Cristina Trois

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The impact of landfills on the environment has come under increasing scrutiny in recent years due to the confounding effects of climate change and water scarcity. There is an urgent need to reduce from landfills the greenhouse gas emissions that cause climate change, and to provide effective treatment solutions for waste, thereby diverting it from landfills. With an estimated 80 million tonnes of plastic waste entering the world’s oceans annually, the accumulation of marine plastic has become a global crisis. Plastic pollution threatens food safety and quality, human health and coastal tourism, and contributes to climate change. For these reasons, there is an urgent need to explore a bioplastic biorefinery process. This review paper examines the potential of organic waste as an alternative carbon source in the efficient and feasible microbial production of polyhydroxyalkanote (PHA) and polyhydroxybutyrate (PHB), which are precursors for bioplastic. More specifically, this paper presents a concept for a bioplastic biorefinery from a technological perspective, based on data from previous studies. Biofuel production processes are also assessed with the aim of integrating these processes to construct a bioplastic waste biorefinery. Garden refuse and food waste have been shown to be feasible feedstocks for the production of PHA and PHB in singular processes. Diverting these wastes away from landfills will significantly ease the environmental impacts currently associated with their disposal.

Significance:

• A bioplastic biorefinery is a viable alternative to treat municipal organic waste.

• Several biofuel production processes can be integrated into a bioplastic biorefinery system.

• Organic waste is poorly managed in South Africa, resulting in greenhouse gas emissions.

• Several barriers and considerations must be overcome before implementing the technology at full scale.

Introduction

Large-scale plastic manufacturing began approximately 70 years ago, and since then an estimated 8.3 billion tonnes of plastic has been manufactured.1 This sum is growing at an accelerated pace.2 Most of this plastic cannot be reprocessed efficiently on a worldwide scale, and therefore it still exists in some form.1,2 The ubiquity of plastic waste on the earth’s surface has prompted some to argue that it might be regarded as a geological indication of the Anthropocene era because of its prevalence.4 Because of advances in waste management systems over the last few decades, more end-of-life alternatives for plastic have become available, and collection rates have increased as a result.1 The ultimate destination of many plastic goods is still unknown, particularly in underdeveloped nations.5 Some of the reasons for this include a lack of global statistics, a lack of official collecting mechanisms in many areas, and unreported waste disposal, including unlawful dumping and unsupervised burning.2 Current estimates of where today’s plastics will be discovered in 20 years’ time reveal that the vast majority have been thrown away, including all packaging. There will be some recycling (mostly downcycling) or incineration of plastics, but the bulk will end up in landfills, and some may become unmanaged litter and end up in the ocean. Packaging is a major contributor to litter and ocean plastic, especially in developed countries. After plastic has entered the ocean, it is almost impossible to remove, which means it quickly accumulates.6 By 2050, approximately 20% of global oil consumption may be devoted to plastics manufacturing, resulting in 15% of greenhouse gas (GHG) emissions. Plastics are also expected to outnumber fish in the seas by then, according to some projections.7 Up to 12 million tonnes of plastic ends up in the ocean each year, and 50% of marine litter is made up of single-use plastic products.8 The indiscriminate use of fossil fuels continues to increase atmospheric carbon dioxide levels. It is therefore imperative that alternatives such as bioplastics are investigated. ‘Bioplastic’ can have several different meanings, including (i) biobased but not biodegradable, (ii) biodegradable but not biobased, and (iii) biodegradable and biobased, as illustrated in Figure 1. This paper will focus on bioplastics that are biobased and biodegradable. Biobased plastics are generally considered to be plastics that are produced from biological or organic material. For this reason, organic waste has been proposed as a feedstock for the production of bioplastics in a biorefinery setup.

A biorefinery is described as ‘The sustainable processing of biomass into a spectrum of commercial goods (food, feed, materials, and chemicals) and energy (fuels, electricity, and/or heat)’ by the International Energy Agency (IEA) Bioenergy Task 42.9 A biorefinery may also be a concept, a facility, a process, a plant, or even a cluster of facilities that combine many different disciplines of expertise, such as chemical engineering, chemistry, biology, biochemistry, biomolecular engineering and others.9 Biorefineries are similar to traditional oil refineries that produce a variety of goods and fuels from petroleum. The IEA emphasises that a biorefinery not only meets the demand for biobased products with functional qualities comparable to those produced from fossil resources, but also provides a distinct advantage by addressing problems of sustainability in all areas – economic, social and environmental. It uses renewable biomass as a feedstock, and reduces biobased product manufacturing costs through economies of scale and the development of green technology. Biorefineries are versatile enough to be used

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all over the world due to the diversity of local wastes, sugar cane, excess food, straw and aquatic biomass, as well as the biomass component of municipal solid refuse, all of which are potential feedstocks for a biorefinery. Biorefineries have the added advantage of producing carbon-neutral products such as certain biofuels, which ultimately have the potential to reduce the carbon footprint.

Because organic solvents have a high recovery rate and purity while still being inexpensive, this technique is often used to extract PHA from residual biomass.

The annual manufacturing capacity of PHA was 900 000 tonnes in 2015. The price of PHA is a significant deterrent to its gaining a larger portion of the market share. PHA is considerably more costly than other biopolymers. In 2014, the price range was between ZAR67 and ZAR79 per kilogram, which was much higher than the prices of other well-established biodegradable and biobased polymer materials. Prices are anticipated to drop if the quantities produced surpass the pilot production scale, because of the savings that come from manufacturing at a larger scale. The price of PHA is also affected by the price of raw materials and the method of extraction. As a result, raw material costs are critical, accounting for up to 50% of total manufacturing costs. The prices of refined sugars or fatty acids/lipids, which are presently used in industrial processes, are just as variable as the raw material costs. They are heavily reliant on oil prices, which have been rising steadily for decades. A search for low-cost raw materials for PHA synthesis has been launched to become less dependent on this significant cost. For this reason, organic waste appears to be an attractive feedstock for this process.

**OFMSW as a potential feedstock for bioplastic**

The organic fraction of municipal solid waste (OFMSW) has long been demonstrated as a suitable feedstock for many different bioprocesses. However, there is a dearth of knowledge on the production of bioplastic building blocks from OFMSW. This section aims to present the potential of OFMSW as a feedstock for bioplastic production.

OFMSW contains several waste streams, including fruit and vegetable waste, food waste and garden refuse. According to the Food and Agriculture Organization (FAO), 780 million tonnes of fruit and vegetable waste are produced each year.14 South Africa is a significant fruit-growing country, with annual citrus, grape and apple production totalling 2.1 million, 1.8 million, and 0.79 million tonnes, respectively. Figure 2 shows the respective proportions of these fruits in the primary fruit production of South Africa. Large amounts of waste are produced during the processing of these fruit streams. For example, 25–35% of processed apples, 50% of citrus, and 20% of grapes are projected to be wasted.15 The bulk of fruit and vegetable waste is generated during the harvesting and processing phases in developing countries, whereas little waste is produced during the consumption stage.16

**Building blocks for bioplastics**

Polyhydroxyalkanoates (PHA) are microbial storage compounds that may be used as polymers for various applications after extraction, compounding and extrusion. The polymer used in the product may be recycled together with other plastics once it has been used. As with other synthetic materials, PHA may be broken down by aerobic and anaerobic bacteria found in soil and water, which makes it less of a concern during its end-of-life phase. The shift to a post-fossil carbon world necessitates the development of PHA as a viable alternative to traditional plastics. PHA granules are carbon and energy sources for a wide variety of bacteria that can manufacture them intracellularly. When an excess of carbon is available at the same time as a restriction in one or more nutrients, the PHA granules are often generated under unbalanced nutritional circumstances. Nitrogen or phosphorus is usually the medium's only limitation with respect to polymer synthesis. The organism's metabolism shifts from growth to PHA accumulation as a result of this restriction. These compounds are synthesised by a wide variety of microorganisms, including members of the phyla Gram-negative eubacteria, Gram-positive eubacteria, archaea and microalgae. Only the genus Pseudomonas produces mcl-PHA, or medium-chain length PHA. Polymers may range in molecular weight from 50 kDa to 1000 kDa, with a molecular weight distribution of 100–30 000 monomers, depending on the culture circumstances. PHA molecules aggregate within cells into granules due to their apolar nature. Every cell has at least one granule that is passed down via the DNA. Cellular polymer loads may be very high when the granules are combined. Once the cells are full of PHA, all that remains is to remove the polymer and convert it into usable plastic.
methods that are now accessible on an industrial scale throughout the world.20 Meanwhile, half of the world’s population lives in cities, where waste management is critical for dealing with the massive amounts of food waste produced every day. Innovative solutions are needed to deal with this ever-increasing waste type.

A 2013 study estimated that the annual wasteage of food in South Africa amounts to 9.04 million tonnes per annum.21 Additionally, fruit and vegetable waste is also produced in huge amounts owing to agricultural activities, supermarkets and wholesale marketplaces. In South Africa, a significant portion of this waste is currently diverted to landfills where it poses serious environment problems. The organic material decomposes and forms GHG, such as methane, which is released into the atmosphere. More concerning is the cost associated with the disposal of food waste, with a 2015 study estimating disposal costs at over ZAR3 billion.22 A more recent estimate, taking into account disposal, clean up and effects on livelihoods, indicated that plastic pollution cost South Africa a staggering ZAR885 billion in 2019.23 Furthermore, and more alarmingly, some of the biggest cities in South Africa (Johannesburg, Tshwane and Cape Town) have less than 10 years left before their landfills are rendered incapacitated.24

OFMSW is generally considered a very rich carbon source for various bioprocesses. For instance, garden refuse, and fruit and vegetable waste are lignocellulosic-based biomass, meaning that they are composed of cellulose and hemicellulose bound by a stiff lignin polymer. Monomeric glucose and acetic acid units make up cellulose and hemicellulose respectively. This indicates the material’s fermentability, either indirectly via enzyme catalysis, or directly through microbial decomposition. However, the lignin layer is regarded as extremely resistant to degradation, which significantly hampers process yields. For this reason, pretreatment is often a required step in a lignin-based biorefinery in order to enhance the digestibility of the material. Several pretreatment technologies exist and have been applied to a vast array of feedstocks. The type of pretreatment used can have significant impacts on process feasibility and economics.

South Africa has not established a separated waste collection plan. For this reason, OFMSW arrives at transfer stations mixed and contaminated with other microbial species. This is an important consideration, because ‘dirty’ OFMSW can significantly hamper the process dynamics in many ways. One concern is the introduction of competing microorganisms, which could reduce the productivity of the microbes employed in either saccharification or the fermentation process. Another concern is the cross-contamination of microbes, resulting in premature degradation of the feedstock and thereby reducing the calorific value of the waste. The microbial dynamics of the process therefore require further elucidation to provide clarity on the feedstock quality.

**Bioplastic biorefineries**

As defined earlier, biorefineries are made up of several different processes that function together to valorise a feedstock and produce multiple products. In recent decades, a multitude of studies have evaluated singular bioprocesses to produce either biofuels (such as bioethanol, biomethane, biohydrogen and biodiesel) or bioproducts (such as pharmaceuticals, enzymes and bioplastic monomers) from organic material. Through analysing the process flow chart of applicable bioprocesses, several singular processes can be strategically combined to create a theoretical bioplastic biorefinery. At the same time, it is important to understand the feedstock and its by-products through every stage of the process in order to ensure its complete valorisation.

PHA manufacturing cannot be industrialised without paying careful attention to the techno-economic environment, for example, the competing uses of raw materials or resources such as land in order to drive it towards economic viability. About a decade ago, the increased production of biofuels sparked a global debate about land use in relation to ‘food versus fuel’. Since then, a worldwide perspective has been required for every new biobased product. Bioplastics need only a tiny fraction of the world’s agricultural land to produce enough feedstock. The amount of land currently being used for the manufacturing of bioplastics is insignificant. However, bioplastics will be increasingly required in the future as the economy moves away from fossil fuels, which could raise the amount of land required 500-fold. A land usage of 5% of global agricultural land or 15% of the world’s arable land would be considered to be unacceptably wasteful. Accordingly, substituting bioplastics for petrochemical-derived plastics would require considerably less land than at present. No additional acreage is needed if waste products are utilised as carbon sources. It is possible to turn by-products into a wide range of goods. In reality, an increasing number of stakeholders view industrial by-products as stepping stones to a biobased economy.25 By increasing competition, the most efficient usage method would benefit. These by-products must be the most competitive usage method for the long-term development of PHA. Cascadic usage is the most essential notion in relation to the utilisation of waste products and virgin biomass. This implies that a biorefinery, which uses biomass holistically, incorporates multiple bio-processes. As described above, the biorefinery is an integrated biobased industry that utilises a variety of technologies to produce products such as chemicals, biofuel, food and feed ingredients, as well as other biomaterials, fibres, heat and power, all with the goal of maximising added value along the three pillars of sustainability.26 Thus, PHA synthesis would be one stage in the cascading use of biomass, similar to the present plastic manufacturing process in an oil refinery, as previously stated. A biorefinery in Brazil, for example, uses sugar cane to make sugar (sucrose), ethanol and a compound called poly(3-hydroxybutyrate) (PH3H), another building block for bioplastics.27 Bioprocess molasses from the sugar crystallisation stage is transformed to PH3H in this biorefinery. An ethanol distillation by-product called long-chain alcohols is used to extract the polymer from the cells. Bagasse, a waste product, is burned to generate process energy. The polymer can be manufactured inexpensively and with sufficient purity using this improved method.

Several studies have examined the production of bioplastic from organic waste, as summarised in Table 1. Ebrahimian et al.28 explored the production of both biofuels and bioplastics from OFMSW. These authors subjected the feedstock to an acetic acid catalysed ethanol organosolv pretreatment at 120 °C for 60 min to enhance enzymatic hydrolysis. The remaining solid residue following hydrolysis was channelled towards methane production through anaerobic digestion, yielding 23.1 L. The pretreated hydrolysate (containing 498.5 g glucose/kg) was fermented with Enterobacter aerogenes PTCC 122 and yielded 139.1 g 2,3-butanediol, 98.3 g ethanol, 28.6 g acetic acid, 71.4 L biohydrogen, and 40 g PHA. Colombo et al.30 reported on the enhanced production of PHA from OFMSW by employing a mix of microbial consortia from activated sludge. This study optimised the organic acid production, resulting in 151 g/kg, and consequently optimised the PHA fermentation process yielding 223 g/kg. The production of PHA from OFMSW was also investigated at a pilot scale.31 A combined treatment of OFMSW and sewage sludge was explored in a fed-batch system, resulting in 65 g PHA/kg total volatile solids. This is an important study, as it indicates the feasibility of setting up a large-scale bioplastics bioprocess from OFMSW. Another pilot-scale study analysed the production of PHA from OFMSW at high pH and ammonia concentrations.32 These authors found that the highest PHA accumulation of 77 wt% occurred as pH increased towards 9 and the ammonia concentration was 500 mg/L.

### Table 1: Polyhydroxyalkanoate (PHA) yields from various studies employing the organic fraction of municipal solid waste (OFMSW) as a feedstock

| Feedstock                  | Inoculum                          | Yield                      | Reference                           |
|----------------------------|-----------------------------------|----------------------------|------------------------------------|
| OFMSW                      | Enterobacter aerogenes PTCC 22    | 40 g PHA/kg                | Ebrahimian et al.28                 |
| OFMSW                      | Activated sludge                  | 223 g PHA/kg               | Colombo et al.29                    |
| OFMSW–sewage sludge        | –                                 | 65 g/kg total volatile solids | Valentino et al.30                  |
| OFMSW                      | –                                 | 77 wt% PHA                 | Mulders et al.31                    |
| OFMSW                      | Anaerobic digester effluent       | 45% PHA                    | Martin-Rylas et al.32               |
Several biofuel processes, such as biomethane, biohydrogen and bioethanol, result in volatile fatty acid (VFA) production as a by-product. VFAs are precursor compounds that are further metabolised by microorganisms to produce PHA.

There are many technologies that can be integrated into a bioplastic biorefinery, as illustrated in Figure 3. Some of these include anaerobic digestion, dark fermentation and alcoholic fermentation.

Anaerobic digestion (AD) is one of the most commonly employed technologies in bioprocess systems owing to its ease of use, scale up and economic outlook. Moreover, AD has the potential to solve one of the most pressing problems confronting modern society: the management of OFMSW. Despite the fact that OFMSW is a potential energy source, obstacles such as the organic fraction’s diversion (requiring expensive and complicated equipment), alternative treatment costs and process dependability have hampered landfill diversion. AD allows valuable organic waste, for example garbage that would otherwise end up in landfills, to be utilised, although more research is required to enhance process dependability and economic advantages. Campuzano and Gonzalez-Martinez demonstrated the potential of OFMSW and an adaptive inoculum in an AD system, reporting a methane yield of 339 NL/kg volatile solids. Another study examined methane production from OFMSW-based bioethanol effluent and realised a yield of 212 mL/g volatile solids. This study has very explicit implications, as it has been demonstrated that multiple biofuels can be produced from OFMSW in a single system.

Another technology is dark fermentation, which is most commonly associated with the production of biohydrogen. This process entails the biochemical breakdown of organic material through complex mixed microbial consortia to produce biohydrogen and an array of VFAs such as propionic and butyric acid. Currently, this process is hampered by relatively lower yields compared to other conventional processes such as fuel cells and steam reforming. Similar to AD, dark fermentation has the advantage of employing a wide range of organic matter as a feedstock, because the complex inoculum possesses the ability to break down the material. Elsamadony and Tawfik illustrated the biohydrogen potential of OFMSW, obtaining a yield of 2.05 mol/mol carbohydrate. Another study explored the production of acetone, butanol, ethanol and hydrogen from OFMSW, yielding 114.1 g, 43.8 g, 15.1 g and 97.5 L, respectively. This study also demonstrated the feasibility of producing multiple products from OFMSW in a biorefinery system. Another commonly considered technology is alcoholic fermentation, the main process responsible for the production of bioethanol. Several studies have examined and reported on the optimal process conditions for high yields. In addition, numerous feedstocks have been employed in the production process. One of the bottlenecks of this process is the requirement of a pretreatment stage to enhance the release of fermentable sugars from organic feedstocks such as garden refuse and agricultural waste. Saccharomyces cerevisiae is the most commonly employed inoculum in this process, while other species such as Pichia stipitis have also been considered. Fermentation is an integral part of the majority of waste biorefineries because bioethanol is a high-value fuel.

A schematic for a bioplastic biorefinery is illustrated in Figure 4. In essence, the OFMSW feedstock undergoes pretreatment to enhance feedstock digestibility. The solid residue biomass may be suitable for anaerobic digestion, thus producing methane and a VFA-rich effluent. The hydrolysate from the pretreatment can then be directed towards dark fermentation to produce biohydrogen as well as an effluent containing VFAs. At this point, two biofuels have been produced. The VFA effluent from both processes can be pooled and further fermented with an appropriate inoculum to produce PHA. This biorefinery system therefore has the capability to add value to organic waste by producing three products. These products have the potential to ease the burden that current conventional plastic places on the environment, and to mitigate the effect that fossil fuel burning and OFMSW dumping has on GHG emission and climate change. In this sense, coupling a biofuel process with a PHA production process enhances the economic and environmental outlook for the process.

As identified earlier, there is currently a lack of studies that explicitly discuss a bioplastic biorefinery from OFMSW. The technologies that would be required in this biorefinery have all been well studied and investigated from a singular process perspective. Other studies have looked at the production of multiple biofuels in a biorefinery system while exploiting OFMSW as a feedstock. In understanding the process requirements and dynamics, it is possible to construct a system that employs OFMSW, where different fractions of the waste (either separated or after enzymatic hydrolysis) are diverted to different processes. This could result in the integration of bioplastic production processes coupled with biofuel processes.

Table 2: Proposed biorefinery scenarios for the primary production of bioplastics

| Scenario | Feedstock | Technology | Products | Reference |
|----------|------------|------------|----------|-----------|
| 1        | Organic fraction of municipal solid waste | Fermentation, dark fermentation | PHA, ethanol, hydrogen | Ebrahimian et al.24 |
| 2        | Food waste | Fermentation, alcoholic fermentation | PHA, ethanol | Kiran and Liu25 |
| 3        | Food waste | Fermentation, alcoholic fermentation | PHA, ethanol | Alamanou et al.26 |
| 4        | Garden refuse | Anaerobic digestion, fermentation | PHA, methane | Perin et al.41 |

PHA, polyhydroxyalkanoate

**Figure 3:** Schematic of potential biorefinery technologies that are capable of being coupled with bioplastic production.

**Figure 4:** A simplified schematic of a bioplastic biorefinery.
Some potential scenarios are outlined in Table 2, based on modified studies that have been surveyed. In all these studies, the production of PHA was not considered, although PHA production could easily replace one or more other processes requiring a carbon source. For instance, Ebrahimian et al. reported the production of acetone, biobutanol, ethanol and hydrogen from OFMSW. The processes responsible for acetone and biobutanol could be removed and replaced with PHA production, because both processes require a carbon source. It would also be necessary to employ a microbial strain capable of metabolising the specific carbon source. Based on preliminary data from the system, it might also be necessary to balance the processes with sufficient organic material to provide the desirable yields. A life cycle assessment of bioplastics in South Africa was conducted by Harding et al. They found that bioplastics such as polyhydroxybutyrate (PHB) were superior in all life cycle categories among all plastic alternatives, such as polypropylene (PP) and polyethylene. Furthermore, PP production was found to release 80% more CO₂ compared to PHB, while ozone depletion was almost 50 times lower with PHB production. In the South African context, this is a clear indication that bioplastic will play an integral role in combating climate change. The integration of such a biorefinery in South African municipalities could significantly reduce the amount of waste that ends up in landfills, thus contributing to landfill space savings and the reduction of GHG emission from both landfills and processes employing conventional fuels. The South African Research Chairs Initiative (SARChI) Waste and Climate Change Group at the University of KwaZulu-Natal is poised to address many of these questions. The first stage will be the assessment of a laboratory-scale bioplastic biorefinery, taking into account life cycle and techno-economic analysis. The process will then be analysed using the Waste Resource Optimization Scenario Evaluation (WROSE™) model to assess its impacts through the evaluation of several key indicators, including GHG emissions; potential for waste diversion from landfills and related savings; technical and economic feasibility for scale up; job-creation potential; social acceptability; health risks associated with the jobs created; and institutional indicators for implementation. These data could provide critical insight into the feasibility of bioplastic biorefineries as a waste management tool.

Conclusions

Plastic pollution is a major environmental problem around the world, and impacts on almost all ecosystems. South Africa alone accounts for about 10 million tonnes of plastic waste, with an associated cost of ZAR885 billion, taking into account clean up, disposal costs and the impact on certain livelihoods. South Africa is also facing several challenges on the organic waste disposal front owing to the diminished capacity of many municipal landfills. Furthermore, the disposal of organic waste to landfills poses many problems, including the release of GHG that plays a pivotal role in climate change. For this reason, by coupling these two problems of plastic and organic waste together, it may be possible to produce a more environmentally friendly plastic using organic waste as a feedstock. Several studies conducted around the world have indicated the feasibility of this process. In order to construct this process to be environmentally and economically viable, a biorefinery system might be the best option, so that complete valorisation of the feedstock occurs, thereby producing PHA and multiple biofuels such as biomethane and biohydrogen. These fuels have the potential to offset the current carbon footprint trajectory, thus acting as a stabilisation wedge for climate change.

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Competing interests

We have no competing interests to declare.

Authors’ contributions

Both authors were responsible for the conceptualisation of the paper. PM was responsible for the writing process.

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