



Sustainable bio-plastic production through landfill methane recycling



Karthigeyan Chidambarampadmavathy^{a,c}, Obulisamy Parthiba Karthikeyan^{a,c,d},
Kirsten Heimann^{a,b,c,*}

^a College of Science and Engineering, James Cook University, Townsville 4811, Queensland, Australia

^b Comparative Genomics Centre, James Cook University, Townsville 4811, Queensland, Australia

^c Centre for Sustainable Tropical Fisheries and Aquaculture, James Cook University, Townsville 4811, Queensland, Australia

^d Sino-Forest Applied Research Centre for Pearl River Delta Environment (ARCPE), Department of Biology, Hong Kong Baptist University, Kowloon Tong, Hong Kong SAR

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ABSTRACT

Plastics are an indispensable part of day-to-day life. Environmental implications of these non-biodegradable plastics in landfills raise major concerns. Use of biodegradable plastics is the best alternative as they are environmental friendly, with great recycling potential, and can be produced using renewable resources such as waste materials, methane (CH₄) and simple carbon sources. Whilst the biodegradable plastics are eco-friendly, they pose a risk of emitting CH₄ under anaerobic conditions in landfills. As a cradle-to-cradle approach, landfill CH₄ could be effectively used for biodegradable plastic production by methanotrophs. This review briefly reviews approaches to plastic disposal, alternatives to plastic waste management and outlines issues arising. The focus of the review is to examine the potential for cost-effective production of polyhydroxybutyrate (PHB) using methanotrophs for manufacturing biodegradable plastics. The data input into this analysis is derived from Australian landfill CH₄ emissions, the average PHB content of methanotrophs and applied to a case-scenario in Sydney, Australia. The results suggest that this approach to biodegradable plastic production can be economically viable and price-competitive with synthetic plastics. In our case study, landfills were sized small, medium and large (5,000, 35,000 and 230,000 t of waste per year, respectively). In small landfills, 162 t of CH₄ can be recovered to produce 71 t of PHB per year, whilst in large landfills 7,480 t of CH₄ can be recovered to produce 3,252 t of PHB. The cost of PHB production can be reduced to 1.5–2.0 AUD meeting the market value of synthetic plastic by increasing production volumes through building a centralised extraction and refinement facility suitable for large metropolitan cities.

1. Plastic production, types and characteristics

Plastics are polymers synthesized chemically via polymerization from petroleum products [1] containing long chains of monomers [2]. The word ‘plastic’ is derived from the Greek word “plastikos”, meaning that materials are pliable for moulding into required shapes [3]. The development of the plastic industry over the last 60–70 years was driven by versatility, low weight, durability, and low costs. Plastics are indispensable commodities in our day-to-day life, with amounts required predicted to increase along with unprecedented human population growth. Annual production of plastics has increased dramatically from 1.5 million tonnes (1950) to 300 million tonnes (as of 2013) [4] and a four- to five-fold market growth has been predicted by the end of this century [5,6]. Eight percent of global oil and gas is used for plastic production, 4% as feedstock and ~3–4% as energy for manufacturing and transportation [7]. Fossil fuel resources

are non-renewable, as they take millions of years to form naturally, and continued use will result in depletion. It is estimated that 20–25% of current crude oil production (93 million barrels per day) [8] will be required for plastic-manufacturing by the end of this century [9].

There are various types of plastics widely used in many applications; for example in packaging products for food, pharmaceuticals, cosmetics, detergents and chemicals, in building materials, as well as in hygienic products [10,11]. Fifty percent of plastics are intended for single-use disposable applications [7]. In general, plastics are divided into two groups; (i) thermo-softening plastics, which can be re-moulded into different shapes rendering them ideal for recycling which includes polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS) polyvinylchloride (PVC), polycarbonate (PC) and nylon and (ii) thermos-setting plastics, which cannot be modified or re-softened, for example polyurethane (PU), epoxy resins and unsaturated polyester resin [12,13]. Commonly used plastics for

* Corresponding author at: College of Science and Engineering, James Cook University, Townsville 4811, Queensland, Australia.
E-mail address: kirsten.heimann@jcu.edu.au (K. Heimann).

packaging along with their annual production and applications are summarized in [Supplementary Table S1](#). In general, all plastic packaging is made of one of the following six resins, either polyethylene terephthalate, high density polyethylene, polyvinyl chloride, low density polyethylene, polypropylene or polystyrene, identifiable by numbering from 1 to 6 inside a triangle of clock-wise arrows (S1) called the resin identification code (RIC). All other resins, which are also recyclable, are grouped under the number 7 or “other” [14,15]. Representatives of this group of plastics are acrylonitrile butadiene styrene (ABS), polyurethane, nylon, polycarbonates and phenolics. A complete overview of properties and applications of plastics can be found elsewhere [3,12,16–18].

2. Plastic disposal and challenges in recycling plastics

One of the prime problems with the use of plastics is their long-lasting effects on the environment [19]. Most of the synthetic polymers are extremely durable and can remain in the environment for up to thousands to millions of years, depending on the type of plastic [20]. Large quantities of synthetic plastics have entered the environment for the past 70 years due to cheap mass production and have become one of the most common and persistent pollutant. There are many options available for the disposal of plastics like landfilling, incineration and energy recovering, gasification and pyrolysis for converting plastic to synthetic gas or fuel, recycling and reuse [7], each with its own disadvantages and environmental hazards. A variety of methods have been identified for decomposing synthetic plastics e.g. chemical depolymerisation to yield non-harmful molecules, or re-processing by mechanical recycling (shredding, melting and re-moulding) or through biological degradation (enzymatic conversion into compostable organic material). These methods are, however, quite complex and face challenges on large scales. At present, recycling, landfill disposal and incineration are the most widely adopted methods for disposal of plastics in Australia ([Fig. 1a](#)).

2.1. Recycling

Recycling is one of the primary option available to reduce environmental impacts associated with the disposal of plastics, reducing carbon dioxide (CO₂) emissions, fossil fuel use and minimizing landfill deposition [7,21,22]. However, many plastic resins are difficult to recycle and sorting of recyclable plastics can be challenging, involving extensive labor costs [22,23]. Furthermore, recycled plastics yield different products, as recycling into the same product with the same quality is not possible because the process alters the chemical properties of the original material. Recycling of plastics results in shortening of the polymer chains, with weakened strength and durability, making it unsuitable for high-end use [21].

2.2. Incineration

Incineration is a common management practice for plastics, mainly applied to reduce the footprint of landfill facilities. The process is, however, very expensive and dangerous [7]. Furthermore, the potential release of hazardous substances such as dioxins, polychlorinated biphenyls and furans into the atmosphere is of concern ([Fig. 1a](#)) [24]. Remediation technologies with strict environmental regulations (e.g. National Pollutant Discharge Elimination System, Toxic Substances Control Act Clean Air Act) are presently in use, where incineration is the primary mode of waste management [25]. For example, in modern incinerators, the released gas is subjected to a high temperature (~850 °C) to thermally crack these pollutants [26]. More details on current technologies used to treat these environmental pollutants are reviewed elsewhere [27,28]. Also, plastic incineration generates greenhouse gases (GHGs) such as methane (CH₄) and CO₂, which are thought to lead to worldwide climate change. When compared to landfill disposal (254 g of CO₂ kg⁻¹ plastic), incineration emits more CO₂ (343–4605 g of CO₂ kg⁻¹ plastic) [29]. Thus in terms of environmental footprint incineration is not a best option for treating/managing plastics.

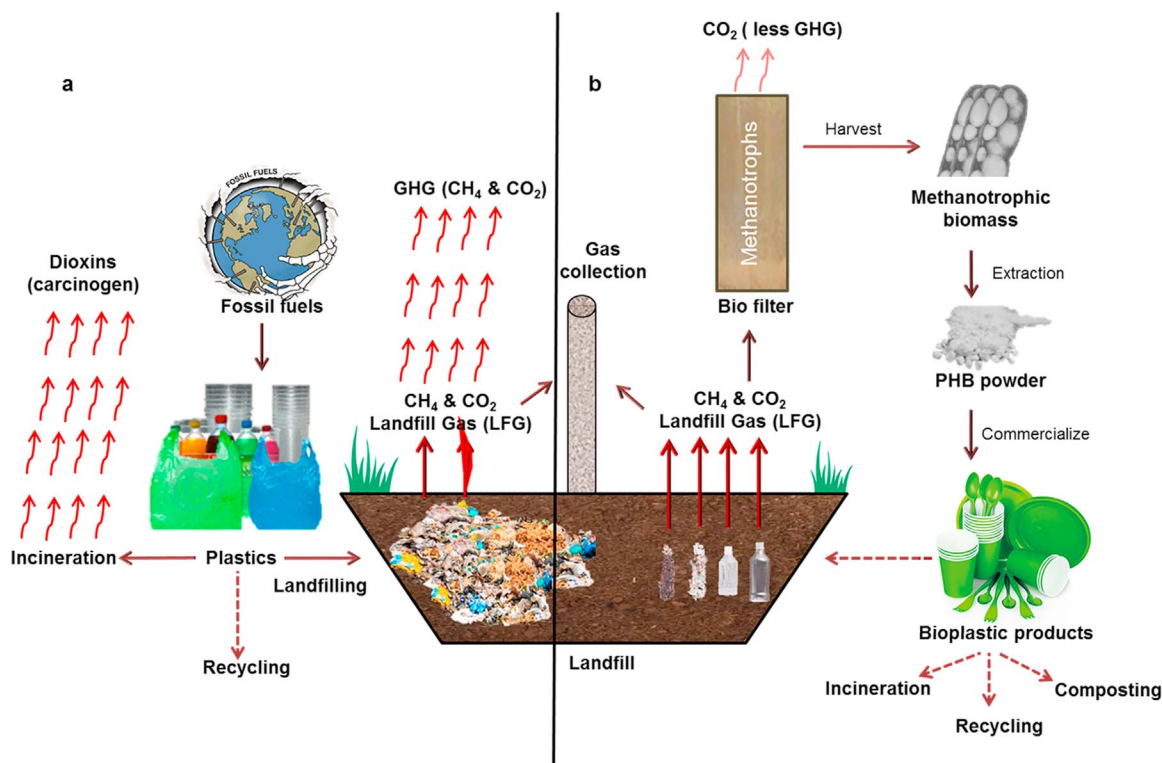


Fig. 1. End of life landfill degradation of a) plastics and b) bio-plastics.

2.3. Landfill disposal

Annually, millions of tonnes of plastics (60% of the produced plastic) are being disposed of in landfill [7]. Landfilling is the most convenient approach for waste management, but space constraints in developed countries and countries with high population densities limits this management practice [23]. Most plastic packaging and non-recyclable plastics are disposed-off in landfills. Globally, plastic accounts for 18–20% of the waste volume in landfill [30,31]. From a sustainability perspective, there are some major drawbacks to waste management of plastics in landfills (Fig. 1a), i.e. leachates are toxic adversely affecting soil microbial flora, leading to soil infertility and/or ground water and water supply contamination [32,33] and the toxins and/or the packaging of organic waste in bio-inert and impervious plastic bags result in persistence of easily biodegradable waste [19,34]. Even targeted degradation in composting facilities still requires the separation of degradable plastics from non-degradable ones.

As stated above, in addition to the commonly adopted methods for plastic disposal, other methods like gasification/pyrolysis and mechanical biological treatment are also in use. Gasification/pyrolysis gained significant attraction recently due to capacity of converting a variety of plastics to synthetic gas/petrochemical fuels, requiring only high temperatures to thermally crack the plastics [11,25,26,35,36]. Gasification at around 600–800 °C results in oil and condensable gases and the mixture is passed through a gasifier with air and steam at 1200–1500 °C to obtain synthetic gas (composed of hydrogen, carbon monoxide, CO₂, CH₄ and steam). In contrast, during the pyrolytic conversion of plastic, materials are thermally cracked (~500 °C) in the absence of oxygen to generate a combination of condensable vapours, which upon cooling, form various petrochemical fuels (oils, chars and waxes). Unlike incineration, gasification/pyrolysis does not allow formation of dioxins and or any other pollutants [26]. However these techniques are expensive and the synthetic gas/petrochemicals produced in large-scale application requires purification [35]. Mechanical biological treatment, as the name suggests, is a combination of mechanical and biological treatment [37], whereby the recyclable wastes are segregated and then biologically treated before being landfilled. Biological treatment usually involves composting/anaerobic digestion of the wastes left after mechanical treatment [37]. While this treatment is very effective for mixed waste (containing cardboard, glass, metals, degradable and non-degradable plastics, rubber, and biodegradable waste), it is not a best method for treating plastic-only containing wastes.

Considering some of the drawbacks of current practices and the growing demand for plastics, there is an urgent need for environmentally and economically sustainable alternatives, which should aim to simultaneously reduce the exploitation of limited feed fossil petroleum resources.

3. Alternatives to conventional plastics

For the past 30–40 years, research has focused on alternative methods for producing plastics using renewable materials.

Table 1
Energy requirement and CO₂emissions for synthetic plastics and bio-plastics.

Type of plastic	Synthetic plastics							Bio plastics	
	HDPE	LDPE	PP	Nylon	PET	PS	PVOH	PLA	PHB
Energy requirement (MJ/kg of polymer)	80	80.6	85.9	120	77	87	102	57	44.7
CO ₂ emissions (kg of CO ₂ eq/kg of polymer)	4.8	5.1	3.4	7.6	4.93	6.0	2.7	3.8	2.6

HDPE: High-density polyethylene, LDPE: Low-density polyethylene, PP: Polypropylene, PET: Polyethylene terephthalate, PS: Polystyrene, PVOH: Polyvinyl alcohol, PLA: Polylactic acid, PHB: Poly-3-hydroxybutyrate.

Economically sustainable production of biologically derived plastics has gained renewed interest, as this approach promises to solve landfilling problems of non-degradable plastics and reduce fossil fuel use and greenhouse gas emissions.

Bio-plastics are defined as polymeric materials derived from renewable carbon sources that naturally decompose naturally to CO₂ and water under aerobic conditions or CH₄, CO₂ and water under anaerobic conditions [38,39], making them sustainable and eco-friendly. The theoretical concept behind this approach is the cradle-to-grave-to-cradle approach stemming from landfilling of recyclable bio-plastics (Fig. 1b). One of the main challenges for bio-plastics is retention of the physico-chemical properties of petroleum-derived plastics, whilst keeping the properties of bio-compatibility and bio-degradability [40,41]. Bio-plastics are currently being produced using three approaches; (i) use of natural polymers (e.g. starch-based plastics), (ii) polymerization of bio-based monomers and oligomers *via* fermentation or conventional chemical processes (e.g. polylactic acid), and (iii) use of microbial polymer (polyhydroxyalkanoates) synthesis.

The two most promising bio-plastics that are considered as viable alternatives to synthetic plastics are polylactic acid (PLA) and polyhydroxyalkanoate (PHA).

3.1. Polylactic acid

PLA is a type of aliphatic thermo-softening polyester derived from the chemical polymerization of D- and L-lactic acid obtained by the fermentation of sugar beet, maize (corn), sugarcane or wheat [42,43]. PLA plastics have similar properties to petroleum-derived PET with a glass transition temperature (*T_g*) of 55 °C, offering improved resistance to moisture penetration of packaged materials [44]. PLA is currently utilized in the production of loose-fill packaging, food packaging, beverage containers, and disposable food service tableware items. PLA is also used extensively in biomedical applications, such as sutures, stents, dialysis devices, drug capsules, and are being evaluated as a matrix for tissue engineering [45–47]. PLA degrades quickly in composting facilities 4–6 weeks), similar to paper [48]. Although production requires 20–50% less fossil fuels compared to petro-plastics [39,49], energy consumption is higher than for PHA (Table 1). At present, PLA is 25% more expensive than its petroleum-based counterpart, PET [31].

3.2. Polyhydroxyalkanoates (PHA)

Polyhydroxyalkanoates (PHA) are aliphatic polyesters produced by microbes via conversion of various carbon substrates for intracellular carbon and energy storage [40,41,50]. PHAs are mainly classified based on the number of carbon atoms and the type of monomeric units, e.g. polymers with 3–5 carbon atoms are considered short chain-length PHA, whilst medium chain-length PHA contain 6–14 carbon atoms [41].

Among the bio-degradable plastics, PHAs have similar material properties to conventional plastics [51], with physical [52,53] and mechanical properties being tunable by changing co-polymer contents

[50,52]. PHA can be used in a wide range of short-term packaging applications, such as plastic films for bags and other diverse packaging applications, containers and paper coatings, in disposable articles (personal care products, surgery clothes), upholstery [50,53]. As PHAs are biodegradable and immunologically inert, they have promising future medical applications, despite expensive production [40,50,53].

The most commonly studied PHA is poly-3-hydroxybutyrate (PHB), produced by several Gram-positive and Gram-negative bacteria, including methanotrophs/ methane-oxidizing bacteria [50,53–57]. PHB polymers have desirable characteristics similar to PP such as moisture resistance, water insolubility, oxygen impermeability, resistance to UV weathering, indefinite stability in air and a higher optical purity than PP [41,58].

Taken production into consideration, being a microbial carbon and energy store, PHBs are suitable for a full cradle-to-cradle waste management approach (Fig. 1b). The total energy requirement and the amount of CO₂ emitted to produce a kilo of polymer for both synthetic and bio-plastic is compared in Table 1. Compared to commercially available bio-degradable and synthetic plastics, production of PHB requires only 44.7 MJ/kg being 50–70% lower than synthetic plastics and emitting 2.6 CO₂eq/kg of polymer, whilst HDPE, LDPE, PP, Nylon, PET, PS production emits 4.8, 5.1, 3.4, 7.6, 4.9, and 6.0 CO₂eq/kg polymer, respectively [52,59].

3.3. Synthesis of polyhydroxyalkanoates

PHA are synthesized via bacterial fermentation, in genetically modified plants, and using enzymatic catalysts in cell-free systems. At present, microbially synthesized PHA production, i.e., PHB, is the only method available at industrial-scale. Microbial PHBs are commercially produced using a number of carbon-sources as feedstocks such as wheat bran, whey, molasses, cane starch, palm oil, cassava waste, sucrose, glucose [49,52], which constitute more than 30–50% of the production cost [60]. Feedstock provision for PHB-producing microbes is therefore the limiting and cost-prohibitive factor for mass production of microbial PHB. As the price of PHB is strongly influenced by the substrate and bacteria used, costs of PHB can range from USD 4–16 per kg of polymer. For example, PHB produced from *Cupriavidus necator* (formerly *Alcaligenes eutrophus*) costs USD 16 per kg of polymer, which is more expensive than polypropylene, whilst production using recombinant *Escherichia coli* reduces costs to USD 4 per kg of polymer, which is comparable to other bio-degradable plastic materials such as PLA and aliphatic polyesters. Despite production cost challenges, the global bio-plastic production is expected to increase from 890,000 t to 2.5 million metric tonnes (Mt) from 2012 to 2017 [61], expected to reach 1.5–4.4 Mt by 2020, globally [60].

Production area requirements for feedstocks also hamper viability [60,61], as increase in production could lead to competition for arable land between food and feedstock crops. According to the Food and Agriculture Organization (FAO) of the United Nations, world food prices reached record highs in 2010 with a Food Price Index of 214.7 points comparable to the food crisis in 2008 (213.5 points) [62].

Given the above, re-routing of unutilized/wasted CH₄, a potent greenhouse gas from landfills and other sources, as a carbon source for PHB production (Table 2) may have significant advantages for reducing production costs associated with feedstock, land use, and energy requirements [54,56,63,64]. Using CH₄-fed methanotrophs/ methane-oxidizing bacteria for PHB production has significant advantages, such as sequestering the greenhouse gases CH₄ and CO₂ (60% of oxidized CO₂ from CH₄ is incorporated for biomass growth [65]), higher accumulation of PHB (maximal ~70% (i.e., 0.70 g g⁻¹ dry biomass) and more than 50–56% on an industrial-scale) [54,56].

4. Case study of PHB production from methane

As shown in Fig. 1b, like synthetic plastics, bio-plastics can also be recycled, incinerated and landfilled or composted. Landfilling of bio-plastics is considered the best option because gases released from landfills usually contain 30–70% of CH₄ and 20–50% of CO₂, but the gas composition changes with landfill age [63]. Release of CH₄ due to the degradation of bio-polymers induces a cradle-to-cradle production approach, as the emitted CH₄ can be effectively used for renewed PHB production (Fig. 1b).

In order to evaluate the theoretical merits of rerouting CH₄ for PHB production using methanotrophs, a case study was carried out based on Australian landfill waste management practices. Solid waste management in Australia relies on landfilling (Fig. 2). A total of 1168 landfills exist in Australia, with highest landfill densities in New South Wales (Table 3). Apart from landfills, there are 872 transfer stations where wastes are sorted for recycling and 806 resource-recovery facilities where wastes are processed using mechanical, biological or thermal transformation technologies in addition to sorting [66]. Landfills in Australia vary greatly in size; small landfills receive on average 5,000 t of wastes, whereas large modern waste gas-managed landfills receive more than 100,000 t of waste annually [66,67].

Annually, 48 million tonnes of solid waste are generated in Australia, of which 40% (~20 Mt) are disposed in landfills. Of the 20 Mt of waste, 14 Mt are organic waste [66], releasing 0.65–1.53 t of carbon dioxide equivalent (CO₂eq) per tonne of waste [67]. Methane emissions from solid waste are estimated based on the first order decay method [68] and, in general, a 25 times multiplier is applied (CH₄ has a 100 year-warming potential 25 times that of CO₂) [69,70] for converting CH₄ emissions to CO₂eq to create a comparative base for all greenhouse gas emissions. Thus, 14 Mt of waste emit 9.1–21.42 Mt of CO₂eq, resulting in an average of 15.26 Mt of CO₂eq (i.e. 0.6 Mt of CH₄ are emitted annually in Australia). In order to effectively use the emitted CH₄ for PHB production, the following key factors have to be considered:

Life of each landfill is estimated to be 30 years and CH₄ can be collected from the time of opening to 30–50 years after closure of each landfill [67]. CH₄ emission will vary depending on the climate and the age of the landfill. Using gas collection systems from well-engineered landfills, a maximum of 95% with an average of 75% of emitted CH₄ can be recovered [67]. The energy required to run a PHB production plant can be derived using 18–26% of the captured CH₄, leaving 74–82% for use as feedstock for PHB production [61,63]. Using 1.13 g of CH₄, 0.5–0.6 g of PHB can be produced [56,63].

The following factors were used to calculate one tonne PHB production from CH₄.

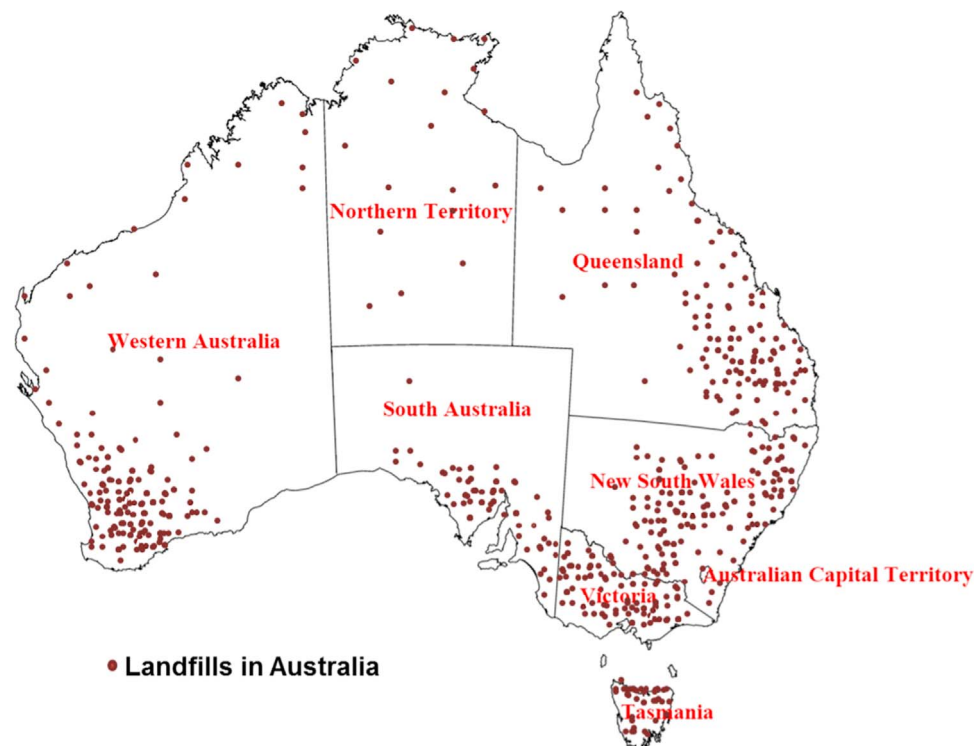
- One tonne organic waste releases 0.65–1.53 t of CO₂eq (average 1.09 t of CO₂eq). Thus multiplying 14 Mt of waste and with average 1.09 t of CO₂eq emission results in 15.26 Mt of CO₂eq i.e., 0.6 Mt of CH₄ (total CO₂eq emission divided by 25, the global warming potential of CH₄ for 100 years [69,70]).
- Of the 0.6 Mt of CH₄, 0.45 Mt of CH₄ can be recovered based on 75% CH₄ recovery.
- To produce one tonne of PHB at a yield rate of 0.56 gPHB.g⁻¹CH₄, 1.79 t of CH₄ are required as a carbon source for production and another 0.52 t of CH₄ for energy (Table 4) for the production process [61]. Thus the production of one tonne PHB requires a total of 2.3 t of CH₄. Based on 0.45 Mt of total CH₄ input and the ratio of CH₄ required for production and energy, a maximum of ~196 ktonnes of PHB can be produced annually.

For a small landfill, with an average annual disposal of 5,000 t of wastes, 163 t of CH₄ can be recovered annually and, using the same model calculations applied before, can yield 71 t of PHB, whilst medium and large landfills with average annual disposal of 35, 000

Table 2

Sustainability of bio-based plastics: general comparative analysis.

Bio Polymer	Energy requirement (MJ/kg polymer)	Yield	Advantages/Disadvantages
PLA	57 [52,59]	–	<ul style="list-style-type: none"> ● GMO crops pose unknown risk to the environment [78]. ● PLA uses organic tin and 1-octanol for lactic acid polymerization. 1-octanol is toxic to the environment [79,80].
PHB from sugars	44.7 [52,59]	0.34 [81] ($\text{g}_{\text{PHB}} \cdot \text{g}^{-1}_{\text{glucose}}$)	<ul style="list-style-type: none"> ● Uses agriculture products as feedstock including GMO crops, posing land usage conflicts which could also increase food prices. GMO microorganism, like GMO crops might pose an unknown risk to the environment [78,79].
PHB from CH_4	37.4 [63]	0.50 [64] ($\text{g}_{\text{PHB}} \cdot \text{g}^{-1}_{\text{CH}_4}$)	<ul style="list-style-type: none"> ● No feedstock costs, as it uses waste CH_4 emitted from landfills, intensive agriculture and coal mines. ● Completely renewable and with significant advantages over corn/sugar based plastic [54,56,82].

**Fig. 2.** Distribution of landfills in Australia.**Table 3**

Australia's waste management infrastructure.

Source: Blue Environment and Randell Environment Consulting [66]. **NSW:** New South Wales, **Vic:** Victoria, **QLD:** Queensland, **WA:** Western Australia, **SA:** South Australia, **NT:** Northern Territory, **ACT:** Australian Capital Territory.

Jurisdiction	NSW	VIC	Qld	WA	SA	Tasmania	NT	ACT	Total Facilities
Number of Landfills	369	92	265	187	117	19	118	1	1168
Resource Recovery Facility	121	233	88	86	2471	14	10	6	806
Transfer Station	166	239	236	26	133	67	4	1	872

Table 4

Base factors for net PHB calculation.

CH_4 recovery[*] (%)	75%
Yield of PHB^{**} ($\text{g}_{\text{PHB}} \cdot \text{g}^{-1}_{\text{CH}_4}$)	0.56
Amount of CH_4 required for PHB production^{***} (per tonne)	As carbon source 1.78
	As energy 0.52

^{*} On an average 75% of CH_4 can be recovered [67].

^{**} 1.13 g of CH_4 , can produce 0.56 g of PHB.

^{***} Refer Roland-Holst et al. [61] for the model calculation.

and 230,000 t of wastes and 1138 and 7480 t of CH_4 recovery can produce 495 and 3252 t of PHB (refer to Tables 4 and 5 for base-factor inputs).

Listewnik and co-workers [64] published a process design for synthesis of PHB using methanotrophs fed with natural gas (96% CH_4) as a carbon source. Estimated costs to produce 1 kg of PHB in a plant producing 500 t PHB per year was AUD ~10.50 (Table 6).

However, by increasing production of PHB, the cost can be considerably reduced. Criddle and co-workers [31] estimated that the costs per kg of PHB can be reduced to 1.5–2.0 AUD based on using natural gas as a feedstock and increasing the production volumes of

Table 5
Annual waste disposal in Australian landfills.

Landfill size	Amount of waste disposed (tonnes/year)	Assumed annual disposal (tonnes/year)	CH ₄ emissions (tonnes/year)	Average CH ₄ recovery (tonnes/year)	Net PHB production (tonnes/year)	Energy used (tonnes/year)
Small	< 10,000	5,000	216	163	71	65
Medium	10,000–100,000	35,000	1518	1138	495	455
Large	> 100,000	230,000	9973	7480	3252	2993

Annual waste disposal data were given based on BDA group and Blue Environment and Randell Environment Consulting [66,67].

Table 6
Estimated production costs for 1 kg of PHB using CH₄ as feed stock.
Source: Listewnik et al. [64].

Type of cost	Value in AUD*
Amortization and financing	2.9
Service and maintenance	0.16
Energy and water	2.5
Raw materials (methane/natural gas)	2.15
Auxiliary materials	1.0
Personnel costs	1.8
Total	10.51

* Australian Dollars (AUD) value given is calculated based on rate of Euros in 2007.

Table 7
Market prices for various polymers.
Source: Criddle et al. [31]. All prices are in AUD, market value based on 2012.

Polymer Type	PS	PP	PET	PLA	PHB (sugar)	PHB (CH ₄)
Market Price (\$/kg)	1.87–2.14	1.38–1.43	1.7–1.9	2.0–2.5	3.5–5.0	1.5–2.0

PHB from 500 t to 5,000 or 10,000 t per year. This is in contrast to a recent estimated production cost of 4.6–7.6 AUD per kg of PHB in a 100,000 t plant, which could be further reduced to 3.6–6.1AUD per kg of PHB using thermophilic methanotrophs instead of moderate temperature strains, the latter adding cost due to required temperature control of the process [71]. It should be noted, however, that the PHB content of thermophilic methanotrophs has not been established. Furthermore, most of the thermophilic methanotrophs use the Ribulose Monophosphate (RuMP) pathway, while PHB synthesis in methanotrophs is linked to the serine pathway [57]. Because of these limitations, the techno-economic model analysis of Criddle and co-workers was employed [31], basing calculations on realistic PHB content estimates (0.56 gPHB g⁻¹CH₄) [31,54,64] to discuss production costs of PHB from CH₄ emissions from Australian landfills.

A comparison with the market price of PLA and other petroleum-based polymers confirms that PHB production using CH₄ could be an attractive option even to date (Table 7), when prices for the raw material are most likely considerably lower than they would be once fossil fuel reserves become severely limiting, resulting in higher prices for material inputs into non-degradable plastics. Based on current prices, PHB production in small and medium landfills would be possible, but the low volumes of PHB produced would result in non-competitive pricing of the product. However, larger landfills can produce a minimum of 3252 t of PHB making the process more

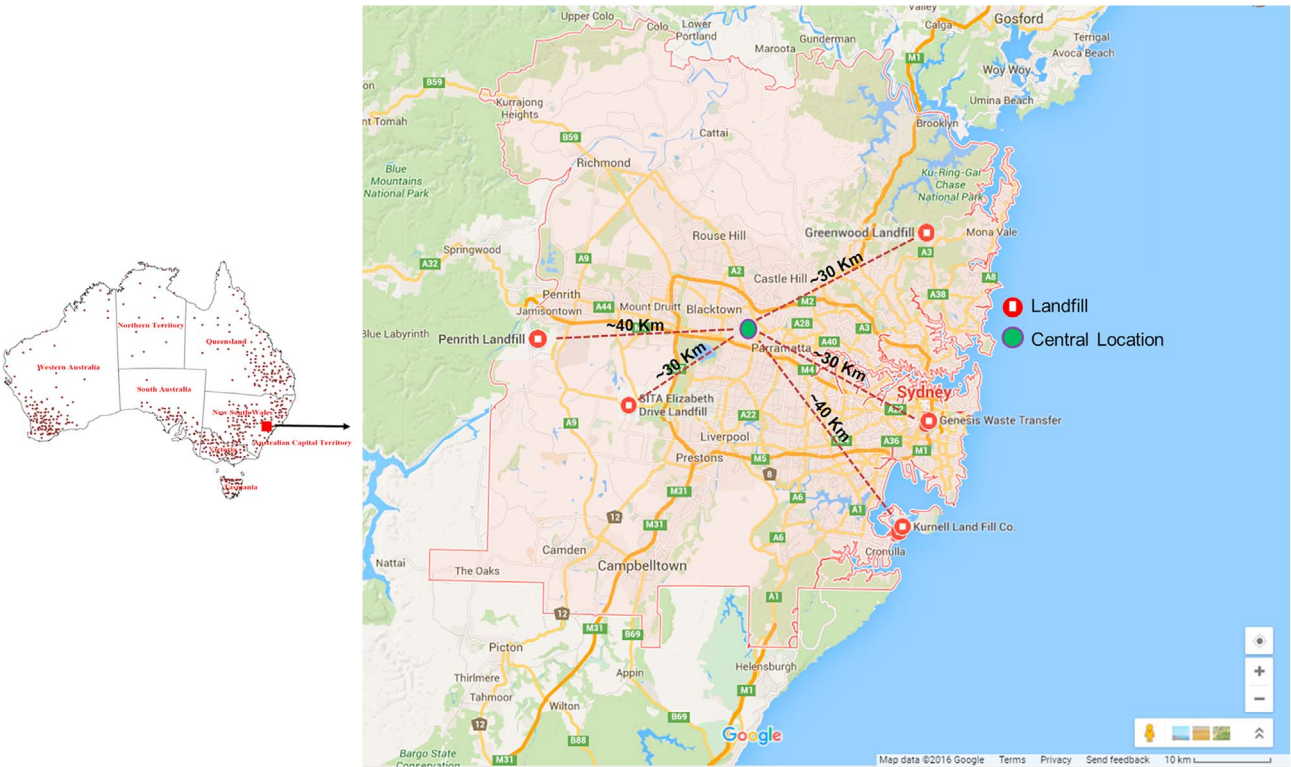


Fig. 3. Model scenario for a centrally located biomass receiving PHB extraction and refining facility in Sydney, Australia.

economical. In large metropolitan areas, it may be possible to create a central PHB extraction and refining facility, which would allow to pool PHB containing biomass from a variety of landfills, which should make the product cost-effective even based on today's low prices. Major metropolitan cities with high population densities like Sydney, Melbourne, Perth, and Brisbane have many large landfills. Hence, we tested this concept, applying it to a case scenario for Sydney.

Sydney is one of the largest and most populated Australian city (4.67 million people as of 2012) [72]. The daily amount of waste produced per person in Australia is 2.2 kg per person [73]. Of this, nearly ~50% is organic waste [74], resulting 1.1 kg per person per day of waste suitable for CH₄-rerouting into PHB production. This yields ~5000 t of waste per day or 1.8 Mt per year. Almost 75% of generated wastes in Sydney are disposed of primarily in five larger landfill (Fig. 3), or smaller or medium landfills in and around it (not considered in this case study). Life cycle analyses for biofuel and bio-refinery feedstock indicate a maximum 50–80 km (km) radius for a central biomass receiving facility as economical and energy-sustainable considering transport logistics [75–77]. Therefore, as the price for PHB is somewhat comparable to that for biofuels, this case analysis assumed a planned central PHB extraction and refining facility within a radius of ~50 km for the five larger landfills (Fig. 3). The biomass grown, using emitted CH₄ at these five larger landfills, can be transported to this central location for extraction of PHB yielding ~25,500 t per year (assumed PHB production based on above calculations) which would ensure PHB can be produced at a market value of 1.5–2.0 AUD [31].

5. Recommendations

Plastics are a critical component of our day-to-day lives. Based on predicted population growth and finite fossil fuel resources, economically and environmentally sustainable production of bio-degradable bioplastics using completely renewable resources will be mandatory for future generations. Microbial production of PHB can meet the future requirements only, if produced from microbes using waste GHGs, as this avoids competition for agricultural food products and land use. The Australian case study, presented here, shows that economically viable production is possible in Australia, even without including any carbon tax offsets, which offers an integrated approach to landfill management. The case study also shows that for production of PHB with competitive market pricing, designing a centralized extraction and refining facility for pooling of PHB-containing biomass will be necessary. In addition, to market value generated through PHB production, reductions in GHG emissions could provide another incentive for modernizing landfill management, should Australia re-consider its stance on carbon tax or equivalent incentives/penalties for GHG polluters.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at <http://dx.doi.org/10.1016/j.rser.2016.12.083>.

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