be a one-to-one substitution between the two when adjusted based on total available nitrogen. This is further complicated by the lack of consistency in compost quality and composition, particularly for composted OFMSW.[36] Favoino and Hogg (2008) estimate that a one-time application of 10 tonnes of compost has the potential to displace 190 kg of nitrogen and save from 160 kWh up to nearly 1600 kWh of energy, although it is unclear if this refers to primary or secondary energy.[34] Although the exact value of synthetic nitrogenous fertilizer use that can be offset through compost application is uncertain, there does appear to be broad consensus that fertilizer application,and thus upstream emissions from its production, can be reduced through compost application to agricultural land. Application of nitrogenous fertilizer also contributes to GHG emissions after it is applied through N_2O fluxes to the atmosphere. The extent to which compost application can reduce these fluxes, if at all, is uncertain and dependent on local conditions, the existing soil biological community, fertilizer characteristics, compost characteristics, and management practices.[34,37] For example, Ryals and Silver (2013) did not observe significant changes to soil CH_4 or N_2O fluxes after compost application.[38] In the absence of consistent empirical evidence that suggests otherwise, the most defensible assumption in life-cycle assessment models may be that compost application does not positively or negatively impact CH_4 or N_2O fluxes from agricultural soils.

The degree to which compost application results in greater carbon sequestration or residence time in soils is highly uncertain. Soil organic matter, which generally refers to the organic fraction of soil excluding undecayed animal and plant matter, is particularly important to crop growth because it directly impacts nitrogen availability, soil water retention, and other physical soil properties.[35,39] However, the long-term stability of that material remains an open question; the soil science community began transitioning away from the concept of humus as stable and resistant to decomposition.[40] Researchers should exercise caution in using soil carbon sequestration factors for compost, particularly if the data is sourced from literature published prior to the recent shift in scientists' understanding of what kinds of molecules can be metabolized by the soil microbial community. Studies do suggest that when compost is applied to grasslands, the increase in net primary productivity results in an increase in above- and below-ground carbon stocks.[34,41] For example, in the analysis of compost application to California grasslands presented in Breunig et al. (2019), a one time application of compost was assumed to provide an additional 0–4.7 tonnes of soil carbon per hectare of amended land and the increase in soil carbon was estimated to last 30 years.[41] The question of how this accumulated carbon remains in soils remains largely unresolved because of limits in the scientific community's understanding of soil microbial communities. Even the lignin fraction of plants, which was long thought to limit decomposition of plant litter, can degrade more quickly than other components of plants under favorable conditions.[42] For this reason, it is challenging to recommend an approach to accounting for soil organic carbon impacts associated with compost application. As with CH_4 or N_2O fluxes, the most defensible assumption in the near-term may be that compost application does not result in any net accumulation of carbon in soils.

B4.2 Comparing Composting Emissions to Landfill Emissions

The most common alternatives to composting depend on both the type of material in need of management (e.g., OFMSW, manure, yard waste, digestate) and the country in which it is being managed. For example, the U.S. landfills most of its OFMSW while manure may be left on land or stored in open lagoons.[43,44] For OFMSW, there is a clear consensus among the majority of life-cycle emissions studies that composting organic waste results in lower net GHG emissions relative to landfilling.[4,45] There is far less consensus on the actual emissions footprint of landfilling different types of organic wastes.

The GHG footprint of landfills is dominated by fugitive CH_4 emissions, even for those with gas capture systems in place, and there are many different strategies for measuring these emissions.[46,47] Even with accurate measurements, it is difficult to draw a causal link between specific types and quantities of waste sent to landfills and the resulting emitted CH_4. For this reason, commonly used emission factors are based on lab experiments, in which anaerobic decomposition of specific types of materials is stimulated to determine decay rates and CH_4 generation, and those values are later adjusted to account for landfill gas capture rates.

One of the most frequently cited sources for GHG emission factors for landfilling is the U.S. Environmental Protection Agency's Waste Reduction Model (WARM),[48] which is based on experimental work by Barlaz (1998) and De la Cruz and Barlaz (2010).[49,50] However, these experiments simulated enhanced landfilling, where decomposition is purposefully accelerated. Barlaz (1998) measured methane emissions with the decay rate of material under optimal conditions in the laboratory. Organic materials were shredded, seeded, and incubated at 40 ºC, and were supplied with phosphate and ammonia to ensure that degradation was not nutrient-limited. Leachate (i.e., residual liquid) was also neutralized and subsequently recycled to the reactor to ensure that the pH did not slow microbial activity. These experimental decay rates represent an upper bound rather than what would occur in a typical industrial scale landfill, where waste does not routinely undergo shredding, nor are other measures taken to neutralize the pH or supply nutrients to speed up degradation. In a similar example, De la Cruz and Barlaz (2010) estimated feedstock specific decay rates using data from Eleazer et al. (1997),[51] which simulated enhanced landfilling like Barlaz (1998). Realistic decay rates are important for estimating fugitive emissions because, in landfills with gas capture systems, most fugitive CH_4 emissions occur in the time between when waste is placed in the landfill and the individual cell is capped. Faster decay rates result in more fugitive CH_4 emissions.

It is possible that reliance on measured values from enhanced landfilling experiments has resulted in systematic overestimation of CH_4 emissions and the extent of material degradation in landfills. For instance, based on Barlaz (1998), WARM uses an emission factor of 6.38×10^{-2} kg of CH_4 per wet kg of food waste landfilled, in contrast to the emission factor of 1.30×10^{-2} kg of CH_4 per wet kg of food waste as measured by Behera et al. (2010).[52] In the Behera et al. study, food waste leachate sourced from a food waste recovery plant was filtered through a sieve and directly fed into an anaerobic reactor in a lab. Food waste leachate, which is a dense liquid, is a suitable proxy for raw food waste, since it makes up 70–90% of the food waste material collected and stored at food waste recovery plants. The measured emission factor from this experiment was about 80% lower than that estimated by Barlaz. Because of the overestimation associated with enhanced landfilling in the Barlaz study, the reduced emission factor from Behera et al. (2010) was preferred for use in Nordahl et al. (2020).[4] Even when considering the reduced landfill emission factor, composting still presents as the favorable option for handling OFMSW, with a per-tonne CH_4 emission factor (8.79×10^{-4} kg of CH_4 per wet kg of OFMSW, Table 3.1) that is two orders of magnitude lower than that assumed by WARM or found in Behera et al. (2010) for landfilling.

B5 Ammonia Emissions

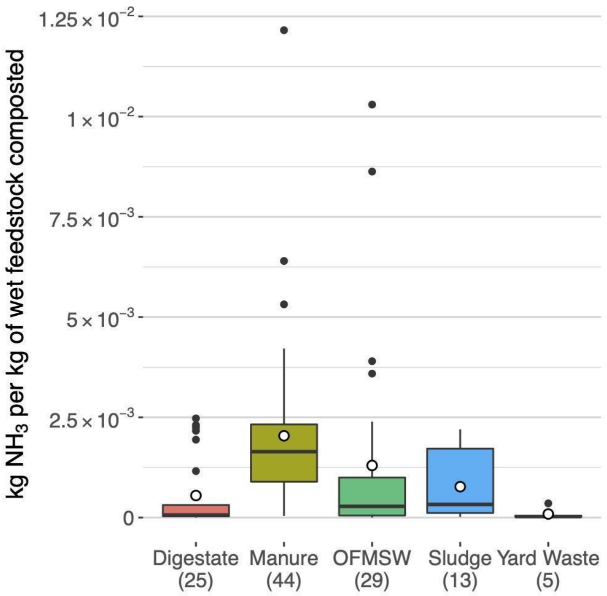

Figure B2. Distributions of NH_3 emission factors for composting reported in the literature

The sample size (n) of data points contributing to each boxplot is indicated in the x-axis labels. The mean values for the boxplot data is indicated by the open point symbols, while outliers are shown as closed circles.

References

(1) U.S. EPA. *Inventory of U.S. Greenhouse Gas Emissions and Sinks 1990-2018*; EPA 430-R-20–002; United States Environmental Protection Agency, 2019.

(2) Gilmore, E. A.; Heo, J.; Muller, N. Z.; Tessum, C. W.; Hill, J. D.; Marshall, J. D.; Adams, P. J. An inter-comparison of the social costs of air quality from reduced-complexity models. *Environmental Research Letters* **2019**, *14*, 074016.

(3) Tschofen, P.; Azevedo, I. L.; Muller, N. Z. Fine particulate matter damages and value added in the US economy. *Proc Natl Acad Sci USA* **2019**, *116*, 19857–19862.

(4) Nordahl, S. L.; Devkota, J. P.; Amirebrahimi, J.; Smith, S. J.; Breunig, H. M.; Preble, C. V.; Satchwell, A. J.; Jin, L.; Brown, N. J.; Kirchstetter, T. W.; et al. Life-cycle greenhouse gas emissions and human health trade-offs of organic waste management strategies. *Environ. Sci. Technol.* **2020**, *54*, 9200–9209.

(5) Büyüksönmez, F. Full-scale VOC emissions from green and food waste windrow composting. *Compost Sci. Util.* **2012**, *20*, 57–62.

(6) Zhu-Barker, X.; Bailey, S. K.; Paw U, K. T.; Burger, M.; Horwath, W. R. Greenhouse gas emissions from green waste composting windrow. *Waste Manag.* **2017**, *59*, 70–79.

(7) Kumar, A.; Alaimo, C. P.; Horowitz, R.; Mitloehner, F. M.; Kleeman, M. J.; Green, P. G. Volatile organic compound emissions from green waste composting: Characterization and ozone formation. *Atmos. Environ.* **2011**, *45*, 1841–1848.

(8) EPA. *Measurement of gaseous emission rates from land surfaces using an emission isolation flux chamber*; EPA/600/8-86/008; U.S. EPA, 1986.

(9) Czepiel, P.; Douglas, E.; Harriss, R.; Crill, P. Measurements of N_2O from composted organic wastes. *Environ. Sci. Technol.* **1996**, *30*, 2519–2525.

(10) Hellmann, B.; Zelles, L.; Palojarvi, A.; Bai, Q. Emission of climate-relevant trace gases and succession of microbial communities during open-windrow composting. *Appl. Environ. Microbiol.* **1997**, *63*, 1011–1018.

(11) Beck-Friis, B.; Pell, M.; Sonesson, U.; Jönsson, H.; Kirchmann, H. Formation and emission of N_2O and CH_4 from compost heaps of organic household waste. *Environmental Monitoring and Assessment* **2000**, *62*, 317–331.

(12) Hao, X.; Chang, C.; Larney, F. J. Carbon, nitrogen balances and greenhouse gas emission during cattle feedlot manure composting. *J. Environ. Qual.* **2004**, *33*, 37–44.

(13) Andersen, J. K.; Boldrin, A.; Samuelsson, J.; Christensen, T. H.; Scheutz, C. Quantification of greenhouse gas emissions from windrow composting of garden waste. *J. Environ. Qual.* **2010**, *39*, 713–724.

(14) Colón, J.; Cadena, E.; Pognani, M.; Barrena, R.; Sánchez, A.; Font, X.; Artola, A. Determination of the energy and environmental burdens associated with the biological treatment of source-separated Municipal Solid Wastes. *Energy Environ. Sci.* **2012**, *5*, 5731–5741.

(15) Chen, R.; Wang, Y.; Wang, W.; Wei, S.; Jing, Z.; Lin, X. N_2O emissions and nitrogen transformation during windrow composting of dairy manure. *J. Environ. Manage.* **2015**, *160*, 121–127.

(16) Hellebrand, H. J. Emission of nitrous oxide and other trace gases during composting of grass and green waste. *Journal of Agricultural Engineering Research* **1998**, 265–275.

(17) Amon; Amon, Th.; Boxberger; Alt, Ch. Emissions of NH_3, N_2O and CH_4 from dairy cows housed in a farmyard manure tying stall (housing, manure storage, manure spreading). *Nutrient Cycling in Agroecosystems* **2001**.

(18) Fukumoto, Y.; Osada, T.; Hanajima, D.; Haga, K. Patterns and quantities of NH3, N2O and CH4 emissions during swine manure composting without forced aeration—effect of compost pile scale. *Bioresour. Technol.* **2003**, *89*, 109–114.

(19) Paillat, J.-M.; Robin, P.; Hassouna, M.; Leterme, P. Predicting ammonia and carbon dioxide emissions from carbon and nitrogen biodegradability during animal waste composting. *Atmos. Environ.* **2005**, *39*, 6833–6842.

(20) Amlinger, F.; Peyr, S.; Cuhls, C. Greenhouse gas emissions from composting and mechanical biological treatment. *Waste Manag. Res.* **2008**, *26*, 47–60.

(21) Maeda, K.; Hanajima, D.; Morioka, R.; Toyoda, S.; Yoshida, N.; Osada, T. Mitigation of greenhouse gas emission from the cattle manure composting process by use of a bulking agent. *Soil Sci. Plant Nutr.* **2013**, *59*, 96–106.

(22) Hrad, M.; Binner, E.; Piringer, M.; Huber-Humer, M. Quantification of methane emissions from full-scale open windrow composting of biowaste using an inverse dispersion technique. *Waste Manag.* **2014**, *34*, 2445–2453.

(23) Vergara, S. E.; Silver, W. L. Greenhouse gas emissions from windrow composting of organic wastes: Patterns and emissions factors. *Environmental Research Letters* **2019**, *14*, 124027.

(24) Preble, C. V.; Chen, S. S.; Hotchi, T.; Sohn, M. D.; Maddalena, R. L.; Russell, M. L.; Brown, N. J.; Scown, C. D.; Kirchstetter, T. W. Air pollutant emission rates for dry anaerobic digestion and composting of organic municipal solid waste. *Environ. Sci. Technol.* **2020**, *54*, 16097–16107.

(25) Brown, S.; Kruger, C.; Subler, S. Greenhouse gas balance for composting operations. *J. Environ. Qual.* **2008**, *37*, 1396–1410.

(26) Jiang, T.; Schuchardt, F.; Li, G.; Guo, R.; Zhao, Y. Effect of C/N ratio, aeration rate and moisture content on ammonia and greenhouse gas emission during the composting. *Journal of Environmental Sciences* **2011**, *23*, 1754–1760.

(27) Chowdhury, M. A.; de Neergaard, A.; Jensen, L. S. Potential of aeration flow rate and bio-char addition to reduce greenhouse gas and ammonia emissions during manure composting. *Chemosphere* **2014**, *97*, 16–25.

(28) Sun, X.; Lu, P.; Jiang, T.; Schuchardt, F.; Li, G. Influence of bulking agents on CH_4, N_2O, and NH_3 emissions during rapid composting of pig manure from the Chinese

Ganqinfen system. *J. Zhejiang Univ. Sci. B* **2014**, *15*, 353–364.

(29) Unable to find information for 6489453.

(30) Boldrin, A.; Andersen, J. K.; Møller, J.; Christensen, T. H.; Favoino, E. Composting and compost utilization: accounting of greenhouse gases and global warming contributions. *Waste Manag. Res.* **2009**, *27*, 800–812.

(31) CalRecycle. *SB 1383 Infrastructure and Market Analysis*; California Department of Resources Recycling and Recovery, 2019.

(32) Al-Bataina, B. B.; Young, T. M.; Ranieri, E. Effects of compost age on the release of nutrients. *International Soil and Water Conservation Research* **2016**, *4*, 230–236.

(33) Sullivan, D. M.; Bary, A. I.; Nartea, T. J.; Myrhe, E. A.; Cogger, C. G.; Fransen, S. C. Nitrogen availability seven years after a high-rate food waste compost application. *Compost Sci. Util.* **2003**, *11*, 265–275.

(34) Favoino, E.; Hogg, D. The potential role of compost in reducing greenhouse gases. *Waste Manag. Res.* **2008**, *26*, 61–69.

(35) Weindorf, D. C.; Muir, J. P.; Landeros-Sánchez, C. Organic compost and manufactured fertilizers: economics and ecology. In *Integrating Agriculture, Conservation and Ecotourism: Examples from the Field*; Campbell, W. B.; Lopez Ortiz, S., Eds.; Issues in agroecology – present status and future prospectus; Springer Netherlands: Dordrecht, 2011; Vol. 1, pp. 27–53.

(36) Eggerth, L. L.; Diaz, L. F.; Chang, M. T. F.; Iseppi, L. Chapter 12 Marketing of composts. In *Compost science and technology*; Waste Management Series; Elsevier, 2007; Vol. 8, pp. 325–355.

(37) Dambreville, C.; Hallet, S.; Nguyen, C.; Morvan, T.; Germon, J.-C.; Philippot, L. Structure and activity of the denitrifying community in a maize-cropped field fertilized with composted pig manure or ammonium nitrate. *FEMS Microbiol. Ecol.* **2006**, *56*, 119–131.

(38) Ryals, R.; Silver, W. L. Effects of organic matter amendments on net primary productivity and greenhouse gas emissions in annual grasslands. *Ecol. Appl.* **2013**, *23*, 46–59.

(39) Johnston, A. E. Soil organic matter, effects on soils and crops. *Soil Use & Management* **1986**, *2*, 97–105.

(40) Lehmann, J.; Kleber, M. The contentious nature of soil organic matter. *Nature* **2015**, *528*, 60–68.

(41) Breunig, H. M.; Amirebrahimi, J.; Smith, S.; Scown, C. D. Role of digestate and biochar in carbon-negative bioenergy. *Environ. Sci. Technol.* **2019**, *53*, 12989–12998.

(42) Hall, S. J.; Huang, W.; Timokhin, V. I.; Hammel, K. E. Lignin lags, leads, or limits the decomposition of litter and soil organic carbon. *Ecology* **2020**, *101*, e03113.

(43) Niles, M. T.; Wiltshire, S. Tradeoffs in US dairy manure greenhouse gas emissions, productivity, climate, and manure management strategies. *Environ. Res. Commun.* **2019**,

1, 075003.

(44) EPA. *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-2020 – Annex 3 Part B*; U.S. Environmental Protection Agency, 2022.

(45) Lou, X. F.; Nair, J. The impact of landfilling and composting on greenhouse gas emissions--a review. *Bioresour. Technol.* **2009**, *100*, 3792–3798.

(46) Mønster, J.; Kjeldsen, P.; Scheutz, C. Methodologies for measuring fugitive methane emissions from landfills - A review. *Waste Manag.* **2019**, *87*, 835–859.

(47) Duren, R. M.; Thorpe, A. K.; Foster, K. T.; Rafiq, T.; Hopkins, F. M.; Yadav, V.; Bue, B. D.; Thompson, D. R.; Conley, S.; Colombi, N. K.; et al. California's methane super-emitters. *Nature* **2019**, *575*, 180–184.

(48) EPA. Waste Reduction Model (WARM). **2018**.

(49) Barlaz, M. A. Carbon storage during biodegradation of municipal solid waste components in laboratory-scale landfills. *Global Biogeochem. Cycles* **1998**, *12*, 373–380.

(50) De la Cruz, F. B.; Barlaz, M. A. Estimation of waste component-specific landfill decay rates using laboratory-scale decomposition data. *Environ. Sci. Technol.* **2010**, *44*, 4722–4728.

(51) Eleazer, W. E.; Odle, W. S.; Wang, Y.-S.; Barlaz, M. A. Biodegradability of Municipal Solid Waste Components in Laboratory-Scale Landfills. *Environ. Sci. Technol.* **1997**, *31*, 911–917.

(52) Behera, S. K.; Park, J. M.; Kim, K. H.; Park, H.-S. Methane production from food waste leachate in laboratory-scale simulated landfill. *Waste Manag.* **2010**, *30*, 1502–1508.

Appendix C

Supplementary Information

Complementary Roles for Mechanical and Solvent-Based Recycling in Low-Carbon, Circular Polypropylene

C1 Virgin Polypropylene Production

Virgin polypropylene production involves two main processes: propylene monomer synthesis and conversion to polypropylene (PP). Propylene production happens alongside petroleum refining and the production of several other products, including other olefins like ethylene. In fact, propylene is often considered a byproduct of ethylene production.[1] The main production process is thermal cracking or steam pyrolysis of fossil feedstocks.[2] Propane is cracked to produce propylene and ethane is cracked to make ethylene. The "cracking" refers to the breaking of C-H bonds to allow for double bonds.

The process starts with feeding saturated hydrocarbons and steam to a hydrocracker where they are heated to ~1000ºC.[2] In the case of olefin production, lighter phase feedstocks like LPG or light naphtha are preferable.[1] To stop the reaction after sufficient heating, the cracked products are cooled with heat exchangers generating high pressure steam. The gas stream goes through a centrifugal compressor to remove fuel oil and then undergoes hydrogen sulfide removal. Lastly, fractional distillation separates the reaction products.

Propylene is converted into PP via free-radical polymerization usually with Ziegler-Natta (Z-N) or metallocene catalysts.[1,2] There are multiple processes used by industry to make polypropylene including gas-phase polymerization and solution or liquid-phase polymerization.[2] In gas-phase polymerization, propylene vapor is mixed with the catalyst in a fluidized bed reactor.[2,3] The reactor is typically kept at 80-90ºC with a pressure of 90-25 atm. Any gaseous propylene that does not react is added back to the feed stream. The result of the reaction is solid PP which is then dried and pelletized. Liquid-phase polymerization is conceptually similar except the feedstock propylene is liquid and the reactor is tubular.

We use mass and energy flow data for PP production, inclusive of both propylene synthesis and conversion to polymer, from a 2011 American Chemistry Council (ACC) report for analysis.[2]

C2 Sourcing PP Waste for Recycling Feedstocks

C2.1 Plastic Waste Sorting

Our analysis begins with plastic waste sorting at an MRF. The efficacy and associated benefits of most recycling processes are highly dependent on feedstock composition and require waste materials to be sorted for reclamation.[4] Initial sorting occurs at MRFs, where recyclable waste is separated by material type (e.g., plastics, fibers, metals, glass). Generally, MRFs also separate plastics by polymer type using optical sorters and near infrared (NIR) technology. Most MRFs in the U.S. today primarily target and bale PET and high-density polyethylene (HDPE), with a particular focus on bottles, during sorting.[5,6] Other polymers, including PP, can also be targeted for selective recovery using the same NIR technology. However, PP is more commonly baled with other non-PET, non-HDPE plastics in mixed bales. These mixed bales are typically called #3-7 bales referring to the resin identification codes for polyvinyl chloride (#3), low-density polyethylene (#4), PP (#5), polystyrene (#6), and other plastics (#7). While #1 and #2 plastics, PET and HDPE respectively, are typically targeted for separate recovery, sorting efficiencies at MRFs are imperfect and some of these materials may end up in Mixed #3-7 bales. Mixed #3–7 bales include rigid product forms but exclude film plastics, which are harder to separate and recycle.

To represent current practices across most of the U.S., we assume the initial input to any PP recycling process is a mixed #3–7 bale (Figure C1) rather than a PP-specific waste bale from an MRF. Non-target (non-PP) materials are then separated out by float-sink separation and routed for disposal. If a wider variety of polymer types, including PP, become attractive candidates for recycling, MRFs may add new “lines” to recover these materials separately. Alternatively, so-called secondary MRFs may be constructed to take in #3–7 bales and further separate the material [7]. Neither of these developments will dramatically impact the energy footprint of recycling. The energy footprint of sorting at MRFs (4.7–7.8 kWh of electricity per tonne of waste throughput) is small compared to the thermal and electrical energy needed during mechanical or advanced recycling processes.[4] Because only 3.7% of current MRF throughput are #3–7 plastics,[8] we assume the MRF energy and GHG impacts that can be allocated to #3–7 bales are negligible.

C2.2 Feedstock Type

The assumed input for all of our recycling scenarios is a mixed #3-7 bale, the most typical bale containing PP available from MRFs. We initially assumed the bale composition reported by the Association of Plastic Recyclers (APR) in a 2017 presentation,[9] which was derived from a 2015 APR report which goes into further detail on MRF bale compositions.[10] This included an “Other” category of 11% but instead of having unknowns, we assumed the maximum allowable contamination of paper (2%), metals (assumed to be aluminum) (2%), and liquids (assumed to be water) (1%) as given by the APR's model bale specifications.[11] The remaining unknown component (a total of 6%) was distributed evenly to PET, HDPE and PP. The final composition used for this study is presented in Figure C1 below. In reality, bales will vary considerably from MRF to MRF. Furthermore, sorting operations and subsequently, the composition of outgoing MRF bales, are constantly evolving with changing market conditions; as the price of different materials and polymers change, MRFs modify operational configurations and bring in new equipment to recover valuable, salable material.

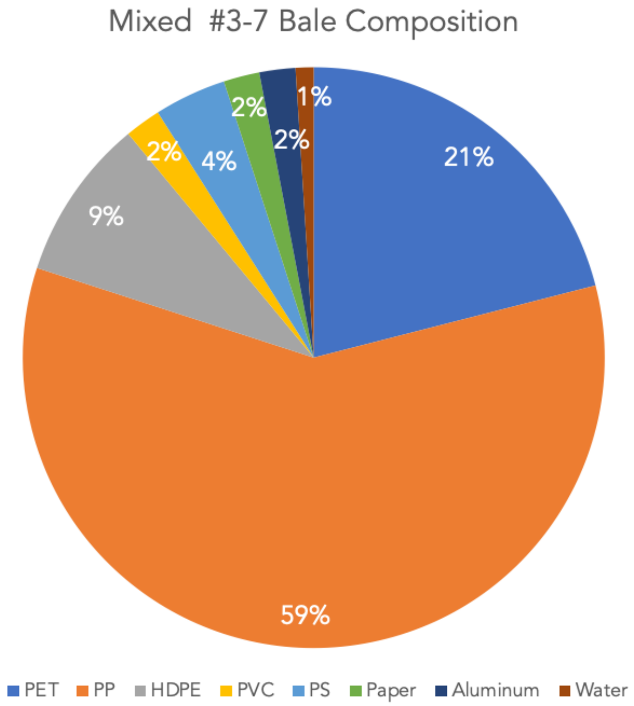

Figure C1. Mixed #3-7 bale composition
Adapted from 2015 APR report.[9]

C2.3 Feedstock Transportation

We assume that on average, transportation of material from site of sorting/separation (i.e. MRFs) to PP recyclers (mechanical or otherwise) includes 0.66 tonne-km of truck transport and 0.25 tonne-km of rail transport per kg of incoming material to recyclers.[12] We assume flatbed trucking for truck transport.

C3 Mechanical Recycling

C3.1 Limitations of mechanical recyclate

PP can only undergo traditional mechanical recycling a few times before irrevocably degrading to unusable materials in circular manufacturing. PP has methyl groups on every other carbon atom, making it prone to chain scission during mechanical grinding and high shear rates during melt processing.[13] In contrast, polyethylene (PE), the most common plastic, can withstand over 30 recycling cycles.[13,14] Repeated mechanical recycling of PP results in reduced molecular weight, increased crystallinity, reduced impact resistance, and increased opacity.[15–19] Because of this physical degradation and difficult-to-separate impurities in plastic waste streams, the material produced from mechanical recycling is usually of lower quality than virgin resin.[20] Mechanically recycled PP has inferior physicochemical properties than virgin PP. For most applications, PP recyclate must be blended with virgin polymer to ensure sufficient material performance.[15,21–23] For some uses including automotive applications and food-safe packaging, which have stricter thermal, chemical, and mechanical property requirements for safety reasons, PP recyclate can be deemed entirely unsuitable.[20,22,24,25]

C3.2 Substitution factors for mechanical recyclate

Some studies account for the imperfect substitution between mechanically recycled and virgin PP by using a substitution factor (Figure C2). For example, 1 kg of mechanically recycled PP can be treated as functionally equivalent to 0.7–1.0 kg of virgin PP.[26–32] Some studies may use terms like "substitution factor" or "substitution ratio" in reference to technical recovery efficiency of a recycling process (e.g., mass maintained versus lost during recycling process),[27,33,34] market substitution factor (e.g., additional material required to make a given product with lower-grade recyclates versus virgin-grade polymer),[26,27,31,33,34] value-corrected market substitution factor or displacement rate (e.g., price-based ratio describing market uptake of recyclate),[30,34,35] and blending limits (e.g., when blending with virgin material, the maximum allowable recycled content to avoid excessive quality loss) (Figure C2).[23] For the purposes of conducting a life-cycle assessment, clearly defining an appropriate functional unit (i.e., virgin material displaced per unit of waste plastic versus per unit of recyclate) is critical.

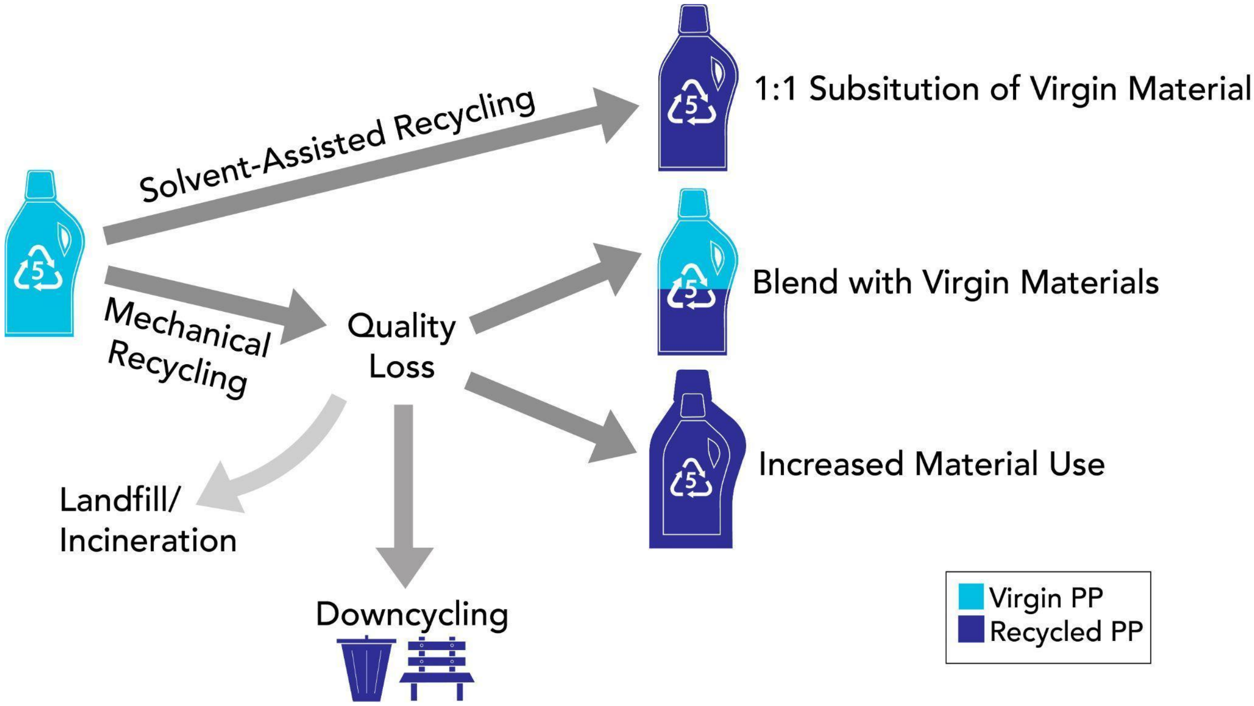

Figure C2. Substituting virgin polypropylene with recycled polypropylene
This figure depicts how recycled material can substitute virgin material use and the limits to perfect substitution for mechanical recyclates. Note that both recycling processes have mass losses which are not depicted in this figure; only loss to quality is included.

This study does not apply a substitution factor because these values are uncertain and product-specific. There are no currently available, robust estimates for how much virgin resin production has actually been offset by recyclates.[36] While technical process efficiencies are well understood and can be measured directly for a particular recycling method, market substitution factors and blending limits are under-reported and variable depending on application and product type.[37] Furthermore, the amount of recycled PP that the market could absorb is likely still much greater than the quantity available today. Until the industry-wide capacity for blending recycled PP has been reached for all product and application types, recycled PP can offset virgin material use on a 1:1 basis unless increased material use is also required (Figure C2).

C3.3 Process Modeling

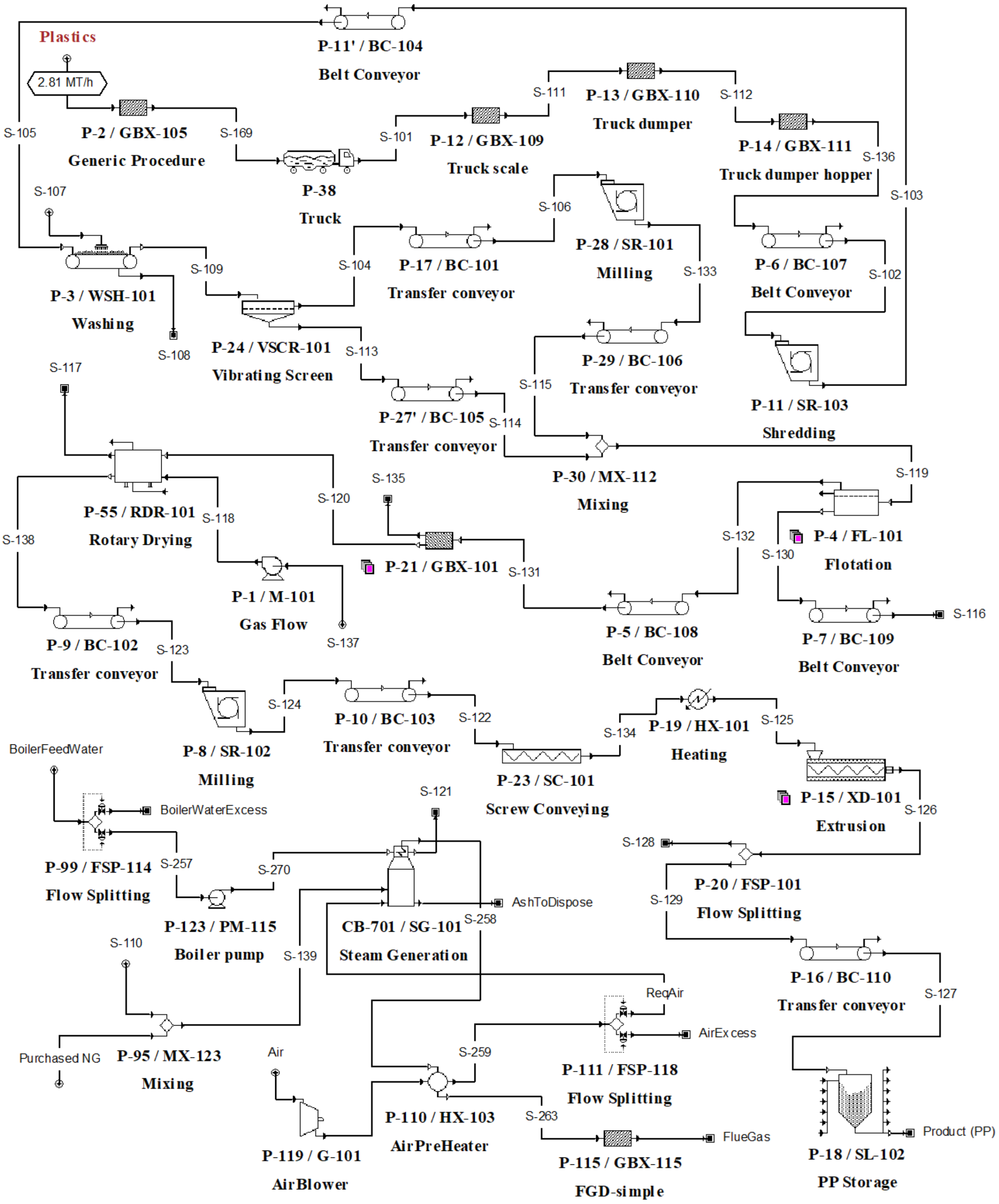

Figure C3. Overview of process model developed in SuperPro Designer

C3.4 Energy Consumption Results

The energy consumption results, normalized in units of kWh per tonne, are presented in Figure C4 and Table C1 below. Our modeling efforts in SuperPro Designer yield full material and energy balance data for mechanical recycling processes. These results are only used in the LCA of solvent-assisted recycling to model pretreatment to PP dissolution; they are not used in the LCA of PP mechanical recycling because real-life facility-scale data is available in literature.[12]

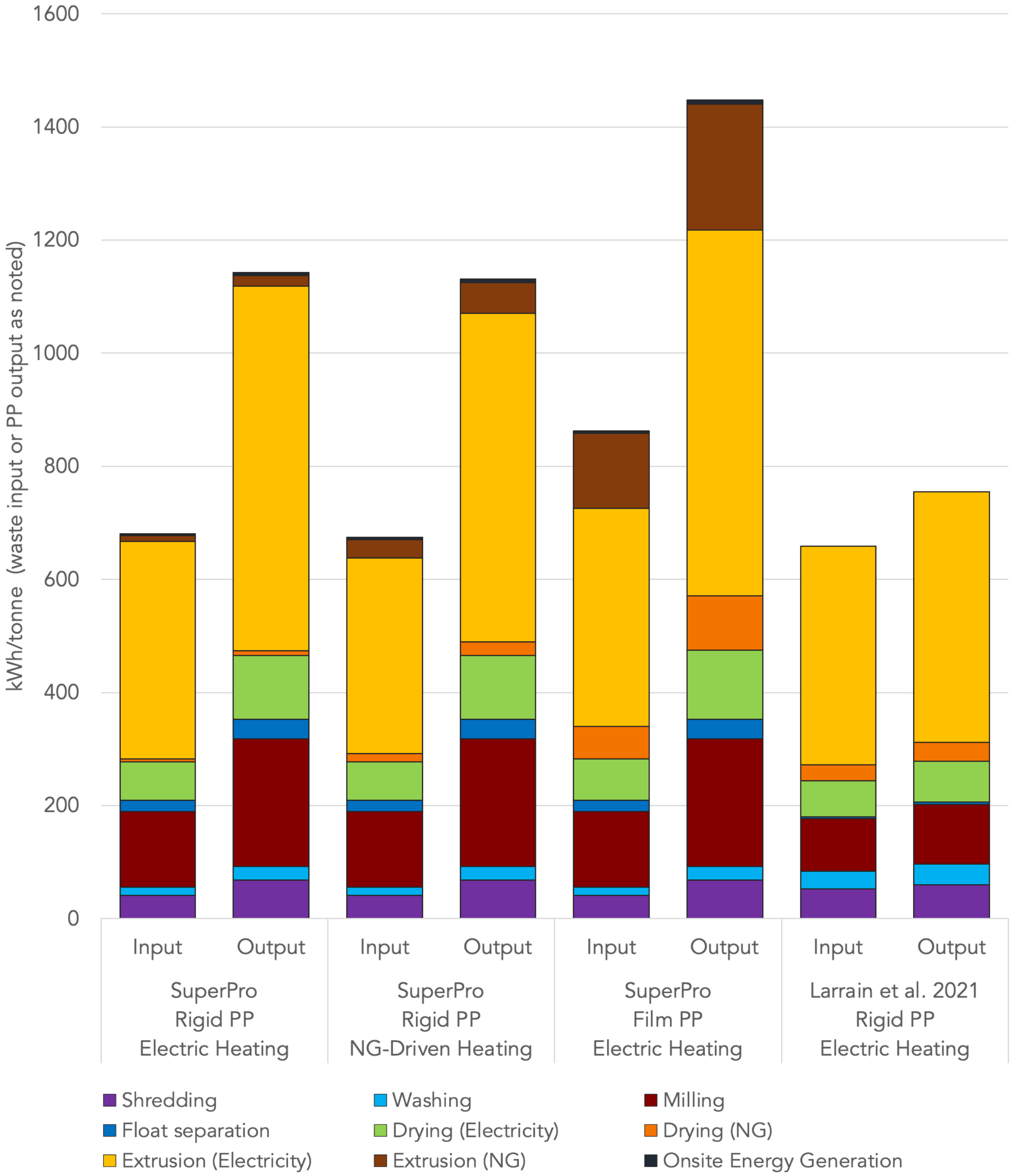

Figure C4. Mechanical recycling energy consumption by unit process

Table C1. Mechanical recycling energy consumption by unit process (tabulated data for Figure C4)

Model	Feedstock	Heating Scenario	Shredding	Washing	Milling	Float separation	Drying (Electricity)	Drying (NG)	Extrusion (Electricity)	Extrusion (NG)	Onsite Energy Generation	Total	Units
SuperPro	Rigid PP	Electric Heating	41	15	134	20	68	5	385	11	3	681	kWh/tonne input
			68	25	225	34	114	8	645	18	6	1142	kWh/tonne output
SuperPro	Rigid PP	NG-Driven Heating	41	15	134	20	68	14	346	33	3	674	kWh/tonne input
			68	25	225	34	114	24	581	55	6	1131	kWh/tonne output
SuperPro	Film PP	Electric Heating	41	15	134	20	73	57	386	133	4	863	kWh/tonne input
			68	25	225	34	123	96	647	223	7	1447	kWh/tonne output
Larrain et al. 2021	Rigid PP	Electric Heating	53	32	92	4	63	28	387	0	0	659	kWh/tonne input
			60	36	106	4	72	33	443	0	0	754	kWh/tonne output

C4 Life Cycle Assessment

C4.1 LCA Scenarios

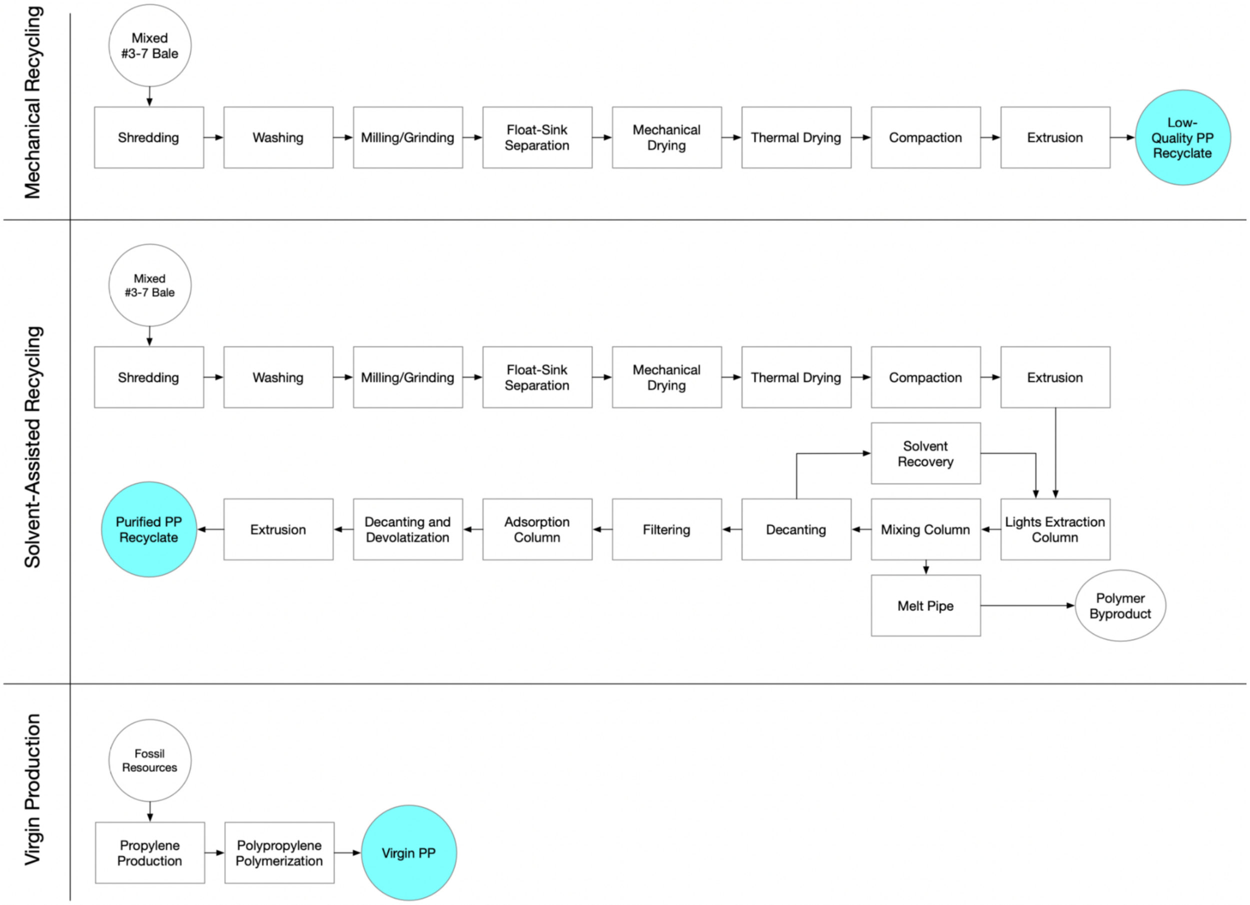

Figure C5. LCA scenario processes

Table C2. Life-cycle inventory (LCI) descriptions by scenario

Scenario	Type of Direct Requirements
Mechanical Recycling (main data source: [12])	Transportation (trucking and rail), Energy (electricity and natural gas), Washing agent (NaOH)
Solvent-Assisted Recycling (main data source is confidential, but reflects pilot-scale operations in the U.S.)	Transportation (trucking and rail), Energy (electricity and natural gas), Solvent (butane), Other process materials (NaOH, silica, alumina), Antioxidant additives (assume pentaerythritol and irgafos 168 as proxies), Antistatic additives (assume stearic acid as proxy),

	Crude oil offset (polymer byproduct stream is assumed to offset crude oil production based on energy content)
Virgin Production (main data source: [2])	Raw material extraction (crude oil and natural gas), Energy (electricity, natural gas, other petroleum-based fuels), Transportation (trucking and barge)

C4.2 LCA Model: Calculations

The basic math upon which the LCA model is built is provided below:

$$(I - A)X = Y$$

N = total number of unit processes
A = NxN input-output matrix with life-cycle inventories for each unit process (non-zero data listed in Table C4)
I = NxN identity matrix
Y = N length vector w/ direct requirements for scenario analysis
X = N length vector w/ life-cycle requirements for scenario analysis
E = N length vector w/ emission factors for each unit process (emission factors by pollutant type are provided in Table C3)

$$X = \begin{bmatrix} x_i \vdots x_N \end{bmatrix} \qquad E = \begin{bmatrix} e_i \vdots e_N \end{bmatrix}$$

$$LifeCycle\ Emissions = \sum_i^N x_i * e_i$$

C4.3 LCA Model: Data

Table C3. LCA model: emission factors

All values are in units of kg of pollutant (given by column name) per unit indicated in the unit process name. These are not necessarily life-cycle emission factors and only definitely include direct emission impacts (in several cases, this means fugitive emissions associated with fuel combustion); full life-cycle impacts must be assessed through the model using this data along with IO data from Table C4. When possible, GHG emission factors are separated by pollutant type; in cases where this is not possible, total GHG impact in CO_2 equivalence is given by the CO_2 column while the CH_4 and N_2O columns are marked with zeros.

Unit Process	CO_2	CH_4	N_2O	Source
alumina.kg	1.93E+00	3.54E-03	4.70E-05	GREET 2020 [38]
barge.mt_km	2.22E-02	0.00E+00	0.00E+00	Cohon et al. 2010 [39]
butane.MJ	5.16E-03	2.24E-07	4.35E-08	GREET 2021 [40]
caco3.kg	1.61E-03	5.15E-08	3.19E-08	GREET 2020 [38]

coal.MJ	6.97E-04	1.39E-04	8.73E-09	GREET 2018 [41]
crudeoil.MJ	2.62E-03	8.24E-05	3.12E-08	GREET 2019 [42]
diesel.MJ	5.09E-03	3.93E-08	2.02E-07	GREET 2020 [38]
electricity.MRO.kWh	6.03E-01	7.54E-05	1.09E-05	Ou and Cai 2020 [43]
electricity.NGCC.kWh	4.05E-01	7.46E-06	7.36E-07	Ou and Cai 2020 [43]
electricity.nuclear.kWh	0.00E+00	0.00E+00	0.00E+00	NA
electricity.TRE.kWh	4.20E-01	3.60E-05	5.00E-06	Ou and Cai 2020 [43]
electricity.US.kWh	4.05E-01	7.00E-06	4.00E-06	Ou and Cai 2020 [43]
electricity.WECC.kWh	3.27E-01	3.41E-05	5.03E-06	Ou and Cai 2020 [43]
flatbedtruck.mt_km	1.24E-01	0.00E+00	0.00E+00	Cohon et al. 2010 [39]
formaldehyde.kg	0.00E+00	0.00E+00	0.00E+00	GREET 2021 [40]
gasoline.MJ	9.23E-03	3.12E-06	8.64E-08	Lu et al. 2016 [44]
gaspipeline.mt_km	0.00E+00	0.00E+00	0.00E+00	NA
glycerin.kg	1.91E-01	4.90E-04	2.69E-06	GREET 2018 [41]
h2.kg	1.06E+01	5.98E-02	4.00E-05	Spath and Mann 2001 [45]
irgafos168.kg	4.68E-01	0.00E+00	0.00E+00	Assumed tris(2,4-ditert-butylphenyl) phosphite as proxy; Ecoinvent v3.9 [46]
landfill_inorganics_wet.kg	0.00E+00	0.00E+00	0.00E+00	WARM v14 [47]
landfill_mixedMSW.wet_kg	0.00E+00	2.87E-02	0.00E+00	WARM v14 [47]
landfill_mixedorganics.wet_kg	0.00E+00	2.18E-02	0.00E+00	WARM v14 [47]
lime.kg	1.11E+00	9.54E-07	9.86E-08	GREET 2020 [38]
liquidpipeline.mt_km	0.00E+00	0.00E+00	0.00E+00	NA
lpg.kg	5.22E-01	1.45E-03	8.28E-06	GREET 2021 [40]
marinetanker.mt_km	6.91E-03	8.02E-08	0.00E+00	Cohon et al. 2010 [39]
methanol.kg	4.05E-01	4.67E-03	7.19E-06	GREET 2021 [40]
na_brine.kg	5.47E-02	1.19E-06	6.20E-07	GREET 2020 [38]
naoh.kg	4.68E-01	8.09E-06	4.32E-06	GREET 2020 [38]
naturalgas_combust.MJ	5.04E-02	0.00E+00	0.00E+00	Based on stoichiometry of methane combustion; Assumes perfect oxidation
naturalgas_select.MJ	0.00E+00	0.00E+00	0.00E+00	NA
naturalgas.conventional.MJ	4.06E-03	8.70E-05	2.60E-08	GREET 2020 [38]
naturalgas.shale.MJ	3.80E-03	9.19E-05	2.59E-08	GREET 2020 [38]
o2.kg	0.00E+00	0.00E+00	0.00E+00	NA
pentaerythritol.kg	3.31E-01	0.00E+00	0.00E+00	Ecoinvent v3.9 [46]
rail.mt_km	1.86E-02	0.00E+00	0.00E+00	Cohon et al. 2010 [39]
refgas.MJ	0.00E+00	0.00E+00	0.00E+00	NA
rfo.MJ	4.00E-03	8.74E-06	6.83E-08	GREET 2019 [42]
silica.kg	0.00E+00	0.00E+00	0.00E+00	GREET 2022 [48]
stearic_acid.kg	1.08E-01	0.00E+00	0.00E+00	Ecoinvent v3.9 [46]

tankertruck.mt_km	8.46E-02	0.00E+00	0.00E+00	Cohon et al. 2010 [39]
uranium.kg	1.12E+01	0.00E+00	0.00E+00	Parker et al. 2016 [49]
LPG_combust_industrialboiler.MJ	6.45E-02	1.01E-06	4.56E-06	GREET 2020 [38]
diesel_combust_industrialboiler.MJ	7.41E-02	1.88E-07	8.70E-07	GREET 2020 [38]
residualoil_combust_industrialboiler.MJ	8.06E-02	3.06E-06	1.62E-06	GREET 2020 [38]
gasoline_combust_industrialboiler.MJ	6.88E-02	2.84E-06	5.69E-07	GREET 2020 [38]
offgas_combust_industrialboiler.MJ	5.77E-02	3.04E-06	5.88E-07	GREET 2020 [38]

Table C4. LCA model: input-output matrix relationships

Our LCA model uses a physical units-based input-output matrix that is populated with life-cycle inventories for each unit process/product included. The relevant non-zero values are included in this table. Each unit process/product is listed with a unit. The value indicates the amount of the upstream/downstream requirement in its listed unit required to make 1 unit of the primary unit product/process. If any unit processes from Table C3 is not included in this table, there are no appreciable upstream/downstream impacts for that parameter.

Unit Process	Upstream/Downstream Requirements	Value**	Source
electricity.US.kWh			
	coal.MJ	2.59E+00	Ou and Cai 2020 [43]
	diesel.MJ	6.75E-03	Ou and Cai 2020 [43]
	rfo.MJ	4.51E-02	Ou and Cai 2020 [43]
	electricity.US.kWh	4.90E-02	Ou and Cai 2020 [43]
	naturalgas_select.MJ	2.77E+00	Ou and Cai 2020 [43]
	electricity.nuclear.kWh	2.15E-01	Ou and Cai 2020 [43]
flatbedtruck.mt_km			
	diesel.MJ	1.78E+00	Cohon et al. 2010 [39]
	flatbedtruck.mt_km	2.50E-01	Cohon et al. 2010 [39]
rail.mt_km			Cohon et al. 2010 [39]
	diesel.MJ	2.68E-01	Cohon et al. 2010 [39]
	rail.mt_km	2.50E-01	Cohon et al. 2010 [39]
naturalgas_select.MJ			
	naturalgas.conventional.MJ	8.35E-01	Burnham 2018 [50]
	naturalgas.shale.MJ	1.65E-01	Burnham 2018 [50]
diesel.MJ			
	rfo.MJ	3.11E-02	GREET 2020 [38]
	refgas.MJ	5.81E-02	GREET 2020 [38]
	crudeoil.MJ	1.00E+00	GREET 2020 [38]
	electricity.US.kWh	8.93E-04	GREET 2020 [38]

	h2.kg	1.08E-04	GREET 2020 [38]
	naturalgas_select.MJ	5.19E-02	GREET 2020 [38]
	tankertruck.mt_km	3.53E-03	GREET 2020 [38]
	liquidpipeline.mt_km	2.10E-02	GREET 2020 [38]
	butane.MJ	9.92E-05	GREET 2020 [38]
naoh.kg			
	coal.MJ	6.01E-01	GREET 2020 [38]
	rfo.MJ	2.11E-02	GREET 2020 [38]
	electricity.US.kWh	1.67E+00	GREET 2020 [38]
	naturalgas_select.MJ	7.28E+00	GREET 2020 [38]
	tankertruck.mt_km	4.26E-01	GREET 2020 [38]
	rail.mt_km	1.26E+00	GREET 2020 [38]
	na_brine.kg	5.83E+00	GREET 2020 [38]
butane.MJ			
	naturalgas_select.MJ	1.00E+00	NA
crudeoil.MJ			
	diesel.MJ	3.06E-03	GREET 2018 [41]
	rfo.MJ	2.04E-04	GREET 2018 [41]
	crudeoil.MJ	2.04E-04	GREET 2018 [41]
	electricity.US.kWh	1.08E-03	GREET 2018 [41]
	gasoline.MJ	4.08E-04	GREET 2018 [41]
	naturalgas_select.MJ	1.26E-02	GREET 2018 [41]
	tankertruck.mt_km	5.49E-03	GREET 2018 [41]
	liquidpipeline.mt_km	2.85E-02	GREET 2018 [41]
	rail.mt_km	2.42E-02	GREET 2018 [41]
	barge.mt_km	4.39E-03	GREET 2018 [41]
	marinetanker.mt_km	1.10E-01	GREET 2018 [41]
silica.kg			
	electricity.US.kWh	5.17E-05	GREET 2022 [48]
	flatbedtruck.mt_km	8.00E-02	General assumption (50 mi by truck)
	rail.mt_km	8.00E-01	General assumption (500 mi by rail)
pentaerythritol.kg			
	electricity.US.kWh	4.16E-01	Ecoinvent v3.9 [46]
	naturalgas_select.MJ	4.00E+00	Ecoinvent v3.9 [46]
	formaldehyde.kg	1.04E+00	Ecoinvent v3.9 [46]
irgafos168.kg			
	electricity.US.kWh	1.25E+00	Assumed tris(2,4-ditert-butylphenyl) phosphite as proxy; Ecoinvent v3.9 [46]

	naturalgas_select.MJ	6.45E+00	Assumed tris(2,4-ditert-butylphenyl) phosphite as proxy; Ecoinvent v3.9 [46]
stearic_acid.kg			
	glycerin.kg	-1.08E-01	Ecoinvent v3.9 [46]
	electricity.US.kWh	4.16E-01	Ecoinvent v3.9 [46]
	naturalgas_select.MJ	2.15E+00	Ecoinvent v3.9 [46]
electricity.TRE.kWh			
	coal.MJ	2.04E+00	Ou and Cai 2020 [43]
	diesel.MJ	1.91E-04	Ou and Cai 2020 [43]
	rfo.MJ	9.01E-03	Ou and Cai 2020 [43]
	electricity.TRE.kWh	4.90E-02	Ou and Cai 2020 [43]
	naturalgas_select.MJ	4.03E+00	Ou and Cai 2020 [43]
	electricity.nuclear.kWh	1.13E-01	Ou and Cai 2020 [43]
rfo.MJ			
	rfo.MJ	2.70E-02	GREET 2019 [42]
	refgas.MJ	3.73E-02	GREET 2019 [42]
	crudeoil.MJ	1.00E+00	GREET 2019 [42]
	electricity.US.kWh	3.71E-04	GREET 2019 [42]
	h2.kg	1.20E-05	GREET 2019 [42]
	naturalgas_select.MJ	2.43E-02	GREET 2019 [42]
	tankertruck.mt_km	3.53E-03	GREET 2019 [42]
	liquidpipeline.mt_km	2.10E-02	GREET 2019 [42]
	butane.MJ	7.20E-05	GREET 2019 [42]
gasoline.MJ			
	rfo.MJ	9.28E-02	Lu et al. 2016 [44]
	refgas.MJ	9.26E-02	Lu et al. 2016 [44]
	crudeoil.MJ	1.00E+00	Lu et al. 2016 [44]
	electricity.US.kWh	1.44E-02	Lu et al. 2016 [44]
	h2.kg	5.26E-05	Lu et al. 2016 [44]
	naturalgas_select.MJ	6.27E-02	Lu et al. 2016 [44]
	tankertruck.mt_km	3.53E-03	Lu et al. 2016 [44]
	liquidpipeline.mt_km	2.10E-02	Lu et al. 2016 [44]
	butane.MJ	6.44E-02	Lu et al. 2016 [44]
barge.mt_km			
	diesel.MJ	3.20E-01	Cohon et al. 2010 [39]
	barge.mt_km	2.50E-01	Cohon et al. 2010 [39]
liquidpipeline.mt_km			
	electricity.US.kWh	1.84E-02	Scown et al. 2012 [51]
coal.MJ			

	diesel.MJ	2.30E-03	GREET 2018 [41]
	rfo.MJ	2.53E-04	GREET 2018 [41]
	electricity.US.kWh	2.09E-03	GREET 2018 [41]
	gasoline.MJ	1.89E-04	GREET 2018 [41]
	naturalgas_select.MJ	5.82E-05	GREET 2018 [41]
	flatbedtruck.mt_km	2.55E-03	GREET 2018 [41]
	rail.mt_km	5.34E-02	GREET 2018 [41]
	barge.mt_km	2.24E-02	GREET 2018 [41]
electricity.nuclear.kWh			
	electricity.US.kWh	1.97E-03	Warner and Heath 2012 [52]
	naturalgas_select.MJ	1.52E-01	Warner and Heath 2012 [52]
	uranium.kg	3.83E-08	Warner and Heath 2012 [52]
naturalgas.conventional.MJ			
	diesel.MJ	3.09E-03	Burnham 2018 [50]
	rfo.MJ	2.56E-04	Burnham 2018 [50]
	electricity.NGCC.kWh	2.98E-04	Burnham 2018 [50]
	gasoline.MJ	2.56E-04	Burnham 2018 [50]
	naturalgas.conventional.MJ	4.81E-02	Burnham 2018 [50]
	gaspipeline.mt_km	7.66E-02	Burnham 2018 [50]
naturalgas.shale.MJ			
	diesel.MJ	2.95E-03	Burnham 2018 [50]
	rfo.MJ	2.44E-04	Burnham 2018 [50]
	electricity.US.kWh	2.95E-04	Burnham 2018 [50]
	gasoline.MJ	2.44E-04	Burnham 2018 [50]
	naturalgas.shale.MJ	4.71E-02	Burnham 2018 [50]
	gaspipeline.mt_km	7.66E-02	Burnham 2018 [50]
refgas.MJ			
	crudeoil.MJ	1.00E+00	NA
h2.kg			
	electricity.US.kWh	2.69E-01	Spath and Mann 2001 [45]
	naturalgas_select.MJ	1.43E+02	Spath and Mann 2001 [45]
	gaspipeline.mt_km	1.21E+00	Spath and Mann 2001 [45]
tankertruck.mt_km			
	diesel.MJ	1.22E+00	Cohon et al. 2010 [39]
	tankertruck.mt_km	2.50E-01	Cohon et al. 2010 [39]
na_brine.kg			
	rfo.MJ	1.28E-01	GREET 2020 [38]
	electricity.US.kWh	2.56E-01	GREET 2020 [38]
	naturalgas_select.MJ	7.91E-01	GREET 2020 [38]
marinetanker.mt_km			
	rfo.MJ	1.00E-01	Cohon et al. 2010 [39]

	marinetanker.mt_km	2.50E-01	Cohon et al. 2010 [39]
formaldehyde.kg			
	electricity.US.kWh	1.50E-01	GREET 2021 [40]
	methanol.kg	1.20E+00	GREET 2021 [40]
methanol.kg			
	electricity.US.kWh	8.05E-03	GREET 2019 [42]
	naturalgas_select.MJ	6.93E+00	GREET 2019 [42]
	tankertruck.mt_km	9.04E-03	GREET 2019 [42]
	gaspipeline.mt_km	9.05E-01	GREET 2019 [42]
	o2.kg	3.80E-01	GREET 2019 [42]
uranium.kg			
	lime.kg	2.91E+00	Parker et al. 2016 [49]
	diesel.MJ	2.39E-09	Parker et al. 2016 [49]
	electricity.WECC.kWh	2.37E+01	Parker et al. 2016 [49]
	electricity.MRO.kWh	4.35E+01	Parker et al. 2016 [49]
	gasoline.MJ	3.30E-11	Parker et al. 2016 [49]
	naturalgas_select.MJ	1.16E-06	Parker et al. 2016 [49]
	flatbedtruck.mt_km	1.17E+01	Parker et al. 2016 [49]
	lpg.kg	2.53E+00	Parker et al. 2016 [49]
	landfill_mixedorganics.wet_kg	1.68E-01	Parker et al. 2016 [49]
	landfill_mixedMSW.wet_kg	1.22E-01	Parker et al. 2016 [49]
electricity.NGCC.kWh			
	electricity.NGCC.kWh	6.50E-02	Ou and Cai 2020 [43]
	naturalgas_select.MJ	7.20E+00	Ou and Cai 2020 [43]
electricity.WECC.kWh			
	coal.MJ	1.92E+00	Ou and Cai 2020 [43]
	diesel.MJ	9.37E-04	Ou and Cai 2020 [43]
	rfo.MJ	1.59E-02	Ou and Cai 2020 [43]
	electricity.WECC.kWh	4.90E-02	Ou and Cai 2020 [43]
	naturalgas_select.MJ	2.53E+00	Ou and Cai 2020 [43]
	electricity.nuclear.kWh	8.83E-02	Ou and Cai 2020 [43]
electricity.MRO.kWh			
	coal.MJ	4.80E+00	Ou and Cai 2020 [43]
	diesel.MJ	3.72E-03	Ou and Cai 2020 [43]
	rfo.MJ	1.02E-01	Ou and Cai 2020 [43]
	electricity.MRO.kWh	4.90E-02	Ou and Cai 2020 [43]
	naturalgas_select.MJ	2.49E+00	Ou and Cai 2020 [43]
	electricity.nuclear.kWh	1.62E-01	Ou and Cai 2020 [43]
lime.kg			
	caco3.kg	1.88E+00	GREET 2020 [38]

	coal.MJ	3.57E+00	GREET 2020 [38]
	diesel.MJ	7.43E-02	GREET 2020 [38]
	rfo.MJ	3.24E-02	GREET 2020 [38]
	electricity.US.kWh	5.70E-02	GREET 2020 [38]
	naturalgas_select.MJ	2.25E-01	GREET 2020 [38]
	tankertruck.mt_km	1.93E-01	GREET 2020 [38]
	lpg.kg	4.19E-02	GREET 2020 [38]
caco3.kg			
	coal.MJ	3.66E-03	GREET 2020 [38]
	diesel.MJ	1.30E-02	GREET 2020 [38]
	rfo.MJ	1.64E-03	GREET 2020 [38]
	electricity.US.kWh	2.44E-04	GREET 2020 [38]
	gasoline.MJ	2.56E-03	GREET 2020 [38]
	naturalgas_select.MJ	1.22E+00	GREET 2020 [38]
	flatbedtruck.mt_km	8.00E-02	GREET 2020 [38]

C4.4 Sensitivity Analysis

To capture uncertainty and variability in process yield, energy consumption, and transportation impacts for the PP recycling scenarios, we conduct 10,000 Monte Carlo simulations using triangular probability distributions as given by Table C5. For all parameters, except for energy consumption for mechanical recycling, the mode is equivalent to the original model value. Because the original energy use data for mechanical recycling is not broken down by pretreatment and extrusion,[12] we opted to use the SuperPro Designer results for the modes instead of the original model data. The total energy consumption for mechanical recycling used in the original LCA falls within the aggregated ranges in Table C5. For mechanical processes, we use specific minimum and maximum values from SuperPro Designer and Larrain et al. (2021).[53] For all other parameter probability distributions, the maximum and minimum values are equally spaced from the mode.

Table C5. Recycling scenarios’ sensitivity analyses: probability distributions for Monte Carlo simulations

Scenario	Parameter	Values				Sources		
		mode	max	min	units	mode	max	min
Solvent-Assisted Recycling	Pretreatment electricity consumption	281.0	287.0	243.5	kWh/tonne input	SuperPro (rigids, electric extrusion)	SuperPro (films, NG-driven extrusion)	Larrain et al. (2021) [53]
	Pretreatment natural gas consumption	16.4	102.4	16.4	MJ/tonne input	SuperPro (electric extrusion)	Larrain et al. (2021) [53]	SuperPro (electric extrusion)
	Process yield	x	x + 3 %	x - 3 %	%	Confidential	N/A	N/A
	Electricity consumption	x	x + 10 %	x - 10 %	kWh/tonne input	Confidential	N/A	N/A
	Natural gas consumption	x	x + 10 %	x - 10 %	MJ/tonne input	Confidential	N/A	N/A
	Byproduct stream	x	x + 10 %	x - 10 %	N/A	Confidential	N/A	N/A
	Transportation (trucking)	0.77	mode + 5 %	mode - 5 %	tonne-km/ kg input	Franklin Associates (2018) (12)	N/A	N/A
	Transportation (rail)	0.29	mode + 5 %	mode - 5 %	tonne-km/ kg input	Franklin Associates (2018) (12)	N/A	N/A
Mechanical Recycling	Pretreatment electricity consumption	281.0	287.0	243.5	kWh/tonne input	SuperPro (electric extrusion)	SuperPro (films, NG-driven extrusion)	Larrain et al. (2021) (53)
	Pretreatment natural gas consumption	16.4	102.4	16.4	MJ/tonne input	SuperPro (electric extrusion)	Larrain et al. (2021) [53]	SuperPro (electric extrusion)
	Process yield	85	87	55	%	Franklin Associates (2018) (12)	Larrain et al. (2021) [53]	SuperPro (electric extrusion)
	Electricity consumption for extrusion	384.7	386.9	209.5	kWh/tonne input	SuperPro (electric extrusion)	Larrain et al. (2021) [53]	Total from Franklin Associates (2018)[12] minus pretreatment from Larrain et al. (2021)[53]
	Natural gas consumption for extrusion	38.3	694.4	0.0	MJ/tonne input	SuperPro (electric extrusion)	Total from Franklin Associates (2018)[12] minus pretreatment from Larrain et al. (2021)[53]	Larrain et al. (2021) [53]
	Transportation (trucking)	0.77	mode + 5 %	mode - 5 %	tonne-km/ kg input	Franklin Associates (2018) [12]	N/A	N/A
	Transportation (rail)	0.29	mode + 5 %	mode - 5 %	tonne-km/ kg input	Franklin Associates (2018) [12]	N/A	N/A

Table C6. Boxplot data for virgin polypropylene production (Figure 4.1 in main text)

Source	**GHG emission factor (kg CO_{2e} per tonne PP output)**
Franklin Associates [12]	1840
Franklin Associates 2011 [2]	1860
Franklin Associates 2021 [54]	1548
Nicholson 2021 (film) [55]	2500
Nicholson 2021 (IM) [55]	2700

C5 LCA Results: Tabulated Data

Table C7. Life-cycle greenhouse gas impacts from virgin production and recycling (data for Figure 4.2)

	Mechanical Recycling	Solvent-Assisted Recycling	Virgin Production
Raw Materials	0.0	0.0	394.5
Preprocessing*	1.7	139.1	0.0
Solvent	0.0	11.7	0.0
Process Materials	0.0	103.2	0.0
Byproducts	0.0	-43.9	0.0
Direct Emissions	0.0	0.0	253.5
Electricity	250.8	607.4	290.0
Natural Gas	53.5	652.3	527.1
Non-NG Fuels	3.8	0.0	792.4
Transportation	168.7	160.6	91.8

*Preprocessing for solvent-assisted recycling includes much of what is considered part of the main process for mechanical recycling. This includes electricity and natural gas use. For mechanical recycling, preprocessing includes washing agents. Units for all values are kg CO_{2eq} per tonne PP produced.

Table C8. Emission factors forecast for PP production and recycling (data for Figure 4.4)

	CA, Mid			CA, Low Renewable Cost			US Ave, Mid			US Ave, Low Renewable Cost		
Year	Mechanical Recycling	Solvent-Assisted Recycling	Virgin Production	Mechanical Recycling	Solvent-Assisted Recycling	Virgin Production	Mechanical Recycling	Solvent-Assisted Recycling	Virgin Production	Mechanical Recycling	Solvent-Assisted Recycling	Virgin Production
2022	354.6	1271.0	2264.2	351.6	1262.0	2262.0	466.2	1602.6	2345.9	465.9	1601.8	2345.7
2024	331.3	1201.7	2247.1	332.8	1206.3	2248.3	419.8	1464.9	2311.9	410.2	1436.2	2304.9
2026	318.8	1164.5	2238.0	317.4	1160.5	2237.0	394.0	1388.2	2293.1	374.4	1329.7	2278.6
2028	296.3	1097.8	2221.5	284.6	1062.8	2212.9	341.9	1233.3	2254.9	301.0	1111.8	2225.0
2030	269.4	1017.7	2201.8	256.3	978.8	2192.2	311.3	1142.2	2232.5	278.3	1044.2	2208.3
2032	263.3	999.6	2197.4	252.4	967.1	2189.4	302.4	1115.9	2226.0	270.6	1021.3	2202.7
2034	258.8	986.3	2194.1	251.3	963.9	2188.6	291.9	1084.5	2218.3	264.1	1002.0	2197.9
2036	255.4	976.1	2191.6	249.5	958.7	2187.3	286.0	1067.1	2214.0	259.7	988.8	2194.7
2038	251.7	965.2	2188.9	248.5	955.7	2186.5	282.2	1055.8	2211.2	260.2	990.2	2195.0
2040	249.1	957.3	2186.9	247.8	953.4	2186.0	278.6	1045.1	2208.6	266.4	1008.9	2199.6
2042	247.2	951.6	2185.5	247.3	952.0	2185.6	276.9	1040.0	2207.3	271.9	1025.1	2203.6
2044	246.0	948.2	2184.7	246.5	949.8	2185.1	277.4	1041.6	2207.7	275.6	1036.1	2206.3
2046	245.5	946.7	2184.3	245.9	947.9	2184.6	282.5	1056.6	2211.4	270.3	1020.3	2202.4
2048	245.1	945.6	2184.0	246.2	948.9	2184.9	288.4	1074.1	2215.7	259.5	988.2	2194.5
2050	244.7	944.3	2183.7	243.2	939.8	2182.6	294.6	1092.6	2220.2	249.6	959.0	2187.3

The units for all values are kg CO_{2eq} per tonne of PP output.

References

(1) Posch, W. Polyolefins. In *Applied plastics engineering handbook*; Kutz, M., Ed.; Elsevier, 2011; pp. 23–48.

(2) Franklin Associates. *Cradle-to-grave life cycle inventory of nine plastic resins and four polyurethane precursors*; Plastics Division of the American Chemistry Council, 2011.

(3) Khan, M. J. H.; Hussain, M. A.; Mujtaba, I. M. Polypropylene production optimization in fluidized bed catalytic reactor (FBCR): statistical modeling and pilot scale experimental validation. *Materials (Basel)* **2014**, *7*, 2440–2458.

(4) Pressley, P. N.; Levis, J. W.; Damgaard, A.; Barlaz, M. A.; DeCarolis, J. F. Analysis of material recovery facilities for use in life-cycle assessment. *Waste Manag.* **2015**, *35*, 307–317.

(5) G.A.A. Inc. *2016-2017 Database on material recovery facilities and mixed waste processing facilities in the U.S., with updates*; Governmental Advisory Associates, Inc., 2019.

(6) Ma, J.; Tominac, P. A.; Aguirre-Villegas, H. A.; Olafasakin, O. O.; Wright, M. M.; Benson, C. H.; Huber, G. W.; Zavala, V. M. Economic evaluation of infrastructures for thermochemical upcycling of post-consumer plastic waste. *Green Chem.* **2022**.

(7) Hundertmark, T.; Prieto, M.; Ryba, A.; Simons, T. J.; Wallach, J. *Accelerating plastic recovery in the United States*; McKinsey & Company, 2019.

(8) Smith, R. L.; Takkellapati, S.; Riegerix, R. C. Recycling of plastics in the United States:

plastic material flows and polyethylene terephthalate (PET) recycling processes. *ACS Sustain. Chem. Eng.* **2022**, *10*, 2084–2096.

(9) APR. Recycling Rigid Plastics Beyond Bottles: Non-Bottle Containers, 2017.

(10) Moore Recycling Associates Inc. *National Mixed Rigid PlasticBale Composition Study*; The Association of Plastic Recyclers, 2015.

(11) APR. *Model Bale Specifications: 3-7 Bottles and ALL Other Rigid Plastics*; The Association of Plastic Recyclers.

(12) Franklin Associates. *Life cycle impacts for postconsumer recycled resins: PET, HDPE and PP*; The Association of Plastic Recyclers, 2018.

(13) Schyns, Z. O. G.; Shaver, M. P. Mechanical recycling of packaging plastics: A review. *Macromol. Rapid Commun.* **2021**, *42*, e2000415.

(14) Oblak, P.; Gonzalez-Gutierrez, J.; Zupančič, B.; Aulova, A.; Emri, I. Processability and mechanical properties of extensively recycled high density polyethylene. *Polym. Degrad. Stab.* **2015**, *114*, 133–145.

(15) Gustavo Barbosa, L.; Piaia, M.; Henrique Ceni, G. Analysis of impact and tensile properties of recycled polypropylene. *IJME* **2017**, *7*, 117–120.

(16) da Costa, H. M.; Ramos, V. D.; de Oliveira, M. G. Degradation of polypropylene (PP) during multiple extrusions: Thermal analysis, mechanical properties and analysis of variance. *Polym. Test.* **2007**, *26*, 676–684.

(17) De Santis, F.; Pantani, R. Optical properties of polypropylene upon recycling. *ScientificWorldJournal* **2013**, *2013*, 354093.

(18) Luijsterburg, B.; Goossens, H. Assessment of plastic packaging waste: Material origin, methods, properties. *Resources, Conservation and Recycling* **2014**, *85*, 88–97.

(19) Vidakis, N.; Petousis, M.; Tzounis, L.; Maniadi, A.; Velidakis, E.; Mountakis, N.; Papageorgiou, D.; Liebscher, M.; Mechtcherine, V. Sustainable additive manufacturing: mechanical response of polypropylene over multiple recycling processes. *Sustainability* **2020**, *13*, 159.

(20) Ragaert, K.; Delva, L.; Van Geem, K. Mechanical and chemical recycling of solid plastic waste. *Waste Manag.* **2017**, *69*, 24–58.

(21) Gu, F.; Hall, P.; Miles, N. J.; Ding, Q.; Wu, T. Improvement of mechanical properties of recycled plastic blends via optimizing processing parameters using the Taguchi method and principal component analysis. *Materials & Design (1980-2015)* **2014**, *62*, 189–198.

(22) Ladhari, A.; Kucukpinar, E.; Stoll, H.; Sängerlaub, S. Comparison of properties with relevance for the automotive sector in mechanically recycled and virgin polypropylene. *Recycling* **2021**, *6*, 76.

(23) Gu, F.; Guo, J.; Zhang, W.; Summers, P. A.; Hall, P. From waste plastics to industrial raw materials: A life cycle assessment of mechanical plastic recycling practice based on a real-world case study. *Sci. Total Environ.* **2017**, *601–602*, 1192–1207.

(24) De Tandt, E.; Demuytere, C.; Van Asbroeck, E.; Moerman, H.; Mys, N.; Vyncke, G.; Delva, L.; Vermeulen, A.; Ragaert, P.; De Meester, S.; et al. A recycler's perspective on

the implications of REACH and food contact material (FCM) regulations for the mechanical recycling of FCM plastics. *Waste Manag.* **2021**, *119*, 315–329.

(25) Eriksen, M. K.; Christiansen, J. D.; Daugaard, A. E.; Astrup, T. F. Closing the loop for PET, PE and PP waste from households: Influence of material properties and product design for plastic recycling. *Waste Manag.* **2019**, *96*, 75–85.

(26) Bora, R. R.; Wang, R.; You, F. Waste polypropylene plastic recycling toward climate change mitigation and circular economy: energy, environmental, and technoeconomic perspectives. *ACS Sustain. Chem. Eng.* **2020**, *8*, 16350–16363.

(27) Astrup, T.; Fruergaard, T.; Christensen, T. H. Recycling of plastic: accounting of greenhouse gases and global warming contributions. *Waste Manag. Res.* **2009**, *27*, 763–772.

(28) Lazarevic, D.; Aoustin, E.; Buclet, N.; Brandt, N. Plastic waste management in the context of a European recycling society: Comparing results and uncertainties in a life cycle perspective. *Resources, Conservation and Recycling* **2010**, *55*, 246–259.

(29) Hossain, M. U.; Ng, S. T.; Dong, Y.; Amor, B. Strategies for mitigating plastic wastes management problem: A life cycle assessment study in Hong Kong. *Waste Manag.* **2021**, *131*, 412–422.

(30) Van Eygen, E.; Laner, D.; Fellner, J. Integrating high-resolution material flow data into the environmental assessment of waste management system scenarios: the case of plastic packaging in Austria. *Environ. Sci. Technol.* **2018**, *52*, 10934–10945.

(31) Beigbeder, J.; Soccalingame, L.; Perrin, D.; Bénézet, J.-C.; Bergeret, A. How to manage biocomposites wastes end of life? A life cycle assessment approach (LCA) focused on polypropylene (PP)/wood flour and polylactic acid (PLA)/flax fibres biocomposites. *Waste Manag.* **2019**, *83*, 184–193.

(32) Mercante, I. T.; Bovea, M. D.; Ibáñez-Forés, V.; Arena, A. P. Life cycle assessment of construction and demolition waste management systems: a Spanish case study. *Int. J. Life Cycle Assess.* **2012**, *17*, 232–241.

(33) Faraca, G.; Martinez-Sanchez, V.; Astrup, T. F. Environmental life cycle cost assessment: Recycling of hard plastic waste collected at Danish recycling centres. *Resources, Conservation and Recycling* **2019**, *143*, 299–309.

(34) Vadenbo, C.; Hellweg, S.; Astrup, T. F. Let's be clear(er) about substitution: a reporting framework to account for product displacement in life cycle assessment. *Journal of Industrial Ecology* **2017**, *21*, 1078–1089.

(35) Rigamonti, L.; Grosso, M.; Sunseri, M. C. Influence of assumptions about selection and recycling efficiencies on the LCA of integrated waste management systems. *Int. J. Life Cycle Assess.* **2009**, *14*, 411–419.

(36) Smith, E.; Bilec, M. M.; Khanna, V. Evaluating the global plastic waste management system with markov chain material flow analysis. *ACS Sustain. Chem. Eng.* **2023**.

(37) Viau, S.; Majeau-Bettez, G.; Spreutels, L.; Legros, R.; Margni, M.; Samson, R. Substitution modelling in life cycle assessment of municipal solid waste management.

Waste Manag. **2020**, *102*, 795–803.

(38) Wang, M.; Elgowainy, A.; Lee, U.; Bafana, A.; Benavides, P.; Burnham, A.; Cai, H.; Dai, Q.; Gracida, U.; Hawkins, T.; et al. Greenhouse gases, Regulated Emissions, and Energy use in Technologies Model ® (2020 Excel). *Argonne National Laboratory (ANL), Argonne, IL (United States)* **2020**.

(39) Cohon, J. L.; Cropper, M. L.; Cullen, M. R.; English, M. R.; Field, C. B.; Greenbaum, D. S.; Hammitt, J. K.; Henderson, R. F.; Kling, C. L.; Krupnick, A. J.; et al. *Hidden costs of energy: unpriced consequences of energy production and use*; National Academies Press: Washington, D.C., 2010.

(40) Wang, M.; Elgowainy, A.; Lee, U.; Bafana, A.; Banerjee, S.; Benavides, P.; Bobba, P.; Burnham, A.; Cai, H.; Gracida, U.; et al. Greenhouse gases, Regulated Emissions, and Energy use in Technologies Model ® (2021 Excel). *Argonne National Laboratory (ANL), Argonne, IL (United States)* **2021**.

(41) Wang, M.; Elgowainy, A.; Lee, U.; Benavides, P.; Burnham, A.; Cai, H.; Dai, Q.; Hawkins, T.; Kelly, J.; Kwon, H.; et al. Greenhouse gases, Regulated Emissions, and Energy use in Transportation Model ® (2018 Excel). *Argonne National Laboratory (ANL), Argonne, IL (United States)* **2018**.

(42) Wang, M.; Elgowainy, A.; Lee, U.; Benavides, P.; Burnham, A.; Cai, H.; Dai, Q.; Hawkins, T.; Kelly, J.; Kwon, H.; et al. Greenhouse gases, Regulated Emissions, and Energy use in Transportation Model ® (2019 Excel). *Argonne National Laboratory (ANL), Argonne, IL (United States)* **2019**.

(43) Ou, L.; Cai, H. *Update of emission factors of greenhouse gases and criteria air pollutants, and generation efficiencies of the U.S. electricity generation sector*; Argonne National Laboratory (ANL): Argonne, IL (United States), 2020.

(44) Lu, Z.; Han, J.; Wang, M.; Cai, H.; Sun, P.; Dieffenthaler, D.; Gordillo, V.; Monfort, J.-C.; He, X.; Przesmitzki, S. Well-to-Wheels Analysis of the Greenhouse Gas Emissions and Energy Use of Vehicles with Gasoline Compression Ignition Engines on Low Octane Gasoline-Like Fuel. *SAE Int. J. Fuels Lubr.* **2016**, *9*, 527–545.

(45) Spath, P.; Mann, M. *Life Cycle Assessment of Hydrogen Production via Natural Gas Steam Reforming*; NREL/TP-570-27637; National Renewable Energy Laboratory, 2001.

(46) Wernet, G.; Bauer, C.; Steubing, B.; Reinhard, J.; Moreno-Ruiz, E.; Weidema, B. The ecoinvent database version 3 (part I): overview and methodology. *Int. J. Life Cycle Assess.* **2016**, *21*, 1218–1230.

(47) EPA. *Documentation for Greenhouse Gas Emission and Energy Factors Used in the Waste Reduction Model (WARM v.14)*; Office of Resource Conservation and Recovery, U.S. Environmental Protection Agency, 2018.

(48) Wang, M.; Elgowainy, A.; Lee, U.; Baek, K.; Bafana, A.; Benavides, P.; Burnham, A.; Cai, H.; Cappello, V.; Chen, P.; et al. Greenhouse gases, Regulated Emissions, and Energy use in Technologies Model ® (2022 Excel). *Argonne National Laboratory (ANL), Argonne, IL (United States)* **2022**.

(49) Parker, D. J.; McNaughton, C. S.; Sparks, G. A. Life Cycle Greenhouse Gas Emissions from Uranium Mining and Milling in Canada. *Environ. Sci. Technol.* **2016**, *50*, 9746–9753.

(50) Burnham, A. *Updated Natural Gas Pathways in the GREET1_2018 Model*; Argonne National Laboratory, 2018.

(51) Scown, C. D.; Nazaroff, W. W.; Mishra, U.; Strogen, B.; Lobscheid, A. B.; Masanet, E.; Santero, N. J.; Horvath, A.; McKone, T. E. Lifecycle greenhouse gas implications of US national scenarios for cellulosic ethanol production. *Environmental Research Letters* **2012**, *7*, 014011.

(52) Warner, E. S.; Heath, G. A. Life Cycle Greenhouse Gas Emissions of Nuclear Electricity Generation. *J. Ind. Ecol.* **2012**, *16*, S73–S92.

(53) Larrain, M.; Van Passel, S.; Thomassen, G.; Van Gorp, B.; Nhu, T. T.; Huysveld, S.; Van Geem, K. M.; De Meester, S.; Billen, P. Techno-economic assessment of mechanical recycling of challenging post-consumer plastic packaging waste. *Resources, Conservation and Recycling* **2021**, *170*, 105607.

(54) Franklin Associates. *Cradle-to-gate life cycle analysis of polypropylene (PP) resin*; American Chemistry Council, 2021.

(55) Nicholson, S. R.; Rorrer, N. A.; Carpenter, A. C.; Beckham, G. T. Manufacturing energy and greenhouse gas emissions associated with plastics consumption. *Joule* **2021**.